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MAGNETOSTRICTION OF THE SPIN-PEIERLS COMPOUND $CuGeO_3$

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The longitudinal and transverse magnetostriction, λ, of the spin-Peierls compound $CuGeO_3$ in the $b-c$-plane was measured in magnetic fields up to 20 T both above and below the transition temperature $T_{sp} = 14.3$ K. It was found that for a given crystallographic direction the value of magnetostriction is weakly dependent on the magnetic field direction. In the uniform U-phase at $T \geq T_{sp}$, λ was negative and approximately equal in the b and c directions, while in the dimerized D-phase at $T < T_{sp}$, λ was positive, and $\lambda_b > \lambda_c$. At low temperatures, λ sharply increased at the magnetic field induced transition from dimerized to magnetic M-phase. Experimental data allow estimation of the stress derivatives of the antiferromagnetic intrachain exchange interaction parameter and of the stress dependencies of the critical field of the $D-M$-phase transition.

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The cuprate, $CuGeO_3$, possessing an orthorhombic crystal structure [1] is considered to be the first inorganic compound experiencing the spin-Peierls transition [1]. It is essential that the crystal structure of $CuGeO_3$ be formed by Cu-O and Ge-O crystallographic elements aligned in the c direction, as shown in Fig. 1. Thus, this structure contains one-dimensional chains of Cu^{2+} ions located at the center of edge sharing rectangles of oxygen atoms. These chains are separated by rigid chains of GeO_4 tetrahedra. Between the Cu^{2+} ions in a chain, a strong antiferromagnetic superexchange Cu^{2+}-O^{2-}- Cu^{2+} interaction occurs. The antiferromagnetic intrachain exchange parameter is estimated in different reports to be $J_c \approx 120-180$ K [3, 4], while the ratios of interchain exchange parameters are: $J_b = 0.1 J_c$, and $J_a = -0.01 J_c$ [3].

At the spin-Peierls transition, the uniform antiferromagnetic quantum chain (U-phase) becomes unstable due to an underlying lattice distortion which causes quasi one-dimensional dimerization of the chain (D-phase). Below the transition temperature, an energy gap opens between a singlet ground state and a band of triplet excited states [3].

The spin-Peierls transition temperature in $CuGeO_3$ $T_{sp} = 14.3$ K [1]. The transition at T_{sp} is a second-order phase transition and is accompanied by anomalies in specific heat [5], elastic moduli [6], magnetic susceptibility [1], etc. Neutron diffraction measurements indicate that below T_{sp} the lattice periods double in the a and c directions due to alteration of Cu^{2+} interionic distances [7]. The lattice deformation at T_{sp} is accompanied by anisotropic anomalies of thermal expansion

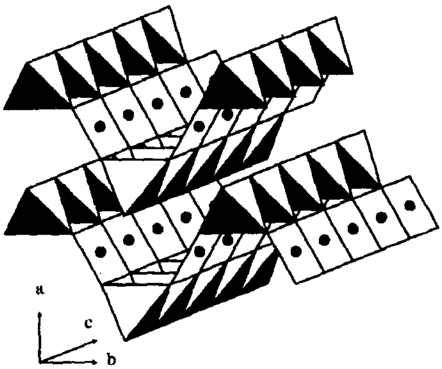

Fig.1. The crystal structure of $CuGeO_3$. The black circles represent the Cu^{2+} ions. The O^{2-} ions are situated at the rectangles corners and corners of tetrahedra. The Ge^{4+} ions are situated at the centers of tetrahedra

coefficients [8]. Application of a magnetic field results in stepwise magnetization along all axes [9] indicating phase transitions at H_t from the dimerized D-phase to either the uniform U-phase at $T > 0.7 \div 0.8\ T_{sp}$, or to the magnetic incommensurate M-phase at $T < 0.7 \div 0.8\ T_{sp}$ [10].

Measurements of magnetostriction provide information regarding the role of magnetoelastic interactions and, in particular, allow estimation of the influence of these interactions on magnetic-field induced structural transformations in $CuGeO_3$. This report presents observations of longitudinal $\lambda_{\parallel}$ and transverse $\lambda_{\perp}$ magnetostriction in single crystals of $CuGeO_3$ in the $b-c$-plane. The samples studied were grown by two different techniques, i.e., either from stoichiometric melts using a floating zone technique associated with an image furnace, or by using a slow cooling technique with melts containing excess Cu oxide. Both longitudinal $\lambda_{\parallel}$ and transverse $\lambda_{\perp}$ magnetostriction of single crystals of $CuGeO_3$ along the b and c axes was measured between 4 and 14 K in pulsed magnetic fields up to 20 T using a quartz platelet piezosensor glued to the samples [11]. The accuracy in determination of the absolute value of magnetostriction was about 15%. The results obtained in samples of different origin coincide within this experimental error. The data presented below were obtained on samples grown by slow cooling technique.

The field dependencies of longitudinal $\lambda_{\parallel}$ and transverse $\lambda_{\perp}$ magnetostriction measured in the b and c crystallographic directions are shown in Fig. 2 and 3. It appears that $\lambda_{\parallel}$ and $\lambda_{\perp}$ in the b and c directions are negative above T_{sp} and positive below this temperature. At $T < T_{sp}$ the field dependencies of magnetostriction show peculiarities due to field induced transitions from the D-phase into either the M-phase or U-phase. These transitions are abrupt at low temperature and gradual at higher temperature. The critical fields H_t measured were practically the same as those found by magnetization measurements [9]. Some decrease of magnetostriction was seen at $H > H_t$.

The data obtained show that for a given crystallographic direction the values of longitudinal $\lambda_{\parallel}$ and transverse $\lambda_{\perp}$ magnetostriction are of the same sign and of approximately same value. The difference to some extend is attributable to anisotropy of the gyromagnetic factor, $g_b = 2.27$ and $g_c = 2.07$ [12]. The values of magnetostriction measured in the b and c directions differ in the D-phase but are close to each other in the U-phase.

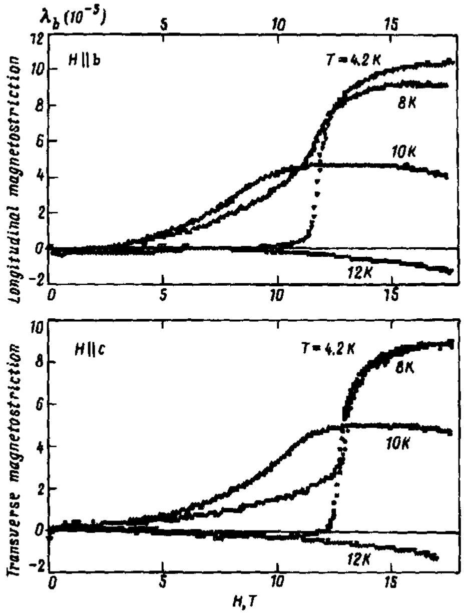

Fig.2. The field dependencies of the longitudinal $\lambda_{\|}$ and transverse $\lambda_{\perp}$ magnetosriction of spin-Peierls compound $CuGeO_3$ along the b axis.

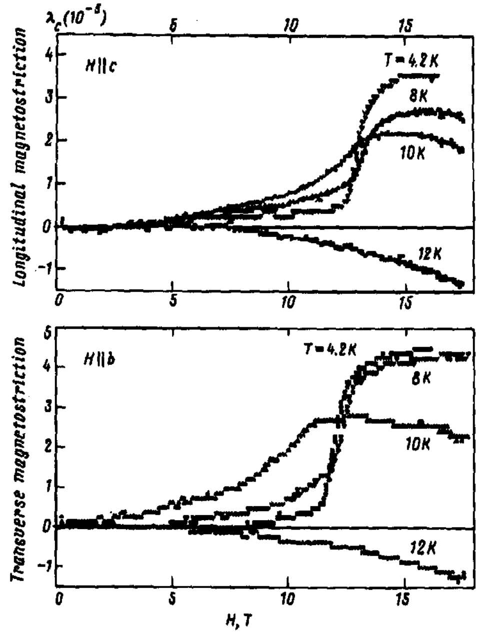

Fig.3 The field dependencies of the longitudinal $\lambda_{\|}$ and transverse $\lambda_{\perp}$ magnetosriction of spin-Peierls compound $CuGeO_3$ along the c axis

At $T = 4.2$ K the magnetic field induced strains at the $D - M$ transition are $\Delta\lambda_b^{\|} = 10.3 \cdot 10^{-5}$, $\Delta\lambda_b^{\perp} = 8.8 \cdot 10^{-5}$ and $\Delta\lambda_c^{\|} = 3.1 \cdot 10^{-5}$, $\Delta\lambda_c^{\perp} = 4.2 \cdot 10^{-5}$. These values are close to spin- Peierls spontaneous strains previously observed at this temperature: $\varepsilon_a = 5.0 \cdot 10^{-5}$, $\varepsilon_b = -7.3 \cdot 10^{-5}$, and $\varepsilon_c = -1.7 \cdot 10^{-5}$ [8].

The thermodynamic approach based on the Clapeyron-Clausius equation allows evaluation of the stress derivative of the critical magnetic field H_t of the $D - M$ first order transition. It can be written as:

$$dH_t/d\sigma_i = -V\Delta\lambda_i/\Delta m_i \qquad (1)$$

where σ_i is the stress, V is the lattice unit per Cu atom, and $\Delta\lambda_i$ and Δm_i are the jumps of magnetostriction and magnetization at the $D - M$ transition in the i-direction. The longitudinal magnetostriction data combined with the results of magnetization measurements [9] show that $dH_t/d\sigma_b \approx -28$ T/GPa and $dH_t/d\sigma_c \approx -9$ T/GPa at $T = 4.2$ K. The transverse magnetostriction data give $dH_t/d\sigma_b \approx -26$ T/GPa and $dH_t/d\sigma_c \approx -11$ T/GPa. Note, that negative stress derivatives of H_t correspond to positive pressure dependence of this field.

The same approach applied to results of magnetostriction measurements in the paramagnetic U-phase allows to evaluate the stress derivative of the antiferromagnetic exchange interaction parameter J_c.

$$\frac{dJ_c}{d\sigma_i} = -\frac{V\lambda_i}{(m_i/m_s)^2} \qquad (2)$$

where m_i is the magnetization in a given crystallographic direction, and m_s is the magnetization of saturation.

The measurements give that at $T \geq T_{sp}$ the magnetostriction is proportional to the square of magnetization. Our data show that $dJ_c/d\sigma_b = 14$ K/GPa and $dJ_c/d\sigma_c = 23$ K/GPa

In summary, magnetostriction of the spin-Peierls compound $CuGeO_3$ was measured. The magnitude of strains at the magnetic-field-induced $D-M$ transition were comparable with spontaneous thermal strains accompanying the $U-D$ transition. The stress derivatives of the critical magnetic field of the $D-M$ transition and of interaction exchange parameter were determined. To describe quantitatively the magnetoelastic phenomena in spin-Peierls compounds an appropriate theoretical approach should be developed. At $T < T_{sp}$, Cu^{2+} ions couple in dimers. Two exchange integrals come into play in the c direction in the D-phase, and the simple approach used above is untenable. The M-phase also possesses a complicated structure incommensurate with an underlying crystal lattice thus also preventing application of that approach to analysis of magnetostriction data.

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