Magnetic asymmetry in photoemission from Fe(100) with linearly polarized synchrotron radiation

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We have measured partially angle-integrated photoemission with linearly polarized undulator radiation on the valence band and on the $3p$ and $3s$ core levels of iron in an Fe(100) single crystal. A large asymmetry is measured for $3p$ core levels when reversing the magnetization direction perpendicular to the photoemission plane. A smaller but clear asymmetry is also measured in the valence ($3d$) band spectrum, and no asymmetry is measured for the $3s$ core levels. The results are explained by the transverse spin polarization of the near-normal-emission photoelectrons which is induced by the light p polarization and by spin-orbit interaction in the initial state. From the line-shape change of the $3p$ core level as a function of magnetization reversal the fine structure of the $3p$ multiplet is derived. This effect provides a very efficient diagnostic of the surface magnetization direction allowing to measure magnetic ordering at surfaces similarly to the Kerr effect, but in a chemically specific and intrinsically surface sensitive way as shown by examples on ferromagnetically coupled and antiferromagnetically coupled Fe/Cr interfaces.

Magneto-optical effects in the optical spectrum or in the x-ray spectrum are generally not suitable for the study of surface magnetism, unless adsorbate magnetic layers are studied. On the other hand the measure of ejected electrons from primary processes, such as photoemission and Auger decay, is intrinsically surface sensitive and best suited for the experimental study of surface physics.[1] It was recently shown that magneto-optical-like effects are also observed in photoemission from core levels, and that these phenomena can represent a unique tool for surface magnetism.[2–4] The first experiments were done with circular polarized synchrotron radiation on ferromagnetic Fe(110):[2] a maximum intensity asymmetry of about 2.5% was measured on the $2p_{3/2}$ photoelectron peak upon magnetization reversal. Spin-polarization selectivity is not a unique feature of circular polarized radiation in a photoemission experiment: Spin polarization of photoelectrons has been demonstrated to arise from atoms[5,6] and from solid surfaces[7] when excited by circularly polarized, linear polarized as well as unpolarized radiation, provided that a chirality is built in the geometry of the experiment. The spin polarization for Ar $3p$ photoelectrons was measured as a function of the angle between the linear polarization vector and the photoelectron momentum, identifying directions that yield 50% spin polarization.[6] Recent results of unpolarized UV photoemission of Pt(100) surfaces have shown that for off-normal and off-grazing incident radiation the valence-band spectra are spin polarized as measured by a Mott detector after energy selection of the normally emitted photoelectrons.[7] Recently Roth *et al.*[8] have measured the normal photoemission of the $3p$ core level from a Fe(100) surface with off normal p-polarized light both in the spin-integrated and spin-resolved modes. The experiment showed an asymmetry in the intensity of the spin-orbit and exchange split $3p$ photoelectron peaks dependent upon the reversal of the magnetization in the direction perpendicular to the photoemission plane. Reference 8 gave direct evidence of a magnetic asymmetry in core-level photoemission from a ferromagnet by using linearly polarized synchrotron radiation. We present and discuss here our partially angle-integrated photoemission measurements on Fe(100) which give similar results on Fe $3p$ to the angle-resolved experiment of Roth *et al.*[8] and extends to $3s$ and valence-band states. We derive the fine structure of the Fe $3p$ core level, by a fitting procedure on the unmagnetized and magnetization-dependent spectra. Finally we provide examples of application of the magnetization dependence of the core-level spectra excited by linear polarized light to magnetic interfaces: Fe/Cr(100) and Fe/Cr/Fe(100).

EXPERIMENTAL

The experimental geometry is given in Fig. 1. A Fe(100) sample was mounted vertically (y axis) closing the gap of a horseshoe magnet in an ultrahigh-vacuum spectrometer ($P = 1 \times 10^{-10}$ mbar). The magnetization could

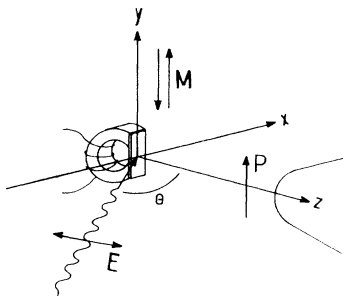

FIG. 1. Geometry of the experiment: The MACII analyzer has a circular entrance slit perpendicular to its axis which lies on the sample normal.

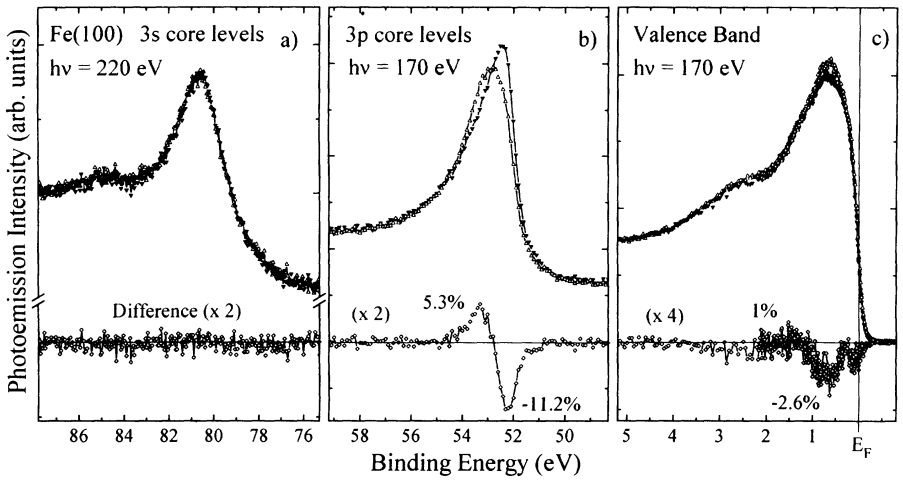

FIG. 2. Fe(100) photoemission spectra as measured with magnetization in the upward direction (upward empty triangles) and downward direction (downward solid triangles). The curves on the bottom are difference curves (up-down magnetization) with quoted percent values. The valence bands were normalized at 5 eV binding energy, where spin polarization is still high (Ref. 16), for displaying the difference. The maximum peak-to-peak asymmetry for Fe $3p$ is 16.5% and for the Fe $3d$ band is 3.6%.

be imposed parallel or antiparallel to the y-axis direction by applying a current to the magnet coil. Linearly polarized radiation from the *SU7* undulator of the SuperAco positron storage ring at LURE was monochromatized by a 10-m toroidal grating monochromator and impinged the sample at 50° off normal. The photoelectrons were collected by a MAC-II Riber analyzer whose axis lies along the sample normal. The scattering plane is not uniquely defined, since the circular entrance slit of the analyzer defines a family of planes that make angles from $-22°$ to $+22°$ with respect to the horizontal plane containing the light polarization vector and the analyzer axis (sample normal). We will conventionally call the horizontal plane the photoemission plane. An Fe(100) single crystal was surface cleaned by sputter-annealing cycles until a sharp low-energy-electron-diffraction (LEED) pattern was observed and C contamination was reduced to a fraction of monolayer. Fe buffer layers of 10–20 Å thickness were epitaxially grown by e-beam evaporation onto the substrate held at 200 °C in a residual gas pressure always lower than 10^{-9} mbar. No oxygen or silicon contamination was detected after growth. *In situ* magnetic hysteresis curves were measured by detecting the dependence of the spin polarization of the secondary electrons upon the external magnetic field with a 100-KV Mott scatterer. Cr was epitaxially grown onto Fe(100) by e-beam deposition onto the substrate held at 150–250 °C, the thickness being monitored with a calibrated quartz microbalance. A monolayer is defined as the surface atomic concentration of a (100) plane of bcc Fe. The trilayer structure was obtained by epitaxy of 7.5 Fe monolayers (1×1 LEED pattern) onto a buffer layer of Cr (11 monolayers) epitaxially grown onto a bulk single-crystal Fe(100) surface. Photon energies of 170 and 220 eV were used to acquire the photoelectron spectra. The overall energy resolution was 350 meV for the valence-band and $3p$ spectra and 500 meV for the $3s$ spectra. The samples were at room temperature during the photoemission measurements, unless indicated in the figures. The photoemission spectra were accumulated in two modes: (a) the field was reversed every scan and then spectra were summed up (at least ten scans per magnetization direction); (b) the field was inverted at every data point during a single scan. Both methods gave identical results.

RESULTS

Figure 2 collects the raw data on Fe(100) as a function of magnetization direction, and the difference obtained by

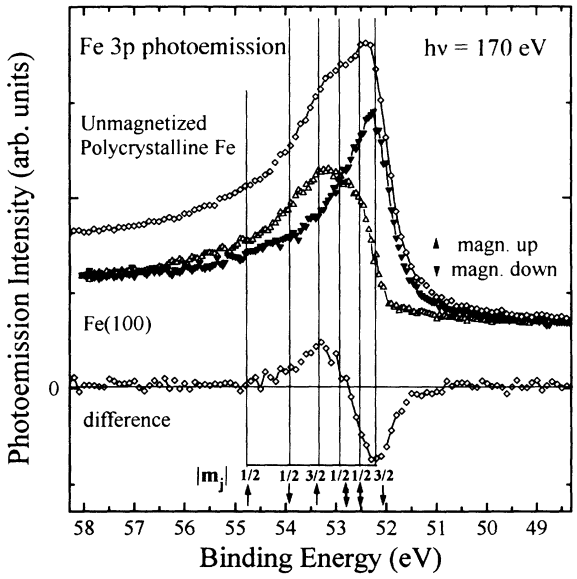

FIG. 3. Fe $3p$ photoemission analysis: the features of the Fe(100) multiplets were enhanced by subtracting an arbitrary (equal) intensity of unpolarized spectrum from the experimental line shapes of each magnetization. This may account for the incomplete magnetization of the surface (or rather by the presence of magnetic domains at 180 degrees) and is a useful procedure to deduce the fine-structure levels. The obtained line shapes (open and full triangles) were normalized at 106 and 115 eV kinetic energy and are compared to the spectrum of unmagnetized polycrystalline Fe measured with the same apparatus. The sum of the two difference curves fits the unmagnetized polycrystalline spectrum. The bottom curve is the experimental difference curve from the central panel of Fig. 2.

subtracting from the spectra measured with the magnetization in the upward vertical direction (as determined *in situ* by Mott scattering) those measured with magnetization in the downward vertical direction. The first panel shows the 3*s* core level with its exchange-split satellite at -4.5 eV,[9] no difference is detected between the two magnetization directions. The central panel shows the 3*p* core levels, which display a large difference on the leading peak. The third panel shows the valence-band spectra, as measured with a photon energy that gives large sensitivity to the 3*d* states:[10] A difference is measured across the *d* band.[11] The spectra were normalized to the background intensity just above the Fermi level and to the intensity at -5 eV from the Fermi level, i.e. at the bottom of the *d* band, in order to show the 3*d* band-shape

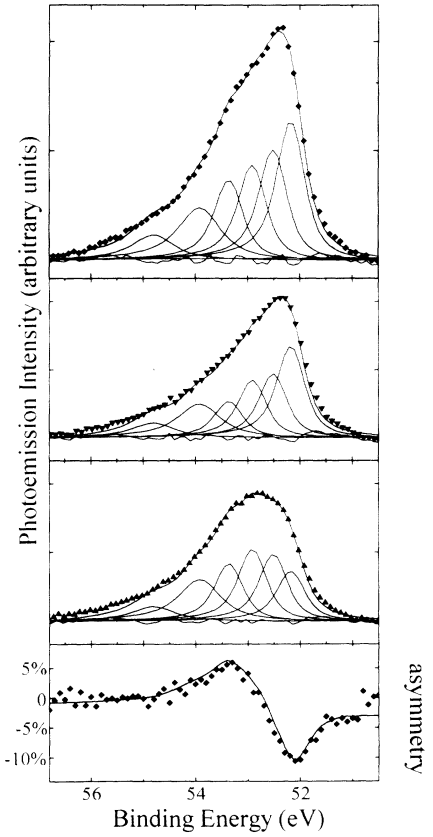

FIG. 4. Analysis of the Fe 3*p* peak shape. Lorenzian-Gaussian peaks were optimized in energy around the fine-structure levels as deduced from Fig. 3. Their intensity and width were fitted to the unmagnetized spectrum (top panel), and the magnetization-dependent spectra (central panels). The bottom curve is the experimental asymmetry (**I** *up* $-$**I** *down*)/(**I** *up* $+$**I** *down*) of the Fe 3*p* intensity (symbols) compared with the asymmetry of the fitted curves (continuous). In each panel the symbols represent the data (after integral background subtraction) and the continuous lines represent the best fit and the individual components. The intensity variations of each component with the magnetization reversal allow to recognize its $\mathbf{m}_j$ value, according to Cherepkov's cross sections for spin polarization in the angular distribution of photoelectrons (Refs. 5, 12, and 25).

difference which peaks at -0.7 eV. The asymmetry defined as (**I** *up* $-$**I** *down*) /(**I** *up* $+$*I down*), where **I** *up* (**I** *down*) is the photoemission intensity measured for in-plane magnetization in the up (down) *y* direction, is 16.5% (peak to peak) for Fe 3*p* and 3.6% for Fe 3*d*.

The line-shape change of the 3*p* core level as a function of magnetization reversal allows one to recognize the fine structure of the multiplet, which cannot be distinguished in the unmagnetized spectrum. *In situ* Mott polarimetry measured a surface magnetization reduced with respect to the saturation value for a single domain surface, but no hysteresis at right angles with respect to the magnetization direction, which indicates the presence of some 180° rotated magnetic domains. This implies that the measured Fe 3*p* line shapes are the sum of asymmetric and "unpolarized" contributions, where the "unpolarized contributions" represent the intensity from the mutually opposite domains and that the analysis of the absolute value of the measured asymmetry cannot be done. The line shapes of Fig. 3 are obtained by subtracting from the

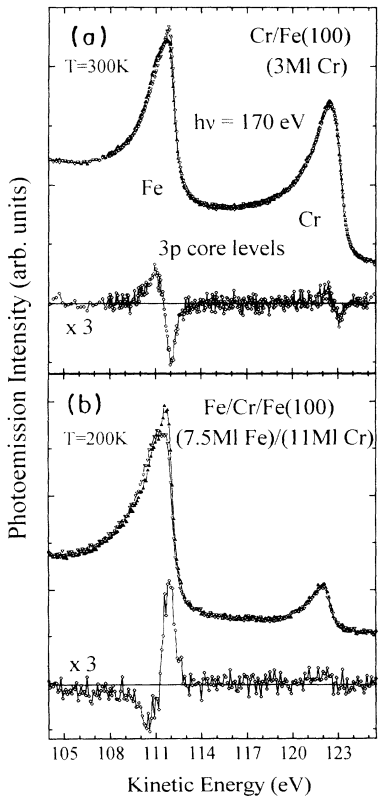

FIG. 5. (a) Fe 3*p* and Cr 3*p* photoemission spectra for three monolayers of Cr epitaxially grown on Fe(100) obtained with opposite magnetization directions (up solid triangles and down empty triangles). The difference signal is shown, multiplied by 3, at the bottom. (b) Spectra from a trilayer structure formed by an overlayer of Fe (7.5 monolayers), an intralayer of Cr (11 monolayers), and the Fe(100) bulk substrate. The reversed line shapes as a function of magnetization (up full triangles and down open triangles) is evident from the difference curve which is shown, multiplied by 3, at the bottom.

magnetization-dependent spectra an equal amount of unpolarized line shape: This procedure is useful to enhance the peaks and shoulders of the fine structure of the $3p$ multiplet, which is indicated by a grid. The experimental difference curve is reproduced at the bottom of the figure. Figure 4 shows the deconvolution of the Fe $3p$ photoemission line shape for unmagnetized Fe and for in-plane magnetized Fe (y direction, up and down). The bottom panel displays the experimental asymmetry and the asymmetry calculated from the fitted line shapes Figure 5 displays the Fe and Cr $3p$ core photoemission spectra and the differences measured for 3 monolayers of Cr epitaxially grown onto the Fe(100) surface, and for a trilayer structure of 7.5 monolayers Fe/11 monolayers Cr/Fe(100).

DISCUSSION

The evidence of a large magnetic signal for core-level photoelectrons which can be measured with highly intense linear polarized synchrotron-radiation sources, and highly efficient cylindrical electron energy analyzers represents a breakthrough in surface science: Up to now the only magnetic information on clean surfaces could be obtained by measuring spin-polarized photoelectrons or scattered electrons, and circular polarized photoemission dichroism, two approaches that are highly inefficient due to the need of spin polarimeters and/or circular polarized photon sources or polarized electron sources.

The rationale for understanding the origin of the magnetic asymmetry measured with linear polarized light is provided by the theoretical work of Cherepkov,[5] and by the experiments of Schönhense[6] and Roth *et al.*[8] We briefly recall here the analogy and difference with respect to circular photoemission dichroism.[2,3] In both cases the photoemission final state is a high-energy free-electron-like state noninteracting with the electronic system and, in the case of a remanently magnetized sample, not affected by the magnetic field which is confined inside the sample, i.e., one must include the interactions between the initial state level and the rest of the excited electronic system directly in the photoionization process.[3] In core-level photoemission with circular polarized light a spin-polarized core hole is created by the action of the photon spin on the orbital momentum of the core level which has a fine-structure splitting ($l>0$).[5] The photoelectrons have a *longitudinal* spin polarization and a magnetic asymmetry can be measured for the exchange-split photoelectron peaks of a ferromagnetic surface according to the parallel (antiparallel), orientation of magnetization and photon spin.

Linear polarized radiation cannot produce a net spin polarization of the total photoelectron yield, but a high degree of spin polarization of the photoelectrons is predicted[5] and measured[6] for particular directions of photoemission. The phenomenon is called angular distribution of spin polarization of the photoelectrons and is a part of the photoionization process itself when the spin-orbit interaction is present in the initial state ($l>0$). The electrons photoemitted by linearly polarized light can only be *transversely* polarized: The highest degree of polarization attainable is measured when $\mathbf{s}\perp\mathbf{e}$ and $\mathbf{s}\perp\mathbf{p}$, where $\mathbf{s}$ is the spin direction, $\mathbf{e}$ the photon polarization, and $\mathbf{p}$ the photoelectron momentum.[5] The sign of the spin polarization for a given photoemission direction depends also on $\mathbf{J}$: The atomic angular distribution of the spin-polarized photoemission calculated by Cherepkov, and measured by Schönhense, implies a sign reversal when $3p_{3/2}$ or $3p_{1/2}$ electrons are excited.[5,6] The angular distribution of the spin-polarized photoelectrons originates an *intensity asymmetry* in the $l>0$ core levels of ferromagnetic surfaces when the spin polarization induced by the photoionization process and the sample magnetization are aligned parallel or antiparallel. This is because the exchange interaction splits on the energy scale the core-hole states according to their spin and the corresponding photoelectrons are distributed over a range of final-state kinetic energies with relative splittings larger, or of the same order, of the experimental energy resolution. In spin-integrated photoemission from magnetically disordered surfaces the spin-polarized multiplets appear largely overlapping in energy (Fig. 3). The magnetic intensity asymmetry induced by linear polarized light resolves the core-hole multiplet states indicating that the $3p$ photoemission process retains to a large degree an atomiclike character in spite of the complexity of the core-valence interactions.

Fine structure of Fe $3p$ photoemission

The analysis of the experimental photoemission line shapes of Fe $3p$ is shown in Fig. 4. The fine structure of "equivalent atomiclike initial states" for the Fe $3p$ transitions, derived by observing the relative enhancements of peaks and shoulders as a function of the magnetization reversal in the spectra of Fig. 3, is used to attempt a fit of the photoemission lineshapes. A sextuplet of levels with the experimental Gaussian width (350 meV) and adjustable Lorenzian width is set at the fine-structure positions indicated by the analysis of Fig. 3. The intensity of the randomly magnetized polycrystalline Fe $3p$ line shape was fitted first, as shown in the top panel of Fig. 4. The best fit confirms the energy positions (within 30 meV) of the levels and indicates a Lorenzian broadening of 420 meV for the top four levels and of 800 meV for the two deepest peaks. By keeping the energy positions and widths fixed the magnetization-dependent spectra for Fe(100) were then fitted by the sextuplet, obtaining the relative enhancement or attenuation of the intensity of each component. The experimental asymmetry is compared with the asymmetry of the fit in the bottom panel. The sign of the variation of intensity of each peak as a function of the field reversal allows to recognize the $\mathbf{m}_j$ character of each component either by comparing with an atomic calculation,[12] or by comparison with the spin-resolved photoemission data which showed that the leading edge of Fe core levels has minority spin character.[13] This independent information sets the signs for the whole multiplet. The $\frac{3}{2}$ character is unambiguously attributed to the $3p$ leading edge by considering that both spin-orbit and Zeeman splitting do push up in energy the $\frac{3}{2}$ terms. The fine structure of Fe $3p$ inferred from our analysis shows six levels that behave as a pure spin-down (minori-

ty) state of $3p_{3/2}$ character at 52.2 eV binding energy, two mixed-spin states of $\frac{1}{2}$ character at 52.5 and 52.9 eV, a pure spin-up (majority) states of $3p_{3/2}$ character at 53.3 eV, a spin-down state of $3p_{1/2}$ character at 53.9 eV, and a spin-up state of $3p_{1/2}$ character at 54.8 eV. The level ordering indicates that the spin-spin (exchange) interaction dominates. The atomiclike level scheme probed by circular magnetic photoemission dichroism for the Fe $2p$ core levels from Fe (110) (Ref. 2) (spin-orbit separation of the order of 13 eV) also showed similar splitting and ordering of the $\mathbf{m}_j$ levels.[14]

As stressed above the measured final states are well in the continuum (110 eV above threshold), they do not interact with the remaining electronic system, and are *not affected by the magnetic field* which is confined inside the sample. This is the important difference between the present effect and the Kerr effect, or the polarized photon-emission spectra from atoms in a magnetic field. Due to this difference one can develop our discussion of the spectra as due to the fine structure of the photoemission initial state only, i.e., the one-hole spectrum of the $3p$ core level.

Relativistic calculations for the hyperfine-field splitted ground state of the $3p$ levels[15] largely deviate from our photoemission fine structure: The ground-state multiplet calculation shows a 50% larger width and a larger separation between $3p_{3/2}$ and $3p_{1/2}$ levels than measured. We stress that in the photoemission from bcc Fe the fine structure reflects also all the solid-state correlations between core hole and extended bands.

Fe 3*s* spectra

No light-induced spin-polarization effects are allowed in $3s$ photoemission, ($l=0$) except for the predicted Fano effect at the Cooper minimum of $3s$ photoionization.[16] The absence of asymmetry in the $3s$ data is a confirmation that final-state effects at the energies of our experiment are not involved with the presently discussed phenomenon.

Fe valence-band spectra

The valence-band photoelectron energy distribution curves were obtained with linearly polarized radiation excitation energy of 170 eV, and are partially integrated in space by the cylindrical analyzer: They must be considered spectra of the one-hole density of states due to k-space broadening. In this case the interband-transition and the mirror symmetry arguments put forward to explain spin polarization in valence-band photoemission from centrosymmetric surfaces are not important.[17] The transverse spin-polarization effect of p-polarized light acts also, albeit reduced, for $l=2$ and $l=3$ initial states[5] and the spin-orbit interaction in the $3d$ initial state is present, therefore the ingredients for a magnetic asymmetry in the p-polarized near-normal photoemission exist without implying band symmetries. It should be noted that valence bands narrow at the surface as a general phenomenon connected with the reduced neighbor coordinations of surface atoms. The d bands become therefore more localized and the orbits can partially relax, due to the reduced symmetry with respect to the bulk. On this ground the relaxation at the surface of the quenching of the orbital magnetic moment was predicted[18] and recently measured in the case of Ni.[4] The spin-orbit interaction of the $3d$ states at the surface should therefore, in principle, be different with respect to the bulk and favor the observation of the present effect.

Applications to the study of magnetic surfaces

When three monolayers (4.2 Å) of Cr are epitaxially grown onto Fe(100) a clear difference is observed on the substrate Fe $3p$ signal and a small difference is also measured on the Cr $3p$ spectrum. Epitaxial (100) layers of Cr on Fe(100) are known to order ferromagnetically in-plane and to couple antiferromagnetically along the [100] direction.[19–23] According to this understanding the only uncompensated difference in the Cr $3p$ signal should arise from the surface of the three-monolayer film. The line shape of the difference curve is similar to that of the Fe $3p$. The results of Fig. 5(a) suggest, by this analogy, that the Cr surface is ferromagnetically coupled to the Fe substrate. Fe/Cr multilayers have been widely studied, since they show oscillatory perpendicular magnetic coupling.[19–22] The spectra of the $3p$ core levels for the trilayer Fe/Cr(11 monolayers)/Fe(100) in Fig. 5(b) show a large asymmetry reversed of the Fe $3p$ peak, which is a clear measure of the Fe-Fe antiferromagnetic coupling across the Cr buffer layer for an odd number of Cr interlayers.[21] *In situ* spin-polarization measurements support these conclusions and will be presented elsewhere.[24] Here we want to stress the efficiency of the $3p$ core-level asymmetry as a method for measuring with intrinsic surface sensitivity the magnetic order at surfaces and interfaces.

CONCLUSIONS

In summary, we have measured a photoemission difference in the intensity of spin-orbit and exchange-split final states which depends on the reversal of the magnetization direction perpendicular to the incidence plane, in agreement with the recent findings of Roth *et al.*[8] The transverse spin polarization of the near-normal photoelectrons, which is due to p polarization of the light and spin-orbit interaction in the initial state, originates the observed phenomenon. This fact implies that an atomiclike sextuplet analysis of the $3p$ core-level photoemission final state is possible, as we have shown in this paper. Furthermore this effect, as measured with standard high-luminosity synchrotron-radiation photoemission experiments, is by far the most efficient diagnostic of the surface magnetization direction and can be exploited for surface experiments, likewise the Kerr effect for bulk magnetism, including lateral and time resolution. The study of the temperature dependence of the surface magnetic order, in a chemically specific way, becomes possible since the Debye-Waller broadening of the core-level photoemission is still small at the Curie temperatures of the ferromagnetic metals and alloys. It is clear that the data quality in terms of statistics, energy resolution, and

effective tunability of the sampling depth of such spectroscopy is superior to the more difficult circular dichroism in photoemission and spin-resolved photoemission. The quantitative understanding of the linear p-polarization effect and its explicit connection with fundamental physical quantities requires $\mathbf{m}_j$-resolved calculations of the angular distribution of spin-polarized electrons and photoionization cross sections.[25] We have shown how this effect allows for an easy measurement of the magnetization of surface and interface layers in a classic interface experiment, Cr/Fe(100) and in a trilayer experiment, Fe/Cr/Fe(100) where the magnetic order of the surface layer coupled to the substrate via the intermediate layer must be measured in an intrinsically surface sensitive manner.

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