Macroscopic spin dynamics of spin-glasses with remanence and anisotropy

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Real spin-glasses possess both anisotropy and remanence. To interpret spin wave, electron spin resonance (ESR), and related experiments on such systems, one must develop an appropriate macroscopic spin dynamics, thus extending the hydrodynamic theory of Halperin and Saslow. It is found convenient to employ a local spin coordinate system represented by $\hat{n}$ and θ, so that the macroscopic variables are $(\hat{n}, \theta)$ and the magnetization $\vec{m}$. The equations of motion, for small disturbances about the remanence $\vec{m}_0$, are analyzed. Longitudinal spin waves and transverse spin waves are obtained; for pure uniaxial anisotropy, the transverse spin waves possess an anisotropy-induced gap. For a static field $\vec{H}$ along $\vec{m}_0$ (so $\vec{m} = \vec{m}_0 + \chi_{\parallel}\vec{H}$), the ESR line satisfies $\omega = \gamma H + \omega_a$, where $\omega_a \propto m^{-1}$. The phenomenological free energy is made consistent with these results. The effective transverse susceptibility $\tilde{\chi}_{\perp}$, for small transverse fields, is also calculated. It is noted that measurement of m_0, $\chi_{\parallel}$, and the zero-field values of $\tilde{\chi}_{\perp}$ and the ESR frequency completely determine the parameters of the theory, so that a measurement of the field dependence of $\tilde{\chi}_{\perp}$ can serve as a check on the theory. Spin waves at finite wave vectors, and the effects of remanence on the low-temperature specific heat are also discussed. It is noted that, on the basis of the ESR data, one may conclude that there is an exchange coupling between $\vec{m}_0$ and $\chi_{\parallel}\vec{H}$, for small transverse fields.

I. INTRODUCTION

Recently, Halperin and the author developed the hydrodynamic theory of spin-glasses and other noncollinear spin systems.[1] Anisotropy and external fields were ignored, and systems with remanence were only briefly discussed. Since real spin-glasses exhibit both anisotropy and remanence, and since they are studied in the presence of magnetic fields, the work of Ref. 1 has only a limited validity. Most particularly, being a hydrodynamic theory it is valid only for long-wavelength disturbances ($\vec{k} \approx \vec{0}$, where $\vec{k}$ is the wave vector), and then only if the following conditions are satisfied. First, the predicted frequencies ω_k must be much lower than any "internal" frequencies not describable by the hydrodynamic variables (so that, e.g., from perturbation theory, the frequencies ω_k will not be significantly affected by a coupling to the internal frequencies). Second, they must be much lower than the inverse of the characteristic equilibration time of the system (so that the parameters employed in the hydrodynamic theory, which assumes local equilibrium, do not depend upon frequency). Because the gap induced by anisotropy dominates the $\vec{k} \approx \vec{0}$ behavior of spin systems, the theory of Ref. 1 does not apply near $\vec{k} = \vec{0}$, even if the above two conditions would be satisfied. Only if a certain "window" of wave vectors develops, for $|\vec{k}|$ not so small that anisotropy dominates yet not so large that the long-wavelength expansion becomes invalid, can the results of Ref. 1 be expected to apply to real systems without remanence.

In fact, some of the most interesting properties of spin-glasses are associated with remanence. The remanent magnetization of a field-cooled spin-glass [or thermoremanent magnetization (TRM)], the remanent magnetization of a zero-field-cooled spin-glass to which a field has been applied [or isothermal remanent magnetization (IRM)], and the temporal decay of the remanent magnetization, are all properties that are not completely understood.[2] Furthermore, spin-glasses often show displaced hysteresis loops and sharp jumps in the magnetization on going from one branch of the loop to another.[3] A recent study on CuMn summarizes much of what is known about hysteresis in spin-glasses, and points to the existence of a well-defined anisotropy constant for the spin-glass state.[4] (See the work of Kouvel[5] and of Iwata *et al.*,[6] for additional studies bearing on anisotropy.)

Because real spin-glass systems possess remanence and anisotropy, it is not clear that the theory of Ref. 1 can be subjected to a clear-cut experimental test. On the other hand, recent electron-spin-resonance (ESR) measurements by Monod and Berthier on CuMn provide a probe of the $|\vec{k}| = 0$ mode of a spin-glass with remanence and anisotropy.[7] These authors find that the ESR frequency is well described by the form

$$\omega = \omega_0 + \omega_a \ , \tag{1.1}$$

where $\omega_0 = \gamma H$ is the Larmor frequency (γ is the

gyromagnetic ratio and H is the applied field), and $\omega_a \propto m^{-1}$ (m is the total magnetization). It is one of the major purposes of this paper to explain this result. This requires extending the theory of Ref. 1 to include an external field as well as remanence and anisotropy. Besides the ESR data, the magnetization induced by a small static transverse field is also considered. If these experiments are performed, in addition to the ESR work, the theory can be put to a stringent test, since its parameters will be overdetermined.

Before the theory can be presented, a number of questions, both experimental and theoretical, must be discussed. Section II contains a review of experimental work bearing on remanence, anisotropy, and other questions pertinent to the present investigation. Section III considers the relationship of the hydrodynamic approach to other approaches. Section IV provides the basic theory, both the equations of motion and the free energy, upon which the present paper is based. In Sec. V the spin waves are obtained (both longitudinal and transverse), and their implications are discussed. Section VI considers the transverse response of the system, for small transverse fields. In Sec. VII an estimate of the domain wall thickness is made, supporting the view that there are long-range correlations in the system. The significance of disorder for explaining the unusual properties of the spin-glass state is also discussed.

II. EXPERIMENTAL EVIDENCE FOR LONG-RANGE CORRELATIONS

Because the ESR experiments[7] have been performed on CuMn, we will focus attention on experiments pertaining to CuMn. We begin by noting that, since the ESR experiments observe the free-electron g factor, the Mn ion is probably in an S state, which should have a weak crystal-field interaction, and low single-ion anisotropy.[8]

Furthermore, despite the vastly differing environments of individual Mn ions, the line is sufficiently narrow that the ions must all have about the same effective anisotropy field acting on them. This can be understood if the spins are coupled together through a strong exchange interaction. Thus, any weak, localized, anisotropy fields acting on isolated ions produce an averaged effect on the system, which is strongly coupled as a whole.

The NMR experiments of Alloul provide strong support for this view.[9,10] Alloul has studied the zero-field Cu NMR of the first- and fourth-nearest neighbors of Mn in powdered CuMn (1%).[9] Using a spin-echo technique, he observes an enhancement of the pulsed rf field and of the signal intensity, as the remanence m_0 increases. This he interprets in terms of a strengthening of the external torque $(\vec{m}_0 \times \vec{H})$ relative to a fixed anisotropy torque that hinders the rf rotation of the Mn spins producing the local field $\vec{H}_L$ at the Cu nuclei. (This $\vec{H}_L$, which is primarily due to the neighboring Mn, determines the position of the resonance.) Such an argument holds only if the spins are strongly coupled to one another, feeling an averaged anisotropy torque. In addition, because he finds an enhancement factor which is uniform over the entire system, Alloul concludes that the spins are coupled over macroscopic distances. Another consequence of such reasoning is that one can define an effective anisotropy field, H_A, which should vary as m_0^{-1}. Indeed, this is found experimentally. A detailed discussion of these points is presented in Ref. 10.

Another indication that the spins are coupled over macroscopic distances comes from the hysteresis studies of Ref. 3. There it was found that a crystal of CuMn, showing sharp hysteresis loops, would occasionally develop states of zero net magnetization which, when pulled across a field coil, gave evidence for two oppositely magnetized macroscopic domains. (It is likely that the small m_0, and thus the small field energy, is responsible for the large size of what appear to be domains.)

From these experiments, we conclude that the spins in a spin-glass are strongly coupled to one another, with a correlation which extends over macroscopic distances, and with an anisotropy energy which is an average over the anisotropy energies for individual ions. Furthermore, for the small transverse rf fields employed in Ref. 7, the anisotropy acts upon the total magnetization, not just the remanent magnetization. This must be built into the free energy employed in Sec. IV. Note that hydrodynamics, being a phenomenological theory, cannot explain the underlying physics of the systems it describes. Thus, for spin-glasses with anisotropy and remanence, it is only possible to implement the anisotropy in a phenomenological fashion, not to explain it; this must be done by another, more microscopic theory.

III. THEORETICAL REVIEW

Until the observation, by Cannella and Mydosh,[11] of a sharp cusp in the susceptibility of a spin-glass (AuFe) as a function of temperature, there was no strong indication that the spin-glass phase represented a qualitatively new state of matter. (Previous to this work, only broad susceptibility maxima had been observed, a result explained by Ref. 2 in terms of the remanent properties of spin-glasses.) Edwards and Anderson have described the spin-glass system in terms of an order parameter which involves the long-time memory of each spin for its initial orientation.[12] Implicit in this picture is the view that the system possesses a very complex type of long-range

order, and thus the spin-glass transition is a true thermodynamic transition. It is this viewpoint that was taken in Ref. 1.[13]

The approach of Edwards and Anderson was to construct a mean-field theory, applicable near T_g, the spin-glass transition temperature. Another approach, taken by Walker and Walstedt,[14] has been to study the ground-state properties of the spin-glass Hamiltonian. This is taken to be of the form

$$\mathcal{H} = -\frac{1}{2}\sum_{ij} J_{ij}\vec{S}_i\cdot\vec{S}_j \ , \tag{3.1}$$

where the $\vec{S}_i$ are spins on a lattice (not necessarily periodic), and the J_{ij} are coupling constants between sites. The specific form taken by Walker and Walstedt was

$$J_{ij} = A\cos(2k_0 r_{ij}+\phi)/r_{ij}^3 \ , \tag{3.2}$$

the asymptotic form of the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction. (Here A is a material-dependent parameter which may be taken by comparing with experiment, k_0 is the Fermi wave vector of the host material, r_{ij} is the separation between $\vec{S}_i$ and $\vec{S}_j$, and γ is a phase.) For a sufficiently dilute system, where the value of $2k_0 r_{ij}$ is typically very large, $\cos(2k_0 r_{ij}+\gamma)$ is very like a random number chosen between -1 and $+1$. As such, J_{ij} scales as the concentration c, and this has led to a number of scaling laws which are very well satisfied.[15]

Because the work of Ref. 14 provides us with much of our knowledge about the properties of spin-glasses with the RKKY Hamiltonian of Eqs. (3.1) and (3.2), it is worthy of an extended discussion. In that work, large periodically repeated cubes of an fcc lattice were taken, with $N=96$ and 324 spins randomly chosen to occupy either 0.3 or 0.9% of the sites. Thus, the equilibrium configurations (EC's) were periodically repeated. It was found that there were many nonidentical but nearly degenerate EC's, each of which "seem to consist of different arrangements of nearly identical regions containing ~ 20 spins." Once a given (periodic) EC was determined, its $\vec{k}=\vec{0}$ spin waves were determined. These were found to have (on the scale of the histogram) an apparently continuous distribution of frequencies from zero to a maximum frequency. Furthermore, on the average, the "localization index" increased with frequency. (Note that the localization index of Ref. 14 does not give the spatial extension of a given mode, but rather provides a measure of the number of spins participating in that mode.) Treating the spin waves as bosons, and calculating their specific heat C_m, it was found that, despite a decidedly nonconstant density of states, a nearly linear specific heat was found over a wide range of temperature, in reasonable agreement with experiments then current.[16] It should be noted that this involved temperatures sufficiently high that the modes contributing to C_m were relatively localized.

To understand better the low-frequency end of the spin-wave spectrum of Ref. 14, it is useful to review some of the predictions of Ref. 1, which specifically studied this regime.[17] There it was shown that the hydrodynamic modes associated with a nearly uniform, slowly varying, rotation of the spin system have a linear dispersion relation, with a velocity[1]

$$v = \gamma(\rho_s/\chi)^{1/2} \ . \tag{3.3}$$

Here ρ_s is the spin stiffness constant, whose upper limit ρ_s^0 is given by[1]

$$\rho_s^0 = (18V)^{-1}\sum_{ij} r_{ij}^2 J_{ij}\langle\vec{S}_i\cdot\vec{S}_j\rangle \ , \tag{3.4}$$

(with V the volume) and χ is the magnetic susceptibility.

To compare the results of Refs. 1 and 14, one must be sure that the EC's considered in Ref. 14 have no net magnetization, otherwise certain of the predictions of Ref. 1 must be modified. It turns out that the EC's employed in Ref. 14 did indeed have a small net magnetization. One of the consequences was that a low- (but nonzero-) frequency mode was produced, as is seen in the inset to Fig. 2 of Ref. 14. As m_0 approached zero, it was found that the frequency of that mode also approached zero.[18] Thus, for no remanence, the model considered in Ref. 14 gives three pairs of zero-frequency modes when $\vec{k}=\vec{0}$, in agreement with the predictions of Ref. 1 for $\vec{k}=\vec{0}$.

Additional information is difficult to extract, for a number of reasons: first, the spin-wave velocity cannot be directly determined because the $\vec{k}\neq\vec{0}$ excitations were not studied; second, because of the finite sample size, the system may not appear to be isotropic, nor may its density of states per spin saturate to the bulk value.

The following should be noted. For a spin-glass with no remanence or anisotropy, the results of Ref. 1 (linear dispersion) would predict that $C_m \propto T^3$ at low T. No evidence for this has been seen to date.[19,20] Either the temperatures studied so far have been too high (i.e., the localized modes are dominating C_m, due to their large density of states) or the effects of remanence and anisotropy cannot be neglected, or both.

IV. EQUATIONS OF MACROSCOPIC SPIN DYNAMICS

Because the presence of anisotropy gives the spin-wave spectrum a gap, so that the $\omega\tau\to 0$ limit may not be taken, the theory that follows is not a truly hydrodynamic theory. However, since the $kl\to 0$ limit may be taken (where l is either a characteristic mean free path or internal dimension of the system),

we are still studying the macroscopic dynamics of the system.

In order to construct a hydrodynamic (or here, macroscopic) theory, one must be able to determine the macroscopic variables which will enter into the theory. When there is no remanence, the macroscopic variables for a spin-glass are the magnetization $\vec{m}$ and the average infinitesimal "angle" $\vec{\theta}$ by which the spins are rotated from equilibrium. (To appreciate the difference between $\vec{m}$ and $\vec{\theta}$, consider a collinear antiferromagnet. If an external field $\vec{H}$ is applied perpendicular to the initial alignment axis, a magnetization develops by a tipping of the spins toward $\vec{H}$, but $\vec{\theta}$ is zero. On the other hand, if the alignment axis is rotated, then $\vec{m}$ remains zero but $\vec{\theta}$ is nonzero.) If $\vec{S}_j$ represents the jth spin, then $d\vec{S}_j = \vec{\theta} \times \vec{S}_j$ represents the change in $\vec{S}_j$ caused by the rotation by $\vec{\theta}$. We take $\vec{\theta} = 0$ in equilibrium.

Rotations may also be represented by a right-handed triad of unit vectors. When there is remanence, it is convenient to employ such a representation, where the equilibrium value of one of the triad, $\hat{n}$, is taken along $\hat{m}_0$, the remanence direction. Rather than employ the other two unit vectors, which would overspecify the system, we use an additional angle θ, measured with respect to an arbitrary, but fixed, direction in the plane perpendicular to $\hat{m}_0$. Because of the uniaxial symmetry of the remanent spin-glass, the absolute value of θ should be irrelevant, so we take $\theta = 0$ in equilibrium. When the system is disturbed from equilibrium, so $\vec{\theta}(=0) \rightarrow \vec{\theta} + d\vec{\theta}(=d\vec{\theta})$, we have $d\hat{n} = d\vec{\theta} \times \hat{n}$ and $d\theta = d\vec{\theta} \cdot \hat{n}$, so that $d\vec{\theta} = \hat{n} d\theta + \hat{n} \times d\hat{n}$.

The equations of motion satisfied by $\vec{m}$ and $\vec{\theta}$, in the absence of remanence, anisotropy, and an external field $\vec{H}$, were derived in Ref. 1. They are

$$\partial m_\alpha / \partial t = -\gamma \delta f / \delta \theta_\alpha \ , \tag{4.1}$$

$$\partial \theta_\alpha / \partial t = \gamma \delta f / \delta m_\alpha \ , \tag{4.2}$$

where f is the free energy density,

$$\delta f / \delta \theta_\alpha \equiv \partial f / \partial \theta_\alpha - \vec{\nabla} \cdot [\partial f / \partial (\vec{\nabla} \theta_\alpha)] \ , \tag{4.3}$$

and similarly for $\delta f / \delta m_\alpha$. To include anisotropy and remanence, it is necessary only to change the form of f; the equations of motion are unaffected.

Intuitively, we may appreciate these equations as follows. Equation (4.1) is a statement that the (spin) angular momentum is driven by the torque, the torque being written in terms of the negative angular derivative of the free energy. Equation (4.2) is a statement that the orientation of the spins is driven to align with the internal field. This is more easily seen by considering

$$\partial \hat{n} / \partial t = (\partial \vec{\theta} / \partial t) \times \hat{n} = \gamma \hat{n} \times (-\delta f / \delta \vec{m}) \ .$$

Since $\delta f / \delta \vec{m}$ is conventionally known as the internal field $\vec{h}$, this equation says that $\vec{h}$ causes $\hat{n}$ to precess opposite to the $\gamma \hat{n} \times \vec{H}$ precession caused by an external field $\vec{H}$. (In equilibrium, $\vec{h} = \vec{H}$.[1])

In Ref. 1, the free energy was taken to be of the form

$$f_0 = \tfrac{1}{2} \chi^{-1} \vec{m}^2 + \tfrac{1}{2} \rho_s (\vec{\nabla} \vec{\theta})^2 \ ,$$

which satisfies

$$df_0 = \chi^{-1} \vec{m} \cdot d\vec{m} + \rho_s \vec{\nabla} \theta_\alpha \cdot d(\vec{\nabla} \theta_\alpha) \ ,$$

and thus gives

$$\delta f / \delta m_\alpha = \chi^{-1} m_\alpha, \quad \delta f / \delta \theta_\alpha = -\rho_s \nabla^2 \theta_\alpha \ .$$

To include anisotropy, which is defined with respect to a remanence $\vec{m}_0 = m_0 \hat{z}$, we must do a number of things. For one, χ and ρ_s should be converted to tensors. In addition, an explicit anisotropy energy, depending upon $\hat{n}$ and θ, must be incorporated. The hysteresis measurements of Ref. 7 indicate that this anisotropy has a uniaxial component. For most purposes it will be sufficient to write the anisotropy free energy f_{an} as

$$f_{an} = K_\perp \hat{n}_\perp^2 + K_\parallel \theta^2 \ , \tag{4.4}$$

applicable for small $\hat{n}_\perp$ (defined with respect to $\hat{z}$, so $\hat{n}_\perp \approx \hat{n} - \hat{n}^{(0)} = \hat{n} - \hat{z}$) and small θ. For large $\hat{n}_\perp$ and large θ, one may continue to use the $(\hat{n}_\perp)^2$ form, but θ^2 should be replaced by a trigonometric function like $\sin^2\theta$. A truly uniaxial system will show no dependence on θ, corresponding to $K_\parallel = 0$.

Simply modifying f_0 to make χ and ρ_s tensors, and adding in f_{an} is still not sufficient to provide an appropriate free energy, for the remanence further complicates the issue. If $m_\parallel \equiv \vec{m} \cdot \hat{z}$ is changed, then $df = h dm_\parallel$. If the internal field h differs from its equilibrium value $h^{(0)}$ by only a small amount, then $m_\parallel$ differs from its equilibrium value $m_\parallel^{(0)}$ by

$$dm_\parallel = \chi_\parallel (h - h^{(0)}) = \chi_\parallel dh \ . \tag{4.5}$$

Since $h^{(0)} = H$,[1] this means that, if H is changed by dH, $dm_\parallel = \chi_\parallel dH$. This empirical relation holds, for constant $\chi_\parallel$, over a wide range of values for H.[2,5] Thus we will employ $m_\parallel^{(0)} = m_0 + \chi_\parallel H$, where m_0 is the remanence magnetization.

Even letting $\chi^{-1} m_\parallel dm_\parallel \rightarrow h dm_\parallel$ is still not sufficient to provide us with the desired free energy. This is because we wish to make the final requirement that, in the absence of f_{an}, it should not cost any energy to rotate $\vec{m}_\parallel$ to some other direction. This will be necessary to explain the work of Ref. 7. This means that the transverse part of the magnetic energy must have the form

$$\tfrac{1}{2} \chi_\perp^{-1} (\vec{m}_\perp - \vec{\theta} \times \vec{m}_\parallel)^2 \ .$$

In this way, if a (small) rotation by $\vec{\theta}$ in spin space is

performed, so that

$$\vec{m}_\perp = \vec{\theta} \times \vec{m}_\parallel (= \vec{\theta} \times \hat{z} m_\parallel), \quad \hat{n}_\perp = \vec{\theta} \times \hat{z} \ ,$$

the magnetic part of the free-energy density is unchanged. (Note that such a free energy implies that $\vec{m}_0$ and $\chi_\parallel \vec{H}$ are exchange coupled for small transverse fields.) It is assumed, of course, that $\vec{m}_\parallel$ is small.

Thus we will employ an f whose differential is given by

$$\begin{aligned} df = {} & \tfrac{1}{2}\chi_\perp^{-1} d(\vec{m}_\perp - \vec{\theta} \times \vec{m}_\parallel)^2 + h dm_\parallel \\ & + \tfrac{1}{2}\rho_{s\perp} d(\vec{\nabla}\vec{\theta}_\perp)^2 + \tfrac{1}{2}\rho_{s\parallel} d(\vec{\nabla}\theta)^2 \\ & + K_\perp d(\vec{\theta}_\perp^2) + K_\parallel d(\theta^2) \ . \end{aligned} \tag{4.6}$$

In the above, we used

$$n_x = (\hat{n}_\perp)_x = \theta_y, \quad n_y = (\hat{n}_\perp)_y = -\theta_x, \quad \theta = \theta_z \ , \tag{4.7}$$

so that $(\hat{n}_\perp)^2 = (\vec{\theta}_\perp)^2$.

The following should be noted. For a wide range of values of cooling fields H_c, and applied fields H, Kouvel has found that $\chi_\parallel$ appears to remain nearly constant.[5] Thus $\chi_\parallel$ for $H_c \neq 0$ is rather close to the (isotropic) χ for $H_c = 0$. We would expect the same to be true for $\chi_\perp$, yet for small fields, $\chi_\perp$ is found to be much enhanced over $\chi_\parallel$. In the next section we will show how Eq. (4.6) leads, for small fields to an enhanced effective susceptibility $\tilde{\chi}_\perp$, due to the possibility that $\vec{m}_\parallel$ can rotate into the direction of an applied transverse field $\vec{H}_\perp$. For large fields, after $\vec{m}_\parallel$ has been tipped along $\vec{H}_\perp$, the transverse differential susceptibility is found[5] to equal $\chi_\parallel$.

To include the effect of the magnetic field, we go to a rotating frame of reference. The extra magnetic free-energy density is $-\vec{m}\cdot\vec{H}$, in the rest frame. The extra term in the rotating-frame free-energy density is $-\vec{\omega}\cdot\vec{S}$, where $\vec{\omega}$ is the angular velocity of the rotating frame, and $\vec{S} = \vec{m}/\gamma$ is the spin-angular-momentum density. We choose these terms to cancel: That is, $\vec{\omega} = -\gamma\vec{H}$. In that way, Eq. (4.6) gives the free-energy density in the rotating frame, and Eqs. (4.1) and (4.2) give the equations of motion in the rotating frame. However, the equations of motion in the laboratory frame are given, with $\vec{\omega} = -\gamma\vec{H}$, by

$$d\vec{m}/dt = \partial\vec{m}/\partial t + \vec{\omega} \times \vec{m} \ , \tag{4.8}$$

$$d\vec{\theta}/dt = \partial\vec{\theta}/\partial t + \vec{\omega} \ , \tag{4.9}$$

$$d\hat{n}/dt = \partial\hat{n}/\partial t + \vec{\omega} \times \hat{n} = \partial\vec{\theta}/\partial t \times \hat{n} + \vec{\omega} \times \hat{n} \ , \tag{4.10}$$

$$d\theta/dt = \hat{n}\cdot d\vec{\theta}/dt \ . \tag{4.11}$$

In the following sections we will apply these equations to various situations.

V. SPIN WAVES

We now study the spin-wave modes of this system, for $\vec{H} = H\hat{z}$.

A. Longitudinal modes

Equations (4.8), (4.1), and (4.6) yield

$$dm_z/dt = -\gamma(2K_\parallel\theta - \rho_{s\parallel}\nabla^2\theta) \ . \tag{5.1}$$

Further, Eqs. (4.11), (4.2), (4.6), and (4.5) yield

$$d\theta/dt = \gamma(h - H) = \gamma\chi_\parallel^{-1}\delta m_z \ . \tag{5.2}$$

The uniform equilibrium solution to these equations is obtained from $\delta m_z = 0$ and $\theta = 0$. Combining Eqs. (5.1) and (5.2) gives

$$d^2\theta/dt^2 = -\gamma^2\chi_\parallel^{-1}(2K_\parallel\theta - \rho_{s\parallel}\nabla^2\theta) \ . \tag{5.3}$$

For $\theta \propto \exp[i(\vec{k}\cdot\vec{r} - \omega t)]$, Eq. (5.3) yields

$$\omega^2 = \gamma^2\chi_\parallel^{-1}(2K_\parallel + \rho_{s\parallel}k^2) \tag{5.4}$$

or

$$\omega^2 = \omega_{af}^2 + v_\parallel^2 k^2 \ , \tag{5.5}$$

where

$$\omega_{af}^2 = \gamma^2(2K_\parallel/\chi_\parallel) \tag{5.6}$$

gives the uniform resonance frequency ω_{af}, and

$$v_\parallel^2 = \gamma^2(\rho_{s\parallel}/\chi_\parallel) \tag{5.7}$$

gives the longitudinal sound velocity $v_\parallel$. This mode is analogous to the mode occurring in longitudinal antiferromagnetic resonance[21]; hence the subscript af. ω_{af} and $v_\parallel$ will be estimated in Sec. V B. Note that for an antiferromagnet there is also a transverse resonance[21] with $\omega^2 = \omega_{af}^2 + \gamma^2 H^2$; this is not applicable to the transverse modes of the present system, which will now be discussed.

B. Transverse modes

The linearized equations of motion are given by

$$\begin{aligned} dm_x/dt = {} & -\gamma[\chi_\perp^{-1} m_\parallel (m_y - n_y m_\parallel) \\ & - 2K_\perp n_y + \rho_{s\perp}\nabla^2 n_y] + \gamma m_y H \ , \end{aligned} \tag{5.8}$$

$$\begin{aligned} dm_y/dt = {} & -\gamma[\chi_\perp^{-1}(-m_\parallel)(m_x - n_x m_\parallel) \\ & + 2K_\perp n_x - \rho_{s\perp}\nabla^2 n_x] - \gamma m_x H \ , \end{aligned} \tag{5.9}$$

$$dn_x/dt = \gamma(H - m_\parallel/\chi_\perp) n_y + \gamma\chi_\perp^{-1} m_y \ , \tag{5.10}$$

$$dn_y/dt = -\gamma(H - m_\parallel/\chi_\perp) n_x - \gamma\chi_\perp^{-1} m_x \ . \tag{5.11}$$

[In the above, Eq. (4.7) has been employed to eliminate $\theta_\perp$.] Letting $m_+ = m_x + i m_y$ and $n_+ = n_x + i n_y$,

these equations become

$$dm_+/dt = -i\gamma(H - m_\parallel/\chi_\perp)m_+ - \gamma(m_\parallel^2/\chi_\perp + 2K_\perp - \rho_{s\perp}\nabla^2)n_+ \ , \quad (5.12)$$

$$dn_+/dt = -i\gamma(H - m_\parallel/\chi_\perp)n_+ - i\gamma\chi_\perp^{-1}m_+ \ . \quad (5.13)$$

For $m_+,\ n_+ \propto \exp[i(\vec{k}\cdot\vec{r} - \omega t)]$, we get

$$[\omega - \gamma(H - m_\parallel/\chi_\perp)]m_+ - \gamma(m_\parallel^2/\chi_\perp + 2K_\perp + \rho_{s\perp}k^2)n_+ = 0 \ , \quad (5.14)$$

$$-\gamma\chi_\perp^{-1}m_+ + [\omega - \gamma(H - m_\parallel/\chi_\perp)]n_+ = 0 \ . \quad (5.15)$$

The roots are given by

$$\omega_\pm = \gamma(H - m_\parallel/\chi_\perp) \pm \gamma[(m_\parallel/\chi_\perp)^2 + (2K_\perp + \rho_{s\perp}k^2)/\chi_\perp]^{1/2} \ . \quad (5.16)$$

If $m_\parallel^2 >> 2K_\perp\chi_\perp$, then

$$\omega_+ \approx \gamma H + \gamma(K_\perp + \tfrac{1}{2}\rho_{s\perp}k^2)/m_\parallel \ , \quad (5.17)$$

$$\omega_- \approx \gamma H - 2\gamma m_\parallel/\chi_\perp \ . \quad (5.18)$$

Note that $|\omega_- - \gamma H| >> |\omega_+ - \gamma H|$ in this case. Monod and Berthier see a $k = 0$ mode which is described by Eq. (5.17), but not one described by Eq. (5.18). This may arise either because $|\omega_-|$ is too high in frequency to be observed with their apparatus, or because it is a solution which has become unphysical because it is of so high a frequency that it merges with the continuum of localized states,[14] and therefore it violates the first of the two criteria given in the initial paragraph of this paper.

Before discussing the implications of Eqs. (5.17) and (5.18), note that ω_+ corresponds to $\vec{m}_\perp = \vec{\theta}_\perp \times \vec{m}_\parallel$. In other words, for ω_+ the induced $\vec{m}_\perp$ is solely caused by the rotation $\vec{\theta}_\perp$ acting on $\vec{m}_\parallel$; whereas, for ω_- the induced $\vec{m}_\perp$ is equal and opposite to its contribution from $\vec{\theta}_\perp$. Effectively, ω_+ corresponds to a magnetic analog of an acoustic phonon, with the internal variables $\vec{m}_\perp$ and $\vec{\theta}_\perp \times \vec{m}_\parallel$ nearly completely in phase; whereas ω_- corresponds to the magnetic analog of an optical phonon, with $\vec{m}_\perp$ and $\vec{\theta}_\perp \times \vec{m}_\parallel$ nearly completely out of phase.

The mode whose frequency is described by ω_+ corresponds to transverse ferrimagnetic resonance. (In the past it has often been associated with transverse antiferromagnetic resonance.[8,22]) From the data of Monod and Berthier,[7] it is possible to extract a value for $K_\perp$, which is $K_\perp = 2.1 \times 10^2$ erg/cm^3. Note that $2K_\perp\chi_\perp/m_0^2 \approx 0.3$, consistent with the assumption made in deriving Eq. (5.17). (For the sample of Ref. 7, $m_0 \approx 0.53$ erg/G cm^3, and we take $\chi_\perp \approx \chi_\parallel \approx 2 \times 10^{-4}$ emu/cm^3.) For $H = 0$, Eq. (5.18) gives $|f_-| \approx 16$ GHz, thus indicating where the second ESR line should lie, if it does not overlap the continuum of localized states.

We now consider what may be expected for $k \neq 0$. To do so we need ρ_s, the exchange stiffness constant. In Eq. (3.3) an expression is given which provides an upper bound for ρ_s. Preliminary calculations indicate that ρ_s is down from this upper bound by a factor of 2 or 3.[18] Hence ρ_s can be estimated. We will do this by employing $\rho_s = \chi(v^2/\gamma^2)$ and an upper-bound estimate for the spin-wave velocity v. Specifically, Huber and Ching derive an approximate upper bound for v, appropriate to an RKKY interaction, given by[23]

$$v_{ub}^2 = 2\pi^2 S(S+1)cA^2/27\hbar^2(2k_0)^5 \ . \quad (5.19)$$

Here c is the number of spins per unit volume, $A = A'(2k_0)^3$ is the amplitude of the RKKY interaction of Eq. (3.2), and k_0 is the Fermi wave vector of the host material. For CuMn (1%) we have $c = 8.45 \times 10^{20}$ cm^{-3}, $A' = 9.5 \times 10^{-37}$ erg cm^3,[14] and $k_0 = 1.36 \times 10^8$ cm^{-1}.[24] With $S = \frac{5}{2}$ this gives $v_{ub} = 1.1 \times 10^6$ cm/sec. If we take ρ_s to be down from its upper bound by a factor of 4, then we may estimate that v is down from v_{ub} by a factor of 2, so that $v \approx 5.5 \times 10^5$ cm/sec. With this value, and the value $\chi \approx 2 \times 10^{-3}$ emu/mole for MnCu (0.97%),[14] or $\chi \approx 1.6 \times 10^{-4}$ emu/cm^3, we obtain $\rho_s \approx 1.3 \times 10^{-7}$ erg/cm. If we take this value to be appropriate to $\rho_{s\perp}$ in Eq. (5.17), then $\rho_{s\perp}k_c^2 \approx 2K_\perp$ for $k_c \approx 5.7 \times 10^4$ cm^{-1}. Since only values of k above $\sim 10^6$ cm^{-1} can be observed, this means that Eq. (5.17) is inappropriate for the interpretation of a neutron scattering measurement. Indeed, for such large values of k we probably have

$$(\rho_{s\perp}/\chi_\perp)k^2 >> (2K_\perp/\chi_\perp) + (m_\parallel/\chi_\perp)^2 \ ,$$

so that Eq. (5.16) yields

$$\omega_\pm \approx \gamma(H - m_\parallel/\chi_\perp) \pm v_\perp k \ , \quad (5.20)$$

where

$$v_\perp \equiv \gamma(\rho_{s\perp}/\chi_\perp)^{1/2} \ . \quad (5.21)$$

For $H = 0$, $v_\perp = 5 \times 10^5$ cm/sec, and $k = 10^6$ cm^{-1}, one finds $v_\perp k = 5 \times 10^{11}$ sec^{-1}; whereas, for $m_\parallel \approx 0.5$ emu and $\chi_\perp \approx 2 \times 10^{-4}$ emu, one finds $-\gamma m_\parallel/\chi_\perp \approx -5 \times 10^{10}$ sec^{-1}. Thus, in this case, the remanence shift should be about one-tenth of the smallest observable energy. (Note that ω_+ shifts down and ω_- shifts up.) In terms of frequency, we have $f_+ = (\omega_+/2\pi) \approx 70$ GHz. This is about 60 times as high as the 1.2-GHz ESR frequency observed by Monod and Berthier.[7] Since their line already had a noticeable linewidth, and since they observed an increased linewidth as the ESR frequency increased (caused by decreasing m_0), so large a value of frequency as 70 GHz might be accompanied by a large amount of line broadening. The splitting of ω_+ and ω_-, proportional to m_0, might be a useful signature in identifying what might otherwise be a hopelessly broadened line.

We now consider the position of the longitudinal

resonance line. Let us begin by making the unlikely assumption that $K_{\parallel} \approx K_{\perp}$, thus overestimating the value of $K_{\parallel}$. With $K_{\perp} \approx 210$ erg/cm^3 and $\chi_{\perp} \approx \chi_{\parallel} \approx 2 \times 10^{-4}$ esu, Eq. (5.6) gives that $f_{af} \equiv \omega_{af}/2\pi \approx 4.4$ GHz. At such a frequency, we would expect the line to be broadened. If, in addition, $\rho_{s\parallel} \approx \rho_{s\perp}$, then $v_{\parallel} \approx v_{\perp}$. Hence, for observable values of k, neutron scattering would measure

$$\omega \approx v_{\parallel} k + \omega_{af}^2/2v_{\parallel}k \ ,$$

the second term being about a twentieth of the first term for $k = 10^6$ cm^{-1}.

However, it is far more likely that $K_{\parallel} \approx 0$ (otherwise the system would possess some element of biaxial symmetry, and there is currently no evidence that typical spin-glasses possess such symmetry). In this case $\omega_{af} \approx 0$ and thus $\omega \approx v_{\parallel}k$. At $k = 10^6$ cm^{-1}, this corresponds to an energy of 0.3 meV, or a frequency of 80 GHz. Our observations about the broadening of the transverse spin wave probably apply here also, despite the different symmetry; in other words, broadening may be significant. Note that this line should be unaffected by H, and should lie between the two transverse waves at $v_{\perp}k \pm \gamma(H - m_{\parallel}/\chi_{\perp})$. It is worth noting that, if $K_{\parallel} = 0$, the longitudinal spin wave is a true hydrodynamic mode, with $\omega \to 0$ as $k \to 0$.

We close this discussion with some considerations on the possible effects of remanence and anisotropy on the specific heat C_m. In an earlier section we noted that, for no remanence or anisotropy, $C_m \propto T^3$ at low T, due to the hydrodynamic modes with linear dispersion. With remanence and anisotropy, the transverse modes develop a gap Δ, and thus they contribute a term to C_m which varies as $\exp(-\Delta/T)$ at low T. For large remanence, where these modes are observable by ESR,[7] they probably lie lower in energy than the localized modes, and thus dominate C_m. Since $\Delta \propto m_0^{-1}$ for large m_0, the low-temperature C_m should show a dependence on m_0. To our knowledge, no measurements of C_m for $m_0 \neq 0$ have been performed.

VI. STATIC TRANSVERSE RESPONSE

If the free energy given by Eq. (4.6) is correct, then it is possible to obtain the magnetization induced by a small magnetic field applied transverse to the direction of field cooling. Performing such an experiment (to obtain the *effective* value of the transverse susceptibility) should provide an additional constraint on the parameters of the system. We will assume that $\hat{z}$ is the remanence direction and we will take $\vec{H} = \hat{x}H_x + \hat{z}H_z$, where H_x will be assumed so small that $\hat{n}_{\perp}$ is also small. The equilibrium conditions are obtained by requiring that $dm_{\alpha}/dt = 0$, $dn_{\alpha}/dt = 0$, $d\theta/dt = 0$.

The equations for dm_x/dt, dm_z/dt, dn_x/dt, and $d\theta/dt$ yield $\delta m_z = 0$, $m_y = 0$, $n_y = 0$, and $\theta = 0$. The equations for dm_y/dt and dn_y/dt take the form

$$0 = dm_y/dt = -\gamma(H_z - m_{\parallel}/\chi_{\perp})m_x - \gamma(2K_{\perp} + m_{\parallel}^2/\chi_{\perp})n_x + \gamma m_{\parallel}H_x \ , \quad (6.1)$$

$$0 = dn_y/dt = -\gamma m_x/\chi_{\perp} - \gamma(H_z - m_{\parallel}/\chi_{\perp})n_x + \gamma H_x \ . \quad (6.2)$$

Their solution is

$$n_x = (2m_{\parallel}/\chi_{\perp} - H)(2m_{\parallel}/\chi_{\perp} - H + 2K_{\perp}/m_{\parallel})^{-1}(m_x/m_{\parallel}) \ , \quad (6.3)$$

where

$$m_x = \frac{\chi_{\perp} + m_{\parallel}^2/K_{\perp} - m_{\parallel}\chi_{\perp}H_z/2K_{\perp}}{1 + m_{\parallel}H_z/K_{\perp} - \chi_{\perp}H_z^2/2K_{\perp}} H_x \ . \quad (6.4)$$

In the absence of anisotropy ($K_{\perp} \to 0$) these yield $m_x \to m_{\parallel}H_x/H_z$, $n_x \to H_x/H_z$, as expected. In the limit as $H_z \to 0$, we have

$$m_x \to (\chi_{\perp} + m_0^2/K_{\perp})H_x \ . \quad (6.5)$$

Thus, for $H_z = 0$, the effective transverse susceptibility is given by

$$\tilde{\chi}_{\perp} \equiv \partial m_x/\partial H_x = \chi_{\perp} + m_0^2/K_{\perp} \ . \quad (6.6)$$

We believe it is likely that $\chi_{\perp} \approx \chi_{\parallel}$. Kouvel's studies on CuMn (see, e.g., Fig. 3 of Ref. 5) indicate that the high-field transverse susceptibility ($\partial m_x/\partial H_x$) is the same as the high-field longitudinal susceptibility ($\partial m_z/\partial H_z$). This point can be tested, for small transverse fields by direct measurement of m_0, $\tilde{\chi}_{\perp}$, $\chi_{\parallel}$, and the zero-field resonance frequency. The theory involves the four parameters m_0, $\chi_{\parallel}$, $\chi_{\perp}$, and $K_{\perp}$, and the four measurements can serve to determine them. For finite H_z, Eq. (6.4) can provide a check on the theory.

VII. SOME OBSERVATIONS ON THE NATURE OF THE SPIN-GLASS STATE

There is a great deal of evidence that spin-glasses possess both uniaxial anisotropy (the usual kind) and unidirectional anisotropy (associated with the direction of the remanence).[5,6] If we may assume that the domain walls are determined only by the uniaxial anisotropy energy, then we may employ the following result for the domain wall thickness[25]

$$Na \approx (k_B T_c/K_{\perp}a)^{1/2} \ .$$

Here a is the characteristic separation of magnetic ions and T_c is the transition temperature. For CuMn (1.35%) we take $a \approx 10^{-7}$ cm, $K_{\perp} \approx 210$ erg/cm^3, and $T_c \approx 15$ K, and we obtain $Na \approx 10^{-5}$ cm $= 10^3$ Å. Since domains themselves must be much larger than

the domain wall thickness, we conclude that domains must be much larger than this dimension. This is consistent with the work discussed in Sec. II.

The evidence for macroscopic domains leaves one somewhat perplexed, because it invalidates the magnetization cloud picture, based on the Néel model of ferromagnetic grains.[26,27] This picture has the advantage that it can explain a large number of the properties of the TRM and IRM, as functions of field H, temperature T, and time t. Despite the evidence against the presence of small weakly interacting magnetization clouds, it would be attractive if certain aspects of this picture could be retained. In this regard, note that computer experiments on Ising spin-glasses, which have no spin waves, reproduce most of the observed qualitative properties of the remanence.[28–30] Thus, for Heisenberg spin-glasses, it would appear that details of the spin-wave spectrum are not particularly relevant for the unusual remanence behavior of such systems. Rather, it would make sense if the "nearly identical subregions containing ~ 20 spins,"[14] which may also appear for Ising spin-glasses, were candidates, within the framework of the Néel model, for the magnetization clouds which may be employed to explain the remanence behavior of spin-glasses. However, this remains to be seen.

It should be noted that such subregions exist only because the system is disordered (or, to be more precise, it has the complex spin-glass order). There is other evidence, of an experimental nature, that the property of disorder, rather than noncollinearity or "frustration" (or both) is the key to the unusual remanence behavior of spin-glasses. (By "frustration" one refers to the fact that the molecular fields set up by the neighbors of a given spin do not all point in the same direction, thus leaving the spin with some uncertainty as to the direction in which it should point. See Toulouse.[31]) We refer now to the work of Kouvel and Kasper on the magnetic properties of the disordered face-centered-cubic alloy $(Ni_{1-x}Fe_x)_3Mn$.

For $x \lesssim 0.4$, high-temperature-susceptibility data[32] give $\theta > 0$, whereas $\theta < 0$ for $x \gtrsim 0.4$. In addition, for $x \lesssim 0.5$, the alloys possess remanence and a displaced hysteresis loop,[32] but elastic neutron scattering for $x = 0$ gives no evidence for periodic long-range order.[33] All this is consistent with the spin-glass state. For $x \gtrsim 0.5$, the alloys possess no remanence,[33] and elastic neutron scattering indicates a periodic long-range order.[34] This is consistent with an antiferromagnetic state. However, x-ray diffraction does not reveal the tetragonal distortion expected for a collinear antiferromagnetic structure, but it is consistent with a noncollinear tetrahedrally coordinated (TC) spin structure of cubic symmetry (with the spins pointing along different $\langle 111 \rangle$ directions).[34] We will accept this latter interpretation. Thus, as the parameter x is increased, the system goes from the spin-glass state to the TC state. On the one hand, both of these states possess noncollinearity and frustration; on the other hand, one state is periodic whereas the other possesses the complex spin-glass order. Only this latter state possesses an unusual magnetic behavior.

In closing, the reader's attention should be brought to the early "exchange-anisotropy" model of Kouvel,[35] which qualitatively explains the hysteresis curves and torque measurements for materials we would now call spin-glasses. Although the underlying microscopic picture presented there may not be accurate, the model itself makes a number of qualitatively correct predictions, and for this reason the model deserves more attention than it has received.

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