Journal of Magnetism and Magnetic Materials 121 (1993) 163–166
North-Holland

Layer-dependent magnetic properties at Fe surfaces using spin-polarized electron emission spectroscopy (SPEES)

C. Rau, N.J. Zheng and M. Lu

Department of Physics and Rice Quantum Institute, Rice University, Houston, TX 77251-1892, USA

We have studied the angle-resolved energy distribution of spin-polarized electrons emitted during ion-surface scattering. At clean Fe surfaces, electron spin polarization values (ESP) of up to 48% are found. Varying the scattering angle of the ions from 1° to 45°, we find that the layer-dependent, average ESP at the topmost surface layer is enhanced by 32% compared to that of the bulk and that the mean free path of electrons with energy 4–10 eV is about 3 atomic distances. The absence of a magnetically dead layer at Fe surfaces covered with one monolayer of O is revealed by the existence of a nonzero surface ESP.

The layer-dependent two-dimensional ferromagnetic properties of the 3d-transit ion metals continue to attract wide scientific attention. Strongly enhanced surface magnetic moments were predicted for a series of transition metal overlayers, interfaces and superlattices [1,2]. Furthermore, questions about the origin of interface magnetism, coupling through nonmagnetic and magnetic interlayers and magnetic exchange coupling between magnetic layers fascinate many scientists [3,4]. This arises from the fact that these systems are of pivotal interest both for fundamental studies and for technological applications.

Grazing-angle ion-surface interaction provides a powerful means to study surface magnetic, electronic and chemical properties of magnetic systems [5,6].

Recently, we reported on experimental data using spin-polarized, angle-resolved electron emission spectroscopy (SPEES) during reflection of H^+, He^+ and Ne^+ ions at various magnetic and nonmagnetic surfaces [6,7]. Investigating the electron spin polarization (ESP) P of electrons emitted from Ni(110) during grazing-angle surface reflection of H^+ and He^+ ions, we find that the angle-resolved energy distribution (ARED) of the emitted electrons is *significantly* different (absence of secondary electron cascades) from that of electron-induced secondary electron emission spectra. It shows a series of characteristic peaks which can be linked, either to various electronic processes where spin-split, local surface electronic states of Ni(110) and electronic states of the reflecting particles are involved, or to spin-polarized, element-specific Auger electron transitions occurring after ion-induced core electron excitation.

In this paper, using SPEES at clean and O-covered Fe surfaces, we report on the layer-dependence of magnetic properties. At grazing angles of incidence, ions cannot penetrate a flat surface, they are specularly reflected and, therefore, probe the physical properties of the *topmost* surface layer. Increasing the angle of incidence α, ions can penetrate the surface. Therefore, changing α from 1° up to 45° enables us to vary the *probing depth* of the incident ions from the topmost surface layer to deeper layers. It further allows us to link SPEES data to electron- or ion-induced electron spectra which are predominantly recorded for $\alpha = 45°$ [8–10]. For $\alpha = 45°$, our spectra exhibit the well-known, low-energy cascade maximum at 2 eV and a peak at around 45 eV due to the emission of spin-polarized, element-specific Auger MVV electrons [11,6] with an average ESP of 30% which is close to the bulk magnetization of Fe (28%).

Correspondence to: Prof. C. Rau, Department of Physics, Rice University, Houston, TX 77251-1892, USA. Tel.: +1-713-285-5417; telefax: +1-713-527-9033; e-mail: RAU@RICEVM1.RICE.EDU.

Varying α from 1° to 45° and using the ESP of electrons emitted at high energies ($\approx$ 10 eV above the vacuum level) as a measure of the average net magnetization, we find that at clean Fe surfaces the surface magnetization (33%) is *enhanced* by approximately 32% compared to the bulk value (25%). For low-energy ($\approx$ 4 eV) electrons, we find ESP values of up to 48% which can be linked to Stoner excitations. For electrons emitted from the topmost layer of Fe surfaces covered with one monolayer of O, we find nonzero ESP values, which indicates the absence of a magnetically dead surface layer.

Experimental details are reported elsewhere [12,6]. For the SPEES experiments presented here, surface scattering of 25 keV Ne^+ ions is used to study the emission of spin-polarized electrons occurring during particle-surface interaction. We note that $P > 0$ is related to a predominance of majority-spin electrons (ESP parallel to the target magnetization), and $P < 0$ refers to a predominance of minority-spin electrons (ESP antiparallel to the target magnetization) [4]. The ESP is measured at 2×10^{-10} mbar.

Fig. 1 gives for $\alpha = 1°$ the ARED and the ESP as function of the energy E (above the vacuum level) of electrons emitted from clean (solid line) and O-covered (dashed line) Fe surfaces. For clean Fe, the ARED of the emitted electrons peaks at around 4 eV. At O-covered Fe, we find a strong increase in the intensity of emitted electrons compared to that of clean Fe.

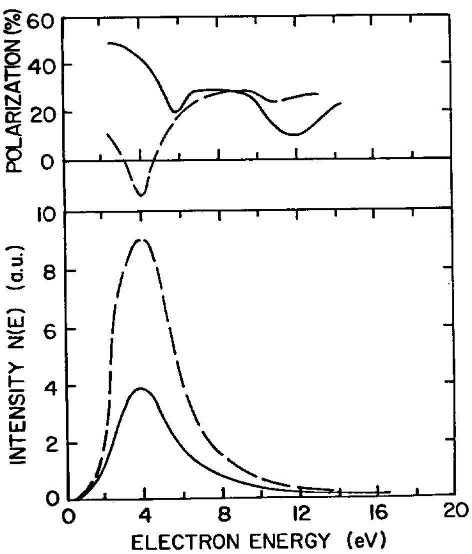

Fig. 1. ARED and ESP as function of the energy E of electrons emitted from clean (solid line) and O-covered (dashed line) Fe surfaces for 25 keV Ne^+ ions and $\alpha = 1°$.

As regards the ESP of electrons excited at the topmost surface layer of clean Fe, we observe an increase of the average ESP from $P = (33 \pm 2)\%$ for $E = 10$ eV to $P = (48 \pm 2)\%$ for $E = 4$ eV. These values are far above the average bulk magnetization value of Fe which amounts to 28%. For Fe surfaces with one monolayer of O, the average ESP of electrons with E = 10 eV remains nearly unchanged ($(32 \pm 2)\%$), whereas for electrons with $E = 4$ eV, the ESP changes from (48 ± 2) to $-(14 \pm 2)\%$ eV indicating the absence of a magnetically dead surface layer.

We note that the distance of closest approach of the ions towards the Fe surface is characterized by the energy component $E_\perp = E_0 \sin^2\alpha \approx E_0\alpha^2 = 8.2$ eV for $\alpha = 1°$. Using appropriate surface potentials [4], it can be shown that the Ne^+ ions are specularly reflected and do not penetrate the Fe surfaces.

At present, it is of considerable interest whether the measured ESP of the emitted electrons reflects the layer-dependent net magnetization of a material. From ion- [10,13,14,6] and electron-induced electron spectra [9], there is evidence that the ESP of electrons emitted at high energies ($\approx$ 10 eV above the vacuum level) scales roughly with the average net magnetization.

For $\alpha = 8°$, $E_\perp$ amounts to 32.8 eV where the ions can penetrate the topmost surface layer and are capable of exciting electrons from the second layer. The shape of the ARED of the emitted electrons is similar to that of fig. 1 with the peak maximum shifted to 5 eV. As regards the ESP of the electrons emitted from clean Fe, we observe an increase of the average ESP from P = 29% for $E = 10$ eV to $P = 50\%$ for $E = 4$ eV. For the O-covered Fe surface, the average ESP of electrons with E = 10 eV remains unchanged ($(32 \pm 2)\%$), whereas for electrons with $E = 4$ eV, the ESP changes from $(50 \pm 2)\%$ to $(14 \pm 2)\%$ eV.

In the following, we discuss our experimental data obtained for $\alpha = 1°$ and 8° in connection

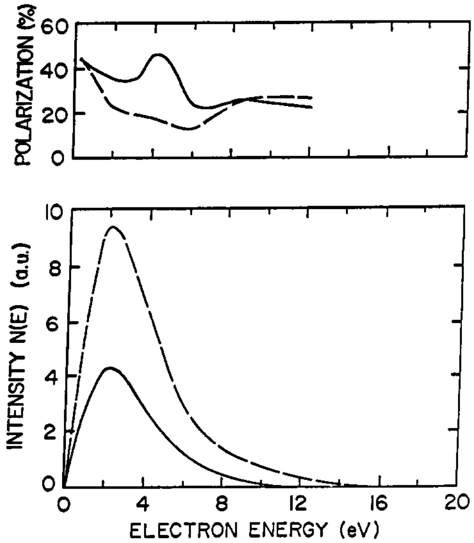

Fig. 2. ARED and ESP as function of the energy E of electrons emitted from clean (solid line) and O-covered (dashed line) Fe surfaces for 25 keV Ne^+ ions and $\alpha = 45°$.

with our SPEES data using $\alpha = 45°$ where bulk layers are probed.

Fig. 2 gives for $\alpha = 45°$ the ARED and the ESP of electrons emitted from clean (solid line) and O-covered (dashed line) Fe surfaces. For $\alpha = 45°$, $E_\perp$ amounts to 12.5 keV where the ions can deeply penetrate into the solid and are capable of exciting electrons from bulk layers. The ARED of clean and O-covered Fe surfaces is similar to that obtained in electron-induced secondary electron emission experiments [8–10] and peaks at 2 eV. For O-covered Fe surfaces, we observe again a strong increase in the intensity of the emitted electrons. We note that for electrons excited in bulk layers, electron cascading and multiple scattering are the dominant processes occurring during electron transport to the surface which cause the well-known 2 eV peak in electron- or ion-induced electron spectra. This is consistent with our data for $\alpha = 1°$ and $\alpha = 8°$ where the ARED of the emitted electrons peaks at higher energies (around 4–5 eV) showing that electron cascading and multiple scattering processes are predominantly negligible.

As regards the ESP of the emitted electrons, we observe an increase of the average ESP from $P = (25 \pm 2)\%$ for $E = 10$ eV to $P = (45 \pm 2)\%$ for $E = 4$ eV. For the O-covered Fe surface, the average ESP of electrons with $E = 10$ eV remains unchanged $((25 \pm 2)\%)$, whereas for electrons with $E = 4$ eV, the ESP changes from (25 ± 2) to $(15 \pm 2)\%$ eV.

Changing α from 45° to 1° which corresponds to a reduction in the probing depth from deep lying layers, where bulk physical properties are probed, to the topmost surface layer, we find an increase in the ESP of "high-energy" electrons from 25 to 33%. This would imply that for polycrystalline Fe surfaces, the net magnetization increases in going from the bulk to the surface. Assuming that the ESP of electrons emitted at high energies (≈ 10 eV) scales roughly with the average net magnetization, it is tempting to correlate this surface enhancement of the ESP, which amounts to approximately 32%, to the influence of theoretically predicted magnetic surface states which cause enhancements of the magnetization of 32% and 20% for Fe(100) and Fe(110) surfaces [1,2].

From our data, it is obvious that the ESP of electrons emitted from clean Fe surfaces increases with decreasing electron energy from the high-energy region (≈ 10 eV) to the low-energy (≈ 3–4 eV) part of the ARED. For $\alpha = 1°$, the average ESP increases from 33 to 48% which amounts to a 45% enhancement, for $\alpha = 8°$, the average ESP increases from 29 to 50% which amounts to an enhancement of the ESP of $\approx$ 70%, and for $\alpha = 45°$, the average ESP increases from 25 to 45% which amounts to an enhancement of the ESP of $\approx 80\%$.

We believe that these enhancements of the ESP are not only due to the spin dependence of the electron mean free path [15] which is caused by an excess of unfilled minority-spin d states over unfilled majority-spin d states in which excited spin-polarized electrons can be scattered during transport to the surface. Assuming that electron transport processes are less important for electrons excited at the surface layer than for electrons excited at subsurface and deeper layers, we correlate the enhancement (45%) of the ESP for $\alpha = 1°$ to Stoner excitations across the ferromagnetic exchange gap which occur during inelastic exchange scattering of minority-spin electrons. We note that the role of Stoner excitations was successfully discussed in the literature to explain electron- and ion-induced secondary electron spectra [9,10,16]. For $\alpha = 8°$ and 45°, we find ESP enhancements of 70 and 80%, which can be explained by taking in addition into account the spin dependence of the electron mean free path and assuming that the mean free path is approximately 3 atomic distances.

At present, due to the use of polycrystalline surfaces in our first SPEES experiments with Fe, we refrain ourselves from discussing the fine structure of our ESP spectra, which can be associated with details of the spin-polarized, *k-dependent* band structure above the vacuum level [17].

Finally, we discuss the behavior of the ESP of electrons emitted from O-covered Fe surfaces (see also figs. 1 and 2). For high-energy electrons, the ESP amounts to 32% and remains unchanged for $\alpha = 8°$ and reduces to 25% for $\alpha = 45°$. For low-energy electrons, the ESP decreases from 48%, obtained at the clean Fe surface for $\alpha = 1°$, to approximately -14% for $\alpha = 1°$, 14% for $\alpha = 8°$ and 15% for $\alpha = 45°$. These findings clearly reveal the absence of a magnetically dead layer at the surface and suggests the existence of spin-split electronic bands in the occupied and unoccupied parts of the band structure of O-covered Fe surfaces [11,18,19].

In conclusion, we state that the experiments presented here give clear evidence that SPEES is a powerful technique to study *layer-dependent* magnetic properties. Provided that a refined theory of SPEES is available, the analysis of ARED and especially ESP spectra from angle-resolved SPEES experiments can give more detailed information about the layer-dependence of magnetic properties of clean and O-covered surfaces and also important and fundamental information about electron excitation processes at magnetic surfaces.

This research was supported by the National Science Foundation, by the Welch Foundation and by the Texas Higher Education Coordinating Board.

References

[1] C.L. Fu, A.J. Freeman and T. Oguchi, Phys. Rev. Lett. 54 (1985) 2700.
[2] J.W. Krewer and R. Feder, Physica B (1991) 135, and refs. cited therein.
[3] S.S.P. Parkin, N. More and K.P. Roche, Phys. Rev. Lett. 64 (1990) 2304.
[4] C. Rau, J. Magn. Magn. Mater. 30 (1982) 141.
[5] Y. Wang, P.M. Levy and J.L. Fry, Phys. Rev. Lett. 65 (1990) 2732, and refs. cited therein.
[6] C. Rau, K. Waters and N. Chen, Phys. Rev. Lett. 64 (1990) 1441.
[7] C. Rau, N.J. Zheng, M. Rösler and M. Lu, in: Ionization of Solids by Heavy Particles, ed. R. Baragiola (Plenum, New York, 1992) in press.
[8] R.A. Baragiola, E.V. Alonso and A. Oliva-Florio, Phys. Rev. B 61 (1979) 121.
[9] M. Landolt, in: Polarized Electrons in Surface Physics, ed. R. Feder (World Scientific, Singapore, 1985) chap. 9, and refs. cited therein.
[10] J. Kirschner, in: Surface and Interface Characterization by Electron Optical Methods, eds. A. Howie and U. Valdre (Plenum, New York, 1988) p. 297.
[11] M. Landolt, Appl. Phys. A41 (1986) 83, M. Landolt, R. Allensbach and M. Taborelli, Surface Sci. 178 (1986) 311.
[12] C. Rau and K. Waters, Nucl. Instr. and Meth. B40 (1989) 127.
[13] J. Kirschner, K. Koike and H.P. Oepen, Phys. Rev. Lett. 59 (1987) 2099.
[14] J. Kirschner, K. Koike and H.P. Oepen, Vacuum 41 (1990) 818.
[15] D. Penn and P. Apell and S.M. Girvin, Phys. Rev. Lett. 55 (1985) 518.
[16] J. Glaser and E. Tosatti, Solid State Commun. 52 (1984) 905.
[17] E. Tamura and R. Feder, Phys. Rev. Lett. 57 (1986) 759.
[18] A. Clarke, N.B. Brookes, P.D. Johnson, M. Weinert, B. Sinkovic and N.V. Smith, Phys. Rev. B 41 (1990) 9659.
[19] J. Chen, M. Drakaki and J.L. Erskine, Phys. Rev. B 45 (1992) 3636.