Scaling of the field effect on the magnetic relaxation about the Curie point of EuS

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Above T_c and also in the homogeneous ferromagnetic phase below T_C, susceptibility measurements between 1.6 MHz and 1.5 GHz in parallel static fields up to 0.5 kOe reveal Lorentzian shapes in ω. In the dipolar critical region on both sides of T_C, the field and temperature effects on the resulting kinetic coefficient of the magnetization scale with the isothermal susceptibility. We discuss this feature phenomenologically based on a recent mode-coupling result valid in the limit $H=0$ and $T>T_C$.

I. INTRODUCTION

In the realm of critical phenomena, almost insulating europium sulfide is serving as one of the preferred experimental testing grounds[1–11] for the theoretical predictions on the magnetic properties near the Curie temperature of real Heisenberg ferromagnets.[12–19] Due to the small magnetocrystalline cubic anisotropy of EuS,[20,21] it is only the classical dipole-dipole interaction, present in all ferromagnets, which has to be taken into account in addition to the well-known isotropic exchange[22] between the localized $S=\frac{7}{2}$ Eu^{2+} spins. As the most significant effect on the static behavior, we recall the saturation of the magnetic susceptibility

$$\chi^{\alpha}_{\mathbf{q}\to 0}(T)=\left[\frac{\xi(T)^{-2}+q^2}{q_d^2}+\left[\frac{q^{\alpha}}{q}\right]^2\right]^{-1},\quad \alpha=t,l \tag{1}$$

for the longitudinal magnetization modes, $\mathbf{M_q}\|\mathbf{q}$, when passing the line $\xi(T)^{-2}+q^2=q_d^2$. In contrast, the transverse modes $\mathbf{M_q}\perp\mathbf{q}$ remain critical to drive the ferromagnetic transition through their diverging correlation length $\xi(T)$. According to early renormalization-group work by Aharony and Fisher,[14] the boundary of this dipolar anisotropic critical region is determined by the dipole wave number $q_d^2=\lambda_d/J_2$, where $\lambda_d=\mu^2/v_0$ and J_2 denote the dipolar energy and the second moment of the exchange interaction, respectively. In fact, this static crossover from the isotropic to the dipole regime has been discovered on EuS using a polarized neutron technique.[5]

Another dramatic effect of the dipolar anisotropy has been predicted for the relaxation rate Γ_0 of the homogeneous magnetization $M_{q=0}$ above T_C. Approaching T_C in the isotropic (exchange dominated) critical region, Γ_0 should speed up proportional to $\chi_0^{3/4}$,[12,15–17] due to a reduction of the Anderson-Weiss exchange narrowing[23] whereas in the ultimate dipolar region, where the homogeneous susceptibility,

$$\chi_0\equiv(q_d\xi)^2\ , \tag{1a}$$

is large, $\chi_0\gg 1$, the thermodynamical slowing down $\Gamma_0\sim 1/\chi_0$ should take place.[13,15] This asymptotic behavior and also the crossover between the speeding up near $\chi_0=1$ [see Eq. (1)] has been first detected on EuS.[1] It was demonstrated[8] that these data were in quantitative agreement with numerical mode-coupling (MC) work for the dipolar Heisenberg ferromagnet[18] covering the full dynamical dipolar crossover.

The present paper is devoted to a first examination on the influence of a magnetic field on the relaxation rate of the parallel magnetization Γ_0 in the dipolar region above and below the Curie temperature of a Heisenberg ferromagnet. Basically, the effect of the field is (i) to break the symmetry of $\mathbf{M}_{q=0}$, (ii) to suppress the fluctuations of the magnetization components, $M^{\|}$ and $M^{\perp}$, parallel and perpendicular to $\mathbf{H}$, thus preventing the corresponding susceptibilities from criticality at T_C, and (iii) to induce a Larmor precession of $\mathbf{M_q}$ about the internal field H_i, which modifies the dynamics of $M^{\perp}_{\mathbf{q}}$. Usually, the so-called thermodynamic effect, which enters Γ_0 via the homogeneous susceptibility $\chi_0(T,H)$, is eliminated by considering the kinetic Onsager coefficient:[12,15–17]

$$L_0(T,H)\equiv\Gamma_0(T,H)\chi(T,H)=\frac{\lambda_d}{T}\int d\tau\langle\dot{S}^z_0(\tau)\dot{S}^z_0(0)\rangle\ . \tag{2}$$

This quantity only depends on the action of the fluctuations on the dipolar spin flipping mechanism, $\dot{S}^z=i[H_d,S^z]/\hbar$, since the spin-lattice coupling is much smaller.

In the isotropic critical region of the dipole Heisenberg ferromagnet $CdCr_2Se_4$ it was observed[24,25] that as soon as

the Larmor frequency, $\omega_L \equiv \gamma H_i$, exceeded the characteristic frequency, i.e., the decay rate of the critical fluctuations, $\Gamma^t_{q=\xi^{-1}} = D\xi(T)^{-2}$, the critical speeding up of Γ_0 was stopped. This suppressed the height of the $\Gamma_0(T,H)$ maximum and shifted it to higher temperatures from which, e.g., the spin-diffusion constant D could be extracted.[25,26]

For the dipole region above T_C, Maleev[15] argued that the Larmor motion will not influence L_0 as long as ω_L remains smaller than the dipole characteristic frequency, that is, the decay rate of the longitudinal fluctuations, $\Gamma^1_{q<q_d}$. We will check this conjecture and, moreover, we will also investigate the effect of temperature on L_0 on both sides of the Curie temperature in finite applied fields. To date, no detailed theory is available on the dynamics of the homogeneous susceptibility Heisenberg ferromagnets, in which a macroscopic magnetization is present. Such theory has also been called for existing data on iron.[27]

II. EXPERIMENTAL RESULTS

Using a UHF bridge technique described elsewhere,[28] the field dependence of the real part of the dynamic susceptibility has been measured at constant temperatures between 15 and 19 K and fixed frequencies from 1.6 MHz to 1.9 GHz. The amplitude of the rf field generated within helix resonators was kept small to guarantee the limit of the linear response. External magnetic fields were applied parallel to both the rf field and to the long axis of a needle shaped single crystal with demagnetization coefficient $N_z = 0.069$.

No indication of magnetic hysteresis appeared neither above nor below T_C. As examples, field sweeps taken at T_C are shown in Fig. 1(a). At all temperatures and fields the frequency dependence of the dispersion can be well described by the real part of a Debye function for $\omega \leq \Gamma_0$:

$$\chi'(\omega;T,H) = \frac{\chi(T,H)}{1+(\omega/\Gamma_0)^2} , \tag{3}$$

as exemplified by Fig. 1(b). These analyses define the static susceptibilities $\chi(T,H)$ of the equilibrium state to which the magnetization relaxes and the corresponding relaxation rates $\Gamma_0(T,H)$. With increasing field, $\chi(T,H)$ falls below the isothermal susceptibility $\chi_T(T,H)$ of EuS,[7] reaching a minimum of 0.8 χ_T at T_C. Using the thermodynamic relation $\chi_S^{-1} = \chi_T^{-1} - \mu_0(\partial H/\partial T)^2_M T/C_M$ and existing data on the specific heat C_M,[29] and the magnetic equation of state $H(M,T)$ for EuS,[7] we could identify $\chi(T,H)$ with the adiabatic susceptibility of the spin system. This reduction of χ to χ_S is due to the fact that the dipolar relaxation mechanism acts within the spin system only, while coupling to the lattice is achieved at much lower frequencies.

Slight deviations from the Lorentzian shape [Eq. (3)] appear near to the transition to the domain state about $H = N_z M_S(T)$. This region will not be considered here. However, it is perhaps worth noting that Shini and Hashimoto[4] observed similar deviations close to T_C of EuS in zero field, who assigned the non-Lorentzian shape to nonlinear dynamics of unknown origin. Since the static zero-field susceptibility deviates from the critical power law in the same temperature interval, $(T - T_C) = 0.002\ T_C$, we attribute these deviations to sample inhomogeneities smearing the transition and the relaxation times. The magnetization dynamics within the domain state $H < NM_s$ is being investigated separately.[10]

From the fitted susceptibilities and relaxation rates we evaluate the kinetic Onsager coefficient $L_0(T,H)$ [Eq. (2)], being the quantity of central interest for the magnetization dynamics. Due to the definition, $L_0 = \lim_{\omega\to 0} i\omega/[\chi(\omega)^{-1} - \chi(0)^{-1}]$,[30] L_0 represents a material constant being independent of sample shape and depending only on intrinsic dynamical processes as outlined above. Figure 2(a) demonstrates that approaching T_C from above, the applied field increasingly suppresses the kinetic coefficient, whereas below the Curie temperature L_0 is rising again. Since L_0 is predicted to depend on the internal magnetic field $H_i = H - N_z M$ rather than

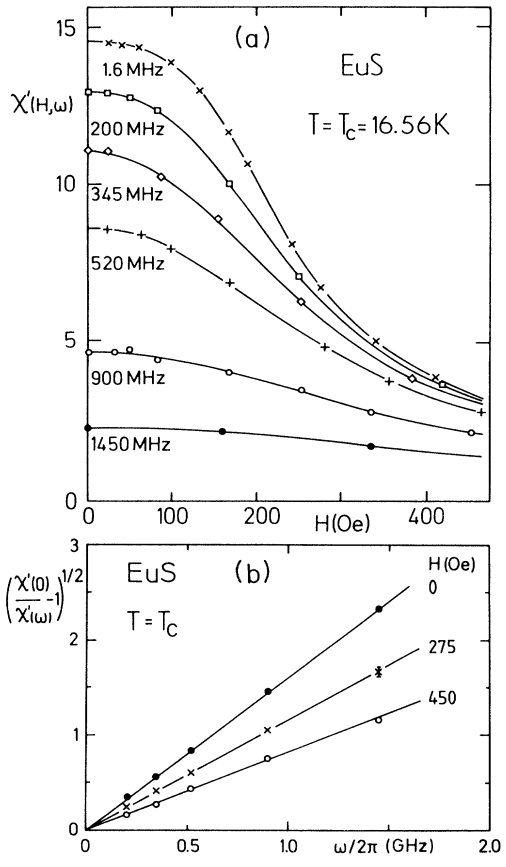

FIG. 1. (a) Dispersion of the dynamic susceptibility of an EuS ellipsoid ($N_z = 0.069$) between 1.6 and 1450 MHz at T_C in external fields $\mathbf{H} \| [100]$ up to 500 Oe. Solid lines are drawn as guides to the eye. (b) Lorentz analyses of $\chi'(\omega)$ at $T = T_C$ in different external fields using Eq. (2); the slopes define the inverse relaxation rates Γ_0^{-1}.

on the applied field H,[30] it is suggestive to associate this recovery of L_0 at given H in Fig. 2(a) with the disappearance of H_i near the transition to the domain state.

A very appealing phenomenological description of the effects of field and temperature on the kinetic coefficient can be achieved by analyzing them in terms of the internal isothermal susceptibility, $\chi_T^i=(\chi_T^{-1}-N_z)^{-1}$. Figure 2(b) clearly demonstrates the interesting feature that all data from the dipolar critical regimes on the paramagnetic side of T_C, and also some from the ferromagnetic (monodomain) side close to T_C, collapse on a single curve. This implies that both the temperature and field dependences of L_0 are fully accounted by $\chi_T^i(T,H)$, being discussed in more detail in the following section.

Another interesting feature emerging from Fig. 2(a) is that within the uncertainty of the data interpolation, the kinetic coefficients assume the same value, $L_0(T_d,H_i)=L_0(T_C,O)\simeq 61$ GHz, at temperatures T_d, where the demagnetizing field of the spontaneous magnetization is just reaching the applied field, i.e., $N_zM(T_d)\equiv H$. There one expects the sample to form domains setting the macroscopic internal field to zero at all temperatures below T_d. It is noteworthy that very recently Kapoor[31] found the same value, $L_0=60(5)$ GHz, on two different EuS samples ($N_z=0.1$ and 0.33) at the domain boundary for 4.2 K, which indicates that L_0 does not vary for all $T\leq T_C$.

Finally we also note that the susceptibility scaling of the kinetic coefficient ceases to be valid near T_d. This feature is accompanied by a breakdown of the static magnetization scaling.[7] Since as a rule critical dynamics reacts much more sensitively than static critical behavior to perturbations, the violation of the susceptibility scaling for the dynamical property L_0 is not unexpected.[32]

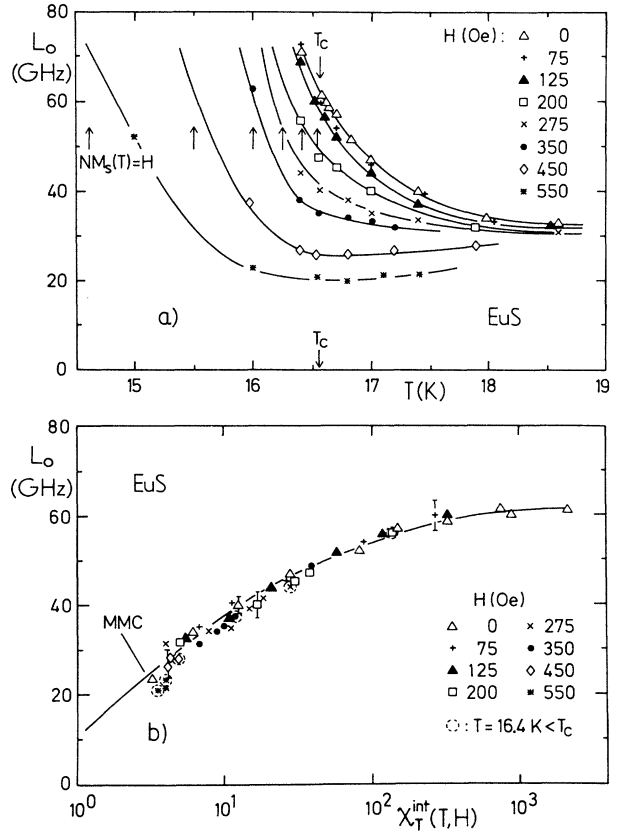

FIG. 2. Kinetic coefficient of the homogeneous magnetization, L_0, (a) about $T=T_C=16.56$ K in different external fields; arrows mark the transitions to the domain state (M_s=spontaneous magnetization); solid lines are drawn as guides to the eye, interpolating to data beyond 80 GHz. (b) Data from (a) vs the internal isothermal susceptibility $(\delta M/\delta H_i)_T=\chi_T^{int}$. Results from the monodomain state below T_C are encircled; the solid line represents the mode-mode coupling (MMC) calculation (Ref. 18) for the kinetic coefficient at zero field above T_C.

III. DISCUSSION AND CONCLUSIONS

First of all, we recall the essential feature of the scaling property of the kinetic coefficient in Fig. 2(b), which includes the zerofield data for $T\geq T_C$. Since the latter are in quantitative agreement with the prediction of the MC theory by Frey and Schwabl,[18] one has to answer the question why the presence of a finite magnetization does not change the susceptibility scaling of L_0 within our range of H-T values about the Curie temperature. To follow this line, we begin with summarizing the physics behind the MC result valid for the paramagnetic regime and $H=0$.

In the Lorentzian limit appropriate to our experimental results, the kinetic coefficient of the homogeneous magnetization is generally defined by Eq. (2). The dipole-dipole interaction generates the torques on S_0^z,[15]

$$\dot{S}_0^z(\tau)=-\frac{\lambda_d}{\hbar}\sum_{\mathbf{q}}S_{\mathbf{q}}^l(\tau)\left[\frac{q^\mu}{q}\epsilon_{\mu z\nu}S^\nu_{-\mathbf{q}}(\tau)\right], \tag{4}$$

where $\epsilon_{\mu z\nu}$ denotes the Levi-Civita tensor. Generally, the MC approximation is based on the factorization of the four spin correlation functions in Eq. (4), which considers only two mode couplings and results in

$$L_0=\left[\frac{\lambda_d}{\hbar}\right]^2\int_0^\infty d\tau\sum_{\mathbf{q}}\chi_{\mathbf{q}}^{(l)}(\tau)\chi_{\mathbf{q}}^t(\tau)\ . \tag{5}$$

If there is no magnetization, usually an exponential decay of both the longitudinal and transverse modes can be assumed, $\chi_{\mathbf{q}}^\alpha(\tau)=\chi_{\mathbf{q}}^\alpha\exp(-\Gamma_{\mathbf{q}}^\alpha\tau)$, leading immediately to

$$L_0=\left[\frac{\lambda_d}{\hbar}\right]^2\sum_{\mathbf{q}}\frac{\chi_{\mathbf{q}}^l\chi_{\mathbf{q}}^t}{\Gamma_{\mathbf{q}}^l+\Gamma_{\mathbf{q}}^t}\ . \tag{5a}$$

Using the scaling properties of $\chi_{\mathbf{q}}^l$ and $\chi_{\mathbf{q}}^t$ [Eq. (1)] and of the relaxation rates $\Gamma_{\mathbf{q}}^l$ and $\Gamma_{\mathbf{q}}^t$ in Eq. (5), the $\mathbf{q}$ summation can be carried out to give[18]

$$L_0(T)=L_d[1+(q_d\xi)^{-2}]^{-7/4}\ . \tag{6}$$

While the term within the brackets accounts for the dramatic dynamic dipolar crossover outlined in the In-

troduction, the prefactor

$$L_d = 0.83\gamma \left[\frac{kT_C q_d^3}{\mu_0} \right]^{1/2} \Lambda \tag{6a}$$

depends only slightly on temperature via the tabulated function $\Lambda[q_d\xi(T)] \leq 1$ with $\Lambda(T_C)=1$.[33] This result is depicted by the solid line in Fig. 2(b). Physically, this crossover to the thermodynamic slowing down, L_d=const., close to T_c is associated with the saturation of both the static susceptibility $\chi_{\mathbf{q}}^l$ and the relaxation rate $\Gamma_{\mathbf{q}}^l$ of the longitudinal magnetization modes, whereas those of the transverse modes remain critical. Both saturation effects have been observed on EuS by elastic[6] and inelastic[11] scattering of polarized neutrons.

In the presence of a magnetization, $\mathbf{M}=M\mathbf{e}_z$, none of this information is available in the dipolar region, neither of EuS nor of similar ferromagnets. Basically, in the torque [Eq. (4)], one has to differentiate between contributions from spin fluctuations parallel ($\delta S_{\mathbf{q}}^{\parallel}$) and perpendicular ($\delta S_{\mathbf{q}}^{\perp}$, spin waves) to $\mathbf{M}$. As regards the dynamics of their susceptibilities, only the spin waves acquire an additional Larmor term leading to $\Gamma_{\mathbf{q}}^{\alpha,\perp} = (L_{\mathbf{q}}^{\alpha,\perp} + i\gamma M)/\chi_{\mathbf{q}}^{\alpha,\perp}$, where $\alpha = l,t$. Since in Eq. (5a), the denominator in the sum is governed by the fast longitudinal decay, γM has to be compared to the kinetic coefficient of these longitudinal modes. To give an example, near T_C we infer from the zero-field data $L_{\mathbf{q}}^l = 61$ GHz, which corresponds to $M=3$ kOe, being larger than the maximum magnetization of 2 kOe in the experiment. Hence, it is perhaps plausible to assume that a dynamic effect of the magnetization on L_0 is absent. The static susceptibilities of the parallel and spin-wave fluctuations evolve from Eq. (1) by introducing different correlation lengths $\xi^{\parallel}$ and $\xi^{\perp}$ (see, e.g., Ref. 34). While in the dipolar region above and below T_C both susceptibilities are not known, they recently have been calculated numerically at low temperatures.[35] The scaling property of $L_0(T,H)$ in terms of $\chi_T(T,H) = [q_d\xi_{\parallel}(T,H)]^2$ implies that in the $\mathbf{q}$ summation in Eq. (5a) the difference between the parallel and spin-wave susceptibilities does not play a role, at least within the dipolar region probed here. Summarizing, the field effect on the kinetic coefficient can completely be described by inserting the field-dependent parallel susceptibility $\chi_T^i = \delta M/\delta H_i = (q_d\xi)^2$ into Eq. (6).

Of course, these rather general arguments are not completely satisfying, and a theoretical analysis, like an extension of the numerical MC approach[18] to finite magnetizations, is highly demanded. Perhaps such theory can also explain the other interesting feature indicating that below T_C near the phase boundary, i.e., at almost zero internal field, the kinetic coefficient acquires the same value as at T_C. It would be interesting to see whether this result can be related to one or the other of the two correlation lengths, $\xi_{\parallel}$ and $\xi_{\perp}$. Both are expected to diverge in zero internal field for all temperatures below T_C,[34] provided any crystalline anisotropy can be ignored. To check this experiment in the dipolar region of a real ferromagnet like EuS, neutron diffraction measurement of the parallel and spin-wave susceptibilities would be also of great value. If, e.g., below T_C, they reveal large correlation lengths, $\xi_{\parallel}, \xi_{\perp} \gg q_d^{-1}$, the magnetization dynamics would be dominated by the dipolar anisotropy in the whole ferromagnetic phase.

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