Ultrasonic study of the Yb-based heavy fermion compound YbRh$_2$Zn$_{20}$

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Abstract. We report ultrasonic measurements on the high quality single crystal of the Yb-based heavy fermion compound YbRh$_2$Zn$_{20}$ over a temperature range from 200 K to 0.5 K. A shallow, but clear minimum was observed in the temperature dependent elastic constants $C_{11}$, $(C_{11} - C_{12})/2$ and $C_{44}$ around 15 K, probably attributed to the ground state and low-lying excited states of Yb$^3$ in the cubic CEF. We discuss the low-temperature elastic properties and possible energy level scheme of localized 4$f$ state of Yb$^3$ ions in YbRh$_2$Zn$_{20}$.

CEF ground state developed at the low temperatures and physical parameters relating to a quadrupolar moment in YbRh$_2$Zn$_{20}$.

1. Introduction
In recent years, heavy fermion behavior in intermetallic materials containing Yb ions has been attractive in the strongly correlated electron systems since the discovery of a new Yb-based heavy fermion series, YbTr$_2$Zn$_{20}$ (Tr: Co, Rh and Ir) which is close to a magnetic quantum critical point (QCP).[1] So far heavy fermion (HF) behavior has been reported in some Yb-based compounds, such as YbAs, Yb$_3$Pt$_4$, YbAuCu$_4$ and YbRh$_2$Si$_2$.[2-5] HF are characterized by a large effective renormalized quasiparticle mass as reflected in strongly enhanced electronic specific heat coefficient $\gamma$, coefficient of $T^2$ resistivity $A$ and spins susceptibility at low temperature. At present, a large number of examples with respect to HF systems are of Ce and U based systems, and they have been investigated energetically from the both experimental and theoretical viewpoints.

YbRh$_2$Zn$_{20}$ crystallizes in the cubic CeCr$_2$Al$_{20}$ ($Fd\bar{3}m$ space group) structure.[1,6,7] 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. YbRh$_2$Zn$_{20}$ is known as a heavy fermion compound with linear coefficient of specific heat, $\gamma \sim 500$ mJ/mol K$^2$.[1] The temperature dependence of the 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 for YbRh$_2$Zn$_{20}$. [1] The large values of $\gamma$ and $A$ lead to the small Kondo temperature $T_K$ to be 16 K. No indication of either magnetic order or superconductivity were found in YbRh$_2$Zn$_{20}$ in the temperature down to 20 mK.
In this paper we would like to report elastic properties of YbRh$_2$Zn$_20$ by means of ultrasonic measurements, as a key ingredient explaining observed elastic anomalies, which determines the ground state and low-lying excited states of Yb$^{3+}$ in this system.

2. Experiment
Single crystal sample of YbRh$_2$Zn$_20$ was prepared by the Zn self-flux method. X-ray diffraction measurement on the prepared sample shows no extra peaks corresponding to the impurity phases, giving evidence that YbRh$_2$Zn$_20$ has a cubic structure with the lattice parameter $a = 14.150$ Å. The specimen used for the present ultrasonic measurement was cut into a rectangular shape with two axes along the $\langle 100 \rangle$ and $\langle 110 \rangle$ directions, which has a size of $2.2 \times 2.3 \times 2.3$ 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 $10 \sim 30$ 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 $\rho = 8.716$ g/cm$^3$ was used, which was derived from the above lattice constant. The details of both the crystal growth and the experimental conditions were described elsewhere. [8,9]

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 CEF effect, via the magnetoelastic interaction, the quadrupolar response of the 4$f$ ions. The elastic constants, as the strain susceptibility, measure the diagonal (Curie terms) and off-diagonal (Van Vleck terms) quadrupolar matrix elements. If a system has a quadrupolar - active state in the ground state or/and low-lying excited states of Yb$^{3+}$, one expects a strong temperature dependence of symmetry elastic constants $C_T$ according to [10-13]

$$C_T(T) = C_T^{(0)}(T) - \frac{N g_T^{(0)}(s)(T)}{1 - g_T^{(0)}(s)(T)}$$

(1)

Here, $C_T^{(0)}$, $\chi_T^{(s)}$ and $N$ are the background elastic constant, the number of Yb ions per unit volume, the corresponding strain susceptibility, respectively. $g_T$ and $g_T^{(0)}$ are the fitting free parameters, mean the magnetoelastic coupling constant, and the q = 0 interaction quadrupoles in the Hamiltonian

$$H = - \sum_i g_T O_T(i) \varepsilon_T(i) - \sum_i g_T^{(0)} O_T(i) O_T(i)$$

(2)

where, $\varepsilon_T$ denotes the symmetry strain, $O_T$ is the corresponding quadrupolar operator.

4. Experimental Results and Discussions
Figure 1 shows the temperature dependence of elastic constants, $C_{11}$, $(C_{11} - C_{12})/2$ and $C_{44}$. 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 77 K and 4.2 K of YbRh$_2$Zn$_20$ are listed in Table 1. The $C_{11}$ was measured by the longitudinal ultrasonic waves with the frequencies of 10 of 30 MHz propagating along the $\langle 100 \rangle$ axis with polarization along the $\langle 100 \rangle$ axis. On the other hand, the $(C_{11} - C_{12})/2$ and $C_{44}$ were measured by the
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 YbRh$_2$Zn$_{20}$ at both 77 and 4.2 K.

| Temperature | $C_{11}$ | $(C_{11} - C_{12})/2$ | $C_{44}$ | $C_B = (C_{11} + 2C_{12})/3$ | $\gamma = C_{12}/(C_{11} + C_{12})$ |
|-------------|----------|-----------------------|----------|-----------------------------|----------------------------------|
| 77 K        | 115.3    | 32.3                  | 26.3     | 72.2                        | 0.305                            |
| 4.2 K       | 115.7    | 32.4                  | 26.3     | 72.5                        | 0.306                            |

The YbRh$_2$Zn$_{20}$ data exhibit near normal behavior at high temperature: a stiffening with decreasing temperature. However, a shallow, but clear minimum was found in the temperature dependence of elastic constants $C_{11}$, $(C_{11} - C_{12})/2$ and $C_{44}$ at around 15 K, most probably ascribable to the 4f electronic state and lowest crystalline electric field (CEF) energy of the Yb ions. It should be noted that the low-temperature elastic behavior is markedly different from that of YbCo$_2$Zn$_{20}$. A pronounced elastic softening, which is proportional to the inverse temperature, was observed in the elastic constants $C_{11}$, $(C_{11} - C_{12})/2$ and $C_{44}$ at low temperatures in YbCo$_2$Zn$_{20}$, reflecting a difference of the ground state of Yb ion between them.[12]

Here, we discuss the ground state property of Yb ion in YbRh$_2$Zn$_{20}$. The eightfold-degenerate $J = 7/2$ Hund’s rule ground state multiplet of the Yb$^{3+}$ ion splits into two doublets $\Gamma_6$, $\Gamma_7$ and a quartet $\Gamma_8$ in a cubic CEF. The present results strongly suggest the ground state of Yb$^{3+}$ ion to be doublet $\Gamma_6$ or $\Gamma_7$. The quartet $\Gamma_8$ is most probably precluded to be the ground state since a
softening toward low temperature proportional to the reciprocal temperature is not observed in the temperature dependence of the both \((C_{11} - C_{12})/2\) and \(C_{44}\). This clear distinctness generally helps us to determine the 4\(f\)-ground state of rare-earth ion in rare-earth compounds.

Unfortunately there has been no information concerning the CEF parameters in YbRh\(_2\)Zn\(_{20}\) so far. It is thus impossible to discuss the CEF level scheme in detail at present. However, the characteristic minimum was observed in the temperature dependent \((C_{11} - C_{12})/2\) and \(C_{44}\), providing us an informative key sign. The solid black lines in Fig. 2 are theoretically best fits based on the formula (3), where we set a level scheme of Yb\(^3\) ion to reproduce reasonably the characteristic minimum appearing around 15 K with CEF level scheme: \(\Gamma_6 (0 \text{ K}) - \Gamma_7 (30 \text{ K}) - \Gamma_8 (70 \text{ K})\). It should be noted that we were, however able to obtain reasonable fits with \(\Gamma_7 (0 \text{ K}) - \Gamma_6 (30 \text{ K}) - \Gamma_7 (70 \text{ K})\). The dashed lines are the background of the unperturbed term. A detailed fitting to the obtained data suggests that the CEF ground state and first excited one are both doublet state \(\Gamma_6\) or \(\Gamma_7\). It is noted that the second exited state has little influence on the behavior. Thus, the total energy difference cannot be determined explicitly by the present study. The coupling constants \(g'\Gamma_3 = 0.005 \text{ K}, \ g\Gamma_3 = 59.66 \text{ K}\) and \(g'\Gamma_5 = 0.04 \text{ K}, \ g\Gamma_5 = 130.8 \text{ K}\) are obtained in the case of the \(\Gamma_6\) doublet ground state for YbRh\(_2\)Zn\(_{20}\). The values of \(g'\Gamma_3\) and \(g'\Gamma_5\) are positive, which suggest that the inter-site interaction of the \(\Gamma_3\) and \(\Gamma_5\) quadrupoles are of ferro-quadrupolar type. These small magnitude may prevent a quadrupolar phase transition in YbRh\(_2\)Zn\(_{20}\).

5. Summary
In summary, the Van Vleck-type elastic anomaly in the temperature dependent both \((C_{11} - C_{12})/2\) and \(C_{44}\) of YbRh\(_2\)Zn\(_{20}\) indicates a doublet ground state \(\Gamma_6\) or \(\Gamma_7\) separated by \(\sim 15 \text{ K}\) from a first excited quartet state \(\Gamma_8\). To determine the ground state explicitly, the details of the CEF schematic level will be fully understood after detailed experiments such as neutron scattering, nuclear magnetic resonance and resonant X-ray scattering measurements.

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