Magnetic first-order phase transition and crossover associated with random anisotropy in crystalline $Dy_xY_{1-x}Al_2$

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The low-temperature ($T=0$ K) first-order phase transition, predicted to drive systems with both weak random and uniform cubic anisotropy from a correlated spin glass to a ferromagnet, has been observed. At higher temperatures the transition is to a quasi- or random ferromagnet. The transition occurs at a concentration $x_t=0.62\pm0.01$ in the *crystalline* Laves-phase compounds $Dy_xY_{1-x}Al_2$. At concentrations above x_t, a line of transition is observed between the paramagnetic and random ferromagnetic phases, as found previously. This line is characterized by the crossover exponent $\phi_\Delta=0.80\pm0.08$, in good agreement with prediction. Two multicritical points have been identified at $x_t=0.62\pm0.01$, $T_t=29.5\pm0.1$ K and at around $x_t'=0.87$, $T_t'=45.4$ K.

It has been argued extensively that random magnetic anisotropy (RMA) tends to destroy long-range ferromagnetic order in real, three-dimensional materials.[1] The equation of state for such systems has been deduced from renormalization-group techniques and scaling arguments by Aharony and Pytte,[2] and via the phenomenological approach of Chudnovsky and co-workers.[3] Scaling arguments suggested that, at temperatures below the ordering temperature of the homogeneous system, *spontaneous* magnetization appears abruptly only as the strength D of the random interaction vanishes. Similarly, the phenomenological model predicts exponential decay of ferromagnetic correlations for all nonzero RMA. A refinement of the scaling approach considers the stabilizing effect of a uniform, or coherent, cubic anisotropy.[2,4] The first-order low-temperature transition persists but now occurs at a finite values of the RMA strength. The condition for such transition is [4] $B_4/J_0 \approx \Delta^{\theta_{4\Delta}}$, where $\Delta=(D/J_0)$,[2] D is the RMA axial crystal-field parameter, J_0 is the ferromagnetic exchange constant, B_4 is the strength of the cubic crystal field, and $\theta_{4\Delta}$ is a crossover exponent varying between 2.0 and 2.15 from 0 K to the transition temperature T_c.[4]

In this paper we demonstrate that cubic anisotropy indeed stabilizes the low-T spontaneous magnetization for sufficiently weak RMA but that, as the RMA strengthens, it gives way abruptly to a spin-glass-like phase (correlated-spin-glass (CSG), in the notation of Chudnovdsky and co-workers[3]) with long-range ferromagnetic correlations. We have chosen as a model system, the diluted, crystalline intermetallic compounds $Dy_xY_{1-x}Al_2$. The parent compound, $DyAl_2$, is a well studied Heisenberg-like ferromagnet with a Curie temperature $T_c=61.5\pm0.1$ K. The easy magnetization direction is along $\langle 100\rangle$.[5] Dilution with Y adds randomness to the cubic anisotropy, and strongly modifies the magnetic behavior.[6] In Fig. 1, we show the phase diagram (solid circles) deduced from the combined data of low-frequency ac susceptibility measurements and low-field magnetometer studies,[7] and where we should point out the presence of a new magnetic phase, labelled as a quasi- or random ferromagnet (RFM). The features of this phase will be discussed later on. The nature of the phase transition along the PM-CSG and PM-RFM line was treated previously in some detail.[7,8] Here, we focus on the crossover exponent that determines the position of

the PM-RFM line and on the observation of the first-order low-temperature ($T=0$ K) boundary between ferromagnetic and correlated-spin-glass phases. In what follows we will combine the phenomenological model of Chudnovdsky and co-workers[3] with a spin-wave approach, developed by del Moral and Cullen,[9] to analyze the results.

Our first goal is to locate the CSG-RFM phase boundary. For a truly ferromagnetic system, the low-field magnetization does not exceed H_a/N, where H_a is the applied field and N is the demagnetization factor of the sample. As the temperature is reduced through T_c, the magnetization increases to this value and then remains constant, giving rise to a typical "kink point." Measurements were performed by cooling samples for which $0.35 \leq x \leq 1$ in fields between 1 and 15 Oe. Samples with $x=1$ and 0.914 exhibited a clear kink point at T_c, reaching there the demagnetizing field limited magnetization. Samples with $x=0.828$, 0.660, and 0.625, while showing a knee at T_c, reach the demagnetization-limited value only in the limit $T=0$ K (see Fig. 2). As we showed previously,[7,8] the transition in these samples is that expected for a random anisotropy ferromagnet. For lower concentrations ($x=0.513$, 0.468, 0.417, and 0.370) both the kink anomaly and an extrapolation to the demagnetization limit were absent.[8]

To search for further evidence for long-range order, neutron-scattering measurements were performed on the

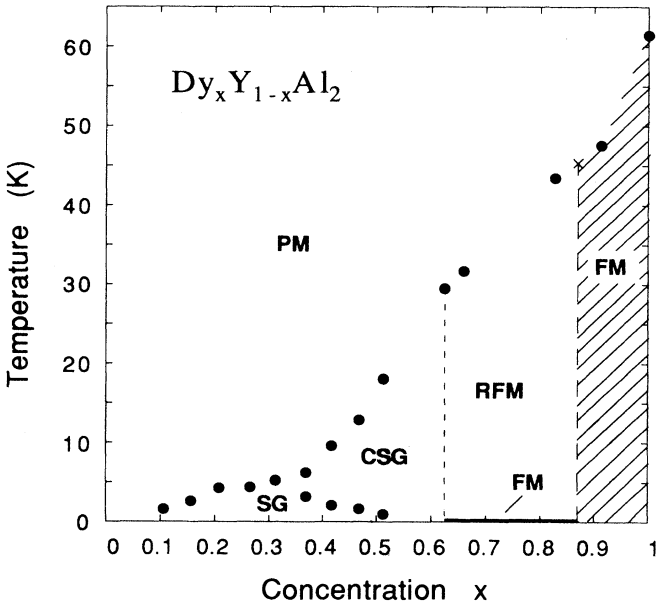

FIG. 1. Magnetic phase diagram for the *crystalline* cubic Laves-phase $Dy_xY_{1-x}Al_2$ series. PM, SG, CSG, RFM, and FM are, respectively, the paramagnetic, spin-glass, correlated-spin-glass, random-ferromagnetic, and ferromagnetic phases. The dashed vertical phase-boundary line is only schematic and ends up in a multicritical point (●). The horizontal dark segment signals the concentration interval where only at 0 K a spontaneous magnetization develops. The hatched region signals the pure ferromagnetic (FM) phase, and the cross (×) indicates the approximate position of another multicritical point.

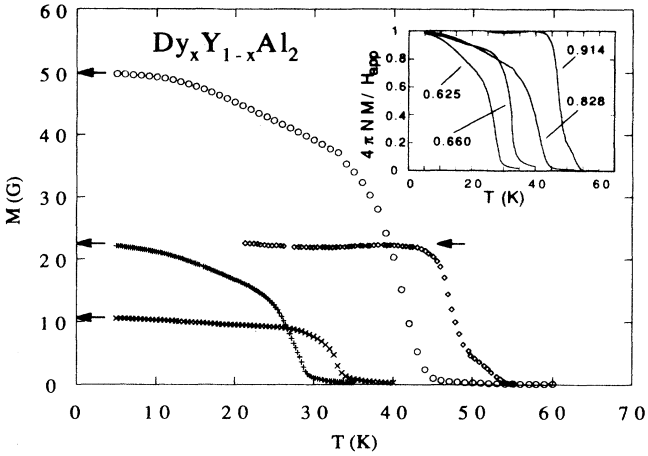

FIG. 2. Magnetization isofields for Dy concentrations $0.625 < x < 0.914$. (◇), $x=0.914$; (○), $x=0.828$; (×), $x=0.660$; (+), $x=0.625$. The applied magnetic field was 13.0 Oe for all the concentrations, except for the $x=0.914$ sample where $H_{app}=8.67$ Oe. The arrows signal the demagnetizing factor limited magnetizations. Inset: isofields of the low-field magnetizations normalized by the demagnetizing field limited values, $H_{app}/4\pi N$.

D1b instrument at the Institute Laue-Langevin (Grenoble). Remarkably, ferromagneticlike Bragg intensity was observed to arise in all samples for which $x \geq 0.4$. Further, a comparison of the width of the (111) powder line above and below T_c showed no change; both were resolution limited. This places a lower limit of $\xi \cong 1500$ Å on the ferromagnetic correlation length. To the contrary, small-angle neutron-scattering experiments pose an upper limit of $\xi \cong 700$ Å at the CSG regime, for the $x=0.35$ compound.[10] However, as pointed out by Chudnovsky and co-workers,[3] the ferromagnetic correlation length could be much larger than the distance R_a over which the anisotropy direction changes. Explicitly, it has been shown by del Moral and Cullen[9] that for weak RMA magnets, the transverse correlation length measured in neutron scattering is given by

$$\xi_\perp \cong \frac{\sqrt{15}}{2}\left[\frac{R_a/a}{\Delta}\right]^{1/2} a \ , \tag{1}$$

where a is the lattice constant. According to this model,[9] $\xi_\perp=[A/(\delta+g\mu_B H_a)]^{1/2}$, where the cubic coherent anisotropy has been neglected.[7] g is the Landé factor, A is the spin-wave (sw) stiffness constant, i.e., $A=(\frac{1}{2})J_0Sa^2$, and δ is the $\underline{k}=0$ wave-vector magnon gap, given by $\delta=(\frac{2}{15})(D^2/J_0)(R_a/a)^{-1}$. Formally this value results from a renormalization of the magnon energy which takes into account sw scattering effects and that, after a perturbation calculation up to order D^2 for the magnon energy, gives $\varepsilon(\underline{k})=Ak^2+g\mu_B H_a+\delta$. Substitution of δ expression into $\xi_\perp$ expression gives Eq. (1) for zero applied magnetic field. Physically, the expression obtained for δ can be easily understood by the following argument. For infinite J_0 all spins are aligned along the molecular field regardless the strength of D, but for J_0 finite, a sim-

ple random-walk calculation shows that the rms spin deviation angle towards the local easy axes directions, over a distance R_a, amounts $\sqrt{\langle\theta^2\rangle}=D/J_0$. This deviation is shared by all the spins along R_a and therefore the energy gain per spin due to the rotation amounts $(D^2/J_0)(R_a/a)^{-1}$.

Going back, again, to the neutron-diffraction results, while the correlation length remains large, the ferromagnetic contribution to the Bragg *intensity* suggests a loss of long-range order. Figure 3 shows the magnetic moment μ_{Dy}/μ_B, determined from the 2 K neutron data. The decrease below $x\cong 0.6$ is a clear indication that the moments are not fully aligned within the coherence volume, i.e., within the above mentioned CSG state.

A second feature derived from the neutron-diffraction data also indicates a collapse of long-range order near $x=0.6$. It was observed that for $x>0.62$, a more or less abrupt contraction of the lattice occurs below the ordering temperature $T_c(x)$, as shown in Fig. 4 for $x=0.828$. Instead, the contraction Δa_{mag} completely disappears for $x\leq 0.62$ as shown in Fig. 3; note that the variation of $\Delta a_{mag}(x)$ is quite steep near $x=0.625$. The origin of such distortions is likely magnetostrictive, inasmuch as the value of $[\Delta a_{mag}/a(2\text{ K})]\cong -0.3\times 10^{-3}$ for $x=1$, compares roughly with the measured cubic spontaneous magnetostriction, $\lambda[001]=-1.1\times 10^{-3}$, in a single crystal of $DyAl_2$.[11] Also below $x=0.62$, the thermal expansion between $T_c(x)$ and room temperature is abruptly reduced, giving way to an invarlike behavior below $x\cong 0.40$;[10] the reason for such behavior remains unclear.

All the above pieces of experimental evidence point to the following scenario for the magnetic phase diagram displayed in Fig. 1. At temperature $T=0$ K, we have a

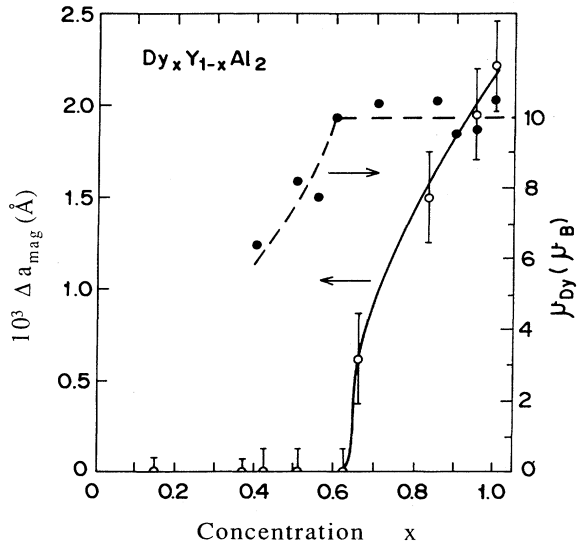

FIG. 3. The variation with concentration x of the Dy^{+3} ordered magnetic moment, μ_{Dy} (in Bohr magnetons), at $T=2$ K, as determined form the Bragg neutron-diffraction lines magnetic intensities, for the $Dy_xY_{1-x}Al_2$ compounds (solid circles). The magnetostrictive distortion, between 2 K and $T_c(x)$, as function of the Dy concentration x. The error bars are indicated. The continuous and dashed lines are guides to the eye.

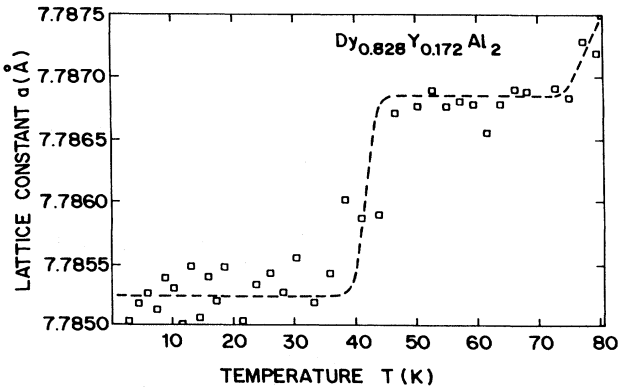

FIG. 4. The temperature variation of the lattice parameter a for the $Dy_{0.828}Y_{0.172}Al_2$ compound.

first-order magnetic phase transition at $x=0.62$ from a CSG phase to a ferromagnetic (FM) phase, i.e., with nonnull spontaneous magnetization, such FM phase remaining so up to $x=1$. Notice that above $x=0.62$ the measured magnetic moment at 2 K reaches its full value of $10\mu_B/Dy^{3+}$. For $0.625\leq x<0.914$, although there exists a contribution to the magnetic neutron scattered intensity and a spontaneous magnetostriction appears at $T_c(x)$, the lack of a demagnetizing limited magnetization points out to only a quasi- or random ferromagnet (RFM). Moreover, the merging of the PM-CSG and PM-RFM lines of transitions[7,8] with the CSG-RFM first-order transition line points out to a multicritical point at $x_t=0.62\pm 0.01$ and $T_t=29.5\pm 0.1$ K (see Fig. 1). On the other hand, at $x\geq 0.914$ the magnetization reaches the demagnetizing limited value for temperatures $T\leq T_c$, pointing out to a true ferromagnetic phase. Therefore a magnetic first-order transition line should also exist for some concentration within the interval $0.828<x_t'<0.914$, with a multicritical point at x_t' and at a temperature T_t', $43.4<T_t'<47.5$ K.

To proceed, it is important to determine Δ. For RMA systems, Chudnovsky and co-workers,[3] and del Moral and Cullen,[9] showed that the magnetization approaches its absolute saturation value M_s according to

$$\frac{M_s-M(T,H)}{M_s}=\frac{1}{15}\Delta\left[\frac{R_a}{a}\right]^3\left[\frac{\lambda M_0}{H+H_c}\right]^{1/2}+\chi_p H\ , \tag{2}$$

where H is the internal applied field, H_c the coherent cubic anisotropy field, χ_p, the residual paramagnetic susceptibility, M_0, the saturation magnetization at temperature T (Refs. 8 and 12), and λ, the mean-field constant, related to T_c (e.g., $\lambda\cong 116.8\ T_c$, adimensional, for $x=0.828$).[7,8,12] Previously, we exploited this relation to determine $\Delta(R_a/a)^3$; a plot of the values for the full range of samples is shown in the inset of Fig. 4. If we assume that the increase is primarily due to the change of Δ with concentration, and not in R_a as we shall justify below, we note that the concentration axis of Fig. 1 can, to a good approximation, be considered linear in the parameter Δ, and thus resembles the phase diagram de-

duced by Goldschmidt and Aharony[4] for nonzero cubic anisotropy. However, the phase diagram displayed by the present series of compounds (see Fig. 1) is far more complex than the predicted one.[4] Moreover, the knowledge of $\Delta(R_a/a)^3$, the lower bound of $\xi_\perp$ and the use of (1) allow us to estimate Δ and R_a separately. For instance, for $x=0.828$ and at $T=5$ K and $H=5$ T, we can take $\xi_\perp \cong 1500$ Å, $M_0=963$ G, $\lambda=5.07\times10^3$, $H_c=11.0$ kG, and $\chi_p=0.83\times10^{-4}$ G/Oe, obtaining $R_a \approx 10\,a$ (≈ 78 Å) and $\Delta \approx 0.05$, this value being in good agreement with the theoretical estimates.[7]

Finally, we consider whether the shape of the PM-RFM phase boundary is consistent with our RMA interpretation. According to the scaling analysis of Aharony and Pytte[2] and of Goldschmidt and Aharony,[4] such magnetic properties as the susceptibility ought to have the extended scaling form,

$$\chi(t,H,\Delta)=|t|^{-\gamma} f\left[\frac{H}{|t|^{\phi}},\frac{\Delta}{|t|^{\phi_\Delta}}\right], \tag{3}$$

where γ and ϕ are ferromagnetic exponents, and ϕ_Δ is the RMA crossover exponent. In zero field, the critical line is clearly determined by

$$\Delta \sim |t_s|^{\phi_\Delta}. \tag{4}$$

The reduced shift in transition temperature $t_s=[T_{co}-T_c(\Delta(x))]/T_{co}$ is plotted vs $\Delta(R_a/a)^3$ on a log-log plot in Fig. 5. The data for $x>0.62$ are well described by a line corresponding to $\phi_\Delta=0.80\pm0.08$. This result justifies the above made assumption of a R_a practically independent of x in the range $0.6 \lesssim x \lesssim 0.9$. In fact, taking into account Eq. (4), the observed log-log linear variation of Fig. 5 would only hold if $(R_a/a)^3$ also scales with some power of $t(x)$, say t^{ϕ_a}. But this eventual dependence has not any physical meaning and ought to be discarded. On the other hand, a direct determination of R_a,[13] is at the present questionable.

We have calculated ϕ_Δ from a combination of $\epsilon-$ expansion and $1/N$ expansion results to obtain[4,14]

$$\phi_\Delta=\frac{(4-d)}{(d-2)}\left[1-8\frac{(5-d)}{4d}\frac{S_{4-\varepsilon}}{N}+\cdots\right], \tag{5}$$

where $S_{4-\varepsilon}=\varepsilon/2-\varepsilon^2/4+O(\varepsilon^3)$. For $d=N=3$, we obtain $\phi_\Delta=0.88$, within experimental uncertainty the same as obtained above. However, the estimated value, using the scaling law $\phi_\Delta=\beta_\Delta+\gamma_0$, where γ_0 is a ferromagnetic exponent, for the amorphous strong-RMA alloy DyNi (Ref. 15), is much higher, $\phi_\Delta \simeq 2.4$.

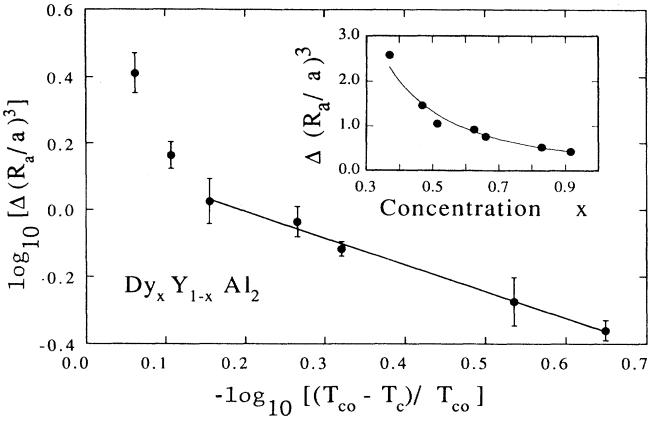

FIG. 5. Double log scaling of the random anisotropy field $\Delta=(D/J_0)^2$ modulo $(R_a/a)^3$ vs the reduced shift in transition temperature $[T_{co}-T_c(x)]/T_{co}$ (see text for full meanings); the slope of the line is the crossover exponent, $\phi_\Delta=0.80\pm0.08$, associated with Δ. In the inset is shown the actual concentration x dependence of $\Delta(R_a/a)^3$, for the $Dy_xY_{1-x}Al_2$ crystalline compounds.

We conclude that further evidence from neutron scattering and the approach to saturation supports our assertion that the $Dy_xY_{1-x}Al_2$ Laves-phase system is an example of a weak RMA system, with ferromagnetism partially stabilized by uniform cubic anisotropy. The neutron-scattering data confirm that the ferromagnetic coherence length is long in the correlated-spin-glass phase but that the moments are not completely aligned, even at 2 K. The substantial magnetostriction effect observed in the more concentrated samples, its disappearance above T_c and its absence for $x<0.62$, all support the notion that increasing randomness results in a first-order collapse of the $T=0$ spontaneous magnetization at a critical value of the RMA strength.

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