

Investigations of the sublattice magnetizations $M_{\text{sub}}(T)$ in antiferromagnets with fourth-order exchange interactions: $\text{Eu}_x\text{Sr}_{1-x}\text{Te}$

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Abstract. We present a neutron scattering study of the temperature and composition dependence of the MnO-type superstructure reflection intensities in the diamagnetically diluted antiferromagnetic compounds $\text{Eu}_x\text{Sr}_{1-x}\text{Te}$. In these materials antiferromagnetic biquadratic and ferromagnetic three-spin interactions have been identified recently. These fourth-order non-Heisenberg interactions are able to create their own order parameter which is believed to govern the order of the transverse moment components and which, hence, is directed perpendicular to the common Heisenberg order parameter. The observed MnO-type diffraction intensities originate in the sublattice magnetizations, $M_{\text{sub}}(T)$, of both order parameters. Due to the different composition dependencies for biquadratic interaction processes ($\sim x$) and three-spin interaction processes ($\sim x^2$), the ferromagnetic three-spin interactions dominate for $x > x_c = 0.85$, while for $x < 0.85$ the antiferromagnetic biquadratic interactions dominate. Associated with this sign change in the fourth-order interaction sum the transverse order parameter changes from the antiferromagnetic MnO type for $x < 0.85$ to ferromagnetic for $x > 0.85$. This is noticed as a sudden decrease of the low-temperature MnO scattering intensities at $x_c = 0.85$. Although susceptibility measurements reveal clearly a ferromagnetic component for $x > 0.85$ no ferromagnetic Bragg intensities were observed in standard neutron scattering spectra using EuTe powder samples. We explain this by the competition of antiferromagnetic biquadratic and ferromagnetic three-spin interactions whereby a disturbed ferromagnetic superstructure may be generated which gives rise also to weak MnO-type diffraction lines. It is found that the resulting $M_{\text{sub}}(T)$ obeys a T^2 law until a temperature as large as $0.75T_N$ irrespective of the nature of the transverse order parameter. The T^2 law must, hence, be common to both types of order parameter showing that the fourth-order interactions re-define the spin dynamics of both completely. From the linear composition dependence of the normalized T^2 coefficient the existence of three-spin interactions is again confirmed.

PACS. 75.30.Et Exchange and superexchange interactions – 75.30.Kz Magnetic phase boundaries (including magnetic transitions, metamagnetism, etc.) – 75.40.Cx Static properties (order parameter, static susceptibility, heat capacities, critical exponents, etc.)

1 Introduction

In two recent publications [1,2] it was shown how second-order (bilinear) and fourth-order interactions can be distinguished with measurements of the linear susceptibility χ_1 and the cubic susceptibility χ_3 . Both susceptibilities are defined by

$$B_i = \frac{1}{\chi_1} \cdot m + \frac{1}{\chi_3} \cdot m^3 + \dots \quad (1)$$

Here B_i is the external magnetic field converted to its value inside the sample and m is the reduced magnetization. It is easy to show that in the high temperature limit fourth-order exchange interactions (which comprise a

product of four-spin operators such as biquadratic, three-spin and four-spin interactions) give rise to a Curie-Weiss law for χ_3 . According to the nomenclature used in equation (1) the Curie-Weiss laws of the linear susceptibility χ_1 and the cubic susceptibility χ_3 read

$$\chi_1 = \frac{C_1}{T - \Theta_1} \quad \text{with} \quad C_1 = \frac{g(S+1)\mu_B}{3k_B} \quad (2)$$

$$\chi_3 = \frac{C_3}{T - \Theta_3} \quad \text{with} \quad C_3 = \frac{10g(S+1)^3\mu_B}{9[(S+1)^2 + S^2]k_B}. \quad (3)$$

It was furthermore shown in reference [2] that Θ_1 is given by a weighted sum of second-order (Heisenberg) exchange interactions and fourth-order exchange interactions while

Θ_3 is given exclusively by fourth-order exchange interactions. This opens a way to evaluate a quantitative measure for those interactions by an analysis of the curvature of the paramagnetic isotherms according to equation (1) and looking for the Curie-Weiss law of χ_3 . Note that in the absence of fourth-order interactions a Curie law is observed for χ_3 with $\Theta_3 \equiv 0$.

The two Curie-Weiss laws of equations (2, 3) mean that molecular field approximation predicts two order-disorder transitions associated either with a divergence of the linear susceptibility χ_1 at $T = \Theta_1$ or with a divergence of the cubic susceptibility χ_3 at $T = \Theta_3$. Moreover, a divergence of χ_1 at a temperature different from the divergence of χ_3 implies molecular field critical exponents which are, in fact, observed in GdMg, a material with sizable fourth-order interactions [3]. It should be remarked that in the Heisenberg model both susceptibilities diverge at one sole critical temperature.

Since a collinear spin system can order only once it has been argued in reference [3] that the second ordering transition driven by the fourth-order interactions affects the transverse moment components. In fact, these moment components are the only degrees of freedom left for an additional ordering process if the longitudinal moment components are already ordered. Though the transverse spin components perform highly correlated motions there is no finite expectation value or spontaneous magnetic order of the transverse magnetization in the absence of fourth-order exchange interactions *i.e.* in the Heisenberg model. Fourth-order interactions are therefore assumed to break the rotational symmetry of the uniaxially ordered Heisenberg spin system.

On the other hand, the molecular field result given by equation (3) is too simple to provide a clear idea on the nature of the second ordering transition at $T = \Theta_3$. We are therefore free to interpret this molecular field hint and have to test the resulting implications experimentally. Evidently also equation (1) gives no explicit indication for a perpendicular arrangement of the two order parameters. This is not to be expected since equation (1) is a scalar relation referring to the high-temperature limit where B_i and m are parallel.

The aim of this communication is to show that the neutron scattering data described here are consistent with the earlier propounded hypothesis [3] of two order parameters generated by second-order and fourth-order exchange interactions, respectively. In particular there are two conclusions to be drawn from our proposed interpretation of the Curie-Weiss law of χ_3 : first, any marginal value of the fourth-order interactions *i.e.* $|\Theta_3| > 0$ should be able to generate a novel ordered phase to be attributed to the transverse moment components. In general, this phase should have a finite transition temperature different from the conventional (Heisenberg) ordering temperature at $T = \Theta_1$. Second, the ordering type of the transverse moment components should be ferromagnetic for $\Theta_3 > 0$ and antiferromagnetic for $\Theta_3 < 0$. Both features have been observed in the diamagnetically diluted antiferromagnetic

compounds $\text{Eu}_x\text{Sr}_{1-x}\text{Te}$ [4, 5] and will be substantiated here with the microscopic method of neutron scattering.

It should be kept in mind that the class of fourth-order interactions comprises a number of different and competing individual interaction processes. The ordering structures generated by these interactions will therefore not be of the ideal collinear type. If we call an ordered state ferromagnetic because of $\Theta_3 > 0$ or antiferromagnetic if $\Theta_3 < 0$ this does not mean that simple collinear structures are given. Disturbed spin structures apply in particular for the conventional order parameter which is the result of all (conflicting) second-order and fourth-order interactions. Molecular field calculations have provided some criteria for the rise of non-collinear ordering structures [6–9]. The here evaluated ordered moment per Eu^{2+} of much less than $7\mu_B$ shows, in fact, that the MnO-type order is strongly perturbed. An analysis using idealized structure models is therefore inadequate. A very dramatic observation in this respect was made in $\text{GdAg}_{1-x}\text{Zn}_x$ [10]. In this ternary alloy system ferrimagnetically ordered phases owing to fourth-order interactions were identified with magnetization, calorimetric and dilatometric measurements without the observation of corresponding neutron scattering intensities.

A discrimination between biquadratic, three-spin and four-spin interactions can be achieved by a polynomial decomposition of the experimental $\Theta_3(x)$ function in mixed crystals of the magnetic material of interest and an isomorphic diamagnetic material such as $\text{Eu}_x\text{Sr}_{1-x}\text{Te}$ or $\text{Eu}_x\text{Sr}_{1-x}\text{S}$ [2]. Provided a perfect statistical distribution of magnetic and diamagnetic ions [11] and constant interaction parameters the coefficients of the linear, quadratic and cubic term in $\Theta_3(x)$ are proportional to the biquadratic, three-spin and four-spin interactions (the validity limits of this power series expansion for smaller x values will be outlined in the Appendix). It turned out, that in $\text{Eu}_x\text{Sr}_{1-x}\text{Te}$ biquadratic interactions are antiferromagnetic and three-spin interactions are ferromagnetic [1, 2]. Four-spin interactions seem to be of minor importance and could be neglected in the data analysis. Since the probability for biquadratic interactions is proportional to x while the probability for three-spin interactions is proportional to x^2 (see Appendix), the relative weight of both interactions changes with composition in $\text{Eu}_x\text{Sr}_{1-x}\text{Te}$. This gives rise to a particular composition of $x_c = 0.85$ at which both interaction types compensate in the high-temperature limit. Although a material with $\Theta_3 = 0$ would meet the requirements of the Heisenberg model, the individual microscopic interaction processes of the biquadratic and three-spin type will still have a finite strength even at $x_c = 0.85$. The order parameters, $M_{\text{sub}}(T)$, sense these interactions and therefore they will show no ideal Heisenberg properties even at $x_c = 0.85$. Only a quantity which is defined by the high temperature average of all fourth-order interaction processes can be expected to conform to the Heisenberg model predictions at x_c . This is the case for the critical field $B_c^{\parallel}(T \rightarrow 0)$ which gives the phase boundary to the paramagnetic phase. In the limit $T \rightarrow 0$ the moments are nearly ferromagnetically aligned along

$B_c^{\parallel}(T)$ and sample the high-temperature average value of the fourth-order interactions. In fact, a crossover from the predominant T^2 law to Bloch's $T^{3/2}$ law could be observed for $B_c^{\parallel}(T \rightarrow 0)$ on approaching x_c [5].

The order parameters react in a quite different way on the sign change of the fourth-order interaction sum at x_c *i.e.* the sign change of $\Theta_3(x)$ at x_c . The longitudinal (Heisenberg) order parameter, O_2 , being defined mainly by second-order interactions but to a smaller extent also by fourth-order interactions, remains antiferromagnetic for all compositions since $\Theta_1(x)$ is always negative. The Néel temperature of this order parameter, $T_N^{\parallel}(x)$, as well as the critical field value $B_c^{\parallel}(T = 0)$ exhibit only a very faint change in slope at x_c [5]. This shows that longitudinal and transverse ordering phenomena do virtually not interfere.

The transverse order parameter, O_4 , on the other hand, is defined exclusively by $\Theta_3(x)$ and changes from antiferromagnetic for $x < 0.85$ to ferromagnetic for $x > 0.85$. For $\Theta_3 < 0$ *i.e.* $x < 0.85$ a second critical field curve, $B_c^{\perp}(T)$, and a second Néel line, $T_N^{\perp}(x)$, are clearly observed in magnetization measurements [4,5]. As we will show here a discontinuous decrease of the low-temperature MnO-type scattering intensities is observed at x_c due to a changing order of the transverse spin components from antiferromagnetic for $x < x_c$ to ferromagnetic for $x > x_c$. This shows that the observed MnO-type scattering intensities are effected by both ordering structures. While $T_N^{\parallel}(x)$ and $T_N^{\perp}(x)$ are different for $x < 0.85$, $T_C^{\perp}(x)$ and $T_N^{\parallel}(x)$ seem to coincide for $x > 0.85$ according to the susceptibility measurements [5].

In 1930, Bloch calculated the low temperature behaviour of the spontaneous magnetization for the Heisenberg ferromagnet and found a $T^{3/2}$ law [12]. For the ferromagnets CrBr_3 , EuS and GdMg it was shown experimentally that the fourth-order exchange interactions change Bloch's $T^{3/2}$ law into a T^2 law [3,5]. These interactions seem to be decisive for the spin dynamics of both order parameters not only for $T \rightarrow 0$ but also in the critical temperature range [5]. By analogy, the antiferromagnetic Heisenberg order parameter *i.e.* the sublattice magnetization $M_{\text{sub}}(T)$ should likewise be modified by the fourth-order exchange interactions. For antiferromagnets a T^2 law has been calculated for $M_{\text{sub}}(T)$ as a first-order approximation for $T \rightarrow 0$ if only bilinear interactions are considered and if the ground state is assumed to be of the Néel type [13]. These assumptions are not correct in the presence of fourth-order exchange interactions and two mutually perpendicular order parameters. Nevertheless, it is observed here that $M_{\text{sub}}(T)$ exhibits a T^2 law, but this law holds up to $0.75T_N$. Such a large validity range cannot be expected for the series expansion used in calculating the leading T^2 term for conventional antiferromagnets. Since the T^2 law observed for $M_{\text{sub}}(T)$ expands over a temperature range which is as large as that one for the spontaneous magnetization of ferromagnets, it is natural to consider this law also as a consequence of the fourth-order interactions. The strong composition

dependence of the normalized T^2 coefficient, $b(x)$, confirms this view, showing that the total magnetic couplings are not simply proportional to x but contain one further term proportional to x^2 . The x^2 term is characteristic for three-spin interactions which occur with a probability proportional to x^2 (see Appendix). Also in $\text{Eu}_x\text{Sr}_{1-x}\text{S}$ the observed x^2 term in $\Theta_1(x)$ could be shown to originate in three-spin interactions while the coefficient of the linear term is given by bilinear and biquadratic interactions [2].

2 Neutron diffraction experiments

Two different sets of neutron diffraction measurements have been performed on single crystal material. A first run of measurements has been done on instrument D10 of the Institut Laue-Langevin in Grenoble using a neutron wavelength of 0.127 nm. A final set of measurements including a vertical magnetic field has been obtained on instrument E1 of the Hahn-Meitner-Institut in Berlin using a wavelength of 0.24 nm. Due to the strong absorption of Europium at this large wavelength the penetration depth for the neutrons is only of the order of 0.1 mm. Therefore, thin platelets have been cleaved from larger single crystal ingots. These rectangular pieces can easily be oriented according to their (100) cleavage planes. It is therefore possible to cover larger areas with oriented single crystal pieces and to compensate in this way for the small penetration depth. The observed line broadening due to misorientations of the composite sample is not substantially larger than the inherent mosaic spread. The latter is as large as two degrees for uncleaved material.

In order to calibrate the observed MnO-type superstructure reflection intensities against the nuclear Bragg intensities five $\text{Eu}_x\text{Sr}_{1-x}\text{Te}$ powder samples have been measured additionally on instrument D1B at ILL. Using powder samples, intensity errors due to extinction effects can be avoided. Such effects could clearly be noticed in the single crystal experiments.

Figure 1 shows the normalized sublattice magnetization as function of the squared reduced temperature for one $\text{Eu}_x\text{Sr}_{1-x}\text{Te}$ sample with $x = 0.85$. The sublattice magnetization is obtained as the square root of the integrated Bragg intensity of the magnetic (1/2 1/2 1/2) superstructure reflection. It should be recalled that the as evaluated $M_{\text{sub}}(T)$ contains contributions from both types of order parameters. It can clearly be seen that the variance of $M_{\text{sub}}(T)$ follows a T^2 law over a temperature range up to $(T/T_N)^2 = 0.53$ *viz.* $T/T_N = 0.73$. Considering that $T_N = 8.1$ K for $x = 0.85$, the T^2 law holds up to $T = 5.9$ K. This can only be rationalized if the interaction responsible for the T^2 law is of the order of 5.9 K. Anisotropy and dipole-dipole coupling are evidently much too small to account for such a large energy.

Biquadratic and three-spin interactions have both the right magnitude. In reference [1] it was shown that $\Theta_3 = +2.8$ K for EuTe and -2.9 K for $\text{Eu}_{0.65}\text{Sr}_{0.35}\text{Te}$. Considering furthermore that $\Theta_3 = 0$ for $x = 0.85$ and that only biquadratic and three-spin interactions are important

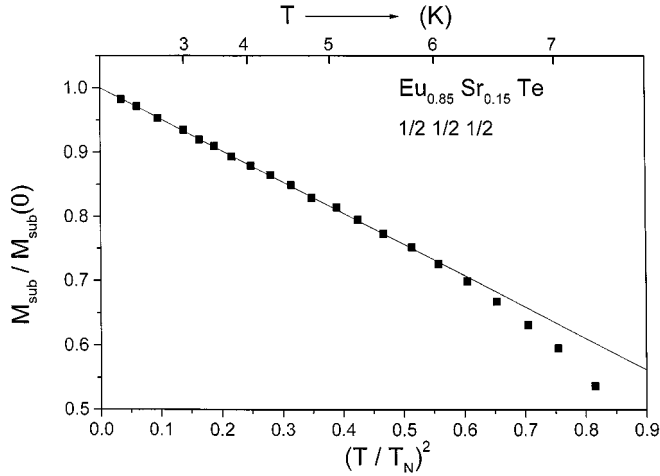


Fig. 1. Normalized sublattice magnetization (square root of integral intensity) of the antiferromagnetic (1/2 1/2 1/2) reflection for $\text{Eu}_{0.85}\text{Sr}_{0.15}\text{Te}$ versus square of reduced Néel temperature. The observed T^2 law holds up to $0.7T_N$.

[2,4], a fit of a quadratic function for $\Theta_3(x)$ results into

$$\Theta_3(x) = -17.6 \cdot x + 20.6 \cdot x^2.$$

It is very surprising that both coefficients are even larger than the Néel temperature of EuTe ($T_N = 9.8$ K). This shows that the competition between antiferromagnetic bi-quadratic interactions and ferromagnetic three-spin interactions is very strong but both interactions contribute with a reduced rate only to T_N^{\parallel} . From reference [2] it results that three-spin interactions contribute the strongest to T_N^{\parallel} with a rate of $\approx 1/3$. Moreover, it becomes clear that fourth-order exchange interactions are strong enough to define the spin dynamics for all temperatures.

Figure 2 compares the normalized sublattice magnetizations as function of the squared reduced temperatures for three samples with $x = 1$ (EuTe), $x = 0.85$ and $x = 0.65$. If the effective interactions would change proportional to x *i.e.* if the exchange integrals are assumed to be constants no differences should be visible in this scaled representation. It can, however, clearly be seen that the slope of the T^2 law gets steeper with dilution. This feature we attribute to the existence of three-spin interactions which, ideally, contribute to the total interaction strength proportional to x^2 (see Appendix).

In Figure 3 we show the composition dependence of the coefficient $b(x)$ of the T^2 term defined by

$$\frac{M_{\text{sub}}(T)}{M_{\text{sub}}(0)} = 1 - b(x) \cdot \left(\frac{T}{T_N}\right)^2 + \dots \quad (4)$$

As can be seen from Figure 3, $b(x)$ changes in a nearly linear way with composition. This is characteristic for three-spin interactions and confirms again that four-spin interactions are less important. Otherwise $b(x)$ should contain one further term proportional to x^2 . A similar conclusion has been drawn from an analysis of the measured “biquadratic” molecular field constant $B_{ex}^q(x)$ which includes the effects of all fourth-order interaction processes

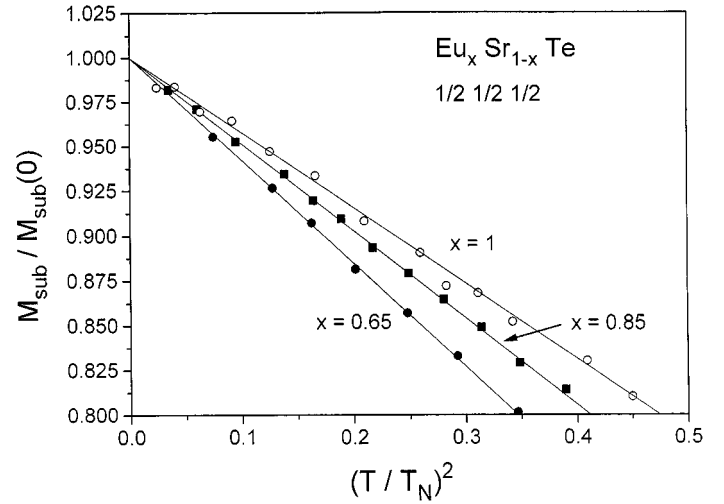


Fig. 2. Comparison of the temperature dependence of the normalized sublattice magnetizations for three $\text{Eu}_x\text{Sr}_{1-x}\text{Te}$ samples. The different slopes indicate an x^2 -dependence in the effective interactions.

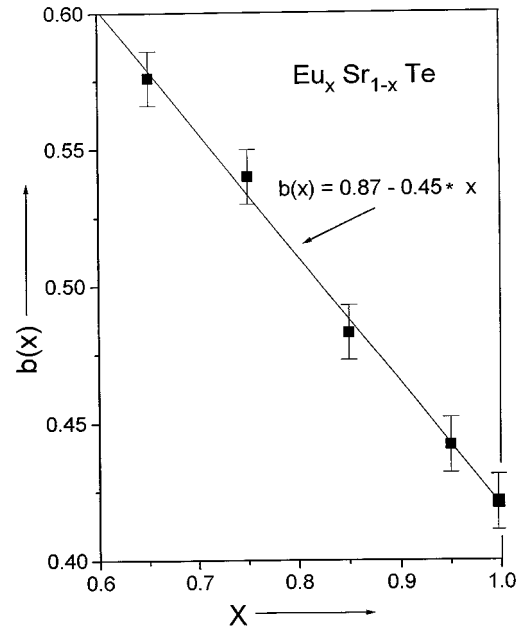


Fig. 3. Composition dependence of the normalized T^2 coefficient, $b(x)$, defined by equation (4). The linear x -dependence verifies the existence of three-spin interactions and shows that four-spin interactions are negligible.

[2,4]. Fitting a linear function through the data points in Figure 3 leads to $b(x) = 0.87 - 0.45 \cdot x$. From this result we have to conclude that without three-spin interactions the coefficient b would be 0.87.

For EuTe ($x = 1$) three-spin interactions change b from 0.87 to 0.42. In other words, three-spin interactions reduce $b(x)$ and keep therefore the sublattice magnetization high with increasing temperature. This stabilization is a much surprising effect of a ferromagnetic three-spin interaction in an antiferromagnet. On the other hand, the possible

ordered moment configurations due to three-spin exchange interactions are not sufficiently investigated theoretically at present (see however Refs. [9, 14]).

Another detail with Figure 3 is worth mentioning: there is no definite anomaly noticeable at $x_c = 0.85$ where the order of the transverse moment components changes from ferromagnetic for $x > x_c$ to antiferromagnetic for $x < x_c$. The insensitivity of $b(x)$ against this phase change shows again that the spin dynamics of both order parameters must be very similar.

For $x < x_c$ the situation is particularly clear. According to the magnetic measurements [4, 5] we are dealing with two antiferromagnets in the same material. Both have clearly different Néel temperatures (T_N^{\parallel} and T_N^{\perp}) and considerably different critical field curves (B_c^{\parallel} and B_c^{\perp}). Since only half-integer scattering lines of the MnO type are observed in neutron diffraction we have to conclude that the associated order parameters, O_2 and O_4 , are mostly of the MnO-type and give rise to the same type of diffraction lines. The due restriction is that the observed MnO-type scattering intensities do not correspond to the fully ordered magnetic moment of Eu.

For $x > x_c$, on the other hand, the half-integer scattering lines should sample the antiferromagnetic Heisenberg order parameter O_2 individually if O_4 is ferromagnetic. This idea must be questioned in view of missing ferromagnetic Bragg lines and a rather small decrease of the absolute MnO-type scattering intensities at $x_c = 0.85$ where the transverse order changes from antiferromagnetic to ferromagnetic.

In order to avoid extinction effects which are clearly noticed in single crystal studies the absolute intensity measurements have been performed on powder samples. Figure 4 shows the evaluated ordered moment per Eu atom obtained by a calibration of the half-integer scattering intensities against the nuclear Bragg intensities using the known scattering lengths of the elements involved. Although the absolute values of the ordered moments are subject to considerable uncertainties, the relative errors in the data in Figure 4 are not larger than the size of the symbols.

There are several important details with the results of Figure 4 to be stressed. First, for all compositions too small ordered moments result by the much too weak MnO-type scattering intensities. This we consider as a consequence of the antiferromagnetic biquadratic interactions which require perpendicular moment orientations. Though we term both antiferromagnetic, bilinear and biquadratic interactions are in conflict with each other such that the MnO-type of order will be disturbed. This is thought to be the reason for the reduced scattering intensities.

Second, a linear intensity increase is observed with composition x for the samples with $x = 0.9, 0.95$ and $x = 1.0$. This we explain by three-spin interactions. The positive slope then means that even though three-spin interactions are ferromagnetic, they support the antiferromagnetic order of the conventional order parameter. This seems to be a contradiction but we must consider that also a ferromagnetic biquadratic interaction would

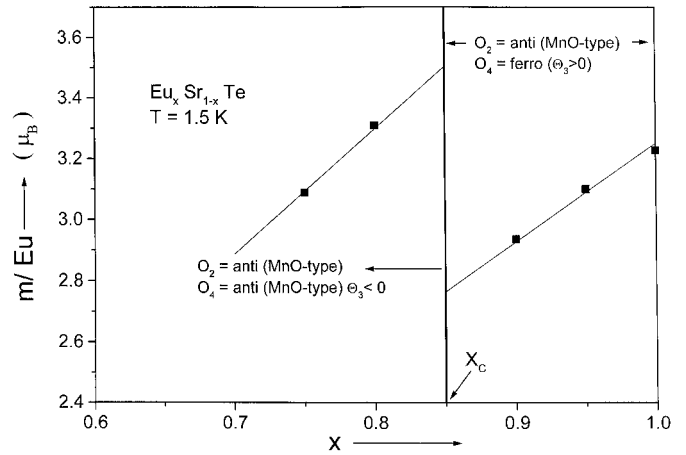


Fig. 4. Ordered antiferromagnetic saturation moment per Eu atom sharing the MnO-type of order as function of composition in $\text{Eu}_x\text{Sr}_{1-x}\text{Te}$. The relative errors of the data are not larger than the size of the symbols. At x_c the total fourth-order interaction changes from antiferromagnetic for $x < x_c$ to ferromagnetic for $x > x_c$.

support the conventional antiferromagnetic order of the magnetic ground state because due to the quadrature parallel and antiparallel orientations are both energetically preferred. It must be noted that the just given interpretation is in keeping with the result for $b(x)$ given in Figure 3. This quantity also showed that the ferromagnetic three-spin interactions support the antiferromagnetic order in that they reduce $b(x)$ and stabilize, hence, the MnO-type magnetic order for increasing temperatures.

An alternative explanation for the strong decrease of the MnO-type scattering intensities with diamagnetic dilution would be given by random site effects. The observed linear decrease of the ordered moment seems, however, much too strong for a diamagnetically diluted Heisenberg system. Computer simulations have shown that the ordered saturation moment stays at its full value for considerable dilution concentrations [15].

The observed linear increase of the ordered moment with increasing x for $x > 0.85$ as well as for $x < 0.85$ is such that for $x \rightarrow 0$ virtually no ordered moment will result by extrapolation. This means that the observed ordered moment is stabilized nearly exclusively by three-spin interactions. In other words, in the absence of three-spin interactions virtually no diffraction intensities would be observed. Apparently the antiferromagnetic biquadratic interactions are strong enough to destroy the MnO-type magnetic order required by the antiferromagnetic bilinear interactions nearly completely. Only the ferromagnetic three-spin interactions support this order and give rise to at least weak MnO-type scattering intensities.

Third, the change in order of the transverse moment components from antiferromagnetic for $x < x_c$ to ferromagnetic for $x > x_c$ is noticed by a sudden decrease of the antiferromagnetically ordered moment. This is what we expect but the observed effect corresponds only to $0.6 \mu_B$ of the total moment of $7 \mu_B$ and is therefore very weak.

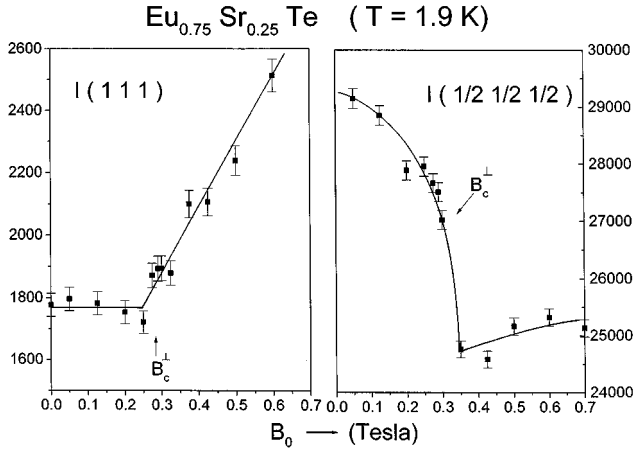


Fig. 5. Field dependence of ferromagnetic (1 1 1) and antiferromagnetic (1/2 1/2 1/2) scattering intensity for an $\text{Eu}_{0.75}\text{Sr}_{0.25}\text{Te}$ sample at $T = 1.9$ K revealing the phase transition at $B_c^\perp(T)$. In contrast to the magnetization which shows a linear field dependence no field induced magnetic scattering intensities are observed for the (1 1 1) reflection in the field range $0 < B_0 < B_c^\perp$.

Of course, one should be careful in drawing quantitative conclusions from data with unclear absolute values.

For all samples with $x < 0.85$ a second critical field curve $B_c^\perp(T)$ is observed in magnetization measurements [4,5]. This phase boundary can also be observed with neutron scattering. Figure 5 shows the field dependence of the integrated scattering intensities of one ferromagnetic (111) and one antiferromagnetic (1/2 1/2 1/2) diffraction line at a temperature of 1.9 K for one sample with $x = 0.75$. The magnetic field was vertical *i.e.* perpendicular to the scattering plane in these experiments. A sudden decrease of the antiferromagnetic (1/2 1/2 1/2) intensity near the phase transition at B_c^\perp can clearly be noticed. This intensity decrease is due to a vanishing magnetic order of the transverse moment components. Assuming that for fields $B_0 > B_c^\perp$ the antiferromagnetic order of the transverse moment components has disappeared completely, the diffraction intensity of this order parameter can be estimated to be 0.17 of that of the longitudinal order parameter. The ratio of the ordered transverse and longitudinal moments is therefore 0.41. In contrast to this, the decrease of the MnO scattering intensity at x_c which must also be ascribed to the disappearance of the transverse antiferromagnetic phase (see Fig. 4) is much less than that one observed at B_c^\perp in Figure 5.

In the limit $B_0 \rightarrow 0$, the observed (111) scattering intensity in Figure 5 is given by nuclear scattering processes only for the sample with $x = 0.75$. No field induced additional magnetic scattering intensity appears in the field range up to the critical field $B_c^\perp(T = 1.9 \text{ K}) = 0.3$ tesla. This is much surprising since it is at variance with the magnetization $m(B_0)$ which exhibits a linear increase with field in the range $0 < B_0 < 0.3$ T (see Fig. (3) of Ref. [5]). The slope of the $m(B_0)$ curve is only slightly smaller for $B_0 < B_c^\perp$ than for $B_0 > B_c^\perp$. It could reasonably be expected that the ferromagnetic (111)

intensity should increase in the same way the antiferromagnetic (1/2 1/2 1/2) intensity decreases in the field range $0 < B_0 < B_c^\perp$.

We interpret the suppression of the ferromagnetic scattering intensities as due to a very inhomogeneous spin order of the field induced ferromagnetic component such that diffraction conditions are not fulfilled. This inhomogeneity may be caused by the competing individual interaction processes within the class of fourth-order interactions, but also by some geometrical incompatibility of a transverse ferromagnetic component coexisting with a much stronger longitudinal antiferromagnetic component on the fcc lattice. By the same reason no ferromagnetic Bragg intensities seem to be observed for the samples with $x > 0.85$ for which the resulting fourth-order interactions are ferromagnetic. For EuTe the rise of the ferromagnetic Bragg intensities with field is also delayed compared to the rise of the magnetization.

3 Conclusions

A T^2 law has been observed for the low-temperature behaviour of the sublattice magnetizations $M_{\text{sub}}(T)$ of all diamagnetically diluted antiferromagnets $\text{Eu}_x\text{Sr}_{1-x}\text{Te}$. This conforms to early zero field NMR measurements on EuTe which also revealed a T^2 law for the hyperfine field [18]. Since the hyperfine field samples the local polarization it can be assumed to be proportional to $M_{\text{sub}}(T)$.

In the present work $M_{\text{sub}}(T)$ was obtained from the scattering intensities of the MnO-type magnetic reflections with half-integer indices. Since ordering structures due to second-order (bilinear) and fourth-order interactions contribute to these MnO-type magnetic reflections a superposition of two order parameters is observed. As a consequence, the observed T^2 law must apply to both types of order parameters. The unusually large validity range of the T^2 law, we believe, shows that this law has a different origin than the T^2 law calculated in first approximation for the Heisenberg antiferromagnet with a Néel ground state [13]. We consider the T^2 law of $M_{\text{sub}}(T)$ as a consequence of the fourth-order interactions. These interactions are known to dominate the spin dynamics of both order parameters [3] leading to the same temperature dependence for both. Among the class of fourth-order interactions individual interaction processes exist which are strong enough to stabilize the T^2 law even for the Heisenberg order parameter (O_2) up to $0.75 T_N^\parallel$ [1,2].

It is very surprising that for the spontaneous magnetization of ferromagnets (such as EuO , EuS , GdMg and CrBr_3) and the sublattice magnetization of antiferromagnets the same T^2 law is observed over a comparably wide temperature range. The common exponent might be fortuitous but considering that, due to the quadrature, the biquadratic interaction energies are the same for ferromagnetic and antiferromagnetic moment orientations, identical exponents appear not unreasonable. This would further support the view that the T^2 law originates in fourth-order interactions [5].

This work was intended to use the microscopic method of neutron scattering to confirm the hypothesis that a second ordering structure is generated by the fourth-order exchange interactions. For $x < 0.85$ the high-temperature average of all fourth-order interactions is antiferromagnetic ($\Theta_3 < 0$) and, consistently, a second antiferromagnetic phase noticed by a second critical field curve $B_c^\perp(T)$ is observed in magnetization [4,5] and neutron scattering measurements. Due to the different and competing individual interaction processes both ordering structures are considerably perturbed and give rise to rather weak half-integer Bragg reflections of the MnO-type. From the observed decrease of the antiferromagnetic (1/2 1/2 1/2) scattering intensity at B_c^\perp it could be estimated that the transverse order parameter is 0.41 of the longitudinal order parameter in the case of a sample with $x = 0.75$.

From the composition dependence of the evaluated MnO-type saturation moment indications are obtained that the antiferromagnetic biquadratic interactions destroy the basic MnO order. These interactions have the same composition dependence as the bilinear interactions ($\sim x$) and reduce the ordered moment per Eu^{2+} by a constant amount for all compositions. In contrast to this the ferromagnetic three-spin interactions seem to support the antiferromagnetic order of the conventional order parameter giving rise to an ordered moment per Eu^{2+} increasing linearly with x . We could estimate that without the existence of ferromagnetic three-spin interactions virtually no MnO scattering intensities would be observed. Since antiferromagnetic biquadratic interactions require a perpendicular moment orientation they are in conflict with the antiferromagnetic Heisenberg interactions and perturb the MnO-type order. On the other hand, ferromagnetic three-spin interactions seem to act similar like ferromagnetic biquadratic interactions in that they support the antiferromagnetic order of the ground state. Of course, a second energy minimum occurs at the ferromagnetic orientation. The terms ferromagnetic and antiferromagnetic have therefore a completely different meaning than usual if they refer to ordered structures induced by the fourth-order exchange interactions.

By analogy, for $x > 0.85$ the transverse order parameter should be ferromagnetic since $\Theta_3 > 0$. The rise of this ferromagnetic component is noticed only indirectly by a sudden decrease of the MnO-type scattering intensities at $x_c = 0.85$ (see Fig. 4). This decrease with composition is surprisingly small compared to the loss of the scattering intensity as function of field at B_c^\perp associated with the disappearance of the antiferromagnetic transverse order parameter for the samples with $x < 0.85$. A positive identification of the ferromagnetic order for $x > 0.85$ using neutron scattering failed. Powder diffraction spectra on EuTe showed virtually no ferromagnetic Bragg lines [4].

On the other hand, we cannot exclude that the postulated ferromagnetic transverse order has a very uncommon superstructure and contributes also to the observed MnO-type scattering intensities. This explanation appears highly speculative but it is not unlikely. In particular, three-spin interactions are able to stabilize spin

orientations between next-nearest neighbour spins over one intermediate spin with a rather unimportant orientation thus creating a ferromagnetic superstructure with a period of twice the lattice constant. In this way we can think also of a transverse ferromagnetic order, the scattering intensities of which add to the half-integer MnO diffraction lines of the conventional order parameter. The relatively small drop of the ordered moment at $x_c = 0.85$ (see Fig. 4) could then be explained by the additional MnO-type scattering intensities originating in this ferromagnetic transverse superstructure for $0.85 < x < 1.0$.

Additionally it must be considered that for $x > 0.85$ the ferromagnetic three-spin interactions dominate over the antiferromagnetic biquadratic interactions. No simple or collinear spin order can be expected under such a competition. This will reduce the observed scattering intensities in general. The postulated “ferromagnetic” order parameter might therefore exist even if there are no conventional Bragg diffraction intensities observed in standard neutron scattering spectra. The missing scattering intensities might be observable only as a diffuse component which is difficult to observe for these strongly absorbing samples.

Though the existence of an antiferromagnetically ordered phase due to fourth-order interactions could be confirmed with neutron scattering on account of a second critical field and the observation of additional MnO-type diffraction intensities, no information was obtained on the orientation of this ordered component relative to the conventional order parameter. Up to now it was shown only for GdMg with neutron scattering [17] and magnetization measurements [3] that both ordered components are perpendicular to each other. Further magnetic systems need to be investigated in order to establish the mutually perpendicular orientation of second-order (O_2) and fourth-order (O_4) order parameters as a general principle.

Appendix

It is generally accepted that the paramagnetic Curie-Weiss temperature $\Theta_1(x)$ is in good approximation a linear function of the concentration x of the magnetic moments provided that 1) magnetic and non-magnetic ions are distributed at random, 2) that the interaction parameters are constants *i.e.* do not change with composition x and 3) that three-spin and four-spin interactions are absent. $\Theta_1(x)$ will deviate from the linear relation $\Theta_1(x) = x * \Theta(1)$ only for concentrations x which are sufficiently small such that the probability for any reference spin to find an interaction partner within the reach of the exchange interactions tends to zero. In the following we will give an estimate for this lower bound of x .

As a matter of definition $\Theta_1(x)$ gives the total interaction a reference spin has with all its interaction partners around. Since it is known that the exchange interactions in the Eu chalcogenides are restricted to nearest and next-nearest neighbours [19] only a coordination of $z = 12 + 6$ has to be considered. Θ_1 is then given by the threefold product consisting of the probability that the reference

site will be occupied by a magnetic atom ($\sim x$) times the average number of interaction partners within the interaction radius, times the exchange constant to these partners (for simplicity we will assume that the exchange constants to all neighbour shells are equal). Using Poisson statistics *i.e.* assuming a continuous lattice the probability of finding one of the z coordination sites occupied by a magnetic moment is given by

$$w_1 = (1 - e^{-z*x}). \quad (\text{A.1})$$

With $z = 18$ the probability w_1 reaches $w_1 \rightarrow 1$ for $x \rightarrow 1$ with a sufficient accuracy. Since the incorporation of the magnetic ions on the z coordination sites is considered as completely independent events occurring with the same probability the average number of interaction partners is given by

$$N_z = z * (1 - e^{-z*x}) \quad (\text{A.2})$$

Θ_1 is therefore proportional to $x * N_z$.

As can easily be verified, the expression in brackets is > 0.99 for $x > 0.25$. For $x > 0.38$ the approximation $\Theta_1 \sim z * x$ holds to within an error of smaller than 10^{-3} . Since we are interested in the composition range with a long-range magnetic order *i.e.* in samples with $x \geq 0.5$ the approximation $\Theta_1 \sim z * x$ is sufficiently correct. On the other hand, for concentrations with $z*x \ll 1$ *i.e.* $x \ll 0.056$ the expression in brackets can be linearized and now $\Theta_1 \sim x^2$. In this limit the magnetic interactions are restricted to isolated magnetic pairs which occur with a probability $\sim x^2$.

It is evident that the probability that three spins are close enough to interact with each other is given by the probability for pair formation ($\sim x^2$) times the probability that both partners have one third interaction partner which is given by the squared expression of equation (A.1). Also in this case $(w_1)^2$ approaches unity for the aforementioned concentrations x . The evaluation of the correct coordination numbers for three-spin interactions requires, however, more elaborate geometrical considerations for each lattice type (see for instance Ref. [20]).

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