Photoelectron diffraction in magnetic dichroism: Surface live magnetic layers in fcc Fe/Co(001)

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We combine photoelectron diffraction and magnetic dichroism in photoemission to obtain magnetic and structural information regarding ultrathin films on an element-resolved basis. Crystallinity-induced effects in the magnetic dichroism of 2*p* core level photoelectrons present valuable structural information regarding magnetically inhomogeneous films. The location of ferromagnetic layers in structurally relaxed fcc Fe films (eight monolayers) grown at room temperature on Co/Cu(001) is found to be at the surface, with nonferromagnetic layers underneath. Tetragonally distorted fcc Fe films (one and three monolayers) are found to be entirely ferromagnetic. [S0163-1829(98)05348-X]

Magnetic and structural properties of magnetic thin films are intimately connected, and a considerable amount of experimental and theoretical work has been dedicated to studying this interrelation.[1] Especially in ultrathin films these properties may vary over the thickness of the film. In particular, it is known that both the structure and magnetism of film interfaces and surfaces may very well deviate from those of the interior of the film.[2] This becomes even more important when a film is in contact with another ferromagnetic film. For the study of such systems a method is thus needed that permits us to obtain information about the distribution of magnetic properties within a single film of the same element.

One possibility for achieving this is to make use, on the one hand, of the structural information intrinsic to the angular distribution of photoelectrons, which is governed by scattering events of the photoemitted electrons at the surrounding atom cores. This effect can be employed for structural investigations (photoelectron diffraction).[3] The change of the spectral intensity distribution of the photoelectrons upon magnetization reversal, on the other hand, yields information about the magnetic properties; in the case when core levels are probed, even with elemental resolution. This is termed magnetic dichroism in photoemission, and is a widely used technique for magnetic characterization.[4–12] It seems obvious that the combination of both photoelectron diffraction and magnetic dichroism, which implies recording the angular *and* spectral distribution of photoelectrons,[13,14] should allow us to study structural and magnetic properties of thin films and surfaces at the same time, i.e., give structural information on a magnetic basis, or magnetic information on a structural basis.

We have chosen ultrathin γ-Fe films on Co/Cu(100) to demonstrate the applicability of this approach. Fcc-Fe, which can be stabilized in ultrathin films by the choice of appropriate substrates, is a system in which striking differences between magnetic properties of film surface and interior have been observed. Fe films of a certain thickness, deposited at room temperature directly on Cu(001), exhibit a nonferromagnetic behavior of the inner film layers, but ferromagnetism at the film surface.[15–18] Bringing this film into contact with a ferromagnetic underlayer, in our case Co, raises the question of the magnetic phases that will then be present in the fcc Fe overlayer. It is particularly interesting to see whether a magnetic material, Fe, may possibly be nonferromagnetic in contact with another magnetic material. If this should be the case it has to be checked whether there is also a ferromagnetic layer at the surface. Previous studies using the magneto-optical Kerr effect (MOKE) and magnetic circular dichroism in x-ray absorption (XMCD) have indeed shown that in a thickness interval between 5 and 11 ML Fe films on top of Co/Cu(001) exhibit a strongly reduced integral magnetic signal compared to that of thinner or thicker films.[19–21] From oxygen adsorption experiments[19,20] and XMCD (Ref. 19) it has been concluded that the observed remaining Fe ferromagnetic response should be due to deeper layers around the Fe/Co interface. We will show unambiguously in this paper that the observed decrease of ferromagnetic response in Fe/Co/Cu(001) is neither due to a nonferromagnetic film on top of an induced-ferromagnetic interface nor to a uniform reduction of Fe magnetic moment. We can instead show that these films consist indeed of a ferromagnetic layer on top of nonferromagnetic Fe underlayers, ferromagnetically aligned to the Co substrate.

The idea of our experiment is sketched in the inset of Fig. 1(a). The angle between the incident photons ($\mathbf{q}$), and the outgoing photoelectrons ($\mathbf{k}$) is fixed at 45°. The magnetization $\mathbf{M}$ is switched between up and down, being perpendicular to the scattering plane defined by $\mathbf{q}$ and $\mathbf{k}$. The angle α between the surface normal $\mathbf{n}$ and the electron detection direction ($\mathbf{k}$) is then varied by rotating the sample about the magnetization axis. Thereby the rotation of the sample does not change the angles between the three vectors $\mathbf{q}$, $\mathbf{k}$, and $\mathbf{M}$, which govern the magnetic dichroism signal in the atomic limit.[22] For a structurally disordered sample, or for free atoms, a variation of α would thus have no influence on the measured dichroism. Any change of the dichroism as a function of α must be related to the crystallinity of the sample, represented by the surface normal $\mathbf{n}$,[5] and can be considered a consequence of photoelectron diffraction.[12] Recent experiments have demonstrated the magnitude of this effect, which in the presented geometry can even lead to a sign reversal of the observed dichroism signal by photoelectron diffraction.[13] In the energy range covered by our experiment ($\approx 450-550$ eV electron kinetic energy) photoelectron diffraction due to forward scattering dominates by far,[3,13] caus-

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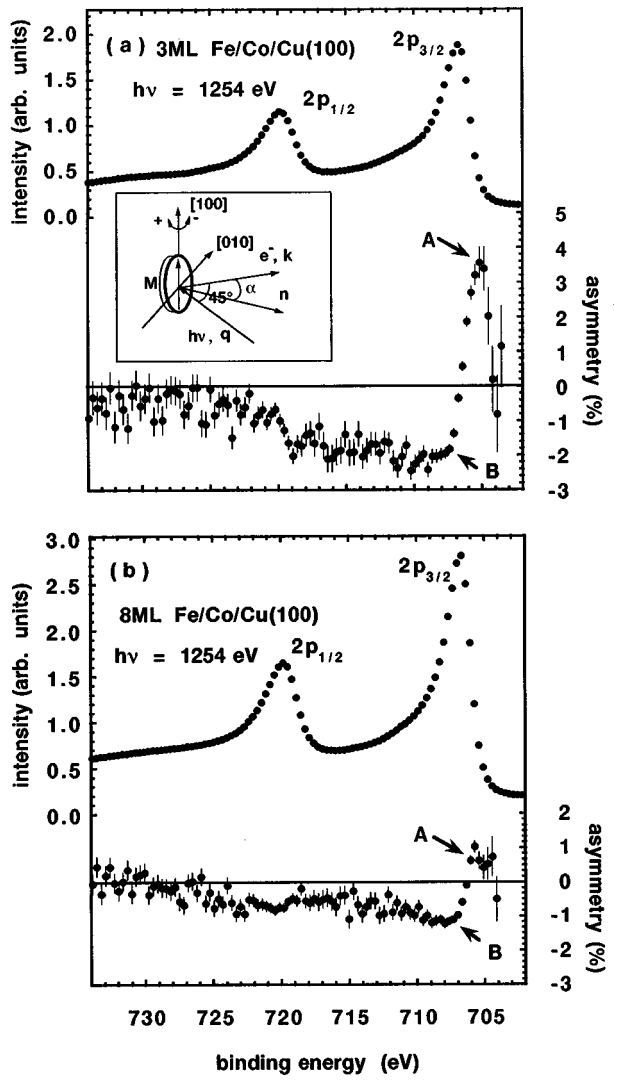

FIG. 1. (a) Fe 2*p* photoemission spectra and dichroic asymmetry of 3 ML Fe/5 ML Co/Cu(100) for an emission angle $\alpha = 0°$. Inset: Experimental setup, showing the directions of the magnetization **M**, light incidence **q**, electron emission **k**, and surface normal **n**. (b) Fe 2*p* photoemission spectra and dichroic asymmetry of 8 ML Fe/5 ML Co/Cu(100) for an emission angle $\alpha = 0°$.

ing diffraction effects in thicker films, whereas in monolayer films this diffraction effect becomes negligible, and a more atomiclike behavior is observed. This has been checked to be true in a thickness-dependent study of Co films, which will be published elsewhere.[14]

The experiment was carried out using a Mg-K_α x-ray tube ($\hbar\omega = 1253.6$ eV), and a hemispherical electron energy analyzer with an angular resolution better than 3°. The overall energetic resolution of the experiment was about 1 eV, and the angular reproducibility of the sample manipulation 0.2°. Co and Fe films were grown on a Cu(001) substrate at room temperature by electron bombardment of cobalt and iron wires of high purity. Photoemission spectra were taken as two interleaved sequences of individual scans with opposite magnetization directions along the [100] direction of the substrate. The magnetization was reversed before each scan by a magnetic field generated by a current pulse through a coil inside the chamber. The films were checked to be in full magnetic saturation by MOKE. Hysteresis loops measured in the longitudinal geometry exhibited a rectangular shape, and showed a reduction of the Kerr signal of about 15% when going from 3 ML to 8 ML Fe on 5 ML Co/Cu(001), which is consistent with results of other groups.[20,21] Photoemission spectra of 1 and 3 ML Fe/5 ML Co were recorded at room temperature, and spectra of 8 ML Fe/5 ML Co both at room temperature and 200 K. We found no significant deviation of the spectra for both temperatures; here we present the latter because of better statistics. We report the dichroism in terms of the intensity asymmetry, defined as $(I_+ - I_-)/(I_+ + I_-)$, where I_+ and I_- are the photoemission intensities for opposite sample magnetization.

Figure 1(a) displays photoemission spectra of the Fe 2*p* level for 3 ML Fe on Co/Cu(001) at $\alpha = 0°$ obtained by summing I_+ and I_- (topmost curve). Depicted are the raw data with just a constant background subtracted. The corresponding dichroic asymmetry is shown below. Its shape is in good agreement with asymmetries presented in the literature.[4,5,10,11] A prominent plus/minus feature at the $2p_{3/2}$ level is observed (labeled *A* and *B*), while at the $2p_{1/2}$ level a weaker minus/plus feature appears. The absolute value of the asymmetry of the latter in the present measurement is somewhat lower compared to, for example, Fig. 1 of Ref. 10, which was measured on bcc Fe with Al-K_α radiation. This may be related to experimental uncertainties in the determination of the asymmetry baseline, which is very sensitive to, e.g., small variations of x-ray intensity. For a quantitative analysis of the asymmetry we therefore used the peak-to-peak asymmetry at the $2p_{3/2}$ level (difference of asymmetry at position indicated by *A* and *B* in Fig. 1), which is largely independent of baseline uncertainties.

Results of the 8 ML Fe/Co film are qualitatively very similar to those of the 3 ML film, but show a higher intensity and a smaller dichroism. This is seen from Fig. 1(b), where Fe 2*p* photoemission intensity and asymmetry of 8 ML Fe/5 ML Co/Cu(001) are shown on the same scales as for 3 ML at the same emission angle $\alpha = 0°$. The measured increase of the $2p_{3/2}$ intensity between 3 and 8 ML of Fe by a factor of 1.55 ± 0.05 in the intensity corresponds to a mean free path of the 500 eV photoelectrons in Fe of 3.5 ± 0.4 ML ($\approx 6-7$ Å), in good agreement with previously published values.[23] In contrast, the asymmetry should stay the same if nothing but the thickness was changed. The decrease of the asymmetry proves that the decrease of the MOKE signal (which integrates over the whole layer stack) is due to a loss of net magnetization in the Fe film at higher thickness. The comparison of the sign of Fe 2*p* with the Co 2*p* dichroic asymmetry (not shown here) shows that the net Fe magnetization is aligned parallel to the magnetization of the Co underlayers. This rules out the possibility that an antiferromagnetic coupling of the Fe film as a whole to the Co layer causes the reduction of the Kerr signal, and confirms that a significant portion of the Fe film is not ferromagnetic.

To obtain structural information from photoelectron diffraction the electron emission angle α with respect to the surface normal is now varied, and the behavior of both the photoemission intensity and asymmetry as a function of α is measured. The *intensity* as a function of α is the usual photoelectron diffraction effect,[3] and contains element-selective structural information about the Fe film. Since in our geom-

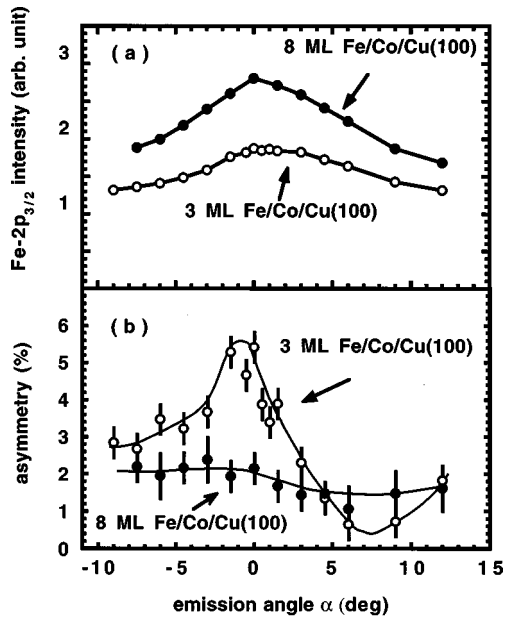

FIG. 2. (a) Intensity of the Fe-$2p_{3/2}$ photoemission peak as a function of the emission angle α for 3 ML Fe (open circles), and 8 ML Fe (filled circles) on 5 ML Co/Cu(001). (b) Fe-$2p_{3/2}$ peak-to-peak asymmetry between points *A* and *B* (cf. Fig. 1) as a function of the emission angle α for 3 ML Fe (open circles), and 8 ML Fe (filled circles) on 5 ML Co/Cu(001). The lines in (b) are guides to the eye.

etry (cf. inset of Fig. 1) a variation of α does not change the relative orientation of light incidence, magnetization, and electron emission, the resulting angular variation of the magnetic *asymmetry* is due to crystallinity induced effects only, and thus apart from the magnetic information contains also structural information about the magnetic layers only. For evenly magnetized films, the angular variation in the case of 8 ML Fe is expected to show qualitatively the same behavior as for 3 ML, with possibly stronger effects due to the higher number of layers.

In Fig. 2 this dependence of the Fe-$2p_{3/2}$ intensity (a) and peak-to-peak asymmetry (b) on the emission angle α is shown. Filled and open circles refer to 3 and 8 ML Fe on Co/Cu(001), respectively. Both films exhibit an intensity maximum at normal emission ($\alpha=0°$), indicative of [001] forward scattering in epitaxially ordered films. From inspection of the angular behavior of the corresponding asymmetries [Fig. 2(b)], the following important observation becomes obvious: Whereas the 3 ML film displays a strong angular variation of the dichroism, it is essentially constant in the case of 8 ML Fe, even if a very weak modulation may still be present. As discussed before, a strong angular variation of the dichroism is due to diffraction effects, related mainly to forward scattering. Compared to the case of 3 ML Fe, this angular variation is strongly damped in the 8 ML Fe film, where the experiment reveals an almost vanishing dependence of the asymmetry on the emission angle α with a value of about 1.6% [Fig. 2(b)]. This cannot be attributed to structural disorder in the film, since the Fe-$2p_{3/2}$ photoemission intensity [Fig. 2(a)] shows a clear maximum at normal direction. The portion of Fe photoelectrons which contribute to the dichroism, i.e., which stem from ferromagnetically ordered Fe atoms, are thus not subject to forward scattering. They must consequently originate from atoms in the topmost two layers at the surface, because in an fcc(001) crystal these are the only ones that do not have scatterers in the [001] emission direction. This leads to the conclusion that within the probing depth of our experiment the ferromagnetism of the 8 ML Fe film is restricted to the two topmost atomic layers. The absence of any diffraction effects in the magnetic dichroic asymmetry of the 8 ML Fe/5 ML Co/Cu(001) sample shows thus unambiguously that there is a magnetic live layer *at the surface* of the Fe film on nonferromagnetic underlayers.

The weak modulation still visible in the curve of 8 ML Fe in Fig. 2(b) may be a hint towards the presence of some amount of ferromagnetic Fe in deeper layers, e.g., at the Fe/Co interface. It may, however, also be attributed to the minor contribution of nonforward scattering to the photoemission signal, the size of which at the present electron energies is expected to be about one order of magnitude weaker than that of forward scattering.[3,13]

As a final consideration, one could think that the focusing effect of forward scattering, which leads to the intensity enhancement at normal emission [cf. Fig. 2(a)] similarly should lead to a reduction of the asymmetry around $\alpha=0°$. In the simple picture, where one assumes the enhancement of forward emission to be solely due to photoelectrons from deeper, and, in our interpretation, nonferromagnetic layers, such a reduction should be the inverse of the intensity curve of Fig. 2(a), and amount to about 35%. The experimental points of the 8 ML Fe film in Fig. 2(b) do not seem to follow such a reduction at normal emission; the experimental uncertainty, indicated by the error bars, however, may easily mask such an effect.

For a more quantitative analysis of the present data we compare the dichroism of the 8 ML Fe/Co film, which shows no diffraction effect and represents thus the atomiclike contribution, with the atomiclike contribution of the 3 ML Fe film, which is entirely magnetized. For that we assume that in the 3 ML film the diffraction effect imposes a modulation on the dichroism as a function of the emission angle around the atomiclike contribution.[13,14] Taking hence the average between the extrema of the 3 ML curve of Fig. 2(b) as the atomiclike contribution yields about 3.2%. This also agrees with the dichroism of a 1 ML Fe film where we have measured, in a similar experiment, a value of 3.4%, and with the dichroism calculated in Ref. 24. The dichroism in photoemission from a magnetic layer of thickness t_{mag} is reduced by a factor of $[1-\exp(-t_{mag}/\lambda)]/[1-\exp(-t_{tot}/\lambda)]$ if this layer sits on top of a nonferromagnetic film of thickness t_{nm} of the same material ($t_{tot}=t_{mag}+t_{nm}$). Comparing the dichroism in 8 ML Fe ($\approx 1.6\%$) and the dichroism for the atomic model ($\approx 3.2\%$), this factor can be calculated to be about 0.5. Taking $\lambda=3.5$ ML as before, for $t_{tot}=8$ ML we can calculate the thickness of the magnetic layer t_{mag} to be about 2 ML. There is, however, a considerable uncertainty in that estimate because of the separation of atomic and diffraction contributions, possible differences in the magnetic moments, and in 2*p* multiplet splitting in the different films.

It has been proposed that a magnetic ordering could be induced in the bottom Fe layer of Fe/Co/Cu(001) due to coupling to the Co at the Co/Fe interface.[19–21] Because in 8 ML Fe the signal of the bottommost Fe layer contributes

only about 4% to the total Fe photoemission intensity, we cannot exclude this possibility of ferromagnetism in the bottom layer from the photoemission measurements. From comparison of our MOKE data of 0, 3, and 8 ML Fe films on Co/Cu(001), however, the total amount of ferromagnetic Fe in the 8 ML film can be determined to be 1.8 ± 0.3 ML equivalent. Under the assumption that the Fe film is saturated by the lower fields of the MOKE coil (≈ 1500 Oe) as it is by the field pulses applied during the photoemission measurements, there can be only a very small contribution to the Fe ferromagnetism from the Fe/Co interface. What can unequivocally be excluded is the possibility of a nonmagnetic surface, and the possibility of a fully magnetized Fe film with a reduced magnetic moment. Both the absence of diffraction in magnetic dichroism in angle-resolved photoemission, and the strength of the dichroism signal are only consistent with the presence of ferromagnetic ordering at the surface, and no ferromagnetic ordering in deeper layers.

In summary we have shown that studying diffraction effects in magnetic dichroism in photoemission provides a new tool to obtain structural and magnetic information simultaneously, on an element resolved basis. This method is shown to be particularly useful to study magnetism of thin films, where the use of techniques which provide integral information are often not sufficient. We have employed this method to investigate fcc Fe films grown on 5 ML fcc Co/Cu(001). The different angular behavior for 3 ML and 8 ML Fe films shows clearly that 3 ML are entirely magnetized, ferromagnetically aligned with the Co substrate, whereas in 8 ML there is a ferromagnetic layer located at the surface with nonferromagnetic layers underneath. We find thus the magnetic phases of fcc Fe/Co/Cu(100) to be qualitatively identical to the ones of Fe/Cu(100).

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[1] See, for example, *Magnetism and Structure in Systems of Reduced Dimension*, Vol. 309 of *NATO Advanced Study Institute Series B: Physics*, edited by. R. F. C. Farrow, B. Dieny, M. Donath, A. Fert, and B. D. Hermsmeier (Plenum Press, New York, 1993), and references therein.

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