LINEAR AND NONLINEAR DYNAMIC SUSCEPTIBILITIES OF WEAKLY ANISOTROPIC FERROMAGNETS: SCALING BEHAVIOR BOTH ABOVE T_c AND OUTSIDE THE CRITICAL REGION

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Some results of the experimental study of the behavior of linear and nonlinear dynamic susceptibilities of weakly anisotropic ferromagnets both in the paramagnetic phase and ordered one are reported. In order to avoid demagnetization effects single-crystal samples of the $CdCr_2Se_4$ type to form rings were used. Experiments were performed in the frequency range $10^2 - 10^7$ Hz.

The main result of the study of the critical dynamics of the homogeneous magnetization is an observation of two temperature regions, — scaling and anomalous, — above the Curie point. In the scaling region the observed behavior of linear and nonlinear susceptibilities is in satisfactory agreement with the conclusions of the scaling theory. However, the results obtained in the anomolous region adjoining directly T_c, disagree very strongly with the scaling theory predictions.

The main result of the study of the uniform dynamics of nearly isotropic ferromagnet in the ordered phase consists in an observation of the power-like behavior of a linear susceptibility at frequencies higher than the two-magnon threshold. We attribute the observed anomalous low-frequency scaling behavior to the influence of dipolar forces. It is concluded that the *ideal* infinite isotropic ferromagnet would be in a "critical" state at any temperature, i.e. the magnetically ordered state would be unstable. In *real* materials the anisotropy and the finite-size effects cut off this divergence.

Critical dynamics of homogeneous magnetization

Experimentally, the critical dynamics of a ferromagnetic substance undergoing a second-order phase transition is investigated, as a rule, by two mutually complementary methods: with the aid of inelastic neutron scattering and by measuring the susceptibility in an alternating magnetic field in the case when $\lambda \ll L$ (λ is the wavelength of the field and L is the characteristic sample dimension). Critical neutron scattering yields primarily an information about the dynamics of the inhomogeneous magnetization, $\vec{M}(\vec{q},\omega)$. The relaxation of the homogeneous magnetization, $\vec{M}(\vec{0},\omega)$, which is the order parameter, is due to the dipole forces, and its behavior in the critical region is usually investigated by radio-frequency methods. Consider a sample kept at a temperature $T > T_c$ and subjected to an alternating magnetic field $h = h_0 \cos \omega t$. The demagnetization factor of this sample is assumed to be zero. Therefore, when the field amplitude is sufficiently low, in the absence of a static magnetic field, the magnetization can clearly be represented as a series

$$m(h) = \chi_0(\omega) h_0 + \chi_3(\omega) h_0^3 + \chi_5(\omega) h_0^5 + ... \quad (1)$$

Here, $\chi_0(\omega)$ is the usual dynamic susceptibility and $\chi_{2n+1}(\omega)$, where n = 1,2,3..., are nonlinear dynamic susceptibilities. According to a predictions of scaling theory in the case of an isotropic ferromagnet the dependences of χ_0 and χ_{2n+1} on the relative temperature $\tau = (T - T_c)/T_c$ are described by

$$\chi_0 \propto \tau^{-4/3}, \chi_3 \propto \tau^{-14/3}, \chi_5 \propto \tau^{-24/3}, ... \quad (2)$$

It should be stressed that the expansion (1) is valid only on condition that the magnetic field amplitude is sufficiently small: $g\mu_B h_0 \ll kT_c\tau^{5/3}$, where the standard symbols for the g factor and the Bohr magneton μ_B are used. Clearly, this fairly rigid condition must be satisfied, when comparisons of experimental and theoretical results are made. In such a comparison it is essential to allow also for another important circumstance. The expressions (2) are currently available only for the static limit. Therefore, in experimental studies of the nonlinear susceptibility in terms of higher harmonics of the magnetization it is necessary to ensure also that the frequency of an alternating magnetic field be low compared with the characteristic energy of critical fluctuations. According to the current ideas, one can distinguish two temperature ranges depending on the relative importance of the dipole forces: $4\pi\chi_0 \ll 1$ (exchange range) and $4\pi\chi_0 \gg 1$ (dipole range). In the exchange range the characteristic energy of critical fluctuations is $\Omega_e \approx kT_c\tau^{5/3}$ and the reciprocal of the relaxation time of a homogeneous magnetization is governed by the dipole forces and depends on τ as $\Gamma_H \propto \tau^{-1}$. In this case the susceptibitily χ_{2n+1} is independent of ω on condition that $\omega \ll \Gamma_H \ll \Omega_e$

In the dipole range the characteristic energy of critical fluctuations and the reciprocal relaxation time Γ_0 of the homogeneous magnetization are quantities of the same order and the amplitudes χ_{2n+1} are independent on ω on condition that $\omega \ll \Gamma_0$.

The results of our investigations [1–4] indicate a complex pattern of critical phenomena in the paramagnetic phase of weakly anisotropic cubic ferromagnets $CdCr_2S_4$ and $CdCr_2Se_4$. The data obtained about the behavior of the linear and nonlinear susceptibilities above T_c suggest that in the range $T > T_c$ we can distinguish two ranges of values of the static susceptibility scaling and anomalous, differing in respect of the behavior of the susceptibility. In the scaling range, corresponding to $4\pi\chi_0 \approx 25$, the observed behavior of the linear susceptibility, of the amplitude χ_3 associated with a four-particle spin correlation function, and of the critical dumping factor Γ_0 are in satisfactory agreement with the conclusions reached in a theory based on dynamic and static similarity considerations. In the range of temperatures adjoining directly the Curie point (anomalous range) the dynamic susceptibility behaves anomalously as a function of ω and τ and metastable formations of nature not yet understood may be postulated. The phenomena observed in the anomalous range are generally very similar to the situation encountered in spin glasses. In fact, the logarithmic frequency dependence of the real part of the susceptibility (the rule "$2/\pi$") had hitherto been reported only for spin glasses and the phenomenon of a residual second harmonic signal, observed in experiments involving cooling in a static magnetic field which is then switched off, are to some extent analogous to the thermoremanent magnetization characteristic of the spin glass state. The characteristic feature of the high harmonics in this spin-glass-like range of temperatures was a weak fall of the amplitudes of the harmonics with increasing harmonic number. An analysis of the harmonic spectra indicated that above T_c a large fraction of the field energy was transferred to higher harmonics.

Recently, the critical behavior of the linear and nonlinear components of the dynamic susceptibility of the cubic manganite $Nd_{0.77}Ba_{0.23}MnO_3$ [5] were investigated. We concluded that there are two temperature regions, scaling and anomalous, above the Curie point. The scaling region is characterized by a satisfactory agreement of the scaling theory with the experimentally determined behavior of the linear and nonlinear susceptibilities as in the case of Heisenberg magnet of the $CdCr_2Se_4$ type. At the same time it is found that the hysteresis of the second harmonic of magnetization in dc magnetic field arises in the anomalous region above T_c. This phenomenon is associated with the formation of macroscopic ferromagnetic (ferrimagnetic) domains in the paramagnetic matrix.

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Anomalous spin dynamics in isotropic ferromagnet

It is known that the absorption of the 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. Weak, so-called relativistic interactions (anisotropy, dipolar forces, etc.) violate the total spin conservation law providing the uniform relaxation.

The uniform longitudinal susceptibility of the isotropic Heisenberg ferromagnet with dipolar forces below T_c has been studied by B.P. Toperverg and A.G.Yashenkin (Phys.Rev.B **48**, 16505 (1993)) within the 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 the infra-red divergence (IRD) of the form $\chi(\omega \to 0) \propto iT/\omega$. At $H_i \neq 0$ one has the threshold for $\chi''(\omega)$ at $\hbar\omega = 2g\mu_B H_i$ (the minimal energy of creation of two magnons), and 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 ω^{-1}-singularity is the nonphysical one since it leads to the nonzero value of the absorption function $\dot{Q}_\omega \propto \omega\,\chi''(\omega)$ at $\omega = 0$ and thus the sample would be heated by DC field.

The low-frequency ($\omega/2\pi \sim 10^3 - 10^6$ Hz) uniform longitudinal susceptibility of the nearly isotropic cubic ferromagnet $CdCr_2Se_4$ ($T_c \approx 128\,\mathrm{K}$) [6] was experimentally studied. The measurements have been performed mostly at $T = 82\,\mathrm{K}$, i.e. outside the critical region near T_c. At frequencies higher than the two-magnon threshold the power-like behavior of $\chi(\omega)$ with the exponent $\rho(T = 82\ \mathrm{K}) = 0.28 \pm 0.02$ was observed. This exponent is found to be slowly varied 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. The description of $\chi(\omega, H)$ in scaling terms is also proposed. One can think that there exist other Bose systems with "weak" violation of some conservation law which should exhibit the anomalous low-energy behavior.

The sample in the shape of a ring was used. Both the constant field H_e and the alternating one $h_0 \cos\omega t$ were applied along the ring. In such a 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 the domains formation the conditions $H_e > h_0 > H_c$ were controlled.

Frequency dependence of both the imaginary and the real parts of the susceptibility as well as of their ratio χ''/χ' were measured at $H_e = 8.16$ and 32 mOe. The response is large,

$\chi'_{\max} \sim 10^2$. Furthermore, $\chi'(\omega)$ is the 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 $\chi''(\omega) \propto \omega$.

The most interesting feature of $\chi(\omega)$ is related to (relatively) high frequencies. In Fig. 1 one sees that within the wide frequency range the susceptibility demonstrates the power-like behavior. 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 it to the fact that actually the power-like 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 arises due to demagnetization and anisotropy. When H_e is large this diminishing is negligible while at $H_e = 8$ mOe it becomes influent.

The above consideration allows us to prove the principal statement of the present work. Indeed, we have shown that the low-frequency crossover scale to $\omega^{-\rho}$-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.

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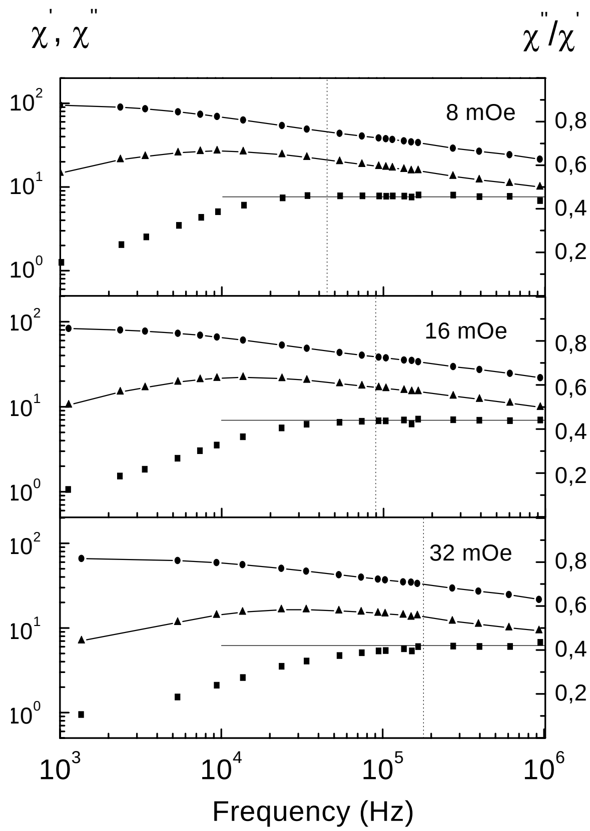

Figure 1: Real (circles) and imaginary (triangles) parts of the longitudinal susceptibility and their ratio (squares) versus frequency measured at various values of the external field ($T = 82$ K). At high enough frequencies the quantity χ''/χ' is seen to be constant

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