INFORMATION TO USERS

While the most advanced technology has been used to photograph and reproduce this manuscript, the quality of the reproduction is heavily dependent upon the quality of the material submitted. For example:

- Manuscript pages may have indistinct print. In such cases, the best available copy has been filmed.

- Manuscripts may not always be complete. In such cases, a note will indicate that it is not possible to obtain missing pages.

- Copyrighted material may have been removed from the manuscript. In such cases, a note will indicate the deletion.

Oversize materials (e.g., maps, drawings, and charts) are photographed by sectioning the original, beginning at the upper left-hand corner and continuing from left to right in equal sections with small overlaps. Each oversize page is also filmed as one exposure and is available, for an additional charge, as a standard 35mm slide or as a 17”x 23” black and white photographic print.

Most photographs reproduce acceptably on positive microfilm or microfiche but lack the clarity on xerographic copies made from the microfilm. For an additional charge, 35mm slides of 6”x 9” black and white photographic prints are available for any photographs or illustrations that cannot be reproduced satisfactorily by xerography.
Gamma-ray optics of multi-beam Borrmann modes with possible applications to nuclear lasing

Hutton, J. Timothy, Ph.D.

Rice University, 1986
PLEASE NOTE:

In all cases this material has been filmed in the best possible way from the available copy. Problems encountered with this document have been identified here with a check mark ✓.

1. Glossy photographs or pages
2. Colored illustrations, paper or print
3. Photographs with dark background
4. Illustrations are poor copy
5. Pages with black marks, not original copy
6. Print shows through as there is text on both sides of page
7. Indistinct, broken or small print on several pages ✓
8. Print exceeds margin requirements
9. Tightly bound copy with print lost in spine
10. Computer printout pages with indistinct print
11. Page(s) ________ lacking when material received, and not available from school or author.
12. Page(s) ________ seem to be missing in numbering only as text follows.
13. Two pages numbered _______. Text follows.
14. Curling and wrinkled pages
15. Dissertation contains pages with print at a slant, filmed as received ✓
16. Other ____________________________________________

University
Microfilms
International
RICE UNIVERSITY

GAMMA-RAY OPTICS OF MULTI-BEAM BORRMANN MODES
WITH POSSIBLE APPLICATIONS TO NUCLEAR LASING

by

J. TIMOTHY HUTTON

A THESIS SUBMITTED
IN PARTIAL FULFILLMENT OF THE
REQUIREMENTS FOR THE DEGREE

DOCTOR OF PHILOSOPHY

APPROVED, THESIS COMMITTEE:

G. T. Trammell, Chairman
Professor of Physics

J. P. Hannon
Professor of Physics

W. L. Wilson, Jr.
Professor of Electrical and
Computer Engineering

Houston, Texas
August, 1986
ABSTRACT

GAMMA-RAY OPTICS OF MULTI-BEAM BORMANN MODES
WITH POSSIBLE APPLICATIONS TO NUCLEAR LASING

by

J. TIMOTHY HUTTON

There is a possibility that an isomer can be found which could be used to obtain a sufficient population inversion density to bring about nuclear lasing. If it can be realized, a gamma ray laser ("graser") will be a new source of radiation in the ~ 1A region of unexcelled spectral brightness and coherence, and can be expected to have revolutionary consequences for physics, biology, and chemistry.

The result of our research is that the effects of crystallinity are of crucial importance in considerations regarding gamma-ray lasers, bringing in new elements to laser theory which could very well prove decisive in determining whether grasing can or cannot be achieved.

Certain multi-beam Bormann eigenmodes of the radiation field in a perfect crystal have much stronger coupling to the emitting nuclei, and much less photoabsorption, than modes in the amorphous case. These modes may be selectively fed by nuclear transitions of the proper multipolarity. A crystal containing such emitters would require orders of magnitude less population inversion density in order to lase than would an amorphous sample of similar material.

Multi-beam Bormann modes also have several applications to single-photon optics, particularly the problem of anomalous emission of a γ-ray by an internal source of multipolarity M1 or higher. While
this effect can occur for a 2-beam Borrmann mode, even stronger anomalous emission is possible into multi-beam Borrmann modes. In addition, certain multi-beam Borrmann modes can anomalously transmit resonant radiation through a Mössbauer crystal. (This cannot occur for the 2-beam modes, except for the case of an E1 transition, since the Borrmann mode couples strongly to the nuclei and thus will be strongly absorbed at resonance.)
ACKNOWLEDGEMENTS

I would like to thank the members of my committee, but particularly George Trammell and Jim Hannon for their patience and invaluable assistance. I would also like to thank George Baldwin, James Farrell, and Dean Taylor of Los Alamos National Laboratory for their helpful discussions. Most of all, I would like to thank my wife for her support and for putting up with me while all this was going on.
# TABLE OF CONTENTS

<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>ABSTRACT</td>
<td>ii</td>
</tr>
<tr>
<td>ACKNOWLEDGEMENTS</td>
<td>iv</td>
</tr>
<tr>
<td>LIST OF ILLUSTRATIONS</td>
<td>vii</td>
</tr>
<tr>
<td>I. INTRODUCTION</td>
<td></td>
</tr>
<tr>
<td>II. GENERAL THEORY ON MULTI-BEAM MODES</td>
<td>8</td>
</tr>
<tr>
<td>Crystal Diffraction Theory</td>
<td>8</td>
</tr>
<tr>
<td>Multi-Beam Eigenmodes</td>
<td>21</td>
</tr>
<tr>
<td>General Structure of the Multi-Beam Modes</td>
<td>21</td>
</tr>
<tr>
<td>Symmetry Properties of the Eigenmodes</td>
<td>27</td>
</tr>
<tr>
<td>Finding the Eigenvalues</td>
<td>31</td>
</tr>
<tr>
<td>The Borrmann Effect and Anomalous Emission</td>
<td>37</td>
</tr>
<tr>
<td>III. PROPERTIES OF 2-BEAM BORRMANN MODES</td>
<td>42</td>
</tr>
<tr>
<td>IV. PROPERTIES OF MULTI-BEAM BORRMANN MODES</td>
<td>56</td>
</tr>
<tr>
<td>Borrmann Modes Corresponding to 1-D Representations</td>
<td>57</td>
</tr>
<tr>
<td>Modes Formed on Regular Polygons</td>
<td>57</td>
</tr>
<tr>
<td>Modes Which Are Not Formed on Regular Polygons</td>
<td>76</td>
</tr>
<tr>
<td>Borrmann Modes Corresponding to 2-D Representations</td>
<td>82</td>
</tr>
<tr>
<td>V. NUCLEAR LASING IN A BORRMANN MODE</td>
<td>94</td>
</tr>
<tr>
<td>Conditions for Lasing to Occur</td>
<td>94</td>
</tr>
<tr>
<td>Effects of Changes of $g_A$ and $g_N$</td>
<td>97</td>
</tr>
<tr>
<td>Lasing Mode Selection</td>
<td>99</td>
</tr>
<tr>
<td>Angular Divergence of Multi-Beam Borrmann Modes</td>
<td>105</td>
</tr>
<tr>
<td>VI. APPLICATIONS TO SINGLE PHOTON OPTICS</td>
<td>108</td>
</tr>
<tr>
<td>The Anomalous Emission Effect</td>
<td>108</td>
</tr>
<tr>
<td>Anomalous Transmission in the Presence of Resonant Absorbers</td>
<td>113</td>
</tr>
<tr>
<td>VII. CONCLUSION</td>
<td>117</td>
</tr>
</tbody>
</table>
LIST OF ILLUSTRATIONS

1. a) Crystal Geometry Used in Developing the Dynamical Theory. b) Radiation Channels for a Plane Layer 9
2. Diagram Showing Two Wavevectors Which Satisfy the Bragg Condition for Reflecting From a Particular Set of Planes Within a Crystal 17
3. General Structure of a Multi-Beam Mode 22
4. Reflecting Planes Which Correspond to a Particular Reciprocal Lattice Vector 23
5. Multi-Beam Modes Possible in Crystals With 3 and 4-fold Symmetry Axes 24
6. Mode Formed in a Cubic Crystal When All Eight Corners of a Cube Simultaneously on the Ewald Sphere 26
7. Standing Wave Electric Fields in the 2-Beam Borrmann and Anti-Borrmann Modes at Exact Bragg 38
8. Two-Beam Mode Geometry 42
9. Two-Beam Eigenmodes 44
10. General Structure of the Multi-Beam Modes 56
11. Excitation Patterns of the 3, 4 and 6-Beam \( |m_o>_{m} \) Borrmann Modes 58
12. Plot of \( g_{A,11} \) and \( g_{A,21} \) vs. Cone Angle For the 3, 4, and 6-Beam \( |m_o>_{m} \) Borrmann Modes 63
13. Plot of \( g_{N,10} \) and \( g_{N,20} \) vs. Cone Angle For the 2, 3, \( \frac{N}{2} \), and 6-Beam \( |m_o>_{m} \) Borrmann Modes 66
14. Excitation Patterns For the \( |2\alpha>_{y} \) and \( |2\pi>_{y} \) Borrmann Modes 67
15. Plot of \( g_{A,11} \) and \( g_{A,21} \) vs. Cone Angle For the \( |2\alpha>_{y} \) and \( |2\pi>_{y} \) Borrmann Modes 69
16. Plot of \( g_{N,20} \) and \( g_{N,21} \) vs. Cone Angle For the \( |2\alpha>_{y} \) and \( |2\pi>_{y} \) Borrmann Modes 70
17. Excitation Patterns For the \( |3\alpha>_{6} \) and \( |3\pi>_{6} \) Borrmann Modes 73
18. Plot of \( g_{A,11} \) and \( g_{A,21} \) vs. Cone Angle For the \( |3\alpha>_{6} \) and \( |3\pi>_{6} \) Borrmann Modes 75
19. Plot of $g_{N,30}$ and $g_{N,31}$ vs. Cone Angle For the $|3\sigma>_{6}$ and $|3\pi>_{6}$ Borrmann Modes . 76

20. Examples of 8-Beam Eigenmodes Which Can Be Formed About a 4-Fold Axis . 78

21. 8-Beam Eigenmodes Which Couple to E2 Transitions . 79

22. Plot of $g_{N}$ vs. the Rotation Angle, $\varepsilon$, For Coupling E2 Transitions to the 8-Beam Modes Shown in Fig. 21 . 80

23. 4-Beam Basis States Which Span the Space Corresponding to the 2-Dimensional Irreducible Representation of $C_{4v}$ . 83

24. 4-Beam Electronic Eigenmodes in the Subspace Spanned by the 2-Dimensional Irreducible Representation of $C_{4v}$ . 86

25. Plot of the Cross Term Occurring in the Expansion of $\tilde{F}_{N1,0}$ in the $|\psi_{AB}\rangle$ Basis as a Function of Cone Angle . 88

26. 6-Beam Electronic Borrmann Modes Which Couple to $M = \pm2$ Transitions . 92

27. Diagram Showing the Effects of Decreasing $g_{A}$ or Increasing $g_{N}$ on the Gain of a Steady State Lasing System . 98

28. Cut Through the Reciprocal Lattice of an $^{57}$Fe Crystal . 101

29. Plot of $K$ vs. $\Delta n$ For Both an Off-Bragg Mode and the Best Case 8-Beam Mode in $^{57}$Fe . 102

30. Angular Divergence Properties of the Eigenmodes of Amorphous, As Well As 2-Beam and Multi-Beam Borrmann Lasers . 106

31. A Simple Experiment to Verify the Anomalous Emission Effect . 109

32. An Absorption Experiment to Measure $g_{N}$ in a Borrmann Mode . 111

33. An Experiment to Demonstrate Anomalous Transmission of Multi-Beam Borrmann Modes Through a Crystal Containing Resonant Nuclei . 114

34. 4-Beam Borrmann Modes . 115

B1. Procedure for Projecting Out the Eigenmodes in the Two-Beam Case . 134
<table>
<thead>
<tr>
<th>Section</th>
<th>Title</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>B2</td>
<td>Two-Beam Eigenmodes</td>
<td>135</td>
</tr>
<tr>
<td>C1</td>
<td>Feynmann Diagrams for Elastic Scattering From the Electrons</td>
<td>137</td>
</tr>
<tr>
<td>C2</td>
<td>Partial Photoelectric Cross Sections for Z = 81 as a Function of Energy</td>
<td>147</td>
</tr>
<tr>
<td>D1</td>
<td>A Two-Beam Borrmann Mode</td>
<td>153</td>
</tr>
<tr>
<td>D2</td>
<td>Feynmann Diagrams For Photoabsorption by an Electron and For Resonant Emission From a Nucleus</td>
<td>157</td>
</tr>
</tbody>
</table>
I. INTRODUCTION

There is a possibility that an isomer can be found which could be used to obtain a sufficient population inversion density to bring about nuclear lasing. If it can be realized, a gamma ray laser ("graser") will be a new source of radiation in the ~1A region of unexcelled spectral brightness and coherence, and can be expected to have revolutionary consequences for physics, biology, and chemistry. For an excellent review of past results as well as current avenues of research in this field see Ref. 1.

Such a device would differ from a laser in the visible region in several important ways. A γ-ray laser will need to rely upon the Mössbauer effect\(^2\) in order to prevent excessive broadening of the narrow nuclear resonance lines, hence the emitting nuclei must be tightly bound in a solid. (In contrast, for an x-ray laser, the natural line width is so large, ~1eV, that the doppler broadening is not significant, even for a hot plasma.) The nuclear transitions must be those of medium or heavy nuclei. Photoelectric cross sections are usually large for these atoms at the gamma-ray energies of interest, so that absorption becomes an important consideration. This is quite different from the case of visible light lasers where the lasing material is largely transparent and absorption is not a serious problem. Furthermore, in optical lasers, mirrors can be used for mode selection, while for the graser, no mirrors in the ordinary sense exist.
The isomer required for such a laser must possess a lifetime sufficiently long to allow fabrication of the device. It must also have a nearby, short lifetime lasing level to which it could be rapidly transferred by suitable pumping radiation. Whether such an isotope, with a highly forbidden level which has a rapidly decaying, higher energy level nearby, exists is not known at present. Neither is it known whether a method can be devised to effect transfer rapidly enough in order to achieve a sufficiently large population inversion density while maintaining the conditions for an appreciable Mössbauer effect so that lasing can occur.

The result of our research is that the effects of crystallinity are of crucial importance in considerations regarding gamma-ray lasers, bringing in new elements to laser theory which could very well prove decisive in determining whether grasing can or cannot be achieved.

The normal modes of the electromagnetic field in an amorphous sample can be taken to be individual plane waves. There is no definite phase relationship between the normal modes and the positions of the atoms, since these are distributed randomly within the sample, and, for a given frequency (if there is no Zeemann splitting), all modes will be degenerate; that is they will scatter in the same manner and be photoabsorbed at the same rate.

In single crystals, however, the atoms are arranged periodically. When the Bragg condition is satisfied, certain plane waves will be scattered one into another, so that they no longer individually constitute eigenwaves of the crystal. Rather the eigenwaves will be
linear superpositions of these plane waves, which, when the Bragg condition is satisfied exactly, will have the character of travelling waves along one direction and standing waves perpendicular to that direction. Because the atoms are arranged periodically, these standing waves will have a definite phase relationship to each atomic site, and the eigenwaves will generally be absorbed at different rates. In contrast, in the amorphous case (or off-Bragg in the crystal) there is no definite phase relationship between the wave field and the atoms in any linear combination of plane waves, and all such combinations will be absorbed at the same rate as a single plane wave (per unit pathlength). When the Bragg condition is satisfied, the periodicity of the crystal removes the degeneracy present in the amorphous case.

Some of the eigenmodes of the radiation field in a perfect crystal have much stronger coupling to the emitting nuclei, and much less photoabsorption, than modes in the amorphous case. These modes may be selectively fed by nuclear transitions of the proper multipolarity. A crystal containing such emitters would thus require orders of magnitude less population inversion density in order to lase than would an amorphous sample of similar material.

When a nucleus in a crystal emits a γ-ray, it is emitted as a spherically diverging wave in all directions. The waves emitted in directions which make Bragg angles with a set of crystal planes are partially reflected from each plane so that the various reflected partial waves are approximately in phase and give rise to a large reflected wave, which also suffers reflections, and so on. If † is
the reciprocal lattice vector for the reflecting planes then the wavelets emitted with wave vectors \( \mathbf{K}_0 \) are reflected into those with wave vectors \( \mathbf{K}_1 = \mathbf{K}_0 + \mathbf{q} \), which in turn are reflected (by \(-\mathbf{q}\)) back into \( \mathbf{K}_0 = \mathbf{K}_1 - \mathbf{q} \), and so on.

The resulting wave field consisting of the coherent superpositions of plane waves \( \mathbf{K}_0, \mathbf{K}_1 \) will, in general, consist of superpositions of the "Borrmann" and "non-Borrmann" crystalline eigenmodes.\(^3\)\(^4\) The Borrmann modes have nodes in the \( \hat{E} \) field at the atomic sites and are absorbed by the electrons only very weakly. The other modes have strong electric fields at the atomic site, and are absorbed much more strongly. The Borrmann mode propagates much further in the crystal than an ordinary plane wave and thus (depending on the coupling) can stimulate emission (lasing) from many more excited nuclei than can waves emitted in non-Bragg directions.

In certain cases, depending on the multipolarity of the transition, particular nuclei will selectively feed only certain eigenmodes. If the nuclear transition is electric dipole (E1), then the coherent superposition of the resultant wave fields gives zero for the Borrmann modes, and these modes cannot be fed by these emitters. If, however, the nuclear emission is M1 or E2 (as most Mössbauer transitions are), or of higher multipolarity, then there can be strong emission into the Borrmann modes. This is referred to as the "anomalous emission effect" for waves emerging from single crystals emitted by M1 and higher nuclear transitions.\(^5\)\(^6\)
Lasing requires that the gain from stimulated emission exceed the losses from nonresonant absorption, and as was shown by Hannon and Trammell for 2-beam Borrmann modes, the anomalous emission effect should decrease the required inversion population density for lasing by one or two orders of magnitude.

Multi-beam Borrmann modes, which consist of the coherent superposition of 3 or more plane waves in the crystal, have properties which are even more advantageous for grasing. These modes can have smaller absorption coefficients, and, more importantly, larger coupling to the nuclear emitters, as well as sharper collimation, and hence require even less population inversion density for lasing, than do corresponding 2-beam modes.

The theory we work out, in addition to its applications to γ-ray lasers, also has new and interesting applications in single-photon optics. The anomalous emission of a γ-ray by an internal source into a Borrmann mode is a problem of interest for its own sake. This effect can occur for a 2-beam Borrmann mode so long as the nuclear transition has multipolarity M1 or higher. Even stronger anomalous emission can occur into multi-beam Borrmann modes. In addition, certain multi-beam Borrmann modes can anomalously transmit resonant radiation through a Mössbauer crystal. (This cannot occur for the 2-beam modes, except for the case of an E1 transition, since the Borrmann mode couples strongly to the nuclei and thus will be strongly absorbed at resonance.)
Most importantly, this work makes a new contribution to the field of crystal optics, and as such, is worth pursuing regardless of whether or not it has any immediate practical applications.

Anomalous transmission of x-rays in a multi-beam mode was first observed by Borrmann in 1965. Since then there has been considerable interest in the x-ray photoabsorption properties of these modes. The problem of interest here is the coupling of these modes to resonant nuclei within the crystal. The presence of such nuclei, in general, changes both the eigenmodes and eigenvalues of the radiation field, and introduces many new features into the problem. Briefly, some of these new features which are of interest for grasing are:

i) $g_N$, the nuclear coupling coefficient to the Borrmann mode, is $\propto m$, where $m =$ number of beams in the mode (for example, for modes which couple well to M1 transitions, $g_N = \frac{m \sin^2 \theta}{\cos \theta}$). Thus, for instance, the coupling of the resonant nuclei to a six beam mode will be three times larger than the coupling to a 2-beam mode at the same angle.

ii) $g_A$, the atomic absorption coupling coefficient, $\propto (\langle x^2 \rangle / \chi^2)^n$ in a Borrmann mode, where $\langle x^2 \rangle / \chi^2 \ll 1$, and $n$ is a positive integer. (For example, for the above mentioned M1 symmetry modes, $g_A = \frac{m}{2} (1 + \delta_{m,2}) \frac{\sin^2 \theta}{\cos \theta} \frac{\langle x^2 \rangle}{\chi^2}$.)

iii) The crystal acts as an etalon at the Bragg angles, confining the radiation in a manner similar to single mirrors, and increasing the effective pathlength in the lasing medium.
iv) The angular divergence δθ of the wave field in the Borrmann mode is determined by the "micro-geometry" of the crystalline planes, not by the "macro-geometry" of the lasing system. The beams emerging in a multi-beam Borrmann mode are collimated by Bragg reflection to an angular divergence of the order of 10^-5 rad, regardless of the size and shape of the lasing sample (so long as it is large enough to allow formation of the Borrmann modes).

Section II of this thesis begins with an overview of the theory of multi-beam diffraction, describes the general structure of the multi-beam eigenmodes, and finally, discusses both the Borrmann and Anomalous Emission Effects. In order to illustrate how the calculations are carried out, the properties of 2-beam Borrmann modes are worked out in detail in Section III. The properties of the various multi-beam Borrmann modes are presented in Section IV, while Section V contains a discussion how these eigenmodes relate to the problem of γ-ray lasing in a Mössbauer crystal. The single-photon applications of this work are discussed in Section VI, along with possible experiments which could be performed to test our results. The conclusion is contained in Section VII. This is followed by four appendices, the first of which gives explicit expressions for the first few vector spherical harmonics. The second contains a discussion of the application of group theory to the eigenvalue problem, while the third contains a derivation of the multipole expansions of the electronic and nuclear scattering amplitudes. Finally, the fourth details two alternate methods by which the coupling factors, g_A and g_N, may be calculated.
II. GENERAL THEORY OF MULTI-BEAM MODES

Crystal Diffraction Theory

We wish to find the eigenmodes and corresponding eigenvalues of the radiation field in a perfect crystal which contains resonant Mössbauer nuclei. That is, we seek waves which propagate through the crystal as \( \psi(\vec{r}) = \psi_0 e^{ik'z} \), where \( \psi_0 \) is some function, independent of \( z \), and \( \vec{k}' = k' \hat{z} \) is the complex propagation vector of the eigenmode. As we shall see, certain eigenmodes made up of several plane waves which simultaneously satisfy the Bragg condition with each other can exhibit much less photoabsorption and couple much more strongly to resonant nuclei than do the eigenmodes off-Bragg or in an amorphous medium.

The diffraction of \( \gamma \)-rays within crystals has historically been treated using two distinct, but equivalent, approaches. The approach that is followed in this thesis is that of Hannon and Trammell,\(^5,34\) which treats the scattering by individual plane layers within the crystal in the manner used by Darwin\(^38\) to treat the x-ray case. The Russian workers in this field\(^39\) follow the treatment of Ewald and von Laue.\(^40,41\)

In Darwin's approach, the crystal is assumed to consist of a series of planes of unit cell thickness, as is shown in Fig. 1a. Each plane consists of an infinite 2-dimensional periodic array of unit cells located on a 2-D Bravais lattice. In general, each unit cell may contain a basis of several atoms, although, for reasons we shall
Fig. 1. a) Crystal geometry used in developing the dynamical theory. b) Radiation channels for a plane layer.

see later, the discussion in this thesis will be primarily restricted to the case where there is only one atom per unit cell.

Consider a wave incident upon such a plane layer in the direction $\vec{k}_0$. The amplitude of the wave elastically scattered by the plane into some other direction, for example $\vec{k}_1$, will be negligible unless all the wavelets scattered by each individual unit cell in the plane add constructively in the direction $\vec{k}_1$; that is $(\vec{k}_1)_{xy} = (\vec{k}_0)_{xy} + \vec{t}_1$, where $\vec{t}_1$ is a reciprocal lattice vector of the plane. The component of $\vec{k}_1$ normal to the plane (along $\hat{z}$) will then be equal to $\pm \varepsilon_1 = \pm [k_0^2 - ((\vec{k}_0)_{xy} + \vec{t}_1)^2]^{1/2}$. If $\varepsilon_1$ is real, then $\vec{k}_1$ is an "open channel" which may be "fed" by waves scattered with the proper phase from successive planar layers. If $\varepsilon_1$ is imaginary, the wave in the $\vec{k}_1$ channel decays as $e^{-|\varepsilon_1||z|}$. $\vec{k}_1$ is then a "closed channel" and for our present purposes may be neglected.

A family of open radiation channels, $\{\vec{k}_0\}$, consisting of a finite number of wavevectors having components $((\vec{k}_x)_{xy}, \pm \varepsilon_0)$, where
\[(\mathbf{R}_s)_{xy} = (\mathbf{R}_0)_{xy} + \mathbf{t}_s \quad (1)\]

and

\[g_s = \left[ k_0^2 - |(\mathbf{R}_0)_{xy} + \mathbf{t}_s|^2 \right]^{1/2}, \quad g_s \text{ real,} \quad (2)\]

is thus associated with any given initial direction, \(\mathbf{R}_0\) (see Fig. 1b). Note that there is nothing special about the direction, \(\mathbf{R}_0\). The entire family, \(\{\mathbf{R}_0\}\), may be generated by the above procedure from any one of its members.

The scattering from an individual planar layer in the x-ray or \(\gamma\)-ray case is usually very small and may be treated in the Born approximation, neglecting multiple scattering within a single plane.\(^{34,35}\) (The small corrections due to intra-planar multiple scattering are developed in Ref. 34.) In the Born approximation, a wave with wavevector \(\mathbf{R}_s^+\) incident upon a planar layer at \(z = 0\) with amplitude \(\hat{A}^s = A^s e^{i(\mathbf{R}_s^+ \cdot \mathbf{r} - \omega t)}\) will be scattered by that plane so that the amplitude of the total coherent elastic wave at a position \(\mathbf{R}\) is \(\hat{A}(\mathbf{R}, t) = \hat{A}(\mathbf{R}) e^{-i\omega t}\), where\(^{42}\)

\[\hat{A}(\mathbf{R}) = \hat{A}^s(\mathbf{R}) + \hat{A}^{sc}(\mathbf{R})\]

\[= A^s e^{i\mathbf{R}_s^+ \cdot \mathbf{r}} + i \sum s' \sum_{n=\pm} \hat{A}_{s'}^{n+} A^{s+} e^{i\mathbf{R}_{s'}^{n+} \cdot \mathbf{r}}. \quad (3)\]

The scattered wave, \(\hat{A}^{sc}(\mathbf{R})\), thus consists of a superposition of waves in a finite number of "open" plane wave channels with wavevectors \(\mathbf{R}_s = ((\mathbf{R}_0)_{xy} + \mathbf{t}_s, \pm g_s)\).
\( \tilde{F}_{n' n}^{s'} s \) is the planar scattering amplitude matrix for scattering from an \((s')\) channel into an \((s' n)\) channel. Letting \( \varepsilon_1^{m n}, \varepsilon_2^{m n} \) \((n = + \text{ or } -)\) denote any convenient orthogonal basis perpendicular to \( \hat{k}_{s n} \), \( \tilde{F} \) is given by

\[
\begin{pmatrix}
(\tilde{F}_{n' n}^{s'} s)_{11} & (\tilde{F}_{n' n}^{s'} s)_{12} \\
(\tilde{F}_{n' n}^{s'} s)_{21} & (\tilde{F}_{n' n}^{s'} s)_{22}
\end{pmatrix}
\]

where

\[
(\tilde{F}_{n' n}^{s'} s)_{ij} = \frac{\lambda_{0 n'}}{\cos \theta_{s'}} f_u(k_{s', n'}, \varepsilon_1^{m'n'}; k_{s n}, \varepsilon_2^{m n}).
\]

\( n' \) is the number density (in \( \text{cm}^{-2} \)) of unit cells in the scattering plane, while

\[
\cos \theta_{s'} = [1 - (\varepsilon_s / |k_0|)^2]^{1/2}
\]

so that \( \varepsilon_s \) is the angle that \( \hat{k}_{s, n'} \) makes with the normal to the scattering planes.

\( f_u \) is the unit cell scattering amplitude given by

\[
f_u(k_{s', n'}, \varepsilon_1^{m'n'}; k_{s n}, \varepsilon_2^{m n}) = \sum_{\nu} f_u(k_{s', n'}, \varepsilon_1^{m'n'}; k_{s n}, \varepsilon_2^{m n}) \varepsilon^{i(k_{s', n'} - k_{s n}) \cdot \hat{p}_y}.
\]
where $f_\nu$ is the scattering amplitude of the $\nu^{th}$ atom, located at $\hat{r}_\nu$, and the sum runs over all the atoms in the unit cell.

We may take $f_\nu = f_{N,\nu} + f_{A,\nu}$, with $f_{N}$ being the resonant nuclear scattering amplitude of the $\nu^{th}$ atom, while, in the usual x-ray notation, $f_{A} = f_{O} + \Delta f' + if''$ gives the contribution due to the electrons. $f_{O} = -r_0ZF(2\theta)$ is the scattering amplitude coming from the $A^2$ term in the S-matrix expansion, while $\Delta f'$ and $f''$ are the real and imaginary parts of the anomalous dispersion contribution (coming from the $\hat{\beta} \cdot \hat{A}$ terms) respectively. (An additional contribution to $f_N$ occurs due to screening effects, but it is generally small and may be neglected here.) Expressions for $f_N$ and $f_A$ are derived in Appendix C.

For the x-ray and $\gamma$-ray case, the planar scattering amplitudes are quite small ($\approx 10^{-3}$). (This is in contrast to the case of low energy electron diffraction (LEED), where the $F$'s are typically of order unity.) Since each individual planar layer scatters so weakly, appreciable scattered waves can only be built up by the coherent superposition of waves coming from several layers. It follows that the wave scattered from any particular layer may be neglected without appreciably altering the resultant scattered wave. The wave will thus propagate through the crystal with a wavevector which is very nearly equal to its value in free space.

Let us denote the wave incident from above upon the $m^{th}$ plane of the crystal in the $s+$ channel (that is, with a wavevector having a positive $\hat{z}$ component, $\hat{s}_g$) by
\[ \hat{\mathbf{r}}_m^{s+} = \{a_{1,m}^{s+}e_{1}^{s+} + a_{2,m}^{s+}e_{2}^{s+}\} e^{i\mathbf{R}_m^{s+}\cdot \hat{\mathbf{r}}_m} \]

\[ = \{p_{1,m}^{s+}e_{1}^{s+} + p_{2,m}^{s+}e_{2}^{s+}\} e^{i(\mathbf{R}_m^s)_{xy}\cdot \hat{\mathbf{r}}_m}, \quad (8) \]

where \( \hat{\mathbf{r}}_m = (\hat{\mathbf{r}}_m)_{xy} + md \hat{\mathbf{z}} \). \( p_{1,m}^{s} = a_{1,m}^{s+}i\gamma_m^{md} \) thus gives the amplitude of the wave in the \( \mathbf{R}_m^{s+} \) channel, polarized along \( e_{1}^{s+} \), that is incident upon the \( m^{th} \) layer. Similarly, let us denote the wave incident upon the \( m^{th} \) plane from below in the \( s^{-} \) channel by

\[ \hat{\mathbf{r}}_m^{s-} = \{M_{1,m}^{s}e_{1}^{s-} + M_{2,m}^{s}e_{2}^{s-}\} e^{i(\mathbf{R}_m^s)_{xy}\cdot \hat{\mathbf{r}}_m}. \quad (9) \]

The wave incident upon the \( m^{th} \) plane in the \( \mathbf{R}_m^{s+} \) channel, \( \hat{\mathbf{r}}_m^{s+} \), is related to the waves incident upon the \( (m-1)^{th} \) plane by

\[ p_{1,m}^{s} = e^{i\gamma_m^{d}}[p_{1,m-1}^{s} + i \sum_{s'} \sum_{j} (R_{s+}^{s'})_{ij} p_{j,m-1}^{s'} \]

\[ + i \sum_{s'} \sum_{j} (R_{s-}^{s'})_{ij} M_{j,m-1}^{s'}]. \quad (10i) \]

That is, the wave incident upon the \( m^{th} \) plane in the \( \mathbf{R}_m^{s+} \) channel consists of the wave incident upon the \( (m-1)^{th} \) plane in that channel, plus the forward scattering of that wave, plus the amplitudes for the waves in each of the other channels incident upon the \( (m-1)^{th} \) plane being scattered into the \( \mathbf{R}_m^{s+} \) channel, all propagated to the \( m^{th} \) plane by the phase factor \( e^{i\gamma_m^{d}} \). A similar relation holds relating the wave incident on the \( m^{th} \) plane in the \( \mathbf{R}_m^{s-} \) channel (from below) to the waves incident on the \( (m+1)^{th} \) plane.
\[ M^S_{i,m} = e^{i\kappa_{sd}[M^S_{i,m-1} + i \sum_{s'} \sum_j (F^S_{-+})_{ij} M^S_{j,m-1} + i \sum_{s'} \sum_j (F^S_{+-})_{ij} P^S_{j,m-1}]] \] (10ii)

Note that the position of the plane, \( z = md \) within the \( m \)th planar layer is totally arbitrary. If the wavefield is known at one mathematical plane within the crystal, Eqs. 10 allows us to determine the wavefield at any other mathematical plane a distance \( md \) away, \( m \) arbitrary, and hence at any place in between. Changing the location of the planes within the unit cell merely changes the phase of the structure factor in Eq. 7. If there is only one atom per unit cell, the planes are typically chosen to coincide with the atomic planes.

Writing \( \mathbf{P}^S_m \) and \( \mathbf{M}^S_m \) for the column vectors

\[
\mathbf{P}^S_m = \begin{pmatrix} P^S_{1,m} \\ P^S_{2,m} \end{pmatrix} \quad \mathbf{M}^S_m = \begin{pmatrix} M^S_{1,m} \\ M^S_{2,m} \end{pmatrix}
\] (11)

the dynamical equations for Mössbauer optics (Eqs. 10) become

\[
\mathbf{P}^S_m = e^{i\kappa_{sd}[\mathbf{P}^S_{m-1} + i \sum_{s'} F^{SS'}_{-+} \mathbf{P}^S_{m-1} + i \sum_{s'} F^{SS'}_{+-} \mathbf{M}^S_{m-1}]} \] (12ii)

and

\[
\mathbf{M}^S_m = e^{i\kappa_{sd}[\mathbf{M}^S_{m+1} + i \sum_{s'} F^{SS'}_{-+} \mathbf{M}^S_{m+1} + i \sum_{s'} F^{SS'}_{+-} \mathbf{P}^S_{m+1}]} \] (12ii)
We can find solutions (eigenmodes) to these iterative equations of the form

\[ \hat{P}_m(k'_Z) = \hat{P}_{k'_Z} e^{ik'_Zd} \]  

(131)

\[ \hat{M}_m(k'_Z) = \hat{M}_{k'_Z} e^{ik'_Zd}. \]  

(1311)

Any general solution may then be expressed in terms of these eigenmodes as

\[ \hat{P}_m = \sum_{k'_Z} \hat{P}_{k'_Z} e^{ik'_Zd} \]  

(141)

\[ \hat{M}_m = \sum_{k'_Z} \hat{M}_{k'_Z} e^{ik'_Zd}. \]  

(1411)

Substituting Eqs. 14 into Eqs. 12 yields the set of coupled equations

\[ (e^{-i(\xi_s-k'_Zd)} - 1)\hat{P}_{k'_Z} - i \sum_{s'} \hat{P}_{s's'} \hat{P}_{k'_Z} - i \sum_{s'} \hat{P}_{s's'} \hat{M}_{k'_Z} = 0 \]  

(151)

\[ (e^{-i(\xi_s-k'_Zd)} - 1)\hat{M}_{k'_Z} - i \sum_{s'} \hat{P}_{s's'} \hat{P}_{k'_Z} - i \sum_{s'} \hat{P}_{s's'} \hat{P}_{k'_Z} = 0. \]  

(1511)

For the above set of coupled equations to have a solution, it must be that the determinant of the coefficients gives zero. If there are \( m \) channels in the family, this dispersion equation will yield the \( 2m \) eigenvalues, \( k'_Z \). Substituting these values back into Eqs. 15 will then yield the eigenmodes.
A considerable simplification occurs for the cases of \( \gamma \)-ray and x-ray scattering, however, since the planar scattering amplitudes, \( \bar{P}_{Ss'}^{ss'} \) are so small. A nontrivial solution to Eqs. 14 will only exist when \( \left( e^{-i(g_{S}-\eta k_{Z}^{i})d} - 1 \right) \) is small (of the same order as \( \bar{F} \)). There will thus always be two values of \( k_{Z}^{i} \) (one for each eigenpolarization) associated with each plane wave channel such that \( k_{Z}^{i} = g_{S} \). In general, however, there is no fixed relationship between \( g_{S} \) and \( g_{S'}^{s'=s} \). Each \( \left( e^{-i(g_{S'}^{s'=s}-\eta k_{Z}^{i})d} - 1 \right) \) will then be quite large (compared to the \( \bar{F}'s \) and the \( \bar{F}_{s} \) channel will not be strongly coupled to any of the other channels. Thus, for these two values of \( k_{Z}^{i} \), neglecting terms of order \((\bar{F})^{2}\), Eqs. 15 reduce to the eigenvalue equation involving only forward scattering,

\[
\bar{P}_{Ss}^{ss'} \bar{g}_{k_{Z}^{i}}^{s'} = - (g_{S} - \eta k_{Z}^{i})d \bar{F}_{k_{Z}^{i}}^{s}
\]  

(16)

Eq. 16 gives the result for the "plus" channels; an identical equation holds for the "minus" channels. Solving this equation will yield the two, generally non-orthogonal, off-Bragg eigenmodes for this channel.\(^{36}\) If there is only a single isotropic resonance, \( F^{ss} = F^{I} \), and all such off-Bragg eigenmodes will be degenerate, with \( (\eta k_{Z}^{i} - g_{S})d = F, \ \eta = \pm \). In particular, they will all decay as \( \exp[\text{Im}(F^{ss})|z|/d] \) as they travel through the crystal. Any linear combination of these eigenmodes will also be an eigenmode, with the same degenerate eigenvalue.
Fig. 2. Diagram showing two wavevectors which satisfy the Bragg condition for reflection from a particular set of planes within a crystal. Note that these reflecting planes are not, in general, the scattering planes used to derive the dynamical theory.

If, however, the wave in one of the channels very nearly satisfies the Bragg condition for some set of planes within the crystal, as is shown in Fig. 2, then, for the case where one of the waves is in a "plus" channel, either

$$d(g_s - g_{s'}) = 2n\pi \quad s' \text{ a "plus" channel} \quad (17\,\text{i})$$

or

$$d(g_s + g_{s'}) = 2n\pi \quad s' \text{ a "minus" channel} \quad (17\,\text{ii})$$

To see this, note that the Bragg condition may be written as

$$k_{s'}n - k_{s^+} = \frac{1}{c} = \frac{1}{p} + \frac{2n\pi}{d}, \quad (18)$$
where \( \mathbf{r}_c \) is a crystal reciprocal lattice vector, while \( \mathbf{r}_p \) is a planar reciprocal lattice vector. Substituting expressions for \( \mathbf{r}_{s,n} \) (Eq. 1) into Eq. 18 then yields Eqs. 17 immediately.

If two waves, say \( \mathbf{r}_0 \) and \( \mathbf{r}_1 \), satisfy the Bragg condition, there will be four values of \( k_z' \) for which both \( (e^{-i(\mathbf{g}_n \cdot \mathbf{r}_0)\mathbf{k}_z'} - 1) \) and \( (e^{-i(\mathbf{g}_n \cdot \mathbf{r}_1)\mathbf{k}_z'}) - 1 \) are small, while each \( (e^{-i(\mathbf{g}_{s=0,1} \cdot \mathbf{r}_0)\mathbf{k}_z'}) - 1 \) is still of order unity. The waves in these two channels, \( \mathbf{r}_0 \) and \( \mathbf{r}_1 \), will be strongly coupled and, again neglecting terms of order \((\tilde{F})^2\), Eqs. 15 reduce (for the case where \( \mathbf{r}_0 \) and \( \mathbf{r}_1 \) are both "plus" channels) to the following pair of coupled equations

\[
[(e^{-i(\mathbf{g}_0 \cdot \mathbf{k}_z')} - 1)|\tilde{F}_{k_z}^0| - i\tilde{F}_{k_z}^0 - i\tilde{F}_{k_z}^1|\tilde{F}_{k_z}^0| = 0
\]

and

\[
-i\tilde{F}_{k_z}^0 + [(e^{-i(\mathbf{g}_1 \cdot \mathbf{k}_z')} - 1)|\tilde{F}_{k_z}^1| - i\tilde{F}_{k_z}^1 - i\tilde{F}_{k_z}^0|\tilde{F}_{k_z}^1| = 0.
\]

Neither the wave in \( \mathbf{r}_0 \) nor the wave in \( \mathbf{r}_1 \) by themselves will constitute eigenwaves, rather the eigenwaves will be linear combinations of these two waves.

Eqs. 19 may be easily solved to obtain these eigenmodes (see Ref. 36). As an example, consider the physically important simple case of two "plus" channels with \( \theta_0 = \theta_1 = \theta \), which very nearly satisfy the Bragg condition for reflection from some set of planes in a perfect crystal. (This corresponds to symmetric Laue transmission when one of the channels is being fed from outside the crystal.) Off-resonance, the nuclei do not interact with the radiation, and this
case reduces to a simple x-ray problem. Choosing the polarization vectors to be $\hat{\mathbf{n}}_s$, which lies in the plane containing $\hat{\mathbf{r}}_0$ and $\hat{\mathbf{r}}_1$, and $\hat{\mathbf{s}}_s$, which lies out of that plane (see Fig. 3 below), the amplitudes of the waves in the $\hat{\mathbf{r}}_0$ and $\hat{\mathbf{r}}_1$ channels in the two $\sigma$-polarized eigermodes are related by

$$p_{k'_\perp}^1 = -\frac{\delta + \beta}{F^{01}} p_{k'_\perp}^0$$  \hspace{1cm} (201)

and

$$p_{k'_\perp}^1 = -\frac{\delta - \beta}{F^{01}} p_{k'_\perp}^0$$  \hspace{1cm} (201i)

with

$$k'_{\perp}^d = \omega_0 d - \delta + F^{00} \pm \beta. $$  \hspace{1cm} (21)

In these equations, $\delta = (k_d \sin \theta) \Delta \theta$, $\beta = [\delta^2 + (F^{01})^2]^{1/2}$ with $\text{Im} \beta \geq 0$, while, for the $\sigma$-polarization case, if the frequency is well above any atomic absorption frequencies,

$$F^{00} = F^{11} = \frac{\lambda \omega_0 n d}{\sin \theta} \left( -r_0 Z + \frac{i\sigma_A}{4\pi} \right)$$  \hspace{1cm} (221)

and

$$F^{01} = F^{10} = -\frac{\lambda \omega_0 n d}{\sin \theta} \left( -r_0 Z \ell(28) + \frac{i\sigma_A}{4\pi} \right) r_D(\hat{\mathbf{r}}_1 - \hat{\mathbf{r}}_0).$$  \hspace{1cm} (221i)

In Eqs. 22, $n$ is the number density of unit cells (in cm$^{-3}$), $Z$ is the
number of electrons, $r_0$ is the classical electron radius, $F(2\theta)$ is the atomic form factor, $\sigma_A$ is the total electronic cross section which at these energies may be taken to be the photoelectric cross section, $\sigma_{pe}$, and finally, $f_D(\vec{R}_1 - \vec{R}_0) = \exp\left[-\frac{|\vec{R}_1 - \vec{R}_0|^2 <x^2>}{2}\right]$ is the Debye-Waller factor, where $<x^2>$ is the mean thermal and zero point motion of the atom about its equilibrium position. The results for $\pi$-polarization are identical except for a polarization factor of $\cos2\theta$ which occurs in $F^{01}$. There is no polarization mixing in this case. These expressions for $F$ are derived in Appendix C.

Note that the eigenwaves are no longer degenerate. Off-Bragg, nearly every rotation and reflection of the wave field (or equivalently, of the crystal) possible in 3-dimensional space will leave the crystal invariant; that is, an off-Bragg eigenwave will remain an eigenwave after any such rotation or reflection which does not cause the wave to satisfy the Bragg condition in its new orientation. (Since the angular width of each Bragg reflection is so narrow, $10^{-5}$ to $10^{-4}$ rad, this set of symmetry operations will be very nearly the entire set of rotations and reflections possible in 3-dimensional space.) When the wave satisfies the Bragg condition, however, one no longer has this freedom; in this case, only certain particular rotations and reflections will be symmetry operations (see below and Appendix B). When the Bragg condition is satisfied, the periodicity of the atoms in the crystal breaks the symmetry present in the off-Bragg or amorphous case, and removes the degeneracy of the eigenmodes.
If several plane waves simultaneously satisfy the Bragg condition with each other, that is each pair of them satisfy Eqs. 17, the exponential factor \( e^{-i(g_0 - n k_1) d} \) associated with each such wave will be small, so that the wave in each channel will be strongly coupled to that in each of the others. While perhaps algebraically tedious when the number of coupled beams is large, the solution to Eqs. 15 in this case is straightforward, and may be easily accomplished on a computer.

Considerable physical insight may be obtained, however, without resorting to the direct reduction of the above set of coupled equations. As we shall see, by limiting the consideration to that point in phase space where the Bragg condition is satisfied exactly for all the reflections involved, the eigenmodes may be obtained directly from symmetry considerations in most of the cases of interest. Once the eigenmodes are known, the eigenvalues may be easily obtained by taking expectation values.\(^{45}\)

**Multi-Beam Eigenmodes**

General Structure of the Multi-Beam Modes

As was shown above, waves in two "plus" channels, \( \hat{\xi}_0 \) and \( \hat{\xi}_1 \), satisfy the Bragg condition for reflection from a set of crystal planes corresponding to the reciprocal lattice vector \( \hat{\bf t}_0 = \hat{\bf t}_p + \frac{2\pi}{d} \) provided
\[ d(g_0 - g_1) = 2n\pi \]  

(23)

Equivalently, the Bragg condition may be written as

\[ \vec{K}_1 - \vec{K}_0 = \mathbf{t}_c. \]  

(24)

that is, the difference between the two wavevectors is very nearly equal to a crystal reciprocal lattice vector.

The same criteria apply if several plane waves simultaneously satisfy the Bragg condition; the difference between the wavevectors of any two will be nearly equal to a reciprocal lattice vector, and, in general, each wave will be reflected into each of the others. Such a

![Diagram](image)

Fig. 3. General Structure of a multi-beam mode. All the wavevectors, \( \vec{K}_s \), lie on the surface of a cone whose base is defined by the circle containing the reciprocal lattice points involved in the reflections. The half-apex angle of the cone, \( \theta \), is not in general a Bragg angle. There are two polarization vectors associated with each beam, denoted \( \mathbf{t}_s \), which lies in the plane of the cone base (along \( \phi \) if the cone axis is along \( \hat{z} \)), and \( \mathbf{t}_s \), which lies out of the plane (along \( \theta \)). The 4-beam case is shown, but similar modes exist for other numbers of beams.
"multi-beam mode" can be formed if all the reciprocal lattice points involved are coplanar and lie on a circle. The wavevector of the each "beam" which forms the mode, \( \mathbf{k}_s \), lies on the surface of a cone whose base is defined by the circle containing the reciprocal lattice points involved, as is shown in Fig. 3. The 2-beam case, involving a single Bragg reflection, is a limiting case of this, as can be seen from Fig. 4. Note that the half-apex angle of the cone will not necessarily be a Bragg angle in the general multi-beam case.

The two linearly independent polarization vectors associated with each beam may be chosen as \( \hat{\mathbf{e}}_s \), which lies in the plane of the cone base (along \( \hat{\mathbf{e}} \) if the cone axis lies along \( \hat{\mathbf{z}} \)), and \( \hat{\mathbf{e}}_s \), which lies out of the plane (along \( \hat{\mathbf{e}} \)), also as shown in Fig. 3.
Fig. 5 Multi-beam modes possible in crystals whose reciprocal lattices contain 3 and 4-fold symmetry axes. The points denote a plane through the reciprocal lattice. Any circle in this plane which intersects more than one reciprocal lattice point can form the base of the cone defining a set of modes. The modes with the most beams occur when the center of the circle lies on a high symmetry axis of the plane. This figure shows the case where that point is a reciprocal lattice point; similar modes occur when the center of the circle coincides with on an interstitial point.

Two-beam modes are possible along any axis which is perpendicular to a reciprocal lattice vector. Modes with more than 2 beams may be obtained when the axis of the cone is aligned with some high-symmetry axis of the crystal. (It is also possible to obtain multi-beam modes about low-symmetry axes, but the number of beams available in such modes is limited. The modes of most interest will occur along high-symmetry axes.)

The modes that are allowed for a given choice of cone axis may be found by drawing all possible circles of radius less than $1/\lambda$ (the length of $\Lambda$) on a plane of reciprocal lattice points perpendicular to that axis, as is illustrated in Fig. 5. Any circle which intersects more than one lattice point forms a possible base for a cone defining
a set of modes. The planes which contain these circles, perpendicular to the symmetry axis of the mode and of unit cell thickness, are those which are chosen as the scattering planes defined in Figs. 1 and 2. This is equivalent to choosing the 2 axis of the crystal to lie along the symmetry axis of the mode.

The two cases illustrated in Fig. 5 are those for 3 and 4-fold axes of the crystal's reciprocal lattice. It can be seen from the figure that axes with three-fold symmetry can support modes with 6 or 12 beams, while four-fold axes can support modes with 4 or 8 beams. Modes with even more beams occur at larger radii.

Note that, since all the reciprocal lattice vectors involved lie within a single plane of the reciprocal lattice, it is the symmetry of that plane, rather than the symmetry of the lattice as a whole which determines the properties of the mode. In particular, note that any slice of unit cell thickness through the reciprocal lattice perpendicular to a 3-fold axis will be a 2-dimensional hexagonal lattice. Such a 2-D lattice possesses 6-fold, as well as 3-fold, rotation axes. If the symmetry axis of the mode passes through a 6-fold rotation axis of this plane, it will be this 6-fold symmetry, rather than the 3-fold symmetry of the crystal as a whole, which determines the properties of the mode.

There are other multi-beam modes in which the reciprocal lattice vectors are not coplanar, but it is only possible to satisfy the Bragg condition for these modes at particular values of the frequency. These modes occur when the Ewald sphere happens to simultaneously intersect two sets of reciprocal lattice points, where each set by
Fig. 6. Mode formed in a cubic crystal when all eight corners of a cube simultaneously lie on the Ewald sphere. Such a mode would form a radiation trap if it could be excited by an internal source.

...itself is coplanar. (The Ewald sphere is a sphere of radius $1/\lambda$ whose surface intersects the reciprocal lattice point at the origin. The Bragg condition will automatically be satisfied for the reflection connecting the origin to any other reciprocal lattice point which also lies on the surface of the sphere.) Since this can only happen if the Ewald sphere has a particular radius, these modes can only be excited by particular discrete frequencies, which are unlikely to correspond to the resonant frequency of any nuclear transition. For this reason, aside from the comments below, the consideration in this thesis will be limited to modes in which all the reciprocal lattice vectors are coplanar.

Nevertheless, such modes have some striking properties which would be of great interest for the lasing problem if the difficulties with excitation could be overcome. A particularly interesting example occurs when the Ewald sphere is centered as a reciprocal lattice point...
in a cubic crystal. If the radius of the sphere is equal to the length of a reciprocal lattice vector along the (111) direction, it will intersect eight points which form a cube, as is shown in Fig. 6. A similar mode exists if the radius of the sphere is equal to the length of a reciprocal lattice vector along (100). The sphere then intersects six points which form an octahedron.

If such a mode could be excited by an internal source the energy would be trapped in the crystal, since each beam is always reflected back into the center. Eventually the energy would diffuse out, but such a mode could serve to store resonant radiation for a period of time long compared to the natural lifetime of the emitter. A laser utilizing such a mode would necessarily have a very high gain, since it would essentially be a multi-pass system, similar to those optical lasers which use mirrors to reflect the radiation back into the lasing cavity.

Symmetry Properties of the Eigenmodes

Let us consider the symmetry properties of an m-beam mode, in which all the reciprocal lattice vectors involved are coplanar. If the Bragg condition is satisfied exactly for all the reflections in the mode, all m wavevectors will make exactly the same angle \( \theta \) with the \( \hat{z} \) axis, where this axis is chosen to lie along the symmetry axis of the mode. All \( m \) values of

\[
g_s = [k_0^2 - ((k_s)_{xy})^2]^{1/2}
\]

will thus be equal. Choosing as a basis the set of state vectors \( \{|\pi_s\rangle, |\sigma_s\rangle\} \) (where, for example, \( |\pi_s\rangle \) denotes a plane wave of unit amplitude with
wavevector \( \vec{k}_s \), polarized along \( \hat{\mathbf{s}} \), we may write Eqs. 15 within the 2m-dimensional subspace defined by this m-beam mode as the single eigenvalue equation

\[
[(e^{-i(g - k_d')d} - 1)\mathbf{I} - i\vec{F}]|P_{k_z}\rangle = 0,
\]

(25)

or, expanding the exponential and neglecting terms of order \([(g - k_d')d]^2 = [\vec{F}]^2\), as

\[
\vec{F} |P_{k_z}\rangle = \frac{\lambda_0}{\cos \theta} (\vec{r}_A + \vec{r}_N)|P_{k_z}\rangle = (k_d - g)d |P_{k_z}\rangle.
\]

(26)

In these equations, \( |P_{k_z}\rangle \) denotes a 2m-dimensional column vector whose components are \( \vec{r}_{k_s, s'}^+ \), \( 1 < s < m \), with \( \vec{r}_s^+ = \hat{\mathbf{s}}_s \) or \( \hat{\mathbf{t}}_s \) (only "plus" channels occur in these multi-beam modes with our choice of \( \hat{\mathbf{z}} \) axis). \( \vec{F} \) now denotes a 2m×2m matrix whose components are \( \langle \vec{r}_s^+ | \vec{F}^+ | \vec{r}_s^+ \rangle \).

We see from Eq. 26 that when the Bragg condition is satisfied exactly, the multi-beam eigenmodes will be those of the planar scattering operator, \( \vec{F} \), alone. That is, the symmetry group of \( \vec{F} \) will also determine the symmetries of the eigenmodes. A detailed discussion of the application of group theory to the eigenvalue problem, with particular emphasis of these multi-beam modes, is contained in Appendix B.

The consideration in this thesis will be limited to the case of a cubic crystal (where there are no electric field gradients) with one atom per unit cell, where the ground state electronic wave function of
the atom is spherically symmetric (see Appendix C). In this case, if there is no Zeemann splitting, \( \vec{F} \) within this subspace, at exact Bragg, will possess the symmetry of the polygon of reciprocal lattice vectors which forms the base of the mode. That is, those rotations and reflections which leave the polygon invariant will also leave \( \vec{F} \) invariant. The symmetry group of the polygon is in turn that group of rotations and reflections about the symmetry axis of the mode which leave the 2-D plane of reciprocal lattice containing the cone base invariant.

The symmetry group of an \( m \)-beam mode whose cone axis is centered on an \( n \)-fold axis of the reciprocal lattice plane which contains its base will be \( C_{nv} \), the group of rotations and reflections which leave a regular \( n \)-sided polygon invariant.\(^{46}\) Each of the \( 2m \) eigenmodes which have the general structure of this \( m \)-beam mode will transform like a particular row of a particular irreducible representation of this group. Eigenmodes which transform like the \( \lambda \gamma \)-dimensional irreducible representation, \( \Gamma^{\gamma} \), will be \( \lambda \gamma \)-fold degenerate.

As is shown in Appendix B, if \( n = m \), so that the base of the mode forms a \textbf{regular} \( m \)-sided polygon, the set of state vectors \( \{ |\pi_{\alpha}\rangle, |\alpha_{\beta}\rangle \} \) defines a regular representation of the symmetry group, \( C_{nv} \), and each irreducible representation will appear among the eigenmodes a number of times equal to its dimension. One dimensional irreducible representations will appear only once, and the eigenmodes which correspond to them may be fully determined by symmetry arguments alone.
In general, for these modes, \( m = \alpha n \), where \( \alpha \) is an integer. Each irreducible representation, \( \Gamma^\nu \), thus appears \( \alpha \lambda^\nu \) times among the eigenmodes. If \( \alpha = 2 \), the 1-dimensional irreducible representations appear twice, and as we shall see, the corresponding eigenmodes may still be determined solely by symmetry arguments. These two cases will constitute the vast majority of the eigenmodes of interest for the lasing problem.

Two-dimensional irreducible representations always appear at least twice among the eigenmodes. (No irreducible representations with dimension higher than two occur in \( C_{nv} \).) Symmetry considerations are thus insufficient to fully determine the eigenmodes in this case. As will be shown in Sections IV and V, these modes are not of great interest for the lasing problem. They do, however, have properties which are of great interest for some single-photon problems.

If there is Zeemann splitting caused by a magnetic field applied along the symmetry axis of the mode, reflections will no longer be valid symmetry operations and the symmetry group of the mode will reduce to \( C_n \), the cyclic group of rotations which leave a regular \( n \)-sided polygon invariant. To see this, expand \( \tilde{r}_n \) in vector spherically harmonics (see Appendix C) and note that, under reflection about a plane containing the \( \hat{z} \) axis, rotated an angle \( \phi \) relative to the \( \hat{x} \) axis, \( \hat{y} \) \( \ell \), \( m \) goes over to \( e^{iM(\pi-2\phi)} \hat{y}_L,-M \). Reflections thus have the effect of interchanging the \( \pm M \) nuclear transitions. If there is no Zeemann splitting these two transitions are degenerate, and the reflection will have no observable effect. If, however, there is Zeemann splitting these two transitions are no longer degenerate. A
mode which couples strongly to the $M = +1$ transition would, upon reflection, have to couple to the $M = -1$ transition. Since the energies of the two transitions are no longer the same, the mode will no longer be in resonance after the reflection.

$M = 0$ transitions constitute a special case since they go over into themselves under reflection. Eigenmodes which couple to $M = 0$ transitions when there is no Zeeman splitting will remain good eigenmodes even if a magnetic field is applied along the symmetry axis of the mode. Since $C_n$ is a cyclic group, all of its irreducible representations are 1-dimensional. It follows, therefore, that the eigenmodes of the non-Zeeman split case which couple to $M = 0$ transitions must transform like 1-dimensional irreducible representations of $C_{nv}$.

Finding the Eigenvalues

The method by which the eigenmodes may be obtained from symmetry arguments is detailed in Appendix B. Once the eigenmodes are known, it is a simple matter to obtain the eigenvalues by taking expectation values of $\bar{F}$. From above these eigenvalues are given by

$$\delta k'_z = k'_z - g = \delta k'_A + \delta k'_N = \frac{\lambda_{0n}}{\cos \theta} \left[ \frac{\langle P_{k'_z} | P_A | P_{k'_z} \rangle}{\langle P_{k'_z} | P_{k'_z} \rangle} + \frac{\langle P_{k'_z} | P_N | P_{k'_z} \rangle}{\langle P_{k'_z} | P_{k'_z} \rangle} \right].$$

(27)
Off-Bragg in a crystal containing a single isotropic resonance with no Zeemann splitting, \( \delta k'_{z} = f^{00} = \lambda_{0}n[f_{A}(0) + f_{N}(0)] \), where \( f_{A}(0) \) and \( f_{N}(0) \) are the electronic and nuclear contributions to the forward scattering amplitude of a single isolated atom, respectively. Well above any electronic absorption frequencies,

\[
f_{A}(0) = -r_{0}Z + i \frac{\sigma_{A}}{4\pi\lambda}.
\]

(28)

Off-resonance, the eigenwave will decay as \( e^{-\mu_{A}z/2} \) on travelling a distance \( z \) through the crystal, where

\[
\frac{\mu_{A}}{2} = \text{Im}(\delta k'_{A})_{OB} = \text{Im}(k'_{z})_{OB} = \frac{n\sigma_{A}}{2}.
\]

(29)

Similarly, near resonance, interaction with the nuclei will give rise to an additional contribution to the absorption of

\[
\exp[-\frac{\sigma_{N}}{X^{2} + 1} n_{z}],
\]

(30)

where \( \sigma_{N} \) is the nuclear cross section at exact resonance and

\[
X = 2(E_{n} - E_{0} - \hbar \omega)/\Gamma \text{ (see Appendix C).}
\]

The strength of the coupling between the multi-beam eigenmodes and the resonant nuclei and electrons determines how rapidly these eigenmodes are absorbed within the crystal. As we shall see, this coupling strength has particular relevance to the lasing problem. In
order to compare the absorption properties of the various multi-beam eigenmodes, let us choose the simple, degenerate off-Bragg case as a standard, and define electronic and nuclear coupling factors, $g_A$ and $g_N$, so that

$$g_A = \frac{\text{Im}(\delta k_A')_{MB}}{\text{Im}(\delta k_A')_{OB}} \quad (311)$$

and

$$g_N = \frac{\text{Im}(\delta k_N')_{MB}}{\text{Im}(\delta k_N')_{OB}} \quad (311i)$$

Thus

$$g_A = \frac{2\lambda_0}{\cos \theta} \frac{\text{Im}(\langle \tilde{f}_A \rangle)}{g_A} \quad (321)$$

and

$$g_N = \frac{2\lambda_0}{\cos \theta} \frac{x^2 + 1}{g_N} \text{Im}(\langle \tilde{f}_N \rangle). \quad (321i)$$

Off-Bragg, $g_A = g_N = 1$.

These coupling factors relate the exponentiation rate of a multi-beam mode propagating down the 2 axis to that of an off-Bragg mode, also propagating down the 2 axis. The factor $1/\cos \theta$ occurs in $g$ due to the increase in pathlength of the radiation in the multi-beam mode as compared to the off-Bragg mode, since each beam in the multi-beam mode is travelling at an angle $\theta$ relative to 2. Note,
however, that at exact Bragg, the direction of energy transport in the multi-beam mode is along \( \hat{z} \), not along the individual beams.

Mössbauer transitions occur between states of well defined angular momentum and parity. In addition, in the energy region of interest here (10 - 100 keV), photoabsorption is primarily electric dipole in nature. Vector spherical harmonics therefore form a natural set of basis functions in which to express \( \bar{r}_A \) and \( \bar{r}_N \). The multipole expansions of these two scattering amplitudes are developed in Appendix C.

As stated above, \( f_A = f_0 + \Delta f' + if'' \), where \( f_0 \) comes from the \( A^2 \) term in the S-matrix expansion, while \( \Delta f' \) and \( f'' \) are the real and imaginary parts of the anomalous dispersion contribution, respectively. Since \( f_0 \) is purely real, only \( f'' \) will contribute to \( g_A \).

Expanding \( \text{Im}(\bar{r}_A) \) in multipoles yields

\[
\text{Im}(\bar{r}_A) = \sum_{L,\lambda} \text{Im}(f_{A, L\lambda}) f_D(\hat{r}_f - \hat{r}_i) \left\{ \frac{8\pi}{2L+1} \right\} \sum_M |Y_{LM}^{\hat{r}_f}(\hat{k}_f)| \langle Y_{LM}^{\hat{r}_i} |, \tag{33}
\]

where

\[
f_D(\hat{r}_f - \hat{r}_i) = \exp(- |\hat{r}_f - \hat{r}_i|^2 \left\langle \frac{x^2}{2} \right\rangle) \tag{34}
\]

is the Debye-Waller factor, and

\[
\text{Im}(f_{A, L\lambda}) = \frac{\sigma_{A, L\lambda}}{4\pi\lambda}. \tag{35}
\]

\( \sigma_{A, L\lambda} \) is the photoelectric cross section for absorbing waves of multipolarity \( L \) and parity \( \lambda \). (The assumption of spherical atoms is
implicit in Eqs. 33 thru 35, so that \( f_{A,L\lambda} \) does not depend upon \( M \). See Appendix C for details.

The photoabsorption parameter in a multi-beam mode may now be written as

\[
\mu_A = n \sum_{L, \lambda} g_{A,L\lambda} \sigma_{A,L\lambda},
\]

where

\[
g_{A,L\lambda} = \frac{1}{\cos \theta} \frac{8\pi}{2L + 1} \sum_M \frac{\langle \hat{Y}_{LM}(\hat{k}_F) \rangle \langle \hat{Y}_{LM}(\hat{k}_1) \rangle \langle \hat{Y}_{LM}(\hat{k}_1) \rangle |P_{k_z} \rangle}{\langle P_{k_z^1} |P_{k_z} \rangle}.
\]

The nuclear coupling factor may be expressed in a similar manner, but since the nuclear transition occurs between particular states of well defined total angular momentum and parity, only one set of values for \( L \) and \( \lambda \) need be included in the sum (or perhaps two, if the Mössbauer transition occurs as a mixture of multipoles, say M1 and E2).

If there is no Zeemann splitting, then

\[
\bar{r}_N = \frac{f_O}{x - 1} \sum_M \frac{\langle \hat{Y}_{LM}(\hat{k}_F) \rangle \langle \hat{Y}_{LM}(\hat{k}_1) \rangle}{\langle \hat{Y}_{LM}(\hat{k}_1) \rangle \langle \hat{Y}_{LM}(\hat{k}_F) \rangle},
\]

where

\[
f_O = \frac{\sigma_N}{4\pi \hbar}
\]
and \( o_N \) is the nuclear cross section at exact resonance,

\[
o_N = \frac{2\pi f}{(1 + a)(1 + a)} \frac{g_n}{g_o}.
\]

(40)

In this equation, \( a \) is the internal conversion coefficient, \((1 + a)\) represents inhomogeneous broadening, \( f \) is the Mössbauer factor, while \( g_n = 2j_n + 1 \) and \( g_o = 2j_o + 1 \) are the spin degeneracies of the two nuclear levels involved in the transition. Thus

\[
ge_{N,L\lambda} = \frac{1}{\cos \theta} \frac{8\pi}{2L + 1} \sum M \frac{\langle P_{k_z} | \hat{Y}_{LM}^{i} (k_f) \rangle \langle \hat{Y}_{LM}^{o} (k_L) | P_{k_z} \rangle}{\langle P_{k_z} | P_{k_z} \rangle}.
\]

(41)

If there is Zeemann splitting, \( g_N \) cannot be defined so easily, since the attenuation rate of an off-Bragg mode now depends upon its direction of propagation relative to the magnetic field axis (see Appendix B). The physical quantity of interest remains \( k_z' \), however, and the attenuation rate of the multi-beam mode may be easily calculated from Eq. 27.

Two other methods by which \( g_A \) and \( g_N \) may be calculated are presented in Appendix D.
Let us examine the absorption properties of these modes in greater detail. If single-plane wave modes are chosen as a basis, the diagonal elements of $\tilde{F}$ will be equal to the off-Bragg value of $\delta k_2$, $f^{00}$. The trace of this matrix is thus $2mf^{00}$. Performing a similarity transformation to a basis consisting of the eigenmodes will diagonalize $\tilde{F}$, but will leave its trace unchanged. Thus the average value of $\delta k_2$ within any $2m$-dimensional subspace defined by an $m$-beam mode must be equal to the value of $\delta k_2$ off-Bragg. (This is a general result which applies at all angles, not just when the Bragg condition is satisfied exactly.)

The average values of both $g_A$ and $g_N$ within the subspace will thus be equal to $1/\cos\theta$. (Again, the path length factor occurs since the beams in the mode are traveling at an angle $\theta$ relative to the $\hat{z}$ axis.) Some modes will thus be absorbed much faster per unit path length than off-Bragg modes, while some will be absorbed much more slowly. Similarly, some modes will couple much more strongly to the resonant nuclei than will the off-Bragg modes, while some modes will couple much more weakly. Indeed, as we shall see, some modes will not couple at all to nuclear transitions of certain multipolarity.

Let us return to the 2-beam example discussed above. To first order, the photoabsorption rate is proportional to the absolute square of the electric field at the equilibrium site of each atom in the crystal. The squares of the electric fields in the two $\sigma$-polarized eigenmodes are given by
\[ |E_+|^2 = |E_0|^2 e^{-2\text{Im}(k_1^\parallel z \left( \frac{\delta - \beta}{\beta F_0} \cos \frac{\vec{r} \cdot \vec{k}_1}{2} + \frac{1}{2i \beta} \left( \frac{\delta + \beta + F_0}{\beta F_0} \right) e^{-\frac{i \vec{r} \cdot \vec{k}_2}{2}} \right)^2} \] (421)

and

\[ |E_-|^2 = |E_0|^2 e^{-2\text{Im}(k_1^\parallel z \left( \frac{\delta + \beta}{\beta F_0} \sin \frac{\vec{r} \cdot \vec{k}_1}{2} + \frac{1}{2i \beta} \left( \frac{\delta + \beta + F_0}{\beta F_0} \right) e^{-\frac{i \vec{r} \cdot \vec{k}_2}{2}} \right)^2}. \] (4211)

At exact Bragg (\(\delta = 0\)), the electric field consists of a pure standing wave in the \(xy\) plane, along with a traveling wave along \(\hat{z}\). The field in the \(k_1^\parallel\) eigenmode has a node in this standing wave at the equilibrium site of the atom, as is shown in Fig. 7. This mode will not couple strongly to the inner shell electrons, and consequently, photoabsorption in this mode is suppressed; that is, this mode will be much more weakly attenuated than will modes off-Bragg or in an amorphous sample. The field in the \(k_1^\parallel\) eigenmode, on the other hand, has an antinode in the standing wave at the site of the atom. This mode will be absorbed much more strongly than will an off-Bragg mode.

---

**Fig. 7.** Standing wave electric fields in the 2-beam Borrmann and anti-Borrmann modes at exact Bragg.
The $k'_l$ mode is referred to as the "Borrmann" mode, while the $k'_r$ mode is termed the "Anti-Borrmann" mode.\textsuperscript{37}

Photoabsorption in the Borrmann mode is not strictly zero since the atom oscillates about its equilibrium position and thus samples the nonzero electric field on either side of the node. Taking this into account yields (for the electric dipole term)

$$g_{A,11} = \frac{1}{\cos\theta} (1 - f_D) = \frac{2\sin^2\theta}{\cos\theta} \frac{<x^2>}{\chi^2}, \quad (43i)$$

where $<x^2>$ is the mean thermal and zero point motion of the atom about its equilibrium position. For the anti-Borrmann mode,

$$g_{A,11} = \frac{1}{\cos\theta} (1 + f_D) = \frac{2}{\cos\theta} \left[1 - \sin^2\theta \frac{<x^2>}{\chi^2}\right]. \quad (43ii)$$

(There are additional finite size effects which account for the higher order terms in Eq. 37. These are discussed in Sections III and IV, as well as in Appendix D.)

If there is more than one atom per unit cell, none of the eigenmodes will, in general, be true Borrmann modes, in that the electric field will not be zero at the equilibrium site of every atom in the crystal.\textsuperscript{47} Therefore, only the case of one atom per unit cell will be considered in this thesis. All the discussion herein will apply to the case of more than one atom per unit cell to the extent
that photoabsorption from one resonant atom in each unit cell is dominant.

Turning now to the nuclear coupling, note that the matrix elements of the nuclear scattering amplitude, \( \text{Im}(\vec{r}_N) \), may be obtained from those of \( \text{Im}(\vec{r}_A) \) by replacing \( q_A \) by \( q_N/(x^2 + 1) \) and by letting \( f_D + 1 \) (see above and Appendix C). The nuclear coupling factor between an E1 emitter and the 2-beam Borrmann mode thus becomes

\[
\delta_{N,11} = \frac{1}{\cos \theta} (1 - 1) = 0
\]

(44)

This is a general result. A Borrmann mode cannot excite E1 Mössbauer transitions, and hence will not be absorbed by such nuclei. The magnetic field and EFG, on the other hand, both have an antinode at the site of each atom in the 2-beam Borrmann mode, so that this mode will be strongly absorbed by M1 and E2 nuclear transitions.

A source atom, that is an excited Mössbauer nucleus, located at a lattice site within a crystal, will emit spherically outgoing wave, in all directions. In particular, it will emit into the directions \( \hat{x}_0 \) and \( \hat{x}_1 \) necessary to feed the Borrmann and anti-Borrmann modes considered above. By applying the reciprocity theorem, we see that E1 Mössbauer transitions cannot feed the Borrmann mode, since a nucleus with an E1 transition will not absorb the wave in that mode. E1 transitions will, however, feed the anti-Borrmann mode. Conversely, M1 and E2 emitters will strongly feed the Borrmann mode. This ability of M1 and higher multipole Mössbauer transitions to feed the weakly
attenuated Borrmann mode is a phenomenon called the "anomalous emission" effect.\textsuperscript{6,7}

As we shall see, Borrmann modes exist for the multi-beam cases in which the suppression of photoabsorption is even stronger than it is in the 2-beam case considered above. As in the 2-beam case, these modes only couple to particular multipole Mössbauer transitions, but never to E1, since, by definition, a Borrmann mode has a node in the electric field at the site of each atom.
III. PROPERTIES OF 2-BEAM BORRMANN MODES

In order to illustrate how the coupling factors are calculated for the various eigenmodes, consider the simple case of a 2-beam mode, where only two wavevectors, for example $\mathbf{k}_1$ and $\mathbf{k}_2$, satisfy the Bragg condition. The two wavevectors lie opposite each other on the surface of a cone, whose base contains the reciprocal lattice vector of the reflection, as is shown in Fig. 8. In this case, the half-apex angle of the cone is the Bragg angle, through this is not in general true for the $m$-beam case. The polarization vectors for each plane wave may be chosen as $\hat{\sigma}_g$ and $\hat{\tau}_g$, where $\hat{\sigma}_g$ lies in the plane defined by the base of the cone (along $\hat{\varphi}$ at each wavevector) and $\hat{\tau}_g$ lies out of the plane, perpendicular to $\hat{\sigma}_g$ (along $\hat{\varphi}$).

Fig. 8. Two-Beam Mode Geometry. The figure on the right shows a projection down onto the plane of the cone base.
The two plane waves are strongly coupled since they satisfy the Bragg condition; neither wave by itself constitutes an eigenwave of the crystal. Rather, these eigenmodes are linear combinations of $\mathbf{F}_1$ and $\mathbf{F}_2$. Since there are two linearly independent polarizations for each beam, there will be four linearly independent eigenmodes.

If there is no Zeeman splitting, the symmetry group of $\mathbf{F}$ at exact Bragg is $C_{2v}$, the point group consisting of the identity, one 2-fold rotation ($c_2$) and two reflections, $a_v$ through a plane containing $\mathbf{F}$ and the cone axis, and $a'_v$ through a plane perpendicular to it, also containing the cone axis. The set of vectors $[|\pi_0\rangle, |\sigma_0\rangle]$ defines a regular representation of this group, and since all the irreducible representations are 1 dimensional, each irreducible representation will be represented once among the eigenmodes; these eigenmodes may thus be uniquely determined from symmetry considerations. The resulting eigenmodes are shown in Fig. 9. (This case is worked out as an example in Appendix B.)

The notation used for the eigenmodes in this thesis, following that of Ref. 26, is $|n\epsilon_m\rangle$, where $m$ is the number of beams and $\epsilon = \pi$ or $\sigma$ refers to the polarization of the beams which make up the mode. The relative phase between adjacent beams in the mode is given by $2\pi n/m$. Thus, for example, $|2\sigma_2\rangle$ denotes the 2-beam mode in which the $\sigma$ polarization of each beam is excited in phase, as shown in Fig. 9.

Note that only one of these four modes, $|2\sigma_2\rangle$, is a Borrmann mode. The electric field in this mode (at exact Bragg) may be written as
Fig. 9. Two-Beam Eigenmodes.

\[ \dot{E}_B = \frac{E_0 e^{ikzx}}{\sqrt{2}} (\hat{\sigma}_1 e^{ik_1x} + \hat{\sigma}_2 e^{ik_2x}). \]  

At the origin, this gives \( \dot{E}_B = E_0 (\hat{\sigma}_1 + \hat{\sigma}_2)/\sqrt{2} = 0. \) All the other modes have a nonzero \( \dot{E} \) at the origin. (The inplane components of \( \dot{E} \) cancel for the mode \(|2\pi\rangle_2\), but the \( \hat{z} \) components do not.) In general, there will be several Borrmann modes for each multi-beam case, each mode with different absorption and coupling properties.

If a magnetic field is applied along the symmetry axis of the cone, so that there is Zeeman splitting of the nuclear levels, reflections containing that axis will no longer leave \( \vec{F} \) invariant. The symmetry group reduces to \( C_2 \), the cyclic point group consisting of the identity and one 2-fold rotation about the cone axis. The order of this group is 2, half that of \( C_{2v} \).
In this case, there is no longer any reflection symmetry to distinguish between the $\varphi$ and $\pi$ modes, and either set, $\{|\pi_2\rangle\}$ or $\{|\varphi_2\rangle\}$, defines a regular representation of the reduced group. Since the $C_2$ is cyclic, all the irreducible representations are 1 dimensional. Each irreducible representation will occur twice among the eigenmodes. $|2\pi_2\rangle$ and $|2\varphi_2\rangle$ belong to the same irreducible representation, as do the pair $|1\pi_2\rangle$ and $|1\varphi_2\rangle$. The new eigenmodes may be found by diagonalizing $\tilde{F}$ within each of these 2x2 subspaces. In general, the eigenmodes calculated without Zeeman splitting will be strongly mixed by a magnetic field applied along the symmetry axis of the mode.

$|2\pi_2\rangle$ and $|2\varphi_2\rangle$, however, will not be mixed by the Zeeman splitting of either E1 or M1 nuclear transitions. $|m\pi_\pm\rangle$ and $|m\varphi_\pm\rangle$ modes only couple to the $M = 0, \pm m, \pm 2m, \ldots$ components of nuclear transitions, where $M = m_n - m_0$ is the change in the $\hat{z}$ component of the angular momentum of the nuclear state due to the transition. E1 and M1 nuclear transitions only contain $M = 0, \pm 1$ components, so that $|2\pi_2\rangle$ and $|2\varphi_2\rangle$ can only couple to the $M = 0$ transition. $\tilde{\chi}_{10}$, however, goes over into itself under reflection (see above), and the eigenmodes which couple only to this term in $\tilde{F}_N$ will remain good eigenmodes in the presence of Zeeman splitting.

Alternatively, this may be seen by noting that $\tilde{\chi}_{10}(\hat{k})$ lies entirely along $\hat{\theta}$ (see Appendix A), and is thus orthogonal to $|2\pi_2\rangle$. $|2\pi_2\rangle$, therefore, does not couple to M1 transitions. Similarly, $\tilde{\chi}_{10}(\hat{k})$ lies entirely along $\hat{\phi}$ and is orthogonal to $|2\varphi_2\rangle$. $|2\varphi_2\rangle$ does not couple to E1 transitions. In each case (E1 or M1 mixing), one of
the two modes has a zero eigenvalue for the transition, and the two modes will not be mixed. $|2\pi \rangle_2$ and $|2\sigma \rangle_2$ will remain good eigenmodes even in the presence of Zeeman splitting caused by a field applied along the symmetry axis of the cone.

This can be seen physically by noting that $M = 0$ transitions correspond classically to either a magnetic or electric dipole oscillating only along the $z$-axis. Changing the sign of the magnetic field merely introduces a global phase shift which is of no physical consequence. Therefore, the eigenmodes calculated including reflections as good symmetry operators (group $C_{2v}$) will remain eigenmodes of the reduced group, $C_2$.

These modes, $|2\pi \rangle_2$ and $|2\sigma \rangle_2$, will, however, be mixed by the Zeeman splitting of an E2 or M2 transition, since in this case, they will couple to the $M = \pm 2$ components, which contain contributions from both $\pi$ and $\sigma$ polarizations. This is a general result; $|m\pi \rangle_m$ and $|m\sigma \rangle_m$ will not be mixed by the Zeeman splitting of any nuclear transition with $L < m$, but will be mixed by the $\pm m, \pm 2m, \ldots$ components if $L \geq m$.

Consider the E1 contribution to photoabsorption in the 2-beam Borrmann mode, $|2\sigma \rangle_2$. Within this 2-beam subspace, the imaginary part of the E1 atomic scattering amplitude is given in the $\{|\pi_3 \rangle, |\sigma_3 \rangle\}$ basis by
\[
\text{Im}(\tilde{f}_A,_{11}) = \frac{\sigma_{A,11}}{4\pi}\begin{pmatrix}
1 & 0 & \zeta & 0 \\
0 & 1 & 0 & -f_D \\
\zeta & 0 & 1 & 0 \\
0 & -f_D & 0 & 1 \\
\end{pmatrix}
\]

where \( \zeta = (-\alpha^2 + s^2)f_D \), \( \alpha \) and \( s \) referring to \( \cos\theta \) and \( \sin\theta \), respectively. \( \sigma_{A,11} \) is the E1 partial photoelectric cross section.

Transforming to a new basis consisting of the eigenmodes, \(|n\epsilon>2\rangle\), \( n=1,2 \), \( \epsilon = \pi,\sigma \) and again writing \( \text{Im}(\tilde{f}_A,_{11}) \) yields the diagonal matrix

\[
\text{Im}(\tilde{f}_A,_{11}) = \frac{\sigma_{A,11}}{4\pi}\begin{pmatrix}
1 + \zeta & 0 & 0 & 0 \\
0 & 1 - f_D & 0 & 0 \\
0 & 0 & 1 - \zeta & 0 \\
0 & 0 & 0 & 1 + f_D \\
\end{pmatrix}
\]

Notice that the trace of the matrix remains unchanged, as it must since a change of basis is simply a similarity transformation of the matrix. The average photoabsorption within this 2-beam subspace is thus equal to that of an off-Bragg mode; some of the 2-beam modes will be attenuated more strongly than off-Bragg, and some less.
In the Borrmann mode, \(|\sigma_1\rangle\) and \(|\sigma_2\rangle\) are excited in phase with equal amplitude, i.e. \(2<2\sigma|\sigma_2\rangle \approx (0,1,0,1)\). Taking the expectation value of \(\tilde{r}_{A,11}\) yields

\[
\text{Im}(r_{A,11}) = \frac{\text{Im}(2<2\sigma|\tilde{r}_{A,11}|2\sigma\rangle_2)}{2<2\sigma|2\sigma\rangle_2} = (1 - f_D) \frac{\sigma_{A,11}}{4\pi\chi},
\]

so that

\[
\sigma_{A,11} = \frac{1}{\cos^2\theta}(1 - f_D) \frac{2\sin^2\theta \langle x^2 \rangle}{\chi^2}.
\]

Higher order terms may be included by evaluating contributions from higher multipoles. The imaginary part of the atomic scattering amplitude for M1 photoabsorption is given by the matrix

\[
\text{Im}(\tilde{r}_{A,10}) = \frac{\sigma_{A,10}}{4\pi\chi}
\]

\[
\begin{pmatrix}
1 & 0 & -f_D & 0 \\
0 & 1 & 0 & \zeta \\
-f_D & 0 & 1 & 0 \\
0 & \zeta & 0 & 1
\end{pmatrix}
\]

which yields for the Borrmann mode
\[ g_{A,10} = \frac{1}{\cos \theta} \left( 1 + (-c^2 + s^2) f_D \right) \]

\[ = \frac{2 \sin^2 \theta}{\cos \theta} + \frac{2 (\cos^2 \theta - \sin^2 \theta)}{\cos \theta} \langle x^2 \rangle. \]  

(48)

Note that this contribution is not suppressed, that is, the leading term in \( g_{A,10} \) is not proportional to \( \langle x^2 \rangle / x^2 \). The magnetic field is large at the equilibrium site of the atom in the Borrmann mode, so the first order contribution does not depend upon the motion of the atom. Nevertheless, the magnetic dipole photoabsorption cross section is usually several orders of magnitude smaller than the electric dipole and quadrupole cross sections (see Appendix C), and this term may generally be neglected.

The first important correction to Eq. 46 will be E2 term. The scattering amplitude operator for this multipole is given by

\[
\begin{pmatrix}
|\pi_1> & |\sigma_1> & |\pi_2> & |\sigma_2>
\end{pmatrix}
\begin{pmatrix}
1 & 0 & \xi & 0 \\
0 & 1 & 0 & \zeta \\
\xi & 0 & 1 & 0 \\
0 & \zeta & 0 & 1
\end{pmatrix}
\]

\[
\text{Im}(\tilde{r}_{A,21}) = \frac{g_{A,21}}{4\pi} f_D
\]

where \( \xi = [6s^2c^2 - (c^4 + s^4)]f_D \) and \( \zeta = (-c^2 + s^2)f_D \), as before. This yields for the Borrmann mode
\[ g_{A,21} = \frac{2\sin^2 \theta}{\cos \theta} + \frac{2(\cos^2 \theta - \sin^2 \theta)}{\cos \theta} \langle x^2 \rangle \tag{49} \]

Note that E2 photoabsorption is not suppressed in this mode either; the electric field gradient is also large at the equilibrium site of the atom. The E2 cross section is somewhat smaller than the cross section for E1 photoabsorption (again, see Appendix C), so that the contribution of this term to \( \mu_A \) will be comparable to that of the suppressed E1 contribution.

To calculate \( g_N \) for the case of an M1 transition with no Zeemann splitting, note that \( f_{N,10} \) is given by

\[
\begin{pmatrix}
|\pi_1\rangle & |\varphi_1\rangle & |\pi_2\rangle & |\varphi_2\rangle \\
1 & 0 & -1 & 0 \\
0 & 1 & 0 & \xi \\
-1 & 0 & 1 & 0 \\
0 & \xi & 0 & 1
\end{pmatrix}
\]

where now \( \xi = (-c^2 + s^2) \). This yields for the Borrmann mode

\[
g_{N,10} = \frac{1}{\cos \theta} (1 - \cos^2 \theta + \sin^2 \theta) = \frac{2\sin^2 \theta}{\cos \theta}. \tag{50}
\]

\( g_{N,10} \) is large because the magnetic field in this mode is large at the site of the nucleus. The factor of 2 occurs because there are two
beams which are adding coherently to produce that field. As will be seen later, coupling factors for m-beam modes are always proportional to m, due to the coherent interaction of m beams with the atoms of the crystal.

For coupling factors of the 2-beam Borrmann mode to nuclear transitions of other multipoles, see Ref. 6.

To treat the fully Zeemann split case, note that the nuclear scattering amplitude matrices for the three M1 transitions \((M = 0, \pm 1)\) are given by

\[
(\tilde{f}_{N,10})_{+1} = \frac{1}{2} f_{0}^{\pm}(m_{0}, +1, m_{n})
\]

\[
\begin{pmatrix}
|\pi_{1}\rangle & |\sigma_{1}\rangle & |\pi_{2}\rangle & |\sigma_{2}\rangle \\
1 & -ic & -1 & +ic \\
+ic & c^{2} & -ic & -c^{2} \\
-1 & +ic & 1 & -ic \\
-ic & -c^{2} & +ic & c^{2}
\end{pmatrix}
\]

\[
(\tilde{f}_{N,10})_{0} = f_{0}^{0}(m_{0}, 0, m_{n})
\]

\[
\begin{pmatrix}
0 & 0 & 0 & 0 \\
0 & s^{2} & 0 & s^{2} \\
0 & 0 & 0 & 0 \\
0 & s^{2} & 0 & s^{2}
\end{pmatrix}
\]
\[
(f_{N,10})^{-1} = \frac{1}{2} f_0'(m_o,-1,m_n) \begin{pmatrix}
1 & +ic & -1 & -ic \\
-il & c^2 & +ic & -c^2 \\
-1 & -ic & 1 & +ic \\
+ic & -c^2 & -ic & c^2
\end{pmatrix}
\]

where

\[
f_0'(m_oM_m) = f_0 \frac{2L + 1}{2J_n + 1} \frac{c^2(J_oL_j_n^i m_oM_m)}{\chi(m_oM_m) - 1}.
\]

(51)

Taking the expectation value of either \((f_{N,10})^+\) or \((f_{N,10})^-\) in the Borrmann mode gives identically zero, so that

\[
(k_{z;N,10}^+ = (k_{z;N,10}^-) = 0.
\]

(52)

The Borrmann mode does not couple to these transitions. Taking the expectation value of \((f_{N,10})_0\), however, yields

\[
\text{Im}[(f_{N,10})_0] = 2 \sin^2 \theta f_0'(m_o,0,m_n)
\]

(53)

so that

\[
(k_{z;N,10}^0 = \lambda_o n \frac{2 \sin^2 \theta}{\cos \theta}.
\]

(54)
The 2-beam Borrmann mode only couples to the $M = 0$ transition of $M_1$. This is easily understood by noting that the two $\phi$ plane wave channels which make up the mode are excited in phase, which is exactly the sort of radiation pattern produced by a classical magnetic dipole oscillating along the symmetry axis of the cone (i.e. classical oscillation which corresponds to the $M = 0$ transition).

The radiation pattern produced by a $M = \alpha$ transition will change sign every $\phi = \pi/\alpha$. A mode will only couple to that transition if the excitations in the various beams which make up that mode change sign accordingly. Thus, while the Borrmann mode ($\ket{2\pi}_2$ in Fig. 9) does not couple to $M = \pm 1$ transitions, both $\ket{1\pi}_2$ and $\ket{1\sigma}_2$ will. (Neither of these are Borrmann modes, however.)

If there is Zeemann splitting, $\ket{1\pi}_2$ and $\ket{1\sigma}_2$ will be strongly mixed in the neighborhood of the $M = \pm 1$ resonances. The matrix for $(\tilde{r}_{N,10})_{+1}$, for example, in this subspace is

$$
(\tilde{r}_{N,10})_{+1} = \frac{f_0'(m_0',+1,m_n)}{2} \begin{pmatrix}
1 & -ic \\
ic & c^2
\end{pmatrix}
$$

Neglecting the interaction with the electrons for the moment, diagonalizing this matrix yields the eigenmodes

$$
\ket{\psi_1} \propto \{ \ket{1\pi}_2 + i \cos \theta \ket{1\sigma}_2 \}
$$

and
\[ |\psi_2\rangle = |\cos\theta|\pi_2 \rangle + |\sin\theta_2\rangle, \]  
\[ k'_z;1 = 0 \]  
\[ k'_z;2 = \lambda_o n f_0(m_o,0,m_n) \frac{1 + \cos^2\theta}{\cos\theta}, \]

respectively. The eigenmodes for the \( M = -1 \) transition may be obtained by simply taking the complex conjugate Eqs. 55. Neither of these modes couples to the \( M = 0 \) transition, so that they will not be mixed by this term.

The above expressions for the eigenmodes only apply in the limit that the nuclear contribution is much stronger than the contribution from the electrons. The true eigenmodes for the case of \( M = \pm 1 \) mixing must be found by diagonalizing \( f_A + f_N \); they will thus depend upon the relative strength of these two contributions.

If a mode couples to an \( M = \pm \alpha \) transition, it will also couple to transitions with \( M = \pm (\alpha + n \beta) \), where \( \beta \) is the number of beams in the mode, and \( n \) is any positive integer. Thus \( |2\alpha\rangle \) and \( |2\pi\rangle \) both couple to the \( M = \pm 2 \) components of an E2 transition, as well as the \( M = 0 \) component. If there is Zeemann splitting, they will be strongly mixed at the resonance energies of the \( M = \pm 2 \) transitions, and neither of the resulting eigenmodes will be a good Borrmann mode. The non-Zeemann split results will still remain valid, however, in the vicinity of the \( M = 0 \) resonances, provided the energy levels are
sufficiently split so that only one transition contributes at any given energy.
IV. PROPERTIES OF MULTI-BEAM BORRMANN MODES

Let us now consider the multi-beam Borrmann modes. Reviewing the results of Section II, we know that such an m-beam mode will consist of m coherent plane waves (beams), whose wavevectors, denoted \( \mathbf{R}_1 \) through \( \mathbf{R}_m \), lie on the surface of a cone, the base of which contains all the reciprocal lattice vectors involved in the reflections. The angle which each beam makes with the cone axis, \( \Theta \), is not, in general, a Bragg angle. The direction of energy propagation in the mode is down the symmetry axis, rather than along the individual beams. The general structure of these modes, as well as the notation used to describe them, is shown in Fig. 10.

There are two independent polarization vectors associated with each plane wave channel, \( \mathbf{R}_s \), which may be chosen to be \( \hat{\sigma}_s \) and \( \hat{\pi}_s \), where \( \hat{\sigma}_s \) lies in the plane of the reciprocal lattice vectors (along \( \hat{\sigma} \) if the cone axis is denoted as \( \hat{z} \)), and \( \hat{\pi}_s \) lies out of the plane.

![Diagram](image)

**Fig. 10.** General structure of the multi-beam modes, as well as the notation used to describe them.
perpendicular to both \( \hat{a}_3 \) and \( \hat{X}_1 \) (along \( \hat{z} \)). There are thus 2m eigenmodes within this subspace. As in the 2-beam case, only some of these eigenmodes will be Borrmann modes. The Borrmann modes will be of primary interest for both the lasing problem and for most single photon problems; the following analysis will, therefore, be restricted primarily to them.

**Borrmann Modes Corresponding to 1-D Representations**

**Modes Formed on Regular Polygons**

If the \( m \)-beam mode is centered about an \( m \)-fold rotation axis of the reciprocal lattice plane which contains its base, the \( m \) beams will be symmetrically distributed around the surface of the cone and the reciprocal lattice points involved in the reflections will lie at the vertices of a regular \( m \)-sided polygon. If there is no Zeemann splitting, the set of vectors \( \{|\pi_3\rangle, |\sigma_3\rangle\} \) will form a basis in this subspace which defines a regular representation of the symmetry group of the mode, \( C_{nv} \). The one-dimensional irreducible representations will thus appear only once among the eigenmodes, and the structure of these modes may be determined solely by symmetry.

Each point group, \( C_{nv} \), possesses either two (if \( n \) is odd) or four (if \( n \) is even) 1-dimensional irreducible representations. The eigenmodes obtained from these irreducible representations in the \( m \)-beam case are precisely analogous to the four eigenmodes found in
the 2-beam case. These eigenmodes will again be denoted $|m\pi\rangle_m$, $|m\sigma\rangle_m$, $|\frac{m\pi}{2}\rangle_m$, and $|\frac{m\sigma}{2}\rangle_m$.

$|m\pi\rangle_m$ and $|m\sigma\rangle_m$ are modes in which either all the waves of $\pi$ polarization, or all the waves of $\sigma$ polarization, are excited equally and in phase. At the origin, $|m\pi\rangle_m$ possesses a non-zero electric field along the $\hat{z}$-axis, and is never a Borrmann mode, regardless of the number of beams. $|m\sigma\rangle_m$, on the other hand, is always a Borrmann mode.

$|\frac{m\pi}{2}\rangle_m$ and $|\frac{m\sigma}{2}\rangle_m$ are antisymmetric $\sigma$ and $\pi$ modes, in which either all the waves of $\sigma$ polarization or all the waves of $\pi$ polarization are again excited equally, but with neighboring beams $\pi$ out of phase. Since the phase of each beam relative to the others must be uniquely defined, such modes obviously exist only when $n$ is even. For the 2-beam case, $|\frac{m\sigma}{2}\rangle_m$ and $|\frac{m\pi}{2}\rangle_m$ both have nonzero in-plane components of $\mathbf{E}$ at the origin, so they are not Borrmann modes. For $n \geq 4$, however, these modes are Borrmann modes.

![Excitation patterns](image)

Fig. 11. Excitation patterns of the 3, 4, and 6 beam $|m\sigma\rangle_m$ Borrmann modes.
The excitation patterns for the 3, 4, and 6-beam $|m_0\rangle_m$ modes are shown schematically in Fig. 11. These modes couple well to $M = 0$ transitions, but not to $M = \pm 1$ or $\pm 2$. The 3-beam mode will couple to $M = \pm 3$ transitions, however. In general, $|m_0\rangle_m$ and $|m\rangle_m$ modes couple to $M = 0$, $\pm m$, $\pm 2m$ transitions, etc.

These modes contain waves of $\hat{z}$ polarization exclusively, and, since the $M = 0$ transition of E1 lies entirely along $\pi$, will not couple to E1 transitions (i.e. they are Borrmann modes). The next $M$ transition which can excite them is $\pm m$. Thus, $|m_0\rangle_m$ modes will not couple at all to any electric nuclear transition with $L < m$ (i.e. $|3\sigma\rangle_3$ will couple to E3 via the $M = \pm 3$ transitions, but will not couple to E2).

If a mode does not couple to a particular multipole nuclear transition, the corresponding contribution to photoabsorption in that mode will be suppressed. To see this, note that, aside from the leading multiplicative constants, the only difference between the operators $\text{Im}(\vec{r}_{A,L\lambda})$ and $\text{Im}(\vec{r}_{N,L\lambda})$ is the presence of the Debye-Waller factor, $f_D$, in $\text{Im}(\vec{r}_A)$ (see Appendix C); that is, $\text{Im}(\vec{r}_N)$ may be obtained from $\text{Im}(\vec{r}_A)$ by replacing $\sigma_A$ by $\sigma_N/(x^2 + 1)$ and letting $f_D = 1$. Now, $f_D$ may be approximated as $1 - W$, where $W = (\Delta R)^2 <x^2>/2$. If the expectation value of $\text{Im}(\vec{r}_{N,L\lambda})$ is zero for a particular eigenmode, the expectation value of $\text{Im}(\vec{r}_{A,L\lambda})$ must be such that the leading term in the expansion of $f_D$ cancels, leaving a term proportional to $W$, at most. Since $W$ is small, photoabsorption of this multipole is suppressed.
The E1 term in the photoabsorption expansion is the contribution due to a point electric dipole located at the center of mass of the atom. If the atom did not oscillate about its equilibrium position, this term would strictly be zero in a Bollmann mode. The term of order $W$, coming from the Debye-Waller factors in $f_{A,11}$, gives the first correction due to this oscillation.

Let the Debye-Waller factors be denoted as

$$f_{D1} = \exp[-(\vec{r}_j - \vec{r}_{j+1}) \frac{<x^2>}{2}].$$  \hspace{1cm} (57)

Note that $f_{D1}$ is independent of $j$ by symmetry. Calculating the E1 contribution to photoabsorption then yields the following:

$$g_{A,11} = \frac{1}{\cos \theta} (1 - f_{D1}) \hspace{1cm} 3\text{-beam} \hspace{1cm} (58i)$$

$$g_{A,11} = \frac{1}{\cos \theta} (1 - f_{D2}) \hspace{1cm} 4\text{-beam} \hspace{1cm} (58ii)$$

$$g_{A,11} = \frac{1}{\cos \theta} (1 - f_{D1} - f_{D2} + f_{D3}) \hspace{1cm} 6\text{-beam} \hspace{1cm} (58iii)$$

Upon expansion, each of these expressions reduces to (to order $W$)

$$g_{A,11} = \frac{m \sin^2 \theta}{2} \frac{<x^2>}{x^2} \hspace{1cm} (59)$$
Compare this with Eq. 47, and note the additional factor of two in the 2-beam case. There is a difference between the symmetry properties of the 2-beam \(|m_0\rangle_m\) mode and those with 3 beams or more. In the 2-beam case, the standing wave pattern in the crystal has nodal planes, while in the multi-beam modes, there are nodal lines. Thus, the symmetry of the field sampled by the electrons as the atoms vibrate about their equilibrium positions is different in the two cases. This difference in symmetry between the 2-beam and the multi-beam cases accounts for the additional factor of 2.

Another way to see that this must be the case is to note that the E1 polarization response goes as \(\hat{c}_f \cdot \hat{c}_i\) (see Appendix C); thus it cannot connect the two \(|2\sigma_2\rangle\) modes which coherently make up \(|4\sigma_4\rangle\). To this order (E1), the two 2-beam modes act independently; they therefore will exponentiate at the same rate. Thus \((g_{A,11})_{4\text{-beam}}\) must equal \((g_{A,11})_{2\text{-beam}}\), hence the additional factor of 2.

The M1 contribution to photoabsorption in these modes has the form

\[
\begin{align*}
g_{A,10} &= \frac{1}{\cos \theta} (1 - (c^2 - 2s^2)f_{D1}) & \quad \text{3-beam} & \quad (60i) \\
g_{A,10} &= \frac{1}{\cos \theta} (1 + 2s^2f_{D1} f_{D2} - (c^2 + s^2)f_{D2}) & \quad \text{4-beam} & \quad (60ii) \\
g_{A,10} &= \frac{1}{\cos \theta} (1 + (c^2 + 2s^2)f_{D1} - (c^2 - 2s^2)f_{D2} - (c^2 - s^2)f_{D3}) & \quad \text{6-beam} & \quad (60iii)
\end{align*}
\]
where $c$ and $s$ refer to $\cos \theta$ and $\sin \theta$, respectively. The leading term in each of these expressions is

$$g_{A,10} = \frac{m \sin^2 \theta}{\cos \theta}.$$  \hspace{1cm} (61)

These modes couple to M1 transitions; M1 photoabsorption is, therefore, not suppressed. It may, however, generally be neglected, since the cross sections for this process are several orders of magnitude smaller than those for E1 (see Appendix C).

The E2 term gives the first correction due to the finite size of the electron shells. These multi-beam modes do not couple to E2 nuclear transitions, and their E2 contribution to $g_A$ will thus also be small. The calculation of the E2 contribution to photoabsorption yields:

$$g_{A,21} = \frac{1}{\cos \theta} \left( 1 - f_D \right) = \frac{3}{2} \frac{\sin^2 \theta}{\cos \theta} \frac{\langle x^2 \rangle}{\kappa^2}$$ \hspace{1cm} 3-beam \hspace{1cm} (62i)

$$g_{A,21} = \frac{1}{\cos \theta} \left( 1 - 2\sin^2 \theta f_{D1} - (\cos^2 \theta - \sin^2 \theta) f_{D2} \right)$$

$$= 2 \cos \theta \sin^2 \theta \frac{\langle x^2 \rangle}{\kappa^2}$$ \hspace{1cm} 4-beam \hspace{1cm} (62ii)

$$g_{A,21} = \frac{1}{\cos \theta} \left( 1 + (\cos^2 \theta - \sin^2 \theta) f_{D1} - f_{D2} - (\cos^2 \theta - \sin^2 \theta) f_{D3} \right)$$
\[ = 3 \cos \theta \sin^2 \frac{x^2}{\lambda^2} \]

6-beam \( (62iii) \)

Notice that in each case, \( g_{A,21} \) is proportional to \( \frac{x^2}{\lambda^2} \), i.e. E2 photoabsorption is suppressed. Compare this with \( g_{A,21} = 2\sin^2 \theta / \cos \theta \) for the mode \( |2\theta_2 \rangle \) (Eq. 49), which couples well to E2 transitions. The electronic coupling factors, \( g_{A,11} \) and \( g_{A,21} \),

![Graph](image)

Fig. 12. Plot of \( g_{A,11} \) and \( g_{A,21} \) vs. cone angle for the 3, 4, and 6-beam \( |mo\rangle_m \) Borrmann modes. The \( [\cos \theta]^{-1} \) path length contribution has been factored out for clarity. \( \xi = \frac{x^2}{\lambda^2} \).
are plotted as a function of cone angle, \( \theta \), in Fig. 12 for the 3, 4, and 6-beam \( |m_0 \rangle_m \) Borrmann modes.

Calculation of the M1 nuclear coupling factor, \( g_{N,10} \), for these modes yields

\[
g_{N,10} = m \, \frac{\sin^2 \theta}{\cos \theta}.
\]

Note that this agrees with Eq. 50 for the 2-beam case. No additional factor of 2 occurs in the 2-beam \( g_N \) since this is a first order effect, and does not depend upon the vibration of the nuclei.

Except for the \( [\cos \theta]^{-1} \) path length term, \( m \) beams, acting coherently with the proper symmetry, can couple up to \( m \) times as well as a single beam to the \( M = 0 \) transition of an M1 oscillator. This is a general result; the coupling of any \( m \)-beam mode to a nuclear transition is proportional to \( m \).

In the presence of Zeemann splitting, the eigenvalue for these modes in the vicinity of the \( M = 0 \) magnetic dipole resonance becomes

\[
(k_z')_0 = \lambda_0 n f'(m_0,0,m_n) \, \frac{m \sin^2 \theta}{\cos \theta},
\]

where \( f'(m_0,0,m_n) \) is given by Eq. 51. These modes do not couple to the \( M = \pm 1 \) resonances.

The coupling factors for M2 and M3 oscillators are
$$\varepsilon_{N,20} = \frac{3}{2} m \sin^2 \theta \cos \theta$$

(65)

and

$$\varepsilon_{N,30} = \frac{9}{16} \frac{\sin^2 \theta}{\cos \theta} (45 \cos^4 \theta - 15 \cos^2 \theta + 2)$$

3-beam case

(66i)

$$\varepsilon_{N,30} = \frac{3}{8} m \frac{\sin^2 \theta}{\cos \theta} (5 \cos^2 \theta - 1)^2.$$ 4 and 6-beam cases

(66ii)

The reason that \(\varepsilon_{N,30}\) does not follow the pattern for the mode \(|3\sigma\rangle_3\)
is that this mode couples to \(M = \pm 3\) transitions, which are allowed for\(M_3\), as well as \(M = 0\). The presence of the two additional terms changes the form of \(\varepsilon_N\). A similar thing happens for the 2-beam mode, coupled to an \(M_2\) oscillator, where

$$\varepsilon_{N,20} = 4 \sin^2 \theta \cos \theta,$$

(67i)

instead of

$$\varepsilon_{N,20} = 3 \sin^2 \theta \cos \theta,$$

(67ii)

as one would expect from Eq. 65. Again, the 2-beam mode couples to the \(M = \pm 2\) transitions of \(M_2\), as well as the \(M = 0\).

As stated above, the first electric transition to which these modes will couple is \(E_m\), so that, for example, the mode \(|3\sigma\rangle_3\) will not couple to \(E_2\), but will to \(E_3\), with

$$\varepsilon_{N,31} = \frac{45 \sin^4 \theta}{16 \cos \theta},$$

(68)
while the 4-beam mode couples to $E_4$ with

\[ \xi_{N,41} = \frac{7}{2} \sin^6 \theta. \]  

(69)

$\xi_N$'s for coupling the 2, 3, 4, and 6-beam $|m_o \geq m$ Borrmann modes to M1 and M2 oscillators are plotted as a function of cone angle in Fig. 13.

---

Fig. 13. Plot of $\xi_{N,10}$ and $\xi_{N,20}$ vs. cone angle for the 2, 3, 4, and 6-beam $|m_o \geq m$ Borrmann modes. The $[\cos \theta]^{-1}$ path length contribution has again been factored out for clarity.
Fig. 14. Excitation patterns for the $|2\sigma\rangle_4$ and $|2\pi\rangle_4$ Borrman modes.

The excitation patterns for the modes $|2\sigma\rangle_4$ and $|2\pi\rangle_4$ are shown in Fig. 14. Like the $|4\sigma\rangle_4$ mode, these may be thought of as being made up of two coherent $|2\sigma\rangle_2$ or $|2\pi\rangle_2$ modes, but now added $\pi$ out of phase. The phase of the excitation changes sign every $\pi/2$ radians around the mode, so that these modes couple to $M = \pm 2, \pm 6 (m+2)$, etc. transitions, but not to $M = 0$ or $M = \pm 1$. These modes cannot be fed by E1 or M1 oscillators and photoabsorption of both these multipoles will be suppressed. The expressions for $g_{A,11}$ and $g_{A,10}$ are given by:

$$(g_{A,11})_\sigma = (g_{A,10})_\pi = \frac{1}{\cos^8} (1 - f_{D2}) = 2 \frac{\sin^2 \theta}{\cos^8} \frac{<x^2>}{\chi^2}$$  \hspace{1cm} (701)$$

$$(g_{A,11})_\pi = (g_{A,10})_\sigma = \frac{1}{\cos^8} \left[ 1 - 2\sin^2 \theta f_{D1} - (\cos^2 \theta - \sin^2 \theta) f_{D2} \right]$$

$$= 2 \cos^8 \frac{\sin^2 \theta}{\chi^2} \frac{<x^2>}{\chi^2}.$$  \hspace{1cm} (7011)$$
The corresponding expressions for $g_{A,21}$ are given by:

$$g_{A,21} = \frac{1}{c} \left( 1 + 2s^2f_{D1} - (c^2 - s^2)f_{D2} \right)$$

$$= 4 \frac{s^2}{c} + 2 \frac{1 - 3s^2}{c} \frac{x^2}{\lambda^2} \quad |2\sigma\rangle_4 \text{ mode} \quad (71i)$$

$$g_{A,21} = \frac{1}{c} \left( 1 - 4s^2c^2f_{D1} - (c^4 - 6s^2c^2 + s^4)f_{D2} \right)$$

$$= 4s^2c + 2(c^4 - 4s^2c^2 + s^4) \frac{x^2}{\lambda^2} \quad |2\pi\rangle_4 \text{ mode}, \quad (71ii)$$

where, again, $c$ and $s$ denote $\cos\theta$ and $\sin\theta$, respectively. Again, the M1 contribution to photoabsorption may be neglected in this case.

The photoabsorption parameters, $g_{A,11}$ and $g_{A,21}$, are plotted in Fig. 15 as a function of cone angle, $\theta$, for the $|2\sigma\rangle_4$ and $|2\pi\rangle_4$ Borrmann modes.

As stated above, neither $|2\sigma\rangle_4$ or $|2\pi\rangle_4$ couple to E1 or M1 nuclear transitions. They do, however, couple to E2 and M2. For the case of no Zeemann splitting, the coupling factors are given by

$$(g_{N,21})|2\sigma\rangle_4 = (g_{N,20})|2\pi\rangle_4 = 4 \frac{\sin^2\theta}{\cos\theta} \quad (72i)$$

and

$$(g_{N,20})|2\sigma\rangle_4 = (g_{N,21})|2\pi\rangle_4 = 4 \sin^2\theta \cos\theta. \quad (72ii)$$

The nuclear coupling factors, $g_{N,20}$ and $g_{N,21}$, are plotted in Fig. 16
Fig. 15. Plot of $g_{A,11}$ and $g_{A,21}$ vs. cone angle for the $|2\sigma\rangle_4$ and $|2\pi\rangle_4$ Borrmann modes. The $|\cos\theta|\bar{\xi}$ path length contribution has been factored out for clarity. $\xi = \langle x^2 \rangle / \langle x \rangle^2$.

as a function of cone angle, $\theta$, for the $|2\sigma\rangle_4$ and $|2\pi\rangle_4$ Borrmann modes.

Even though they each couple to E2 and M2 oscillators, such transitions will not mix these two modes. $|2\sigma\rangle_4$ and $|2\pi\rangle_4$ correspond to different irreducible representations of the group $C_{4v}$. Indeed, if the cross term is calculated, one finds that

$$4<2\sigma|\tilde{f}_{N,20}|2\pi\rangle_4 = 4<2\sigma|\tilde{f}_{N,21}|2\pi\rangle_4 = 0.$$  \hspace{1cm} (73)
Fig. 16. Plot of $g_{N,2\alpha}$ and $g_{N,2\pi}$ vs. cone angle for the $|2\alpha\rangle_4$ and $|2\pi\rangle_4$ Bornmann modes. The $[\cos \theta]^{1/2}$ path length contribution has been factored out for clarity.

If there is Zeemann splitting, however, these modes will be mixed by the $M = \pm 2$ components of E2 or M2 transitions, since they both belong to the same irreducible representation of the point group $C_4$.

The expressions given in Section III for the mixing of the $|1\pi\rangle_2$ and $|1\sigma\rangle_2$ modes by the $M = \pm 1$ components of E1 transitions may be generalized to the case of the mixing of $|m\pi\rangle_m$ and $|m\sigma\rangle_m$ by the $M = \pm \frac{m}{2}$ components of $E \frac{m}{2}$ and $M \frac{m}{2}$ transitions. In each case, the matrix of $(\hat{r}_{N,\lambda})_{\pm L/2}$ in the $|m\pi\rangle_m$ and $|m\sigma\rangle_m$ basis is given by
\[ (\tilde{r}_{N,L1})_{\pm L/2} = \mathcal{D}_{L,m}(\theta) f' \]

\[
\begin{pmatrix}
  c^2 & -ic \\
  ic & 1
\end{pmatrix}
\]

for electric transitions, and by

\[ (\tilde{r}_{N,L0})_{\pm L/2} = \mathcal{D}_{L,m}(\theta) f' \]

\[
\begin{pmatrix}
  1 & -ic \\
  ic & c^2
\end{pmatrix}
\]

for magnetic transitions, where

\[
\mathcal{D}_{L,m}(\theta) = \frac{8\pi m}{2L+1} |\tilde{r}_{L,\pm L/2,\hat{e}}|^2 = \frac{8\pi m}{2L+1} |\tilde{r}_{L,\pm L/2,\hat{p}}|^2. \tag{74}
\]

For the 4-beam case, \( \mathcal{D}_{2,4}(\theta) = 2\sin^2\theta \).

Neglecting the interaction with the electrons for the moment and diagonalizing these matrices for the case \( M = \pm L/2 \) yields, for the electric case, the following eigenmodes:

\[
|\psi_1\rangle = \left\{ |\frac{m}{2}\pi\rangle_m - ic\cos\phi |\frac{m}{2}\rangle_m \right\} \tag{75i}
\]

and

\[
|\psi_2\rangle = \left\{ ic\cos\phi |\frac{m}{2}\pi\rangle_m - |\frac{m}{2}\rangle_m \right\}, \tag{75ii}
\]

with eigenvalues
\[ (k_{z,N}^1)_1 = 0 \]  
\[ (k_{z,N}^1)_2 = \frac{\lambda_0 n}{\cos \theta} \mathcal{D}_{L,m} (\theta) f_0^* (1 + \cos^2 \theta), \]  
(76i)  
(76ii)

respectively. For the magnetic case, the eigenmodes are

\[ |\psi_1\rangle = \{i \cos \theta |\frac{m}{2}\rangle_m + |\frac{m}{2}\rangle_m \} \]  
(77i)

and

\[ |\psi_2\rangle = \{|\frac{m}{2}\rangle_m + i \cos \theta |\frac{m}{2}\rangle_m \}, \]  
(77ii)

again with eigenvalues

\[ (k_{z,N}^1)_1 = 0 \]  
(78i)

and

\[ (k_{z,N}^1)_2 = \frac{\lambda_0 n}{\cos \theta} \mathcal{D}_{L,m} (\theta) f_0^* (1 + \cos^2 \theta), \]  
(78ii)

respectively. In each case, the modes are strongly mixed, so that one of the resulting eigenmodes couples strongly to the transition, while the other does not couple at all.

To find the true eigenmodes, it is necessary to diagonalize \( \vec{r}_A + \vec{r}_N \), not just \( \vec{r}_N \). The eigenmodes and eigenvalues will thus depend upon the relative strength of the electronic and nuclear contributions to \( \vec{r} \). Note, however, that since \( |\frac{m}{2}\rangle_m \) and \( |\frac{m}{2}\rangle_m \) are both Borrmann
modes, the electronic contributions to their eigenvalues are both small. Whatever the relative strength of the electronic and nuclear scattering amplitudes, the resulting eigenmodes will not couple strongly to the electrons, and photoabsorption will be suppressed.

Similar results can be found for the modes $|3\sigma\rangle_6$ and $|3\pi\rangle_6$. The excitation patterns for these modes are shown schematically in Fig. 17. These modes may be thought of as two coherent $|3\sigma\rangle_3$ or $|3\pi\rangle_3$ modes, added $\pi$ out of phase, so that they couple only to $M = \pm 3, \pm 9, \pm 15$, etc. transitions. Thus, E1, M1, and E2 photoabsorption are all suppressed, with the coupling factors being given by

\[
(g_{A,11})_0 = (g_{A,10})_\pi = \frac{1}{3} \left( 1 - f_{D1} - f_{D2} + f_{D3} \right) = \frac{3}{4} \frac{\sin^4 \theta}{\cos \theta} \frac{\langle x^2 \rangle^2}{\lambda^4}
\]

(791)

Fig. 17. Excitation patterns of the $|3\sigma\rangle_6$ and $|3\pi\rangle_6$ Borrmann modes.
\[ (g_{A,11})_\pi = \frac{1}{c} \left( 1 - (c^2 + 2s^2)f_{D1} - (c^2 - 2s^2)f_{D2} + (c^2 - s^2)f_{D3} \right) \]
\[ = \frac{3}{4} \sin^4 \theta \cos \theta \frac{\langle x^2 \rangle}{x^4} \]  
\[ (7911) \]

and

\[ (g_{A,21})_\pi = \frac{1}{c} \left( 1 - (c^2 - s^2)f_{D1} - f_{D2} + (c^2 - s^2)f_{D3} \right) \]
\[ = 3 \sin^2 \theta \frac{\langle x^2 \rangle}{\cos \theta} \]  
\[ (801i) \]

\[ (g_{A,21})_\pi = \frac{1}{c} \left( 1 - (c^4 + 3s^2c^2 + s^4)f_{D1} - (c^4 - 7s^2c^2 - s^4)f_{D2} + (c^4 - 6s^2c^2 + s^4)f_{D3} \right) \]
\[ = 3 \sin^2 \theta \cos \theta \frac{\langle x^2 \rangle}{x^2} \]  
\[ (801ii) \]

Again, the M1 contribution may be neglected. \( g_{A,11} \) and \( g_{A,21} \) for these modes are plotted as a function of cone angle in Fig. 18.

The first nuclear transitions to which these modes couple are E3 and M3. The coupling factors for the case of no Zeemann splitting are

\[ \langle g_{N,31} \rangle |\Delta > = \left( g_{N,30} \right) |3\pi > = \frac{45}{8} \sin^4 \theta \]  
\[ (81i) \]

and

\[ \left( g_{N,30} \right) |\Delta > = \left( g_{N,31} \right) |3\pi > = \frac{45}{8} \sin^4 \theta \cos \theta \]  
\[ (81ii) \]
Fig. 18. Plot of $g_{A,11}$ and $g_{A,21}$ vs. cone angle for the $|3\sigma>_6$ and $|3\pi>_6$ Borrmann modes. The $[\cos \theta]^2$ path length contribution has been factored out for clarity. $\xi = <x^2>/\pi^2$.

These functions are plotted in Fig. 19.

In the presence of Zeemann splitting, these modes will be mixed by the $M = \pm 3$ components of E3 or M3 transitions, exactly as discussed above. For these transitions, $\mathcal{D}_{3,6}(\theta) = \frac{45}{16} \sin^4 \theta$. Again, both the resulting eigenmodes will be Borrmann modes.
Fig. 19. Plot of $g_{N,30}$ and $g_{N,31}$ vs. cone angle for the $|3\sigma>_{6}$ and $|3\pi>_{6}$ Borrmann modes. The $[\cos^2 \theta]$ path length contribution has been factored out for clarity.

Modes Which Are Not Formed on Regular Polygons

If the m-beam mode is centered about an n-fold rotation axis of the plane of reciprocal lattice points which contains its base, with $n < m$, the m beams in a mode will not be distributed uniformly around the surface of the cone (see, for example, the 8 beam modes shown in Fig. 5), and the mode will not possess the full rotation and reflection symmetry implied by the number of beams. Instead, the symmetry of the mode will be that of the axis around which it is formed; i.e. 8, 12, and 16-beam modes formed around a 4-fold axis possess 4-fold rotation symmetry, whereas a 12-beam mode about a
6-fold axis will possess 6-fold rotation symmetry. If there is no Zeeman splitting, the symmetry groups of these modes will be $C_{4v}$ and $C_{6v}$, respectively. If the axis possesses n-fold rotational symmetry the one-dimensional irreducible representations will appear $m/n$ times among the eigenmodes. As will be seen below, if they only appear twice, it is still possible to determine the eigenmodes by symmetry. If they appear more than twice, the eigenmodes will depend upon the exact form of $\tilde{F}$, and it is necessary to diagonalize this matrix within this subspace.

These modes may be thought of as linear combinations of several modes which possess the basic symmetry of the rotation axis. An 8-beam mode, for example, is made up of two 4-beam modes, rotated relative to each other by an angle $\varepsilon$, as is shown schematically in Fig. 20. Considered by itself, each of the n-beam modes which go into making up the m-beam mode are degenerate, that is, they will have the same eigenvalues, $g_A$ and $g_B$. (Herein lies the difference between these modes and those which correspond to 2-D irreducible representations. As we shall see, those subspaces are always spanned by a pair of modes which have the same symmetry properties but which are not degenerate in $g_A$, one being a Borrmann mode and the other an anti-Borrmann mode.)

Consider first the simple case of the subspace of the 8-beam geometry spanned by the two $|4_0\rangle_4$ modes, each of which couples well to M1 oscillators through the $M = 0$ transition, precisely the case which is shown in Fig. 20. For simplicity, denote the two $|4_0\rangle_4$ modes by
Fig. 20. Examples of 8-beam eigenmodes which can be formed about a 4-fold axis. Note that each mode may be described as a linear superposition of two 4-beam modes, in this case two \( |\sigma_+\rangle \) modes, rotated relative to each other by an angle \( \epsilon \).

\( |\sigma\rangle \) and \( |\sigma'\rangle \), respectively. Then by symmetry, regardless of the detailed structure of \( \widetilde{F} \),

\[
\langle \sigma | \widetilde{F} | \sigma \rangle = \langle \sigma' | \widetilde{F} | \sigma' \rangle = \alpha
\]

(821)

and

\[
\langle \sigma | \widetilde{F} | \sigma' \rangle = \langle \sigma' | \widetilde{F} | \sigma \rangle = \beta.
\]

(821i)

The eigenvalues are \( k_{\alpha,\pm}^n = \frac{\lambda_0 n}{\cos \theta} (\alpha \pm \beta) \), while the corresponding eigenmodes are

\[
|\sigma_{\pm}\rangle = \{|\sigma\rangle \pm |\sigma'\rangle\}.
\]

(83)

\( |\sigma_\pm\rangle \) does not couple to M1 transitions, since it contains equal amounts of \( +\sigma \) and \( -\sigma \) polarizations. \( |\sigma_+\rangle \) will couple to M1, however. The coupling factors for this mode are
\[ \delta_{N,10} = 8 \frac{\sin^2 \theta}{\cos \theta} \]  
(841)

and

\[ \delta_{A,11} = 4 \frac{\sin^2 \theta}{\cos \theta} \frac{\langle x^2 \rangle}{x^2}, \]  
(8411)

in agreement with the general results for symmetric \( \sigma \) modes, Eqs. 63 and 59. These coupling factors do not depend upon \( \epsilon \), since \( M = 0 \) transitions have no \( \phi \) dependence.

The eigenmodes in the 2x2 subspace spanned by two antisymmetric \( \sigma \) modes (\( |2\sigma \rangle \)), for simplicity denoted \( |\sigma_+ \rangle \) and \( |\sigma_- \rangle \), may be found in a similar manner. These eigenmodes are shown in Fig. 21. Both of these eigenmodes couple to E2 oscillators through the \( M = \pm 2 \) transitions. These transitions do have a \( \phi \) dependence, so the coupling factors for these modes do depend upon the rotation angle, \( \epsilon \). These coupling factors are given by

Fig. 21. 8-beam eigenmodes which couple to E2 transitions. Note that as the rotation angle \( \epsilon \) is increased past \( \pi/4 \) the two modes transform into each other.
\[
(g_{N,21})_{8\text{-beam}} = [(g_{N,21})_{4\text{-beam}}] \cdot (1 \pm \cos 2\epsilon)
\]

\[
= 4 \frac{\sin^2 \theta}{\cos \theta} \{1 \pm \cos 2\epsilon\}
\tag{85}
\]

and

\[
(g_{A,21})_{8\text{-beam}} = [(g_{A,21})_{4\text{-beam}}] \cdot (1 \pm \cos 2\epsilon)
\]

\[
= 2 \frac{\sin^2 \theta}{\cos \theta} \frac{<x^2>}{x^2} \{1 \pm \cos 2\epsilon\}.
\tag{86}
\]

As can be seen from Fig. 21, the two eigenmodes,

Fig. 22. Plot of \(g_N\) vs. the rotation angle, \(\epsilon\), for coupling E2 transitions to the 8-beam modes shown in Fig. 21. \(g_0 = 4\sin^2 \theta/\cos \theta\) is the coupling factor for a single \(|2\sigma>\) mode. Since the two eigenmodes transform into each other under \(\epsilon + \epsilon + \pi/4\), \(g_N\) is only unique over the range \(0 \leq \epsilon \leq \pi/4\).
\[ |\alpha\rangle = \{ |2\alpha\rangle_u + |2\alpha\rangle_y \} \text{ and } |\alpha\rangle = \{ |2\alpha\rangle_u - |2\alpha\rangle_y \}, \]

transform into each other under \( \epsilon + \epsilon + \pi/4 \), so that only the range \( 0 \leq \epsilon \leq \pi/4 \) need be considered. \( g_N \) for the two eigenmodes is plotted in Fig. 22 as a function of \( \epsilon \) over this range. The coupling is best for small \( \epsilon \), where the mode closely resembles a single \( |2\alpha\rangle_y \) mode. This becomes less and less true as \( \epsilon \) is increased, hence the decrease in \( g_N \).

It is interesting to note that when \( \epsilon \to 0 \), Eq. 85 does not give the value for \( g_N \) appropriate for a single \( |2\alpha\rangle_y \) mode (Eq. 72i), but rather twice this value. This is because Eq. 85 was calculated assuming an 8-beam geometry. This is not valid when \( \epsilon = 0 \), since then there are only four beams. The ambiguity arises in determining when one actually has 8 distinct beams, rather than four. The angular width of each beam is of the order of the Borrann width, \( W_B \sim 10^{-5} \) rad), and so long as \( \epsilon > W_B \) the adjacent beams will be distinct and may be treated as such. In practice this problem does not arise due to the discrete nature of the reciprocal lattice; adjacent beams are always separated by a reciprocal lattice vector, so that \( \epsilon \) can never approach zero.

It is not possible, in general, to calculate the structure of the eigenmodes directly from symmetry for cases where the 1-D irreducible representations appear more than twice. It is often possible to make an educated guess, however. For example, the 12 beam symmetric \( \sigma \) mode \( (|12\alpha\rangle_{12}) \) is an eigenmode both of \( (\tilde{r}_N)_{M_1} \) and \( (\text{Im}(\tilde{r}_A))_{E_1} \). It is not in general an eigenmode of \( \text{Re}(\tilde{r}_A) \), however, due to the presence of the form factors, \( F(\theta) \). Nevertheless, if the nuclear contribution dominates, the true eigenmode will be very nearly \( |12\alpha\rangle_{12} \).
Modes such as these, where the 1-D irreducible representations appear more than twice, will necessarily have a large number of beams (12 or more) and will occur only for very large cone base diameters (see Fig. 5). They will only be open in most crystals if the energy of the nuclear transition is quite large. For example, the first such mode available in an Fe lattice is a 12-beam mode for which the radius of the cone base is 11.0Å⁻¹. This mode would require a resonant energy exceeding 21.6keV to excite. The second such mode (also with 12 beams) occurs at 15.5Å⁻¹, which corresponds to 30.5keV. The third mode, which has 16 beams, lies at 25.0Å⁻¹ and needs an energy greater than 49.2keV to excite. The Mössbauer fraction for transitions at such large energies will be quite small, which will severely limit the chances of achieving lasing, even with the enhancement produced by the coherent interaction of many beams. Such modes are therefore not likely to be of great interest for the lasing problem.

**Borrmann Modes Corresponding to 2-D Representations**

As stated above, all the point groups, Cₘᵥ, possess either two or four 1-dimensional irreducible representations. If m is greater than 2, they also possess one or more 2-dimensional irreducible representations. As will be seen below, modes corresponding to these representations are not of particular interest for lasing, but do have properties of great interest for some single photon problems.
In the case of an \( n \)-beam mode centered about an \( n \)-fold rotation axis, these 2-dimensional irreducible representations will appear \( \frac{2m}{n} \) times among the eigenmodes (see Appendix B). These eigenmodes will always be doubly degenerate, so that they can be divided up into two sets of \( \frac{2m}{n} \) modes each, where all the modes in each set have the same symmetry properties, and each mode in the set has a corresponding mode in the other set with which it is degenerate. Since there are always at least two modes in this subspace which have the same symmetry type, these eigenmodes cannot be determined solely from symmetry. It is always necessary to diagonalize \( \hat{F} \) within one of these \( \frac{2m}{n} \)-dimensional subspaces.

For purposes of illustration, let us consider the 4-beam modes which couple to \( M = \pm 1 \) transitions. In this case, \( m = n = 4 \), and there will thus be two pairs of eigenmodes within this subspace, the two members of each pair having exactly the same symmetry properties. The character table of the 2-dimensional irreducible representation which corresponds to this subspace (see Appendix B) is:

\[
\begin{array}{cccc}
|\pi_y\rangle & |\sigma_x\rangle & |\pi_x\rangle & |\sigma_y\rangle \\
\end{array}
\]

Fig. 23. 4-beam basis states which span the space corresponding to the 2-dimensional irreducible representation of \( C_{4v} \).
where the operations of the group are shown on top; \( E \) denotes the identity, \( C_2 \) a rotation by \( \pi \), \( 2C_4 \) rotations by \( \pm \pi/2 \), and \( 2\sigma_v, \gamma \), the four reflections, through the horizontal, the vertical, and the two diagonals. A set of basis vectors which span this subspace are shown in Fig. 23.

The notation used above for the modes which transform like 1-dimensional irreducible representations becomes somewhat tedious in this case, so these basis vectors will be denoted as \( |\sigma_{x,y}\rangle \) and \( |\pi_{x,y}\rangle \), where \( x \) and \( y \) refer to the axis along which the excited beams lie.

Note that \( |\sigma_{x}\rangle \) and \( |\pi_{y}\rangle \) change sign under reflection through the \( x \)-axis, but do not change sign under reflection about the \( y \)-axis, while the opposite is true of \( |\sigma_{y}\rangle \) and \( |\pi_{x}\rangle \). Thus \( |\sigma_{x}\rangle \) and \( |\pi_{y}\rangle \) correspond to functions of one symmetry type, while \( |\sigma_{y}\rangle \) and \( |\pi_{x}\rangle \) correspond to the other.

The \( E^1 \) electronic contribution to the atomic scattering amplitude well above any electronic absorption frequencies is approximately

\[
\tilde{f}_A = (-Zr_0 F(\theta) + \frac{ig_A}{4\pi\lambda} f_D(\Delta\xi)) \hat{e}_r^* \hat{e}_i

= \mathcal{A}(\theta) \hat{e}_r^* \hat{e}_i, 
\]

(87)

where \( F(\theta) \) is the form factor for \( \lambda^* \cdot A \) scattering and \( f_D \) is the Debye
Waller factor. In the 4-beam case, only three values of $\Delta k$ occur, so there will only be three values of $I$ of interest, denoted $I_0$, $I_1$, and $I_2$, corresponding to forward scattering, scattering to a beam at $\Delta \phi = \pm \pi/2$, and at $\Delta \phi = \pi$, respectively. The matrix corresponding to $f_A$ in this basis is then given by

$$
\begin{pmatrix}
I_0 + (c^2 - s^2)I_2 & -2cI_1 \\
-2cI_1 & I_0 + I_2 \\
0 & 0 & I_0 + (c^2 - s^2)I_2 & -2cI_1 \\
0 & 0 & -2cI_1 & I_0 + I_2
\end{pmatrix}
$$

Taking the determinant of this matrix yields the eigenvalues

$$
k_{z,0}^2 = \frac{\lambda_0^2}{\cos^2 \theta} \left[ I_0 + c^2 I_2 - \frac{1}{2} \left( (I_0 + c^2 I_2)^2 - (I_0^2 + 2c^2 I_0 I_2 + (c^2 - s^2)I_2^2 - 4c^2 I_1^2) \right) \right]^{1/2}.
$$

In general, no special relationship exists between the three form factors, $F(0)$, $F(\theta_1)$, and $F(\theta_2)$, and it is difficult to simplify this equation further. Therefore, in order to illustrate the behavior of the modes, let us take $F(\theta) = f_D = 1$. While this approximation is wholly unjustified physically, it avoids tedious algebra which would only obscure the symmetry properties of the modes, and will not alter the main points of the argument.
In this approximation, \( \tilde{r}_A \) reduces to a constant times the matrix

\[
\begin{pmatrix}
2\alpha^2 & -2\alpha & 0 & 0 \\
-2\alpha & 2 & 0 & 0 \\
0 & 0 & 2\alpha^2 & -2\alpha \\
0 & 0 & -2\alpha & 2 \\
\end{pmatrix}
\]

which has eigenvalues of \( k_{z,-}^I = 0 \) and \( k_{z,+}^I = 2(1 + \cos^2\theta) \). Each 2x2 irreducible subspace is thus spanned by a Borrmann mode (with \( k_z^I = 0 \)).

![Diagram](image)

\[
\begin{align*}
|\pi_y\rangle & = \begin{pmatrix} 0 \\ 0 \end{pmatrix} \\
|\sigma_x\rangle & = \begin{pmatrix} 0 \\ 1 \end{pmatrix} \\
|\pi_x\rangle & = \begin{pmatrix} 1 \\ c \end{pmatrix} \\
|\sigma_y\rangle & = \begin{pmatrix} c \\ -1 \end{pmatrix}
\end{align*}
\]

\[
|\psi_{B1}\rangle = \begin{pmatrix} 0 \\ 0 \end{pmatrix}, \quad |\psi_{AB1}\rangle = \begin{pmatrix} 0 \\ 1 \end{pmatrix}, \quad |\psi_{B2}\rangle = \begin{pmatrix} -c \\ 0 \end{pmatrix}, \quad |\psi_{AB2}\rangle = \begin{pmatrix} 1 \\ 0 \end{pmatrix}
\]

Fig. 24. 4-beam electronic eigenmodes in the subspace spanned by the 2 dimensional irreducible representation of \( C_{4v} \). The components of each vector in the \( |0,\pi_x, y\rangle \) basis (in the approximation that all form factors are neglected) are given below each diagram.
in this approximation) and an anti-Borrmann mode. These eigenmodes are shown schematically in Fig. 24.

These two modes will be the correct eigenmodes off-resonance (within the approximation made above, \( F(\theta) = f_D = 1 \) and neglecting the E2 and higher order contributions). The presence of any resonant nuclear contribution to \( \tilde{F} \) other than E1, however, will invariably mix these two modes, with the result that neither of the eigenmodes at resonance will be a good Borrmann mode. To see this, let us now introduce an additional term into the scattering amplitude corresponding to resonant nuclear scattering by a transition of multipolarity which couples well to the Borrmann mode, for example M1. (These modes couple well to \( M = \pm 1 \) transitions.) In the interest of simplicity, assume that there is no Zeemann splitting. In the basis of \( |\Psi_{B1}\rangle \) and \( |\Psi_{AB1}\rangle \), \( \tilde{F}_{N,10} \) is given by a constant times the matrix

\[
\begin{pmatrix}
\frac{2s^4}{1+c^2} & \frac{4cs^2}{1+c^2} & 0 & 0 \\
\frac{4cs^2}{1+c^2} & \frac{8c^2}{1+c^2} & 0 & 0 \\
0 & 0 & \frac{2s^4}{1+c^2} & \frac{4cs^2}{1+c^2} \\
0 & 0 & \frac{4cs^2}{1+c^2} & \frac{8c^2}{1+c^2}
\end{pmatrix}
\]
The cross term is plotted in Fig. 25 as a function of cone angle. Note that, except at $\theta = 0$ and $\pi/2$, there will be strong mixing between the $E1$ Borrmann and anti-Borrmann eigenmodes. Neither of the eigenmodes at resonance will possess strongly suppressed photoabsorption. In addition, the eigenmodes, and thus $g_A$ and $g_N$, will be strongly dependent upon the relative strength of the electronic and nuclear contributions in $\tilde{F}$.

This is true in general for all the modes which correspond to 2-dimensional irreducible representations. These representations always occur at least twice among the eigenmodes. Off-resonance, half the eigenmodes in this subspace will be Borrmann modes; the rest will be anti-Borrmann modes. The presence of any strong nuclear contribution, other than $E1$, which couples to these modes will invariably mix them, so that none of the eigenmodes at resonance will

![Graph](image-url)

Fig. 25. Plot of the cross term occurring in the expansion of $\tilde{f}_{N1,0}$ in the $|\psi_{B,AB}\rangle$ basis as a function of the cone angle, $\theta$. 
exhibit strongly suppressed photoabsorption. In addition, the structure of these eigenmodes will depend upon the relative strength of the electronic and nuclear contributions to \( \tilde{F} \). As will be seen in Section V, these two characteristics make these modes unsuitable candidates for lasing, and a crystalline graser will instead lase into one of the modes corresponding to 1-dimensional irreducible representations considered above.

Even though this is the case, it is instructive to consider the effect of Zeemann splitting along the symmetry axis in these modes. Again using the 4-beam geometry as an example, the symmetry group reduces from \( C_{4v} \) to \( C_4 \), the cyclic group of 4-fold rotations. In the basis, \( |\sigma_x, y\rangle \) and \( |\pi_x, y\rangle \), the matrices for the M1 scattering amplitude operators in this subspace are given by

\[
\begin{pmatrix}
1 & -c & -i & -ic \\
-c & c^2 & ic & ic^2 \\
i & -ic & 1 & c \\
ic & -ic^2 & c & c^2
\end{pmatrix}
\]
\[
\begin{pmatrix}
1 & -c & i & ic \\
-c & c^2 & -ic & -ic^2 \\
-i & ic & 1 & c \\
-ic & ic^2 & c & c^2
\end{pmatrix}
\]

while \((\vec{r}_{N,10})_0 = 0\), since these modes do not couple to \(M = 0\) transitions. Note that \((\vec{r}_{N,10})_{+1} + (\vec{r}_{N,10})_{-1} = \vec{r}_{N,10}\), provided \(X_{+1} = X_{-1}\), i.e. no Zeemann splitting.

In this basis, as one would expect due to the lowering of the symmetry, \((\vec{r}_{N,10})_{\pm 1}\) are not block diagonal; \(|\sigma_{x,y}\rangle\) and \(|\pi_{x,y}\rangle\) do not possess the symmetry of the new representations, and thus are not a convenient basis for expanding \(\vec{r}\). Basis vectors of the proper symmetry which span this subspace are easily shown to be either equal excitations of all 4 \(\sigma\) polarizations or all 4 \(\pi\) polarizations, with the phase either increasing or decreasing by \(\pi/2\) around the circle, denoted respectively \(|\sigma_{\pm}\rangle\) and \(|\pi_{\pm}\rangle\), so that

\[
|\sigma_{\pm}\rangle = \{|\sigma_{x}\rangle \pm i|\sigma_{y}\rangle\}
\]

and

\[
|\pi_{\pm}\rangle = \{|\pi_{x}\rangle \pm i|\pi_{y}\rangle\}.
\]

There is no longer any distinction between the symmetry of the \(\sigma\) and \(\pi\) polarizations, since reflections are no longer valid symmetry operations. All the irreducible representations are now 1
dimensional, but each of them occurs twice. The non-zero matrix
elements again occur in 2×2 irreducible subspaces, spanned either by
the pair of states |0+⟩ and |π+⟩ or the pair |0−⟩ and |π−⟩. Indeed,
in this basis the M1 scattering amplitudes become:

\[
\begin{pmatrix}
2 & -2ic & 0 & 0 \\
2ic & 2c^2 & 0 & 0 \\
0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 \\
\end{pmatrix}
\]

\[
(\tilde{t}_{N,10})^1_1 = \frac{f_0}{x_+ - i} 
\]

\[
(\tilde{t}_{N,10})^{-1}_1 = \frac{f_0}{x_- - i} 
\]

Note that the "plus" modes only couple to the M = +1 transition, while
the "minus" modes only couple to the M = -1 transition.

The above comments about the unsuitability of such modes for
lasing remain true in the case of Zeeman splitting. Either operator,
\((\tilde{t}_{N,10})^\pm_1\), will mix the electronic Borrmann and anti-Borrmann modes,
so that all the eigenmodes exhibit strong photoabsorption.
Fig. 26. 6-beam electronic Borrmann modes which couple to $M = \pm 2$ transitions. The relative amplitudes of the excitations of each beam depend upon the form factors which occur in $f_A$.\textsuperscript{11}

While modes which correspond to 2-D irreducible representations are not of interest for the lasing problem, they are of great interest for some single photon optics problems. Any pair of modes (one Borrmann and the other non-Borrmann) which do not couple to the Mössbauer transition in the crystal will not be mixed by that transition. The strong suppression of photoabsorption in the Borrmann mode will be retained, even in the presence of the resonant nuclei. Radiation in these modes, along with that in certain of the modes corresponding to 1-dimensional irreducible representations, can be anomalously transmitted at resonance through quite thick slabs of Mössbauer crystals with transitions of the proper multipolarity.

The 6-beam eigenmodes of $\bar{f}_A$ which couple to $M = \pm 2$ transitions are of particular interest in this regard, since the Borrmann modes in this subspace, shown schematically in Fig. 26, exhibit particularly strong suppression of the photoabsorption, with an E1 coupling factor given by
\[ g_{A,11} = \frac{1}{\cos \theta} \left[ 1 - 2f_{D1} + 2f_{D2} - f_{D3} \right] \]

\[ = \frac{3 \sin^6 \theta}{2 \cos \theta} \left( \frac{\langle x^2 \rangle}{x^2} \right)^2. \]  

(90)

These modes, and their non-Borrmann mates, do not couple to \( M = 0, \pm 1 \) transitions, and they will not be mixed by M1 nuclear transitions, i.e. for each mode, \( g_{N,10} = 0 \). **Both electronic and M1 nuclear absorption will be simultaneously suppressed in these Borrmann modes**.

If a crystal of M1 Mössbauer nuclei, say \( ^{57}\text{Fe} \), for example, were illuminated by resonant radiation at a 6-beam point, so that these modes were excited, a portion of the radiation would be anomalously transmitted through quite a thick slab, just as in the ordinary 2-beam Borrmann effect in a nonresonant crystal.
V. NUCLEAR LASING IN A BORMANN MODE

**Conditions for Lasing to Occur**

It has been shown\(^6\) that, due to the anomalous emission effect, the critical population inversion density required to induce lasing at threshold into a 2-beam Bormann mode of a Mössbauer crystal is substantially smaller than that required to induce lasing in an amorphous sample of the same material (or equivalently, off-Bragg in the crystal), so long as the emitters are of multipolarity M\(^1\) or higher. As we shall see below, multi-beam Bormann modes have properties which are even more favorable for lasing than are those of the 2-beam Bormann modes.

While the structure of these multi-beam modes may be quite complex, with many individual beams coherently combining to constitute the mode, they will be automatically fed by emitters of the proper multipolarity substitutionally implanted within the crystal. The imbedded emitters emit spherically, with radiation emerging in all directions, so that there is always radiation emitted into whatever is the correct set of directions necessary to excite the multi-beam mode.

The Schawlow-Townes\(^{48}\) steady-state lasing condition is

\[
K > 0, \quad (91)
\]

where \(K\) is the net gain in intensity per unit length of the sample.
Following Baldwin et al., the net gain per unit length for a crystal containing resonant Mössbauer nuclei is given by

$$K = K_0 - \mu_A$$  \hspace{1cm} (92)

where

$$K_0 = \frac{2\pi\alpha^2 t}{(1+\alpha)(1+\beta)} \frac{\Gamma_1}{\Gamma_1 + \Gamma_2} \frac{\Gamma_1}{\Gamma_1 + \Gamma_2} g_N \left( n_1 - \frac{g_1 n_2}{g_2 n_2} \right)$$  \hspace{1cm} (93)

$$= \sigma^i_N g_N \Delta n.$$  

In these equations, $\sigma^i_N = (g_2/g_1)\sigma_N$ is the nuclear emission cross section at exact resonance, $\Delta n$ is the population inversion density, and $g_N$ is the nuclear coupling factor introduced previously. $n_i$, $\Gamma_i$, and $g_i$, $i = 1,2$, are the number density, natural width, and spin degeneracy of the upper and lower lasing levels, respectively. $\mu_A$ is the usual photoabsorption parameter, given by

$$\mu_A = \sigma_A g_A n_0.$$  \hspace{1cm} (94)

The Schawlow-Townes lasing condition applies only when the population inversion density, $\Delta n$, remains constant in time. In the more practical case, where the inversion density decays with time, the initial inversion must be much larger than that predicted by Eq. 88 for lasing to occur. In fact, under these conditions, the system will quite likely decay by superradiant, rather than stimulated, emission. While the coupling factors, $g_N$ and $g_A$, figure
prominently in these processes as well, the detailed behavior of such systems is not well understood at present. The role which multi-beam Borrmann modes play in such processes will not be addressed in this thesis, but will be the subject of our future research. Only the steady-state case will be considered here.

According to the semiclassical linearized theory of Lamb and co-workers,\textsuperscript{52} a spontaneously emitted $\gamma$-ray will be amplified by a factor $\exp(Kl)$ in traversing a length $l$ through the sample. If the lasing material is in the form of a thin amorphous needle of length $l$ and width $a < l$ with $Kl \gg 1$, it will lase in the "end fire" mode, the radiation emerging with an angular spread $\delta \theta = \text{the larger of } a/l$ (geometry) or $a/l$ (diffraction).\textsuperscript{1} Crystalline needles will lase into Borrmann modes, as will be seen below.

The two coupling factors, $g_N$ and $g_A$, determine which radiation mode of the crystal is most favored for lasing. The favored lasing mode will be that which has the largest $Kl$ product, and will therefore tend to be the mode for which the coupling to the emitters is maximized while, at the same time, the coupling to the electrons is minimized. Such a mode would have the best gain and the lowest photoabsorption. It is possible to find such modes only because Mössbauer transitions exist with multipolarity other than $E1$. Photoabsorption is primarily electric dipole in this energy region, and any mode which has a node in the electric field at the site of the atom will have reduced photoabsorption. Such a mode would not couple to an $E1$ emitter, but will couple well to certain emitters of higher multipolarity.
Effects of Changes of $g_A$ and $g_N$

The effects of changes in the coupling factors, $g_N$ and $g_A$, are shown in Fig. 27. The net gain, $K$, is given by Eqs. 92-94. Setting $K=0$, we obtain the threshold condition on the population inversion density,

$$(\Delta n)_t = (g_A g_A n_0)/(g_N g_N).$$

(95)

Note that the $1/\cos \theta$ path length factor, which occurs in both $g_N$ and $g_A$, cancels out in this expression; at threshold, the improvement in nuclear coupling due to this increased path length is exactly balanced by the resulting increase in photoabsorption.

The threshold population inversion density may be reduced either by decreasing photoabsorption (reducing $g_A$) or by improving the nuclear coupling (increasing $g_N$). A practical graser, however, will have to operate with a gain well above threshold, at a population inversion density $(\Delta n)_p \gg (\Delta n)_t$. Reducing $g_A$ will give a lower threshold, but will not change the slope of the line $K$ vs. $\Delta n$, as is shown in Fig. 27b. $\Delta n_p$, will be reduced by the same amount as the reduction in threshold. Increasing $g_N$, however, both lowers the threshold and increases the slope, as shown in Fig. 27c. $\Delta n_p$ is then reduced not only by the amount of the reduction in threshold, but by an additional amount proportional to the increase in the slope of the gain curve.
Fig. 27. Diagram showing the effects of decreasing $g_A$ or increasing $g_N$ on the gain of a steady state lasing system. The net gain vs. population inversion density curve for some arbitrary mode is plotted in a). The level of gain necessary to make a practical laser, $K_p$, implies the need to exceed a particular level of inversion density, $\Delta N_p$. The same curve with reduced photoabsorption ($g_A \rightarrow g_A' < g_A$) is shown in b). Note that the slope of the curve remains unchanged. Both the threshold density, $\Delta n_t$ and $\Delta n_p$ are reduced by the same amount. The same curve with increased coupling to the nuclei ($g_N + g_N' > g_N$) is shown in c). Note that the slope is now increased, resulting in a much lower value of $\Delta n_p$.

Of course $g_N$ and $g_A$ may not be varied independently since they are determined by the symmetry properties of the eigenmodes, the relative strength of the electronic and nuclear contributions to the scattering amplitude, and the cone angle. Nevertheless, any increase in $g_N$ between two modes will, in general, be much more significant.
than will a similar decrease in $g_A$. The favored lasing mode will thus tend to be that which has the largest value of $g_N$.

**Lasing Mode Selection**

The mode into which a steady-state system will lase, out of all the possible modes available, will be that which has the largest gain-length product, $K\lambda$, where $\lambda$ is the length of the sample along the mode axis. The $\gamma$-ray laser, as it is generally envisioned, would consist of a needle-like sample, sufficiently long to ensure that $K\lambda$ is much greater for the modes along that axis than for any off-axis modes. This would enhance the "end-fire" modes (those aligned along the sample axis) and cause the radiation to emerge from the ends of the sample.

If the sample is amorphous, there will only be one such mode, consisting of a single plane wave channel aligned directly along the needle axis. For this mode, $g_N = 1$. A crystalline sample will have many such modes, at various cone angles, each with a different value of $g_N$. Which mode will lase then depends upon the the multipolarity of the emitter, the number of beams in each mode, as well as the cone angle.

The Borrmann and anti-Borrmann modes corresponding to 2-dimensional irreducible representations (which always occur at least twice among the eigenmodes) are always strongly mixed by any nuclear transition of multipolarity other than $E1$ to which they couple. None of the resulting eigenmodes will be good Borrmann modes and the losses
due to photoabsorption in these modes will be quite high. These modes are, therefore, unlikely to be of interest for the lasing problem.

Borrmann modes which correspond to 1-D irreducible representations, on the other hand, are either not mixed by the presence of resonant nuclei, or if they are mixed (as in the case of the \( | \frac{m}{2} \rangle \) and \( | \frac{m}{2} \rangle \) modes in the presence of Zeeman splitting) remain good Borrmann modes. It is these modes, therefore, which will be favored for lasing.

It seems likely that the lasing transition of any practical graser will be either M1 or E2. Transitions of higher multipolarity have much longer lifetimes, and consequently, much narrower energy widths. Any amount of inhomogeneous broadening in such a sample would shift the nuclei off resonance by an amount sufficient to deter lasing. The discussion in the remainder of this section will therefore concentrate on modes which couple well to M1 and E2 emitters. Similar remarks will hold for modes which couple well to higher multipoles, should such a case prove practical.

Both of these emitters couple well to 2-beam Borrmann modes, with

\[
S_N = 2 \frac{\sin^2 \theta}{\cos \theta}.
\]  

(96)

in each case. If only 2-beam modes were available, as would likely be the case if the needle axis was not aligned along some high symmetry axis of the crystal, the sample would lase into the mode with the largest cone angle.
If the crystal is grown along a high symmetry axis several multi-beam modes also become available. The multi-beam Borrmann modes which couple best to M1 emitters are the symmetric $\omega$ modes, denoted $|\omega\rangle_m$. For these modes,

$$g_N = m \frac{\sin^2 \theta}{\cos \theta}. \quad (97)$$

The mode which is favored will be that at the largest angle with the most number of beams.

As an example, consider the case of $^{57}$Fe, which has a 14.4 keV M1 Mössbauer transition. Fe is a body-centered cubic crystal with a

![Diagram of reciprocal lattice](image)

Fig. 28. Cut through the reciprocal lattice of an $^{57}$Fe crystal, perpendicular to the (100) axis. The 8-beam mode shown has $g_N = 23.0$, and is the best case for this crystal.
lattice constant $a = 2.87\text{Å}$; its reciprocal lattice is therefore face-centered cubic.

The best case mode about a high symmetry axis turns out to lie along $(100)$. A cut through the reciprocal lattice perpendicular to $(100)$ is shown in Fig. 28. The outermost circle has a radius in reciprocal space of $1/x = 7.30\text{Å}^{-1}$. The lasing mode of an $^{57}\text{Fe}$ crystal grown along the $(100)$ axis will be the 8-beam mode just inside the outer circle. The circle which contains these eight reciprocal lattice points has a radius of $6.93\text{Å}^{-1}$, so that this mode has a cone angle of $\theta = 71.7^\circ$.

![Graph](image)

**Fig. 29.** Plot of $K$ vs. $\Delta n$ for both an off-Bragg mode and the best case 8-beam mode in $^{57}\text{Fe}$, as well as for a corresponding (same $g$) 2-beam mode.
\( \frac{\langle x^2 \rangle}{\langle x \rangle^2} = 0.2 \) for Fe at room temperature. Calculating \( g_N \) and \( g_A \) for this mode using this value yields \( g_N = 23.0 \) and \( g_A = 2.57 \). Note that this mode actually has larger photoabsorption than an off-Bragg mode (for which \( g_A = 1 \)), even though it is a Borrmann mode. This is largely due to the \([\cos \theta]^{-1}\) path length contribution. (Calculating the photoabsorption per unit path length, the relevant parameter is \( g_A \cos \theta = 0.81 \). Thus, photoabsorption is still suppressed.) The improved coupling to the nuclei more than outweighs the increased photoabsorption, however, so that this mode will be the favored lasing mode.

This can be easily seen in Fig. 29, where \( K \) vs. \( \Delta n \) lines are plotted for both this 8-beam mode and an off-Bragg mode. In addition, the line for a corresponding 2-beam (same cone angle) mode is included for comparison. The threshold in the 8-beam mode is reduced by a factor of 10 relative to that of the off-Bragg mode, while the slope of the gain curve is 23 times greater. Thus the 8-beam mode will reach a usable gain at much lower inversion densities than will the off-Bragg mode.

The best 2-beam case for \( ^{57}\text{Fe} \) is the mode corresponding to the (622) reflection, with a cone angle of 84°. Due to this large cone angle, this mode has \( g_N = 20 \) and \( g_A = 3.8 \). Thus, by choosing the axis of the needle appropriately, the crystallinity of the sample allows for a certain amount of freedom in the choice of the lasing mode. In this case, the mode with the best gain may be obtained by growing the crystal along the (100) axis. If, on the other hand, it is desirable to limit the number of beams emerging from the crystal, the axis of
the needle could be aligned along some low symmetry axis of the crystal, perpendicular to (622), which would, instead, enhance the 2-beam mode.

Rotating the cone axis around the (622) reflection, it is possible to pick up two additional reflections, for example the (442) and the (2,-2,0). This results in a 4-beam mode whose cone axis lies along (1,1,-4), with the same cone angle as the 2-beam mode, 84°. While (1,1,-4) is not a symmetry axis of the crystal, it is a 2-fold axis of the plane of reciprocal lattice vectors which contain the (442) and (2,-2,0) reflections. The symmetry group of the mode is thus C2v, and, as shown in Sect. IV, |4o>4 will be one of the eigenmodes. This mode has the largest value of gN available in 57Fe, gN = 40. Cutting a crystalline needle along this low-symmetry axis with sufficient precision to favor this mode may be difficult, however.

M1 transitions have particularly good properties for lasing, since they couple well to the symmetric o mode regardless of the number of beams. There is considerably less freedom in the choice of the lasing mode for the case of E2 transitions, since these transitions only couple to modes aligned along 4-fold axes. They couple well to the 4-beam antisymmetric o modes (|2o>4), as well as to the modes spanned by these antisymmetric modes in the 8, 12, and 16 beam cases, particularly if the reciprocal lattice points are closely grouped into four sets (for example, ε small in Fig. 21). As an example, if the antisymmetric o mode of the 8-beam geometry
considered above (θ = 71.7°) were coupled to an E2 emitter, the resulting coupling factors would be g_N = 18.4 and g_A = 2.05.

Angular Divergence of Multi-Beam Borrmann Modes

An additional advantage that the multi-beam Borrmann modes of a Mössbauer crystal have over the single plane wave modes of an amorphous sample is the natural collimation of the emerging radiation obtained in these modes. The angular divergence of the wave field emerging from an amorphous needle, or in an off-Bragg mode of a crystalline needle, is determined either by the physical collimation introduced by the size of the needle or by the diffraction limit, whichever is greater. Such a needle of length l and diameter d cannot produce a beam collimated to better than Δθ = d/l. Making the diameter too small, however, will introduce a large angular divergence due to diffraction, with Δθ = λ_0/d. The best possible case occurs when these two effects are comparable. This requirement therefore serves to limit the size of the needle which may be used. In particular, for a given length, the diameter must be d = \sqrt{λ_0}.

The angular divergence of radiation in Borrmann modes, however, is not determined by the size and shape of the sample, but rather is limited by the natural collimation which takes place during Bragg diffraction, as is shown in Fig. 30. In a 2-beam Borrmann mode, Bragg diffraction limits the angular divergence in the direction perpendicular to the reflecting planes (along θ) to Δθ = \Omega_B, the Borrmann width, typically 10^{-5} rad or less, regardless of the
Fig. 30. Angular divergence properties of the eigenmodes of amorphous, as well as 2-beam and multi-beam Borrmann lasers. While lasing into a 6-beam mode is shown, other multi-beam modes are similar.

thickness of the crystal in that direction (so long as the crystal is sufficiently thick to form the Borrmann modes, i.e. a few primary extinction lengths, typically 10μm). The angular divergence in the
other direction (along $\phi$), however, is still determined by either geometry or the diffraction limit.

The radiation from the multi-beam (more than 2) Borrman modes emerges in well defined beams which occur at the intersections of the various 2-beam modes which make up the multi-beam mode. The angular divergence is thus limited in both directions to $\Delta \theta = W_B$, regardless of the size and shape of the lasing sample.
VI. APPLICATIONS TO SINGLE-PHOTON OPTICS

The theory of the coupling of multi-beam Borrmann modes to resonant nuclear emitters also has applications to problems in single-photon optics. These application include:

1) The Anomalous Emission Effect
2) Anomalous Transmission in the Presence of Resonant Absorbers

The Anomalous Emission Effect

The electronic Borrmann effect (anomalous transmission of x-rays through a crystal) has been thoroughly investigated, both for the 2-beam case and for the multi-beam cases considered here. The expressions for $g_A$ given in this thesis for those cases in which the eigenmodes are determined solely by symmetry are thus well understood.

New features arise, however, with the inclusion of resonant nuclear transitions, both in the presence of the new coupling factors, $g_N$, and in the strong mixing which can occur among modes which correspond to the same irreducible representation. Nuclear transitions of multipolarity higher than E1 will couple strongly to certain Borrmann modes of the crystal. While the anomalous emission of radiation into the Borrmann modes by substitutionally implanted Mössbauer nuclei has not as yet been observed, it is hoped that the recent interest in γ-ray lasers will stimulate research in this area and lead to a verification of these results.
There are several ways in which such experiments could be done, either by substitutionally implanting excited nuclei within the crystal and looking directly for anomalous emission, or, using the reciprocity theorem, by observing the equivalent absorption problem. The simplest approach would be to take two thick crystals (thick relative to an off-Bragg absorption length), and substitutionally implant parent isomers of two different Mössbauer nuclei into a small region of the upper surface the crystals; one crystal being implanted with nuclei which decay to an E1 emitter, and the other with nuclei which decay to an emitter of higher multipolarity, say M1, as is shown in Fig. 31. The energies of the Mössbauer transitions would have to be high enough to allow Bragg reflections in the crystal. The experiment could then be performed by simply setting each crystal on a

![Diagram](image)

**Fig. 31.** A simple experiment to verify the anomalous emission effect.
piece of photographic film in a dark room and waiting until the films are exposed.

In this geometry, there is a thick region of crystal between the resonant emitters and the film, and only radiation emitted into the Borrmann modes should get through. The M1 emitters will couple strongly to these modes and radiation from them should register as spots (in the case of multi-beam modes) or lines (in the case of 2-beam modes) on the film at the proper positions for the Bragg peaks. (The reason the doped area must be small is that the film will pick up an image of the doped region, and if that area is large there will be strong smearing of the Bragg peaks on the film.)

Recoilless emission from the E1 emitters, on the other hand, will not feed the Borrmann mode. The radiation reaching the film in this case will thus be down by at least a factor \(1 - f\) compared to the radiation from the M1 emitters, resulting in spots and lines on the film of much lower intensity.

Alternatively, the same experiment could be performed using only one crystal doped with a parent whose decay scheme contains two Mössbauer transitions, one E1 and one of higher multipolarity, at different energies. The radiation from the two transitions could be distinguished by the different Bragg angles of the two reflections. (It would be particularly important to have a point-like source in this case.) The patterns corresponding to the higher multipole emission will again be of much higher intensity than those corresponding to the E1 transition.
While this film experiment would serve to demonstrate the essential features of the anomalous emission effect, it may be difficult to obtain a quantitative measure of $g_H$ using this method. To do so, it may be more convenient to perform an absorption experiment using singles counting detectors, rather than photographic film. Such an experiment is illustrated schematically in Fig. 32. Instead of doping the crystal with parent nuclei at the top, nuclei with resonant Mössbauer transitions are substitutionally implanted into the bottom layers of a thick crystal (thick relative to an off-Bragg absorption length). In this case there would be no need to restrict the physical area of the doped region.

![Diagram](image)

Fig. 32. An absorption experiment to measure $g_H$ in a Bormann mode. Only the lower layers of the crystal are doped with absorbers. By the time the radiation reaches this layer, all modes other than Bormann modes have been absorbed out by the electrons.
The crystal is then illuminated by a divergent source of resonant radiation from the top, exciting either the simple 2-beam modes or the more complicated multi-beam geometries. (Using a divergent source eliminates the need for careful alignment and will greatly facilitate exciting multi-beam modes.) If only one beam of a multi-beam geometry is fed, several eigenmodes will be excited simultaneously. The incoming beam may be expanded in terms of these eigenmodes to determine how much each eigenmode is excited as

$$|\hat{R}_1, \hat{e}_1\rangle = \sum_{k_2^*} |P_{k_2^*} \rangle \langle P_{k_2^*} |\hat{R}_1, \hat{e}_1\rangle,$$

(98)

where $|P_{k_2^*}\rangle$ are the eigenmodes for this geometry. Since the crystal is thick, the radiation in the non-Borrmann modes would be absorbed before it reaches the doped layer, so that the resonant atoms would only be exposed to radiation in the Borrmann modes. The transmitted radiation would be observed by counters placed below the crystal. The alignment of the crystal could be checked by feeding off-resonance, where any radiation which excites a Borrmann mode would be transmitted. As the incident radiation passes through resonance, one should observe a pronounced reduction in the intensity of the transmitted beams, corresponding to strong absorption of radiation in some of the Borrmann modes by the implanted resonant nuclei, provided the resonance has multipolarity $M1$ or higher. If the implanted nuclei has an $E1$ transition, no increase in absorption should be observed. A quantitative determination of $g_N$ may then be made by comparing the
transmitted intensity off resonance to that on resonance, knowing the density of implanted nuclei.

**Anomalous Transmission in the Presence of Resonant Absorbers**

One cannot have anomalous transmission at resonance in a 2-beam mode through a crystal containing substitutionally implanted resonant nuclei if the resonant transition is other than E1. If the transition is E1, the nuclei do not couple to the Borrmann mode, so that mode is transmitted. If the transition has any other multipolarity, however, the nuclei will couple strongly to the Borrmann mode. There will be no anomalous transmission in this case, since the Borrmann mode will be strongly absorbed by the nuclei, while the anti-Borrmann mode will be strongly absorbed by the electrons.

This is not true, however, for the multi-beam geometries. In each such case there exist Borrmann modes which do not couple to particular transitions with multipolarity higher than E1; the symmetric \( \sigma \) modes (\( |m\sigma>_m \)), for example, do not couple to any electric multipole less than E1.

In the 4 and 6-beam cases, antisymmetric \( \sigma \) and \( \pi \) modes occur (\( |\frac{m}{2}\sigma>_m \) and \( |\frac{m}{2}\pi>_m \)) which do not couple to any transition whose multipole order is less than \( m/2 \) (\( |2\pi>_m \) does not couple to M1, for example). Finally, there are 6-beam modes, corresponding to two-dimensional irreducible representations, which couple to \( M = \pm 2 \) transitions, and thus do not couple to M1. If such modes could be excited at resonance in a crystal containing M1 Mössbauer nuclei,
Fig. 33. An experiment to demonstrate anomalous transmission of multi-beam Borrmann modes through a crystal containing resonant nuclei. On resonance, only the multi-beam Borrmann modes will be transmitted.

these modes would be anomalously transmitted, just as in the ordinary electronic Borrmann case.

To do an experiment of this kind would require a crystal of resonant nuclei which was thick relative to an anti-Borrmann absorption length, ensuring nearly complete absorption of the anti-Borrmann modes by the electrons. If such a crystal were illuminated at resonance by a divergent source of radiation, as shown in Fig. 33, any radiation which excited 2-beam modes would be absorbed within the crystal, either by the electrons in the case of the anti-Borrmann modes, or by the nuclei in the case of the Borrmann modes. In multi-beam points, however, those Borrmann modes which did not couple to the nuclei would be transmitted with minimal absorption.
Fig. 34. 4-beam Borrmann modes. Only the first three would couple to a σ polarized beam incident along $k_i$. All but the first would couple to an M1 transition of a nucleus at a lattice site.

To illustrate this, consider a particular set of 4-beam modes in a cubic crystal containing M1 resonant nuclei, for example $^{57}$Fe. (In practice, $^{57}$Fe may not not a good choice since its isotopic abundance is low; making a sufficiently thick crystal out of it could prove prohibitively expensive.) There are four Borrmann modes in this geometry, three of which would be excited by a particular beam with σ polarization incident upon the crystal at a four-beam point, as illustrated in Fig. 34. Off resonance, all three will be transmitted, with the radiation emerging in four spots below the crystal. Excitation of 2-beam Borrmann modes near the 4-beam point will also be observed. As the beam is passed through resonance, this 2-beam
excitation will be absorbed by the nuclei in the crystal, and will no longer be transmitted. In addition, two of the 4-beam Borrmann modes couple strongly to M1 transitions, so that on resonance only the $|2\sigma\rangle_4$ mode would be transmitted. There will be a sharp drop in the intensity at the detectors due to the absorption of the other 4-beam modes.

A similar experiment could be done at a 6-beam point. In this case, there are seven Borrmann modes, six of which could be excited by a single $\sigma$ polarized beam incident upon the crystal surface. Three of these modes would strongly couple to M1 emitters at resonance, but the other three would be anomalously transmitted. Experiments of this type should clearly demonstrate the multipolar characteristics of the various eigenmodes of these multi-beam cases.
VII. CONCLUSION

The results of our analysis are that the effects of crystallinity are of crucial importance regarding Y-ray lasers, and could well prove decisive in determining whether or not grasing can or cannot be achieved. Certain multi-beam Bormann eigenmodes of perfect crystals, in which several plane waves are excited coherently, have lower photoabsorption and couple much more strongly to resonant nuclear transitions of multipolarity M1 or higher than do the eigenmodes off-Bragg or in an amorphous sample. These multi-beam modes offer the possibility of achieving lasing with orders of magnitude less population inversion density than would be required in the case of an amorphous sample.

The coupling factors (gN's) between these multi-beam modes and the nuclei are always proportional to m, the number of beams. Furthermore, each multi-beam mode has a definite multipole character, in that it will only couple to certain M components of nuclear transitions. Those that couple to the M = 0 components of magnetic transitions (\(|m_o>\_m\) modes) exhibit the maximum possible nuclear coupling (for a given cone angle), regardless of the how the beams which make up the mode are arranged in space. M1 nuclear transitions are thus particularly attractive as candidates for lasing.

The beams which make up a multi-beam mode are not oriented along the sample axis, but rather at an angle \(\theta\) relative to it, so that the path length of the radiation is increased by a factor \(1/cos\theta\). gN is thus proportional to this factor, and, since the number of beams
available also increases with $\theta$, the modes favored for lasing will be
those with many beams at large cone angles.

While the structure of these multi-beam modes may be quite
complicated, with many individual beams coherently combining to constitute
the mode, they will be automatically fed by emitters of the proper
multipolarity substitutionally implanted within the crystal. The
imbedded emitters emit spherically, with radiation emerging in all
directions, so that there is always radiation emitted into whatever is
the correct set of directions necessary to excite the multi-beam mode.

The multi-beam eigenmodes also possess a high degree of natural
collimation, with the radiation emerging from the crystal in a number
of well defined beams each with angular divergence of the order of the
Borrmann width ($\sim 10^{-5}$ rad). The number of beams in the lasing mode
may be selected to some extent by choosing the crystal axis along
which the symmetry axis of the lasing sample is aligned.

All of these advantages are obtained automatically as soon as one
goes to a crystalline sample geometry. If a sufficient population
inversion can be achieved without destroying the properties of the
crystal necessary for the Borrmann and Mössbauer effects, the crystal
should emit into the mode with the largest gain-length product, which,
if the crystal axis is chosen appropriately, will one of the Borrmann
modes discussed above.

Multi-beam Borrmann modes are also of interest for several
single-photon optics problems. If only a small number of resonant
nuclei are implanted within the crystal, so that nuclear absorption
may be neglected, radiation emitted into the Borrmann modes by the
nuclei will emerge after passing through sections of the crystal much thicker than an off-Bragg absorption length. This "anomalous emission" effect can occur in both 2-beam and multi-beam modes, but because of the greatly enhanced nuclear coupling possible in the latter case, the multi-beam modes may show a much stronger effect.

In addition, the multi-beam modes offer the possibility of anomalous transmission at resonance through a crystal containing a large number density of resonant nuclei. This phenomenon can only occur in the 2-beam case if the nuclear transition is E1; if the emitter has any other multipolarity, the Borrmann mode will be strongly absorbed the nuclei, while the non-Borrmann modes will be strongly absorbed by the electrons, with the result that no significant radiation is transmitted. Multi-beam Borrmann modes exist, however, that do not couple M1 or E2 transitions, and radiation in such modes can be anomalously transmitted at resonance in such a case.

If a γ-ray laser can be produced, it will be an important new source of coherent radiation in the Mössbauer region and could prove quite useful as a laboratory tool. There are, however, a great many obstacles to be overcome before such lasing can be achieved. The optics of multi-beam modes of crystals containing resonant Mössbauer nuclei remains an interesting problem, however, that is worth pursuing for its own sake, regardless of whether or not the theory has any immediate practical applications.
APPENDIX A - Vector Spherical Harmonics

The notation used for the vector spherical harmonics, \( |\vec{Y}_{L,M}\rangle\), in this thesis is that of Akhiezer and Berestetskii, *Quantum Electrodynamics*.\(^{53}\) They are defined in spherical coordinates \((r, \theta, \phi)\) by the relations

\[
\langle \hat{r} | \vec{Y}_{j,m} \rangle \cdot \hat{e} = +i \langle \hat{r} | \vec{Y}_{j,m} \rangle \cdot \hat{\phi} = \frac{1}{\sqrt{j(j+1)}} \frac{3Y_{jm}}{\sin \theta} \tag{A1}
\]

\[
\langle \hat{r} | \vec{Y}_{j,m} \rangle \cdot \hat{\phi} = -i \langle \hat{r} | \vec{Y}_{j,m} \rangle \cdot \hat{\theta} = \frac{1}{\sqrt{j(j+1)}} \frac{1}{\sin \theta} \frac{3Y_{jm}}{\partial \theta} \tag{A2}
\]

\[
\langle \hat{r} | \vec{Y}_{j,m} \rangle \cdot \hat{\theta} = \langle \hat{r} | \vec{Y}_{j,m} \rangle \cdot \hat{\phi} = 0, \tag{A3}
\]

where \(Y_{j,m}\) is the ordinary scalar spherical harmonic.

The following is a tabulation of \(\vec{Y}_{L,M}(\hat{k}) = \langle \hat{r} | \vec{Y}_{L,M} \rangle\) for the first few multipoles, up to \(M3\).

For \(E1\):

\[
\vec{Y}_{1,0}(\hat{k}) = -\left(\frac{3}{8 \pi}\right)^{1/2} \sin \theta \tag{A4i}
\]

\[
\vec{Y}_{1,\pm1}(\hat{k}) = \left(\frac{3}{16 \pi}\right)^{1/2} \pm i \phi [\pm \cos \theta - i \phi] \tag{A4ii}
\]

120
For M1:

\[ \mathcal{F}_{1,0}(k) = i(\frac{3}{8\pi})^{1/2} \sin\hat{\phi} \quad (A5i) \]

\[ \mathcal{F}_{1,\pm 1}(k) = (\frac{3}{16\pi})^{1/2} e^{\pm i\phi} [\hat{\theta} \pm i\cos\hat{\phi}] \quad (A5i1) \]

For E2:

\[ \mathcal{F}_{2,0}(k) = -(\frac{15}{32\pi})^{1/2} \sin 2\hat{\theta} \quad (A6i) \]

\[ \mathcal{F}_{2,\pm 1}(k) = (\frac{5}{16\pi})^{1/2} e^{\pm i\phi} [i\cos 2\hat{\theta} - i\cos\hat{\phi}] \quad (A6i1) \]

\[ \mathcal{F}_{2,\pm 2}(k) = (\frac{5}{16\pi})^{1/2} e^{\pm 2i\phi} [\sin\cos\hat{\theta} \pm i\sin\hat{\theta}] \quad (A6i11) \]

For M2:

\[ \mathcal{F}_{2,0}(k) = i(\frac{15}{32\pi})^{1/2} \sin 2\hat{\phi} \quad (A7i) \]

\[ \mathcal{F}_{2,\pm 1}(k) = (\frac{5}{16\pi})^{1/2} e^{\pm i\phi} [\cos\hat{\theta} \pm i\cos\hat{\phi}] \quad (A7i1) \]

\[ \mathcal{F}_{2,\pm 2}(k) = (\frac{5}{16\pi})^{1/2} e^{\pm 2i\phi} [\mp \sin\hat{\theta} \pm i\sin\cos\hat{\phi}] \quad (A7i11) \]
For E3:

\[
\psi_{3,0}(k) = -\left(\frac{21}{64\pi}\right)^{1/2} \sin\theta \left(5\cos^2\theta - 1\right) \hat{\phi}
\]  
(A8i)

\[
\psi_{3,\pm1}(k) = \left(\frac{7}{125\pi}\right)^{1/2} e^{\pm i\phi} \left[\hat{r}(15\cos^3\theta - 11\sin\theta) \hat{\theta} - i(5\cos^2\theta - 1) \hat{\phi}\right]
\]  
(A8ii)

\[
\psi_{3,\pm2}(k) = \left(\frac{35}{125\pi}\right)^{1/2} e^{\pm 2i\phi} \left[\hat{r}(2\sin\theta - 3\sin^3\theta) \hat{\theta} \pm 2i\sin\theta \cos\theta \hat{\phi}\right]
\]  
(A8iii)

\[
\psi_{3,\pm3}(k) = \left(\frac{105}{125\pi}\right)^{1/2} e^{\pm 3i\phi} \left[\sin^2\theta (\hat{r} \cos\theta - i\hat{\phi})\right]
\]  
(A8iv)

For M3:

\[
\psi_{0,0}(k) = i\left(\frac{21}{64\pi}\right)^{1/2} \sin\theta \left(5\cos^2\theta - 1\right) \hat{\phi}
\]  
(A9i)

\[
\psi_{0,\pm1}(k) = \left(\frac{7}{125\pi}\right)^{1/2} e^{\pm i\phi} \left[\hat{r}(5\cos^3\theta - 1) \hat{\theta} \pm i(15\cos^3\theta - 11\sin\theta) \hat{\phi}\right]
\]  
(A9ii)

\[
\psi_{0,\pm2}(k) = \left(\frac{35}{125\pi}\right)^{1/2} e^{\pm 2i\phi} \left[\hat{r}2\sin\theta \cos\theta \hat{\theta} - i(2\sin\theta - 3\sin^3\theta) \hat{\phi}\right]
\]  
(A9iii)

\[
\psi_{0,\pm3}(k) = \left(\frac{105}{125\pi}\right)^{1/2} e^{\pm 3i\phi} \left[\sin^2\theta (\hat{r} \pm i\cos\theta) \hat{\phi}\right]
\]  
(A9iv)
APPENDIX B - Group Theory

A problem which occurs quite often in physics is that of determining the eigenvalues and eigenmodes of some quantum mechanical operator. If the system under consideration possesses certain symmetries, this task may be greatly facilitated by group theoretical considerations, which will allow one to restrict the choice of possible eigenmodes, and in some cases, determine those eigenmodes directly, with very little effort. While this appendix does not contain a detailed and rigorous derivation of the theory of group representations, those results from that theory which have particular application to the eigenvalue problem are discussed and applied to the particular problem of interest here. Those readers who desire a more in-depth derivation of group theory should consult Refs. 54 and 55.

Consider an invariant subspace, \( S \), of an operator \( \vec{F} \) which is spanned by the basis \( \{ |\xi_i> \} \), a set of \( \lambda \) linearly independent state vectors. By invariant subspace, I mean that \( \vec{F} \) acting on any vector in the subspace always gives another vector in the subspace, i.e.

\[
\vec{F} |\xi_i> = \sum_{j=1}^{\lambda} a_j |\xi_j>, \forall i. \tag{B1}
\]

We wish to find the eigenmodes and eigenvalues of \( \vec{F} \) within \( S \).

Let \( G' = \{ R_j \} \) denote the set of symmetry operators (rotations, reflections, inversions, etc.) which commute with \( \vec{F} \). In our case, these will be the rotations and reflections which leave the plane of reciprocal lattice which contains the base of a particular \( m \)-beam mode

123
unchanged. If \(|\psi\rangle\) is an eigenmode of \(\tilde{F}\), then \(R_j|\psi\rangle\) will also be an eigenmode of \(\tilde{F}\), with the same eigenvalue. These operators are thus said to leave \(\tilde{F}\) invariant.

This set of operators, \(G'\), forms a group; that is, the set possesses the following four properties:

1) The set is closed; the successive application of any two symmetry operators in \(G'\) is equivalent to another operator in \(G'\).

2) The operations are associative; that is \((R_iR_j)R_k = R_i(R_jR_k)\).

3) The set contains the identity element, usually denoted \(E\), for which \(ER_j = R_jE = R_j\), for every \(R_j\).

4) The inverse of each operator is also a member of the group.

The properties of this group may be fully specified by giving the group multiplication table, which lists the result of all ordered pairs of group operations.

The operators, \(R_j\), in \(G'\) act upon the reciprocal lattice. This group is isomorphic with the group \(G = \{A_j\}\), where \(A_j\) operate on state functions within the crystal, so that, if \(f(\tilde{r}) = \langle \tilde{F}|f\rangle\), then \(A_jf(\tilde{r}) = f(R_j^{-1}\tilde{r}^*\). The operators, \(A_j\), also commute with \(\tilde{F}\). For the case of interest here, \(|f\rangle\) is the state of the radiation field within the crystal, \(\tilde{F}\) is the planar scattering amplitude operator at exact Bragg, while the \(A_j\)’s rotate (reflect) \(|f\rangle\) relative to the axes of the reciprocal lattice.
A particular choice of basis vectors in $S$ defines a representation of the group, denoted $\Gamma(G) = \{\Gamma(A_j)\}$, which consists of a set of $\lambda \times \lambda$ unitary matrices, one for each $A_j$, such that

$$A_j |\xi_i\rangle = \sum_{k=1}^{\lambda} \Gamma(A_j)_{ki} |\xi_k\rangle.$$  \hspace{1cm} (B2)

$\Gamma(G)$ obeys the matrix multiplication table for the group.

Since the set of basis vectors, $\{|\xi_i\rangle\}$, are chosen arbitrarily, $\Gamma(G)$ will in general be reducible. By an alternate choice of basis vectors, the subspace $S$ may be broken down into a series of smaller subspaces, each of which is also invariant under $\tilde{F}$. This choice of basis vectors will define another representation of the group, $\Gamma'(G)$, related to $\Gamma(G)$ by a similarity transformation. The two representations are thus equivalent. Each matrix $\Gamma'(A_j)$ in $\Gamma'(G)$ will have the same form, consisting of one $\lambda \mu \times \lambda \mu$ block on the diagonal for each $\lambda \mu$-dimensional invariant subspace in $S$, with all other elements zero.

The set of $\lambda \mu \times \lambda \mu$ matrices from any given block also forms a representation of the group. If such a block cannot be further reduced in the above manner, this set of $\lambda \mu$-dimensional matrices is said to form an irreducible representation. The number and dimensionality of the irreducible representations for a given group are completely determined by that group's multiplication table. Such representations may be uniquely defined up to a similarity transformation.
Any representation of the group may be built up out of irreducible representations as

\[ \Gamma = \sum_{\nu} a_{\nu} \Gamma^{\nu}, \]  

(B3)

where \( a_{\nu} \) denotes the number of times that the \( \nu \)th irreducible representation, \( \Gamma^{\nu} \), appears in \( \Gamma \). (Note that Eq. B3 does not imply matrix addition, but is meant to denote the construction of a set of matrices of dimension \( \sum_{\nu} a_{\nu} \lambda_{\nu} \) in which the \( \nu \)th irreducible representation occurs \( a_{\nu} \) times along the diagonal.) In the new basis, \( \{|\phi^{\nu}_{i}\rangle\} \), in which \( \Gamma \) is block diagonal,

\[ A_{j} |\phi^{\nu}_{i}\rangle = \sum_{k=1}^{\lambda_{\nu}} \Gamma^{\nu}(A_{j})_{ki} |\phi^{\nu}_{k}\rangle, \]  

(B4)

where now \( 1 \leq i \leq \lambda_{\nu} \).

The character of a representation is defined as the set of traces of the matrices of that representation, denoted

\[ \chi^{\nu}(A_{j}) = \text{Tr}(\Gamma^{\nu}(A_{j})) \]  

(B5i)

\[ \Xi(A_{j}) = \text{Tr}(\Gamma(A_{j})). \]  

(B5ii)

\( a_{\nu} \) is given in terms of the character of \( \Gamma \) as

\[ a_{\nu} = \frac{1}{h} \sum_{j} (\chi^{\nu}(A_{j}))^{*} \Xi(A_{j}), \]  

(B6)

where \( h \) is the order of the group.
Now let us calculate the matrix elements of $\bar{F}$ is the $\{|\phi_i^{αν}\rangle\}$ basis. Since all the $A_j$ are unitary operators they leave scalar products invariant; that is

$$\langle A_j f | A_j g \rangle = \langle f | A_j^{-1} A_j | g \rangle = \langle f | g \rangle.$$  \hspace{1cm} (B7)

Thus

$$\langle \phi_i^{α'ν'} | \phi_i^{αν} \rangle = \frac{1}{h} \sum_j \langle A_j \phi_i^{αν} | A_j \phi_i^{α'ν'} \rangle$$

$$= \frac{1}{h} \sum_j \sum_k \sum_k' (r^{νν'}(A_j)_k' |1|)^* r^{νν}(A_j)_k | \langle \phi_k^{α'ν'} | \phi_k^{αν} \rangle.$$  \hspace{1cm} (B8)

The irreducible representations obey the following "grand orthogonality relation" -

$$\sum_j (r^{νν'}(A_j)_k' |1|)^* r^{νν}(A_j)_k = \frac{h}{λ_ν} δ_{νν'} δ_{kk'} δ_{11'},$$  \hspace{1cm} (B9)

so that

$$\langle \phi_i^{α'ν'} | \phi_i^{αν} \rangle = δ_{νν'} δ_{11'}, \frac{1}{λ_ν} \sum_k \langle \phi_k^{α'ν'} | \phi_k^{αν} \rangle$$

$$= δ_{νν'} δ_{11'}, r^{ν}_{1α'},$$  \hspace{1cm} (B10)

independent of $i$!

Now, $\bar{F} | \phi_i^{αν} \rangle$ transforms as the $i^{th}$ row of $r^ν$, since
\[ \tilde{F} | A_j \phi_q^\nu \rangle = \tilde{F} \sum_k r^\nu(A_j)_{k1} | \phi_k^\nu \rangle = \sum_k r^\nu(A_j)_{k1} \{ \tilde{F} | \phi_k^\nu \rangle \} \]

or

\[ A_j \{ \tilde{F} | \phi_1^\nu \rangle \} = \sum_k r^\nu(A_j)_{k1} \{ \tilde{F} | \phi_k^\nu \rangle \}. \quad (B11) \]

Thus

\[ \langle \phi_1^\alpha' \nu' | \tilde{F} | \phi_1^\nu \rangle = \delta_{\nu \nu'} \delta_{\alpha \alpha'} f^\nu_{\alpha \alpha'} \quad (B12) \]

where

\[ f^\nu_{\alpha \alpha'} = \langle \phi_1^\alpha' \nu' | \tilde{F} | \phi_1^\nu \rangle = \langle \phi_2^\alpha' \nu' | \tilde{F} | \phi_2^\nu \rangle = \ldots \quad (B13) \]

This may be written as

\[ \tilde{F} | \phi_1^\nu \rangle = \sum_{\beta=1}^{a^\nu} g^\nu_{\alpha \beta} | \phi_1^\beta \nu \rangle, \quad (B14) \]

where

\[ f^\nu_{\alpha \alpha'} = \sum_{\beta=1}^{a^\nu} g^\nu_{\alpha \beta} I^\nu_{\beta \alpha'}, \quad (B15) \]

(Note that if \{ | \phi_1^\nu \rangle \} is orthonormal, then \( I^\nu_{\beta \alpha'} = \delta_{\beta \alpha'} \) and \( f^\nu_{\alpha \alpha'} = g^\nu_{\alpha \alpha'} \).)

Thus we see from Eq. B14 that \( \tilde{F} \) can only couple states of the same symmetry type. If \( a^\nu = 1 \), so that the \( \nu \)th irreducible representation only occurs once in \( \Gamma \), group theory will yield the \( \lambda^\nu \) degenerate eigenmodes in this subspace of \( S \) directly. If \( a^\nu > 1 \), we must diagonalize \( \tilde{F} \) within the subspace \{ | \phi_1^\nu \rangle, 1 \leq \alpha \leq a^\nu \}, for one particular value of \( i \). This will to determine all of the \( a^\nu \) sets of \( \lambda^\nu \)-fold degenerate eigenfunctions given by
\[ |\psi^<_V> = \sum_a \zeta^<_V|\phi^<_V>, \quad \text{(B16)} \]

where \( \zeta^<_V \) are independent of \( i \).

If the irreducible representations are known, the special symmetry functions which block diagonalize \( \bar{F} \), \( \{|\phi^<_V>\} \), may be obtained by projecting out that portion of an arbitrary function within \( S \) which transforms like each row of each irreducible representation using the projection operator

\[ p^<_Vk_i = \frac{\lambda^<_V}{n} \sum_j (\Gamma^<_V(A_j)_{ki})^* A_j. \quad \text{(B17)} \]

Now

\[ p^<_Vk_i|\phi^<_V> = \delta^<_VV\delta^<_II, \quad |\phi^<_V>, \quad \text{(B18)} \]

so that, starting with an arbitrary function \( |f> = \sum_a \sum_v \sum_i \alpha^<_V|\phi^<_V> \)

\[ p^<_Vk_k|f> = \sum_a \alpha^<_V|\phi^<_V>, \quad \text{(B19)} \]

a function which transforms like the \( k^{th} \) row of \( \Gamma^<_V \). If \( \Gamma^<_V \) appears \( a^<_V \) times in \( \Gamma \), \( a^<_V \) linearly independent such functions may be obtained in this manner by using \( a^<_V \) different functions \( |f> \).

If only the characters of the irreducible representations are known, similar but less detailed results may be obtained by using the projection operator.
\[ \rho^\nu = \sum_k P^\nu_{kk} = \frac{\lambda^\nu}{\pi} \sum_j (\chi^\nu(A_j))^* A_j. \] (B20)

Now let us apply these results to the particular case of interest here, that of determining the eigenmodes of the planar scattering operator for a particular m-beam mode geometry. As was shown in Section II, the appropriate symmetry group for the case of an m-beam mode centered around an n-fold axis of the 2-D reciprocal lattice corresponding to a scattering plane will be \( C_{nv} \) if there is no Zeemann splitting. In such a case, the polygon forming the base of the cone of the wavevectors in the mode will possess n-fold symmetry; \( C_{nv} \) is the group of rotations and reflections which leave that polygon invariant. If there is Zeemann splitting, reflections are no longer valid symmetry operations, and the group reduces to \( C_n \).

Consider first the case where \( n = m \), so that the base of the cone formed by the wavevectors is a regular m-sided polygon, and choose for a basis the set of state vectors, \( \{|\pi_i\rangle, |\phi_i\rangle\} \), where, for example, \( |\pi_i\rangle \) denotes a plane wave of unit amplitude with wavevector \( \mathbf{k}_i \), polarized along \( \pi_i \). Clearly this choice of basis vectors spans the subspace defined by the m-beams of the mode. The representation defined by this choice of basis vectors is regular, which may be seen by noting that the character of this representation is such that \( ^5 \)

\[ \Xi(A_j) = 0, \quad A_j = E \] (B21i)

\[ \Xi(E) = h. \] (B21ii)
As a specific example, consider the 2-beam case. This representation of $C_{2v}$ is given by

$$E = \begin{pmatrix}
1 & 0 & 0 & 0 \\
0 & 1 & 0 & 0 \\
0 & 0 & 1 & 0 \\
0 & 0 & 0 & 1
\end{pmatrix}$$

$$C_{2} = \begin{pmatrix}
0 & 0 & 1 & 0 \\
0 & 0 & 0 & 1 \\
1 & 0 & 0 & 0 \\
0 & 1 & 0 & 0
\end{pmatrix}$$

$$\sigma_v = \begin{pmatrix}
0 & 0 & 1 & 0 \\
0 & 0 & 0 & -1 \\
1 & 0 & 0 & 0 \\
0 & -1 & 0 & 0
\end{pmatrix}$$
\[
\sigma'_v = \begin{pmatrix}
1 & 0 & 0 & 0 \\
0 & -1 & 0 & 0 \\
0 & 0 & 1 & 0 \\
0 & 0 & 0 & -1
\end{pmatrix}
\]

where \(E\) denotes the identity, \(C_2\) is a rotation by \(\pi\), and \(\sigma_v\) and \(\sigma'_v\) are the two reflections in the plane. This representation may be seen to be regular by inspection.

Applying Eq. B6 yields, for a regular representation,

\[
a_v = \frac{1}{\hbar} \left( \chi^v(E) \right)^\dagger \Xi(E) = \frac{1}{\hbar} \lambda_v h = \lambda_v.
\]  \(\text{(B22)}\)

That is, the number of times a given irreducible representation appears in a regular representation is equal to that irreducible representation's dimension. Thus, for this case (\(n = m\) and no Zeemann splitting), there will be one eigenmode which transforms like each 1-dimensional irreducible representation, and two eigenmodes which transform like each row of each 2-dimensional one.

If \(n = m\), but there is Zeemann splitting, so that the symmetry group is \(C_m\), then either set, \(|\pi_i>\) or \(|\sigma_i>\), by itself defines a regular representation. In this case, \(a_v = 2\lambda_v\), and there will be two eigenmodes which transform like each 1-dimensional irreducible representation. In general, for these modes, \(m = \alpha n\), where \(\alpha\) is an integer, and the representation defined by the basis \(|\pi_i>, |\sigma_i>\) may
be regarded as being built up out of a regular representations (or 2α if there is Zeemann splitting). Each irreducible representation of dimension λυ will thus appear αλυ (or 2αλυ) times among the eigenmodes. In all cases the eigenmodes will be λυ-fold degenerate.

As an example of how the eigenmodes are determined, let us consider a 2-beam mode in the case of no Zeemann splitting. The group is then C₂v which has four 1-dimensional irreducible representations, each of which will occur only once among the eigenmodes. The character table of this group is

<table>
<thead>
<tr>
<th>C₂v</th>
<th>E</th>
<th>C₂</th>
<th>αυ</th>
<th>α'υ</th>
</tr>
</thead>
<tbody>
<tr>
<td>A₁</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>B₂</td>
<td>1</td>
<td>-1</td>
<td>-1</td>
<td>1</td>
</tr>
<tr>
<td>A₂</td>
<td>1</td>
<td>1</td>
<td>-1</td>
<td>-1</td>
</tr>
<tr>
<td>B₁</td>
<td>1</td>
<td>-1</td>
<td>1</td>
<td>-1</td>
</tr>
</tbody>
</table>

Since these irreducible representations are all 1-dimensional, the above table also gives the representations themselves. The eigenmodes may be obtained by applying the projection operator Pₓk (Eq. B17) to an arbitrary function in the subspace. This procedure is illustrated in Fig. B1 for the case where the initial function is |α₁>. The resulting eigenmodes are shown in Fig. B2. Thus, the mode |2αₐ>₂ corresponds to the irreducible representation A₂. |1α₂> to B₂, etc. Once the eigenmodes have been obtained in this manner, it is a simple matter to find the eigenvalues by taking expectation values.
Fig. B1. Procedure for projecting out the eigenmodes in the two-beam case. The initial vector shown is $\sigma_1$. The operators shown at the top are the identity (E), rotation by $\pi$ ($C_2$), and two reflections ($\sigma_v$ and $\sigma_v'$). The action of the operation, multiplied by the character of that class, is shown schematically under each operator for all four irreducible representations, $A_1$, $B_2$, $A_2$, and $B_1$. The resulting eigenmode (unnormalized) is the sum of all four contributions, and is shown on the right. Note that $A_1$ and $B_1$ applied to $\sigma_1$ yields zero; modes which transform like these two representations contain only $\pi$ polarization vectors and can be obtained by exactly the same procedure starting with $\pi_1$. 
$|2\pi\rangle_2$  $|2\sigma\rangle_2$  $|1\pi\rangle_2$  $|1\sigma\rangle_2$

Fig. B2. Two-Beam Eigenmodes.
APPENDIX C - Multipole Expansion of the Scattering Amplitude

Electronic Contribution to Coherent Elastic Scattering

The interaction of a photon with an atom in a crystal is described, in the interaction picture, by the scattering matrix, which is defined as\textsuperscript{56}

\[
S_{o\rightarrow f} = \langle \psi_f | U(-\omega,\omega) | \psi_o \rangle \\
= \delta_{o\rightarrow f} + 2\pi i \delta(E_f - E_o) \xi f(R_f, \epsilon_f; R_o, \epsilon_o), \tag{C1}
\]

where \(| \psi_o \rangle\) and \(| \psi_f \rangle\) are the initial and final states of the system,

\[
| \psi_o \rangle = | \psi_o \rangle_x | \hat{R}_o; \epsilon_o \rangle = \prod_{e1} | \psi_{o,e1} \rangle_x | \chi_o \rangle_x | \hat{R}_o; \epsilon_o \rangle \\
| \psi_f \rangle = | \psi_f \rangle_x | \hat{R}_f; \epsilon_f \rangle = \prod_{e1} | \psi_{f,e1} \rangle_x | \chi_f \rangle_x | \hat{R}_f; \epsilon_f \rangle \tag{C2}
\]

In these expressions, \(| \chi \rangle\) denotes the vibrational state of the atom, and the internal atomic state has been taken to be a product of individual electron states. For the coherent elastic scattering case of interest here, \(| \psi_o \rangle = | \psi_f \rangle\).

\(f(R_f, \epsilon_f; R_o, \epsilon_o)\) is the scattering amplitude while \(\xi\) is a constant equal to \(2\pi\hbar c^2/Vw\) in CGS units. \(U(-\omega,\omega)\) is the evolution operator for the system in the limit \(t_o \rightarrow -\omega\) and \(t_f \rightarrow +\omega\), given by

136
\[ U(-\infty, \infty) = \mathbb{I} - \frac{1}{\hbar} \int_{-\infty}^{\infty} H_I(t)dt \]
\[ + \frac{(-1)^2}{\hbar^2} \int_{-\infty}^{t} \int_{t}^{\infty} H_I(t)H_I(t')dt\cdot dt' + \ldots \] (C3)

where \( H_I \) is the interaction Hamiltonian

\[ H_I(t) = e^{iH_a(t)} \left[ \frac{-1}{c} \int \mathbf{j}_e(\mathbf{x}) \cdot \mathbf{A}(\mathbf{x}, t) d\mathbf{x} \right] e^{-iH_a t}. \] (C4)

In this equation, \( H_a \) is the Hamiltonian of the unperturbed electron system, so that \( H_a |\psi_0\rangle = E_0 |\psi_0\rangle \). \( \mathbf{j}_e(\mathbf{x}) = e\mathbf{e}_\mathbf{e}(\mathbf{x} - \mathbf{x}_e) \) is the electron current, while \( \mathbf{A} \) is the quantized vector potential of the radiation field, given by

\[ \mathbf{A}(\mathbf{x}, t) = \sum_{\mathbf{k}, \alpha} \frac{1}{\sqrt{\xi}} \hbar_\alpha \left[ a_{\mathbf{k}, \alpha} e^{-i(\mathbf{k} \cdot \mathbf{x} - \omega t) - a_{\mathbf{k}, \alpha}^\dagger e^{i(\mathbf{k} \cdot \mathbf{x} - \omega t)} \right] \] (C5)

Fig. C1. Feynmann diagrams for elastic scattering from the electrons.
The dominant contribution to elastic scattering will come from the lowest order Feynmann diagrams which contribute to this process. In this case, these are the second order diagrams shown in Fig. C.1. Thus, (setting \( n = 1 \))

\[
S_{0\rightarrow f} = (-1)^2 \sum_{\psi_n} \int_{-\infty}^{t} dt' |\psi_n(t)| \langle \psi_n | H(t') | \psi_o \rangle \langle \psi_n | H(t') | \psi_o \rangle.
\]

where

\[
|\psi_n\rangle = |\psi_n\rangle \times \text{photon state}
\]

\[
= |\psi_n, j\rangle \prod_{j \neq i} |\psi_o, e_1 \rangle \times |\chi_n\rangle \times \text{photon state}.
\]

Now \( \hat{A} \) acts only on the photon states; contracting these states onto \( \hat{A} \) yields the two terms shown in Fig. C.1. After integrating over the time coordinates the contribution to \( S \) from the first diagram is

\[
S_{0\rightarrow f,r} = \lim_{n \to 0} (-1) 2\pi \delta(\Delta E) \varepsilon e^2 \times
\]

\[
\sum_{\psi_n} \frac{\langle \psi_o | \hat{a} \cdot \hat{e}_f e^{-i \hat{K}_f \cdot \hat{x}} | \psi_n \rangle \langle \psi_n | \hat{a} \cdot \hat{e}_o e^{i \hat{K}_o \cdot \hat{x}} | \psi_o \rangle}{E_o - E_n + \omega + i\eta},
\]

yielding a contribution to \( f(\hat{K}_f, \hat{e}_f; \hat{K}_o, \hat{e}_o) \) of

\[
f_r = -e^2 \lim_{n \to 0} \sum_{\psi_n} \frac{\langle \psi_o | \hat{a} \cdot \hat{e}_f e^{-i \hat{K}_f \cdot \hat{x}} | \psi_n \rangle \langle \psi_n | \hat{a} \cdot \hat{e}_o e^{i \hat{K}_o \cdot \hat{x}} | \psi_o \rangle}{E_o - E_n + \omega + i\eta},
\]
where the subscript in Eqs. C8 and C9 refers to resonant. The contribution from the second diagram is nonresonant and is given by

\[
f_{nr} = -e^2 \sum \frac{\langle \psi_o | \hat{a}^\dagger \hat{e}_o e^{i \hat{k}_f \cdot \hat{x}} | \psi_n \rangle \langle \psi_n | \hat{a} \hat{e}_f e^{-i \hat{k}_o \cdot \hat{x}} | \psi_o \rangle}{E_o - E_n + \omega}.
\]

These are the scattering amplitudes for a particular atom in the crystal. To obtain the coherent elastic scattering amplitude per atom, expressions B9 and B10 must be averaged over the distribution of initial atomic and vibrational states within the crystal.

Now \( \hat{x} = \hat{x}_o + \hat{u} + \hat{r} \), where \( \hat{x}_o \) is the equilibrium position of the center of mass of the atom, \( \hat{u} \) is its deviation from equilibrium, and \( \hat{r} \) is the internal coordinate of the electron relative to that center of mass. If there is only one atom per unit cell, all the \( \hat{x}_o \) are equivalent, and this position may be chosen as the origin within the unit cell.

The vibrational states may now be contracted on \( \hat{u} \) yielding the Debye-Waller factor.

\[
\sum \langle x_o | e^{-i \hat{k}_f \cdot \hat{u}} | x_n \rangle \langle x_n | e^{i \hat{k}_o \cdot \hat{u}^\dagger} | x_o \rangle = \langle x_o | e^{\frac{-i (\hat{k}_f - \hat{k}_o) \cdot \hat{u}}{2}} | x_o \rangle = e^{-\frac{1}{2} \Delta^2 \langle u^2 \rangle} = f_D,
\]

using closure and the fact that the scattering occurs in a time much
shorter than the vibrational period of the atom. (An average over the initial vibrational states of the atoms is implicit in Eq. C11.)

The only significant contribution from the negative energy electron states will come from $E_0 - E_n \pm \omega = 2mc^2$. These states give a contribution, which in the usual x-ray notation is denoted $f_o$, of

$$f_o = \frac{-e^2}{2mc^2} f_D \times$$

$$\langle \psi_o \mid [\hat{x} \cdot \hat{r}_f (\hat{x} \cdot \hat{r}_o) + (\hat{x} \cdot \hat{r}_o) (\hat{x} \cdot \hat{r}_f)] e^{-i(\hat{r}_f - \hat{r}_o) \cdot \hat{x}} \rangle \psi_o \rangle.$$  \hspace{1cm} (C12)

where $|\psi_o\rangle$ now contains only the internal electron states. Now,

$$(\hat{x} \cdot \hat{r}_f) (\hat{x} \cdot \hat{r}_o) = \hat{r}_f \cdot \hat{r}_o + i\hat{x} \cdot (\hat{r}_f \times \hat{r}_o),$$ \hspace{1cm} (C13)

so that

$$f_o = \frac{-e^2}{2mc^2} f_D \hat{r}_f \cdot \hat{r}_o \langle \psi_o \mid \sum_j e^{-i(\hat{r}_f - \hat{r}_o) \cdot \hat{x}_j} \rangle |\psi_o\rangle$$

$$= -r_o f_D \hat{r}_f \cdot \hat{r}_o F(\theta).$$ \hspace{1cm} (C14)

$f_o$ is the instantaneous Coulomb interaction coming from the $A^2$ term, nonrelativistically, and is purely real. It is equal to $-r_oZ$ in the forward direction, but falls off with increasing $\theta$ due to interference effects since the atom is not a point scatterer. Its value at $\theta = \pi$ may be only a few electrons. A multipole expansion of $F(h)$ will not converge rapidly due to this large variation with $\theta$ and this term is better included in the analysis in an analytic form. Nevertheless, it
is instructive to carry out this expansion for comparison with later results.

Following the notation of Ref. 53,

$$\hat{c}_{ee} \hat{A}_{L M}^\lambda = \sum_{L M \lambda} (\hat{c}_{\Gamma} \hat{Y}_{L M}^\lambda (\hat{k})) \hat{A}_{L M}^\lambda (\hat{r})$$ \hspace{1cm} (C15)$$

where

$$\hat{a}_{L M}^1 = \frac{1}{\sqrt{2L+1}} \left[ \frac{L}{2L+1} g_{L+1}(\omega r) + \frac{L+1}{2L+1} g_{L-1}(\omega r) \right] \hat{Y}_{L M}^{+1}$$

$$+ \frac{\sqrt{L(L+1)}}{2L+1} \left[ g_{L-1}(\omega r) - g_{L+1}(\omega r) \right] \hat{Y}_{L M}^{-1}$$

$$= A \hat{Y}_{L M}^{+1} + B \hat{Y}_{L M}^{-1},$$ \hspace{1cm} (C16i)$$

$$\hat{a}_{L M}^0 = g_{L}(\omega r) \hat{Y}_{L M}^0,$$ \hspace{1cm} (C16ii)$$

and

$$g_{L}(\omega r) = (2\pi)^{3/2} (2L+1)^{1/2} \frac{J_{L+1/2}(kr)}{\sqrt{kr}}.$$ \hspace{1cm} (C17)$$

Substituting this expansion into Eq. C14 and remembering that the process still consists of a transition to an excited state and back down, so that there will be no cross terms in L,L', M,M', or \lambda,\lambda', yields the expression

$$f_o = -r_o f D \sum_{L M \lambda} (\hat{c}_{\Gamma} \hat{Y}_{L M}^\lambda (\hat{k}_{\Gamma}))^* (\hat{c}_{\Omega} \hat{Y}_{L M}^\lambda (\hat{k}_{\Omega})) \langle \psi_0 | \hat{a}_{L M}^\lambda \hat{a}_{L M}^\lambda \psi_0 \rangle.$$ \hspace{1cm} (C18)$$
To obtain the coherent elastic scattering amplitude, $f_0$ must be averaged over the distribution of atomic ground states in the crystal. If either $\psi_0$ is spherical (filled or half filled shells only), or there is no statistically preferred orientation of the atomic state in the crystal, the matrix element in Eq. C18 will not depend upon $M$.

For the magnetic terms, the matrix element becomes

$$
\langle \psi_0 | s_L^2(\omega r) | x_{LM}^0 \rangle^2 | \psi_0 \rangle = \int dr_0(r) g_L^2(\omega r) \int d\Omega | x_{LM}^0 \rangle^2
$$

$$
= \frac{8\pi}{2L+1} \langle g_L^2(\omega r) \rangle,
$$

(C19)

so that

$$(f_0)_{L,0} = -r_0 f_D g_L^2(\omega r) \frac{8\pi}{2L+1} \sum_M (\hat{\epsilon}_F \cdot \hat{x}_{LM}^0(\hat{r}_F)) (\hat{\epsilon}_O \cdot \hat{x}_{LM}^0(\hat{r}_O)).$$

(C20)

Evaluating the sum over $M$ for the magnetic dipole case ($L = 1$) yields

$$
\frac{8\pi}{3} \sum_M (\hat{\epsilon} \cdot \hat{x}_{1,1}^0(\hat{r}_F)) (\hat{\epsilon}_O \cdot \hat{x}_{1,1}^0(\hat{r}_O)) = (\hat{r}_F \times \hat{\epsilon}_F) \cdot (\hat{r}_O \times \hat{\epsilon}_O),
$$

(C21)

as expected.

For the electric multipoles, the component of $f_0$ is given by

$$(f_0)_{L,1} = -r_0 f_D \xi_L \frac{8\pi}{2L+1} \sum_M (\hat{\epsilon}_F \cdot \hat{y}_{LM}^1(\hat{r}_F)) (\hat{\epsilon}_O \cdot \hat{y}_{LM}^1(\hat{r}_O)),
$$

(C22)

where

$$
\xi_L = \int dr \rho(r) [A^*A + B^*B],
$$

(C23)
A and B being defined by Eq. C16i. Evaluating this for \( L = 1 \) yields

\[
(f_0)_{1,1} = -r_0 f_D \xi_1 \hat{e}_r \hat{e}_o, \tag{C24}
\]

again, as expected.

Due to its large energy denominator, the positive energy contribution from \( f_{nr} \) may generally be neglected in comparison that of \( f_r \). In the usual x-ray notation the real and imaginary parts of the contribution from the resonant term are denoted \( f' \) and \( f'' \), respectively. This resonant term may be written as

\[
f_r = e^2 f_D \lim_{n \to 0} \sum_{| \psi_n >} \frac{\psi_0 f_r e^{-i \hat{e}_r \cdot \hat{r}} \psi_n \psi_0 f_0 e^{i \hat{e}_0 \cdot \hat{r}'} | \psi_n >}{x_n - i n}
\]

\[
= e^2 f_D \lim_{n \to 0} \sum_{| \psi_n >} \frac{\psi_0 | \psi_0 > \psi_n | \psi_0 >}{x_n + i n} \frac{x_n + i n}{x_n^2 + n^2}, \tag{C25}
\]

where \( x_n = E_n - E_0 - \omega \).

The dominant contribution to this term will come from transitions to continuum intermediate electron states. Letting

\[
\sum_{| \psi_n >} \int dE_n \rho(E_n) \int dX_n \rho(X_n) \tag{C26}(X_n)
\]

gives
\[ f_r = \Delta f' + i f'' = e^2 f_D \left[ P.V. \int dX_n \rho(X_n) \langle |\rangle \langle \rho \rangle \frac{1}{x_n^2} \right] \]

\[ + i \lim_{n \to 0} \int dX_n \rho(X_n) \langle |\rangle \langle \rho \rangle \frac{n}{x_n^2 + n^2}, \tag{C27} \]

where P.V. refers to principle value.

\( \Delta f' \) and \( f'' \) are the real and imaginary parts of the anomalous dispersion, respectively. The imaginary part will determine the photoabsorption. To evaluate this term, note that

\[ \lim_{n \to 0} \frac{n}{x_n^2 + n^2} = \delta(x_n), \tag{C28} \]

and that the sum over the intermediate states may be written as

\[ \sum \delta E_n \sum \sum_{j_n m_n} \]

\[ |\psi_n\rangle \langle \rho | j_m n \]

so that

\[ f'' = e^2 f_D \sum_{LM\alpha} \left( \tilde{\epsilon}_{\alpha} \cdot \tilde{\alpha}_L^\dagger(\tilde{\gamma}_\alpha) \right) \left( \tilde{\epsilon}_\delta \cdot \tilde{\alpha}_L^\dagger(\tilde{\gamma}_\delta) \right) \times \]

\[ \sum_{j_0 m_0} \sum_{j_n m_n} |\langle j_n m_n | \tilde{\alpha}_L(\tilde{\gamma}) | j_0 m_0 \rangle|^2 \tag{C29} \]

where the matrix element is understood to be evaluated at exact resonance, \( E_n = E_0 + \omega \), and the sum over \( j_0, m_0 \) is over all the occupied shells.
Applying the Wigner-Eckhart theorem gives

\[ \langle j_n^m | \hat{a} \cdot \hat{a}^\dagger_{LM} (\vec{\hat{r}}) | j_o^m \rangle = C(j_o L | j_n^m ; m_o M_m \rangle \langle j_n^m | \hat{a} \cdot \hat{a} | j_o^m \rangle. \]  \hspace{1cm} (C30)

If the ground state has spherical symmetry so that the sum over \( m_o \) is complete,

\[ \sum_{m_o} (C(j_o L | j_n^m ; m_o M_m \rangle)^2 = \frac{2j_n + 1}{2L + 1}, \]  \hspace{1cm} (C31)

so that

\[ [f^n]_{L, \lambda} = e^2 f_D \{ \sum_M (\hat{\epsilon}_f \cdot \hat{Y}^\dagger_{LM}(\vec{r}_f))^\ast (\hat{\epsilon}_o \cdot \hat{Y}^\dagger_{LM}(\vec{r}_o)) \} \times \]

\[ \sum_{j_o j_n} |\langle j_n | \hat{a} \cdot \hat{a} | j_o \rangle|^2 \frac{2j_n + 1}{2L + 1}. \]  \hspace{1cm} (C32)

The partial cross section may be obtained by applying the optical theorem

\[ \sigma_{tot} = 4\pi \Im(f(\vec{r}_o, \hat{\epsilon}; \vec{r}_o, \hat{\epsilon})). \]  \hspace{1cm} (C33)

In this energy region, \( \sigma_{tot} = \sigma_{pe} \). Using

\[ \sum_M |\hat{\epsilon} \cdot \hat{Y}^\dagger_{LM}(k)|^2 = \frac{2L + 1}{8\pi} \]  \hspace{1cm} (C34)

yields

\[ (\sigma_{pe})_{L \lambda} = \frac{1}{2} e^2 \sum_{j_o j_n} (2j_n + 1) |\langle j_n | \hat{a} \cdot \hat{a} | j_o \rangle|^2. \]  \hspace{1cm} (C35)
Thus

\[ [f''_{\lambda}]_{L,\lambda} = \frac{(a_{\text{pe}})^{L,\lambda}}{4\pi \lambda} f_D \frac{8\pi}{2L + 1} \sum_M (\varepsilon_f \cdot \hat{\lambda}_M(\hat{\kappa}_f))^* (\varepsilon_o \cdot \hat{\lambda}_M(\hat{\kappa}_o)). \]  

(C36)

Evaluating this expression for \( L = 1 \) yields

\[ [f''_{\lambda}]_{L,\lambda} = \frac{(a_{\text{pe}})^{L,\lambda}}{4\pi \lambda} f_D \varepsilon_f^* \varepsilon_o \]  

for E1 and

\[ [f''_{\lambda}]_{L,\lambda} = \frac{(a_{\text{pe}})^{L,\lambda}}{4\pi \lambda} f_D (\hat{k}_f \times \varepsilon_f)^* (\hat{k}_o \times \varepsilon_o) \]  

for M1.

The reduced matrix element in Eq. C35 may be related to the radial matrix element which occurs in internal conversion coefficient calculations.\(^{57}\) Values for these matrix elements have been published by Band et al.\(^{58,59}\) and by Rose.\(^{60}\) In Band's notation,

\[ (a_{\text{pe}})^{L,\lambda} = 2\pi x^2 (2L + 1) \sum_{\kappa',\kappa} (\text{Re}(M_\kappa))^2, \]  

(C38)

where \( M_\kappa \) are the published matrix elements.

Partial photoelectric cross sections for \( Z = 81 \) are plotted in Fig. C2 as a function of energy. Note that the M1 contribution is several orders of magnitude less than those of E1 and E2 over the range of energies for which there is an appreciable Mössbauer effect.
Fig. C2. Partial photoelectric cross sections for Z = 81 as a function of energy. The points were calculated from the tabulated matrix elements of Band et al.58, 59

This difference is even larger for lower Z atoms. The M1 contribution to photoabsorption may thus be neglected in most multi-beam Borrmann modes.

Nuclear Contribution to Coherent Elastic Scattering

The nuclear scattering amplitude may be obtained from the scattering matrix exactly as was done for the electronic case,36 with the result that
\[ f_N = \frac{1}{n} \sum_{n} \frac{1}{E_n - E_o - \omega - i\Gamma/2} \langle o | -\frac{1}{c} \int d\vec{x} j_n(\vec{x}) \cdot \hat{\epsilon}_\omega e^{-i\vec{K}_o \cdot \vec{x}} | n \rangle \times \]

\[ \langle n | -\frac{1}{c} \int d\vec{x} j_n(\vec{x}) \cdot \hat{\epsilon}_\omega e^{i\vec{K}_o \cdot \vec{x}} | o \rangle \]  

\[
(C39)
\]

where \( j_n(\vec{x}) \) is the nuclear current, \( \Gamma \) is the width of the resonance and the initial and intermediate states are given by \( |o\rangle = |x_o, j_o, m_o\rangle \) and \( |n\rangle = |x_n, j_n, m_n\rangle \). Again making the substitution \( \vec{x} = \vec{x}_o + \vec{u} + \vec{r} \) and using Eq. C15 yields

\[ \langle n | j_l | o \rangle = \langle x_b | e^{i\vec{K}_o \cdot \hat{u}} | x_o \rangle < j_n | m_n \rangle \sum_M \hat{\epsilon}_\omega \cdot \hat{\gamma}_{LM}(k_o) \times \]

\[ C(j_oLj_n; m_o; m_n) \langle j_n | j_o \rangle, \]  

\[
(C40)
\]

where the values of \( L \) and \( \lambda \) are fixed by the values of \( j_n \) and \( j_o \) and the parity of the initial and final states. The reduced matrix element may be evaluated by noting that, by Fermi's Golden Rule,

\[ \Gamma_Y = \frac{1}{\tau} = R_{o+q} \]

\[ = \frac{1}{2\pi} \sum_{m_o} \sum_{M} \int d\Omega |\langle j_n | m_n \rangle |^{-1} \int d\vec{x} j_n(\vec{x}) \cdot \hat{\epsilon}_\omega e^{i\vec{K}_o \cdot \vec{x}} | j_o | m_o \rangle |^2 \]

\[ = \frac{1}{2\pi} \sum_{m_o} \sum_{M} \int d\Omega |\hat{\epsilon}_\omega \cdot \gamma_{LM}(k) |^2 \times \]

\[ C^2(j_oLj_n; m_o; m_n) \langle j_n | j_o \rangle |^2, \]  

\[
(C41)
\]
But
\[ \sum \int d\Omega |\hat{e}_o \cdot \hat{r}_{LM}(\hat{k})|^2 = 1, \tag{C42} \]
and
\[ \sum_{m_o^M} c^2(j_o^L j_n; m_o^M m_m^M) = 1, \tag{C43} \]
so that
\[ |\langle j_n | | j_o \rangle|^2 = \lambda_o \gamma. \tag{C44} \]

Substituting these expressions back into Eq. C39 yields
\[ f_N = 2\lambda_o \frac{\gamma}{\Gamma} \sum_{x_o^m M} \langle x_o | e^{-i \hat{k}_f \cdot \hat{u}} | x_o \rangle \langle x_o | e^{i \hat{k}_o \cdot \hat{u}} | x_o \rangle \times \]
\[ \frac{c^2 (j_o \cdot \hat{r}_{LM}(\hat{k}_o)) (\hat{e}_o \cdot \hat{r}_{LM}(\hat{k}_f))^{*}}{X_n - 1}, \tag{C45} \]

where \( X_n = 2(E_n - E_o - \omega)/\Gamma \). Now \( E_o = (E_o)_N + \epsilon_o \), where \( (E_o)_N \) is the energy of the nuclear level and \( \epsilon_o \) is the energy of the vibrational state. Similarly, \( E_n = (E_n)_N + \epsilon_n \). \( X_n \) contains the expression \( \epsilon_n - \epsilon_o \gg \Gamma \). Therefore, the only intermediate vibrational state which will contribute significantly is \( |x_o \rangle \).

To obtain the coherent scattering amplitude, Eq. C45 must again be averaged over the distribution of ground states in the crystal. Performing this average over the vibrational matrix elements yields the Mössbauer factor...
\[ f = \langle e^{-i \hat{K}_F \cdot \mathbf{u}} \rangle \langle e^{i \hat{K}_O \cdot \mathbf{u}} \rangle. \]  

(C46)

In effect, since resonant scattering occurs over a time much longer than the characteristic vibration time of the atom in the crystal lattice, the nucleus loses all "memory" of its location at the time the incident \( \gamma \)-ray was absorbed by the time the outgoing \( \gamma \)-ray is emitted.\(^6\) The Mössbauer factor is thus independent of the scattering angle.

To perform the average over the initial nuclear state, Eq. C45 must be summed over \( m_o \) and divided by \( 2j_o + 1 = g_o \), the spin degeneracy of the ground state. If there is no Zeemann splitting, \( X_n \rightarrow X \), and using Eq. C31 yields the final result

\[
\gamma_N = \frac{x}{2} \frac{f}{1 + \alpha} \frac{g_n}{g_o} \frac{1}{x^2 + 1} \left[ \frac{8\pi}{2L + 1} \sum_M (\hat{e}_f \cdot Y^2_{LM}(\hat{k}_f)) (\hat{e}_o \cdot Y^2_{LM}(\hat{k}_o)) \right],
\]  

(C47)

where \( \alpha = (\Gamma - \Gamma_Y)/\Gamma_Y \) is the internal conversion coefficient. If there is inhomogeneous broadening, \( 1 + \alpha \) is replaced by \( (1 + \alpha)(1 + a) \).

Applying the optical theorem and using Eq. C34 yields the resonant nuclear cross section

\[
\sigma_N = 2\pi x^2 \frac{f}{1 + \alpha} \frac{g_n}{g_o} \frac{1}{x^2 + 1}.
\]  

(C48)

This is the cross section for absorption. For the emission process,
summing over the final states and averaging over the initial ones yields

$$\alpha_N = 2\pi x^2 \frac{f}{1 + \alpha} \frac{1}{x^2 + 1}. \quad \text{(C49)}$$

Substituting Eq. C48 back into Eq. C46 allows the scattering amplitude to be written as

$$f_N = \frac{\alpha_N}{4\pi} \frac{x + i}{x^2 + 1} \frac{8\pi}{2L + 1 M} \sum \left( \hat{\epsilon}_f \cdot \hat{\gamma}_LM(\hat{k}_f) \right) \left( \hat{\epsilon}_o \cdot \hat{\gamma}_LM(\hat{k}_o) \right). \quad \text{(C50)}$$

If there is Zeemann splitting, it is not possible to pull the energy denominator out of the sum over $M$, and in this case, the cross section will depend upon the angle between the incident radiation and the quantization axis of the nuclear state.
APPENDIX D - Alternate Methods of Calculating $g_A$ and $g_N$

There are two additional methods by which $g_A$ and $g_N$ may be calculated, provided the eigenmodes are known. These are (1) by calculating the Poynting vector, and (2) by using Fermi's Golden Rule. In this appendix, these two methods are dealt with in turn, with particular emphasis on the simple 2-beam case, which is presented as a worked example.

**Poynting Vector Approach**

Consider a 2-beam Borrmann mode in a simple cubic crystal, of lattice constant $a$, with one atom per unit cell, as is shown in Fig. D1. The two $\hat{z}$-polarized plane wave channels, $\hat{k}_1$ and $\hat{k}_2$, are excited equally, and in phase, so that:

\[
\hat{k}_1 = k_x \hat{x} + k_z \hat{z} \quad \quad \hat{k}_2 = -k_x \hat{x} + k_z \hat{z} \quad \quad (D1i)
\]

\[
\hat{E}_1 = \hat{E} \hat{y} \quad \quad \hat{E}_2 = -\hat{E} \hat{y} \quad \quad (D1ii)\)
\]

\[
\hat{B}_1 = E(\cos \theta \hat{x} + \sin \theta \hat{z}) \quad \quad \hat{B}_2 = E(-\cos \theta \hat{x} + \sin \theta \hat{z}) \quad \quad (D1iii)
\]

The electric and magnetic fields in the (normalized) Borrmann mode may be written as

\[
\hat{E}_B = \frac{1}{\sqrt{2}} (\hat{E}_1 e^{i\hat{k}_1 \cdot \hat{r}} + \hat{E}_2 e^{i\hat{k}_2 \cdot \hat{r}}) \quad \quad (D2i)
\]

and

152
Fig. D1. A 2-beam Borrmann mode, showing the two plane wave channels, $\vec{K}_1$ and $\vec{K}_2$, as well as the reciprocal lattice vector, $\vec{t}$.

$$\vec{B}_B = \frac{1}{\sqrt{2}} (\vec{B}_1 \epsilon^{\vec{K}_1 \cdot \vec{r}} + \vec{B}_2 \epsilon^{\vec{K}_2 \cdot \vec{r}}).$$  \hspace{1cm} (D2i1)

The time averaged Poynting flux is given by

$$\vec{S} = \frac{c}{2\pi} [\vec{E}_B \times \vec{B}_B]$$

$$= \frac{1}{2} \frac{c}{2\pi} [\vec{E}_i \times \vec{B}_i + \vec{E}_j \times \vec{B}_j] \epsilon^{\vec{(K}_1 - \vec{K}_j) \cdot \vec{r}}.$$  \hspace{1cm} (D3)

We wish to calculate the total time-averaged flux going through a Wigner-Seitz cell. By symmetry, only the $\hat{z}$ component will survive upon integration over the area of the cell normal to $\vec{S}$. Now, from Eqs. D2i,11, $S_2$ is given by

$$S_2 = \frac{c}{2\pi} \frac{1}{2} E^2 \cos \theta (1 - \cos(2k_x x))^2,$$  \hspace{1cm} (D4)

so that the flux going through each Wigner-Seitz cell is
\[ S_{WS} = \int_{WS} SdA_{WS} \]
\[ = \frac{c}{2\pi} a E^2 \cos \theta \int_{-a/2}^{a/2} [1 - \cos(2k_x x)] dx \]
\[ = \frac{c}{2\pi} a^2 E^2 \cos \theta, \quad (D5) \]

where \( A_{WS} = a^2 \) is the cross sectional area of the Wigner-Seitz cell, normal to \( \hat{z} \).

Off resonance, \( S_{WS} \) will decrease with \( z \) as \( \exp(-\mu_A z) \), where \( \mu_A \) is the attenuation parameter due to interaction with the electrons. At these energies (=10-100 keV), \( \mu_A \) is almost entirely due to photoabsorption, primarily from the inner K or L shell electrons (so long as the energy is above either the K or L shell edge, respectively, so that these electrons can participate). From Fermi's Golden Rule, the rate at which photons are absorbed by any given atom is

\[ R_{abs} = \frac{\langle S \rangle \alpha_{pe}}{\hbar \omega} = \frac{c}{2\pi} \frac{\alpha_{pe} \langle E^2 \rangle}{\hbar \omega}, \quad (D6) \]

where \( \langle S \rangle \) is the average Poynting flux seen by the atom and \( \langle E^2 \rangle \) is the mean-square E field at the atomic site. Since photoabsorption is primarily due to inner shell electrons, and since these electron shells are small, the field in the neighborhood of the origin may be approximated as \( \langle E^2 \rangle \approx 2E^2 k_o^2 \langle x^2 \rangle \), where \( \langle x^2 \rangle \) is the mean thermal and zero point motion of the nucleus about its equilibrium position. This
is equivalent to assuming that the atom absorbs like a point source. Since, in reality, the K-shell has a finite physical extent, this corresponds to neglecting terms due to the finite size of the atom, of the order of the size of the K-shell. These terms will generally be smaller than the $<x^2>$ term, however, as will be shown below.

From Eq. D6, it can be seen immediately that the rate at which energy is absorbed from the field by each atom is \( \frac{c}{2\pi} \langle E^2 \rangle \sigma_{pe} \). The atoms occur at intervals of \( z \) along the \( z \) axis, so that

\[
\frac{dS_{ws}}{dz} = \frac{c}{2\pi} \frac{\langle E^2 \rangle \sigma_{pe}}{a} = -\mu_A S_{ws},
\]

or

\[
\mu_A = \frac{c}{2\pi} \frac{\langle E^2 \rangle \sigma_{pe}}{a S_{ws}} = \frac{2k_e^2 \langle x^2 \rangle \sigma_{pe}}{a^3 \cos \theta}.
\]

Now, \( \mu_A = g_A \sigma_{pe} \), where \( n = a^{-3} \), so that

\[
g_A = \frac{2k_e^2 \langle x^2 \rangle}{\cos \theta} = 2 \frac{\sin^2 \theta \langle x^2 \rangle}{x^2},
\]

as before.
To calculate $g_N$, for example for the case of $M1$ nuclear transitions, which couple to the magnetic field at the origin, note that

$$g_N = \frac{|B_B|^2}{B_{pw}^2} \frac{1}{\cos \theta}$$  \hspace{1cm} (D10)

where $B_B$ is the amplitude of the magnetic field in the Borrmann mode and $B_{pw} = E$ is the amplitude of the magnetic field in a plane wave channel. Now $B_B = \frac{1}{\sqrt{2}} (2E \sin \theta)$, so that

$$\langle g_N \rangle_{M1} = 2 \frac{\sin^2 \theta}{\cos \theta}$$  \hspace{1cm} (D11)

again, as before.

This method is not very convenient for calculating $g_N$ for higher multipole transitions, and also becomes cumbersome for modes with many beams, particularly when dealing with eigenmodes which have a more complicated structure than the one used here. It does serve to demonstrate the validity of the results, however, and provides some physical insight into the problem.
Fermi's Golden Rule

The Feynmann diagrams for the processes of photoabsorption by an electron and emission by the resonant nuclei are shown in Figs. D2a and D2b, respectively. We wish to calculate the transition matrix elements for each process, coupling the electrons and the nuclei to the single photon state which corresponds to the eigenmode of the radiation field of interest. Comparing the cross section obtained in this way with the corresponding cross section for a single plane wave channel will yield expressions for $g_A$ and $g_N$.

Addressing first the calculation of $g_A$, note that the interaction Hamiltonian between the radiation field and the atomic electrons is\[ \hat{H}_i = \frac{-1}{c} \int d\mathbf{x} \ j_\mu(\mathbf{x}) \ A_\mu(\mathbf{x}). \] (D12)

The transition of interest will occur between a single photon state $|k, \epsilon\rangle$ and the excited state $|l_j, m_j\rangle$ with quantum numbers corresponding to the transition, and the final state $|l_n, m_n\rangle$ of the electron from which the photon was absorbed.

Fig. D2. a) Feynmann diagram for photoabsorption by an electron. The incident photon ionizes the electron, leaving behind a hole in the K electron shell. b) Feynmann diagram for resonant emission from a nucleus.
(|ψ⟩) of the radiation field and the vacuum (|0⟩) accompanied by movement of a K-shell electron (|K⟩) into a continuum state (|c⟩).
(For the most part, the energies of interest are well above the K-edge, where photoabsorption is primarily due to these electrons; if not, the L shell calculation is similar.) During this process, the nucleus may change its vibrational state from the ground state (|x₀⟩) to some excited state (|x_n⟩). The transition matrix element, M_{fo}, for this process is thus

\[ M_{fo} = -\frac{\hbar}{c} \langle x_n | c | 0 \rangle \int d\vec{x} j_\mu(\vec{x}) A_\mu(\vec{x}) |ψ⟩|K⟩|x_0⟩. \]  

(D13)

For a single plane wave channel, |ψ⟩ = |\vec{K}, \epsilon⟩, so that

\[ \langle 0 | A_\mu(\vec{x}) |ψ⟩ = \left[\frac{2\pi c}{Vω}\right]^{1/2} \epsilon e^{i\vec{K} \cdot \vec{x}}, \]  

(D14)

where V is the (large) normalization volume, and \(R = 1\). Now \(j_\mu(\vec{x}) = j_\mu(\vec{x}_e) δ(\vec{x} - (\vec{x}_n + \vec{x}_e))\). Thus

\[ M_{fo} = -\frac{\hbar}{c} \left[ \right]^{1/2} \langle x_n | c | j(\vec{x}_e) \cdot \epsilon e^{i\vec{K} \cdot (\vec{x}_n + \vec{x}_e)} |K⟩|x_0⟩. \]  

(D15)

By Fermi's Golden Rule, the transition rate, R, is given by

\[ R = 2\pi |M_{fo}|^2 ρ_{E_p} d\Omega, \]  

(D16)
where $\rho_{FP, dQ}$ is the density of final states, which for a free electron, is $2mV|\vec{p}|/(2\pi)^3$. Thus

$$R = \frac{(2\pi)^2}{\omega V} |\langle c | \vec{J}(|\vec{x}_e| \cdot \hat{e} e^{i\vec{k} \cdot \vec{x}_n} |K\rangle|^2 |\langle x_n | e^{i\vec{k} \cdot \vec{x}_n} |x_0\rangle|^2 \rho_{FP, dQ}. \quad (D17)$$

Summing over the final states, $|x_n\rangle$ and $|c\rangle$, yields

$$R_{tot} = \frac{(2\pi)^2}{\omega V} \int dQ \rho_{FP, dQ} |\langle c | \vec{J}(|\vec{x}_e| \cdot \hat{e} e^{i\vec{k} \cdot \vec{x}_n} |K\rangle|^2, \quad (D18)$$

since $\sum_{x_n} |\langle x_n | e^{i\vec{k} \cdot \vec{x}_n} |x_0\rangle|^2 = |\langle x_0 | x_0\rangle = 1$. The photoabsorption cross section is simply, $\sigma_{pe} = R_{tot}[V/c]$, where $c/V$ is the photon flux.

Now consider photoabsorption in a 2-beam Borrman mode. In this case, $|\psi_B\rangle = \frac{1}{\sqrt{2}} [ |\vec{k}_1, \sigma_1\rangle + |\vec{k}_2, \sigma_2\rangle$. $M_{fo}$ is now given by

$$M_{fo} = \frac{-1}{c} \left[ \begin{array}{c} 1/2 \langle x_n|c| \vec{J}(|\vec{x}_e)| \end{array} \right]$$

$$\frac{1}{\sqrt{2}} (e^{i\vec{k}_1 \cdot (\vec{x}_n + \vec{x}_e)} + e^{i\vec{k}_2 \cdot (\vec{x}_n + \vec{x}_e)} \rangle |K\rangle |x_0\rangle. \quad (D19)$$

Now, $\vec{k}_1 \cdot (\vec{x}_n + \vec{x}_e) = k_z (z_n + z_e) \pm k_x (x_n + x_e)$. The term in $k_z$ is common and will not appear in $|M|^2$. For this 2-beam case, $\sigma_1 = \hat{y}$ and $\sigma_2 = -\hat{y}$. Thus the term $\frac{1}{\sqrt{2}} | \rangle$ in Eq. D19 reduces to

$$\frac{1}{\sqrt{2}} = \sqrt{2} i \hat{y} \sin[k_x (x_n + x_e)]$$

$$\frac{1}{\sqrt{2}} = \sqrt{2} i \hat{y} k_x (x_n + x_e) \quad (D20)$$
for small values of $x_n$ and $x_e$. Again,

$$R_{\text{tot}} = 2\pi \sum_{|c\rangle} \sum_{|x_n\rangle} |M_{fo}|^2. \quad (D21)$$

Substituting Eqs. D19 and D20 into Eq. D22 and summing over $|x_n\rangle$ yields

$$R_{\text{tot}} = \xi 2k_0^2 \sum_{|c\rangle} \langle K|\langle x_o|x_{n^*}y(x_n+x_e)|c\rangle\langle c|j_y(x_n+x_e)|x_o\rangle|K\rangle, \quad (D22)$$

where $\xi$ contains all factors of $2\pi, c,$ etc., and is identical to the similar factor which occurs in the off-Bragg case.

Assuming a cubic environment, terms like $\langle x_o|x_n|x_o\rangle = 0$, and

$$R_{\text{tot}} = \xi 2k_x^2 \sum_{|c\rangle} \left| \langle c|j_y|K\rangle \right|^2 \langle x_n^2 \rangle + \langle c|j_yx_e|K\rangle \right|^2. \quad (D23)$$

The cross section is $\sigma_{2B} = R_{\text{tot}}/\mathcal{F}_B$, where $\mathcal{F}_B$ is the photon flux in the Bornmann mode, $\mathcal{F}_B = \frac{c}{V} \cos \theta$. From above, the cross section for a single plane wave is $\sigma_{1B} = \frac{\sqrt{V}}{c} \xi \sum_{|c\rangle} \left| \langle c|j_y|K\rangle \right|^2$. Thus

$$g_A = \frac{\sigma_{2B}}{\sigma_{1B}} = \frac{2k_x^2}{c^2 \cos \theta} \left[ \langle x_n^2 \rangle + \frac{\xi(1)}{\xi(0)} \right], \quad (D24)$$

where $\xi(a) = \sum_{|c\rangle} \left| \langle c|j_yx_e^a|K\rangle \right|^2$.

The first term in Eq. D24 is identical to the expression obtained by the Poynting vector calculation above, and gives the 1st order contribution to $g_A$ due to the atom sampling the electric field as it
vibrates about its equilibrium position. The second term gives a correction due to the finite physical extent of the electron shells. Even if the nucleus did not deviate from its equilibrium position, the electrons would still see a non-zero electric field, since the field is only strictly zero at the origin.

The magnitude of the finite size correction may be easily estimated by making several approximations. Using non-relativistic free electron wave functions for the final state \( |c\rangle \), (which will give an over-estimate of \( g_A \)) and setting \( \mathbf{f}(\mathbf{x}_e) = \frac{q}{m} \mathbf{p} \) yields

\[
\zeta(\alpha) = \int d\mathbf{n}_P \, \rho_{E_P} \, d\mathbf{n} \, |I_\alpha|^2, \tag{D25}
\]

where

\[
I_\alpha = \langle c | \mathbf{j}(\mathbf{x}_e) \cdot \mathbf{f} \, x^3 | k \rangle = \xi' \, p_y \int d^3 \mathbf{r} \, e^{-i \mathbf{p} \cdot \mathbf{r}} \, x^3 \, \exp\left(-\frac{Z}{a_0}\right). \tag{D26}
\]

In this equation, \( a_0 \) is the Bohr radius, \( Z \) is the atomic number of the nucleus, and \( \mathbf{r} = \mathbf{x}_e \).

Performing the integration for \( I_0 \), and using the fact that \( I_1 = 133_p \, I_0 \) yields, after some algebra, the final result,

\[
\frac{\zeta(1)}{\zeta(0)} = \frac{16}{5} \left(\frac{a_0}{Z}\right)^2 \frac{(p a_0/Z)^2}{[1 + (p a_0/Z)^2]^2}, \tag{D27}
\]

where \( p \) is the magnitude of the momentum of the final electron. If
screening is neglected, and $E_k = z^2 / (2ma_0^2)$ is used for the energy of the K-shell electron, Eq. D27 may be written as

$$\frac{\xi(1)}{\xi(0)} = \frac{16}{5} \left( \frac{a_0}{z} \right)^2 \frac{E_k}{E_{\gamma}} (1 - \frac{E_k}{E_{\gamma}}).$$

(D28)

Using values for $^{57}\text{Fe}$ gives $\xi(1)/\xi(0) = 3.1 \times 10^{-4} \text{Å}^2$, which is to be compared to $\langle x_n^2 \rangle = 1.4 \times 10^{-3} \text{Å}^2$ at $T=0$ (or $4.2 \times 10^{-3} \text{Å}^2$ at room temperature). Thus, for $^{57}\text{Fe}$, the finite size contribution in a 2-beam Borrmann mode is at most 20% of the Debye-Waller term.

This approach may also be applied to modes with more than two beams. In particular, calculating the finite size correction directly for the mode $|4\sigma_4\rangle$ gives zero to first order, as one might perhaps expect knowing that E2 photoabsorption is suppressed for $|4\sigma_4\rangle$. For this mode,

$$M_{f0} = \langle x_n | c | \frac{J_x}{2} \left[ -e^{ik_2 \cdot \hat{x}} + e^{ik_4 \cdot \hat{x}} \right]$$
$$+ \frac{J_y}{2} \left[ e^{ik_1 \cdot \hat{x}} - e^{ik_2 \cdot \hat{x}} \right] |K>|x_o\rangle,$$

(D29)

where $\hat{x} = \hat{x}_n + \hat{x}_e$. Expanding, and keeping $\hat{x}$ to first order yields

$$M_{f0} = \langle x_n | c | \left[ -iJ_xk_x(y_n + y_e) + iJ_yk_x(x_n + x_e) \right] |K>|x_o\rangle.$$

(D30)

After performing the integration of Eq. D25, we find that
so that the contribution from the electron coordinates cancels out to this order. To obtain a non-zero finite size correction for this mode by this method, it would be necessary to keep terms of order \( x_e^2 \). In doing so, the algebra rapidly becomes quite tedious, and in this case, the expansion of \( \bar{F} \) by multipoles is a much better approach.

Calculating the finite size contribution directly in the \( |2\sigma\rangle_4 \) mode yields a transition matrix element

\[
M_{f0} = \langle \chi_n | \langle c | \left[ \frac{J_x}{2} \left[ e^{i \hat{r}_2 \cdot \hat{x}} - e^{i \hat{r}_4 \cdot \hat{x}} \right] \\
+ \frac{J_y}{2} \left[ e^{i \hat{r}_1 \cdot \hat{x}} - e^{i \hat{r}_2 \cdot \hat{x}} \right] \right] |K\rangle \langle \chi_o |, \quad (D32)
\]

which is to be compared to Eq. D29 for the \( |4\sigma\rangle_4 \) mode. Note the sign change in the term proportional to \( J_x \). Expanding, and keeping \( \hat{x} \) to first order yields

\[
M_{f0} = \langle \chi_n | \langle c | \left[ iJ_x k_x (y_n + y_e) + iJ_y k_x (x_n + x_e) \right] |K\rangle \langle \chi_o |. \quad (D33)
\]

Since both terms now have the same sign, rather than cancelling out, the first order finite size correction for \( |2\sigma\rangle_4 \) is twice that for an equivalent 2-beam mode, so that for this mode (in comparison with Eq. D24)
\[(g_A)_{2o>4} = \frac{2}{\cos \theta} \langle x_n^2 \rangle + 2 \frac{\xi(1)}{\xi(0)} \].

(D34)

Recall that the leading term in E1 photoabsorption for the mode \( |3o>6 \) goes as \( \langle x^2 \rangle^2 \). Calculating \( g_A \) for this mode using this approach yields \( M_{fo} = M_{fo,j_x} + M_{fo,j_y} \), where

\[ M_{fo,j_x} = \frac{1}{2\sqrt{2}} \langle x_n | \langle c | J_x \left( e^{i \hat{k}_2 \cdot \hat{x}} - e^{i \hat{k}_3 \cdot \hat{x}} + e^{i \hat{k}_5 \cdot \hat{x}} - e^{i \hat{k}_6 \cdot \hat{x}} \right) | x_o \rangle | K \rangle \].

(D35i)

and

\[ M_{fo,j_y} = \frac{1}{2\sqrt{6}} \langle x_n | \langle c | J_y \left( 2e^{i \hat{k}_1 \cdot \hat{x}} - e^{i \hat{k}_2 \cdot \hat{x}} - e^{i \hat{k}_3 \cdot \hat{x}} \right. \]
\[ + 2e^{i \hat{k}_4 \cdot \hat{x}} - e^{i \hat{k}_5 \cdot \hat{x}} - e^{i \hat{k}_6 \cdot \hat{x}} \left. \right) | x_o \rangle | K \rangle \].

(D35ii)

Expanding either \( M_{fo,j_x} \) or \( M_{fo,j_y} \) to first order in \( \hat{x} \) gives zero. To obtain an expression for \( g_A \), it is necessary to carry the calculation to second order in \( \hat{x} = \hat{x}_n + \hat{x}_e \). Unfortunately, the algebra becomes nearly intractable, and in this case, a better approach is to diagonalize \( \tilde{F} \) by multipoles.

Turning now to the nuclear coupling, the amplitude for emission into a single plane wave channel by a transition from the state \( |j_n,m_n> \) to the state \( |j_o,m_o> \) is given by

\[ M_{fo} = -\frac{1}{c} \langle m_o | \langle R, \hat{c} | \hat{f}(\hat{x}_n) \cdot \hat{A}(\hat{x}_n) | 0 \rangle | m_n \rangle \]
\[
\begin{align*}
&= -\frac{1}{c} \left( \frac{2\pi c^2}{\omega V} \right)^{1/2} \langle m_o | \hat{J}(\hat{x}_n) \cdot \hat{e} e^{-i \hat{R} \cdot \hat{x}_n} | m_n \rangle \\
&= -\frac{1}{c} \left[ \right]^{1/2} C(j_o, l, j_n; m_o, M, m_n) \sqrt{\lambda_o \gamma} \ \mathcal{Y}_{LM}^{(\lambda)}(\hat{R}) \cdot \hat{e},
\end{align*}
\]

(see Appendix C) where \( C(j_o, l, j_n; m_o, M, m_n) \) is the Clebsch-Gordan coefficient, \( \gamma \) is the radiative width of the transition, \( \lambda_o \) is the wavelength of the emitted radiation, and \( \mathcal{Y}_{LM}^{(\lambda)}(\hat{R}) \) is a vector spherical harmonic,\(^{53}\) the first few of which are tabulated in Appendix A.

If there is no Zeeman splitting, the total transition rate is obtained by summing over the final states and averaging over the initial ones, so that

\[
R = \frac{1}{2j_n + 1} \sum_{m_n m_o} \frac{(2\pi)^2}{\omega V} |C(\ ; \ ;)|^2 \lambda_o \gamma \ |\mathcal{Y}_{LM}^{(\lambda)}(\hat{R}) \cdot \hat{e}|^2.
\]  

(D37)

Changing the sum over \( m_n \) and \( m_o \) to a sum over \( M \) and \( m_o \) \((m_o = m_n + M)\) and using the identities

\[
\sum_{m_o} |C(\ ; \ ;)|^2 = \frac{2j_n + 1}{2L + 1}
\]  

(D38)

and

\[
\sum_{M} |\mathcal{Y}_{LM}^{(\lambda)}(\hat{R}) \cdot \hat{e}|^2 = \frac{2L + 1}{8\pi}
\]  

(D39)

yields
\[ R = \xi'' \sum_M |\tilde{\gamma}_M(\lambda)(R) \cdot \tilde{e}|^2, \]  
(D40)

\[ \alpha \xi'' = \frac{2L + 1}{8\pi} = \frac{\lambda \rho\Gamma_Y}{\omega V} \]  
(D41)

where \( \xi'' = \frac{(2\pi)^2\lambda \rho\Gamma_Y}{\omega V(2L + 1)} \). Again, the cross section, \( \sigma_N \), is obtained by dividing \( R \) by the photon flux, \( c/V \).

For the 2-beam Borrmann mode, Eq. D40 becomes

\[ R = \xi'' \sum_M \frac{1}{2} |\tilde{\alpha}_1 \cdot \tilde{\gamma}_M(\lambda)(R_1) + \tilde{\alpha}_2 \cdot \tilde{\gamma}_M(\lambda)(R_2)|^2. \]  
(D42)

Dividing by the photon flux in the Borrmann mode again gives the cross section, so that

\[ \sigma_n = \frac{\sigma_{2B}}{\sigma_{1B}} = \frac{1}{\cos \theta} \frac{R_{2B}}{R_{1B}} \]

\[ = \frac{1}{\cos \theta} \frac{1}{2} |\tilde{\alpha}_1 \cdot \tilde{\gamma}_M(\lambda)(R) + \tilde{\alpha}_2 \cdot \tilde{\gamma}_M(\lambda)(R_2)|^2. \]  
(D43)

Evaluating this equation for the case of an M1 transition (\( \lambda=0, L=1 \)) yields

\[ \sigma_{N,10} = 2 \frac{\sin^2 \theta}{\cos \theta} \]  
(D44)

as before.
REFERENCES


38. C. G. Darwin, Phil. Mag. 27, 315 (1914); 27, 675 (1914); 43, 800 (1922).


41. M. von Laue, Röntgenstrahleninterferenzen (Becker and Erler, Liepzig, 1941), Chapt. V.


43. See, for example, R. W. James, The Optical Principles of the Diffraction of X-Rays (Ox Bow Press, Woodbridge, Conn. 1982).


45. Neither the angular divergence of the emerging beams or the physical extent of the wave field produced within the crystal by a single nuclear decay may be calculated in this manner, however. These results depend on the scattering in the vicinity of the exact Bragg point, as well as the scattering at that particular point. To answer these questions, it is necessary to solve the full set of coupled equations (Eqs. 16). This will be a subject of our future research.


47. It is possible to form true Borrmann modes in crystals with more than one atom per unit cell only if the atoms are located at the proper positions and the mode axis is aligned along a particular crystal axis. For example, such modes are possible in diamond structure if the mode axis is aligned along the unique (111) displacement axis of the crystal. In this case, both atoms in the unit cell will lie on the nodal lines of the electric field in the mode. This will not be true if the mode is aligned along any other (111) axis.


53. A. I. Akhiezer and V. B. Berestetskii, Quantum Electrodynamics (Interscience, N.Y. 1965).


56. See, for example, J. J. Sakurai, Advanced Quantum Mechanics (Addison-Wesley, London, 1967).


