Magnetic interface formation at Cr/Fe(100) and Fe/Cr/Fe(100): Magnetic dichroism in photoemission study

Giancarlo Panaccione* and Fausto Sirotti
Laboratoire pour l'Utilisation du Rayonnement Electromagnetique, CNRS-CEA-MESR, F-91405 Orsay, France

Elisabetta Narducci
Instituto Nazionale di Fisica della Materia, Dipartimento di Fisica, Università di Genova, I-16146 Genova, Italy

Giorgio Rossi
Laboratorium für Festkörperphysik, Eidgenössische Technische Hochschule–Zürich, CH-8093 Zürich, Switzerland and Instituto Nazionale di Fisica della Materia, Dipartimento di Fisica, Università di Modena, I-41110 Modena, Italy
(Received 24 May 1996)

The early stages of the growth of Cr/Fe(100) and Fe/Cr/Fe(100) interfaces have been investigated by magnetic dichroism in photoemission of Fe 3*p* and Cr 3*p* core levels as measured from chiral experiments employing linearly polarized synchrotron radiation. Evidence is obtained for a 30% larger magnetic moment of interface Cr atoms with respect to Cr atoms belonging to epitaxial ultrathin films and a 40% magnetic moment enhancement of top Fe interface atoms in the Fe/Cr/Fe(100) trilayer. The kinetic growth conditions (450 K) lead to a uniform overlayer growth, without intermixing, but dominated by islanding. As a consequence the formation of a single-surface ferromagnetic domain for Fe/Cr/Fe(100) is frustrated up to two Fe monolayer (ML) thickness. The line shape of Fe 3*p* photoemission in the frustrated regime is consistent with the presence of in-plane magnetic order at 90° with respect to the substrate magnetization direction. The appearance of photoemission magnetic dichroism for Fe overlayer thicknesses exceeding 2 ML is interpreted as due to domain rotation towards the direction antiparallel to the Fe substrate magnetization. [S0163-1829(97)03601-1]

I. INTRODUCTION

Stacking of alternate layers of Cr and Fe makes an artificial solid with intrinsically anisotropic electronic properties, among which great importance is given to the giant magnetoresistance effect.[1] As a function of the thickness of antiferromagnetic (AF) Cr layers separating next-neighboring Fe layers the magnetic coupling between ferromagnetic (FM) Fe layers is parallel or antiparallel. The oscillations of the coupling follow a long period and a short period which are thought to be related to the shape of the Fermi surface.[2]

Due to the prototypical value of the Fe/Cr/Fe structure for the understanding of magnetically dependent electron transport, a large number of experiments, models, and theoretical descriptions have been produced in recent years. An overall agreement has been reached on the double periodicity of the magnetic coupling and on the general behavior of the giant magnetoresistance, but open questions and discrepancies remain in the detailed description of the magnetic behavior of the atoms at the interface between Fe(100) and Cr and between Fe and Cr(100) as the multilayer grows. In particular, open questions are the magnitude of the magnetic moments of Fe and Cr at and close to the interface and surface region, and the magnetic coupling of the first layers, together with its dependence on growth conditions, i.e., on the morphology, perhaps metastable, assumed by the interfaces at the early stages of formation.

Cr and Fe have the same bcc lattice at room temperature and very similar lattice parameters (2.87 Å and 2.88 Å for Fe and Cr, respectively[3]), so that epitaxial growth is possible with either one or the other as a substrate. Fe is FM at ordinary temperature and Cr is AF with a slightly incommensurate AF structure oriented along the [100] direction.[4] Cr is often defined as a layered AF solid since it can be viewed as a stacking of ferromagnetically ordered (100) planes, antiferromagnetically coupled one to the next. Antiparallel coupling across the interface and a strong enhancement (up to sevenfold) of the Cr surface magnetic moment, with respect to the bulk Cr value of $0.59\mu_B$, were predicted for an ordered Cr monolayer on Fe(100).[5,6] Total energy calculations for the Fe/Cr/Fe system showed that the number of the Cr layers dictates the parallel or antiparallel coupling between the Fe overlayer and substrate separated by the Cr buffer.[2,7,8] The Fe magnetic moments in the Fe/Cr/Fe multilayers are predicted to be similar to that of bulk Fe.[8]

A number of experimental results were apparently inconsistent, being strongly influenced by the actual thicknesses of the layers grown in different experiments, by the growth conditions which determine the degree of epitaxial order and of atomic mixing at the interface, and also by the incertitude of the methods used for measuring the magnetic moments. Extreme values for the Cr magnetic moment at the Cr/Fe(100) interface have been quoted from *in situ* magnetometer measurements giving $\mu_{Cr} \geqslant 4\mu_B$ for submonolayer thicknesses and $\mu_{Cr} \sim 3\mu_B$ after completion of the first monolayer;[9] in the same experiment, the authors inferred from their data a delayed onset of the Cr AF stacking. Spin-resolved photoemission and energy loss spectroscopies of Cr at the interface with Fe have indicated $\mu_{Cr} \sim 1.8\mu_B$,[10] an enhancement with respect to bulk Cr (Ref. 11) or a moment similar to bulk Cr.[12] X-ray absorption dichroism experiments on multiple interfaces showed basically the bulk value for

Cr.[13] The Fe magnetic properties at the Fe/Cr(100) and Fe/Cr/Fe stacks are strongly influenced by the effective structural properties of the AF Cr substrate, as shown from magneto-optical Kerr effect results.[14] Both enhancement or reduction of the surface Fe magnetic moment are predicted, and some theoretical and experimental results suggested the growth of a magnetically dead layer.[13,15–17] Very clear spin-resolved electron microscopy images show the AF coupling of Fe layers across Cr layers of variable thickness, with monolayer resolution.[18–20] These experiments also showed the incommensurability of the Cr AF order with the lattice, together with a ''defect'' in the layered antiferromagnetism of Cr in the 0–4 atomic layers range. Moreover, bond frustrations and interface roughness, depending on growth conditions, can either suppress the short coupling period oscillations in the Cr overlayers or modify the orientation of Fe overlayers, as proposed from theoretical calculations.[15,21,22] Superconducting quantum interference device (SQUID) measurements and polarized neutron reflectivity showed for Cr/Fe superlattices the importance of biquadratic interlayer magnetic coupling, when a magnetic frustration is present,[23,24] confirming the prediction of Ref. 20. The above results indicate that the structure of the *real* surfaces and interfaces plays a fundamental role in the magnetic order and coupling. Any experimental methods that integrate the information on magnetic order and magnetic moment lead therefore inevitably to gross errors.

The aim of the present work was to study the magnetic properties in the Cr/Fe(100) and Fe/Cr/Fe(100) interfaces with magnetic dichroism in photoemission. The advantage of photoemission experiments on magnetic interfaces comes from the sensitivity to the chemical species and to the surface atoms. By exploiting magnetic dichroism in chiral experiments with linearly polarized synchrotron radiation [LMDAD (Refs. 25 and 26)] on core levels, one can take advantage, with respect to the previous spin-resolved experiments, of the much higher counting rates and therefore of the better statistics which is attainable, allowing one to deepen the interpretation by separating the effects of magnetic order from the information on magnetic moments at a qualitative, but fruitful level.[27,28] Cr(100) layers display dichroism in photoemission since the surface contributes approximately 1/3 of the total signal which is not completely averaged by the exponentially damped underlayer contributions.

II. LMDAD METHOD

The LMDAD effect has been described in several recent experimental and theoretical papers.[25–31] We refer for the definition of the experimental geometry and for description and application of the atomic model interpretation to Refs. 27 and 29. Here it is important only to recall that (1) the sign of the LMDAD dichroism, i.e., its plus-minus feature, defines the parallel-antiparallel magnetic alignment between overlayer and substrate with respect to a standard ferromagnetic sample. (2) The magnitude of the dichroism is proportional to $\langle M_{\mathrm{surf}} \rangle$, i.e., to the order parameter of the ferromagnetic surface; it vanishes at the Curie temperature and/or for unmagnetized samples. LMDAD is therefore sensitive to in-plane disorder and domains: A reduced LMDAD signal implies a reduction of the magnetic order along the axis defined by the chirality of the experiment.[28] Knowing that photoelectron diffraction effects can strongly influence the obtained dichroism,[32] experimental results can be compared only when measured in a fixed geometry and for a fixed photon energy as done in the present work. (3) The presence of magnetic order can be observed from dichroism experiments even if field averaging is done naturally by 180° domains or by antiferromagnetic ordering. In fact the line shape of the field-averaged spectra depends on the alignment of the magnetic moments, i.e., of the magnetic core hole states, independently of the degree of polarization (magnetization).[33] In this case variable chirality experiments can be performed in order to probe the existence of magnetic order. (4) The LMDAD spectra are determined by the energy splitting of the magnetic core hole sublevels, which is proportional to the magnetic moment.[34]

III. EXPERIMENT

LMDAD experiments were performed on the Swiss-French beam line SU3 and on the SU7 beam line at the SuperAco storage ring in LURE (Orsay). In both cases, the electron energy analyzers were placed at 45° with respect to the direction of the linearly polarized synchrotron radiation from standard planar undulators impinging onto the sample. Angular acceptances were, respectively, of $\pm 1°$ (SU3) and $\pm 22°$ (SU7). The overall energetic resolution was ~ 100 meV for the experiments using 120 eV photons (SU3) and $\sim$ 250 meV for 150 eV photons (SU7).

All measurements presented in this paper were obtained using a [100]-oriented Fe 3% Si single crystal as a substrate, mounted to close the gap of a soft iron yoke.[26,34] The Fe(100) surfaces were prepared by Ar^+-ion sputtering and annealing cycles. In order to avoid the surface segregation of bulk impurities (Si, C, and S) the final iron surfaces were obtained either by a mild sputtering-annealing cycle or by homoepitaxy of a thin iron overlayer onto a well-ordered but C-segregated Fe(100) surface. Fe and Cr were evaporated by electron bombardment from high purity rods, with a typical deposition rate of 0.5 and 0.2 Å/min, respectively, and in a pressure below 2×10^{-10} mbar. The thickness of the deposit was monitored by a quartz crystal oscillator and verified by the Cr 3*p* and Fe 3*p* photoemission intensities. Analysis of low-energy electron diffraction (LEED) patterns suggested that layer-by-layer growth is favored at high temperature ($\sim$600 K), confirming the findings of Ref. 20. However, in order to minimize interdiffusion, the growth was performed at a substrate temperature of 450 K. No trace of contaminants was detected before and after each evaporation. Valence band spectra were measured to control the surface cleanliness during the experiment. The base pressure was 3×10^{-11} mbar.

The sample was magnetically saturated by current pulses through the winding of the electromagnet. All spectra were measured in remanence. Both spin polarization data, obtained from a 100 kV Mott detector on the same Fe single crystal and mounting, and *in situ* Kerr-effect measurements showed a squared hysteresis loop as well as 100% remanence. Linear magnetic dichroism in the LMDAD mode was measured in the chiral geometry described in Refs. 26 and 34, obtaining two mirror experiments by reversing the sign

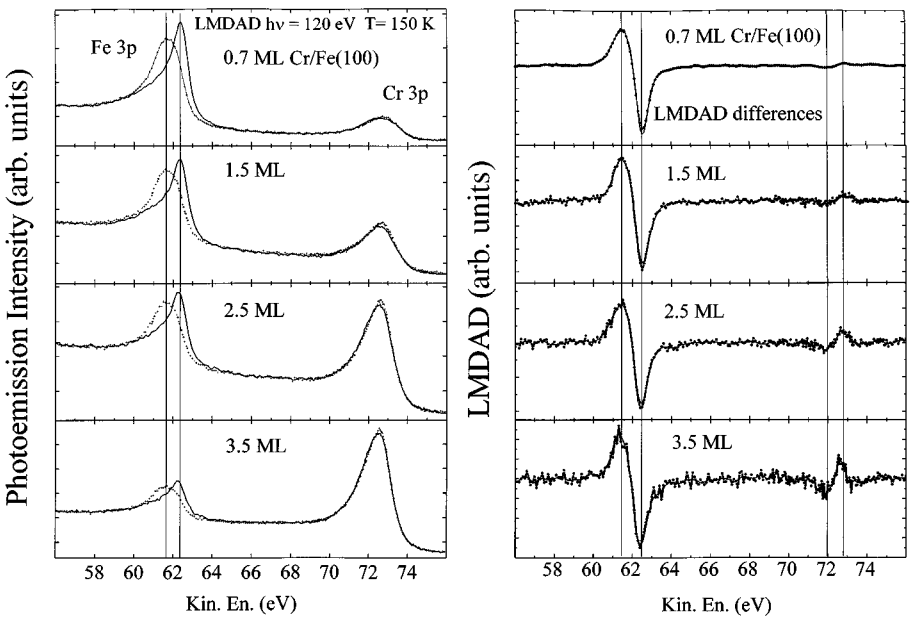

FIG. 1. Left: LMDAD spectra for the two magnetization directions (crosses and continuous curves) of the Fe and Cr 3*p* core level as a function of the Cr coverage on the Fe(100) surface (from up to down), as measured in the same fixed chiral geometry, for a photon energy of 120 eV at 150 K of temperature. The vertical bars indicate the energy positions of the maxima. Right: LMDAD difference curves, corresponding to the magnetization-dependent spectra, for Fe and Cr 3*p*. Solid circles are experimental data and solid lines are the smoothed functions.

of the magnetization direction, which was parallel to the Fe(100) surface and perpendicular to the scattering plane defined by the photon beam and by the photoelectron momentum vector. The LMDAD magnetic asymmetry is defined as $A_{\mathrm{LMDAD}}=(I_{\mathrm{up}}-I_{\mathrm{down}})/(I_{\mathrm{up}}+I_{\mathrm{down}})$, where $I_{\mathrm{up\ (down)}}$ are the photoemission intensities measured for the imposed magnetization in the upward (downward) direction.

IV. RESULTS

The left panel of Fig. 1 presents the 3*p* core level spectra for Fe and Cr as measured in the two mirror experiments, as a function of the Cr coverage on the Fe(100) surface. The corresponding LMDAD difference curves, representing the LMDAD dichroism, are shown in the right panel of the same figure. The vertical bars identify the different peak positions for the two core levels. The opposite behavior, i.e., the reversed plus-minus feature, of the Cr dichroism with respect to the one of Fe indicates that the dominant contribution is from Cr antiferromagnetically coupled to the Fe(100) substrate.[27] At 2.5 and 3.5 monolayers (ML) one sees a small energy shift of both Fe and Cr 3*p* peaks and a marked narrowing of the Cr 3*p* LMDAD curve. This effect is better shown in Fig. 2 where the Cr 3*p* LMDAD spectra, after normalization, at 1.5 ML of coverage and at 3.5 ML are compared, both aligned to the positive asymmetry peak: The width of the Cr LMDAD spectrum of the 1.5 ML coverage is larger by 35% with respect to the 3.5 ML spectrum. Within the scheme of the atomic model and according to Fe 2*p* LMDAD data, the positive and negative peaks of the asymmetry correspond to the energy of the $m_J=\pm 3/2$ sublevels.[27,29] The width of the Cr dichroism, i.e., the $m_J=\pm 3/2$ energy splitting, is plotted in the bottom panel of Fig. 3 versus the Cr thickness on Fe(100), after normalization to the photoemission peak area. Within the error bars the submonolayer and monolayer data are equal, but a sharp decrease of the width is measured at 2.5 ML, and starting from 3.5 ML a constant value is reached, up to thicker Cr films. The same analysis for the Fe 3*p* core level splitting is shown in the top panel of Fig. 3, where a similar result of reduction of the $m_J=\pm 3/2$ splitting is observed for Cr coverages larger than 1.5 ML. Figure 4 presents the Fe/Cr/Fe(100) interface, for 1.5 ML of Fe on top of a 12 ML Cr film grown onto Fe(100). From the comparison with the magnetization-dependent spectra for the Cr/Fe(100), we observe that (a) the coupling between the Fe overlayer and the Fe(100) substrate, across the Cr layer, is dominantly antiferromagnetic, as shown by the reversal of the sign of Fe LMDAD; also the Cr LMDAD signal is reversed, showing that Fe is the magnetic driver in the Fe/Cr interface; (b) the degree of magnetic order is small in the iron overlayer, which has a small LMDAD signal. The Cr LMDAD dichroism width is within the errors of the same order of the thick layer, as indicated in the right part of Fig. 4, but the value of the Fe 3*p* splitting is different with respect to the value of the Cr/Fe(100) interface in the Cr monolayer regime. In fact, the widths of the Fe LMDAD dichroism for the Fe(100) clean surface and for the $\leqslant 1$ ML Cr/Fe(100) interface have comparable values, but the LMDAD width for the 1.2 ML Fe/12 ML Cr/Fe(100) trilayer is $\sim 30\%$ larger. The same enhancement in the value of the Fe splitting was observed in previous experiments,[26,35] whose results are reported in Fig. 3 (circles). Finally, Fig. 5 shows the evolution of the Fe 3*p* LMDAD splitting (bottom panel) and the Fe 3*p* normalized LMDAD (top panel) as a function of the Fe coverage in the Fe/Cr/Fe(100) system. After the first coverage with no LMDAD signal, starting from 1.5 ML a large Fe LMDAD splitting is found, followed

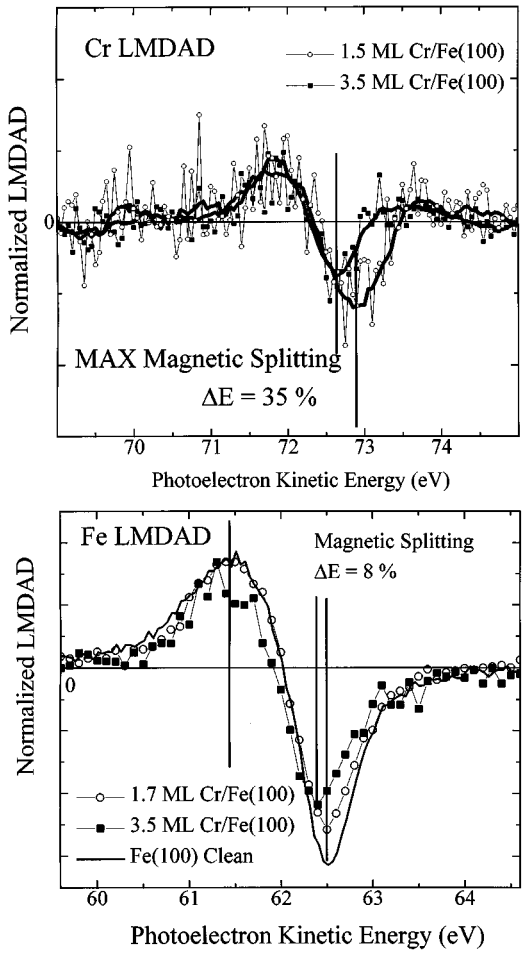

FIG. 2. Top: comparison between the Cr 3*p* LMDAD curves for 1.5 ML (open circles) and 3.5 ML (solid squares) coverage in the Cr/Fe(100) interface. Solid curves are smoothed functions. The normalized dichroism curves are reversed in sign and both arbitrarily aligned on one side of the curve, to better show the difference of the splitting value. The vertical bars indicate the peak position: A difference of about 35% in the width of the dichroism is recognizable. Bottom: same comparison for the 3*p* dichroism of Fe. Open circles, 1.5 ML of Cr; solid squares, 3.5 ML of Cr; solid curve, Fe(100) clean surface. The LMDAD curves are aligned on the same side of dichroism. The reduction of the magnetic splitting is of 8%.

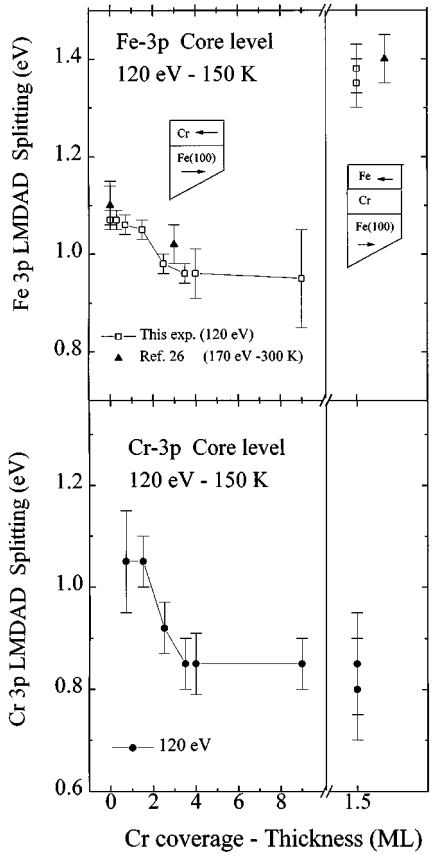

FIG. 3. Top: evolution of the 3*p* LMDAD splitting of Fe (open squares) as a function of the Cr coverage in the Cr/Fe(100) and Fe/Cr/Fe(100) interfaces. Solid triangles are the results of a previous experiment on the same interface (Ref. 26). The insets show the direction of coupling, parallel or antiparallel to the substrate Fe(100), in the bilayer and trilayer systems. Bottom: evolution of the 3*p* LMDAD splitting of Cr as a function of the coverage in the Cr/Fe(100) and Fe/Cr/Fe(100) interfaces.

by a reduction of the splitting towards the value of the Fe(100) clean surface.

V. DISCUSSION

A. Cr/Fe(100)

The changes of the photoemission peak shape and width can have several origins including bonding disorder, with unresolved chemical shifts arising from inequivalent sites, and size-dependent core hole screening effects. In addition, in a magnetic material the variations of the magnetic moment at the surface or at an interface are directly reflected in the energy splitting of the core hole magnetic sublevels and, therefore, in the energy width of the magnetic dichroism spectrum. We will discuss below the relative changes in the dichroism width as defined above independently of the small chemical shifts that are observed at the interface formation.

The exponential attenuation of the Fe signal through the Cr overlayer in Fig. 6 excludes the occurrence of an extended intermixing for our growth conditions, but the little magnetic order for the Fe monolayer deposited on top of the 12-layer Cr film (Fig. 5) indicates a stepped Cr surface, which in turn suggests that the growth of Cr is in large islands rather than layer by layer. We must observe that the lack of well-behaved layer-by-layer growth in our conditions frustrates the correct development of layered antiferromagnetism in the range of small coverages investigated (up to 3.5 ML). We can in fact infer the presence of a Cr structural disorder from the results of Fig. 1, where the obtained Cr dichroism shows for all the coverages the plus-minus feature that corresponds to an antiparallel Cr alignment with respect to the Fe substrate. The breakdown of the layered antiferromagnetism behavior is a signature of a stepped and frustrated Cr layer.[20] By assuming that (i) the spin-orbit interaction is fixed and (ii) the splitting between the $m_J = +3/2$ and the $m_J = -3/2$ sublevels, which are the two pure spin-orbit states of the multiplets,[31] varies linearly with the strength of the exchange interaction, one can interpret the changes of the width of the LMDAD curve reported in Fig. 3 as being *proportional* to the relative variations of the surface magnetic moment. Figure 3 shows that in the range 0–3.5 Cr ML on Fe(100), the Cr $m_J = \pm 3/2$ splitting value decreases from 1.05 ± 0.05 eV

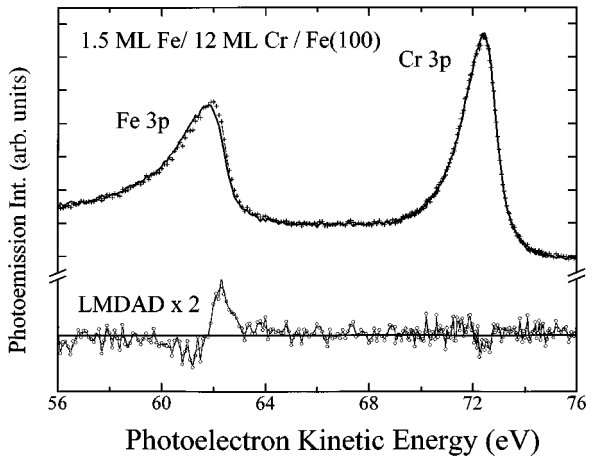

FIG. 4. LMDAD 3*p* photoemission spectra as a function of magnetization reversal (crosses and solid curve) for the Fe/Cr/Fe(100) trilayer system ($h\nu = 120$ eV, $T = 150$ K). The plus-minus feature of the dichroism curve (open circles) is reversed for both Fe and Cr, showing an antiparallel orientation of the Fe top layer with respect to the Fe substrate. Also the Cr LMDAD dichroism is reversed with respect to the one measured for the same coverage as a free terminated layer. The solid curve in the LMDAD dichroism is a smoothed function.

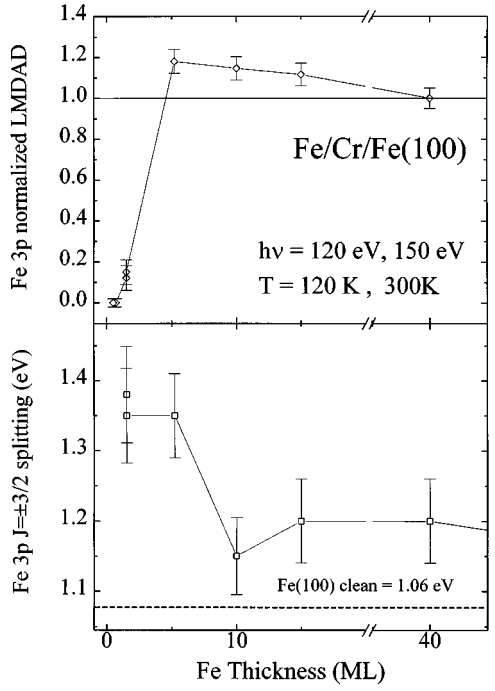

FIG. 5. Top: evolution of the normalized dichroism (ND) of the Fe 3*p* LMDAD in a Fe/Cr/Fe(100) trilayer system (open diamonds with error bars), as a function of the Fe top layer coverage. The value of 1 corresponds to the normalized dichroism of the Fe(100) clean surface. Bottom: evolution of the Fe 3*p* LMDAD splitting in the same trilayer system as a function of the Fe top layer coverage (squares with error bars). The dashed line indicates the value of the Fe(100) clean surface splitting. The ND is obtained by dividing the integral of the Fe photoemission peak at each coverage and referred to the standard spectrum of the clean Fe(100) surface; this was measured at 120 eV of photon energy and 150 K of temperature.

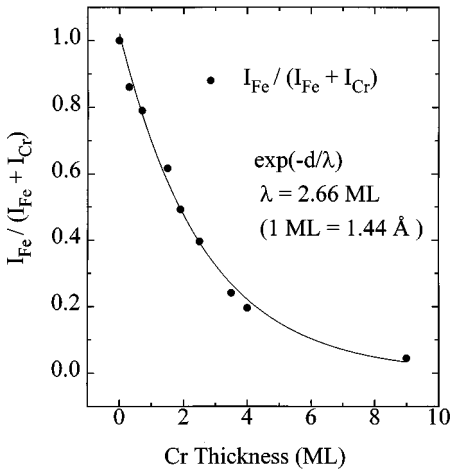

FIG. 6. Total photoemission intensity of Fe 3*p* core levels, divided by the sum of the total intensity of Cr and Fe 3*p* (solid circles), as a function of the Cr coverage in the Cr/Fe(100) interface. The solid curve is the fitted exponential function.

to 0.85 ± 0.05 eV. This relative reduction of 35% in the splitting as the coverage exceeds the first monolayer is the signature of an enhanced *interface* magnetic moment of the Cr atoms in contact with Fe. The splitting value of about 0.85 eV cannot be representative of the Cr bulk magnetic moment, considering also that the thickness range over which the reduction of the magnetic splitting occurs is affected by the island growth mode: Signals from first, second, and third Cr layers are added. Nevertheless, it appears that the magnetic moment changes gradually at least through three layers before stabilizing at the value which is measured up to 12 layers in this experiment.[22] The measured enhancement of μ_{Cr} at the surface of Fe(100) is large, but definitely smaller than some values reported before,[9,10] or predicted by theory [theoretical predictions are referred to $T = 0$ K and for a perfect (100) Cr monolayer].

The Fe 3*p* LMDAD splitting is basically unaffected by the adsorption of the first monolayer of Cr, but a reduction of about the 10% is observed for higher Cr coverages. The simultaneous reduction for both the Fe and Cr LMDAD splittings ($\propto \mu_{Fe}$ and μ_{Cr}) suggests a change in the magnetic properties of the whole interface region at a ''critical'' thickness of 1.5–2 ML of Cr. This range of thicknesses is the onset of the ferromagnetic order of Cr, in qualitative agreement with Turtur and Bayreuther[9] and Alvarado and Carbone.[16] The interface between Fe(100) and a single monolayer of Cr is different from the interface between Fe(100) and an AF stacked Cr film: The latter case implies a reduction of the Fe moments near the surface while the former case does not.

More insight into the magnetic order of Fe in the interface region, below the Cr overlayer, can be obtained by using the LMDAD normalized dichroism (ND) and by plotting it against the $m_J = \pm 3/2$ splitting.[36] As we discussed above and in Ref. 37 the width and the ND of the LMDAD are not fully independent as one can test by applying the atomic model and calculating the LMDAD spectra when the $m_J = \pm 3/2$ splitting is artificially varied. The wider the splitting, the larger is the ND since the opposite dichroic intensities overlap less and less. Conversely, if the splitting is reduced to

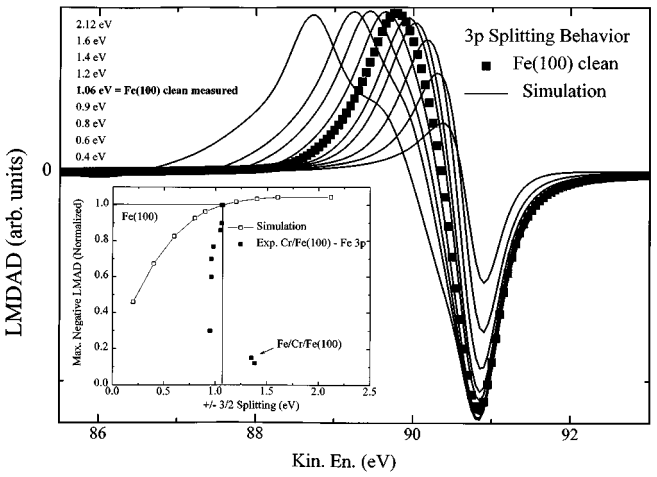

FIG. 7. Simulation of the LMDAD behavior as a function of the $m_J = \pm 3/2$ splitting value, in the scheme of the atomic model of Ref. 29. Solid squares are the LMDAD of the measured Fe(100) clean surface. Inset: Comparison between the simulation (open squares) and the Fe 3*p* experimental data for the Cr/Fe(100) interface (solid squares) for the maximum negative asymmetry in the normalized LMDAD dichroism curve vs splitting value. The normalized value equal to 1 corresponds to the Fe(100) clean surface as measured. Data for the Fe/Cr/Fe(100) are also indicated by the arrow, showing the largest splitting value, as well as the minimum of normalized dichroism.

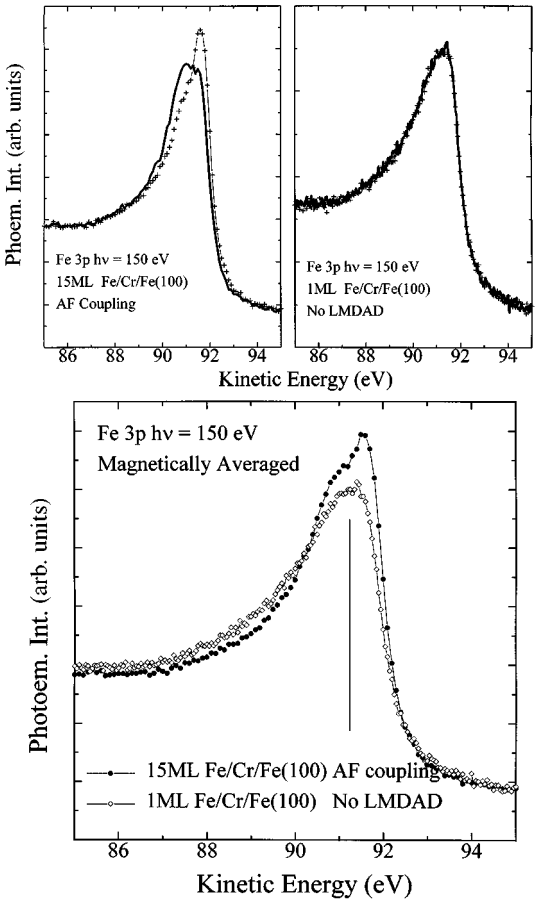

FIG. 8. Top: Fe 3*p* magnetization-dependent spectra for two different Fe coverages in the Fe/Cr/Fe(100) trilayer, $h\nu = 150$ eV, $T = 300$ K; the lower coverage does not show any LMDAD (right panel). The spectra for 15 ML do show LMDAD (left) and correspond to AF (i.e., antiparallel) coupling with respect to the Fe(100) substrate. Bottom: comparison between the magnetization-averaged spectra for the two Fe films. The line shapes show a marked difference between the zero LMDAD spectra and the AF coupled ferromagnetic overlayer spectra.

zero, i.e., in the case of degenerate hole sublevels in the absence of magnetic moment, also the ND is reduced to zero. Such a calculation is represented in Fig. 7 by the solid squares and lines. One sees that for typical changes of the magnetic splitting (changes of the magnetic moment by $\pm 30\%$), the related changes in the ND are less than 10%. On the other hand, changes of the order parameters, i.e., of the ND, do not influence the splitting.[37] In Fig. 7 we compare also the calculated values with the data for Fe 3*p* of the Fe(100) surface covered by increasing thickness of Cr. The data (open squares) show a reduction of Fe ND for two monolayers of Cr, followed by a reduction of the splitting as seen in Fig. 3, and by a sharp reduction of the ND. The ND reduction is large and independent on the magnetic moment of the Fe substrate. This effect is further proof of the perturbation in the Fe near-interface layers of the substrate. The layer nearest the interface, i.e., basically the only one contributing to the photoemission in the data point for 8 ML Cr/Fe(100), has a severely reduced magnetic order in the direction probed by our experiment. Intermixing at the level of a single interface double layer (Fe-Cr) cannot be excluded, as well as a rotation of the Fe moments forming an extended domain wall with the antiparallel oriented Cr interface layer.

B. Fe/Cr/Fe(100)

Interface magnetic effects are seen from the Fe/Cr/Fe results too (Figs. 4 and 5). Data from Fig. 5 show that the first monolayer of Fe grown on the 12-layer Cr buffer has a large splitting, but a small degree of alignment of the moments along the substrate magnetization (parallel or antiparallel). This is consistent with the results of Alvarado and Carbone who measured zero spin polarization for Fe growing on a Cr(100) epitaxial film up to 2 ML Fe thickness.[16] From Fig. 5, we observe that the maximum ND is reached for 5 ML; this value corresponds to both high order and higher splitting with respect to the standard reference spectrum of the Fe(100) clean surface (i.e., $m_{J\pm 3/2} = 1.06$ eV and ND=1). At higher coverages the ND converges to the standard value. Correspondingly, the bottom panel of Fig. 5 shows that the $m_J = \pm 3/2$ splitting starts very high as soon as it can be measured, and then decreases, still remaining higher than the Fe(100) substrate for relatively high thicknesses. These results indicate that the magnetic moment of the top Fe film of the Fe/Cr/Fe(100) is enhanced and that the onset of a Fe magnetic order antiparallel aligned to the Fe(100) substrate is for ~ 1.5 ML thickness of the Fe top layer. In fact, Fig. 5 and the magnetization-dependent spectra of Fig. 8 show that for submonolayer and monolayer Fe thicknesses no LMDAD is measured. The absence of LMDAD has been confirmed by performing experiments at 150 K, which excludes the hypothesis of a strongly reduced Curie temperature for the Fe overlayer, in agreement with Ref. 16. The spectra do not present LMDAD, but this does not correspond to a narrow-

ing of the photoemission peaks. All these findings suggest that the magnetic moments are oriented differently.

As we discussed above, based on indirect evidence, the Cr surface is highly stepped, implying the existence of terraces of different height determining antiparallel Cr surface domains to which the Fe top monolayer should couple antiferromagnetically. The Fe top layer would therefore break itself into domains with many in-plane Néel walls, which is an energetically unfavorable situation. Nevertheless, if this was the case, the Fe 3*p* photoemission spectrum would look just like the field average of the usual LMDAD spectra. In fact in this hypothesis the quantization axis of the 180° domains would still be parallel to the magnetization axis of the substrate. The Fe top layer would be unmagnetized, but its moments would be still aligned along the perpendicular direction to the photoemission plane; so the spectrum would have the same line shape as a field-averaged spectrum of the Fe substrate. As a matter of fact, the spectra for submonolayer and monolayer coverages of the top Fe layer (i.e., the one showing no LMDAD) are quite different from the field average of the iron substrate spectra, as easily observable in the bottom panel of Fig. 8, and qualitatively resemble the line shape measured in the nonchiral geometry which can be obtained by rotating the quantization axis (the magnetization) in the scattering plane. Based on the present set of data we can make the following statements: (a) The magnetic splitting of Fe 3*p* is present from the submonolayer regime, (b) the absence of LMDAD cannot be explained by 180° domains aligned with the substrate quantization axis, and (c) the spectra are compatible with the hypothesis of a nonchiral effective geometry of the experiment, obtained by a 90° rotation of the surface quantization axis, either within the surface plane or perpendicular to it, the LMDAD being zero in both cases.

The hypothesis of perpendicular magnetization was put forward by Alvarado and Carbone to explain the lack of spin polarization at less than 2 ML of Fe coverage.[16] Although it is a possibility, it implies a large anisotropy which for almost relaxed quasiepitaxial layers is not expected. In-plane 90° rotation may occur due to biquadratic interlayer coupling. The exchange energy E_{ex}, which describes the coupling between layers, is proportional to both the bilinear J_1 and the biquadratic coupling J_2, i.e., $E_{\mathrm{ex}} \propto J_1 \cos\theta + J_2 \cos^2\theta$, where θ is the angle between the magnetization direction of two layers.[38,39] When $J_2 < 0$, as possible in the presence of interface roughness and of terraces of opposite magnetization, a 90° orientation of two magnetic adjacent layers may occur, instead of 0° or 180°.[23,24,40] The square lattice structure of the (100) surface allows 90° domains with inequivalent anisotropy energy. The behavior shown in Fig. 5 can be interpreted then as representing the rotation of the surface iron magnetization from 90° (biquadratic interlayer coupling) to 180° (antiferromagnetic bilinear coupling) as the Fe film thickness crosses 1.5–2 ML. The magnetic moment of Fe atoms is enhanced by 30% independently of the direction of the film magnetization, a result which is in qualitative agreement with the theoretical analysis of Stoeffler and Gautier for spin-frustrated systems.[22]

The experimental evidence of enhanced magnetic moments near the interface and of a thickness-dependent orientation of the surface magnetization axis shows how delicate the energy balance is for the magnetic coupling through Cr spacer layers, at least if these present a rough surface. From Fig. 4 one also observes the AF coupling between the Cr and top Fe layers; this means that as the ferromagnetic order of iron sets in, the rough Cr interface becomes magnetically ordered. This effect stores some extra energy in the Cr buffer layer, which influences the subsequent coupling oscillation. The rotation of the surface iron magnetization into the 180° direction is due to the prevalence of exchange coupling over anisotropy, i.e., to a fine energy balance which can be easily modified by any extra energy term like strain or surface impurities. The difficulty of reproducing fully consistent experimental results when different growth conditions and substrates are employed is therefore easily understood.

VI. CONCLUSIONS

We have shown that the interfaces between Cr and Fe(100) and between Fe and Cr/Fe(100) as grown in conditions that optimize both the degree of structural order (less than perfect) and the suppression of atomic intermixing at the interface are dominated by antiferromagnetic coupling through the interface, unless magnetic frustration arises from interface roughness. The Cr magnetic moments at the Fe(100) surface are enhanced with respect to the ones in the thin film regime (3–12 ML). The Fe magnetic moment at the Fe/Cr/Fe(100) surface appears enhanced by 30% with respect to the Fe(100) surface value. The complexity of the magnetic behavior of the interface involves both the growing overlayer and the substrate near interface layers. Beyond the changes of magnetic moments, the observed changes of magnetic order of the substrate can be qualitatively described as the formation of a magnetic domain wall, between substrate and overlayer, extended over several atomic planes. The energy balance governing the formation of the interface magnetic wall, or extended magnetic interface, includes anisotropy, epitaxial strain, roughness, impurities, and of course exchange interlayer coupling. The balance may favor 90° domains when spin frustration is large, as appears to be the case for the Fe monolayer on Cr/Fe(100).

ACKNOWLEDGMENTS

We gratefully acknowledge M. Sacchi for fruitful discussions. This work was partially supported by the EC, under the HCM program. G.R. thanks H.C. Siegmann for continuous support.

*Present address: Institut de Physique, Univ. Neuchâtel, CH-2000, Switzerland.

[1] M.N. Baibich, J.M. Broto, A. Fert, F. Nguyen van Dau, F. Petroff, P. Etienne, G. Creuzet, A. Friedrich, and J. Chazelas, Phys. Rev. Lett. **61**, 2472 (1988).

[2] Y. Wang, P.M. Levy, and J.L. Fry, Phys. Rev. Lett. **65**, 2732 (1990).

[3] C. Kittel, *Introduction to Solid State Physics*, 5th ed. (Wiley, New York, 1976).

[4] In this work we will not consider the long periodicity due to this

incommensurability, the analysis being in the few monolayers regime.
[5] R.H. Victora and L.M. Falicov, Phys. Rev. B **31**, 7335 (1985).
[6] C.L. Fu, A.J. Freeman, and T. Oguchi, Phys. Rev. Lett. **54**, 2700 (1985).
[7] P.M. Levy, K. Ounadjela, S. Zhang, Y. Wang, C.B. Sommers, and A. Fert, J. Appl. Phys. **67**, 5914 (1990).
[8] J. Xu and A.J. Freeman, Phys. Rev. B **47**, 165 (1993).
[9] C. Turtur and G. Bayreuther, Phys. Rev. Lett. **72**, 1557 (1994).
[10] Zhongde Xu, Y. Liu, P.D. Johnson, and B.S. Itchkawitz, Phys. Rev. B **52**, 15 393 (1995).
[11] T.G. Walker, A.W. Pang, and H. Hopster, Phys. Rev. Lett. **69**, 1121 (1992).
[12] F.U. Hillebrecht, Ch. Roth, R. Jungblut, E. Kisker, and A. Bringer, Europhys. Lett. **19**, 711 (1992).
[13] Y.U. Idzerda, L.H. Tjeng, H.J. Lin, C.J. Gutierrez, G. Meigs, and C.T. Chen, Phys. Rev. B **48**, 4144 (1993).
[14] A. Berger and H. Hopster, Phys. Rev. Lett. **73**, 193 (1994).
[15] A. Vega, C. Demangeat, H. Dreyssé, and A. Chouairi, Phys. Rev. B **51**, 11 546 (1995).
[16] S.F. Alvarado and C. Carbone, Physica B **149**, 43 (1988).
[17] C. Carbone and S.F. Alvarado, Phys. Rev. B **36**, 2433 (1987).
[18] J. Unguris, R.J. Celotta, and D.T. Pierce, Phys. Rev. Lett. **67**, 140 (1991).
[19] J. Unguris, R.J. Celotta, and D.T. Pierce, Phys. Rev. Lett. **69**, 1125 (1992).
[20] J. Unguris, Joseph A. Stroscio, R.J. Celotta, and D.T. Pierce, Phys. Rev. B **49**, 14 564 (1994).
[21] D. Stoeffler and F. Gautier, Phys. Rev. B **44**, 10 389 (1991).
[22] D. Stoeffler and F. Gautier, J. Magn. Magn. Mater. **147**, 260 (1995).
[23] S. Adenwalla, G.P. Felcher, Eric. E. Fullerton, and S. Bader, Phys. Rev. B **53**, 2474 (1996).
[24] Eric E. Fullerton, K.T. Riggs, C.H. Sowers, S.D. Bader, and A. Berger, Phys. Rev. Lett. **75**, 330 (1995).
[25] Ch. Roth, F.U. Hillebrecht, H. Rose, and E. Kisker, Phys. Rev. Lett. **70**, 3479 (1993).
[26] F. Sirotti and G. Rossi, Phys. Rev. B **49**, 15 682 (1994).
[27] G. Rossi, F. Sirotti, and G. Panaccione, in *Core Level Spectroscopies for Magnetic Phenomena: Theory and Experiment*, Vol. 345 of NATO *Advanced Study Institute, Series B: Physics*, edited by P. Bagus, G. Pacchioni, and F. Parmigiani (Plenum, New York, 1995).
[28] G. Panaccione, Ph.D. thesis, Univ. Paris VI and LURE, Orsay, France, 1995; G. Panaccione, F. Sirotti, and G. Rossi, J. Electron Spectrosc. Relat. Phenom. **76**, 189 (1995).
[29] G. Rossi, F. Sirotti, N.A. Cherepkov, F. Combet-Farnoux, and G. Panaccione, Solid State Commun. **90**, 557 (1994).
[30] G. van der Laan and B.T. Thole, Solid State Commun. **92**, 427 (1994); B.T. Thole and G. van der Laan, Phys. Rev. B **50**, 11 474 (1994).
[31] G. van der Laan, Phys. Rev. B **51**, 240 (1995), and references therein.
[32] F.U. Hillebrecht, H.B. Rose, T. Kinoshita, Y.U. Idzerda, G. van der Laan, R. Denecke, and L. Ley, Phys. Rev. Lett. **75**, 2883 (1995).
[33] Ch. Roth, H. Rose, F.U. Hillebrecht, and E. Kisker, Solid State Commun. **86**, 647 (1993).
[34] F. Sirotti, G. Panaccione, and G. Rossi, Phys. Rev. B **52**, R17 063 (1995).
[35] G. Rossi, G. Panaccione, and F. Sirotti, in *Magnetic Ultrathin Films, Multilayers and Surfaces*, edited by E. Marinero *et al.*, MRS Symposia Proceedings No. 384 (Materials Research Society, Pittsburgh, 1995), p. 447.
[36] The ND was obtained after a background subtraction, taking into account the single-peak intensities in the total photoemission spectra, i.e., magnetically averaged. The ND value was normalized to different temperatures and corrected for the two (120 eV and 150 eV) photon energies. The ND of the clean Fe(100) surface measured at 120 eV photon energy was taken equal to 1 as a reference value. It is well known that the LMDAD dichroism signal is energy dependent, as shown theoretically in Refs. 29 and 31 and also experimentally (Ref. 28). It is then necessary, when comparing two values of the ND obtained at different photon energies, to take into account this energy dependence. We used the *measured* value of the LMDAD on the Fe(100) clean surface to obtain the presented normalization.
[37] G. Rossi, F. Sirotti, and G. Panaccione (unpublished).
[38] M.D. Stiles, Phys. Rev. B **48**, 7238 (1993).
[39] J. Unguris, R.J. Celotta, and D.T. Pierce, J. Magn. Magn. Mater. **127**, 205 (1993).
[40] J.C. Sclonzewsky, Phys. Rev. Lett. **67**, 3172 (1991).