

Structure and local magnetic moments of epitaxial $\text{Fe}(100)_{100-x}\text{Co}_x$ alloy films determined by photoelectron diffraction and magnetic linear dichroism

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We discuss the relation between a photoelectron diffraction (PED) related core-level magnetic-field-induced intensity asymmetry (“magnetic dichroism”) and the dependence of the structure and the local magnetic moments on $\text{Fe}_{100-x}\text{Co}_x/\text{Ag}(001)$ epitaxial ultrathin alloy films. Data are obtained using a standard laboratory x-ray source. The $2p$ core levels of Fe and Co at any of the alloy compositions show PED patterns indicative of bcc environments of the emitter atoms. The magnetic dichroism of the Fe $2p_{3/2}$ core level increases with increasing x , whereas it is virtually constant for the Co $2p_{3/2}$ level. Both dependencies scale well with the calculated variation of the local magnetic moment with alloy composition. [S0163-1829(98)04725-0]

I. Introduction. The interplay between structural and magnetic properties is of great interest for testing theoretical predictions and for optimizing magnetic devices based on thin-film technology. A very important property of magnetic materials is the magnetic moment of its constituents. Frequently, core-level photoelectron spectroscopic methods are used to study the local magnetic moments. Element specificity is due to the specific binding energies of the core levels. The large and easily observable splitting in the Fe and Mn $3s$ core-level photoelectron spectra was attributed to arise from the exchange interaction between the unpaired $3d$ valence electrons and the core electrons.¹ However, van Acker *et al.*² report that no relation exists between the $3s$ splitting and the magnetic moment. Jordan *et al.*,³ based on a study of FeNi alloys, arrive at the opposite result.

More specific core-level photoelectron spectroscopic methods use the fact that symmetry is broken due to the presence of the magnetization. Consequently, the core-level photoemission lines from a magnetized ferromagnet show a spin polarization.⁴ Hence more direct information on the element-specific magnetic moment⁵⁻⁷ is obtained. Whereas the aforementioned studies used synchrotron radiation, Klebanoff *et al.*⁸ demonstrated that this method can be used successfully with a standard laboratory x-ray source. However, due to the low brightness and the low efficiency of the spin detector, this method is relatively slow. With the increasing availability of synchrotron radiation, magnetic circular dichroism (MCD) in photoabsorption at core-level thresholds⁹ is frequently used currently to get information on the magnetic moment. Magnetic circular dichroism in photoemission¹⁰ also gives information on the magnetic moment. K -edge MCD in combination with magnetic extended x-ray absorption fine structure was used to investigate bcc Fe-Co alloys by Pizzini *et al.*¹¹ Braicovich *et al.*¹² have recently correlated the magnetic circular dichroism in resonant soft-x-ray inelastic scattering with the compositional dependence of the magnetic moments. Tobin *et al.* used MCD to study an fcc $\text{Fe}_3\text{Co}_1/\text{Cu}(001)$ sample.¹³

The photoemission methods described so far require sophisticated experimental equipment such as a synchrotron (to obtain the circularly polarized radiation) or a spin analyzer.

An alternative but quite efficient method is based on the so-called magnetic linear dichroism in angle-resolved photoemission (“MLDAD”):¹⁴ the photoemission intensity with linearly p -polarized radiation depends on the direction of the magnetization. Hillebrecht and Herberg¹⁵ showed that the MLDAD persists when using *unpolarized* radiation from a standard laboratory x-ray tube, due to the presence of the p -polarized component.

In an atomic picture, the MLDAD is proportional to the atomic magnetic moment.¹⁶ In single-crystalline materials, photoelectron diffraction (PED) gives rise to a new kind of intensity asymmetry, as we have shown recently.^{17,18} At photoelectron energies above 400 eV this is even the major source for the asymmetry.¹⁸ The intensity asymmetry shows a left-to-right asymmetry with respect to forward scattering peaks, which are observed along low-index crystallographic directions, i.e., the dichroism oscillates as a function of the emission angle. This effect is easily accessible when studying $3p$ and $2p$ transition-metal core levels using standard laboratory unpolarized x-ray sources (e.g., Mg or Al K_α). The intensity asymmetry pattern could be reproduced in a PED calculation,¹⁸ and a simple heuristic explanation of the effect is given there. Since the peak-to-peak asymmetry is of the order of 10%, it is experimentally relatively accessible and can be used for element-specific magnetometry. By taking energy distribution curves (EDC’s) for a large range of emission angles and for each magnetization direction, the PED pattern and the dichroism are measured simultaneously. They together provide information on the crystal structure and on magnetic properties. In the present work, we apply this method to ultrathin-film alloys. We chose the well-known bcc $\text{Fe}_{100-x}\text{Co}_x$ system, here synthesized by epitaxial growth on an $\text{Ag}(001)$ substrate. In this well-studied system, the average magnetization shows a remarkably different compositional dependence than the individual local moments. We show that the magnetic dichroism of the components agrees well with the dependencies of the local magnetic moments on the alloy composition, rather than with the average magnetization.

II. Experiment. The magnetic dichroism caused by PED is relatively large and easy to measure by a conventional x-ray photoemission spectroscopy spectrometer. We used a CLASS 150 spectrometer (manufacturer VSW), equipped

with a Mg K_α x-ray source. Figure 1 shows the experimental geometry. The spectrometer and the x-ray source make an angle of 45° with respect to each other. The magnetization is aligned perpendicular to the plane of incidence and photoelectrons emitted parallel to the incidence plane are detected. The polar acceptance direction (α) of the photoelectrons with respect to the crystal axes is varied by rotating the sample around an axis *parallel* to the magnetization direction. At each preset value of α the samples are alternatingly oppositely magnetized by short magnetic field pulses of about 80 Oe prior to taking each EDC. The films, obtained by co-evaporation of Fe and Co onto an Ag(001) substrate at room temperature from e -beam heated rods, had been chosen to be 20 atomic layers thick. Fe films of this thickness grown on Ag(100) are well known to be close to 100% remanently magnetized. From the smooth variation magnetic dichroism that we observe over the whole concentration range we conclude that the same is true for the bcc FeCo alloy ultrathin films.

The magnetic dichroism $A = (I_1 - I_2)/(I_1 + I_2)$ of the atom at the binding energy (E_b) is proportional to $\mathbf{M} \cdot (\mathbf{k} \times \mathbf{E}) \cdot (\mathbf{k} \cdot \mathbf{E})$.¹⁹ I_1 and I_2 (E_b) are the intensities obtained for the two opposite directions of the magnetic moment (\mathbf{M}), \mathbf{k} refers to the photoelectron wave vector, and \mathbf{E} to the electric-field vector. When the angle α is changed, the geometrical relation between \mathbf{M} , \mathbf{k} and \mathbf{E} remains constant. Any change in the dichroism with α will therefore be a solid-state effect.

With a change in the alloy composition the line-to-background ratio also changes. Since we are concerned with determining the intensity asymmetry of core-level lines for different alloy compositions, background subtraction is mandatory. Here, we subtract constant backgrounds as determined by the intensities at the onset of the $2p_{3/2}$ core-level EDC's prior to calculating the dichroism. Since we are evaluating the region of the EDC's in the high binding-energy region, a more sophisticated background like a Shirley background yields virtually the same results. Since A tends to diverge at energies where the background-subtracted intensity becomes small due to the increasing dominance of the statistical error, in plots displaying the binding-energy dependence we present the dichroism by plotting the intensity difference normalized to the maximum in $I_1 + I_2$. The normalized intensity difference is equal to the asymmetry at the binding energy where $I_1 + I_2$ has its maximum value. In plots showing the angular dependency of the dichroism at the binding energy where the dichroism is maximal we show the asymmetry, since the maximum occurs near the intensity maximum. From a future theory modeling the dichroism data both quantities could equally well be calculated and compared with the experimental data.

So far, there is no simple quantitative picture on the relation between structural and magnetic properties and the angular dependence of the intensity asymmetry. Therefore, a PED theory is required to extract the full amount of information from the angular-dependent data. However, the dependence of the dichroism on α can be averaged out by integrating over the emission angle. Since the left-right asymmetry is not perfect, the resulting asymmetry in the energy distribution curves is different from zero. We have shown recently that such angular integrated data obtained from Fe(001) compare well with data from an amorphous

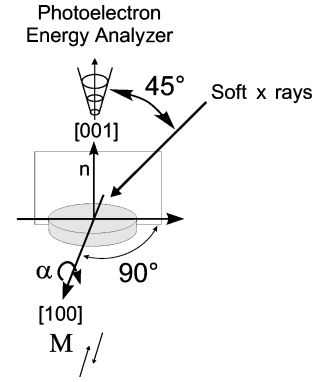


FIG. 1. The experimental geometry. Unmonochromatized Mg K_α radiation falls onto the sample at a fixed angle of 45° with respect to the acceptance cone of the photoelectron spectrometer. The radiation incidence plane is parallel to the [001] direction of the sample. The sample is rotated about the [010] direction to vary the angle (α) between the surface normal (n) and the photoelectron collection direction that defines the photoelectron wave vector (k). The magnetization is switched between [010] and [0 $\bar{1}$ 0] directions.

Fe-based metallic glass where PED effects are absent and that the energy dependence of the angular-integrated curve is close to that calculated for a single Fe atom.²⁰

III. Results and discussion. Energy distribution curves have been measured for the Fe and the Co $2p_{3/2}$ and $2p_{1/2}$ core levels for emission angles α ranging from -20° to $+70^\circ$. For any of the studied alloy compositions, we observe well-defined photoelectron diffraction patterns for each of those core levels. Because of their higher intensities, only the $j=3/2$ sublevels are evaluated here. Figure 2(a) shows the angular distributions of the photoelectron intensities ($I_1 + I_2$) and Fig. 2(b) that of the intensity asymmetries for the Fe and Co $2p_{3/2}$ sublevels for an Fe₈₄Co₁₆/Ag(001) alloy film. Well-defined peaks in the intensity are observed at emission angles corresponding to the [001] (0°), [012] (18.4°), [013] (26.6°), [011] (45°), and [021] (63.4°) forward scattering directions. The peaks corresponding to the [013] and [012] directions merge into a single broader peak. The PED patterns of the Fe and of the Co are virtually independent on the alloy composition, as seen by comparing with data obtained from an Fe₃₁Co₆₉/Ag(001) sample [Figs. 3(a) and 3(b)]. However, the Co PED angular dependence differs slightly from that of the Fe for all Co concentrations. This difference might be caused either by the different kinetic energies of the Co and Fe core-level lines or by potentially different local environments of an Fe and a Co emitter.

The asymmetry changes sign in going from $\alpha = -10^\circ$ towards $\alpha = +10^\circ$ and oscillates further with increasing angle. The pattern can be viewed as a virtually symmetrical oscillation superimposed on an offset, which decreases from about +2% at normal emission to some smaller value at large emission angles. The oscillation amplitudes at larger angles are reduced from those around normal emission ($\alpha = 0$). Since the surface sensitivity is enhanced at larger emission angles, the data suggest a diminished surface magnetization. The angular dependencies are virtually independent on the alloy composition and compare qualitatively with those observed in pure Fe/Ag(001) films.

Figures 4(a) and 4(b) show the intensity and normalized intensity difference distributions of the Fe $2p$ core level after

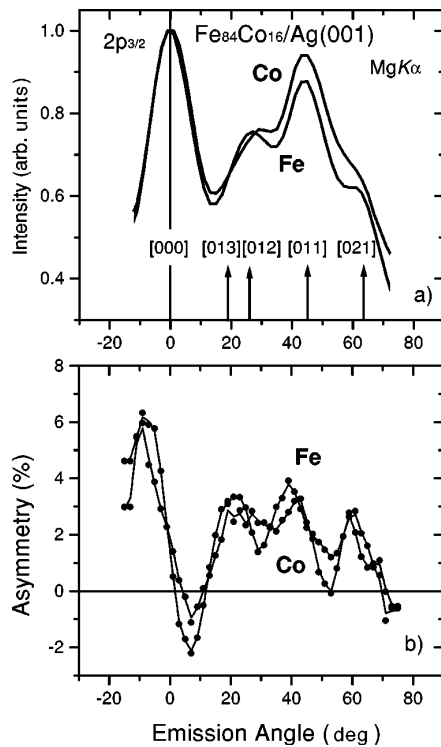


FIG. 2. The angular dependency of the magnetization-averaged photoelectron intensity (a) and of the intensity asymmetry (b) for the $2p_{3/2}$ core levels of Fe and Co, respectively, for an epitaxial $\text{Fe}_{84}\text{Co}_{16}/\text{Ag}(001)$ ultrathin film. The intensity is taken at the core-level intensity-peak binding energy whereas the asymmetry is taken at the maximum asymmetry binding energy (which is 0.5 eV less than that of the intensity peak, independent on the emission angle). Indicated are emission angles corresponding to low-index crystallographic directions.

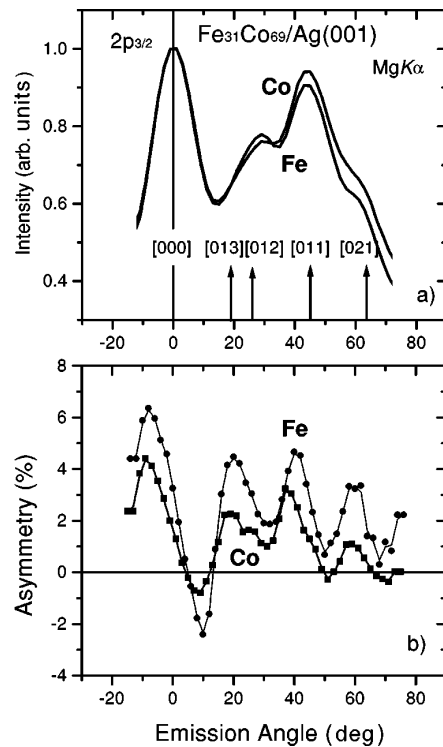


FIG. 3. The same as Fig. 2, but for an epitaxial $\text{Fe}_{31}\text{Co}_{69}/\text{Ag}(001)$ film.

having averaged over three asymmetry oscillations ($-10^\circ \leq \alpha \leq 56^\circ$). The maximum asymmetry obtained thereby amounts to about 2%, slightly larger than the value obtained from a pure Fe/Ag(001) evaporated under the same conditions. Figure 5(a) compiles the peak-to-peak normalized intensity difference values of the Co and the Fe $2p_{3/2}$ core levels obtained in this way for each of the alloy compositions. When the Co concentration increases from 16% to 70%, the Fe $2p_{3/2}$ core-level dichroism increases, whereas that of the Co $2p_{3/2}$ remains virtually constant. These experimentally determined compositional dependencies of the element-specific *dichroism* compare well with the dependencies of the Fe and Co *spin magnetic moments* in bcc $\text{Fe}_{100-x}\text{Co}_x$ random alloys calculated by Ebert and Battocletti²¹ and to some extent with polarized neutron-scattering data²² (in the latter, only few data points exist with comparatively large error bars). The calculations by Ebert and Battocletti include relativistic effects and also give the orbital moments, which are small compared to the spin magnetic moments. The orbital moment of Co is larger than that of the Fe. Both remain virtually constant with increasing Co concentration rather than to increase as does the Fe spin magnetic moment. Due to the smaller spin magnetic moment of Co as compared to that of Fe the average saturation magnetization decreases by 20% in going from $x=16$ to $x=70$. Turek *et al.*²³ calculate similar dependencies of the local and of the average moments; however, the orbital moment is not

determined. In the MCXD measurements by Pizzini *et al.*¹¹ on this system an approximate scaling of their Fe-specific A_{2^*} near-edge intensity with the Fe moment upon increasing Co concentration is observed. However, the Co (A_2) signal did not saturate. Hence the MLDAD method presented here appears to give a more straightforward access to the local magnetic moments.

Our measurements are performed in the magnetic remanent state. It is well known from the literature and from our own magnetic microscopy measurements that Fe/Ag(001) films thicker than about 5 monolayers are fully remanently magnetized along [100] after applying a weak magnetic field. The observed smooth behavior of the dichroism as a function of Co concentration suggests that this remains true for our FeCo alloy films. The *ratio* of the Fe to Co dichroism [see Fig. 5(b)] is even unaffected by the degree of remanence since both quantities are determined simultaneously. Also this ratio compares well with that extracted from Ebert and Battocletti's theory. The agreement strongly corroborates again that the MLDAD is proportional to the element-specific local magnetic moments.

Notably, the compositional dependencies of the dichroism signals do not follow the trend of the average magnetization [cf. Fig. 5(a)]. In contrast, in a temperature-dependent study of an Fe(3% Si)(001) sample, Sirotti *et al.*²⁴ conclude that the dichroism scales with the temperature dependence of the average magnetization. The reduced magnetization at elevated temperatures is considered to be caused by transverse fluctuations of the local magnetic moment. Accordingly, the independence of the dichroism on the average magnetization in the FeCo alloys shows that transverse fluctuations do not cause or accompany the decrease of the magnetization with increasing Co concentration.

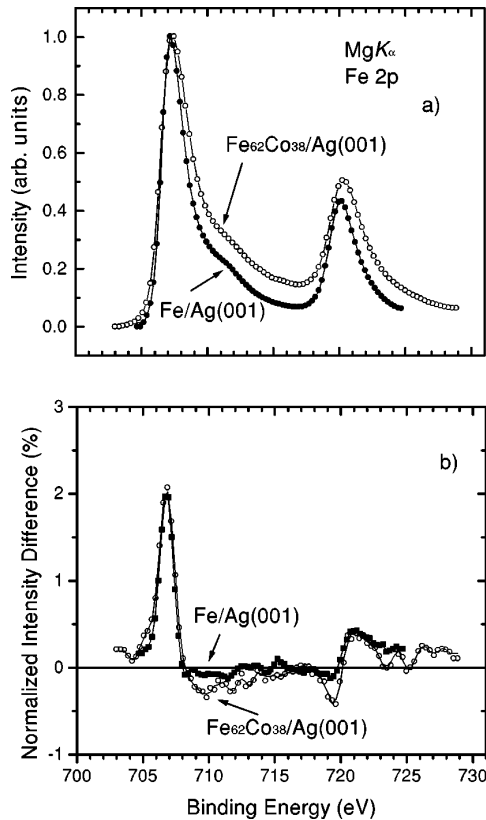


FIG. 4. Energy distribution curves of the intensity (a) and of the normalized intensity difference (b) of the Fe $2p_{3/2}$ core level obtained from an epitaxial $\text{Fe}_{62}\text{Co}_{38}/\text{Ag}(001)$ film with Mg K_{α} radiation. (c) Data from the same sample, but for the Co $2p_{3/2}$ core level.

IV. Conclusions. We show that structural and element-specific magnetic properties of a composite material can be determined by photoelectron diffraction and the related magnetic linear dichroism in angle-resolved core-level photoemission using a conventional unpolarized laboratory x-ray source. We find that the dependencies of the Fe and Co $2p_{3/2}$ core-level dichroism in epitaxial $\text{Fe}_{100-x}\text{Co}_x/\text{Ag}(001)$ alloy films scale with the compositional dependence of the local magnetic moments rather than with the average magnetization.

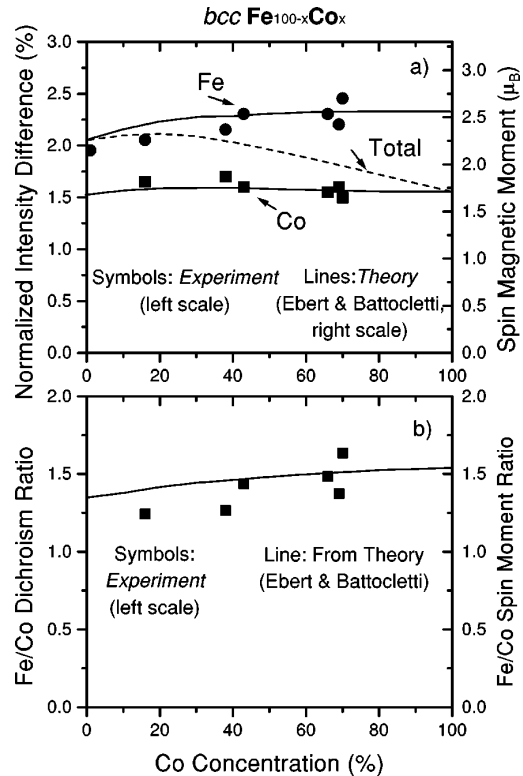


FIG. 5. (a) The peak-to-peak values of the normalized intensity differences of the $2p_{3/2}$ core levels of Fe and Co, respectively, for different compositions of the $\text{Fe}_{1-x}\text{Co}_x/\text{Ag}(001)$ ultrathin films. The data are compared with the calculated values of the spin magnetic moments of Ebert and Battocletti (Ref. 21). (b) The ratios of the data in (a).

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