Elastic softening and phase transition characteristics in YbPtGe

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Abstract
YbPtGe with the orthorhombic ε-TiNiSi-type structure is reported as a heavy-fermion ferromagnet with $T_C = 5.4$ K. Recent research suggested that its magnetic anisotropy can be ascribed to the crystal electric field (CEF) effect which acts on the eight-fold multiplet of Yb$^{3+}$ ions. In order to clarify the CEF effect and to investigate the magnetic phase transition in this compound, ultrasonic measurements of YbPtGe and related theoretical calculation have been performed. The transverse elastic modulus $C_{66}$ exhibits a softening characteristic with a peak at 180 K while the longitudinal modulus $C_{33}$ shows a continuous hardening in the whole temperature range. According to the theoretical strain-susceptibility fitting of the transverse elastic modulus $C_{66}$ by using the CEF parameters reported by Katoh et al., we argue the softening at higher temperatures (>50K) to be a result of quadrupole interaction between the CEF ground and excited Kramers doublets. With further decreasing the temperature below $T_C$, $C_{66}$ mode exhibits a step-like softening which is quite different from that of the $C_{33}$ mode and the possible mechanism is discussed.

Keywords: YbPtGe, Elastic modulus, elastic softening, ferromagnetic phase transition

1 Introduction

Recently, the physical properties of a series of Yb-based YbTGe ($T =$ transition metal) are well investigated [1-4] and YbPtGe with the orthorhombic ε-TiNiSi-type structure (space group $Pnma$) is a typical one of them. The lattice parameters of YbPtGe are $a = 6.863$ Å, $b = 4.305$ Å, and $c = 7.510$ Å. It is reported that YbPtGe shows a ferromagnetic transition at $T_C = 5.4$ K.

At high temperatures, the magnetic susceptibility of YbPtGe obeys the Curie-Weiss law above 100 K and the effective magnetic moment obtained of 4.48$\mu_B$ is close to the value of 4.53$\mu_B$ expected for the free Yb$^{3+}$ ion [2]. Consequently, the magnetic anisotropy in the higher-temperature range can be ascribed to the CEF effect. The eight-fold multiplet derived from the total angular momentum
For the Yb$^{3+}$ ion, the $J=7/2$ state splits into four doublets by the CEF effect. By theoretical fitting of specific heat and magnetic susceptibility [1], Katoh et al. obtained the CEF parameters of $B_2^0 = -2.6$ K, $B_2^2 = 1.2$ K, $B_4^0 = 0.23$ K, $B_4^2 = 0.09$ K, and $B_4^4 = 0.53$ K in a first approximation which treats only the second- and fourth-order terms of the orthorhombic CEF model. The first-, second-, and third-order excitation energies of the CEF levels from the ground doublet were estimated to be 196 K, 242 K, and 378 K, respectively.

Below 20 K, the temperature dependence of the electrical resistivity $\rho(T)$ of the $a$-, $b$-, and $c$-axes show a broad peak at approximately 12 K, which suggests the incoherent Kondo scattering of conduction electrons by Yb ion. The large background of $C_m(T)$ between 10 and 20 K also could be explained by the Kondo contribution. Besides, the magnetic entropy $S_m(T)$ at $T_C = 5.4$ K is 52% of $R\ln2$ which is expected for a doublet ground state, suggesting that the release of magnetic entropy is suppressed by the Kondo effect. The estimated electronic specific heat coefficient $\gamma$ is 209 mJ/(mol·K$^2$). These revealed that the YbPtGe is a heavy-fermion compound with a ferromagnetic transition. To investigate the phase transition at $T_C$ and clarify the CEF effect in YbPtGe, we carried out ultrasonic measurements on a single-crystalline sample and related theoretical calculation was performed.

2 Experimental

Single crystal of YbPtGe was grown by the Bridgman method [5]. The elastic moduli $C_{33}$ and $C_{66}$ were measured as a function of temperature from 3 to 300 K using the phase comparison-type pulse echo method [6]. 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 13.19 g/cm$^3$, where $v$ is the sound velocity. We estimated the absolute value of $v$ at 4.2 K by using the relation $v = 2l/\tau$, where $l$ is the sample length and $\tau$ is the time interval between pulse echoes.

3 Results and discussion

Fig. 1 and Fig. 2 show the variation of longitudinal elastic modulus $C_{33}$ and transverse modulus $C_{66}$ versus temperature for YbPtGe from 3 to 300 K respectively. At temperatures above $T_C$, $C_{33}$ increases monotonically with decreasing temperature, indicating an elastic hardening characteristic before the phase transition. Unlike the $C_{33}$, at higher temperatures, the transverse elastic modulus $C_{66}$ first exhibits hardening with decreasing $T$, then begins to show softening at 180 K, and this softening stops with further decreasing the temperature at about 30 K.

For the elastic moduli below $T_C$, abrupt elastic hardening is observed in $C_{33}$ mode, while a step-like softening around $T_C$ is observed in transverse mode $C_{66}$. We can discuss the origin for the shape of anomaly by assuming the coupling between the strain and a magnetic order parameter. Based on our assumption, the order parameter can be estimated as magnetic moment, which is a vector, and the strain is a second-rank tensor. Since the free energy $F_C$ should be scalar, we firstly estimate the coupling linear in the strain and quadratic in the order parameter, which is the magnetostriction Hamiltonian. Rehwald calculated a step-like behavior in the elastic modulus around $T_C$ with decreasing the temperature assuming the magnetostriction Hamiltonian [7]. The abrupt hardening in $C_{33}$ at $T_C$ indicates that the $\varepsilon_{zz}$ strain couples the quadratic product of the magnetic order parameter. However, $C_{66}$ shows the step-like softening at $T_C$, suggesting a different type of coupling between the $\varepsilon_{xy}$ strain and the magnetic order parameter. The measurements of all elastic moduli of YbPtGe are...
underway. We will discuss the magnetically ordered structure taking into account the variation of elastic anomalies in all elastic moduli elsewhere [8].

**Figure 1:** Temperature dependent longitudinal elastic modulus $C_{33}$. The inset shows the $T$ dependence of $C_{33}$ below 10 K.

**Figure 2:** Temperature dependent transverse elastic modulus $C_{66}$. The inset displays the $T$ dependence of $C_{66}$ below 10 K.
As it is reported that the CEF effect exists in this compound [2], we suppose that the softening characteristic of $C_{66}$ below 180 K might arise from the CEF effect. To obtain the detail information of CEF in YbPtGe and its affection on the elastic moduli, we carried out a theoretical fitting for the non-ordered state of the transverse elastic modulus $C_{66}$ by using the CEF parameters reported by Katoh et al. as we mentioned above[1], $|g| = 102$ K, and $g' = 7.2$ K. We considered the effective Hamiltonian $H_{\text{eff}}$ for the elastic moduli and the Hamiltonian of CEF as follows,

$$H_{\text{eff}} = H_{\text{CEF}} - g O_{xy} \varepsilon_{xy} - g' \langle O_{xy} \rangle O_{xy} \quad (1)$$

$$H_{\text{CEF}} = B^2_2 O_2^0 + B^2_2 O_2^0 + B^4_2 O_4^0 + B^4_4 O_4^0 \quad (2)$$

Where $\varepsilon_{xy}$, $g$, and $g'$ are a strain, a strain-quadrupole coupling constant, and a quadrupole–quadrupole coupling constant, respectively. The $T$ dependence of the elastic modulus, $C_{66}(T)$, is represented by the following equation[9],

$$C_{66}(T) = -\frac{\chi_s(T)}{1-g \chi_s(T)} C_0(T) \quad (3)$$

Where $\chi_s$ is the so-called strain susceptibility and $N_0 (=1.83 \times 10^{28} \text{ m}^{-3})$ is the number density of Yb ions per unit volume at room temperature. The fitting result of elastic modulus $C_{66}$ is represented as the solid curve shown in Fig. 3. Although $C_{66}$ increases linearly when cooling down to about 200 K from 250 K, which is similar to the behavior described by an expression given by Varshni[10], based on our analysis for similar materials within all the temperature range before, here the background stiffness will be assumed as $C_0(T) = a + bT^2 + cT^4$[11,12].

![Figure 3: (Color online) T dependences of the elastic modulus $C_{66}$. The red solid and blue broken curves are the calculated curve and the background stiffness, respectively.](image-url)
We can see in Fig. 3 that the fitting curve of $C_{66}$ perfectly matches the experimental data above 60 K. This result indicates that the softening in $C_{66}$ originates from a quadrupole interaction. However, the ground doublet $\Gamma_5$ of YbPtGe contains no quadrupole degeneracy, which is very similar to the phenomenon we discovered in YbIrGe which has the same structure and similar CEF effect as YbPtGe. Accordingly, the softening of YbPtGe should also be due to the indirect quadrupole interaction between the ground doublet and the excited doublets as we discussed in YbIrGe [11]. However, the low temperature range (below 60 K), $C_{66}$ could not be reproduced by the CEF parameters we used because these CEF parameters only comprise the second- and fourth-order terms of Hamiltonian, that means sixth-order CEF term should be taken into consideration. To clarify the CEF level scheme, ultrasonic measurements of other modes ($C_{11}$, $C_{22}$, $C_{44}$, and $C_{55}$) are in progress.

To reproduce the elastic moduli, we are planning a theoretical fitting by using all terms of the orthorhombic CEF, in addition of this, the theoretical fitting together with magnetic susceptibility and the specific heat Schottky anomaly of YbPtGe is also in progress. [8]

4 Conclusion

Ultrasonic measurements have been performed on a single crystal of the heavy-fermion compound YbPtGe from 3 to 300 K. We measured the elastic moduli $C_{33}$ and $C_{66}$ in order to study the CEF effect and the phase transition at $T_C$ in this compound. The longitudinal elastic modulus $C_{33}$ displays a hardening feature while the transverse elastic modulus $C_{66}$ shows elastic softening which begins at 180 K and disappears at 30 K with decreasing temperature. The longitudinal elastic modulus $C_{33}$ and transverse elastic modulus $C_{66}$ show a step-like hardening and softening around $T_C$, respectively. The variation of the elastic anomaly may be understood by the form of coupling between the strain and the magnetic order parameter. The result of theoretical strain-susceptibility fittings reveals that the indirect quadrupole interaction between the ground doublet and the excited doublets can explain the elastic softening phenomenon in YbPtGe, although all doublets have no quadrupole degeneracy.

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