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Magnetostriction of the spin-Peierls cuprate $CuGeO_3$

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Abstract. The isothermal, longitudinal magnetostriction of the spin-Peierls cuprate $CuGeO_3$ has been measured along the three orthorhombic directions up to 14 Tesla by means of a high resolution capacitance dilatometer. For all three axes we observe anomalies at both the dimerized/incommensurate (D/I) and the dimerized/uniform (D/U) transition whose sizes and signs differ. A precise H-T phase diagram is determined from the field and temperature dependence of the lattice constants, which roughly agrees with theoretical predictions. At the D/I transition the magnetostriction shows a jumplike behavior and a hysteresis indicating a first order transition. From the jumps of the magnetostriction at the D/I transition we estimate considerable, uniaxial stress dependences of the critical magnetic field H_c, which correlate with those of the spin-Peierls transition temperature. A finite magnetostriction is also resolved within the high temperature uniform phase of $CuGeO_3$ for all three lattice directions. These data show a pronounced, strongly anisotropic spin-lattice coupling in $CuGeO_3$ and allow to derive the uniaxial pressure dependences of the magnetic susceptibility. Based on our findings the relevance of different structural parameters for the magnetic exchange interaction is discussed.

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I. Introduction

The discovery of a spin-Peierls transition in the inorganic cuprate $CuGeO_3$ by Hase et al. [1] renewed the interest for this magnetoelastic transition occurring in quasi one-dimensional, antiferromagnetic insulators. $CuGeO_3$ crystallizes in an orthorhombic structure with the space group *Pbmm* at room temperature [2]. The unit cell contains two Jahn-Teller-elongated CuO_6 octahedra forming the edge sharing Cu-O(2) chains oriented parallel to the c axis. Many properties of $CuGeO_3$ are well described by a model of spin-$\frac{1}{2}$ Heisenberg chains showing a spin-Peierls transition at $T_{SP} \simeq 14.3$ K. Intensive experimental studies during the last three years revealed many characteristic features expected for this transition, i.e. the rapid drop of the susceptibility for fields along all lattice directions below the phase transition [1], the structural distortion which transforms the uniform chain into a chain of alternating distances [3], the gap in the magnetic excitation spectrum [4] etc.. Similar to organic spin-Peierls systems such as $MEM(TCNQ)_2$ [5] or TTFCuBDT [6], the theoretical predictions of Cross and Fisher [7, 8] seem to be the most applicable theory at present. Indeed, it turns out that the low temperature properties, e.g. the H-T phase diagram, are well described by this theory. However, striking discrepancies between theory and experiment are present when considering the behavior of the uniform phase, i.e. for $T > T_{SP}$. For example the temperature dependence of the magnetic susceptibility does not agree with the theoretical calculations for the $S = \frac{1}{2}$ uniform Heisenberg antiferromagnetic chains [1]. To describe the broad maximum of the susceptibility around 60 K a coupling constant of $J \sim 88$ K is necessary. However, for this and for any other choice of J, the agreement between theory and measurement is poor in the entire temperature range [1]. Recently it was shown that a much better agreement is obtained when assuming in addition a significant antiferromagnetic exchange between next nearest neighbors (J') [9, 10] which competes with the nearest neighbor interaction. Moreover, it was argued that this frustration of the quasi one-dimensional, antiferromagnetic exchange is an important source for the occurrence of the spin-Peierls transition in $CuGeO_3$ [10, 11], i.e. one may speculate that the mechanism of the transition markedly differs from that in the conventional spin-Peierls theories [7, 12, 13].

In order to study the coupling between magnetic and lattice degrees of freedom in $CuGeO_3$ we have measured the isothermal, longitudinal magnetostriction of a $CuGeO_3$ single crystal, i.e. the change of the lattice constants as a function of an external magnetic field at constant temperature. From these data measured in a large temperature and field range information on properties of

both the low temperature phases and the uniform phase is obtained.

For the uniform phase it is possible to extract the spin-lattice coupling. The presence of a finite magnetostriction is an important precondition for the application of the Cross Fisher [7] and all other earlier theories [12, 13] to the spin-Peierls transition in a given compound. However, measurements of the magnetostriction do not only allow to show the bare presence of the spin-lattice coupling. The structural parameters which are most relevant for the magnetic exchange and the amount of their influence can also be determined, in particular when measuring the anisotropic magnetostriction of a single crystal. The knowledge of these structural parameters yields information on the spin-phonon coupling and is important to interpret the dimerization of the magnetic exchange in the dimerized phase.

In the ordered phases at low temperatures the field dependence of the lattice constant is determined by the structural changes at the different phase transitions in the characteristic spin-Peierls H-T phase diagram. These changes are expected to be large for $CuGeO_3$, since various studies of the lattice constants as a function of temperature using diffraction [14–16] and high resolution capacitive methods [17] yield that the spin-Peierls transition leads to large, strongly anisotropic, spontaneous strains, which scale with the order parameter of the transition. Thus the magnetostriction at low temperatures allows to study the field dependence of these spontaneous strains and their changes at the field driven phase transitions.

The paper is organized as follows. After a brief presentation of the experimental technique we illustrate the thermodynamic description of the magnetostriction of a solid and its relationship to the magnetic properties in Sect. III. In particular, we will focus on the relationship between the magnetostriction and the basic properties of a compound undergoing a spin-Peierls transition. Our experimental results are presented and discussed in Sect. IV and the main findings are summarized in Sect. V.

II. Experimental

The experiments were performed on a single crystal of $CuGeO_3$ of about $6.0 \times 5.0 \times 8.25\,mm^3$ cut from a large cylindrical crystal grown by a floating zone technique [18]. For all three lattice directions the isothermal, longitudinal magnetostriction, i.e. the change of the relative length of the sample as a function of an external magnetic field at constant temperature $\frac{\Delta L}{L}(H)$ (L: length of the sample) was measured by means of a high resolution capacitance dilatometer. The dilatometer is fixed in the center of a superconducting magnet. The sample is attached to the moveable plate of a plate capacitor whereby the sample's length change results in a variation of the capacitance, which is resolved with high accuracy (better than $5 \cdot 10^{-7}$ pF). The magnetic field is sweeped continuously with a rate of 0.25 T/min, while the temperature is stabilized with a deviation ΔT smaller than 0.02 K by a software PID-controller. The magnetostriction measurements on $CuGeO_3$ were carried out up to 14 Tesla in the temperature range between 2.5 K and 60 K. For calibration of the dilatometer, which shows a small temperature independent magnetostriction, measurements on Si and Mo were performed. The accuracy of the absolute values of the magnetostriction at the maximum field is better than $5 \cdot 10^{-7}$. The relative resolution determined from the scatter of the data in a single measuring run is more than two orders of magnitude higher.

III. Magnetostriction of a spin-Peierls compound

The reversible magnetostriction of any magnetic system is thermodynamically equivalent to the stress dependence of its magnetization. By introducing the elastic constants in the thermodynamic equations the magnetostriction measures the initial strain dependence of the magnetization in the limit of zero strain. This strain dependence of the magnetization is a fundamental property of a magnetic system yielding information on details of the magnetic exchange in a solid. In principle it may also be measured directly by applying an external stress and observing the change of the magnetization. However, in most cases it is much more reliable to measure the magnetostriction and to extract the stress dependence of the magnetization from thermodynamic relations. In particular, it is possible to determine small uniaxial pressure dependences of the magnetic susceptibility via the magnetostriction.

It is easily seen from the differential of the free enthalpy $dG = V\,dp - M\,dH - S\,dT$ that the volume magnetostriction $\omega = \frac{\Delta V}{V}(H)$ is related to pressure and magnetization in the following very simple way:

$$\frac{\partial \omega}{\partial H} = -\frac{\partial m}{\partial p}\bigg|_{p=0} = \kappa \frac{\partial m}{\partial \omega}\bigg|_{p=0}, \tag{1}$$

where m denotes the magnetization per unit volume, p the pressure, and $\kappa = -\frac{1}{V}\frac{\partial V}{\partial p}$ the compressibility. In single crystals one usually does not measure the volume magnetostriction but the length change of the sample along a well defined crystallographic direction. The obtained isothermal magnetostriction coefficients are related to the uniaxial pressure dependences of the magnetization. This relationship reads

$$\frac{\partial \varepsilon_{ij}}{\partial H_a}\bigg|_{\sigma, T, H_a = 0} = \frac{\partial m_a}{\partial \sigma_{ij}}\bigg|_{H, T, \sigma_{ij} = 0}. \tag{2}$$

where ε_{ij} and σ_{ij} are strain and stress, respectively, and m_a denotes the magnetization in a direction after applying an external field H_a along the same lattice direction. In experiment the length changes, i.e. the diagonal components ε_{ii}, are measured, which are related to the uniaxial pressure dependences $\partial/\partial p_i \equiv -\partial/\partial \sigma_{ii}$. Note that in the general case, i.e. without considering special symmetries of the lattice structure or the magnetization, nine different magnetostriction coefficients have to be measured to determine the volume magnetostriction. Apart from the longitudinal effects measured in this study, there are two transversal magnetostriction coefficients for each lattice direction obtainable, if the magnetic field is applied perpendicular to the measuring direction.

From the description given so far it is apparent that the magnetostriction measures the strength of the coupling between elastic and magnetic degrees of freedom. Thus it yields important information on basic properties of a compound undergoing a spin-Peierls transition. For a more quantitative description of the magnetostriction in spin-Peierls compounds it is necessary to distinguish between the behavior above and below the spin-Peierls transition.

A. Temperatures above T_{SP} $(H = 0) \equiv T_{SP}^0$

The presence of a finite magnetostriction within the uniform phase is an important precondition for the occurrence of a spin-Peierls transition. As a starting point of the theoretical treatments of this transition it is assumed that the antiferromagnetic coupling J within the one-dimensional chains is a function of the strain ε. Obviously, this implies that the magnetic susceptibility χ, which is determined by J, also depends on lattice strains and stress. Thus a magnetostriction, i.e. a finite pressure dependence of the magnetization (or susceptibility), is necessarily expected for any spin-Peierls compound.

To obtain a more quantitative description of the magnetostriction we consider an expansion of the free energy density f in powers of the magnetic field and the stress. Within the uniform phase of a spin-Peierls system there is no spontaneous magnetization and the magnetization has central symmetry. Thus f is a function of even powers of H only and for an isothermal process we get

$$\begin{aligned} f = &- \Theta + \Xi - \tfrac{1}{2} m_{ab} H_a H_b - \tfrac{1}{4} m_{abcd} H_a H_b H_c H_d \\ &- \tfrac{1}{2} m_{abij} H_a H_b \sigma_{ij} - \tfrac{1}{4} m_{abcdij} H_a H_b H_c H_d \sigma_{ij} \\ &- \tfrac{1}{4} m_{abijkl} H_a H_b \sigma_{ij} \sigma_{kl} \cdots , \end{aligned} \tag{3}$$

where the summation is understood with respect to each suffix which enters twice in a term, and where $a, b, \ldots, i, j, \ldots$ take the values of 1, 2, 3. Θ is a function of stresses and temperature only, whereas Ξ and m's are functions of temperature only. Neglecting higher-order terms in the magnetic field the susceptibility tensor reads

$$\chi_{ab} = - \frac{\partial^2 f}{\partial H_b \partial H_a} = \frac{\partial m_a}{\partial H_b} = m_{ab} + m_{abij} \sigma_{ij} \cdots . \tag{4}$$

Magnetization measurements up to ~ 25 T [19, 20] show that this expression is sufficient to describe the susceptibility of $CuGeO_3$ at ambient pressure for moderate fields. The non-linear effects neglected in (4) become important for extremely high fields only [21].

The magnetostriction, i.e. the magnetic field dependent part of the strain tensor, is given in leading order by

$$\varepsilon_{ij} = - \frac{\partial f}{\partial \sigma_{ij}} = \frac{1}{2} m_{abij} H_a H_b + \cdots . \tag{5}$$

As will be shown below, the experimental findings in the uniform phase of $CuGeO_3$ are well described by this expression, where the magnetostriction $\Delta L/L$ is proportional to H^2.

Combining Eqs. (4) and (5) the relationship between the magnetostriction within the uniform phase and the uniaxial stress dependence of the susceptibility is apparent:

$$\left. \frac{\partial \chi_{ab}}{\partial \sigma_{ij}} \right|_{\sigma_{ij} = 0} = m_{abij} = \frac{1}{H_b} \frac{\partial \varepsilon_{ij}}{\partial H_a} . \tag{6}$$

Thus, from measurements of $\varepsilon_{ii}(H)$ it is possible to determine the uniaxial pressure dependences of the diagonal components of the magnetic susceptibility tensor. For a spin-Peierls compound with Heisenberg $S = \frac{1}{2}$ chains an isotropic susceptibility is expected. Taking into account the slightly different gyromagnetic ratios g along the three lattice directions, this isotropic magnetic susceptibility is indeed observed in $CuGeO_3$. Therefore the volume magnetostriction can be calculated without the transverse effects in this particular case.

B. Temperatures below T_{SP}^0

The general connection between the magnetostriction and the stress dependence of the magnetization also holds below T_{SP}. However, the simple treatment presented in the last section is no longer valid. Due to the well known decrease of the spin-Peierls order parameter Φ, i.e. the dimerization of the lattice, with increasing magnetic field, the magnetization increases nonlinearly with H. Vice versa, the magnetization at a given field or temperature is a function of $\Phi(T, H)$. The transition temperature T_{SP} and as a consequence $\Phi(T, H)$ also depends on external stress. For $CuGeO_3$ these pressure effects are known to be extremely large and strongly anisotropic [17]. Thus, below T_{SP} a magnetostriction occurs which is related to the magnetic field and pressure dependence of the spin-Peierls order parameter. An approach for the discussion of the magnetostriction below T_{SP}^0 is possible, when starting from the spontaneous strains which have been found for $CuGeO_3$ in zero magnetic field [15, 17]. As has been discussed in detail by Winkelmann et al. [17] these spontaneous strains are closely related to the uniaxial pressure dependences of T_{SP}. This relationship is expressed in the Ehrenfest or the similar Pippard relation. The uniaxial pressure dependences of the transition temperature scale with the anomalies of the thermal expansion ($\Delta\alpha_i$) according to

$$\left. \frac{\partial T_{SP}}{\partial \sigma_{ii}} \right|_H = - v T_{SP} \frac{\Delta \alpha_{ii}}{\Delta c_\sigma} , \tag{7}$$

where v denotes the unit volume and Δc_σ the anomaly of the specific heat (at constant pressure) at a second order phase transition. Hence spontaneous strains occur when decreasing the temperature below T_{SP}^0, since the spin-Peierls transition temperature T_{SP}^0 in $CuGeO_3$ is pressure dependent. By definition, these spontaneous strains are given by the temperature integral of the anomalous contributions of the thermal expansion coefficients.

A phenomenological description of these spontaneous strains is possible when assuming a coupling between the spin-Peierls order parameter and lattice strains in the

Landau free enthalpy

$$G = \int \mathrm{d}V \{A(\nabla\Phi)^2 - h\Phi + a(T - T_c)\Phi^2 + u\Phi^4 + \cdots + \tfrac{1}{2}c_{ijkl}\varepsilon_{ij}\varepsilon_{kl} + \zeta_{ij}\Phi^2\varepsilon_{ij}\}, \quad (8)$$

where $\frac{1}{2}c_{ijkl}\varepsilon_{ij}\varepsilon_{kl}$ is the usual elastic energy contribution (c_{ijkl} is the stiffness tensor) and $\zeta_{ij}\Phi^2\varepsilon_{ij}$ corresponds to the lowest order nonvanishing coupling term between the order parameter and strain. The condition of a stress free sample $\frac{\partial G}{\partial \varepsilon_{ij}} = 0$ gives

$$-\Phi^2\zeta_{ij} = \tfrac{1}{2}\, c_{ijkl}\varepsilon_{kl}, \quad (9)$$

which relates the order parameter to the spontaneous strains, which occur for finite Φ, i.e. below T_{SP}. The expression in Eq. 9 means that a reduction of the spin-Peierls order parameter as a function of an external field H leads to a reduction of the spontaneous strains. Therefore, at the field driven transitions between the dimerized and the uniform phase a contribution to the magnetostriction is expected, whose size and anisotropy corresponds to that of the large spontaneous strains of the dimerized phase.

At low temperatures and high fields a further magnetic phase characterized by an incommensurate lattice distortion occurs in spin-Peierls compounds. At the corresponding field driven phase transition one may also expect anomalies of the magnetostriction. However, since at this transition the meaning of the order parameter changes, it is impossible to extract any predictions for the magnetostriction from the treatment within the Landau theory discussed so far. However, when considering the close relationship between spontaneous strains and uniaxial pressure dependences it is possible to estimate the anomalies of the lattice constants at the D/I transition. Similar to the anomalies of the thermal expansion (see Eq. 7) the jumps (kinks) of the magnetostriction at field driven first (second) order phase transitions are related to the uniaxial pressure dependences of the corresponding transition fields. In the case of first-order transitions, the jumps are related to the stress dependence of the critical magnetic field H_c via the Clausius-Clapeyron relation:

$$\left.\frac{\partial H_c}{\partial \sigma_{ii}}\right|_{T,\sigma_{ii}=0} = -\frac{\Delta\varepsilon_{ii}}{\Delta m} \quad \text{with} \quad H \parallel m, \quad (10)$$

where Δm denotes the jump of the isothermal magnetization at H_c. For a second-order phase transition one can extract the pressure dependence of H_c from the kinks of the magnetostriction and the magnetization using an Ehrenfest relation.

Following the theoretical descriptions of spin-Peierls compounds, which predict an universal, i.e. pressure independent H-T phase diagram, the signs of the magnetostriction jumps at the discontinuous D/I transition can be easily predicted. If T_{SP} decreases (increases) as a function of uniaxial pressure the theory predicts a corresponding decrease (increase) of H_c. Since the magnetization increases at the D/I transition, a negative (positive) pressure dependence of the critical field H_c corresponds to a decrease (increase) of the lattice constant at the D/I transition.

IV. Results and discussion

We have studied the change of length $\varepsilon_{ii}(H) \equiv \frac{\Delta L}{L}(H)$ along the a, b, and c axes as a function of an external magnetic field up to 14 T applied parallel to the measured direction in a temperature range between 2.5 K and 60 K.

The results of the measurements along the a axis for temperatures below T_{SP}^0 are displayed in Fig. 1. The upper part shows the magnetostriction of the a axis between 4.5 K and 11.0 K. At low temperatures the change of the length is rather small below about 12 T. In the field region between 12 T and 13 T there is a strong, jumplike reduction of the lattice constant with a maximum value of $\frac{\Delta L}{L} \approx -22 \cdot 10^{-6}$ indicating the transition between the dimerized (D) and the magnetic, incommensurate (I) phase [22]. These length jumps are a characteristic property of the D/I transition in $CuGeO_3$ and give evidence for a first order character of this transition. As shown in Fig. 2, the length jumps $\Delta\frac{\Delta L}{L}$ decrease with increasing temperature and disappear above 11.0 K. The latter is also visible in the raw data of the magnetostriction at temperatures between 11 K and 14 K plotted in the lower part of Fig. 1. Above 11.0 K there is still a strong reduction of the lattice constant as a function of field, but in contrast to the findings at lower temperatures it decreases continuously, i.e. without any jumps. This continuous decrease of the length above 11.0 K or more precisely its change of slope signals the D/U transition, which is theoretically expected to be of second order. Thus, by comparing the shape of the magnetostriction curves above and

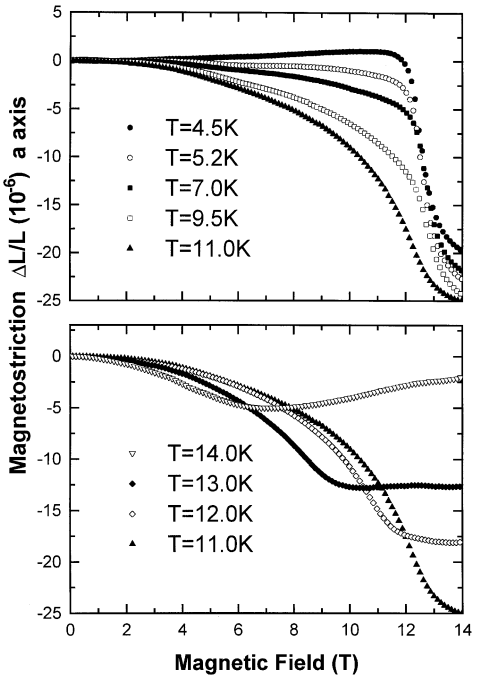

Fig. 1. Magnetostriction of the a axis in the temperature region between 4.5 K and 14.0 K. Below 11.0 K the magnetostriction is jumplike indicating a first order D/I transition, whereas above 11.0 K the change in length is continuous reflecting a second order D/U transition

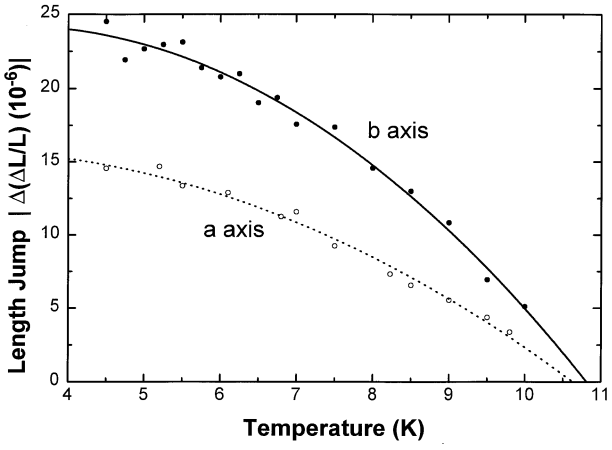

Fig. 2. Length jump $\Delta\frac{\Delta L}{L}$ of the a and b axis versus temperature. The length jumps decrease with increasing temperature and disappear above 11.0 K. The solid and broken lines are guides to the eye

below 11.0 K, respectively, it is possible to distinguish between field driven D/I and D/U transitions. Moreover, from the shape of the anomalies a first-order character of the D/I transition is indicated in the entire temperature range up to 11 K.

Besides the shape of the anomalies there are further differences of the magnetostriction below and above 11 K, respectively. At higher temperatures the phase transition shifts to lower fields with increasing temperature, whereas the transition field of the D/I transition hardly depends on temperature. In addition, the total magnetostriction up to 14 Tesla scarcely varies in the temperature range below 11 K, whereas it strongly reduces with increasing temperature above 11 K. This means that the lattice constants in the D and I phases show a similar temperature dependence, whereas those in the D and U phases markedly differ. Note that the latter is just a consequence of the continuous temperature dependent development of the spontaneous strains in the D phase discussed in the last section.

These findings are confirmed by our data of the magnetostriction along the two other lattice directions. Figure 3 presents the corresponding data for the b axis in the temperature region between 3.0 K and 14.0 K. Though the temperature dependence is similar to that found along the a axis, there is apparently a strong anisotropy. Whereas the magnetostriction of the a axis is negative in the temperature range below T_{SP}^0, the b axis shows a positive magnetostriction below T_{SP}^0 in the whole field region up to 14 T. Moreover, the maximum effect along the b axis is about twice as large as the maximum value of the magnetostriction along the a axis. The magnetostriction measured along the c direction (Fig. 4) is similar to that along the b axis. Again the lattice constant increases with increasing field in the entire temperature range below T_{SP}^0. But there is, however, a quantitative difference: the maximum magnetostriction is about three times smaller at lower temperatures and roughly six times smaller near T_{SP}^0.

A comparison of the three axes at $T = 4.5$ K and at $T = 13.0$ K showing a D/I and a D/U transition, respect-

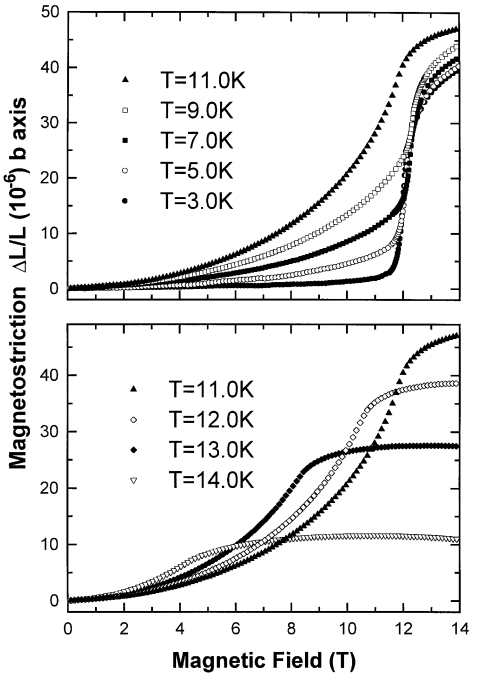

Fig. 3. Magnetostriction of the b axis in the temperature region between 3.0 K and 14.0 K

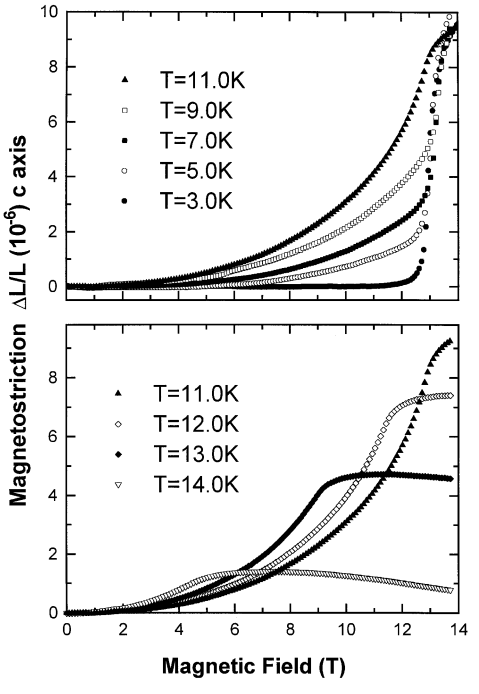

Fig. 4. Magnetostriction of the c axis in the temperature range between 3.0 K and 14.0 K

ively, is presented in Fig. 5. The arrows in the figure mark the critical fields H_c of the phase transitions, which are defined at the maximum in the field derivative of the magnetostriction. The different critical fields arise from the anisotropic g-values and coincide by scaling the

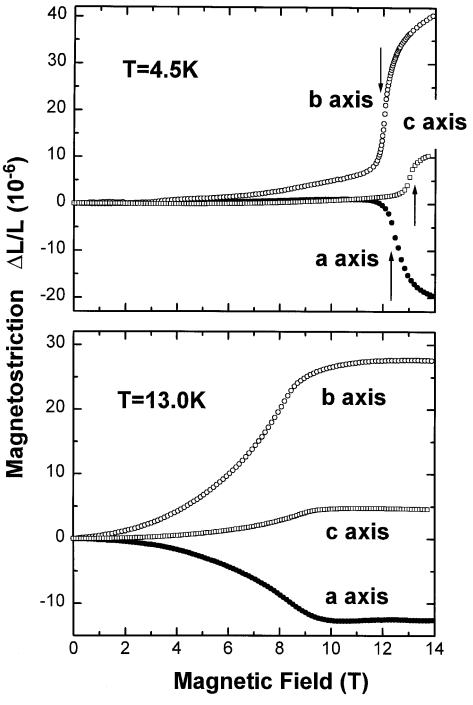

Fig. 5. Anisotropic magnetostriction of the *a*, *b* and *c* axis at 4.5 K and 13.0 K showing a D/I and D/U transition, respectively. The arrows mark the critical fields H_c of the phase transitions, which are defined at the maximum of the field derivative $\partial \frac{\Delta L}{L} / \partial H$

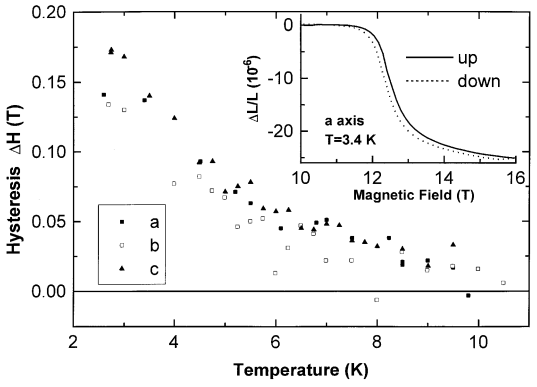

Fig. 6. Hysteresis $\Delta H = H_c^{up} - H_c^{down}$ versus temperature. The inset shows the magnetostriction with increasing (line) and decreasing (dots) field in the vicinity of the D/I transition at 3.4 K

H-axis with the g-values as determined from ESR ($g_a = 2.15$, $g_b = 2.23$, $g_c = 2.05$) [9].

Before discussing the H-T phase diagram as revealed from our data we analyze again the D/I boundary, which has been identified to be of first order by the shape of the magnetostriction curves. Further strong evidence for a first-order transition is obtained from a hysteresis of the magnetostriction observed along all directions. Up to now we have only presented data measured with increasing field, which slightly differ from those detected with decreasing field as shown in the inset of Fig. 6. This hysteresis is observable only at temperatures below 11.0 K and occurs in the vicinity of the D/I transition. Thus our data yield two different critical fields H_c^{up} and H_c^{down}. The hysteresis $\Delta H \equiv H_c^{up} - H_c^{down}$ as a function of the temperature is plotted in Fig. 6. At 2.5 K ΔH amounts to about 0.18 T. The hysteresis decreases with increasing temperature and disappears at 11.0 K. In agreement with the disappearance of the jumplike behavior of the magnetostriction, the hysteresis also yields a change from a first to a second-order transition at 11.0 K.

Hysteresis effects at the D/I transition are also present in organic spin-Peierls systems such as $MEM(TCNQ)_2$ [5] or TTFCuBDT [6]. But in contrast to our findings in $CuGeO_3$, $\Delta H(T)$ is a linear function of temperature in the case of $MEM(TCNQ)_2$ and in TTFCuBDT even a negative curvature of the hysteresis as a function of T has been found. Moreover, the reduced extrapolated hysteresis $\frac{\Delta H(T=0\,\mathrm{K})}{T_{SP}^0}$ of all organic systems are very similar (≈ 0.12 T/K). In contrast to that, $CuGeO_3$ shows a three times smaller reduced hysteresis. In the organic systems a hysteresis could, despite its larger value, only be resolved for rather small reduced temperatures. It disappears e.g. at $t = \frac{T}{T_{SP}^0} = 0.67$ and at t = 0.5 for $MEM(TCNQ)_2$ [5] and TTFCuBDT [6], respectively. This indicates a change of the D/I boundary from first to second order. In the case of $CuGeO_3$, a first-order character of the D/I transition is signaled by a hysteresis for the entire D/I phase boundary up to 11.0 K (see the phase diagram in Fig. 7). Although the hysteresis at the D/I transition seems to be a common feature of all spin-Peierls compounds, it is obvious, that each system shows slight differences in $\Delta H(T)$. At present it is not clear, whether these differences are a consequence of different intrinsic properties or just a consequence of lattice imperfections.

Both the hysteresis of the magnetostriction below 11.0 K in $CuGeO_3$ as well as its jumplike change show a first order D/I transition as expected from the theory of Cross and Fisher [7, 8]. From this finding one should, however, not exclude the description within soliton theories which predict a continuous D/I transition [23–25]. The observed weakly first-order character of the phase transition might also be caused by pinning of solitons at lattice defects [23–25].

From our magnetostriction data and from thermal expansion measurements in fixed magnetic fields up to 16 T (for details see [26]), which have been carried out with the same high resolution capacitance dilatometer, a precise H-T phase diagram of $CuGeO_3$ is obtained. It is shown in the upper part of Fig. 7. The magnetostriction reveals a very sensitive probe of the D/I transition, because the boundary is crossed almost perpendicular (see Fig. 7). Vice versa, the U/I transition is only obtained with high precision from the thermal expansion measurements. The D/U transition can be observed by both methods and the temperature (field) dependent transition fields (temperatures) give the same boundary in perfect agreement. Since the magnetostriction and thermal expansion are static quantities, their anomalies are very reliable indicators of "true" phase boundaries. In particular the obtained phase boundaries are not influenced by relaxation effects, which have been observed e.g. in χ_{ac} measurements of organic spin-Peierls compounds [27].

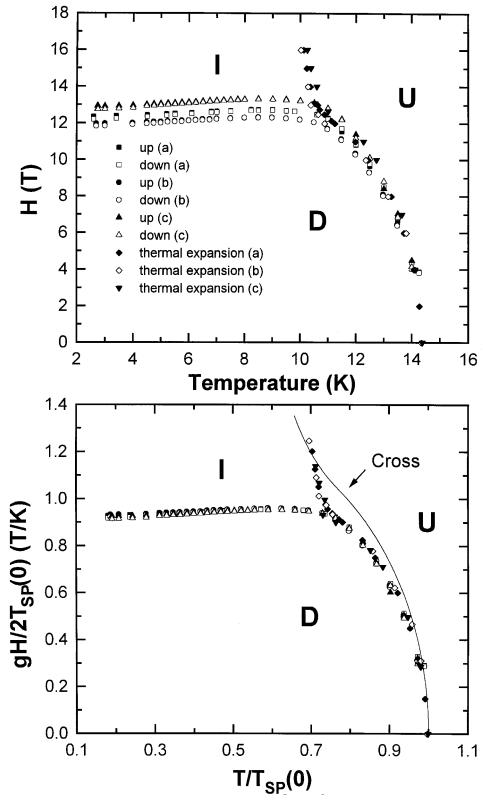

Fig. 7. Upper panel: H-T phase diagram with H parallel to the three principal axes. Lower panel: H-T phase diagram in terms of reduced variables. The solid line represents the theoretical prediction of the Cross theory

The magnetic field was applied parallel to the three axes and we get three slightly different phase diagrams, which reflect the anisotropy of the g-values as mentioned above. In the lower part of Fig. 7 we show the same data on reduced field $h = \frac{gH}{2T_{SP}}$ and temperature $t = \frac{T}{T_{SP}}$ scales, where h takes into account the anisotropic g-values. In this representation the phase boundaries obtained for the three field directions fall onto a single line. Moreover, the magnetic phase diagram of $CuGeO_3$ agrees qualitatively with the theoretical predictions for a spin-Peierls system. A detailed comparison of this experimental phase diagram to several theoretical predictions [8, 13, 23, 24, 25, 27] is given elsewhere [26]. We mention that the best agreement is obtained with the results of the Cross-Fisher theory (solid line in Fig. 7). But as shown in Fig. 7 there are also significant deviations from these calculations, which, however, vanish nearly completely when reducing the calculated transition fields by about 10%. In other words, the influence of the magnetic field destabilizing the dimerzied phase in $CuGeO_3$ appears to be $\sim 10\%$ larger than that expected from theory.

Whereas the influence of a magnetic field on the spin-Peierls transition is isotropic apart from the slightly different gyromagnetic ratios, i.e. nearly independent on the direction of the applied field, there is a pronounced anisotropy of the magnetostriction jumps along the three lattice directions (see Figs. 2, 5). This anisotropy of the magnetostriction correlates with that of the anomalies found in the thermal expansion coefficients at T_{SP}^0 [17, 11]. In these data the largest effect is also found along the b axis and the smallest one in the c direction. Moreover, as well as in the magnetostriction, the anomalies of the thermal expansion coefficients are positive for the b and c axes, whereas the a axis shows an opposite behavior. The absolute values of the spontaneous strains at low temperatures as determined from the thermal expansion data in zero magnetic field amount to about 4.5, -7.4 and -2.0×10^{-5} for the a, b, and c axis, respectively [11]. Note that for all three directions the jumps of the magnetostriction at the D/I transition correspond to a reduction of about 50% of these spontaneous strains of the D phase, which are defined with respect to the lattice constants of the U phase. This means that there are reduced but still large spontaneous strains in the incommensurate phase, which we have also observed in thermal expansion measurements in fixed external fields [26]. Moreover, the anisotropy of the spontaneous strains in the D and I phases is very similar.

As discussed in Sect. IIIB, the correlation between the magnetostriction jumps and the anomalies of the thermal expansion is expected for a spin-Peierls compound. Both scale with the uniaxial pressure dependences of the phase boundaries in the universal H-T phase diagram, i.e. they are determined by a single quantity, namely the anisotropic strain order parameter coupling. To allow for a quantitative comparison with the theoretical prediction of a universal, pressure independent spin-Peierls phase diagram, we have calculated the uniaxial pressure dependences using Eq. 10 . and assuming the magnetization jumps as reported in [19]. In Table 1 the obtained uniaxial pressure dependences of the transition fields are presented. For uniaxial pressure along the a axis, the critical magnetic field H_c considerably decreases, whereas uniaxial stress along the b and c axis leads to the opposite effect. These pressure induced changes of H_c coincide with corresponding changes of T_{SP}^0. Uniaxial pressure causes either a simultaneous increase or decrease of the transition temperatures and critical fields separating the D phase from the U and I phases, respectively. In other words, uniaxial pressure either stabilizes or destabilizes the D phase with respect to both, temperature and magnetic field.

A quantitative comparison with the theory can be drawn by considering the temperature dependence of the D/I phase boundary displayed in the H-T phase diagram. Apparently, for temperatures below the tricritical point the critical field H_c is almost constant and corresponds to the magnetic field H^* at the tricritical point. According to the Cross-Fisher theory, H^* is proportional to T_{SP}^0 [8]. Moreover, theory predicts this scaling for all spin-Peierls compounds and thus also at finite pressure. Therefore the same scaling is expected for the uniaxial pressure dependences of H^* and T_{SP}^0, i.e. the universality of the H–T phase diagram implies that

$$\frac{1}{T_{SP}^0}\frac{\partial T_{SP}^0}{\partial p_i} = \frac{1}{H^*}\frac{\partial H^*}{\partial p_i}. \tag{11}$$

Due to the lack of detailed measurements of the magnetization jumps or kinks in $CuGeO_3$, we cannot compare in

Table 1. Left: Uniaxial pressure dependences of the critical field H_c at the dimerized/incommensurate phase transition as determined from Eq. (10) at different temperatures given. The magnetization jumps are taken from [19]. The error of the values amounts to about 20%. Right: Relative values of the uniaxial pressure dependences of $T_{SP}(H = 0)$ and $H_c(T = 4.2\ \mathrm{K})$. The former are determined from the anomalies of the thermal expansion and the specific heat using Eq. 7 [17]. Within experimental error the values agree to those reported and discussed in [17]

Crystal axes	$\frac{\partial H_c}{\partial p_i}\left(\frac{T}{\mathrm{GPa}}\right)$			$\frac{1}{T_{SP}^0}\frac{\partial T_{SP}^0}{\partial p_i}\left(\frac{1}{\mathrm{GPa}}\right)$	$\frac{1}{H^*}\frac{\partial H_c}{\partial p_i}\left(\frac{1}{\mathrm{GPa}}\right)$
	$T = 4.2$ K	8.0 K	9.0 K		T = 4.2 K
a	− 5.0	− 3.4		− 0.26	− 0.40
b	6.4		4.6	0.50	0.54
c	2.0	1.9		0.11	0.16

detail the experimental findings in $CuGeO_3$ to the simple theoretical prediction in Eq. (11) at present.

The values of the relative uniaxial pressure dependences of H_c at temperatures well below the tricritical point, which are also given in Table 1, roughly agree with those of T_{SP}^0. However, we emphasize that the error of the pressure dependences of H_c is quite large, since magnetostriction and magnetization have not been measured on the same crystal. Within the large error, the data are consistent with a pressure independent, universal H-T phase diagram, where the only parameter is the transition temperature T_{SP}^0. In order to investigate the theoretical prediction, more detailed studies of the magnetization of our crystals are planned.

The similarity between the anisotropies of the magnetostriction and those of the thermal expansion are predicted by theory for a spin-Peierls system. Further similarities are, however, not expected. Remarkably, the anisotropy of the absolute values of the magnetostriction correlates with that found for the uniaxial compressibilities of $CuGeO_3$ measured by Adams et al. [28]. Their x-ray diffraction studies at room temperature reveal that the lattice response to pressure is highly anisotropic. The *b* axis is most and the *c* direction least compressible. The ratios of the compressibilities of the three lattice constants roughly agree with those of the absolute values of the magnetostriction at low temperatures. This similarity between the magnetostrictive effects below T_{SP}^0 and the lattice compressibilities means that the anisotropy of the strain dependences of the properties of the spin-Peierls phase is much smaller than that of the corresponding stress dependences. An accurate calculation of the strain dependences is not possible at present, since not all elastic constants of $CuGeO_3$ have been measured yet. Nevertheless, the similar anisotropy of the compressibilities and the magnetostriction indicate that nearly the same increase of H_c and T_{SP}^0 is obtained by uniaxial pressures, which cause the same relative decrease of the lattice constant *b* and *c*, respectively. The absolute value for the strain dependence along the *a* axis is also similar to those along the *b* and *c* directions, but for the *a* axis the sign differs. This means that the data are consistent with nearly isotropic absolute values of the strain dependences, but a pronounced anisotropy remains with respect to the signs of the effects.

The similar absolute values estimated for the strain dependences of H_c or T_{SP}^0 may indicate that the spin-Peierls transition in $CuGeO_3$ depends rather on bond angles than on bondlengths in a particular lattice direction. For example, a reduction of the Cu–Cu distance within the magnetic chains is not the only origin of the strong pressure dependences of T_{SP}^0 and H_c.

As visible in Fig. 8, a small but significant magnetostriction is also resolved above the spin-Peierls transition T_{SP}^0. This finite magnetostriction measures the spin-lattice coupling, which is a precondition for the spin-Peierls transition. In other words the data in Fig. 8 are a measure of the change of the magnetic interaction in $CuGeO_3$ as a function of lattice distances as we will discuss below.

Remarkably, there is a striking correlation between the magnetostriction above and below T_{SP}^0. For all three lattice constants the sign of the magnetostriction changes at T_{SP}^0. Below T_{SP}^0 the field induced length changes are positive along the *b* and *c* axes and negative for the *a* direction. Within the U phase we find a negative magnetostriction along the *b* and *c* axes and a positive one along the *a* direction. The anisotropy of the absolute values of the magnetostriction above and below T_{SP}^0 is very similar. The field driven changes in the lattice constants in the U phase scale with the increase (decrease) of the lattice constants at the field induced D/I phase transition.

This correlation means that the magnetoelastic coupling in the uniform phase is related to the pressure dependence of the spin-Peierls transition. As we discuss in detail in [11] this correlation is not expected at all within conventional theories of the spin-Peierls transition. But this behavior can be explained, if one assumes that T_{SP}^0 in $CuGeO_3$ is strongly affected by a frustration of the magnetic exchange [11] as suggested in recent theoretical studies [9, 10].

Let us now analyze the magnetostriction above T_{SP}^0 along the lines described in Sect. IIIA. In leading order a quadratic field dependence of the lattice constant for this paramagnetic U phase is expected (see Eq. 5). Figure 9, for example, shows the magnetostriction of the *b* axis as a function of the squared magnetic field in the temperature range between 18.0 K and 60.0 K. Apparently the magnetostriction above the spin-Peierls transition follows the expected behavior and one concludes that the pressure dependence of χ is magnetic field independent for $H \leq 14$ T. Using Eq. (6) our data allow to calculate the uniaxial pressure dependences of the magnetic susceptibility. The results are plotted in Fig. 10 as a function of temperature. Obviously, the different signs of the magnetostriction correspond to different signs of the uniaxial pressure dependences of the

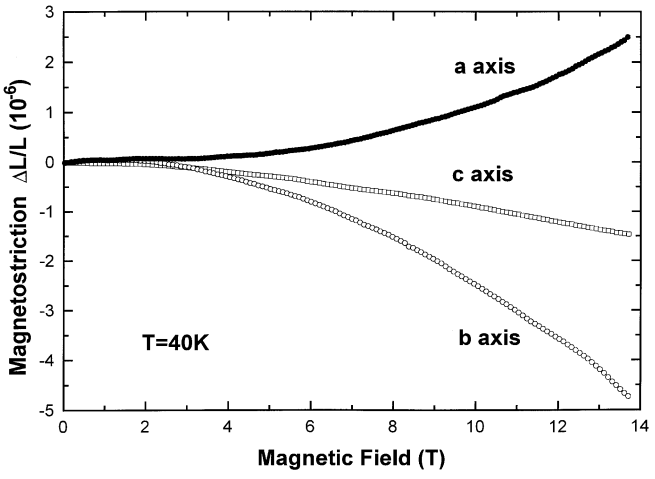

Fig. 8. Magnetostriction of the three lattice constants at 40 K

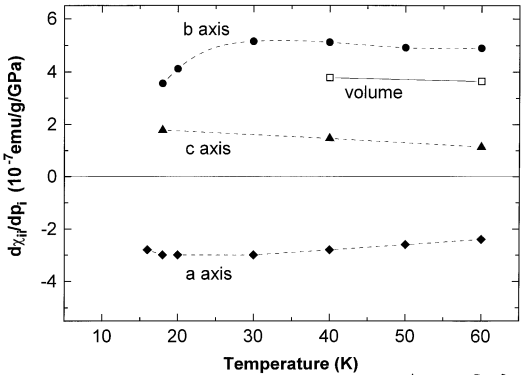

Fig. 10. Uniaxial pressure dependences of the magnetic susceptibility in the uniform phase as detected from the magnetostriction using Eq. (4) versus temperature. "Volume" denotes the estimated hydrostatic value assuming that the anisotropy with respect to the direction of the magnetic field arises form the different g-value only

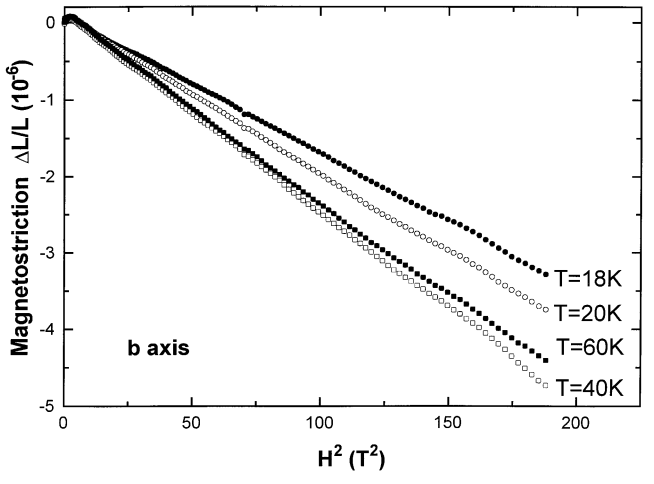

Fig. 9. Relative lattice constant b as a function of H^2 for different temperatures within the uniform phase.

magnetic susceptibility. Uniaxial pressure along the a axis leads to a reduction, whereas pressure in c or b direction causes an enhancement of χ. The pressure effects on the susceptibility are fairly large. For example, the increase for uniaxial pressure applied along the b axis displayed in Fig. 10 amounts to about 5%/GPa.

Taking into account that the anisotropy of the susceptibility with respect to the direction of the applied magnetic field only arises from the different g-values, the hydrostatic pressure dependence is also extractable from our data. It is obtained by a (g-value) weighed sum of the uniaxial pressure dependences. The result also shown in Fig. 10 can be compared with measurements of the susceptibility at finite pressure. Such measurements on powdered samples have been reported by Takahashi et al. [29] for a pressure of 1.2 GPa. Consistent with our findings, the susceptibility increases with pressure. Moreover, the pressure induced increase is hardly temperature dependent up to 60 K. From the data of Takahashi et al. [29] a hydrostatic pressure derivative of the susceptibility of about $\frac{\partial\chi}{\partial p} = (5 \pm 1) \cdot 10^{-7}$ emu/g GPa is obtained at $T \simeq 60$ K, if we assume a linear increase of χ as a function of pressure. This value is slightly larger than the value we extract from the magnetostriction, which amounts to $3.6 \cdot 10^{-7}$ emu/g GPa. This might indicate that the susceptibility increases nonlinearly with pressure. Note that in theories of the spin-Peierls transition a linear spin-lattice coupling is assumed. Strong nonlinearities of this coupling, which usually also imply nonlinear pressure dependences of the susceptibility, would question the application of these theories to $CuGeO_3$. However, more experimental studies are necessary to investigate the possibility of a nonlinear pressure dependence of χ. Within the accuracy of the experimental results known so far, one can neither prove nor exclude this nonlinearity. Our data for the limit of vanishing pressure and those at 1.2 GPa have been obtained on different samples. Moreover, the absolute values of the susceptibility in $CuGeO_3$ are strongly affected by defects, whose number may increase under pressure. The latter has to be considered in particular for $CuGeO_3$, since there are indications that the pressure transmitting medium strongly influences the properties measured as a function of pressure [30]. Considering these uncertainties there is a fair agreement between the pressure dependences of the susceptibility calculated from the magnetostriction and that measured at finite pressure. Further studies of the susceptibility and structure as a function of pressure are desirable in order to decide whether a nonlinear spin-lattice coupling affects the spin-Peierls transition in $CuGeO_3$.

Now we turn to the discussion of possible microscopic origins of the magnetostriction in the U phase, i.e. the sources of the anisotropic stress dependences of the magnetic susceptibility. The only reason for a significant pressure dependence of χ in the quasi one-dimensional antiferromagnet $CuGeO_3$ is a pressure dependent magnetic exchange interaction between the Cu^{2+} ions.

Since we observe a finite magnetostriction for all three lattice constants we conclude that this interaction does not only depend on distances along the magnetic chains. Vice versa, the uniaxial pressure dependences found

parallel to the one-dimensional chains, i.e. $\| c$, are much smaller than those parallel to the a and b axis. A finite uniaxial pressure dependence of χ for $p \perp c$ is expected since the electronic structure of $CuGeO_3$ leads to a Cu–O–Cu superexchange path which is not parallel to the c axis (e.g. [10]).

We mention that a smaller anisotropy is revealed, if we consider strain instead of stress dependences of the susceptibility, i.e. when taking into account the anisotropic lattice compressibilities as mentioned above. However, the qualitative behavior, which we will discuss below, remains unchanged. Apart from the limited knowledge of the pressure induced structural changes, a further problem arises from the temperature dependence of the susceptibility of $CuGeO_3$. It is well known that χ does not follow the calculations for one-dimensional Heisenberg chains [1, 10, 9]. In the case of a one-dimensional Heisenberg chain the pressure dependences of χ would directly give the corresponding ones of the magnetic exchange interaction constant (J). In particular, the maximum value of χ (χ_{max}), which is found at about 60 K in $CuGeO_3$, is proportional to $1/J$ i.e. a pressure induced increase of χ_{max} of 5% would correspond to a decrease of J by 5%.

For $CuGeO_3$ this simple correlation does not hold. In particular χ is not a function of a single parameter. There is strong evidence for a significant antiferromagnetic exchange between next nearest neighbors (J') which competes with the nearest neighbor exchange interaction [10, 9]. Moreover, the susceptibility may also be affected by the magnetic coupling perpendicular to the chain direction ($J_\perp$), which amounts to about $0.1J$ along the b axis [31]. Thus, for the maximum value of the susceptibility in $CuGeO_3$ one may assume an expression of the form

$$\chi_{max} = \chi(60\ K) \propto \frac{1}{J} \cdot f\left(\frac{J'}{J}, \frac{J_\perp}{J}\right), \tag{12}$$

where f denotes an unknown function. Since 60 K is significantly larger than $k_B J_\perp$, the influence of the interchain coupling on χ_{max} is weak [10] and will be neglected in the following. From numerical studies of the J–J' model it is found that χ_{max} increases with increasing J' (see e.g. [9, 10]), i.e. with increasing frustration of the intrachain magnetic exchange.

There are two sources of a pressure induced increase of χ_{max}: a decrease of J and/or an increase of J'. According to our data such an increase of χ_{max} occurs as a function of hydrostatic pressure and indeed recent neutron scattering data [32] confirm our qualitative argumentation based on Eq. (12). Neglecting J' Nishi et al. extract a strong decrease of J as a function of pressure from the flattening of the dispersion curve.

Now let us discuss the anisotropic influence of pressure on the magnetic exchange signalled by our measurements. A simple reason for the significant influence of the next nearest neighbor exchange on the magnetic properties of $CuGeO_3$ is a suppressed nearest neighbor exchange constant J. J is e.g. about one order of magnitude smaller in $CuGeO_3$ than in the cuprate superconductors. This small value of J is a consequence of the geometrical arrangement of the Cu and O atoms in $CuGeO_3$ [10, 33, 34]. In particular, the small J can be traced back to the nearly 90 degree Cu–O–Cu bond angle ($\gamma \simeq 98°$) [34] along the one-dimensional chains. It is well known that there is a weak ferromagnetic exchange for a 90° and a much stronger antiferromagnetic exchange for a 180° exchange path (Goodenough-Kanamori-Anderson rules). As a consequence J increases with increasing γ giving rise to a significant spin-lattice coupling. As a function of uniaxial pressure bond angles do change and thus a pronounced and anisotropic pressure dependence of J is expected in $CuGeO_3$. Moreover, there is no reason to assume a similar strong influence of small structural changes on the value of J' (see e.g. the discussion in [10]). Therefore we will only consider the pressure dependence of J in the following qualitative discussion.

Our findings for uniaxial pressure applied parallel to the c and the a axes can be explained very easily in the framework described so far. From the structural arrangement it is apparent that uniaxial pressure along the c axis causes a reduction of the Cu–Cu distance and therefore a reduction of the Cu–O–Cu bond angle. Thus one expects a decrease of the antiferromagnetic coupling and consequently an increase of the magnetic susceptibility. This is confirmed by our data, which reveal a positive uniaxial pressure dependence of χ for $p \| c$. Note that our data imply that J increases with increasing lattice parameter c, i.e. with increasing distance between neighboring Cu atoms. Thus the singlets in the spin-Peierls phase are formed by the pair of Cu atoms whose distance increases at the spin-Peierls transition.

The bond angle γ must also be considered for the pressure effects along the other lattice directions. Both the x and y components of the structural position of the O atoms mediating the superexchange differ from those of the Cu atoms. Therefore one expects an increase of γ for uniaxial pressure applied along both the a and the b axes. Hence uniaxial pressures applied perpendicular to the chains should cause an increase of J and consequently a decrease of χ_{max}. This is found from our data for the a axis, whereas the uniaxial pressure along the b axis leads to the opposite effect.

Hence, the largest uniaxial pressure dependence of χ, which is observed for $p \| b$, deviates from the behavior expected in our simple treatment. We emphasize that a closer inspection of the structure does not yield any plausible reason for a decrease of γ by applying uniaxial pressure along the b axis. Moreover, it is obviously impossible to explain the largest uniaxial pressure dependence via elastic reactions due to the induced changes of the other lattice constants. Thus we conclude that the largest uniaxial pressure dependence of the susceptibility of $CuGeO_3$ can not be traced back to the γ dependence of J alone.

Considering bond distances within the CuO sublattice does not yield any plausible argument for a strong decrease of J as a function of uniaxial pressure along the b axis neither. Structural data show that there is a strong thermal contraction of the lattice constant b, whereas the short CuO bondlength is temperature independent [34]. This strongly indicates that uniaxial pressure along the b axis does not affect the bond distances in the magnetic CuO ribbons significantly. Moreover, it is very difficult to

reconcile the signs and sizes of the magnetostriction along the a and c axes, if one assumes a strong influence of the CuO bondlength on the susceptibility.

Thus, one may conclude that our assumptions concerning the relevance of the different magnetic exchange constants for the pressure dependence of χ are not valid. On the one hand $J_\perp$ may significantly influence the maximum value of χ and determine its pressure dependence, in particular for $p \parallel b$. On the other hand there may be a strong pressure dependence of J'. At present we can not exclude these possibilities. However, note that the magnetostriction measures an important precondition of the spin-Peierls transition, i.e. the spin-lattice coupling. Therefore both interpretations of the large magnetostriction of the b axis in the U phase would imply that an additional spin-phonon coupling, which takes into account the strain dependence of J' or $J_\perp$, has to be incorporated in a realistic theory of the spin-Peierls transition in $CuGeO_3$.

However, there remains a possibility to explain the large magnetostriction of the b axis neglecting the pressure dependences of $J_\perp$ and J' completely as we have done for the two other lattice directions. One has to assume that J also depends on the structure of the GeO sublattice or, more precisely, its bonding to the magnetic CuO ribbons. This possibility was discussed recently in a theoretical work by Geertsma et al. [33]. In this work it is argued that side groups may alter the Goodenough-Kanamori-Anderson rules and therefore make a 90° superexchange antiferromagnetic. In $CuGeO_3$ there are covalent bonds between the O atoms in the magnetic chains and the surrounding Ge atoms. The calculations of Geertsma et al. yield that the magnetic exchange between neighboring Cu atoms depends on the hybridization of the Ge and O orbitals, since the usual antiferromagnetic superexchange is suppressed due to nearly 90° Cu–O–Cu bond angle. Unfortunately, both the structural changes as a function of uniaxial pressure as well as the quantitative influence of Ge–O bond length and Cu–O–Ge bond angle on J are not known precisely at present. Qualitatively, an influence of pressure along the b axis on the Ge–O bond, which is relevant in the calculations of Geertsma et al., can be inferred from the temperature dependent change of the structure [34]. Model calculations are in progress to study whether a pressure dependent hybridization of Ge and O orbitals does explain the large uniaxial pressure dependence of χ for $p \parallel b$.

V. Conclusions

We have presented measurements of the isothermal, longitudinal magnetostriction of the spin-Peierls cuprate $CuGeO_3$ along the three orthorhombic directions for magnetic fields up to 14 T. The magnetostriction of $CuGeO_3$ below T_{SP}^0 is determined by the anisotropic strain order parameter coupling, which is introduced to describe the spontaneous strains in the dimerized phase. For low temperatures the magnetostriction and thus the field dependence of the spin-Peierls order parameter of the dimerized phase is very small. However, large and strongly anisotropic anomalies of the lattice constants occur at both kinds of field induced transitions, the dimerized/incommensurate phase transition below 11 K and the dimerized/uniform transition for $11\,\mathrm{K} < T < T_{SP}^0 = 14.3\,\mathrm{K}$. The jumplike behavior of the magnetostriction at the D/I transition indicates a first-order character of this phase transition. This is confirmed by a hysteretic behavior of the lattice constants present at the entire D/I phase boundary. The jumps of the magnetostriction at the D/I transition reflect a pronounced decrease of the spontaneous strains at this phase transition. Finite spontaneous strains are also present in the incommensurate phase. The anisotropy of these strains is similar to that found in the dimerized phase.

Precise H-T phase diagrams for fields oriented parallel to all lattice constants are presented, which clearly reflect the anisotropic g-values. In terms of reduced variables the magnetic phase diagram is in fair agreement with the theory of Cross, though systematic deviations from the predictions are also revealed.

From the jumps of the magnetostriction at the D/I boundary we estimate considerable, uniaxial stress dependences of the critical field H_c at this transition. These pressure dependences correlate with the uniaxial pressure dependences of the spin-Peierls transition temperature T_{SP}^0. For $p \parallel b$ and $p \parallel c$ uniaxial pressure stabilizes the dimerized phase, whereas $p \parallel a$ destabilizes the D phase. Within the present accuracy, the data are consistent with the theoretically expected pressure independent H-T phase diagram in reduced variables, which is determined by T_{SP}^0 only.

A finite, strongly anisotropic magnetostriction could also be resolved well above the spin-Peierls transition temperature. These data show the presence of a pronounced spin-lattice coupling in $CuGeO_3$, which is an important precondition for the occurrence of a spin-Peierls transition. The magnetostriction in the uniform phase is proportional to H^2 as expected for the leading order of an expansion of the free energy.

Using thermodynamic relations considerable uniaxial pressure dependences of the magnetic susceptibility in the uniform phase are determined from the magnetostriction. They amount to several %/GPa and differ in sign and size for the three lattice directions. Our findings indicate that the magnetic exchange in $CuGeO_3$ depends rather on bond angles than on bond distances. Based on our data we have presented a qualitative description of the spin-lattice coupling in $CuGeO_3$. The decrease of J with increasing lattice constant c as well as the opposite effect along the a axis can be easily traced back to a Cu–O–Cu bond angle dependence of the nearest neighbor magnetic superexchange along the one-dimensional chains. However, the largest uniaxial pressure dependence of the magnetic susceptibility, which we observe for $p \parallel b$, has another origin. A pressure dependent infuence of side groups on the nearest neighbor exchange is the most plausible way to interpret our findings for $p \parallel b$. However, at present we cannot exclude the relevance of a spin-lattice coupling due to strain dependences of other magnetic exchange constants, i.e. the interchain coupling or the next nearest neighbor exchange.

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