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Uniaxial Pressure Dependence of Spin-Peierls Transition Temperature in $CuGeO_3$

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Neutron diffraction experiments were performed applying uniaxial pressure along the *a*-axis of a single crystal of $CuGeO_3$. We observed an overall decrease of the spin-Peierls transition temperature (T_{sp}) with increasing uniaxial pressure with dT_{sp}/dp= -5.0 K/GPa in contrast to the application of hydrostatic pressure, where an increase of T_{sp} was observed. The uniaxial pressure dependencies of T_{sp} were estimated along each principal axis by Winkelmann *et al.* from the thermal expansion coefficients and the specific heat via the Ehrenfest relation. According to these results the uniaxial pressure along the *a*-axis strongly suppresses T_{sp} with dT_{sp}/dp = -4.2 K/Gpa. Our experimental result is consistent with the estimation within error.

KEYWORDS: spin-Peierls, one-dimensional magnetism, $CuGeO_3$, neutron scattering, pressure

§1. Introduction

The spin-Peierls (SP) transition of inorganic compound $CuGeO_3$ was discovered in magnetic susceptibility measurements by Hase *et al.*[1)] in 1993. Dynamical magnetic excitation was observed with neutron inelastic scattering[2)] and lattice dimerization was observed by x-ray and neutron diffraction[3)] and electron diffraction.[4)] Many experimental studies were performed but microscopic spin-lattice coupling mechanism is not yet fully clarified until now. The crystal structure of $CuGeO_3$ is orthorhombic and group symmetry is P_{bmm} at room temperature (RT). A pair of CuO_2 chains and GeO chains along the *c*-axis are in the unit cell. Above spin-Peierls transition temperature (T_{sp}) with decreasing temperature lattice constant *a* and *b* are contracting but *c* is elongated, and the absolute temperature variation of *b* is especially very large.

The anisotropic thermal expansion coefficients α_i (i = 1, 2, 3) of $CuGeO_3$ were measured along the each principal axis and the extremely anomalous temperature dependences of them were observed by Winkelmann *et al.*[5)] Sharp jump of them, $\Delta\alpha_i$, observed at T_{sp} that accompanying the sudden contraction of *b* and *c* but an elongation of *a*. These phenomena at T_{sp} were analyzed with the specific heat (C_p) via the Ehrenfest's relation at the second order phase transition in thermodynamics, which is $dT_{sp}/dp = VT_{sp}\Delta\alpha/\Delta C_p$ where p is pressure and V is unit volume. The uniaxial pressure dependence of T_{sp} is calculated from the thermal expansion coefficients along the each principal axis. They predict the increase of T_{sp} under hydrostatic pressure to be 4.5 K/GPa with ΔC_p = 1.62 J/mole·K.[5)] Indeed in the measurement of T_{sp} by neutron diffraction under the hydrostatic pressure the value of 4.5 K/GPa is observed by Katano *et al.*[6)] If the uniaxial pressure is loaded along the *a*-axis direction, the abrupt suppression of T_{sp} is expected with dT_{sp}/dp = -4.2 K/GPa. It may be possible that SP phase disappears and another magnetic phase would appear if the uniaxial pressure along the *a*-direction is beyond 3.4 GPa.

§2. Experiments

The $CuGeO_3$ single crystal was grown along the *a*-axis by floating zone method (FZ) which size is 5 mm diameter and 7 mm high. $CuGeO_3$ has a cleavage surface including *b* and *c*-axes. A single crystal was pressed along the *a*-axis using aluminum pistons and the three kinds of springs with different spring constants (2.6, 10.2, 128 kgf/mm) to keep the pressure constant. The strength of the uniaxial press was determined by the contraction length of the spring. The sample pressure cell is made from 40 mm thick Al metal transmitting the neutron beam. It was set on the cold tip of 4K-refrigerator of a helium-gas circulation type and was cooled down to 4 K.

Neutron diffraction experiments were performed with (h, k, h) scattering plane inclined the *a*-axis about 31 degrees toward the *c*-axis on a high Q resolution spectrometer ISSP-HQR with two-axis mode installed in the guide hall (T1-1 port) of the JRR-3M at JAERI in Japan. Other experimental conditions were as horizontal collimators to Guide tube (15') - open (20') - 40' and the incident neutron wavelength 2.45 Å with the monochromator of Pyrolytic graphite (PG) (002) reflection. A PG filter was set before the sample to remove higher order contaminations. A temperature sensor of Si-diode was put in 3 mm diameter hole in an Al block within 10 mm distance from the sample. The temperature of the sample was controlled accurately within 0.01 degree by heater wound around the pressure cell and by adjusting the Joule-Thomson valve. Measuring time was 5 minutes and waiting time to obtain the thermal equilibrium also 5 minutes at each temperature.

§3. Results and Discussions

The unit cell of the crystal structure below T_{sp} is ($2a$, b, $2c$) where (a, b, c) are the lattice constants of $CuGeO_3$

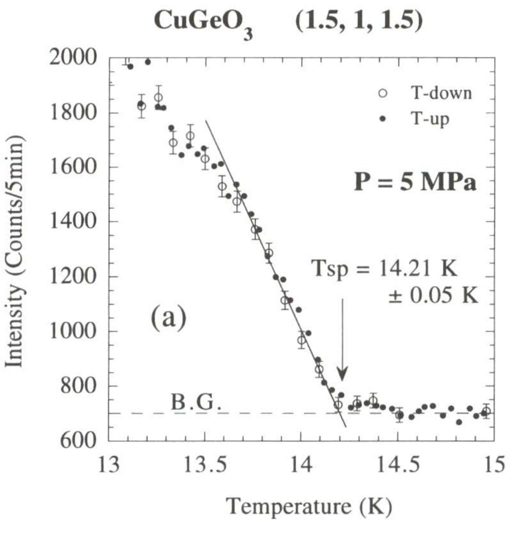

Fig. 1. The temperature dependence of the peak intensity of (1.5, 1, 1.5) superlattice peak under uniaxial pressure. The spin-Peierls transition temperatures are determined to 14.21 ± 0.05 K and 13.55 ± 0.05 K at the pressure of 5 MPa and 128 MPa, respectively. The measurements at decreasing temperature are represented by open circles and squares, and these at increasing temperature are done by closed circles and triangles. The measurements of the temperature dependence were repeated several times.

at RT. The temperature dependence of the peak intensity of superlattice reflection (1.5, 1, 1.5) applying the uniaxial pressure $p = 5$ MPa and 128 MPa along the *a*-axis are shown in Fig. 1. Background intensity was estimated in averaging the counting rate above T_{sp}. The thermal hysteresis between the increasing and decreasing temperature is not recognized within 0.05 K near T_{sp} at both processes. T_{sp} was determined at crossing point between the constant background line and the fitted line to the increasing intensity with decreasing temperature. The error of Tsp was determined as the allowance at crossing points of line with several kinds of slope. T_{sp} shifts to 14.21 (5) K and 13.55 (5) K at the pressure of 5 MPa and 128 MPa, respectively, where T_{sp} is 14.06 (10) K at ambient pressure. The uniaxial pressure dependence of T_{sp} is plotted in Fig. 2. When a linear fit was applied to the whole experimental data, the uniaxial pressure dependent coefficient was determined to -5.0 (8) K/GPa as

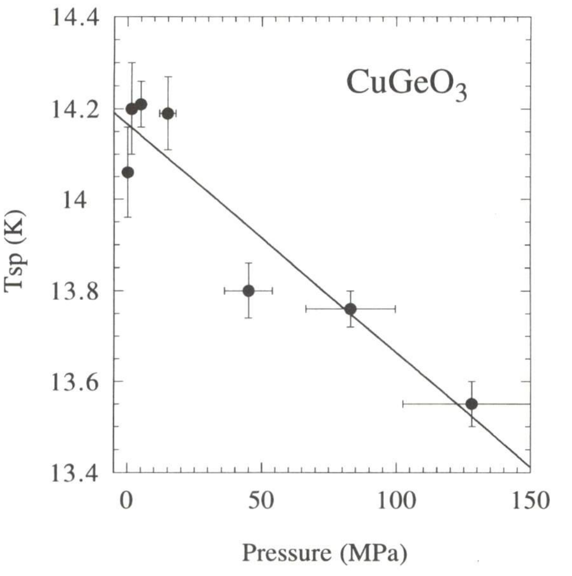

Fig. 2. The spin-Peierls transition temperature versus the uniaxial pressure along the *a*-axis . Solid line is a linear fit with the coefficient -5.0 ± 0.8 K/GPa.

shown by solid line in Fig. 2. This experimental value is consistent with the calculated value -4.2 (5) K/GPa on the bases of Ehrenfest's relation. The studies of doping effects on the SP state were reported with many kinds of impurity atoms, Zn, Mg, Ni, Si, etc. [7-9] T_{sp} of $CuGeO_3$ decreases abruptly with increasing doping concentration because the impurity atoms affect to disturb not only the lattice dimerization but the spin singlet formation locally. In contrast to the impurity doping the pressure gives homogeneous effect to the lattice.

Three data points below 20 MPa seem to indicate an initial increase of T_{sp} from the ambient pressure value. However, these statistics is not enough to discuss. We need more detailed experiment in the near future.

§4. Summary

The suppression of T_{sp} in $CuGeO_3$ under uniaxial pressure along the *a*-axis is observed by neutron diffraction. The experimentally determined coefficient -5.0 (8) K/GPa is consistent with the coefficient -4.2 (5) K/GPa derived from the analysis using the Ehrenfest's relation. Our observation is the first evidence of the decrease of T_{sp} in $CuGeO_3$ under uniaxial pressure.

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