# Magnon dispersion of the dipolar ferromagnet EuS near the zone center

P. Böni

Labor für Neutronenstreuung, Eidenössische Technische Hochschule und Paul Scherrer Institut, CH-5232 Villigen PSI, Switzerland

M. Hennion

Laboratoire Léon Brillouin, Centre d'Etudes Nucléaires de Saclay, F-91191 Gif-sur-Yvette Cedex, France

J. L. Martínez

Consejo Superior de Investigaciones Científicas, Instituto de Ciencia de Materiales, Facultad de Ciencias Cantoblanco,

E-28049 Madrid, Spain (Received 24 February 1995)

The dispersion of the spin waves in EuS has been investigated by means of neutron scattering. In zero field and at small momentum transfer  $\mathbf{q}$ , the energy  $\hbar\omega_q$  of the spin waves is proportional to q in agreement with the predictions of spin-wave theory that includes demagnetization effects. By application of a magnetic field H || q, the spin waves attain a finite mass, i.e., a gap is induced and  $\hbar\omega_q$  becomes proportional to  $q^2$ . Using polarization analysis we show that the degeneracy between spin waves at  $0.87T_c$  polarized along and transverse to  $\mathbf{q}$  is lifted due to the depolarizing fields. The transverse spin-wave modes are the Goldstone modes and diverge at  $T_c$ , whereas the longitudinal spin waves do not diverge. This situation resembles the lifting of the degeneracy between the longitudinal and transverse fluctuations in the

paramagnetic phase. Both the dynamics and the statics are in agreement with linear spin-wave theory.

# I. INTRODUCTION

The spin-wave dispersion curves of many isotropic ferromagnets have been investigated over large areas of the Brillouin zone using various neutron-scattering techniques. The measured dispersion curves from insulating materials like EuS are reasonably well understood in terms of localized magnetic moments,<sup>1</sup> whereas band models have been proven to be rather successful in describing the spin dynamics in itinerant ferromagnets like Fe and Ni at low temperatures.<sup>2</sup> Moreover, it was demonstrated convincingly that the renormalization of the spin waves close to  $T_c$  follows the laws of dynamical scaling.<sup>3,4</sup>

Interestingly the critical behavior of true isotropic ferromagnets cannot be investigated in the limit  $q \rightarrow 0$  because the magnetic moments induce (long-range) anisotropic dipolar fields that decay like  $1/r^3$  and dominate the exchange interactions at small q. As a measure of the strength of the dipolar interactions one can define a dipolar wave number  $q_d$  that can be inferred from the inverse correlation length  $\kappa(T)$  and the homogeneous internal susceptibility above  $T_c$ ,  $\chi(q=0,T)$ , via the relation  $q_d^2 = \kappa^2 \chi(0,T)$  or by equating the dipolar width of the spin-wave band to the spin-wave energy at  $q_D$ ,  $g\mu_B\mu_0M = Dq_D^2$ . Here g is the gyromagnetic ratio,  $\mu_B$  is Bohr's magneton,  $\mu_0$  is the induction constant, M(T,H)is the magnetization, and D is the stiffness constant. If the dipolar interactions are included in the Hamiltonian for a Heisenberg model one arrives at the following dispersion relation for the spin-wave excitations:<sup>5,6</sup>

$$\hbar\omega_q = \{(E_q + g\mu_B\mu_0 H)[E_q + g\mu_B\mu_0 H]$$

$$+g\mu_B\mu_0 M(T,H)\sin^2\theta_q]\}^{1/2}.$$
(1)

Here  $E_q$  designates the exchange energy, H the magnetic field, and  $\theta_q$  is the angle between **M** and the momentum transfer with respect to the nearest Bragg peak, **q**. The term containing H is responsible for the Zeeman splitting  $E_H$ , and the term containing M is due to the dipolar interactions. In most ferromagnets, like Ni  $[q_d \simeq 0.013 \text{ Å}^{-1}$  (Ref. 7)], the dipolar energy  $E_d = g\mu_B\mu_0 M(T,H)$  is so small that it has not been directly observed in neutron scattering. By contrast in EuS,  $q_d = 0.25 \text{ Å}^{-1}$  is large and dipolar effects have been included in the data analysis,<sup>1,8</sup> although the effects were small, because the spin-wave measurements have only been performed at such large q that the dipolar contribution appeared as a constant energy, i.e., a "dipolar gap" (like a Zeeman gap), added to the exchange energy  $E_q$ (see Fig. 6 in Ref. 3). However, the more complicated qdependence of the dispersion curve given by Eq. (1) has not been determined so far.

Another important consequence of the dipolar interactions is the lifting of the isotropy of the paramagnetic fluctuations close to  $T_c$ . Here the dipolar fields prevent the longitudinal (long-wavelength) fluctuations ( $\delta S || q$ ) from criticality, while the transverse fluctuations ( $\delta S || q$ ) diverge.<sup>9</sup> Moreover the longitudinal fluctuations are strongly damped.<sup>10</sup> We have observed similar effects in EuS below  $T_c$  namely the longitudinal spin waves and the

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transverse spin waves were not identical.<sup>11,12</sup>

The goal of the present paper is to show conclusively that the behavior of the transverse spin waves  $(\delta S \perp q)$  is in accordance with spin-wave theory that includes dipolar interactions.<sup>6</sup> In particular we show that  $\hbar \omega_q$  depends linearly on q in zero field for  $q \rightarrow 0$ , whereas for reasonably large H a gap opens at  $q \simeq 0$ . Using polarization analysis we show that the spin waves with transverse and longitudinal polarization have different cross sections similar as for the paramagnetic fluctuations above  $T_c$ .

# **II. EXPERIMENT**

The neutron-scattering experiments were performed on the triple-axis spectrometer 4F1 located at the cold source of the Orphée reactor at the Laboratoire Léon Brillouin in Saclay. The isotopically enriched sample <sup>153</sup>EuS was composed of roughly 100 single crystals, aligned such that the overall mosaic was  $\eta \simeq 0.8^{\circ}$ . It was mounted inside a closed-cycle cryostat mounted between the pole pieces of an electromagnet. Most unpolarized measurements were conducted near [000] and [200], along the [100] direction with  $\mathbf{q} \| \mathbf{H}_{ext}$ , using neutrons with fixed incident energies  $2.28 \le E_i \le 4.06$  meV and various collimations depending on the conflicting requirements of resolution and intensity. The polarized beam measurements were performed near [200] along the [100] and [010] directions in a vertical field  $\mathbf{H}_{ext} \perp \mathbf{q}$  of 164 mT. The flipping ratio was 10, i.e., the polarization of the instrument was 0.82.

In zero external field the sample is composed of domains pointing along the [111] easy direction. Hence  $\langle \sin^2 \theta_q \rangle = \frac{2}{3}$ . The anisotropy field at T = 0 K is  $\mu_0 H_{111} \simeq 2.5$  mT and its influence on the spin dynamics

can be neglected. The saturation magnetization of EuS is  $\mu_0 M = 1.53T$ .<sup>1</sup>

#### **III. THEORY**

In this section we follow the development of linear spin-wave theory by Lovesey for a Heisenberg ferromagnet.<sup>6,13</sup> The dispersion relation for an isotropic ferromagnet is given by

$$\epsilon_q = g\mu_B\mu_0 H + 2S[J(0) - J(\mathbf{q})] . \tag{2}$$

 $J(\mathbf{q})$  is the spatial Fourier transform of the exchange interactions. If dipolar interactions are added to the Hamiltonian the dispersion relation becomes

$$i\omega_q = (A_q^2 - |B_q|^2)^{1/2} = [\epsilon_q(\epsilon_q + 2|B_q|)]^{1/2},$$
 (3)

where

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$$A_q = \epsilon_q + |B_q| \tag{4}$$

and  $B_q$  is given for all but the extreme value q=0 (Ref. 14) by

$$B_a = \frac{1}{2}g\mu_B\mu_0 M(T,H)\sin^2\theta_a \exp(-2i\phi_a) .$$
 (5)

The angles  $\theta_q$  and  $\phi_q$  define the orientation of **q** in Cartesian coordinates taking 0z along the easy axis of magnetization, e.g.,  $q_x = |\mathbf{q}| \sin \theta_q \cos \phi_q$ . Similarly one can define the orientation of the scattering vector  $\mathbf{Q} = \tau + \mathbf{q}$  with respect to the easy axis of magnetization by means of the angles  $\theta$  and  $\phi$ , e.g.,  $Q_x = |\mathbf{Q}| \sin \theta \cos \phi$ , where  $\tau$  is a reciprocal-lattice vector.

The cross section for a dipolar ferromagnetic has been derived by Lovesey<sup>13</sup> starting from the work of Keffer<sup>15</sup> for unpolarized neutrons:

$$\frac{d^{2}\sigma}{d\Omega dE'} = r_{0}^{2} \frac{k'}{k} \left[ \frac{1}{2} gF(\mathbf{Q}) \right]^{2} \exp[-2W(\mathbf{Q})] \\ \times \frac{S}{2} \frac{(2\pi)^{3}}{v_{0}} \sum_{\mathbf{q},\tau} \left[ (1 + \hat{Q}_{z}^{2}) \frac{A_{q}}{\hbar \omega_{q}} + (1 - \hat{Q}_{z}^{2}) \frac{B_{q}}{\hbar \omega_{q}} \cos 2(\phi - \phi_{q}) \right] \\ \times n_{q} \delta(\hbar \omega + \hbar \omega_{q}) \delta(\mathbf{Q} + \mathbf{q} - \tau) + (n_{q} + 1) \delta(\hbar \omega - \hbar \omega_{q}) \delta(\mathbf{Q} - \mathbf{q} - \tau) .$$
(6)

 $\hat{Q}_z$  is the projection of  $\mathbf{Q}/|\mathbf{Q}|$  on the direction of magnetization. For the rest of the notation see Ref. 13. Equation (6) is identical with the expression for an isotropic ferromagnet if  $B_q$  is set equal to 0. The dipole forces greatly complicate the angular dependence of the intensity of the scattered neutrons.

Starting from the expression, Eq. (6), we consider expressions for the scattered intensity for some experimental situations of interest and in the limit  $\hbar\omega \ll k_B T$ . In the following N is a constant.<sup>16</sup>

(1) We discuss first two geometries with **M** perpendicular to the scattering plane  $(1+\hat{Q}_z^2=1-\hat{Q}_z^2=1)$ . The in-

tensity of neutrons scattered by spin waves with transverse polarization  $(\delta S \perp q)$  is obtained by setting q ||Q| in Eq. (6):

$$I_T(q) = N \frac{k_b T}{\epsilon_q} . \tag{7}$$

 $I_T(q)$  is identical with the expression for a ferromagnet without dipolar interactions and it diverges in zero field like  $1/q^2$ . These are the Goldstone modes. By setting  $q \perp Q$  one obtains, for the longitudinal spin waves ( $\delta S \parallel q$ ), 10 144

$$I_L(q) = N \frac{k_b T}{\epsilon_q + 2|B_q|} . \tag{8}$$

 $I_L(q)$  is smaller than  $I_T(q)$  and does not diverge for  $q \rightarrow 0$  even in zero field as long as  $B_q \neq 0$ . Although the spin-wave energy  $\hbar \omega_q$  is independent of the polarization  $(\theta_q = 90^\circ)$ , the denominators are in neither case given by  $\hbar \omega_q$  as might be expected in analogy to an isotropic ferromagnet.

(2) Next we discuss the situation for a sample in zero field  $(\langle \sin^2 \theta_q \rangle = \frac{2}{3})$  with  $\mathbf{q} || \mathbf{Q}$ . Here  $\phi = \phi_q$ :

$$I_{\rm up}(q) = N \frac{k_B T}{\hbar \omega_q} \frac{4A_q + 2B_q}{3\hbar \omega_q} \ . \tag{9}$$

In an applied vertical field Eq. (9) evolves into Eq. (7), i.e., from the multidomain structure to complete field alignment. Note that  $I_i(q)/k_b T$ , i = T, L, is proportional to the wavelength-dependent susceptibility  $\chi_{sw}^i(q)$ .

#### **IV. RESULTS**

# A. Unpolarized neutron scattering

Figure 1 shows the temperature dependence of the spin waves at  $(0.12\ 00)$  at  $T = 12.5\ K\ (q = 0.125\ Å^{-1})$ . The elastic background, mostly due to incoherent scattering from the sample, and the sample holder have been determined at 75, 125, 181, and 240 K and have already been subtracted. With increasing field the spin-wave peaks move initially to smaller energy and then to higher energy. This behavior cannot be simply explained in terms of a Zeeman shift that would always increase the energy of the spin waves with increasing field.

In order to shed more light on the physics behind we have extended the measurements to smaller and to larger q. Because of the complicated form of the scattering cross section, Eq. (6) the measured spectra have been fitted with a scattering function  $S(q,\omega)$  appropriate for an isotropic ferromagnet, convoluted with the four-dimensional resolution function of the spectrometer:

$$S(q,\omega) = \frac{4}{3} N_{\rm iso} \frac{k_B T}{\hbar \omega_q} \frac{1}{2\pi} \left[ \frac{\Gamma_q}{(\hbar \omega - \hbar \omega_q)^2 + \Gamma_q^2} + \frac{\Gamma_q}{(\hbar \omega + \hbar \omega_q)^2 + \Gamma_q^2} \right].$$
(10)

 $\chi^2$  was minimized by varying the constant  $N_{\rm iso}$ ,  $\hbar\omega_q$ , and  $\Gamma_q$ . Any departure of the data from the behavior of an isotropic ferromagnet is revealed in a q dependence of  $N_{\rm iso}$  as well as in changes of the dispersion curve. It became apparent that the spin waves are not damped within the resolution limits of the spectrometer, hence, linear spin-wave theory is applicable.

The resulting spin-wave dispersions are shown in Fig. 2. In zero-field,  $\hbar\omega_q$  has definitely not a parabolic shape as expected for the exchange part of an isotropic ferromagnet at small q. With increasing field, a gap opens up near q=0 and the dispersion curves become more parabolic. Figure 3 shows that our zero-field data are



FIG. 1. Field dependence of transverse spin waves at  $\zeta = 0.12$ and  $T = 0.72T_c$ . With increasing field, the peaks move initially to smaller energies, then to higher energies. 10 mon corresponds to a counting time of 4.1 min.



FIG. 2. Field dependence of the dispersion for transverse spin waves at different temperatures. Note the almost linear dependence of the dispersion at small q in zero field (thick solid lines).

consistent with the data of Bohn *et al.*, that were obtained at T = 12 K from a sample that was assembled from the same crystals.<sup>1</sup> The solid line in Fig. 3 represents a fit to the data using as parameters the exchange constants  $J_1$  and  $J_2$  and the dipolar energy. The latter value is  $E_d \sin^2 \theta_q = 0.087 \pm 0.009$  meV, which is in excellent agreement with the calculated value  $E_M = 0.088$  meV (from the bulk magnetization at 12 K, and  $\sin^2 \theta_q = \frac{2}{3}$ ).

The interpretation of the data in the light of Eq. (1) is straightforward. With increasing field the magnetic domains align progressively along the momentum transfer  $\mathbf{q}=\mathbf{Q}$ . Therefore the dipolar term, containing  $\sin^2\theta_q$  decreases. Because of depolarization effects, however, the internal field H remains initially zero and no Zeeman gap is induced. As soon as  $\mathbf{M} || \mathbf{q}$ , the demagnetization effects vanish and H increases.

To put this scenario on a more quantitative basis we have fitted the dispersion curves directly to the expression  $\hbar \omega_q = [(Dq_2 + E_H) \cdot (Dq^2 + E_H + E_d \sin^2 \theta_q)]^{1/2}$  treating the stiffness D, Zeeman gap  $E_H$ , and the dipolar "gap"  $E_d \sin^2 \theta_q$  as fitting parameters. The fits (Table I) indicate that  $E_H$  is zero within error bars for  $H^{\text{ext}} \le 72$ mT whereas  $E_d = 0$  for 420 mT. In particular, at small qand in zero field the spin-wave energy extrapolates to 0



FIG. 3. Dispersion of EuS for  $0.72T_c$  for **q** along the [100] direction and H=0. The solid circles are taken from Bohn *et al.* (Ref. 1) and the open circles are from this work. The solid line is a fit to the data, including nearest and next-nearest exchange interactions and the dipolar term.

for  $q \rightarrow 0$ . From the fits we deduce a stiffness  $D = 2.31 \pm 0.10$  meV Å<sup>2</sup>. D does not depend on field because H is rather small. For large H, however, D can become field dependent.<sup>17</sup>

Additional measurements were performed at  $0.96T_c$ and  $0.965T_c$  in zero field. The spin waves renormalize, as expected, and the dipolar gap decreases. The values  $\sin^2\theta_q (\neq \frac{2}{3})$  have been obtained from the measured dipolar contribution to  $\omega_a$ .

The reduced magnetization  $m = M/M_0$  of EuS was already measured many years ago, using neutron diffraction.<sup>18</sup> We deduce  $m \simeq 0.746$ , 0.401, and 0.353 for T = 0.72, 0.95, and  $0.965T_c$ , respectively.<sup>19</sup> Using  $M_0 = 1.53$  T from Ref. 1 we obtain for the dipolar energies (in zero field),  $E_d \sin^2 \theta_q$ , the values 0.088, 0.047, and 0.042, meV, respectively. The values for 0.72 and 0.965 $T_c$  are reasonably close to the experimental results (Table I) whereas at  $0.95T_c$  there is a disagreement, maybe due to the combined effects of errors in temperature and in determining the rather small energies.

Finally we discuss the Zeeman splitting of the spin waves. The effective field  $H^c$  deduced from  $E_H$  is systematically lower than  $H^{ext}$ , as expected due to the demagnetizing field effects. The difference

TABLE I. Stiffness D, Zeeman gap  $E_H$ , and dipolar contribution  $E_d \sin^2 \theta_q$  vs the external field  $H^{\text{ext}}$  ( $1J = 6.241 \times 10^{21}$  meV).  $\chi^2$  designates the quality of the fit.

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$T (T_c)$	$\mu_0 H^{\text{ext}}$ (mT)	$D (meV Å^2)$	$E_H$ (meV)	$E_d \sin^2 \theta_q$ (meV)	$\sin^2 \theta_q$	χ²
0.72	0	2.35±0.15	fixed at 0	0.077 = 0.010	$\frac{2}{3}$	1.03
0.72	72	$2.33 {\pm} 0.11$	$0.005 {\pm} 0.006$	$0.018 {\pm} 0.017$	0.16	0.87
0.72	144	$2.26{\pm}0.08$	$0.011 {\pm} 0.002$	$0.012 {\pm} 0.002$	0.10	0.76
0.72	420	$2.30{\pm}0.05$	$0.041 {\pm} 0.002$	$0.000 {\pm} 0.000$	0	1.08
0.72	560	fixed at 2.31	$0.062 {\pm} 0.002$	fixed at 0	0	
0.95	0	1.19±0.14	fixed at 0	$0.032{\pm}0.009$	$\frac{2}{3}$	2.09
0.965	0	$0.83{\pm}0.16$	fixed at 0	$0.040 {\pm} 0.013$	$\frac{2}{3}$	0.36



FIG. 4. Field and q dependence of the constant  $N_{\rm iso}$  that is related to the energy integrated spin-wave intensity at  $0.72T_c$ . The solid lines are calculations as explained in the text.

 $\mu_0(H^{\text{ext}} - H^c) = 57 \text{ mT}$  can be obtained with

$$\mu_0 M(0.72T_c, H) \simeq \mu_0 M(0.72T_c, 0) = 1.14 \text{ T},$$

and a demagnetizing factor  $D_N = 0.05$ . This latter value is reasonable for our platelike single crystals.

Figure 4 shows the q dependence of the fitted constant  $N_{\rm iso}$  [Eq. (10)]. In zero field (solid circles)  $N_{\rm iso}$  decreases slowly with decreasing q. With increasing field the fluctuations are suppressed at small q and  $N_{\rm iso}$  decreases. In contrast, with increasing H,  $N_{\rm iso}$  is enhanced at large q with respect to H=0, because the magnetic domains align progressively along H, hence  $1+\hat{Q}_z^2$  increases from 1 to 2 [see Eq. (6)].

We explained above that because of the complex angular dependence of the cross section it is impossible to fit the measurements directly to Eq. (6). However, in order to demonstrate the internal consistency of our data we have calculated the expected constant  $N_{\rm iso}$  using Eq. (9) on the basis of the numerical values quoted in Table I and compared them in Fig. 4 with the experimentally determined values. Note that there is only one free parameter involved in this calculation, namely an overall scaling factor. It is gratifying that the agreement between calculation and experiment is so excellent.

#### **B.** Polarized neutron scattering

As already mentioned in the Introduction the dipolar anisotropy lifts the degeneracy of the longitudinal and transverse spin fluctuations above  $T_c$ . In order to detect such effects below  $T_c$  we have performed polarized neutron scattering in the ferromagnetic phase at 0.79, 0.87, and  $0.98T_c$ . Note, that all spin-wave scattering is spin flip because the external field was chosen to be perpendicular to the scattering plane. Figure 5 shows some measurements performed at  $(2-\zeta 00)$  and  $T=0.87T_c$  in an external field of 164 mT. The polarization of the spin waves is transverse to **q** because  $\mathbf{Q} || \mathbf{q}$ . An elastic peak with an amplitude of 4 counts/7.5 M has already been subtracted from the data.

The deconvolution of the data (solid lines) on the basis of Eqs. (6) and (7) yields spin-wave energies  $\hbar\omega$  that are compatible with the unpolarized beam data presented above, when the renormalization of the spin waves is properly taken into account. Note that we have included during the fitting procedure the Zeeman splitting  $E_H = 0.014$  meV, caused by the internal field  $\mu_0 H \simeq 120$ mT. Moreover we have used a unique normalization constant N for the three measurements.

In contrast, the data sets for the longitudinal spin



FIG. 5. Transverse spin waves measured at  $(2-\zeta 00)$  in a vertical field of 164 mT and  $T=0.87T_c$ . The solid lines are fits to the data, including the four-dimensional instrumental resolution function. 7.5 mon corresponds to a counting time of 7 min.

TABLE II. q dependence of the different magnetic modes in a dipolar ferromagnet.  $\kappa_z$  and  $\kappa$  are the inverse correlation lengths below and above  $T_c$ , respectively. The term followed by a question mark is speculative.

M · · · q	$T < T_c$	$T = T_c$	$T > T_c$
<b>M</b>   q	$\propto \frac{2\chi_{sw}^{T} + \chi_{z}^{L}}{\alpha^{2} + \frac{1}{\alpha^{2} + \mu^{2} + \alpha^{2}}} $ (?)	$\frac{2\chi_p^T + \chi_p^L}{\propto \frac{2}{\alpha^2} + \frac{1}{\alpha^2 + \alpha^2}}$	$\propto \frac{2\chi_{p}^{T} + \chi_{p}^{L}}{\alpha^{2} + \alpha^{2}} + \frac{1}{\alpha^{2} + \alpha^{2} + \alpha^{2}}$
M⊥q	$ \begin{array}{c} q  q  + \kappa_z + q_D \\ \chi_{sw}^T + \chi_{sw}^L + \chi_z^T \\ \propto \frac{1}{2} + \frac{1}{2 + 2} + \frac{1}{2 + 2} \end{array} $	$\frac{q}{2\chi_p^T + \chi_p^L} \propto \frac{2}{2} + \frac{1}{2}$	$\alpha \frac{2}{2} + \frac{1}{2} + $
	$\propto \frac{1}{q^2} + \frac{1}{q^2 + q_D^2} + \frac{1}{q^2 + \kappa_z^2}$	$\propto \frac{1}{q^2} + \frac{1}{q^2 + q_d^2}$	$\propto \frac{1}{q^2 + \kappa^2} + \frac{1}{q^2 + \kappa^2}$



FIG. 6. Longitudinal spin waves measured at  $(2\zeta 0)$  in a vertical field of 164 mT and  $T=0.87T_c$ . The solid lines have been calculated using linear spin-wave theory, including the dipolar interactions. The other lines are explained in the text. 22.5 mon corresponds to a counting time of 21 min.

waves measured at  $(2\zeta 0)$  have a rather different shape (Fig. 6). The same background as in Fig. 5 has been subtracted from the data. The most significant difference to the transverse data is the enhanced intensity at E = 0 that increases even slightly with decreasing T (not shown). It is most likely caused by spurious nuclear and magnetic scattering from the [200] Bragg peak due to the still rather large mosaic of the sample. In fact the nonspin-flip scattering at (20.120) and E = 0 has an amplitude of the order of 510 counts, or more. Therefore due to the not perfect polarization of the neutron beam,  $P \simeq 0.82$ , at least 50 counts contribute to the spin-wave spectrum. At larger q the contributions are smaller.

In order to show that the intensity of the scattered neutrons depends on the polarization of the spin waves, we have used the fit parameters  $\hbar\omega$  and N from the transverse spectra (Fig. 5) and calculated the expected intensities for the longitudinal spin waves for three different models.<sup>20</sup> Note, that  $\hbar\omega_q$  does not depend on the polarization of the spin waves within the framework of linear spin-wave theory, since  $\theta_q = 90^\circ$ .

(1) If we assume that EuS is isotropic, i.e.,  $B_q = 0$ , then the calculated intensity for the longitudinal spin waves is too large (dashed line in Fig. 6). Therefore EuS is indeed anisotropic, in contrast to a weak dipolar  $[q_d = 0.01 \text{ Å}^{-1}$ (Ref. 21)] ferromagnet such as Pd<sub>2</sub>MnSn.<sup>11</sup>

(2) If we use the dipolar wave number  $q_D = 0.18$  Å<sup>-1</sup> from Lovesey<sup>6</sup> that is based on Refs. 8 and 15 then the calculated and the fitted spectra agree reasonably well (solid line in Fig. 6).

(3) If we use the dipolar wave number  $q_d = 0.245$  Å<sup>-1</sup> from measurements of the paramagnetic fluctuations,<sup>10</sup> then the predicted intensity is too small (thin dashed line).

The simulations indicate that the longitudinal spectra can be well described by spin-wave theory, when a dipolar wave number  $q_D \simeq 0.18$  Å<sup>-1</sup> is used. This number is smaller than  $q_d$  as determined in the paramagnetic phase, however, it is roughly compatible with the number  $q_D = (E_d/D)^{1/2} = 0.22 \pm 0.02$  Å<sup>-1</sup> as determined from the unpolarized beam experiments (Table I).

# V. DISCUSSION

We have shown in the previous section that the dipolar interactions have important effects on the spin-wave dispersion and the magnetic scattering intensities of an isotropic ferromagnet. In particular, the zero-field dispersion relation becomes linear for  $q \rightarrow 0$ , in agreement with theory<sup>5,6</sup> [Eq. (1)] and the intensities of the spin waves with transverse and longitudinal polarization are different. Under application of a field a gap opens up at q=0 and the intensity decreases at small q as expected. The magnitude of the Zeeman gap and of the dipolar term agree with the bulk values. Therefore, the present experiments are in agreement with the existing spin-wave theories. We remark that it is possible to characterize the orientation of the domains in a field by determining the spin-wave dispersion in the dipolar regime and then extracting the term  $\langle \sin^2 \theta_q \rangle$ . Therefore **M** and *D* can be measured at the same time on the same sample and  $q_D$  can be determined.

On the basis of renormalization group theory<sup>22</sup> and previous experiments<sup>9</sup> it is known that the paramagnetic susceptibility is given by ( $\kappa$  is the inverse correlation length for  $T > T_c$ )

$$\chi_p^i(q) \propto \frac{1}{q^2 + \kappa^2 + \delta_{i,L} q_d^2} , \qquad (11)$$

i.e., that the longitudinal fluctuations (i = L) do not diverge. In contrast, the transverse fluctuations (i = T)diverge and are responsible for the phase transition at  $T_c$ . The present experiments in the ferromagnetic phase show now, that a similar behavior is observed in the ferromagnetic phase. The corresponding spin-wave susceptibility can be obtained by dividing Eqs. (7) and (8) by D, yielding

$$\chi_{\rm sw}^{i}(q) \propto \frac{1}{q^2 + q_H^2 + \delta_{i,L} q_D^2}$$
 (12)

Note that we designate the dipolar wave number above  $T_c$  with  $q_d$ , and below  $T_c$  with  $q_D$ . The magnetic wave vector is given by  $q_H = (E_H/D)^{1/2}$  and it is zero when no field is applied. Obviously, the q dependences of  $\chi_p^i$  and  $\chi_{sw}^i$  are equivalent. In the paramagnetic phase, the dipolar effects become weaker with increasing T, because the size of the correlated areas decreases. Hence the dipolar fields decrease too. Below  $T_c$ , however, there is long-range order and the dipolar effects are important at all temperatures.

In addition to the spin-wave scattering we have also investigated the parallel fluctuations  $\chi_z^T$ , i.e.,  $\delta S || \mathbf{M}$ , with transverse polarization  $\delta S \perp \mathbf{q}$ . The spectral shape of  $\chi_z^T(\mathbf{q},\omega)$  is quasielastic, similar as in Ni (Ref. 23) and some features have already been discussed elsewhere.<sup>24</sup> In analogy to Ni,  $\chi_z^T(\mathbf{q})$  is expected to diverge like  $1/(q^2 + \kappa_z^2)$  for  $q \gg \kappa_z$ . Such a behavior has also been observed in EuO by Als-Nielsen *et al.*<sup>18</sup> In the limit  $q \ll \kappa_z$  the susceptibility is expected to diverge like  $1/(q\kappa_z)$ .<sup>25</sup>

scattering and is the subject of further studies.

In Table II we collect the q dependences of different polarization modes of susceptibilities above and below  $T_c$ for an isotropic ferromagnet with dipolar interactions for  $q \gg \kappa_z$ . They have the effect that the longitudinal spin waves attain a mass, therefore reducing the number of Goldstone modes from 2 to 1 as predicted by Prokovsky.<sup>26</sup> In order to have a smooth transition of the susceptibilities at  $T_c$ , we speculate that the intensity of the longitudinal susceptibility  $\chi_z^L$  of the parallel fluctuations is also reduced with respect to the transverse fluctuations  $\chi_z^T$ . Our preliminary experiments for measuring  $\chi_z^L$  failed indeed, possibly because of the too low intensity.

The reduction of the number of Goldstone modes by 1 has very recently also been inferred from NMR and lowfrequency ac-susceptibility measurements in EuS by Kötzler et al.<sup>27</sup> In addition they verified the singularity  $\chi_z \propto H^{-1/2}$  that has been predicted theoretically many times. From the amplitude of the singularity they deduced  $q_D = 0.27$  Å<sup>-1</sup>. This value is clearly incompatible with our measurements that are well described by  $q_D \simeq 0.18$  Å<sup>-1</sup>, that is also smaller than the value reported in Ref. 10 for  $T > T_c$ . This discrepancies must be resolved in further theoretical work and with more accurate experiments. In passing, we note, that a mild T dependence of  $q_D$  is expected because the critical exponents for D and for M are not identical, i.e.,  $q_D^2 = g\mu_B\mu_0M/D \propto (T_c - T)/(T_c)^{\nu\eta} (\nu\eta = 0.024).^{28}$ 

The experimental situation for dipolar ferromagnets is now as follows. Above  $T_c$  the dynamics and statics of the magnetic fluctuations are rather well understood on the basis of mode-mode coupling theory.<sup>29</sup> However, below  $T_c$ , a good understanding is still missing. Whereas our experiments indicate that the essential features of the wavelength-dependent susceptibility are understood, the dynamical aspects are not clear, at least not close to  $T_c$ . Linear spin-wave theory predicts energies for the magnetic excitations that are independent of the polarization. However, close to  $T_c$  this can no longer be true, because, when approaching  $T_c$  from above, the energy scale (i.e., the linewidth) of the longitudinal fluctuations is larger than that of the transverse fluctuations. Something similar is expected to happen below  $T_c$  for the spin waves and for the fluctuations along the magnetization direction. Experiments to investigate these questions are being performed and we are awaiting further theoretical progress in this direction.

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