Experimental Demonstration of the Existence and Subsequent Breakdown of Triad Dynamics in the Spin-Glass CuMn

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ESR measurements of a CuMn spin-glass performed as a function of frequency and magnetic field angle confirm that there is a mode splitting as predicted by the triad model. At large field angles neither the triad nor vector models are consistent with the data for any assignment of anisotropy constants, suggestive of a breakdown in the assumption of a rigid spin rotation. However, at 180° the data are well described by a vector model. Determinations of the anisotropy constants by various experimental methods are discussed.

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The existence of macroscopic anisotropy energy in the metallic Rudderman-Kittel-Kasuya-Yosida spin-glasses has been demonstrated by several experimental techniques including hysteresis,[1] transverse susceptibility,[2,3] torque,[4,5] NMR,[6] and ESR.[7,8] However, the anisotropy constants determined by these techniques are not in agreement. Most important, the macroscopic description of spin-glass dynamics has not been experimentally established. In this Letter we present ESR measurements as a function of frequency and magnetic field angle which exhibit a mode splitting thereby confirming the existence of a third mode. This new mode implies that the macroscopic internal degrees of freedom must be represented as an orthonormal triad, as suggested by Halperin and Saslow,[9] as opposed to a unit vector.[8] We find that our ESR data are satisfactorily described by the triad model for modest angular displacements of the remanent magnetization from the field-cooled position, $\theta_r \lesssim 90°$, provided that T is much less than the spin-glass transition temperature T_g. For larger rotation angles, $90° < \theta_r < 180°$, there is no choice of anisotropy constants that correctly describe all the data. Paradoxically, at $\theta_r = 180°$ the observed frequency versus resonant field relationship is in excellent agreement with a vector model incorporating both unidirectional and uniaxial anisotropy constants.

The vector model was introduced to interpret the observed ESR and dc magnetization for the CuMn spin-glass under conditions where the applied field was parallel to $\hat{N}$, the direction of H_c, the magnetic field in which the sample was cooled.[8] The free energy, as extended to include both a unidirectional (K_1) and uniaxial (K_2) anisotropy constant, is given by

$$F = (1/2\chi)(\vec{M} - M_r\hat{n})^2 - \vec{M}\cdot\vec{H} - K_1(\hat{n}\cdot\hat{N}) - \tfrac{1}{2}K_2(\hat{n}\cdot\hat{N})^2, \quad (1)$$

where χ is the susceptibility, $\hat{n}$ is associated with the order parameter, M_r is the remanent magnetization with direction $\hat{n}$, and $\vec{H}$ is the applied magnetic field at angle θ_H measured from $\hat{N}$. This free energy implies that the total magnetization is $\vec{M} = \chi\vec{H} + M_r\hat{n}$.

The equations of motion for $\vec{M}$ and $\hat{n}$, obtained from this free energy, predict two ESR modes, $\omega^{\pm}$, whose dependence upon the various parameters was confirmed for the applied field parallel to $\hat{N}$. However, measurements of the field at resonance, H_r, for the ω^+ mode as the field direction was rotated away from $\hat{N}$ were not satisfactorily explained.

The triad model was developed by Saslow,[10] and Henley, Sompolinsky, and Halperin.[11] The free energy is given by

$$F = (1/2\chi)(\vec{M} - M_r\hat{n})^2 - \vec{M}\cdot\vec{H} - K_1\cos\theta - \tfrac{1}{2}K_2\cos^2\theta, \quad (2)$$

where θ is defined by $\mathrm{Tr}R = 1 + 2\cos\theta$, where R is the rotation matrix for the orthonormal triad of vectors describing the motion of $\vec{M}_r$. The solutions of the corresponding dynamical equations now contain, in addition to the $\omega^{\pm}$ modes, a third mode, ω_L, which is longitudinal.

To gain further insight and test the microscopic theories of the spin-glass state, it is necessary to determine which model is representative of real spin-glasses. The equilibrium value of θ_r as a function of H and θ_H is identical for the two models provided that $K_1 + K_2\cos\theta > 0$. Hence, no differentiation may be made by static measurements in this regime.[12] In contrast, the dynamical behaviors of the two models differ since the triad model contains a longitudinal resonance. At $\theta_H = 0$, the transverse ($\omega^{\pm}$) modes of both models are identical, and the longitudinal mode (ω_L) is located at $[(K_1 + K_2)/\chi]^{1/2}$. A direct observation of the longitudinal mode at this angle is extremely

difficult since ω_L is independent of the applied field. However, for finite values of θ_H there is a mixing of the longitudinal and transverses modes whenever they cross. Observation of the consequent splitting at such crossings constitutes a specific signature of the triad model.

The ESR measurements were made with a variable-frequency homodyne reflection spectrometer with a sample which formed part of the center conductor of a coaxial cavity. The magnetization was measured *in situ* by the method of Schultz *et al.*[8]

In Fig. 1 we present the spectrometer frequency as a function of magnetic field at resonance, H_r, for three different angles (θ_H). The solid and dashed lines are calculated from the triad and vector models, respectively.[13] In Fig. 1(a), at $\theta_H = 0$, the two calculations are identical for the $\omega^{\pm}$ modes. From these data and the measured values of M_r and χ we obtain the sum of the two anisotropy constants $K^+/\chi = (K_1 + K_2)/\chi = (7.4 \pm 0.2) \times 10^6$ erg/cm^3. For modest angles neither theory is significantly dependent upon the distribution of K^+ between K_1 and K_2, and in Figs. 1(b) and 1(c) the theoretical curves are calculated by using $K_1 = K^+$ and $K_2 = 0$, as suggested by the results of Hippert, Alloul, and Fert.[3] In Fig. 1(b), $\theta_H = 30°$, and in Fig. 1(c), $\theta_H = 60°$, the data clearly exhibit the mode splitting as predicted by the triad model. We believe that this is the first demonstration of the existence of the longitudinal mode in a spin-glass.

The data for Fig. 1 were taken at the lowest temperature (1.4 K) we could conveniently obtain and correspond to $T/T_g \cong 0.04$. We have taken similar data up to 8 K, and while it is clear that the model splitting still exists, there are increased deviations from the theoretical triad curve.

As θ_H is increased past 60° the agreement with the triad theory deteriorates rapidly even at the lowest temperature. For the region $90° \lesssim \theta_H < 180°$, there is no satisfactory way to describe the data for any choice of K_1/K_2. We suggest that for this situation the assumption of a rigid spin rotation, inherent in both models, breaks down in a manner yet to be understood. Surprisingly, we find that at $\theta_H = 180°$ the entire frequency versus field relation is accurately described by the vector model as shown in Fig. 2 with $K^-/\chi = (K_2 - K_1)/\chi = (0.85 \pm 0.1) \times 10^6$ erg/cm^3.[14] We also show the prediction of the triad model for the same choice of K^- and note that we could find no choice of K_2/K_1 which would allow even a remotely satisfactory fit to the data. (The experimental uncertainties are less than the dot size.) The additional curve in the right-hand lower corner of Fig. 2 is calculated for the triad model with $K_2 = 0$, as inferred from Hippert, Alloul, and Fert,[3] and is clearly inconsistent with the data.

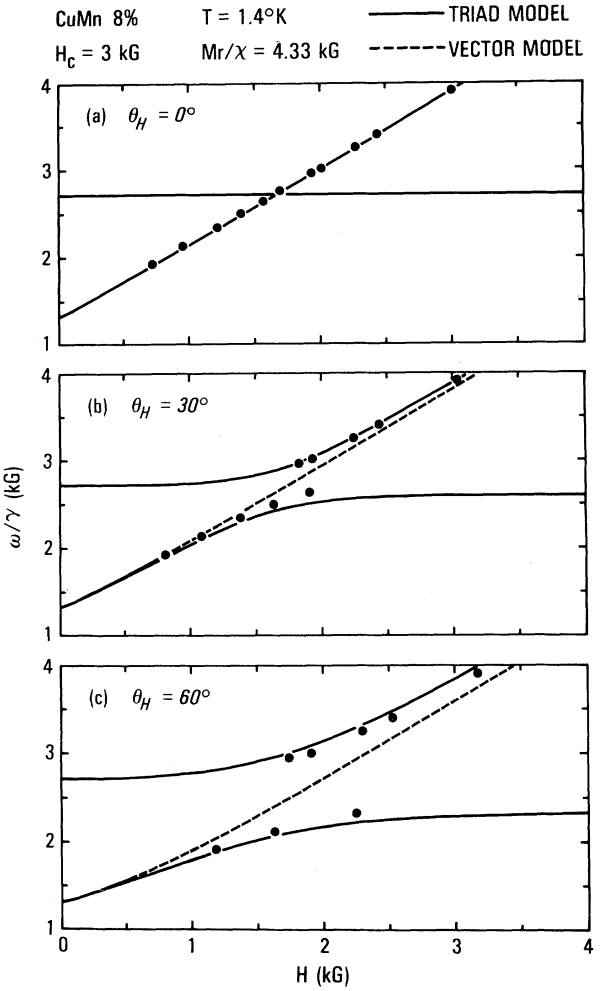

FIG. 1. Spectrometer frequency vs the magnetic field at resonance for three angles, θ_H, of the applied field with respect to the field for cooling, H_c. In (a), $\theta_H = 0$, the transverse modes for both theories are identical and a fit to the data yields a total anisotropy constant $K^+/\chi = (7.4 \pm 0.2) \times 10^6$ erg/cm^3. The triad model predicts a longitudinal resonance at $\omega_L/\gamma = (K^+/\chi)^{1/2} = 2.72$ kG in addition to the observed transverse mode. In (b), $\theta_H = 30°$, and (c) $\theta_H = 60°$, the triad model (solid line) and the vector model (dashed line) are both shown under the assumption of only a unidirectional anisotropy (i.e., $K_2 = 0$). The data clearly exhibit a mode splitting, confirming the prediction of the triad model.

The splitting shown in Fig. 1 is between the ω^+ and ω_L modes. The mode splitting predicted by the triad model in Fig. 2 is between ω^+ and ω^-.

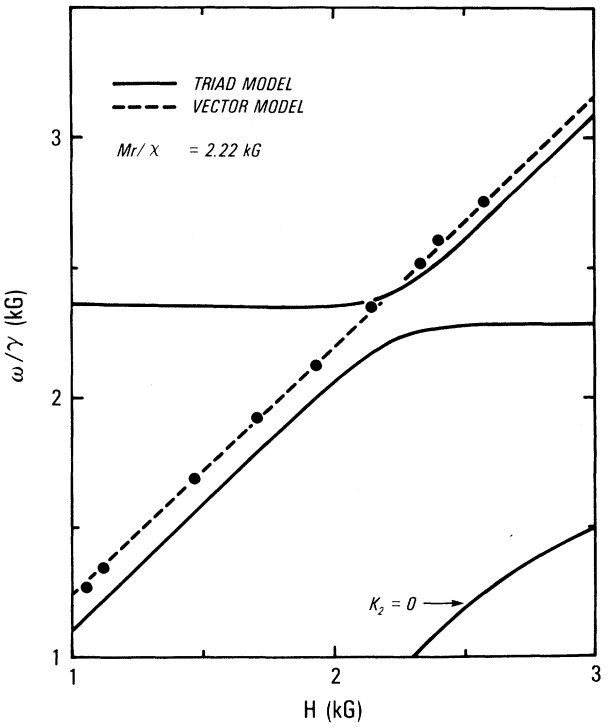

FIG. 2. Spectrometer frequency vs the magnetic field at resonance for a CuMn 8% sample after cooling to 4.2 K in a field, H_c, of 1.5 kG. The magnetic field is applied antiparallel to H_c (i.e., $\theta_H = 180°$). The vector model (dashed line) yields an excellent fit to the data with $K^-/\chi = (K_2 - K_1)/\chi = (0.85 \pm 0.1) \times 10^6$ erg/cm^3. The triad model (solid line) exhibits a splitting of the two transverse modes, $\omega^\pm$, and cannot be fitted to the data with any value of K^-. The experimental uncertainty is less than the dot size shown. A representative fit is shown here with the same K^- as above. The severe breakdown of the triad model with only unidirectional anisotropy ($K_1/\chi = 4.1 \times 10^6$ erg/cm^3; $K_2 = 0$) is illustrated by the curve labeled $K_2 = 0$.

Although we have observed the ω^- mode in CuMnNi samples, we have not yet been able to observe the ω^- mode in pure CuMn.

Given the quality of fit by the vector model at $\theta_H = 180°$, we have examined the extent to which it applies at other angles. The vector model not only does not apply for the range of angles as shown in Fig. 1, but deviates within a few degrees from 180° despite the excellent fit exactly at 180°. We have taken resonance data near $\theta_H = 180°$ where the angle is arrived at via rotation, inversion, or rotation in one plane followed by back rotation in a perpendicular plane. We found no difference in the results.

Only the sum of the anisotropy constants, K^+, can be measured if θ_r is near zero, and all the values reported for the various techniques[2-8] are in agreement. However, in order to determine K_1 and K_2 separately it is necessary to perturb the system to larger angles, where measurements made by different techniques are definitely not in agreement. We review these measurements in the light of the results presented here to provide a summary for the current experimental situation concerning anisotropy in spin-glasses.

(1) $\chi_\perp(z)$: In these experiments the transverse ac susceptibility is measured as a function of a dc field parallel to $\hat{N}$. This technique[2] also determines K^+ and K^- at $\theta_r = 0$ and 180°, respectively. These values agree with those from our ESR measurements. Note that this agreement spans a frequency range from $\simeq 10$ to 10^{10} Hz.

(2) Torque: The restoring torque measurements of both groups[4,5] are in agreement with each other, but are in disagreement with ESR and transverse-susceptibility results. For a low Mn concentration (4.7%) at low temperature (1.5 K) the data are hysteretic after modest rotation angles and, consistent with our large-angle ESR data, suggest nonrigid rotations of the spin system. For high-concentration samples (20% Mn), the torque data are reversible and may be fitted over the entire angular range with $K_2/K_1 \cong 0.1$. Fert and Hippert[5] performed a rotation of the magnetic field by 180° in one plane followed by small rotations in a perpendicular plane and found significant differences in the torque. However, after back-rotation angles of only about 10° the system rapidly reverts to the original torque curve. The torque in this narrow transition region was essentially zero. They ascribe this as evidence for some metastable triad aspect of the system, which is difficult to reconcile with their assignment of only a K_1 and with $K_2 = 0$.[12]

(3) $\chi_\perp(y)$: Hippert, Alloul, and Fert have recently reported measurements of the transverse ac susceptibility as a function of a dc field applied perpendicular to $\hat{N}$. This technique allows the determination of the K_1/K_2 ratio for smaller values of θ_r than the other methods discussed. Their data support the assignment $K_2 = 0$ for θ_r up to 35°, but did not test larger angles. This was the basis for our choice of $K_2 = 0$ for the curves of Fig. 1. However, the confirmation of the mode splitting between the ω^+ and ω_L modes does not depend on any K_1/K_2 assignment for the range of angles shown in Fig. 1.

We conclude that the current experimental situation supports the description of the CuMn spin-

glass by the triad model with only unidirectional anisotropy provided that $T/T_g \ll 1$ and $\theta_r \lesssim 90°$. At larger values of θ_r there is presently no description which encompasses all of the data though at exactly 180° the vector model with unidirectional and uniaxial anisotropy fits surprisingly well. Given the discrepancy between theory and experiment at large angles it may not be appropriate to infer any specific interpretations about the spin-glass state from measurements made under such conditions until further clarification.

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[12]If $K_1 + K_2 \cos\theta < 0$ then there exists a "π rotated state," where $\theta = \pi$ and $\theta_r = \theta_H$, in the triad model. Observation of this state in an equilibrium experiment would distinguish between the two models. However, if $K_1 > K_2$ this state is completely unstable with respect to changes in θ. W. M. Saslow, Phys. Rev. B 26, 1483 (1982).

[13]The resonant frequencies for the triad model were calculated by using Eq. (4.3) of Ref. 11. For the vector model, the equations of motion were obtained from Eq. (1). These were solved to yield the eigenvalue equation for the resonance frequencies.

[14]At the lowest temperature the ESR data at $\theta_H = 180°$ imply that $K^- \simeq 0$. In this case the vector and triad models are indistinguishable. The data in Fig. 2 were taken at a temperature high enough to be able to distinguish between the two models yet low enough that the system still obeyed the triad model at small θ_H.

Enhanced Paramagnetism and Spin Fluctuations in Expanded Liquid Cesium

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^{133}Cs NMR has been used to probe the volume-dependent static and dynamic spin susceptibility of expanded liquid cesium. Data taken over the temperature range 55–1400 °C cover the density range $1.03 < \rho < 1.92$ g cm^{-3}. For $\rho \lesssim 1.5$ g cm^{-3}, both the isobaric temperature dependence and the isothermal pressure dependence of the Knight shift change sign and the spin susceptibility becomes increasingly enhanced. Analysis of the nuclear relaxation rates at low densities indicates changes in the q-dependent susceptibility suggestive of a metallic antiferromagnet above its ordering temperature.

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Liquid alkali metals, expanded by heating toward the liquid-gas critical point, exhibit interesting and unusual properties. Conduction-electron densities nearly five times lower than those of ordinary metals can be achieved before metallic properties give way to a metal-nonmetal transition close to the critical point.[1,2] As elemental, monovalent metals they closely resemble the hypothetical expanded lattice of hydrogen or alkali atoms considered by Mott[3] in his original discussion of the metal-nonmetal transition.[4] Although similar in some respects to dilute metals obtained by solution of donors in a nonmetallic host, e.g., heavily doped semiconductors, liquid-met-

