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Universality of the dipolar dynamic crossover of cubic ferromagnets above T_c

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In the critical regions of EuS, $CdCr_2S_4$, and $CdCr_2Se_4$, the relaxation rates of the magnetization, $\Gamma_{q=0}(T)$, quantitatively obey the crossover from speeding up to thermodynamical slowing down, recently calculated by Frey and Schwabl using the coupled-mode approach. Some deviations from this universality occurring in EuO and Ni indicate that, in addition to Heisenberg exchange and dipole-dipole interactions, further effects gain importance, which appear to be absent in $\Gamma_{q>0}(T_c)$.

I. INTRODUCTION

Our present experimental knowledge of the order-parameter dynamics near the Curie temperature T_C of ferromagnets stems from two main sources: inelastic neutron scattering, probing the decay rates Γ_q of the magnetization fluctuations $\mathbf{M}_q$ at finite wave vectors, and high-frequency magnetic susceptibility, which measures the relaxation rate Γ_0 of the macroscopic order parameter $M_{q=0}$. The early neutron data, e.g., for Fe,[1] could be well explained in terms of the so-called Resibois-Piette (homogeneous) scaling function, $\gamma(q\xi(T))=\Gamma_q(T)/q^{5/2}$ ($\xi=$ correlation length), which results from Kawasaki's mode-mode coupling approach (MMC, see, e.g., Ref. 2) and considers the decay of the mode $\mathbf{M}_q$ into others by means of the Heisenberg exchange. However, more recent high-resolution neutron work on Fe (Ref. 3) revealed significant deviations from this Heisenberg scaling occurring at small wave vectors. As a possible source of this effect the magnetic dipole interaction has been proposed,[4] which separates the fluctuations into critical transverse ($\mathbf{q}\perp\mathbf{M}_q$) and noncritical longitudinal ($\mathbf{q}\parallel\mathbf{M}_q$) ones.

This dipolar anisotropy above T_C has been evidenced experimentally for EuS and EuO using a polarized neutron technique:[5] The suppression of the longitudinal susceptibility by the depolarizing dipolar fields, as compared to the critical transverse one, could be accounted for by the form

$$\chi_q^{\alpha}(T,q_d)=[(\xi^{-2}+q^2)/q_d^2+\delta^{\alpha,L}]^{-1},\quad \alpha=L,T. \tag{1}$$

Here q_d denotes the characteristic dipolar wave number, given by the ratio between the dipolar coupling and the exchange stiffness. For the archetype Heisenberg ferromagnets, the q_d's have been estimated in Ref. 6 and are reproduced in Table I below.

Very recently, Frey and Schwabl[7] included the dipolar interaction into the MMC equations and solved them numerically for the transverse and longitudinal relaxation energies

$$\hbar\Gamma_q^{\alpha}(T,q_d)=\Lambda q^{5/2}\gamma^{\alpha}\left(\frac{1}{q\xi},\frac{q_d}{q}\right). \tag{2}$$

The scaling functions γ^{α} contain q_d as a second scaling variable and attain the isotropic Resibois-Piette limit for small q_d/q. As a matter of fact, Frey and Schwabl were able to reproduce — at least qualitatively — the characteristic phenomena which Mezei[3] found for $\Gamma_q^T(T)$ of Fe at wave numbers about q_d.

Aiming at an additional and perhaps more quantitative check of their approach, they suggested to compare their

TABLE I. Critical and background contributions to kinetic coefficients of homogeneous relaxation above T_C, relaxation coefficients of transverse fluctuations at T_C, and dipolar wave numbers.

	EuS	EuO	$CdCr_2S_4$	$CdCr_2Se_4$	Ni	Fe	Co
T_C (K)	16.56	69.10	84.40	127.8	627.2	1044	1388
$\hbar L_d$ (μeV)	38(2)	24(3)	5.9(3)	4.4(2)	3.0(6)[a]	...	...
$\hbar L_{bg}$ (μeV)	1.8	0.64	0.01[b]	0.01[c]	...	...	...
$\hbar\Gamma_q^T/q^{2.5}$ (meV Å$^{2.5}$)	2.1(3)[d]	8.7(7)[e]	...	...	350[f]	130[g]	300(30)[h]
q_d (Å^{-1})	0.25[i]	0.15[i]	0.058[c]	0.034[c]	0.013[i]	0.033[i]	0.025[i]

[a]Reference 9.
[b]Reference 13.
[c]Reference 12.
[d]P. Böni, G. Shirane, H. G. Bohn, and W. Zinn, J. Appl. Phys. **61**, 3397 (1987).
[e]F. Mezei, J. Magn. Magn. Mater. **45**, 67 (1984).
[f]Reference 17.
[g]Reference 3.
[h]C. J. Glinka, V. J. Minkiewicz, and L. Passell, Phys. Rev. B **16**, 4084 (1977).
[i]Reference 6.

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result in the homogeneous ($\mathbf{q}=\mathbf{0}$) limit of Eq. (2),

$$\hbar\Gamma_0^g(T,q_d) = \Lambda q_d^{5/2}\gamma_0^g\left(\frac{1}{q_d\xi}\right), \tag{3}$$

with existing experiments. This idea is rather appealing, since the crossover function γ_0^g has been calculated explicitly[7]

$$\gamma_0^g(x) = \gamma_0(x)\frac{(1+x^2)^{-7/4}}{\chi_0^g(x)}, \tag{3a}$$

where $\gamma_0(x)$ depends only weakly on x with $\gamma_0(0)=2.6$.[8] Moreover, γ_0^g is directly related to the kinetic Onsager coefficient. $L_0 \equiv \Gamma_0^g\chi_0^g$, which depends neither on α nor on the sample demagnetization[9] and is readily available from a number of experimental investigations.[9-15]

II. DATA ANALYSIS

The most direct access to the Onsager coefficient of the order parameter is measuring the longitudinal dynamic susceptibility of ellipsoidal samples in the limit $\omega \to 0$,

$$L_0 = \lim_{\omega\to 0} i\omega/\{[\chi_0^L(\omega)]^{-1} - (\chi_0^L)^{-1}\}. \tag{4}$$

At some distance from T_C, where the isothermal susceptibility χ_0^L decreases, the relaxation rates Γ_0^L speed up and hence the sensitivity drops, it is often more convenient to measure Γ_0^L via the linewidth of the field-sweeping electron-spin-resonance (ESR), $\Delta H = (\delta H/\delta\omega)\Gamma_0^L$. This method yields the desired zero-field relaxation rate, provided the field-induced Larmor frequency of the spins $\omega = \gamma H$ remains slower than the decay rate of the critical fluctuations $\Gamma_{q=\xi}^T$.[12,16]

Both methods have been applied to EuO (Ref. 11) and EuS,[9,14] while $CdCr_2Se_4$ (Ref. 12), $CdCr_2S_4$, and Ni (Ref. 9) have been studied by ESR alone. In order to extract the contribution by the critical fluctuations to L_0, one has to determine the noncritical background which in the critical regions of all ferromagnets (except for Ni) is very small, $L_{bg} \ll L_{cr}$ (see Table I). Without examining in detail the mechanisms behind L_{bg}, we just mention that it may result from spin coupling to phonons[16,18] or to noncritical, i.e., short-wavelength modes, via pseudodipole[19,20] interactions.

The critical parts of the L_0's are presented in Fig. 1. To date, the discussion of L_{cr}[9,11,16,18-23] concentrated either to the critical speeding up, $L_{cr} \sim (q_d\xi)^{7/2}$, occurring in the so-called exchange critical region, $(q_d\xi)^2 \ll 1$, or to the limiting dipolar region, $(q_d\xi)^2 \gg 1$, where the dipolar anisotropy governs the statics *and* dynamics of the spin fluctuations and leads to a saturation of L_{cr}. Now we are in the position to consider the full crossover between both regions by comparing it to the prediction of the MMC theory, Eq. (3),

$$L_{cr} = L_d\frac{\gamma_0(x)}{\gamma_0(0)}(1+x^2)^{-7/4}. \tag{5}$$

Following Eq. (1), $x^{-2} \equiv (q_d\xi)^2$ has been identified with the (internal) homogeneous susceptibility, $\chi \equiv \chi_0^T = \Gamma(T/T_c - 1)^{-\gamma}$, being well known for all ferromagnets. To adjust the data to Eq. (5), the parameter L_d was fixed by L_{cr} in the center of the crossover region ($x=\chi=1$),

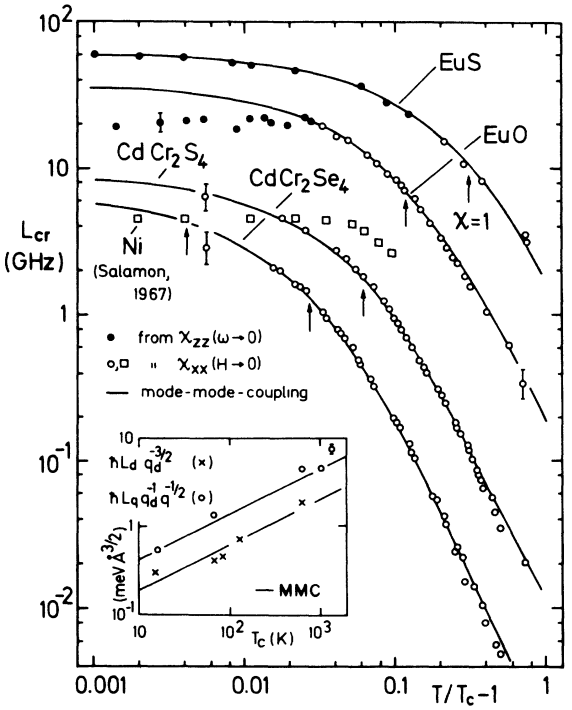

FIG. 1. Critical part of the kinetic Onsager coefficient for the spin dynamics above T_C of $CdCr_2Se_4$ (Ref. 12), $CdCr_2S_4$ (Ref. 13), EuO (Ref. 11), EuS [● (Ref. 9), o (Ref. 14)], and Ni (Ref. 10). Full lines represent fits to the mode-mode (MM) dipolar crossover function (Refs. 7 and 8). Inset: relaxation parameters of above ferromagnets including Fe and Co determined from rates $\Gamma_{q=0}$ ($T \geq T_C$) and $\Gamma_{q>0}$ ($T = T_C$).

and in fact, an inspection of Fig. 1 reveals that —except for Ni and to some extent for EuO, as well—the observed crossovers are very well explained by Eq. (5). Very striking is the agreement for EuS close to T_c. The only parameters of the fits, L_d, are listed in Table I.

III. DISCUSSION

First, with regard to the overall agreement between the observed and calculated dynamical crossovers for the *nonmetallic* ferromagnets it is quite tempting to compare the absolute magnitude of the relaxation parameter L_d with the predicted value[7]

$$\hbar L_d = \Lambda q_d^{5/2}\gamma_0(0), \tag{6}$$

with[7,23]

$$\Lambda q_d = \frac{g\mu_B}{2\pi^2}\sqrt{k_B T_C}. \tag{6a}$$

Indeed, the inset of Fig. 1, where we have plotted the measured $\hbar L_d$ normalized to $q_d^{3/2}$ against T_C, shows that the expected $\sqrt{T_C}$ law is fairly well obeyed. The full line close to the data corresponds to $\hbar L_d/q_d^{3/2} = 0.062\sqrt{T_C}$ meV Å$^{3/2}$K$^{-1/2}$ following from Eq. (6) with $g=2$ and $\gamma_0(0)=2.6$.[8] Hence the MMC theory quantitatively explains the relaxation of the homogeneous ($\mathbf{q}=\mathbf{0}$) magnetization in the entire critical region of the disordered phase. The minor differences to the data are most

likely due to some uncertainties in the dipolar wave numbers q_d.

We note that similar agreement exists for the relaxation rate of the transverse magnetic fluctuations at T_C, $\Gamma_q^T(T_C)$, inferred from inelastic neutron scattering outside the dipolar region, $q > q_d$ (see Table I). To demonstrate this, we have also indicated the kinetic coefficient of the measured damping rates $L_q \equiv \Gamma_q^T \chi_q$ normalized to $q_d q^{1/2}$. For this ratio one expects according to the MMC ansatz of Eq. (2)

$$\frac{L_q(T_C)}{q_d q^{1/2}} = \frac{\Gamma_q^T(T_C)}{q^{5/2}} q_d = \Lambda q_d \gamma^T(0,0) , \tag{7}$$

with $\gamma^T(0,0)=5.12$. Using the available coefficients, $\Gamma_q^T(T_C)/q^{5/2}$ (Table I), we find reasonable agreement with the $\sqrt{T_C}$ law predicted by Eq. (6a). Moreover, the full line calculated from Eq. (7) explains the data on an absolute scale, which comprises both nonmetallic (EuS, EuO) and metallic ferromagnets (Ni, Fe, Co). According to the underlying theory, at $q > q_d$ this damping is entirely due to the decay of $\mathbf{M_q}$ into two other modes by means of the Heisenberg exchange, so that one can conclude that additional interactions, e.g., between magnetic fluctuations and conduction electrons, play no role.

On the other hand, a more-detailed examination of the critical behavior of the $\mathbf{q}=\mathbf{0}$ Onsager coefficient of EuO and Ni (Fig. 1) reveals distinct deviations from the universal dipolar crossover function. Discussing at first EuO, we note that the data break off from the curve below $T/T_C - 1 = 0.02$ with a slight tendency to decrease. This behavior of EuO was observed independently by Dunlap and Gottlieb[24] and contrasts sharply to that of EuS, where L_{cr} steadily rises as predicted. Figure 2 elucidates the different critical dynamics of the two homologues presenting $L_0(T)$ on linear scales and including data from the ferromagnetic phase.[25] Below T_C, the kinetic coefficient of EuS continues to grow and saturates around 200 GHz at $T \to 0$, whereas for EuO this effect is almost completely suppressed. Since no theory on the critical dynamic of the $\mathbf{q}=\mathbf{0}$ magnetization below T_C is known to us, we have to confine ourselves to some qualitative remarks.

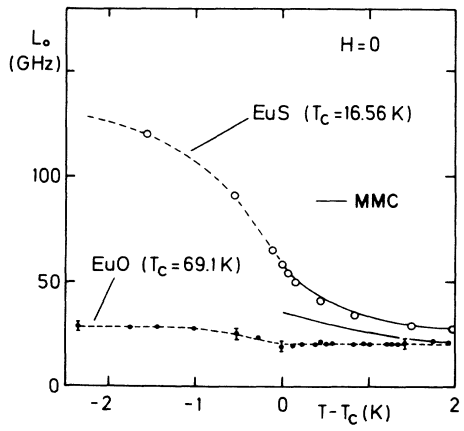

FIG. 2. Kinetic Onsager coefficient about T_C of EuS and EuO: full lines are the coupled-mode results, dashed lines are guides to the eye.

First of all, it is not yet clear whether in the present case of zero external field the relaxation arises from domain-wall motion or intradomain spin relaxation. Due to the large intradomain susceptibility of these ferromagnets with weak cubic anisotropy,[26] both relaxation mechanisms lead to the same, maximum susceptibility $\chi(\omega \to 0) = 1/N$, being observed for EuS and EuO as well. The only information on spin dynamics below T_C arises from previous ferromagnetic resonance (FMR). At 4.2 K, dramatic increases of the FMR linewidth of EuO with rising conductivity have been measured and associated with imperfections due to the Eu excess in the highly conducting specimen.[27] On the other hand, EuS exhibits much less variation in stoichiometry and is characterized by FMR linewidths much smaller than in EuO. We could confirm this behavior for the present samples yielding FMR linewidths of 800 and 70 Oe at 4.2 K and 24 GHz for EuO and EuS spheres, respectively.[25] It is not unlikely that local fields extending from the vacancies perturb the criticality of the long-range fluctuations close to T_C.

Turning to Ni, the most remarkable feature emerging from Fig. 1 is the missing dipolar crossover of L_{cr} around $\chi = 1$ corresponding to $t \equiv T/T_C - 1 = 0.004$. In contrast, the crossover from a speeding up to an essentially noncritical L takes place at higher temperatures, i.e., about $t = 0.05$. Since the data were evaluated from EPR at 24 GHz,[10] we first check whether the field-induced Larmor precession perturbs the dynamics of the critical fluctuation. According to neutron data[17,28] at $t = 0.05$, their decay rate amounts to $\Gamma_q = \xi^{-1} = 10^3$ GHz so that this effect can safely be neglected. Another possible mechanism behind the crossover shift in Ni could be the pseudodipole interaction. Since this force is much stronger and of shorter range than the classical dipolar interaction, it provides a spin coupling to short-wavelength modes. Thus one may expect a crossover to noncritical Onsager coefficient[21] at higher temperature. As for EuO one should consider the effect of sample imperfections for Ni where internal strains associated with dislocations can give rise to local fields. Stress-induced anisotropy of the critical fluctuations above T_c of Ni has been observed by diffuse neutron scattering.[29] Along with this, the effect of field-induced anisotropy on the critical ESR linewidth of Ni remains to be investigated. Unless the influence of all these possible mechanisms on the homogeneous relaxation of Ni has been considered, the consistency of L_0 with the $\sqrt{T_C}$ law in Fig. 1 appears more or less accidental.

IV. CONCLUSIONS

Our comparison between the order-parameter relaxation rates $\Gamma_q(T)$ of cubic ferromagnets and current results of the coupled-mode theory,[7,8] considering Heisenberg exchange plus the magnetic dipole interaction, leads to the following conclusions.

(i) All existing data on the decay rates of the transverse fluctuations at the critical point, $\Gamma_{q>q_d}^T(T_C)$, are in close numerical agreement with the MMC theory. This result identifies the isotropic exchange as the only damping mechanism under operation in nonmetallic as well as for

metallic ferromagnets.

(ii) The crossover of the relaxation rates of the order parameter, $\Gamma_{q=0}(T > T_C)$, from critical speeding up to conventional slowing down close to T_c of the nonmetals EuS, $CdCr_2S_4$, and $CdCr_2Se_4$ is exactly explained by the MMC calculations, which implies the dominating role of the classical dipolar force on their dynamics.

(iii) Deviations from the dipolar crossover of $\Gamma_{q=0}(T > T_C)$ established under (ii) have been evidenced on EuO and Ni, for which sample imperfections (e.g., O vacancies in EuO, internal stress for Ni) may be one possible source.

Future work should resolve the effects of pseudodipolar forces and conduction electrons on the critical fluctuations and hence on $\Gamma_{q=0}$ above T_C of the metallic ferromagnets. Even more unsatisfying is the situation in the ferromagnetic critical region, where only few data are available and a theory is still lacking.

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