Critical two- and three-spin correlations in EuS: An investigation with polarized neutrons

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The critical magnetic scattering has been investigated in EuS by means of small-angle scattering with polarized neutrons using an inclined magnetic field geometry, allowing the determination of three-spin correlation functions. Two contributions to the critical magnetic scattering $I_\Sigma(q)=I^\uparrow(q)+I^\downarrow(q)$ and $\Delta I(q)=I^\uparrow(q)-I^\downarrow(q)$ were studied for temperatures near $T_C=16.52$ K. The $I^\uparrow(q)$ and $I^\downarrow(q)$ are the scattering intensities for the incident neutron beam polarized along (↑) and opposite (↓) to the magnetic field. The symmetric contribution, namely $I_\Sigma(q)$, comes from the pair-spin correlation function. The scattering intensity is well described by the Ornstein-Zernike expression $I_\Sigma(q)=A(q^2+\kappa^2)^{-1}$, where $\kappa=\xi^{-1}$ is the inverse correlation length of the critical fluctuations. The correlation length ξ obeys the scaling law $\xi=a_0\tau^{-\nu}$, where $\tau=(T-T_C)/T_C$ is the reduced temperature, $a_0=0.17$ nm, and $\nu=0.68\pm0.01$. The difference contribution $\Delta I(q)$ is caused by the three-spin chiral dynamical spin fluctuations that represent the asymmetric part of the polarization dependent scattering. The q dependence of $\Delta I(q)$ follows closely $1/q^2$. $\Delta I(q)$ depends on the temperature as $\tau^{-\nu}$ with $\nu=0.64\pm0.05$. The exponents ν as determined by means of the static measurements by ξ and the dynamic measurements (using the chirality) are in excellent agreement with each other, demonstrating the internal consistency of the theory and the experiment. Therefore, our results confirm the principle of the critical factorization, which is known as Polyakov-Kadanoff-Wilson operator algebra.

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I. INTRODUCTION

The static and dynamic properties of Heisenberg ferromagnets above the Curie temperature T_C have been investigated in detail both experimentally and theoretically and are considered to be well understood. The parameters defining the static properties of the ferromagnets in the critical range agree well with the theoretical critical exponents, for example, α for the specific heat capacity, β for the magnetization, γ for the suscuptibility, ν for the correlation length of the pair correlation function, etc. The dynamic properties for systems with an isotropic exchange interaction are well interpreted in terms of the dynamic scaling hypothesis.[1,2] In its simple form this hypothesis states that all physical variables are described by homogeneous scaling functions, which depend only on the single parameter $q\xi(T)$ with ξ as the correlation length of the fluctuating order parameter.

The complication arises when the relativistic interactions, such as dipolar forces or anisotropic exchange, are considered. As was shown in Ref. 3, despite their weakness, the relativistic interactions cannot be taken into account in the frame of the perturbation theory. It is also shown that they considerably change the critical dynamics. Thus, dipolar forces should gain inportance somewhere in a so-called dipole critical regime, where $\xi^{-1}=\kappa$ and q are small compared to the dipole number q_d. For each ferromagnet this quantity measures the strength of the dipolar interaction relative to the exchange interactions. The effects of the dipolar forces on the dynamics of the ferromagnet have been taken fully into account in Refs. 3–5. An excellent agreement of the theory[4,5] with the experiments was obtained for the critical slowing down of $\Gamma(q)$ observed for $q\to0$ and $T\to T_C$ on the transverse fluctuations in Fe (Ref. 6) and on the transverse and longitudinal fluctuations in EuS (Ref. 7). The most clear result of these studies is the crossover from $\Gamma(q>q_d)\sim q^{5/2}$ in the exchange-dominated regime to $\Gamma(q\ll q_d)\sim q^2$ in the dipolar regime.

It is also well known that the presence of the high-order correlations is generally strongly enhanced in the critical region near T_C. The even-order correlations, involving four spins etc., change the critical exponent of the pair correlation function from the mean-field value $\nu\approx1/2$ to that for critical scaling $\nu\approx2/3$. The odd-order correlation functions do not contribute to the static part of the magnetic susceptibility because this would contradict the energy conservation laws with respect to the time inversion. Nevertheless the odd correlation functions may contribute to the dynamic part of the susceptibility. In lowest order, triple-vertex and triple-spin fluctuations associated with it were introduced by Maleyev[8] and studied in detail by Lazuta.[9] As was shown in Ref. 8, and experimentally confirmed in Ref. 10, the triple vertex $([\mathbf{S}_x\times\mathbf{S}_y]\mathbf{S}_z)$ is a new entity lacking in the static theory of phase transitions. The triple dynamic spin fluctuations were detected for the first time by Okorokov *et al.*[10,11] in the scattering of polarized neutrons in iron. In the limit of small magnetic fields the theory predicts that the temperature dependence of the scattering cross section connected to the triple vertex is determined by a factor $(\kappa a)^{-1}\propto\tau^{-2/3}$. This theoretical prediction was confirmed by experiment, where the dependence of $\tau^{-0.67\pm0.07}$ was found. Thus, this investigation has allowed testing experimentally the confluence rules of correlations[1] that are equivalent to the Polyakov-Kadanov-Wilson (PKW) algebra.[12–14]

The triple-vertex and the three-spin correlation (TSC) function are objects showing chirality that can be directly studied by means of polarized neutrons. It was shown recently[15,16] that not only static but also dynamic chirality can be investigated by means of polarized neutron scattering using (i) a special inclined geometry of the applied magnetic field (**H** is inclined to the wave vector **q**) or (ii) directly by means of triple-axis techniques.[17] It was shown in[18–20] that in the case of ferromagnets (in the inclined geometry) neutron scattering from the three-spin fluctuations leads to the appearance of an asymmetric contribution to the polarization-dependent cross section. The symmetric part of the magnetic scattering is connected to the pair-correlation function. Thus, both correlation functions can be measured in the same experiment.

The present paper aims to study the triple vertex and the TSC function along with the pair-correlation function in the localized ferromagnet EuS in the critical region $T \geqslant T_C$. Both functions are measured using small-angle scattering of polarized neutrons in the inclined experimental geometry of the magnetic field. Both contributions are studied as a function of the temperature and the magnetic field. Firstly, we will present the critical scaling behavior of the pair-correlation function. We find that the temperature dependence of the correlation length in zero field obeys the scaling law $\xi \sim \tau^{-\nu}$, where $\nu = 0.68 \pm 0.02$, in agreement with previous work.[21] From experiments in a magnetic field at $q < q_d$ and T close to T_C we derive the dynamic index $z = 2.1 \pm 0.1$, which is close to the theoretically predicted value $z = 2$. The results of our study of the pair-correlation function are in good agreement with those obtained previously.[7,21] On this basis the scattering intensity attributed to the triple-correlation function is studied. It is shown that in the limit of the small fields the scattering increases with increasing field, showing the scaling temperature behavior proportional to $\tau^{-\nu}$ with $\nu = 0.64 \pm 0.04$. These results are close to those obtained in the itinerant ferromagnet Fe.[10]

The importance of studying the high-order correlation functions should be pointed out because in many cases the physical properties of a system of particles are defined by interaction of numerous neighboring particles. As a consequence, not only the pair-correlation functions but also the many-point (triplet, etc.) correlation functions play fundamental roles in descriptions of statistical properties of such systems and are important for condensed matter research.[22] Numerous studies on this subject involved different theoretical considerations and computer simulations.[23]

The paper is organized in the following way. Section II gives a theoretical description of the method (SAPNS in the inclined geometry) and the derivation of the correlation functions that can be measured with SAPNS in the critical region. Section III describes the sample and the experimental setup. The results from small-angle scattering of polarized neutrons (SAPNS) and their interpretation are given in Sec. IV. The results are summarized in Sec. V.

II. THEORETICAL BACKGROUND

It is well known that the neutron magnetic scattering is determined by two-spin correlation functions that are directly related to the imaginary part of the wavelength dependent susceptibility $\chi(\mathbf{q},\omega)$. In zero magnetic field χ is a symmetric second-rank tensor and the neutron scattering intensity does not depend on the neutron polarization. In a magnetic field **H**, the tensor attains antisymmetric parts. If **H** is along the z direction we have $\langle S_x S_y \rangle \neq \langle S_y S_x \rangle$ and the cross section depends on $\mathbf{P}_0$. In small **H**, the antisymmetric part is proportional to **H** and, as the Zeeman interaction is a product of **H** and the total spin $\Sigma \mathbf{S_R}$, the antisymmetric part of the scattering is determined by the three-spin correlation function $(\langle [\mathbf{S}_x \times \mathbf{S}_y] \mathbf{S}_z \rangle)$.[9,15,16] This chiral part of the neutron scattering is static for helimagnets and dynamic for ferromagnets. Indeed, the transverse components of the spin in ferromagnets (S_x and S_y), saturated in the z direction, are always related to the excitations.

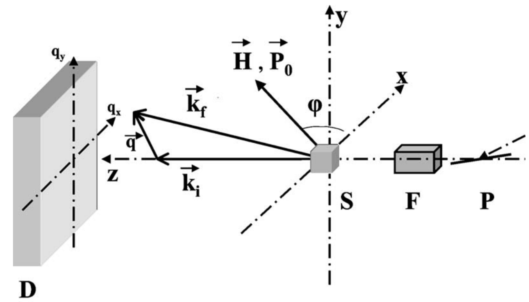

FIG. 1. Schematic outline of the SAPNS experiment in the inclined geometry: P denotes the polarizer, F the spin flipper, S the sample, D the two-dimensional position-sensitive detector. The magnetic field **H** is inclined at an angle $\varphi = 45°$ with respect to the incident beam $\vec{k}_i$ in the (xz) plane.

For small-angle scattering the chiral cross section has the following form:[15,16]

$$\sigma_{ch}(\mathbf{q},\omega) = (2r^2 P_0 T/\pi\omega)(\hat{q}\cdot\hat{h})^2 \, \mathrm{Im}\, C(\mathbf{q},\omega), \qquad (1)$$

where $\hat{h}$ and $\hat{q}$ are unit vectors along the magnetic field **H** and momentum transfer **q**, respectively, and ω is the energy transfer. In this expression we have taken into account that both the dynamical chirality $\mathbf{C}(\mathbf{q},\omega)$ and the neutron polarization $\mathbf{P}_0$ are directed along **H**, and $\mathbf{C} = \hat{h}\mathrm{C}$.

In a small-angle neutron scattering (SANS) experiment, only elastic components of momentum transfer **q**, i.e., perpendicular to the incident wave vector $\mathbf{k}_i$, are visible in the detector: $q_\perp = k_i(\theta_x + \theta_y)$. An inelastic, parallel to $\mathbf{k}_i$, part of the momentum transfer $q_\parallel = q_z = k_i\omega/(2E)$ cannot be detected. Thus, no energy analysis is performed in a conventional SANS experiment but integration over the energy is automatically performed. Therefore, in theory, Eq. (1) has to be integrated over all energies of the scattered neutrons. Recalling that $\mathrm{Im}\, C(\mathbf{q},\omega)$ is an even function of ω,[9] the integrated chiral cross section becomes zero if there is no ω-odd term in the factor $(\hat{h}\cdot\hat{q})^2$ of Eq. (1). Such an ω-odd term appears if **H** is inclined with respect to the incident beam at an angle φ (Fig. 1). Then, for the inclined field, we have

$$(\hat{q}\cdot\hat{h})^2 = \frac{(2E\theta)^2\cos^2\varphi + \omega^2\sin^2\varphi + 2E\theta\omega\sin 2\varphi}{(2E\theta)^2+\omega^2}. \quad (2)$$

Obviously, only the third term is ω odd. Hence, the ω-integrated chiral cross section is given by

$$\sigma_{ch}(\theta) = \frac{2}{\pi}r^2P_0T\sin 2\varphi\int_{-\infty}^{\infty}\frac{d\omega 2E\theta\,\mathrm{Im}\,C(\mathbf{q},\omega)}{(2E\theta)^2+\omega^2}. \quad (3)$$

This integral can be evaluated in the critical paramagnetic region $T>T_C$. According to scaling theory all physical parameters have a scaling dimensionality determining the general form of the corresponding correlation function. For example, the two-spin correlation function has the form

$$G(q) = \frac{1}{(\kappa a)^{2-\eta}}F\left(\frac{q}{\kappa}\right) \simeq \frac{1}{a^2(q^2+\kappa^2)}, \quad (4)$$

where κ is the inverse correlation length of the critical fluctuations defined as $\kappa=\tau^{\nu}/a$. Here, $\tau=|T-T_C|/T_C$ is a reduced temperature, $\nu\approx 2/3$ is the critical exponent of the correlation length and a is a length scale of order of the lattice spacing. The Ornstein-Zernicke expression on the right-hand side of Eq. (4) is valid for $\eta \ll 1$. This is the case for three-dimensional (3D) spin systems. Therefore, we will neglect η below.

In a magnetic field, we have to compare the energy of the magnetic field $g\mu_BH$ with the energy of the critical fluctuations $k_BT_C(\kappa a)^{5/2}$ in order to determine the condition for the weak-field $f\ll 1$ and strong-field regime $f\gg 1$, respectively, where $f=g\mu_BH/k_BT_C(\kappa a)^{5/2}$ is a dimensionless number.

In a weak field the chiral scattering is proportional to **H**. Therefore, its scaling dimensionality is determined by the product $fG(q)$ and we obtain

$$\mathrm{Im}\,C(q,\omega) = \frac{g\mu_B\mathbf{H}}{k_BT_c(\kappa a)^{9/2}}F\left[\frac{q}{\kappa},\frac{\omega}{\Omega(q)}\right], \quad (5)$$

where $\Omega(q)=k_BT_C(qa)^{5/2}$ is the characteristic energy of the critical fluctuations with momentum $\mathbf{q}$. Equation (5) is valid for ferromagnets in the exchange approximation.[3]

The dynamical chirality is a three-spin correlation function and it may be considered as a result of the scattering of critical fluctuations on the uniform magnetic field.[16] From this point of view it is clear that $C(\mathbf{q})$ is a function of two momenta, namely the momentum of the fluctuation, $\mathbf{q}$, and the momentum of the field, $\mathbf{q_H}=0$. The principle of critical factorization was formulated by Polyakov[12–14] and is known as Polyakov-Kadanoff-Wilson operator algebra. It states that in any multispin correlation function, the dependence on the largest momentum $\mathbf{q}(q\gg\kappa)$ appears as a factor $(q/\kappa)^{-5+1/\nu}\Phi[\omega/\Omega(q)]$. In our case, putting $\nu\approx 2/3$ we obtain

$$\mathrm{Im}\,C(q,\omega) = \frac{g\mu_BH}{T_c(qa)^{7/2}(\kappa a)}\Phi\left[\frac{\omega}{\Omega(q)}\right]. \quad (6)$$

In this expression we have $q=k_i[\vartheta^2+(\omega/2E)^2]^{1/2}$. The dependence of q on ω may be neglected in the quasielastic approximation, if the residence time of the neutron in a region of the size of the order of $1/q$ is much smaller than the characteristic lifetime of the fluctuation of the same size $\hbar/\Omega(q)$. The corresponding condition can be expressed as

$$q \ll q_{in} = a^{-1}(2E/k_BT_cka)^{2/3}. \quad (7)$$

Hence we can replace q by $k_i\vartheta$ and neglect ω in the denominator of Eq. (3). Therefore, the chiral cross section becomes

$$\sigma_{ch}(\theta) = \frac{2}{\pi}r^2P_0\sin 2\varphi\frac{g\mu_BH}{2E(ka)\theta^2}\frac{1}{\kappa a}\,\mathrm{sgn}(\theta). \quad (8)$$

Often, it is convenient to normalize Eq. (8) by the symmetric cross section $\sigma(\theta)=(2/3)r^2G(q)$ and one obtains

$$\hat{\sigma} = \frac{\sigma_{ch}(\theta)}{\sigma(\theta)} = AP_0\sin 2\varphi\frac{g\mu_BH}{E}\frac{k}{\kappa}\,\mathrm{sgn}(\theta). \quad (9)$$

Here, A is a constant of the order unity. We will use Eq. (9) for analyzing our experimental results in the next paragraph.

III. EXPERIMENTAL

The isotopically enriched sample ^{153}EuS was assembled from approximately 100 small EuS crystals on a machined aluminum substrate. The anisotropy axes of all crystals were aligned in one direction to within a precision of 0.75°.[24] The average size of a single crystal is 1–2 mm^2. The SAPNS experiment was performed at the SANS-2 scattering facility of the FRG-1 research reactor in Geesthacht (Germany). The schematic outline of the experiment is given in Fig. 1. A polarized beam of neutrons with an initial polarization $P_0=0.95$, a wavelength $\lambda=0.58$ nm ($\Delta\lambda/\lambda=0.1$), and a divergence 10 mrad were used. The scattered neutrons were detected in a q range 0.30 to 2.5 nm^{-1} with a position sensitive detector composed of 256×256 pixels. The scattering was measured in a temperature range 14 K$<T<$50K, i.e., from below to far above $T_C=16.55$ K. The external magnetic field 1 mT$<\mathbf{H}<$200 mT was applied at an angle of $\phi=45°$ with respect to the incident beam $\mathbf{k_i}$ (the inclined geometry). The adiabatic condition for the transmission of polarized neutrons was sufficiently satisfied to obey the relation $\mathbf{P}_0\|\mathbf{H}$. Two scattering intensities $I^{\uparrow}(q)$ and $I^{\downarrow}(q)$ were measured for the incident neutron beam polarized along (↑) and opposite (↓) to the magnetic field.

The method of the inclined geometry allows distinguishing the two contributions to the magnetic scattering: the symmetric polarization-independent (SPI) and asymmetric polarization-dependent (APD) scattering. First, we separated the magnetic critical scattering from the T-independent, nonmagnetic contribution. Following the standard procedure we determined the pure magnetic scattering by subtracting from the measured intensity the nonmagnetic background as measured at $T\gg T_C$, i.e.,

$$I_m(\mathbf{q}_\perp,T) = I(\mathbf{q}_\perp,T) - I(\mathbf{q}_\perp,50\text{ K}). \quad (10)$$

As noted above, $\mathbf{q}_\perp$ is a projection of $\mathbf{q}$ onto the detector plane. In the folowing text we will omit the subscript "⊥." To separate the SPI term from the APD one, we take the sum of the measured intensities $I_\Sigma(q)=I_m(P_0,q)+I_m(-P_0,q)$ and average it over 2π for each $|q|=\sqrt{q_x^2+q_y^2}$. As a consequence,

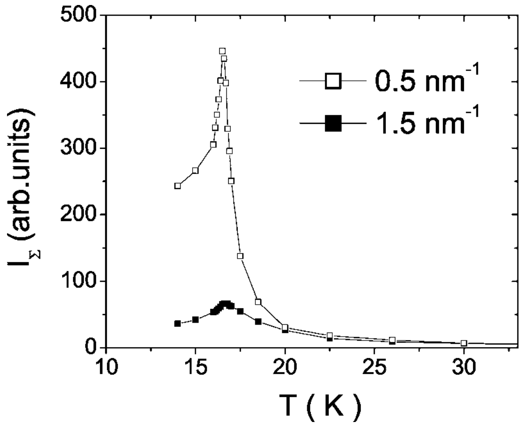

FIG. 2. Temperature dependence of magnetic scattering cross section I_m at a residual magnetic field $\mathbf{H}=1.4$ mT at $q=0.5$ and $1.5\ \mathrm{nm}^{-1}$.

the asymmetric part is averaged out and only the SPI part survives. It is related to the pair-correlation function. The polarization dependent part of the scattering $\Delta I(q)=I(P_0,q)-I(-P_0,q)$ is asymmetric. The asymmetry is directly related to the direction of $\mathbf{H}$. In the particular case of $\mathbf{H}$ being in the (xz) plane (Fig. 1), the asymmetry is most pronounced along the x component of the momentum transfer q_x due to the selection rules. Thus the asymmetric contribution was extracted by taking the difference of the measured intensities $I(\pm P_0,\pm q_x)$,

$$\Delta I(q)=\frac{1}{4}[I(P_0,q_x)-I(-P_0,q_x)] + \frac{1}{4}[I(-P_0,-q_x)-I(P_0,-q_x)]. \tag{11}$$

For the analysis of the data, the SPI and the APD contributions were attributed to the pair-correlation function and the triple-correlation function, respectively.

IV. RESULTS

A. Pair-correlation function

Figure 2 shows the temperature dependence of the magnetic intensity I_m for two different momentum transfers. It is clearly seen that the intensity has a pronounced maximum at $T\approx T_C$. It is more pronounced for small q than for large q. This behavior demonstrates the appearance of critical fluctuations and an increase of the correlation length as T approaches T_C from low and high temperature. The q dependence of the intensity I_m is treated in a standard way using the Ornstein-Zernike expression

$$I_m(q)=\frac{Z_m}{q^2+\kappa^2}, \tag{12}$$

where $\kappa=\xi^{-1}$ is the inverse correlation length. The parameters Z_m and κ have been obtained from a least-squares fit to the data using Eq. (12). The temperature dependence of Z_m and κ are shown in Figs. 3(a) and 3(b) for small

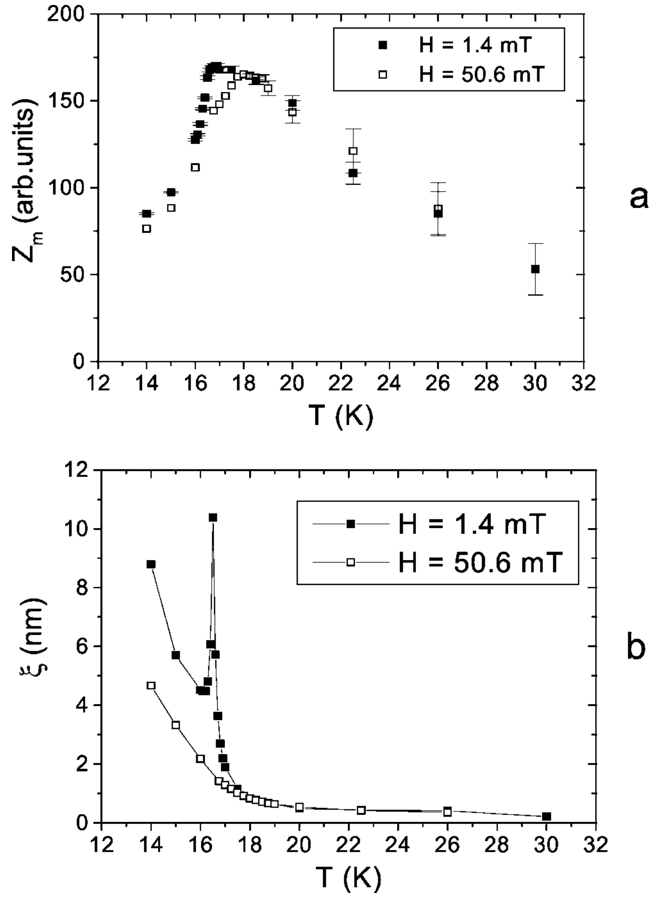

FIG. 3. Temperature dependence of the parameter Z_m (a) and the correlation length ξ (b) for two values of the magnetic field: $\mathbf{H}=1.4$ and 50.6 mT.

($H=1.4$ mT) and large ($H=50$ mT) fields, respectively. The parameter Z_m exhibits a smooth growth when T approaches T_C from the high-temperature side. In principle, $Z_m=\chi(0)\kappa^2$ should increase like $(T_C/T)^{4/3}$ (see, for example, Ref. 25). Then Z_m becomes almost constant in the critical regime at $T_C<T<T_C(1+\tau)$ with $\tau<0.1$. Z_m decreases sharply as soon as the temperature crosses the critical temperature $T_C=16.52$ K, which implies enhancement of the ferromagnetic domain structure. Z_m does not depend on the magnetic field in the whole temperature range except close to T_C. The correlation length ξ shows a sharp maximum as the temperature approaches T_C [Fig. 3(b)]. Below T_C, ξ decreases first as the contribution of the longitudinal fluctuations decreases[24] with decreasing T and increases again due to domain formation. When the magnetic field is applied, the maximum at T_C vanishes and the transition is smeared out. The τ dependence of the inverse correlation length κ is shown in Fig. 4 on a log-log scale. The parameter κ obeys the scaling law $\kappa=(a)^{-1}(\tau)^{\nu}$ in a range of $0.005<\tau<0.2$, with $a=0.17\pm0.01$ nm and $\nu=0.67\pm0.02$. The obtained parameters are close to those obtained in Ref. 21, where they were found to be $a=0.19$ nm and $\nu=0.70\pm0.02$. There is a clear crossover at $\tau=0.2(T\simeq 20\ \mathrm{K})$ to the noncritical regime.

As seen in Fig. 3, the magnetic field affects the correlation length ξ in the close vicinity of T_C. Figure 5 shows the mag-

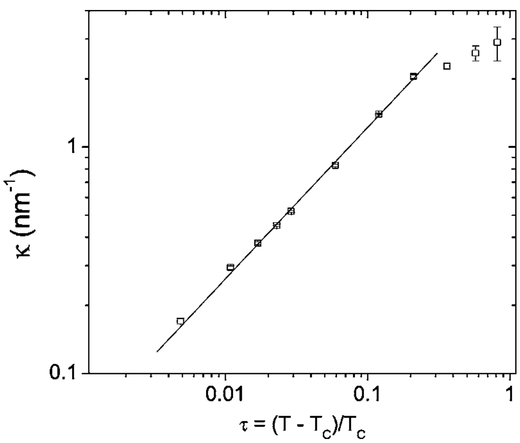

FIG. 4. Inverse correlation length κ as a function of the reduced temperature $\tau=(T-T_C)/T_C$ on a log-log scale at $\mathbf{H}=1.4$ mT.

netic field dependence of the correlation length ξ at $T=16.55$ $(\tau\rightarrow 0)$, 17 $(\tau\sim 0.1)$, 18 $(\tau>0.1)$, and 19 K. The value of ξ depends strongly on the field for $\tau\rightarrow 0$, while it has a weak dependence at $\tau\sim 0.1$, and almost no dependence at $\tau>0.1$. As was noted above, the parameter Z_m has little or no change with H. The effect of the magnetic field on the correlation length of the critical fluctuations can be understood in terms of the balance between the energy of the magnetic field $g\mu H$ and that of the critical fluctuations $k_BT_C(\kappa a_0)^z$ with $\kappa=\xi^{-1}$. Here the dynamic index is $z=2$ in the dipolar dominated regime for $q<q_d$ and $z=5/2$ in the exchange-dominated regime for $q>q_d$.[4,5] The dipolar wave number for EuS was found $q_d=2.2(5)$ nm^{-1} in Ref. 26. For $g\mu H\gg k_BT_C(\kappa_{(H=0)}a_0)^z$, the correlation length is renormalized as a function of the magnetic field: $\kappa(H)a_0=(g\mu H/T_C)^{1/z}$. The observed behavior of κ at $T\approx T_C$ is a result of the crossover to the strong field regime. The fit gives a value for the parameter $1/z=0.48\pm 0.02$, which is close to the theoretical value in the dipolar regime where $1/z=0.5$.

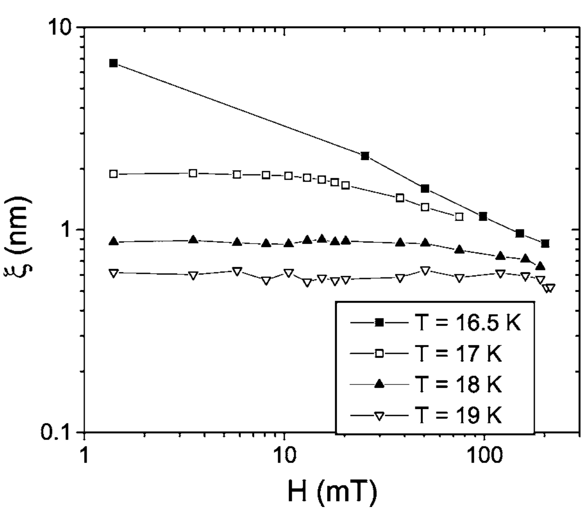

FIG. 5. Inverse correlation length $\kappa=\xi^{-1}$ as a function of the magnetic field as measured at temperatures $T=16.55$, 17, 18, and 19 K.

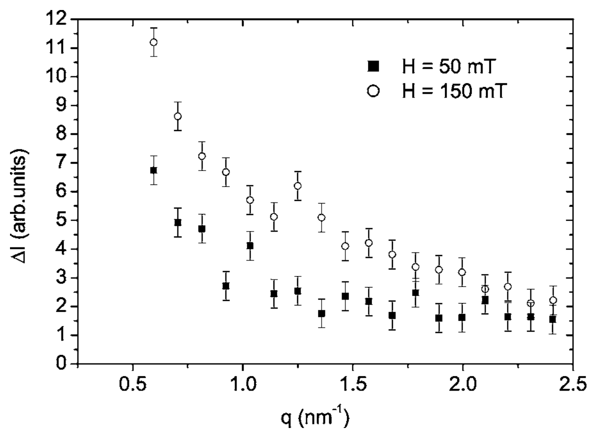

FIG. 6. q dependence of the asymmetric part of the SAPNS difference intensity ΔI for magnetic fields $\mathbf{H}=50$ and 150 mT at $T=16.55$ K.

The observed renormalization of $\kappa_c(H)$ indicates that for $q\leqslant\kappa_c(H)$ the energy of the critical fluctuations $\Omega=k_BT_C(\kappa_H a_0)^z$ is determined by the field $\mathbf{H}$. For $q\geqslant\kappa(H)$ the energy is equal to $\Omega_c=k_BT_C(qa_0)^z$ and the magnetic field can be considered to be a weak perturbation.

B. Three-spin correlation function

Figure 6 shows typical examples of the asymmetric scattering $\Delta I(q)$ at the magnetic field $\mathbf{H}=50$ and 150 mT and $T=16.55$ K. The analytical expression for $\Delta I(q)$ given by Eq. (8) is known in a rough approximation and is valid within the range $\kappa<k_i\theta<q_{in}$, where the inelastic characteristic momentum q_{in} [Eq. (7)] is of the order of 10 nm^{-1} for EuS. The remarkable features of this APD scattering contribution are the following: (1) It appears only when the inclination angle φ between the magnetic field $\mathbf{H}$ and the incident beam direction $\mathbf{k_i}$ is not equal to 0 or $\pi/2$. Its appearance in this inclined geometry implies the dynamical nature of the scattering. (2) $\Delta I(q)$ changes sign when the scattering angle θ changes sign. (3) The scattering depends on the scattering angle as θ^{-n}. (4) It increases with the applied magnetic field $\mathbf{H}$. (5) It vanishes for non-polarized neutrons $(P_0=0)$. All these features clearly identify this scattering to arise from the triple vertex correlations.

It is sometimes convenient to normalize the asymmetric scattering by the magnetic scattering $\hat{\sigma}=\Delta I(q)/I_\Sigma(q)$ [Eq. (9)]. $\hat{\sigma}$ is presented in Fig. 7 as a function of q. The figure demonstrates that it is constant at small q and in low fields and has a tendency to increase at large q. Therefore, the q dependences of $\Delta I(q)$ and $I_\Sigma(q)$ are equivalent at small q, while they are different at large q. The last feature may be connected with the dipolar interactions of the spin system at $q=q_d\approx 2.2$ nm^{-1}. The restricted q range of measurements does not allow us, however, to make more definite conclusions on the q dependence of the asymmetric scattering. Still, in a small q range, $\Delta I(q)$ is proportional to q^{-2} as predicted by the theory in Eq. (8).

In order to quantify the temperature and magnetic field dependence of ΔI we have averaged $\Delta I(q)$ over q ranges

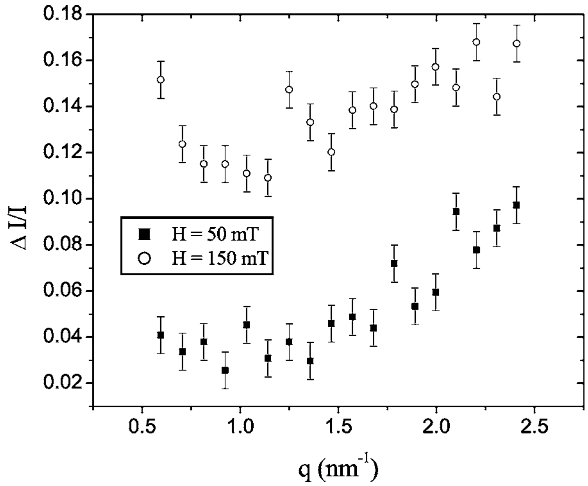

FIG. 7. Normalized value of $\Delta I/I_\Sigma$ in magnetic fields $\mathbf{H}$=50 and 150 mT at T=16.55 K.

within the limits (i) $0.50<q<1$ nm^{-1}; (ii) $1<q<1.5$ nm^{-1}; and (iii) $1.5<q<2.3$ nm^{-1}. Figure 8 shows the averaged values versus H for T=16.55 K. For all q values, $\langle\Delta I\rangle$ increases with increasing field and saturates above $H\sim 150$ mT. The increase of $\langle\Delta I\rangle$ is clearly related to the range of H where $q>\kappa_H$ (i.e., to the weak field regime) and it saturates as soon as $\kappa_H\sim q$. This observation demonstrates the validity of the weak field approximation for the concept described above.

The values $\langle\Delta I\rangle$ are shown in Fig. 9 versus T for H=50 mT. The theory predicts that the value $\langle\Delta I\rangle$ depends on T as $\tau^{-\nu}$. The parameters obtained from the fit of the experimental data are: ν=0.62(0.05) for q=1.85 nm^{-1}; ν=0.65(0.03) for q=1.25 nm^{-1}, and ν=0.64(0.04) for q=0.75 nm^{-1}. The points at $T=T_C$ do not follow the scaling law $\tau^{-\nu}$. This may be related to the crossover to the strong field regime and/or to demagnetization effects. According to

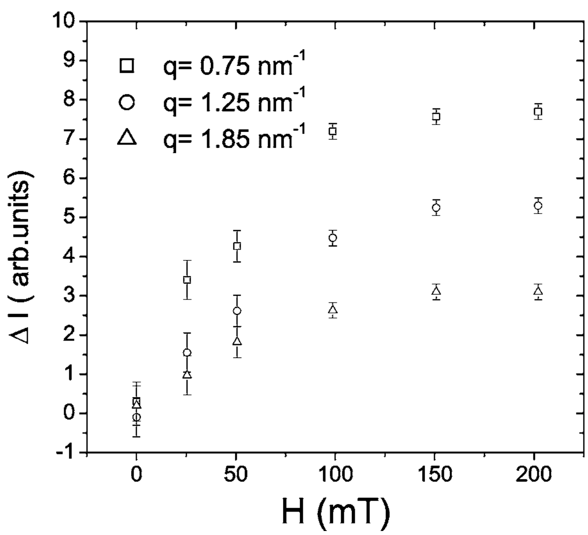

FIG. 8. Magnetic field dependence of $\langle\Delta I\rangle$ for three different q values q=0.75 nm^{-1}, q=1.25 nm^{-1}, and q=1.85 nm^{-1} at T=16.55 K.

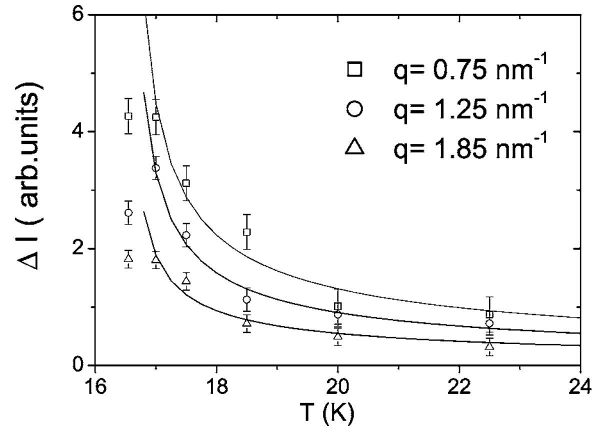

FIG. 9. Temperature dependence of $\langle\Delta I\rangle$ for three different q values q=0.75 nm^{-1}, q=1.25 nm^{-1}, and q=1.85 nm^{-1} in a field $\mathbf{H}$=50 mT.

Eq. (8), the observed increase of the intensity ΔI is proportional to $\tau^{-\nu}=\xi$ at $q>\kappa$. It is interesting to note that the intensity attributed to the pair-correlation function $I_\Sigma\sim 1/(a\kappa)^2=\tau^{-2\nu}=\xi^2$ for $q\ll\kappa$. The factor of 2 difference between the exponents for the symmetric and antisymmetric scattering clearly proves the completely different origin of the observed scattering contributions. On the other hand, the exponents ν as determined by means of the static measurements by ξ and the dynamic measurements (using the chirality) are in excellent agreement with each other, demonstrating the internal consistency of the theory and the experiment.

V. CONCLUSION

The ferromagnetic to paramagnetic phase transition in EuS has been investigated by means of small-angle scattering of polarized neutrons using the inclined geometry of the applied field $\mathbf{H}$ with respect to the neutron wave vector $\mathbf{k_i}$. This geometry of the experiment allows separating two contributions to the critical scattering: (i) the symmetric polarization-independent part attributed to the pair-correlation function and (ii) the asymmetric polarization-dependent part attributed to the triple vertex $[\mathbf{S}_x\times\mathbf{S}_y]\mathbf{S}_z$ and its related triple-correlation function.

The behavior of both parts of the scattering has been studied as a function of temperature and magnetic field: (1) The pair-spin correlation function was deduced with its amplitude and the correlation length $\xi(T,H)$. At zero field the correlation length obeys the scaling law $\xi=a_0\tau^{-\nu}$ with $a_0=0.17\pm0.01$ and $\nu=0.68\pm0.02$. The magnetic field strongly influences the critical fluctuations near T_C, so that the correlation length is suppressed by the field as $\xi(H)=a_0(g\mu_B H/T_C)^{1/z}$ with the dynamic index $z=2.1\pm0.1$, which is close to the theoretically predicted value $z=2$ for the dipolar regime $\kappa<q_d$. (2) The asymmetric polarization-dependent part of the scattering was unambiguously identified as arising from the three-spin correlation function. The specific features of the scattering may be summarized as follows: (i) it is θ antisymmetric and P_0 dependent; (ii) it appears only in the inclined geometry and implies the dynami-

cal and chiral nature of the fluctuations. The analytical expression for this scattering is known within the weak-field approximation and for the limited q range $\kappa < q < q_{in}$. The theory predicts that the scaling behavior of the scattering cross section is proportional to $\tau^{-\nu}$ with $\nu \simeq 2/3$. The experiment confirms the theoretical predictions, yielding values $\nu = 0.62 \pm 0.05$, $\nu = 0.65 \pm 0.03$, and $\nu = 0.64(\pm 0.04)$ for different q values within the above-mentioned range. A similar experiment has been performed on pure iron.[10] Our data and their analysis support the conclusions of the work.[10] (3) The obtained results together with those reported in Ref. 10 provide the experimental proof for the confluence rules governing the correlations,[12] which is equivalent to the Polyakov-Kadanov-Wilson operator algebra. Moreover, to the best of our knowledge, there is no other way to test this theory except to study the dynamical chirality of spin systems. In all other cases only the pair-correlation functions are studied.

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