Eur. Phys. J. B **8**, 207–216 (1999)

THE EUROPEAN PHYSICAL JOURNAL B

EDP Sciences

On the T^2 Bloch law in magnets with fourth-order exchange interaction

R.M. Mueller, U. Köbler, and K. Fischer

Institut für Festkörperforschung, Forschungszentrum Jülich, 52425 Jülich, Germany

Received 23 July 1998 and Received in final form 12 October 1998

Abstract. For the ferromagnets EuS and GdMg, in which fourth-order exchange interactions (*i.e.* biquadratic, three-spin and four-spin interactions) have been identified, the deviation of the spontaneous magnetization with respect to the $T = 0$ value is shown to follow a T^2 law instead of the famous $T^{3/2}$ law expected for a Heisenberg ferromagnet. Moreover, the observed T^2 law holds for temperatures as large as $0.8T_C$ and the extrapolated magnetization value for $T \to 0$ does not conform to ferromagnetic saturation. This is because the fourth-order exchange interactions generate a second order-parameter which is assumed to govern the order of the transverse moment components. These moment components have a finite expectation value for $T \to 0$ at the expense of the Heisenberg order parameter. Like the spontaneous magnetization, the critical field curves $B_c(T)$ of the metamagnet EuSe and the antiferromagnet EuTe also start decreasing with a T^2 term for $T \to 0$. It is argued that the T^2 law is a consequence of the fourth-order exchange interactions. This is shown experimentally by a study of the critical field curves $B_c^{\parallel}(T \to 0)$ pertinent to the longitudinal (Heisenberg) order-parameter in the diamagnetically diluted antiferromagnets $Eu_xSr_{1-x}Te$. In this solid solution series a particular composition of $x_c = 0.85$ exists at which the different fourth-order interaction processes compensate each other in the high temperature average. As a consequence, an $Eu_xSr_{1-x}Te$ sample with $x = 0.85$ meets the requirements of a Heisenberg antiferromagnet at least if a quantity is considered for which the high-temperature average over all fourth-order interactions is decisive. This seems to be the case for the critical field curve $B_c^{\parallel}(T \to 0)$ which gives the phase boundary to the paramagnetic phase. In fact, a crossover from a T^2 to a $T^{3/2}$ law is observed for $B_c^{\parallel}(T \to 0)$ on approaching x_c. This, we believe, shows the frequently observed T^2 law is caused by the fourth-order interactions.

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 the year 1930, Bloch, using the spin-wave concept, showed theoretically that the spontaneous magnetization of a Heisenberg ferromagnet with bilinear exchange interactions to nearest neighbours decreases like $M(T) = M(0)[1 - a \cdot T^{3/2} + \cdots]$ in first approximation [1]. In 1961, Gossard, Jaccarino and Remeika attempted to verify Bloch's theoretical prediction for the newly discovered insulating ferromagnet $CrBr_3$ [2]. Gossard *et al.* evaluated the spontaneous magnetization of $CrBr_3$ on account of the ^{53}Cr hyperfine field using zero-field NMR frequency measurements. These data revealed that the spontaneous magnetization could not be described adequately by only a $T^{3/2}$ term but that the next higher $T^{5/2}$ term and one anisotropy field had to be included in the fit in order to reproduce the experimental data correctly.

In view of frequently observed T^2 laws either for the critical field curves of EuSe and EuTe [3] or the spontaneous magnetization of GdMg [4], all of which are materials in which fourth-order exchange interactions (*i.e.* biquadratic, three-spin and four-spin interactions) have been identified, we re-plotted the ^{53}Cr NMR frequencies of reference [2] over T^2 (see Fig. 1) and observed that a single T^2 term is able to describe the experimental NMR data quite well. Unfortunately, the measurements of reference [2] were limited to the temperature range $1.2 \leq T \leq 4.2$ K and therefore the T^2 law is only confirmed to hold for a T/T_C ratio of 4.2 K/37 K = 0.11.

The same situation observed for $CrBr_3$ is also found for EuS. Figure 2 gives the zero-field ^{153}Eu NMR resonance frequencies taken from reference [5] but also plotted *vs.* T^2 for the same temperature range of $1.2 \leq T \leq 4.2$ K. Again, the ^{153}Eu hyperfine field of EuS obeys a T^2 law at

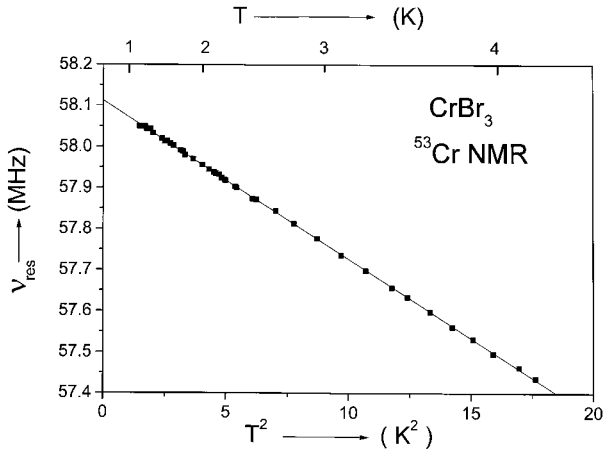

Fig. 1. ^{53}Cr zero-field NMR frequencies of the ferromagnet $CrBr_3$ taken from reference [2] but plotted *vs.* squared absolute temperature. Assuming the hyperfine field sampled by the NMR signal to be proportional to the spontaneous magnetization, a single T^2 term (straight-line) is sufficient to describe the temperature dependence of the spontaneous magnetization.

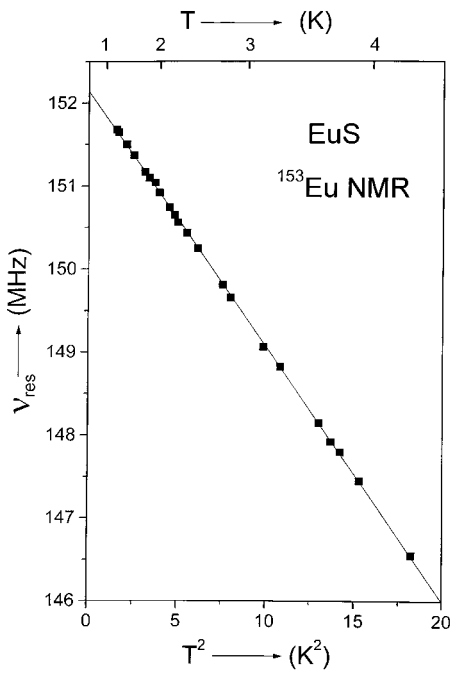

Fig. 2. ^{153}Eu zero-field NMR frequencies of the ferromagnet EuS taken from reference [5] but plotted *vs.* squared absolute temperature. Since bilinear and biquadratic exchange interactions are assumed to generate two perpendicular ferromagnetic order-parameters in EuS [8] the observed T^2 law (straight-line) should be common to both order-parameters. The T^2 law holds at least until a $\frac{T}{T_C}$ ratio of $\frac{4.2\ \mathrm{K}}{16.6\ \mathrm{K}} = 0.26$.

least until a T/T_C ratio of 4.2 K/16.3 K = 0.26 with high precision.

GdMg is another ferromagnet for which the spontaneous magnetization was reported to exhibit a T^2 law over a temperature range as large as $0.8T_C$ [4]. Since in EuS [6] and GdMg [7] appreciable fourth-order interactions have been identified it is to be assumed the T^2 law is a consequence of these interactions. This we want to demonstrate in the present work with investigations of the critical field $B_c^{\parallel}(T)$ of the antiferromagnetic longitudinal (Heisenberg) order-parameter in the diamagnetically diluted antiferromagnets $Eu_xSr_{1-x}Te$.

In recent measurements of the transverse susceptibility of GdMg it was shown that the class of fourth-order exchange interactions generates a second order-parameter which is oriented perpendicular to the common bilinear (Heisenberg) order-parameter [7]. This conclusion is consistent with earlier neutron scattering experiments [4]. In GdMg the Curie temperature of the longitudinal (Heisenberg) order-parameter is $T_C^{\parallel} = 110$ K but the Néel temperature of the transverse order-parameter is $T_N^{\parallel} = 91$ K. Therefore, in measurements in which both order-parameters are not distinguished by a magnetic field or some other means as is the case in zero-field NMR measurements, a mixture of both order-parameters will be measured. As a consequence, the observed T^2 law must be common to both order-parameters. This argument is at least correct if both order-parameters have a similar ordering type as is the case for EuS in which both order-parameters are ferromagnetic [3,6,8]. In GdMg, on the other hand, the transverse order-parameter is antiferromagnetic and, as a consequence, contributes not much to the longitudinal magnetization measurements. It is therefore easily possible to align the ferromagnetic component by a magnetic field and to measure its properties separately with conventional field-parallel magnetization measurements. The T^2 law observed in this way for GdMg can be assumed to be a property of the ferromagnetic bilinear (Heisenberg) order-parameter.

Although there exists a number of theoretical studies dealing with biquadratic and three-spin interactions on a mean field level [9–11] there is presently, to the best of our knowledge, no theory available which would explain the T^2 law. We therefore tried to prove experimentally whether the T^2 law originates in the fourth-order interactions. To this end a Heisenberg ferromagnet with definitely no fourth-order exchange interactions is needed. Such a material is not known.

In the following section we will briefly review the two methods used for the identification of fourth-order exchange interactions in the diamagnetically diluted antiferromagnetic compounds $Eu_xSr_{1-x}Te$. Both methods infer this information from an analysis of the curvature of the magnetic isotherms as function of a magnetic field and provide average values over all individual fourth-order interaction processes. It turned out that the observed ferromagnetic total fourth-order interaction sum pertinent to all Europium monochalcogenides [3] is composed of much stronger antiferromagnetic biquadratic interactions and slightly dominating ferromagnetic three-spin interactions [6]. To a good approximation it can be assumed that biquadratic interactions vanish proportional to x in a diluted system but three-spin interactions vanish proportional to x^2 (the limitations of this assumption for small

x values will be outlined in the Appendix of the following article). Therefore, there exists one particular composition of $x_c = 0.85$ at which antiferromagnetic biquadratic and ferromagnetic three-spin interactions cancel in the high temperature limit though they still have finite strengths microscopically [12]. The critical field curve $B_c^{\parallel}(T)$ pertinent to the Heisenberg order-parameter, being the phase boundary to the paramagnetic phase, can be expected to be defined by the paramagnetic average over bilinear (second-order) and fourth-order interactions because the magnetic moments are nearly ferromagnetically aligned along $B_c^{\parallel}(T \to 0)$. Since the fourth-order interactions vanish for $x \to 0.85$ only the Heisenberg interactions should be active at $x_c = 0.85$. In fact, we will show here that this is correct for $B_c^{\parallel}$ and that $B_c^{\parallel}(T \to 0)$ exhibits a crossover from a T^2 law to a $T^{3/2}$ law on approaching $x_c = 0.85$.

On the other hand, the transverse order-parameter is defined exclusively by the fourth-order interactions and should react in a direct way on the sign change of the fourth-order interaction sum at $x_c = 0.85$. In fact, for $x < 0.85$, where the fourth-order interaction sum is negative, a second antiferromagnetic phase is observed in addition to the conventional antiferromagnetic phase present for all $Eu_xSr_{1-x}Te$ samples [12]. The spin structure of both ordering types is essentially of the MnO type but the Néel temperatures of the transverse order parameter are slightly lower than the Néel temperatures of the longitudinal order parameter [12] (see Fig. 13). Moreover, the critical field values $B_c^{\perp}(T \to 0)$ of the transverse order parameter are only about $0.3T$ in contrast to the critical field values $B_c^{\parallel}(T \to 0)$ of the longitudinal order parameter which are in the range 6–7 tesla.

For the samples with $x > 0.85$, for which the fourth-order interaction sum is ferromagnetic, indications are given for a ferromagnetic order of the transverse order-parameter. This shows that the ordering type of the transverse order-parameter is defined by the sign of the fourth-order interaction sum *i.e.* Θ_3, pretty much as the order of the longitudinal order-parameter is defined essentially by the conventional Heisenberg interaction sum *i.e.* Θ_1. In other words, the transverse ordering processes have the character of an order-disorder phase transition even if they occur at a temperature where a Heisenberg order parameter already exists. It appears therefore plausible that they concern the disordered transverse moment components. This was shown explicitly only for GdMg up to now [7]. A more detailed investigation of the nature of both order-parameters will be the subject of two forthcoming articles [8,13].

2 Identifying fourth-order exchange interactions

Information on the fourth-order interaction strength is provided by the curvature of the magnetic isotherms as function of the applied magnetic field. It has been observed long ago [14] that for temperatures $T \ll T_N$ the magnetization curve of EuTe is not a linear function of the applied magnetic field as might be anticipated for an antiferromagnet with only bilinear interactions but instead it increases faster than linearly with field until the critical field $B_c^{\parallel}$ where the system undergoes a second-order transition into the paramagnetic state (see Fig. 3). Computer simulations revealed also a linear magnetization curve for $T = 0$ if only bilinear interactions are considered, irrespective of the range of these interactions [15].

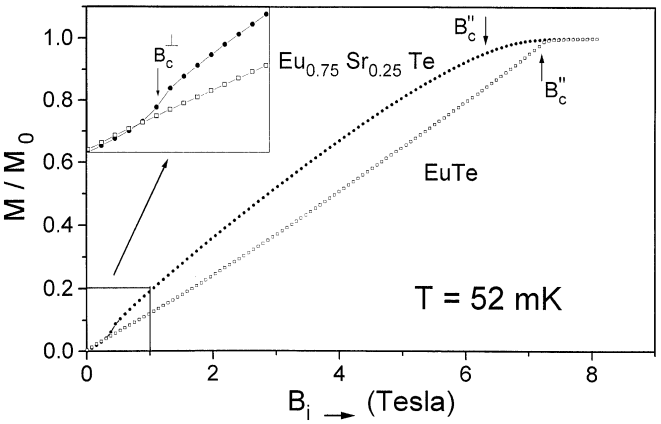

Fig. 3. Low-temperature magnetization curves of EuTe and $Eu_{0.75}Sr_{0.25}Te$ showing different curvatures owing to a ferromagnetic (EuTe) and an antiferromagnetic ($Eu_{0.75}Sr_{0.25}Te$) fourth-order interaction sum. In the antiferromagnetic case the critical field $B_c^{\perp}$ of the transverse order-parameter appears additionally at ~ 0.35 tesla (see inset). The critical field $B_c^{\parallel}$ of the longitudinal order-parameter can clearly be seen only in the differentiated magnetization curves (see Fig. 5).

Using a simple molecular field treatment it can easily be shown [12] that for $T \ll T_N$ the curvature of the magnetization curves can be described in terms of a "biquadratic" molecular field constant B_{ex}^q which includes all fourth-order interaction processes according to:

$$B_i = -B_{ex}^l \, m - B_{ex}^q \, m(2m^2 - 1). \qquad (1)$$

Here B_i means the applied magnetic field converted to its value inside the sample, B_{ex}^l means the bilinear (Heisenberg) molecular field constant and m the reduced magnetization. Equation (1) shows that already the linear relation between field and magnetization is affected by the fourth-order interactions but that the curvature of the magnetization curves is due exclusively to the fourth-order interactions. In the case of EuTe the non-linear part of the magnetization curve favours a ferromagnetic moment orientation (the magnetization increases faster than linearly with field, see Fig. 3) and therefore the resulting fourth-order interaction sum is to be termed ferromagnetic. As can also be seen from Figure 3 the sense of the curvature of the magnetization curve has changed for the $Eu_{0.75}Sr_{0.25}Te$ sample. The fourth-order interactions now oppose a parallel moment orientation *viz.* delay magnetic saturation and are therefore called antiferromagnetic. This change in sign was shown to result from dominating antiferromagnetic biquadratic interactions in the diluted

samples but by dominating ferromagnetic three-spin interactions in the concentrated materials including EuTe [6].

Consistent with an antiferromagnetic fourth-order interaction sum for $x < 0.85$ a second anomaly to be ascribed to the critical field $B_c^{\perp}$ of the transverse order-parameter appears in the $x = 0.75$ magnetization curve in Figure 3 at a field value of about 0.35 tesla (see inset). This anomaly is shown more clearly in Figure 10.

The just outlined analysis of the curvature of the magnetic isotherms for $T \ll T_N$ agrees reasonably well with results of a molecular field analysis of the curvature of the high-temperature paramagnetic isotherms. In the paramagnetic phase the curvature of the magnetic isotherms is described by the cubic susceptibility χ_3 according to

$$B_i = \frac{1}{\chi_1}\, m + \frac{1}{\chi_3}\, m^3 + \cdots \qquad (2)$$

As was outlined in references [3,6] not only the linear susceptibility χ_1 but also the cubic susceptibility χ_3 obeys a Curie-Weiss law at sufficiently high temperatures. The associated Curie-Weiss temperature Θ_3 gives the high temperature average over all fourth-order interaction processes. The similar structure of equations (1, 2) shows furthermore that Θ_3 is given only by the fourth-order interactions but Θ_1 – the Curie-Weiss temperature of the linear susceptibility χ_1 – is given by all interactions. This has been shown more rigorously by a high-temperature expansion in reference [6].

The appearance of a second Curie-Weiss term in equation (2) can be taken as indication for a second order-disorder phase transition in addition to the conventional one which is driven by a divergence of the linear susceptibility χ_1. This second ordering process, being driven by a divergence of χ_3, occurs at Θ_3 (in molecular field approximation) and concerns the transverse moment components as revealed from investigations of GdMg [4,7].

Both methods of analysis according to equations (1, 2) deliver an average over all fourth-order interaction processes. $\Theta_3(x)$ gives the high temperature average but also the molecular field constant evaluated from the low temperature magnetization curve averages over all spin orientations between antiparallel for zero field and parallel at the critical field $B_c^{\parallel}$. Both parameters are related by the cubic Curie constant C_3 and agree reasonably well [3], even quantitatively. The results for $B_{ex}^q(x)$ obtained from an analysis of magnetization curves at $T = 50$ mK are given in Figure 4.

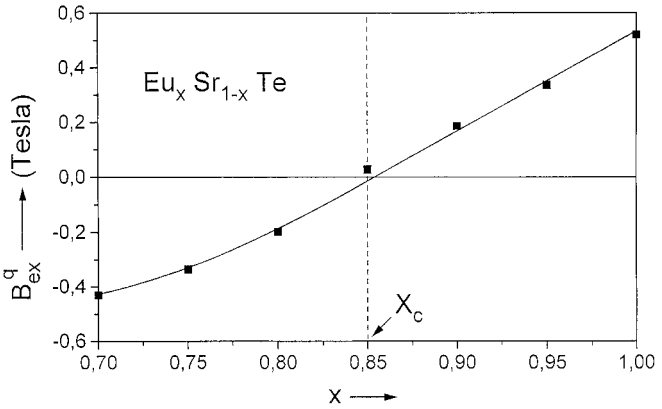

Fig. 4. Biquadratic molecular field constant $B_{ex}^q(x)$ obtained by an analysis of the curvature of low-temperature magnetization curves as are seen in Figure 3. $B_{ex}^q > 0$ means a ferromagnetic fourth-order interaction sum, $B_{ex}^q > 0$ an antiferromagnetic fourth-order interaction sum. $B_{ex}^q(x)$ includes all effects from biquadratic, three-spin and four-spin interactions. The solid line is a guide to the eye.

3 Measurements of the critical field $B_c^{\parallel}(T,\ x)$ of the bilinear order-parameter

Once it is known there are two order-parameters it is important to distinguish them experimentally in order to evaluate their properties individually. This will be a difficult task in neutron scattering experiments if both order parameters give rise to the same type of diffraction lines as is the case in $Eu_xSr_{1-x}S$ and $Eu_xSr_{1-x}Te$. If both order-parameters are antiferromagnetic ($x < 0.85$) they can easily be identified on account of their different critical fields ($B_c^{\perp}$ and $B_c^{\parallel}$) or their different ordering transitions ($T_N^{\perp}$ and $T_N^{\parallel}$). On the other hand, for $x > 0.85$ the ferromagnetic transverse order-parameter will only increase the magnetization values without producing a field induced phase transition similar to $B_c^{\perp}$. Therefore, the observed anomaly at $B_c^{\parallel}(T, x > 0.85)$ can safely be attributed to the antiferromagnetic longitudinal order-parameter.

It has been observed long ago that the critical field curve $B_c^{\parallel}(T)$ of EuTe starts decreasing for finite temperatures with a T^2 term instead with a $T^{3/2}$ term [16]. These early measurements were, however, only conducted to a lowest temperature of 0.5 K which turned out to be not sufficiently low to evaluate the T^2 coefficient correctly, causing wrong conclusions as regards the origin of the T^2 law.

In contrast to the longitudinal (Heisenberg) spontaneous magnetization of ferromagnets such as GdMg and EuS which exhibits a T^2 law until a temperature as large as $0.8T_C$ the asymptotical T^2 law of $B_c^{\parallel}(T \to 0)$ is observed to hold only until $0.1T_N$ [3]. This we attribute to the fact that the behaviour of $B_c^{\parallel}(T)$ is defined by the paramagnetic average over all fourth-order interactions and this average is one order of magnitude smaller than the individual fourth-order interaction processes which seem to be decisive for the low-temperature behaviour of both order-parameters (note that $B_c^{\parallel}$ is not the order parameter). Additionally, the fact that $B_c^{\parallel}(T = 0,\ x)$ is fairly well described by the combined action of bilinear ($B_{ex}^l(x)$) and biquadratic ($B_{ex}^q(x)$) molecular field constants (see Fig. 9) and that $B_{ex}^q(x)$ conforms well to the high-temperature average of the fourth-order interaction sum given by $\Theta_3(x)$ makes us confident that the x and T dependence of $B_c^{\parallel}(T,\ x)$ should be given by the quantities $\Theta_3(x)$ *viz.* $B_{ex}^q(x)$. In particular, for $x \to x_c = 0.85$

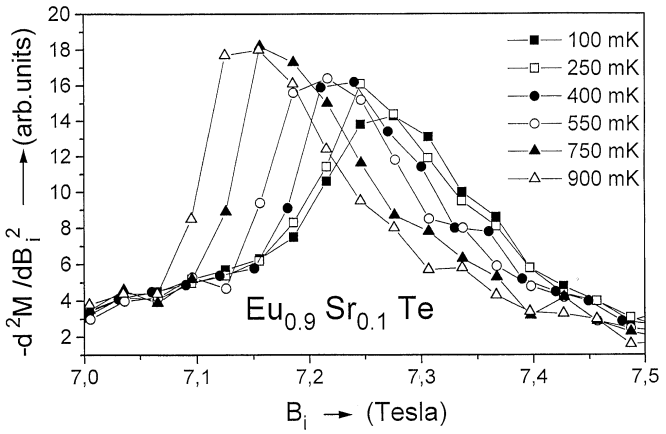

Fig. 5. Second derivatives of magnetization with respect to field which most accurately define the critical field $B_c^{\parallel}$ for the longitudinal (Heisenberg) order-parameter. As $B_c^{\parallel}$ the mean height of the left hand flank has been chosen.

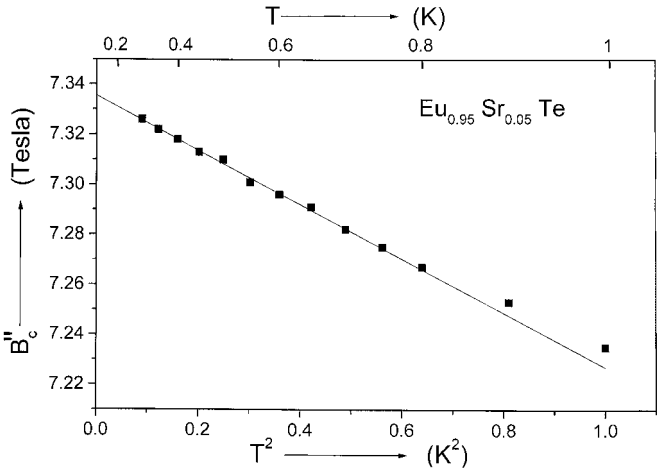

Fig. 6. Critical field $B_c^{\parallel}$ of the longitudinal (bilinear) order-parameter for one $Eu_xSr_{1-x}Te$ sample with $x = 0.95$ *vs.* squared temperature. Below 0.8 K $B_c^{\parallel}$ shows a T^2 law like that observed for the critical fields of EuSe and EuTe [3].

Heisenberg behaviour can be expected for $B_c^{\parallel}(T \to 0)$ since $\Theta_3(0.85) = 0$.

Magnetization measurements have been performed by means of an inductive method using excitation frequencies of 30 Hz. A small concentric transformer was wound with an astatic pair of secondary windings. The needle-shaped, single crystalline samples with typical weights of 20 mg were obtained by cleaving from a larger ingot and placed as a core in one of the two antisymmetric secondary coils, thus producing an unbalance secondary signal which was passed through a 1:100 low noise transformer before being fed into a lock-in amplifier. Low temperatures have been attained with a ^{3}He-^{4}He dilution refrigerator furnished with resistance thermometers which were calibrated against a NBS SRM 768 Temperature Reference Standard and against the susceptibility signal of a Cerium-Magnesium-Nitrate (CMN) probe used to interpolate between the SRM 768 superconducting transitions.

Magnetization curves such as shown in Figure 3 have been obtained by a numerical integration of the ac-susceptibility signal. In those curves the critical field $B_c^{\parallel}$ cannot be localized unambiguously, especially for the diamagnetically diluted samples. Only the second derivative of magnetization with respect to field exhibits a distinctive but smeared discontinuity and allows a sufficiently accurate identification of the phase transition at $B_c^{\parallel}$. This is demonstrated in Figure 5 which gives the second derivative of magnetization as a function of field. In the evaluation of the temperature dependence of $B_c^{\parallel}(T)$ it is very important to choose homologous points in each d^2M/dB_i^2 curve being measured at a different temperature. As $B_c^{\parallel}$ we have chosen the half height of the steep left hand flanks in Figure 5. Since the relative variation of $B_c^{\parallel}(T)$ within the investigated temperature range is only ≈ 0.015 very homogeneous samples are required for these studies.

Figure 6 displays one $B_c^{\parallel}(T)$ curve *vs.* T^2 for one $Eu_xSr_{1-x}Te$ sample with $x = 0.95$. As was observed for EuTe in reference [3], $B_c^{\parallel}(T)$ decreases with a T^2 law in the limit $T \to 0$ also for the $x = 0.95$ sample. This behaviour changes on approaching the critical composition of $x_c = 0.85$.

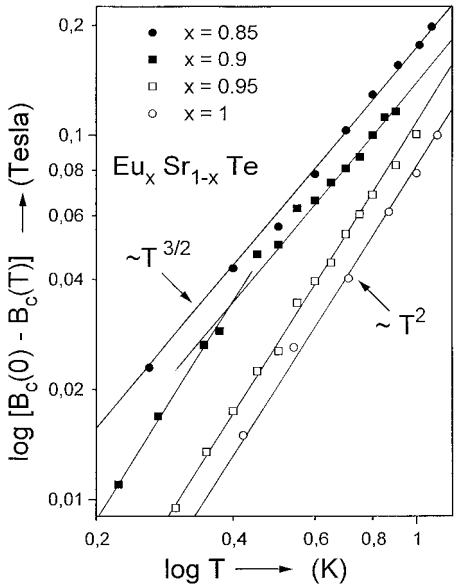

Fig. 7. log-log plot of the variation of $B_c^{\parallel}(T)$ with respect to $B_c^{\parallel}(T = 0)$ as function of temperature revealing a crossover behaviour from a T^2 dependence for $x = 1$ to a $T^{3/2}$ dependence for $x = 0.85$.

In the case of the four investigated samples with $x \geq 0.85$ the classical crossover behaviour seems to hold, as can be seen in Figure 7, a log-log plot is showing the variation of $B_c^{\parallel}(T)$ with respect to the $B_c^{\parallel}(T = 0)$ value as a function of temperature. Note that we make use here of the convention $B_c^{\parallel}(T) = B_c^{\parallel}(0) - b \cdot T^2$. For pure EuTe the T^2 law holds over a temperature range as large as 1.2 K. In the case of the $x = 0.95$ sample the T^2 law is

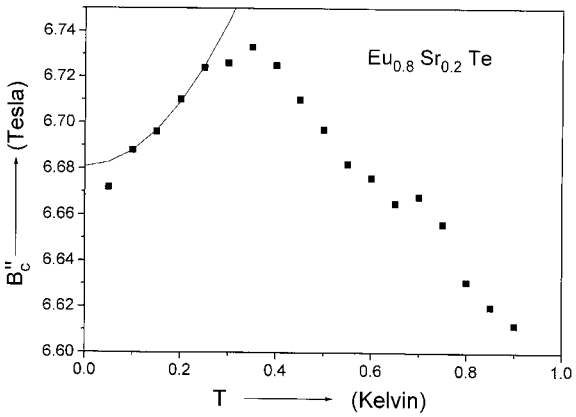

Fig. 8. Critical field curve $B_c^{\parallel}(T)$ of the longitudinal (bilinear) order-parameter for one sample with $x = 0.8$ for which the fourth-order interaction sum is antiferromagnetic. For all samples with $x < 0.85$ $B_c^{\parallel}(T)$ decreases again for $T \to 0$. Solid line is guide to the eye.

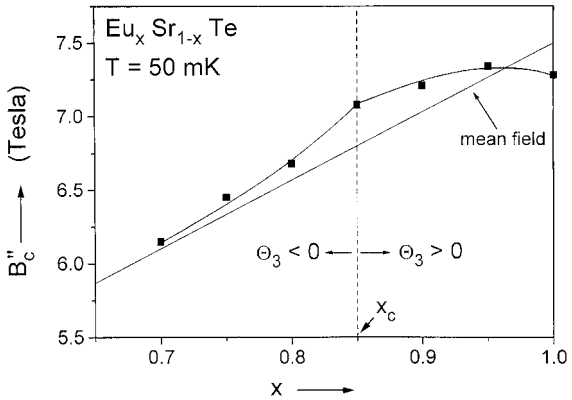

Fig. 9. Critical field values $B_c^{\parallel}(50\text{ mK},\ x)$ taken as the $T \to 0$ limit as function of composition x. The straight solid line gives the calculated mean field behaviour using the biquadratic molecular field values $B_{ex}^{q}(x)$ from Figure [4]. At the critical composition of $x_c = 0.85$, where the transverse order-parameter changes from ferromagnetic ($x > 0.85$) to antiferromagnetic ($x < 0.85$), only a weak kink is noticeable in the longitudinal critical field $B_c^{\parallel}(T = 0,\ x)$.

restricted to $T < 0.85$ K and for the $x = 0.90$ sample to $T < 0.44$ K. This reduction of the absolute validity range of the T^2 law is much greater than that for the Néel temperature $T_N^{\parallel}$ which changes only between 9.9 K for EuTe and 8.8 K for $Eu_{0.9}Sr_{0.1}Te$. As a consequence, the limited T^2 range cannot be correlated with the decreasing bilinear interactions. Moreover, the $x = 0.90$ data in Figure 7 show the typical crossover phenomenon to a high temperature exponent of 3/2 in agreement with classical spin wave theory [17]. This observation makes clear the T^2 law must be attributed to the non-Heisenberg interactions. The range of validity of the T^2 law decreases on approaching $x_c = 0.85$ where the paramagnetic average of the fourth-order interactions tends to zero and is replaced on the high temperature side by the $T^{3/2}$ law. As a consequence, the $B_c^{\parallel}(T)$ data of the $x = 0.85$ sample obey a $T^{3/2}$ law over the whole observed temperature range.

This shows the $B_c^{\parallel}(T \to 0)$ behaviour is dominated by the fourth-order interactions which turn out to be crucial for the spin dynamics. This is very surprising since the absolute value of $B_c^{\parallel}$ is clearly dominated by the conventional bilinear interactions apart from smaller modifications due to fourth-order interactions. Note also that for $T \to 0$ the moments are not aligned perfectly parallel at $B_c^{\parallel}(T = 0)$ [3].

Figure 8 displays the $B_c^{\parallel}(T)$ data for one sample with $x = 0.8$ for which the fourth-order interaction sum is antiferromagnetic. For all samples with $x < 0.85$ it is observed the $B_c^{\parallel}(T \to 0)$ curve decreases again for $T \to 0$ such that the coefficient $b(x)$ of a fitted T^2 law would be negative. In Figure 8 this is indicated by a solid line serving as guide to the eye. It should be noted that such a decreasing behaviour is possible for the critical field but thermodynamically prohibited for an order-parameter which must increase steadily as $T \to 0$.

The behaviour of $B_c^{\parallel}(T = 0,\ x)$ is worth discussing. Figure 9 shows the $B_c^{\parallel}(x)$ data for the lowest temperature of 50 mK which can be taken as the limit $T \to 0$. In molecular field approximation the critical field $B_c^{\parallel}$ is given according to equation (1), by $-B_{ex}^{l}(x) - B_{ex}^{q}(x)$ assuming $m \approx 1$ at $B_c^{\parallel}(T = 0)$. At this point care is required with respect to the signs of both molecular field constants. For EuTe, for instance, B_{ex}^{l} is antiparallel but B_{ex}^{q} is parallel to the external field near $B_c^{\parallel}$, where the system has nearly approached magnetic saturation. At zero field both molecular fields are antiparallel to the external field.

The effective biquadratic molecular field constant $B_{ex}^{q}(x)$ is composed of contributions from biquadratic interactions, which are, like the bilinear interactions, proportional to x, and of contributions from three-spin interactions, which are proportional to x^2 (see Fig. 4). Both interaction types seem to have similar consequences such that they can be comprised in one single molecular field constant. In the here interesting composition range of $0.7 \leq x \leq 1.0$ this quadratic x-dependence can be linearized by $B_{ex}^{q} = 3.33\ x - 2.83$ with B_{ex}^{q} given in tesla (compare Fig. 4) which together with $B_{ex}^{l} = 8x$ results in a critical field of $B_c^{\parallel}(T = 0,\ x) = 4.67x + 2.83$. It is important to note that the slope of the critical field as function of composition is changed from 8 to 4.67 by the fourth-order interactions. Figure 9 compares the experimental $B_c^{\parallel}(T = 0,\ x)$ data with the molecular field calculation (straight line). It can be seen that the molecular field approximation gives a surprisingly good fit to the experimental results. At the critical composition of $x_c = 0.85$, where the fourth-order interaction sum changes from ferromagnetic for $x > 0.85$ to antiferromagnetic for $x < 0.85$ the experimental $B_c^{\parallel}(T = 0,\ x)$ data exhibit an only weak kink. This event is a weak reaction caused by the changing magnetic order of the transverse order-parameter showing

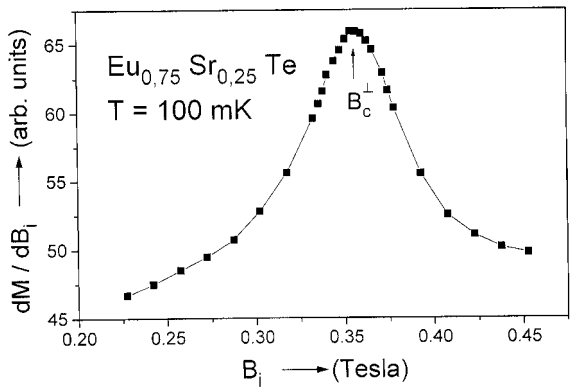

Fig. 10. First derivative of field parallel magnetization (ac susceptibility) as function of field showing the anomaly at the critical field $B_c^{\perp}$ of the transverse order-parameter as observed for the samples with $x < 0.85$, for which the fourth-order interaction sum is antiferromagnetic.

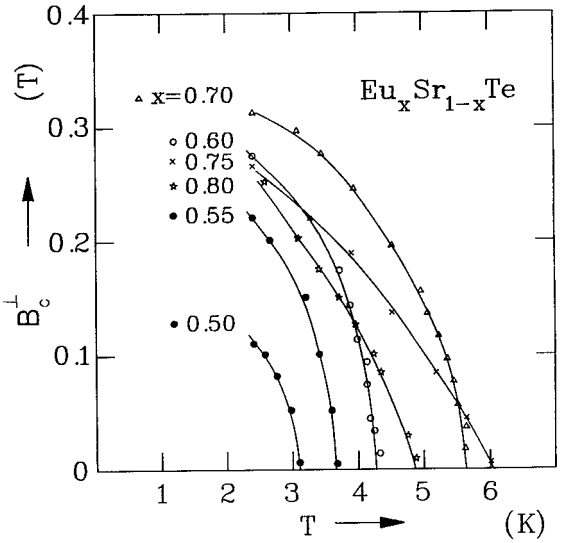

Fig. 11. Critical field curves $B_c^{\perp}(T)$ of the transverse order-parameter for $Eu_xSr_{1-x}Te$ samples with $x < 0.85$, obtained from field-parallel magnetization measurements. For these samples the fourth-order interaction sum, as given by $B_{ex}^q(x)$ in Figure 4, is negative (antiferromagnetic). The $B_c^{\parallel}(T)$ curves of the longitudinal order-parameter occurring at higher temperatures are not shown for reasons of clarity.

that longitudinal and transverse ordering processes interfere very little. The same conclusion can also be drawn from the very weak anomaly displayed at x_c by $T_N^{\parallel}(x)$. $B_c^{\parallel}$ and $T_N^{\parallel}$ are two quantities of the Heisenberg order-parameter being defined mainly by the bilinear interactions but to some extend also by fourth-order interactions (see Figs. 9 and 13). Nevertheless the spin dynamics of the bilinear (longitudinal) order-parameter is clearly dominated by the fourth-order interactions as is demonstrated by the observed low temperature T^2 law.

For the $Eu_xSr_{1-x}Te$ samples we studied the transverse order-parameter is like the longitudinal one of the antiferromagnetic MnO type for $x < 0.85$ ($\Theta_3 < 0$) but the critical field values $B_c^{\perp}$ are considerably smaller than the $B_c^{\parallel}$ values of the longitudinal order-parameter (compare Figs. 9 and 11). This is surprising since the corresponding ordering temperatures $T_N^{\perp}$ and $T_N^{\parallel}$ are very similar (Fig. 13). On the other hand, the observed $B_c^{\perp}(T = 0)$ values conform to the biquadratic molecular field constants B_{ex}^q displayed in Figure 4.

Figure 10 shows the differentiated magnetization anomaly (ac measurement) observed along $B_c^{\perp}$ for one sample with $x = 0.75$. This graph gives the derivative of the magnetization anomaly as is shown in the inset of Figure 3. Performing those longitudinal magnetization measurements for different temperatures, the complete critical field curves $B_c^{\perp}(T)$ can be obtained as are shown in Figure 11. It must be considered as characteristic that the susceptibility anomalies along $B_c^{\perp}(T)$ disappear on approaching $T_N^{\perp}$ (*i.e.* for $B_o \to 0$) and that no anomaly at all is observed in longitudinal zero field susceptibility measurements at $T_N^{\perp}$ as one might expect for a transverse ordering process. Interestingly, also zero-field magnetic specific heat measurements give no indication of $T_N^{\perp}$ [18]. Therefore, $T_N^{\perp}$ must be evaluated by the extrapolation $B_c^{\perp}(T,\ B_o \to 0)$ if longitudinal magnetization measurements are performed.

By analogy, the transverse order-parameter can be expected to be ferromagnetic for $x > 0.85$ where $\Theta_3(x) > 0$ holds. Also for $x > 0.85$ zero-field specific heat measurements reveal no anomaly in addition to the well-known Néel temperature $T_N^{\parallel}$. The only irregularity observed is that the critical magnetic behaviour of the magnetic specific heat of EuTe does not conform to the Heisenberg model prediction at $T_N^{\parallel}$ [18]. This might be a consequence of the fact that $T_N^{\parallel}$ and $T_C^{\perp}$ are nearly equal (see Figs. 12, 13).

Whenever there is one ferromagnetic and one antiferromagnetic order-parameter the ferromagnetic one can preferentially be aligned by the application of moderate magnetic fields and can be distinguished using longitudinal (field parallel) magnetization measurements. This orientational process will only be limited by the antiferromagnetic domains.

Figure 12 compiles a number of ac susceptibility measurements for EuTe for the transverse susceptibility $\chi_{\perp}$ (upper panel) and the longitudinal (field parallel) susceptibility $\chi_{\parallel}$ (lower panel) for different values of the static longitudinal magnetic field $B_o^{\parallel}$. From the unusually strong field dependence of both susceptibilities for temperatures just below the longitudinal Néel temperature $T_N^{\parallel}$ it can be concluded that the Curie temperature $T_C^{\perp}$ of the transverse order-parameter must be very close to $T_N^{\parallel}$. Whether $T_C^{\perp}$ and $T_N^{\parallel}$ really are equal cannot be decided unambiguously in view of the limited temperature resolution of these measurements.

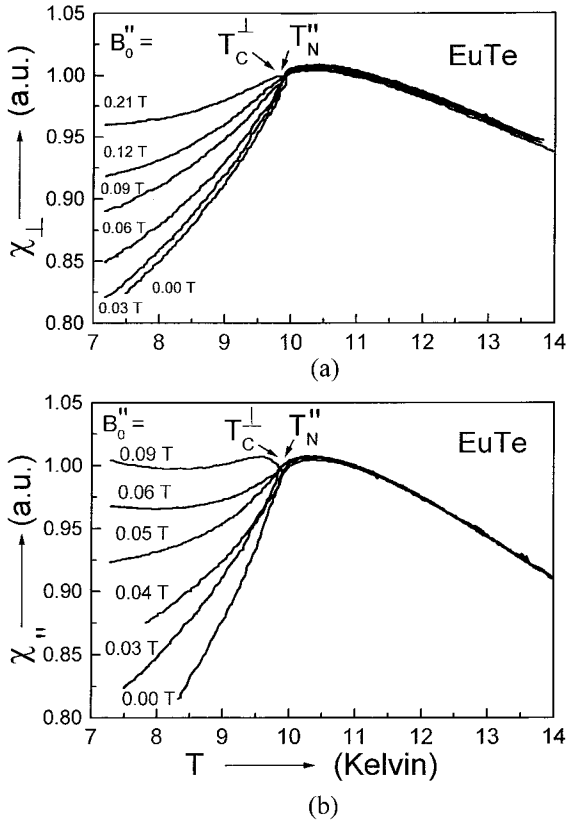

Fig. 12. Transverse ac susceptibility $\chi_\perp$ (upper panel) and longitudinal (field parallel) ac susceptibility $\chi_\parallel$ (lower panel) for EuTe for different values of the static longitudinal field $B_o^\parallel$. The strong field dependence just below $T_N^\parallel$ shows that $T_C^\perp$ and $T_N^\parallel$ must be very close to each other. Due to the magnetic domains and the associated distribution of moment orientations no ideal ferromagnetic behaviour in $\chi_\parallel$ and no ideal antiferromagnetic behaviour in $\chi_\perp$ results. Note that the strong field dependence below $T_N^\parallel$ is absent for the samples with $x < 0.85$.

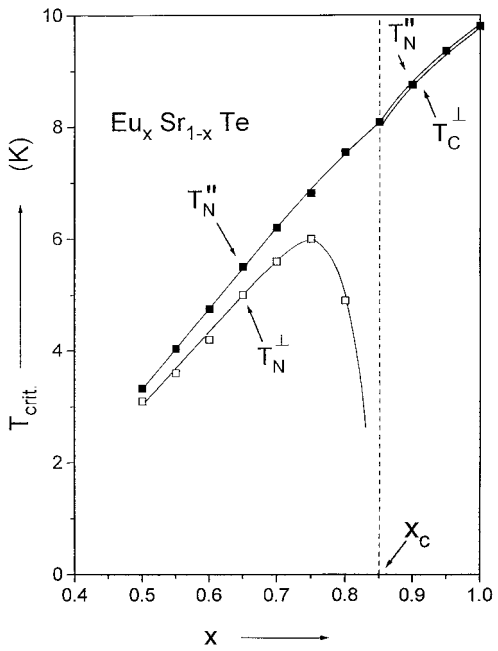

Fig. 13. Zero-field phase diagram of $Eu_xSr_{1-x}Te$ showing the critical temperatures of both order-parameters. $T_N^\parallel$ is the Néel temperature of the longitudinal (Heisenberg) order-parameter. $T_C^\perp$ is the Curie temperature of the transverse (non-Heisenberg) order-parameter in the range of a ferromagnetic fourth-order interaction sum ($x > 0.85$) while $T_N^\perp$ is the Néel temperature of the transverse order-parameter in the range $x < 0.85$ where the fourth-order interaction sum is antiferromagnetic. The $T_N^\perp$ values have been obtained by extrapolating the $B_c^\perp$ curves of Figure 11 to $B_o \to 0$.

The main difference between the two susceptibilities is a much stronger field dependence of $\chi_\parallel$ compared to $\chi_\perp$ for $T < T_N^\parallel$. This behaviour indicates a new type of anisotropy associated with the perpendicular configuration of two order-parameters caused by a gradual rotation of the ferromagnetic component into the field direction. Similar observations were made also on GdMg using neutron scattering methods [4]. Note that a field dependent ac susceptibility means a non-linear magnetization curve. It should furthermore be noted that the field parallel susceptibility $\chi_\parallel$ is isotropic, *i.e.*, does not depend on the crystallographic direction [18]. The field induced difference between $\chi_\parallel$ and $\chi_\perp$ is therefore also independent of the crystallographic orientation.

A stronger field dependence of $\chi_\parallel$ conforms to our expectation that the ferromagnetic transverse magnetization will gradually turn into the field direction with increasing field and will thereby increase the $\chi_\parallel$ values accordingly. This process is limited to field values below 0.075 tesla. For fields in excess of 0.075 tesla the magnetization is a much slower function of field (compare with Fig. 3) and the $\chi_\parallel(T)$ curves do increase only gradually with field. This change of behaviour is evidently caused by the magnetic domains which can no longer rotate for fields > 0.075 T [19]. It is particularly interesting to observe that for this marginal field value $\chi_\parallel(T)$ increases immediately below $T_N^\parallel$ for decreasing temperatures, a behaviour clearly showing the ferromagnetic character of the non-Heisenberg order parameter.

In contrast to $\chi_\parallel$ the $\chi_\perp$ increases much slower with field; even at the limiting field $B_o = 0.21$ tesla, above which $\chi_\perp$ is only weakly field dependent, all $\chi_\perp(T)$ curves decrease clearly with decreasing temperature below $T_N^\parallel$. The $\chi_\perp(T)$ curves conform therefore much better to the anticipated behaviour of an antiferromagnetic order-parameter. Of course, $\chi_\parallel$ does not show ideal ferromagnetic behaviour and $\chi_\perp$ also fails to show ideal antiferromagnetic behaviour. We attribute this also to the magnetic domains [19] which prevent an ideal orientation of both order-parameters and mix their properties. It is very important to note that the strong field dependence of the susceptibilities shown in Figure 12 is absent for the samples with $x < 0.85$ for which the transverse order-parameter is antiferromagnetic ($\Theta_3 < 0$). One further detail which is worth mentioning is the fact that in

the zero field measurements $\chi(T_N)/\chi(0) = 3/2$ is found as expected for a perfect antiferromagnet. The transverse ferromagnetic order-parameter does therefore not affect the longitudinal zero field susceptibility measurements. This must also be attributed to the existence of an isotropic distribution of antiferromagnetic domains. To orient the spontaneous magnetization some "demagnetization" field must be applied.

We should admit that the postulated ferromagnetic order of the transverse order-parameter cannot be of the conventional type since in neutron diffraction measurements on EuTe powder samples no definite ferromagnetic Bragg intensities can be observed (compare Fig. 12 of Ref. [15]); however, the MnO superstructure reflection intensities which are a signature of both antiferromagnetic order-parameters for $x < 0.85$ suddenly decrease at x_c where the transverse order-parameter changes from antiferromagnetic for $x < 0.85$ to ferromagnetic for $x > 0.85$ [13]. This is at least an indirect indication for the phase change of the transverse moment components.

In Figure 13 we show the composition dependence of the transition temperatures of both order-parameters. Note that $T_N^{\parallel}(x)$ shows only a very weak change in slope at $x_c = 0.85$. It is particularly interesting that the ordering temperatures of the transverse (non-Heisenberg) order-parameter appear to be attracted by the Néel line $T_N^{\parallel}(x)$ of the longitudinal (Heisenberg) order-parameter except for $x_c = 0.85$. The fact that the transverse order-parameter changes from ferromagnetic for $x > 0.85$ to antiferromagnetic for $x < 0.85$ in accordance with the sign change of $\Theta_3(x)$, a quantity which gives the interaction strength in the disordered state, shows clearly that these ordering phenomena are of the order-disorder type and are caused exclusively by the fourth-order interactions. The very small interference of both ordering processes supports the postulated orthogonality of the associated order parameters.

4 Conclusions

In the diamagnetically diluted antiferromagnetic composition series $Eu_xSr_{1-x}Te$ fourth-order (non-Heisenberg) exchange interactions are now quantitatively known [3,6,12]. This information comes from molecular field analyses of the curvature of the magnetic isotherms as function of the applied magnetic field either in the limit $T \ll T_N$ or $T \gg T_N$. For $T \ll T_N$ a biquadratic molecular field constant can be inferred from the curvature of the magnetization curves. B_{ex}^q gives the sum of all fourth-order exchange interactions in terms of a molecular field. For $T \gg T_N$ the cubic susceptibility χ_3 describes the curvature of the paramagnetic isotherms and from the observed Curie-Weiss law of χ_3 a Curie-Weiss temperature Θ_3 is obtained which represents again an averaged measure for the strength of the fourth-order exchange interactions. Both quantities are related by the cubic Curie constant C_3 and agree surprisingly well [3]. This is a non-trivial result. Evidently Θ_3 gives the high-temperature average of all fourth-order interactions. Also in the evaluation of B_{ex}^q from low-temperature magnetization curves in which the spin configuration changes from antiferromagnetic for $B_o = 0$ to nearly ferromagnetic for $B_o = B_c^{\parallel}$ an averaging process over all fourth-order interactions is performed.

Biquadratic and three-spin interactions can be distinguished by investigating the composition dependence either of $B_{ex}^q(x)$ or $\Theta_3(x)$. Both quantities contain a negative term proportional to x due to antiferromagnetic biquadratic interactions and a positive term proportional to x^2 due to ferromagnetic three-spin interactions (see Appendix of following article) [6]. Both individual interaction processes are one order of magnitude larger than the average value over all these interactions observed in the compact material EuTe.

In reference [7] it was argued that the class of fourth-order interactions governs the magnetic ordering processes of the transverse moment components which order independently of the longitudinal moment components. The ordering types of both order-parameters are given essentially by the signs of the Curie-Weiss temperatures Θ_1 and Θ_3 of the linear susceptibility χ_1 and the cubic susceptibility χ_3, respectively. For instance, in the composition series $Eu_xSr_{1-x}Te$ Θ_1 is negative for all compositions x but Θ_3 is negative only for $x < 0.85$ and therefore both order-parameters are antiferromagnetic for $x < 0.85$. The transverse (non-Heisenberg) order-parameter is distinguished by considerably smaller critical field values $B_c^{\perp}$ but only slightly smaller Néel temperatures $T_N^{\perp}$.

As a consequence, it is very important to discriminate between both order-parameters by choosing suitable geometrical conditions in the magnetization measurements. In zero-field NMR measurements a mixture of both order-parameters is observed since these measurements are performed for a state with an isotropic distribution of magnetic domains. For EuS, for which both order-parameters are ferromagnetic ($\Theta_1 > 0$, $\Theta_3 > 0$) [3,8], the NMR measurements revealed a T^2 law for the variance of the spontaneous magnetization with respect to the $T = 0$ value. The T^2 law must, as a consequence, be common to both order-parameters. This is confirmed with magnetization measurements on GdMg, a material with a ferromagnetic bilinear order-parameter and an antiferromagnetic transverse order-parameter. In such a system the antiferromagnetic transverse order-parameter does not contribute much to the longitudinal (field-parallel) magnetization measurements and therefore the bilinear (longitudinal) order-parameter can be measured individually. These measurements confirm the T^2 law for the Heisenberg order-parameter up to temperatures as large as $0.8T_C$ [4,7]. The spin dynamics of both order parameters is therefore dominated by the fourth-order interactions.

Since there is at present no theory available which would explain the T^2 law this work aimed to verify experimentally that the T^2 law originates in the fourth-order interactions. The only way of doing is the investigation of the low-temperature behaviour of the critical

field $B_c^{\parallel}(T \to 0,\ x)$ in the diamagnetically diluted antiferromagnetic compounds $Eu_xSr_{1-x}Te$. The behaviour of the critical field $B_c^{\parallel}$, being the phase boundary to the paramagnetic phase, can be assumed to be defined by the paramagnetic average of all interactions since for $T \to 0$ the spins are nearly ferromagnetically saturated at $B_c^{\parallel}$. At $x_c = 0.85$ the paramagnetic average over the fourth-order interactions passes zero because antiferromagnetic biquadratic interactions and ferromagnetic three-spin interactions compensate at x_c. Under this condition only the bilinear Heisenberg interactions remain active. Consistent with this, a crossover from a T^2 law to a $T^{3/2}$ could be observed for $B_c^{\parallel}(T \to 0,\ x)$ on approaching $x_c = 0.85$. The Bloch $T^{3/2}$ law is hence an exception and the T^2 law the rule.

The combination antiferromagnetic-antiferromagnetic of order-parameters ($x < 0.85$) is particularly clear since both order-parameters can clearly be distinguished on account of their different critical fields, but for $x > 0.85$, where the longitudinal order-parameter is still antiferromagnetic but the transverse order-parameter is ferromagnetic, no direct indication of long range ferromagnetic order is given by the available powder neutron diffraction spectra [12]. Some ideas to unravel this puzzle are outlined in reference [13] (following article).

We gratefully acknowledge fruitful discussions with E. Müller-Hartmann, University of Cologne. The technical assistance of Mrs. Ch. Horriar-Esser and Mr. B. Olefs are also very much appreciated.

References

1. F. Bloch, Z. Phys. **61**, 206 (1930).
2. A.C. Gossard, V. Jaccarino, J.P. Remeika, Phys. Rev. Lett. **7**, 122 (1961).
3. U. Köbler, R.M. Mueller, L. Smardz, D. Maier, K. Fischer, B. Olefs, W. Zinn, Z. Phys. B **100**, 497 (1996).
4. P. Morin, J. Pierre, D. Schmitt, D. Givord, Phys. Lett. A **65**, 156 (1978).
5. M. Neusser, H. Lütgemeier, W. Zinn, J. Magn. Magn. Mat. **4**, 42 (1977).
6. E. Müller-Hartmann, U. Köbler, L. Smardz, J. Magn. Magn. Mat. **173**, 133 (1997).
7. U. Köbler, R.M. Mueller, W. Schnelle, K. Fischer, J. Magn. Magn. Mat. **188**, 333 (1998).
8. U. Köbler, R.M. Mueller, Th. Lorenz, J.P. Brown, K. Fischer, J. Magn. Magn. Mat. (to appear).
9. V.M. Matveev, E.L. Nagaev, Sov. Phys. Sol. State **14**, 408 (1972).
10. R.G. Munro, M.D. Giradeau, J. Magn. Magn. Mat. **2**, 319 (1976).
11. J. Adler, J. Oitmaa, J. Phys. C **12**, 575 (1979).
12. U. Köbler, I. Apfelstedt, K. Fischer, W. Zinn, E. Scheer, J. Wosnitza, H. von Löhneysen, Th. Brückel, Z. Phys. B **92**, 475 (1993).
13. U. Köbler, A. Hoser, H.A. Graf, M.-T. Fernandez-Diaz, K. Fischer, Th. Brückel, Eur. Phys. J. B **8**, 217 (1999).
14. I.S. Jacobs, S.D. Silverstein, Phys. Rev. Lett. **13**, 272 (1964).
15. R.M. Nowotny, K. Binder, Z. Phys. B **77**, 287 (1989).
16. W. Figueiredo, S.R. Salinas, Phys. Lett. A **83**, 231 (1981).
17. K. Muramatsu, J. Phys. Soc. Jpn **50**, 3207 (1981).
18. E. Scheer, J. Wosnitza, H. von Löhneysen, Z. Phys. B **85**, 79 (1991).
19. J.W. Battles, G.E. Everett, Phys. Rev. B **1**, 3021 (1970).