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Effect of Hydrostatic Pressure on the Resistivity of Single Crystal Holmium

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The pressure effects of the c-axis and the basal plane resistivities of holmium have been investigated. The Néel temperature T_N of 131 K below which the spiral spin ordering develops is decreased with increasing pressure at a rate of -0.47 K/kbar. It is for the first time found that the spiral to conical spin structure transition temperature T_C is decreased with pressure at a rate of -0.10 K/kbar. The pressure dependencies of the residual, phonon and spin disorder ($\rho_{sd}(P,T)$) resistivities for both the a- and c-axes are estimated; $\rho_{sd}(P,T)$ for the a-axis is a decreasing function of pressure and scales fairly well with the reduced temperature T/T_N. Anomalous resistivity rise for the c-axis below T_N is analyzed on the basis of the theories by Miwa and Elliott and Wedgwood. It is indicated that the correction factor to the resistivity due to the magnetic superzone boundary gap as a consequence of incommensurate magnetic ordering is enhanced by pressure.

KEYWORDS: pressure effect, resistivity, superzone gap, Ho, single crystal, magnetic transition temperatures

1. Introduction

Many investigations on the effect of pressure on the magnetic properties of heavy rare earth metals[1-11] have been devoted to examine the indirect exchange interactions via conduction electrons, but most of experiments have been done using the inductive method and limited to above 77 K. It is well known that there are two magnetic transitions in Tb to Tm rare earth metals.[12,13] Below a Néel temperature T_N they show an antiferromagnetic structure with a periodic (incommensurate) arrangement of magnetic moments along the hexagonal c-axis. At lower temperatures there exists a transition to a structure with a ferromagnetic component below the Curie temperature T_C. For most heavy rare earths T_N has been reported to be decreased with increasing pressure, but the results are found to be scattered through the literatures.[1-11] Furthermore, there has been no report on the effect of pressure on the lower temperature transition T_C of Ho and Tm.

On the other hand, the effect of pressure on the transport properties, particularly the electrical resistivity, has not very often been investigated in heavy rare earths. This is partly due to the fact that high-accuracy resistivity measurement under hydrostatic pressure condition has been hampered by the instability to maintain good ohmic contacts to the sample and by the difficulty to introduce more than four electrical leads into the sample cell. However, the resistivity measurement under pressure may give invaluable information on the magnetic spin (disorder) scattering, particularly unusual temperature dependence of resistivity below T_N arising from the formation of magnetic superzone as a consequence of incommensurate magnetic ordering, as well as magnetic ordering temperatures, $T_N(P)$ and $T_C(P)$. Recently a neutron diffraction study in Ho under pressure[11] revealed that the wave vector $\boldsymbol{q}$ of the spiral arrangement below T_N was appreciably changed and suggested that the exchange interaction V was largely suppressed by pressure; $\Delta V/V = -12$ % at $P = 21$ kbar. It is, therefore, expected that the gap opening is so sensitive to pressure in Ho. There has been no reports concerning high pressure experiment not only on resistivity but also on $T_C(P)$ in Ho.

In the present paper, we report the precise high pressure resistivity measurements on holmium single crystals up to 60 kbar. The effects of pressure on both the magnetic ordering temperatures, T_N and T_C, and the anomalous behavior of the resistivity arising from the energy gap opening have been investigated.

2. Experimental

Single crystalline holmium samples were grown by the strain-anneal method. The starting Ho ingot with purity of 99.9+ % was wrapped by a Ta sheet, sealed into an evacuated quartz tube and subsequently annealed for 3 days at 1,420 K. The samples thus obtained were cut into the form of cube of $3 \times 3 \times 3$ mm^3 for magnetic measurements and rectangular parallelepipeds with dimensions of $0.4 \times 0.4 \times 6$ (or 2) mm^3 for electrical resistivity measurements.

The d.c. magnetization was measured by a SQUID magnetometer in the temperature range between 4.5 K and 300 K. The electrical resistivity measurements at pressures were made from 4.2 to 300 K by means of a conventional four-probe method. Voltage and current leads of fine gold wires were spot welded to the sample. Two high-accuracy nanovoltmeters (Keithley 182) were used to measure the sample voltage. The temperature is determined using a calibrated Au + 0.07 at. % Fe vs. Chromel thermocouple and a RuO_2 thermometer. Hydrostatic pressure was generated by

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using the following two devices: A clamp-type piston-cylinder cell and a cubic-anvil. The former was employed for simultaneous resistivity measurement parallel to the a-axis and to the c-axis on the two single crystals under pressures lower than 25 kbar using transmitting medium of 1:1 mixture of n-pentane and isoamylalcohol. The latter was used for higher pressures up to 60 kbar with 4:1 methanol and ethanol fluid.

3. Results

Weak field magnetization, σ, and the reciprocal magnetic susceptibility, $1/\chi$, are plotted as a function of temperature T in Fig. 1. Since the position of the cusp which indicates the onset of antiferromagnetic ordering is field dependent, the Néel temperature T_N was determined by the extrapolation to $\sigma_{H=0}$ in the isofield curves for the a-axis sample. Value determined in this way is 131 K. On the other hand, there is no anomaly around T_N in the c-axis data indicative of any onset of antiferromagnetism.

For the anomaly associated with the spiral to cone structure transition in the σ -T curve, a small peak is seen around the Curie temperature T_C ~ 20 K for the a-axis sample. The c-axis sample shows the sharp rise in the σ -T curve below T_C.

The $\chi(T)$ curves follow a Curie-Weiss law at higher temperatures as shown in Fig.1. The deduced effective Bohr magneton numbers and the paramagnetic Curie temperatures which are presented in Fig.1 are close to those of Strandburg *et al.*.[12)]

The resistivity versus temperature curves for the a-axis and the c-axis crystals under various pressures are shown in Fig.2 (a) and (b), respectively. The residual resistivities ρ_0 which are pressure dependent as will be shown in Fig.6 are subtracted. Correction of the resistivity for the changes in sample dimensions by pressure was made by using the compressibility data obtained from elastic measurement [14)] above the Néel temperature. The a-axis resistivity decreases

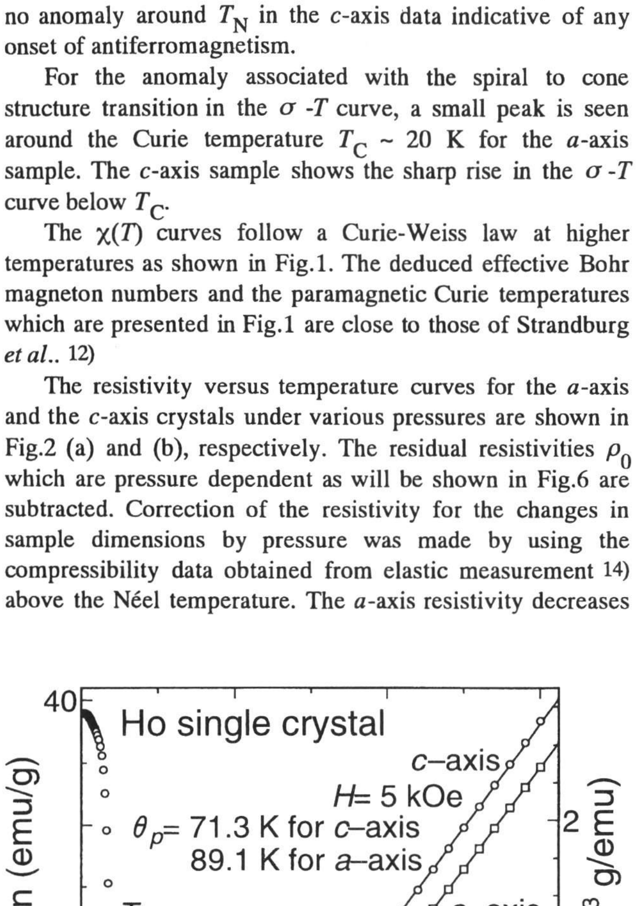

Fig.1 Magnetization and reciprocal magnetic susceptibility of Ho single crystals as a function of temperature. The dashed lines represent linear fits through the experimental data.

below T_N as the magnetic order increases in a similar way to that observed in Ni. Néel temperatures were determined from the intersection points of linear extrapolations of the $\rho(T)$ curves above and below the transition. The T_N at P=0 kbar determined in this way is 131 K in agreement with magnetization measurements. It is found that the Néel point is decreased with increasing pressure. And the spin disorder resistivity is also decreased with pressure. The Curie temperature T_C was determined from the points in which appreciable change in slope appears in the $\rho(T)$ curve.

For the c-axis resistivity, it is seen that there is a sharp rise below T_N and a maximum is followed at lower temperatures (Fig.2 (b)). We define the Néel temperature for the c-axis sample by the intersection of linear $\rho(T)$ curves in the paramagnetic region below 200 K and the sharp rise below the transition as illustrated in Fig. 2(a). The T_C was determined in the same way as for the a-axis.

4. Discussion

4-1 Pressure dependence of magnetic ordering temperatures; $T_N(P)$ and $T_C(P)$

The variation of T_N with pressure up to 20 kbar is presented in Fig.3. It is found that with increasing pressure the Néel temperature is decreased with upward curvature. The initial pressure derivative of T_N is - 0.47 K / kbar for both the a-axis and the c-axis. This rate should be compared with previous works listed in Table 1. Our results are very closed to that obtained by Bloch and Pauthenet[1,2)], McWhan *et al.*.[3)] and Achiwa *et al.*[11)] Figure 4 gives the higher pressure results up to 60 kbar for the c-axis sample together with the data obtained by a piston cylinder. T_N changes smoothly as a function of pressure. Any unusual phenomena such as pressure-induced structural transitions do not occur.

In Fig.5 is shown the variation of T_C with pressure up to 20 kbar for the a-axis and the c-axis samples. The Curie temperature is also a decreasing function of pressure. The initial pressure derivative of T_C is - 0.10 K / kbar. It should be noted that this is the first pressure experiment on the spiral to cone structure transition temperature in holmium. For Tb,[6)] Dy[4,7)] and Er,[7)] negative dT_C /dP was also reported, but any interpretations have not been given for this point.

4-2 Pressure dependence of residual resistivity, phonon resistivity and spin disorder resistivity; $\rho_0(P)$, $\rho_{ph}(P)$ and $\rho_{sd}(P)$.

Miwa [15,16)] and Elliott and Wedgwood [17,18)] developed the theories to account for the anomalous temperature dependence of resistivity below T_N of the heavy rare earth metals. They took into account the effect of the spiral spin structure as well as the fluctuations in the spin system to calculate the resistivity. In their theories the resistivities in the c-plane and along the c-axis are respectively given by

$$\rho_a(T) = \rho_0' + \rho_{ph}'(T) + \rho_{sd}'(T)\,, \tag{1}$$

$$\rho_c(T) = \frac{\rho_0 + \rho_{ph}(T) + \rho_{sd}(T)}{1 - \Gamma M}\,, \tag{2}$$

where ρ_0 is the residual resistivity, $\rho_{ph}(T)$ the phonon term and $\rho_{sd}(T)$ is the spin disorder resistivity which may be approximated[19)] as

$$\rho_{sd}(T) = A V^2(1 - M^2)\,. \tag{3}$$

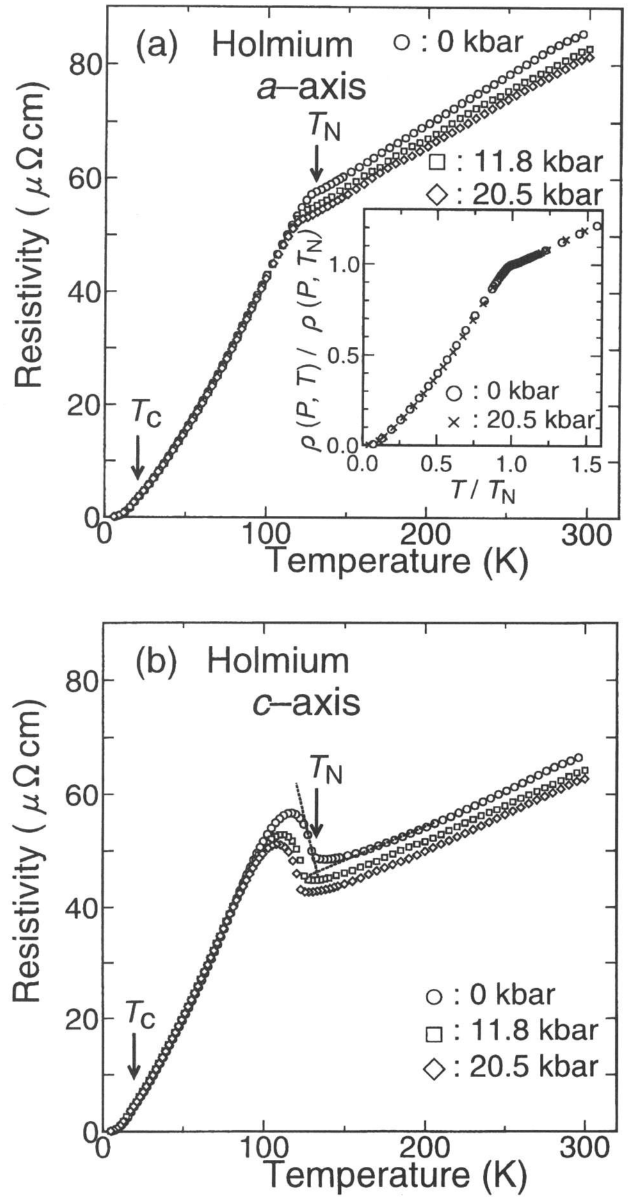

Fig.2 Electrical resistivity of Ho single crystal as a function of temperature and pressure: (a) for the *a*-axis; the inset gives the reduced resistivity as a function of reduced temperature, and (b) for the *c*-axis.

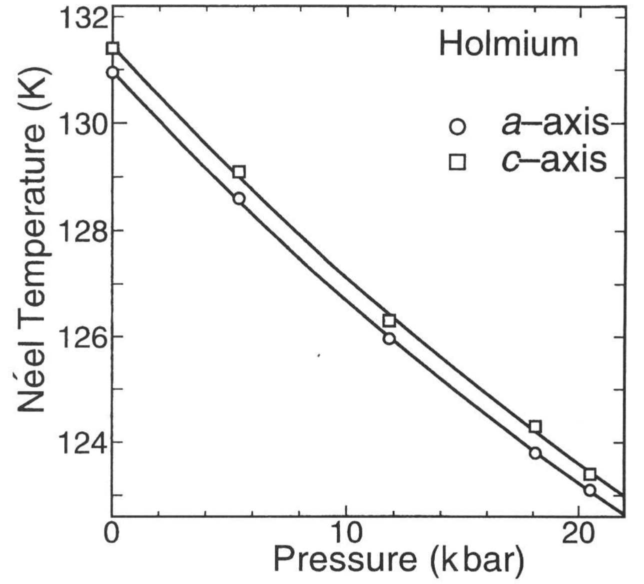

Fig.3 The variation of the Néel temperature with pressure up to 20 kbar for the *a*-axis and the *c*-axis Ho single crystals.

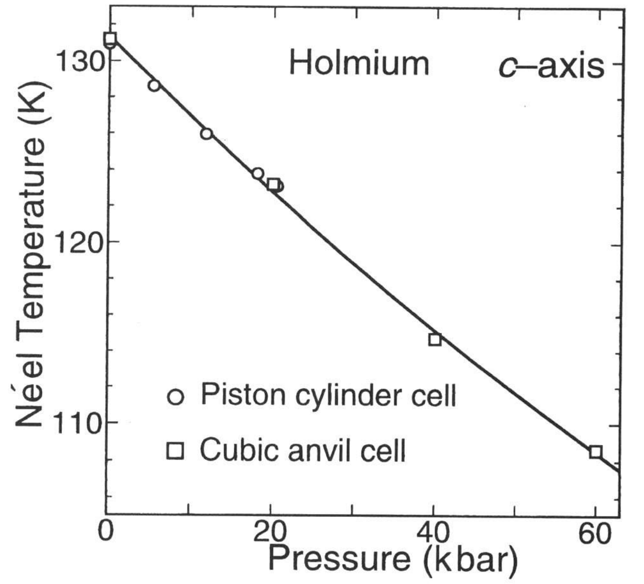

Fig.4 The variation of the Néel temperature with pressure up to 60 kbar for the *c*-axis Ho single crystal.

Γ is the correction factor to the resistivity due to the change in the Fermi surface brought about by the new superzone boundaries and $M(T)$ is the normalized magnetization. V is an effective exchange energy between a conduction electron spin and 4f electron spin S. It is noted that eq.(1) express the resistivity when the effect of gap opening is absent.

At T= 0 K, $\rho_{ph}(T)$ is 0. Moreover M should be 1 and then $\rho_{sd}(T)$ be zero, which gives

$$\rho_c(0) = \frac{\rho_0}{1 - \Gamma}. \tag{4}$$

The residual resistivity ρ_0 was determined by a least square fit of the data below 10 K to the T^3 law. The results are given in Fig.6. ρ_0 for both directions at P= 0 are about twice as large as those of Strandburg *et al.*; 12) 5.22 μΩcm for the *a*-axis and 7.17 μΩcm for the *c*-axis. It is found that ρ_0 is increased with pressure for the *c*-axis while for the *a*-axis it is decreased; relative pressure derivative of ρ_0 is - 3.5 × 10 $^{-3}$ kbar $^{-1}$ and + 3.7 × 10 $^{-3}$ kbar $^{-1}$, respectively. Positive pressure dependence for the *c*-axis suggests that Γ is enhanced with increasing pressure. However, the fact that there exists the spiral-to-cone structure transition at T_C = 20 K does not allow definite estimation of the change in Γ with pressure from the $\rho_0(P)$ data.

Figure 7 shows the pressure dependence of the slope of the phonon resistivity against temperature, $d\rho_{ph} / dT$. Here we assume that a T independent spin disorder resistivity, ρ_{sd}, and a linear ρ_{ph} - T relation hold for the paramagnetic region. It is found that the phonon resistivity is suppressed by pressure; the variation of the slope with pressure is larger for the *a*-axis by a factor of 3 than that for the *c*-axis. This suppression of ρ_{ph} should be attributed to an increase in the

Table 1. Pressure coefficients of the Néel temperature T_N and Curie temperature T_C of Ho.

Reference	Method	dT_N/dP (K/kbar)	dT_C/dP (K/kbar)	P range (kbar)
Bloch and Pauthenet[1,2]	ac susceptibility	-0.45 ± 0.15		0 - 6
McWhan and Stevens [3]	ac susceptibility	-0.48 ± 0.01		4 - 71
Kawai *et al.* [8]	electrical resistance	-0.5 ± 0.1		0 - 18
Umebayashi *et al.* [9]	neutron diffraction	-0.33 ± 0.05		0 - 6
Okamoto *et al.* [10]	ac susceptibility	-0.40		0 - 5
Achiwa *et al.* [11]	neutron diffraction	-0.48		0 - 21
Present study	electrical resistance	-0.47 ± 0 .01	-0.10 ± 0.02	0 - 20/60

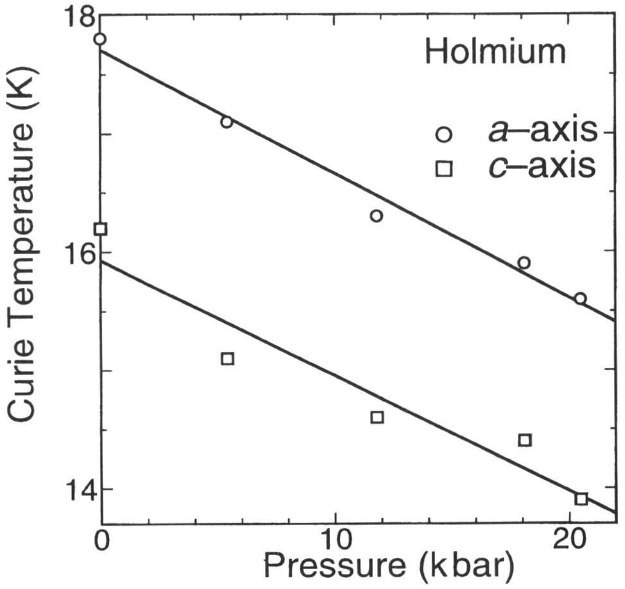

Fig.5 The variation of the Curie temperature with pressure up to 20 kbar for the *a*-axis and the *c*-axis Ho single crystals.

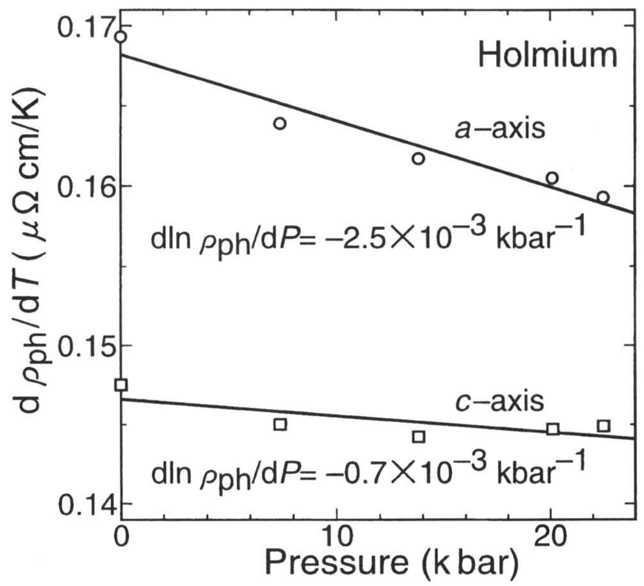

Fig.7 The variation of the slope of the phonon resistivity against temperature, $d\rho_{ph}/dT$, with pressure up to 20 kbar for the *a*-axis and the *c*-axis Ho single crystals.

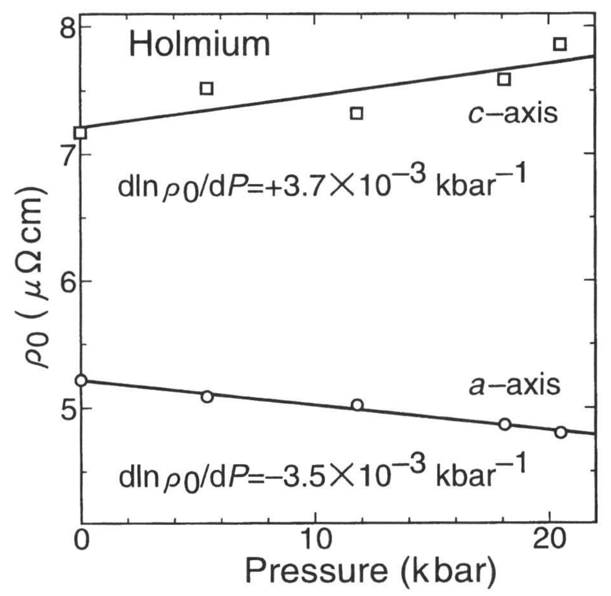

Fig.6 The variation of the residual resistivity ρ_0 with pressure up to 20 kbar for the *a*-axis and the *c*-axis Ho single crystals.

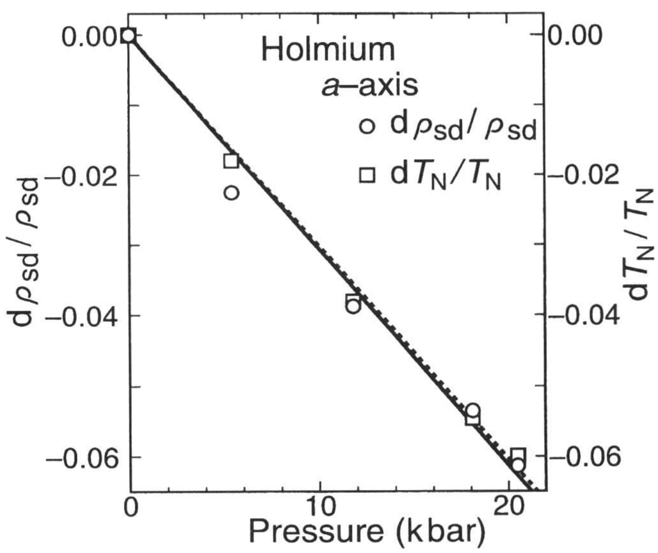

Fig.8 Relative changes in the spin disorder resistivity, $d\rho_{sd}$ / ρ_{sd}, and the Néel temperature, dT_N/T_N, with pressure up to 20 kbar for the *a*-axis Ho single crystal.

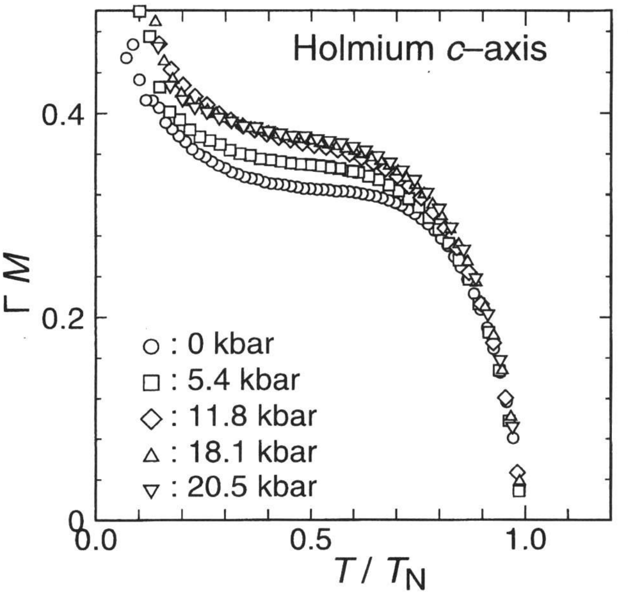

Fig.9 The variation of ΓM with the reduced temperature at various pressures up to 20 kbar for the c-axis Ho single crystal.

Debye temperature with pressure in the Grüneisen-Bloch model. However we can not explain the difference in the pressure response of ρ_{ph} for these two principal axes.

To estimate the effect of pressure on the spin disorder resistivity may give an indispensable basis from which the volume dependence of the exchange interaction between the conduction electron spin and the localized 4f spin can be determined in holmium. The a-axis resistivity is expressed by eq.(1). The $\rho_{\mathrm{sd}}(T)$, which is pressure dependent, is obtained by subtracting both the residual resistivity and the phonon resistivity from the observed data as

$$\rho_{\mathrm{sd}}(P,T) = \rho(P,T) - \rho_0(P) - \rho_{\mathrm{ph}}(P,T). \qquad (5)$$

Very interestingly, if $\rho_{\mathrm{sd}}(P,T)$ for the a-axis is normalized to its $\rho_{\mathrm{sd}}(P,T_N)$, it scales with $T/\ T_N$ for temperatures both above and below the Néel temperature. The scaling behavior is illustrated in the inset of Fig.2(a) where the resistivities at 0 kbar and 20.5 kbar are plotted as representatives. This finding suggests that the pressure dependence of these two parameters, ρ_{sd} and T_N, should be related to each other through the change in the exchange energy with pressure. Figure 8 shows relative changes in the $\rho_{\mathrm{sd}}(T_N)$ and T_N with pressure. It is found that the spin-disorder resistivity at T_N is also a decreasing function of pressure and that its relative change is the same as that of the Néel temperature, - 3.0 × 10^{-3} kbar^{-1}. Using eq.(3), where A is assumed to be independent on pressure, $\mathrm{d}\ln V\ /\ \mathrm{d}P$ is given - 1.5 × 10^{-3} kbar^{-1}, i.e., $\Delta V/V$ = - 3 % at P=20 kbar. This rate is compared with the results of the neutron diffraction work by Achiwa *et al.*[11] They claimed $\Delta V/V$ = - 12 % at P=21 kbar from the analysis of the pressure dependence of spiral turn angle on the basis of the Miwa's theory.[16] The difference in the values of $\Delta V/V$ determined by two ways is not so surprising. As Miwa pointed out in ref. 16), the theory is based on the free electron model. Furthermore the contours of equal turn angle which Achiwa *et al.* used for the estimation of $V(P)$ can not be drawn quite uniquely.

4-3 Pressure dependence of the enhancement factor Γ.

The determination of the spin disorder resistivity $\rho_{\mathrm{sd}}(T)$ for the c-axis is another main problem. As the $\rho_{\mathrm{sd}}(T_N)$ for the c-axis is also a decreasing function of pressure as seen in Fig.2(b) and the $\rho_{\mathrm{sd}}(P,T)$ for the a-axis scales very well with $T/\ T_N$, the $\rho_{\mathrm{sd}}(P,T)$ for the a-axis is used to determine the c-axis $\rho_{\mathrm{sd}}(P,T)$. At $T=T_N$ the ΓM in the denominator of the right hand side of eq.(2) is 0 and the c-axis $\rho_{\mathrm{sd}}(P,T_N)$ is scaled with the a-axis $\rho_{\mathrm{sd}}(P,T_N)$. Thus the measured $\rho_c(P,T)$ data are fitted as

$$\rho_c(P,T) = \frac{\rho_0(P) + \rho_{\mathrm{ph}}(P,T) + \rho_{\mathrm{sd}}(P,T/T_N)}{1 - \Gamma M}. \qquad (6)$$

Thus the parameter ΓM is determined. The results are shown as a function of reduced temperature in Fig.9. ΓM is found to be increased largely up to 11.8 kbar and to be less sensitive to pressure in higher pressure region. The rapid increase in ΓM with decreasing temperature is found approximately at T_C. Keeping in mind that M is the normalized magnetization and if $M(T)$ is assumed to be pressure independent for the localized spin system, we may conclude that the quantity Γ is increased with increasing pressure; 0.32 at P= 0 to 0.4 at P= 20 kbar.

In the theories,[15-18] the total energy of the conduction electrons depends on both the magnitude of the energy gap along new Brillouin zone boundaries, $\Delta = VSMF(\boldsymbol{\tau})$, in which $F(\boldsymbol{\tau})$ is the structure factor, and the positions of the new superzone boundaries, $2\boldsymbol{l} = \boldsymbol{\tau} + \boldsymbol{q}$, where $\boldsymbol{\tau}$ is any reciprocal lattice vector and $\boldsymbol{q}$ is the wave vector which describes a spiral spin structure. The correction factor ΓM is given by a function of the above two terms as in eq.(32) in ref.18),

$$\Gamma M = \sum_l \delta\left(\frac{|l|}{k_f}, \Delta(l)\right)\left(\frac{l_z}{l}\right)^2. \qquad (7)$$

Notations are referred to ref. 18). If we accept the analysis of $\rho_{\mathrm{sd}}(T_N)$ and T_N (P) made in section *4-2*, Δ is also a decreasing function of pressure, which suggests that the change with pressure of the position of the new Brillouin zone boundaries must be considered for detailed interpretation of the gap formation. In Fig. 6 of ref.19), ΓM was calculated on the basis of the free electron model to result in that ΓM is increased as Δ is decreased. Thus we conclude that the enhancement of ΓM with pressure may be interpreted by the existing theories.[15-18] However, the detailed geometry of Fermi surface and its distortion by pressure are still unknown in Ho.

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