Elastic softening in the orthorhombic compound YbPdGe

Isao Ishii1, Yoshihito Noguchi1, Hiroki Goto1, Xiaojuan Xi1, Shuhei Kamikawa1, Koji Araki2, Kenichi Katoh2, and Takashi Suzuki1

1Hiroshima University, Higashihiroshima, Hiroshima, Japan
2National Defense Academy, Yokosuka, Kanagawa, Japan
ish@hiroshima-u.ac.jp, tsuzuki@hiroshima-u.ac.jp

Abstract
The Yb-based compound YbPdGe with the orthorhombic structure shows a ferromagnetic phase transition at 11.4 K. YbPdGe has localized 4f-electrons of Yb3+ ions and the crystal electric field (CEF) effect at high temperatures. On the other hand, a heavy-fermion state is reported at low temperatures, indicating that YbPdGe is a heavy-fermion ferromagnet. To investigate the phase transition at 11.4 K and the CEF effect, we carried out ultrasonic measurements on a single-crystalline sample. Characteristic elastic softening below 170 K is observed at high temperatures in the transverse elastic modulus $C_{66}$ in contrast to the longitudinal modulus $C_{22}$ without the softening. In the orthorhombic compound YbIrGe, the transverse moduli exhibit a similar elastic softening originating from the CEF effect, suggesting that the softening of $C_{66}$ in YbPdGe is also caused by the CEF effect. At the phase transition, $C_{22}$ and $C_{66}$ show abrupt elastic hardening and softening, respectively, due to the magnetostriction.

Keywords: YbPdGe, crystal electric field effect, heavy-fermion ferromagnet, ultrasound, elastic modulus

1 Introduction

The Yb-based compounds with Yb3+ ions have attracted much attention as a counterpart of the Ce-based compounds with Ce3+ ions. In these compounds, physical properties originating from the competition between the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction and the Kondo interaction are investigated. In the Yb-based compounds YbMX ($M$ : transition metal; $X$ = Ga and Ge) with the orthorhombic TiNiSi-type structure (space group $Pnma$), some compounds are reported as heavy-fermion magnets, such as YbNiGa, YbIrGe, and YbPtGe [1-5].

YbPdGe with the orthorhombic YbAuSn-type structure (space group $Imm2$) is also suggested as a heavy-fermion ferromagnet with the transition at $T_C = 11.4$ K, and it has four Yb sites [6-8]. The specific heat shows a clear peak at $T_C$. The magnetization in a magnetic field $H$ along the $b$-axis
increases steeply with increasing $H$ and saturates at 0.2 T. In contrast, the magnetization enhances gradually in $H$ along the $a$- and $c$-axes. In addition to the magnetization measurements, neutron diffraction measurements recently performed propose that the phase transition at $T_C$ is ferromagnetic ordering with magnetic moments in parallel with the $b$-axis [9].

At high temperatures, the magnetic susceptibility follows the Curie–Weiss law above about 100 K, and a relatively large magnetic anisotropy is observed [8]. The effective magnetic moments estimated are close to the value of the free Yb$^{3+}$ ion, suggesting that the crystal electric field (CEF) effect acts on its physical properties. Here, the eight-fold multiplet of the Yb$^{3+}$ ion splits into four doublets by the orthorhombic CEF.

Meanwhile, the Kondo effect is reported by electrical resistivity measurements. The temperature $T$ dependence of the magnetic resistivity shows a $-\ln T$ behavior between 150 and 300 K [8]. At low temperatures, the magnetic specific heat increases below 15 K. The electronic specific heat coefficient is estimated as 150 mJ/(mol K$^2$). The released magnetic entropy is equal to 0.7$R\ln2$ at $T_C$ and reaches $R\ln2$ at around 30 K, where $R$ is the gas constant. From these results, YbPdGe possesses the heavy-fermion state at low temperatures and is regarded as a heavy-fermion ferromagnet.

We previously carried out ultrasonic measurements on the orthorhombic compound YbIrGe which has a similar physical properties with YbPdGe. YbIrGe also has a localized character of 4$f$-electrons of Yb$^{3+}$ ions at high temperatures and the heavy-fermion state at low temperatures. We found characteristic softening of the elastic moduli due to an indirect quadrupole interaction between the ground doublet and the excited doublets under the CEF, though all doublets have no quadrupole degeneracy [10]. Elastic hardening is also observed below the temperature of antiferromagnetic transitions, suggesting a magnetostrictive coupling between a strain and magnetic order parameters. In this work, we measured the $T$ dependence of the elastic moduli on a single-crystalline sample using an ultrasonic technique in order to investigate the ferromagnetic transition at $T_C$ and the CEF effect in YbPdGe.

2 Experimental

Single crystal of YbPdGe was grown by the Bridgman method [6]. The lattice parameters of YbPdGe are $a = 4.334$ Å, $b = 20.506$ Å, and $c = 7.526$ Å. The $T$ dependence of the elastic moduli $C_{22}$ and $C_{66}$ is measured between 4.2 and 300 K using the phase comparison-type pulse echo method [11]. The modulus $C_{22}$ is the longitudinal mode propagating along the $b$-axis. The $C_{66}$ is the transverse mode propagating along the $b$-axis with the polarization direction along the $a$-axis. The frequency of ultrasound was 96 MHz for $C_{22}$ and 111 MHz for $C_{66}$. The $C_{ii}$ was calculated using an equation $C_{ii} = \rho v^2$ with a room-temperature mass density $\rho = 10.49$ 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 $v = 2lt$, where $l$ is the sample length and $t$ is the time interval between pulse echoes.

3 Results and discussion

The $T$ dependence of the longitudinal elastic modulus $C_{22}$ is shown in Fig. 1. The modulus $C_{22}$ increases monotonically with decreasing $T$. With further decrease in $T$, abrupt softening is detected just at $T_C$, and then $C_{22}$ exhibits elastic hardening down to 4.2 K, as shown in the inset of Fig. 1. The phase transition at $T_C$ is of the second-order because of no hysteresis in the $T$ sweep. A plausible origin of the abrupt softening at $T_C$ is owing to the change in specific heat at $T_C$ through the Ehrenfest relation. We can estimate the strain $\varepsilon$ dependence of $T_C$ along the $b$-axis using the equation [12]:
where $V_{mol} = 4.03 \times 10^{-4} \text{ m}^3/\text{mol}$ is the molar volume and $\Delta C_p(T_C) = 5.53 \text{ J/(mol K)}$ is the change in isobaric specific heat at $T_C$ [8]. From our result, the magnitude of the abrupt softening of $C_{22}$ at $T_C$, $\Delta C_{22}(T_C)$, is equal to $-2.53 \times 10^{-2} \text{ GPa}$. We calculated the absolute value of the $\varepsilon$ dependence of $T_C$, $|dT_C/d\varepsilon| = 142.2 \text{ K}$, along the $b$-axis. This value is the same order as the value of $|dT_C/d\varepsilon| = 128.8 \text{ K}$ along the $c$-axis in YbIrGe [10].

**Figure 1:** $T$ dependence of the longitudinal elastic modulus $C_{22}$. The inset represents the same data in an expanded scale below 15 K.

**Figure 2:** $T$ dependence of the longitudinal elastic modulus $C_{66}$. The inset represents the same data in an expanded scale below 15 K.
Figure 2 shows the $T$ dependence of the transverse elastic modulus $C_{66}$. The modulus $C_{66}$ exhibits monotonic hardening above 170 K. On the contrary, $C_{66}$ turns into decrease below 170 K. YbPdGe has localized 4f-electrons of Yb$^{3+}$ ions and is under the CEF at high temperatures. As mentioned in Sec. 1, in the Yb-based compound YbIrGe which forms the orthorhombic structure, the transverse moduli behave a similar elastic softening. We carried out theoretical analyses based on the CEF effect for the transverse elastic moduli, the magnetic susceptibility, and the Schottky specific heat between 10 and 300 K in YbIrGe. It is manifested that the softening originates from an indirect quadrupole interaction between the ground doublet and the excited doublets under the CEF [10]. In the case of YbIrGe, we could fit the data perfectly because of a single Yb site. Since YbPdGe has four Yb sites, it is difficult to estimate the CEF effect exactly although elastic softening can be explained by the CEF effect in principle. Considering a similarity of physical properties between YbPdGe and YbIrGe, the softening of $C_{66}$ in YbPdGe will be caused by the CEF effect. YbPdGe also has the indirect quadrupole interaction between the ground doublet and the excited doublets, though all doublets are Kramers doublets and have no quadrupole degeneracy.

With further decreasing $T$, the softening due to the CEF effect stops around 20 K, and then $C_{66}$ shows elastic softening below $T_C$ down to 4.2 K, as shown in the inset of Fig. 2. The elastic hardening below $T_C$ is observed in the longitudinal modulus $C_{22}$. The phase transition at $T_C$ is reported as ferromagnetic ordering [8,9]. These softening in $C_{66}$ and hardening in $C_{22}$ below $T_C$ propose that a strain strongly couples to a magnetic order parameter. The difference of elastic behavior below $T_C$ between $C_{66}$ and $C_{22}$ is owing to a difference of the predominant coupling between the strain and the magnetic order parameter [13].

4 Conclusion

We performed ultrasonic experiments on the heavy-fermion ferromagnet YbPdGe, which has the orthorhombic structure. Characteristic elastic softening between 20 and 170 K is observed in the transverse modulus $C_{66}$ in contrast to the longitudinal modulus $C_{22}$ without the softening at high temperatures. The softening probably arises from the indirect quadrupole interaction between the ground doublet and the excited doublets under the orthorhombic CEF. We found abrupt elastic softening just at $T_C$ in $C_{22}$, and estimated the strain dependence of $T_C$ along the $b$-axis. The elastic hardening of $C_{22}$ and softening of $C_{66}$ are detected below $T_C$, suggesting the strong coupling between the strain and the magnetic order parameter.

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References