Spin-dependent inelastic electron scattering on Ni(110)

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The dependence on primary energy, angle, and temperature of spin-polarized electron-energy-loss spectra from Ni(110) has been investigated. It is shown that the spin asymmetries are strongly energy and angle dependent. The behavior can be accounted for in a simple model in which the inelastic intensity consists of two contributions: A strongly energy- and angle-dependent dipole contribution (purely direct scattering) is superimposed on an isotropic-impact scattering mechanism, which is dominated by exchange scattering. Temperature-dependent spectra do not reveal any changes in the electronic structure between room temperature and the Curie temperature.

I. INTRODUCTION

Electron-energy-loss spectroscopy (EELS) is now being widely applied in surface physics and chemistry.[1] The work has mainly concentrated on vibrational excitations of adsorbates, and more recently it has also become possible to study phonon-dispersion curves of clean metal surfaces. For small scattering angles (close to specular) the energy-loss processes can be described by classical dipole theory since the electron is interacting with the long-range electric fields far above the surface which are generated by the atomic motions. Further off specular (in the so-called impact scattering regime) a quantitative theoretical description is much more complicated and requires in general a full microscopic description including multiple scattering.[2]

Very little attention has been paid to the low-energy electronic excitations of metallic samples, which are always present as a small, smooth background.[3] On magnetic materials, a spin-polarized version of EELS (SPEELS) offers the opportunity to study magnetic excitations at the surface. With the earlier remarks about dipole versus impact scattering in mind, it seems clear that SPEELS should be performed in the impact regime, since long-range dipole scattering does not involve the spin of the electron. The question then arises whether the data can be interpreted in simple models involving, e.g., transitions within a spin-polarized band structure (as experimentalists would like to do) or whether even for a qualitative understanding a full microscopic scattering theory (which is not available at present) is needed. The experimental data published so far on this question are scarce. Based on early work on Ni by Kirschner *et al.*,[4] where large changes in the SPEELS spectra as a function of primary energy were reported, Mills[5] presented a simple theory of electronic losses which included both dipole scattering, and impact scattering contributions. The latter were described phenomenologically. The analysis involves interference between the two in the near specular regime as a source of spin asymmetry in the loss spectrum. Recently Venus and Kirschner published angle dependent SPEELS data on Fe and report on finding evidence for this mechanism.[6]

In this paper we present data on spin asymmetries in SPEELS on Ni as a function of primary energy and angle. It is shown that the results can be understood within a simple model by a superposition of long-range dipole contributions (centered around specular) and an isotropic background of impact scattering, which contains large exchange scattering contributions. Also, temperature-dependent SPEELS data are presented, ranging from room temperature to close to T_c. No changes in the magnetic excitation spectrum are discernible over the energy-loss range investigated (100–600 meV).

II. EXPERIMENT

Spin-polarized electron scattering experiments can be performed in different ways: using a spin-polarized beam and measuring the difference in scattering intensity upon reversal of the polarization of the beam; measuring the polarization of the scattered beam by using an unpolarized primary beam; or in the "complete" experiment combining a polarized beam with polarization analysis of the scattered electrons. Although our experimental setup provides both a source of spin-polarized electrons and a spin detector, in the present study on the angle and energy dependence of spin asymmetries, the spin detector (a high-energy Mott detector) was not used, and intensities were measured by a channeltron electron multiplier, instead.

The spin-polarized electron source is a standard GaAs source. A hemispherical electrostatic deflector is used to monochromatize the beam. Another hemispherical deflector serves as the energy analyzer. The sample is a Ni(110) picture frame single crystal. The scattering geometry is shown in Fig. 1. The total scattering angle is 90°. Off-specular spectra can be taken by rotating the sample. The Ni(110) surface was cleaned by extended Ne-ion bombardment and annealing cycles combined with short heating periods in 10^{-7} Torr oxygen, followed by high-temperature flashes. Cleanliness and surface order were checked by Auger electron spectroscopy and low-energy electron diffraction. The base pressure in the vacuum system was in the low 10^{-11} Torr range.

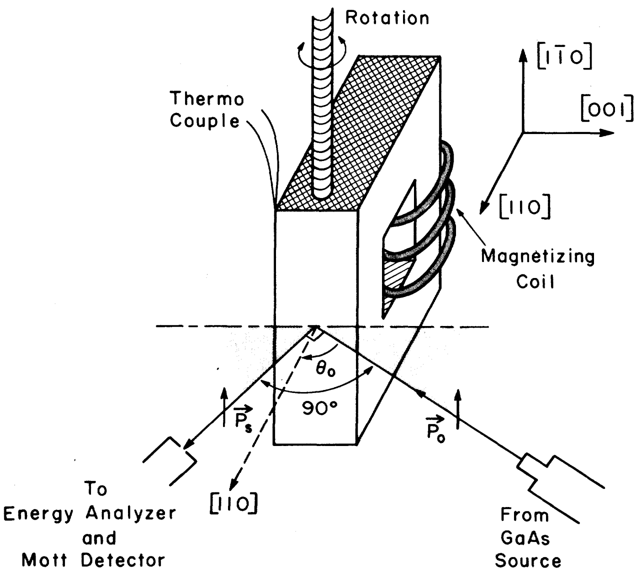

FIG. 1. Schematic of the scattering geometry.

III. RESULTS

A. Energy and angle dependence

In Fig. 2 we show the intensity and spin asymmetry

$$A=\frac{1}{|P_0|}\frac{I_+-I_-}{I_+-I_-} \tag{1}$$

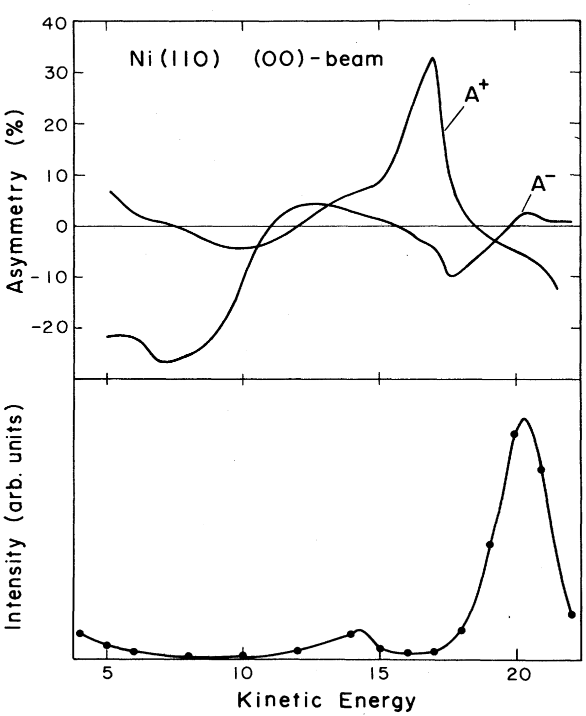

FIG. 2. Intensity (bottom panel) and spin asymmetries (upper panel) for elastic scattering in specular direction as function of beam energy.

for the elastically scattered specular beam over the energy range 4–22 eV. The prominent intensity maximum around 20 eV is the first primary Bragg maximum. The intensity changes by a factor of 30 from 17 to 20 eV. We use this narrow energy range later to investigate the behavior of the spin asymmetries in inelastic scattering. The elastic asymmetries A^+, A^- in Fig. 2 are for magnetization up or down, respectively. They contain contributions due to exchange and spin-orbit interactions.[7] As is generally the case, there is a strong correlation between the asymmetry and the intensity, asymmetry maxima being associated with intensity minima. The very sharp drop visible in A^+ is due to the Bragg maximum.

In order to unambiguously establish the inelastic scattering mechanisms we show in Fig. 3 data from a "complete" experiment, i.e., where the spin polarizations of the scattered electron were also measured. These data were taken at 12-eV primary energy and 20° off specular. Data taken at 10° and 40° off specular are not significantly different.

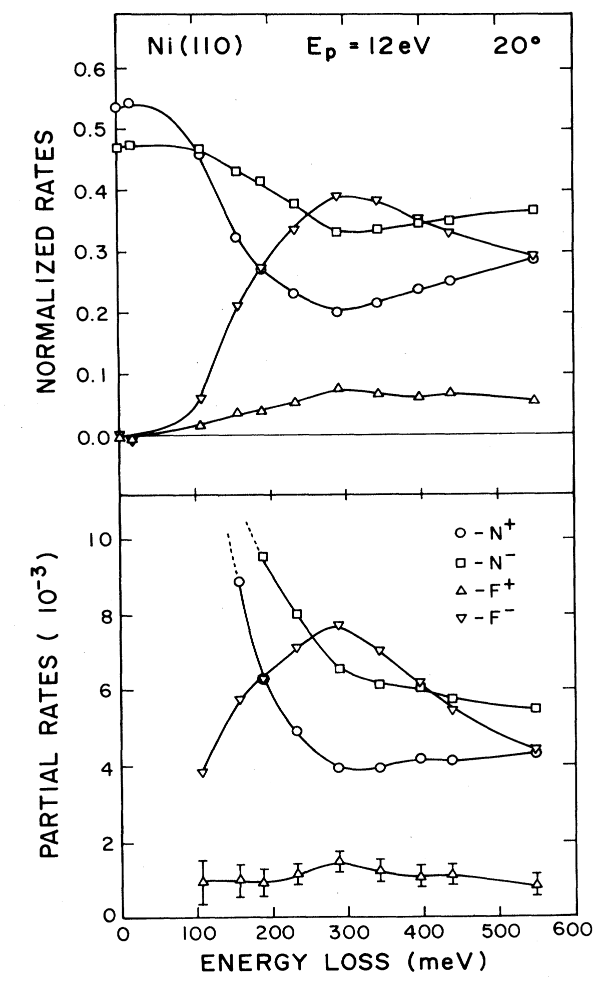

FIG. 3. Nonflip and flip rates as a function of energy loss for 12 eV primary energy, 20° off specular; partial rates normalized to elastic intensity (lower panel); and rates normalized to unity (upper panel).

The upper panel shows the energy-loss rates (N^+, N^-, F^+, F^-) normalized to unity at every energy point (this is a convenient way of presenting the data). The N and F refer to nonflip and flip, the + and − stand for spin-up and -down (of the incoming electron), respectively. The lower panel shows the absolute rates (normalized to the elastic intensity). These data have been discussed in more detail elsewhere.[8] We should emphasize that the elastic scattering is purely nonflip and that in the inelastic regime exchange scattering dominates for these scattering conditions. The differences in both the nonflip and the flip rates lead to the large negative spin asymmetries, making the loss probability for spin-down electrons more than three times larger than for spin-up electrons for energy losses around 300 meV. The energy resolution in this experiment was 80 meV. In the asymmetry spectra shown later the energy resolution was increased to 35–40 meV.

Figure 4 shows a series of asymmetry spectra as a function of primary energy taken in specular geometry. The strong dependence on energy is obvious: The magnitude of the asymmetry values around 300-meV energy loss decrease from −35% at 16.5 eV to zero at 19-eV primary energy. We recall that this is the range where the strong rise in the low-energy electron diffraction (LEED) curve due to the Bragg peak occurs. In order to quantitatively analyze this behavior, we plot in Fig. 5 the ratio of the inelastic intensity (at 300-meV loss) to the elastic intensity versus the elastic intensity. These values are shown as open circles. The inelastic intensity I_{in} is well described by a straight line: $I_{in} = I_{imp} + \alpha I_{el}$. Within classical dipole theory a proportionality is expected since the scattering consists of an elastic reflection followed or preceded by a small-angle inelastic event. The additional intensity term I_{imp} signals the presence of nondipole contribution. If these two processes are independently superposed the resulting asymmetry is given by an intensity-weighted average

$$A = \frac{A_{el}\alpha I_{el} + A_0 I_{imp}}{\alpha I_{el} + I_{imp}} . \quad (2)$$

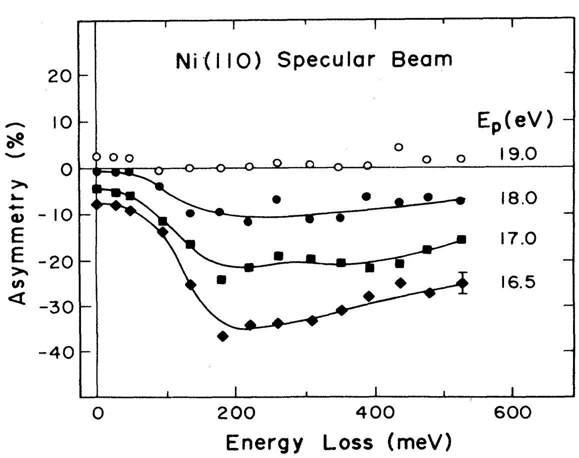

FIG. 4. Inelastic spin asymmetry spectra for different primary energies taken in specular geometry.

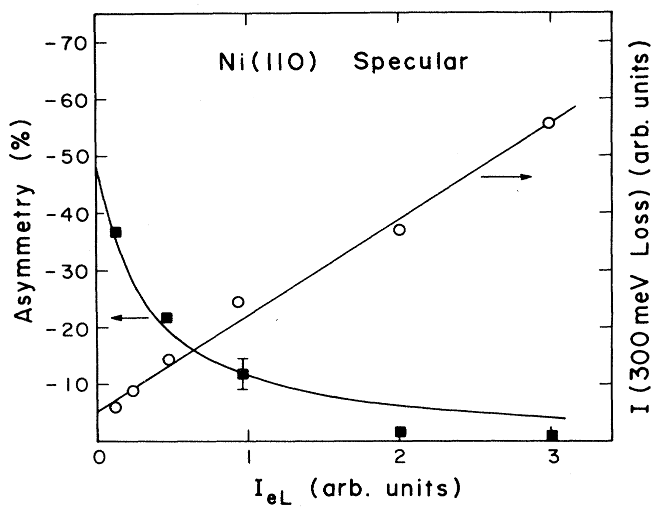

FIG. 5. Measured inelastic intensity at 300-meV energy loss (open circles) vs elastic intensity (the energy range is 16–19 eV). The solid squares are the measured asymmetries around 300-meV loss. The curve represents the calculated asymmetry according to Eq. (1) (see text).

This analysis assumes that the impact scattering does not involve an elastic diffraction. Since the elastic asymmetry is generally quite small (especially where the elastic intensity is large) a good approximation is

$$A = \frac{A_0 I_{imp}}{\alpha I_{el} + I_{imp}} , \quad (3)$$

where A_0 is the asymmetry in the absence of any dipole contributions. A_0 is found experimentally (see further) to be $\simeq -0.55$. In order to test this model we plot in Fig. 5 the measured asymmetries (solid squares) (around 300-meV loss) versus the elastic intensity and compare them to the calculated asymmetries (curve) based on Eq. (3). The agreement is very good. This model assumes the interference term discussed by Mills is small. In Mills theory the interference term is responsible for the difference between the rates N^+ and N^- in Fig. 3. At 300-meV energy loss this difference is rather small compared to the dominant spin-flip rate F^-.

If this simple model describes the primary energy dependence so well, it might also describe the angular dependence. Figure 6 shows the elastic and inelastic (at 300 meV again) intensities as a function of the angle away from specular. The primary energy is 19 eV, i.e., close to the Bragg maximum. The elastic intensity (solid circles) falls off approximately exponentially until it starts to level off at around 15°. The angular width (full width at half maximum, FWHM) of the beam is about 4°. The inelastic intensity levels off somewhat earlier (around 10°). The solid squares are the measured asymmetries around 300-meV energy loss (the curve connecting those points is a guide for the eye only). This shows how the asymmetry rapidly emerges away from specular direction. The open squares are again the calculated asymmetries based on the superposition of dipole and impact contributions, i.e.,

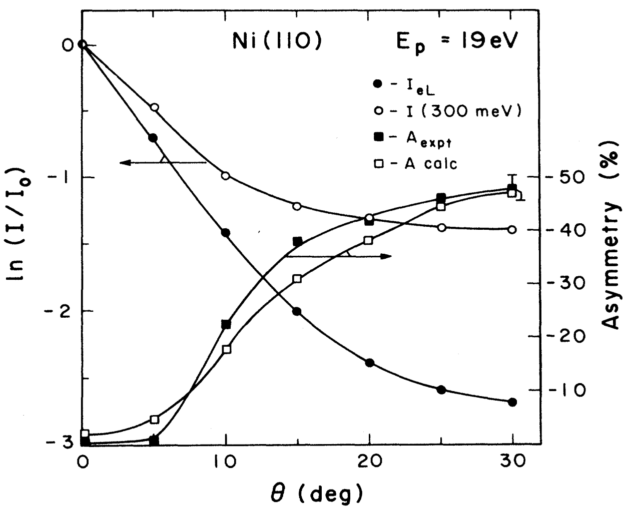

FIG. 6. Angle dependence of the elastic and inelastic intensity and the asymmetry at primary energy of 19 eV (see text).

Eq. (3) (the curve is a guide for the eye only). Note that the asymmetries saturate again around values of -0.5, i.e., very similar to the maximum found in the primary energy dependence. We tried to find situations (by going to lower primary energies and further off specular) with higher asymmetries; -55% was the maximum asymmetry measured under all conditions, e.g., cleanest sample, furthest off specular, lowest primary energy. Since all the data were taken at room temperature, where the surface magnetization is decreased by 10–20% with respect to the $T=0$ value, we should get asymmetries of up to 60–65% at low temperatures. This would make the energy-loss probability (for 300-meV energy loss) ≈ 4.5 times larger for spin-down electrons.

A theoretical explanation of the value of A_0 would have to take into account the details of the band structure around E_F and the relative strength of the possible scattering mechanism (direct and exchange).

The asymmetries over a larger energy-loss range (2 eV) are shown in Fig. 7 at 16.5 eV primary energy for specular and 10° off-specular scattering. The asymmetries decrease smoothly towards higher energies with no signs of additional structures.[9]

B. Temperature dependence

The temperature dependence of the electronic structure of the model itinerant ferromagnet Ni is still a controversial topic. The behavior of spin-split states in angle resolved photoemission spectroscopy, e.g., is very different for Ni and Fe. While Fe shows a temperature-independent exchange splitting (local band theory), for Ni a merging of the spin-split peaks is observed. This could be interpreted as a Stoner-type behavior. But it is also not inconsistent with local band theory.[10] The apparent difference between Fe and Ni could be due to the much smaller exchange splitting in Ni. SPEELS is a new and independent technique used to study this question. If

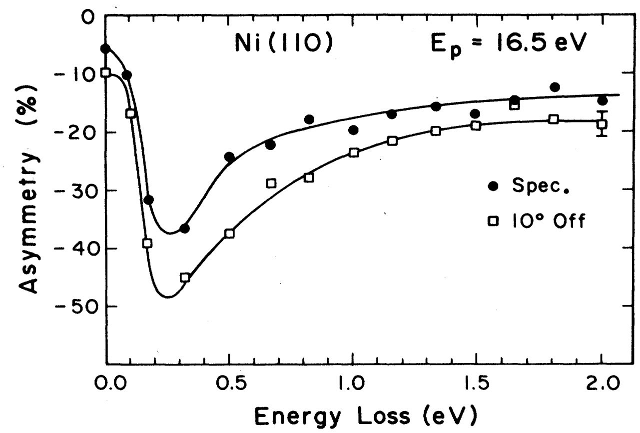

FIG. 7. Inelastic asymmetry spectrum over a 2-eV energy-loss range for specular and 10° off-specular geometry.

there is a reduction in the exchange splitting with increasing temperature one should expect a shift of the asymmetry to lower energies (mainly due to the shift of the F^- contribution). If, on the other hand, there is no significant change in the exchange splitting, the shape of the asymmetry spectrum should be temperature independent. The asymmetries then are expected to approach zero at T_c according to

$$A(E,T)=A(E,0)M_{\text{eff}}(T) , \quad (4)$$

where M_{eff} is an effective surface magnetization (averaged over the probing depth). At T_c the asymmetries have to vanish due to the loss of long-range order (spin-orbit asymmetries can still be present). In Fig. 8 we show a series of asymmetry spectra between room temperature and $0.97T_c$. As expected the asymmetries approach zero as T approaches T_c. At T_c (spectrum not shown) they are zero within the statistical uncertainties, showing that at least under these conditions spin-orbit effects are negli-

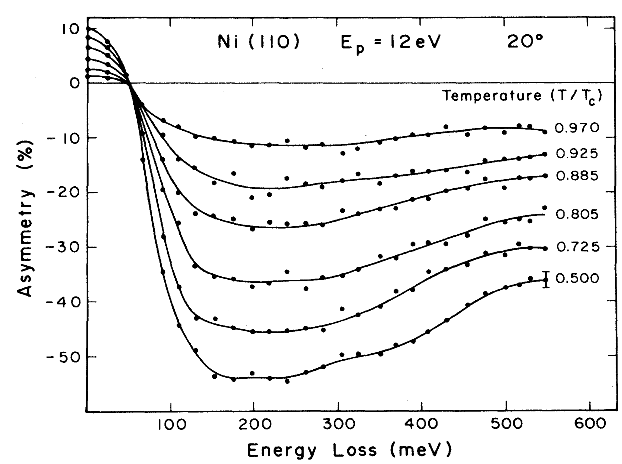

FIG. 8. Temperature-dependent inelastic asymmetry spectra at 12-eV primary energy, 20° off specular.

gible. For a quantitative analysis it is more instructive to plot the asymmetries at fixed energy loss as a function of temperature. This is done in Fig. 9. These data are taken from different runs with fewer energy points but better statistics than in Fig. 8. Also shown for comparison is the temperature dependent of the elastic asymmetry (changed sign and multiplied by 4). All the curves show, within the statistical error, the same temperature dependence. This rules out any significant temperature-dependent changes in the electronic structure and is consistent with local band theory, i.e., a temperature-independent exchange splitting. We also show in Fig. 9 the bulk magnetization curve. We note that the observed temperature dependence of the asymmetries is very similar to the temperature dependence of the spin polarization of low-energy secondary electrons.[11] This is not too surprising, since it is exactly these spin-dependent inelastic processes which determine the spin polarization of the secondaries. Therefore, a similar probing depth is expected. It is also interesting to note that the magnetic probing depth in elastic and inelastic scattering appear to be about the same. While our main conclusion about the temperature independence of the exchange splitting agrees with a recent similar study by Kirschner and Langenbach,[12] their reported asymmetries show a quite different temperature dependence. We do not understand the origin of this discrepancy at present.

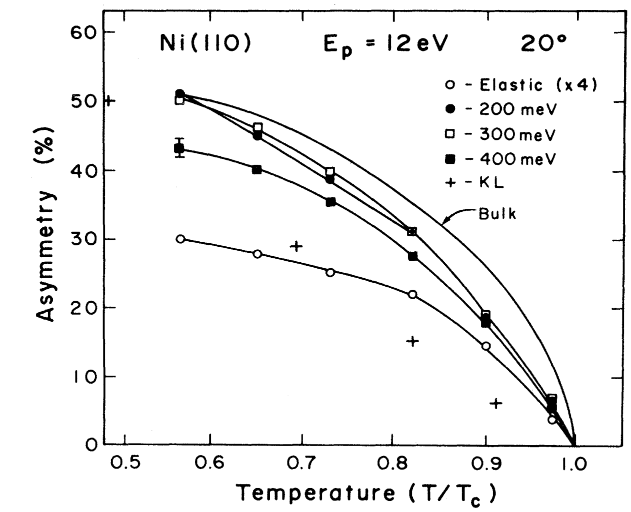

FIG. 9. Measured asymmetries at various loss energies vs reduced temperature. The crosses are from Ref. 12. For comparison, the bulk magnetization curve is also shown.

IV. CONCLUSIONS AND OUTLOOK

We have shown by spin polarized EELS on Ni that inelastic electron scattering due to electron-hole pair excitations can be described by a superposition of long-range dipole scattering, which is strongly centered around specular, and by an *isotropic* impact scattering mechanism, which is dominated by exchange scattering. Spin asymmetries of up to -65% (at low T) lead to a strongly spin-dependent electron mean free path. A microscopic theory is needed in order to explain the observed asymmetry spectra. Temperature-dependent spectra show no signs of a variation of the exchange splitting. It would still be highly desirable to perform the complete experiment at and above T_c. Preliminary data[13] at $1.1T_c$ show that spin-flip scattering is still very strong at 300-meV energy loss, consistent with the persistence of local moments and a spin-split electronic structure in the paramagnetic phase of Ni.

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