Temperature-Induced Reversal of Magnetic Interlayer Exchange Coupling

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(Received 18 December 2007; published 6 June 2008)

For epitaxial trilayers of the magnetic rare-earth metals Gd and Tb, exchange coupled through a nonmagnetic Y spacer layer, element-specific hysteresis loops were recorded by the x-ray magneto-optical Kerr effect at the rare-earth M_5 thresholds. This allowed us to quantitatively determine the strength of interlayer exchange coupling (IEC). In addition to the expected oscillatory behavior as a function of spacer-layer thickness d_Y, a temperature-induced sign reversal of IEC was observed for constant d_Y, arising from magnetization-dependent electron reflectivities at the magnetic interfaces.

DOI: 10.1103/PhysRevLett.100.227203 PACS numbers: 75.70.Cn, 78.70.Ck, 78.20.Ls

Thin magnetic layers coupled by magnetic exchange interaction across a nonmagnetic spacer layer have been studied extensively ever since the first observations of antiferromagnetic (AFM) coupling in such systems [1–4]. Widespread applications came through the associated giant magnetoresistance effect (GMR) [3], which made such layered magnetic structures essential elements in advanced reading heads of magnetic storage devices. The interlayer exchange coupling (IEC) behind these effects was found to oscillate with the thickness of the nonmagnetic spacer layer [5,6]. This observation can be explained by the Rudermann-Kittel-Kasuya-Yosida (RKKY) model, in which the oscillation period of the IEC is given by the length of the extremal vectors that connect parallel sections of the Fermi surface of the spacer layer [7]. Dependences of the IEC on other characteristic parameters of the magnetic layers, such as thickness [8] and composition [9], as well as thickness of capping layers [10] have also been investigated.

In the past few years, several studies dealt with the temperature (T) dependence of magnetic coupling through metallic as well as insulating layers [11,12]. The standard RKKY theory of IEC, considering constant magnetization of the magnetic layers, leads to a rather weak T dependence of the coupling [13]. For metallic spacer layers, thermal broadening of the Fermi edge causes a minor weakening of the coupling strength. For insulating spacer layers, a similarly weak increase in coupling strength is expected due to thermal population of conduction-band states. Thermal excitation of spin waves [14] has also been proposed to explain some of the experimental observations [15]. In a few cases, sizable T dependences of IEC have been reported, e.g., for Co/Cu/Co [16], and unusual temperature behaviors have been observed for ferromagnets exchange coupled to antiferromagnets, like NiO/Cu/NiFe [17], or ferrimagnets, like GdFe/Gd [18]. However, no T-induced sign reversal of IEC between ferromagnetic (FM) layers has been reported so far.

In this Letter, we report on a strong T dependence of IEC in epitaxial Gd/Y/Tb trilayers on W(110) that even leads to a T-induced sign reversal of IEC for a given spacer-layer thickness. This novel effect is explained by magnetization-dependent electron reflectivities at the interfaces of Y with Gd and particularly with Tb. This causes strong temperature effects on the amplitude and on the *phase* of the oscillatory coupling strength as a function of spacer-layer thickness, leading to the observed sign reversals.

Heavy rare-earth metals are interesting magnetic materials due to their different magnetic properties despite similar crystalline and electronic structures. Gd and Tb (including Y) crystallize in the hexagonal close-packed structure with lattice parameters that differ by less than 2%. The spherical charge distribution of the $4f^7$ shell of Gd leads to a small magnetic anisotropy and hence to small coercive fields in epitaxial films of good crystalline quality. On the other hand, Tb has a large magnetic anisotropy due to its aspherical charge distribution caused by a large atomic orbital momentum ($L = 3$). It is thus expected that the magnetization of the softer Gd layer in the trilayer structures can be selectively reversed.

Epitaxial Gd/Y/Tb/W(110) trilayers were grown in ultrahigh vacuum on a W single-crystal substrate by metal-vapor deposition. Typical deposition rates were 1 to 4 monolayers (ML) per minute. The crystallinity of the layers was checked by low-energy electron diffraction. The thicknesses of the layers were $d_{Tb} = 10$ nm, $d_{Gd} = 3.5$ nm, with d_Y ranging from 0.3 to 3.3 nm. Trilayers with both constant d_Y and a wedge-shaped Y spacer layer were studied. The as-grown films were annealed at temperatures known from previous studies to result in smooth layers without significant interdiffusion [19]. Measurements of resonant soft x-ray reflectivity with circularly polarized x rays were performed *in situ* using the UE52 and UE56/1 beam lines at BESSY (Berlin). The specularly reflected intensity was detected by a Si photodiode mounted on a home-built $\theta - 2\theta$ goniometer inside the vacuum chamber. X-ray magneto-optical Kerr effect (XMOKE) hysteresis loops were recorded by sweeping an external magnetic field, produced by a rotatable magnet [20], along the substrate [110] direction that corresponds to the $[1\bar{1}00]$ easy axis of magnetization of epitaxial Tb/W(110) films [19].

The Gd/Y/Tb trilayers were cooled in an external magnetic field to ensure saturation of the magnetically hard Tb layer. Figure 1(a) shows typical reflectivity spectra of a remanently magnetized trilayer. The large magnetic contrast allows us to perform XMOKE measurements in an element-specific way by selecting the appropriate photon energy and by varying the applied magnetic field. Typical hysteresis loops are displayed in the inset of Fig. 1(a), reflecting the widely different coercivities of Gd and Tb layers due to the large difference in anisotropies. This renders it possible to reverse only the magnetization of the softer Gd layer by applying a magnetic field not strong enough to influence the magnetization of the Tb layer. In these trilayers, the exchange coupling between the two magnetic layers acts as an effective bias field (exchange bias) that needs to be overcome in order to reverse the magnetization of the softer layer. The resulting shift of the Gd hysteresis curve with respect to zero field is a measure of the coupling strength [21]. As an example, Fig. 1(b) displays Gd hysteresis loops taken at two different temperatures for $d_Y = 1.5$ nm: The data clearly show that the exchange bias changes sign from positive at 11 K to negative at 174 K.

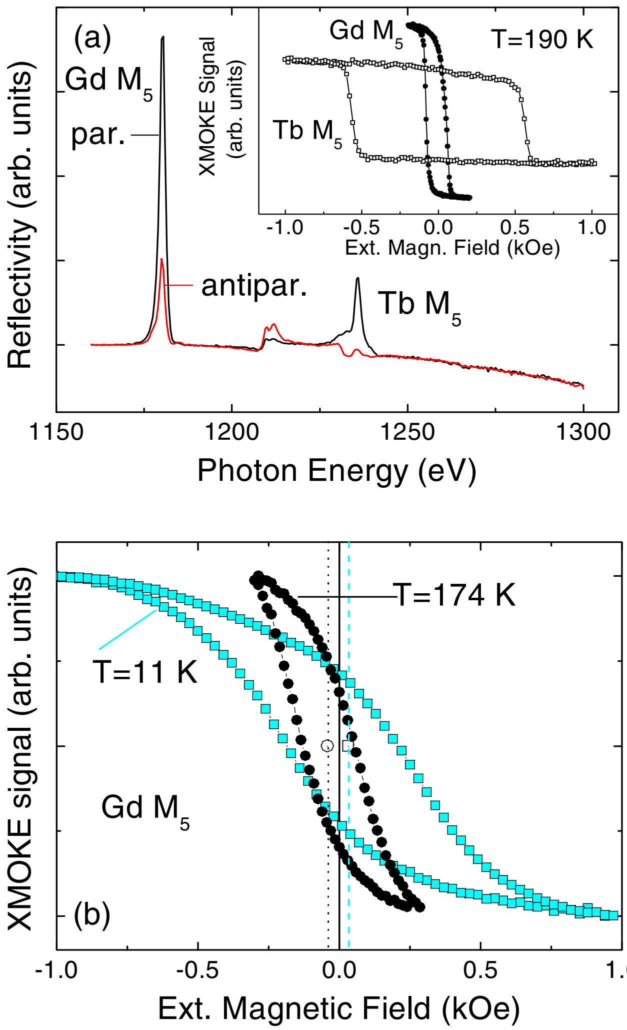

FIG. 1 (color online). (a) Soft x-ray reflectivity spectra in the region of the $M_{4,5}$ thresholds of Gd and Tb recorded from a Gd/Y/Tb/W(110) trilayer with $d_Y = 1.5$ nm at $T = 20$ K. The incidence direction of the circularly polarized x rays was nearly parallel and antiparallel to the in-plane sample magnetization. The inset shows element-specific hysteresis curves measured at photon energies corresponding to the Gd and Tb M_5 reflectivity maxima, respectively. (b) The Gd M_5 hysteresis curves, measured at two different temperatures, are shifted due to exchange bias. For clarity, the centers of mass are marked with an open dot (at $T = 174$ K) and an open square (at $T = 11$ K), showing negative and positive shifts, respectively.

This striking T dependence of the exchange bias was studied for $11\ \mathrm{K} \leq T \leq 220\ \mathrm{K}$ and for various d_Y. Figure 2 shows the T dependence of the exchange bias for two trilayers with fixed d_Y: For $d_Y = 1.5$ nm, the coupling changes from FM to AFM at ≈ 80 K, while for $d_Y = 2.2$ nm, the opposite sign change from AFM to FM is observed at ≈ 125 K. In both samples, the exchange bias vanishes at ≈ 220 K.

To investigate the sign change of the exchange bias more systematically, i.e., as a function of T and d_Y, trilayer structures with a wedge-shaped Y-spacer layer were studied. The results are summarized in Fig. 3, where the coupling strength J between the magnetic Tb and Gd layers is displayed as a function of d_Y for various T. Here, J is calculated as $J = H_b M_{\mathrm{Gd}} d_{\mathrm{Gd}}$, where the exchange bias H_b was determined from the shifts of the Gd hysteresis loops and d_Y was varied by measuring at different positions along the Y wedge (with an error $\Delta d_Y = \pm 0.1$ nm, due to the precision of $\Delta z = \pm 0.1$ mm in the vertical position of the sample in the focused x-ray beam). An oscillatory behavior is evident for all temperatures, and the oscillation amplitudes decrease with increasing d_Y; in addition, the coupling strength decreases with increasing T, as expected for metallic spacer layers [13]. The curves oscillate around $J = 0$, demonstrating the absence of a

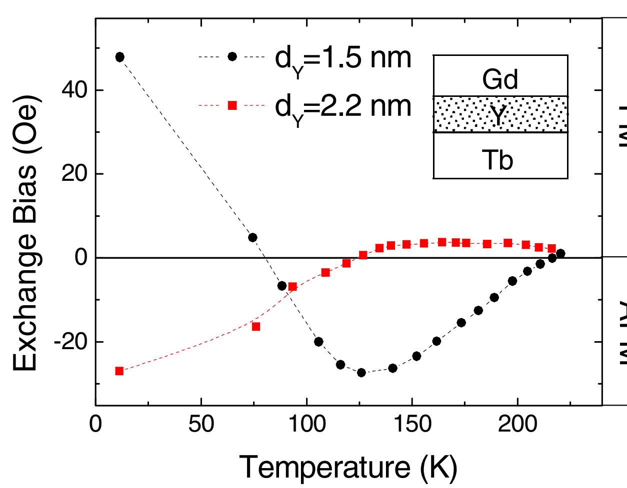

FIG. 2 (color online). Exchange bias as a function of temperature for Gd/Y/Tb/W(110) trilayers with two different d_Y. In both cases, the coupling changes sign with temperature.

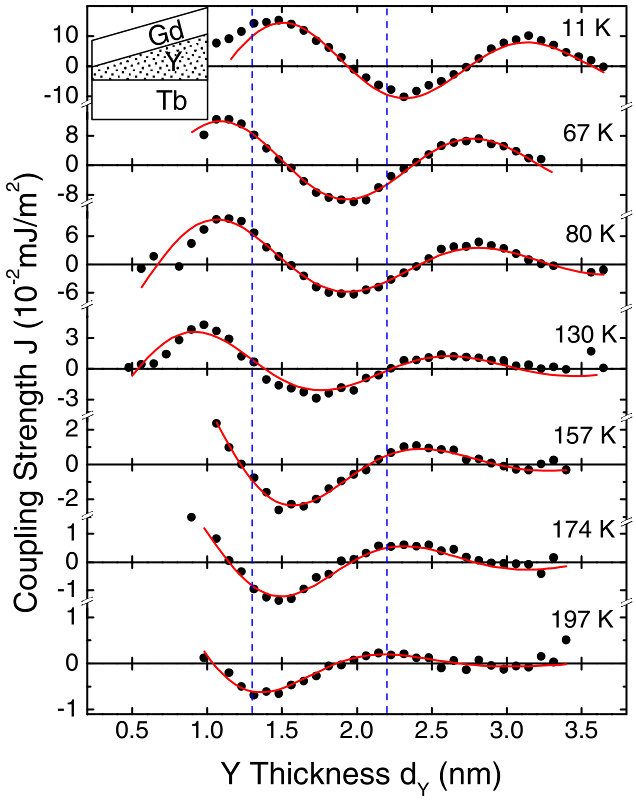

FIG. 3 (color online). Coupling strength J between Gd and Tb layers for a Gd/Y/Tb/W(110) trilayer with a wedge-shaped Y layer. Solid lines represent fits of Eq. (1) to the data. Note the scale changes on the ordinate for different T.

significant contribution of magnetostatic Néel coupling in our system [22].

The novel result, evident from Fig. 3, is contained in the significant, T-induced *phase change* of the oscillatory $J(d_Y)$ curves. This phase change causes the unusual T dependence of IEC, including the sign reversals with T for constant d_Y (see Fig. 2). Further data for the T dependence of the IEC for other values of d_Y can be obtained from Fig. 3.

The decay of the amplitude is predicted by RKKY theory [13] to follow a d_Y^{-2} law for large d_Y, in agreement with the present results for $d_Y > 1.5$ nm. To describe the data also in the range $d_Y < 1.5$ nm, we used an exponential dependence of the form $J \propto e^{-\beta d_Y}$. A fit of the data in Fig. 3 with the phenomenological expression [23]

$$J(d_Y) = Ae^{-\beta d_Y}\mathrm{Im}(e^{2\pi i d_Y/\lambda}e^{-i\phi}), \qquad (1)$$

where the amplitude A, the decay constant β, the period λ, and the phase ϕ are taken as T-dependent adjustable parameters, results in the relevant parameters λ and ϕ. The fit describes the experimental curves in Fig. 3 rather well, leading to an oscillation period of $\lambda = (1.5 \pm 0.1)$ nm, independent of T and in good agreement with the value of 1.6 nm determined from the length of the extremal vector in [0001] direction of Y metal that connects parallel sections of the Fermi surface, as obtained in a recent band-structure calculation [24].

We now extract the T dependences of the amplitudes and phases of the oscillations displayed in Fig. 3. As shown in Fig. 4, both the amplitude, $J_{\max} = A\exp(-\beta d_Y)$, plotted for $d_Y = 1.3$ and 2.2 nm (see dashed vertical lines in Fig. 3) and the phase ϕ reveal strong changes with T that cannot be explained by the standard RKKY theory [13] or the spin-wave model [14]. We therefore postulate that the magnetizations of the layers have an intrinsic influence on the coupling itself, as they are changing significantly in the studied T range, particularly for Tb metal (with a Curie temperature $T_C^{\mathrm{Tb}} = 220$ K as compared to $T_C^{\mathrm{Gd}} = 293$ K).

The origin of the indirect magnetic interlayer coupling through a metallic layer can be understood within the picture of multiple spin-dependent reflections of the valence electrons inside the quantum well formed by the magnetic-nonmagnetic interfaces [23]. The difference Δr of the complex reflection coefficients for electrons of opposite spins causes a polarization of the valence band of the spacer material that mediates magnetic coupling. The phase accumulation model leads to an expression for the coupling strength at $T = 0$ K, $J(d_Y, 0)$, which contains a factor $\Delta R_t e^{2iq_F d_Y}$ [23], with $\Delta R_t = |\Delta R_t|e^{i\phi} = \Delta r_{\mathrm{Tb}}\Delta r_{\mathrm{Gd}}$ (here, the contribution of a single Fermi wave vector q_F is considered). The reflection coefficient at a magnetic interface depends on the exchange splitting of the valence bands of the magnetic material. Assuming a linear dependence in first-order approximation and considering

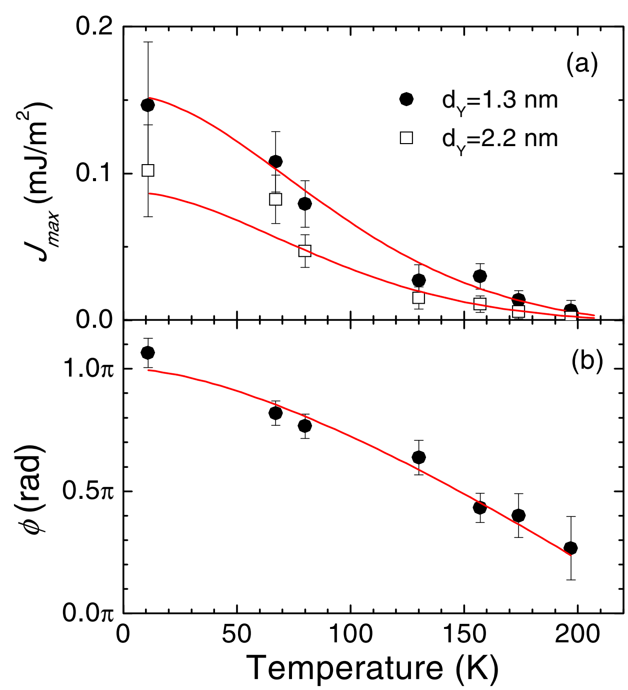

FIG. 4 (color online). T dependence of (a) $J_{\max}$ (for $d_Y = 1.3$ and 2.2 nm) and (b) phase ϕ for Gd/Y/Tb trilayers, as extracted from data in Fig. 3. For the fits, see text.

an exchange splitting proportional to the saturation magnetization M_S, as given by the Stoner model, we obtain

$$\Delta r(T) = [r^{\uparrow}(T) - r^{\downarrow}(T)] \propto M_S(T). \quad (2)$$

In this way, a T dependence is introduced in the model through the phases and amplitudes of the reflection coefficients. Considering contributions from both magnetic layers, the coupling strength J will be approximately proportional to an effective magnetization $M^*(T) = M_{\rm Tb}(T)M_{\rm Gd}(T)/[M_{\rm Tb}(0)M_{\rm Gd}(0)]$ [for $M_{\rm Tb}(T)$ and $M_{\rm Gd}(T)$, see Ref. [25]]. In addition, the T dependence arising from thermal broadening of the Fermi edge has to be taken into account, given by $F(cT) = cT/\sinh(cT)$ [13]. Here, $c = ad_Y + b$, where a is a bulk term and b an interface term. We obtain $a = 0.000\,18$ $(\text{Å K})^{-1}$ from the experimental oscillation period λ (following Ref. [13]). The interface term b is independent of d_Y [26,27] (see below). Including the described temperature effects on the spin-dependent electron reflection coefficients, we obtain

$$J(d_Y, T) \propto \exp(2iq_F d_Y + i\phi)F(cT)M^*(T). \quad (3)$$

The amplitude of coupling, $J_{\max}$, is well described by this expression, as shown in Fig. 4(a). A simultaneous fit of $J_{\max}(T) = J_{\max}(0)F(cT)M(T)$ to the data results in $J_{\max}(0) = (0.15 \pm 0.03)$ and (0.09 ± 0.03) mJ/m^2 for $d_Y = 1.3$ and 2.2 nm, respectively, and $b = (0.015 \pm 0.002)$ K^{-1}. These values are comparable to those found for the Co/Ru/Co and Co/Cu/Co systems [27].

The strong T dependence of the IEC reported in this work is caused by the T-dependent phase of the oscillations of $J(d_Y)$. It is thus relevant to study this phase in more detail. Figure 4(b) represents the T dependence of the phase ϕ determined by least-squares fits of the $J(d_Y, T)$ curves in Fig. 3. This strong T dependence of ϕ is caused by changes of the complex total reflectivity ΔR_t with T. In first-order approximation and similar to the case of $|\Delta R_t|$, we assume that ϕ depends linearly on the effective magnetization $M^*(T)$. Accordingly, $\phi(T) = \phi_0 + \alpha M^*(T)$ was fitted to the data in Fig. 4(b), leading to $\phi_0 = (0.03 \pm 0.08)\pi$ and $\alpha = (0.97 \pm 0.10)\pi$. The relation $\phi(T) = \pi \cdot M^*(T)$ is thus compatible with the experimental data within the error bars. It is tempting to interpret this result analogously to the reflection of traveling waves from free and fixed ends of a vibrating rope, with phase changes of 0 and π, respectively. The vanishing magnetic exchange splitting of the valence bands at T_C is intuitively expected to result in a reflection of the electron waves with zero phase change. While the value of ϕ close to π at $T = 0$ K is possibly accidental, its vanishing at the Curie temperature of Tb strongly supports our assumption that the variations in magnetizations of the magnetic layers cause the observed strong T dependence of IEC in the present case.

In summary, the observed T-induced sign reversal of magnetic interlayer exchange coupling is described by the thermal variations of the magnetizations of the two magnetic layers, which lead to strong effects on amplitudes and phases of the spin-dependent electron reflectivities at the interfaces. This new effect might find practical applications in temperature-sensitive devices through the GMR associated with a reversal of IEC. The working temperature range can be tuned by selecting suitable materials and layer thicknesses.

Work supported by the DFG Project No. KA 564/10-1. The authors thank H.-Ch. Mertins, K. Godehusen, and J. Nötzold for experimental support. J. E. P. acknowledges financial support by the A.-v.-Humboldt Foundation and by the MEC program “Ramón y Cajal.”

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