Magnetic Coupling of Surface Adlayers: Gd on Fe(100)

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The magnetic ordering of very thin Gd layers on Fe(100) has been characterized with use of the element-specific magnetic resolving power of spin-polarized Auger spectroscopy. We find antiparallel coupling of Gd to the Fe substrate, and identify surface-specific temperature dependence of the Gd and Fe sublattice magnetizations. The magnetic correlation length in Gd above its ordering temperature is obtained.

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Rare-earth–transition-metal (RE-TM) compounds[1] have found successful applications, in particular as permanent magnets and high-density magnetic data-storage media.[2] Their ferrimagnetic properties are essentially determined by the antiparallel magnetic coupling of the RE $4f$ (and $5d$) spin moment to the TM $3d$ moment. This seems to be a general, though hardly understood, phenomenon, for it occurs in ordered as well as amorphous alloys. A relevant question is what structural conditions are necessary for this magnetic coupling to occur. The present investigation sets out to make contact between RE-TM compounds and "atomic engineering" by studying a very thin RE film on a TM single-crystal surface. The model system here is Gd on Fe(100), and the following points will be addressed: (i) What is the magnetic coupling of a very thin Gd layer to the single-crystalline Fe substrate? (ii) How does the magnetic adlayer influence the temperature dependence of the surface magnetization? (iii) What is the magnetic behavior of a thick Gd film; in particular, what is the range of magnetic correlation above the Gd Curie temperature?

SPAES, spin-polarized Auger-electron spectroscopy, finds its natural application in this realm because of its potential to measure element-specific, local magnetizations at surfaces. Detailed knowledge of the Auger processes is not required here. It is, however, another important aspect of the present investigation and will be discussed in detail elsewhere.[3]

The experimental setup is the one used in previous studies.[4] The magnetized single-crystalline Fe(100) surface is irradiated with unpolarized electrons of 2500 eV at 70° off normal, the secondary electrons emitted in normal direction are energy analyzed by means of a cylindrical mirror analyzer, and the degree of spin polarization P is determined by a Mott detector. P is defined as $P=(n_\uparrow-n_\downarrow)/(n_\uparrow+n_\downarrow)$, where $n_\uparrow$ ($n_\downarrow$) denotes the number of electrons with magnetic moment parallel (opposite) to the Fe magnetization. A liquid-N_2 heat-exchange system and radiation heating make it possible to vary the sample temperature between 150 and 900 K. The Fe(100) surface is prepared by cycles of Ne^+ bombardment at grazing incidence and heating to 900 K. Polycrystalline Gd films of various thicknesses (0.3 to 30 Å) have been slowly evaporated from a W filament, with a rate of 0.5–1 Å/min. The absolute determination of the film thickness by comparing Gd *NNN* and Fe *MMM* Auger signals is reliable only within $\simeq 50\%$; the relative error between films of different thicknesses, however, is much smaller, about 10% to 15%. We note that the overlayers grow homogeneously: As the Auger signal of Gd increases, the Fe signal decreases correspondingly. Moreover, the low-energy electron diffraction pattern of Fe (100) vanishes for films of $\simeq 2.5$ monolayer thickness ($d\simeq 5$ Å), indicating that island formation or alloying does not occur. Furthermore, variation of the substrate temperature during evaporation between 150 and 600 K did not alter the characteristics of the Gd films. An additional proof of good film quality and cleanliness is the fact that all Gd overlayers order ferromagnetically independent of film thickness. Epitaxial growth, on the other hand, is not possible because of different symmetries and the too large lattice mismatch of Fe and Gd.

The Auger as well as true secondary-electron spin polarizations of clean Fe(100) have recently been discussed,[5] and an analysis of Gd Auger spin polarization will soon be published.[3] Here, we would like to compare, in a qualitative manner, Fe and Gd Auger spin polarizations in order to obtain local, element-specific magnetic information.

Figure 1 depicts the spin polarization of electrons emitted through the strongest Fe and Gd Auger decay channels, obtained for a Gd film of $\simeq 1$ monolayer thickness ($d\simeq 2.4$ Å) on the Fe(100) substrate at $T=150$ K. The spin polarization of the Fe $M_{23}M_{45}M_{45}$ Auger line at 43 eV exhibits a *positive* peak, whereas the two most intense Gd lines at 103 and 133 eV, the $N_{45}O_{23}N_{67}$ and the resonant $N_{45}N_{67}N_{67}$ decays,[6] respectively, are strongly *negative* even in the raw data. This unambiguously shows that Gd is magnetically ordered and that the coupling of Gd to the Fe substrate is antiparallel: The *MMM* spin polarization of pure Fe is established to be positive,[5] $P_{\text{eff}}=37\%$ in the main line, i.e., majority electrons are predominantly emitted, and for the resonant *NNN* line of Gd we expect also positive polarization, $P_{\text{eff}}=100\%$ in the simplest picture assuming spin conservation. Thus the sign of the raw data of Fig. 1 clearly proves

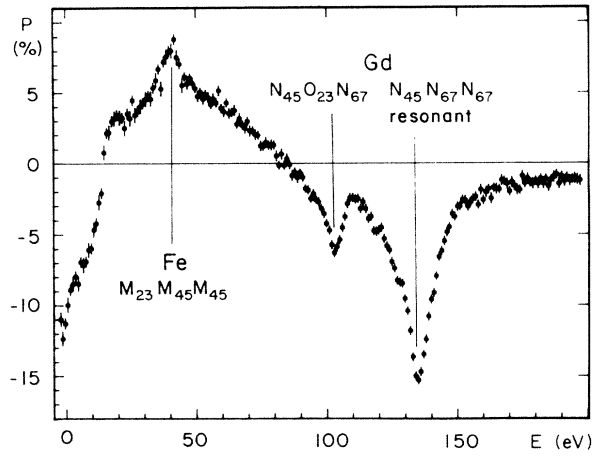

FIG. 1. Spin polarization vs kinetic energy of secondary electrons from a Gd film on Fe(100), excited with primary electrons of 2500 eV. $T = 150$ K; the film thickness is $\simeq 1$ monolayer (2.4 Å).

that the Gd overlayer couples antiferromagnetically to the Fe substrate.

Proper background subtraction in intensity and spin polarization yields $|P_{\text{eff}}| = 77\%$ for the resonant Gd $N_{45}N_{67}N_{67}$ (^7F) transition. Considering admixture of weaker polarized *NNV* and *NVV* as well as spin-flip transitions, we conclude that the Gd $4f$ moments are *fully* aligned opposite to the Fe magnetization at zero temperature. P_{eff} remains unchanged when we reduce the Gd coverage to the detection limit of $\frac{1}{10}$ monolayer, which means that even isolated Gd atoms or small patches chemisorbed on Fe(100) align their $4f$ moments opposite to the Fe magnetization. We conclude therefore that the RE-TM coupling is established by a single RE atom chemisorbed on a TM surface. Hence, the geometrical arrangement of the neighboring atoms is not of crucial importance for the antiparallel coupling, and in particular, no three-dimensional RE-TM coordination or RE-RE coupling is necessary.

The relevance of the present experiment is the establishment of the antiferromagnetic coupling in a well defined arrangment of TM substrate and RE overlayer. Antiparallel coupling between TM and RE electron spins has already been observed in a great number of alloys,[1] and amorphous films[7] like GdCo or TbFe. In contrast to these systems with complicated nearest-neighbor geometry, our model system provides a tractable test case for spin-polarized metal-metal interface calculations.

The strict coupling of the Gd $4f$ moment to the Fe $3d$ moment might lead to an interesting application: Gd atoms distributed on a surface can be used as a "magnetic marker" of the magnetism of the topmost surface layer. Then the polarization signal in magnetic-domain readout is amplified by the strong RE magnetic moments.

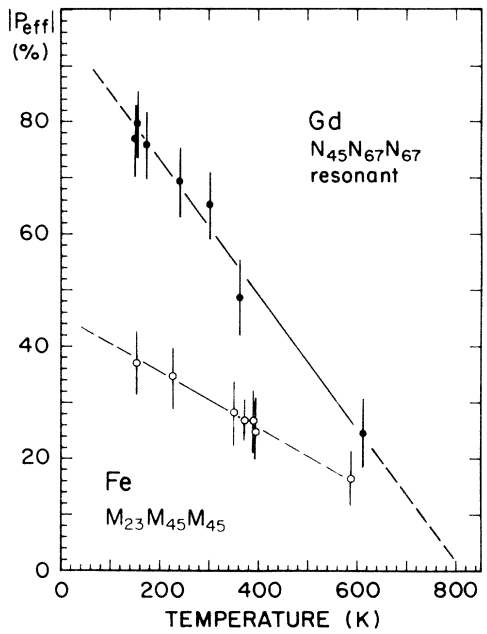

FIG. 2. Absolute value of the effective spin polarization of the resonant $N_{45}N_{67}N_{67}$ Gd Auger line at 133 eV (full circles) and of the Fe $M_{23}M_{45}M_{45}$ line at 43 eV (open circles) vs temperature, for $\simeq 1$ monolayer (2.4 Å) of Gd on Fe(100).

A second point of relevance is that SPAES allows us simultaneously to record sublattice magnetizations in composite systems. Until now, similar information could be gained by related techniques like spin-polarized photoemission[8] and magneto-optic Kerr effect.[9] They are, however, sensitive to the $3d$ transition-metal sublattice only.

The technique of SPAES goes one step further: At any temperature, the magnetization of *both* "sublattices" can be recorded at the same time, distinguishing the two species by choosing the respective Auger kinetic energy. On the Fe *MMM* line at 43 eV, we clearly see a hysteresis loop typical for Fe as discussed by Allenspach *et al.*,[10] and an inverted loop is observed on the Gd *NNN* line at 133 eV, with the same coercivity and shape. These observations confirm that the Gd moments closely follow the magnetic behavior of the Fe substrate, at least at sufficiently low temperature. Let us now discuss the surface magnetic properties by considering the thermodynamic behavior of the magnetic adlayer. In Fig. 2, the temperature dependence of the resonant Gd $N_{45}N_{67}N_{67}$ and the Fe $M_{23}M_{45}M_{45}$ Auger spin polarization is shown for a film of $\simeq 1$-monolayer thickness, $d \simeq 2.4$ Å. The data on the Gd *NNN* line represent the magnetization of the outermost layer, which consists of Gd. P_{eff} can be considered as the Gd sublattice magnetization of the Gd-Fe interface bilayer. Its temperature dependence indicates that the Gd-Fe surface sheet has a magnetic or-

dering temperature in the vicinity of 800 K, which is considerably higher than the Curie temperature of bulk Gd, $T_C(\mathrm{Gd}) = 293$ K, but likewise far below the one of bulk Fe, $T_C(\mathrm{Fe}) = 1041$ K. Moreover, we see that the relation between polarization and temperature is nearly linear in the entire range investigated. This relates to the surface, since it is clearly distinct from the bulk magnetization curve. Both observations lead to the conclusion that the magnetic coupling of the surface sheet to the Fe bulk is relaxed due to the presence of the Gd moments.

The T dependence of the Gd adlayer should be compared to the corresponding T behavior of the topmost Fe layer underneath. The lower curve in Fig. 2 gives the effective Auger polarization of the Fe *MMM* decay at 43 eV. The data, at the present state of accuracy, suggest a linear behavior for 150 $K < T < 600$ K, which also points to a reduced Curie temperature. Because of the small probing depth[11] of $\simeq 5$ Å at this energy we are essentially detecting the magnetization of the Fe sublattice in the interfacial Fe-Gd bilayer. We note that future refined measurements will provide invaluable information with regard to two different aspects: First, the surface T dependence in Fig. 2 could be compared to the Fe near-bulk magnetization revealed by the Fe $L_3M_{45}M_{45}$ Auger polarization at 703 eV, with a probing depth of approximately 20 Å.[11] Second, experiments at elevated temperatures open possibilities of determining surface critical exponents.[12] The details of the behavior, in particular the question of the surface phase transition, remain a topic of future study.

The third point of interest is the behavior of thicker Gd films, where the RE-RE coupling comes into play. A film of thickness $d \simeq 30$ Å is found to order magnetically below $T_C(\mathrm{Gd})$ with its magnetization pointing in opposite direction to the magnetization of the Fe substrate and, while going around a hysteresis loop, even opposite to the driving magnetic field. We note that for a film thickness considerably below the width of a domain wall, the RE-TM coupling at the interface is strong enough to keep the entire film in its reversed magnetic state. Above $T_C(\mathrm{Gd})$, the intermediate thickness range is interesting: The effective Gd Auger spin polarization is then probing the one-dimensional magnetic correlation length $\xi(T)$ of the RE-RE coupling, i.e., the distance from the oriented interfacial Gd layer to where the magnetization has dropped to $1/e$ of its value. Note that ξ may be different from the three-dimensional magnetic correlation length,[13] because with the present technique we are averaging over all the Gd moments of each layer parallel to the surface. As an illustration, in Fig. 3, we present the resonant $N_{45}N_{67}N_{67}$ Auger spin polarization versus film thickness, for a temperature well above $T_C(\mathrm{Gd})$, $T = 360$ K. We see a polarization which is constant for

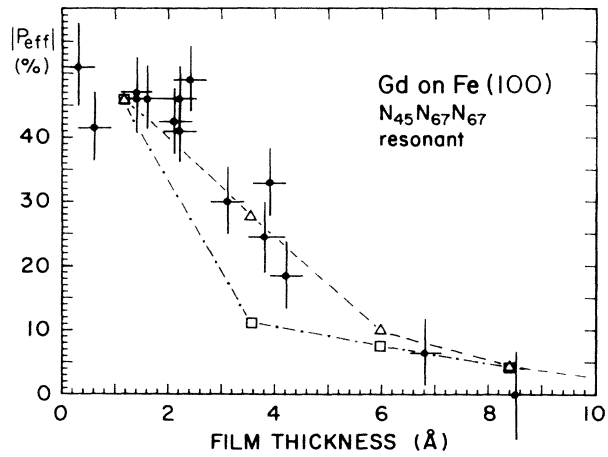

FIG. 3. Absolute value of the effective spin polarization of the resonant $N_{45}N_{67}N_{67}$ Gd Auger line at 133 eV vs film thickness; $T = 360$ K. Circles, experiment. Triangles, polarization profile calculated in the mean-field approximation as described in text; 1 monolayer = 2.4 Å. The polarization of the first layer has been normalized to the experimental value. Squares, same as triangles, but with a negative exchange constant between topmost and second Gd layer. Straight lines have been drawn through the calculated profiles.

films of $d \leq 2.4$ Å, and rapidly decreasing for thicker films. The interpretation is obvious: The first Gd layer couples magnetically to the Fe substrate through the strong antiferromagnetic Gd-Fe coupling, whereas the weak Gd-Gd exchange allows the orientation of the moments to persist within a characteristic correlation length ξ. From Fig. 3, therefore, the correlation length in Gd films above $T_C(\mathrm{Gd})$ can be deduced: The magnetization profile within the Gd adlayers is calculated within the mean-field approximation[14] respecting the boundary condition of a magnetically ordered Gd layer at the interface. The spin-polarization signal then is an average of emission from all the Gd layers atop the Fe crystal, weighted with the universal attenuation lengths of hot electrons in metals.[11] With the assumption of $T_C = T_C(\mathrm{Gd})$ for the film Curie temperature we find agreement between calculated polarization profile and experiment; see Fig. 3. The calculated magnetization profile then yields a correlation length $\xi = 2.5 \pm 0.5$ Å at $T/T_C = 1.22$.

In a recent experiment, Weller *et al.*[15] observed antiparallel ordering of the topmost Gd surface layer to the underlying bulk Gd. The present data do not show any indication of similar behavior. Taking the same model as above, but with a negative exchange constant between topmost and second Gd layer, we obtain a modified polarization profile as shown in Fig. 3. We ascribe the discrepancy to the different structures of the films, polycrystalline in the present case, epitaxially grown in Ref. 15.

These qualitative results show that SPAES provides a means of retrieving the magnetic correlation length $\xi(T)$ and surface-specific magnetic behavior.

A last remark concerns the peculiar polarization behavior at very low energies in Fig. 1. We find that below 20 eV P dramatically decreases with decreasing energy and assumes negative values. In this energy range, all $3d$ ferromagnets investigated so far exhibit a polarization enhancement arising from exchange scattering between low-energy hot electrons and spin-polarized $3d$ conduction electrons.[16-18] Also pure polycrystalline Gd was found to exhibit a similar P enhancement.[3] The observed negative polarization at 1 eV does not, however, correspond to a weighted average of Fe and Gd electrons, since the escape depth is very large, > 50 Å,[11] and hence most of the cascade electrons stem from the positively polarized Fe substrate. We ascribe the unexpected polarization spectrum of Fig. 1 at very low kinetic energy to the fact that the exchange scattering of hot electrons with the Gd $4f$ electrons is much stronger than with the Fe $3d$ electrons. Thus, during the escape process, the initially strongly polarized Fe $3d$ electrons are depolarized, and even inversely polarized by the ferromagnetic Gd moments present in the overlayer. This is quite analogous to depolarization by paramagnetic moments.[19,20] We note that this effect is only operative at low energies where the exchange-scattering cross section is large. The remaining part of the spin-polarization spectrum above $\simeq 20$ eV is not affected at all, and this is essential for comparing magnetic moments at different atomic sites using Auger electrons at higher kinetic energies.

To summarize, we have investigated the magnetic coupling of a very thin Gd adlayer on a Fe(100) surface, with the conclusion that individual Gd atoms chemisorbed on Fe(100) strongly couple antiferromagnetically to the substrate. A Gd adlayer of monolayer thickness seems to induce a Gd-Fe surface Curie temperature of $\simeq 800$ K, and the Gd sublattice magnetization exhibits roughly linear temperature dependence in the range $0.2T_{C_s} < T < 0.8T_{C_s}$. Thicker Gd films of 12 atomic layers order magnetically below the Gd Curie temperature with magnetization opposite to the one of the Fe substrate, demonstrating that the Gd-Fe coupling across the interface is strong enough to keep thin films in reversed magnetic state and that the Gd-Gd coupling within the film is still ferromagnetic. Films of intermediate thickness (1–5 monolayers) allow us to determine the magnetic correlation length of the Gd-Gd coupling above T_C(Gd). We wish to emphasize that SPAES is a promising technique for magnetic characterization particularly because of its ability to record sublattice magnetizations near or at surfaces.

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