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## EFFECTS OF PRESSURE ON THE HELICAL TURN ANGLE OF HOLMIUM

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**Abstract.** – The pressure dependence of the helical turn angle of a holmium single crystal has been determined by neutron diffraction up to 2.1 GPa in the temperature range 10 K to  $T_N$ . It is found that the energy gap of conduction electrons at the magnetic superzone boundary decreases with increasing pressure on the basis of the free electron model.

It is generally accepted that helical magnetic ordering in rare-earth metals and alloys is attributed to the indirect RKKY exchange interaction [1] between localized 4f spins through the conduction electrons (s-f exchange interaction). The helical turn angle corresponds to the wavevector that gives the maximum  $q$ -dependent susceptibility  $\chi(q)$  of conduction electrons; the angle is hence sensitive to the band character. Elliot and Wedgwood [2] studied, on the basis of the free electron model, the redistribution of conduction electrons across the energy gaps at the boundary of the superzone produced by the helical magnetic order. For rare earth metals, however, the situation differs from that of the free electron model. Recently, Kaino and Kasuya [3] showed the effect of the non-linear s-f exchange interaction through a flat part of the Fermi surface, on the helical-ferromagnetic phase transition in rare-earth alloys.

The properties of the 5d6s band character such as the geometry of the Fermi surface, the number of carriers, the density of states at the Fermi level etc. are important factors in understanding the exchange interaction mechanism in rare-earth metals. The effects of pressure on the helical turn angle and on the magnetic ordering temperature can be observed through a change in the conduction band; the effects will give important information, although the precise experimental data of the Fermi surface are not yet sufficient for rare-earth metals.

A high-pressure cell of hot-pressed  $\text{Al}_2\text{O}_3$  cylinder was designed by Onodera *et al.* [4]. A single crystal of holmium prepared with a usual strain-anneal method was placed in an aluminium micro-capsule together with a single crystal of NaCl; the pressure was calibrated by measuring the lattice compression of NaCl. The neutron diffraction experiment under high pressure was carried out by using a double-axis diffractometer at KUR; the wavelength of monochromatized neutrons was 1.006 Å, the  $\lambda/2$  contamination being not more than 0.2 %.

The temperature dependence of helical turn angles was determined in the temperature range 10 K to  $T_N$  by measuring the separation of the magnetic satellites of the (002) nuclear reflection. The results for the pressure range 0.1 MPa to 2.1 GPa are plotted in figure 1. The pressure dependence of the turn angles is largest at low temperatures;  $T_N$  at 2.1 GPa is 122.0 K, which gives a pressure dependence of  $dT_N/dP = -4.8$  K/GPa.

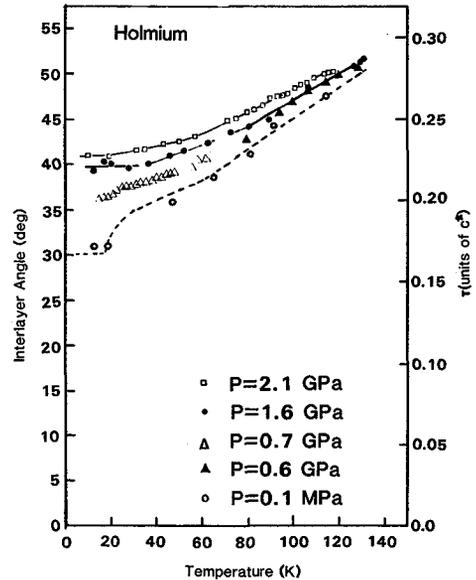


Fig. 1. – Temperature dependence of the helical turn angle for holmium at several pressures together with the experimental results by Koehler *et al.* [5] at 0.1 MPa (dashed line) and by Umebayashi *et al.* [6] at 0.6 GPa (▲). Solid lines are just guides to the eye.

According to the free-electron model, the temperature variation of helical turn angles depends on the two parameters [7]: the energy gap at the magnetic

superzone boundary  $\omega = MV/E_F$  and the spin-disorder scattering parameter  $\gamma = (9\pi/8)(S^2 - M^2)(V/E_F)^2$ . Here  $S$  and  $M$  are the spin magnetic moments at 0 and  $T$  K respectively,  $V$  the exchange potential and  $E_F$  the Fermi energy of conduction electrons. In figure 2, the helical turn-angle is plotted along  $t = T/T_N$  line on the "equal turn-angle contours" on the  $(M^2, (S^2 - M^2))$  plane given by Miwa [7]. On this plot, the variable on the abscissa is not  $M^2$  but  $(V/E_F)^2$  because the exchange potential is modified by the pressure. Consequently, the variation of helical turn angle at any pressure can be expressed by one straight line. The lines for different pressures have different inclinations but converge at the (0,4) point in the figure. At 2.1 GPa the line crosses the abscissa at (3,0) instead of (4.0) at 0.1 MPa. This means that the squared exchange-potential at 2.1 GPa is 75 % of that at 0.1 MPa.

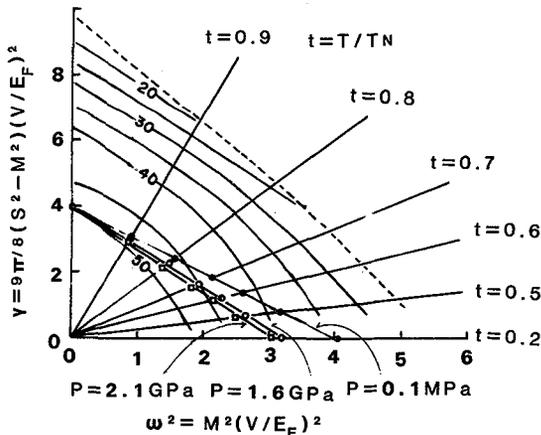


Fig. 2. - Contours of equal turn angles (in degree) at 0.1 MPa plotted in  $(M^2, (S^2 - M^2))$  plane at 0.1 MPa (Miwa [7]). The turn angles at higher pressures are plotted along  $t = T/T_N$  line where  $(S^2 - M^2)/M^2 = \text{constant}$  but the  $(V/E_F)$  is modified with pressure in the same figure.

The pressure dependence of helical turn angles is almost linear above 30 K; below 30 K, on the other hand, the initial derivative of the turn angle with pressure becomes large and seems to approach a smaller constant value above 0.7 GPa.

The non-linear pressure dependence of the helical turn angle below 30 K would reflect a rapid disappearance of the flat Fermi surface when the pressure increases. This suggests that the non-linear s-f exchange interaction with a flat Fermi surface predicted by Kaino and Kasuya for holmium below 20 K disappears at high pressures.

Below 20 K the helical turn angles are temperature independent; the lock-in turn angles are  $31^\circ$ ,  $36^\circ$ ,  $40^\circ$  and  $41^\circ$  at 0.1 MPa, 0.7 GPa, 1.6 GPa and 2.1 GPa, respectively. Recent experiments of magnetic scattering with synchrotron radiation have shown the presence of spin slips in the helical magnetic structure of holmium [8]. That is, a clustering of the helical turn angles was observed around 20 K,  $30^\circ$  (wavevector =  $2/10$ ),  $33.33$  ( $5/27$ ) and  $32.73$  ( $2/11$ ) and the last peak was due to a modulation of the lattice by spin defect. All the observed lock-in turn angles  $31^\circ$ ,  $36^\circ$  ( $2/10$ ),  $40^\circ$  ( $2/9$ ) and  $41^\circ$  should be commensurate with the crystal lattice. The turn angles  $31^\circ$  and  $41^\circ$  can be attributed to a structure with a spin defect for every 36 layers: the values respectively correspond to 3 periods of 12 layers,  $6/35$ , and 4 periods of 9 layers,  $8/35$ . The resolution of the present experiment is not sufficient for distinguishing a periodic spin slip from a random one.

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