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Uniaxial-Pressure-Induced Release of Magnetic Frustration in a Triangular Lattice Antiferromagnet YbCuGe

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Abstract: We have studied the effect of geometrical frustration on the antiferromagnetic order in the Yb-based triangular lattice compound YbCuGe below $T_N = 4.2$ K by the measurements of magnetization and specific heat under hydrostatic and uniaxial pressures. By applying hydrostatic pressure P up to 1.34 GPa, T_N hardly changes. By contrast, T_N increases as P is applied along the hexagonal a axis, while T_N decreases by the application of P along the c axis. The increase of T_N only for $P \parallel a$ suggests the release of the frustration inherent in the triangular lattice of Yb ions of this compound.

Keywords: uniaxial pressure; antiferromagnetic order; magnetic frustration; Yb compound; magnetization; specific heat



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1. Introduction

Cerium (Ce)-based and Ytterbium (Yb)-based intermetallic compounds have a great deal of interest in the field of magnetism because they exhibit a variety of physical behaviors, such as an anomalous magnetic ordering, superconductivity, and non-Fermi-liquid behavior due to the competition between the Kondo effect and the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction [1,2]. The competition can be controlled easily by applying pressure. In fact, such unusual behaviors have been realized in many of Ce and Yb compounds under pressure [2–6].

On the other hand, magnetic properties of compounds with geometrically frustrated lattices, such as triangular, honeycomb, kagome, and pyrochlore lattices, have received considerable attention from both theorists and experimentalists [7,8]. The geometrical frustrations give rise to a number of unusual behaviors, such as suppression of magnetic ordering temperatures, multiple-phase transitions, complex magnetic structures, and spin liquid states [7,8]. In rare-earth-based intermetallic compounds with frustrated lattices, the long-range nature of the RKKY interaction has been believed to overcome the effect of frustration. However, the interplay among frustrated anisotropic exchange interactions, crystalline electric field (CEF) anisotropy, and Kondo effect arises the partial disordered magnetic state, multiple-phase transition, and unusual heavy-fermion state in Ce- and Yb compounds with the quasikagome lattices, CePdAl [9], CeRhSn [10,11], CeIrSn [12], and YbAgGe [13] as well as that with the honeycomb lattice CePt₆Al₃ [14]. For example, in CePdAl below $T_N = 2.7$ K, one-third of Ce ions remain in a paramagnetic state due to the geometrical frustration of the quasikagome lattice [9]. Another example is a heavy-fermion compound CePt₆Al₃ in which Ce atoms form a honeycomb lattice in a trigonal basal plane [13]. Theoretically, a quantum spin liquid state is expected to be realized in

the stuffed honeycomb lattice that interpolates between the triangular and honeycomb lattices [15,16].

We focus on YbCuGe as a candidate of a frustrated Yb-based intermetallic compound. This compound crystallizes in the hexagonal NdPtSb-type structure in which Yb atoms form a triangular lattice in the c plane, as shown in Figure 1 [17]. The magnetic susceptibility $\chi = M/B$ above 100 K for both $B \parallel c$ and $B \parallel a$ follows the Curie-Weiss law expected for the trivalent Yb ion [18]. The easy magnetization direction along the c axis can be explained by the CEF effect on a single Yb ion. The broad maximum of the magnetic specific heat at 50 K is reproduced as a Schottky anomaly by the CEF model [18]. This compound shows an antiferromagnetic order at $T_N = 4.2$ K, where a sharp peak appears in the specific heat $C(T)$ and a shoulder in χ_a and a hump in χ_c . The Sommerfeld coefficient is as small as 5.2 mJ/K²mol [18], suggesting the Kondo effect to be very weak. However, $C(T)$ shows a broad tail above T_N and the magnetic entropy at T_N is 28% of $R \ln 2$. These results hint to the effect of a magnetic frustration in the triangular lattice of trivalent Yb ions in this compound. In this study, to reveal the magnetic frustration, we have measured the magnetic susceptibility and specific heat under uniaxial and hydrostatic pressures P . When P is applied along the a axis, the magnetic frustration should be released due to the distortion of the regular triangle of Yb ions in the c plane, while the frustration remains under the uniaxial pressure of $P \parallel c$ and hydrostatic pressure, which keep the triangle equilateral. The comparison of the results under the uniaxial and hydrostatic pressures will be presented and discussed.

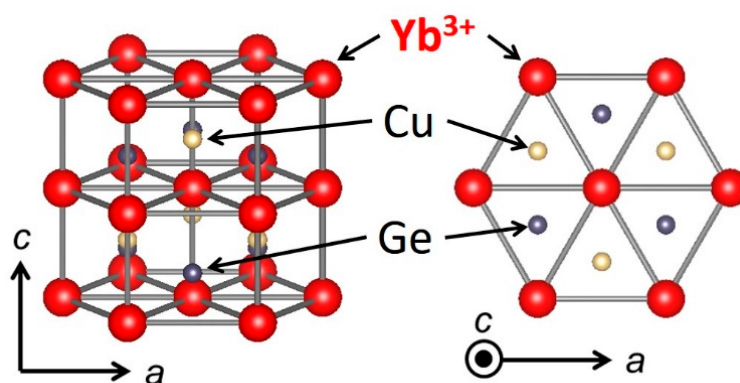


Figure 1. Crystal structure of YbCuGe with a triangular lattice of Yb atoms in the c plane of the hexagonal NdPtSb-type structure.

2. Experimental Procedures

Single-crystalline samples of YbCuGe were prepared by the Bridgman method in a sealed tungsten crucible, as reported previously [18]. The crystal structure of YbCuGe has been confirmed by X-ray diffraction measurements using a single crystal and powder samples, as reported in the previous paper [17]. The X-ray powder diffraction measurement on the powdered single crystals detected no impurity phases [18]. The crystals were oriented by the Laue method and shaped by the spark erosion for the measurements of magnetic susceptibility $\chi = M/B$ and specific heat C . At hydrostatic and uniaxial pressures, χ was measured by using a commercial superconducting quantum interference device magnetometer (Quantum Design, MPMS, San Diego, CA, USA). For hydrostatic pressures up to 1.34 GPa, a piston-cylinder pressure cell made of NiCrAl alloy and Daphne oil 7373 as a pressure transmitting medium were used. For uniaxial pressure of the configurations $P \parallel B$ and $P \perp B$, two type of pressure cells were constructed [19]. Uniaxial pressures were applied on a sample plate of 0.4–0.7 mm in thickness by a homemade pressure cell made of ZrO₂. The applied pressure was determined by measuring the superconducting transition temperature T_c of a piece of tin [20], which was placed at the lower part of the pressure cell.

The C under uniaxial pressures was measured up to 0.45 GPa by using a home-made ac calorimeter in the temperature range of 0.5 to 10 K [21]. Figure 2 shows the pressure cell and sample setting for the ac calorimeter under uniaxial pressures [22]. Thereby, we used disk-shape samples of 2 mm in diameter and 0.5 mm in thickness. Two RuO₂ chip resistors as a thermometer and a heater, respectively, were mounted on the fringe of the Cu-Be plate. To thermally isolate the Cu-Be plate and sample from the ZrO₂ piston, diamond powder of a 0.25- μ m diameter was placed on the top of the piston. The sample of YbCuGe and the sample of In metal used as a pressure gauge are placed in the cell in the vertical direction, as shown in Figure 2. Therefore, the pressure for the sample should be equal to that for the In metal. The pressure for the In metal was determined from the pressure dependence of the superconducting transition temperature of In metal [23]. The superconducting transition was measured by the diamagnetic signal of ac magnetic susceptibility.

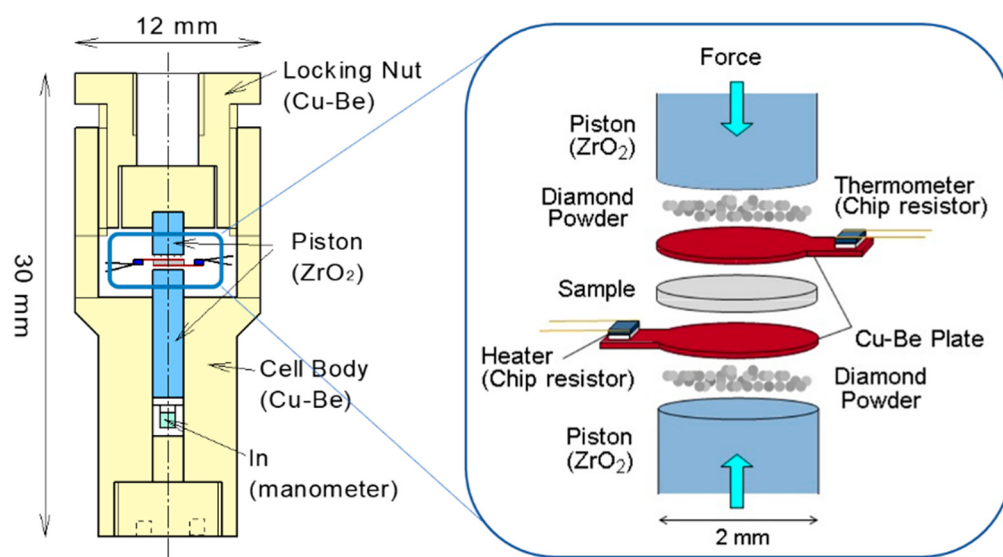


Figure 2. Schematic drawing of the ac calorimeter under uniaxial pressure.

3. Results and Discussion

Figure 3 shows the temperature dependence of $\chi = M/B$ of YbCuGe under hydrostatic and uniaxial pressures. The maximal temperature of $\chi(T)$ for Figure 3a was taken as the Néel temperature T_N . The value of T_N for Figure 3b–d was taken as the temperature where the two lines extrapolated from above and below the kink intersect. For hydrostatic pressure, T_N hardly changes, while T_N under uniaxial pressure displays opposite variations, a decrease for $P \parallel c$, and an increase for $P \parallel a$. The increase of T_N only for $P \parallel a$ suggests the release of the frustration inherent in the triangular lattice of Yb ions of this compound. The variations of $T_N(P)/T_N(0)$ for hydrostatic and uniaxial pressures are plotted in Figure 5, which will be discussed later. Note that the value of χ increases with increasing hydrostatic pressure, while χ decreases for uniaxial pressures both $P \parallel c$ and $P \parallel a$.

To confirm the opposite variations of T_N for $P \parallel c$ and $P \parallel a$, we have measured the specific heat under uniaxial pressures. As shown in Figure 4a, with increasing $P \parallel c$, the peak of the specific heat C divided by temperature C/T becomes broader and the peak temperature shifts to a lower temperature range. For $P \parallel a$ in Figure 4b, on the contrary, the peak temperature shifts to a higher temperature range. The T_N was determined by considering the entropy balance above and below T_N , as shown in Figure 4.

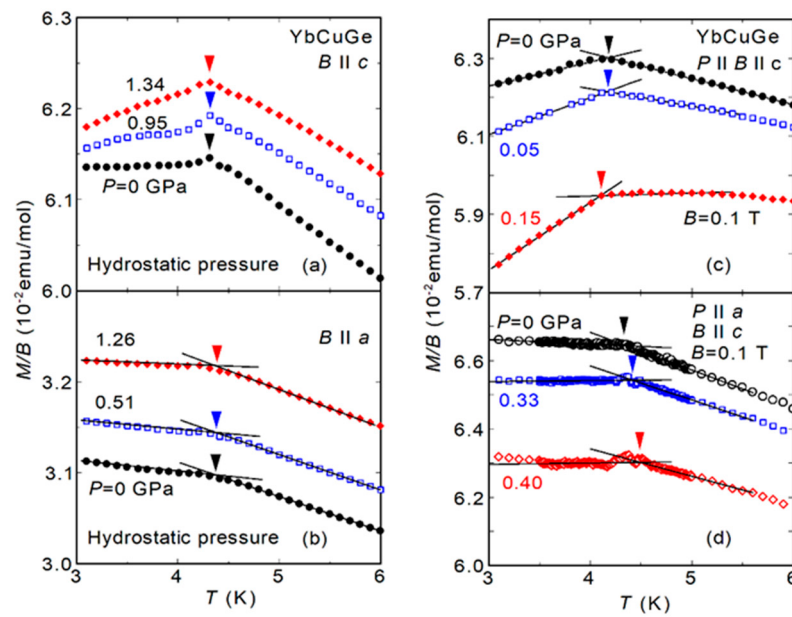


Figure 3. Temperature dependence of the magnetic susceptibility M/B of YbCuGe under hydrostatic pressures (a,b) and uniaxial pressures (c,d). The data at 0.33 GPa are shifted downward by 0.1×10^{-2} emu/mol to avoid the overlap with the data for 0 GPa. The arrows indicate the antiferromagnetic ordering temperature T_N , where T_N in (b–d) is taken as the intersection of the two lines above and below T_N .

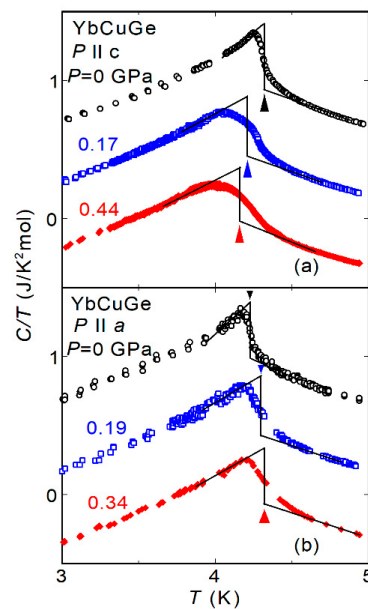


Figure 4. Temperature dependence of the specific heat divided by temperature C/T of YbCuGe under uniaxial pressures applied along the c axis (a) and a axis (b). The arrows indicate the antiferromagnetic ordering temperature T_N . The data sets at various pressures are shifted downward by $0.5 J/K^2 \cdot mol$ for clarity.

The uniaxial pressure dependences of T_N are compared in Figure 5 together with that for hydrostatic pressure. We plot the values of T_N normalized by the value at $P = 0$ because T_N ($P = 0$) depends slightly on the samples. Now, we will discuss the pressure dependence of T_N in terms of the change only in the exchange interaction J between Yb ions with pressures. Thereby, a change in the Kondo effect is neglected because the Kondo effect is

sufficiently weak even at an ambient pressure, as mentioned above. At first, under both hydrostatic pressure and $P \parallel c$, the equilateral nature of the triangle should hold. However, T_N for hydrostatic pressure hardly changes while T_N decreases for $P \parallel c$. Under hydrostatic pressure, the shrinkage along both a and c axes may enhance both J_a along the a -axis and J_c along the c -axis as manifested in the increment of χ , which is shown in Figure 3a,b. However, the enhancement of J_a may strengthen the magnetic frustration effect. Therefore, the unchanged T_N for the hydrostatic pressure may result from the compensation between the two effects of enhanced exchange interaction and strengthened magnetic frustration. On the other hand, for $P \parallel c$, the lattice expands along the a -axis, as shown in the inset of Figure 5, while it shrinks along the c -axis. Therefore, it would be expected that J_a decreases while J_c increases. However, the decrease of the value of χ as shown in Figure 3c suggests that the net value of J decreases with increasing $P \parallel c$. Therefore, the suppression of J may lead to the decrease in T_N .

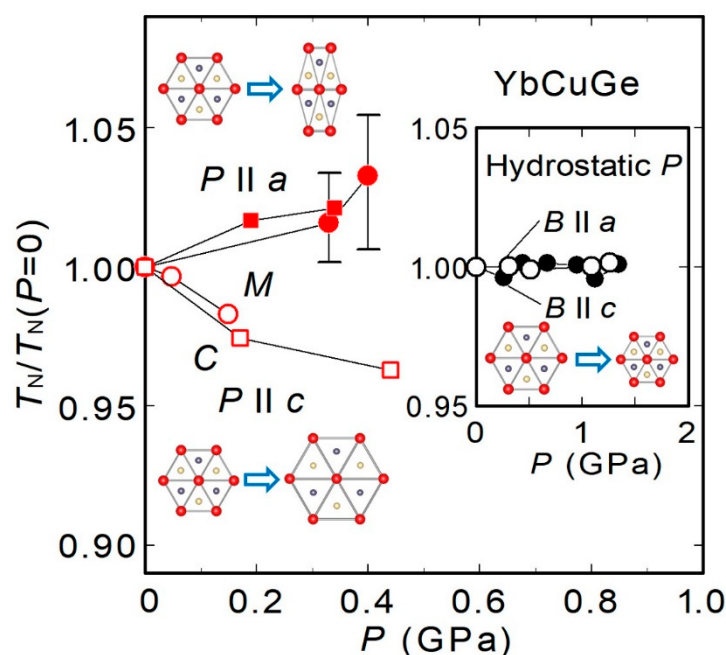


Figure 5. Uniaxial pressure dependence of the antiferromagnetic ordering temperature T_N of YbCuGe, where the data of T_N are normalized by the value at ambient pressure. The inset shows the hydrostatic pressure dependence of T_N . The circles and squares denote the data obtained from the specific heat C and magnetization M measurements, respectively. The schematic changes in the triangular configuration in the hexagonal c plane under uniaxial and hydrostatic pressures are drawn in the main frame and inset, respectively. The balls in red, yellow, and black denote the Yb, Cu, and Ge atoms, respectively.

For $P \parallel a$, the regular triangle in the c plane would be distorted, as shown in the inset of Figure 5. In this case, the Yb-Yb distance parallel to the pressure decreases but that nonparallel to the pressure in the c plane increases. This makes it difficult to guess how the value of J_a would change. However, for $P \parallel a$, the value of χ decreases for $B \parallel c$ (Figure 3d) as well as for $B \parallel a$ (not shown). This fact suggests that the net value of J_a decreases for $P \parallel a$. Nevertheless, T_N for $P \parallel a$ increases, which should result from the release of the magnetic frustration in the triangular lattice of this compound. A similar release of the frustration under uniaxial pressure has been found in an Fe-based triangular lattice system [24]. To confirm this conjecture on YbCuGe, neutron diffraction measurements under pressures are highly desirable.

4. Conclusions

To reveal the effect of magnetic frustration in the Yb-triangular-lattice antiferromagnet YbCuGe with $T_N = 4.2$ K, we have studied the response of the magnetic susceptibility and specific heat to uniaxial and hydrostatic pressures. It is found that T_N increases by the application of P only along the a -axis, which violates the symmetry of the triangular lattice. This finding indicates the release of the frustration inherent in the triangular lattice of Yb ions of this compound.

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