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Longitudinal spin fluctuations in the nearly isotropic ferromagnet $CdCr_2Se_4$: Scaling behavior outside the critical region

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The uniform dynamics of nearly isotropic cubic ferromagnet $CdCr_2Se_4$ in the ordered phase is investigated. It is found that the longitudinal susceptibility behaves as $\chi \propto \omega^{-\rho}$ with $\rho \approx 0.28$. The low-frequency crossover to $\omega^{-\rho}$ dependence is induced by the internal magnetic field H_i. We attribute the observed anomalous behavior to the influence of dipolar forces and propose a description of $\chi(\omega, H_i)$ in terms of phenomenological scaling. [S0163-1829(99)50526-2]

It is known that the absorption of a uniform external magnetic field in a Heisenberg ferromagnet is absent. It stems from the fact that the operator of the total spin of the system commutes with the exchange Hamiltonian. The weak so-called relativistic interactions (anisotropy, dipolar forces, etc.) violate the total spin conservation law providing uniform relaxation.[1]

The uniform longitudinal susceptibility of the isotropic Heisenberg ferromagnet with dipolar forces below T_c has been studied in Ref. 2 within a framework of the linear spin-wave theory (LSWT), i.e., with only two-magnon intermediate states taken into account. At finite temperatures the susceptibility in zero internal field H_i demonstrated an infrared divergence (IRD) of the form $\chi(\omega \to 0) \propto iT/\omega$. At $H_i \neq 0$ one has a threshold for $\chi''(\omega)$ at $\hbar\omega = 2g\mu_B H_i$ (the minimal energy of creation of two magnons), and a ω^{-1} dependence at $2g\mu_B H_i \ll \hbar\omega \ll \omega_0 \langle S \rangle$. Here $\omega_0 = 4\pi(g\mu_B)^2 v_c^{-1}$ is the characteristic dipolar energy, $\langle S \rangle$ is the mean atomic spin, $g\mu_B$ and v_c are the effective magnetic moment and the volume per one magnetic atom, respectively.

The mentioned IRD originates from specific "weak" violation of the total spin conservation law by dipolar forces upon which the number of magnons in an elementary scattering event may change while the magnon spectrum remains gapless. Note that the ω^{-1} singularity is a nonphysical one since it leads to a nonzero value of the absorption function $\dot{Q}_\omega \propto \omega\chi''(\omega)$ at $\omega = 0$ and thus the sample would be heated by a dc field.

Meantime, ω^{-1} dependence has been obtained within the LSWT. The intermediate states with a larger number of excitations yield more singular contributions to $\chi(\omega)$. Hence, in order to describe the limit $\omega \to 0$, the summing of a series of divergent terms is needed. This problem is not solved yet;[3] however, it is natural to assume that the result will be a powerlike function of ω. Taking into account analytical properties of $\chi(\omega)$ one can suggest two variants of the behavior at $\omega \to 0$:

$$\chi(\omega \to 0) \propto \chi_0 + A(i/\omega)^\rho, \tag{1}$$

$$\chi(\omega \to 0) \propto \chi_0 - A_1(\omega/i)^{\rho_1}, \tag{2}$$

where χ_0 is the static susceptibility, and ρ, ρ_1 are real numbers. While the latter scenario corresponds to elimination of the IRD, the former one resembles the behavior of susceptibility at the critical point[4,5] and means instability of the system since it results in a divergence of the spin-spin correlator K at large times, $K(t \to \infty) \propto t^\rho$. Both these scenarios (except for the case $\rho_1 = 1$) should be regarded as anomalous because usually for generalized susceptibilities one could expect $\chi'(\omega \to 0) \approx \chi_0$ and $\chi''(\omega \to 0) \propto \omega$.

In this paper we present experimental evidence that isotropic ferromagnets below T_c behave according to the *first* scenario. More precisely: the *ideal* infinite isotropic ferromagnet at $H_i = 0$ would be in a "critical" state at any temperature. In *real* materials the anisotropy and the finite-size effects cut off this divergence.

We studied the low-frequency ($\omega/2\pi \sim 10^3 - 10^6$ Hz) uniform longitudinal susceptibility of a nearly isotropic cubic ferromagnet $CdCr_2Se_4$ ($T_c \approx 128$ K) in weak external magnetic fields using both the resonance and the phase methods.[6] The measurements have been performed mostly at $T = 82$ K, i.e., outside the critical region near T_c. At frequencies higher than the two-magnon threshold we observed a powerlike behavior of $\chi(\omega)$ with the exponent $\rho(T = 82\ \mathrm{K}) = 0.28 \pm 0.02$. This exponent is found to be slowly varying with T. We show that the characteristic frequency scale is determined by the internal magnetic field and identify the principal relaxation mechanism as the spin-wave one.[7] A description of $\chi(\omega, H)$ in scaling terms is also proposed. We argue that there exist other Bose systems with a "weak" violation of some conservation law which should exhibit an anomalous low-energy behavior.

We used a sample in the shape of a ring $\phi 5.4 \times \phi 2.0 \times 2.1$ mm^3. The plane of the ring coincided with the [111] plane of the crystal. Both the constant field H_e and the alternating one $h_0 \cos \omega t$ were applied along the ring. In such geometry the lines of magnetic field were closed, and the field was almost completely concentrated inside the sample. It allowed us to observe extremely narrow hysteresis with the coercive field $H_c \sim 5$ mOe. In order to avoid domain formation and to minimize nonlinearities [$\chi(h_0)$ dependence] we controlled the conditions $H_e > h_0 > H_c$.

Frequency dependence of both the imaginary and the real parts of the susceptibility as well as of their ratio χ''/χ' measured at $H_e = 8$, 16, and 32 mOe is shown in Fig. 1. The response is seen to be large, $\chi'_{max} \sim 10^2$. Furthermore, $\chi'(\omega)$

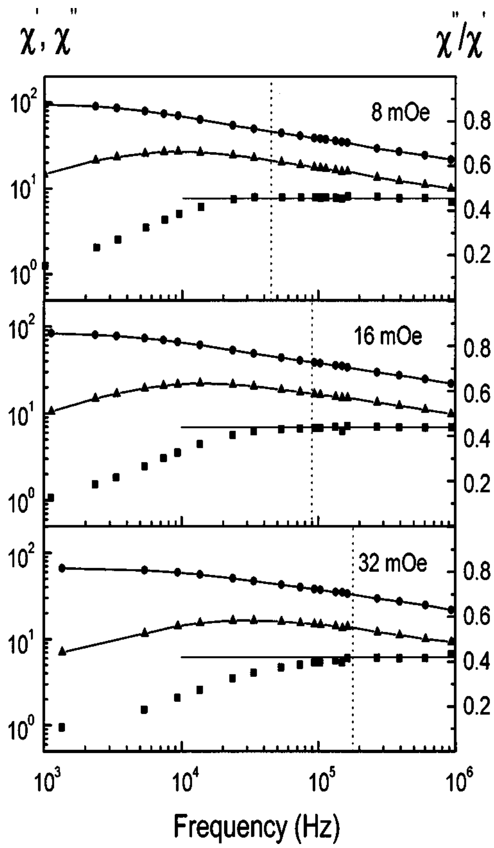

FIG. 1. Real (circles) and imaginary (triangles) parts of the longitudinal susceptibility and their ratio (squares) versus frequency measured at various values of the external field ($T=82$ K). At high enough frequencies the quantity χ''/χ' is seen to be constant which agrees with Eq. (1) and suggests the absence of a χ_0 term there.

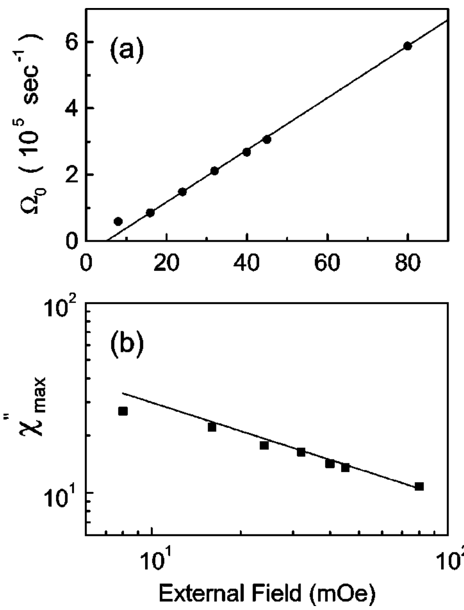

FIG. 2. Top panel: circles correspond to frequency of maximum in ω dependence of χ'' (which we associate with the characteristic frequency scale Ω_0) at various values of the external field. The line is the fit by Eq. (4). Bottom panel: the log-log plot for the maximal value of $\chi''(\omega)$ versus the external field (squares). The line is $H^{-1/2}$ dependence.

is a decreasing function while $\chi''(\omega)$ has a maximum in the considered frequency range. Note that the region $\omega/2\pi \sim 10^3-10^6$ Hz is usually associated with the asymptotic regime $\omega \to 0$ when one could expect $\chi' \approx \chi_0$ and $\chi'' \propto \omega$.

The most interesting feature of $\chi(\omega)$ is seen at (relatively) high frequencies. Figure 1 shows that within a wide frequency range the susceptibility demonstrates a powerlike behavior and can be well fitted by Eq. (1). The coefficient A in this expression is found to be *H independent*.

Equation (1) can be rewritten in the form

$$\chi(\omega) \propto \chi_0 + A|\omega|^{-\rho}[\cos \pi\rho/2 + i \operatorname{sgn} \omega \sin \pi\rho/2]. \quad (3)$$

The value of the exponent $\rho(T=82\ \mathrm{K}) = 0.28 \pm 0.02$ extracted from the slopes in ω dependences of both χ' and χ'' coincided with the one found from the ratio $\chi''/\chi' = \tan \pi\rho/2$. It means that the term χ_0 in Eq. (3) can be omitted.

Methodically, the frequency range in our experiment has been limited from above at $\omega/2\pi \sim 1$ MHz. We believe that the same $\omega^{-\rho}$ dependence of the susceptibility takes place at higher frequencies as well. On the other hand, the region of this behavior expands to low frequencies with the decrease of H_e. The dotted vertical lines in Fig. 1 mark the quantity $2g\mu_B H_e/h$, i.e., the two-magnon threshold neglecting the difference between external and internal fields. At $H_e = 32$ mOe the region of $\omega^{-\rho}$-dependence is limited from below by $\omega \gtrsim 2g\mu_B H_e/\hbar$ while at lower fields this region is somewhat wider. We attribute this to the fact that actually the powerlike behavior occurs at $\omega \gtrsim 2g\mu_B H_i/\hbar$, and in our experiment a small diminishing of the field in the sample $\Delta H \simeq 5$ mOe appears due to demagnetization and anisotropy. When H_e is large this diminishing is negligible while at $H_e = 8$ mOe it becomes significant.

Another peculiarity of susceptibility represented in Fig. 1 is the presence of a broad maximum for $\chi''(\omega)$. It is natural to associate the position of this maximum with the characteristic frequency scale Ω_0. In Fig. 2(a) we plotted the dependence of Ω_0 on external field. We see that Ω_0 is a *linear* function of H_e which can be written in the form

$$\hbar\Omega_0 \approx 0.45 g\mu_B(H_e - \Delta H), \quad (4)$$

where ΔH is the quantity defined above. Hence, the scale Ω_0 is induced by the *internal* magnetic field. Note that the point $\Omega_0(H_e = 8\ \mathrm{mOe})$ in Fig. 2(a) lies higher than the straight line. Apparently, at such weak fields the coercive ($H_c \sim 5$ mOe) and the nonlinear ($h_0 = 7$ mOe) effects already come into play.

The above consideration allows us to prove the principal statement of the present work. Indeed, we have shown that the low-frequency crossover to the $\omega^{-\rho}$ dependence is induced by the internal field H_i. Then in the ideal situation the susceptibility at $H_e = H_i = 0$ would reveal the ''critical'' behavior up to $\omega = 0$, i.e., the magnetically ordered state would be unstable. In reality, the finite-size effects and the anisotropy limit the value of $\chi(\omega=0)$ and stabilize the system.

Our analysis demonstrates that the investigated longitudinal susceptibility can be represented in the form $\chi(\omega, H_e) = \chi_0(H_i) F(\omega/\Omega_0, H_i/H_0)$, with H_0 being some characteristic field scale; however, the dependence of function F on its second argument in the considered range of parameters

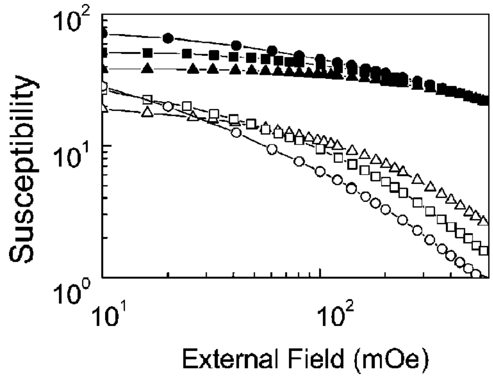

FIG. 3. Field dependence of χ' (solid symbols) and χ'' (open symbols) measured at frequencies 9 KHz (circles), 30 KHz (squares), and 94 KHz (triangles).

is quite weak. Hence, let us for a moment neglect it. Then the maximum value of $\chi''(\omega)$ will be determined by a field dependence of the static susceptibility, $\chi''_{max}(H_e) \propto \chi_0(H_i)F''(1) \propto \chi_0(H_i)$. In Fig. 2(b) we see that χ''_{max} behaves as a power of H_e (again, the deviations appear only in weak fields) with the exponent very close to $-1/2$. It agrees with both the theoretical predictions[8,9] and the experimental measurements[10] for the longitudinal static *spin-wave* susceptibility, $\chi_0^{SW} \propto H_i^{-1/2}$.

Until now we discussed mostly the cases $\omega \gg \Omega_0$ and $\omega \sim \Omega_0$. Unfortunately, the region $\omega \ll \Omega_0$ cannot be analyzed in detail with the use of our frequency measurements only. In order to fill in this gap we investigated the field dependence of χ measured at $\omega/2\pi$=9, 30, and 93 KHz. All the curves presented in Fig. 3 are decreasing functions of H_e. At low fields χ' and χ'' decrease with increasing ω. At high fields χ' ceases to depend on ω while the imaginary parts of the susceptibility attain parallel powerlike asymptotes. The narrow range of the high-field asymptotic behavior available experimentally allowed us only to estimate the character of susceptibility decay. We found that $\chi' \propto H^{-0.5}$ and $\chi'' \propto \omega H^{-1.3}$ in this region. It supports our assumption about ω/H_i scaling since we observed $\chi'(\omega/\Omega_0 \to 0) \to \chi_0(H_i)$ and $\chi''(\omega/\Omega_0 \to 0) \to 0$. Also, the behavior of χ' in high fields confirms that we actually deal with spin-wave dynamics.

Our data can be summarized as follows

$$\chi(\omega,H_e) = \chi_0(H_i)F(\omega/\Omega_0, H_i/H_0). \tag{5}$$

Here $\chi_0 \propto H_i^{-\mu}$ and $\Omega_0 \propto H_i^{\lambda}$. The function F has the asymptotes

$$F(x \gg 1, y) \propto \alpha(y)(i/x)^{\rho}, \tag{6}$$

$$F(x \ll 1, y) \propto 1 + i\alpha(y)x, \tag{7}$$

with $\alpha(y) \propto y^{\mu-\rho}$. The experimental values of the exponents are found to be $\lambda = 1.00 \pm 0.03$, $\rho = 0.28 \pm 0.02$, and $\mu \simeq 0.5$. Note that the case $\mu = \rho$ would correspond to the typical situation of one-parametrical dynamical scaling.[11] Our case is more involved due to the presence of the additional field scale H_0; however, the value $\mu - \rho \simeq 0.2$ is small and the ''extra'' field dependence of susceptibility appears to be quite weak (although visible). Our data do not allow us to judge confidently about the value of the characteristic field H_0 and we can only conclude that it apparently exceeds 100 mOe.

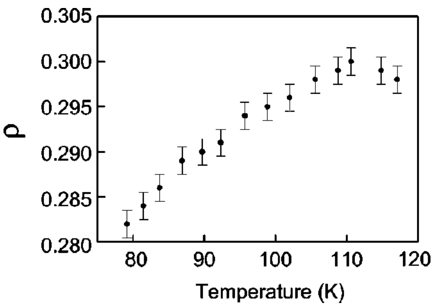

FIG. 4. Temperature variation of the exponent ρ.

The complex of measurements reported above have been performed at $T = 82$ K. We also studied $\chi(\omega)$ at other temperatures within the interval 78 K$< T < T_c$. These data reproduce the main features of $\chi(T = 82$ K) including the principal fact—existence of a wide frequency range with a powerlike behavior of the susceptibility. Furthermore, we found[12] that the dynamical exponent ρ varies with temperature (see Fig. 4). In particular, far away from T_c it slowly increases with T. When T_c is approached, the exponent $\rho(T)$ demonstrates the tendency to saturation (and, possibly, to a further drop inside the critical region). One can expect that there exists a smooth crossover between our scaling regime taking place far away from T_c, and the ordinary scaling behavior inside the critical region near T_c. The investigation of the latter one is beyond the scope of the present study.

Our data reveal that the effective anisotropy field in the sample did not exceed a few mOe. Unfortunately, no direct measurements of the anisotropy constant for $CdCr_2Se_4$ in the studied temperature interval are known to us. On the other hand, the control experiment with yttrium iron garnet (YIG) where this constant is definitely much larger than in $CdCr_2Se_4$ demonstrates the *absence* of scaling properties of the longitudinal susceptibility in the investigated range of parameters.

We attribute the observed unusual behavior of $\chi(\omega,H)$ in $CdCr_2Se_4$ to the influence of dipolar forces. On the LSWT level, the problem manifests itself as the IRD (Ref. 2) caused by (i) the magnon number nonconservation and (ii) the gapless character of the magnon spectrum. Meanwhile, the same ω^{-1} divergence has been obtained for the staggered susceptibility of the *Heisenberg antiferromagnet*[13] where the magnon spectrum is also gapless and the staggered magnetization is not conserved. Also, the density-density correlator of a weakly interacting *condensed Bose-Einstein gas* demonstrates the same feature due to the nonconservation of a number of (off-condensate) particles and the linear phononlike character of the excitation spectrum.[14]

Thus, there exists a class of Bose systems with the ''weak'' violation of the conservation law which exhibit the phenomenon of IRD.[15] While the further scenario (the elimination or the survival of the divergence at $\omega \to 0$) may vary from one system to another because of the different structure of the interparticle interaction, the reminiscence of the IRD should remain in all these cases as some kind of scaling behavior observable within a certain frequency interval. We believe that comparative studies of such systems could give interesting results.

In conclusion, we observed an anomalous low-frequency scaling behavior of the longitudinal susceptibility in a nearly isotropic ferromagnet $CdCr_2Se_4$ outside the critical region near T_c. We attribute this behavior to the influence of the dipole-dipole interaction.

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