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Magnetic coupling in Fe/Cr/Fe(001) by spin-resolved empty-state spectroscopies

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Abstract

We report on a study of empty electronic states in Cr/Fe(001) films and a trilayer as obtained by means of spin-polarized inverse photoemission and absorbed current spectroscopies: both spectroscopies are sensitive to the magnetic character of the Fe film. In relatively thick (>7 ML) Cr films, our data do not reveal any polarization dependence; on the contrary, in Fe/Cr/Fe(001), we observe a reversal of the magnetic ordering of the Fe overlayer with respect to the buried Fe substrate as a function of the Cr spacer thickness, confirming the oscillating transition from FM to AF coupling between the Fe layers. © 2000 Elsevier Science B.V. All rights reserved.

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1. Introduction

Magnetic transition metals separated by thin non-magnetic layers exhibit an exchange coupling, whose ferromagnetic (FM) or antiferromagnetic (AF) character depends on the spacer thickness. In particular, Cr/Fe(001) multilayers have attracted considerable theoretical [1] and experimental [2] studies. For this system, the oscillating FM or AF coupling between the two Fe layers with varying Cr interlayer thicknesses is governed by a period that strongly depends on the growth temperature, i.e. on the film quality [3,4]. Well-ordered films, grown at 300°C, display a period of about 2 monolayers (ML) superposed on a longer one of about 12 ML.

In room-temperature-grown samples, the 'short' period oscillation is completely absent, while the 'long' period oscillation is still observable. Theoretical predictions, obviously suited for ideal systems, sketch a magnetization profile in which the alignment of successive Fe layers can be determined in terms of the trend towards AF coupling between adjacent layers within the Cr film and, at the same time, AF coupling between Cr and Fe atoms at the interfaces. This determines a FM (AF) coupling between Fe layers separated by an odd (even) number of ML of the Cr spacer. Experimental results reveal an opposite relation between the even or odd number of Cr layers and the Fe magnetic coupling; such a discrepancy is probably due to the formation of a mixed Cr–Fe phase at the interface, which delays the onset of FM–AF oscillations [5,6].

The occupied electronic states of Cr/Fe systems have been investigated by several groups, also using spin-resolved techniques [2,7]. However, the empty electronic states in magnetic systems are also particularly interesting, since they contain the

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unbalanced minority-spin holes that make up the magnetic moment. Moreover, since, in 3*d* ferromagnets, there are fewer unoccupied than occupied *d* states, in the empty region, the data analysis is simplified by the minor presence of overlapping states. Thus, we have addressed the study to the empty portion of the band structure above the Fermi level (E_F) by means of spin-polarized inverse photoemission and spin-polarized absorbed current spectroscopies (spin-polarized IPE and AC, respectively). In the present paper, we focus on Cr/Fe(001) films, with a relatively large thickness (>7 ML), and on the corresponding Fe/Cr/Fe(001) trilayers, while results on the early stages of the Cr/Fe(001) interface formation and ultra-thin films will be discussed elsewhere [8].

2. Experimental

The experiments were performed in an ultra-high-vacuum system provided with standard growth and surface characterization equipment [9] as well as with a spin-polarized electron gun [10].

The starting point for all the samples studied was a clean and ordered Fe(001) substrate obtained by annealing at 600°C a 700 ML thick Fe film grown on a clean MgO(001) single crystal. During the Cr deposition, the iron substrate was kept at 300°C in order to promote the formation of a well-ordered Cr film, while the Fe overlayer was deposited at room temperature, so as to avoid any intermixing at the Cr/Fe interface. The evaporation, led at a rate of 1.5 Å/min, was monitored by a carefully calibrated quartz-crystal microbalance and checked by X-ray photoemission spectroscopy (XPS) quantitative analysis; XPS data also indicated the absence of impurities within the detection limit (a few per cent of a ML). The low-energy electron diffraction (LEED) pattern after the Cr deposition maintained the 1×1 symmetry typical of the Fe(001) surface, according to the strong structural similarity of the two metals, even if an increase in the LEED background indicated a degradation of the surface order which appeared, however, to be independent of the Cr film thickness. Samples are magnetized in situ along the in-plane [100] easy axis direction of the Fe substrate by means of a current pulse sent through a coil surrounding the crystal. Measurements were then taken in magnetic remanence, as usual in electron spectroscopies. All the spectra reported here were taken at normal incidence and at room temperature. The polarization of the electron beam ($P=25\pm2\%$), produced by a negative affinity GaAs photocathode, could be switched from parallel to antiparallel with respect to the sample magnetization.

IPE spectra were collected in the isochromat mode, i.e. by varying the beam energy and detecting only photons of a fixed energy (in our case, $h\nu=9.4$ eV), among those emitted in the electron decay towards the empty states. AC spectra were recorded simultaneously with the IPE ones by measuring the current running to the ground from the sample (for details on the experimental apparatus, see Refs. [10,11]).

3. Results and discussion

Spin-resolved spectra for the clean Fe(001) surface and for a 11 ML thick Cr/Fe(001) film are shown in Fig. 1. The spin-integrated IPE spectrum from a Fe(100) surface (not shown) exhibits two peaks in the 2 eV energy range above E_F, as well as a broad unresolved double structure at around 10 eV [12,13]. In the spin-resolved spectra presented here, continuous lines ('spin up' acquisition channel) indicate the data obtained in the case of an antiparallel alignment between the sample magnetization and the primary beam polarization, that is when the spin of the incoming electrons is parallel to the spin of the majority electrons inside the Fe substrate, whereas dotted spectra ('spin down' channel) are acquired reversing the beam polarization. Thus, the structures B1 and C1 (B2 and C2) appearing in the spin-up (spin-down) channel for the clean surface have to be attributed to transitions towards majority (minority) states [14,15] and constitute clear evidence of the sample magnetic ordering. When Cr is deposited, the substrate contribution is attenuated, and new structures can arise, related to the interface and/or the overlayer. Thus, in thin-film spectroscopy, a careful data analysis is needed in order to disentan-

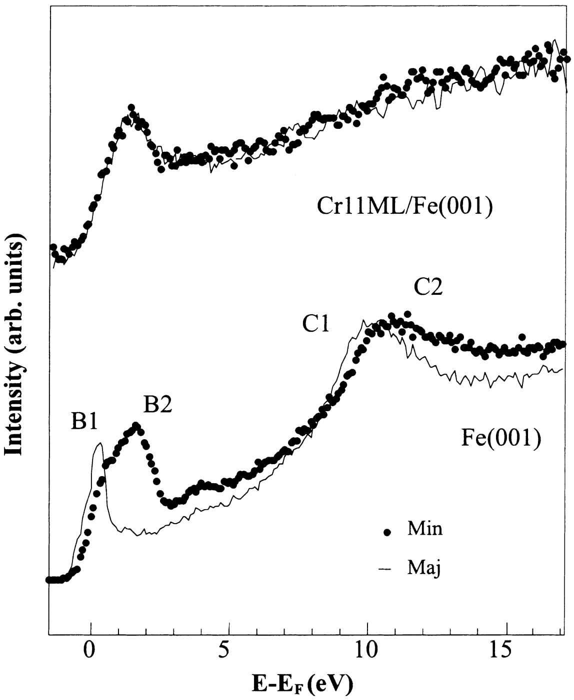

Fig. 1. IPE spectra for the Fe(001) surface (bottom) and a Cr/Fe(001) system with a 11 ML thick Cr spacer (top). Continuous lines refer to transitions between majority (with respect to the Fe substrate) states, and dotted lines to transition between minority states.

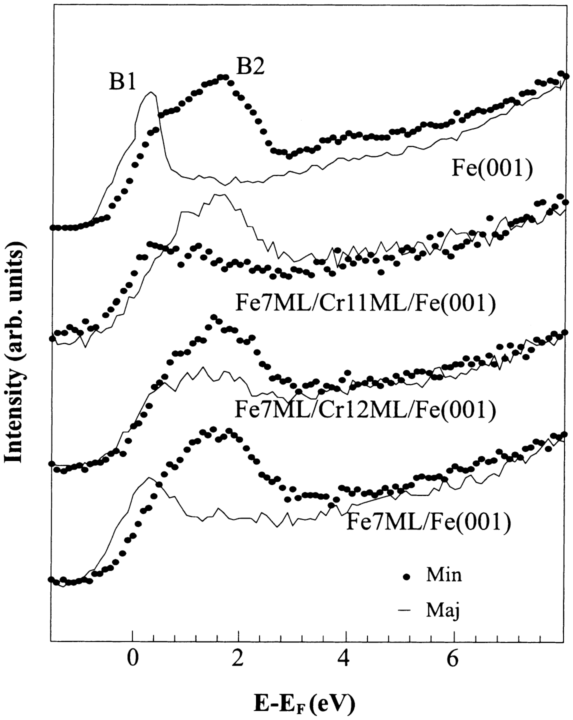

Fig. 2. IPE spectra taken (from top to bottom) form: an annealed Fe(001) surface, two Fe/Cr/Fe(001) multilayers with a 11 and 12 ML thick Cr spacer and a 7 ML thick Fe/Fe(001) homoepitaxial film as grown. The majority (continuous lines) or minority (dotted lines) character of the spectral features is again referred to the Fe substrate.

gle the substrate from the interface and overlayer contribution. However, due to the extremely small value of the electron inelastic mean free path in low d-occupancy metals [16], a Cr film (>7 ML thick) can completely mask the substrate. The latter contribution can thus be neglected, and the spectra can be interpreted in terms of the overlayer electronic structure alone. As shown in Fig. 1, the Cr film spectrum becomes dominated by a peak lying at ≈ 1.4 eV, present in both spin channels, i.e. the data do not show any polarization dependence. The peak energy is consistent with a transition towards empty states near the H_{25} point of the Cr bulk band structure [17]. The absence of any polarization dependence, however, can be explained in terms of the AF coupling between adjacent Cr layers within the film, as outlined by experimental and theoretical results: contributions to the IPE spectrum brought about by successive Cr layers with opposite magnetization add up to an unpolarized feature, washing out any polarization dependence, as indeed can be seen in Fig. 1.

Even though the magnetization profile of the bare film does not show up in our spectra, it does affect the magnetic properties of the Fe overlayer grown on top of it, which in turn influence the spectra from multilayers. The IPE results for different Fe and Fe/Cr/Fe systems are collected in Fig. 2, where spectra for the Fe 7 ML/Cr 11 ML/Fe(001) and the Fe 7 ML/Cr 12 ML/Fe(001) trilayers are shown: as above, the spin character is again referred to majority electrons of the Fe(001) surface whose spectra are also reported for direct comparison. When considering the Fe/Cr/Fe trilayer spectra, we note first that the Fe overlayer is thick enough (7 ML) to hinder any sizeable contribution from the underlying Cr film. The present measurements can then be interpreted in terms of a pure Fe contribution, while the role of the Cr spacer is to mediate the exchange interaction with the substrate. By looking at the polarization dependence of the related spectra, a commutation between spin-up and spin-down channels, i.e. the reversal of the spin character of the B1 and B2 features, can be clearly seen when the Cr spacer thickness is increased from 11 to 12 ML. This is direct evidence of the switching from FM to AF coupling between the topmost Fe film and the buried Fe substrate when adding a single Cr layer to the spacer. The coincidence of both the 'short' and the 'long' period of the FM–AF transition when the Cr spacer thickness is increased from 11 to 12 ML makes these systems very well suited for observing the switching of the magnetic coupling between Fe layers. It has to be noted, however, that the remanent magnetization configuration is not always indicative of the sign of the interlayer exchange coupling. In fact, anisotropy-driven alignments can sometime mask exchange coupling effects during the magnetization process [18]. This does not seem to apply to the well-studied Cr/Fe system, in particular for present data that show a definite sign reversal when adding a single Cr layer.

The interchanging of the spectral shapes shown in Fig. 2 is accompanied by a strong attenuation of structure B1. IPE is a very demanding technique in terms of the structural order of the sample under investigation, and its sensitivity can be considerably lowered by a degraded surface order. As already reported [11,12], in Fe(001), this is particularly relevant for the structure B1. This is also shown in Fig. 2, where the bottom spectrum refers to a homoepitaxial Fe film, which presents a B1 peak considerably reduced with respect to the annealed surface spectra at the top of the same figure. Thus, we can attribute the discrepancies observed comparing the spectra of Fig. 2 to a progressive decrease in surface order when increasing the overall multilayer thickness.

A further independent confirmation of the above findings comes from AC spectroscopy. Owing to the exchange band splitting, spin-up and spin-down electrons impinging on the crystal encounter a different band structure: this determines a spin asymmetry in the AC spectra that is related to the sample magnetic order [13,19]. Such an asymmetry is defined as the ratio between the difference and the sum of the absorbed current for spin-up and spin-down incident electrons [13,19]. The AC asymmetry spectra for the substrate and the trilayers are shown in Fig. 3. It turns out that in Fe(001), spin-up primary electrons with an energy of 8–10 eV (referred to E_F) cannot find any empty states to accommodate themselves into the solid at variance with spin-down electrons, whereas between 14 and 16 eV, only spin-up electrons can enter the crystal [13,19]. This determines the characteristic oscillations in the AC spin asymmetry, which are again strictly connected to the topmost Fe film magnetic ordering. In this case, the FM–AF coupling oscillations in the Fe/Cr/Fe trilayers are revealed by the sign reversal of the asymmetry spectra. Going towards a greater overall multilayer thickness, the spin asymmetry oscillations become progressively less pronounced; this effect is again related to a decreased surface order, and it has already been observed in different magnetic systems [20].

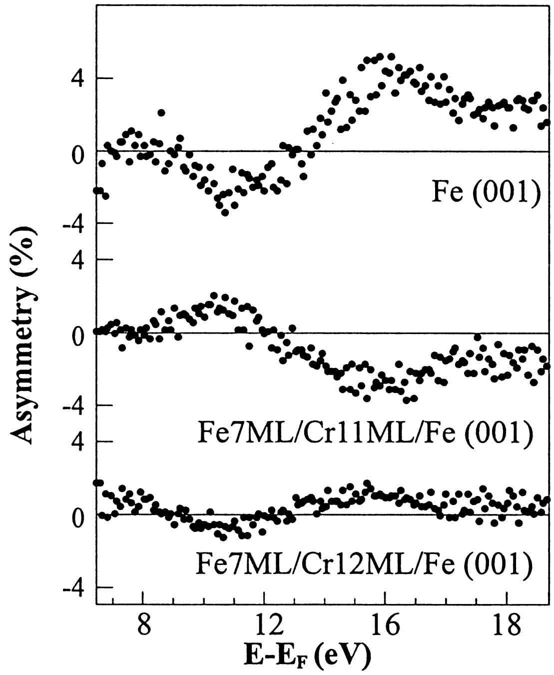

Fig. 3. AC spin asymmetry spectra for the Fe(001) surface and two Fe/Cr/Fe multilayers.

In conclusion, we have studied the empty states in Cr/Fe systems by spin-resolved spectroscopies, which have been proved to be very sensitive to the surface magnetization. In particular, we have monitored the magnetic character of the topmost Fe film in Fe/Cr/Fe(001) trilayers, observing a reversal of the magnetic ordering of the Fe overlayer with respect to the buried Fe substrate when going from an odd to an even number of ML for the Cr spacer.

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