Remanent Magnetization of a Simple Ferromagnet

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We have measured the remanent magnetization of single-crystal EuS from 10^{-4} to 10^4 sec after removing an applied field. Using a recent model for magnon relaxation on finite domains, the data yield size-scaling exponents of $\theta=-0.10\pm0.02$ and $\zeta=0.669\pm0.004$, in excellent agreement with exact theoretical predictions. As a function of temperature, the remanent magnetization is a maximum above the Curie transition, and, for some thermal histories, becomes negative in the ferromagnetic regime. The behavior is consistent with a percolation backbone that is negatively coupled to the finite domains.

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Relaxation of the remanent magnetization in ferromagnetic materials has previously been attributed to rotation of individual domains, wall motion between domains, or relaxation of a continuum of internal degrees of freedom. Most models of domain rotation [1,2] consider activation over a smooth distribution of barrier heights, resulting in logarithmic time dependences, for which the 1949 measurements of Street and Woolley [3] remain the most cited evidence [4,5]. Power-law behavior is predicted by scaling theories for domain growth [6] and internal dynamics [7]. Other treatments yield Kohlrausch-Williams-Watts stretched-exponential relaxation [8]. All of these empirical formulas have divergent slope at short times, and hence they can only be approximations valid over a limited range. Complete numerical solution of semiclassical Ising [9] and Heisenberg [10] models generally exhibit relaxation times that increase with domain size. We have measured the magnetic relaxation of ferromagnetic EuS from 10^{-4} to 10^4 sec after removing an applied field. The quality and range of the data are sufficient to demonstrate significant deviations from all of these previously proposed relaxation mechanisms.

A recent model for relaxation of quantized excitations (magnons, phonons, polaritons, etc.) on a distribution of finite domains has been shown to give excellent agreement with observed magnetic relaxation in spin glasses [11], stress relaxation in ionic glasses [12,13], and dielectric susceptibility of glass-forming liquids [14,15]. The model is based on standard domain-size distributions and elementary finite-size quantization, thus providing a common link between fundamental excitations and observed dynamic response. Here we show that the remanent magnetization of single-crystal EuS is dominated by finite-size quantized magnons.

Macroscopic ferromagnetic samples segregate into mesoscopic domains to reduce their magnetic dipolar energy. The lowest-energy domains (Goldstone modes), which form as metastable droplets in an otherwise ordered system, generally have a positive surface tension and a negative energy term proportional to volume. Minimizing the free energy leads to a domain-size distribution of the form [16–18] $n_s \sim s^{-\theta}\exp(-s^{\zeta})$. The size-scaling exponents, $\theta=-\frac{1}{9}$ and $\zeta=\frac{2}{3}$, are exact theoretical predictions for isotropic low-energy excitations, regardless of local structure or nature of interaction.

We make the reasonable assumption that mesoscopic domains have homogeneously extensive static susceptibility, but size-dependent relaxation rate (w_s), so the net relaxation becomes $M(t) \propto \sum_{s=0}^{\infty}[sn_s]\exp(-tw_s)$. For activated (Arrhenius-like) behavior the relaxation rate is $w_s \propto \exp(-\delta E/k_BT)$. The most important feature of our model is that we consider the relaxation of finite-size quantized dispersive excitations, for which the energy of an individual excitation (Δ) is distributed throughout its domain, producing an average energy-level spacing per particle of $\delta E=\Delta/s$. This is true regardless of the dispersion relation since densities of states are always proportional to volume ($\delta N/\delta E \propto V \propto s$). Using $x \propto s$, the net relaxation becomes

$$M(t)=M_i\int_0^{\infty}[x^{10/9}\exp(-x^{2/3})]\exp(-tw_{\infty}e^{-C/x})dx\,, \tag{1}$$

where M_i is proportional to the initial magnetization, w_∞ (the relaxation rate of an infinite domain) is related to an attempt frequency, and $C \propto \Delta/k_BT$ we define as a correlation coefficient. For weakly interacting particles ($\Delta \approx 0$) or high temperatures, the correlation coefficient is small and relaxation is Debye-like (single exponential), whereas for $|C| \gg 1$ the response is extremely broad.

Ferromagnetic samples cooled from the paramagnetic regime in zero external field contain domains oriented in all possible directions, resulting in no net magnetization. At finite temperatures, the magnetic moment of each domain is reduced from saturation by magnon excitations. The average magnon density is uniform for all zero-field-cooled domains, but the level of excitation in field-cooled domains depends on their orientation. "Aligned" domains have a reduced density of magnons, so that their net internal energy *increases* after H is removed, resulting in $C \propto \delta E > 0$. We define aligned domains by $C > 0$; they need not be aligned with H but in general will be oriented with the local field. Similarly "antialigned" domains are defined by their initially higher level of excitation, which *decreases* during relaxation, producing $C \propto \delta E < 0$. The observed relaxation of EuS may be divided into distinct aligned and antialigned temperature regimes.

Although Eq. (1) describes the relaxation of isotropic low-energy excitations without reference to specific models, we find that the temperature-dependent behavior of EuS can best be explained using percolation theory [19]. Percolation theory, with its inherent randomness, has rarely been applied to homogeneous crystals. Dynamic correlations, however, are quite independent of structural order. For example, nonequilibrium liquids exhibit anomalous dynamic correlations over optical length scales [20]. Similarly, neutron-scattering measurements reveal significant dynamic correlations in glasses [21] and random magnetic systems [22,23] well above their freezing temperatures. In EuS, dynamic magnetic correlations persist to at least 3 times the ferromagnetic transition temperature [24].

For magnetic systems, we define a dynamically correlated domain as a region where spins share a common relaxation rate. Quantum mechanically, a domain may correspond to a region where electrons form a single many-bodied system. Thermodynamically, a domain is a microcanonical ensemble which, during relaxation, need not be in equilibrium with neighboring domains or the thermal bath. The existence of persistent, spatially heterogeneous, dynamically correlated domains has recently been demonstrated in a glass-forming liquid using NMR spectroscopy [25].

For percolation we assume that a given spin is dynamically correlated to one of its neighbors with random probability p. When p is less than the critical concentration for bond percolation (p_c) there are only finite domains of correlated spins, whereas for $p > p_c$ there is an infinite backbone in addition to finite domains. Percolation theory provides specific predictions for the distribution of finite domains; for $p > p_c$ in three dimensions [16,18] $n_s \propto s^{1/9}\exp[-(C's)^{2/3}]$, where $C' \propto |p-p_c|^{1/\sigma}$ and $\sigma = 0.45$. Using $x = C's$, the correlation coefficient in Eq. (1) becomes $C = C'\Delta/k_BT$.

Europium sulfide has been extensively investigated as an ideal Heisenberg system [26], with a small anisotropy constant and classic Curie transition at $T_C = 16.57 \pm 0.02$ K. We observed no anisotropy dependence from magnetic measurements of two spherical single crystals [27] in three distinct orientations. Magnetization as a function of time was measured using a SQUID magnetometer coupled to a high-speed voltmeter. Measurements were made by applying a field ($H = 9.0$ Oe) at ~ 25 K, field cooling to the measurement temperature (T), and then removing H and recording the magnetization as a function of time. The absolute magnitude of the remanence was determined by measuring the magnetization while warming before reapplying H. With no current to the solenoid, the "zero"-field-cooled magnetization indicated a residual field of ~ 0.8 mOe, which was subtracted to obtain the best value for the absolute remanence. A small background relaxation in the 0.1–1-msec range was also compensated for. The primary uncertainty was due to instrumental drifts at long times.

Figure 1 is a plot of the remanent magnetization versus logarithm of time for nine temperatures between 4.2 and 18.2 K. The curvature of the data (signifying deviation from logarithmic behavior) may be divided into two temperature regimes. For $T > T_B = 17.75 \pm 0.1$ K, $M(t)$ exhibits portions of an entire S-shaped curve, whereas for $T < T_B$ only negative curvature is visible over the available time window. For $T > T_B$, no simple power law can fit the data since $d^2(t^\alpha)/d(\ln t)^2$ cannot change sign. Equation (1) with $C > 0$ (solid curves) gives excellent agreement throughout the fit range (10^{-3}–10^2 sec) and extrapolates well to shorter and longer times. The positive correlation coefficient indicates aligned domains, and the quality of fit demonstrates that other relaxation mechanisms need not be considered.

The curvature of $M(t)$ vs $\log(t)$ changes abruptly at T_B, and remains negative for all $T < T_B$. Fitting these data requires $C < 0$. Most of the variation in the magnitude of the correlation coefficient is due to the explicit $1/T$ divergence ($|C| \sim 1400/T$), but $C \approx 75$ at 17.8 K changes to $C \approx -75$ at 17.7 K. Simultaneously the relaxation rate of an infinite domain jumps from 10^4 sec^{-1}

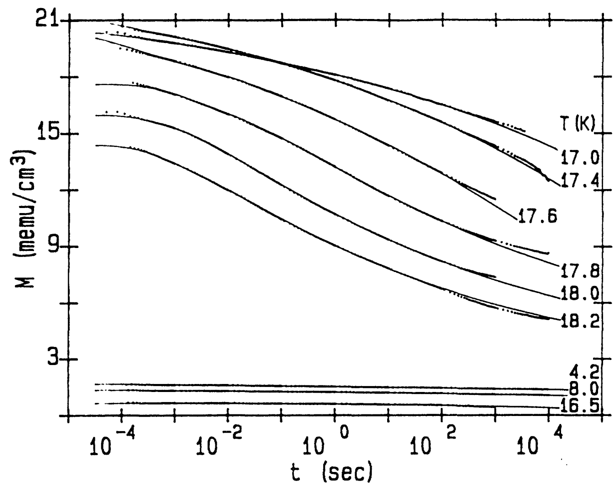

FIG. 1. Remanent magnetization of EuS as a function of time after removing $H = 9.0$ Oe. Above $T_B = 17.75$ K, S-shaped curvature indicates "aligned" relaxation, whereas for $T < T_B$ only $d^2M/d(\ln t)^2 < 0$ can be seen, indicative of "antialigned" behavior. The observed curvature remains negative for $T < T_C = 16.57$ K, but the magnitude of the remanence is much smaller in the ferromagnetic regime. Best fits using Eq. (1) (solid curves) give good agreement with the data.

at 17.8 K, where w_∞ is the fastest rate in the system (smaller domains have larger energy-level spacing, and hence relax more slowly), to 10^{-11} sec^{-1} at 17.7 K where $C<0$ makes w_∞ the slowest relaxation rate. We suggest that the crossover from aligned to antialigned behavior may be due to freezing of semiclassical internal degrees of freedom. At high temperatures, domains align with the local field to reduce their net internal energy. In the vicinity of T_B, reorientation becomes inhibited, and the domains freeze into slowly relaxing higher-energy configurations. This picture is consistent with the fact that above 18.2 K the quality of fits using Eq. (1) deteriorates rapidly with increasing temperature, indicating the onset of alternative relaxation mechanisms. We could not distinguish any cooling-rate or time dependence of the field-cooled state; evidently freezing occurs sharply in the vicinity of T_B.

The best previously proposed relaxation function for $T<T_B$ is a simple power law with $\alpha>0$. Indeed, fits using $M(t)=M_a+M_b t^\alpha$ [which has the same number of adjustable parameters as Eq. (1)] produce small chi-squared deviations (χ_α^2). Nevertheless, for $T<T_B$ the average χ^2 using Eq. (1) is significantly smaller, $\ln(\chi^2/\chi_\alpha^2)=-0.030\pm0.018$. Several facts emphasize that Eq. (1) is the correct function. Most of the uncertainty in $\ln(\chi^2/\chi_\alpha^2)$ comes from the vicinity of T_B. For 20 temperatures below T_C, $\ln(\chi^2/\chi_\alpha^2)=-0.008\pm0.003$ and χ^2 was smaller than χ_α^2 for all but one of these measurements. Furthermore, the values of α necessary to fit the data (≈ 0.1 near T_B, ≈ 0.03 at 4.2 K) are an order of magnitude smaller than standard exponents predicted for traditional dynamic scaling or domain growth. Most convincing, however, is that over the range from 10^{-3} to 10^{1} sec, the quality of the data allows the size-scaling exponents to be set free as additional adjustable parameters, so that size scaling is determined solely by $\delta E \propto 1/s$. At 27 temperatures from 4.2 to 18.2 K we obtain $\theta=-0.10\pm0.02$ and $\zeta=0.669\pm0.004$. Excellent agreement with exact theoretical predictions [17,19] ($\theta=-\frac{1}{9}$ and $\zeta=\frac{2}{3}$) confirms the presence of a percolation distribution of finite domains *and* relaxation rates that vary exponentially with inverse size.

The relaxation rate of an average-sized domain can be obtained directly from the fitting parameters using $\bar{w}\equiv w_\infty \exp(-C/\bar{x})$, where $\bar{x}=(\frac{19}{6})^{3/2}$. In the ferromagnetic regime, $\bar{w}$ increases exponentially with increasing temperature [Fig. 2(a)]. At T_C, $\bar{w}$ exhibits a sharp minimum, characteristic of critical slowing. Smooth continuity of $\bar{w}$ to above T_B (solid circles) is indicative of the subtle crossover from antialigned to aligned behavior.

The percolation correlation length (average radius of finite domains) is given by $\xi \propto (\Delta/|CT|)^{\sigma\nu}$, where $\sigma=0.45$ and $\nu=0.88$. With the physically reasonable assumption of temperature-independent interaction between spins (Δ), ξ is roughly constant throughout the ferromagnetic regime [Fig. 2(b)]. A vague maximum in the vicinity of T_C suggests that ξ may be weakly related to the critically divergent thermodynamic pair-correlation function.

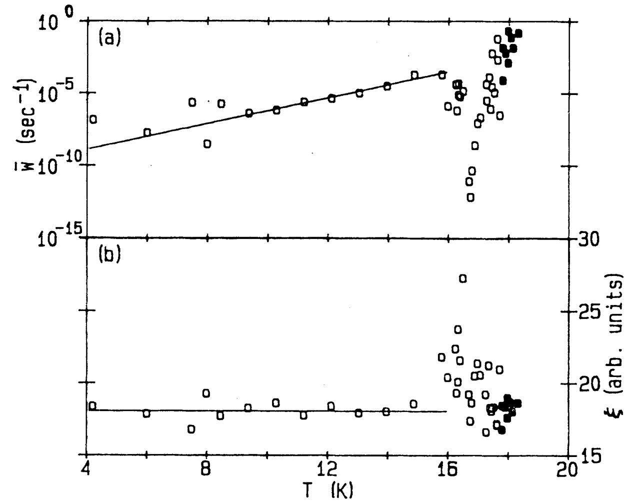

FIG. 2. Temperature dependence of (a) the relaxation rate of an average-sized domain $\bar{w}=w_\infty\exp(-C/\bar{x})$ and (b) correlation length $\xi\propto(\Delta/|C|)^{\sigma\nu}$. Solid lines below T_C indicate linear temperature dependence of $\log(\bar{w})$ and constant ξ in the ferromagnetic regime. Smooth continuity to $T>T_B$ (solid circles) demonstrates the subtlety of the crossover from "antialigned" to "aligned" behavior.

Remanent magnetization as a function of temperature was measured by applying H to the sample at ~ 25 K, field cooling to a starting temperature (T_0), and then removing H and recording the magnetization during warming or cooling. When warmed from $T_0=3$ K [solid curve in Fig. 3(a)] the remanent magnetization exhibits a local minimum in the vicinity of T_C, and conspicuous maximum *above* the Curie transition. Presumably this remarkable behavior has not been previously reported because its observation requires a very small residual field ($<10^{-3}H$). The behavior is consistent with a remanent magnetization that is due primarily to finite percolation domains, which for $p>p_c$ are compact ($D=3$), and hence they may order at higher temperatures than the infinite backbone which is fractal ($2.5\le D_{bb}\le 3$). The backbone dominates the field-cooled magnetization, ac susceptibility, and thermodynamic behavior, but has surprisingly little remanence by 100 μsec after H is removed.

When field cooled to $T_0=16$ K [dashed curve in Fig. 3(a)] the remanent magnetization again exhibits a local minimum in the vicinity of T_C. For $T_0=17.4$ K, however, the remanence becomes *negative* below T_C [Fig. 3(b)]. This behavior is consistent with an infinite backbone that orders abruptly at T_C. When field cooled below T_C, the backbone aligns in the direction of H, producing a net positive remanence. Whenever $H=0$ above T_C, negative coupling causes the backbone to align oppositely to the finite domains, resulting in net negative remanence below T_C. This is verified in Fig. 3(c), where

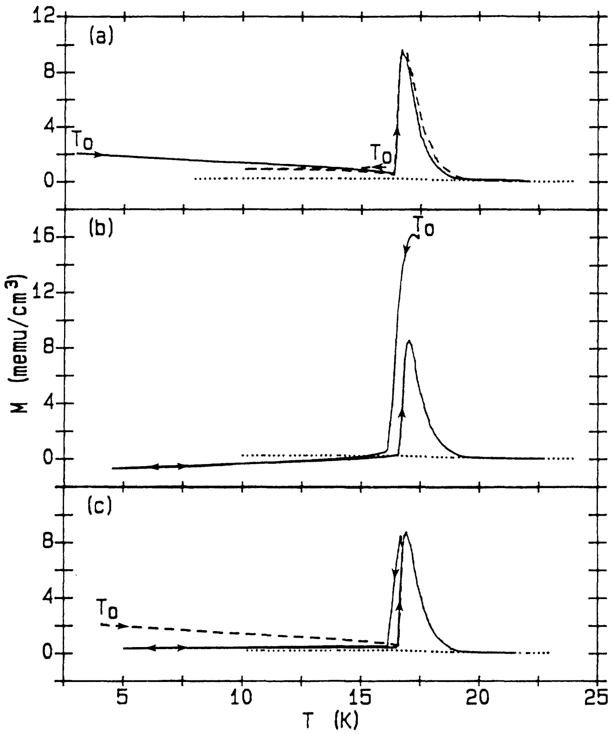

FIG. 3. Remanent magnetization of EuS for various thermal histories. (a) When H is removed in the ferromagnetic regime ($T_0=3$ K, solid curve; $T_0=16$ K, dashed curve) the remanence exhibits a local minimum in the vicinity of T_C and conspicuous maximum above the Curie transition. (Dots are continuation to "zero"-field cooled without SQUID reset, demonstrating negligible residual field and absolute magnitude of the remanence.) (b) For $T_0=17.4$ K, the remanence becomes negative below T_C. (c) For $T_0=4$ K, the remanence was positive during warming to 17 K (dashed curve), then reverses upon subsequent cooling below T_C (solid curve).

the sample was field cooled to $T_0=4$ K, exhibited positive remanence while warming to 17 K (dashed curve), then reverses upon subsequent cooling below T_C. The remanence reversal concomitant with T_C confirms the presence of negatively coupled, *distinct* magnetic systems in this simple ferromagnet.

In conclusion, magnetic relaxation measurements provide strong evidence that the remanent magnetization of single-crystal EuS is dominated by magnons on a universal distribution of finite domains. As a function of temperature the small, sometimes negative, remanence below the Curie transition is consistent with a percolation backbone that is negatively coupled to the finite domains. The backbone dominates equilibrium behavior, but has surprisingly little influence on the remanent magnetization.

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