Pressure Effect on Magnetic Transitions of CrSb_2 and $\text{Cr}_{0.8}\text{Fe}_{0.2}\text{Sb}_2$

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We have examined the pressure effect on the Néel temperature for CrSb_2 and $\text{Cr}_{0.8}\text{Fe}_{0.2}\text{Sb}_2$ by measuring the temperature dependence of electrical resistivity ρ . The Néel temperature T_N of both compounds decreases linearly with increasing pressure. The pressure derivative of T_N , dT_N/dp are evaluated -1.0 K/kbar for CrSb_2 and -0.6 K/kbar for $\text{Cr}_{0.8}\text{Fe}_{0.2}\text{Sb}_2$. [pressure effect, Néel temperature, insulating magnetic compound, electrical resistivity, CrSb_2]

1. Introduction

Transition metal dipnictide CrSb_2 has an orthorhombic marcasite type structure as shown in Fig. 1. It is a semiconductor with energy gap $E_G = 0.14$ eV [1] and antiferromagnet with the Néel temperature $T_N = 273$ K [2]. According to the neutron diffraction study [2], the magnetic moment of the Cr atom is $1.94 \mu_B$ at 80 K. The magnetic moments are coupled ferromagnetically within (011) plane of chemical cell and the magnetic moments in the adjacent planes are antiparallel. The effects of substitution of Fe for Cr on the magnetic properties were investigated by Kjekshus et al. [2]. Lattice parameters a , b , c and unit cell volume V decrease with increasing Fe content x . The Néel temperature T_N decreases with x . Antiferromagnetism of $\text{Cr}_{1-x}\text{Fe}_x\text{Sb}_2$ disappears near $x=0.5$. These results suggest that the Néel temperature decreases with decrease

of unit cell volume or application of pressure. However, many investigations show that T_N for insulating antiferromagnets increases with pressure. It is uncertain whether the decrease of T_N and the disappearance of antiferromagnetism with x is due to the chemical effect or the volume effect by Fe substitution for Cr. In this study, we have examined the pressure effect on T_N for CrSb_2 and $\text{Cr}_{0.8}\text{Fe}_{0.2}\text{Sb}_2$ to obtain an information for the volume dependence of T_N in $\text{Cr}_{1-x}\text{Fe}_x\text{Sb}_2$.

Fig.1. The crystal structure of CrSb_2 and $\text{Cr}_{0.8}\text{Fe}_{0.2}\text{Sb}_2$.

Fig.2. The temperature dependence of the electrical resistivity ρ for CrSb_2 .

2. Experimental

Sintered samples of CrSb_2 and $\text{Cr}_{0.8}\text{Fe}_{0.2}\text{Sb}_2$ were prepared by solid-state reaction from stoichiometric mixture of Cr (99.99%), Fe (99.9%) and Sb (99.9999%). The mixture was heated at 650 °C in a vacuum silica tube for two weeks. The reaction products were then reground, pressed into pellets and heated again at 600 °C. The X-ray diffraction shows that the prepared samples are in single phase. The lattice parameters are obtained to be $a=6.0184$ Å, $b=6.8604$ Å, $c=3.2670$ Å, $u=0.180$ and $v=0.364$ for CrSb_2 and $a=5.9915$ Å, $b=6.7956$ Å, $c=3.2535$ Å, $u=0.179$ and $v=0.362$ for $\text{Cr}_{0.8}\text{Fe}_{0.2}\text{Sb}_2$. These lattice parameters of CrSb_2 and $\text{Cr}_{0.8}\text{Fe}_{0.2}\text{Sb}_2$ are in agreement with those in Ref.[2]. The hydrostatic pressure was applied to the sample in a Teflon pressure cell filled with a Fluorinert by using the piston-cylinder type device. The pressure change of the Néel temperature was determined by measuring the temperature dependence of electrical resistivity ρ under various pressures.

Fig.3. The pressure dependence of T_N for CrSb_2 .

3. Experimental results

Figure 2 shows the temperature dependence of ρ for CrSb_2 at various pressures. As seen in the figure, ρ decreases with increasing temperature and the slope of ρ - T curve changes through T_N . The Néel temperature T_N was defined as the temperature of the intersection of interpolations of the curve above and below T_N as shown in the figure. It was found that ρ increases with pressure. Figure 3 shows the pressure dependence of T_N for CrSb_2 . The Néel temperature decreases almost linearly with pressure.

The pressure derivative and coefficient of T_N are evaluated as $dT_N/dp = -1.0$ K/kbar and $(1/T_N)(dT_N/dp) = -3.6 \times 10^{-3}$ 1/kbar for CrSb_2 . Figure 4 shows the temperature dependence of ρ for

Fig.4. The temperature dependence of the electrical resistivity ρ for $\text{Cr}_{0.8}\text{Fe}_{0.2}\text{Sb}_2$.

$\text{Cr}_{0.8}\text{Fe}_{0.2}\text{Sb}_2$ at various pressures. As seen in the figure, ρ

Fig.5. The pressure dependence of T_N for $\text{Cr}_{0.8}\text{Fe}_{0.2}\text{Sb}_2$.

decreases with increasing temperature and the slope of p - T curve changes through T_N as well as the case of CrSb_2 . Figure 5 shows the pressure dependence of T_N for $\text{Cr}_{0.8}\text{Fe}_{0.2}\text{Sb}_2$. The Néel temperature decreases almost linearly with pressure. The pressure derivative and coefficient of T_N are evaluated as $dT_N/dp = -0.6$ K/kbar and $(1/T_N)(dT_N/dp) = -3.8 \times 10^{-3}$ 1/kbar for $\text{Cr}_{0.8}\text{Fe}_{0.2}\text{Sb}_2$. It should be noted that the pressure coefficient of T_N of both compounds is almost same.

Fig.6. The x dependence of V and T_N . Triangle and circle are the data in Ref.[2] and [5], respectively.

4. Discussion

Figure 6 shows the x dependence of V and T_N [2,5]. From the results in the figure, we can estimate the pressure derivative of T_N to be $dT_N/dp = -5.4$ K/kbar, if we assume that the decrease of T_N with x is due to only the decrease of V with x . The pressure shift of T_N estimated above is much larger than those of observed directly in this experiment. Therefore, the decrease of T_N with x

is considered to be mainly due to the chemical effect by Fe substitution for Cr.

It is known that many non-metallic antiferromagnets of transition metal compounds show the positive pressure derivative of T_N as mentioned. Bloch reported that there is an empirical rule for the relationship between the magnetic transition temperature T_N and pressure in most insulating magnetic compounds: $dT_N/dp = (10/3) \kappa T_N$, where κ expresses compressibility [3]. The main part of the exchange interactions of insulating antiferromagnets is the superexchange interaction. In that case, the Néel temperature T_N is expressed by the following equation,

$$T_N = A (t^2/U),$$

where A is constant, t is transfer integral and U is Coulomb interaction, respectively. Kanomata et al. have reduced $dT_N/dp = (10/3) \kappa T_N$ by taking account of the pressure dependence of t ($dt/dp > 0$) on the basis of the above equation, where they assumed for dU/dp to be zero [4]. If dU/dp is positive, we can expect the negative pressure derivative of T_N as the results of the competition between dt/dp and dU/dp . Therefore, the negative dT_N/dp observed in this study suggests that the effect of dU/dp overcomes that of dt/dp in the present compounds.

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References

- [1] K.Adachi, K.Sato, and M.Matsuura, *J.Phys.Soc.Jpn.*, **26**, 906 (1969).
- [2] A.Kjekshus, P.G.Peterzens, and T.Rakke, A.F.Andersen, *Acta Chemica Scandinavia*, **A33**, 469 (1979).
- [3] D.Bloch, *J.Phys.Chem.Solids*, **27**, 881 (1966).
- [4] T.Kanomata, K.Shirakawa, and T.Kaneko, *J.Phys. Soc.Jpn.*, **54**, 334 (1985).
- [5] T.Harada, to be published.