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Macroscopic dynamical theory for spin-glasses above T_g

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We argue that, due to magnetic-field- and random-anisotropy-induced order, spin-glasses at "high" frequencies have the same number of macroscopic variables above as below T_g (i.e., magnetization, spin triad, and anisotropy triad); spin-glasses behave like paramagnets only as $T/T_g \rightarrow \infty$. A macroscopic theory is constructed and applied to ESR. One longitudinal and two transverse modes, all with $\omega \sim H$, are predicted. However, the longitudinal and the non-Larmor-like transverse modes are perhaps so broad as to be unobservable. The Larmor-like transverse mode can exhibit an effective g-value shift, if the induced variables are not strongly damped.

One of the more unyielding problems in the area of spin-glasses (SG's) is the anomalous electron spin resonance (ESR) line observed for temperatures T above the susceptibility cusp T_g. There is both an anomalously large shift $\Delta\omega$ and width Γ (where $\Delta\omega \equiv \omega - \gamma H$, with ω the center of the resonance, γ the gyromagnetic ratio, and H the static applied field). Two recent works, one theoretical and one experimental, have helped focus on the nature of the problem. Levy, Morgan-Pond, and Raghavan[1] have computed $\Delta\omega$ and Γ for SG's, assuming that for $T > T_g$ they have the same macroscopic symmetry as paramagnets, so that the magnetization $\vec{m}$ is the only macroscopic variable. Comparison with the data of the UCLA group (Mozurkewich, Elliot, Hardiman, and Orbach[2]) shows good agreement for $T > 2T_g$, but for $T < 2T_g$ the predicted shift and width are smaller than observed.

The non-negligible size of the shift is a strong indication that the system has a more complex symmetry than an ordinary paramagnet. [Some other systems where this is true are liquid ^{3}He (Ref. 3), solid ^{3}He (Ref. 4), and SG's for $T < T_g$.[5,6]] For this reason we have examined some simple Heisenberg spin systems which, like SG's, have frustrated and disordered exchange bonds. A system of three classical spins subject to the Hamiltonian

$$\mathscr{H} = J(\vec{S}_1 \cdot \vec{S}_2 + \vec{S}_1 \cdot \vec{S}_3 + \alpha \vec{S}_2 \cdot \vec{S}_3) - \gamma \vec{H} \cdot (\vec{S}_1 + \vec{S}_2 + \vec{S}_3)$$

is sufficiently complex to illustrate the more complex symmetry of a spin-glass. For $J > 0$, $\alpha = 1$, and $H = 0$, this system is said to be frustrated, because there is no way for $\vec{S}_3$ to satisfy the conflicting information it gets if $\vec{S}_1$ is up and $\vec{S}_2$ is down. At low temperatures, the mean-field solution gives spontaneous ordering in a plane, with the spins at 120° angles to one another. At high temperatures, and $H \neq 0$, each spin satisfies $S = (C/T)H$, where the Curie constant C is independent of J or H. Indeed, above the transition temperature T_c, all the spins have the same local susceptibility χ. Letting $0 < \alpha \neq 1$ simulates, in addition to frustration, the effects of disorder. Above T_c, defined by $1 + \alpha r_c - 2r_c^2 = 0$, where $r \equiv (CJ/\gamma T)$, the mean-field solution yields $S_2 = S_3$ and

$$S_1/S_2 = \chi_1/\chi_2 = [1 - r(2-\alpha)](1-r)^{-1} \quad .$$

Evidently the local susceptibilities are not all the same.[7] Indeed, if $\alpha = 1 + \epsilon$, $\epsilon \ll 1$, then $(S_1/S_2) \rightarrow -2$ as $T \rightarrow T_g^+$, so S_1 can align oppositely to the other spins.

Clearly this system has at least as many dynamical variables as a two-sublattice ferrimagnet. This latter has the property that its magnetization $\vec{m}$ can change both by an overall rotation $d\vec{\theta}_\perp$ about an axis normal to $\vec{m}$ and by internal polarization $d\vec{m}_{\rm pol}$. Thus, for two-sublattice ferrimagnets, our simple three-spin system, and *(a fortieri)* spin-glasses, the macroscopic dynamical variables include both $\vec{m}$ and $d\vec{\theta}_\perp$. (Note, however, that because the local susceptibilities are all equal as $T \rightarrow \infty$, $d\vec{\theta}_\perp$ must become a redundant variable as $T \rightarrow \infty$.)

If we now include a weak, random, microscopic anisotropy, the spins will tip slightly away from $\vec{H}$, and the system will thus be three dimensional [as for a ferromagnet with random anisotropy (FRA)[8]]. Hence, the system must be described macroscopically by $\vec{m}$ and an orthonormal triad $(\hat{n},\hat{p},\hat{q})$ (called the spin triad[9]) which changes on rotations by the three-dimensional angle $d\vec{\theta}$. Moreover, the anisotropy is specified by an orthonormal triad $(\hat{N},\hat{P},\hat{Q})$ (called the anisotropy triad[9]), which changes on rotations by the three-dimensional angle $d\vec{\Theta}$. The anisotropy torque $\vec{\Gamma}$ is determined by the relative orientation of the spin and anisotropy triads, being zero when they coincide. In equilibrium, $\vec{m}$, $\hat{n}$, and $\hat{N}$ align with $\vec{H}$, and the spin and anisotropy triads are aligned.

In what follows we will derive the equations of motion for this system, and apply them to ESR. It turns out that there is a crucial relaxation time U_2 associated with $d\vec{\theta}$. Only for $\omega U_2 \gg 1$ do the new degrees of freedom manifest themselves (for $\omega U_2 \ll 1$ the new degrees of freedom relax to values determined by the instantaneous field); they then provide a natural framework for understanding the shifts observed in transverse ESR. As will be seen, we interpret the experiments to indicate that U_2 is strongly dependent on the applied field, with $\omega U_2 \ll 1$ in the high-field limit.

The procedure for obtaining the macroscopic equations is relatively standard.[9] One begins with the differential of the energy density (including the Zeeman interaction)

$$d\epsilon = T\,dS + (\vec{h} - \vec{H}) \cdot d\vec{m} + \vec{\lambda} \cdot d\vec{\theta} - \vec{\Gamma} \cdot d\vec{\psi} \quad , \qquad (1)$$

where S is the entropy density, $\vec{h}$ the internal field, $\vec{\lambda}$ a field conjugate to $d\vec{\theta}$, and $d\psi_\alpha \equiv d\theta_\alpha - R_{\alpha\beta} d\Theta_\beta$ ($R_{\alpha\beta}$ is the rotation taking the anisotropy triad into the spin triad). The requirements that $dS = 0$ and $d\epsilon = -\vec{H} \cdot d\vec{m}$ under rotations of all of the spins (so that $d\vec{m} = d\vec{\theta} \times \vec{m}$) yields $\vec{\lambda} = -\vec{m} \times \vec{h}$. One next writes down the equations of motion

$$\partial\epsilon/\partial t+\partial_i j_i^\epsilon=0,\ \ \partial S/\partial t+\partial_i j_i^S=R>0\ ,\quad \partial m_\alpha/\partial t=\gamma(\vec{m}\times\vec{H})_\alpha+\gamma\Gamma_\alpha+J_\alpha,\ \ \partial\theta_\alpha/\partial t=\omega_\alpha\ ,\quad \partial\Theta_\alpha/\partial t=\Omega_\alpha\ , \tag{2}$$

and requires that they reproduce the thermodynamics of Eq. (1) at all times. This leads, with $\Gamma'_\alpha\equiv\Gamma_\alpha-\lambda_\alpha=\Gamma_\alpha+(\vec{m}\times\vec{h})_\alpha$, to

$$0\le TR=-\partial_i(j_i^\epsilon-Tj_i^S)-j_i^S\partial_i T-J_\alpha(h_\alpha-H_\alpha)+\Gamma'_\alpha[\omega_\alpha-\gamma(h_\alpha-H_\alpha)]-(\Gamma_\beta R_{\alpha\beta})\Omega_\alpha\ . \tag{3}$$

This is almost identical to the case for $T<T_g$,[9] except that we have not yet specified $\vec{h}$ or $\vec{\Gamma}$, which differ from their $T<T_g$ forms. We now express the unknown thermodynamic fluxes j_i^ϵ, j_i^S, J_α, ω_α, and Ω_α, in terms of the thermodynamic forces $\vec{h}-\vec{H}$, $\vec{\Gamma}'$, and $\vec{\Gamma}\cdot\vec{R}$, subject to the Onsager symmetry principle and the symmetry of SG's for $T>T_g$. This yields

$$\begin{aligned}
&j_i^\epsilon=Tj_i^S,\ \ j_i^S=-(\kappa/T)\partial_i T\ ,\\
&J_\alpha=-\gamma D_{\alpha\beta}(h_\beta-H_\beta)+\gamma G[\vec{m}\times(\vec{\Gamma}\cdot\vec{R})]_\alpha+\gamma G'(\vec{m}\times\vec{\Gamma}')_\alpha\ ,\\
&\omega_\alpha-\gamma(h_\alpha-H_\alpha)-\gamma C(\vec{m}\times\vec{\Gamma}')_\alpha=\gamma E_{\alpha\beta}\Gamma'_\beta+\gamma E'_{\alpha\beta}(\vec{\Gamma}\cdot\vec{R})_\beta-\gamma G'[\vec{m}\times(\vec{h}-\vec{H})]_\alpha\ ,\\
&\Omega_\alpha=-\gamma F_{\alpha\beta}(\vec{\Gamma}\cdot\vec{R})_\beta-\gamma E'_{\beta\alpha}\Gamma'_\beta+\gamma G[\vec{m}\times(\vec{h}-\vec{H})]_\alpha\ .
\end{aligned} \tag{4}$$

These equations differ from those for $T<T_g$ because the system is no longer isotropic, so that terms which were scalars (e.g., D) are now tensors (e.g., $D_{\alpha\beta}$), and vector terms which were previously neglected are now included.[9] All of Eqs. (4) have been written so that the right-hand sides represent dissipative terms. We will not discuss the stability conditions which the dissipative coefficients must satisfy. Note the term $\gamma C(\vec{m}\times\vec{\Gamma}')$, which drives $\partial\theta_\alpha/\partial t$. Typically, it is not significant for $T<T_g$, where $C\approx m_s^{-2}$ (m_s being the saturation magnetization), but it cannot be neglected for $T>T_g$ if we are to ensure the correct (i.e., paramagnetic) high-temperature limit. In that case we have $C\to m^{-2}$ as $T/T_g\to\infty$, so that $\vec{m}_\perp=\dot{\vec{\theta}}_\perp\times\vec{m}$ for the transverse components of $\vec{m}$ and $\vec{\theta}$. Note that C can be obtained in terms of commutators of the microscopic operators for $d\vec{\theta}$.[10]

To complete the theory we now need only $\vec{h}$ and $\vec{\Gamma}$. The latter quantity can be obtained as in the case of the ferromagnet with random anisotropy, by rotating $\vec{m}$ and the spin triad away from equilibrium:[8]

$$\Gamma_\gamma=K_2(\hat{m}\times\hat{N})_\gamma(\hat{m}\cdot\hat{N})+\tfrac{1}{2}K_1[-2(\hat{m}\times\hat{N})_\gamma(\hat{m}\cdot\hat{N})+(\hat{m}\times\hat{N})_\gamma R_{\alpha\alpha}+(\hat{m}\cdot\hat{N})\epsilon_{\alpha\beta\gamma}R_{\alpha\beta}]\ . \tag{5}$$

This result utilizes our earlier assumption that $\vec{m}$ and $\hat{N}$ are collinear in equilibrium. Furthermore, one can show that it is not changed, to first order in deviations from equilibrium, if the unrotated system has a nonzero $\vec{\delta m}_{\rm pol}$. For $\vec{h}$, we assume that

$$\vec{h}=\chi_\parallel^{-1}\hat{n}(\vec{m}\cdot\hat{n})+\chi_\perp^{-1}[\vec{m}-\hat{n}(\vec{m}\cdot\hat{n})]\ . \tag{6}$$

To ensure that $\vec{m}$, $\hat{n}$, and $\vec{H}$ are parallel in equilibrium, we require that $\chi_\parallel\ge\chi_\perp$. Note that the anisotropy is field induced, so we expect that $\chi_\parallel-\chi_\perp\propto\chi_\parallel(m/m_s)^2$. Therefore, for small oscillations, as needed in ESR studies, we must employ the differential susceptibilities. For small deviations from equilibrium, this gives

$$\delta\vec{h}=\chi_\perp^{-1}\delta\vec{m}+\Delta\chi^{-1}(m\delta\hat{n}+\hat{n}\delta m)\ , \tag{7}$$

where $\Delta\chi^{-1}\equiv\chi_\parallel^{-1}-\chi_\perp^{-1}$, $\tilde{\chi}_\parallel^{-1}\equiv\partial(m/\chi_\parallel)/\partial m$, and $\hat{n}$ is considered to point along $\vec{H}$. Note that $m=\vec{m}\cdot\hat{m}$, $\delta m=\delta\vec{m}\cdot\hat{m}$. Thus,

$$\delta\vec{h}_\perp=\chi_\perp^{-1}\delta\vec{m}_\perp+\Delta\chi^{-1}\delta\vec{\theta}_\perp\times\vec{m},\ \ \delta h_\parallel=\tilde{\chi}_\parallel^{-1}\delta m\ . \tag{8}$$

It is straightforward to determine the ESR frequencies, assuming fixed anisotropy, and neglecting dissipation. For the longitudinal variables we have

$$\partial m_\parallel/\partial t=-\gamma K_1\delta\theta_\parallel,\ \ \partial\theta_\parallel/\partial t=\gamma\tilde{\chi}_\parallel^{-1}\delta m_\parallel\ , \tag{9}$$

so that

$$\omega_l=\gamma(K_1/\tilde{\chi}_\parallel)^{1/2}\ . \tag{10}$$

Note that, in terms of field, temperature, and microscopic anisotropy (D), we expect that K_1, $K_2\propto D^2H^2/T^3$, and $\tilde{\chi}_\parallel^{-1}\propto T$, so $\omega_l\propto\gamma(D/T)H$.[11] Thus, the longitudinal resonance will appear to have a g factor down by the (small) ratio D/T. (The temperature dependence is certainly more complex than T^{-1}.)

For the transverse variables we have (with $K_\perp=K_2+K_1/2$)

$$\begin{aligned}
&\partial\vec{m}_\perp/\partial t=\gamma\delta\vec{m}_\perp\times\vec{H}-\gamma K_\perp\delta\vec{\theta}_\perp\ ,\\
&\partial\vec{\theta}_\perp/\partial t=\gamma\chi_\perp^{-1}\delta\vec{m}_\perp+\gamma\Delta\chi^{-1}\delta\vec{\theta}_\perp\times\vec{m}\\
&\qquad+\gamma C\vec{m}\times[-K_\perp\delta\vec{\theta}_\perp\\
&\qquad\qquad+\Delta\chi^{-1}\vec{m}\times(\delta\vec{\theta}_\perp\times\vec{m}-\delta\vec{m}_\perp)]\ ,
\end{aligned} \tag{11}$$

so that the normal-mode frequencies are

$$\omega_\pm=(\gamma/2)\{(H+\alpha m)\pm[(H-\alpha m)^2+4K_\perp/\tilde{\chi}_\perp]^{1/2}\}\ , \tag{12}$$

where $\alpha\equiv K_\perp C+\Delta\chi^{-1}(1-Cm^2)$ and $\tilde{\chi}_\perp^{-1}=\chi_\perp^{-1}+\Delta\chi^{-1}Cm^2$. For $H>>\alpha m,\ (K_\perp/\tilde{\chi}_\perp)^{1/2}$, (12) yields

$$\begin{aligned}
&\omega_+\approx\gamma[H+(K_\perp/\tilde{\chi}_\perp)(H-\alpha m)^{-1}]\ ,\\
&\omega_-\approx\gamma[\alpha m-(K_\perp/\tilde{\chi}_\perp)(H-\alpha m)^{-1}]\ ,
\end{aligned} \tag{13}$$

$$\Delta\omega_+\equiv\omega_+-\gamma H=\omega_A^2\gamma^{-1}(H-\alpha m)^{-1},\ \ \omega_A^2\equiv\gamma^2K_\perp/\tilde{\chi}_\perp\ . \tag{14}$$

Since $K_\perp\propto H^2$, (14) predicts the Larmor-like ω_+ mode to posses an altered g factor.[12] More important, in principle, is the presence of the non-Larmor-like ω_- mode, whose observation would be a strong indication that $\delta\vec{m}_\perp$ and $\delta\vec{\theta}_\perp$ can behave like independent variables.

We now consider the effect of dissipation on the ESR lines. This requires consideration of the regime where the anisotropy is fixed, so $\Omega_\alpha \approx 0$. This permits us to neglect $F_{\alpha\beta}$, $E'_{\alpha\beta}$, and G. However, $D_{\alpha\beta}$, $E_{\alpha\beta}$, and G' cannot be neglected. The first describes ordinary T_1 and T_2 processes $[D_{\alpha\beta}=D_\parallel \hat{m}_\alpha \hat{m}_\beta + D_\perp(\delta_{\alpha\beta}-\hat{m}_\alpha\hat{m}_\beta)$, where $D_\parallel \propto T_1^{-1}$, $D_\perp \propto T_2^{-1}]$; the second describes relaxation of the spin triad due to its own disequilibrium, and the third describes cross relaxation of $\vec{m}$ due to disequilibrium of the spin triad, and vice versa. For simplicity, we will neglect the cross-relaxation terms. Adding the damping terms to (9), and assuming they are small, we find that

$$\Gamma_l = -\mathrm{Im}(\omega_l) \approx \tfrac{1}{2}\gamma(T_1^{-1}+U_1^{-1}) \ , \tag{15}$$

where $T_1^{-1}\propto D_\parallel$ and $U_1^{-1}\propto E_\parallel$ are associated with relaxation of $m_\parallel$ and $\theta_\parallel$. Since ω_l may be rather small, the damping may be relatively large, and thus the longitudinal mode may be difficult to observe.

Adding the relaxation terms to (11), for small damping we obtain

$$\Gamma_- = -\mathrm{Im}(\omega_-) \approx U_2^{-1} + \frac{\omega_A^2}{\gamma^2(H-\alpha m)^2}(T_2^{-1}-U_2^{-1}) \ . \tag{16}$$

where $T_2^{-1}\propto D_\perp$ and $U_2^{-1}\propto E_\perp$ are associated with relaxation of $\vec{m}_\perp$ and $\vec{\theta}_\perp$. Since ω_- varies as H (and thus may not be very large), the damping indicated in (16) may be relatively large, and thus the ω_- mode also may be difficult to observe. (In addition, the ω_- mode probably has only a small component of net magnetization, giving it a weak spectral weight.)

To make contact with data on the ω_+ mode, we note that one can write the transverse solutions, including dissipation, in the form

$$\omega = \gamma H - iT_2^{-1} + \frac{\omega_A^2}{\omega - \gamma\alpha m + iU_2^{-1}} \ . \tag{17}$$

For the ω_+ mode, which is usually well defined (i.e., $\omega >> T_2^{-1}$), and satisfies $\omega >> \alpha m$, (17) yields, with $\tilde{U}_2^{-1} \equiv U_2^{-1} - T_2^{-1}$,

$$\Delta\omega_+ \approx \frac{\omega_A^2\omega}{\omega^2+\tilde{U}_2^{-2}} ,$$
$$\Gamma_+ = -\mathrm{Im}(\omega_+) \approx T_2^{-1} + \frac{\omega_A^2\tilde{U}_2^{-1}}{\omega^2+\tilde{U}_2^{-2}} \ . \tag{18}$$

To compare with the actual data, we must remember that ω depends on the vaule of H; therefore, ω_A and U_2^{-1} change when ω changes. Thus, for the 1–2 GHz regime of Ref. 2, (18) is consistent with the observed apparent g-value shift, if $\omega\tilde{U}_2 >> 1$ in that regime. On the other hand, to be consistent with the 9.6-GHz data (which is taken at much larger values of H), one must have $\omega\tilde{U}_2 << 1$. This implies that U_2^{-1} has a strongly nonlinear dependence on H. (It is customary to assume that T_2 is independent of H.) Detailed study of the full frequency regime from 1 to 10 GHz would permit ω_A and U_2^{-1} to be determined, as functions of H and T.

To summarize, we have worked out a number of implications of magnetic-field– and random-anisotropy–induced order in spin-glasses, for $T > T_g$. This theory differs from two formally similar (but not identical) theories[12,13] in that its choice of macroscopic variables is motivated by a more specific microscopic picture of the SG state for $T > T_g$. It also differs from Ref. 1 in that it includes additional dynamical variables (in particular, for ESR it includes $d\vec{\theta}$). We find that these extra variables are fully manifested only for $\omega U_2 >> 1$, where they lead to what can be interpreted as a g-value shift in the ESR spectrum. A complete experimental study of both the shifts and widths, as a function of T and H, would permit a parametrization of the two most important quantities in the theory: ω_A and U_2.

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