Relaxation-Coupled Order-Parameter Oscillation in a Transverse Ising System

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A novel technique, which employs $\omega\chi'/\chi''$ of the longitudinal susceptibility in place of the shape function $\chi''/\omega\chi(0)$, reveals a coupling of the heavily overdamped soft mode to a relaxation mode in the transverse Ising ferromagnet $LiTbF_4$. Our analysis by exact Mori-Langevin equations suggests that this contribution to the central peak, which becomes temperature dependent only below T_c, arises from a dynamical coupling to the transverse polarization.

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Despite numerous experimental[1-4] and theoretical[5-9] studies the collective motion of transverse Ising systems still poses an open problem.[10] This situation also appears to be unsatisfactory with regard to the formal simplicity of the underlying transverse Ising model (TIM), which originally was proposed by de Gennes[5] to describe the order-parameter motion of hydrogen-bonded uniaxial ferroelectrics:

$$\mathcal{H}_{\rm TIM} = -\tfrac{1}{2}\sum_{i\neq j} J_{ij} S_z^i S_z^j - \Delta \sum_i S_x^i. \tag{1}$$

There, Δ/h constitutes the tunneling frequency of the protons in their double-well potentials, opposing the ordering effect of the interaction J_{ij}. For ferromagnets the transverse field arises from the crystal-field splitting of the ground-state doublet into two nonmagnetic singlets.[3]

For the limiting cases of a weak or a strong transverse field Δ (compared to $\langle J_{ij}^2\rangle^{1/2}$, i.e., the root mean square of the interaction), one simply finds pure relaxational or oscillating behavior of the polarization S_z, respectively. In the intermediate regime, $\Delta \approx \langle J_{ij}^2\rangle^{1/2}$, the excitations become collective which leads to a softening as predicted by the early theories[6] for $T \geq T_c$,

$$\omega_{\mathbf{q}} = (\Delta/h)\{1-[J(\mathbf{q})/\Delta]\langle S_x\rangle\}^{1/2}, \tag{2}$$

in accord with light- and neutron-scattering data, e.g., on ferroelectric potassium dihydrogen phosphate[1,2] and ferromagnetic $Li(Tb_x,Y_{1-x})F_4$.[4] However, the severe damping of the oscillation, $\Gamma_{\mathbf{q}} > \omega_{\mathbf{q}}$, obscured the true nature of the order-parameter dynamics, in particular whether the scattering intensity χ_{zz}''/ω for $\omega \to 0$ [central peak (CP)] simply results from the overdamped soft mode $\omega_{\mathbf{q}}$ or includes extra relaxation mechanisms.[11]

Three-pole approximations of the shape function $F(\omega) = \chi_{zz}''/\omega\chi_{zz}(0)$ indicated that the CP represents an intrinsic property of the TIM,[8] present at all temperatures above T_c. More recently, self-consistent solutions of kinetic equations confirmed this intrinsic three-pole structure of $F(\omega)$, the only exception being the order-parameter mode $S_z(q_\perp \to 0)$ in the presence of dipolar contributions to J_{ij}, for which a true soft mode, i.e., a two-pole shape of $F(\omega)$, has been predicted.[9,10] However, because of the approximate nature of both treatments, those results were considered to be highly speculative, especially close to T_c.[8-10]

This Letter communicates results on $\omega\chi_{zz}'/\chi_{zz}''$ measured along the tetragonal (z) axis of the ferromagnet $LiTbF_4$ ($T_c = 2.87$ K) at microwave frequencies between 0.6 and 33 GHz. The axial crystalline field lifts the $(2J+1)$-fold degeneracy of the free Tb^{3+} ion ($J=6$) giving rise to the $M_J = \pm J$ ground-state doublet. Since the excited levels are at least 170 K apart,[12] at low temperatures the effective spin is $S=\frac{1}{2}$. The tetragonal symmetry mixes a small amount of $M_J = \pm 2$ states to the ground-state doublet, which (i) reduces the g_z value from $g_J J = 18$ to $g_z = 17.85 \pm 0.1$,[12] but obviously does not change the Ising nature ($g_x = g_y = 0$), and (ii) splits the doublet into two (Γ_2) singlets separated by $\Delta = 1.40 \pm 0.14$ K.[12] This latter effect is exactly accounted for by a transverse field acting on the pseudospin S, $\mathcal{H}_T = -\Delta S_x$,[3,12] so that $LiTbF_4$ constitutes an excellent example for the TIM Hamiltonian [Eq. (1)], for which the ordering occurs via the (dominating dipolar[13]) spin-spin interaction $\langle J_{ij}^2\rangle^{1/2} S = 2.0 \pm 0.2$ K.[13]

Inelastic neutron scattering at 4 K revealed a single CP for $F(\omega)$ of width smaller than the instrumental resolution of 10 GHz.[4] The absence of excitonic sidebands around $\omega_{\mathbf{q}}$ has been tentatively assigned to the presence of long-lived ferromagnetic clusters. Our data also incorporate the dispersion part of $\chi_{zz}(\omega)$ and clearly demonstrate that the ordering TIM obeys neither a simple relaxational-type dynamics nor a pure soft mode, but an overdamped softened oscillation around $\omega_0 = \omega_{q=0}$ coupled to a Debye spectrum of width ϕ:

$$\chi_{zz}(\omega) = (\lambda/T)\Delta^2/[\omega_0^2 - \omega^2 + i\omega\gamma(\omega)], \tag{3}$$

$$\gamma(\omega) = \delta^2/(\phi + i\omega) + \Gamma. \tag{3a}$$

This three-peak structure was discovered for the soft acoustic phonon of Nb_3Sn ($T_c = 45$ K) by Shirane and Axe,[14] who ascribed $\gamma(\omega)$ to a coupling to the bath of thermal phonons. For the present TIM our discussion by means of Mori-Langevin equations of motion suggests that the $\gamma(\omega)$ is related to a dynamical coupling of S_y to the narrow ($\phi \ll \Gamma$) Debye spectrum of the polarization S_x associated with the transverse field. Thence, this contribution to the CP appears to be a general characteristic of the TIM, requiring no additional coupling mechanisms. Most interestingly, $\gamma(\omega)$ remains essentially independent of temperature throughout the paramagnetic regime, implying that the CP cannot be associated with long-living ferromagnetic clusters as suggested recently.[4] Moreover, we find the three-peak structure also immediately below T_c, with the coupling constant δ^2 increasing proportional to the square of the spontaneous moment $\langle S_z \rangle$. In a first-order approximation, we can associate this with the kinematical coupling between S_x and S_y mediated by the Larmor precession of **S** around the combined transverse and longitudinal local fields.

We determined the high-frequency dispersion and absorption from the differences between frequencies, $\Delta f/f = \eta\chi'/2$, and between quality factors, $\Delta Q/Q = \eta\chi''$, for filled and empty resonance circuits. The empty state was achieved in a sufficiently high longitudinal magnetic field ($\gtrsim 20$ kOe). Below 5 GHz we employed cavities containing helical or linear inner conductors, while at higher frequencies microwave cavities were used reaching resolutions up to 5×10^{-8}.[15] In all cases, the z axis of the $LiTbF_4$ was aligned parallel to one principal axis of the sample ellipsoid (demagnetization coefficient N) and to the direction of the alternating field. Because of difficulties in fixing the filling factor η in the various cavities to a reliable accuracy, we decided to eliminate η by extracting the ratio $R = f\chi'/\chi''$ from the Δf and ΔQ data. Depending on the magnitudes of Δf and ΔQ, the absolute values of R could be determined to between 0.5 and 3 GHz.

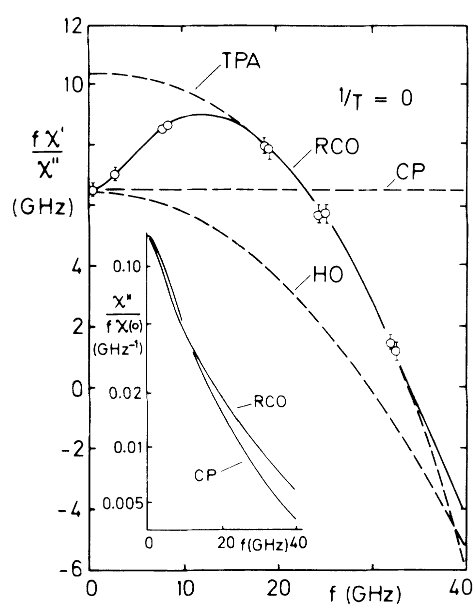

FIG. 1. Frequency dependence of $f\chi'/\chi''$ ratios at infinite temperature determined from extrapolations of the measured $1/T$ behavior (see Fig. 2) and compared to different models: central peak (CP), harmonic oscillator (HO), three-pole approximation (TPA), relaxation-coupled oscillator (RCO). Inset: Dynamic form factors accessible to scattering experiments.

The frequency dependence of R at infinite temperature is displayed in Fig. 1 and compared to different dynamical behaviors. All are special cases of the relaxation-coupled oscillator (RCO) for which Eq. (3) yields ($\omega = 2\pi f$)

$$\frac{\omega\chi'_{zz}}{\chi''_{zz}} = \frac{\omega_0^2 - \omega^2[1-\delta^2/(\phi^2+\omega^2)]}{\Gamma + \phi\delta^2/(\phi^2+\omega^2)]}. \tag{4}$$

ω_0 represents the $q_z/q \to 0$ limit of the electronic excitation, Eq. (2), appropriate to our experiment:

$$\omega_0^2 = (\Delta/\hbar)^2(\lambda/T)(\chi_0^{-1}+N). \tag{4a}$$

At high tempertures, the internal equilibrium susceptibility χ_0 assumes the Curie-Weiss law, $\chi_0 = \lambda/(T-3.5\ \mathrm{K})$, with $\lambda = 8.5$ K,[15] while below 8 K well-known logarithmic factors enter.[16] Figure 1 clearly illustrates the full accord between the data fitted by the RCO model, as well as the failure of the other basic dynamics: (i) $R = \Delta^2/\gamma(\omega=0)$ for Debye relaxation (CP); (ii) $R = (\Delta^2 - \omega^2)/\gamma(\omega=0)$ for harmonic oscillation, predicted by the early RPA approaches[6]; and (iii) $R(\Gamma=0)$, following from the continued-fraction expansion of χ''_{zz}/ω terminated at second order (three-pole approximation),[8] which has to be adjusted at high frequencies. A particular confirmation of the RCO model stems from the excellent agreement of our result, $\Delta/h = 29.4(1.0)$ GHz, with the spectroscopic (ESR) values 30(10) GHz for

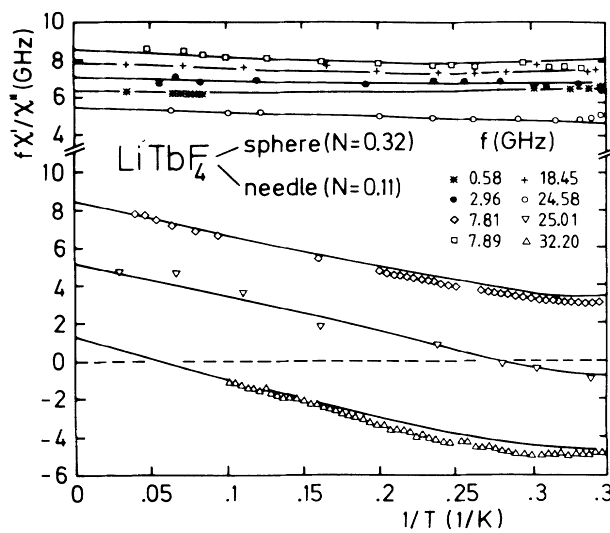

FIG. 2. Effect of temperature on $f\chi'/\chi''$ of two differently shaped samples above $1/T_c = 0.348\ \mathrm{K}^{-1}$. Solid lines were calculated by the RCO model based on the conventional softening of the oscillation frequency $\omega_0 = \Delta(\lambda/T\chi_0)^{1/2}$ and temperature-independent damping.

$LiTbF_4$ and 28.0(0.1) GHz for $Li(Tb_{0.01}Y_{0.99})F_4$.[12] The intrinsic resonance width, $\Gamma/2\pi=94(7)$ GHz, turns out to be much larger than Δ/h, than the width of the relaxation $\phi/2\pi=7.4(5)$, and than the coupling constant $\delta/2\pi=17.3(1.0)$ GHz. We note that $\Gamma/2\pi$ is close to the value $\delta\omega/2\pi=\gamma\delta H_{\rm ESR}\approx 120$ GHz following from ESR linewidths measured at high frequencies ($\omega/2\pi>280$ GHz) and $T\gg T_c$,[12] which seems to support the present analysis in terms of the RCO model.

A consequence of this overdamping, $\Gamma\gg\Delta$, is illustrated in the inset of Fig. 1. It can be seen that a high resolution is necessary in a scattering experiment in order to distinguish the RCO from the simple CP shape for $LiTbF_4$. As an important feature from Fig. 2, it emerges that at finite temperatures above T_c, not only is the RCO shape of $R(\omega)$ conserved, but also the values of the damping parameters Γ, ϕ, and δ. Thus the temperature behavior of $\chi_{zz}(\omega)$ is dictated by the softening of the electronic excitation ω_0 due to the diverging susceptibility χ_0.

For the ferromagnetic state, Fig. 3 displays a strong increase of the zeros in $R(\omega)$, indicating a growth of the excitation energy. This is not unexpected, since now the longitudinal field caused by the spontaneous magnetization adds to the transverse crystal field to speed up the precession of **S**. Analyzing $R(\omega)$, we found that the RCO model explains the shape observed below T_c fairly well. The fit of all data by Eq. (4), a portion of which is displayed in Fig. 3, yielded the following (i) for the equilibrium susceptibility, $\chi_0=0.1(1-T/T_c)^{-1.30(4)}$, which is close to the adiabatic susceptibility determined recently[17]; (ii) for the coupling constant $\delta^2(T)=\delta^2+(\delta_1\langle S_z\rangle)^2$ with $\delta_1/2\pi=80(10)$ GHz if we use $\langle S_z\rangle(T)$ from Als-Nielsen *et al.*[18]; and for the damping parameters Γ, δ, and ϕ, a common, weak temperature factor T/T_c.

In order to elucidate the physical origin of this hitherto neither experimentally nor theoretically observed RCO dynamics of the TIM, we start rather generally with the dynamic susceptibility,

$$\chi_{zz}(\omega)=(\lambda/T)\{[1-\omega(\omega\mathbf{C}+\mathbf{F}+i\mathbf{L})^{-1}]\mathbf{C}\}_{zz}. \quad (5)$$

The tensors are exactly defined within the Mori-Langevin theory[19]: the static correlation, $C_{\mu\nu}=(g_\mu^\dagger,g_\nu)$, the frequency matrix $F_{\mu\nu}=k_BT\langle[g_\mu^\dagger,g_\nu]\rangle/h$, and the Onsager-Casimir matrix $L_{\mu\nu}=(Q\dot g_\mu\,|\,M(\omega)\,|\,Q\dot g_\nu)$, where $Q=1-\sum_{\mu\nu}|g_\mu)(g_\nu|(C^{-1})_{\mu\nu}$ and $M(\omega)$ is the memory operator. The natural observables g_ν for the present TIM, decoupled from the lattice, are the three "adiabatic" components of **S**, $g_\nu=S_\nu-\mathcal{H}(\partial\langle S_\nu\rangle/\partial\beta)/(\mathcal{H}\,|\,\mathcal{H})$ ($\beta=1/k_BT$).[20] Without any approximation, one finds for the sum of frequency and Onsager matrices

$$\mathbf{F}+i\mathbf{L}=\begin{pmatrix} iL_{xx} & i(k_BT\langle S_z\rangle+L_{xy}) & 0\\ i(-k_BT\langle S_z\rangle+L_{yx}) & iL_{yy} & ik_BT\langle S_x\rangle\\ 0 & -ik_BT\langle S_x\rangle & 0\end{pmatrix}. \quad (6)$$

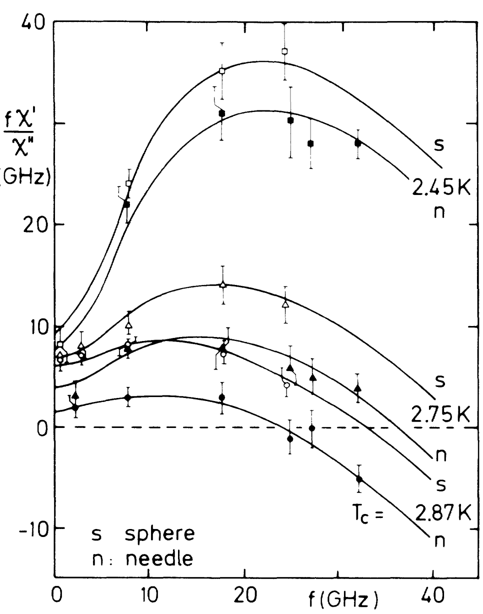

FIG. 3. Frequency dependence of $f\chi'/\chi''$ in the ferromagnetic regime. Solid lines correspond to the RCO model considering an additional coupling between oscillation of S_y and relaxation of S_x, being proportional to $\langle S_z\rangle^2$.

After insertion of this matrix into Eq. (5), a straightforward calculation yields just the RCO susceptibility supposed in Eq. (3), provided that **C** is diagonal. This is true above T_c, where $C_{xx}\approx C_{yy}=\frac14=C_\perp$, $C_{zz}=\chi_0/(4\lambda/T)$ (Ref. 7) and, moreover, $\langle S_x\rangle=\frac12\tanh(\frac12\beta\Delta)\approx C_\perp\beta\Delta$ for $LiTbF_4$.

Above T_c the physical messages obtained by this *Ansatz* are the following:

(i) The large (high frequency) damping of the Larmor precession of **S** around the transverse field, $\Gamma=L_{yy}/C_\perp$, arises solely from the strong decay of the S_y component due to the spin-spin interaction, $\mathcal{H}_{ss}=-\frac12\sum_{i\neq j}J_{ij}S_z^iS_z^j$, since the S_z component is neither changed by $\mathcal{H}_{ss}$ ($[\mathcal{H}_{ss},S_z]=0$) nor by the spin-lattice interaction, which can safely be ignored ($\Gamma_{sl}\leq 10$ Hz[21]).

(ii) The characteristic frequency of the relaxation is defined by $\phi=L_{xx}/C_\perp$, i.e., by the spin-spin decay rate of the S_x component. Comparing the relevant matrix elements, $L_{xx}\sim\langle S_yS_z\,|\,M(\omega)\,|\,S_yS_z\rangle$ to $L_{yy}\sim\langle S_xS_z\,|\,M(\omega)\times|\,S_xS_z\rangle$, one notes that the difference between both arises from the different action of $M(\omega)$ on S_xS_z and S_yS_z, which is plausible because of the presence of the transverse field ΔS_x in $M(\omega)$. A detailed evaluation of this effect apparently requires some theoretical effort, since the ordering interaction and the transverse field are of the same magnitude, so that there are not simple perturbation schemes.

(iii) The positive coupling between S_x and S_y, $\delta^2 = -L_{xy}L_{yx}/C_\perp^2$, arises from the imaginary part of **L** i.e., $L_{xy}(\omega) = iL''_{xy}(\omega) = L_{yx}(\omega)$ in our range of frequencies, since the other alternative, $L_{xy} = L'_{xy} = -L'_{yx}$, appears to be forbidden by time-reversal symmetry.[20] This interpretation is plausible if $L''_{xy}(\omega)$ exhibits a broad maximum within our ω range which then, according to the Kronig-Kramers relations, may be accompanied by a small value of $L_{xy}(\omega)$. Since L''_{xy} vanishes for ω tending both to zero and to infinity, the observed S_x-S_y coupling δ^2 is of purely dynamic origin.

We note that principally the RCO lineshape stems from the lack of any coupling to the z elements in the Onsager matrix **L**. This is an exact consequence of $\mathcal{H}_{\rm TIM}$, which does not provide a coupling of S_z to a dissipative force; hence no residual force for the damping of S_z exists, formally following from $Q\dot{S}_z \equiv 0$. We should also note, however, that the present assignment of the RCO parameters Γ, ϕ, and δ to L'_{yy}, L'_{xx}, and L''_{xy} is just the simplest one. Any other interpretation of the observed line shape would require a more detailed knowledge of the frequency behavior of the complex elements $L_{\alpha\beta}$ $(\alpha,\beta = x,z)$. Below T_c, the general *Ansatz*, Eq. (6), reproduces the observed enhancement, $\delta^2(T) = \delta^2 + (\delta_1\langle S_z\rangle)^2$, with, however, $\delta_1/2\pi = k_BT_c/h \approx 220$ GHz being significantly larger than the experimental value of 80 GHz.

In conclusion, the $\omega\chi'/\chi''$ data presented here demonstrate that the order-parameter dynamics of the TIM ferromagnet $LiTbF_4$ exhibits a relaxation-coupled oscillation, which was not expected from existing experimental and theoretical work. In the entire paramagnetic region, the coupling remains essentially independent of temperature, whereas below T_c it strongly increases. Our analysis in terms of exact Mori-Langevin equations suggests that the observed line shape arises from the TIM symmetry producing a dynamical coupling between the oscillating S_y and relaxing S_x components, the detailed features of which are not yet known.

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