Spin-dependent elastic and inelastic electron scattering from magnetic surfaces (invited)

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Results from scattering experiments, using a polarized electron beam combined with polarization analysis, are presented and discussed. There is no evidence of depolarization in quasielastic scattering for the systems studied [Ni(110), Ni(110)O(2×1), paramagnetic Ni(110), Pt(111), graphite]. In inelastic scattering from graphite for small energy losses the depolarization is found to be weak, whereas for ferromagnetic Fe and Ni surfaces large spin-flip contributions have been reported for small energy losses.

I. INTRODUCTION

Spin-polarized electron spectroscopies have become an important technique for studying the magnetism of surfaces and thin films. As primary excitation sources photons (spin-polarized photoemission spectroscopy), electrons (spin-polarized secondary electron spectroscopy including Auger electrons and energy loss spectroscopy) and metastable atoms, and even ion bombardment can be used to emit spin-polarized electrons from a ferromagnetic surface.[1] In all these types of experiments the question arises as to how much the measured polarization reflects the ground state of the system or whether the polarization can be altered significantly by spin-dependent excitation effects (intrinsic and extrinsic), e.g., by spin-dependent elastic or inelastic scattering of the electrons on their way out of the sample. In order to study the possibility of spin-polarization-altering scattering mechanisms in detail one needs, in addition to a spin-polarization detector, a well-defined initial polarization state. We have, therefore, set up a scattering experiment combining a spin-polarized electron source delivering a beam of known polarization (in magnitude and direction) with a high-efficiency and high-accuracy spin-polarization detector. The experiment can also be done with good energy and angular resolution. While spin-polarized electron scattering experiments with either a polarized beam or a polarization detector have been done for a number of years, the combination of both has been tackled only in the last few years. In this paper we discuss the results from these experiments and their implications on other types of spin-polarized spectroscopies.

II. EXPERIMENT

The scattering geometry in our apparatus is shown schematically in Fig. 1. The transversely polarized electron beam is derived in the usual way by photoemission from a Cs and oxygen treated GaAs crystal using circularly polarized light from a GaAlAs laser diode. Besides the usual 90° electrostatic deflector (quarter sphere) to convert longitudinal into transverse spin polarization, the gun contains an additional 180° hemispherical deflector which serves as a monochromator capable of 10-meV energy resolution. Scattered electrons for a fixed 90° scattering angle are spin analyzed in a high-energy Mott detector (typically run at 115 keV) after passing through another hemispherical electrostatic deflector (identical to the monochromator) for energy analysis. The advantage of this setup is that the primary beam polarization direction, the spin-sensitive direction of the Mott detector, and the magnetization of the sample are always parallel (see Fig. 1) and stay that way even if the sample is rotated for angular-dependent measurements. In this way magnetic effects on the scattering are always at a maximum.[2] A more detailed description of the apparatus will be given elsewhere.[3]

III. RESULTS

A. Spin-polarization changes in elastic scattering

It has long been known that, e.g., an unpolarized electron beam can be highly polarized by reflection from, e.g., a W surface, due to spin-orbit interaction (cf. Ref. 1). This process can be described by spin-dependent reflection coefficients R^+ and R^- for spin-up and spin-down electrons, respectively. A partially polarized electron beam of polarization P_0 and total intensity I_0 can be decomposed into spin-up intensity and a spin-down intensity $I_0^\pm = (1 \pm P_0)I_0/2$. The scattered intensities I^+ and I^- are then given by $I^+ = R^+ I_0^+$ and $I^- = R^- I_0^-$ and the polarization $P = (P_0 + A)/(1 + P_0 A)$ where A is the asymmetry $A = (R^+ - R^-)/(R^+ + R^-)$ which describes the intensity changes upon reversal of the primary beam polarization.[4]

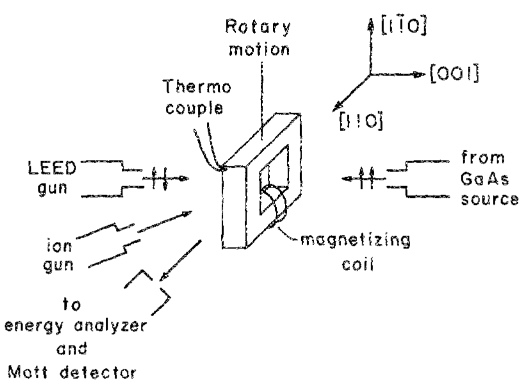

FIG. 1. Schematic of the scattering geometry. The direction of the primary beam polarization, the sample magnetization, and the spin-sensitive axis of the Mott detector are all parallel.

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Now, on a ferromagnetically ordered sample, in general the reflection coefficients depend also on the direction of the magnetization due to the exchange potential. We describe these by N^+ and N^- (where N stands for nonflip). We neglect possible interference effects between spin orbit and exchange. In addition, in general there is the possibility of a spin flip which we describe by F^+, F^-, e.g., F^+ is the probability that an incoming spin-up electron is emerging as a spin-down electron (due to exchange).

In the experiment we measure eight different scattering intensities: The spin-up and spin-down currents for the four possible magnetization/primary-beam-polarization combinations (+ +, + −, − +, − −). These normalized intensities can be written in terms of the reflection probabilities R^α, N^α, F^α as

$$I_\beta^\alpha(\gamma) = \frac{1+\gamma|P_0|}{2} R^\alpha + \frac{1+\alpha\gamma|P_0|}{2} N^{\alpha\beta} + \frac{1-\gamma\alpha|P_0|}{2} F^{-\alpha\beta}, \quad (1)$$

where $\alpha,\beta,\gamma = \pm 1$, and stand for spin-up or down intensity, magnetization up or down, and primary beam polarization up or down, respectively. From these intensities the four polarizations $P_\beta(\gamma)$ can be calculated as

$$P_\beta(\gamma) = \left[\gamma|P_0|\left(1-\frac{2F}{\Sigma}\right) + \frac{\Delta R + \beta(\Delta N - \Delta F)}{\Sigma}\right] \Bigg/ \left(1+\gamma|P_0|\frac{\Delta R + \beta(\Delta N + \Delta F)}{\Sigma}\right), \quad (2)$$

with $F = F^+ + F^-$, $\Delta N = N^+ - N^- \Delta R = R^+ - R^-$, $\Delta F = F^+ - F^-$, and $\Sigma = R^+ + R^- + N^+ + N^- + F^+ + F^-$. The intensity asymmetries upon reversing the primary beam polarization for a completely polarized beam for a given magnetization ($\beta = \pm 1$) are given by

$$A_\beta = [\Delta R + \beta(\Delta N + \Delta F)]/\Sigma. \quad (3)$$

Then $P_\beta(\gamma)$ can be rewritten as

$$P_\beta(\gamma) = \left[\gamma|P_0|\left(1-\frac{2F}{\Sigma}\right) + A_\beta - \beta\frac{2\Delta F}{\Sigma}\right] \Big/ (1+\gamma|P_0|A_\beta). \quad (4)$$

It is interesting to discuss a few special cases of this equation. If the flip rates are zero, it follows that

$$P_\beta = (P_0 + A_\beta)/(1 + P_0 A_\beta), \quad (5)$$

and for $P_0 = 0$ follows $P = A$, a result which has been studied experimentally for spin-orbit asymmetries from W. On a ferromagnet with different flip rates F^+, F^- for $P_0 = 0$ we obtain

$$P_\beta = A_\beta - \beta(2\Delta F/\Sigma). \quad (6)$$

In this case one can, in principle, already detect the difference in flip rates in two separate experiments by measuring the asymmetry with a polarized beam and the scattered polarization using an unpolarized primary beam. Any systematic deviation would be the signature of spin flips. On the other hand, by scattering from a paramagnet the flip probabilities can be expected to be equal ($\Delta F = 0$). Then

$$P = \{P_0[1 - (2F/\Sigma)] + A\}/(1 + P_0 A), \quad (7)$$

and for $P_0 = 0$ it follows $P = A$, always. In this case the complete experiment is needed in order to detect spin flips. The factor $(1 - 2F/\Sigma)$ describes a true "depolarization" of the scattering, i.e., it always tends to decrease the magnitude of P independent of the sign of P_0.

We have performed the measurements on a clean Ni(110) surface and on Ni(110)-O(2×1) for elastic scattering in specular geometry over an energy range from 5 to 30 eV kinetic energy. The results for the clean Ni surface are shown in Fig. 2. In the bottom panel we show the measured asymmetries A^+, A^- for the two sample magnetizations. The upper panel shows the four measured polarizations ($P_0 = \pm 30\%$ and magnetization up and down) (data points) together with the calculated polarization (solid lines) based on the asymmetries only according to Eq. (5). The agreement is excellent, in general the deviations are smaller than 1%, i.e., within the statistics of the measurement. The few cases with deviations of the order of 2%–3% can be attributed to apparatus asymmetries of the Mott detector. The important point is that there are no *systematic* deviations. From this agreement we can put an upper limit on the spin-flip rates by using Eq. (4). We conclude that the total flip processes $F^+ + F^-$ contribute less than 1.5% to the scattering and that the difference in flip rates $F^+ - F^-$ is smaller than 0.5% of the total scattering. Without reproducing the data here we mention that the same limits apply for the case of the Ni(110)O(2×1) surface.[5] Also for the clean Ni(110) surface at and slightly above T_c we found no depolarization within the same limits.

Some comments about the term "elastic" scattering are in order. In a ferromagnetically ordered material no truly elastic spin flip is possible for $q = 0$. The energy resolution in our experiment was kept at 300 meV, so that any magnons

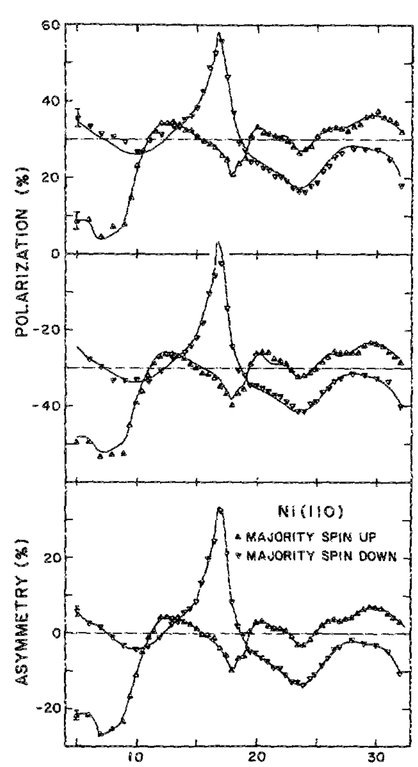

FIG. 2. Comparison of the directly measured spin polarization (symbols) to the calculated polarization (curves) based on the measured asymmetries only, for the four magnetization/primary polarization alignments.

and low-energy Stoner excitations to some extent are included in the measured quasielastic intensity. For Ni above T_c and also in the case of O/Ni system, i.e., in general when the long-range ferromagnetic order disappears but local moments still exist, quasielastic spin flip is still possible, even through the flip rates F^+, F^- (referenced to a fixed spatial axis) must be equal. The signature of spin flip is thus a depolarization of the beam according to $P = P_0[1 - (2F/\Sigma)]$. In a true paramagnet with spin-degenerate energy bands spin flip is possible for arbitrarily small energy loss for $q \neq 0$. However, the available phase space volume goes to zero as $q \to 0$ and $\Delta E \to 0$, so that in this limit $F^+, F^- \to 0$. The only origin of spin dependencies is the spin-orbit interaction. Then the polarization after scattering is given by Eq. (5). Any possible contribution from spin-flip scattering would manifest itself as a systematic lowering of this value. In Fig. 3 we show the comparison between the directly measured polarizations and the calculated polarizations based on the measured asymmetries only (not shown) for scattering from a Pt(111) surface. Again, the agreement is excellent and no systematic deviations indicating spin flip are present. We have shown in detail elsewhere how these data can then actually be used for an accurate self-calibration of a spin polarimeter.[6] Measuring the asymmetries and polarizations simultaneously is essentially equivalent to performing a double scattering experiment. It is due to the data of Fig. 3 that we have confidence in the accuracy of our quoted polarization values on the percent level.

Summarizing our results on elastic (or quasielastic) scattering we can say that in no case have we found any significant contribution due to spin flip. The changes in polarization can be accounted for entirely by spin-dependent (nonflip) reflection coefficients. There is no true depolarization. How far this statement can be generalized to other types of systems, e.g., localized $4f$ moment systems such as Gd above T_c remains to be seen.

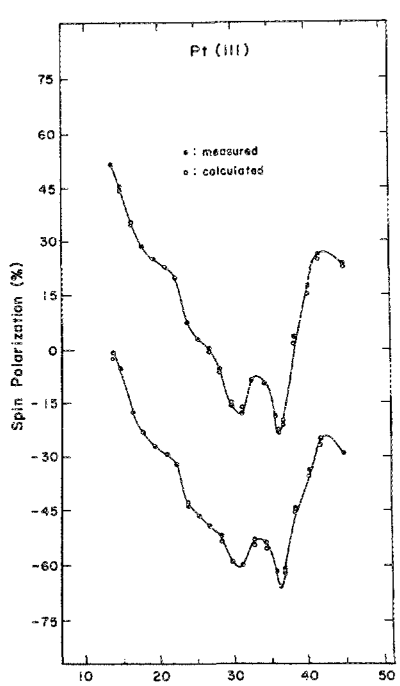

FIG. 3. Same comparison as in Fig. 2 for Pt. Full points: measured polarization; open points: calculated polarizations based on the spin-orbit asymmetry only. The lines are guides to the eye only.

B. Inelastic scattering from ferromagnets

Only a few spin-polarized electron energy loss spectroscopy (SPEELS) experiments on ferromagnetic surfaces have been performed so far.[7] Here we want to discuss only the experiments on $3d$ transition-metal surfaces concerned with electron-hole excitations. Kirschner, Rebenstorff, and Ibach[8] measured asymmetries on an Ni single crystal while Hopster, Raue, and Clauberg[9] measured polarizations on a Fe-based metallic glass. The measured asymmetries on Ni were negative with a broad maximum around 300-meV energy loss, while the measured polarizations on Fe were positive with a maximum around 2 eV energy loss. Taken independently, these experiments then showed only that on Ni incoming spin-down electrons have a higher energy loss probability while the scattered electrons on the Fe sample emerge preferentially with spin-up. Taken together, on the other hand, this is strong evidence for spin-flip scattering. As seen from Eqs. (3) and (6) in the case $\Delta R = 0$ $\Delta N = 0$, $\Delta F \neq 0$, $P = -A$. The fact that the asymmetry was maximum at 300-meV loss energy in Ni and the polarization at ~ 2 eV in Fe, values which correspond to the d-band exchange splittings, was then taken as strong evidence for spin-flip excitations within the d bands, i.e., Stoner excitations. Thus, SPEELS appeared to be a very promising tool to study Stoner excitations, and their temperature dependence in itinerant ferromagnets. Kirschner and co-workers have performed the "complete" SPEELS experiment, i.e., using a polarized beam and polarization analysis, on Fe. The most detailed account of their work was recently given by Venus and Kirschner (VK).[10] Their claim of a "peak" in the flip probability of F^- around 2 eV energy loss is unfortunately not supported by their data. In Fig. 3 of VK they plot the *normalized* partial intensities, for instance, F^-/total intensity. Multiplying this with the intensities from Fig. 3(a) of VK shows that the peak almost completely disappears. The reason for the "peak" in F^-/Σ is to a large extent due to the minimum in the nonflip rates, and the peak position at the exchange splitting of Fe seems rather accidental and may not be directly related to a d-band exchange splitting as done by VK.[11] On the other hand, Dodt *et al.*[12] find real structures in the (unnormalized) flip rates for epitaxial bcc Fe films on Cu_3Au which they attribute to d-band exchange splittings. Also, Idzerda *et al.*[13] report very sharp structures in asymmetry only SPEELS measurements on epitaxial Co films.

We are presently performing SPEELS measurements with 80-meV resolution in Ni(110). We find that the flip-down rate is the dominating loss channel in the range of 250–400 meV energy loss. We take this as evidence that at least to a great extent these losses are due to d-band Stoner excitations. We also find a rather large difference in the nonflip rates, N^- being up to 50% larger than N^+. This difference contributes significantly to the measured scattering asymmetries[8] and may explain the broad tail extending to higher energy losses. We have also for the first time identified inelastic spin-flip scattering on Ni(110) above T_c. A detailed account of this work will be given in a forthcoming publication.

C. Depolarization from paramagnetic materials

To study the effect of depolarization from a paramagnetic surface we took SPEELS data for different primary energy on a graphite surface. Graphite was chosen for experimental convenience. Due to the very low atomic number, spin-orbit effects are negligible; it is easy to clean by flashing and easy to keep clean in UHV. The results are shown in Fig. 4 for two primary energies. The polarization is normalized to the primary beam polarization P_0. Depolarization becomes important only for significant energy losses, above 2.5 eV loss for 10 eV primary energy and above ~ 7 eV for 30 eV energy. In both cases even the electrons emerging at the lowest kinetic energies still have a substantial polarization. These data were taken in specular scattering geometry; spectra taken at 10° off specular do not show significant differences. A theoretical interpretation of these data is not available at present. Glazer and Tosatti[14] have calculated the depolarization due to electron-electron scattering for a free-electron metal. They predict significant depolarizations below about 50 eV primary energy even for arbitrarily small energy losses. With increasing energy loss the polarization goes through zero and then becomes negative (i.e., opposite to the primary beam polarization). In the experimental data in Fig. 4 the effects are much weaker. A negative polarization as predicted is not observed. A theoretical analysis including band structure and matrix element effects is presently underway.[15] In addition, multiple scattering effects for higher energy losses might be important.

IV. CONCLUSIONS

We have found no evidence of any significant spin-flip scattering in elastic scattering. Even though the polarization of an electron beam in general changes in a scattering experiment we have shown that in all cases investigated so far these changes are due solely to spin-dependent reflection coefficients (i.e., to spin-orbit and exchange asymmetries). These energy-dependent asymmetries are reflected, for instance, as structures in the spin polarization spectrum of secondary electrons.

We have shown that in inelastic scattering from graphite there is indeed depolarization, but the effects are weaker than expected and significant only for relatively large energy losses. On the other hand, on ferromagnetic surfaces spin-flip scattering is a large contribution to the inelastic scattering at low energies even for small energy losses. This effect leads to the large spin-polarization enhancement of low-energy secondary electrons.

Perhaps we can summarize our results by saying that it is easy to create a spin-polarized electron beam but much harder to depolarize it.

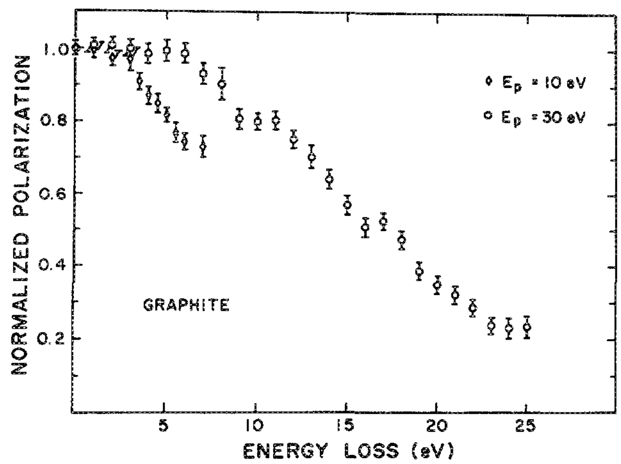

FIG. 4. Normalized spin-polarization P/P_0 for inelastic scattering from graphite for two primary energies.

ACKNOWLEDGMENT

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