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Dynamic theory of ferromagnetic—to—spin-glass transition

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We study the interaction between spin waves and magnetic two-level systems (TLS) in a ferromagnet with frustration. The coupling between the magnons and the TLS is due to the modulation of the angle between spins, by the rearrangement of the spins directions when the system tunnels from one metastable state to another. The magnon-TLS interaction causes an instability of the ferromagnetic phase with decreasing temperature due to the appearance of a soft mode. We associate this instability with the disappearance of long-range ferromagnetic order observed in reentrant ferromagnets.

I. INTRODUCTION

Recently much interest has been devoted to the phenomenon of reentrant ferromagnetism, that is, of systems which exhibit a ferromagnetic—to—spin-glass or paramagnetic transition on cooling.[1–4] This transition has been well characterized recently through a scaling analysis of the magnetization as a function of temperature and applied magnetic field.[3] The scaling hypothesis is obeyed for this kind of transition and allows for a determination of the critical exponents and critical temperature.[3]

Reentrant ferromagnetism has been observed in dilute ferromagnets with a concentration of magnetic atoms near the percolation concentration[3,5] and in concentrated ferromagnets with a high proportion of magnetic components.[1,2,4] The common feature of these materials is that they have additional magnetic excitations, besides the characteristic spin waves of ferromagnets. These additional modes can be observed on the specific heat where they give origin to a large linear term at low temperatures,[6] on the neutron scattering cross section associated with an excess scattering at low frequencies,[4] and on the electrical resistivity where they provide an extra mechanism to scatter the conduction electrons in metallic ferromagnets.[5] Besides, ferromagnetic resonance[5] and neutron scattering[7] give enough evidence for the freezing of these additional modes at sufficiently low temperatures.

In order to understand the phenomenon of reentrant ferromagnetism and the nature of the ferromagnetic instability on cooling, we describe the additional magnetic modes in these materials as two-level systems (TLS) and study the coupling of the spin waves to these extra modes. Owing to the random nature of the systems we are interested in the ensemble of TLS is characterized by a distribution of parameters.

We adopt of the point of view of Anderson, Halperin, and Varma[8] and look at the classical potential energy of the system in a space in which each spin is specified by a set of two angles. We assume that in this $2N$-dimensional configuration space, the classical energy has, in a given direction, local minima which are associated with the metastable ferromagnetic states characterized by distinct spin configurations. Many of these states are accessible to one another through quantum-mechanical tunneling or thermal activation and most probably the transition between them involves the rearrangement in the directions of a small number of spins.[9] In each of the metastable states the spin waves are well-defined excitations. We shall restrict our attention to pairs of adjacent, accessible minima in configuration space, the double-well potentials, which constitute the TLS excitations.[8]

The theory developed in this paper generalizes the model of Anderson, Halperin, and Varma for spin-glasses[8] for the case of reentrant ferromagnets. In both theories the exact nature of the modes described as TLS remains unspecified.

One may think that the origin of magnetic TLS and of the "glassy" topology of the classical energy in configuration space may be quite different in diluted and concentrated reentrant ferromagnets. In fact in the dilute systems we could tentatively identify the TLS modes with the degrees of freedom of the finite clusters which coexist with the infinite cluster near the percolation concentration.[5]

In concentrated reentrant ferromagnets the excess modes and the glassy structure of the classical magnetic energy arise from the competition between ferromagnetic and antiferromagnetic interactions, which is a common characteristic of ferromagnets with reentrant behavior. Indeed, this competition is the essential ingredient for frustration,[10] and as shown by Villain[11] and others[9,12] magnetic systems with frustration exhibit two-level-system types of excitations associated with distinct spin configurations. The existence of these excitations has been demonstrated on several experiments on spin-glasses.[13,14] We point out that even in the dilute reentrant ferromagnets we cannot discard the relevance of frustration as is obvious from the fact that these systems become spin-glasses at low temperatures. Also the similarity on the behavior of dilute and concentrated reentrant ferromagnets[5,4] supports the point of view that frustration is the common ingredient responsible for the glassy behavior of both type of systems.

The essential difference from the dynamical point of view between spin-glasses and the systems we are considering lies in the fact that in spin-glasses TLS excitations are sufficient to explain most of their magnetic properties without being necessary to invoke the existence of spin waves[13,15] which, although predicted theoretically,[16] have not been undisputedly observed.[16] On the other hand, in reentrant ferromagnets the TLS play a complementary role, the relevant excitations being the spin waves. We show, however, that the coupling of the spin waves to the magnetic TLS softens the former excitations and eventually causes an instability of the ferromagnetic phase with decreasing temperature due to the appearance of a soft spin-wave mode. We associate this instability with the disappearance of long-range ferromagnetic order on cooling which characterizes reentrant ferromagnets. It is remarkable that a softening of the spin waves has been directly observed by neutron scattering on these materials,[1] as the temperature is decreased and the spin-glass phase is approached.

II. HAMILTONIAN

The Hamiltonian which describes magnons, two-level systems, and their coupling is given by

$$H=\sum_k \epsilon_k a_k^\dagger a_k+\frac{\epsilon}{2}\sigma^z+\frac{\Delta}{2}\sigma^x+K\sigma^z S^x . \quad (1)$$

A derivation of this Hamiltonian is given in Appendix A. The first term in (1) describes magnons with energy $\epsilon_k=\Delta_0+D_0k^2$, where Δ_0 is a gap, D_0 the spin-wave stiffness, and k the wave vector of the excitations. The $a_k^\dagger, a_k$ are Holstein-Primakoff boson operators. The next two terms in (1) refer to the TLS; ϵ represents the energy difference between metastable ferromagnetic states lying on opposite sides of the double-well potentials and which are characterized by distinct metastable equilibrium directions for the spins. Δ is the quantum-mechanical tunneling frequency which is related to the transition rate from one metastable ferromagnetic state to another. The last term couples the TLS and the spin waves. The coupling arises due to a modulation of the angle between spins, or of the angle they make with a given direction when the system "moves" from one metastable state to another (see Appendix A). In the Hamiltonian above the Pauli matrices, σ^i refers to the TLS, and S^x represents the x component of the spin $\vec{S}$. Higher-order terms which give origin to Raman-type magnon-TLS processes have been negelcted in (1).

On the basis which diagonalizes the energy of the defect, the Hamiltonian (1) can be rewritten as

$$H=\sum_k \epsilon_k a_k^\dagger a_k+(E/2)\sigma^z+(D\sigma^z+M\sigma^x)S^x , \quad (2)$$

where $D=K\epsilon/E$, $M=K\Delta/E$, and $E=(\epsilon^2+\Delta^2)^{1/2}$.

III. FERROMAGNETIC INSTABILITY

Recently we have studied[17] a Hamiltonian such as (2) to take into account the coupling between spin waves and structural defects in a ferromagnetic glass. The effect of the magnon-TLS interaction on the spin wave of frequency ω can be described by a shift $\Delta\omega$ in the energy and a damping Γ of these excitations.[17] One finds from (2), for the total energy shift and damping,[17]

$$\Delta\omega_{\rm res}=\frac{NM^2S}{N_0}\frac{\tanh(E/2k_BT)}{E}\left[\frac{\omega(\omega-\omega_r)}{(\omega-\omega_r)^2+\tau_2^{-2}}+\frac{\omega(\omega+\omega_r)}{(\omega+\omega_r)^2+\tau_2^{-2}}-2\right] , \quad (3)$$

$$\Delta\omega_{\rm rel}=-\frac{4ND^2S}{N_0}\chi_0^{zz}\frac{1}{\omega^2\tau_1^2+1} , \quad (4)$$

$$\Gamma_{\rm res}=\frac{NM^2S}{2N_0}\frac{\tanh(E/2k_BT)}{E}\left[\frac{\omega\tau_2}{(\omega-\omega_r)^2\tau_2^2+1}+\frac{\omega\tau_2}{(\omega+\omega_r)^2\tau_2^2+1}\right] , \quad (5)$$

$$\Gamma_{\rm rel}=\frac{4ND^2S}{N_0}\chi_0^{zz}\frac{\omega\tau_1}{\omega^2\tau_1^2+1} , \quad (6)$$

where

$$\chi_0^{zz}=\frac{1}{4k_BT}\,\mathrm{sech}^2(E/2k_BT)\ .$$

In the equations above $\hbar\omega_r=E$, the energy splitting of a TLS. N and N_0 are the numbers per unit volume of TLS and spins, respectively, and τ_1 and τ_2 are the longitudinal and transverse relaxation time of the TLS.[18] In reentrant ferromagnets TLS relaxation may occur by direct magnon emission and absorption. The longitudinal relaxation time due to these types of processes has been obtained in Ref. 17. The transverse relaxation time τ_2 arises due to the interaction between TLS, which for the systems studied here may be mediated by spin waves.[17] The subscripts "rel" and "res" in the equations above denote the relaxation and resonant contribution for the spin-wave energy shift and damping.[18] In the theory of magnetic resonance these contributions are known as longitudinal and transverse or as the slow and fast relaxation mechanisms.[19]

Since we are dealing with a random system, the expressions above must be averaged over the distribution of energy splittings of the TLS. Let us first consider the resonant contribution. For this purpose we introduce the function $n_0(E)$, which gives the density of states of TLS per unit volume and unit energy and assume this function to be a constant n_0, for E between 0 and $E_{\max}$, and 0 otherwise. This is consistent with the linear temperature dependence of the specific heat observed at low temperatures on the spin-glass phase and which is due to the TLS excitations.[8] We then find for the resonant contribution to the energy shift of the spin wave of frequency ω (Ref. 17)

$$\Delta\omega_{\mathrm{res}}=\frac{2n_0M^2S}{N_0}\left[\frac{\omega^2\tau_2^2}{\omega^2\tau_2^2+1}[\mathrm{Re}\psi(\tfrac{1}{2}+z)-\psi(\tfrac{1}{2})]+\frac{\omega\tau_2}{\omega^2\tau_2^2+1}\,\mathrm{Im}\psi(\tfrac{1}{2}+z)-\left[\ln\frac{E_{\max}}{2\pi k_BT}-\psi(\tfrac{1}{2})\right]\right],\qquad(7)$$

where $\psi(z)$ is the digamma function[20] and $z=\hbar(\tau_2^{-1}-i\omega)/2\pi k_BT$. We are taking the couplings D and M as constants independent of the energy splitting E as is usually assumed in the theory of glasses.[18]

The renormalized spin-wave energy E_k can be written as

$$E_k=\epsilon_k+\Sigma(\hbar\omega=\epsilon_k)\qquad(8)$$

with $\Sigma(\omega)\equiv\Delta\omega_{\mathrm{res}}(\omega)$. Let us make this equation self-consistent; that is, we write

$$E_k=\epsilon_k+\Sigma(\hbar\omega=E_k)\ ,\qquad(9)$$

which amounts to sum an infinite series of diagrams representing single scattering resonant processes of a magnon by a TLS.[21] The critical temperature for the appearance of a soft mode is determined by the condition $E_k\equiv 0$, that is

$$\epsilon_k+\Sigma(0)=0\ ,$$

which gives

$$\Delta_0+D_0k^2=G\left[\ln\frac{E_{\max}}{2\pi k_BT}-\psi(\tfrac{1}{2})\right],\qquad(10)$$

where $G=2n_0M^2S/N_0$ and $\psi(\frac{1}{2})$ is a negative constant. When the temperature is decreased, the first mode to soften is the $k=0$ mode for which the critical temperature is given by

$$k_BT_0=\frac{\alpha E_{\max}}{2\pi}\exp\frac{-\Delta_0}{G}\ ,\qquad(11)$$

where $\alpha=7.121$. The effect of a magnetic field H is to decrease the critical temperature. We find

$$k_BT_0(H)=k_BT_0(H=0)\exp\frac{-g\mu_BHS}{G}\ .\qquad(12)$$

For small fields ($g\mu_BH\ll G$) the effect is linear on the applied magnetic field. Notice that large fields ($g\mu_BH>G$) can push the transition to infinitesimally small temperatures.[22] The damping of the spin waves due to the resonant contribution is given by Eq. (17) of Ref. 17.

Let us make some numerical estimates of the quantities calculated above. Firstly, we assume that $(\alpha E_{\max}/2\pi)$ is of the order of k_BT_C, where T_C is the Curie temperature of the material. This is in agreement with the arguments of Anderson *et al.*[8] regarding the scale on which $n_0(E)$ should be constant to yield a linear specific heat. Also for $\Delta_0=0$ we should get $T_0=T_C$. In glasses $E_{\max}$ is of the order of the glass transition temperature.[18] The quantity n_0 can be obtained from the linear term of the specific heat on the spin-glass phase and turns out to be of order $n_0\cong 10^{35}\ \mathrm{erg}^{-1}\,\mathrm{cm}^{-3}$ (Ref. 7) compared to $10^{33}\ \mathrm{erg}^{-1}\,\mathrm{cm}^{-3}$ due to structural TLS in glasses.[18] We make the assumption that the density of states of magnetic TLS is the same on both ferromagnetic and spin-glass phases. This is expected for a quenched system and from the nature of magnetic TLS on systems with frustration; that is, once the sample is prepared the number of TLS is fixed independently if the system is in a ferromagnetic or spin-glass state. The gap Δ_0 in soft ferromagnets is mainly due to dipolar interactions and can be approximated by $\Delta_0 \sim g\mu_B(4\pi/3)M_S$. It is typically of order 10^{-5} eV for the systems we are interested in.[23] Taking $N_0=10^{22}$ cm^{-3} and $S=1$ as for iron, the only quantity which remains to be estimated in order to obtain T_0 is the coupling constant M. This is a difficult task since M is associated with anisotropic interactions (see Appendix A) which are not well known in these systems. We shall proceed in the opposite direction and from the known values of T_0 we estimate the order of magnitude of the coupling constant M necessary to account for the ferromagnetic instability. From experimental results we take $T_0 \sim T_C/e$ (Refs. 3 and 4) which requires that $\Delta_0/G \sim 1$ so that M turns out to be of order 10^{-3} eV. This value of M implies strong anisotropic interactions although this is not necessary if the TLS involves a reasonably large number of spins since, as shown in Appendix A, the constant K appearing in (1) scales with the total number of spins in the TLS. We point out however that large anisotropies may exist in these materials, as has been shown recently.[24] This order of magnitude for M is consistent with a Dzyloshinskii-Moriya or pseudodipolar anisotropic interaction.[24] In metallic ferromagnetic glasses with reentrant behavior, these interactions may also be due to the presence of metalloid atoms which give origin to anisotropic superexchange interactions between the transition-metal ions.[25]

The relaxation contribution to the shift also decreases the spin-wave energy as can be seen from Eq. (4). In principle this type of process can also lead to an instability of the ferromagnetic phase. The relaxation shift is more difficult to calculate since in general the longitudinal relaxation time of the TLS depends on the energy of the TLS.[17] There is strong experimental evidence from the study of spin-glasses[14] that in systems with frustration TLS tunneling is mostly performed by thermal activation. In this case the relaxation contribution to the energy shift is given by[17]

$$\Delta\omega_{\rm rel} = -\frac{ND^2S}{N_0k_BT}\int P(V)\frac{dV}{\omega^2\tau^2(V)+1}, \quad (13)$$

where $\tau=\tau_0\exp(V/k_BT)$ with τ_0 a constant and V an activation energy with probability distribution $P(V)$. In particular, for a constant distribution of energy barriers from 0 to $V_{\max}$ one gets from (13)

$$\Delta\omega_{\rm rel} = -\frac{D^2SN}{N_0}\left[\frac{1}{k_BT}+\frac{1}{2V_{\max}}\ln\frac{\omega^2\tau_0^2+1}{\omega^2\tau_{\max}^2+1}\right], \quad (14)$$

where $\tau_{\max}$ is the value of τ for $V=V_{\max}$. From this expression we obtain a critical temperature for the mode of $k=0$, with the same arguments used above, $k_BT_0=(D^2Sc/\Delta_0)$, where $c=N/N_0$ is the concentration of defects. This temperature turns out to be roughly of the same order of magnitude of T_0 obtained from the resonant shift, if we use $D\approx 10^{-3}$ eV and $c=0.1$ (see Appendix A). In this case, however, we get a different field dependence for the critical temperature.

IV. FERROMAGNETIC RESONANCE IN REENTRANT FERROMAGNETS

Expressions (3)–(6) and (13), averaged over the distribution of TLS parameters, give the line shift and linewidth of the modes of frequency ω excited in a ferromagnetic resonance experiment in ferromagnets with frustration.[17] Owing to the relaxation contribution, we expect the linewidths to exhibit maxima as a function of temperature. These maxima are in fact observed.[5,23] They are due in part to the shape of the distribution of energy barriers and reflect the freezing of the magnetic TLS on the time scale of the experiment.[18] As shown below in Eq. (15), these linewidths are proportional to the average imaginary part of the dynamic longitudinal susceptibility of the TLS. The physical origin of these maxima is quite simple. The ensemble of magnetic TLS provides a relaxation channel for the ferromagnetic medium. When the temperature decreases the TLS starts to freeze, increasing the lifetime of the ferromagnetic modes excited by the radio frequency, and consequently the linewidth decreases giving origin to a maximum. On the high-temperature side of the maximum there is a kind of "motional" narrowing of the line due to the increasingly rapid relaxation of the TLS. This is essentially the explanation for the linewidth maxima proposed by Coles *et al.*[5] for these systems. The model developed by Sarkissian[26] however considers only the resonant contribution since he assumes an isotropic interaction between spins. Furthermore, he took $\omega_r=0$.

The expression for the relaxation contribution to the linewidth is, in the case of thermally activated processes, given by[17]

$$\Gamma_{\rm rel} = \frac{ND^2S}{N_0k_BT}\int P(V)\frac{\omega\tau}{\omega^2\tau^2+1}dV \quad (15)$$

with $\tau=\tau_0\exp(V/k_BT)$ as before. In the case when the width of the distribution $P(V)$ is much larger than k_BT, a useful approximation can be obtained for the above integral.[27] We find

$$\Gamma_{\rm rel} \cong \frac{ND^2S}{N_0}P(\tilde{V}), \quad (16)$$

where $\tilde{V}=-k_BT\ln(\omega\tau_0)$. The above expression allows for direct determination of the distribution of energy barriers $P(V)$ from the measured linewidths.

Notice that for a constant distribution $P(V)$ the linewidth is temperature independent.

Recently Bhagat *et al.*[28] have shown that in a large class of systems exhibiting TLS freezing, including ferromagnets, the linewidth as a function of temperature can be described by the following expression, in a certain temperature range above the freezing temperature:

$$\Gamma=\Gamma_1 \exp(-T/T_A) , \tag{17}$$

where Γ_1 and T_A are functions of the concentration of magnetic ions. It is easy to see that if we assume a normalized distribution of energy barriers

$$P(V)=\begin{cases} f(V), & V<V_1 \\ (1/V_0)\exp(-V/V_0), & V>V_1 \end{cases} \tag{18}$$

where $f(V)$ is a function joining smoothly to the value of $P(V)$ for $V=V_1$, and with the use of Eq. (16) for $\Gamma_{\rm rel}$ we obtain expression (17) for the linewidth, whenever

$$k_B T > V_1 / |\ln(\omega\tau_0)| ,$$

and $\Gamma_1 = cD^2S/V_0$. Using $\omega/2\pi=10$ GHz, $\tau_0=10^{-13}$ sec, and the value of $T_A=26$ K obtained by Spano and Bhagat[23] for a typical reentrant ferromagnetic glass, we get $V_0 \sim 10^{-2}$ eV. Since Γ_1 is in this case of order 10^3 Oe (Ref. 23) we estimate cD^2 above to be of order 10^{-8} eV^2. Thus expression (17) for the linewidth is a consequence of the exponential tail of the distribution of energy barriers for large activation energies.

The linewidth due to the resonant contribution is given by

$$\Gamma_{\rm res}=(\pi n_0 M^2/N_0)\hbar\omega/2k_B T$$

for $\omega\tau_2 \gg 1$ and $\hbar\omega \ll 2k_B T$.[17] Our estimate of M from the critical temperature obtained above, $M\sim 10^{-3}$ eV is consistent with the observed linewidths in magnetic resonance[23,5] in reentrant ferromagnets since for $\omega/2\pi=10$ GHz and on the range of temperatures investigated, the resonant contribution turns out to be negligible compared to the relaxation contribution to these linewidths.

The shift in the field for resonance due to the resonant contribution can be obtained from Eq. (7). It is given, in the cases $\omega\tau_2 \gg 1$ and $\hbar\omega \ll k_B T$, by[17]

$$H_{\rm res}(T)-H_{\rm res}(T_0)=-\frac{2n_0 M^2 S}{\hbar\gamma N_0}\ln\left[\frac{T}{T_0}\right] , \tag{19}$$

where $H_{\rm res}(T_0)$ is the field for resonance at an arbitrary reference temperature T_0.

The dynamic shifts, that is, these contributions to the shift which vanish with frequency, are positive and generally increase with decreasing temperature for both relaxation and resonant contribution. These dynamic shifts consequently cause a decrease on the field for resonance as the system is cooled. They are given by expressions (10) and (11) of Ref. 17 averaged over the distribution of energy splittings. The static shifts, on the other hand, are negative and should enter the resonant conditions for the ferromagnetic spins as effective anisotropy fields.

In a ferromagnetic glass with frustration one has coexistence of magnetic and structural relaxation and as we have shown in Ref. 17 structural relaxation may also give origin to maxima on the linewidths as a function of temperature. However, in the case of the glasses studied by Spano and Bhagat[23] the anomalies on the linewidth as a function of temperature appear, for the range of temperature investigated, only on the alloys which show a transition from a ferromagnetic to a spin-glass state at low temperatures. These results strongly suggest that the observed anomalous linewidths in these materials are due to magnetic relaxation and are given in Ref. 16 with an appropriate distribution of energy barriers. It is interesting to speculate whether structural and magnetic TLS can in fact be treated in reentrant ferromagnetic glasses as distinct excitations.

V. CONCLUSIONS

We have calculated the effects on the spin-wave propagation of the interaction between these modes and magnetic TLS in ferromagnets with frustration. The coupling between magnetic TLS and magnons arises due to a modulation of the orientation of the spins, by the rearrangement of the spin directions when the system tunnels from one metastable state to another. We have shown that the magnon-TLS interaction causes an instability of the ferromagnetic phase with decreasing temperature due to a softening of the former excitations. Both resonant and relaxation contributions may drive this instability. They predict, however, distinct field dependence for the critical temperature and could be easily identified experimentally.[19]

We have associated the instability at T_0 with the disappearance of long-range ferromagnetic order on cooling observed in systems with competing interactions and which is accompanied by a softening of the spin waves as directly confirmed by neutron scattering experiments. The instability involves an interplay between dipolar forces which give origin to the gap Δ_0 and frustration effects represented by a density of states n_0 of magnetic TLS. The theory presented above shows that a system with a small anisotropy gap and a large density of defects may not sustain long-range ferromagnetic order which, however, can be stabilized by an external magnetic field. In this respect it is worthwhile mentioning some recent experimental results which show that ferromagnetic order may set in on spin-glasses under sufficiently strong applied magnetic fields.[29]

Recently a ferromagnetic—to—spin-glass transition has been associated with the temperature of the maximum of the ferromagnetic resonance (FMR) linewidth.[30] As we have shown this maximum is due to the relaxation contribution, Eq. (15), and occurs at a temperature T_f such that $\omega\tau(T_f)\cong 1$.[18] It should be clear from the results above that the freezing temperature T_f and the temperature of the ferromagnetic instability T_0 are independent quantities such that ferromagnetic order may subsist below T_f (ferro-plus-cluster glass regime, $T_0 < T_f$) or alternatively freezing may occur below T_0.[2] Also the FMR linewidth is proportional to the imaginary part of the dynamic susceptibility of the TLS and not to the real part from which the spin-glass transition temperature is obtained.

Finally, it is important for our analysis that the spin-wave modes whose energy vanish and which are responsible for the collapse of the ferromagnetic phase remain well defined down to T_0. The damping of the spin waves in the limit $\omega \rightarrow 0$ can be easily obtained for the resonant contribution. It is given by $\Gamma_{\text{res}} = (\pi n_0 M^2 / 2N_0 k_B T)\hbar\omega$ and at T_0, with the numerical values used before, one finds $\Gamma_{\text{res}}/\hbar\omega \sim 10^{-3}$ and consequently well-defined magnons in this limit. The relaxation contribution to the damping depends strongly on the relaxation mechanism of the TLS. For thermally activated TLS relaxation, with a constant distribution of energy barriers, as used to obtain (14), we get $\Gamma_{\text{rel}} = (ND^2 S \tau_{\max} / \hbar N_0 V_{\max})\hbar\omega$. At T_0 with $V_{\max} = 2k_B T_0$ (Ref. 18) and the other parameters as used before one gets $\Gamma_{\text{rel}}/\hbar\omega \sim 10^{-2}$ confirming the validity of our approach.

The spin waves which are probed by neutron scattering have much larger energies than the modes which cause the ferromagnetic instability. The behavior of the damping of these excitations can be inferred directly from the FMR linewidth and in fact we expect it exhibits a maximum at the freezing temperature of the TLS due to the relaxation contribution.

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APPENDIX A

Let us consider a system of spins which interact through a Hamiltonian of the general form[31]

$$H = \sum_{\substack{i,j \\ \alpha,\beta}} J_{ij}^{\alpha\beta} S_i^\alpha S_j^\beta , \tag{A1}$$

where i and j run through the spins and $\alpha,\beta = x,y,z$. This Hamiltonian includes the usual isotropic Heisenberg terms and also anisotropic interactions like pseudodipolar, dipolar, etc.[31] In a ferromagnetic system with competing interactions the directions of the spins in the metastable equilibrium are noncollinear due to frustration,[32] and in this case it is useful to rewrite (A1) in a local set of frames in which the local Z direction is taken as the local metastable equilibrium direction of the spin. For simplicity we consider the case of planar spins for which the direction of a given magnetic moment is specified by just one angle[17] θ_i which the spin makes with a fixed direction in the laboratory. After the change of axis we can distinguish three types of terms in the transformed Hamiltonian:

(i) terms of the type

$$\sum_{i,j} J_{ij} g(\theta_i,\theta_j) S_i^z S_j^z ,$$

(ii) terms like

$$\sum_{i,j} J_{ij} [f(\theta_i,\theta_j) S_i^z S_j^x + z(\theta_i,\theta_j) S_i^z S_j^y] ,$$

and finally,

(iii) terms like

$$\sum_{i,j} J_{ij} [h(\theta_i,\theta_j) S_i^x S_j^x + k(\theta_i,\theta_j) S_i^y S_j^y + l(\theta_i,\theta_j) S_i^x S_j^y] .$$

The S_i^α above refer to the components of the spins on the local frames, and for simplicity we took all $J^{\alpha\beta}$ to be equal. On these local frames it is reasonable to take $S_i^z = S$, and we adopt this approximation on the discussion below.

The first type of term determines essentially the energy of a given metastable state and together with contributions arising from local anisotropies give origin to the term $(\epsilon/2)\sigma^z$ in (1) which represents the difference in energy between the two accessible magnetic states lying on opposite sides of the double-well potentials. This difference in energy is associated with different sets of angles θ_i characterizing the local equilibrium directions of the spins in the two distinct metastable states. At this point we introduce the term $(\Delta/2)\sigma^x$ appearing in Hamiltonian (1) and which allows for quantum-mechanical tunneling from one magnetic metastable state to another. The theory can be easily generalized to include thermally activated transitions[33] as is done in the text.

The second type of term arising from (A1) gives origin to magnon-TLS interactions. The functions $f(\theta_i,\theta_j)$ and $z(\theta_i,\theta_j)$ have different values in the distinct metastable states. This difference is accounted for by the interaction terms $K^0S^0\sigma^zS^x$ and $GS^0\sigma^zS^y$, S^0 is the total spin associated with a TLS. Notice that S^0 has been included in the definition of the constant K appearing in Hamiltonian (1) which gives a false impression that (1) is not time-reversal invariant. In (1) we considered only the term proportional to S^x since taking into account the one proportional to S^y merely renormalizes the coupling constants D and M which are not determined from first principles anyway.

Finally, the third type of term gives origin to magnon-TLS interactions described by terms like $B\sigma^zS^xS^x$ or $C\sigma^zS^xS^y$, which are associated with indirect scattering of spin waves by the TLS (Raman processes). They involve two boson operators and shall not be considered here.

The spectrum of spin-wave excitations is obtained from (A1) in a given metastable ferromagnetic configuration. It may also include other contributions arising from local anisotropies. We assume that this spectrum which is described by the dispersion relation $\epsilon_k=\Delta_0+D_0k^2$, remains unchanged and well defined in the different metastable ferromagnetic states. This assumption is essentially the same made on the theory of glasses when one couples phonons to structural defects in the well-known tunneling model.[18]

With regard to the order of magnitude of the couplings D and M, we have shown above that K in Eq. (1) can be written as $K=K^0S^0$, where K^0 is related to an anisotropic exchange interaction and S^0 is the effective spin of a TLS. Let us assume that a TLS involves on the average a rearrangement on the relative orientations of ten spins so that we associate an effective spin $S^0=10S$ with a defect. This is reasonable since it is of the order of the number of nearest neighbors in a three-dimensional structure. In this case, for M and D to be of order 10^{-3} eV one requires $K^0\sim10^{-4}$ eV (with $S=1$ as for iron), which is of the order of a familiar pseudodipolar interaction.[34] The total number N of TLS is given by $N=n_0E_{\max}\sim10^{21}$ cm^{-3} which yields $c=0.1$ as used in the text.

A final remark concerning metastability should be made. When one does the transformation of the spin components to the set of local axis one finds that in the case of planar spins the isotropic Heisenberg exchange interaction J_{ij}^0 gives origin to a term of type (ii), namely[17]

$$\sum_{i,j} J_{ij}^0 \sin(\theta_i-\theta_j) S_i^z S_j^x \;.$$

What we mean by a metastable state is one for which

$$\sum_{j} J_{ij}^0 \sin(\theta_i-\theta_j)=0$$

for all i and that is the reason why effectively the isotropic Heisenberg interaction does not contribute to the coupling K^0.

APPENDIX B

In this Appendix we generalize a cluster model introduced by Sarkissian[26] to take into account anisotropic interactions of the Dzyaloshinskii-Moriya (DM) type between spins. In this model a ferromagnet with concentration of magnetic ions a little above the percolation threshold is viewed as consisting of an infinite island of ferromagnetic spins coexisting with small finite clusters. The infinite cluster has long-range ferromagnetic order and can sustain propagating spin waves. The finite clusters are treated as single entities and represented by two-level systems. This model is interesting on its own and is also useful in clarifying the one studied in the text, which we shall refer to as the glassy model.

The Hamiltonian which describes spin waves in an infinite ferromagnetic cluster interacting with aglomerates of spins represented by TLS is

$$H=\sum_k \epsilon_k a_k^\dagger a_k + J\vec{\sigma}\cdot\vec{S}+\vec{D}\cdot\vec{\sigma}\times\vec{S} \;, \tag{B1}$$

where the spins $\vec{S}$ belonging to the infinite cluster interact with the finite clusters through effectives isotropic exchange and anisotropic DM interactions. The symbols above have the same meaning as in the text.

This Hamiltonian can be studied by the dynamic reaction field method used before. The shift on the energy of the magnon $\Delta\omega$ and its damping Γ are given by

$$\Delta\omega=-\frac{8SN}{N_0}(J^2-D^2)\,\mathrm{Re}[\chi^x(0)-\chi^z(0)] + \frac{8SN}{N_0}[(J^2+D^2)\,\mathrm{Re}\chi_d^x(\omega)+D^2\,\mathrm{Re}\chi_d^z(\omega)] \;, \tag{B2}$$

$$\Gamma=\frac{8SN}{N_0}[(J^2+D^2)\,\mathrm{Im}\chi^x(\omega)+D^2\,\mathrm{Im}\chi^z(\omega)] \;,$$

where $\omega=\epsilon_k/\hbar$. N and N_0 are the number of clusters and of spins in the infinite cluster and $\chi^x(\omega)$

and $\chi^z(\omega)$ are the dynamical transverse and longitudinal susceptibilities of a TLS, respectively.[17] We also defined $\chi_d^\alpha(\omega)=\chi^\alpha(0)-\chi^\alpha(\omega)$, and for simplicity we took $\langle (D^x)^2\rangle=\langle (D^y)^2\rangle=\langle (D^z)^2\rangle=D^2$ where $\langle\ \rangle$ means a configuration average. The dynamical susceptibilities depend on the energy splitting E of the TLS which in this case is given by $E=JS^z$, that is the "Zeeman" energy of the finite cluster on the local molecular field produced by all other spins. The function $n_0(E)$ for the cluster model is given by the distribution of these local molecular fields while in the glassy model it is associated with the energy for the rearrangement of the metastable equilibrium directions of the spins.

The terms in square brackets on the shift represent static and dynamical contributions, respectively. For $J>D$ and $\chi^x(0)>\chi^z(0)$ the static shift is negative and an instability may occur for this cluster model at $k=0$. A linewidth maximum due to the relaxation contribution arising from $\mathrm{Im}\chi^z(\omega)$ also appears in this model.

For $D=0$ we have $\chi^z=\chi^x$, the relaxation contribution for the energy and damping disappears and we obtain essentially Sarkissian's result.[26] In this isotropic case there is no ferromagnetic instability since the static shift is zero, which is a consequence of Goldstone's theorem.[35] A more detailed study of the cluster model which consists in a particular representation of a magnetic two level system will appear in a future publication.[36]

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