Ultrasonic Study of Antiferro-quadrupole Ordering in HoB_2C_2

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Abstract

Elastic properties of the ternary rare earth compound HoB_2C_2 with a tetragonal symmetry have been investigated by means of ultrasonic measurements. The elastic constants of C_{44}, C_{66} and $(C_{11}-C_{12})/2$ exhibit the characteristic softening below about 40 K. In order to analyze quadrupole susceptibility, we assume a pseudo-degeneracy model consisting of a ground state E-doublet and an excited state A or B-singlet at energy $\Delta \sim 5.0$ K. The considerable softening of the elastic constant and ultrasonic attenuation in phase IV between $T_{C1} = 5.9$ K and $T_{C2} = 5.0$ K have been found. The slow relaxation rate of 7×10^{-9} sec suggests that the fluctuation of quadrupole moments is enhanced in phase IV. We also show a H-T phase diagram of HoB_2C_2 under magnetic fields along [100].

Key words: HoB_2C_2; quadrupole ordering; elastic constant; ultrasonic attenuation

Tetragonal HoB_2C_2 shows an antiferro-quadrupole (AFQ) ordering at $T_{C2} = 5.0$ K below the antiferromagnetic (AFM) ordering at $T_{C1} = 5.9$ K [1]. Below T_{C2}, two inter-site interactions for electric quadrupole and magnetic dipole compete each other. Such a reversal of AFQ and AFM transition sequence is a curious behavior. Neutron experiments shows diffused scattering in an orderd phase IV between T_{C2} and T_{C1} [3]. The order parameter of the phase IV is not settled yet. Ultrasound has with great success been employed as a probe of the quadrupole orderings, i.e. AFQ ordering of O_{yz}, O_{zx}, O_{xy} in CeB_6 [2]. In the present work, we have performed the ultrasonic measurements on single crystals of HoB_2C_2.

Fig. 1 represents the relative change of the elastic constant of transverse C_{44}, C_{66} and $(C_{11} - C_{12})/2$ modes with frequencies of 31 MHz. All transverse modes show a considerable softening below about 40 K down to the transition into the AFM ordering phase III at $T_{C2} = 5.0$ K. The C_{44} shows the largest softening of 22 % as compared with 5.5 % for C_{66} and 2.4 % for $(C_{11} - C_{12})/2$. Considering the selection rule for the quadrupole moments at Ho^{3+} site with C_{4h} point group symmetry, we assume a model consisting of a ground E-doublet and an excited A- or B-singlet at energy separated Δ, in order to analyze this softenings based on quadrupole susceptibility [4]. In the case of the excited energy Δ is as small as $T_{C1} = 5.0$ K, the energy level forms pseudo-degeneracy, the C_{44} mode as the quadrupole susceptibility for O_{yz}, O_{zx} proportionals to reciprocal temperature described by $C_\Gamma = C_\Gamma^0(T - T_C^0)/(T - \Theta)$ above $T_{C2} = 5.0$ K.

In the phase IV between T_{C1} and T_{C2}, we have found the abrupt increase of the softenings and shrinkage of the ultrasonic echo. Fig. 2 represents ultrasonic attenuation curve for C_{44} mode as a function of temperature with constant frequencies of 31 MHz and 52 MHz. Inset shows a temperature dependence of relaxation time τ which was calculated from the experimental result by employing of the Debye-type dispersion formula as $\alpha(\omega) = (C_\infty - C_0)\omega^2\tau/2\rho v^3(1 + \omega^2\tau^2)$. We obtain a very slow relaxation rate $\tau = 7 \times 10^{-9}$ sec in the phase IV which is much slower than the rate of phase III and I. Such a slowing down of relaxation rate suggests a

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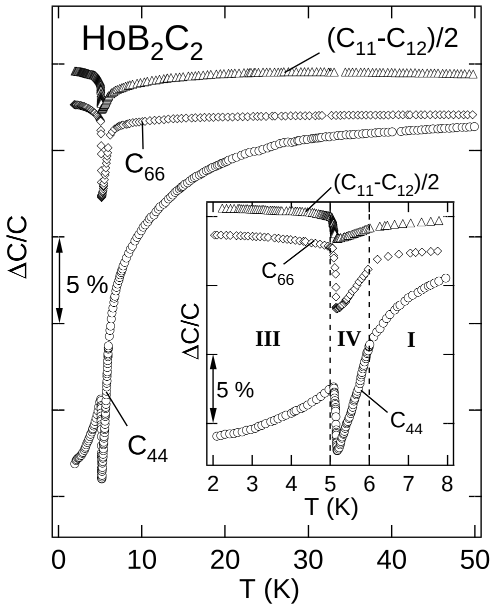

Fig. 1. Relative change of the elastic constants C_{44}, C_{66} and $(C_{11}-C_{12})/2$ of the transverse ultrasonic modes as a function of temperature in HoB_2C_2. Inset shows the detail behavior of these elastic constant change below 8 K. Vertical dashed lines in inset show the boundaries of phase IV.

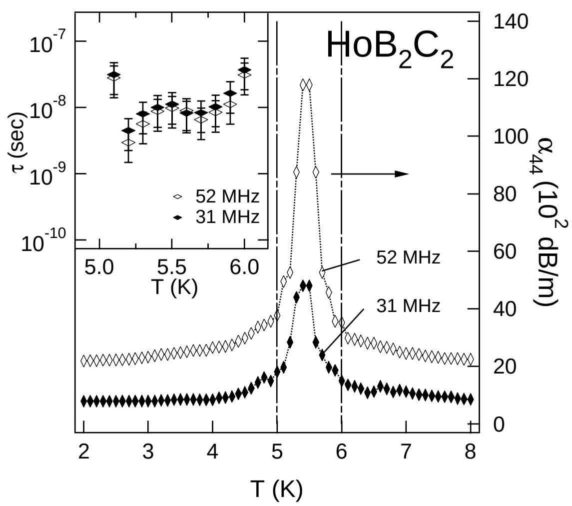

Fig. 2. Temperature dependence of the ultrasonic attenuation coefficient α_{44} in HoB_2C_2 at frequencies of 31 MHz and 52 MHz. Vertical lines show the boundaries of phase IV. Inset shows the calculated relaxation time τ vs temperature in phase IV.

fluctuation of the quadrupole moments $O_{yz}$and O_{zx}.

We have measured magnetic field dependence of the elastic constants of HoB_2C_2 in fields up to 12 T applied along the [100]-axis [5]. The considerable softening in the phase IV is wiped out by the low magnetic field of 1.0 T in both C_{66} and C_{44} modes (not shown). In Fig. 3, we show a magnetic phase diagram of HoB_2C_2

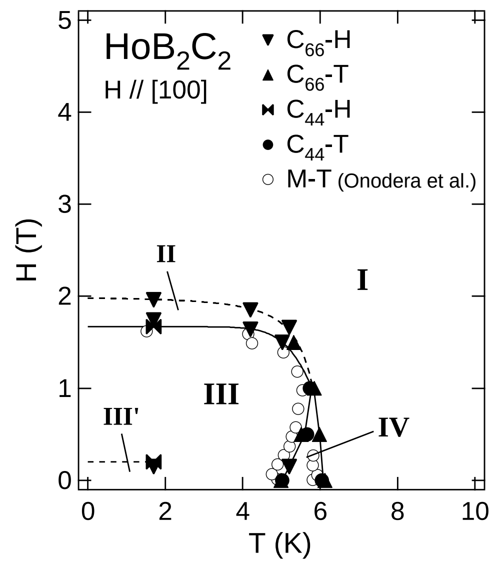

Fig. 3. Magnetic phase diagram of HoB_2C_2 with the fields applied along the [100]-axis. We numberd the phases as follows : I (PM), II (AFQ), III (AFQ+AFM) and IV (unknown). Previous data of magnetization by Onodera et al. are shown for comparison.

for $H//[100]$. The points on the phase diagram are determined by the elastic anomalies in C_{44} mode (solid circles and ribbons) and in C_{66} mode (solid triangles and reverse triangles). Solid and doted lines are guide to eyes. We have found phase II in the vicinity of the phase III-I boundary and sub phase III' in low field and temperature region. There probably exists a tetracritical point of field about 1.0 T at temperature of 5.8 K, which consists of the crossing of the AFQ and AFM phase boundaries. It should be noted that there is no indication for the phase boundary in high field region over 5 T up to 12 T in Fig. 3. It is now in progress that the ultrasonic measurement in magnetic field applied along the [110]- and [001]- axes, thermal expansion and magnetostriction.

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