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[10]See Ref. 1 for a definition of the $\vec{\phi}^{(i)}$ and their relation to $\vec{v}_s$ and $\vec{I}$.

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Magnetic Resonance in a Concentrated Spin-Glass: Experiment and a Phenomenological Model

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The microwave spectrum of amorphous sputtered films of $Gd_{0.37}Al_{0.63}$ exhibits a resonance which moves rapidly to lower fields as temperature is decreased below the paramagnetic Curie temperature at 30 K and which exhibits no anomaly at the spin-glass ordering temperature of 16 K. The shift of the resonance field is interpreted as an effective anisotropy field arising from local demagnetizing fields of the inhomogeneous magnetic system. The theory is also compared to earlier resonance results on CuMn.

In this Letter we report on the microwave resonance spectrum of amorphous GdAl and describe a semiquantitative model for the resonance line positions. The conclusions are of wider interest for the study of spin-glasses or other inhomogeneous magnetic systems because our theory indicates that the resonance line positions act as a sensitive probe of the spatial inhomogeneity of the system while standard magnetic measurements yield information primarily about the spatially averaged properties.

Magnetic measurements[1-3] on an amorphous sputtered film of $Gd_{0.37}Al_{0.63}$ have revealed a paramagnetic Curie temperature of 30 K, indicating a predominance of ferromagnetic exchange interactions. However at 16 K there appeared a sharp maximum in the dc low-field magnetic susceptibility, indicating a spin-glass ordering which presumably arose from the presence of random antiferromagnetic interactions along with the ferromagnetic interactions. Evidence of spatial magnetic inhomogeneity has come from specific-heat measurements,[4] which indicate partially ferromagnetic clusters with dimensions of order 35 Å, while small-angle x-ray scattering measurements[5] indicate structural inhomogeneities of undetermined nature but of comparable dimensions.

Here we describe the magnetic resonance spectrum of the same GdAl material,[6] obtained with a standard Varian electron spin resonance spectrometer at 9.13 GHz. We observed a broad resonance ($\Delta H \sim 1$ kOe) and measured the resonance fields $H_{\parallel}$ and $H_{\perp}$ as a function of temperature with dc fields applied parallel and perpendicular, respectively, to the film plane. At high temperature the resonance corresponds to $g = 2$, as expected for the Gd^{3+} ion. At lower temperatures the parallel and perpendicular resonances move apart because of a well-known demagnetizing-field effect, and below the paramagnetic Curie temperature both resonances plunge steeply to lower fields, as shown in Fig. 1. In this region the resonance line broadens rapidly and can no longer be measured accurately below about 12.5 K. Also shown in Fig. 1 are $M_{\perp}$ and $M_{\parallel}$, the average static magnetizations at the fields $H_{\perp}$ and $H_{\parallel}$, as interpolated from previously reported magnetic measurements.[1]

Qualitatively similar resonance results have been obtained in earlier studies of CuMn alloys by Owen and co-workers[7,8] and by Griffiths,[9] who observed resonance fields shifting downward with decreasing temperature near the magnetic ordering. To interpret the shift, Owen and co-workers[7,8] assumed that CuMn was a simple antiferromagnet giving an antiferromagnetic resonance at

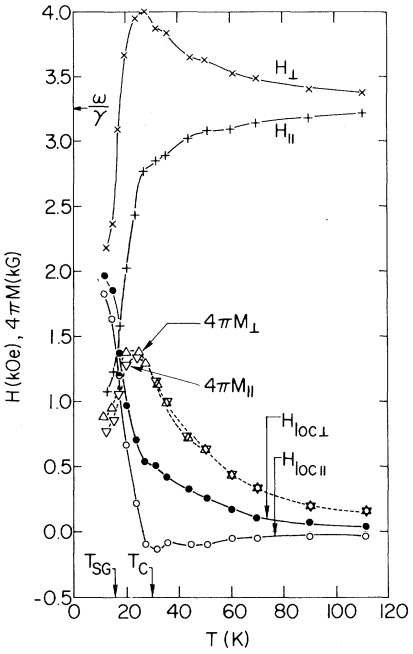

FIG. 1. Magnetic resonance fields $H_\perp$ or $H_\parallel$ at 9.13 GHz magnetizations $M_\perp$ and $M_\parallel$ at $H_\perp$ and $H_\parallel$, respectively, and $H_{\mathrm{loc}\perp}$ and $H_{\mathrm{loc}\parallel}$ from Eqs. (9) and (10) for an amorphous sputtered $Gd_{0.37}Al_{0.63}$ film as a function of temperature. $\perp$ and $\parallel$ refer to the dc applied field perpendicular and parallel to the film plane, respectively. T_{SG} is the spin-glass ordering temperature and T_C is the paramagnetic Curie temperature.

the field

$$H = (\omega^2\gamma^{-2} - H_c^2)^{1/2}, \quad (1)$$

where ω is the angular microwave frequency, γ the gyromagnetic ratio, and H_c the antiferromagnetic critical field. In their theory, H_c is given by $(2H_E H')^{1/2}$ where H_E is an antiferromagnetic exchange field arising from direct exchange between Mn ions and H' is a "conduction-electron field" which arises from the *sd* exchange interaction and which is assumed to act like an anisotropy field. We consider such a theory quite implausible for our amorphous GdAl system. The predominance of ferromagnetic interactions in GdAl, as indicated by the paramagnetic Curie temperature, makes its treatment as a simple antiferromagnet highly artificial. Furthermore, direct exchange between the well-shielded *f* electrons of the Gd^{3+} ions is negligible. The theory is also implausible for CuMn, since CuMn is now also known to be an inhomogeneous magnetic system with a combination of ferromagnetic and antiferromagnetic interactions.[7,10,11]

Here we propose an alternative theory for the downward shift of the resonance fields. Our theory is based on the effect of inhomogeneities, which are likely to be present in any (spin-glass) system with mixed ferromagnetic and antiferromagnetic interactions.[11,12] For example, let us imagine, as shown schematically in Fig. 2, regions enriched in gadolinium or regions where the nearest-neighbor configurations are such as to give a particularly high percentage of ferromagnetic interactions. These regions are surrounded by a matrix in which there are more competing (antiferromagnetic) exchange interactions. We presume a statistical distribution of shapes for the ferromagnetic regions: some spherical, others prolate, others oblate. If the overall distribution is isotropic, there are as many prolate or oblate shapes pointing in one direction as in another. The scale of the structure in Fig. 2 is presumed to be several tens of angstroms, as suggested by the specific-heat and small-angle x-ray scattering measurements. This assumption allows us to use micromagnetic theory to treat the problem, which gives a considerable simplification. However we emphasize that in principle the problem could also be treated on an atomistic scale, in which case we would expect qualitatively similar conclusions.

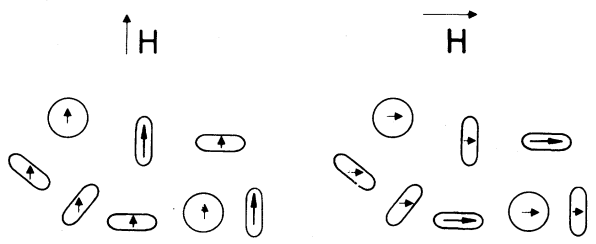

FIG. 2. Schematic inhomogeneous magnetic medium in applied field $\vec{H}$.

Consider now the effect of an applied magnetic field on such a magnetic system near the paramagnetic Curie temperature, so that there is incipient magnetic order in the ferromagnetic regions. Because of the low local demagnetizing fields, the needlelike regions pointing along the applied-field direction will magnetize most strongly.[12] Spheres or orthogonally oriented needles magnetize less strongly because of their larger demagnetizing fields. For example if the incipient "ferromagnetic" regions are described by a volume susceptibility χ, the parallel needles have magnetization χ while the perpendicular ones have magnetization $\chi(1+2\pi\chi)^{-1}H$. As χ diverges near the paramagnetic Curie temperature,

the ratio of the magnetizations in the two regions diverges as $2\pi\chi$. Therefore we crudely approximate the induced static micromagnetic magnetization distributions as $M_z(x,y)$, homogeneous in the z direction along the applied field but spatially varying along the orthogonal x and y axes. That is, we assume that the induced magnetization forms a columnar pattern. As indicated in Fig. 2, the columnar axis follows the applied dc field $\vec{H}$.

In analogy with the columnar anisotropy mechanism of Cargill and Mizoguchi,[13] this field-induced columnar magnetization inhomogeneity can give rise to an effective anisotropy field H_{1oc} which is responsible for the resonance field shift in the following way. Let us suppose the exchange coupling between spins is sufficiently strong that in spite of the inhomogeneity of the system, the spin precession angle θ during resonance is essentially constant over the characteristic distance separating the columns. If this condition is not fulfilled, then the different regions will precess independently and the response can no longer be considered as a single resonance mode. Let us further suppose that during the precession, the local induced magnetization $M(x,y)$ retains its static magnitude, which requires that its relaxation rate be small compared to the precession frequency.

Since the precessing magnetization has a component $M(x,y)\sin\theta$ which is perpendicular to the dc-field direction and which is spatially varying, there must exist a local magnetostatic energy density[13] $\frac{1}{2}H_d\Delta M\sin^2\theta$. Here ΔM is the deviation $M(x,y)-\langle\vec{M}\rangle$ of the magnetization from its spatial average $\langle\vec{M}\rangle$ and H_d is the demagnetizing field $2\pi\Delta M$ of a column. Spatially averaging this energy density and equating it to $K_u\sin^2\theta$, where K_u is the effective anisotropy constant, one finds the effective average anisotropy field $H_{1oc}=2K_u/\langle\vec{M}\rangle$,[13]

$$H_{1oc}=2\pi\langle|M(x,y)-\langle M(x,y)\rangle|^2\rangle/\langle M(x,y)\rangle. \quad (2)$$

For example, in the idealized case where the columnar ferromagnetic regions have magnetization M_1 and occupy a volume fraction V_1 of the sample, while the remainder of the sample has magnetization M_2, one finds

$$H_{1oc}=2\pi V_1(1-V_1)(M_1-M_2)^2/\langle\vec{M}\rangle, \quad (3)$$

where

$$\langle\vec{M}\rangle=(M_1-M_2)V_1+M_2. \quad (4)$$

These results are valid so long as the intercolumnar distance is less than the range over which exchange holds the spins strongly coupled against demagnetizing forces. A measure of this range[6] is the parameter $2\pi\Lambda_0$, where $\Lambda_0=(A/2\pi M^2)^{1/2}$ is a conventional length parameter in micromagnetic theory and A is the exchange stiffness. To estimate the parameter we use expressions for the average exchange stiffness[14]

$$A=\tfrac{1}{6}NJS^2X^2zr^2, \quad (5)$$

the average magnetization

$$M=gS\beta XN, \quad (6)$$

and the Curie temperature in the molecular-field model

$$kT_C=\tfrac{2}{3}zXS_0^2J. \quad (7)$$

Here N is the number of atoms per unit volume, J is the average Gd-Gd exchange constant (defined as $E_{ij}=-2J\vec{S}_i\cdot\vec{S}_j$), S is the average temperature- and field-dependent Gd spin value, S_0 is the zero-temperature value of S $(=\frac{7}{2})$, X is the atom fraction of Gd, z is the average number of nearest neighbors, r is the Gd-Gd interatomic distance, g is the g factor, β is the Bohr magneton, and k is Boltzmann's constant.

Considering the local anisotropy field $\vec{H}_{1oc}$ to act along the columns and hence along the applied-field direction, one can easily generalize the conventional resonance equations[15] to

$$\omega/\gamma=H_\perp-4\pi M_\perp+H_{1oc\perp}, \quad (8)$$

$$\omega/\gamma=[(H_\parallel+H_{1oc\parallel})(H_\parallel+H_{1oc\parallel}+4\pi M_\parallel)]^{1/2}, \quad (9)$$

where $\perp$ and $\parallel$ refer to the applied-field configurations perpendicular or parallel to the film plane, respectively. As before, $M_\perp$ and $M_\perp$ and $M_\parallel$ are the static average induced magnetizations at $H_\perp$ and $H_\parallel$, respectively. We have included $\perp$ and $\parallel$ subscripts on H_{1oc} to allow for the possibility of some structural anisotropy.

Applying these equations to our GdAl sample, we first estimate $2\pi\Lambda_0$ using Eqs. (5)–(7) and the parameters $N=4.5\times10^{22}$ cm^3, $T_C=30$ K, $r=3.5$ Å, $X=0.37$, $g=2$, and $S_0=\frac{7}{2}$. We find $2\pi\Lambda_0=35$ Å, which is comparable to the inhomogeneity scale observed in other experiments.[4,5] This justifies our use of the strongly coupled limit. Next we determine $H_{1oc\parallel}$ and $H_{1oc\perp}$ from Eqs. (8) and (9), with the results shown in Fig. 1. There is a noticeable difference between the two values, which could be explained if more of the needles of Fig. 2 were oriented normal to rather than lying in the film plane. Such an anisotropy is a common feature of materials prepared by thin film deposition techniques. But above and beyond this structural

anisotropy effect, H_{1oc} increases strongly for both parallel and perpendicular configuration below the paramagnetic Curie temperature. This is physically plausible since below this temperature one expects ferromagnetic clusters to be forming and the magnetization inhomogeneity to be increasing. The magnitude of $\vec{H}_{1oc}$ is also plausible. For example, the maximum observed value of H_{1oc} is 1900 Oe at 12.5 K where $4\pi\langle\vec{M}\rangle \sim 880$ G. $Gd_{0.37}Al_{0.63}$ would have $4\pi M = 12\,600$ G if it were fully ferromagnetically aligned and therefore we can expect magnetization deviations $4\pi(M_1 - M_2)$ of this order. If we first assume $M_2 = 0$ and take $4\pi\langle\vec{M}\rangle = 880$ G as observed, we can determine $V_1 = 0.07$ from Eq. (4) and $H_{1oc} \approx 6000$ Oe from Eq. (3). With these assumptions, H_{1oc} is even larger than observed. Working backwards from the observed H_{1oc}, we find $V_1 = 0.02$ and $4\pi M_2 = 630$ G, which are reasonable values. A more quantitative test of the theory must await more detailed structural or magnetic information on the distribution or strength or the local magnetization inhomogeneities.

A similar model can be applied to CuMn. For example, using data of Owen *et al.*[7] and Kouvel[10] on 5.5% Mn in Cu, one has an observed line shift of 1300 Oe at 1.3°K and $4\pi\langle M\rangle \sim 50$ G. The CuMn sample would have $4\pi M \sim 2500$ G if it were fully ferromagnetic with a spin of $\frac{5}{2}$ on each Mn. Assuming a magnetization deviation $4\pi(M_1 - M_2)$ of this order and taking $M_2 = 0$, one predicts $V_1 = 0.02$ and $H_{1oc} \approx 1200$ Oe, in agreement with the observed value. From the observed paramagnetic Curie temperature of 50 K, the range of the exchange coupling can be estimated to be of order 85 Å. While these estimates are at best semiquantitative and while we cannot rule out the earlier interpretation of Owen *et al.*, we believe our inhomogeneity model offers a more natural interpretation of the CuMn resonance shifts.

In conclusion, we have described a semiquantitative model for the resonance line shifts in concentrated spin-glasses. The effect arises from local demagnetizating fields in the inhomogeneous system. These fields are related to the anisotropic dipolar fields of clusters in the model of Holtzberg, Tholence, and Tournier[16] but we have introduced an additional element, namely the field-induced columnar magnetic structure, without which the line shift is hard to explain. Our results offer a new additional means for characterizing messy magnetic systems. For example, as the concentration of Gd increases, the magnetic properties of amorphous GdAl films appear to approach more conventional ferromagnetic behavior.[3] In the crossover region it is hard to say whether the material is a spin-glass or a ferromagnet. We have shown here how the resonance line shifts can measure the degree of inhomogeneity in such a system and thus characterize quantitatively the transition from spin-glass to ferromagnet as a function of composition. A more complete resonance study of such a transition will be presented later.[6]

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