Interfacial Magnetism of Eu/Gd(0001) Studied by Magnetic Circular Dichroism in Photoemission

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We report on the fabrication of an atomically flat heteromagnetic rare-earth interface of 1-ML-Eu(6×6)/Gd(0001), and its element-specific magnetic analysis by means of magnetic dichroism in photoemission, revealing a high net Eu magnetization at low temperatures. This magnetic Eu phase is due to a strong positive interlayer exchange coupling across the interface which overrules the weak negative intralayer coupling between Eu spins in the hexagonal two-dimensional lattice. [S0031-9007(98)05556-2]

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Magnetic coupling in layered metallic structures has become a key issue in thin-film magnetism since the observation of oscillatory exchange coupling across nonferromagnetic spacer layers [1,2]. Although this phenomenon was first discovered in rare-earth (RE) superlattices [1], most studies today deal with transition metal (TM) systems because of their technological relevance to magnetic storage devices. The present theoretical understanding of TM multilayers has been developed on the basis of detailed observations revealing, e.g., short-period oscillations in interlayer coupling strength [3] as well as 90° coupling [4]. For these key findings, the fabrication of high-quality interfaces and the experimental access to atomic layers right at the interface has been essential.

In metallic RE systems, due to the localized nature of the $4f$ electrons even in the solid phase, the exchange coupling is well described by the Heisenberg Hamiltonian $H = -JS_iS_j$, where J denotes the effective coupling strength between the localized $4f$-spin moments S_i and S_j. The exchange coupling in RE metals is indirect, mediated by the $6s$ and $5d$ conduction electrons [5]; it is oscillatory and usually described in a Ruderman-Kittel-Kasuya-Yosida picture. Yet, since the 1980s, little progress has been made in understanding the coupling in RE superlattices. This is due mainly to the high intermiscibility of all trivalent RE metals, which has prevented preparation of atomically sharp interfaces [1,6]. Thus, a magnetic analysis of atomic layers right at a RE interface has not even been attempted so far. As a consequence, the present-day picture of RE interfaces is still based on the first semiempirical model assuming ideally flat interfaces [7].

In the present Letter, we report on the fabrication of an atomically flat heteromagnetic RE interface near a surface and its magnetic analysis by means of magnetic circular dichroism (MCD) in photoemission (PE). The Eu/Gd interface, a text-book example of a Heisenberg system with spin-only localized $4f$ moments, is found to form a thermodynamically stable hexagonal two-dimensional lattice. The temperature dependence of the interface magnetization is monitored by MCD in PE, using $4f$ PE spectra. By comparison with a layer-dependent mean-field calculation, this allows for the first time to determine the strength of interlayer ($J_\perp$) and intralayer ($J_{||}$) exchange coupling for this RE interface.

The PE experiments were performed at the U2-FSGM crossed-undulator beam line of the Berliner Elektronenspeicherring für Synchrotronstrahlung (BESSY), providing soft x rays with ≅45% circular polarization around $h\nu = 48$ eV [8]. The light was incident at an angle of 15° with respect to the film plane, and the photoelectrons were collected around surface normal with a hemispherical electron-energy analyzer. For x-ray absorption (XA) measurements the SX700/III monochromator at BESSY [9] was employed. The XA spectra were recorded in the total-electron-yield mode for different angles of light incidence with respect to the sample surface. 100-Å-thick Gd(0001) films, monitored by a quartz microbalance, were grown epitaxially on W(110) at 270 K by vapor-phase deposition of high-purity Gd metal evaporated from an electron-beam heated Mo crucible. The base pressure in the experimental chamber was 3×10^{-11} mbar, rising to 2×10^{-10} mbar during evaporation. Subsequent annealing at 630 K led to a well-ordered Gd(0001) surface. Thin Eu films were deposited onto the Gd film at 300 K in an analogous way. A 1 monolayer (ML) Eu coverage, Θ_0, could be calibrated via the $4f$-binding energy as well as the shape of the Eu-$4f$ PE spectrum, since the $^7F_6$ binding energy passes through a minimum of (2.45 ± 0.02) eV at Θ_0. For $\Theta > \Theta_0$, two additional features, growing in intensity with Θ, appeared in the PE spectrum, one at each side of the multiplet, which can be assigned to interface and surface Eu atoms, respectively. An analogous behavior had been reported for Yb/Mo(110) [10]. The films were magnetized remanently in plane at 25 K by short field pulses of about 2000 Oe produced by a nearby electromagnet.

The lateral structure of 1-ML-Eu/Gd(0001) at 25 K was studied by low-energy electron diffraction, revealing a 6×6 hexagonal superstructure. Here, the nearest-neighbor (nn) distance between Eu atoms, $a_{\mathrm{nn}}^{\mathrm{Eu}} = 4.3$ Å, was found to be $\cong 20\%$ larger than the one between Gd atoms ($a_{\mathrm{nn}}^{\mathrm{Gd}} = 3.58$ Å), which is a consequence of the larger ionic radius of divalent Eu as compared to trivalent Gd. But also compared to bulk bcc Eu, the nn Eu distance in 1-ML-Eu/Gd(0001) is elongated by 9%. These findings are quite analogous to earlier results for Sm(0001) [11], where a 5×5 hexagonal structure had been found for the divalent (0001) surface of bulk-trivalent Sm metal.

In Fig. 1, Eu-$4f$ and Gd-$4f$ PE spectra of 1-ML-Eu/Gd(0001) are shown for two temperatures, recorded with circularly polarized light and the sample magnetized remanently in plane. Since Eu and Gd both have a half-filled $4f$ shell, analogous ^7F_J final-state PE multiplets are observed, but with different binding energies and multiplet splittings. For an identical magnetic order of the two elements, one would therefore expect identical effects in the Eu and Gd spectra. The Gd-$4f$ PE data in Fig. 1 resemble those of uncovered ferromagnetic Gd(0001) [12], yet without a surface component on the high-binding-energy edge due to the adsorption of 1 ML Eu. The Eu-$4f$ multiplet behaves analogously to that of Gd, i.e., the shapes of both $4f$ multiplets are peaked or rounded depending on sample magnetization. This clearly demonstrates a high net Eu magnetization, i.e., a predominantly parallel orientation of the Eu spins within the Eu adlayer and with respect to the Gd magnetization. This behavior is striking since bulk Eu metal is antiferromagnetic below $T_{\mathrm{N}} = 90.4$ K.

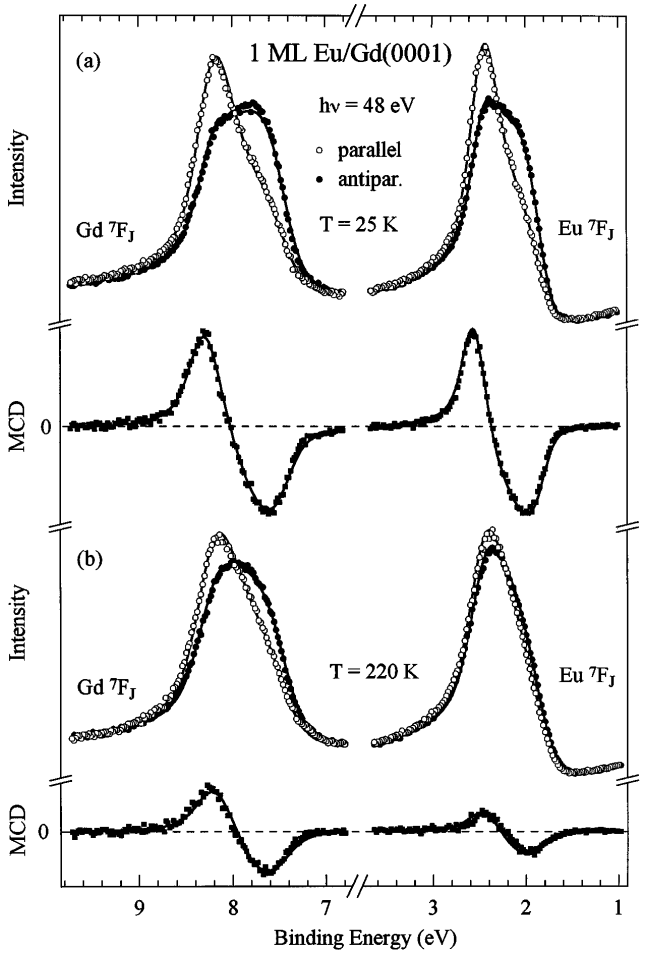

FIG. 1. $4f$ PE spectra of 1-ML-Eu/Gd(0001) at (a) 25 K and (b) 220 K. Open (filled) symbols represent parallel (antiparallel) orientation of sample magnetization and photon spin. The Eu and Gd spectra were normalized to the same overall intensity of the ^7F_J multiplet corrected for different backgrounds. The filled squares reproduce the intensity difference $I_{\mathrm{parallel}} - I_{\mathrm{antipar}}$. The solid curves through the data points represent the results of least-squares fits.

For a comparison of the magnitudes of the magnetization of the Eu overlayer and the underlying Gd film, it is not justified to simply compare the peak-to-peak signal of the MCD spectra, although Eu and Gd give rise to the same ^7F_J final-state multiplet. It has been shown [13] that the intensity of each line in the magnetic linear dichroism (MLD) spectrum, and hence the peak-to-peak MLD signal, is proportional to the expectation value $\langle M^2 \rangle$ of the square of the magnetic quantum number M. Similarly, it can be shown that the intensity of each line in the MCD spectrum is proportional to $\langle M \rangle$. However, this proportionality is only valid if the line and the multiplet parameters, such as the linewidth and the multiplet splitting, do not change. A modification of one of these parameters for constant $\langle M \rangle$ will cause an increase or a decrease of the peak-to-peak MCD signal. Consequently, since the line parameters and the multiplet splittings are different in the ^7F_J multiplets of Eu and Gd, a simple comparison of the magnitude of the MCD signal does not give correct information on the ratio $\langle M \rangle_{\mathrm{Eu}\text{-}4f} / \langle M \rangle_{\mathrm{Gd}\text{-}4f}$. To obtain this ratio, it is necessary to fit the $4f$ PE spectra obtained for Eu and Gd in order to extract the sizes of $\langle M \rangle_{\mathrm{Eu}\text{-}4f}$ and $\langle M \rangle_{\mathrm{Gd}\text{-}4f}$. For this purpose, the Gd spectra plotted in Fig. 1(a) were simultaneously least-squares fitted with a common parameter set, assuming Doniach-Šunjic line shapes for the individual PE lines, convoluted by a Gaussian to account for broadening. The energy separations of the ^7F_J multiplet components were taken from optical data for Eu, expanded in the case of Gd to account for the higher nuclear charge. The relative intensities of the ^7F_J components were obtained by a superposition of the isotropic spectrum I^0, the MCD spectrum I^1, and the anisotropic spectrum I^2, following the notation of Ref. [14], with relative weights obtained (at 25 K) from the PE spectrum of an uncovered Gd surface. The good description of the data with these parameters [solid curves in Fig. 1(a)] shows that the MCD effect in Gd, and hence the magnetization of the Gd film at 25 K, is not influenced by adsorption of an Eu monolayer. Note, however, that the Gaussian linewidth of the Gd-bulk emission is

significantly larger than for a clean Gd surface. The fit results for the 4f PE spectra of the Eu monolayer are also shown in Fig. 1(a), with the Eu-4f MCD signal at 25 K being reduced to (80 ± 10)% of the Gd-4f value; again, an increase in the Gaussian linewidth with respect to the PE spectrum of a thick Eu film was found. This broadening could be caused by variations in the local environment of Eu and Gd atoms in the topmost layers due to the 6×6 superstructure, leading to changes in the local potential and hence in the Gd-4f and Eu-4f binding energies. Since these binding-energy shifts cannot be resolved, they lead only to an increase in the Gaussian linewidth.

The temperature dependences of $\langle M \rangle_{\text{Eu-}4f}$ and $\langle M \rangle_{\text{Gd-}4f}$, extracted from the experimental data by the described fit procedure, are displayed in Fig. 2: With increasing temperature, $\langle M \rangle_{\text{Gd-}4f}$ is reduced, following qualitatively the behavior of bulk Gd. $\langle M \rangle_{\text{Eu-}4f}$, on the other hand, decreases much more rapidly for temperatures between 50 and 100 K.

One possible explanation for the observed rapid decrease of $\langle M \rangle_{\text{Eu-}4f}$ could be a magnetic reconstruction of the Eu adlayer, with the Eu moments changing their orientation from in plane to out of plane. To clarify this point, MCD in XA was employed to determine the orientation of the Eu magnetization relative to the surface normal for two temperatures, 30 and 100 K. The advantage of using MCD in XA is that for an entirely in-plane magnetized sample the angular dependence of the MCD signal follows a simple cosine law [15]. As an example, Fig. 3 shows the XA spectra at the $M_{4,5}$ threshold of 1-ML-Eu/Gd(0001), taken with circularly polarized light incident at an angle of 15° with respect to the sample surface. The angular dependences of the Eu-MCD signals at 30 and 100 K are identical, clearly demonstrating the absence of such a magnetic reorientation.

For the magnetism of the Eu monolayer, two exchange-coupling constants are important: $J_{\|}$ within the Eu layer, and $J_{\perp}$ between Eu and Gd in adjacent interface layers. The fact that the shapes of the Eu-4f and Gd-4f multiplets are peaked or rounded for the same sample magnetization shows clearly that adjacent Eu and Gd interface layers are coupled ferromagnetically, i.e., $J_{\perp} \geq 0$. In order to derive quantitative values for $J_{\|}$ and $J_{\perp}$, the temperature-dependent Eu and Gd data were compared with the results of layer-resolved mean-field calculations [16,17], treating $J_{\|}$ and $J_{\perp}$ as variable parameters. An antiferromagnetic intralayer coupling, i.e., $J_{\|} < 0$, results in a frustration of the Eu spins in the hexagonal reconstructed Eu adlayer and hence in a noncollinear spin structure. The energetically most favorable spin configuration for $J_{\|} < 0$ and $J_{\perp} = 0$ is sketched in Fig. 4(a) for $T = 0$ K. Each Eu moment forms angles of 120° with

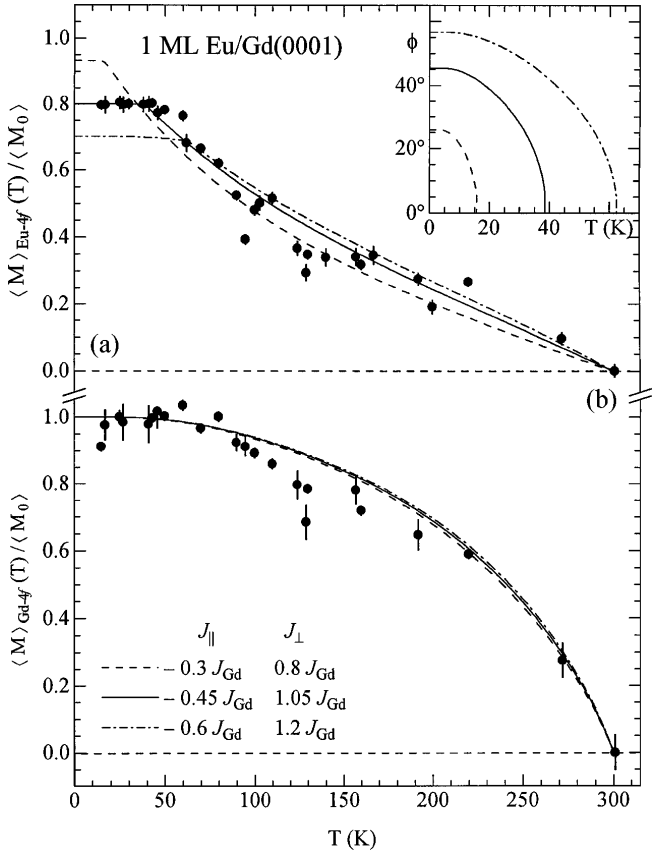

FIG. 2. Temperature dependences of (a) $\langle M \rangle_{\text{Eu-}4f}$ and (b) $\langle M \rangle_{\text{Gd-}4f}$ of 1-ML-Eu/Gd(0001), normalized to the 25-K point of the Gd $\langle M_0 \rangle$. The solid, dashed, and dash-dotted curves through the data points represent the results of a mean-field theory for three different sets of exchange-coupling constants. The inset in (a) gives the temperature dependence of the angle ϕ; for details see text.

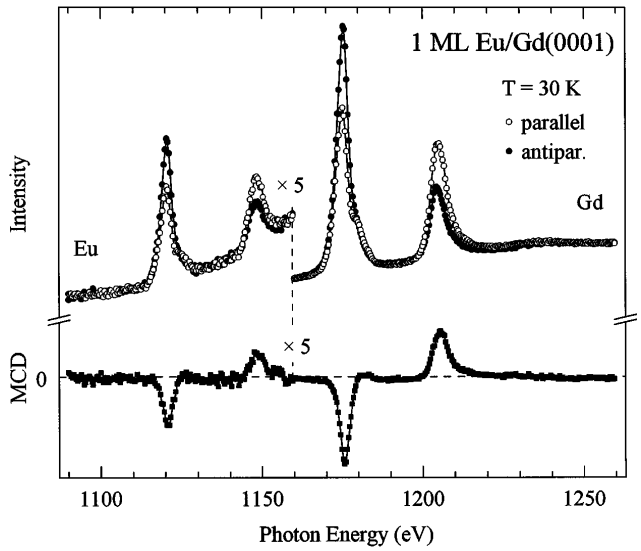

FIG. 3. XA spectra at the $M_{4,5}$ thresholds of 1-ML-Eu/Gd(0001) at 30 K taken with circularly polarized light incident at an angle of 15° with respect to the sample surface. Open (filled) symbols represent parallel (antiparallel) orientation of sample magnetization and photon spin. The filled squares represent the MCD spectrum $I_{\text{parallel}} - I_{\text{antipar}}$.

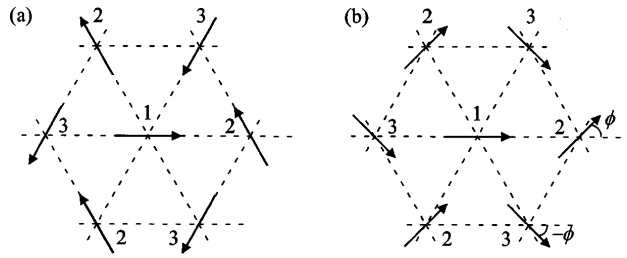

FIG. 4. Eu spin configuration for an antiferromagnetic Eu intralayer coupling, $J_{||}$: (a) Energetically most favorable spin configuration for an antiferromagnetic Eu intralayer coupling, $J_{||}$, and a vanishing Eu-Gd interlayer coupling, $J_{\perp}$. Each Eu moment forms 120° angles with the nearest-neighbor moments. (b) Eu spin configuration for $J_{||} < 0$ and $J_{\perp} > 0$.

the nearest-neighbor moments. For $J_{\perp} > 0$, the magnetic exchange coupling of the Eu spins with the Gd moments in adjacent interface layers leads to a more parallel alignment of the Eu spins, i.e., ϕ is reduced as shown in Fig. 4(b). To formally described this situation, the Eu monolayer is subdivided into three interpenetrating sublattices indicated by the numbers 1, 2, and 3 in Fig. 4. The spins of one sublattice were assumed to be oriented parallel to the magnetization of Gd for $J_{\perp} > 0$, while the Eu spins of the other two sublattices were allowed to rotate in the Eu plane, forming angles of ϕ and $-\phi$ with the Gd moments. For $J_{||} > 0$, a homogeneous magnetization of the Eu layer ($\phi = 0$) was obtained.

The temperature dependences of $\langle M \rangle_{\text{Eu-}4f}$ and $\langle M \rangle_{\text{Gd-}4f}$ were calculated for different values of $J_{||}$ in the range from $-0.6J_{\text{Gd}}$ to $0.6J_{\text{Gd}}$ and then optimized to reach the best agreement between experiment and calculation by varying $J_{\perp}$. J_{Gd} characterizes the exchange-coupling constant of bulk Gd, which is given by the mean-field relation $k_B T_C = Z J_{\text{Gd}}$, where k_B is the Boltzmann constant, $T_C = 294.3$ K is the Curie temperature of bulk Gd, and $Z = 12$ is the bulk coordination number of Gd metal. Figure 2 gives a comparison of the experimental and theoretical results for three ratios of $J_{||}/J_{\perp}$. The best agreement between experiment and theory was reached for $J_{||} = -0.45J_{\text{Gd}}$ and $J_{\perp} = 1.05J_{\text{Gd}}$. Significant poorer agreement was obtained when $J_{||} = -0.3J_{\text{Gd}}$ and $J_{\perp} = 0.8J_{\text{Gd}}$ or $J_{||} = -0.6J_{\text{Gd}}$ and $J_{\perp} = 1.2J_{\text{Gd}}$ were assumed. For $J_{||} = -0.45J_{\text{Gd}}$ and $J_{\perp} = 1.05J_{\text{Gd}}$, the mean-field calculation predicts a constant net Eu magnetization for temperatures up to 38 K and a noncollinear Eu spin structure, as described by the temperature dependence of the angle ϕ given in the inset of Fig. 2(a).

In summary, the magnetic properties of 1-ML-Eu/Gd(0001) were investigated in an element-specific way by MCD in $4f$ PE. The Eu adlayer was found to be magnetically ordered at 25 K, with a large net magnetization parallel to the magnetization of the Gd substrate. The component of the Eu magnetization parallel to the direction of the incoming light was found to be reduced by 10% to 30% with respect to that of the Gd substrate. Estimates for exchange-coupling constants were obtained by comparing the temperature-dependent experimental data with the results of a mean-field calculation. While $J_{\perp}$ was found to be comparable to J_{Gd} of bulk Gd metal, $J_{||}$ was found to be antiferromagnetic. The possible formation of a noncollinear spin structure in the Eu adlayer at temperatures below $\cong$40 K was discussed. The results of this work, in combination with surface and interface shifts of the $4f$ PE lines, open a way for an element-specific and layer-resolved study of the magnetism of interfaces. Work in this direction is being pursued in our laboratory.

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