Magnetic Frustration and Spin–Peierls Transition in $CuGeO_3$

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Summary: In $CuGeO_3$ an unexpected correlation between the pressure dependencies of the spin–Peierls transition temperature and the magnetic susceptibility is observed. It will be shown that this correlation may be drawn back to a pressure–dependent magnetic frustration along the spin chains. In addition, the magnetic–field temperature phase diagram and the field dependence of the structural order parameter are discussed. The spatial modulation of the order parameter in the incommensurate phase continuously changes its character from a soliton–lattice like close to the dimerized incommensurate transition to a sinusoidal one at higher fields.

1 Introduction

The spin–Peierls (SP) transition may occur in one–dimensional (1d) antiferromagnetic (AF) spin chains of half–integer spin with a finite magnetoelastic coupling, i.e. a dependence of the exchange constant J on lattice distortions. Such chains are unstable against dimerization, since a 1d AF spin chain with alternating nearest neighbor (NN) exchanges $[J_{1,2} = J(1 \pm \delta)]$ has a lower groundstate energy than a chain with uniform NN exchange J. With decreasing temperature SP systems undergo a continuous structural transition at T_{SP} from a uniform (U) phase with a gapless spin excitation spectrum to a dimerized (D) phase with a collective non–magnetic singlet groundstate separated by a gap from the excited triplet states. The alternation of J is achieved by a dimerization of the lattice, e.g. alternating NN distances $[a_{1,2} = a(1 \pm A)]$, because the gain in magnetic energy ($\propto \delta^{4/3}$) over–compensates the loss in elastic energy ($\propto \delta^2$).

The SP transition has been widely studied during the 1970's [1] and a well developed theoretical description has been obtained by Cross and Fisher (CF) yielding, e.g., $T_{\mathrm{SP}} \simeq \lambda^2/\omega_0^2$ (λ is the spin phonon coupling constant and ω_0 the bare frequency of a phonon that softens at T_{SP}) [2]. Moreover, a characteristic magnetic–field temperature phase diagram has been calculated. With increasing field T_{SP} continuously decreases and above a critical field a so–called I phase is present at low temperatures. The latter is magnetic and its lattice distortion is spatially modulated ($A = A(n)$; n is the site index along the chains) with a periodicity that is field dependent and, in general, incommensurate (I) with respect to that of the underlying lattice. Different models have been proposed for the character of this modulation ranging from a purely sinusoidal one to a soliton lattice, i.e. a regular array of domain walls. Depending on the model the field–induced transition from the D to the I phase is expected to be of first or second order [2, 3].

The experimental investigations of the SP transition were restricted to a few organic compounds until 1993. Then Hase *et al.* observed this transition in the inorganic compound $CuGeO_3$ [4]. Due to the possibility of growing large high–quality single crystals, $CuGeO_3$ allows for a broad variety of experimental investigations, which were often difficult to perform on the organic compounds. The crystal structure is orthorhombic and the AF spin chains are oriented along the c direction. The latter are formed by Cu^{2+} ions ($S = 1/2$) coupled via a $\simeq 99°$ Cu–O–Cu superexchange. Although many experimental observations in $CuGeO_3$ at least qualitatively agree to the CF theory, there are also clear discrepancies between theory and experiment [5]. For example, the phonon softening expected within the CF theory is not observed [6]. In addition, the model of 1d AF spin chains with NN exchange is not sufficient to describe the magnetism in $CuGeO_3$. On the one hand, an AF coupling $J_b \simeq 0.1J$ along the b direction is observed, i.e. the 1d character of the magnetism ($J \gg J_\perp$) is not very well fulfilled [7, 8]. On the other hand, there is evidence for an AF coupling $J' \simeq 0.35J$ between next nearest neighbors (NNN) along the chains, i.e. for a strong magnetic frustration arising from the competition between the NN and NNN exchange [9, 10].

The influence of the magnetic frustration on the spin–Peierls transition in $CuGeO_3$ will be discussed in more detail in the following sections. Section 2 deals with the (uniaxial) pressure dependencies of T_{SP} and the magnetic susceptibility χ, whereas in Section 3 the magnetic–field dependencies of T_{SP} and of the structural order parameter are considered. It is mentioned that different aspects of this paper have been the subject of previous separate publications [10, 11, 12, 13, 14]. Therefore, only representative experimental data will be shown. For further information concerning, e.g., the experimental techniques or details of the data analysis, it is referred to the original work.

2 SP Transition and Pressure Dependencies

The SP transition in $CuGeO_3$ has been discovered by a drop–like decrease of the magnetic susceptibility χ_i below $T_{SP} \simeq 14.3$ K [4]. This decrease is observed for magnetic fields applied along all three lattice directions ($i = a, b, c$) and is a consequence of the gap Δ in the magnetic excitation spectrum due to the magnetic dimerization. Both inelastic neutron scattering data [8, 7] as well as the low–temperature behavior of χ_i or the specific heat C_p [4, 13] yield $\Delta \simeq 23$ K. The structural dimerization leads to a doubling of the unit cell for $T < T_{SP}$, which

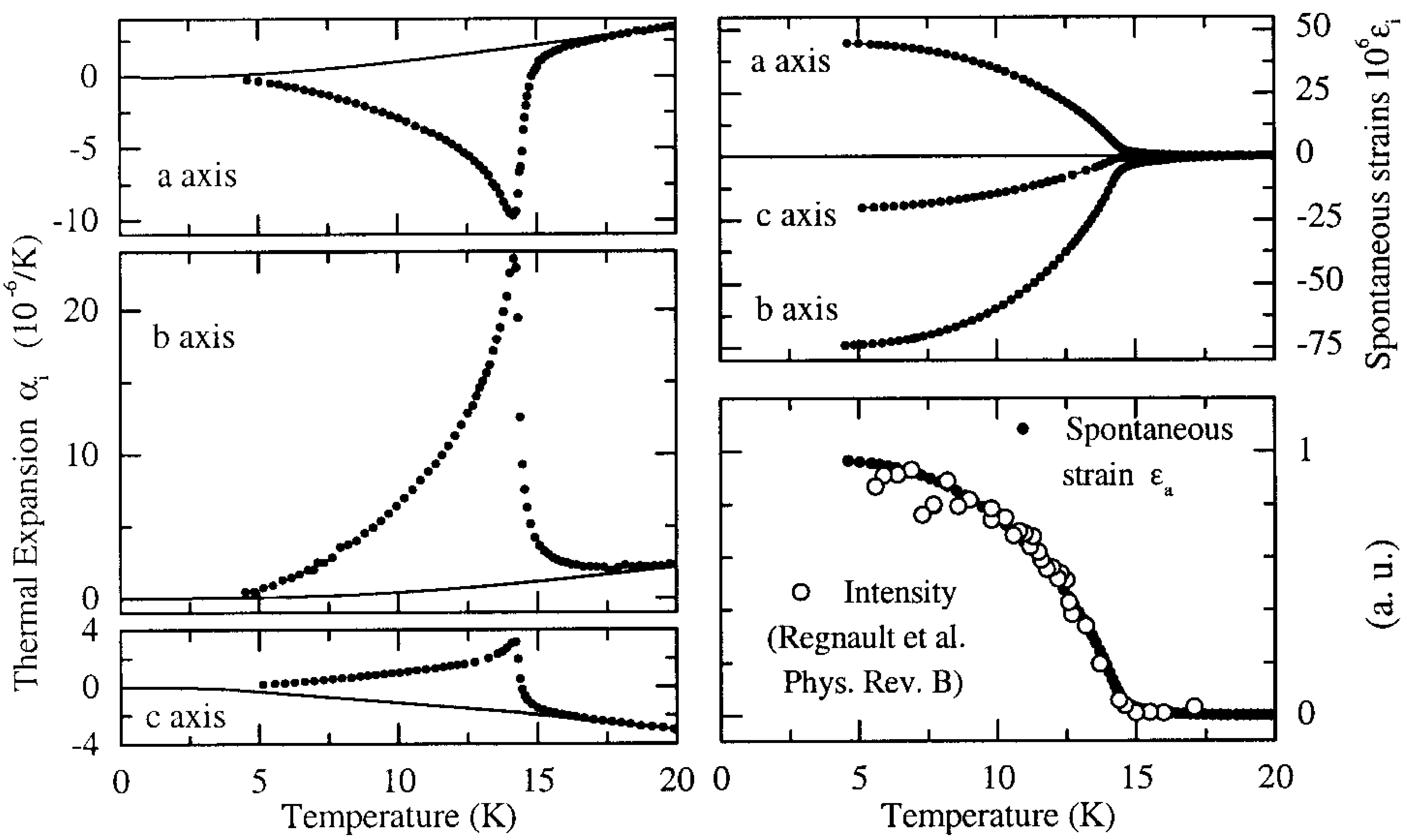

Figure 1 Left: Thermal expansion along the a, b, and c axis of $CuGeO_3$. The solid lines are background extrapolations. Upper right panel: Spontaneous strains ϵ_i along the different axes. Lower right panel: Comparison between ϵ_a and the intensity of a $(1/2, 3, 1/2)$ superstructure reflection [8] (see text).

is observed by superstructure reflections in diffraction experiments [8, 15]. An example is displayed in the lower right panel of Fig. 1. The dimerization is, however, not the only structural change at the SP transition in $CuGeO_3$. In addition, there are large anomalies of the thermal expansion coefficients $\alpha_i = \frac{1}{L_i}\frac{\partial L_i}{\partial T}$ (left panel of Fig. 1). That means, the temperature dependencies of all three lattice constants strongly change at T_{SP} or, in other words, the SP transition is accompanied by the development of spontaneous strains ϵ_i. The latter are shown in the upper right panel of Fig. 1. They are obtained from the α_i by subtracting smooth background extrapolations $\alpha_{i,extr.}$ (solid lines in Fig. 1) to $\alpha_i(T{=}0) = 0$ and integration, i.e. $\epsilon_i = \int(\alpha_i - \alpha_{i,extr.})dT$.

The anomaly of α_a is of the opposite sign and about half (double) as large as that of α_b (α_c). Thus, the respective ϵ_i differ in signs and sizes, but the temperature dependencies of all three $|\epsilon_i|$ are very similar and show the typical behavior of an order parameter. They strongly increase below T_{SP} and saturate with further decreasing T. The SP order parameter is the distortion amplitude A and a Landau–expansion of the free energy yields $\epsilon_i \propto A^2$, when a usual linear–quadratic strain order-parameter coupling is assumed [13]. As shown in Fig. 1 this proportionality is experimentally confirmed by comparing the temperature dependence of ϵ_a to the intensity of a superstructure reflection, which directly

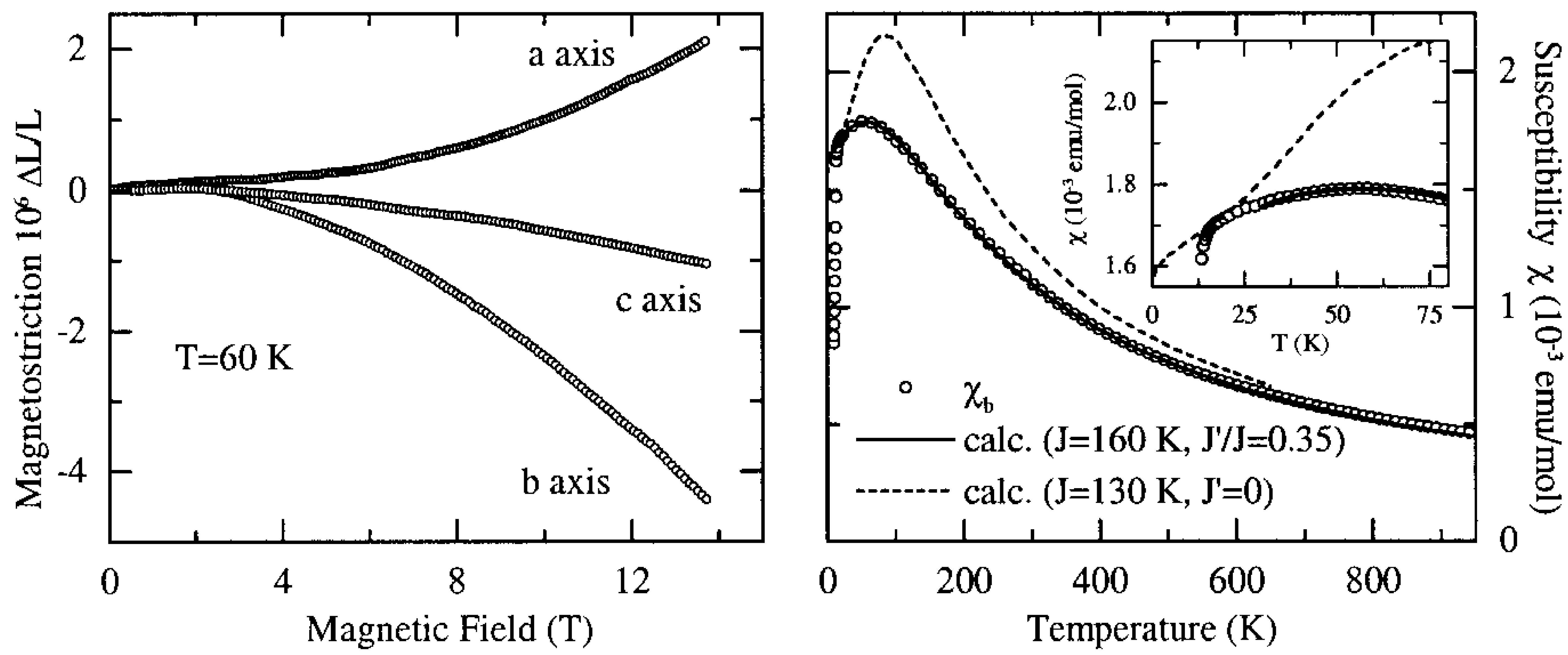

Figure 2 Left: Magnetostriction along the a, b, and c axis. Right: Magnetic susceptibility measured in $H = 1$ T ($||b$). The dashed and dotted lines are calculated susceptibilities for different exchange constants [10]. The inset shows an expanded view up to 80 K (see text).

measures A^2. The 'deeper' reason for the ϵ_i are uniaxial pressure dependencies of $T_{\rm SP}$, which are related to the anomalies of the α_i via the Ehrenfest relations

$$\left.\frac{\partial T_{\rm SP}}{\partial p_i}\right|_{p_i\to 0} = V_m T_{\rm SP}\frac{\Delta\alpha_i}{\Delta C_p}\ . \tag{2.1}$$

V_m denotes the volume per mole and $\Delta\alpha_i$ (ΔC_p) the anomaly size of α_i (C_p) at $T_{\rm SP}$. Equ. (2.1) yields $\frac{\partial T_{\rm SP}}{\partial p_i} \simeq -3.7$, 7.2, and 1.6 K/GPa for pressure along the a, b, and c axis, respectively. The hydrostatic pressure dependence is $\frac{\partial T_{\rm SP}}{\partial p_{hydr.}} = \sum \frac{\partial T_{\rm SP}}{\partial p_i} \simeq 5$ K/GPa in agreement with the results of direct measurements [16].

The $\partial T_{\rm SP}/\partial p_i$ are large and strongly anisotropic. The same anisotropy is found for the magnetostriction in the U phase of $CuGeO_3$ (see Fig. 2). All three crystal axes are significantly magnetic–field dependent. The a axis increases whereas the b and c axes decrease with increasing field and again the change of the a axes is of the opposite sign and about half (twice) as large as that of the b (c) axes. The magnetostrictions $\frac{\Delta L_i}{L_i}$ of the paramagnetic U phase are related to the uniaxial pressure dependencies of the χ_i via the Maxwell relations

$$\frac{1}{H}\left.\frac{\partial \Delta L_i/L_i}{\partial H}\right|_{T,p_i} = -\left.\frac{\partial\chi_i}{\partial p_i}\right|_{T,H} \qquad \text{(with } H||L_i) \tag{2.2}$$

and one obtains $\partial\chi_i/\partial p_i \simeq -0.9$, 1.8, and $0.4\cdot 10^{-3}$ emu/moleGPa for $p_i||a$, b, and c, respectively. Apparently, the $\partial\chi_i/\partial p_i$ and $\partial T_{\rm SP}/\partial p_i$ have the same anisotropy indicating that they are determined by a common parameter.

For 1d AF spin chains with NN exchange the maximum χ_i^{max} of χ_i is proportional to $1/J$, i.e. $\partial \ln J/\partial p_i = -\partial \ln \chi_i^{max}/\partial p_i$. Thus, from the magnetostriction at 60 K, which is close to the χ_i^{max} of $CuGeO_3$ (see Fig. 2), the pressure dependencies $\partial \ln J/\partial p_i \simeq +5$, -10, and -2.5 %/GPa for $p_i \| a$, b, and c, respectively, are derived. A pressure–dependent J is not surprising, since a pre–condition of the SP transition is a finite magnetoelastic coupling. However, the striking correlation between the anisotropy of the $\frac{\partial \chi_i^{max}}{\partial p_i}$ and the $\frac{\partial T_{SP}}{\partial p_i}$ is not expected at all: on the one hand χ_i^{max} depends *only* on J, whereas on the other hand it follows from the CF theory that T_{SP} is *independent* on J (see above).

As mentioned in the introduction, the model of NN exchange is not sufficient to describe the magnetism of $CuGeO_3$. The agreement between the calculated χ and the experimental data is poor, e.g., for $J = 130$ K the absolute value χ^{max} as well as its position T_{max} are significantly too large (see Fig. 2). A much better description of the experimental data is obtained by including a magnetic frustration [9, 10]. As shown by the solid line in Fig. 2 the calculation for $J = 160$ K and $J'/J = 0.35$ perfectly agrees with the experimental χ. The determination of J and J' is discussed in Ref. [10]. In addition, it is shown there that the magnetic entropy calculated for these parameters is consistent with an estimation from the measured C_p. Usually, the influence of J' is studied by exact diagonalization techniques of finite spin chains and due to finite–size effects a comparison with experimental data is, in general, restricted to $T \gtrsim 30$ K. Quite recently, however, a variant of the density matrix renormalization group (DMRG) technique has been used to study frustrated (non–dimerized as well as dimerized) spin chains to lower T. This technique even allows the theoretical investigation of the SP transition in frustrated spin chains and it has been found that both χ and the magnetic entropy calculated for $J = 160$ K and $J'/J = 0.35$ well agree to the respective experimental data for $T < 30$ K, too [17].

Coming back to the pressure dependencies of χ^{max} and T_{SP}. Although χ depends on both J and J', the relation $\chi^{max} \propto 1/J$ is still approximately valid and the magnetostriction is determined by the $\partial \ln J/\partial p_i$. Let us assume for a moment the pressure dependencies of J' to be small (see below). Then the frustration J'/J essentially increases (decreases) to the same extent as χ^{max} raising the question whether T_{SP} also depends on J'/J. At present, calculations about the frustration dependence of T_{SP} do not exist. It is, however, known that for $J'/J \gtrsim 0.24$ there is already a finite gap Δ in the magnetic excitation spectrum even without (lattice) dimerization, i.e. for $\delta = 0$ [18]. The dependence of Δ on both δ and J'/J has been calculated numerically [19]. For $J = 160$ K and $J'/J = 0.35$ these calculations yield $\delta \simeq 0.01$ in order to obtain the experimental $\Delta = 23$ K. Using Fig. 16 of Ref. [19], one obtains that for $J'/J = 0.4$ the same $J\delta$ (corresponding to the same structural distortion) leads to $\Delta \simeq 29$ K. That means, a 14% increase of the frustration causes a 26% increase of Δ. Since a comparable increase of T_{SP} and Δ is reasonable, a pressure–dependent frustration ratio gives a possible explanation for the correlation between the $\partial \chi/\partial p_i$ and the $\partial T_{SP}/\partial p_i$ of $CuGeO_3$. Please note, that in the above example the relative change of Δ is about twice as large than that of J'/J, whereas $\frac{\partial \ln T_{SP}}{\partial p_i} \Big/ \frac{\partial \ln \chi}{\partial p_i} \simeq 4.5$ is obtained experimentally. There are various reasons for this quantitative deviation. Both $J\delta$ as well as Δ/T_{SP} may depend on J'/J and, in addition, the change of J'/J could be stronger than that of χ^{max} due to an additional pressure dependence of J'. It is, however, clear that depending on future results all three points are also possible sources which may contradict the above conclusion.

3 $CuGeO_3$ in Magnetic Fields

The left panel of Fig. 3 shows measurements of α_a, which has been studied up to 28 T. The field dependence of T_{SP} is easily seen from the systematic shift of the anomalies. Moreover, one recognizes that for $H < 12.5$ T the anomaly sizes remain nearly unchanged, whereas for $H > 12.5$ T a significant reduction takes place. This difference is due to the change from D/U in the low–field to I/U transitions in the high–field range. The additional anomalies around 5 K for $H = 12.5$ T arise from the D/I boundary, which is close to this field and has only a weak temperature dependence. The D/I phase boundary is, thus, easier to derive from magnetostriction measurements. As shown by the measurements at 2.2 and 7 K in the upper right panel of Fig. 3 the D/I transition causes an sharp decrease of $\Delta L_a(H)/L_a$. The almost jump–like length changes indicate a first order character of the D/I transition, that is confirmed by different $H_{D/I}$'s derived from measurements with increasing and decreasing field, respectively. The hysteresis amounts to $\simeq 0.2$ T at 2.2 K, continuously decreases with increasing T and vanishes for $T \simeq 11$ K [12]. Above 11 K the field–driven transitions are from the D to the U phase. These transitions are of second order and $\Delta L_a(H)/L_a$ now continuously changes with H as shown by the measurement at 12.5 K. Above $H_{D/U}$ a quadratic increase of $\Delta L_a(H)/L_a$ with H is observed, which is the usual behavior in the U phase as can be seen from the measurement at 20 K. Interestingly, the high–field magnetostriction at 7 K, i.e. in the I phase, also shows this quadratic increase with H. This similarity arises from a saturation of the order parameter as will be discussed below.

The phase diagram of $CuGeO_3$ is plotted in the lower right panel of Fig. 3 together with the calculation of Cross (solid line). The theoretical D/U boundary systematically deviates from the experimental data as shown by the dotted line, which is the solid one after division by 1.12. The need for such a rescaling has been mentioned already by Cross himself: the calculation uses $\chi(T=0)$ of a uniform spin chain instead of $\chi(T=T_{SP})$ leading to an underestimation of the field influence by a factor $\chi(T_{SP})/\chi(0)$ [2]. The rescaling factor can be obtained by comparing the experimental $\chi(T_{SP})$ to the theoretical $\chi(0)$. However, this comparison cannot consider the frustration determined above, since $\chi(0) = 0$

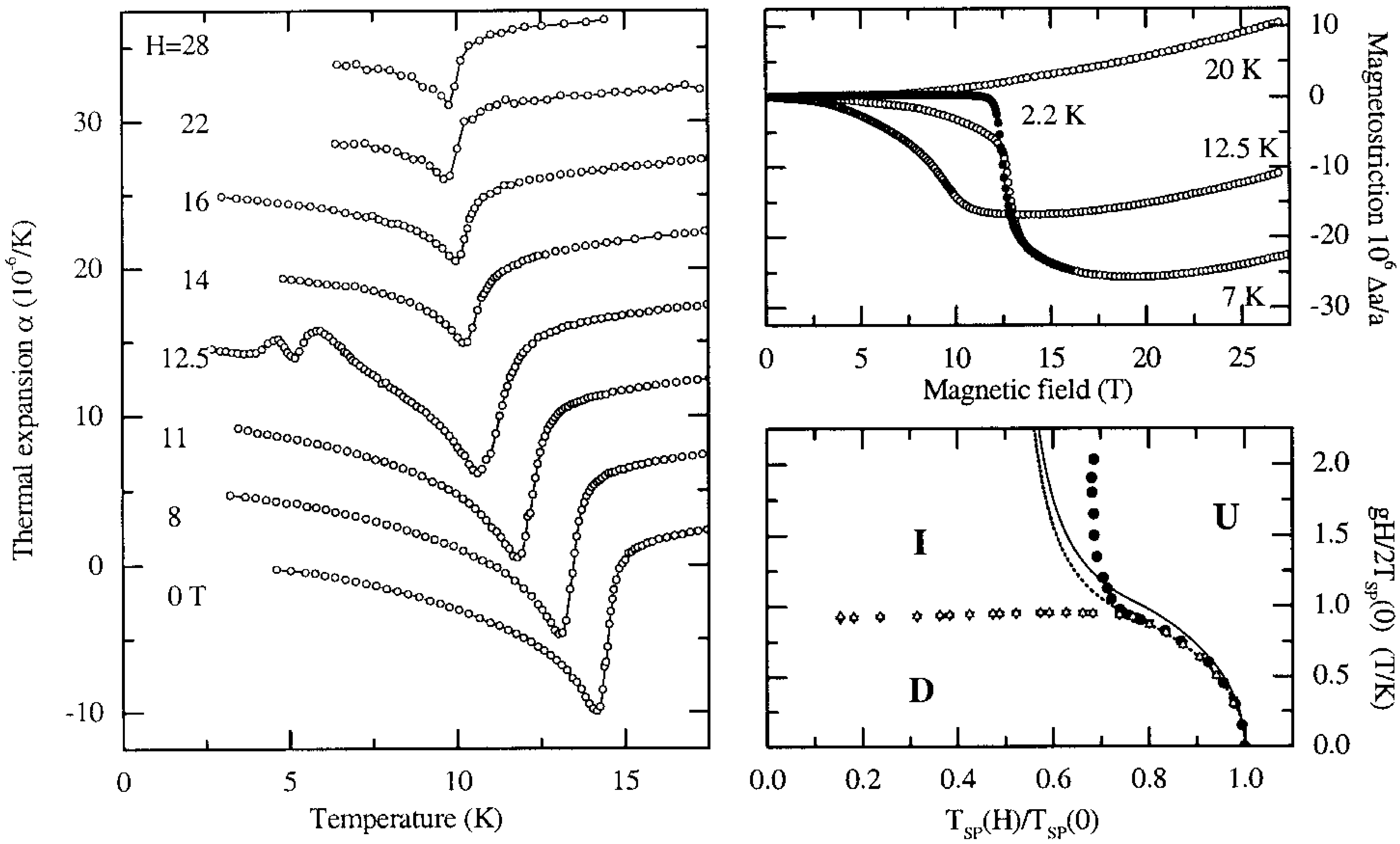

Figure 3 Left: Thermal expansion of the a axis in different magnetic fields. (With increasing H the curves are shifted by $5 \cdot 10^{-6}$/K.) Upper right panel: Magnetostriction of the a axis at different temperatures. Lower right panel: H–T phase diagram in reduced scales. The solid line is the theoretical prediction of Cross [2] and the dotted line shows the same curve divided by 1.12 (see text).

for $J'/J \gtrsim 0.24$, i.e. Cross' approximation $\chi(T_{SP}) \simeq \chi(0)$ is not valid in this case. As shown in the Inset of Fig. 2 the experimental $\chi(T)$ in a small temperature range above T_{SP} is described by $J_{eff} = 130$ K and the rescaling factor amounts to $\chi(14.5\mathrm{K})/\chi(0) \simeq 1.1$. That means, the D/U phase boundary of $CuGeO_3$ perfectly agrees with the calculation of Cross. At first sight, this might seem contradictory to the above discussion of the pressure dependencies of T_{SP}. However, a tentative explanation could be that the relative change $T_{SP}(H)/T_{SP}(0)$ does not explicitly depend on J'/J, although the absolute value of $T_{SP}(0)$ is very sensitive to the frustration. Calculations of the phase diagram including a magnetic frustration are highly desirable in order to check this point, but also with respect to the U/I boundary. The CF theory expects that for high fields $T_{SP}(H)$ saturates at $0.5 \cdot T_{SP}(0)$ [2]. For $H \geq 24$ T a field independence of $T_{SP}(H)$ is actually present in $CuGeO_3$. However, T_{SP} saturates at 9.9 K$\simeq 0.7 \cdot T_{SP}(0)$, i.e. at a higher temperature than expected from the CF theory (see Fig. 3).

Fig. 4 displays the spontaneous strain ϵ_a as a function of field and temperature. For $T \to 0$ the ϵ_a remain nearly unchanged within the D phase, whereas their size is strongly reduced in the I phase. This is a consequence of the spatial modulation $A(n)$ of the order parameter in the I phase leading to a reduced average

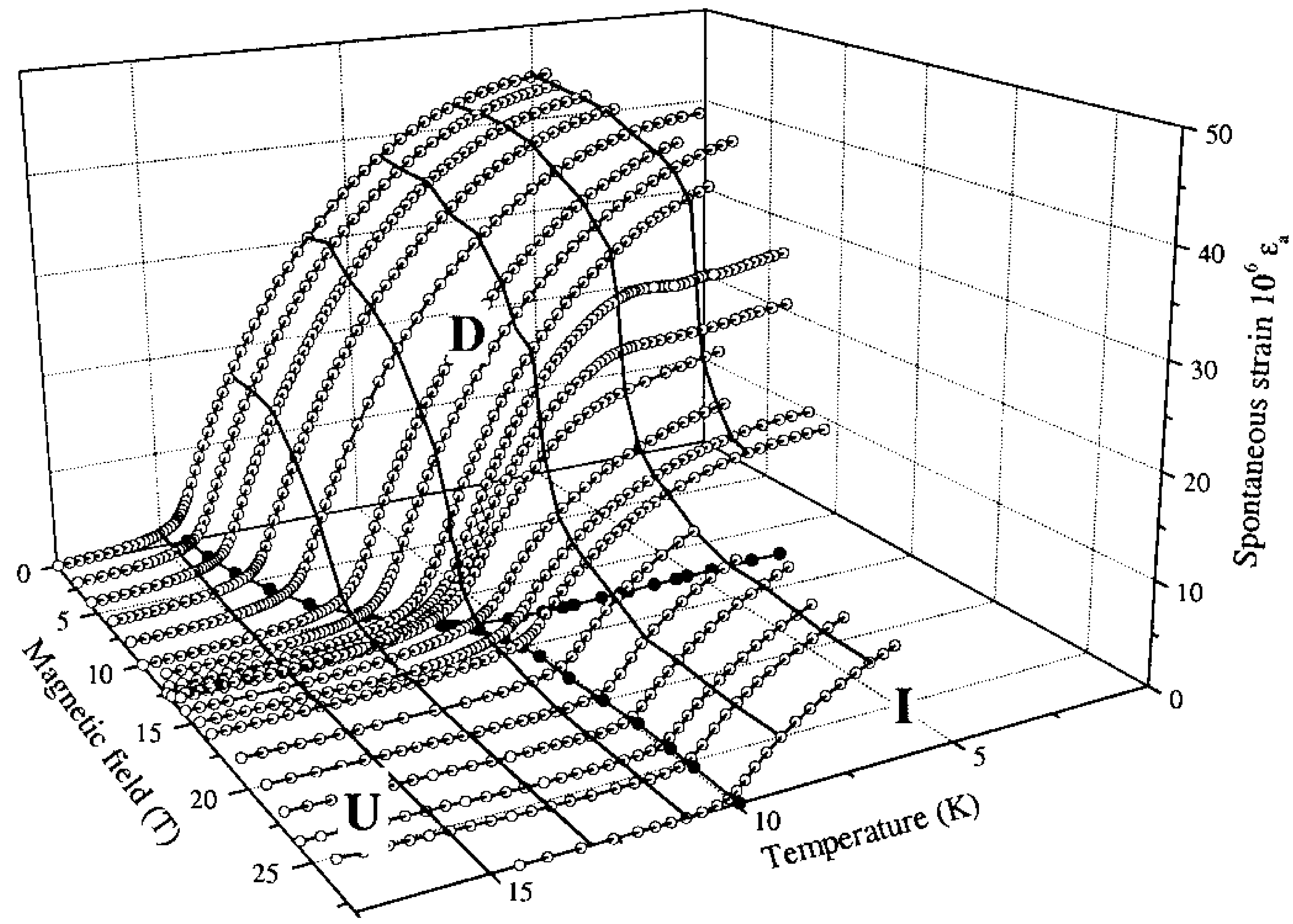

Figure 4 Field and temperature dependence of the spontaneous strain ϵ_a (o). The closed symbols show the phase diagram.

value $\langle A^2(n)\rangle$, which is measured by the macroscopic strain. Although the average value $\epsilon_a \propto \langle A^2(n)\rangle$ does not give direct information about the microscopic character of $A(n)$, it is possible to discriminate between a sinusoidal modulation and a soliton lattice by comparing $\epsilon_a(H)/\epsilon_a(0)$ to $\langle A^2(n,H)\rangle/A^2(0)$ calculated for the different models. For a sinusoidal modulation $A(n) = A_0 \sin[q(H)nc]$ (c is the lattice constant) the main reduction will be due to $\langle \sin^2[q(H)nc]\rangle = 0.5$, which is independent on the wave vector $q(H)$. Therefore, ϵ_a should be reduced by a factor of 2 at $H_{\mathrm{D/I}}$ and a field–dependent $\epsilon_a(H)$ above $H_{\mathrm{D/I}}$ can only arise from a field–dependent $A_0(H)$. This is different for a soliton lattice. In this case, $A(n)$ is reduced around the domain walls over a length scale determined by the correlation length ξ. Since the number of domain walls continuously increases with H, a continuous decrease of $\langle A^2(n,H)\rangle$ to 0 is expected.

Fig. 5 displays $\epsilon_a(H)/\epsilon_a(0)$, which may be obtained either from the thermal expansion or the magnetostriction measurements [13, 14]. The strong decrease of $\epsilon_a(H)/\epsilon_a(0)$ in the low–field range of the I phase is consistent with the soliton picture assuming $\xi = 10c$ (solid line in Fig. 5). However, with further increasing H the decrease of $\epsilon_a(H)/\epsilon_a(0)$ weakens and saturates at $\simeq 0.27$ for $H > 23$ T. This saturation at a finite value is in qualitative disagreement to the soliton picture and indicates a change from a soliton lattice close to $H_{\mathrm{D/I}}$ towards a sinusoidal modulation at higher fields.

This scenario is confirmed by DMRG calculations of the order parameter modulation $A(n)$ in the I phase [14]. As shown in the right part of Fig. 5 the calculated $A(n)$ is soliton–like for $H \simeq 12.5$ T, whereas it looks like a simple sine wave for

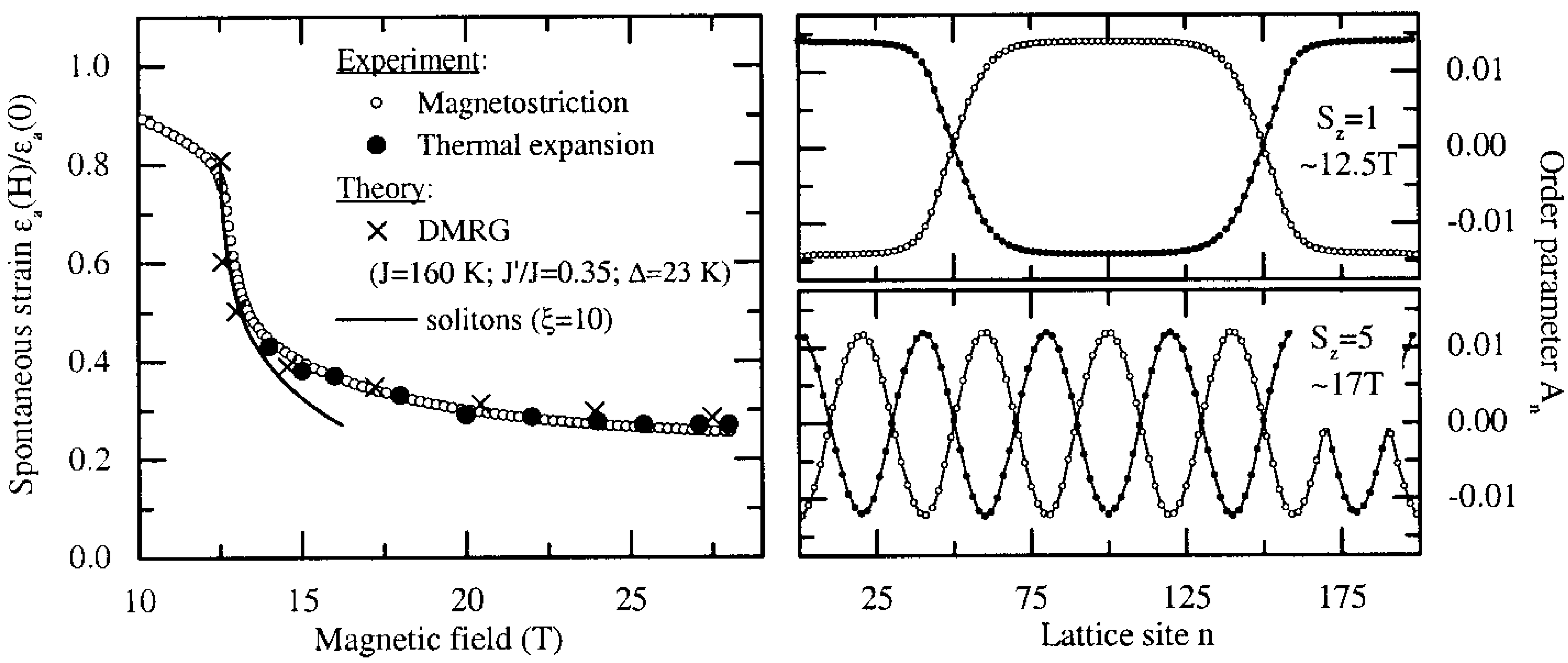

Figure 5 Left: Comparison of $\epsilon_a(H)/\epsilon_a(0)$ derived from thermal expansion (•) and magnetostriction (○) measurements to calculations for a soliton lattice (–) and DMRG results (×). Right: Modulation of the order parameter in the I phase for $H \simeq 12.5$ T (top) and $H \simeq 17$ T (bottom) obtained by DMRG. The open (closed) symbols denote even (odd) lattice sites along the chain direction [14].

$H \simeq 17$ T. The numerical result of $\langle A^2(H,n)\rangle / A^2(0)$ does even quantitatively agree very well to the experimental $\epsilon_a(H)/\epsilon_a(0)$ (see left part of Fig. 5). It is emphasized that the only parameters which enter the calculation are J, J', and Δ and the agreement is obtained using the values given above without additional fit parameters. In other words, from properties determined in the U and D phases of $CuGeO_3$ the experimental $H_{D/I}$ as well as the field dependence of $\epsilon(H)$ in the I phase are quantitatively reproduced. The modulation $A(n)$ has also been calculated using the exchange constants $J = 150$ K, $J'/J = 0.24$ and $J = 120$ K, $J' = 0$. In both cases, a change from a soliton lattice towards a sinusoidal modulation is obtained, too. However, the saturation values amount to 0.33 and 0.45 for $J'/J = 0.24$ and $J' = 0$, respectively. Thus, the good agreement between the experimental and numerical data suggests that the structural modulation of the I phase significantly depends on J'/J.

4 Summary

Various observations in $CuGeO_3$, which are not expected within the usual spin–Peierls theory, may be explained by a model of 1d frustrated spin chains. The susceptibility calculated for the exchange constants $J = 160$ K and $J' = 0.35J$ is in perfect agreement and the magnetic entropy is consistent with the respective experimental data. Using the same parameters numerical calculations quantitatively reproduce the measured field dependence of the averaged order parameter square in the I phase. Moreover, a pressure–dependent frustration ratio J'/J yields a possible explanation for the correlation between the uniaxial pressure dependencies of T_{SP} and χ. It remains, however, to be clarified whether the field dependence of T_{SP} is influenced by the frustration, since the D/U phase boundary of $CuGeO_3$ perfectly agrees to the calculation of Cross (for $J' = 0$).

Acknowledgement

I like to thank B. Büchner, A. Klümper, R. Raupach, F. Schönfeld, and G.S. Uhrig for valuable discussions and J.P. Renard for critical reading the manuscript. This work was supported by the Deutsche Forschungsgemeinschaft through SFB 341.

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