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Coexistence of glassy antiferromagnetism and giant magnetoresistance in Fe/Cr multilayer structures

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Abstract

Using temperature-dependent magnetoresistance and magnetization measurements on Fe/Cr multilayers that exhibit pronounced giant magnetoresistance (GMR), we have found evidence for the presence of a glassy antiferromagnetic phase. This phase reflects the influence of interlayer exchange coupling (IEC) at low temperature ($T < 140$ K) and is characterized by a field-independent glassy transition temperature, T_g, together with irreversible behavior having logarithmic time dependence below a "de Almeida and Thouless" critical field line. At room temperature, where the GMR effect is still robust, IEC plays only a minor role, and it is the random potential variations acting on the magnetic domains that are responsible for the antiparallel interlayer domain alignment.

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Given the established presence of giant magnetoresistance (GMR)-based devices in technology, especially in the multi-billion dollar computer hard disk drive market, it may come as a surprise that there is still an incomplete scientific understanding of the GMR effect [1]. The mechanism for GMR, first observed in single crystalline (1 0 0) Fe/Cr multilayers grown by molecular beam epitaxy [2–4] and subsequently in magnetron-sputtered polycrystalline films [5], relies on spin-dependent scattering [6] and the associated dependence of resistance on the relative orientations of the magnetizations in neighboring layers. It is important to recognize that interlayer exchange coupling (IEC) is not necessarily required for a GMR effect [1]. In a particularly simple manifestation, two neighboring films, separated by a non-magnetic spacer layer, could have different coercive fields, thus giving rise to antiparallel alignment and a GMR effect, as the external field is cycled [7]. Randomness [8,9] and competing interactions such as biquadratic coupling [10,11] can also play a significant role. In this paper we identify (for our films) a glassy antiferromagnetic (GAF) phase which, by marking the influence of IEC at low temperatures, implies that at higher temperatures

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random potential variations rather than IEC are responsible for antiparallel alignment.

Our Fe/Cr multilayer samples have been prepared on silicon substrates by ion beam sputter deposition of separate Fe and Cr targets. Extensive characterization of the deposited multilayers showed distinct compositional and structural modulations with well-defined interfaces and a surface roughness on the order of 5 Å. Ten and 30-layer stacks with the repeat sequence [Fe(20 Å)/Cr(d_{Cr})] are typically deposited and passivated with a 50 Å-thick Cr layer. The Cr spacer thickness d_{Cr} is varied over the range 8–12 Å. The inset of Fig. 1 shows typical GMR traces at 300 and 10 K for both current and magnetic field parallel to the planes of a [Fe(20 Å)/Cr(12 Å)] × 30 sample.

In Fig. 1 we show a selected subset of temperature-dependent field-cooled (FC, open symbols) and zero-field-cooled (ZFC, closed symbols) magnetization data for a 30-layer sample with $d_{Cr} = 12$ Å and a GMR ratio ($(R(0) - R(H))/R(0)$, Fig. 1 inset) of 20.6% at 10 K. The data were taken using a SQUID magnetometer in fields (indicated on the plot) oriented parallel to the layers. At each field the corresponding FC and ZFC curves can be characterized by three distinct temperatures: an irreversibility temperature $T_{irr}(H)$ denoting the bifurcation point below which there is hysteresis (upward arrows), a temperature $T_m(H)$ (downward arrows) denoting the maximum in each of the ZFC curves, and an inflection temperature T_{infl} (vertical dashed line) which marks the inflection point of each FC curve. Evidently T_{infl} is quite robust and independent of field, having a value $T_{infl} = 93.0 \pm 1.4$ K determined to relatively high precision from FC measurements at five different fields spanning the range 50–400 Oe.

Compelling evidence for an interlayer rather than intralayer effect is found in the resistance measurements of Fig. 2 on the same sample. For each datum on this graph, the sample was ZFC to the target temperature, the resistance $R(0)$ measured, and then a field applied to measure the change in resistance $\delta R = R(0) - R(H)$. The ratio $|\delta R/R(0)|$ is plotted against temperature for the fields indicated in the legend. The striking aspect of these data is that although the peaks are not as pronounced as those in the ZFC magnetizations of Fig. 1, their positions in an H–T plot of Fig. 3 (open triangles) show close similarity with respect to the positions of the ZFC peaks (solid circles).

The presence of a spin-glass-like phase is buttressed by our finding that $T_m(H)$ defines a

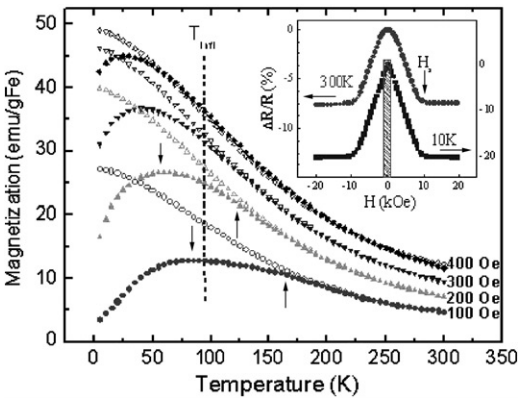

Fig. 1. Magnetization of a multilayer sample ([Fe(20 Å)/Cr(12 Å)] × 30) normalized to the weight of iron plotted as a function of temperature at the indicated fields. The data at each field are taken in pairs: the open(solid) symbols referring to the FC (ZFC) procedure. The vertical arrows and dashed line are described in the text. Inset, dependence of the GMR ratio on applied field for the same film at 300 K (left axis) and at 10 K (right axis).

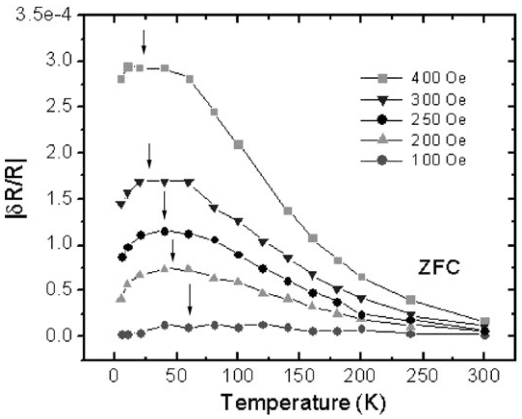

Fig. 2. Temperature dependence of the relative changes in resistance at the fields indicated in the legend for the same sample characterized in Fig. 1. For each data point, the sample was ZFC as described in the text. The vertical arrows indicate the positions of the maxima for each field and define a critical field dependence similar to that defined by the maxima of the ZFC magnetizations in Fig. 1.

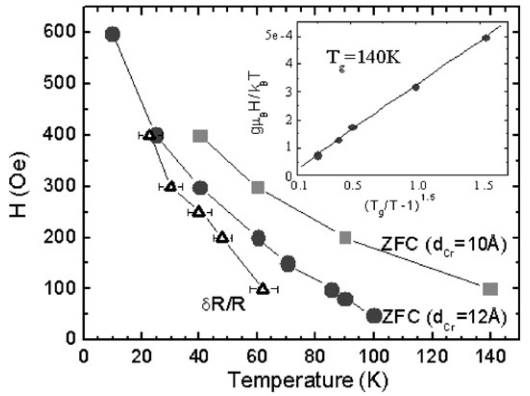

Fig. 3. Critical field lines for the 30 layer [Fe(20 Å)/Cr(12 Å)] (solid circles and open triangles) sample shown in Fig. 1 and for a second 30 layer [Fe(20 Å)/Cr(10 Å)] (solid squares) multilayer sample with smaller Cr spacer thickness. The solid symbols refer to determinations using the experimental $T_m(H)$'s of ZFC magnetizations and the open triangles are determined by similar peaks in the resistance measurements. Inset, plot of the high temperature points (solid circles) showing the AT scaling dependence for spin glasses.

critical field line (solid circles in Fig. 3) which delineates the onset of strongly irreversible behavior and has the de Almeida and Thouless (AT) form [12,13], $H/T \propto (T_g/T-1)^{3/2}$ (inset), where T_g is the spin glass temperature. In Heisenberg spin-glass systems with short-range interactions, the lower critical dimension is four [13] and robustness with respect to fluctuations occurs only in higher dimensions. Accordingly, our determination of AT scaling shown in Fig. 3 is dynamic since the ZFC curves occur on short enough time scales to see a spin-glass signature. In this paper, we choose the definition, $dM_{ZFC}/dT = 0$, used by Binder and Young ([13, pp. 908–909], Fig. 72b) to determine the AT line. Extrapolation of this dynamic line to zero field naturally involves the field-cooled measurement. This is understood by recognizing that for a canonical spin glass $M_{FC} \propto H/T$ for $T > T_g$ and $M_{FC} \propto H^{(1-u)}$ (the exponent $u = 0$ in mean field) for $T < T_g$. The field independent inflection point of $M_{FC}(T)$ (dashed line of Fig. 1) therefore gives a measure of T_g. Experimentally, we find $T_g = 1.51 \times T_{infl} = 140$ K as a best fit parameter to the linear dependence shown in the inset of Fig. 3. We do not choose $T_g = T_{irr}$, since such a choice pertains to "orthodox spin-glass systems" rather than the GAF phase considered here.

An additional and essential ingredient for a glassy phase is the presence of disorder measured by the variance, ΔJ, in the antiferromagnetic (AF) coupling strengths. This variance arises because of the existence of domains and the concomitant constraints imposed at the intralayer level. Within each Fe layer, the moments are subjected to the competing effects of ferromagnetic (FM) (short range) and dipolar (long range) forces, leading to a domain structure. The equilibrium pattern displays orientational randomness due to the pinning effects of local structural imperfections and of the Fe/Cr interfaces. In the following we consider that the total spin of a domain constitutes the elemental entity. These domains lie in the plane of the films with an orientation that remains constrained within the plane for interlayer coupling, J_{AF}, in a direction perpendicular to the layers. Thus, the exchange energy between two Fe 'moments' separated by a spacer layer is of the form $E = J_{AF}\cos(\Psi)$, where Ψ denotes their relative angle. The well-defined orientations imposed by the intralayer constraints will not be consistent, in general, with $\Psi = \pi$ (i.e. with a minimum value of E). Because of the long-range nature of dipolar interactions, lowering the exchange energy requires the overturning of one or of several clusters of Fe moments, which is energetically inhibited at low temperature. In this regime, Ψ behaves like a pseudo-random variable. A realistic estimate for ΔJ can be obtained by assuming a flat distribution for the values of Ψ on the $[0,\ 2\pi]$ interval, leading to $\Delta J = J_{AF}/\sqrt{2}$. At $T > T_g$, IEC is no longer effective and intralayer dipolar interactions dominate.

Many glassy systems, including the one discussed here, show AT like boundaries without being Ising spin glasses to which the theory [13,14] strictly applies. The GAF phase associated with our GMR multilayers is clearly not an Ising system and is more reasonably described in terms of an anisotropic vector model in which the elemental spins, belonging to magnetized domains, are coupled antiferromagnetically in the direction perpendicular to the layers. For such vector glass systems there is an additional degree of freedom in

the order parameter and the true phase boundary is delineated at higher temperatures and fields by a Gabay–Toulouse like (GT) boundary [15]. A more comprehensive viewpoint that facilitates understanding of our experiment can be gleaned from the schematic phase diagram, shown in Fig. 4 for the H–T plane at $J_{AF}/\Delta J > 1$. (Note that the PM phase is not labeled as a FM phase, since in the presence of a field there is no spontaneous symmetry breaking as the temperature is reduced through the Curie temperature.) In simplified terms the GT line (solid) can be thought of as denoting the onset of a phase transition to glassy behavior and the AT line (dotted) as the onset of pronounced irreversibility. The validity of the above description is not restricted to canonical spin glasses and it extends beyond the mean field level. Indeed, for experimental glassy systems the AT and GT boundaries are expected to show up in dynamic measurements [13]. Thus the region sandwiched between the AT and GT lines of Fig. 4 (or equivalently, the approximate region between the peak temperatures T_m and the bifurcation temperatures shown as upward arrows in Fig. 1) manifests weak hysteretic behavior, reflecting irreversibilities due, for example, to

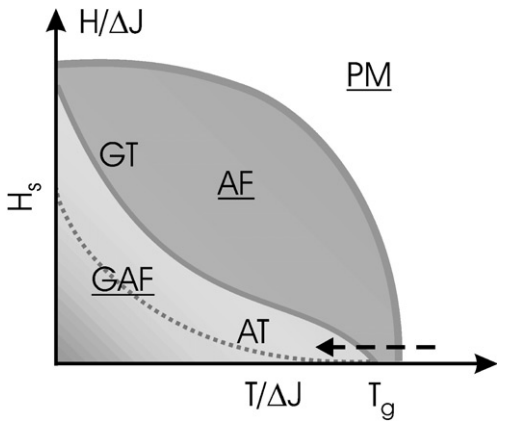

Fig. 4. Schematic of phase diagram in the H–T plane showing the relationship between the GAF, the AF and the paramagnetic (PM) phases. The axes are normalized as discussed in the text. The GT and AT line (dashed) are described in the text. For our samples the disorder is sufficiently large (i.e., $\Delta J \simeq J_{AF}$) and the field sufficiently low to ensure that the presence of an AF phase is obscured on the transition from the PM to GAF phase (horizontal dashed arrow).

pinning of domain walls by local structural imperfections; whereas strong hysteresis sets is below the AT line. The experimental signature of the GT line, which has not been measured here, is a divergence in the transverse AC susceptibility. At $H = 0$ both the AT and GT lines terminate at $T = T_g$, a fixed point which we have associated with our experimentally determined field-independent temperature, T_{infl}.

The following three consequences, confirmed by experiment, are immediately apparent: Firstly, since $T_g \propto J_{AF}$ and $\Delta J \simeq J_{AF}$, it is clear that as T_g increases, the boundary of the GAF phase moves out to higher temperatures and fields. Experimentally this is confirmed in Fig. 3 where the AT line for the sample with $d_{Cr} = 10$ Å (solid squares) has higher critical fields and a correspondingly higher T_g than the sample with 12 Å spacer. A second consequence is that the disorder-induced close proximity of T_g and J_{AF} implies that at low H the presence of an AF phase is obscured on the transition (Fig. 4, horizontal dashed arrow) from the PM to GAF phase. If this were not the case, then the FC DC susceptibility would have a maximum at the AF boundary and then saturate at a smaller value as $T \to 0$. Such maxima are not observed! A third consequence supporting the existence of a GAF phase comes from the scaling of the FC magnetization with H. FC magnetizations including those shown in Fig. 1 reveal that $M_{FC}/H \sim H^{-u}$ as $T \to 0$. Here we find $u = 0.58(2)$ for 5 K magnetization data taken at seven different fields ranging from 100 to 800 Oe, thus confirming behavior characteristic of spin-glass systems below the lower critical dimension [13]. This scaling is an indication that the true thermodynamic T_g is equal to zero. Finally, in addition to hysteresis, we also observe slow relaxations in the magnetization and resistance that are logarithmic in time and which can be explained by invoking constraints on the dynamics imposed by a hierarchy of domain sizes [16,17].

To fully appreciate the role of randomness in multilayers, it is important to recognize the difference between GMR multilayers, in which there is a strong interaction between closely coupled interfaces, and bilayer or trilayer configurations in which such interactions can be ignored

since there are at most only two interfaces. Thus for example, in studies of exchange bias in single FM/AM (Co/CoO) bilayers [9], the onset of exchange bias, which is induced by random interactions [8], is observed to occur at a single temperature, the Neel temperature. By contrast, in our case there are two temperatures: $T_g = 140$ K for glassiness and the Neel temperature (310 K) for bulk Cr where there is a loss of antiferromagnetism but disorder is still important. (In agreement with Barthèlèmy et al. [4], we find that the GMR is not affected by the antiferromagnetism of bulk chromium.) Accordingly, the picture described for FM/AF bilayers [9] is different for closely coupled multilayers where interactions between multiple FM layers and interactions between interfaces should be taken into account. Similar considerations also apply to the magneto-optic Kerr effect (MOKE) and scanning electron microscopy with polarization analysis (SEMPA) studies [18] on Fe/Cr/Fe trilayers and magnetization and FM resonance studies of CoFe/Mn/CoFe trilayers [11], all of which specialize to a specific type of spacer layer and do not include the multilayer interactions responsible for our GAF behavior. Our results are thus complementary yet distinct from the results of bilayer/trilayer experiments.

A consideration of the relevant energy scales and the mutual interactions of the magnetized domains in the Fe layers solidifies this emerging picture of spin-glass-like behavior in GMR multilayers. If adjacent Fe layers of thickness t and saturation magnetization M_s are coupled through an AF exchange J per unit area, then saturation at a field $H = H_s$ occurs when $J = HM_st/4$, a relation found by equating the field energy per unit area, HM_st, to the energy difference, $4J$, between the aligned and antialigned magnetic configurations . We note that a glass temperature near 140 K corresponds to an AF coupling energy $\simeq 10$ meV, in good agreement with theoretical calculations [19,20] for Fe/Cr layers. In the calculation by Fishman and Shi [19] the Fe layers are exchange coupled below the Neel temperature T_n of the Cr spacer and a very strong AF coupling between the Fe and Cr moments at the interface is assumed. In the second calculation by Majumdar et al. [20] magnetoresistance data is well described by a theoretical expression in which RKKY interactions give a best fit AF coupling strength of (70 ± 20) K.

For $T > T_g$, the Fe layers are no longer AF coupled and the expression $J = HM_st/4$ to calculate the IEC is no longer relevant. In its place we use the expression [21,22] $H_s = 4\pi M_s$, to calculate the maximum saturation field necessary to align dipolar-coupled domains within each layer. This expression is valid for both perpendicular and parallel fields [22]. The saturation fields of 10–20 kOe in our samples (Fig. 1 inset) and similar samples reported by others [2,5] are the right order of magnitude for Fe which with a saturation magnetization $M_s = 1700\,\mathrm{Oe/cm^3}$ implies a maximum saturation field $H_s = 4\pi M_s = 21$ kOe. For our three different samples with $d_{Cr} = 8$, 10 and 12 Å we find a linear dependence of H_s on d_{Cr} which extrapolates to the origin ($d_{Cr} = 0$) to a value within 5% of $H_s = 21$ kOe, thus validating our use of this analysis.

At this point we reemphasize that the AF coupling strength is set by the approximate equalities, $T_g \simeq \Delta J \simeq J_{AF}$. Accordingly, randomness effects are dominant enough to ensure that even in the absence of a magnetic field the AF phase is suppressed and only the glass phase exists [13]. The width of the AF slice in Fig. 4 should therefore always be small. The collective interlayer effects appear only for temperatures and fields below the AT line. Accordingly, the natural saturation field for glassy behavior is determined by the AT line. At higher temperatures and fields, intralayer dipolar domain physics dominates. In this region the true saturation field, $H_s = 4\pi M_s$, is a single-layer property determined by dipolar physics.

To associate field scales with energy (or equivalently, temperature), we use the conversion ratio, $2.2\,\mu_B B/k_B T = 1.5\,\mathrm{T/K}$, where the magnetic moment of Fe is $2.2\,\mu_B$. Accordingly, the dipolar interaction strengths measured by H_s, which are balanced by domain wall energies, are on the order of a few Kelvin and hence not strong enough at $T > T_g$ to determine domain orientation. Rather, domain orientation at $T > T_g$ is determined by the much stronger potential variations associated with crystalline anisotropies and the presence of

impurities and defects. The presence of a GAF phase implies that IEC is effective in creating an anti-alignment effect beneficial to a large GMR effect only at low temperatures ($T < T_g$) and low fields ($H < H_{AT}$). The shaded region in the inset of Fig. 1 illustrates just how narrow this region is.

In summary, we show that a heretofore-unrecognized GAF state coexists with GMR in polycrystalline Fe/Cr multilayer stacks. The very presence of this glassy phase sets an energy scale ($T_g = 140$ K) for antiferromagnetic IEC that is well below room temperature. We therefore conclude that, for temperatures greater than T_g, IEC plays only a minor role in forcing the antiparallel interlayer domain orientations that give rise to the ($H = 0$) high resistance state of multilayer Fe/Cr GMR samples. Rather, random potential variations, which constrain domain orientation, must be taken into account to understand GMR in multilayer GMR devices. Since it is relatively easy to sweep a magnetization curve out to the saturation field, H_s, of a few Tesla and miss the small amount of hysteresis bounded by the AT line, we cannot assert that the glassy effects reported here do not apply to multilayers grown by molecular beam epitaxy. If glassy behavior is in fact absent from such multilayers, then smoother interfaces and larger spacer thicknesses may be the cause. The origin of the dependence of H_s on spacer thickness in multilayers as observed here and by others [2,5] as well as the origin of the AF couplings for $T < T_g$ are totally open questions. This contrasts with the bilayer and trilayer cases [8,9,18] for which the AF couplings have a clear source.

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