X-ray resonant exchange scattering from 3d transition metal surfaces

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Abstract

The decay of the elastic X-ray intensity peak versus perpendicular transferred momentum is theoretically studied at energies close to the L_{II} edge, for single crystal Fe surfaces and thin Fe films. For the single crystal surface, large intensity changes are observed upon reversal of the direction of the magnetisation. For thin Fe films, which are known to exhibit perpendicular magnetisation, analogous effects are predicted when, by raising the temperature, the magnetisation becomes in-plane. These changes are sensitive to both direction and magnitude of the surface magnetic moments, making this technique a powerful probe for studying the magnetic properties of 3d surfaces and interfaces.

Keywords: Computer simulations; Iron; Low index single crystal surfaces; Magnetic films; Magnetic interfaces; Magnetic phenomena; Magnetic surfaces; Metal–metal interfaces; Semi-empirical models and model calculations; Superlattices

1. Introduction

The recent development of X-ray resonant exchange scattering (XRES) [1] for the study of magnetic properties and of X-ray crystal truncation rod (CTR) analysis [2,3] for the determination of surface structures, makes it possible to conceive the study of surface magnetic properties by means of X-rays. In a recent theoretical paper [4] a combination of these two techniques has been proposed, and shown to be capable of probing the surface magnetic order of 4f materials via the CTR analysis of the in-plane diffraction spots.

Since the detection of the magnetic order goes via a resonant process, the photon wavelength has to be close to an atomic transition from a spin–orbit split core level and, at the same time, be suitable for diffrac-

tion. The low energy of the L_{II} and L_{III} edges in 3d materials does not satisfy the latter condition. On the other hand, for these materials (Fe, Co, Ni) a different behaviour of the surface magnetization with respect to the bulk is generally observed, making them interesting both from a fundamental and a technological point of view.

In this paper, we aim at showing that it is possible to obtain detailed magnetic surface information by analysing the decay of the elastic peak versus perpendicular transferred momentum at resonance, also for 3d transition metals.

2. Theory

Hannon et al. [1] derived the expression for the total amplitude of coherent elastic scattering to leading order in the weakly relativistic limit:

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$$f = -Zr_0 e_f^* \cdot e_i + f' + if'', \qquad (1)$$

with, in the electric dipole approximation and neglecting crystal field effects:

$$\begin{aligned}
f_{XRES} &= f' + if'' \\
&= \frac{3}{8}\frac{\lambda}{\pi}\Big[\big(F_{11} + F_{1-1}\big)\,(e_f^* \cdot e_i) \\
&\quad - i\big(F_{11} - F_{1-1}\big)\,(e_f^* \times e_i) \times u \\
&\quad + \big(2F_{10} - F_{11} - F_{1-1}\big)\,(e_f^* \cdot u)(e_i \cdot u)\Big],
\end{aligned}$$

$$(2)$$

where e_i and e_f represent, respectively, the ingoing and outgoing photon polarization, u is the direction of the local quantization axis of the magnetic moments and F_{LM} is proportional to the transition probability of order L ($L = 1$ for dipole transitions) divided by a resonant denominator, which is responsible for the huge intensity enhancement. The first term is the usual anomalous charge scattering amplitude, and the second and third term give the magnetic contributions to the resonant exchange scattering amplitude.

Furthermore, it has been shown [2,3] that it is possible to obtain very precise structural surface information by studying the diffraction intensity in between Bragg peaks in the direction of perpendicular momentum transfer, the so-called crystal truncation rod (CTR) analysis. Fasolino et al. [4] have shown that it is possible to extend this technique to study surface magnetism in rare earths. They study the CTR's associated with magnetic reflections at resonance in antiferromagnetic UAs and ferromagnetic Gd and show that the resulting spectra are very sensitive to both direction and magnitude of the surface magnetic moment.

The long photon wavelength at the L_{II} and L_{III} edges of 3d transition metals does not allow to perform diffraction but only reflectivity measurements (which correspond to forward scattering). This way, by changing the energy of the incoming photons [5] or the incident angle, one probes the Ewald sphere in reciprocal space, with radius $q_z = \frac{4\pi}{\lambda}$ along the direction $(0,0,q_z)$, perpendicular to the surface. The decay of the elastic peak intensity versus the perpendicular transferred momentum at energies close to resonance, will yield information about the surface in much the same way as with CTR-analysis. Information about the surface magnetism will emerge around those point

in k-space where the charge scattering is close to zero, since then the magnetic terms dominate the total scattering cross section (1), i.e., at values $q = (0,0,q_z)$, around:

$$q_z = \frac{2\sqrt{2}\pi}{\lambda}, \qquad (3)$$

where $(e_f^* \cdot e_i)$ is zero for p-polarized light.

A description of the resonant process by means of Eq. (2) implies assuming an atomic picture of the magnetic ion. This is not fully appropriate for 3d metals where band structure effects lead to a fractional occupation of the d-shell. In the case of Ni, it has been shown that the ground state of a Ni atom in ferromagnetic nickel can be described as a superposition of several 3d configurations [6]. In the following we assume a Fe^{2+} atomic picture for the ground state. Therefore, our results are only qualitative and a detailed comparison with experiments will require a more sophisticated description of the magnetic ground state.

3. Results and discussion

We analyse the decay of the elastic peak intensity versus perpendicular transferred momentum at energies close to the L_{II} absorption edge (727 eV), where the largest effects have been observed in bulk Fe [5]. The F_{1M} matrix elements are obtained using Cowan's Hartree–Fock and multiplet programs (with relativistic corrections) [7]. We first consider the Fe(001) surface and calculate the scattering intensity with a magnetic field applied perpendicular to the scattering plane in opposite directions. Fig. (1) shows the reflected intensity as a function of q_z for the case of pure charge scattering and when the magnetic contributions to the cross-section are taken into account. One can see that the intensity drops off dramatically near $q_{min} = 0.517$ Å, but also that there is a substantial influence of the magnetic scattering near this point in reciprocal space. The change in intensity upon reversal of the external magnetic field gives rise to the asymmetry ratio

$$R_\pm = \frac{I^- - I^+}{I^- + I^+}, \qquad (4)$$

which we show in Fig. 2 as a function of q_z. I^+ is the intensity of the reflected beam when the magnetisa-
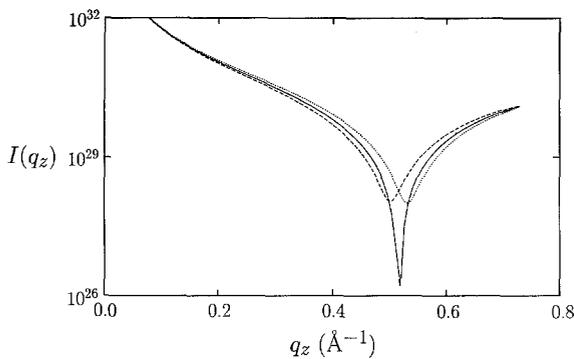
Fig. 1. Decay of the elestic peak intensity $I(q_z)$ at the L_{II} edge ($E = 727$ eV) versus perpendicular transferred momentum q_z. $I(q_z)$ is in arbitrary units. The scattering plane lies along the [100] direction. Thick solid line: resonant charge scattering. Thin solid line: magnetization u in the same direction as ($e_f^* \times e_i$). Dotted: u in the opposite direction.

tion vector u points in the same direction as the cross product of the polarization vectors ($e_f^* \times e_i$); I^- is the intensity for reversed magnetisation.

At the point q_{min}, the asymmetry ratio is zero, but interference effects with the charge scattering terms in the cross section (1) cause $R_\pm$ to rise to considerable values at a distance of about 0.05 Å^{-1} around q_{min}. It has been well-established by self-consistent spin-polarized electronic structure calculations [8–13] and confirmed by photoemission experiments [14,15], that the reduced coordination at a surface
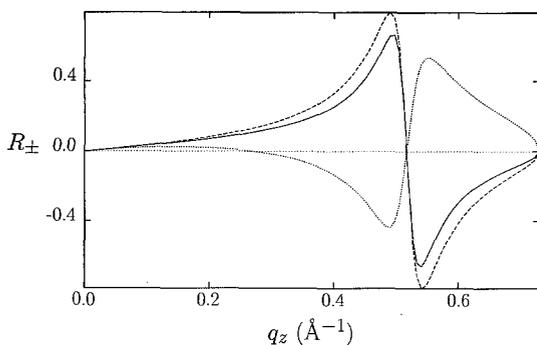

Fig. 2. Asymmetry ratio of the $(0,0,q_z)$ reflection of the Fe (100) surface. Thick solid line: Surface magnetisation equals bulk magnetisation. Dotted line: Enhanced surface magnetisation. Thin solid line: Magnetisation of topmost surface layer reversed. See text.
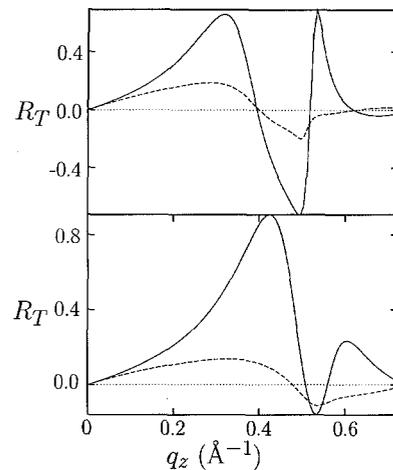

Fig. 3. Temperature asymmetry ratio of the $(0,0,q_z)$ reflection of thin Fe films on Ag(100). Upper figure: 7 Fe layers; lower figure: 5 Fe layers. Solid line: for the in-plane contribution the surface magnetisation vector u_S points in the same direction as ($e_f^* \times e_i$); dotted line opposite direction

generally leads to enhanced magnetic moments. We will use the surface magnetic moments as they are calculated in Ref. [13], who give for the magnitude of the magnetisation of the uppermost surface layers $\mu_S^i = (2.97, 2.35, 2.43, 2.25, 2.24)$, for $i = 1$ through 5, respectively. Starting from the 6th layer the magnetisation retains its bulk value $\mu_B = 2.27$. We mimic the change in surface magnetic moments by multiplying the resonant matrix element in each surface layer by a factor μ_S^i/μ_B. The resulting asymmetry ratio is also shown in Fig. 2. In order to determine the sensitivity of the asymmetry ratio to the direction of the surface magnetic moments, we also show $R_\pm$ when the magnetisation in the topmost Fe-layer is reversed.

We have also studied the asymmetry ratio for epitaxial α-Fe grown on Ag(100). Several experiments [16–20] have shown that the orientation of the magnetic moment for thin Fe films (1 to 7 monolayers) is perpendicular to the film plane. In this case, due to the fact that the cross-product of the polarization vectors in the scattering configuration is in-plane, the occurrence of perpendicular magnetic moments is only reflected in the asymmetry ratio through interference with the scattering from in-plane moments. Since they are not present in this system a zero asymmetry ratio is obtained. However, Qiu et al. [21] have shown
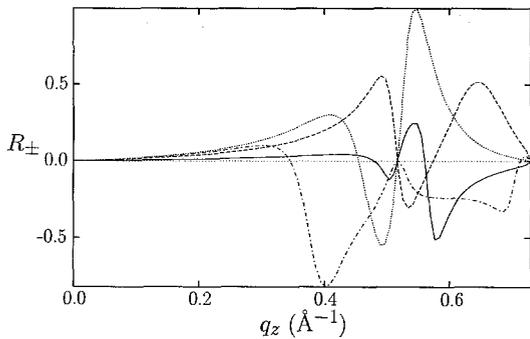
Fig. 4. Asymmetry ratio of the $(0,0,q_z)$ reflection of Fe_nAg_m superlattices. In-plane magnetisation. For the in-plane contribution the surface magnetisation vector u_S points in the same direction as $(e_f^* \times e_i)$. Thin solid line: $(n,m) = (1,5)$; thin dotted line: $(n,m) = (3,3)$; thick solid line: $(n,m) = (5,3)$; Thick dotted line: $(n,m) = (5,5)$.

by means of the magneto-optical Kerr effect that at a temperature $T_R < T_C$, the critical temperature, the magnetisation switches from perpendicular to parallel to the surface. Therefore, we can define the temperature asymmetry ratio

$$R_T = \frac{I_{(T<T_R)} - I_{(T>T_R)}}{I_{(T<T_R)} + I_{(T>T_R)}}, \tag{5}$$

where T is the temperature and T_R is the temperature at which the in-plane remanent magnetic component starts to develop.

When calculating $I_{(T<T_R)}$ (perpendicular magnetisation) only the magnetic term which is quadratic in the moment direction u contributes to the XRES scattering amplitude ; for $I_{(T>T_R)}$ (in-plane magnetisation) only the linear term in u contributes because in this type of experiment u is perpendicular to the scattering plane. A complication in the interpretation might come from the fact that the F_{LM} transition probabilities may change when raising the temperature. In Fig. 3 we show the temperature asymmetry ratio for two different layer thicknesses. It is clear that the temperature asymmetry ratio is very sensitive to the occurrence of perpendicular magnetic moments.

Finally, we show the asymmetry ratio of Fe_nAg_m superlattices for $(n,m) = (1,2)$, $(3,3)$, $(5,3)$ and $(5,5)$ assuming in-plane magnetisation for the Fe layers. In Fig. 4 one can see that the influence of the superlattice composition on the shape of the asymmetry ratio

curve is large.

In conclusion we have shown that XRES can yield information on the surface magnetic structure of 3d metals. The change in reflected intensity is much more sensitive to the direction of the individual surface or interface magnetic moments than it is to their magnitude. Therefore, one can anticipate that the technique we propose will be most suited for the study of magnetic reconstructions or interlayer oscillatory magnetic coupling.

Acknowledgements

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