Polarization transfer in ion-surface scattering
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Chapter 5

Spin-polarized surfaces

5.1 Introduction

Auger Electron Spectroscopy (AES) and Electron Capture Spectroscopy (ECS) are used to investigate whether ion-surface scattering is sensitive to spin polarization effects in surfaces. For these studies we used helium ion beams, which can be produced rather easily with high intensity. In AES, Auger spectra of doubly excited He$^{2\ast}$, formed by the He$^{2+}$-surface interaction, may reveal information on spin polarization effects by changes in the population balance between triplet and singlet He$^{2\ast}$ states. In ECS, the degree of circular polarization of light emitted in the decay of He$^\ast$ triplet states is used to study spin polarization effects. In this chapter our results from spin-polarized surfaces, obtained with both spectroscopy methods, are presented. Firstly, AES was used to study a Cs-covered GaAs(001) surface, for which it is known that it can be spin-polarized relatively easily by optical pumping with circularly polarized laser light (section 5.2). Secondly, a ferromagnetic Ni(110) surface was studied below and above the Curie temperature by means of AES (section 5.3). Finally, magnetically induced spin polarization of the Ni(110) surface was studied with ECS (section 5.4).

5.2 Spin polarization by optical pumping

Auger electrons of excited He$^{2\ast}$ are of relatively low energy ($\sim 35$ eV) and thus sensitive to (stray) magnetic fields. Therefore a non-magnetic, spin-polarized target is preferred. To produce such a target we used the ideas underlying spin-polarized electron sources [73, 74].
GaAs band structure

The concept is best explained via the band structure of GaAs, shown on the left side of figure 5.1. In the figure, the binding energy is plotted versus the wave number $k$ within the first Brillouin zone, near the $\Gamma$ point. Unlike metals, which have a continuous conduction band, semiconductors have a band gap $E_g$ separating the conduction band $E_c$ from the valence band $E_v$. At $T = 0$ K, in a pure (un-doped) semiconductor, the Fermi level $\mu$ is equal to the Fermi energy $E_F$. The position of $\mu$ can be changed by increasing the temperature and/or by doping of the semiconductor. By n-type doping $\mu$ is increased, by p-type decreased. At room temperature the Fermi level normally lies within the band gap and the conduction band is empty.

The quantity $\chi (\sim 4$ eV) is the electron affinity, i.e. the energy difference between the minimum of the conduction band and the vacuum level. In GaAs, the bottom of the conduction band is separated by a band gap $E_g = 1.43$ eV ($\lambda = 870$ nm) from the top of the valence band. For GaAs the wave functions at the valence band maximum and the conduction band minimum, which is at the $\Gamma$ point, have $p$ and $s$ symmetry, respectively. Due to the spin-orbit interaction the sixfold degenerate $p$ band is split into a fourfold degenerate $P_{3/2}$ state and
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a twofold degenerate $P_{1/2}$ state, separated by $\Delta = 0.34 \text{ eV}$ [73].

**Optical pumping**

Electronic spin polarization can be obtained by optical pumping with circularly polarized light, thereby inducing transitions between the $P_{3/2}$ state (valence band) and the $S_{1/2}$ state (conduction band). In order to selectively pump from only the $P_{3/2}$ state the photon energy $E = \hbar c/\lambda$ should be higher than the band gap $E_g$, but not so high as to promote electrons from the $P_{1/2}$ state too. In our experiments a 30 mW GaAlAs diode laser, producing 1.58 eV ($\lambda = 788 \text{ nm}$) photons, was used for the optical pumping. Circularly polarized light was created by a linear polarizer and a quarter wave plate.

Electronic transitions between initial $|\psi_i\rangle$ and final $|\psi_f\rangle$ states have to obey the selection rules for optical dipole transitions, i.e. $\Delta L = \pm 1$ and $\Delta M_L = 0, \pm 1$. In these transitions the spin is not affected, thus $\Delta M_J = \Delta M_L$. Transitions with $\Delta M_J = 0$ are induced by linearly polarized light ($\pi$ light) and transitions with $\Delta M_J = \pm 1$ by circularly polarized light ($\sigma$ light). The transition probability for a transition from state $|\psi_i\rangle$ to state $|\psi_f\rangle$ is given by $|\langle \psi_f | H_{fi} | \psi_i \rangle |^2$, where $H_{fi}$ represents the interaction Hamiltonian [73]. On the right side of figure 5.1 the magnetic substates $M_J$ of the relevant band states are shown. The arrows indicate the transitions from the valence band substates to the conduction band substates, which can be induced by $\sigma^+$ light. The encircled numbers in the arrows are the corresponding relative transition probabilities [73].

**Spin-polarized surface electrons**

To have the spin-polarized electrons available for capture by the He$^{2+}$ projectile ions, further tricks need to be played in order to prevent the spin-polarized electrons from disappearing into the bulk material. By adsorbing a small amount of Cs atoms onto the clean GaAs surface, the electron affinity $\chi$ can be reduced to almost zero. When a Cs atom is adsorbed on the GaAs, it gives up an electron to the GaAs because the ionization potential of Cs is much lower than the work function of GaAs [75]. The adsorbed Cs atom can be thought of as being resonantly ionized. Evaporating a small amount ($\sim 0.5 \text{ ML}$) of Cs onto the GaAs surface leads to the formation of a dipole layer, which effectively reduces the electron affinity near the surface. Electrons in the conduction band are therefore attracted to the surface and almost set free.

In spin-polarized electron sources, the optically pumped electrons are extracted from the cesiated GaAs surface by an applied electrostatic field. Electrons pumped to the $S_{1/2}$ state are thus effectively removed before they can decay back to the $P_{3/2}$ state. In principle, the spin polarization of the $S_{1/2}$
state is then given by the relative transition probabilities. The spin polarization is defined as the relative difference between the number of spin-up \( N_\uparrow \) and spin-down \( N_\downarrow \) electrons, \( i.e. \)

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

The maximum spin polarization of the \( S_{1/2} \) state, determined by the corresponding relative transition probabilities from figure 5.1, is then \( P = (3 - 1)/(3 + 1) = 50\% \). Electron sources producing a spin polarization of 40-50\% have already been reported [73, 74].

![Helium Auger spectrum taken from a clean GaAs(001) surface. The background and the fits to the two peaks are also indicated.](image)

**Figure 5.2:** Helium Auger spectrum taken from a clean GaAs(001) surface. The background and the fits to the two peaks are also indicated.

### Results

To investigate whether spin polarization effects can be studied by means of Auger electron spectroscopy, we used the recipe of Cs adsorption and optical pumping to produce a spin-polarized target. To do so, Auger spectra as a function of Cs coverage were recorded for three cases: (1) laser off, (2) laser on and linearly polarized light \((\pi)\), and (3) laser on and circularly polarized light \((\sigma^+)\). The Cs was evaporated from a Cs dispenser placed about 2 cm in front of the GaAs surface.
To see whether the surface can be ‘activated’ we biased the target. It was found that electron emission started roughly after 2 minutes of Cs evaporation at a dispenser current of 5 A, but only with the laser on. For longer evaporation times the electron emission decayed almost exponentially. This implies that the Cs coverage is $\sim 0.5$ ML after 2 min of evaporation because at this coverage the work function of the Cs/GaAs system reaches its minimum value $[76, 77]$. It is thus around this coverage that spin polarization effects may be expected in the Auger spectra. By means of Low Energy Ion Scattering (LEIS) measurements we determined that after about 1.4 hours this initial Cs coverage ($\sim 0.5$ ML) was removed from the surface due to sputtering by the He$^{2+}$ ion beam. The LEIS measurements also indicated that the Cs coverage changed linearly with time. Thus by taking Auger spectra as a function of time after evaporation, we obtained information as a function of decreasing Cs coverage. (See figure 5.4 for these spectra.)

Figure 5.3: The ratio (peak 1/peak 2) is plotted as a function of the Cs coverage on the GaAs surface.

Surface spin polarization effects should be determined from the difference in the Auger spectra between case (2) and (3). Optical pumping with $\pi$ light leads to equal numbers of spin-up and spin-down electrons in the conduction band. This is most easily understood by realizing that addition of $\sigma^+$ and $\sigma^-$ light leads to $\pi$ light. Only optical pumping with one type of $\sigma$ light can lead to a net spin polarization of the conduction band. Spin polarization of the electrons at the surface should increase the probability for neutralization into
He$^{2+}$ triplet states. In turn, this should lead to an enhanced intensity of the corresponding Auger peak.

Figure 5.4: Helium Auger spectra from Cs/GaAs plotted for different Cs coverages. (laser off)

Figure 5.2 shows an Auger spectrum obtained from a clean GaAs surface. The energy of the He$^{2+}$ ions was $E_{\text{kin}} = 500$ eV, the incidence and detection angles were $\psi = 10^\circ$ and $\theta = 90^\circ$, respectively. The two peaks in the spectrum are known to be due to autoionization (AI) of four doubly excited He$^{2+}(22l')$ states, formed by resonant neutralization and decaying above the surface on the incoming trajectory [65]. Peak 1 at 34.3 eV is due to AI from the $(2s^2)\,^1S$ singlet state and the $(2s2p)\,^3P$ triplet state. Peak 2 at 35.9 eV originates from the $(2p^2)\,^1D$ and $(2s2p)\,^1P$ singlet states. Due to the small differences in their binding energies and due to level broadening close to the surface, the four states emerge as two strong peaks in the Auger spectra.

In figure 5.2 the data analysis procedure is indicated too. From the raw data (dots) a linear background is subtracted (solid line) and the resulting spectrum is fitted by two Gaussian-shaped peaks (dashed lines). The integral of each fitted peak corresponds to the sum of the intensities of the underlying
5.3. TEMPERATURE DEPENDENT SPIN POLARIZATION

Peaks. Spin polarization should enhance the population of the triplet state with respect to the singlet states. Therefore, the intensity of the (2s2p) \( ^3P \) component in peak 1 should increase relative to the intensities of the three singlet components. The ratio of peak 1 and peak 2 might then serve as a 'measure' of surface spin polarization effects.

In figure 5.3 the (peak 1/peak 2) ratio is plotted versus the Cs coverage, for the three different cases: (1) laser off (up-triangles), (2) laser on and linearly polarized light (\( \pi \), squares), and (3) laser on and circularly polarized light (\( \sigma^+ \), circles). The lines connecting the data points serve to guide the eye. From figure 5.3 it can be seen that only when \( \sigma^+ \) light is used, the peak ratio is enhanced and exhibits a maximum value around a Cs coverage of about 0.5 ML. At this coverage the work function is minimum and the optically pumped and polarized conduction band electrons are expected to be located at the surface. For the other two cases the peak ratio is almost constant as a function of Cs coverage. Based on these first results, it seems that AES might indeed be suited for studies on spin-polarized surfaces.

A critical note is still in place here. In figure 5.2 the background subtraction and peak fitting procedure is indicated. In case of a clean GaAs surface, the clear spectrum allows for a reasonable estimate of the background as well as a good fit to the peaks. However, as can be seen in the Auger spectra of figure 5.4, for higher Cs coverage the spectra are more difficult to analyze. For observation of spin polarization effects the Cs coverage is about 0.5 ML and there the analysis procedure is still applicable.

5.3 Temperature dependent spin polarization

Another test system for observing spin effects is a ferromagnetic target such as e.g. Ni(110). Although the target exhibits no macroscopic magnetization at room temperature, on the microscopic level and below the Curie temperature \( T_C \) the electron spins are magnetically ordered within domains. The ferromagnetic ordering (or spin alignment) can be destroyed by increasing the temperature to the Curie temperature. We have chosen a Ni(110) target for our experiments on temperature dependent spin polarization, because its surface is relatively easy to prepare and because the Curie temperature is relatively low (\( T_C = 354^\circ C \)) and thus easily reached.

AES was used to observe changes in the spin polarization as a function of the temperature. The results are shown in figure 5.5. All four spectra are taken at the same scattering conditions: \( E_{\text{kin}} = 100 \text{ eV}, \psi = 15^\circ \) and \( \theta = 90^\circ \). The Auger spectrum taken at room temperature \( (T = 20^\circ C) \) is shown at the bottom. It can be seen that the intensities of peak 1 and peak 2 are about the same. As the temperature is increased, \( T = 200^\circ C \) and \( T = 300^\circ C \), the intensity of peak 1 becomes slightly less. A loss in ferromagnetic
ordering would manifest itself in a reduced probability of neutralization into the He$^{2+}$($2s2p$) $^3P$ triplet state and thus a reduced intensity of peak 1. The upper spectrum, taken at $T = 430^\circ C$ which is well above $T_C$, shows an even clearer reduction of the triplet peak intensity. This spectrum should represent an Auger spectrum obtained from a surface with a completely random distribution of electron spins. Ergo, the experimental results obtained with AES are at least qualitatively consistent with the temperature induced changes in magnetic ordering of electron spins.

Unfortunately, subsequent attempts of probing spin polarization effects induced by an applied magnetic field were less successful. In these experiments, the sample was clamped in a soft iron yoke (section 3.3.5). However, due to stray magnetic fields, it was no longer possible to detect the helium Auger electrons because of their low kinetic energies ($\sim 35$ eV). Therefore Electron Capture Spectroscopy (ECS) was applied to observe magnetically induced spin polarization effects.
5.4 Magnetically induced spin polarization

The principle of ECS [19], i.e. polarization analysis of the fluorescence from the decaying excited projectiles, was treated in chapter 3. For first explorative experiments on surface magnetism, the \( (1s3d)^3D \rightarrow (1s2p)^3P \) transition in excited helium was chosen. This line, from now on abbreviated as \(^3\!D\)-line, has a wavelength of \( \lambda = 587.56 \) nm. The same line has been used in earlier experiments on a non-magnetic Ni(110) surface [78, 79, 80].

![Figure 5.6: MOKE signal versus yoke current. The Ni(110) target reached saturation magnetization at 4A.](image)

In our ECS experiments the magnetic field is applied along the easy axis of magnetization, i.e. the [111] direction, which is perpendicular to the ion beam direction. The ECS geometry is indicated in figure 3.9 and figure 5.8a. The Ni(110) target was demagnetized by an oscillating current with an amplitude exponentially decreasing in time. Zero yoke current (0 A), and thus no magnetic field, corresponds to a demagnetized target. Target magnetization was checked by the Magneto-Optical Kerr Effect (MOKE). In figure 5.6 the MOKE signal is plotted as a function of the yoke current. Saturation magnetization of the Ni(110) target was reached at a yoke current of 4 A.

Before discussing the ECS results, it should be indicated why the degree of circular polarization is linked to the alignment of surface electron spins. This correlation is best explained in three successive steps. Firstly, for the singlet \(^1\!D\)-line, the link between the polarization of light and the distribution over the magnetic substates is made. Secondly, the preferential orientation of the orbital
angular momentum by the scattering conditions is discussed by two models. Finally, the influence of aligning the surface electron spins on the distribution over the magnetic substates is explained for the triplet $^3D$-line.

**The singlet $^1D$ line**

The singlet $(1s3d)^1D \to (1s2p)^1P$ transition or $^1D$-line in helium has a wavelength of $\lambda = 667.82$ nm. The initial $(1s3d)^1D$ state is populated during the He$^+$($1s$)-surface interaction by electron capture of a surface electron $e_2$ and decays via photoemission to the final $(1s2p)^1P$ state. The captured surface electron $e_2$ with spin $s_2$, will orbit the helium core with orbital angular momentum $l_2$. The initial helium electron $e_1$ has $s_1$ and $l_1$.

The initial and final state configurations are given by their spectroscopic notation $^{2S+1}L_J$. The initial $^1D$ state has $l_1 = 0$ and $l_2 = 2$, so $L = l_1 + l_2 = 2$ and $M_L$ varies between $-2$ and $+2$. The selection rules for optical dipole transitions are $\Delta L = \pm 1$ and $\Delta M_L = 0, \pm 1$, thus the final $^1P$ state has $L = 1$ and $M_L = -1, 0$ or $+1$. Final and initial states are both singlet $S = 0$ states thus the spin can be discarded. Therefore, $\vec{J} = \vec{L}$ and $M_J = M_L$, i.e. the state can be specified by $LM_L$. Figure 5.7 shows the relevant $LM_L$ states for the $^1D$-line. Some examples of optical transitions leading to the emission of linearly polarized light ($\pi$ light) and circularly polarized light ($\sigma^\pm$ light) are also indicated. (For the definition of $\sigma^\pm$ see e.g. [47]).

If all initial magnetic substates are equally populated, the emitted light is un-polarized. Any distribution non-symmetric around $M_L = 0$ leads to the emission of elliptically polarized light. For example, if only $M_L = -2$ substates are populated, pure circularly polarized light ($\sigma^-$) will be emitted. If $I(\sigma^+)$ and $I(\sigma^-)$ are the light intensities of the corresponding optical transitions, the
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relative degree of circular polarization $S/I$ is given by

$$ S/I = \frac{I(\sigma^-) - I(\sigma^+)}{I(\sigma^-) + I(\sigma^+)} $$

The quantity $S/I$ is thus related to an asymmetry in the distribution over the magnetic substates $M_L$.

Model interpretations

Emission of circularly polarized light with a considerable degree of polarization has been reported for the $^1D$-line, see e.g. [81] and [20]. Thus it is known that the distribution over the $^1D$ magnetic substates is not centered around $M_L = 0$. This implies that there is a net orientation of the total orbital angular momentum $\vec{L}$ induced by the scattering conditions.

Such a preferential orientation can be explained by the density gradient model [82]. In this model, the ion experiences an asymmetry in the friction force due to the gradient of the electron density outside the surface, which causes the ion’s electrons to rotate. In figure 5.8a the scattering geometry and the electron density $n_e$ are indicated. The gradient $\nabla n_e$ points in the $-\hat{y}$ direction, the ion velocity $\vec{v}_i$ in the $+\hat{x}$ direction. According to the density gradient model, the net angular momentum is then oriented as $\langle \vec{L} \rangle \sim -\vec{\nabla} n_e \times \vec{v}_i$, i.e. along the $-\hat{z}$ direction.

The same preference is found if the neutralization is described as in ion-atom collisions [83]. It can then be argued that if the electron $e_2$ is transferred to the ion at an internuclear distance $\vec{d}$, its classical orbital angular momentum $\vec{L}$ is proportional to $\vec{d} \times -\vec{v}_i$, with $\vec{v}_i$ the ion velocity. The minus sign occurs because
in the ion reference frame the electron moves in the $-\vec{v}$ direction. Within the scattering geometry of our ECS experiment, the captured electron would have $\vec{l}$, and thus $\vec{L}$, oriented in the $-\hat{z}$ direction, as depicted in figure 5.8b.

An orientation of $\vec{L}$ along the $-\hat{z}$ direction implies that $\langle M_L \rangle < 0$, since we have $L_z = \hbar M_L$. If on average mainly substates with $M_L < 0$ are populated, more $\sigma^-$ light than $\sigma^+$ light is emitted. The measured light intensity will have a high degree of circular polarization with $S/I \approx 30\%$. This is in accordance with previous experiments on (de-)magnetized Fe(110) surfaces [19, 81, 20]. It can now be concluded that $S/I$ is related to the distribution over the magnetic substates. The second step is to describe the influence of the surface electron spin $s_2$ on the $M_L$ distribution. This is best explained via the $(1s3d)\, ^3D \rightarrow (1s2p)\, ^3P$ transition, or $^3D$-line, which has a wavelength of $\lambda = 587.56$ nm.

**The triplet $^3D$ line**

Firstly, it is emphasized that the spin $s_2$ of the surface electron $e_2$ is conserved during the electron capture and during the decay. The spin $s_1$ of the initial electron is always randomly distributed and can have either $m_{s1} = -1/2$ or $+1/2$. The alignment of $s_2$ depends on the ordering of the electron spins in the surface. Ideally, $m_{s2}$ can be chosen to be $-1/2$ or $+1/2$ by magnetizing the surface for example.

![Figure 5.9: The relevant $JM_J$ states for the $(1s3d)\, ^3D \rightarrow (1s2p)\, ^3P$ transition (587.56 nm) and the three types of polarization.](image)

The initial $(1s3d)^3D$ state has $L = 2$ and $M_L$ varies between $-2$ and $+2$. A triplet state has additionally $S = s_1 + s_2 = 1$ and $M_S = -1, 0, +1$. By spin-orbit coupling the total angular momentum is $\vec{J} = \vec{L} + \vec{S}$. In this case $J$ can be 1,2 or 3. The magnetic substate $M_J = M_L + M_S$ can vary between $-J$ and $+J$. The spin is not affected by the decay, i.e. $\Delta S = 0$, thus only $\Delta J = \pm 1$ and $\Delta M_J = 0, \pm 1$ transitions are allowed. The final $(1s2p)^3P$ state
has therefore $L = 1$ and $M_L$ varies between $-1$ and $+1$. The total angular momentum $J$ can now be 0, 1 or 2 and the $M_J$ can vary between $-J$ and $+J$. It is clear that the states must now be specified by $JM_J$. Figure 5.9 shows the relevant $JM_J$ states for the $^3\text{D}$-line. The three types of polarized light are also indicated.

The way the distribution is changed by spin polarization of target electrons can be indicated by the following example. If there is a preferential orientation of $\vec{L}$, say $M_L = -2$, then the possible $M_J$ values are given by $M_J = M_L + M_S = -2 + M_S$. For the triplet $S = 1$ state, $M_J$ can then be $-3, -2$ or $-1$. The total magnetic substate $M_S$ can be described in terms of the individual spin components $m_s$ and is given by $M_S = m_{s1} + m_{s2}$. If the spin of $e_2$ is fixed at $m_{s2} = +1/2$ and $e_1$ is random, $M_S$ can be 0, +1 and $M_J$ can be -2 or -1. But when $m_{s2} = -1/2$, $M_S$ can be $-1, 0$ and $M_J$ can be -3 or -2. Thus by aligning the spin of the surface electron, e.g. by an applied magnetic field, the distribution can be shifted towards lower or higher $M_J$ substates. Higher $M_J$ values increase the degree of circular polarization $S/I$, while lower values decrease $S/I$.

The interaction energy $V$ between an electron’s magnetic moment $\vec{\mu}_s = -g_s \mu_B \vec{s}/\hbar$ and an applied magnetic field $\vec{B}$ is given by $V = -\vec{\mu}_s \cdot \vec{B}$ [47]. For the $z$-component of the interaction one obtains $V_z = g_s m_s \mu_B B_z$, with $g_s = 2$, $m_s = \pm 1/2$ and $\mu_B$ the Bohr magneton. The spin $\vec{s}$ will thus align anti-parallel to an applied magnetic field $\vec{B}$, since this requires minimum energy.

If $B_z$ is applied in the $+\hat{z}$ direction, $m_{s2}$ will align anti-parallel to $B_z$ and we have $m_{s2} = -1/2$. If we have $M_L = -2$, $m_{s2}$ and $M_L$ are aligned parallel. The possible $M_J$ values are then $-3$ and $-2$ and the distribution is shifted to higher negative $M_J$ values. Ergo, a magnetic field in the $+\hat{z}$ direction will shift the $M_J$ distribution to the left, $I(\sigma^-)$ is increased and $S/I$ becomes more positive. Vice versa, a magnetic field applied in the $-\hat{z}$ direction will align $m_{s2}$ anti-parallel to $M_L$. In that case the $M_J$ distribution is shifted to the right and $S/I$ is reduced.

The net effect of aligning $m_{s2}$ on $S/I$ depends on the magnitude of $M_L$. The strongest effect is obtained when $M_L = 0$, because then a change from $m_{s2} = -1/2$ to $+1/2$ will ideally change the degree of polarization from $S/I > 0$ to $S/I < 0$. For higher values of $M_L$, changes in $m_{s2}$ will obviously have a smaller effect on $S/I$ (see e.g. [84]). For the $^3\text{D}$-line it can thus be expected that the changes in $S/I$, induced by a reversal of the applied magnetic field, may not be symmetric with respect to the results for a demagnetized surface with random spins.
ECS results for Ni(110)

Figure 5.10 shows an ECS spectrum obtained for a demagnetized Ni(110) target. The light intensity was measured with PDS-V in the $+\hat{z}$ direction, i.e. parallel to the surface and the magnetic field, and perpendicular to the scattering plane $XY$. This geometry allows for the detection of circularly polarized light. The measured light intensity of the $3^D$-line is plotted as a function of the rotation angle of the $\lambda/4$-plate, which rotates from 0 to 360°. Since the data is symmetric about 180°, only half the rotation range is shown. The two Stokes parameters $I$ and $S$ are also indicated. The solid line is a fit of equation (3.15) to the data.

![Figure 5.10: ECS spectrum of a demagnetized Ni(110) surface. ($E_{kin} = 20$ keV, $\psi = 5^\circ$, $3^D$-line)](image)

The use of a perfect $\lambda/4$-wave plate implies $\Delta = 90^\circ$. Substitution of this value for $\Delta$ simplifies equation (3.15) to

$$I^*(\beta, 90) = \frac{1}{2}I + \frac{1}{2}M\cos^2(2\beta) + \frac{1}{4}C\sin(4\beta) - \frac{1}{2}S\sin(2\beta) \quad (5.2)$$

From equation (5.2) it can be seen that $S/I$ can be obtained from a single measurement. At the angles $\beta = 45^\circ$ and $135^\circ$, equation (5.2) only depends on the Stokes parameter $S$, and one obtains respectively

$$I^*(45, 90) = \frac{1}{2}I - \frac{1}{2}S \quad (5.3)$$

$$I^*(135, 90) = \frac{1}{2}I + \frac{1}{2}S \quad (5.4)$$
The relative degree of circular polarization $S/I$ is then given by

$$S/I = \frac{I^*(135, 90) - I^*(45, 90)}{I^*(135, 90) + I^*(45, 90)}$$

Equation (5.5) holds for a perfect $\lambda/4$-wave plate with $\Delta = 90^\circ$. The retardation angle $\Delta$ for our waveplate was determined by taking ECS spectra (see fig. 5.10) for highly $M$-polarized light. For our waveplate a value of $\Delta = 89 \pm 1$ for $\lambda = 587.56$ nm was obtained.

For a more precise value of $S/I$, equation (3.15) must be fit to the data in figure 5.10. From the fit the following relative Stokes parameters are obtained: $M/I = (2.0 \pm 1.0)\%$, $C/I = (-7.7 \pm 1.0)\%$ and $S/I = (28.3 \pm 0.5)\%$. The value obtained for $S/I$ is consistent with previous measurements on Ni(110) [80]. From the small $M$ and $C$ values it is clear that the light was elliptically polarized, but with a high degree of circular polarization $S$.

![Figure 5.11: Light intensity of the $^3D$-line plotted versus time for switching of the electron spins. ($E_{kin} = 12$ keV, $\psi = 2.5^\circ$)](image)

According to equation (5.3) and (5.4), changes in $S/I$ due to an applied magnetic field can be directly observed at $\beta = 45, 135^\circ$. Intensity measurements with the $\lambda/4$-plate fixed at $135^\circ$ were done for three extreme cases: (1) no alignment (0 A), (2) spins aligned in $-\hat{z}$ direction (-5 A), and (3) spins aligned in $+\hat{z}$ direction (+5 A). The results are shown in figure 5.11, where the intensity of the $^3D$-line is plotted as a function of time. The time axis is divided in
periods of 100 s (dotted lines), i.e. 10 s per data point and 10 points in each period. The value of the applied magnetic field, indicated in each period, was kept constant during the period. The dots represent the raw data, the solid line represents the average intensity during each period and serves to guide the eye.

From figure 5.11 it can be seen that the light intensity directly follows the (hard) changes due to the spin reversal. Alignment of the spins in the $-\hat{z}$ direction (-5 A) leads to a small increase in the light intensity. This is consistent with our interpretation, which was discussed above. Aligning the surface electron spins $m_{s2}$ parallel to $M_L$, i.e. in the $-\hat{z}$ direction, leads to an increase in $S/I$. Vice versa, aligning the spins in the $+\hat{z}$ direction (+5 A) leads to a decrease of the intensity. Furthermore, a clear asymmetry with respect to the (0 A) case between the spin directions can be seen. As mentioned above, the asymmetry is due to the spin-orbit coupling. The effect of aligning $m_{s2}$ is most pronounced when the $M_L$ distribution is centered around $M_L = 0$ [84]. For higher $M_L$ substates the effect of aligning $m_{s2}$ becomes less.

![Figure 5.12: Hysteresis plot: light intensity difference versus yoke current (magnetic field).](image)

The same effects were observed when the magnetic field was varied more smoothly. From these results a hysteresis plot is produced. As an example, the results for spin alignment in the $+\hat{z}$ direction are shown in figure 5.12. The intensity difference $I^*(+A) - I^*(0)$ is plotted versus the yoke current. The
arrows between the data points indicate how the loop was traversed. Note that the curve does not retrace itself, i.e. hysteresis effects occur. At the highest magnetic fields the intensity seems to saturate, which is consistent with our MOKE measurement (fig. 5.6).

It can be concluded that these first results are in accordance with the current interpretation. The sign and magnitude of $S/I$ and the observed asymmetry are in line with our expectations. Ergo, it seems that changes in $S/I$ can be induced by changing the direction of surface electrons spins. ECS may thus have the potential of probing magnetic ordering effects in surfaces or thin films.