Elastic constants of U(Ru$_{1-x}$Rh$_x$)$_2$Si$_2$

T Yanagisawa$^1$, H Saito$^2$, T Mayama$^2$, Y Ikeda$^2$, H Hidaka$^2$, M Yokoyama$^3$, H Amitsuka$^2$

$^1$ Creative Research Initiative Sousei, Hokkaido University, Sapporo, Japan.
$^2$ Department of Physics, Hokkaido University, Sapporo, Japan.
$^3$ Department of Physics, Ibaraki University, Mito, Japan.
E-mail: tatsuya@cris.hokudai.ac.jp

Abstract. Ultrasonic measurements on Rh-substitution system U(Ru$_{1-x}$Rh$_x$)$_2$Si$_2$ ($x = 0, 0.02$ and $0.07$) were performed in order to investigate lattice properties for low temperature phase transitions of hidden order (HO) phase and antiferromagnetic (AFM) phase. Elastic constant $C_{11}$ vs. $T$ of the $x = 0.02$ and $0.07$ samples exhibit a depression below $80$ K, which is similar to a softening in the non-doped sample which had already been reported. The $C_{11}$ of $x = 0.02$ exhibits a step-like anomaly at $14.0$ K and an additional softening of $3.2\%$ with minimum at $6.7$ K, which correspond to HO and AFM transitions, respectively. The $C_{11}$ in $x = 0.07$ sample only exhibits a shoulder-like anomaly at around $17$ K and continue to depress down to $4.2$ K.

1. Introduction

A notorious heavy fermion compound URu$_2$Si$_2$ possesses a mysterious phase transition, so-called hidden order (HO), at $T_O = 17.5$ K and becomes superconducting at $T_{SC} \sim 1.2$ K. Since the HO had been discovered over two decades ago [1, 2, 3], a primary order parameter of the HO phase was once thought to be inducing a weak antiferro-magnetic (AFM) order with small moment of $0.02\mu_B/U$. [4] A series of recent experiments under hydrostatic pressures have, however, revealed that HO changes to AFM with first order transition under pressure and the weak AFM in HO is coming from a phase separation, which is independent from HO. [5, 6, 7, 8] After all, what is the primary order parameter for HO? Is it still an open and controversial question. The present study addresses this issue by means of ultrasonic measurements, which can sensitively detect lattice instability in both the HO and the large-moment AFM phases. By substituting Ru with Rh, the HO is suppressed with increase Rh concentration $x \leq 0.03$, while a volume fraction of AFM phase in the HO increases with maximum at around $x = 0.02$. [5] Such competition of HO and AFM in the Rh-doped samples resembles the situation of the pure URu$_2$Si$_2$ under hydrostatic pressures. The inset of Fig.1 represents a temperature-Rh concentration ($T$-$x$) phase diagram of U(Ru$_{1-x}$Rh$_x$)$_2$Si$_2$, which is compiled by elastic anomalies (open circles) in the present ultrasonic measurement and onset temperatures in a temperature variations of the intensity of magnetic Bragg peak in a previously reported neutron scattering (cross). [9] Here the low temperature phase boundaries, highlighted by two-toned color, are guides to the eye and being obscured because the boundaries are expected to end at intermediate Rh-concentrations between $0.01 < x < 0.02$ and $0.03 < x < 0.04$.

The HO is also accurately observed by ultrasonic measurement as an elastic anomaly in
the all elastic constants, longitudinal $C_{11}$, $C_{33}$, and transversal ($C_{11}$-$C_{12}$)/2 and $C_{44}$, $C_{66}$ modes. [10, 11, 12, 13] European and Japanese groups have independently performed the ultrasonic measurements on URu$_2$Si$_2$ in some 15-20 years ago, and after that, any other ultrasonic investigation on the material has not been reported for a while. In the present work, we have resumed the investigation of ultrasonic study on URu$_2$Si$_2$ and Rh-doped ones.

2. Experimental Details

Single crystals of U(Ru$_{1-x}$Rh$_x$)$_2$Si$_2$ were grown by the Czochralski pulling method using a tetra-arc furnace. Elastic constant of the non-doped sample was measured as grown, thus far. Rh-doped samples were vacuum-annealed at 1000 °C for 5 days. A pair of parallel faces was prepared using spark erosion perpendicular to the principal axes [100], [110] and [001]. A phase comparator using double balanced mixer can detect the relative change of sound velocity down to 4.2 K by using $^4$He refrigerator. Piezoelectric LiNbO$_3$ wafers of 36°Y-cut with 100μm thickness were used for generating and detecting longitudinal ultrasonic waves. The absolute value of sound velocity was estimated by monitoring ultrasonic echoes on a digital oscilloscope and calculated by sample length $L$ and mass density $\rho$, which was calculated from the lattice constants. [14]

3. Results and Discussion

Figure 1 shows relative change of elastic constant $C_{11}$ of U(Ru$_{1-x}$Rh$_x$)$_2$Si$_2$ for $x = 0$, 0.02 and 0.07 as a function of temperature. The $C_{11}$-curves are displayed by being shifted against each other. Down arrows and dotted lines indicate characteristic temperatures $T^*$ and $T'$, respectively (see text). Inset shows a transition temperature-Rh concentration ($T$-$x$) phase diagram of U(Ru$_{1-x}$Rh$_x$)$_2$Si$_2$, which is compiled by the present ultrasonic measurement and previous neutron scattering measurements. [9]
These characteristic temperature dependences in a PM phase will be explained phenomenologically by considering a Kondo volume collapse with a strong Grüneisen parameter coupling due to \( c-f \) hybridization effects. [15] The similar temperature dependence of longitudinal elastic constant has also been reported in the typical heavy-fermion compound such as UPt \(_3\). [16] When simply compared the magnitude of softening in U(Ru\(_{1-x}\)Rh\(_x\))\(_2\)Si\(_2\) with \( x = 0 \) and 0.02, the \( x = 0.07 \) seems to exhibit the biggest amount of change. However, a gradient of softening \( \partial C_{11}/\partial T \) at \( \sim 45 \) K changes with Rh-doping as \( \partial C_{11}/\partial T \ (x = 0) < \partial C_{11}/\partial T \ (x = 0.07) < \partial C_{11}/\partial T \ (x = 0.02) \). From this perspective, the softening of \( x = 0.07 \) is more likely to be suppressed by comparing \( x = 0.02 \).

In some heavy-fermion compounds, the characteristic temperature \( T^* \), where the elastic constant \( C_{11} \) takes local maximum, is considered to be linked to Kondo temperature \( T_K \). [15] The systematic change of \( T^* \) with doping Rh in the present work could be comparable with

### Table 1. Comparisons of elastic constant and characteristic temperatures

| Rh concentration \( x \) | \( C_{11} \) at 80 K \( (\times 10^{10}\text{Jm}^{-3}) \) | Softening (%) | Local Min. \( T'(K) \) | Local Max. \( T''(K) \) | \( T_O \) (K) | \( T_M \) (K) |
|---------------------------|---------------------------------|---------------|-----------------|-----------------|----------|----------|
| 0\(^\dagger\)            | 25.9                            | 0.10          | 32.0            | 80.1            | 17.5     | —        |
| 0.02                     | 24.8                            | 0.24          | 27.3            | 83.8            | 14.0     | 6.7      |
| 0.07                     | 24.8                            | 0.31          | \( \sim 17^\dagger \) | 79.6            | —        | —        |

\(^\dagger\)As grown sample
\(^\ddagger\)Using a shoulder as a local minimum
magnetic susceptibility $\chi$, where the similar anomalies (broad maximum) determined at $(T^*)_x = 55$ K, 60 K, and 51.8 K for $x = 0$, 0.025, and 0.05, respectively. [14]

Figure 2 shows detailed behaviors of the $C_{11}$ for $x = 0$, 0.02 and 0.07 below 25 K. The $C_{11}$ of $x = 0$ shows a kink with accompanying a tiny depression at the hidden order $T_O ≈ 17.5$ K, while the $x = 0.02$ shows a step-like depression at $T_O ≈ 14.0$ K. Below $T_O$, the $x = 0$ gradually saturates with hardening, while an additional softening of 3.2% from $10$ K to 6.7 K associated with ultrasonic attenuation is observed in the $x = 0.02$. Any anomaly was not observed in the $C_{11}$ of $x = 0.07$, except a broad shoulder at $T_O ≈ 17$ K. A magnitude of the depression at $T_O$ is $\Delta C_{11} ≈ 6.4 \times 10^7$ Jm$^{-3}$ for $x = 0.02$, and $T_O$ is $\Delta C_{11} ≈ 1.4 \times 10^6$ Jm$^{-3}$ for $x = 0$ (not shown).

The dramatic change of the behavior in $C_{11}$ at $T_O$ implies that a nature of HO will be also changed by Rh-doping. On the other hand, a specific heat of the present system also shows a jump at $T_O$, however, a tendency of a magnitude of the jump shows an opposite $x$-behavior, where the specific-heat-jump decreases with doping Rh concentrations. [17] A mechanism of the enhancement of elastic anomaly in Rh-0.02% doped sample is an open question.

In summary, the elastic constant $C_{11}$ of U(Ru$_{1-x}$Rh$_x$)$_2$Si$_2$ ($x = 0, 0.02, 0.07$) were measured down to 4.2 K. In order to ascertain whether the enhanced elastic anomaly at $T_O$ in the $x = 0.02$ sample is due to isotropic phenomena, which conserve the tetragonal symmetry and accompany a volume change, such as a Kondo effect and/or a $\Gamma_1$ multipole (scholar) order, it must be necessary to verify an anisotropy of the elastic anomalies in the Rh-doped samples by measuring an another longitudinal mode $C_{33}$ and volume conservative transverse $(C_{11}-C_{12})/2$, $C_{44}$ and $C_{66}$ modes. A measurement of elastic constants on the pure URu$_2$Si$_2$ under hydrostatic pressure is also now in progress.

This work is supported by Grant-in-Aid for Young Scientists (B) No.20740192 and for Scientific Research (B) No.19340086 and (S) No.20224015, MEXT, Japan. One of the authors (T.Y.) was supported by Hokkaido Univ. Leader Development System in the Basic Interdisciplinary Research Areas.

References

[1] Palstra T M M, Menovsky A A, van den Berg J, Dirkmaat A J, Kes P H, Nieuwenhuys G J and Mydosh J A 1985 Phys. Rev. Lett. 55 2727
[2] Maple M B, Chen J W, Dalichaouch Y, Kohara T, Rossel C, Torikachvili M S, McElfresh M W and Thompson J D 1986 Phys. Rev. Lett. 56 185
[3] Schlabitz W, Baumann J, Pollit B, Rauchschwalbe U, Mayer H M, Ahlheim U, Bredl C D 1989 Z. Phys. B 62 171
[4] Broholm C, Kjems J K, Buyers W J L, Matthews P, Palstra T M M, Menovsky, A A and Mydosh J A 1987 Phys. Rev. Lett. 58 1467
[5] Amitsuka H, Matsuda K, Kawasaki I, Tenya K, Yokoyma M, Sekine C, Tateiwa D, Kobayashi T C, Kawarazaki S, Yoshizawa H 2007 J. Magn. Magn. Mater. 310 214
[6] Amitsuka H, Sato M, Metoki N, Yokoyma M, Kuwahara K, Sakakibara T, Morimoto H, Kawarazaki S, Miyako Y and Mydosh J A 1999 Phys. Rev. Lett. 83 5114
[7] Matsuda K, Kohori Y, Kohara T, Kawahara K, Amitsuka H 2001 Phys. Rev. Lett. 87 087203
[8] Motoyama G, Nishikawa T, Sato N K 2003 Phys. Rev. Lett. 90 166402
[9] Yokoyma M, Amitsuka H, Itoh S, Kawasaki I, Tenya K and Yoshizawa H 2001 J. Phys. Soc. Jpn. 70 545
[10] Lüthi B, Wolf B, Thalmeier F, Günther M, Sixel W, Bruls G 1993 Phys. Rev. Lett. A 175 237
[11] Lüthi B, Bruls G, Thalmeier F, Wolf B, Finsterbusch D and Kouroudis I 1994 J. Low. Temp. Phys. 95 257
[12] Wölfl B, Sixel W, Graf R, Finsterbusch D, Bruls G, Lüthi B, Knetsch E A, Menovsky A A and Mydosh J A 1994 J. Low. Temp. Phys. 94 307
[13] Kuwahara K, Amitsuka H, Sakakibara T, Suzuki O, Nakamura S, Goto T, Mihalk M, Menovsky A A, de Visser A and Franse J M 1997 J. Phys. Soc. Jpn. 66 3251
[14] Dalichaouch Y, Maple M B, Chen J W, Kohara T, Rossel C, Torikachvili M S and Giorgi A L 1990 Phys. Rev. B 41 1829
[15] Lüthi B 2005 Physical Acoustics in the Solid State (Berin Heidelberg New York: Springer)
[16] Yoshizawa M, Lüthi B, Goto T, Suzuki T, Renker B, de Visser A, Frings P, Franse J J M, 1985 J. Magn. Magn. Mater. 52 413

[17] Yokoyama M et al. To be published