Critical Speeding-Up of Spin-Relaxation in $CdCr_2Se_4$

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The critical behavior of magnetic resonance linewidths has been measured at fixed frequencies between 0.7 and 9.6 GHz. At sufficiently low frequencies, the derived Onsager coefficient of the spin-relaxation rate "*speeds up*" with $\chi^{7/4}$ in the exchange critical region ($\chi << 1$) and flattens around $\chi = 1$ when reaching the crossover to the dipolar region. These results turn out to be quantitative confirmations of current mode-coupling calculations.

During the past few years considerable experimental effort[1-6] has been made to check a classical result of the mode-coupling theory[7,8] predicting the spin-relaxation rate Γ of Heisenberg ferromagnets to increase like $\chi^{3/4}$ when the Curie temperature is approached from the paramagnetic side. Qualitatively spoken, this "critical speeding-up" arises from the growth of the correlation length ξ and of the lifetime of the critical magnetization fluctuations, $\Gamma(q \approx \xi^{-1})$, which diminished the exchange narrowing of the magnetic dipole interaction. More precisely, this effect should be operative in the so-called exchange critical region (characterized by $\chi \ll 1$), where the influence of the dipolar forces on the critical correlations is negligible. Nearer T_C, when χ reaches the order of 1, the dipolar effects come into play, removing the spatial isotropy of the critical fluctuations tions[9] and the dominance of the exchange-induced spin diffusion over their lifetime. As a result, the relaxation rate Γ is expected to change over to the conventional (thermodynamic) slowing down[10-12] $\Gamma \propto \chi^{-1}$ at $\chi \gg 1$. While the latter case has been observed recently on the archetypal Heisenberg ferromagnets EuS[4] and EuO[6], a clear experimental evidence for the critical speeding up at $\chi \ll 1$ is still lacking though several qualitative symptoms for it have been reported. In particular, magnetic resonance linewidths of $CrBr_3$,[1] Ni,[2] EuO[3,6], and Gd[5] indicate some anomalies, which remain, however, significantly weaker than expected. So far, different suggestions exist to explain this discrepancy: (1) The applied magnetic field, inherent in resonance measurements, but not considered by the theory to date, modifies the critical dynamics.[1-3,5] Following Kawasaki,[7] this effect can be ignored as long as the Larmor frequency is small compared to the decay rate of the critical fluctuations, $\gamma B \ll \Gamma(q \approx \xi^{-1})$. (2) The decoupling of four-spin correlations performed by the theory leads to inadequate results,[3,5,7] overestimating the critical divergence of Γ. Using an alternative approach Kawasaki obtained a weaker divergence, $\Gamma \sim \chi^{1/4}$.[13] (3) The relaxation rate has not been measured in the asymptotic exchange critical region,[4] where beside $\chi \ll 1$, also $\xi \gg r_{nn}$ must be valid in order to ensure the presence of long-range correlations. In Table I we give estimates for the ratio $(\xi/r_{nn})^2/\chi$, based on the mean-field approximation.

$$\chi/\xi^2 \approx q_d^2 \equiv G_d/J_2, \tag{1}$$

where $G_d = (g\mu_B)^2/v_{spin}\mu_0 k_B$ measures the strength of the dipolar force and $J_2 = (1/2q^2)\sum_j J_{ij}(\vec{q}\cdot\vec{r}_{ij})^2$ represents the second moment of the magnetic interactions. These ratios which are in good accord with existing neutron data for EuS and EuO,[14] provide some evidence that in EuS, EuO, and Gd the correlations are of comparatively short range at $\chi \lesssim 1$.

In the present work, investigating the critical behavior of the resonance linewidth for $CdCr_2Se_4$,

TABLE I. ξ^2/χ ratios calculates from Eq. (1) by using exchange interactions from the cited references. r_{nn} is the nearest-neighbor distance.

	T_C(K)	$(\xi/r_{nn})^2\chi^{-1}$
EuS	16.5	0.4 [a]
$CrBr_3$	32.6	36 [b]
EuO	69.1	2 [a]
$CdCr_2Se_4$	127.8	58 [c]
Gd	290	3 [d]

[a]Ref. 14. [b]Ref. 15. [c]Ref. 16. [d]Ref. 17.

we want to eliminate two of the former difficulties: (i) According to Table I, one expects the largest $(\xi/r_{nn})^2$ value, guaranteeing the existence of long-range correlations also in the exchange region; (ii) with the principal aim to make the magnetic field effect visible and to reduce it, as far as possible, we systematically vary the microwave frequency from 9.6 GHz down to 0.7 GHz, reaching resonance fields as low as 25 mT —thereby we hope to provide a more reliable test of the mode-coupling approach, which is of general relevance for the calculation of transport coefficients near phase transitions.[7]

Our measurements were made with a sphere [of diameter 0.63(3) mm] carefully shaped from a high-quality single crystal, the preparation of which has been described elsewhere.[18] This sample was placed on the shorted end of a broadband waveguide, detecting the field-dependent absorption by standard techniques. More details will be published later.[19]

The critical behavior of the linewidths is seen in Fig. 1, where only results obtained from

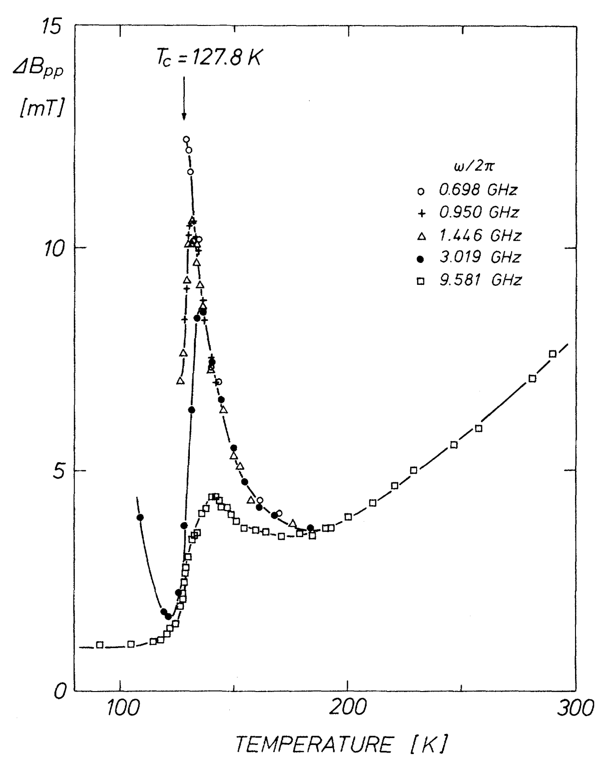

FIG. 1. Peak-to-peak ESR linewidth the Curie temperature of $CdCr_2Se_4$ measured at different microwave frequencies showing the suppression of the critical anomaly with increasing magnetic resonance field. Full lines are drawn as guides to the eye.

Lorentzian resonances are considered, while deformed lines occurring at small fields below T_C (probably due to domain formation) were not taken into account. Discussing the results in detail we concentrate on the paramagnetic part of the critical region. At 9.6 GHz a weak anomaly appears similar as in $CrBr_3$,[1] EuO,[3,19] and Gd.[5] For lower frequencies the linewidth anomaly is appreciably enhanced and shifted towards T_C, reaching a maximum at the smallest frequency (0.698 GHz). Obviously these data form an envelope, being joined by the higher frequencies at some elevated temperatures. Thus it is reasonable to assume that the data on the envelope do not depend on the small applied field, and therefore represent, to a good approximation, the desired zero-field limit. In order to extract the spin-relaxation rate Γ from the measured Lorentzian half-widths ΔB $(=\sqrt{3/2}\,\Delta B_{pp})$ we utilize a relation derived recently[6,19] from the linear response theory. In the special case of a spherical sample, being of interest here, one obtains the simple form

$$\Gamma_s = \frac{\gamma \Delta B}{[1+(\Delta B/B)^2]^{1/2}}. \tag{2}$$

Γ_s still depends on the shape and in order to deal with a true material constant, it is appropriate to introduce the so-called kinetic Onsager coefficient of the homogeneous magnetization[20,21]:

$$L = \Gamma_s \chi_s/\chi_0, \tag{3}$$

where χ_s denotes the external susceptibility of the sphere $[=(\chi^{-1}+\frac{1}{3})^{-1}]$ and $\chi_0 = \frac{1}{3}S(S+1)G_d/k_B T$ is the bare susceptibility. Evaluating L from the measured linewidths with the aid of Eqs. (2) and (3), we employed data for χ_s measured in our laboratory in the low-temperature region ($T \leq 170$ K).[22] The excellent agreement between the Curie temperature, following from a Kouvel-Fisher analysis $T_C = 127.78(10)$ K, and Miyatani's value[23] of 127.7(2) K should be noted. The inset in Fig. 2 shows the high-temperature tail of L deduced from Eqs. (2) and (3) using the χ_s data of Baltzer *et al.*[16] Incorporating also linewidths at $T > 300$ K from Samokhalov *et al.*,[24] which fit our results reasonably well, a slowly (almost linear in T) varying contribution to L is found, which we relate to noncritical relaxation. The existence of such a term was pointed out earlier[7,8] and here it proves to be small: $L_0(T_C) = 2.5$ GHz compared to $L(T_C) \approx 10^3$ GHz. In order to make possible a direct comparison between the critical part of L, i.e., $L_{cr} = L - L_0$, and the current mode-coup-

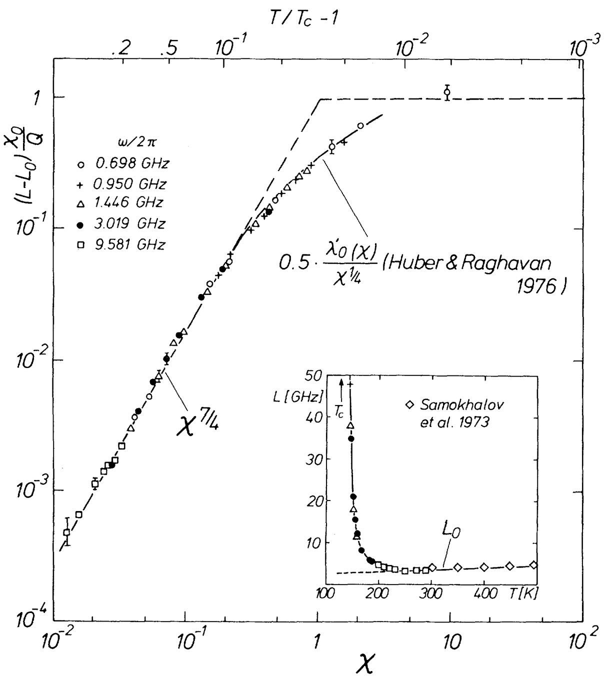

FIG. 2. Critical behavior of the Onsager coefficient of the homogeneous magnetization normalized to Q/χ_0 [cf. Eq. (4)]. The noncritical contribution L_0 is estimated from the high-temperature tail (see inset). Full lines are *ab initio* calculations from mode-coupling results.

ling results[12]

$$L_{\rm cr} = \begin{cases} (Q/\chi_0)\chi^{7/4} & (\chi \ll 1), \quad (4a) \\ Q/\chi_0 & (\chi \gg 1), \quad (4b) \end{cases}$$

we have plotted in Fig. 2 $L_{\rm cr}\chi_0/Q$ against χ. Using $\chi/\xi^2 = q_d^2$ [Eq. (1)] the normalization factor is given by $hQ = (G_d k_B T q_d^3 v_{\rm spin}/8\pi^3)^{1/2}$ and can readily be calculated. It is obvious from Fig. 2 that in the exchange critical regime ($\chi \ll 1$) the measured speeding-up of $L_{\rm cr}$ agrees *quantitatively* with the prediction: The weighted least-squares fit of the data below $\chi = 0.25$ yields $L_{\rm cr}\chi_0/Q = 0.93(9)\chi^{1.74(3)}$. Approaching the dipolar-critical region, $L_{\rm cr}$ starts to leave the asymptotic law and tends to join the expected conventional critical behavior for $\chi \gg 1$ [Eq. (4b)] without, however, reaching it. There, for $\chi \gg 1$, ΔB becomes comparable to the resonance field even at 0.7 GHz and it is impossible to measure the zero-field relaxation rate. Recently, Raghavan and Huber[11] have calculated the critical spin relaxation also up to $\chi \approx 3$ by solving the mode-coupling equation numerically rather than asymptotically. Accepting again Eq. (1), we could adjust their results to our experimental data in the exchange region by adding a simple factor of 0.5. Then, as demonstrated by Fig. 2, this theory also explains quite satisfactorily a considerable portion of the dynamic crossover.

The results presented here might be viewed as complementary to the previous investigations of the critical spin-relaxation in EuS[4] and EuO[6]. In the dipolar region of both systems, the conventional critical behavior of L was found, verifying Eq. (4b). In Table II measured results for Q are compared to the calculated values, thus providing some additional evidence of the reliability of the theoretical approach used.

A final remark should be devoted to the influence of the magnetic field, which is believed to modify the critical dynamics if the decay time of the critical fluctuations comes into the order of the Larmor frequency $\omega_L \approx \Gamma(q \approx \xi^{-1})$.[7] If we estimate $\Gamma(q = \xi^{-1})$, using the results from Ref. 11 (including the adjustment factor 0.5), we find $\Gamma(q = \xi^{-1})$ to reach $\omega_L = 2\pi \times 0.95$, $2\pi \times 1.45$, and $2\pi \times 3.0$ GHz at 132.5, 134, 137 K, respectively. These temperatures are very close to 132(1), 133(1), and 136(1) K where the linewidths corresponding to these frequencies start to leave the "zero-field" envelope.

Summarizing our results, we have reported a systematic study of the critical behavior of the magnetic resonance linewidth of $CdCr_2Se_4$ at microwave frequencies between 0.7 and 9.6 GHz. The pronounced effect of the magnetic field is seen and can be qualitatively understood by help of the Kawasaki condition $\gamma B \approx \Gamma(q = \xi^{-1})$. The kinetic coefficient of the spin relaxation derived from the linewidths measured in the low-field limit exhibits a speeding up, the temperature (i.e., χ) dependence of which is fully consistent with numerical solutions of the mode-coupling equations up to $\chi = 3$,[11] while at $\chi \ll 1$ there is absolute agreement with Finger's asymptotic result.[12] Former difficulties in finding this crit-

TABLE II. Amplitudes of critical spin relaxation [cf. Eq. (4)] of three cubic ferromagnets as calculated from $Q(T_C) = (G_d k_B T_C q_d^3 v_{\rm spin}/8\pi^3)^{1/2}$ and as measured in the given regions (in units of GHz).

	$Q_{\rm cal}$	$Q_{\rm exp}$	Region
Eus	26	55(10)	$\chi >> 1$ [a]
EuO	24	22(2)	$\chi >> 1$ [b]
$CdCr_2Se_4$	3.4	3.1(4)	$\chi << 1$

[a] Ref. 4. [b] Ref. 6.

ical behavior can very likely be ascribed to the use of too high frequencies (i.e., magnetic fields) and of improper ferromagnets, where no true exchange critical regime exists.

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Temperature Dependence of the Plasma Frequency of Two-Component Ionic Fluids

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In two recent moleuclar-dynamics experiments on two-component ionic fluids it was observed that the plasma frequency was shifted with respect to its mean-field value. This effect is explained here on the basis of a microscopic theory. The shift is shown to exhibit a strong plasma parameter dependence.

Recently, the fastly growing literature on Coulomb systems has been enriched with a new molecular-dynamics (MD) study of the time-dependent fluctuations in a H^+-He^{++} mixture.[1] It was observed there that the infinite-wavelength ($k=0$) plasma oscillations occur in this system at a frequency which is shifted both with respect to the mean-field prediction and with respect to the prediction from phenomenological hydrodynamics. The observed oscillation frequency was found to agree nevertheless with a sum-rule analysis predicting a plasma frequency independent of the system's plasma parameter.

In this Letter I analyze the above experiment on the basis of a microscopic theory[2] which has previously been applied to the one-component plasma (OCP)[3-5] and was recently extended to multicomponent Coulomb systems.[6] I consider a system of two mobile species ($\sigma = 1,2$) of mass m_σ, charge e_σ, and average number density n_σ immersed in an inert neutralizing background, and characterized by the plasma parameters $\Gamma_\sigma = (\frac{4}{3}\pi)^{1/3} e_\sigma{}^2 n_\sigma{}^{1/3}\beta$. When $e_1 e_2 > 0$ such a system will be called a binary ionic mixture (BIM) and