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Surface Science 482–485 (2001) 1015–1020

SURFACE SCIENCE

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Spin-polarized (e,2e) spectroscopy of ferromagnetic iron

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Abstract

The scattering of spin-polarized electrons from a ferromagnetic iron surface (1 1 0) is studied using spin-resolved two-electron coincidence spectroscopy ((e,2e) technique) in the reflection mode. The (e,2e) spectra are measured for parallel and antiparallel alignment of the sample's magnetization direction with the polarization vector of the incident electron beam. It is found that the main contribution to the asymmetry of the (e,2e) spectra is due to the exchange interaction; spin-orbit effect is negligible. The sign and the magnitude of the asymmetry are determined by the spin polarization of the sample and by the exchange-induced asymmetry in the electron–electron scattering.

Keywords: Electron–solid interactions, scattering, diffraction; Electron emission; Iron

1. Introduction

A variety of spectroscopic techniques based on the interaction of spin-polarized electrons with a ferromagnetic surface are frequently employed for studying surface magnetism [1]. In particular, the energy and angular-resolved single-electron emission from the valence band following the impact of polarized electrons is utilized for the study of the spin-resolved single-particle electronic structure of ferromagnetism. Manifestations of the electronic correlation in such a single-particle spectroscopy show up as subsidiary features in the spectra. A suitable tool for studying charge and spin-correlation effects more directly is provided by a many-electron coincidence spectroscopy where two or more electrons, emitted from a target upon electron impact, are detected and analyzed in energy and emission angles. The simplest example of the many-particle spectroscopy is the two-electron coincidence experiment, often referred to as (e,2e) [2–4]. Since magnetic properties of surfaces are largely determined by the correlated behavior of the valence electrons it seems natural to envisage the (e,2e) technique in its spin-polarized version for the investigation of magnetism and spin-dependent scattering, as will be done in this work.

The spin-dependence of electronic collisions is basically due to the spin-orbit and exchange interactions [5]. In the present study we focus on the inelastic scattering of low energy electrons from a ferromagnetic surface. If the polarization vector of the incident beam is collinear with the magnetization direction of the sample and perpendicular to the scattering plane both the spin-orbit and exchange interactions contribute to the spin

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0039-6028/01/$ - see front matter
PII: S0039-6028(01)00914-1

asymmetry of the (e,2e) spectrum, i.e. the normalized difference of the two (e,2e) spectra obtained upon reversal of the electron polarization or/and magnetization of the sample. The role of the exchange interaction in the inelastic channel has been studied thoroughly using spin-resolved electron-energy-loss spectroscopy (SPEELS) [6]. Two types of elementary excitations in ferromagnetic crystals were observed with SPEELS: electron–hole pair excitations with spin flip (Stoner excitations) [6] and collective excitations (spin waves) [7]. In SPEELS only one electron resulting from the electron–electron collision is detected. Therefore, more complete information can be gained if, in a (e,2e) experiment, both of the two active electrons are detected. We substantiate this proposition for the case of the spin-polarized (e,2e) reaction from ferromagnetic Fe(1 1 0).

2. Experiment

We performed the (e,2e) experiment in reflection-mode geometry using a low energy (20–30 eV) spin-polarized incident electron beam. As demonstrated previously, the low-energy (e,2e) experiments are highly surface sensitive [8,9] and yield information on the few top-most atomic layers of the sample. The geometrical arrangement of the experiment is shown in Fig. 1. The sample normal, the electron beam and the axes of the two detectors are in the scattering plane. The polarization vector

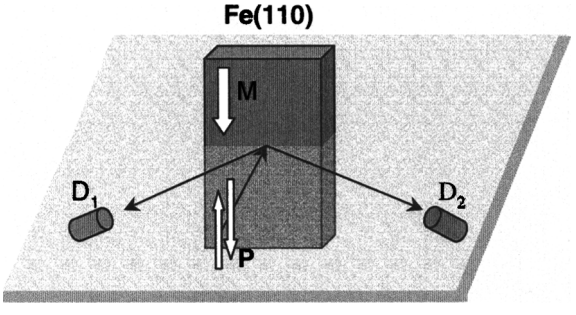

Fig. 1. Geometrical arrangement of the experiment. Detectors (D_1 and D_2), incident beam and surface normal are in the same plane that is perpendicular to the magnetization direction of the sample (M).

of the incident beam and the magnetization direction of the sample are parallel to each other and perpendicular to the scattering plane. We call the spin orientation of the primary beam "up" when its polarization vector is parallel to the majority spin orientation of the sample (antiparallel to the magnetic moment of the sample) and "down" if it is parallel to the minority spin orientation. As a source of spin-polarized electrons we used a strained GaAs multi-layer photocathode activated by Cs deposition and oxygen exposure. The computer controlled Pockels cell polarizes the light of a diode laser ($\lambda = 837$ nm) to obtain right- and left-helicity light that can be used to generate photoelectrons with spin up or down, respectively.

Photoelectrons emitted from the photocathode were deflected by a 90° deflector to convert the originally longitudinally polarized beam into a transversely polarized one. The measurement process is based on the time-of-flight technique and has been described elsewhere [10]. Here we present some experimental details that are important for the present measurements. The single crystal Fe(1 1 0) of size $10.5 \times 3.5 \times 1.5$ mm^3 is mounted on a rotatable holder that allows changing azimuth and polar angles of the sample. The magnetization direction [1 0 0] is oriented perpendicular to the scattering plane. The sample is magnetized using a magnetic yoke with a coil that suppresses stray magnetic fields and allows maintaining single-domain magnetization. All measurements were performed at remanence magnetization of the sample. We used position sensitive micro-channel plate-based electron detectors of 75 mm in diameter. The two detector axes include an angle of 80°. The flight distance from the sample to the detectors was 160 mm. The acceptance angle of each detector was $\pm 13°$. In the present measurements we integrated all events over the acceptance angles of the detectors. The position information on the electron impact was used to correct for the difference in flight paths from the center of the plate to its perimeter, thus improving the energy resolution. The energy resolution of the time-of-flight electron energy analyzers depends on the electron energy to be measured and varied from 0.5 to 1 eV in the range of interest. The average coincidence count rate was 1–3 events per second.

To achieve reasonable statistics the total number of events in one spectrum should be of the order of 10^6 events. The acquisition time for such a spectrum was about 270 h. This long-term measurement requires stability of electronics, constant incident current and good vacuum conditions. To avoid the possible influence of these factors on the measured (e,2e) spectra the polarization of the incident beam has been reversed every 5 s and the data for each polarization were stored in two different files. In spite of the UHV conditions ($p = 5 \times 10^{-11}$ mbar) we found it necessary to interrupt the measurement each 5–7 h for surface cleaning. The cleaning procedure included Ar^+ ion sputtering followed by annealing and, if necessary, oxygen treatment to remove the carbon from the surface. The cleanliness of the surface was monitored by Auger electron spectroscopy.

The essential requirements of our experiment are the high degree of the incident beam polarization and a single-domain magnetization of the sample. To insure that these conditions are fulfilled we measured the electron-energy-loss spectra for spin-up (I_{up}) and spin-down (I_{down}) incident electrons (SPEELS) at 20 eV primary energy and then calculated the intensity asymmetry of Stoner excitations. Fig. 2(a) and (b) shows the SPEEL spectra for spin-up and spin-down primary electrons and the corresponding asymmetry, defined as:

$$A = \frac{I_{up} - I_{down}}{I_{up} + I_{down}}.$$

The value of the asymmetry depends strongly on the cleanliness of the surface, the average magnetization of the sample and the degree of polarization of the primary beam. The asymmetry in the region of the Stoner excitations is measured before and after the coincidence measurements. In the best case we measured $A = 20\%$, otherwise the measured spin asymmetry is in the range of 17%. Assuming a perfectly magnetized sample and a reference value of the asymmetry in the Stoner excitation [11] of $\approx$30%, we estimated the polarization of the incident beam to be about 60–65%.

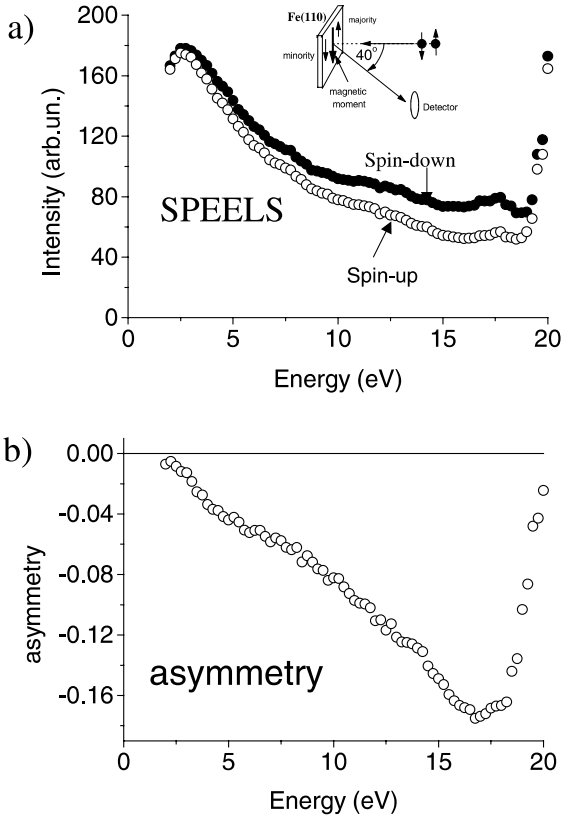

Fig. 2. (a) Geometrical arrangement and electron energy loss spectra (EELS) for the incident spin orientation parallel (spin-up EELS) and antiparallel (spin-down EELS) to the majority spin-orientation of the sample, $E_p = 20.5$ eV. (b) Asymmetry of the EELS.

3. Results

We made measurements for a set of primary electron energies from 23 to 28 eV at normal and off-normal incidence.

The measured two-dimensional time-of-flight spectra were converted into two-dimensional energy distributions of correlated electron pairs, hereafter referred to as the "(e,2e) spectra". Each point in such a distribution corresponds to a correlated electron pair with energies E_1 and E_2 of the two electrons.

Fig. 3(a) shows the difference of the two (e,2e) spectra for incident electrons with spin up and with spin down. The incident electron has a primary energy of $E_p = 26$ eV and the experiment has

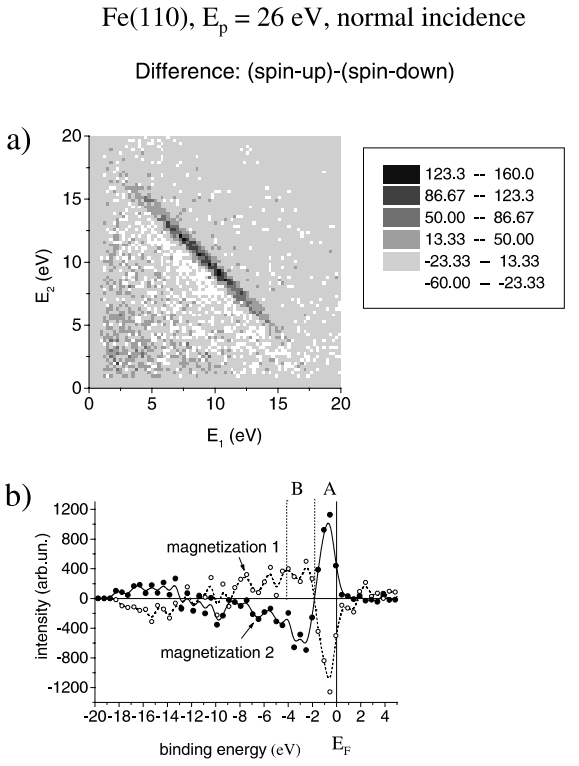

Fig. 3. (a) Difference of two-dimensional distributions of correlated electrons ((e,2e) spectra) measured with the incident spin orientation parallel and antiparallel to the sample magnetization. (b) Difference binding energy distributions for two opposite magnetizations of the sample.

been performed in normal incidence. The initial-state energy E_b of the valence electron is determined from the energy conservation law

$$E_b = E_p - E_{tot} - \varphi, \quad (1)$$

where E_p is the primary electron energy, E_{tot} is the sum energy of the two electrons and $\varphi = 4.8$ eV is the work function of the iron surface. The main contrast in the difference spectrum, shown in Fig. 3(a), occurs along the diagonal line with a constant sum energy of the two correlated electrons $E_{tot} = E_1 + E_2 = 20$ eV. This corresponds to electron pair emission from the vicinity of the Fermi level. From Eq. (1) it is clear that if one collects all the electron pairs with a certain sum energy E_{tot} regardless of the individual electrons' energies E_1 and E_2 (i.e. if we integrate over E_1 while keeping fixed $E_2 = E_{tot} - E_1$), the (e,2e) spectrum can be scanned as function of the initial-state energy E_b. This is done in Fig. 3(b). The spectrum changes sign from positive, just below the Fermi level (region A), to negative, for higher binding energies (below -2 eV) (region B). To ensure that the structure in the difference spectrum is due to the relative orientation of the beam polarization vector and the magnetic moment of the sample the magnetization of the sample was reversed and the same sequence of spin-up and spin-down spectra were measured again. The corresponding difference binding energy spectrum (for opposite magnetization direction of the sample, called "magnetization 1") is shown in Fig. 3(b) by open circles (solid circles are deduced from the measurement of Fig. 3(a)). The reflection symmetry of the spectra for magnetization 1 and magnetization 2 at the zero line intensity indicates that the observed spin-asymmetry is mainly due to the exchange interaction in the electron–electron scattering while the spin–orbit effects are negligible [12].

For a more detailed insight into the spin-dependent scattering dynamics one can analyze the influence of the spin projection of the incoming beam on the sharing of the energy E_{tot} between the two emitted electrons, i.e. one can study the spin-dependent energy correlation spectrum. This kind of study is an advantage of the (e,2e) coincidence technique and can not be performed by conventional single particle spectroscopic methods such as SPEELS.

We focus on two regions of the binding energy which are indicated by *A* and *B* in Fig. 3. Fig. 4(c) shows the spin asymmetry deduced from the energy-sharing spectra for spin-up and spin-down of the incoming electron beam where the sum energy is fixed to $E_{tot} = 20.7 \pm 0.7$ eV (region A). The asymmetry is positive for almost all combinations of energies.

E_1 and E_2 that conserve $E_1 + E_2 = E_{tot}$. When the sum energy of pairs is lowered to $E_{tot} = 19 \pm 1$ eV (region B) (i.e. higher binding energy of the valence electrons) the asymmetry turns negative (Fig. 4(d)). This behavior is also confirmed by the theoretical calculations. The theory encompasses the spin-split band structure (within FPLAPW) and the scattering dynamics via the evaluation of

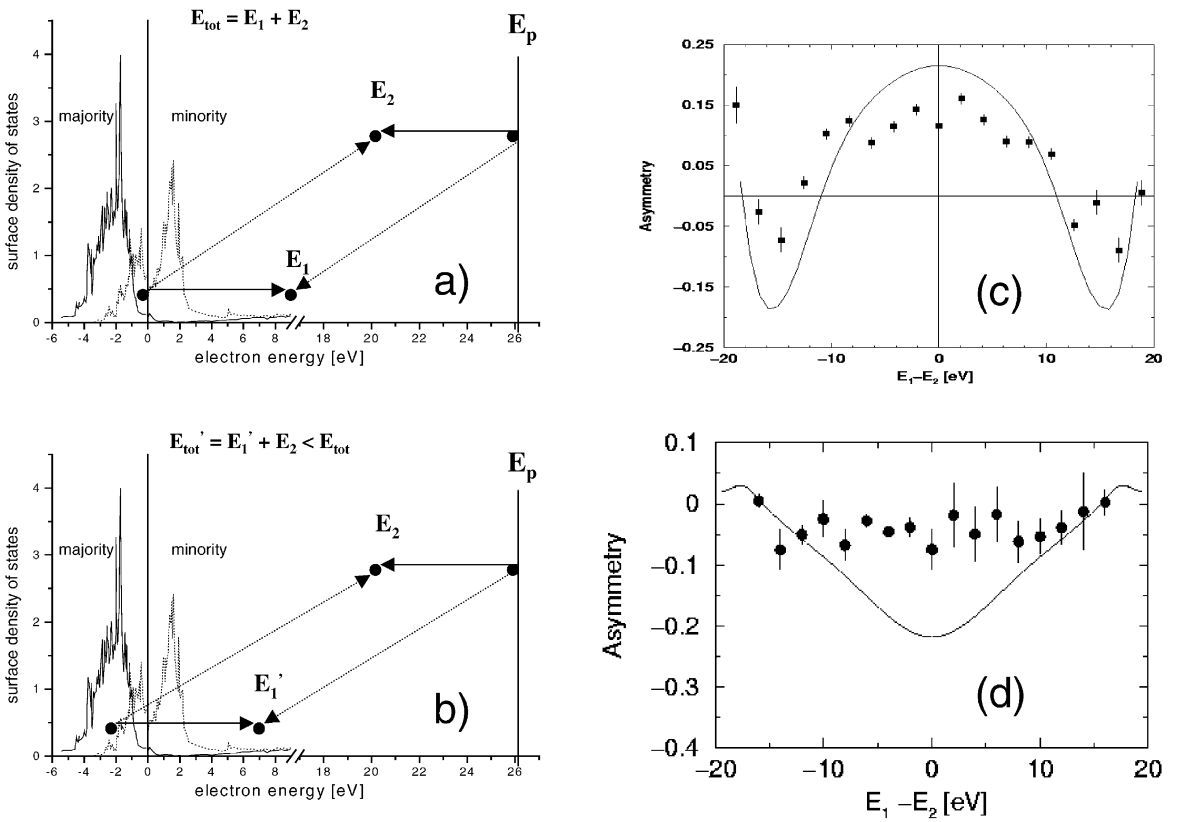

Fig. 4. (a) Energy diagram for the correlated electron pair excitation with electron energies E_1 and E_2 when the valence electron is excited from the vicinity of the Fermi level ($E_b = -0.7 \pm 0.7$ eV) where the surface DOS for minority electrons is higher than for majority ones; (b) the same energy diagram as in (a) but for the valence electron excited from the lower energy level ($E_b = -2.4 \pm 1$ eV) where the surface density of states for majority electrons is higher than for minority ones; (c) and (d) asymmetry of energy sharing distributions of correlated electron pairs with total energy equal to E_{tot} and E'_{tot} respectively. Dots are experimental data, lines indicate the corresponding calculations.

the layer-resolved transition matrix in each spin channel, full details are given elsewhere [12,13]. To uncover the reason for the sign change when comparing Fig. 4(c, d) we consider the variation of the spin-resolved surface density of states (DOS) in iron (see Fig. 4(a, b)) as the binding energy varies.

The binding energy indicated on Fig. 4(a) corresponds to that of Fig. 4(c) while on Fig. 4(b) we marked the binding energy appropriate for Fig. 4(d). As shown previously [12,13], for a certain atomic layer, the normalized difference in the Diff = (DOS(minority) − DOS(majority))/ (DOS (minority)+DOS(majority)) enters as a multiplicative factor in the expression for the spin asymmetry. This factor is positive in case of Fig. 4(c) and negative in case of Fig. 4(d), as evident from Fig. 4(a) and (b). We note that an iron-bulk calculation not only yields different shapes of DOSs but also reversed sign of the normalized difference Diff at the Fermi level. This results in a wrong sign of the calculated spin asymmetry as compared to the measurement.

The agreement between theory and experiment is better in Fig. 4(b) than in (d). This is because the present theory does not account for pair generation accompanied with energy-loss processes. This shortcoming is more serious for pair emission from deeper levels of the conduction bands (as is the case in Fig. 4(d)) than emission from the vicinity of Fermi edge (as in Fig. 4(b)).

4. Conclusions

In summary we studied in this work the electron-pair emission from a ferromagnetic iron (1 1 0) following the impact of a polarized low-energy electron beam. We unraveled the influence of the orientation of the spin-polarization of the incoming electron relative to the orientation of the magnetization of the sample on the electron-pair spectra. These spin effects depend in a characteristic way on the electronic band-structure of the sample as well as on the energies of the emitted electrons. It is one of the aims of future research to refine this coincidence technique to realize the mapping of the spin-split band structure of ordered and amorphous materials, as suggested in Ref. [12].

Acknowledgements

We thank H. Engelhard for his help.

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