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To cite this article: Y Nakanishi et al 2010 J. Phys.: Conf. Ser. 200 012142

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Ultrasonic investigation close to quantum critical point in YbTr$_2$Zn$_{20}$ (Tr: Co, Rh, and Ir)

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Abstract. We performed ultrasonic measurements on high quality single crystals of the Yb-based heavy fermion compounds YbTr$_2$Zn$_{20}$ (Tr: Co, Rh and Ir) over a temperature range from 200 K to 0.5 K, which seem to be close to a quantum critical point (QCP). A sharp contrast of the temperature dependence of elastic constants was found at low temperature among the three compounds, reflecting the 4$f$ electronic state stemmed from Yb ion. The results indicate that a crystalline electric field (CEF) effect seems to be dominant in the systems YbRh$_2$Zn$_{20}$ and YbIr$_2$Zn$_{20}$ at low temperatures. On the other hand, the CEF effect is much less, but an additional effect would be dominant which is most probably ascribable to non Fermi liquid characteristics formed close to the QCP. We discuss briefly each 4$f$ electronic state developed at the low temperatures and physical parameters relating to a renormalized band model in YbTr$_2$Zn$_{20}$ in the framework of a deformation potential approximation.

1. Introduction

Recently, quantum critical phenomena is one of the most interesting issues in strongly correlated electron system. Quantum critical points (QCP) emerging when the continuous-transition temperature of symmetry breakings such as magnetic orders is suppressed to absolute zero have been investigated energetically, since the critical fluctuations induce unusual physical phenomena including unconventional superconductivity.

A new family of lanthanide intermetallic compounds $R$Tr$_2$Zn$_{20}$ ($R$=lanthanide, Tr=transition metal) would appear to be ideal candidates for investigating the above strong electronic correlations. In particular, the Yb-based compounds YbTr$_2$Zn$_{20}$ (Tr: Co, Rh and Ir) have been attracted considerable attention since the discovery of a huge electronic specific heat $\gamma\sim 7900$ mJ/mol K$^2$ in YbCo$_2$Zn$_{20}$.[1] To shed light upon the mechanism how to form such an unusual electronic state a dual nature of localized/itinerant 4$f$ electrons is of great importance.

YbTr$_2$Zn$_{20}$ crystallizes in the cubic CeCr$_2$Al$_{20}$ ($Fd\bar{3}m$ space group) structure.[1,2] The feature is that Yb ion is surrounded by the four nearest neighbors as well as the 12 next-nearest neighbors made up of Zn atoms in this system. YbTr$_2$Zn$_{20}$ are know as heavy fermion compounds with linear coefficient of specific heat, $\gamma\sim 740$ mJ/mol K$^2$, $\sim 540$ mJ/mol K$^2$ and $\sim 7900$ mJ/mol K$^2$. 

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for Tr=Rh, Ir and Co, respectively.[1] The temperature dependence of their electrical resistivity shows metallic behavior and manifest clear $T^2$ temperature dependency at low temperatures. The low temperature magnetic susceptibility correlates well with the electronic specific heat value leading to the Wilson ratio to be 1.3, 1.2 for Tr=Rh, Ir, respectively. [1] Unfortunately there has been no report of the Wilson ratio on Tr=Co. The large values of $\gamma$ and $A$ lead to the small Kondo temperature $T_K$ to be 20 K, 21 K and 1.5 K for Tr=Rh, Ir and Co, respectively.[1] It seems that YbCo$_2$Zn$_{20}$ is the particular system among the three compounds. Accordingly, a sharp contrast of the temperature dependence of elastic constants was found at low temperature among them as mentioned below.

Ultrasonic measurements are one of the most powerful probes since it can provide us distinct responses characteristic of localized $f$ electrons. Indeed, we can obtain important information about quadrupolar degrees of freedom derived from the $4f$ localized nature.[3-5] It is, therefore, worthwhile to search for YbTr$_2$Zn$_{20}$ systems.

In this paper we would like to present elastic properties of YbTr$_2$Zn$_{20}$ studied by ultrasonic measurements, as a key ingredient explaining observed elastic anomalies, which determines the electronic states of Yb$^{3+}$ with strong renormalization of quasi-particles in these systems.

2. Experiment

Single crystals of YbTr$_2$Zn$_{20}$ were prepared by the Zn self-flux method. X-ray diffraction measurements on the prepared samples show no extra peaks corresponding to the impurity phases, giving evidence that YbTr$_2$Zn$_{20}$ has a cubic structure with the lattice parameter $a = 14.005$ Å, $a = 14.150$ Å and $a = 14.165$ Å for Tr=Co, Rh and Ir, respectively.[6] The each specimen used for the present ultrasonic measurements was cut into a rectangular shape with two axes along the $\langle 100 \rangle$ and $\langle 110 \rangle$ directions, which has roughly a size of $2 \times 2 \times 2$ mm$^3$. The sound velocity was measured by an ultrasonic apparatus based on a phase comparison method. Plates of LiNbO$_3$ were used for the piezoelectric transducers. The fundamental resonance frequency of LiNbO$_3$ transducers is 10 ~ 30 MHz. The transducers were glued on the parallel planes of the sample by an elastic polymer Thiokol. The absolute value of the sound velocity was obtained by measuring the delay time between the ultrasonic echo signals with an accuracy of a few percent. In the estimation of the present ultrasonic measurements was cut into a rectangular shape with two axes along the $\langle 100 \rangle$ and $\langle 110 \rangle$ directions, which has roughly a size of $2 \times 2 \times 2$ mm$^3$. The sound velocity was measured by an ultrasonic apparatus based on a phase comparison method. Plates of LiNbO$_3$ were used for the piezoelectric transducers. The fundamental resonance frequency of LiNbO$_3$ transducers is 10 ~ 30 MHz. The transducers were glued on the parallel planes of the sample by an elastic polymer Thiokol. The absolute value of the sound velocity was obtained by measuring the delay time between the ultrasonic echo signals with an accuracy of a few percent. In the estimation of the elastic constant $C = \rho v^2$, the mass density $\rho = 7.730$ g/cm$^3$, 7.906 g/cm$^3$ and 8.716 g/cm$^3$ was used for Tr=Co, Rh and Ir, which was derived from the each lattice constant above mentioned. The details of both the crystal growth and the experimental conditions were described elsewhere. [6]

3. Strain susceptibility

Ultrasonic measurement is one of the most powerful methods of establishing the ground state multiplet of rare-earth ions mainly split by crystalline electric field (CEF) effect, via the magnetoelastic interaction, the quadrupolar response of the $4f$ ions.[3-5] Actually, there are various couplings of lattice distortions and electronic states. In the case of intermetallic rare earth, especially for Heavy Fermion (HF) compounds a deformation potential coupling to conduction electrons plays an important role in their elastic property in addition to the quadrupolar response mentioned above. A large density of states of quasi-particles in the vicinity of Fermi level formed by a strong hybridization, a coupling constant $d$ between the quasi-particles and a relevant elastic strain associated with sound waves becomes significantly strong. [3,7] This situation often brings about an elastic anomaly. For the simplicity, we employed here a rectangular density of states with a total band width $W$. According to the deformation potential approximation with the rectangular density of states, the elastic constant $C_\Gamma$ can be written by

$$C_\Gamma(T) = C_\Gamma^{(0)}(T) - \frac{4d^2}{W} \tanh\left(\frac{\beta W}{4}\right)$$

(1)
Figure 1. The temperature dependence of elastic constant $C_{11}$, for YbCo$_2$Zn$_{20}$, YbRh$_2$Zn$_{20}$ and YbIr$_2$Zn$_{20}$ in zero field. A black circle and solid red line are experimental data and theoretical result, respectively. A dotted line denote a back ground elastic constant $C^{(0)}_{11}$

Table 1. The absolute values of each elastic constants and bulk modulus $C_B=(C_{11}+2C_{12})/3$ and Poisson ratio $\gamma=C_{12}/(C_{11}+C_{12})$ of YbTr$_2$Zn$_{20}$ at 4.2 K.

| Tr  | $C_{11}$ (GPa) | $(C_{11} - C_{12})/2$ | $C_{44}$ (GPa) | $C_B=(C_{11}+2C_{12})/3$ | $\gamma_p=C_{12}/(C_{11}+C_{12})$ |
|-----|----------------|------------------------|----------------|--------------------------|---------------------------------|
| Co  | 118.7          | 30.9                   | 21.5           | 77.5                     | 0.324                           |
| Rh  | 115.7          | 32.4                   | 26.3           | 72.5                     | 0.306                           |
| Ir  | 126.7          | 38.4                   | 27.7           | 75.5                     | 0.283                           |

where, $C^{(0)}_1$, $d$ and $W$ denote a background elastic constant, coupling constant and total band width, respectively. $\beta$ denotes $1/k_BT$.

4. Experimental Results and Discussions

Figure 1 shows examples of the temperature dependence of elastic constant $C_{11}$ for Tr=Co, Rh and Ir. The absolute values of each elastic constant and the calculated bulk modulus $C_B=(C_{11}+2C_{12})/3$ and Poisson’s ratio $\gamma_p=C_{12}/(C_{11}+C_{12})$ from $C_{11}$ and $(C_{11}-C_{12})/2$ at 4.2 K of YbTr$_2$Zn$_{20}$ are listed in Table 1. Only data of $C_{11}$ are shown here. The $C_{11}$ was measured by the longitudinal ultrasonic waves with the frequencies of 10 or 30 MHz propagating along the $\langle 100 \rangle$ axis with polarization along the $\langle 100 \rangle$ axis. All the data exhibit almost normal behavior at high temperature: a stiffening on lowering temperature. However, a strong temperature dependent behavior appears below around 30 K in the elastic constants $C_{11}$ of YbTr$_2$Zn$_{20}$. A slight but clear minimum structure is observed at around 10 K for Tr = Rh and Ir in YbTr$_2$Zn$_{20}$. On the other hand, a significant softening towards low temperatures is observed below 20 K, which is apparently different to the behavior of Tr = Rh and Ir. Solid red lines in Fig. 1 show the calculated results based on the formula (1). These fits yield the important values: background $C_{11}^{(0)}=a+bT$, deformation coupling constant $d$, and band width as summarized in Table 2.
Table 2. Physical parameters of background $C_T^{(0)} = a + bT$, deformation coupling constant $d$, and band width $W$ determined from our experimental data of YbTr$_2$Zn$_{20}$.

|     | $a$ (GPa) | $b$ (GPa/K) | $d^2$ (GPa) | $W$ (K) |
|-----|-----------|-------------|-------------|---------|
| Co  | 124.2     | -0.042      | 0.044       | 27.0    |
| Rh  | 117.8     | -0.027      | 0.006       | 83.3    |
| Ir  | 131.0     | -0.035      | 0.01        | 111.1   |

Here, we discuss the ground state property of Yb ion in YbTr$_2$Zn$_{20}$. In the case of Tr = Rh and Ir, the slight minimum would be originated from the 4f ground state multiplet of the Yb$^{3+}$ ion which can be $\Gamma_6$ or $\Gamma_7$ doublet. Actually, as the CEF effect would be dominant in Tr = Rh and Ir systems at low temperatures, as mentioned in the separated article.[8]

Apart from the CEF effect, we focus on the deformation potential coupling to renormalized quasi-particles in the vicinity of Fermi level for the moment, although the CEF effect is also important at low temperature in these systems. The estimated parameters $d$ and $W$ indicate that a rather narrow band would be formed in the vicinity of Fermi level in Tr=Co. The value is almost 5 times smaller than those of Tr=Rh and Ir. Correspondingly, the coupling constant $d$ becomes larger, suggesting the strong renormalization of quasi particles formed in Tr=Co. However, comparing a factor of the difference of the values $d$ and $W$, it is quite difficult to explain such a huge $\gamma$ value observed in Tr=Co among YbTr$_2$Zn$_{20}$ systems. Additional effect such as valence or magnetic fluctuation would probably contribute to the enhancement of the $\gamma$ value in Tr=Co. This point is a future issue to be solved.

5. Summary
In summary, we have investigated the elastic properties of YbTr$_2$Zn$_{20}$ systems. A marked difference was found in the temperature dependence of the longitudinal elastic constant $C_{11}$ for YbCo$_2$Zn$_{20}$ at low temperatures. The feature would reflect strong renormalization of quasi particles in YbCo$_2$Zn$_{20}$, probably due to non Fermi liquid characteristics formed close to the QCP. This is consistent with the fact that YbCo$_2$Zn$_{20}$ shows such a huge electronic specific heat $\gamma$ value of 7900 mJ/mol K$^2$, being in markedly contrast to that of YbRh$_2$Zn$_{20}$ and YbIr$_2$Zn$_{20}$.

Detailed microscopic experiments such as neutron scattering, nuclear magnetic resonance and resonant X-ray scattering measurements are highly desired in order to obtain the detailed information with respect to the CEF schematic level for each YbTr$_2$Zn$_{20}$ systems.

6. Acknowledgement
The work was performed at the Cryogenic Laboratory, Center for Instrumental Analysis, Iwate University. This work was in part supported financially by the discretionary expenditure of the president of Iwate University.

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