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DOUBLE ELECTRON MUON RESONANCE

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Double Electron Muon Resonance

by

David A. Vanderwater

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APPROVED, THESIS COMMITTEE:

\[\underline{\text{Thomas L. Estle}}\]
T. L. Estle
Professor of Physics
Chairman

\[\underline{\text{S. A. Dodds}}\]
S. A. Dodds
Assistant Professor of Physics

\[\underline{\text{R. B. McLellan}}\]
R. B. McLellan
Professor of Mechanical Engineering and Materials Science

HOUSTON, TEXAS

MAY 1981
ABSTRACT

Double Electron Muon Resonance

by

David A. Vanderwater

Muon-containing defects in insulators and in semiconductors have been studied for several years using the muon spin rotation (μSR) technique. Double electron muon resonance (DEMUR) is an extension to the more common technique. This new double resonance technique can be used to observe EPR transitions of these muonic defects that are not directly observable in normal μSR. This experimental technique has been applied successfully to muonium in quartz. A general theory of the response of two strongly coupled spin 1/2 particles to an intense rf magnetic field, when particularized to muonium in quartz, is found to describe the experimental results in detail.
Acknowledgements

I would like to thank all those, both here at Rice and up at Los Alamos, without whose help this work would not have been so enjoyable to me. Special thanks must first go to Tom Estle for many of the original ideas contained within this thesis.

To the ever pessimistic Stan Dodds, who has taught me never to be completely satisfied, many thanks for all the advice and assistance in actually making everything work. To Bob (the czar) Heffner, without whose leadership no μSR collaboration would be possible at Los Alamos, thanks for keeping such a divergent group working so well together for so long. To Keith Blazey, whose sharp wit was always well aimed, thanks for the many deserved criticisms.

And of course, to the hordes of others who helped with the time consuming process of actually acquiring the data for this experiment, thanks for all your help.
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Introduction

Investigations of muon-containing defects in solids have recently progressed beyond the early superficial surveys of these centers. Detailed studies have been made of the nature of some of the more interesting of the observed muonic defect centers.[1] The work to be discussed here concerns the study, by a new double resonance technique, of positive muons implanted in semiconductors and insulators. This new technique which we have labeled Double Electron Muon Resonance (DEMUR), is capable of measurements that are not possible with any other existing technique.

The relevant properties of the positive muon are listed in table 1. When implanted in a solid, the positive muon, with its mass of approximately 1/9 that of the proton, will behave very much like a light isotope of hydrogen. The analogy between this work and the technologically important problem of hydrogen in semiconductors is obvious. Amorphous silicon-hydrogen alloys have recently been developed with useful semiconducting properties.[2] These materials have been used to construct various types
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<td><strong>Rest Mass</strong></td>
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<tr>
<td>$m_\mu = 105.6595 \text{ MeV}$</td>
</tr>
<tr>
<td>$= 206.7684 \text{ m}_e$</td>
</tr>
<tr>
<td>$= 0.112612 \text{ m}_p$</td>
</tr>
<tr>
<td><strong>Charge</strong></td>
</tr>
<tr>
<td>$+</td>
</tr>
<tr>
<td><strong>Spin</strong></td>
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<tr>
<td>$1/2$</td>
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<tr>
<td><strong>Magnetic Moment</strong></td>
</tr>
<tr>
<td>$\mu_\mu = \frac{en}{2m_\mu c} = 4.49052 \times 10^{-26} \text{ J/T}$</td>
</tr>
<tr>
<td>$\frac{\mu_\mu}{h} \approx 13.3538 \times 10^{-3} \text{ MHz/\text{gauss}}$</td>
</tr>
<tr>
<td><strong>Decay</strong></td>
</tr>
<tr>
<td>$\mu^+ \rightarrow \bar{\nu}_\mu + e^+ + \nu_e$</td>
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<tr>
<td><strong>Lifetime</strong></td>
</tr>
<tr>
<td>$\tau_\mu = 2.1994 \mu\text{sec.}$</td>
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Table 1: Properties of the positive muon, from [8].
of semiconducting devices including solar cells. Any insight into the basic nature of hydrogen or of muon impurities in silicon or germanium could conceivably be useful in the further development of these alloy devices.

Recent photoconductivity studies on hydrogen impurities in germanium show evidence for hydrogen forming complexes with other impurities present in these very pure samples. When positive muons are implanted in germanium, three types of muonic defects are observed. Extensive μSR studies in germanium, and in silicon and diamond where similar muonic centers are observed, have not yet been able to confirm the microscopic structure of these defects. A determination of the basic nature of these muonic centers is desirable; are they simply related to the hydrogenic centers observed in germanium, or are they a totally new type of center not observed in the hydrogen work?

What is μSR?

Muon Spin Rotation (μSR) is a free-precession magnetic resonance technique very similar to free induction decay. In μSR a μ⁺ particle from a spin polarized beam is stopped in a sample, resulting in a μ⁺ imbedded in the sample. This stopping process proceeds initially by scattering from electrons. The spin-dependent forces present in this scattering process are too small to cause
any significant reorientation of the muon spin.[7] For kinetic energies below an energy which for low density gases is approximately 200 eV a fair fraction of the muons will have captured an electron, forming a muonium-like complex. The last portion of the muon momentum is lost by collisions of this muonium atom with the atoms of the target material. This process occurs too rapidly to allow any reorientation of the muon spin. The total time between a muon entering the target and final thermalization is between $10^{-10}$ and $10^{-9}$ seconds.[8]

Obviously this technique requires the use of an intense spin-polarized positive muon beam. Such a beam may be produced from an intense pion source by allowing the pions to decay in flight. In this weak decay process, the positive pion decays into a positive muon and a neutrino. The neutrino, by its very nature, must be longitudinally spin polarized. Since the pion has no spin, to conserve both linear and angular momentum, the muon must be emitted with its spin orientation antiparallel to its momentum direction in the rest frame of the pion.[9] By selecting the portion of the decay muons with a specific momentum direction in the pion rest frame (usually parallel to or antiparallel to the pion momentum) it is possible to achieve muon beams with 80% polarization.[10] In practice this is accomplished by first selecting the pions with a specific momentum, and then after the pion has decayed, by
selecting either the low or the high momentum muons. This double momentum selection results in a muon beam with longitudinal polarization, either with positive or with negative helicity depending upon the portion of the decay muons that are selected.

By placing appropriate detectors in such a beam, a polarized muon may be observed entering and stopping in a sample. This muon enters the sample at a known time, with a known initial spin orientation, is stopped in a time short compared to its lifetime with its spin orientation preserved in the stopping process. At some later time this muon will decay. This decay again involves the weak interaction, the decay products are a positron and two neutrinos. Because this process involves the parity-violating weak interaction, the positron will be emitted preferentially along the direction of the muon spin at the time of its decay.[9] By detecting this decay positron, it is possible to infer the orientation of the muon spin.

By detecting all decay positrons emitted in a given direction, and by measuring the time interval between the muon stop and the detection of the decay positron for each muon that enters the sample, it is possible to measure the time dependence of the component of the spin of an ensemble of muons in the direction of the positron detector. Typically, this data is kept as a time differential spectrum; this spectrum consists of a histogram containing the
number of decay positrons detected as a function of the
time interval between the muon stop and its subsequent
decay. For the present work a stationary magnetic field
was applied transverse to the muon beam direction, and
consequently transverse to the initial spin direction of
the muon. In the presence of such a field, the muon spin
will precess; this precession may be observed in the time
differential spectrum, see figure 1.

The apparatus necessary for such a measurement is
diagrammed in figure 2. The muon, with its spin polariz-
atation along the direction of its momentum, passes through
the M counter signaling the timer to begin counting. If
the muon stops in the sample the F counter will fail to
detect it; if the muon passes through the sample without
stopping, the F counter will detect this and signal a bad
event. At some subsequent time the muon will decay. If
the decay positron passes through the two positron counters
($E_1$ and $E_2$), it will be recorded as a good event and will
increment the histogram channel corresponding to the time
interval between the detection of the muon stop and the
detection of the decay positron.

This technique differs in some fundamental respects
from the more conventional resonance techniques. The first
difference is in the number of magnetic particles present
in the sample at a given time. EPR and other techniques
require a large number of spins to be present in the sample
Figure 1: Typical transverse field μSR data. The number of counts is plotted versus the time. The exponential decay of the muon is responsible for the decay. The oscillations are due to the precession of the muon in a transverse magnetic field.
Figure 2: Schematic representation of a transverse field μSR spectrometer. A longitudinally polarized $\mu^+$ particle enters from the right, triggering the M counter, and failing to trigger the F counter, indicating that it has stopped in the sample. The decay positron may be detected by the two counters $E_1$ and $E_2$. 
(usually $> 10^{12}$/cc.), μSR requires exactly one muon to be present in the sample at a given time. This infinite dilution eliminates any possible effects due to 'like spin' interactions that may occur in NMR and other techniques.

The second major difference between μSR and other magnetic resonance techniques is the time scale characteristic of the measurement. The 2.2 microsecond lifetime of the muon limits μSR measurements to time periods shorter than about 10 microseconds. Experimental considerations limit the resolution of a μSR experiment to about 1 nanosecond. Other resonance techniques, such as NMR, usually make measurements on a much longer time scale; typically a few milliseconds or longer.

The third major difference between μSR and other magnetic resonance techniques is in the mode of detection. In μSR the properties of the muon decay make it possible to directly observe the spin orientation of the muon as a function of time. More conventional techniques usually rely upon more indirect measures of the spin polarization, such as a measurement of the ac magnetic susceptibility of the sample.

Finally μSR is capable of measurements in field regions that are inaccessible to most other magnetic resonance techniques. Because the observed signal in μSR is a result entirely of the properties of the muon decay, and is not dependent upon an rf driving or an external
polarizing field, \( \mu SR \) experiments may be carried out in any convenient magnetic field. \( \mu SR \) experiments, with slight modifications, are even carried out in zero field. In typical NMR experiments field values of at least a few kilogauss are required to make the nuclear transitions accessible to the rf driving field.

In most insulators and semiconductors the muon will, with significant probability, remain as a muonium-like atom. This muon-electron pair will be isolated, the corresponding hole being left elsewhere in the crystal by the final thermalization process. It is this isolated muonium-like defect center that is of interest in the present work.

An isolated muonium atom can be described by a simple isotropic spin Hamiltonian of the form:

\[
\mathcal{H} = g_\mu H^\ast S - g_\mu \mu_\alpha H^\ast I + A S^\ast I ,
\]

where \( S \) and \( I \) are the e- and the \( \mu^+ \) spin operators, and \( H \) is an externally applied magnetic field.\([11]\) The hyperfine interval, \( A \), for the free muonium atom is large, 4463.3 MHz,\([12]\) compared to a typical electronic Zeeman splitting of about 280 MHz for a field magnitude of 100 gauss (see figure 3). Observations of the \( \mu SR \) of such a system are generally done in the low field region (\( g_\mu H \ll |A| \)). In this low field (Zeeman) region the two spin 1/2 particles are strongly coupled together by the dominant hyperfine
interaction, forming an $F = 1$ triplet and an $F = 0$ singlet. Due to an experimental inability to observe any transition whose frequency is larger than a few hundred MHz, only the two $\Delta m_F = 1$ transitions within the $F = 1$ triplet will be observed in the $\mu$SR. Of course, a muonium-like defect in a solid is not so simply described; but in some cases, such as the muonium center in quartz at room temperature, the complications are minor.[13]

Although many muonium-like centers are very similar to free muonium, there are a few centers with observed hyperfine splittings much smaller than the vacuum value of 4463 MHz. The most intensively studied of these centers with small hyperfine couplings are the 'anomalous' muonium centers in the group IV semiconductors. With a much smaller hyperfine interval (typically a few percent of the vacuum value) $\mu$SR observations are not done in the low field region. In the high field (Paschen-Back) limit $F$ is not a good quantum number; in this region the eigenfunctions are single products of electronic and muonic spin functions. The observed transitions for a system in this region are described as electron spin flip ($\Delta m_s = 1, \Delta m_I = 0$), or as muon spin flip ($\Delta m_I = 1, \Delta m_s = 0$).
Figure 3: Typical Breit-Rabi diagram for a system of two coupled spin 1/2 particles. For the case of muonium in the high field region; the electron spin flip (EPR) transitions are $1 \leftrightarrow 4$ and $2 \leftrightarrow 3$, the muonic (μSR) transitions are $1 \leftrightarrow 2$ and $3 \leftrightarrow 4$, the transitions $1 \leftrightarrow 3$ and $2 \leftrightarrow 4$ are forbidden double spin flip transitions.
What is DEMUR?

In the case of free muonium, or of muonium-like centers, the electron spin is coupled to the muon spin by the hyperfine coupling. If the electron spin could be perturbed by some external influence, the effects of that influence might be observable in measurements on the muon spin. In particular, consider the effects on the $\mu$SR of an oscillatory magnetic field applied in such a manner as to resonantly drive one of the EPR transitions of this system. Such a double resonance experiment can be described as the detection of an EPR transition by its effects on a $\mu$SR transition. This new double resonance technique, which we have labeled Double Electron Muon Resonance (DEMUR), will be described in detail together with the results of the first experimental observation of DEMUR.

Solution of the eigenvalue problem for the spin Hamiltonian for free muonium (equation 1) yields 4 eigenstates. In the high field limit ($g\mu_B H \gg |A|$) these 4 states may be described by a simple basis set with the electron and the muon spin aligned parallel to or antiparallel to the field direction. The six transitions between these high field states can therefore be described as electron spin flip (EPR), muon spin flip ($\mu$SR), or double spin flip transitions. Conventional $\mu$SR, because only the muon spin orientation is observed, can detect only those transitions that involve the muon spin flip. DEMUR,
because the electron spin flip transition is driven, can probe some of those transitions to which the $\mu$SR is insensitive. Of course, in any real muonium-like defect system, the Hamiltonian will be more complex; however, as in this simple example, there will be situations that are more suited to DEMUR than to normal $\mu$SR.

As an example of a system more suited to study by DEMUR than by conventional $\mu$SR, consider the 'anomalous' muonium center, Mu*, in silicon. This center, first observed in 1973 [14], was not understood even from a phenomenological viewpoint until 1978 when a thorough $\mu$SR study showed that the observed spectrum could be explained by an axially symmetric spin Hamiltonian.[5] Currently the precise microscopic nature of this center is still largely conjectural. Studies by conventional $\mu$SR were able to determine, with good accuracy, the hyperfine tensor and even the muonic $g$ factors. But, because the transitions most sensitive to variations in the electronic $g$ tensor are not observed, only approximate results for these electronic $g$ factors have been obtained. DEMUR, because it is not limited to observations made entirely on the muon spin, would be far more suited to a measurement of these electronic $g$ factors, than would conventional $\mu$SR.

The very small and very anisotropic hyperfine interaction observed in these Mu* centers indicates that the electron is tightly bound to the muon and to its near
neighbors.[15] Accurate measurements of the electronic Zeeman interaction would yield additional information on the properties of the electronic state, and provide evidence of the structure of these defect centers.
Theory

To drive the EPR transitions of muonium in a solid, an oscillatory magnetic field is used. The theory of DEMUR presented here involves the solution of the time-dependent Schroedinger equation obtained from a standard spin Hamiltonian. This Hamiltonian includes both electronic and muonic Zeeman terms, as well as a hyperfine interaction term. No assumptions concerning the actual form of the electronic g tensor or of the hyperfine A tensor need be made until the results are applied to a specific system. The muonic Zeeman term included here is assumed to be isotropic. This treatment also neglects any relaxation effects that may be present, and the interactions with any nuclear spins in the surrounding host material. These approximations appear to be valid for quartz at room temperature, and for silicon, germanium, and diamond at sufficiently low temperatures.\[5,6\]

Quite generally, for an electron spin of \(1/2\), and excluding any nuclear spins in the host material,\[16\] we have:
\[ H = u_{\beta} H_{\beta} \ast q_{\ast} S + S \ast A \ast I - g_{\mu} u_{\mu} H_{\mu} \ast I \tag{2} \]

where the total field \( H_{\epsilon} \) includes both the dc component, and a uniform linearly-polarized oscillatory portion:

\[ H_{\epsilon} = H_{e} + H_{f} \cos(\omega t + \phi) \tag{3} \]

For convenience, in this calculation the muon stops are assumed to be at \( t=0 \), and \( \phi \) is the phase angle of the oscillatory field at this time. The oscillatory field included here is assumed to be linearly polarized because of the ease of generating such a field, not because of its conceptual simplicity.

The observable quantity in this experiment is the spin polarization of the muon, \( P = 2\langle I \rangle \). The derivation to be presented here will proceed in a very straightforward way: direct calculation of the solutions of the Schrödinger equation for the Hamiltonian presented in equation 2, followed by the calculation of the observable quantity from these solutions. One alternate approach to this problem would be to transform to a reference frame rotating at the applied frequency. Another alternate approach is to make use of the very powerful density matrix formalism, either treating the problem in the laboratory reference frame or in a rotating reference frame. Either of these methods have the added feature that relaxation effects may be included. Since no relaxation is to be expected, at least in the initial experiments, this will not be neces-
sary here. One alternate approach including relaxation effects is illustrated for a simple spin 1/2 system in appendix A.

The solution of the time-dependent Schroedinger equation proceeds by first solving the time-independent problem. By taking $\mathcal{H}_t$ in equation 3 as the stationary component of the total field, a time-independent spin Hamiltonian results. The stationary states of this Hamiltonian have wave functions $\psi_n$ and energies $\hbar \omega_n$ for $n = 1, \ldots, 4$. The solution of the Schroedinger equation for the total time-dependent Hamiltonian can then be taken as a linear superposition of these 4 stationary states:

$$\Psi(t) = \sum_{n=1}^{4} a_n(t) \psi_n e^{-i \omega_n t} , \quad (4)$$

with time dependent coefficients $a_n(t)$.

Before proceeding with the solution of the time-dependent problem, it is helpful to consider further the time-independent case. This time-independent problem is identical to that of calculating the frequencies and amplitudes of the conventional $\mu$SR spectrum for the system represented by this particular spin Hamiltonian. In the present context this time-independent problem is nothing more than that of calculating the initial values of the $a_n(t)$. To specify the $a_n(0)$ it is sufficient to determine the initial state of the spin system. If we assume that the $\mu^+$ beam is 100% polarized (it is actually only approx-
imately 80% polarized), the polarization of the $\mu^+$ is known at $t=0$. The initial polarization of the $e^-$ is unknown and is assumed to be random. The initial state of the spin system is then most easily written in terms of the high field basis functions $|m_s, m_r>$, using random variables to describe the $e^-$ spin state. The coefficients of the eigenfunctions $\Psi_i$ may then be calculated from:

$$a_i(0) = <\Psi_i | \Psi(t=0)> .$$

(5)

A more detailed discussion of the procedure for determining the $a_i(0)$ is included in appendix B.

In DEMUR, as in conventional $\mu$SR, it is the spin polarization of the $\mu^+$ that is detected. The observed quantity $P(t)$, therefore can be calculated from:

$$P(t) = 2 <\Psi(t) | I | \Psi(t)> .$$

(6)

Combining this with equation (4) for $\Psi(t)$, the observed polarization in terms of the $a_i(t)$ and the $\psi_i$ is determined from:

$$P(t) = 2 \sum_{m_s,m_r} a_m^*(t) a_m(t) e^{i\omega_{nm}t} <\psi_m | I | \psi_n> ,$$

where, $\omega_{nm} = \omega_m - \omega_n$.

(7)

A calculation of the results of a conventional $\mu$SR experiment could be accomplished by using equation (7) and the initial values of the $a_i(t)$:
\[ P = 2 \sum_{m,n=1}^{A} a_{m}^{*}(0) a_{n}(0) e^{-i\omega_{mn}t} \langle \psi_{m} | \Omega | \psi_{n} \rangle. \] (8)

The calculation of the DEMUR spectrum requires only knowledge of the \( a_{n}(t) \), the \( \psi_{n} \) and the \( \omega_{n} \). Equation 7 can be broken up into components each resulting from a single pair of states:

\[ P(t) = P_{0}(t) + \sum_{z=1}^{4} P_{z}(t), \]

where,

\[ P_{z}(t) = 2 \sum_{v=2}^{4} |a_{z}(t)|^{2} \langle \psi_{v} | I | \psi_{z} \rangle, \]

and,

\[ P_{z}(t) = 4 Re \left[ a_{z}^{*}(t) a_{j}(t) e^{i\omega_{j} t} \langle \psi_{v} | I | \psi_{j} \rangle \right]. \] (9)

Returning to the time-dependent Hamiltonian of equation 2, the time dependence of the \( a_{n}(t) \) can be deduced from the time-dependent Schroedinger equation. Substituting the expression in equation 4 for the total wave function \( \Psi(t) \) into the full time-dependent Schroedinger equation:

\[ i\hbar \frac{d}{dt} \Psi(t) = \mathcal{H} \Psi(t), \] (10)

yields, in terms of the \( a_{n}(t) \):

\[ i \frac{d}{dt} a_{n} = \sum_{m=1}^{A} a_{m} e^{i\omega_{mn}t} \Omega_{mn} \cos(\omega t + \phi), \]

where,

\[ \Omega_{mn} = \frac{i}{\hbar} \langle \psi_{m} | u_{p} B_{1} \psi_{n} | \psi_{n} \rangle. \] (11)

The solution of these coupled differential equations will determine the time dependence of the \( a_{n}(t) \) coefficients.
From this time dependence, and the initial values of the \( a_x \), the entire DEMUR spectrum may be calculated using equations 7 or 9 for \( \bar{p}(t) \).

These coupled differential equations suggest a very complicated time dependence for the \( a_x(t) \). Before an attempt is made to solve this system of equations some simplifications are possible. The only terms on the right hand sides of equations 11 which will lead to a contribution to the \( a_x(t) \) with appreciable amplitude are those representing transitions whose frequency is nearly resonant with the driving field. The characteristic interaction strength of any of these terms is given by the \( \Omega_{ij} \) matrix element. Only those terms whose total frequency \( (\omega_{ij} \pm \omega) \) has a magnitude comparable to that of the interaction strength for that term will contribute to the results.\(^{[18]} \)

Those terms that satisfy the condition:

\[
|\Omega_{ij}| / |(\omega_{ij} \pm \omega)| \ll 1 \tag{12}
\]

may be neglected as having little effect on the results.

In the simplest case the rf frequency is on or near resonance with a single transition frequency \( \omega_{k\ell} \), which is well separated from any other transition frequency of the system.\(^{[19]} \) Then the differential equations simplify considerably. Taking \( \omega \approx \omega_{k\ell} > 0 \), and labeling the other two stationary states with \( m \) and \( n \) we have:
\[ \frac{d}{dt} a_m = \frac{d}{dt} a_n = 0, \]
\[ i \frac{d}{dt} a_k = \frac{1}{\sqrt{2}} \sum_{k,l} a_{k} e^{-i(\omega - \omega_{kl})t} e^{i\phi}, \]
\[ i \frac{d}{dt} a_{\ell} = \frac{1}{\sqrt{2}} \sum_{k,l} a_{k} e^{i(\omega - \omega_{kl})t} e^{i\phi}. \] (13)

Equations 13 require that \( a_m(t) \) and \( a_n(t) \) be constant. In this approximation, which shall be referred to as the "two-level" calculation, only \( a_k \) and \( a_{\ell} \) are time dependent.

In solving these equations it is convenient to eliminate the dependence of the differential equations on the initial phase of the applied rf field, \( \phi \), by the transformation:

\[ b_\ell(t) = a_\ell(t) e^{-i\phi}, \]
\[ b_k(t) = a_k(t). \] (14)

Equations 13 then become:

\[ i \frac{d}{dt} b_k(t) = \frac{1}{\sqrt{2}} \sum_{k,l} b_{\ell} e^{-i(\omega - \omega_{kl})t}, \]
\[ i \frac{d}{dt} b_\ell(t) = \frac{1}{\sqrt{2}} \sum_{k,l} b_{k} e^{i(\omega - \omega_{kl})t}. \] (15)

The solution of these equations still requires a moderate amount of algebra, but it is at least straightforward.

Before presenting a solution, several features evident from the form of the equations are noteworthy. Since the two equations are coupled, and the initial values of the \( a_k(t) \) are independent of \( \phi \) whereas the initial value of \( b_\ell(t) \) is proportional to \( e^{-i\phi} \); two terms will be present
in the solutions for $b_c(t)$, one independent of $\phi$ and one proportional to $e^{i\phi}$. Assuming an oscillatory solution for the $b_c(t)$ in equation 15, two eigenfrequencies are found for each $b_c(t)$. The solutions, written in terms of $a_k^e$ and $a_k^d$ are:

$$a_k^e = \frac{1}{2} (1 + \frac{\pi}{\sqrt{z^2 + 1}}) a_k^e(0) - \frac{1}{2} \frac{1}{\sqrt{z^2 + 1}} a_k^e(0) e^{i\phi_k} e^{-i\omega t}$$

$$+ \frac{1}{2} (1 - \frac{\pi}{\sqrt{z^2 + 1}}) a_k^e(0) + \frac{1}{2} \frac{1}{\sqrt{z^2 + 1}} a_k^e(0) e^{i\phi_k} e^{i\omega t},$$

$$a_k^d = \frac{1}{2} (1 - \frac{\pi}{\sqrt{z^2 + 1}}) a_k^d(0) - \frac{1}{2} \frac{1}{\sqrt{z^2 + 1}} a_k^d(0) e^{-i\phi_k} e^{i\omega t}$$

$$+ \frac{1}{2} (1 + \frac{\pi}{\sqrt{z^2 + 1}}) a_k^d(0) + \frac{1}{2} \frac{1}{\sqrt{z^2 + 1}} a_k^d(0) e^{i\phi_k} e^{i\omega t},$$

where $\omega_\pm = \frac{1}{2} \left( z \pm \sqrt{z^2 + 1} \right) |\Omega_{kd}|$

$$z = \frac{\omega_\pm - \omega_{kd}}{|\Omega_{kd}|}$$

and $e^{i\phi_{kd}} = \frac{\Omega_{kd}}{|\Omega_{kd}|}. \quad (16)$

When these solutions for $a_c(t)$ are combined with equation 7 for $P$, the observed spectrum may be calculated. The calculated spectrum, separated into components as defined in equations 9, is:

$$P_{kd} = \mathcal{R}_0 \left[ \left( 1 + \frac{z}{\sqrt{z^2 + 1}} \right)^2 C_0 

+ \frac{1}{\sqrt{z^2 + 1}} (1 + \frac{z}{\sqrt{z^2 + 1}}) C_1 e^{i\phi} - \frac{1}{z^2 + 1} C_2 e^{i\phi} \right] e^{i(\omega_{kd} + 2\omega)t}$$
\[
\mathcal{P}_{km} = 2 \mathcal{R} \left\{ \left[ (1 + \frac{\varepsilon}{\sqrt{\varepsilon^2 + 1}}) D_0 - \frac{i}{\sqrt{\varepsilon^2 + 1}} D_1 e^{i\phi} \right] e^{i(\omega_k t + \omega_0) t} + \left[ (1 - \frac{\varepsilon}{\sqrt{\varepsilon^2 + 1}}) D_0 + \frac{i}{\sqrt{\varepsilon^2 + 1}} D_1 e^{i\phi} \right] e^{i(\omega_m t + \omega_0) t} \right\}
\]

\[
\mathcal{P}_{mm} = 4 \mathcal{R} \left\{ a_m^*(0) a_m(0) <\psi_m|\mathbb{I}|\psi_m> e^{i\omega_{mm} t} \right\}
\]

\[
\mathcal{P}_0 = 2 \left[ |a_k(0)|^2 <\psi_k|\mathbb{I}|\psi_k> + |a_\varphi(0)|^2 <\psi_k|\mathbb{I}|\psi_k> + |a_m(0)|^2 <\psi_m|\mathbb{I}|\psi_m> + |a_m(0)|^2 <\psi_m|\mathbb{I}|\psi_m> \right]
\]

\[
+ \left[ <\psi_k|\mathbb{I}|\psi_k> - <\psi_k|\mathbb{I}|\psi_k> \right] \frac{i}{\sqrt{\varepsilon^2 + 1}} \mathcal{R} \left\{ |a_\varphi(0)|^2 - |a_k(0)|^2 - 2\varepsilon C_3 + (|a_k(0)|^2 - |a_\varphi(0)|^2 + 2\varepsilon C_3) e^{i(\omega_\varphi - \omega_\varphi) t} \right\}
\]

where,

\[
C_0 = a_k^*(0) a_\varphi(0) <\psi_k|\mathbb{I}|\psi_k>,
\]

\[
C_1 = (|a_k(0)|^2 - |a_\varphi(0)|^2) e^{-i\phi_{\mathbb{I}}} <\psi_k|\mathbb{I}|\frac{1}{\mathbb{I}}>,
\]

\[
C_2 = a_\varphi^*(0) a_\varphi(0) e^{i\phi_{\mathbb{I}}} <\psi_k|\mathbb{I}|\psi_k>,
\]

\[
C_3 = a_k^*(0) a_\varphi(0) e^{-i\phi_{\mathbb{I}}},
\]

\[
D_0 = a_k^*(0) a_m(0) <\psi_k|\mathbb{I}|\psi_m>.
\]
\[ D_i = a_{x}^{m}(0) \ a_{m}^{x}(0) e^{-i \hbar z} \langle \psi_{k}^{x} | I | \psi_{m}^{x} \rangle . \] (17)

The solutions for \( P_{km}^{x}(t) \), \( P_{\omega m}^{x}(t) \), and \( P_{\omega r m}^{x}(t) \) are all very similar to the solution for \( P_{km}^{x}(t) \) shown here.

The three different types of spectra that may appear in DEMUR are diagrammed in figure 4 for a hypothetical system. First, if the driven frequency \( \omega_{kl} \) is observed in the \( \mu \)SR (\( \omega_{kr} \) in figure 4), it will be split into three components; the middle frequency appearing at the frequency of the oscillatory field \( \omega \) flanked by two lines displaced in frequency by \( \sqrt{z^2 + 1} \ | \Omega_{kl} \). The second type of DEMUR spectrum possible is the case of the observed transition sharing a single eigenstate with the driven transition (\( \omega_{kr} \) in figure 4). This results in a doubling of the single \( \mu \)SR line with a characteristic splitting of \( \sqrt{z^2 + 1} \ | \Omega_{kl} \). Finally, if a transition sharing no endstates with the driven transition is observed in the \( \mu \)SR (\( \omega_{kr} \) in figure 4), then no splitting occurs. From these results it is also evident that the initial phase of the oscillatory field does not in any way affect the frequencies of the DEMUR lines. If the phase can be held constant for all \( \mu^+ \) stops, then the amplitudes of the various DEMUR lines will show a dependence upon this phase.

These results are quite general. The only approximation taken in obtaining them, in addition to neglecting relaxation and the interactions with any nuclear spins of the host, was that of neglecting all but the \( \omega_{kl} \) transition.
Figure 4: Energy levels and frequency spectrum of a hypothetical muonium-like system. These two sets of data show the three types of DEMUR splittings that are possible.
when solving the differential equations. These results do not depend upon the complexity of the spin Hamiltonian, nor upon the details of the energy levels or wave functions; but only upon the transition $\omega_{k2}$ being well separated from all others as stated by equation 12.

As an example, consider the case of an isotropic spin Hamiltonian, of the form:

$$\mathcal{H} = \mu_5 g_5 \mathbf{H} \cdot \mathbf{S} - \mu_\mathbf{I} g_\mathbf{I} \mathbf{H} \cdot \mathbf{I} + \mathbf{A} \cdot \mathbf{S}.$$  \hspace{1cm} (18)

The time-independent eigenvalue problem will yield, in terms of the high field states $|m_\mathbf{S}, m_\mathbf{I}\rangle$:

$$\Psi_1 = |1/2, 1/2\rangle \quad , \quad E = A/4 + \frac{i}{2} (g_5 \mu_5 - g_\mathbf{I} \mu_\mathbf{I}) H$$

$$\Psi_2 = c|1/2, -1/2\rangle + s|-1/2, 1/2\rangle$$

$$, \quad E = -A/4 + A/2 \sqrt{1 + x^2}$$

$$\Psi_3 = |-1/2, -1/2\rangle \quad , \quad E = A/4 - \frac{i}{2} (g_5 \mu_5 - g_\mathbf{I} \mu_\mathbf{I}) H$$

$$\Psi_4 = c|-1/2, 1/2\rangle - s|1/2, -1/2\rangle$$

$$, \quad E = -A/4 - A/2 \sqrt{1 + x^2}$$

where

$$x = (g_5 \mu_5 + g_\mathbf{I} \mu_\mathbf{I}) H/A$$

$$\zeta = \sqrt{\frac{i}{2x} \left( 1 \pm \frac{x}{\sqrt{x^2 + 1}} \right)}$$  \hspace{1cm} (19)

Assume that the stationary field direction is perpendicular to the oscillatory field, and that the beam direction is along that of the oscillatory field:
\[ \mathbb{H}_\perp \mathcal{H}_1 \parallel \text{the beam}. \quad (20) \]

Further assume that no effort to phase lock the \( \mu^+ \) stop with the oscillatory field has been made, and that the oscillatory field is applied at a frequency very near \( \omega_{z3} \):

\[ \phi \text{ is random, } \omega \approx \omega_{z3}. \quad (21) \]

Then the observed Demur spectrum would be:

\[
P_{Z} = 0, \\
P_{Z} = \frac{2\mathcal{N}}{\rho_0} \left( 1 + \frac{Z}{\sqrt{\gamma_{z3}^2}} \right) \left[ (1 - \frac{Z}{\sqrt{\gamma_{z3}^2}}) e^{i(\omega_{z3} - \omega_0) \mathcal{T}} \\
+ (1 + \frac{Z}{\sqrt{\gamma_{z3}^2}}) e^{-i(\omega_{z3} - \omega_0) \mathcal{T}} \right] \\
+ \frac{1}{\mathcal{G}} \left( (1 - \frac{Z}{\sqrt{\gamma_{z3}^2}}) \left[ (1 + \frac{Z}{\sqrt{\gamma_{z3}^2}}) e^{i(\omega_{z3} + 2\omega_0) \mathcal{T}} \\
+ \frac{2}{\sqrt{\gamma_{z3}^2}} e^{i\omega_0 \mathcal{T}} + (1 - \frac{Z}{\sqrt{\gamma_{z3}^2}}) e^{-i(\omega_{z3} + 2\omega_0) \mathcal{T}} \right] \\
+ \frac{1}{\mathcal{G}} \left( (1 - \frac{Z}{\sqrt{\gamma_{z3}^2}}) e^{i\omega_0 \mathcal{T}} + \frac{1}{\mathcal{G}} \left( (1 + \frac{Z}{\sqrt{\gamma_{z3}^2}}) e^{-i(\omega_{z3} - \omega_0) \mathcal{T}} \right) \right] \right)
\]

where

\[ z = \frac{\omega - \omega_{z3}}{\omega_\infty} \]

\[ \omega_{z3} = \frac{1}{2} \left( z \pm \sqrt{z^2 + 1} \right) \omega_\infty \]

\[ \omega_\infty = \left( \sqrt{1 + \frac{Z}{\sqrt{\gamma_{z3}^2}}} g_{\mu \nu} - \sqrt{1 - \frac{Z}{\sqrt{\gamma_{z3}^2}}} g_{\mu \nu} \right) \mathcal{H}. \quad (22) \]

These assumptions would be valid for muonium in quartz at low fields where only \( \omega_{z2} \) and \( \omega_{z3} \) are observable in the
μSR, provided that $|\Omega_{12}|/(\omega_{12} - \omega) << 1$. For quartz, however, because these two observed μSR transitions are often quite close together the conditions assumed in this "two-level" solution may not always be satisfied.

As an example of the extensions to this simple theory that are possible, consider a system in which two transitions sharing a common level are near enough to the driving frequency to be of importance. Because three eigenstates of the time-independent problem are involved this will be labeled the "three-level" solution. To particularize this general problem to a case applicable to muonium in quartz it is sufficient to assume that $\omega_k > \omega_c > \omega_m$ and that the two transitions $\omega_{kl}$ and $\omega_{km}$ appear at nearly the same frequency. Other cases are possible, the solutions of any of these alternatives are only slightly different from the solution to the particular case applicable to quartz presented below.

If two transitions $\omega_{kl}$ and $\omega_{km}$ appear at nearly the same frequency and all others are well separated from this pair, then only terms from the original differential equations involving $(\omega_{kl} - \omega)$ and $(\omega_{km} - \omega)$ need be kept. These assumptions would be valid for muonium in quartz for conditions where the assumptions taken in obtaining the "two-level" results may not be satisfactory.
Retaining only these terms, the differential equations simplify to:

\[
\begin{align*}
    i \frac{d}{dt} a_n &= 0 , \\
    i \frac{d}{dt} a_m &= \frac{i}{2} a_k \Omega_{km}^* e^{i(\omega - \omega_m)t} e^{-i\phi} , \\
    i \frac{d}{dt} a_k &= \frac{i}{2} a_m \Omega_{mn} e^{i(\omega - \omega_m)t} e^{-i\phi} \\
    &\quad + \frac{i}{2} a_k \Omega_{kl} e^{i(\omega - \omega_{kl})t} e^{-i\phi} , \\
    i \frac{d}{dt} a_k &= \frac{i}{2} a_k \Omega_{kl} e^{i(\omega - \omega_{kl})t} e^{-i\phi} .
\end{align*}
\]

(23)

To solve these equations, assume an \( a_i(t) \) of the form:

\[
a_i(t) = \sum_j C_j^i e^{i\omega_j^i t} .
\]

(24)

where the \( C_j^i \) are time-independent coefficients. Using the relation between the \( \omega_j^i \) that results when equations 23 and 24 are combined, the eigenvalue equation for \( \omega_m^j \) becomes:

\[
\begin{vmatrix}
    \omega_m^j - 2\omega + \omega_{kl} + \omega_{km} & \Omega_{kl} e^{-i\phi} & 0 \\
    \Omega_{km}^* e^{i\phi} & \omega_m^j - \omega + \omega_{km} & \Omega_{km} e^{-i\phi} \\
    0 & \Omega_{km}^* e^{i\phi} & \omega_m^j
\end{vmatrix} = 0
\]

(25)

which will result in 3 solutions for \( \omega_m^j \). This then leads to 3 solutions each for \( \omega_k^j \) and \( \omega_k^j \).

From these results, it would at first appear that 9 frequency components would result in the DEMUR spectrum for each of the two pSR transitions involved, \( \omega_{kl} \) and \( \omega_{km} \);
however because the $\omega_1^1$ are not all independent, there are 
a total of only 7 lines resulting from the original pair. 
Similarly observations made near $\omega_1^1$, $\omega_2^1$ or $\omega_{mn}$ would 
result in detection of 3 DEMUR lines where only 1 $\mu$SR line 
exists.

Numerical solutions for this "three-level" calcu-
lation have been obtained for parameters specific to muonium 
in quartz. One set of such results is shown in figure 5 
for experimentally realistic parameters. In figure 5 the 
frequency and amplitudes of the DEMUR results are plotted 
against the applied frequency $\omega$. The 7 DEMUR lines are 
arbitrarily labeled 1-7 as indicated. For conditions 
consistent with the "two-level" calculation, the approp-
riate 5 of these 7 lines differ from the "two-level" results 
in equations 22 by less that 1% in frequency, and by less 
than 10% in amplitude. The additional two lines predicted 
by this more complete theory will occur with amplitudes so 
small as to be virtually unobservable in any real experi-
ment.
Figure 5: Results of the numerical solution of the differential equations applicable to quartz. These figures show the theoretically predicted frequencies and amplitudes for DEMUR observations made with a dc field of 124.8 gauss, and an rf field magnitude of 1.6 gauss. These results include terms from the differential equations involving both $\omega_z$ and $\omega_y$, the "three-level" calculation.
Experimental Apparatus

In order to demonstrate the feasibility of DEMUR, an experiment on muonium in quartz was devised. Quartz, because of its strong muonium signal at room temperature and its nearly isotropic spin Hamiltonian, was an obvious choice as a test system.[13] In some other systems, such as the Mu$^*$ center in silicon, the signal characteristic of the defect center of interest appears only at low temperatures (for Mu$^*$ in Si only below 200 K). The quartz experiment was done at room temperature, in air, with a minimum of apparatus necessary.

This experiment was carried out at the C. P. Anderson Meson Physics Facility (LAMPF), a portion of the Los Alamos Scientific Laboratory (LASL), located in Los Alamos, New Mexico. The Stopped Muon Channel at LAMPF utilizes an intense pion beam to produce a spin polarized muon beam. The particular beam used for this experiment was tuned to select the portion of the muons decaying from the pion beam in the backward direction in the pion rest frame. This "backward decay" momentum selection results in a muon beam with a momentum of 80 MeV/c and its spin aligned parallel
to its momentum.

\( \mu SR \text{ Apparatus} \)

Figure 6 shows the configuration of the \( \mu SR \) spectrometer used to acquire the data for this experiment. The Helmholtz coils used to generate the static magnetic field were arranged to produce an homogenous field in the vertical direction at the position of the sample. The current passing through these coils was controlled to approximately 1 part in 10,000, insuring that the field magnitude did not vary with time. The muon beam, with its spin initially directed along its momentum, entered in the horizontal plane. Counter telescopes were arranged to detect positrons emitted in directions orthogonal to both the beam and the field.

As the muon enters the apparatus it triggers two scintillation detectors, labeled BE\(_1\) and BE\(_2\). The muon then proceeds through detectors B\(_1\) and M, finally reaching the sample. If the muon passes through the sample without stopping, its passage will be detected by the F\(_1\) counter; a signal in this counter indicates a bad event and is not accepted as a \( \mu^+ \) stop. A muon scattered into either the L\(_1\) or the R\(_1\) counter will also signal a bad event. The decay positron emitted at some time after a muon stop is detected by one of two positron telescopes, each consisting of two
Figure 6: Top view of the μSR spectrometer. The μ⁺ beam enters from the right, passing through three collimators (C₁ is not shown), and stops in the sample at the center of the magnet. Note that this apparatus has 2 sets of positron detectors.
scintillation detectors ($R_1$ and $R_2$ or $L_1$ and $L_2$).

The electronics that accompanies this apparatus is diagrammed in figure 7. Signals directly from photomultiplier tubes attached to each of the scintillation detectors are first passed through discriminators, converting them to standard digital pulses; these pulses are then routed to the rest of the logic circuit. As previously described, a "Mu" stop consists of signals in $BE_1$, $BE_2$, $M$, and no signals in $R_1$, $L_1$, or $F_1$ ($Mu = BE_1 * BE_2 * M * (R_1 + L_1 + F_1)$). This "Mu" signal first passes through a 'Pile Up Rejector' (PUR) to insure that only one muon is in the sample at a time, and then is used to start a 'Time to Amplitude Converter' (TAC).

This pile up rejector has two outputs. The 'Busy' output will turn on when a pulse is detected at the input, and remain on for a specified period after this input pulse; if a second pulse arrives at the input while this output is active, the active period is extended until the specified period has elapsed without any input pulses. The second output, the 'Pileup' signal is activated only when two or more input pulses arrive in a time interval shorter than the specified pileup interval.

A decay positron passing through the right positron counters is indicated by signals in $R_1$ and $R_2$ and no signals in $L_1$, $B_1$, $F_1$, $M$ or $BE_2$ ($RI = R_1 * R_2 * L_1 * (B_1 + F_1 + M + BE_2)$). This "RI" signal is used to stop the TAC, and after a
Figure 7: Simplified logic diagram of the μSR spectrometer counting electronics. Note that only the right side is shown; the circuit elements necessary for the detection of decay positrons are repeated for the left side. The one-shots are pulse stretchers, set to output pulses of up to 10's of microseconds duration.
second pileup circuit, is used to "Gate" the 'Analog to Digital Converter' (ADC). If more than one positron appears during the specified positron pileup interval, this pileup circuit will block the gate signal to the ADC, effectively disabling this event. The signal from the TAC, after it is digitized by the ADC, is read into a computer where separate histograms are kept for the right and the left positron telescopes.

DEMUR Apparatus

The experimental requirements of DEMUR differ from those of standard \( \mu \)SR only in the need for an intense rf magnetic field. For the quartz experiment the rf field required was achieved with a resonant circuit, diagrammed in figure 8. A three turn coil, tightly wound around a single crystal quartz sample, together with an air variable capacitor were used as the resonant circuit. This circuit was inductively driven by a series tuned primary coil (tuned to match the 50 ohm impedance of the driving line). To monitor the field amplitude during the experiment, a third coil was placed near the other two; the rf signal from this coil was rectified, filtered and used as a monitor.
Figure 8: Diagram of the circuit used to generate the large rf magnetic fields necessary for the DEMUR experiment in quartz.
This circuit was enclosed in a Faraday cage to isolate the rf fields, and mounted in the μSR apparatus previously described. The circuit was mounted with the rf field direction oriented in the horizontal plane; provisions were made to allow the direction of the field to be moved in this plane by rotating the sample and coil assembly.

This rf circuit was driven by a 40 Watt broadband amplifier whose input came from an rf oscillator. The oscillator was not phase locked to the μ⁺ stops. The resonance of the secondary circuit had a measured Q factor of approximately 200 when critically coupled (coupling was determined from the SWR of the entire circuit). The maximum rf field obtainable in this manner was found to be limited by heating of the circuit; dissipating several watts in the apparatus caused significant heating and drift of the resonance frequency. We were able to operate this apparatus stably at field amplitudes as large as 1.56 Gauss (as determined from the DEMUR results) with frequencies between 150 and 190 MHz. Because the DEMUR splitting is proportional to the field magnitude, the homogeneity of the field produced in this manner was of concern; however no line width beyond the 0.6 MHz instrumental width of our data acquisition system was observed.
Experimental Results

A data set obtained from this apparatus consists of two histograms, each containing the time differential decay spectrum of the $\mu^+$ particle in a given direction. The data were taken over a 1.6 microsecond period and digitized into time bins of 0.5 nanosecond width. The actual resolution of the apparatus was limited to approximately 1 - 2 nanoseconds by time jitter in the counting electronics. A typical data set consisted of data from a total of between 2 and 8 million $\mu^+$ decay events in two histograms.

Such data were obtained on a quartz sample for various rf field magnitudes, at frequencies near each of the two $\mu$SR transitions and off resonance, with the rf field aligned both parallel to and perpendicular to the beam direction, as well as without the rf field. The sample consisted of a single crystal of synthetically grown quartz, cut approximately into a cube 2 - 3 cm. on a side. The sample was mounted with the rf field direction along the crystallographic c axis of the quartz. The large size of the sample eliminated the need to introduce any moderators into the beam to achieve a satisfactory muon stopping
distribution.

This data was analyzed by Fourier transformation, and by fitting. The fit consisted of a linearized least squares fit to a function containing a total of up to 5 frequency components in addition to the \( \mu^+ \) precessional component and background terms.[20] In all cases both methods resulted in identical values to within expected accuracy; all numerical results quoted below are those given by the average of the results of individual fits to each of the two histograms within a data set weighted by the calculated errors. All errors quoted are one standard deviation errors, as determined from these fits.

The spin Hamiltonian for muonium in quartz is very nearly isotropic.[13] The corrections to an isotropic Hamiltonian necessary to account for the anisotropy are small, and may easily be made by incorporating a field dependent hyperfine parameter. The DEMUR calculations were done using an isotropic spin Hamiltonian with an empirically determined hyperfine parameter. Because of the nature of the two low frequency transitions of muonium in quartz, the "three-level" calculation described in the theory section was used. Since, in this experiment no attempt was made to control the phase of the oscillatory field, an average over the phase angle was made.
As previously mentioned, the magnitude of the rf magnetic field was obtained from the DEMUR results. In order to obtain an initial estimate of this field magnitude, and of the actual position of the resonance, the analytical results of the "two-level" calculation were used. These "two-level" results, although not as accurate as the numerical "three-level" results, are sufficiently accurate to allow good estimates of these parameters. A typical DEMUR spectrum for quartz consists of a total of five frequency components; three lines near the applied frequency, and two near the other μSR transition frequency. The "two-level" results predict that the ratio of the amplitudes of the pair of lines composing the doublet, or of any pair of lines from the triplet, will be a function of the parameter z only (see equations 16 and 17). Once either pair of DEMUR lines has been used to determine z, the magnitude of the rf field is easily obtained from the characteristic splitting of either group of lines; and then the actual position of \( \omega_{kl} \) may be determined.

With these two parameters, and the spin Hamiltonian parameters which may be determined from the conventional μSR, the numerical "three-level" results corresponding to these particular experimental conditions may be calculated. Before a final set of parameters is chosen, the experimental results are compared with numerical results for a range of values of each of the parameters near those predicted by
the "two-level" results. It is the result of this adjustment procedure that will be compared with the experimentally determined results.

For simplicity, in this experiment the dc magnetic field was held constant while the rf frequency was varied from one data set to the next. Similar effects could be observed by holding the rf frequency constant and varying the dc field, but the results would be more difficult to interpret. For all data presented here the dc magnetic field was applied perpendicular to both the beam direction and the rf field direction. The field magnitude, as determined from the μSR results, was held at 124.8 Gauss. This field magnitude gives μSR lines at approximately 167.2 MHz and 180.5 MHz for $\omega_{12}$ and $\omega_{23}$, respectively. At this field value an isotropic hyperfine interaction would have an interval of approximately 4620 MHz (this is actually 2.5% higher than the true value due to the slight anisotropy that is present).[13]

Figure 9 shows the Fourier transform power spectra of a μSR data set for quartz at room temperature and a DEMUR data set for the same sample. In part (b) of the figure the rf magnetic field was applied near the upper μSR transition frequency. The splitting characteristic of DEMUR can easily be seen in this transform spectrum. The bars included on part (b) of this figure indicate the results of the "three-level" calculation of the DEMUR
Figure 9: Fourier transform power spectra for muonium in quartz at room temperature. In (a) no rf power was applied, while in (b) an rf magnetic field of 1.6 gauss amplitude was applied at 180.279 MHz. The vertical bars represent the calculated frequencies and powers. The apparent discrepancies between the calculated intensities and the observed Fourier transform are due to counting statistics, and are to be expected.
intensities and frequencies for the experimental conditions.

The results of the Fourier transform of three DEMUR data sets are shown in figure 10. The results of the more complete "three-level" calculation are indicated on the figure as bars. The first set of data (figure 10a) shows the results of the transform of a DEMUR data set with the rf frequency near the lower $\mu$SR transition (167.2 MHz). The three components of this spectrum with frequencies closest to that of the lower $\mu$SR line are characteristic of the transition being irradiated also appearing in the $\mu$SR. The two components in this spectrum appearing near the frequency of the upper $\mu$SR line (180.5 MHz), show the characteristics of the observation of a transition sharing only a single eigenstate with the irradiated transition. In the second data set (figure 10b), the rf power was applied at 173.9 MHz, well away from both $\mu$SR frequencies; no effect on the $\mu$SR spectrum is observed. The third data set (figure 10c) was obtained with the rf frequency near the upper $\mu$SR transition (180.5 MHz), reversing the roles of the two $\mu$SR transitions from that found in the first set. The additional 2 DEMUR lines predicted by the "three-level" calculation are, for all conditions obtainable with this apparatus, unobservable; and so are not indicated with the theoretical results.
Figure 10: Results of DEMUR on quartz. These three Fourier power spectra show the effects of applying an rf magnetic field near (a) the lower $\mu$SR frequency, (b) the mid-frequency, and (c) the upper $\mu$SR transition frequency. For all three sets of data, the dc magnetic field was 124.8 g., and the rf magnetic field was held at 1.0 g. amplitude.
Tables 2 and 3 contain the results of the fits to several sets of DEMUR data along with the results of the numerical "three-level" calculation for appropriate conditions for each set. The results contained in table 2 came from the same three data sets presented in figure 10. The discussion of the contents of table 2, therefore, will be identical to that for figure 10.

Table 3 contains results of data runs with the largest rf fields obtainable from this apparatus. The analyses contained in this table represent three DEMUR spectra taken with the rf field applied near the upper $\mu$SR transition (compare to figure 10c or entry c in table 2). The three sets of data presented in this table show the effects on the intensities of the DEMUR lines of slightly missing the resonance condition. For comparison purposes, these 5 DEMUR lines correspond to the 5 lower frequency lines predicted by the "three-level" results shown in figure 5 (these are labeled 1 - 5 in figure 5).

The first feature to note in these data is that the characteristic DEMUR splitting, although constant within a data set, is not identical for all three data sets. For the first two sets, where the applied frequency is very nearly on resonance ($\nu_{28} = 180.51$ MHz), the splitting is 1.61 MHz; for the third set of data the splitting is 1.86 MHz. This increase in the DEMUR splitting as the applied frequency is moved farther from resonance is predicted by
<table>
<thead>
<tr>
<th>Data Set</th>
<th>Applied Field</th>
<th>Experimental Results</th>
<th>Theoretical Results</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Freq. (MHz)</td>
<td>ampl.</td>
</tr>
<tr>
<td>a</td>
<td>167.301 MHz</td>
<td>166.25±.025 .0133±.0009</td>
<td>166.32 .0124</td>
</tr>
<tr>
<td></td>
<td>1.0 g</td>
<td>167.24±.018 .0187±.0009</td>
<td>167.30 .0189</td>
</tr>
<tr>
<td></td>
<td></td>
<td>168.17±.05 .0064±.0009</td>
<td>168.28 .0072</td>
</tr>
<tr>
<td></td>
<td></td>
<td>179.69±.034 .0098±.0009</td>
<td>179.92 .0091</td>
</tr>
<tr>
<td></td>
<td></td>
<td>180.84±.034 .0100±.0009</td>
<td>180.90 .0108</td>
</tr>
<tr>
<td>b</td>
<td>173.898 MHz</td>
<td>167.11±.011 .0288±.0009</td>
<td>167.05 .0121</td>
</tr>
<tr>
<td></td>
<td>1.0 g</td>
<td>167.15 .0167</td>
<td>173.90 .0016</td>
</tr>
<tr>
<td></td>
<td></td>
<td>173.90 .0016</td>
<td>180.55 .0102</td>
</tr>
<tr>
<td></td>
<td></td>
<td>180.64 .0131</td>
<td></td>
</tr>
<tr>
<td>c</td>
<td>180.510 MHz</td>
<td>166.56±.025 .0115±.001</td>
<td>166.53 .0115</td>
</tr>
<tr>
<td></td>
<td>1.0 g</td>
<td>167.68±.03 .0139±.001</td>
<td>167.68 .0139</td>
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<tr>
<td></td>
<td></td>
<td>179.91±.04 .0081±.001</td>
<td>179.47 .0081</td>
</tr>
<tr>
<td></td>
<td></td>
<td>180.42±.025 .0141±.001</td>
<td>180.51 .0141</td>
</tr>
<tr>
<td></td>
<td></td>
<td>181.51±.05 .0061±.001</td>
<td>181.55 .0061</td>
</tr>
</tbody>
</table>

* The field amplitudes are determined from the magnitude of the DEMUR splittings.

* The theoretical amplitudes are normalized to give the observed total amplitude within each group of lines.

Table 2: Results of DEMUR on quartz. These three data sets show the effects of applying the rf power near (a) the lower pSR transition, (b) the mid frequency, and (c) the upper pSR transition. These three are identical to those appearing in figure 10.
<table>
<thead>
<tr>
<th>Data Set</th>
<th>Applied Field°</th>
<th>Experimental Results (MHz)</th>
<th>Theoretical Freq.</th>
<th>ampl. *</th>
</tr>
</thead>
<tbody>
<tr>
<td>none</td>
<td></td>
<td>167.23±.01 .0323±.001</td>
<td>166.47 .0130</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>180.51±.015 .0233±.001</td>
<td>168.08 .0117</td>
<td></td>
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<tr>
<td>a</td>
<td>180.279 MHz</td>
<td>166.44±.017 .0133±.0006</td>
<td>178.69 .0054</td>
<td></td>
</tr>
<tr>
<td></td>
<td>1.56 g</td>
<td>168.09±.02 .0114±.0006</td>
<td>180.28 .0131</td>
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</tr>
<tr>
<td></td>
<td></td>
<td>178.72±.045 .0050±.0006</td>
<td>181.92 .0079</td>
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</tr>
<tr>
<td>b</td>
<td>180.490 MHz</td>
<td>166.07±.03 .0117±.001</td>
<td>166.38 .0121</td>
<td></td>
</tr>
<tr>
<td></td>
<td>1.56 g</td>
<td>167.75±.024 .0143±.001</td>
<td>178.91 .0076</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>178.80±.048 .0071±.001</td>
<td>180.49 .0143</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>180.39±.025 .0138±.001</td>
<td>182.03 .0072</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>181.84±.038 .0061±.0006</td>
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</tr>
<tr>
<td>c</td>
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</tr>
<tr>
<td></td>
<td>1.56 g</td>
<td>167.49±.01 .0224±.0007</td>
<td>167.61 .0211</td>
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</tr>
<tr>
<td></td>
<td></td>
<td>179.63±.015 .0161±.0007</td>
<td>179.65 .0167</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>181.40±.025 .0101±.0007</td>
<td>181.52 .0095</td>
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<td></td>
<td></td>
<td>183.36 .0013</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

° The field amplitudes are determined from the magnitude of the DEMUR splittings.

* The theoretical amplitudes are normalized to give the observed total amplitude within each group of lines.

Table 3: Results of DEMUR on quartz. The three DEMUR spectra shown here were all taken with the rf power applied near the upper μSR transition frequency. The dramatic effects on the amplitudes of the DEMUR lines of slightly missing the resonance condition are evident.
the analytical "two-level" calculation, as well as the numerical results of the "three-level" calculation. In the "two-level" results the splitting is proportional to $\sqrt{z^2 + 1}$, where $z$ is the relative distance from the resonance condition as defined in the theory section.

The second major feature of this data appears in the intensities of the various DEMUR lines. In the first two data sets the intensities of the two DEMUR lines resulting from a splitting of the $\omega_{12}$ $\mu$SR line are comparable in magnitude. In the third data set these two lines appear with drastically different intensities. The "two-level" results predict that these two lines should occur with identical intensities only when the applied rf frequency is exactly on resonance (the "three-level" results predict that the ratio of the intensities of this pair of lines should be slightly different than 1 on resonance). Similar intensity effects can be observed in the third, fourth and fifth DEMUR lines (the triplet resulting from a splitting of the $\omega_{13}$ $\mu$SR transition).
Conclusions

The results of the DEMUR experiment presented in the previous section are found to be described in every detail by the theory of two strongly coupled spin 1/2 particles as presented in section 2. This theory and the corresponding experiment give a very complete picture of the coherent response of two coupled spins to the application of an intense rf magnetic field. Effects, such as the observation of the 3 DEMUR lines near the applied frequency, are a result of the coherent driving of the muonium center by the rf field. Features analogous to these results are not commonly observed in other double resonance techniques such as ENDOR, INDO, or ELDOR.

One feature of DEMUR that is usually not found in more conventional double resonance techniques is the intensity of the rf fields that were used. It is not that the DEMUR apparatus used was capable of extraordinary field magnitudes, but instead that the dc field used was of a very small magnitude when compared to those necessary for other techniques. For this DEMUR experiment, which was by no means optimized in this regard, the rf field had a
magnitude of 1.25 % that of the dc magnetic field. For comparison, typical NMR experiments, because they require dc magnetic fields generally 10 - 100 times larger than the field used here, have an rf to dc field magnitude ratio of some small fraction of one percent. A DEMUR experiment that was optimized in this regard could have rf field magnitudes larger than 10% of the dc field value.

This very intense rf field is responsible for the coherence effects observed in DEMUR. The response of a multi-level spin system to the presence of a weak oscillatory field is usually described in the context of transition and occupation probabilities. In that context the effects of the oscillatory magnetic field are described by a perturbation of these occupation probabilities. Coherence effects are characterized by these occupation probabilities being driven by the oscillatory field in a coherent fashion. This type of coherent behavior was clearly exhibited by the solutions for the \( a_\xi(t) \) presented in chapter 2. These coherence effects are reflected in the observed DEMUR spectrum, most dramatically, by the triplet of DEMUR lines appearing near the driving frequency.

An additional characteristic of these muonium-like defect centers that is necessary for the observation of coherence effects is the absence of any significant spin relaxation. Relaxation can destroy the phase coherence of the spin system. Frequently spin systems suitable to study
by other techniques have relaxation rates too large to ignore as was done for these muonium-like centers.

The final feature of this DEMUR experiment that is worth noting is that of concentration. DEMUR is in some respects EPR, detected via its effects on $\mu$SR. When considered in this way, DEMUR has allowed the detection of an EPR transition in a sample containing a single active spin. Because $\mu$SR allows only a single muon to be present in the sample at a given time, DEMUR necessarily works with a sample containing exactly one muonium center. Obviously $\mu$SR, and therefore DEMUR, require an ensemble of these muonic centers, one after the other; but for acceptable data at any given time only a single center is present in the sample.

In summary, DEMUR is a new double resonance technique that has characteristics which are unique when compared to other double resonance techniques. The most important of these characteristics are the rich variety of aspects of the coherent response of the spin system to the presence of an applied oscillatory magnetic field. These coherence effects are due to the strength of the rf field obtainable in DEMUR, and to the absence of appreciable relaxation in these muonium-like centers.
What Next?

DEMUR is a new and very interesting technique that has proven to be experimentally feasible. However, the DEMUR technique has not realized its full potential with this single quartz experiment. As previously described there are systems that are intrinsically capable of being probed more deeply by DEMUR that by conventional μSR; it is only when applied to these systems that the full power of DEMUR will be felt.

Currently, the most interesting and most promising of the systems which can be studied in greater depth by DEMUR are the various muonium-like centers in the elemental semiconductors. These centers have been observed in silicon, germanium, and more recently in diamond.[5,6] The observations in diamond are very recent and indicate that this is a difficult system to work with; for the present, considerations will therefore concentrate on silicon and germanium. In these materials two types of muonium-like centers are observed. The first observations made were of the 'normal' muonium center; with its isotropic spin Hamiltonian it is very similar to free muonium. The second center, the 'anomalous' muonium or Mu\(^\circ\) center, is more complex; observations of the Mu\(^\circ\) center are found to be described by an axially symmetric spin Hamiltonian, with
the symmetry axis along one of the 4 \text{<111>} crystallographic directions. The spin Hamiltonians for these centers are outlined in table 4.

The spin Hamiltonian parameters for the 'normal' muonium states in these materials have been well determined from \( \mu \mathrm{SR} \), as have the hyperfine parameters for the 'anomalous' muonium centers. The electronic \( g \) tensors for these \( \mathrm{Mu}^* \) centers, however, are not so easily measured. Because the hyperfine splitting in these centers are of a magnitude comparable to, or smaller than, the electronic Zeeman splitting, the intensities of several of the \( \mu \mathrm{SR} \) lines are very weak.[22] In this moderate field region the Zeeman splitting will tend to decouple the electron and the muon spins. The eigenstates of this system will consist predominantly of the high field basis functions, with the electron and the muon spins parallel to, or antiparallel to, the dc field direction. The transitions which are sensitive to the electronic Zeeman term in the Hamiltonian are those that involve an electron spin flip. It is precisely those transitions that are the most sensitive measure of the electronic \( g \) tensor that are unobservable in conventional \( \mu \mathrm{SR} \) due to their lack of significant intensity. DEMUR can be used to determine the form of the electronic Zeeman interaction, because these electronic transitions are driven with the rf field although they are unobservable in the \( \mu \mathrm{SR} \).
For normal Muonium (Mu):

\[ = g_p \mu_B \cdot S - g_p \mu_B \cdot I + A \cdot S \cdot I \]

For anomalous Muonium (Mu\(^*\)):

\[ = g_1 \mu_B \cdot S + (g_1 - g_1 \cdot \mu_B) \cdot H_z S_z - g_p \mu_B \cdot I + A_1 \cdot S \cdot I + (A_1 - A_1) \cdot S_z I_z \]

where the z direction is one of the 4 \((111)\) crystallographic directions.

<table>
<thead>
<tr>
<th>Material</th>
<th>Normal Mu A</th>
<th>Anomalous Mu(^*) A_1</th>
<th>Anomalous Mu(^*) A_1</th>
</tr>
</thead>
<tbody>
<tr>
<td>Vacuum</td>
<td>4463</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Diamond</td>
<td>3700</td>
<td>168.1</td>
<td>392.5</td>
</tr>
<tr>
<td>Silicon</td>
<td>2012</td>
<td>16.79</td>
<td>92.59</td>
</tr>
<tr>
<td>Germanium</td>
<td>2361</td>
<td>26.8</td>
<td>130.7</td>
</tr>
</tbody>
</table>

Table 4: Spin Hamiltonian parameters for the Mu and the Mu\(^*\) centers in the group IV semiconductors, from [5,6,23].
The apparatus necessary for a DEMUR experiment on these Mu\* centers, would be far more complex than the apparatus used for the quartz experiment. The Mu\* center is observed only at low temperatures (in silicon below 200 K); requiring that any DEMUR apparatus be capable of maintaining the sample at these cryogenic temperatures. The apparatus for the quartz experiment dissipated 10-20 W in the resonant circuit used to generate the rf magnetic field. Any DEMUR apparatus using this type of resonant circuit must be capable of dissipating this amount of power. In particular, if the sample and resonant circuit are both to be maintained at cryogenic temperatures, the refrigeration mechanism must be capable of handling this high heat load.

If sufficiently large rf magnetic fields are to be produced by a resonant circuit of the type shown in figure 8, the sample must not be conductive enough to introduce any resistive losses. The semiconducting samples necessary for observation of the Mu\* centers must be very pure.[24] Semiconducting materials have a temperature dependent resistivity. As the temperature is lowered, an intrinsic semiconductor will become more conductive (the carrier mobility is increasing) until a temperature sufficiently low to begin to freeze out the carriers is reached, after which the resistivity will increase exponentially.[25] To achieve large enough resistivities to eliminate eddy
currents, these semiconducting samples must be run at temperatures even lower than those required for the $\mu$SR. In the case of silicon sample temperatures below approximately 40 K are required. For germanium the temperature must be held below about 5 K.

An additional conductivity related problem may arise due to surface damage. Damage at the surfaces of these materials can form conducting layers, causing resistive losses that can reduce the Q factor of the resonant circuit. Chemical etches have proven to be useful in removing these damaged surface layers from silicon, and from germanium, and would be required for either of these materials.[26]

Figure 11 shows the details of a proposed DEMUR Dewar system for use with silicon or germanium samples. This apparatus is conceived of as a double Dewar with the outer (liquid nitrogen) Dewar wall made of some entirely non-magnetic metal such as brass, and the inner (liquid helium) wall being constructed of glass. By making this inner dewar of an electrically insulating material it is possible to isolate the sample thermally from the resonant circuit used to generate the rf magnetic field. This thermal isolation allows the large amounts of heat generated by the resonant circuit to be dissipated while maintaining the sample at liquid helium temperatures.
Figure 11: Details of the tail section of a proposed DEMUR Dewar for silicon and germanium samples.
The rf circuit used in this apparatus would be electrically identical to that used in the quartz experiment (see figure 8). Because of the difficulty associated with manipulating a variable capacitor at cryogenic temperatures, this resonant circuit would be designed to operate at a fixed frequency. A fixed frequency resonant secondary circuit can be made from a single piece of wire by utilizing the self capacitance of the coil as part of the resonant circuit. Such a resonant secondary coil could be encapsulated together with the primary driving coil, eliminating any microphonic effects. This coil assembly would then be mounted surrounding the inner Dewar and the sample.

The future for DEMUR appears bright. The apparatus described here will be capable of DEMUR measurements of the electronic Zeeman interactions in both silicon and germanium. A DEMUR measurement of this interaction would certainly aid in attempts to determine the microscopic structure of these defects. μSR itself is a young field with prospects for a long and an interesting future. As the μSR field grows and expands, new and different muonium-like centers are likely to be found; some of these will undoubtedly be suited to study by DEMUR.
Appendix A

In chapter 2 the theory of DEMUR was developed using one method; at that time an alternate approach was suggested. The alternate approach is to treat the oscillatory magnetic field by a transformation to a rotating reference frame.[16] Instead of the usual derivation of these equations, a procedure based on the eigenvector coefficients will be used.[16,27]

For simplicity consider a 2 level spin system with eigenstates $\psi_1$ and $\psi_2$, and energies $\hbar \omega_b/2$. This type of system is not equivalent to a DEMUR system, but it is a simple system possessing many of the same characteristics. The total time-dependent wave function for such a system may be written as:

$$\Psi(t) = A_1(t) \psi_1 + A_2(t) \psi_2 .$$

(26)

Note that in this expression the $e^{\pm i\omega_b t}$ terms are incorporated into the $A(t)$.

The application of an oscillatory magnetic field to this system may be described by the Hamiltonian:
\( \mathcal{H} = \gamma H_z \mathbb{J} e^{i \omega t} \) \hspace{1cm} (27)

Taking the eigenstates \( \psi_1 \) and \( \psi_2 \) to be quantized along the z axis and the \( H_z \) field to be in the x direction, the equations of motion may be written as:

\[
d/dt A_1 = iA_1 \omega_s/2 + iA_2 \frac{\gamma}{2\hbar} H_z e^{-i \omega t}, \\
d/dt A_2 = -iA_2 \omega_s/2 + iA_1 \frac{\gamma}{2\hbar} H_z e^{-i \omega t}. \hspace{1cm} (28)
\]

These equations may be written in a simpler form by transforming to a rotating reference frame. To accomplish this, the real variables \( r_1, r_2, \) and \( r_3 \) are constructed:

\[
r_1 = i r_2 = 2A_1^* A_2 e^{i \omega t},
\]

or,

\[
r_1 = A_1 A_2^* e^{i \omega t} + A_1^* A_2 e^{-i \omega t},
\]

\[
r_2 = iA_1 A_2^* e^{i \omega t} - iA_1^* A_2 e^{-i \omega t},
\]

and,

\[
r_3 = A_1 A_1^* - A_2 A_2^* \hspace{1cm} (29)
\]

Constructing the time derivatives of these new quantities:

\[
d/dt r_1 = d/dt(A_1) A_2^* e^{i \omega t} + A_1 d/dt(A_2^*) e^{i \omega t}
+ i \omega A_1 A_2^* e^{i \omega t} + c. c. \hspace{1cm} (30)
\]

and similarly for \( r_2 \) and \( r_3 \). Substituting equations 29 into these expressions yields the new equations of motion:
\[
\frac{d}{dt} r_1 = (\omega - \omega_0) r_2, \\
\frac{d}{dt} r_2 = - (\omega - \omega_0) r_1 + \frac{\gamma H}{\kappa} r_3, \\
\frac{d}{dt} r_3 = - \frac{\gamma H}{\kappa} r_2
\]
or,
\[
\frac{d}{dt} \mathbf{r} = \mathbf{r} \times \Omega
\]
where,
\[
\Omega = \frac{\gamma H}{\kappa} \mathbf{e}_1 + (\omega - \omega_0) \mathbf{e}_3.
\]
(31)

Solutions for these equations are well known from classical mechanics.

The incorporation of a relaxation mechanism into this treatment is simple. Both a longitudinal, \( T_1 \), and a transverse, \( T_2 \), relaxation process may be included by incorporating the phenomenological decay rates. After including these relaxation rates the equations of motion become:

\[
\frac{d}{dt} r_1 = (\omega - \omega_0) r_2 - r_1 / T_2, \\
\frac{d}{dt} r_2 = - (\omega - \omega_0) r - r_2 / T_2 + \frac{\gamma H}{\kappa} r_3, \\
\frac{d}{dt} r_3 = - \frac{\gamma H}{\kappa} r_2 - r_3 / T_1
\]
(32)

where \( T_\rho \) differs from \( T_1 \) because the transformation to the rotating reference frame changes the effective spin temperature.[28] Should a DEMUR system be found that requires the consideration of relaxation, solutions of equations of this form would be capable of doing so.
Appendix B

In chapter 2 a calculation of the DEMUR spectrum was performed. Inherent in these results is a knowledge of the initial spin state of the system. The muon with its spin initially oriented in a known direction is easily included in a description of the system. The electron spin will initially be oriented in a random direction, and is more difficult to include. By the use of a pair of random or distributed variables, the initial spin orientation of the electron may be included in a description of the system. These random variables are then carried through the complete calculation of the DEMUR spectrum, and in a final step the results are averaged to obtain a single spectrum.

Describing the initial state of the electron spin by a superposition of the high field electron spin-functions:

\[ |\chi(0)\rangle = \alpha |m_s=1/2\rangle + \beta |m_s=-1/2\rangle \]  \hspace{1cm} (33)

The spin operator in the direction of the initial electron spin orientation may be described by:
\[ S = \sin \Theta \cos \phi \, S_x + \sin \Theta \sin \phi \, S_y + \cos \Theta \, S_z, \]  
\[ \text{(34)} \]

where \( \Theta \) and \( \phi \) are standard spherical coordinates. The products \( \langle \chi(0) | S | \chi(0) \rangle \) will yield identities for the \( \alpha \) and \( \beta \) coefficients:

\[ \beta \sin \Theta \, e^{-i\phi} + \alpha \cos \Theta = \alpha \]
\[ \alpha \sin \Theta \, e^{i\phi} - \beta \cos \Theta = \beta \]
\[ \text{(35)} \]

One possible set of solutions for these equations are:

\[ \alpha = \sqrt{\frac{1}{2} (1 + \cos \Theta)} \]
\[ \beta = \sqrt{\frac{1}{2} (1 - \cos \Theta)} \, e^{i\phi} \]
\[ \text{(36)} \]

Averaging over \( \Theta \) and \( \phi \), \( \alpha \) and \( \beta \) possess the following properties:

\[ |\alpha|^2 + |\beta|^2 = 1, \quad \alpha^* \beta = \alpha \beta^* = 0, \quad |\alpha|^2 = |\beta|^2 = 1/2. \]
\[ \text{(37)} \]

The simplest set of values for \( \alpha \) and \( \beta \) are:

\[ \alpha = 1, \quad \beta = 0 \]

and \[ \alpha = 0, \quad \beta = 1 \]
\[ \text{(38)} \]

The use of this set of variables corresponds to performing the calculation with the initial electron spin parallel to and antiparallel to the field direction. Of course there are other sets of \( \alpha \) and \( \beta \) that satisfy the requirements of equations 37, but all would give identical final results.
As an example of the use of these distributed variables, consider the case of a muon with its spin initially oriented along the dc field direction. In terms of the high field basis states, $|m_5, m_I\rangle$, the initial state of the system is:

$$\Psi(t=0) = \alpha |1/2, 1/2\rangle + \beta |-1/2, 1/2\rangle. \quad (39)$$

The calculation of the DEMUR spectrum is performed twice, once for each set of the $\alpha$ and $\beta$ variables. The final spectrum is obtained by averaging the results of the two calculations.
References


18. This approximation is commonly taken in any type of resonance calculation. For example, consider a perturbative treatment of:

$$\langle k | \mathcal{H}'(t) | 1 \rangle = \langle k | \mathcal{H}'(0) | 1 \rangle e^{i\omega t},$$

Then to first order the time dependent coefficients take the form:

$$a_k^{(\omega)}(t) = a_k(0) \frac{1}{\omega - i \omega_k} \langle k | \mathcal{H}'(0) | 1 \rangle e^{i\omega t}.$$

19. This is the approximation that has been described in previous work reported in J. A. Brown, et. al., "Detection of EPR Transitions of Muonium in Quartz by Muon Spin Rotation", Phys. Rev. Lett. 43, 1751 (1979).

20. For a discussion of this type of fit see program 11-5 in P. R. Bevington, Data Reduction and Error Analysis for the Physical Sciences, McGraw-Hill 1969. The particular function the data was fit to was:

$$F(t) = a(1) + a(2) t + a(3) e^{-t/\lambda} \{ 1 + \sum_{i=1} \cos[2 \pi \frac{a(4i+3)}{t + a(4i+4)}],$$

where each element in the sum represents one line in the observed spectrum.

22. The intensity of the Mu* signals observed via standard μSR in these materials drops dramatically at low fields. This is presumably due to some broadening effects only important at low fields, such as a ligand nuclear hyperfine splitting that is quenched at larger fields.


