RICE UNIVERSITY

SPIN POLARIZED PHOTOEMISSION FROM GaAs
AND ITS APPLICATION TO ELECTRON POLARIMETRY

by

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

MASTER OF SCIENCE

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Abstract

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Spin polarized electron sources based on photoemission from GaAs are widely used. Several new advanced photocathodes are under development. In the present work we have investigated the use of cathodes based on epitaxially grown $p$-type GaAs(110) layers, which are 100 nm thick, as a source of electrons of known polarization. For photon radiation at 807 nm, we observed an electron spin polarization of $40.0 \pm 2.5 \%$. This is similar to earlier measurements made in other laboratories suggesting that indeed such epilayers can be used as a new derivative standard for calibrating electron polarimeters.
Dedicated to the memory of

Fabio Casartelli
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1. Introduction

An explosion of interest in polarized electron physics has occurred during the past two decades. Polarized electrons have been used to investigate many problems involving solids and surfaces, (including the magnetic properties of surfaces, thin films and multilayers), the fundamental properties of the electron, and of nuclei, (including protons and neutrons). The widespread application of polarized electron techniques has been enabled by the development of new polarized electron beam sources and spin analyzers. These developments have prompted widespread experimental and theoretical interest.

Polarized electron sources based on a variety of different physical principles have been developed, and although further improvements would be beneficial, robust sources already exist. Improved spin analyzers would also be welcomed, as these devices are notoriously inefficient and represent the weak link in any measurement involving polarized electrons.

One difficulty in using polarimeters is the lack of a practical standard for their calibration. Uncertainties associated with calibration make it difficult to compare results obtained in different laboratories. Currently, polarimeters based on Mott scattering have found the widest application because they provide good efficiencies and analyzing power, because they are relatively simple to implement experimentally, and because the physical basis of their operation is well understood. Mott polarimeters calibrated to better than 2 % are uncommon. The typical instrument may not be calibrated to
better than 10%. Developing a useful and practical method for calibrating these types of polarimeters would benefit the field immensely.

To calibrate electron polarimeters one must have a suitable source of electrons of known polarization. This source should be relatively easy to fabricate, be readily accessible, and be well characterized.

Photoemission from GaAs is widely used as a source of polarized electrons and we have participated in a study to determine if epitaxially grown, 100 nm thick \( p \)-type GaAs(110) layers can provide a usable source of electrons of known polarization. Measurements of the electron polarization obtained using such a source have been undertaken in several laboratories other than Rice University, including Stanford Linear Accelerator Center (SLAC), University of California at Irvine, University of Nebraska at Lincoln, and University of Münster, Federal Republic of Germany. These measurements all yield similar values, indicating that thin GaAs films can be used as a derivative standard calibration for electron polarimeters.

The present thesis discusses the character and choice of polarized electron sources (chapter 3) and explains in detail the theory behind spin polarized photoemission from GaAs (chapter 4). Recent advances are reviewed (chapter 5). After a brief description of the experimental apparatus (chapter 6), our results, as well as those from other participating laboratories, are presented (chapter 7).
2. Electron Spin

2.1 History

Electron spin, a purely quantum mechanical property, was first postulated in 1925 by Goudsmit and Uhlenbeck.\textsuperscript{1} Not until 1943 were the first definite polarization effects measured by Shull \textit{et al.}\textsuperscript{2} Over the past two decades, beginning with experiments done in 1967 by Loth and Eckstein, work involving polarization effects in scattering from solids is finally being pursued.\textsuperscript{3,4} This long delay between the initial discovery and the beginning of intense experimental efforts can be explained by the difficulty in measuring electron spin, contrasted with the straightforwardness of measuring the atomic magnetic moment.

Stern and Gerlach developed the first atomic magnetic moment filter in 1921 by passing an atomic beam through an inhomogenous magnetic field.\textsuperscript{5} For electron spin, there exists no analogous filter. A Stern-Gerlach analyzer cannot be used because of the Lorentz force acting on a charged particle in a nonzero magnetic field. The Lorentz force combines with the Heisenberg Uncertainty Principle to result in the loss of any information about the electron spin. Successful experimental techniques to determine electron spin have been either very inefficient or difficult to implement, thereby slowing research in this field. In the early 1970s as technology came to the aid in the form of advanced ultra high vacuum (UHV) polarized electron sources, improved polarimeters, readily available quality laser systems, and a greater understanding of surface physics, spin sensitive research began to bloom.
2.2 Formalization

In classical mechanics, for a particle to have angular momentum, it must rotate about an axis. In quantum mechanics, however, particles can possess an intrinsic angular momentum termed spin. Denoted by the quantum mechanical operator $S$, spin represents an internal degree of freedom which behaves as an angular momentum. Thus, the Cartesian components of spin satisfy the commutation relations

$$\left[S_i, S_j \right] = i\hbar \epsilon_{ijk} S_k.$$  \hspace{1cm} (1.1)

One can express the spin as $S = (\hbar/2)\sigma$, where the Pauli matrices $\sigma$, obtained from the above commutation relations and the condition that $\sigma_z$ is diagonal, are

$$\sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad \sigma_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}, \quad \sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}. \hspace{1cm} (1.2)$$

By using the eigenfunctions $|\alpha\rangle = \begin{pmatrix} 1 \\ 0 \end{pmatrix}$ and $|\beta\rangle = \begin{pmatrix} 0 \\ 1 \end{pmatrix}$ of $\sigma_z$ as a basis, a general spin state can be represented as

$$|\chi\rangle = a_1|\alpha\rangle + a_2|\beta\rangle = a_1\begin{pmatrix} 1 \\ 0 \end{pmatrix} + a_2\begin{pmatrix} 0 \\ 1 \end{pmatrix} = \begin{pmatrix} a_1 \\ a_2 \end{pmatrix}. \hspace{1cm} (1.3)$$

If $\chi$ is normalized to unity, so that $\langle \chi | \chi \rangle = 1$, one has

$$|a_1|^2 + |a_2|^2 = 1. \hspace{1cm} (1.4)$$

The probability of finding a particle in the ‘spin-up’ state $|\alpha\rangle$ (i.e. the probability that a measurement of $S_z$ will yield the value $+\hbar/2$) is given by $|a_1|^2$, while $|a_2|^2$ is the probability of finding the particle in the ‘spin-down’ state $|\beta\rangle$ (i.e. the probability that the value $-\hbar/2$ will be found upon measuring $S_z$).
Measuring the spin direction disturbs the spin state, making it impossible to determine all the spin components simultaneously.

The operator \( S^2 \) does commute with the components of \( S \). One can simultaneously measure its eigenvalue and that of any one of the components of the spin direction. Hence, the statement that "the spin is in the \( \hat{z} \) direction" means more precisely the following: The spin vector lies somewhere on a conical shell in such a way that the magnitude of its component in the \( \hat{z} \) direction is \( \hbar/2 \); the other two components are unknown. It is merely known that \( S_x^2 + S_y^2 + S_z^2 = 3\hbar^2/4 \) (the eigenvalue of \( S^2 \)) as in Figure 2.1.

![Figure 2.1. Spin in the \( \hat{z} \) direction.](image)

The polarization \( P \) is a vector quantity defined as the expectation value of the Pauli spin operator,

\[
P = \langle \chi | \sigma | \chi \rangle.
\]

The degree of polarization \( P_z \) with respect to a given \( \hat{z} \) axis is a scalar value lying between -1 and +1, given by
Therefore the polarization of a beam of electrons relative to the \( \hat{z} \) axis may be written as

\[
P_z = \langle \chi | \sigma_z | \chi \rangle = |a_1|^2 - |a_2|^2.
\]  

(1.6)

Therefore the polarization of a beam of electrons relative to the \( \hat{z} \) axis may be written as

\[
P_z = \frac{N_\uparrow - N_\downarrow}{N_\uparrow + N_\downarrow},
\]  

(1.7)

where \( N_\uparrow \) and \( N_\downarrow \) are the number of electrons with spin parallel and antiparallel to the \( \hat{z} \) axis, respectively.

As a consequence of the above discussion, one notes that a completely unpolarized beam of electrons is indistinguishable from a beam in which one half of the electrons are polarized in one direction and the other half are polarized in the opposite direction. Both beams are totally unpolarized and must be regarded as identical.
3. Evaluation of a Polarized Electron Source

To choose amongst the possible sources of spin polarized electrons, one must define the desired performance characteristics of the source for a particular application. In all sources, we assume that a higher electron spin polarization is preferable, but other considerations exist. This chapter introduces the criteria leading to our choice of source, a GaAs photoemitter.

3.1 Source Characteristics

3.1.1 Figure of Merit

A useful measure in characterizing polarized electron sources is the value \( P^2I \), where \( P \) is the beam polarization and \( I \) is the beam current. This figure of merit applies when counting statistics are the primary source of experimental uncertainty. If systematic errors exist, then achieving an adequately high polarization to mask the errors will be important. Arbitrarily increasing \( I \) to compensate for low polarization is not always possible or preferable. Space charge effects may limit the beam current. High beam currents can damage delicate sample surfaces.\(^7\) A high figure of merit, although quite useful in characterizing sources, is not always sufficient. In many cases a high polarization is necessary as well.

3.1.2 Energy Spread/Width/Distribution

In many experiments it is necessary to have an electron beam which is as close to monoenergetic as possible to permit energy-resolved measurements. In surface experiments, for example, a wide energy
distribution may cause more than one scattering mechanism to be involved in the results. A narrow energy spread could solve the problem, allowing the various mechanisms to be independently excited.

### 3.1.3 Source Current Lifetime and Stability

The source lifetime can be quite sensitive to operating conditions, depending on the source type. A longer source lifetime will mean less maintenance for the source. The current stability, in regards to both the short-term fluctuations during a measurement and the long-term drift, will factor into the confidence of the results.

### 3.1.4 Polarization Stability and Reversal

A very important source characteristic is the stability of the beam polarization. Spin-dependent effects are typically revealed by reversing the beam polarization and observing the asymmetry resulting in some measured parameter. If the polarization is susceptible to instabilities, the results could be confusing or rendered meaningless. Equally important is the ability to reverse the polarization without affecting other beam properties such as intensity, angle, or position. For example, the use of a Wien filter to reverse the polarization will disturb the beam trajectory unless great care is taken. Another consideration for the source is whether the electron polarization is produced transverse or longitudinal relative to the beam direction. The change from longitudinal to transverse polarization and vice versa can be readily achieved through the use of a 90° electrostatic deflector or Wien filter.\(^7-9\)
3.2 Choice of a Polarized Electron Source

In order to be useful a source must be stable and, for many applications, UHV compatible. There exist many different types of sources, each with its own strengths and weaknesses, based on a variety of physical processes. Examples, all of which have been reviewed extensively elsewhere, include (a) photoemission from GaAs or strained GaAs, to be discussed presently; (b) chemi-ionization of optically oriented He atoms in a flowing He afterglow\textsuperscript{10}, or surface Penning Ionization\textsuperscript{11}; (c) photoionization of alkali atoms, known as the Fano effect\textsuperscript{12,13}; (d) photoionization of polarized Li atoms\textsuperscript{14}; and (e) electron ejection from ferromagnetic materials\textsuperscript{15,16} A comparison among the different source types\textsuperscript{9} shows that sources based on photoemission from GaAs have many advantages. The GaAs source, when properly activated, provides a stable beam of monoenergetic electrons, with energy width $\leq 100$ meV\textsuperscript{7,17} and polarization typically ranging from 25 to 45%. The beam polarization can be reversed with no change in beam intensity or trajectory. Although sources based on chemi-ionization of He atoms display many of the same strengths, with other possible advantages, the main drawback is their incompatibility with UHV; typically, this source operates at pressures on the order of 100 mTorr. Thus GaAs photoemission sources have had the most widespread application, and an important parameter defining their performance is the quantum yield of the cathode.

3.3 Quantum Yield

The quantum yield or quantum efficiency of a device provides a measure of the efficiency of the photoemission process. The quantum yield $Y$, is the number of photoemitted electrons per incident photon. If corrections are made for the optical surface reflectance of the device, then the
quantum yield is equal to the number of photoemitted electrons per absorbed photon. Factors which influence the quantum yield include the choice of wavelength of light used for photoemission, surface characteristics, and the photoexcitation depth in the photocathode. Occasionally, a figure of merit $P^2Y$ is defined using the quantum yield rather than the current. This has the advantage of conveying information regarding the source efficiency. Unfortunately, this latter figure of merit is rarely used.
4. Theory of Spin Polarized Photoemission from Gallium Arsenide

The three-step model, first developed by W. E. Spicer, will be used to discuss photoemission from GaAs. The three steps are (1) the photoexcitation of the electrons, (2) their motion through the crystal to the surface, and (3) the emission of the electrons into the vacuum. Attention will be given to the still controversial physics of the activation layer on the GaAs surface critical to achieving an effective negative electron affinity. The theory of the band bending effect in the surface states of doped GaAs will also be examined.

4.1 Photoemission in Semiconductors vs. Metals

Photoexcitation occurs when an electron-hole pair is created by absorption of a photon. The general case of photoemission from a metallic crystal with a bare surface is exhibited in the energy bandstructure diagram in Figure 4.1. $E_F$ is the Fermi level which represents the highest occupied energy state at $T = 0\text{K}$. A photon of energy $h\nu$, when absorbed in the bulk, creates a hot electron that has the possibility of being emitted into the vacuum. The work function $\Phi$ is defined as the potential difference between the crystal’s Fermi level and the vacuum potential level. This is the minimum energy needed to remove an electron from the Fermi level and place it into the vacuum at rest. If $h\nu > \Phi$, then the photoexcited electron can be emitted with an energy equal to the difference between the absorbed energy and the work function. In practice, even if $h\nu > \Phi$, emission does not always occur due to the strong electron-electron collisions and electron-phonon
scattering causing energy loss while the photoexcited electron is in transport to the surface. If upon arriving at the surface its energy is still greater than the vacuum level, then there is a probability that the electron will escape into the vacuum.

Figure 4.1 'Three-step' photoemission process near a metallic surface. The workfunction, \( \Phi \), is the difference between the Fermi level, \( E_F \), and the vacuum level.

![Diagram of photoemission process](image)

Semiconductors, although usually described as insulators with a "narrow" band gap, are distinguishable from insulators because of the existence of photoexcitation.\(^{20}\) The energy gap is a set of states that is not accessible to electrons as shown in Figure 4.2.\(^{21}\) The electrons in the highest filled band, called the valence band, can be excited into the lowest empty band,
the conduction band, so long as the minimum amount of energy necessary to "cross" the band gap is supplied. In other words, when $h\nu > E_g$, where $E_g$ is the width of the energy band gap, photoexcitation can occur. This also explains the transparency of semiconductors to longer wavelengths in the infrared radiation spectrum. Photons with energy smaller than $E_g$ cannot be absorbed since the final state of the electron would lie in the forbidden band.

Figure 4.2 Energy level diagram for a semiconductor. The electron affinity, $\chi$, is the energy necessary to remove an electron from the conduction band minimum into the vacuum.

The low quantum efficiency of metals contrasted with that of semiconductors is caused by two fundamental differences between semiconductors and metals. First, the surface optical reflectance of metallic crystals is quite high relative to semiconductors. The number of photons
reflected from the surface of a semiconductor will be much lower than those that are reflected from a metallic surface given the same operating conditions. Second, the photoexcited electron has a higher chance of suffering an inelastic electron-electron collision in a metal while being transported to the surface for emission. A comparison of the scattering length of electrons, which is a measure of the strength of electron-electron collisions, with the optical penetration length, which indicates the distance from the surface at which the electron is typically photoexcited, reveals that the majority of photoexcited electrons in a metallic crystal will suffer inelastic collisions with other electrons. These collisions significantly reduce the electron’s energy, making photoemission nearly impossible. As will be discussed below, semiconductors have much longer scattering lengths than metals do. The conduction band is essentially empty except for a very few electrons that are thermally excited, and these lie low in the conduction band. The chance of an electron-electron collision is dramatically reduced as a result. These two reasons provide the motivation for using semiconductors in the photoemission process.

4.2 GaAs Energy Band Structure

For the majority of semiconductors, including GaAs, photoemission occurs primarily in the volume of the crystal with only a small contribution from the surface states. To analyze the potential photoexcitation channels, one must only consider the band structure for bulk GaAs.

A complete picture of the energy band structure for semiconductors begins with the notion of Bloch states. The Bloch theorem describes the form of the eigenfunctions of the Schrödinger equation in a periodic crystal lattice, such as GaAs. The eigenfunction, or wavefunction describes
the electrons in the crystal, and thus Bloch's theorem provides a form for
descrribing the motion of the electrons in the crystal. The electronic
wavefunction, or Bloch function, can be written as follows:

$$\psi_{k,\sigma}(r) = u_{k,\sigma}(r)e^{ikr},$$ (4.1)

where $u_{k,\sigma}(r)$ has the same periodicity as the crystal potential, $U(r)$, which is
seen by the electrons in the crystal.

A free electron has a simple quadratic relation between energy, $E$ and
momentum $k$, which is $E = (\hbar k)^2/(2m)$. The periodic crystal potential results
in a much more complex energy band structure for an electron in a lattice. To
determine the band structure for a semiconductor, the wavefunctions $\psi_{k,\sigma}(r)$
are assumed to be of the form given above in Equation 4.1. That is, the
wavefunctions must be Bloch functions. The Bloch functions can be
expressed in any chosen basis, such as the plane wave basis, for example. In a
model called the tight binding approximation or linear combination of
atomic orbitals (LCAO), the Bloch functions are expressed as combinations of
atomic orbitals centered at the lattice sites. Given a group of $N$ identical
atoms separated from each other, the discrete levels will be $N$-fold
degenerate. As the atoms are brought closer together to form the crystal, the
electronic states are perturbed by the neighboring atoms, resulting in the
formation of energy bands and gaps. The valence states are the most strongly
affected. The core electron state wavefunctions, which are localized near the
nucleus, do not overlap as much with the neighboring atoms. With $N$
atoms, the atomic levels will split into $N$ levels. Since typical values of $N$ for
a crystal are on the order of $10^{23}$ atoms, the energy levels are so close that they
form bands, as well as gaps, as shown in Figure 4.3.\textsuperscript{24} It is these bands and
gaps which compose the energy band structure of the semiconductor crystal.
The lattice structure of GaAs is known as the zinc-blende structure. The zinc-blende structure consists of two interpenetrating face-centered cubic (fcc) lattices, one containing Ga atoms at the lattice sites, the other As. The energy band structure for GaAs is shown in Figure 4.4. The energy band diagram maps out the energy levels in the first Brillouin zone of the reciprocal lattice space. For reference, the first Brillouin zone of a fcc lattice is shown in Figure 4.5.
Figure 4.4 Energy band structure of the first Brillouin zone of GaAs.

Figure 4.5 The first Brillouin zone of the reciprocal lattice (k-space).
The properties of the GaAs band structure can be determined by examining the constituent elements. The atomic structures of Ga and As are of the following form:

\[
\begin{align*}
\text{Ga} & \quad 1s^22s^22p^63s^23p^63d^{10}4s^24p^1 \\
\text{As} & \quad 1s^22s^22p^63s^23p^63d^{10}4s^24p^3
\end{align*}
\] (4.2)

The outer valence electrons have \textit{s}- and \textit{p}-type orbital symmetries. Although these are strictly the atomic orbitals for the uncombined elements, it turns out that the electrons in the valence and conduction bands of the semiconductor retain the \textit{s}- or \textit{p}-type symmetry properties.\(^{24}\) At the \(\Gamma\) point, the conduction band minimum and the valence band maximum have \textit{s} and \textit{p} symmetry, respectively. The conduction band minimum is doubly degenerate due to spin. The states are \(|s_{1/2}; m_s = +1/2\rangle\) and \(|s_{1/2}; m_s = -1/2\rangle\). The valence band maximum would be six-fold degenerate were it not for the spin-orbit splitting.

The spin-orbit effect results from the interaction of an electron's spin with its orbital angular momentum. Recall that an electron in a \(\textit{p}\)-state has an orbital angular momentum of \(\hbar\). The spin angular momentum of the electron, \(\pm \hbar/2\), couples with the orbital angular momentum to form the total angular momentum \(j\) which results in a perturbation that removes the degeneracy of the \(\textit{p}\)-levels. In the resulting valence band structure, the four \(j = 3/2\) states, \(|p_{3/2}; m_s = +3/2\rangle\), \(|p_{3/2}; m_s = +1/2\rangle\), \(|p_{3/2}; m_s = -1/2\rangle\), and \(|p_{3/2}; m_s = -3/2\rangle\) are split from the doubly degenerate \(j = 1/2\) states, \(|p_{1/2}; m_s = +1/2\rangle\) and \(|p_{1/2}; m_s = -1/2\rangle\), by \(\Delta = 0.34\ \text{eV}\).\(^{23}\) The resulting bands near the \(\Gamma\) point are shown in Figure 4.6.\(^{25}\) The spin-orbit splitting is crucial to the success of the spin polarized photoexcitation process.
At the $\Gamma$ point, the $p_{3/2}$ states are degenerate. Since $k = 0$, the crystal field does not reduce the symmetry of the point group. As one goes away from this point to areas where $k \neq 0$, the bands are no longer purely $p$-type, but rather a mix of different symmetries, leading to further splitting of the four degenerate states into two bands of two states each as shown in Figure 4.6. The two bands are referred to as the heavy hole and light hole bands according to their effective mass.\(^{22}\) The lifting of the degeneracy at the $\Gamma$ point of these two bands is the goal in the fabrication of more advanced polarized electron source structures which will be discussed in chapter 5.

4.3 Spin Polarized Photoexcitation

Transitions induced by photoemission can be divided into two types, direct and indirect, based on whether the process involves only the photon
absorption, or whether phonon coupling is required. In the direct process, no energy is transferred to or from the crystal lattice during the excitation process, leaving the initial and final crystal momentum unaltered, i.e. $\Delta k = 0$. In an indirect process, the crystal momentum is altered by one or more phonons being emitted or absorbed at the same time as the photon is absorbed, i.e. $k_{\text{final}} \neq k_{\text{initial}}$. This terminology is extended to a description of the bandgap in semiconductors. A direct bandgap occurs when the maximum of the valence band and minimum of the conduction band coincide at the same value of $k$. Thus, in a direct bandgap semiconductor, a direct transition is required to photoexcite with $\Delta k = 0$. The indirect bandgap semiconductor requires an indirect transition for an electron to go from $k_{\text{min}}$ to $k_{\text{max}}$. This cannot be effected with a direct transition.

Spin polarized photoemission occurs in GaAs in the direct bandgap region, where electron transitions occur between states of well-defined momentum. In typical atomic transitions between energy states, the Clebsch-Gordon coefficients are used to determine the selection rules. Due to the well-defined symmetries at the $\Gamma$ point, the Clebsch-Gordon coefficients are also used to determine selection rules for photoexcitation. By shining circularly polarized light at the minimum frequency to overcome the band gap, one can find the results of the transition by calculating the matrix elements.

Using the electric dipole approximation for the photoexcitation of electrons into the conduction band of GaAs, one finds the selection rules $\Delta l = \pm 1$ and $\Delta m_j = \pm 1$, from circularly polarized light, or $\Delta m_j = 0$, from linear polarized light. Consider the degenerate energy states at the valence band maximum ($\Gamma$ point), as in Figure 4.6 (b). Using positive helicity (right handed circular) polarized light (the photon’s angular momentum $\vec{\omega}$ parallel to its
direction of motion $\hat{k}$), the only two transitions which are allowed from the top of the valence band by the selection rules are

$$|p_{3/2}; m_j = -3/2\rangle \rightarrow |s_{3/2}; m_j = -1/2\rangle, \text{ and}$$

$$|p_{3/2}; m_j = -1/2\rangle \rightarrow |s_{3/2}; m_j = +1/2\rangle.$$  

(4.3, 4.4)

The net polarization of the electrons in the conduction band is determined by the transition probabilities for these transitions. The probabilities are calculated using the matrix element for the transition of the form $\langle \Psi_f | H_{\text{int}} | \Psi_i \rangle$, where $H_{\text{int}}$ represents the operator for circularly polarized radiation. Considering only electric dipole transitions and light propagating along the $\hat{z}$ axis, $H_{\text{int}}$ can be written as $(x \pm iy)$, or in terms of the spherical harmonics, as $Y_{m}^{\pm\ell}$, where the positive (+) and negative (-) terms correspond to right-handed and left-handed circular radiation, respectively.\(^5\) The wave functions of the crystal, the Bloch functions, can also be written in terms of the spherical harmonics. The expansion consists of a radial function, an angular function (the spherical harmonics, $Y_{m}^{\ell}(\theta, \phi)$), and a spin function ($|\alpha\rangle$ or $|\beta\rangle$). Table I summarizes the wave functions for the different states, ignoring the radial part which does not affect the transition probabilities in this case. The Clebsch-Gordan coefficients are the coefficients of the basis functions in the expansion.\(^27\)
The calculation of the probability of exciting an electron in the states listed in Equation 4.3 consists of simply calculating the matrix element

\[ \left| \langle s_{1/2}; m_j = -1/2 | Y_1^1 | p_{3/2}; m_j = -3/2 \rangle \right|^2 = 1. \]  

(4.5)

Comparing this to the result for the transition in Equation 4.4, we see that the relative strength of the transitions is 3:1. This results in a predominance of \(|\beta\rangle\)-state electrons in the conduction band leading to the following spin polarization (see Eqn. 1.7):

\[ P = \frac{3 - 1}{3 + 1} = 50\%. \]  

(4.6)

This is the theoretical maximum polarization that can be obtained. For a broad bandwidth light source, photoelectrons could be produced from the \(p_{1/2}\) states as well. The contribution from this transition would be such that the polarization would decrease to be zero; see Figure 4.6 (b).
In the process described above, the electron’s spin state does not actually change in the excitation. The resultant spin polarization is a consequence of the symmetry of the Bloch states, which follows from the symmetry of the crystal. The circularly polarized light cannot affect the spin states of the electrons. It merely imparts the energy and the orbital angular momentum necessary to transfer the electrons into the conduction band. The relative intensities of the possible transitions determine the resultant polarization. The relative intensity of a transition is determined by the number of electrons in that state (of a particular spin) which can be excited according to the selection rules. Thus, crystal symmetry influences the possible transitions and their relative intensities, resulting in a predominance of one spin state.

Note that the outcome is the same for reversal of the circularly polarized light except that the final net spin polarization is reversed. As a consequence of this fact, linear or unpolarized light, which can be described as an incoherent superposition of two oppositely polarized circular waves, results in no polarization.

4.4 Electron Transport

Spin polarized photoelectrons excited into the conduction band must work their way to the surface before being emitted into the vacuum. In typical GaAs, the electrons at the bottom of the conduction band would be about 4 eV below the vacuum level and could not escape from the crystal into the vacuum without further excitation. However, by doping the semiconductor and treating the surface with Cs and O$_2$, it is possible to lower the vacuum level at the surface below the energy of the conduction band minimum in the bulk. This condition is known as negative electron affinity
and will be described in the next section. In this section, we discuss the transport mechanism, the energy loss and depolarization that occur as the electron moves to the surface from inside the bulk.

### 4.4.1 Transport Mechanisms

Initially an electron which is photoexcited into the conduction band has a given amount of energy above the level of the conduction band minimum. These 'hot' (high-energy relative to thermal equilibrium) electrons can lose energy in primarily two ways. First, collisions with lattice phonons rapidly cause the electron to lose a small amount of energy. This process takes place on the order of $10^{-13}$ sec and results in an energy loss on the order of $30 \text{ meV}$.\(^{21,28}\) The second mechanism, which causes greater energy loss, is the inelastic scattering of hot electrons. A collision with another electron can easily cause the photoexcited electron to drop back into the valence band. If electron-electron scattering dominates, the overwhelming majority of the photoexcited electrons would remain trapped in the solid. Fortunately, the number of thermally excited electrons in the conduction band of GaAs is rather low ($10^{-6} \text{ cm}^{-3}$ for $10^{19} \text{ cm}^{-3} p$-doped GaAs at $300 \text{ K}$)\(^{21}\), decreasing the chance for electron-electron collisions. Thus, the primary collision type is phonon scattering. The majority of hot electrons are not inelastically scattered but instead undergo a few phonon collisions as they thermalize to the conduction band minimum.

Once the electrons have thermalized to the conduction band minimum, their diffusion can be halted by recombination with a hole. The minority carrier lifetime is a measure of the mean time before recombination occurs. The electron spends this amount of time diffusing toward the surface. The minority carrier lifetime is an intrinsic property of the material. The
lifetime, \( \tau \), can be expressed more meaningfully using the minority carrier diffusion length, \( L \). The two are simply related through the diffusion constant \( D \) as follows:\textsuperscript{29}

\[
L = (D \tau)^{1/2}.
\]  

For GaAs (with a doping concentration of \( \approx 3 \times 10^{19} \text{ cm}^{-3}, T = 300 \text{K} \)), the diffusion length at the \( \Gamma \) point is estimated at 1.2 \( \mu \text{m} \).\textsuperscript{29} The diffusion length limits the escape depth of the diffusing electrons.

The region from which electrons are photoexcited is determined by the optical absorption length, \( \alpha^{-1} \), (\( \alpha \) is the optical absorption coefficient). For GaAs, \( \alpha^{-1} \approx 1 \mu \text{m} \).\textsuperscript{21} Contrasting this length with the mean free path of hot electrons,\textsuperscript{30} which are on the order of 300 Å, and with the minority carrier diffusion length, which is on the order of 1 \( \mu \text{m} \), one notices that most of electrons reach the surface only if they are hot electrons excited near the surface, or if they thermalize to the conduction band minimum and diffuse to the surface, as described above.

Typically, the diffusion model can be described as the energetic thermal motion of swarms of independent particles.\textsuperscript{21} Since the minority carrier density is so low in \( p \)-type GaAs, the concept of swarms of electrons moving from regions of higher density to those of lower density is called into question. One could more appropriately model the electrons as performing a random walk, arriving by chance in any specific place. R. L. Bell has shown\textsuperscript{21} that the simple diffusion model still applies to the statistical average of the electron density derived from the random walk model. That is, electrons undergoing a random walk can be modeled using the diffusion equation, so long as one only seeks time averaged quantities after an initial short transient
The probability of an electron reaching the surface can be written in terms of the diffusion length and the absorption length as follows:\textsuperscript{31-33}

\[ P_s = \frac{1}{1 + (\alpha L)^{-1}}. \] (4.8)

This result represents the dependence of the transport on the diffusion model.

### 4.4.2 Depolarization Mechanisms

Electrons which reach the surface are not always spin polarized. Equation 4.8, which expresses the probability for photoexcited electrons to reach the surface, does not indicate whether the polarization persists or is lost during the transport process. There are two possibilities for depolarization during transport. The first occurs during thermalization of ‘hot’ electrons, and the second occurs during transport along the conduction band minimum. Depolarization mechanisms have been widely studied\textsuperscript{34,35} and the results suggest that the primary mechanism is determined by the exchange interaction between the electrons and holes.

The polarization of the excited electrons is reduced as a result of the competing effects of the minority carrier lifetime, \( \tau \), and the spin relaxation time, \( \tau_s \). The spin relaxation time incorporates the effects of the depolarization mechanisms into a mean time for the electron’s spin state to be lost. The resultant polarization will then be lowered from the theoretical polarization as follows:

\[ P_L = P_{th} \frac{\tau_s}{(\tau + \tau_s)}. \] (4.9)

This equation is derived from photoluminescence studies, and thus, it is not exactly representative of the polarization from photoemission. To arrive at
an equation for polarization from photoemission, the photoluminescence result must be adjusted to account for details of the photoemission process. Pierce\(^7\) derives the new polarization equation taking into account the finite penetration depth of the light and the diffusion of the excited electrons to the surface, arriving at an equation which simplifies to

\[
P_e = \frac{[\alpha + (D\tau)^{-1/2}] P_{th}}{[\alpha + (DT)^{-1/2}]}\]

(4.10)

where \(T = \tau_s\tau/(\tau_s + \tau)\). The photoemission polarization is higher than the luminescence polarization, \((P_e > P_L)\), because the emitted electrons are less depolarized than the electrons which spend their entire lifetime \(\tau\) in the solid.

One can increase the polarization of the emitted electrons by changing the spin relaxation time, or the effective minority carrier lifetime. The spin relaxation time depends on the doping concentration in the crystal and the temperature, while the minority carrier lifetime depends only weakly on these parameters.\(^7,36,37\) Lowering the temperature increases \(\tau_s\), leading to higher resultant polarization. The drawback to this method is the inconvenience of working with cryogenics in UHV. Another way to increase the polarization is to vary the thickness of the emission layer of GaAs in order to decrease the effective value of \(\tau\). Here the downside is the drop in intensity of the emitted beam. Studies\(^36\) have been performed to show the effect of photoemitter thickness. A compromise between quantum yield and polarization must be reached in order to optimize the figure of merit.

Further depolarization can occur during transport through the surface region and through the activation layer on the surface. These possibilities will be examined in the following sections.
4.5 Negative Electron Affinity and Electron Escape

Once electrons diffuse to the surface, they must have enough energy to overcome the vacuum potential barrier. The energy difference between the conduction band minimum and the vacuum level is the electron affinity, $\chi$, as defined in Figure 4.2. Only electrons with energy $\chi$ above the conduction band minimum can be emitted. Generally, the lower the electron affinity, the higher the probability of emission.

A special case occurs when the electron affinity is made negative. The primary transport mechanism of emitted electrons becomes minority carrier diffusion, rather than hot electron transport. This is due to the fact that the electrons which diffuse along the conduction band minimum now have the required energy to be emitted. The emission probability increases dramatically due to the number of electrons which thermalize to the conduction band minimum compared with those that remain as hot electrons as they travel to the surface. Figure 4.7 illustrates the difference between the energy distribution curves of a conventional (positive electron affinity) emitter and a negative electron affinity (NEA) emitter.30

4.5.1 Band Bending

As previously stated, a necessary condition for electron escape is that the energy of the electrons be higher than the vacuum level. For GaAs, the electron affinity $\chi$ is usually on the order of 4 eV. By coating the surface with a (Cs, O) activation layer, the electron affinity is greatly reduced to values of about 0.5 eV. Additional lowering of $\chi$ results from band bending in the surface region. The combination of these two effects produces the NEA condition.
The band bending in the surface region results from the formation of surface states whose properties are different than those in the bulk. Surface states differ from bulk states in that they are not part of the eigenvalue spectrum of the Hamiltonian of a perfect lattice. Their energies lie outside of the allowed energy range of the bulk crystal lattice band structure. Their wavefunctions are peaked in the surface region. These properties enable the formation of a space charge layer on the surface. 38

The Fermi level of a semiconductor gives a measure of the balance between electrons in the conduction band and holes in the valence band. 39 In heavily doped p-type GaAs, there is an abundance of acceptors and the Fermi level is just above the valence band. At the surface, the excess positive charge resulting from doping will occupy the surface states, creating a surface space charge layer. The space charge layer changes the net charge residing at the surface. The surface states, now containing an excess of positive charge, cause the formation of an depletion region near the surface. With excess positive charges in the surface states (or space charge layer), the net charge in the
depletion region is negative. The result is to 'pin' the Fermi level at the surface. The 'pinning' of the surface potential is illustrated in Figure 4.8. Thus, the Fermi level at the surface does not change as much as it does in the bulk.

![Diagram of band structure](image)

Figure 4.8 (a) Undoped semiconductor band scheme: Fermi level lies midway between the valence and the conduction bands. (b) Band scheme for doped (p-type) semiconductor showing 'pinning' of the Fermi level. The migration of positive charges into the surface states causes a depletion region near the surface resulting in band bending.

The continuity of the band structure manifests itself in the band bending effect shown in Figure 4.8. This is crucial for the negative electron affinity situation. The difference between the bottom of the conduction band in the bulk of the semiconductor and the vacuum potential level can be
defined as an 'effective' electron affinity. The true electron affinity $\chi$ is the difference between the bottom of the conduction band at the surface and the vacuum level. Using the values defined in Figure 4.9, the effective electron affinity can be written as follows:\textsuperscript{21}

$$\chi_{\text{eff}} = \chi - V_{BB},$$ \hspace{1cm} (4.11)

and is shown to be negative in Figure 4.9. In all cases herein, the discussion of NEA implies the effective, and not the true, NEA.

![Diagram of Conduction Band and Depletion Region](image)

Figure 4.9 $p$-type semiconductor showing band bending region caused by the formation of a space charge layer in the surface states. $\chi_{\text{eff}}$, the effective electron affinity, is negative as shown.

The true electron affinity becomes unimportant when the width of the band bending region is small compared to the electron escape length, or diffusion length. The width of this region is determined by the doping
concentration. Higher doping concentrations decrease the width of the band bending region as shown in the following equation:

\[ W = \left(2\varepsilon V_{bb}/eN_A\right)^{1/2} \]  

(4.12)

where \( W \) is the width in cm, \( \varepsilon \) is the dielectric constant (in F/cm), \( V_{bb} \) is the amount of band bending (in V), and \( N_A \) is the acceptor doping concentration (in cm\(^{-3}\)). The band bending distance should be smaller than the optical absorption depth \( \alpha^{-1} \) and the diffusion length \( L \). The width should also be such that the diffusing electrons, which now become hot electrons in the band bending region, do not suffer too many phonon scatterings resulting in energies that are lower than the true electron affinity \( \chi \), which is positive, as shown in Figures 4.8 and 4.9. The scattered electrons lose energy, become trapped in the surface layers, and recombine eventually with holes in the bulk. However, higher dopant concentrations are necessary to decrease the width of the depletion region. These higher doping levels in turn cause more energy loss collisions for the escaping electrons. The doping level, then, must be optimized to provide a balance between these competing factors, diffusion length and escape probability. Doping levels for NEA GaAs photocathodes are on the order of \( 10^{18} \) cm\(^{-3}\). Typical band bending region widths are on the order of 100 Å. The amount by which the bands are bent, \( V_{bb} \) in Figure 4.9, is typically 0.5 eV or more.\(^{21,40} \) This amount alone is not sufficient to obtain the NEA condition.

4.5.2 The (Cs, O) Activation Layer

The full NEA condition necessitates additional lowering of the vacuum level to reduce \( \chi \) below the level achieved by band bending alone. The basic principle involves depositing alkali metals, as well as some
electronegative species, such as oxygen or flourine, onto the surface of the semiconductor. The details of the physics of the activation layer (so named because it ‘activates’ the photocathode by enabling the NEA condition) is the least understood of all the areas discussed, yet it is crucial to the realization of practical sources. This section will present the basic ideas of the proposed theories with the understanding that it is not clear which, if any, are correct. The three basic theories\textsuperscript{41} are the dipole model, the heterojunction model, and the cluster model.

The NEA activation employs the yo-yo method. Cs atoms are generated from Cs dispensers and oxygen is leaked into the chamber via a leak valve. First, Cs is adsorbed on the surface until saturation occurs. Next, oxygen is adsorbed. Photocurrent is monitored during the process to determine when the best condition is achieved. The cesiation and oxidation processes are then repeated several times until the photocurrent does not increase anymore.

The dipole model\textsuperscript{30,42-44} views the cesium and oxygen atoms arranging on the surface in layers of electrical dipoles, resulting in a potential barrier. This model is consistent with the behavior of the GaAs-Cs system, but is unable to explain the decrease of photocurrent that occurs (without a simultaneous increase in the work function)\textsuperscript{45} when a thick layer (4 - 5 ML) of cesium and oxygen is deposited over it.

The heterojunction model\textsuperscript{21,46-48} explains the potential barrier at the surface as a heterojunction between the surface oxide layer and the substrate. Strong support for this model comes from studies\textsuperscript{46,49} suggesting that the photoemission threshold of III-V semiconductors is no longer determined by the band gap when activation is performed. The heterojunction model is consistent with the existence of the interfacial barrier, and has the advantage
that it predicts the change in the nature of the photoemission threshold which the dipole model fails to predict. But, the heterojunction model does not account for the behavior of thick activation layers (as discussed above)\textsuperscript{42,50}, nor does it provide for a minimum work function for thin cesium coatings (< 1 ML).

The final and most recently proposed model to be described is the cluster model.\textsuperscript{45,51} This model relies on the formation of cesium and oxygen clusters at the surface. The clusters would be highly repulsive for photoelectrons, and thus spatially confine the electrons. This would in turn cause an increase in their kinetic energy, thereby effectively lowering the work function. The primary drawback of this model is its failure to account for the interfacial barrier.
5. Advanced GaAs-type Photocathode Structures

In ordinary p-type NEA GaAs photocathodes, the maximum theoretical polarization is limited to 50 % due to the degeneracy of the light-hole and heavy-hole bands at the Γ point, as illustrated in Figure 4.6. Advances in photoemission structures have come in the form of devices fabricated to break the degeneracy of the two bands. If the degeneracy is split, then single band excitation becomes possible, leading to a theoretical electron spin polarization of 100 %. In this section, advances based on the method of strained GaAs are reviewed.

5.1 Strained GaAs Structures

Maruyama, et al.,\textsuperscript{52} observed the first strain-enhanced electron spin polarization in 1991. The structure developed consisted of a thin layer of InGaAs epitaxially grown on a GaAs substrate. The mechanism used to remove the degeneracy of the light hole (lh) and heavy hole (hh) bands is strain-induced changes of the band structure. The strain is produced by lattice mismatch between the epilayer and the substrate as shown in Figure 5.1.\textsuperscript{24} Strain-induced changes of band structure have been extensively studied.\textsuperscript{24,53-55} In some cases, GaAs has been used as the substrate with a In\textsubscript{x}Ga\textsubscript{1-x}As as the epilayer\textsuperscript{52,56}, and in other cases GaAs is used as the epilayer on a GaAs\textsubscript{1-x}P\textsubscript{x} substrate.\textsuperscript{57-59} One more recent study included (Al\textsubscript{y}Ga\textsubscript{1-y})\textsubscript{x}In\textsubscript{1-x}As layers on a GaAs substrate.\textsuperscript{60} Photoelectron spin polarization of over 80 % is readily achieved in almost all cases.

Applying a biaxial compressive strain in the plane of the interface produces a tensile strain along the growth direction. This causes the hh-band
to move up in energy and the lh-band to move down in energy with respect to the degenerate energy level that would exist without the strain. A biaxial compressive strain in the interface plane can be affected if the substrate has a smaller lattice constant than that of the epilayer.

![Figure 5.1 Lattice strains. (a) Larger lattice constant layers formed on smaller lattice constant substrate results in biaxial compression along the interface plane, and tensile strain in the growth direction. (b) Smaller lattice constant layers formed on larger lattice constant substrate resulting in tensile strain along the interface plane, and compressive strain in the growth direction.](image)

The band gap energy of the substrate must be larger than that of the strained epilayer. Otherwise, photoemission occurs not only from the epilayer, but also from the substrate. This also can be accomplished if the lattice constant of the substrate is smaller than that of the epilayer.

The strain must be large enough to clearly separate the hh- and lh-bands from each other, enabling single band excitation. If the strain is too large or if the epilayer becomes too thick, the strain is relieved by dislocations.
The following equation gives the energy splitting $\delta_s$ as a function of the properties of the lattice mismatch:

$$\delta_s = -2u\frac{C_{11}+C_{12}}{C_{11}}\varepsilon$$

(5.1)

where $C_{ij}$ are elastic-stiffness constants, $u$ is the deformation potential, and $\varepsilon = \Delta a/a_0$ is the lattice mismatch ($\Delta a$ is the difference between the unstrained lattice constants of the epilayer $a$ and the substrate $a_0$).

<table>
<thead>
<tr>
<th>Sample</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
</tr>
</thead>
<tbody>
<tr>
<td>$x$</td>
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<td>0.243</td>
<td>0.279</td>
<td>0.244</td>
<td>0.238</td>
</tr>
<tr>
<td>GaAs thickness ($\mu$m)</td>
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<td>0.15</td>
<td>0.11</td>
<td>0.20</td>
<td>0.30</td>
</tr>
<tr>
<td>Lattice mismatch (%)</td>
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<td>0.88</td>
<td>1.01</td>
<td>0.88</td>
<td>0.86</td>
</tr>
<tr>
<td>Measured strain (%)</td>
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<td>85</td>
<td>87</td>
<td>81</td>
<td>61</td>
</tr>
<tr>
<td>$E_{C_{HH}}^c$ (eV)</td>
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<td>1.47</td>
<td>1.48</td>
<td>1.47</td>
<td>1.46</td>
</tr>
<tr>
<td>$E_{C_{LH}}^c$ (eV)</td>
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<td>1.52</td>
<td>1.54</td>
<td>1.52</td>
<td>1.52</td>
</tr>
<tr>
<td>$h_1^{eq}$ (Å)</td>
<td>133</td>
<td>110</td>
<td>92</td>
<td>110</td>
<td>113</td>
</tr>
</tbody>
</table>

Table II. Data from strained GaAs samples.

The characteristics for strained GaAs samples are included in Table II.\textsuperscript{58} The data represents five different samples grown by the Spire Corporation, Bedford, Massachusetts. GaAs was grown onto a GaAs$_{1-x}$P$_x$ substrate. The table summarizes the phosphorus fraction, chosen to assure the proper amount of energy band splitting, the GaAs epilayer thickness, the lattice mismatch, the measured strain, expressed as a percent of the strain for complete lattice accommodation, the calculated gap energies of the heavy- and light-hole bands relative to the conduction band, and the calculated equilibrium critical thickness ($h_1^{eq}$) for five different samples. Surpassing the critical thickness of the epilayer leads to relaxation of the strain during the
heat cleaning process. The measured electron spin polarization and quantum efficiency as a function of excitation photon energy are shown in Figure 5.2. The solid arrow in the figure indicates the heavy-hole band gap energy and the dashed arrow indicates the substrates band gap energy. For excitation energies greater than about 1.7 meV, both the epilayer and the substrate contribute to the photoemission process resulting in a higher quantum efficiency, but lower overall polarization.

![Graph showing polarization and quantum efficiency as a function of photon energy]

**Figure 5.2** Results from strained GaAs. High polarization is consistent with low QE. The solid arrow indicates the energy at which the lh-band excitation is included. The broken arrow indicates the energy at which the spin-orbit split $p_{3/2}$ band is included.

The drawback in using the strained photocathodes is that the high polarization is achieved at the expense of quantum efficiency, as shown in Figure 5.2. For bulk GaAs, quantum yields of up to 10% are not atypical, but
for strained GaAs with polarization around 80 %, the yield is on the order of 0.1 to 0.3 %. This limitation exists because photoexcitation occurs only from the heavy-hole band which is a low density state near the band edge. Also, the critical thickness of the strained layer limits the photoexcitation region. Absorption length and diffusion length are no longer the dominant factors in determining the quantum yield as they were for standard GaAs photoemission.

5.2 Strained GaAs Photocathode with Distributed Bragg Reflector

A novel solution has been recently developed to overcome the problem of low quantum efficiency without lowering the electron spin polarization: A new type of photocathode with a distributed Bragg reflector (DBR). The cathode is similar to the strained GaAs/GaAsP structures just discussed with the addition of a quarter-wave DBR inserted in between the GaAs substrate and the GaAsP buffer layer, as in Figure 5.3. The DBR forms

![Diagram](image)

**Figure 5.3** Strained GaAs on GaAsP substrate layer with DBR inserted. The helicity of the excitation radiation is shown, as well as the relative directions of the angular and linear momentum, \( \hat{\omega} \) and \( \hat{k} \), respectively. The GaAsP is transparent to this radiation because its band gap is larger than the band gap for GaAs.
a Fabry-Perot cavity which increases the absorption of the light by multiple reflections between two parallel mirrors. The DBR is an alternating sequence of quarter-wave layers with different indices of refraction. By choosing the thickness for the layers as $\lambda_b/(4n_h)$ and $\lambda_b/(4n_l)$, where $n_h$ and $n_l$ are the high- and low-reflectivity layers, respectively, the reflectivity of the cavity will be quite high for the region around the wavelength $\lambda_b$. The design used in by Saka, et al, contained 30 pairs of Al$_{0.1}$Ga$_{0.9}$As and Al$_{0.6}$Ga$_{0.4}$As layers with $n_h = 3.52$ and $n_l = 3.20$, respectively. The $\lambda_b$ is 860 nm, the value expected for maximum polarization. From these data the bandwidth $\Delta\lambda_b$ can be calculated. This bandwidth is defined as wavelengths with reflectivity over 90% and is given by

$$\Delta\lambda_b = \frac{4\lambda_b}{\pi} \sin^{-1}\left(\frac{n_h - n_l}{n_h + n_l}\right).$$

(5.2)

This results in $\Delta\lambda_b \sim 52$nm.

The direction of spin polarization of the electrons excited by the reflected light is a consideration in this device. The photoexcited electron will have a spin state determined by the direction of the photon’s angular momentum $\vec{\omega}$. Upon reflection, $\vec{\omega}$ does not change, even though the linear momentum of the photon $\vec{k}$ does reverse directions (see Figure 5.3). Electrons of the same spin state will be excited by the incident or the reflected light, since $\vec{\omega}$ is the same for both. This is a key advantage of the DBR design.

The results obtained by Saka, et al, are shown in Figure 5.4. The three peaks in the quantum efficiency curve correspond to the resonant absorption of the incident light in the Fabry-Perot cavity. The resonance condition, which determines the resonant wavelength $\lambda_R$, is

$$2nL = m\lambda_R,$$

(5.3)
where $n$ is the refractive index, $L$ is the cavity length, and $m$ is an integer.

Figure 5.4 On the left, the polarization vs. wavelength is plotted. The solid circles represent the present structure (strained GaAs with DBR), while the open circles represent the structure without the DBR inserted (strained GaAs only). On the right, the QE vs. wavelength is plotted. The arrows mark the resonant wavelength conditions of the Fabry-Perot cavity. The solid line indicates the QE enhancement with the DBR inserted over that without the DBR.

The enhancement of the quantum efficiency with the DBR is plotted as a solid line. Figure 5.4 shows the polarization of the photocathode with the DBR compared with that of the photocathode without the DBR. The final quantum efficiency is about an order of magnitude larger for ~70% polarization. The research in the area of advanced photocathodes is quite promising.
6. Experimental Methods and Apparatus

In this section, the apparatus used in the present work will be introduced. A general description of the complete UHV system is first given, followed by introductions to the primary apparatus used in the experiment. The experimental apparatus has been described in detail elsewhere, and will be reviewed here for the sake of clarification of upgrades and additions.\textsuperscript{9,62,63}

The complete apparatus is shown in Figure 6.1.\textsuperscript{62} The chamber is divided into two sections, the main chamber and the spin polarized electron source (SPES) chamber. The main chamber consists of an upper region which houses the surface preparation and characterization equipment. The lower chamber is used for data acquisition. The SPES chamber houses the GaAs source and electron transport optics to control the electron beam dynamics. The two sections can be isolated from each other by use of an inline gate-valve\textsuperscript{64}, allowing the exposure of one section to atmosphere while the other remains under vacuum. The main chamber is pumped by two ion pumps with pumping speeds of 400 L/s and 500 L/s respectively. The pumps are connected to the main chamber via a manifold and backed up by a turbomolecular pump\textsuperscript{65} (TMP) and a mechanical roughing pump. The TMP and mechanical pump also provide pumping for the differential seals of the manipulator and Mott rotaries. The SPES chamber is pumped by a smaller 50 L/s ion pump. The normal base pressure is \( \sim 2 \times 10^{-11} \) Torr, measured by a nude ionization gauge in the main chamber. A residual gas analyzer\textsuperscript{66} (RGA) with quadrupole mass spectrometer can measure both the total and constituent partial pressures. The RGA provides background gas
Figure 6.1 Schematic of the experimental apparatus. Only the major parts are illustrated in the diagram. Both the sample and the Mott polarimeter can rotate around the vertical axis independently.
composition, purity information for gases deliberately introduced into the chamber, and leak checking services, by detecting He gas sprayed around the suspected areas.

6.1 Spin Polarized Electron Source (SPES) Chamber

The SPES chamber houses the p-type GaAs photocathode on a retractable support "arm." Heating coils and a water cooling system, which are used to control the source temperature during the cleaning process, are integrated into the support platform. The cesium dispenser is attached above the sample. A sapphire-sealed variable leak valve is used to leak O₂ into the SPES chamber to activate the photocathode surface.

Above the SPES chamber rests the laser optics setup which is responsible for the photoexcitation process. The anchored aluminum stand supports a AlGaAs semiconductor laser which operates at 807 nm wavelength. The typical operating features of this diode laser are a low threshold current (~25 mA) and an operating current of 68 mA for light power output of 15 mW. The rapidly divergent laser beam is collimated by a "fish eye" lens and focused through a telescope to reduce the spot size to less than 1 mm. The collimated beam is circularly polarized by passing it through a linear polarizer and a Mica quarter wave retarder. The final focusing of the beam onto the photocathode is achieved by passing the beam through a 15 cm focal length convex lens, and once inside the SPES chamber, passing it through a 3 mm aperture. The laser beam is directed onto the GaAs surface by micron adjusting screws attached to the aforementioned optics.

The helicity of the circularly polarized laser light can be reversed by merely rotating the quarter wave plate by 90°. Provided that the beam is
directed straight through the center of the retarder, the resulting intensity variation will be negligible.

The GaAs crystals supplied by SLAC are approximately 0.4 mm thick and cleaved to 0.49 cm$^2$ to fit the sample holder. The doping level is $\sim 5 \times 10^{18}$ Be cm$^{-3}$, a level chosen to insure a good band bending in the surface region, while minimizing depolarizing effects. The crystal is chemically etched to remove all carbon traces, followed by an extensive cleaning procedure before installation.$^{71}$ After installation, the crystal is treated by heating in vacuum to $\sim 600$ °C for several minutes. Activation consists of depositing cesium and oxygen onto the crystal surface using a modified "yo-yo method." The procedure consists of constantly monitoring the white light photoemission current while depositing Cs to increase photocurrent. Once the current peaks and decreases to $2/3$ its peak value, oxygen is admitted into the chamber. The oxygen is turned off when the current has again peaked and decreased to $2/3$ of the new peak value. This cycle is repeated until a maximum photocurrent has been obtained.

In the operation mode, the spin polarized photoelectrons are extracted by a 240 eV potential before passing through an aperture and a 90° spherical deflector which changes the direction of polarization of the electron beam from longitudinal to transverse. Transmission of $\sim 45$ % is routinely achieved with a beam width $\sim 2$ mm.$^9$

### 6.2 Hemispherical Energy Analyzer

Energy analysis of the electron beam is achieved using a concentric hemispherical analyzer shown in Figure 6.2.$^{62}$ The analyzer is then coupled to a compact Mott polarimeter. In the energy analyzer, an incident electron
beam is deflected electrostatically by a pair of concentric spherical electrodes. The pass energy, \( E = eV_0 \), is determined by

\[
V = V_0 \left( \frac{R_2}{R_1} - \frac{R_1}{R_2} \right)
\]

(6.1)

where \( V \) is the potential difference across the two hemispheres with radii \( R_1 \) and \( R_2 \), respectively. The energy resolution is given by

\[
\frac{\Delta E}{E} = \frac{\omega}{2R_0} + \frac{\alpha^2}{2}
\]

(6.2)

where \( \omega \) is the width of the entrance and exit apertures, \( \alpha \) is the incident angle, and \( R_0 \) the mean radius of the spheres. The dimensions of the analyzer used in this work are \( R_1 = 37.5 \text{ mm} \), \( R_2 = 62.5 \text{ mm} \), and \( R_0 = 50.0 \text{ mm} \). The entrance and exit apertures have diameters of \( \omega = 3.0 \text{ mm} \). The analyzer is designed to work in the constant absolute-resolution mode with pass

Figure 6.2 Hemispherical Energy Analyzer
6.3 Compact Mott Polarimeter

Accurately measuring the electron spin polarization is the primary goal for this experiment. Mott polarimeters are widely used for this purpose because they provide good efficiencies and analyzing power, and are simple to use and fully UHV compatible. This laboratory has been at the forefront in developing a number of compact Mott polarimeters in recent years.⁷²,⁷³

The physical basis of the Mott polarimeter operation is well understood and can be accurately described using a classical picture of electron scattering.⁷⁴,⁷⁵ Mott polarimeters utilize the left-right scattering asymmetry that results when polarized electrons are scattered at large angles by high-Z atoms. The polarization can be determined from the measured asymmetry provided that the instrumental asymmetry is known.

The high-energy electrons must be incident with a small impact parameter to scatter at large angles from high-Z atoms. A reasonable model of this is high-energy electron scattering off of a bare nucleus of charge Ze. The electron, moving through an electric field produced by the nucleus, produces and experiences a magnetic field in the electron rest frame. This field is proportional to the electron orbital angular momentum \(L = mr \times \mathbf{v}\) as follows:

\[
B = \frac{Ze}{mcr^3}L
\]  

(6.3)

The electron magnetic moment \(\mu_s\) interacts with this magnetic field producing an additional term in the scattering potential,

\[
V_{so} = -\mu_s \cdot B = \frac{Ze^2}{2m^2c^2r^3} L \cdot S.
\]  

(6.4)
The presence of the spin-orbit term $V_{so}$ introduces a dependence in the scattering cross-section $\sigma(\theta)$ which may be written as

$$\sigma(\theta) = I(\theta)[1+S(\theta)P \cdot \hat{n}]$$

(6.5)

where $S(\theta)$ is the asymmetry (Shermann) function, $I(\theta)$ the spin-averaged scattered intensity, and $P$ the incident electron polarization. The unit vector $\hat{n}$ is normal to the scattering plane. The scattering will result in a left-right asymmetry $A(\theta)$ given as

$$A(\theta) = \frac{N_L - N_R}{N_L + N_R}$$

(6.6)

where $N_L$ and $N_R$ are the number of electrons scattered to the left and right, respectively, through an angle $\theta$. Equation 6.5 shows that $N_L$ will be proportional to $N^l_1[1+S(\theta)] + N^l_2[1-S(\theta)]$, while $N_R$ will be proportional to $N^r_1[1+S(\theta)] + N^r_2[1-S(\theta)]$. Inserting this into Equation 6.6 and utilizing Equation 1.7 yields

$$A(\theta) = PS(\theta).$$

(6.7)

If the Shermann function is known, then the polarization can be determined by measuring the scattering asymmetry. This is the crux of Mott polarimeter theory.

Ideally, then, the polarization of the incident beam can then be calculated from the counting rates of the two detectors. However, an experimental setup will contain instrumental asymmetries which must be corrected for, such as different counting efficiencies in the left and right detectors, misalignment of the incident electron beam, or by inhomogeneity of the target. If the instrumental asymmetry cannot be eliminated, then the results must be corrected to take the asymmetry into account. Another
measurement taken with the polarization reversed \((P = -P)\) will eliminate
the instrumental asymmetries provided the incident beam trajectory is not
affected. Reversing the polarization of the electron beam only involves
reversing the helicity of the photoexcitation light. The ratio of count rates in
the left and right detectors is
\[
\frac{R_L}{R_R} = \frac{\eta_L (1 + PS)}{\eta_R (1 - PS)} \delta
\]
(6.8)
where \(\eta_L\) and \(\eta_R\) are the channeltron efficiencies and \(\delta\) is the instrumental
asymmetry which needs to be corrected out of the results. Reversing the
beam results in a ratio of count rates
\[
\frac{R'_L}{R'_R} = \frac{\eta_L (1 - PS)}{\eta_R (1 + PS)} \delta.
\]
(6.9)
The polarization of the incident beam, corrected for the instrumental
asymmetry, can then be calculated from
\[
P = \frac{1}{S} \frac{X - 1}{X + 1}
\]
(6.10)
where
\[
X = \sqrt{\frac{R_L R'_R}{R_R R'_L}} = \frac{1 + SP}{1 - SP}.
\]
(6.11)
The corrections for instrumental asymmetry will be worked out to first
order by the above process. However, asymmetries that are due to temporal
or helicity-related changes in the beam position may still remain.\(^6\) These can
be monitored during the data taking by measuring the false asymmetry
defined as
\[
A_f = \frac{Y - 1}{Y + 1}
\]
(6.12)
where

\[ Y = \sqrt{\frac{R_L R_R}{R'_L R'_R}}. \]  

(6.13)

The false asymmetry should equal zero in the absence of spurious effects.

In the Mott polarimeter used, the incident electrons scatter from a solid rather than a single atom to obtain stronger scattered-electron signals. This results in multiple scattering events in the solid target, which reduce the scattering asymmetry. An "effective" Shermann function \( S_{\text{eff}}(\theta) \), which will be reduced from the single-atom Shermann function, should be used instead. One way to calibrate a Mott polarimeter device is to measure the scattering asymmetry of an electron beam of known polarization, thereby determining the effective Shermann function.

Figure 6.3 Compact Mott Polarimeter
The compact Mott polarimeter\textsuperscript{63} used in this work is shown in Figure 6.3.\textsuperscript{73} The incident electron beam, after passing through the hemispherical energy analyzer at 10 eV, is further accelerated and focused by the input lenses. The operating voltages for the three lenses are 800 V, 500 V, and 1900 V, respectively. Electrons are scattered through $\pm 120^\circ$ by the thorium target which is biased to 20 kV. The recoiling electrons enter a retarding electric field region as they leave the main housing. The scattered electrons are decelerated in this field and only those that are elastically scattered or have suffered small inelastic energy losses are able to overcome the retarding field and be detected by two bell-mouthed channeltrons. The inelastic energy loss window $\Delta E$, the amount of energy that an electron can lose and still be detected, is set by varying the potential applied to the outer apertures and the channeltron cones.

The typical operating parameters for the polarimeter at 20 kV, are an energy loss window of 532 eV, working in a Shermann function of $\sim 0.17$, and bias across the channeltron of $\sim 4100$ V.

6.4 Data Acquisition System

The data acquisition system is based on a Motorola MC68030 microprocessor. Incident electrons are counted by the channeltrons in pulse mode for high efficiency. To take a polarization measurement of the scattered electron beam, the count rates of the channeltrons are measured. This is accomplished by sending the output pulses from the channeltrons through a pulse-shaping circuit which creates a pulse of amplitude 200 - 500 mV and width of $\sim 20$ ns which is AC coupled to fast preamplifiers. The amplified signal is fed through a discriminator and the output pulses are counted in a scalar-timer and sent to the computer.
7. Derivative Standard for Polarimeter Calibration

The calibration of Mott polarimeters is typically performed using a beam of electrons with accurately known spin polarization. While several polarimeters exist that have been absolutely calibrated to 2 %, most research labs do not have the facilities, the funding, or the time to establish the apparatus to calibrate a polarimeter to this accuracy. Therefore, many researchers rely on an accepted calibration value for the design of the polarimeter which they use, having an inherent accuracy of no better than ± 10 %. Such a large uncertainty can limit the progress of research in fields which utilize spin-sensitive techniques. Without properly calibrated instruments, measurements undertaken at different laboratories are difficult to compare. The availability of a derivative standard for polarimeter calibration would be a great asset. Although the polarization of a derivative standard may not be accurately predicted from first principles, it should meet the rest of the criteria for a standard, i.e. be reproducible and be well characterized.

7.1 Testing the Material

The material to be used as the calibration standard was carefully chosen according to the issues discussed in chapter 3, and also with respect to the material’s availability, its cost, and its compatibility with present polarized electron source systems. The photocathode is 100 nm thick GaAs(110) epitaxially grown onto a layer of Al0.3Ga0.7As, which has a larger bandgap of 1.85 eV, preventing emission from anything but the top 100 nm layer (GaAs).
The lattice constants of the two materials are close enough to insure unstrained growth of the GaAs.

The material testing procedure was quite extensive. In the Cathode Test Laboratory (CTL) at SLAC, the source specimens were cut from a single 3" diameter wafer. The uniformity across the wafer and performance of the material under a variety of conditions were tested. Six samples from the wafer were measured in the CTL under various conditions. The polarization at a given wavelength and the quantum efficiency (QE) did not drift more than 2% during the tests. Additional testing was done to determine the effect of quantum efficiency decay on polarization for the samples. The samples were subjected to different mechanisms to cause the QE to decay. The polarization is slightly dependent on the QE decay, but at a rate which is less than the statistical variation of the measurements.

7.2 Results of the Measurements

The results from the CTL testing, as well as the results from the measurements performed in our laboratory at Rice University and elsewhere are displayed in Figure 7.1. At the CTL, 841 nm excitation wavelength was used, chosen due to the high polarization, high QE and insensitivity to changes in the QE achieved at this wavelength. The CTL has assigned a preliminary polarization value of 43.40 ± 0.86 % for QE in the range 1.2 - 0.009 %. Results from U. C. Irvine, also for 841 nm excitation, utilizing a highly accurate polarimeter are reported to be $P = 42.69 \pm 0.33 \%$ at a QE of 0.38 % and $P = 44.66 \pm 0.30 \%$ at a QE of 0.08 %. Preliminary results have been recorded at the University of Nebraska. This group has measured a polarization value of ~ 47 % at an excitation wavelength of 870 nm. This measurement may be slightly higher than the measurements at the CTL, but still agrees to within 5
% of the other measurements. Measurements at University of Münster will be completed presently.

Figure 7.1 Plot of polarization, and QE, vs. wavelength. The results are from the participating laboratories.

Systematic polarization and quantum efficiency measurements were performed on the material sent to our laboratory. The excitation wavelength
for our measurements was 807 nm. We measured a mean polarization value of \(40.0 \pm 2.5\%\). The QE measured was \(\leq 0.1\%\). This QE value is on the low end of that achieved at the CTL. The polarization measurement falls within the statistical spread of the measurements performed by CTL at this wavelength. Our tests indicate that, at 807 nm, the value of polarization is agreed upon to within \(~ 2\%\).

For the activation procedure used in our laboratory, we used oxygen rather than NF3, as the electronegative element. The close agreement of the results in our laboratory with those at the CTL, and elsewhere, indicates that the polarization is not dependent upon the activation procedure, in agreement with previous studies.\(^7\)

At the CTL, the samples were introduced into the UHV system via a vacuum loadlock system. The typical procedure followed by our laboratory, involves opening the chamber to atmosphere to insert the samples, followed by a bake at temperatures up to \(~ 180^\circ\text{C}\). This enables us to reach UHV. The difference in these two procedures does not affect the polarization of the calibration material.
8. Conclusion

The ability of using 100 nm thick GaAs(110) as a derivative standard for calibrating systems possessing a solid state polarized electron source has been investigated. This material shows good promise for application as a derivative standard. For excitation wavelengths at 807 nm, the measured polarization is in the range $P \approx 41 \pm 2 \%$. At 870 nm wavelength the mean polarizations are somewhat high but all lie within the range $P \approx 44 \pm 3 \%$. Thus, at least to within this uncertainty, spin polarized photoemission from GaAs can form the basis of a simple method to calibrate electron spin polarimeters.
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