

Spin- and angle-resolved photoemission study of ultrathin Gd films on Fe(100)

C. Carbone and E. Kisker

Institut für Festkörperforschung der Kernforschungsanlage Jülich, D-5170 Jülich, West Germany

(Received 20 March 1987)

Using spin-resolved photoemission with synchrotron radiation, we have studied the electronic structure of the Gd/Fe(100) interface. For Gd films up to one monolayer thickness, we find a high Gd 4*f*-electron spin polarization which is antiparallel to that of the Fe valence bands. This is the first direct observation of high Gd 4*f*-electron polarization. The main effect of the Gd adsorption on the Fe-derived valence bands is a depolarization. For thick Gd films (> 35 Å) the observed Gd 4*f* polarization is small, even well below the Gd *T_C*.

Rare-earth-transition-metal (RE-TM) compounds are of considerable interest because of their potential use as thermomagnetic recording media.¹ It is known that their magneto-optical activity stems from the spin-split electronic structure² which, therefore, has to be determined in order to understand the magneto-optical effects. Materials primarily considered are, e.g., amorphous alloys of Gd or Tb with Fe or Co. From Kerr-effect measurements it was concluded that the magneto-optical effect in amorphous Gd-Fe films is largely determined by Fe,³ but an understanding in terms of the electronic structure of the compound is lacking since the latter is unknown. Pure Fe and, to less extent, Co have been investigated thoroughly, and their valence-band electronic structures are considered to be well understood. This is not so much the case for pure Gd, which is difficult to investigate experimentally, in part because of its reactivity and the related difficulty of preparing atomically clean surfaces from bulk material. The Gd 4*f* electrons are expected to be fully polarized at *T*=0. In this Rapid Communication, we observe, for the first time, a spin polarization as high as 60% for the Gd 4*f* electrons, at *T*=175 K.

As a step toward the understanding of the electronic structure of RE-TM systems, we have performed in the present work spin-resolved photoemission studies of evaporated Gd films on Fe(100). Films in the monolayer (ML) regime allow us to investigate the Gd/Fe interfacial region. This system has been characterized previously by Allenspach, Taborelli, and Landolt⁴ by spin-resolved Auger electron spectroscopy, which, however, did not uncover the electronic structure. It was shown that the Gd magnetic moment is aligned opposite to the Fe moment, the interface forming a ferrimagnetic system.

Synchrotron radiation in the 30- to 70-eV photon energy range has been used to observe directly the Gd 4*f* level and the Fe-derived valence bands in the same system, for Gd coverages low enough to allow the observation of the Fe valence-band emission originating from the interfacial region. The spectrometer is described in Ref. 5. Gd was evaporated onto a clean Fe(100) surface in an ultrahigh-vacuum environment with a base pressure of 5×10^{-11} Torr. During Gd evaporation from a tungsten basket, the pressure rose to 6×10^{-10} Torr. The sample could be cooled to 170 K, and Gd evaporation was performed at this low temperature. The Gd deposition rate was cali-

brated by means of a commercial quartz microbalance film-thickness monitor. The accuracy in the Gd coverage determination is estimated to be $\pm 20\%$. The evaporation rate was about 1 Å/min. The sharp low-energy electron diffraction (LEED) pattern of the pure Fe(100) substrate becomes gradually worse with Gd adsorption, until LEED spots are hardly visible for a Gd film thickness ≥ 1 ML. We conclude that film growth is not epitaxial for those evaporation conditions.

Spectra have been taken for normal emission and *s*-polarized light, at 70-eV photon energy. Throughout this paper we refer either to the representation of the data in the form of spin-resolved energy distribution curves (SREDC's) $I^\uparrow(E), I^\downarrow(E)$, or to the spin-polarization curve

$$P(E) = [I^\uparrow(E) - I^\downarrow(E)] / [I^\uparrow(E) + I^\downarrow(E)] ,$$

and the spin-averaged intensity curves $I_0(E) = I^\uparrow(E) + I^\downarrow(E)$. Obviously,

$$I^{\uparrow,\downarrow}(E) = 0.5I_0(E)[1 \pm P(E)] , \quad (1)$$

and $\uparrow(\downarrow)$ refers to Fe majority- (minority-) spin electron emission. The Gd magnetic moment is directed antiparallel to that of Fe.⁴ We indicate by an asterisk those spin-dependent quantities which are referred to the local spin-polarization direction of the Gd.

Figure 1(a) shows the spin-averaged angle-resolved EDC after evaporation of 1 ML Gd onto the clean Fe(100) substrate. The simultaneously measured spin polarization is shown in Fig. 1(b), and the corresponding SREDC's in Fig. 2(a). The clean Fe(100) SREDC's taken prior to Gd evaporation are shown in Fig. 2(b).

Gd has the electronic configuration $4f^7(5d6s)^3$. The $4f^7$ shell is half filled and carries a magnetic moment of $7\mu_B$. The peak at about 8-eV binding energy (E_B) in Fig. 1(a) is due to the Gd 4*f* emission.⁶ The Gd 4*f* (spin-averaged) photoemission spectrum is interpreted in terms of the $4f^6$ final-state multiplet structure.⁷ The Gd 5*d6s* valence bands overlap the Fe valence bands, but they give rise to a negligible contribution ($\approx 1\%$) at 70-eV photon energy because of the very low cross section of the Gd 5*d6s* emission as compared to that of the Fe 3*d* emission.⁸ Since the background polarization is dominated by the Fe majority-spin electrons [see Fig. 2(b)], the negative dip in

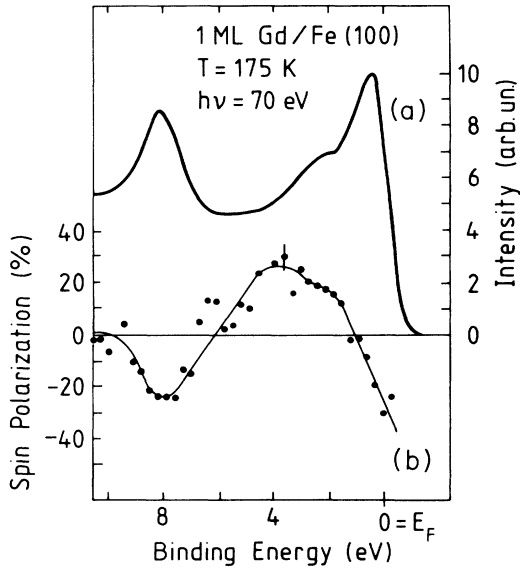


FIG. 1. (a) Spin-averaged energy distribution curve from 1 ML Gd on Fe(100) for normal emission and normal incident light at 70-eV photon energy, $T = 175$ K. (b) Spin-polarization distribution as measured simultaneously with (a).

the spin-polarization curve at around 8-eV binding energy in Fig. 1(b) indicates that the Gd 4*f* moment is aligned opposite to that of the Fe. Accordingly, the Gd 4*f*⁷ emission is observed mostly in the (Fe) minority-spin channel, which is majority-spin emission when referring to the Gd local frame. After subtracting a background of inelastically scattered electrons as indicated in Fig. 2(a), a spin polarization $P^* \approx 60\%$ is obtained for the Gd 4*f* peak. Besides the dominating Gd 4*f* majority-spin (in the Gd local frame) emission, there is a small peak of similar shape at the same binding energy of Gd 4*f* minority-spin (in the Gd local frame) emission. The presence of this peak is an indication of the incomplete polarization of the Gd 4*f* emission.

The clean Fe(100) valence-band SREDC's shown in Fig. 2(b) are understood according to Ref. 5. We identify the minority-spin peak near E_F as due to emission from initial states near Γ_{25}' , and the two majority-spin peaks at 1- and 2.6-eV binding energy (E_B) as due to initial states near Γ_{12}' and Γ_{25}' , respectively. The inelastic background at higher binding energy is small and smooth in both spin channels.

By comparing Figs. 2(a) and 2(b), we see also that the Fe valence-band emission is affected by the Gd adsorbate. The majority-spin SREDC increases near E_F , and the peak at about $E_B = 2.6$ eV decreases relative to the peak at $E_B = 1$ eV upon Gd adsorption. In the minority-spin EDC, a shoulder grows around $E_B = 3$ eV. This corresponds to an overall decrease in spin polarization across the valence-band region.

The question is how to understand the Fe-valence-band-derived SREDC's as obtained from the Gd/Fe interface [cf. Fig. 2(a)]. We will show below that the different appearance of the Fe-valence-band derived

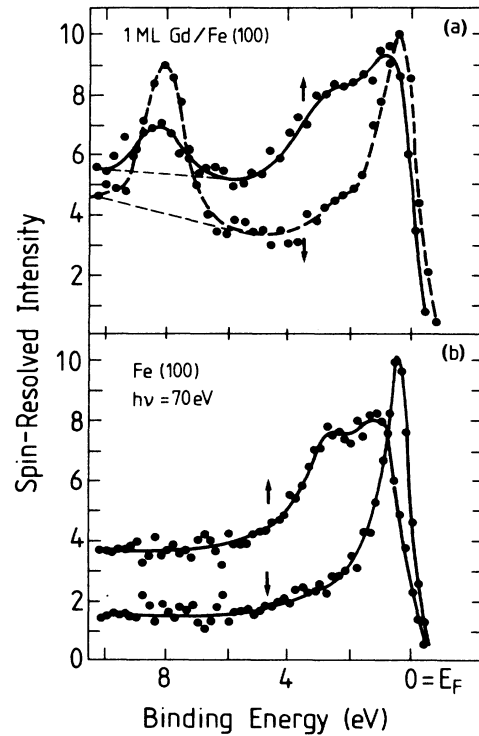


FIG. 2. (a) Spin-resolved energy distribution curves corresponding to Figs. 1(a) and 1(b). (b) Spin-resolved energy distribution curves from clean Fe(100) before Gd evaporation.

SREDC's in Fig. 2(a) as compared to the clean Fe(100) SREDC's [cf. Fig. 2(b)] is essentially due to depolarization. We rescale the spin-polarization curve $P(E)$ obtained for pure Fe(100) by a factor M_r , prior to recalculating the SREDC's by Eq. (1). For a fully magnetized sample, $M_r = 1$. With $M_r = 0.6$, we obtain the SREDC's as shown in Fig. 3 (lines) which are overlaid with the Gd/Fe(100) valence-band data of Fig. 2(a). The agreement between the measured Gd/Fe SREDC's and the calculated SREDC's in the Fe valence-band regime is good. Notably, with the same value of M_r , the observed Gd 4*f* SREDC's in Fig. 2(a) (with the backgrounds subtracted) are obtained from initially fully polarized Gd 4*f* emission. This suggests that the reductions of the Fe and the Gd polarizations are strongly correlated. This is also supported by the fact that the Gd 4*f* and the Fe-derived valence-band polarization change similarly at elevated temperatures.

In the following, we will show that, in principle, two different mechanisms could produce the changes in the Fe-derived valence bands which have been discussed above: (i) a reduced interface magnetization, or (ii) spin-flip scattering.

Because of the success of fitting the Fe-derived SREDC's from the Gd/Fe interface by a mixture of pure Fe(100) SREDC's, the result of Gd adsorption on the clean Fe(100) surface is similar to the effect of elevated temperatures on pure Fe(100) SREDC's.^{9,10} It was shown by calculations that transverse fluctuations of magnetic moments in the presence of short-range magnetic or-

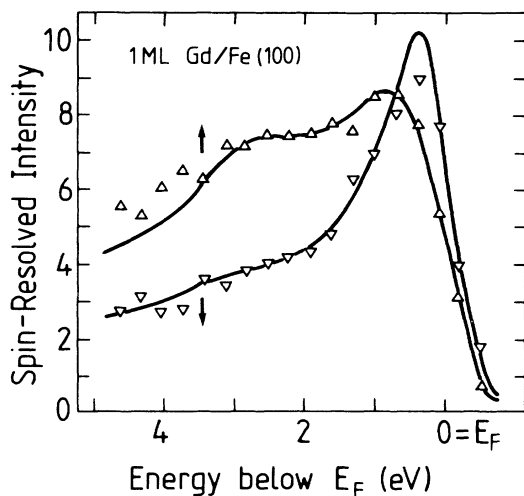


FIG. 3. Comparison of the SREDC's in the valence-band regime from 1 ML Gd on Fe(100) [data from Fig. 2(a)] with the results of a model calculation based on the pure Fe(100) SREDC's from Fig. 2(b). A reduction of the magnetization from 1 to 0.6 is assumed in the calculated curves.

der exceeding 4 \AA can explain the changes observed in the SREDC's of pure Fe(100) when approaching T_C .¹⁰ Therefore, if the depolarization observed here in the Fe-derived SREDC's from the Gd/Fe interface arises because the Gd atoms are forcing the Fe magnetic moments to point into directions deviating from the long-range magnetic order direction, it is likely that short-range magnetic order exists also among the Fe atoms in the interfacial region. The spin-split "local" electronic structure in a frame which rotates with the local magnetization direction ("fluctuating-band model"¹¹) remains largely unchanged.

The other mechanism which would result in depolarization of the Fe-derived valence-band SREDC's is spin-flip scattering. It has been shown recently that the Gd optical excitation spectrum is very complicated because of spin-orbit interaction.¹² In order to interpret the optical conductivity data, d - f exchange excitations with energy $\Delta E \approx 0.7 \text{ eV}$, " s - l " excitations among d electrons ($\Delta E \approx 0.35 \text{ eV}$), and "thermal" excitations of fluctuations of magnetic moments ($\Delta E \approx 0.01 \text{ eV}$) have been invoked.¹² In principle, the excited photoelectrons could be affected by each of the above-mentioned scattering processes. Because of the small energy loss, changes in line shape of the SREDC's might not have been detected in the present experiments.

Strong depolarizing scattering by paramagnetic Gd adlayers on Ge has been observed in threshold photoemission by Meier, Pescia, and Baumberger.¹³ Low-energy excitations similar to the thermal excitations as defined by Besnosov, Eremenko, and Gnezdilov¹² have been sug-

gested to explain the effect. In principle, the scattering length as given by Meier *et al.* would be of the right order of magnitude to explain the depolarization as observed in the present work. However, the energies of the electrons involved are quite different, and a strong energy dependence might be expected for spin-exchange scattering. This shows that further investigations of electron scattering processes in Gd appear to be promising. The fact that the depolarization is the same for the Gd layer and the Fe substrate suggests, however, that electron scattering is not the main process, since overlayer and substrate should show different amounts of depolarization.

Similar to the temperature-dependent studies of pure Fe(100), we observe also a reduction of the peak near E_F in the down-spin EDC (see Fig. 3), which is beyond the simple spin-mixing model. Since the minority-spin peak emission is strongly confined to the surface normal direction in pure Fe(100),⁵ we interpret the reduction of its amplitude as caused by a broadening of the angular distribution due to the magnetic or structural disorder, or due to electron-electron scattering.

Regardless which of the above mentioned depolarizing mechanisms is at work the conclusion is that the "local" Fe valence-band electronic structure in the interfacial region is only weakly affected by the Gd adsorption.

With comparatively thick films, the spin polarization of the Gd $4f$ emission is only 10% at $T = 175 \text{ K}$. Since Gd films of 35 \AA thickness have a Curie temperature close to that of bulk Gd,¹⁴ we have to find a mechanism which could cause the small polarization obtained here from a highly polarized system. It is known that the magnetic anisotropies of Gd are complicated and temperature dependent.¹⁵ Since we are measuring only one component of the spin polarization (parallel to the Fe polarization), these anisotropy effects might prevent the alignment of the Gd film magnetization along this axis. However, also the role of the strong spin-orbit coupling on the quasiparticle electronic structure has still to be explored.¹²

In conclusion, we have observed for the first time highly spin-polarized Gd $4f$ photoelectrons. We are able to observe the Gd $4f$ emission together with the Fe-derived valence-band emission from the ferrimagnetic Gd/Fe(100) interface. The spin-resolved Fe-derived valence band EDC's deviate strongly from those of pure Fe(100) and are similar to those of pure Fe at much higher temperature. This suggests either the presence of transverse fluctuations of the Fe magnetic moments in the presence of short-range order in the interfacial region or depolarizing scattering in the Gd overlayer (or a combination of both). In any case, it is concluded that changes in the local Fe valence-band electronic structure due to the Gd adsorption are minor.

We wish to thank Professor M. Campagna and the Berliner Elektronenspeicherring-Gesellschaft für Synchrotronstrahlung (BESSY) staff for their support.

- ¹For a review, see, e.g., R. J. Gambino and T. R. McGuire, *J. Magn. Magn. Mater.* **54–57**, 1365 (1986).
- ²J. L. Erskine and E. A. Stern, *Phys. Rev. B* **8**, 1239 (1973).
- ³K. Sato and Yuji Togami, *J. Magn. Magn. Mater.* **35**, 181 (1983).
- ⁴M. Taborelli, R. Allenspach, G. Boffa, and M. Landolt, *Phys. Rev. Lett.* **26**, 2869 (1986); R. Allenspach, M. Taborelli, and M. Landolt, *Phys. Rev. B* **34**, 6112 (1986).
- ⁵E. Kisker, K. Schröder, W. Gudat, and M. Campagna, *Phys. Rev. B* **31**, 329 (1985).
- ⁶J. K. Lang, Y. Baer, and P. A. Cox, *J. Phys. F* **11**, 121 (1981).
- ⁷G. K. Wertheim, A. Rosencwaig, R. L. Cohen, and H. J. Guggenheim, *Phys. Rev. Lett.* **27**, 505 (1971).
- ⁸J. J. Yeh and I. Lindau, *At. Data Nucl. Data Tables* **32**, 1 (1985).
- ⁹E. Kisker, *J. Magn. Magn. Mater.* **45**, 23 (1984).
- ¹⁰E. M. Haines, R. Clauberg, and R. Feder, *Phys. Rev. Lett.* **54**, 932 (1985).
- ¹¹V. Korenman, J. Murray, and R. E. Prange, *Phys. Rev. B* **16**, 4032 (1977).
- ¹²A. B. Besznosov, V. V. Eremenko, and V. P. Gnezdilov, *J. Magn. Magn. Mater.* **43**, 243 (1984).
- ¹³F. Meier, D. Pescia, and M. Baumberger, *Phys. Rev. Lett.* **49**, 747 (1982).
- ¹⁴M. Farle and K. Baberschke, *Phys. Rev. Lett.* **58**, 511 (1987).
- ¹⁵M. Ponomarev, *J. Magn. Magn. Mater.* **61**, 129 (1986).