Elastic Softening in the Metallic Antiferromagnet YbCuGe

X J Xi[1], I Ishii[1], Y Noguchi[1], H Goto[1], S Kamikawa[1], K Araki[2], K Katoh[2] and T Suzuki[1,3,4]

[1]Department of Quantum Matter, ADSM, Hiroshima University, Higashihiroshima, Hiroshima 739-8530, Japan
[2]Department of Applied Physics, National Defense Academy, Yokosuka, Kanagawa 239-8686, Japan
[3]Institute for Advanced Materials Research, Hiroshima University, Higashihiroshima, Hiroshima 739-8530, Japan
[4]Cryogenics and Instrumental Analysis Division, N-BARD, Hiroshima University, Higashihiroshima, Hiroshima 739-8526, Japan

E-mail:d140698@hiroshima-u.ac.jp, tsuzuki@hiroshima-u.ac.jp, ish@hiroshima-u.ac.jp

Abstract. The Yb-based compound YbCuGe with a hexagonal structure is reported as a metallic antiferromagnet with T_N = 4.2 K. It is also proposed that this compound contains geometrical magnetic frustration due to the competition of exchange interaction within the quasi-two dimensional Yb plane of which Yb ions form a triangle. Previous research suggested that its magnetic anisotropy can be ascribed to the crystalline electric field (CEF) effect on a single Yb ion. In order to clarify the CEF effect in this compound, ultrasonic measurements and related theoretical fitting have been carried out. The transverse elastic modulus C_{44} exhibits a softening which begins at 120 K and stops at about 20 K. On the other hand, the longitudinal modulus C_{33} mode shows a continuous hardening before T_N. We carried out the theoretical strain-susceptibility calculation of C_{44} by using the CEF parameters reported by Katoh *et al.* According to the fitting results, it can be concluded that the softening at high temperature range can be explained by the quadrupole interaction between the ground and excited Kramers doublets under the hexagonal CEF.

1. Introduction

Yb-based compounds have recently attracted the interest of many researchers, especially the physical properties of a series of compounds YbTGe (T = transition metal) are well investigated [1-4]. Among them, the metallic compound YbCuGe with a hexagonal structure is reported as an antiferromagnet with a phase transition temperature of T_N = 4.2 K [5, 6]. In general, the Kondo interaction which makes a spin-singlet composed of f- and conduction-electrons may be suppressed by the spontaneous magnetization below phase transition temperature. However, comparing with some other compounds in this YbTGe series, such as YbPtGe and YbIrGe [1, 2], YbCuGe compound contains some different properties.

Above 100 K, as reported by Katoh *et al.*, the temperature dependence of the inverse magnetic susceptibility $1/\chi(T)$ of YbCuGe obeys the Curie-Weiss law. The obtained effective magnetic moment μ_{eff} is 4.27 μ_B for $H//a$ and 4.43 μ_B for $H//c$, respectively. These values of μ_{eff} are close to the value of 4.53 μ_B expected for the free Yb^{3+} ion. It is considered that this magnetic anisotropy in the higher-

temperature range is ascribed to the crystalline electric field (CEF) effect on a single Yb ion. The magnetic contribution $C_m(T)$ of the specific heat for YbCuGe shows a broad peak at 50 K. This can be ascribed to the Schottky contribution which is derived from four Kramer's doublets for the total angular momentum J = 7/2 of the Yb^{3+} ion. Katoh *et al.* got the CEF parameters of B_2^0 = -0.7 K, B_4^0 = 0.15 K, and B_4^3 = 0.5 K in the first approximation which treats only the second- and fourth-order terms of the trigonal CEF model as the site symmetry of the Yb ions is trigonal. The first-, second-, and third-excitation energies of the CEF levels from the ground doublet were estimated to be 99 K, 162 K, and 219 K, respectively [6].

At low temperature, the peak of inverse magnetic susceptibility for $H//c$ indicates its antiferromagnetic ordering below 4.2 K. By the results of temperature dependence of the specific heat $C(T)$, the electronic specific heat coefficient γ was estimated as 1.35 mJ/(mol K^2). However, the magnetic entropy $S_m(T)$ at T_N is 28% of that of $R\ln 2$ which suggests that the slow release of magnetic entropy is probably ascribed to the spin fluctuation instead of the Kondo effect because of the small γ value. Moreover, it is also proposed that this compound contains geometrical magnetic frustration due to the competition of exchange interaction within the quasi-two dimensional Yb plane, which forms the triangle of Yb ions. All of these revealed that YbCuGe is a metallic antiferromagnet without heavy fermion character [6]. To investigate the phase transition at T_N and clarify the CEF effect in YbCuGe, we carried out ultrasonic measurements on a single-crystalline sample and related theoretical calculation was performed.

2. Experimental

Single crystal of YbCuGe was grown by the Bridgman method [5]. The elastic moduli C_{33} and C_{44} were measured as a function of the temperature T from 2 to 300 K using the phase comparison-type pulse echo method [7]. Ultrasound with the frequency of 10 or 30 MHz was generated and detected by $LiNbO_3$ transducers glued onto parallel surfaces of a sample. The elastic modulus C_{ii} was calculated using $C_{ii} = \rho v^2$ with a room-temperature mass density ρ = 9.49 g/cm^3, where v is the sound velocity in a sample. We estimated the absolute value of v at 4.2 K by using the relation $v = 2l/t$, where l is the sample length and t is the time interval between pulse echoes.

3. Results and discussion

Fig. 1 shows the T dependence of the longitudinal elastic modulus C_{33} from 2 K to 300 K, and C_{33} with the temperature range around phase transition is shown as the inset in Fig. 1. It can be seen that C_{33} increases monotonically with decreasing temperature down to T_N, indicating an elastic hardening characteristic before the phase transition. With further decreasing the temperature, C_{33} exhibits a step-like softening, and a peak is located at 4.2 K which is corresponding to the antiferromagnetic phase transition temperature as previously reported.

The variation of the transverse modulus C_{44} versus temperature for YbCuGe from 2 to 300 K is shown in Fig. 2. C_{44} exhibits hardening with decreasing T at higher temperatures, and then shows a softening which begins at 120 K. The softening slows down around 30 K and finally stops with further decreasing the temperature to 20 K. For the T below T_N, a similar step-like softening around T_N is observed in the transverse mode C_{44} as shown in the inset of Fig.2. We previously discussed such a step-like softening as a result of magnetostriction [8].
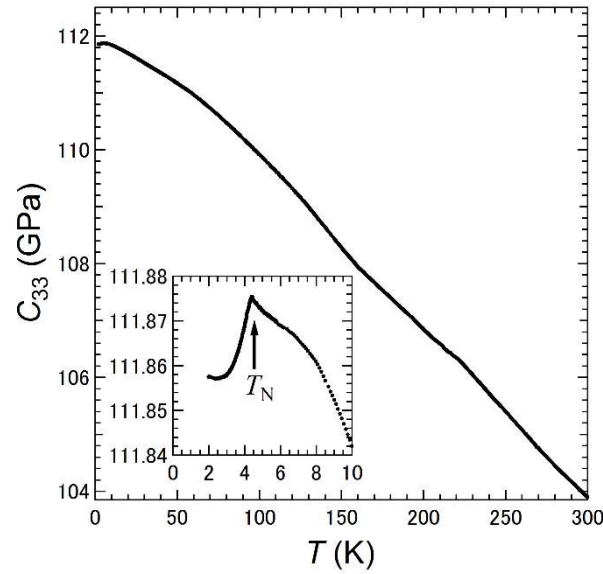
Figure 1: *T* dependence of the longitudinal elastic modulus C_{33}. The inset shows the *T* dependence of C_{33} below 10 K
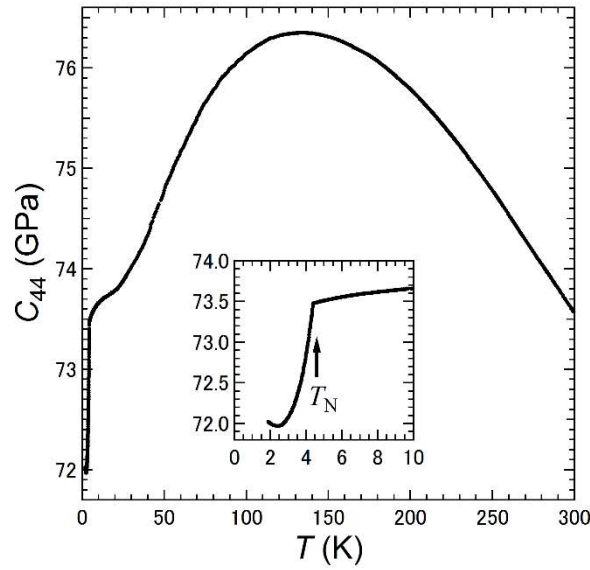
Figure 2: *T* dependence of the transverse elastic modulus C_{44}. The inset represents the *T* dependence of C_{44} below 10 K

We consider that the elastic softening of C_{44} may be due to the CEF effect. To obtain the detailed information, we carried out the theoretical fitting of C_{44} by using the CEF parameters reported by Katoh *et al.* as we mentioned above. We considered the effective Hamiltonian H_{eff} for the elastic modulus and the Hamiltonian of CEF as follows,

$$H_{eff} = H_{CEF} - gO_{yz}\varepsilon_{yz} - g'\langle O_{yz}\rangle O_{yz} \qquad (1)$$

$$H_{CEF} = B_2^0 O_2^0 + B_4^0 O_4^0 + B_4^3 O_4^3 \qquad (2)$$

where ε_{yz}, g, and g' are a strain, a strain-quadrupole coupling constant, and a quadrupole–quadrupole coupling constant, respectively. The T dependence of the elastic modulus, $C_{44}(T)$, is represented by the following equation [9],

$$C_{44}(T) = \frac{-N_0 g^2 \chi_s(T)}{1 - g' \chi_s(T)} + C_0(T) \qquad (3)$$

where χ_s is the so-called strain susceptibility and $N_0 \, (=1.85 \times 10^{28} \, \text{m}^{-3})$ is the number density of Yb ions per unit volume at room temperature. To consider the background stiffness, based on the analysis for the similar materials with CEF effect before, here it is assumed as $C_0(T) = a + bT^2 + cT^4$ [8, 10, 11].
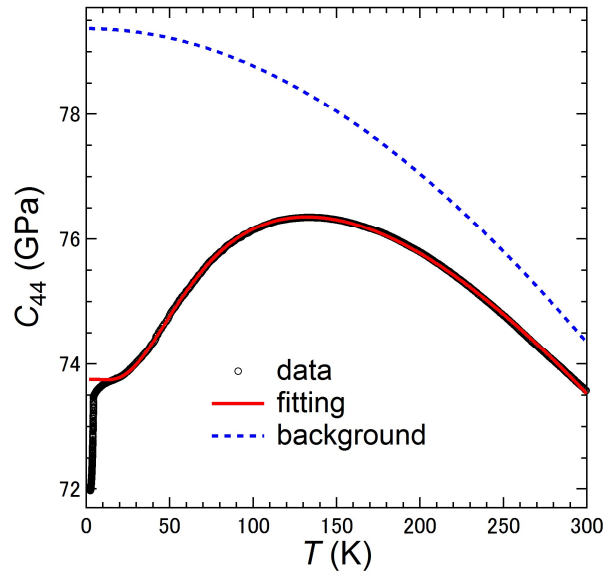
Figure 3: (Color online) T dependence of the elastic modulus C_{44}. The red solid and blue broken curves are the calculated curve and the background stiffness, respectively.

The fitting result of elastic modulus C_{44} is represented as the solid curve shown in Fig. 3. Here, we got the strain-quadrupole coupling constant, and the quadrupole–quadrupole coupling constant as $|g| = 237$ K, and $g' = -0.2$ K, respectively. The theoretical fitting curve of C_{44} is matched well with the experimental data above 20 K, indicating that the softening in C_{44} originates from a quadrupole interaction. However, same as the other YbTGe system, the ground Kramers doublet of YbCuGe also contains no quadrupole degeneracy. This is very similar to the phenomenon we discovered in YbIrGe and YbPtGe which have a similar CEF effect as YbCuGe. Accordingly, comparing with the similar phenomenon we discovered in YbIrGe and YbPtGe, we conclude that the softening phenomenon of YbCuGe should also be due to the indirect quadrupole interaction between the ground doublet and the excited doublets [8, 10]. As shown in Fig.3, for the low temperature range (below 20 K) of C_{44}, the fitting curve shows a tendency to deviate from the experimental data which may arise from spin fluctuation of the phase transition.

However, the inverse magnetic susceptibility, especially for $H//a$, was not reproduced by this CEF parameters which are only considered the second- and fourth-order terms of Hamiltonian [6]. It means that it is necessary to consider the sixth-order terms of the CEF. To clarify the CEF level scheme more precise, ultrasonic measurements of other modes (C_{11} and C_{66}) are in progress. We are planning theoretical fitting of the elastic moduli by combining with the magnetic susceptibility and the Schottky specific heat of YbCuGe by using all terms of the trigonal CEF, and this work is in progress now.

4. Conclusion

Ultrasonic measurements have been performed on the single crystal of metallic antiferromagnet YbCuGe from 2 to 300 K. In order to study the CEF effect and the phase transition at T_N in this compound, the elastic moduli C_{33} and C_{44} were measured. A hardening feature above phase transition temperature is observed in longitudinal elastic modulus C_{33}. On the other hand, the transverse elastic modulus C_{44} shows elastic softening which begins at 120 K and disappears at 20 K with decreasing the temperature. The result of theoretical strain-susceptibility fitting reveals that the elastic softening phenomenon in YbCuGe originates from the indirect quadrupole interaction between the ground doublet and the excited doublets although all doublets have no quadrupole degeneracy.

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References

[1]   Katoh K, Nakagawa S, Terui G and Ochiai A 2009 *J. Phys. Soc. Jpn.* **78** 104721
[2]   Katoh K, Tsutsumi T, Yamada K, Terui G, Niide Y and Ochiai A 2005 *Physica B* **369** 81
[3]   Katoh K, Mano Y, Nakano K, Terui G, Niide Y and Ochiai A 2004 *J. Magn. Magn. Mater.* **268** 212
[4]   Enoki K, Hirose Y, Yoshiuchi S, Sugiyama K, Honda F, Takeuchi T, Yamamoto E, Haga Y, Hagiwara M, Kindo K, Settai R and Onuki Y 2012 *J. Phys. Soc. Jpn.* **81** SB056
[5]   Heying B, Rodewald Ute Ch., Pöttgen R, Katoh K, Niide Y and Ochiai A 2005 *Monatshefte für Chemie* **136** 655
[6]   Katoh K, Maeda M, Matsuda S and Ochiai A 2012 *J. Alloy Compd.* **520** 122
[7]   Lüthi B, Bruls G, Thalmeier P, Wolf B, Finsterbusch D and Kouroudis I 1994 *J. Low Temp. Phys.* **95** 257
[8]   Xi X J, Ishii I, Noguchi Y, Goto H, Kamikawa S, Araki K, Katoh K and Suzuki T 2015 *J. phys. Soc. Jpn.* **84** 124602
[9]   Lüthi B 1980 *Dynamical Properties of Solids* ed G K Horton and A A Maradudin (North-Holland, Amsterdam) Chap. 4
[10]   Ishii I, Noguchi Y, Kamikawa S, Goto H, Fujita T K, Katoh K and Suzuki T 2014 *J. phys. Soc. Jpn.* **83** 043601
[11]   Nohara M, Suzuki T, Maeno Y, Fujita T, Tanaka I and Kojima H 1995 *Phys. Rev. B* **52** 570