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INTERLAYER COUPLING STRENGTH IN SUPERCONDUCTORS,  $2H-TaS_2$  AND  $2H-TaS_2(py)_{1/2}$

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Résumé.- La température de transition et le champ critique supérieur parallèle et perpendiculaire au plan des couches ont été mesurés sur plusieurs monocristaux de  $2H-TaS_2$  et  $2H-TaS_2(py)_{1/2}$ . La supraconductivité dans ces deux types de matériaux est analysée sous l'aspect de la force de couplage intercouche utilisée par Klemm et al. dans la théorie du couplage Josephson.

Abstract.- The transition temperature and the upper critical field parallel and perpendicular to the layer plane are measured on several single crystals of  $2H-TaS_2$  and of  $2H-TaS_2(py)_{1/2}$ . Superconductivity in these two kinds of materials is discussed from the viewpoint of the interlayer coupling strength used in the Josephson coupling theory by Klemm et al.

For several years, we have studied the superconducting properties of single crystals of  $2H-NbSe_2$  by measuring the upper critical field,  $H_{c2}$  /1/, and the specific heat /2/. In the present paper, we describe on the transition temperature,  $T_c$ , and the temperature and angular dependences of  $H_{c2}$  on several single crystals of  $2H-TaS_2$  and of  $TaS_2(py)_{1/2}$ , where the latter means layered single crystal  $2H-TaS_2$  and of  $TaS_2(py)_{1/2}$ , where the latter means layered single crystal  $2H-TaS_2$  containing pyridines intercalated between dichalcogenide planes. We discuss the difference of interlayer coupling strength between these two kinds of superconducting compounds on the basis of our experimental results.

Single crystals of  $2H-TaS_2$  were prepared by a method of chemical vapor transport reactions using iodine as a carrier. X-ray Weissenberg photographs and electron diffraction patterns showed that our crystals are of 2H polytype and gave no indication of mixtures of polytypes such as 1T, 4Hb and 6R. The intercalation procedure of pyridines to the crystal is similar to that by Thompson /3/. The measurements were done by the electrical conduction method down to 0.4 K.

Although  $T_c$  of pure  $2H-TaS_2$  has been reported to be 0.8 to 1.2 K by several researchers, no reliable data exist at present. In figure 1 is shown  $R(T)/R(4.2 K)$  as a function of T for one of our best samples,  $2H-TaS_2$  (RRR = 95), together with  $2H-TaS_2(py)_{1/2}$ . The transition width,  $\Delta T_c/T_c$ , is pretty broad even in the smallest current density for the pure samples, where  $T_c$  is defined by the extrapolation of the middle temperature of the transition to zero current. This result is in contrast

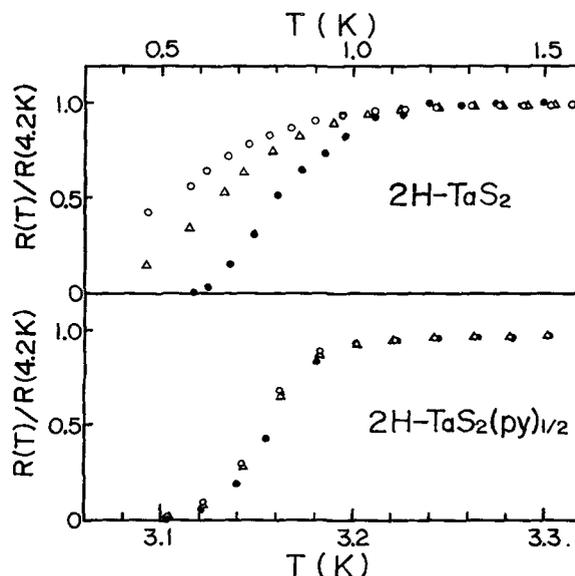


Fig. 1 :  $R(T)/R(4.2 K)$  versus T curves for the pure crystal of  $2H-TaS_2$  and the intercalated crystal of  $2H-TaS_2(py)_{1/2}$ . Results for three currents through samples are shown. For  $2H-TaS_2$ , ( $2H-TaS_2(py)_{1/2}$ ), ●, Δ and ○ are data of 1(1), 5(2) and 10(3) mA, respectively. For  $2H-TaS_2$  of RRR = 95,  $T_c$  is determined to be 0.86 K. (See text). For  $2H-TaS_2(py)_{1/2}$ ,  $T_c = 3.16 K$  and  $\Delta T_c \approx 60 mK$ .

to the relatively narrow  $\Delta T_c/T_c$  of the intercalated compounds. It is considered that the occurrence of the structural transformation such as C. D. W. or the presence of large internal strain introduced in transition between polytypes in making the 2H-structure will give a strong effect to the superconducting to normal transition, especially, in the case of the pure  $2H-TaS_2$ . From such considerations,  $T_c$  of  $2H-TaS_2$  of good quality is expected to be less than

0.9 K, while  $T_c$  of  $2\text{H-TaS}_2(\text{py})_{1/2}$  in  $12.01 \text{ \AA}$ -phase /3/ is about 3.1 to 3.2 K.

In figure 2 is shown  $H_{c2}$  as a function of  $T$  for a  $2\text{H-TaS}_2$  crystal with the highest purity obtained and for a  $2\text{H-TaS}_2(\text{py})_{1/2}$  compound. The data

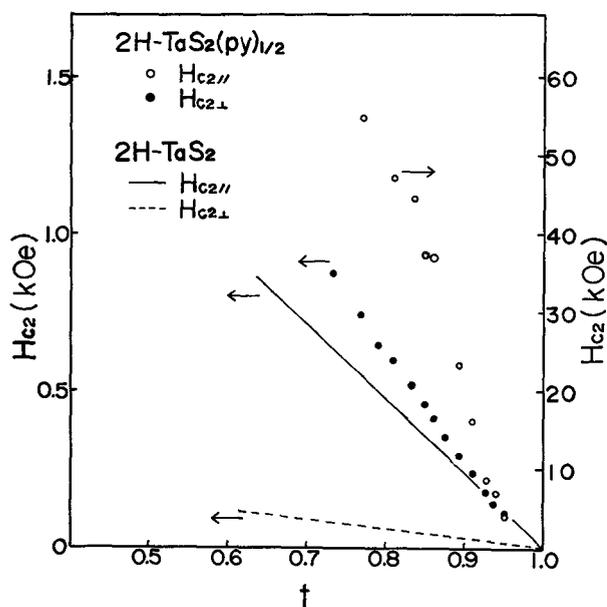


Fig. 2 :  $H_{c2}$  versus ( $t = T/T_c$ ) curves for  $2\text{H-TaS}_2(\text{py})_{1/2}$  and  $2\text{H-TaS}_2$  (the same one as fig. 1), where  $H_{c2//}$  and  $H_{c2\perp}$  mean  $H_{c2}$  parallel and perpendicular to the layer plane, respectively. For  $2\text{H-TaS}_2(\text{py})_{1/2}$ ,  $\circ$  and  $\square$  are  $H_{c2//}$  and  $H_{c2\perp}$ , and for  $2\text{H-TaS}_2$ , — and --- express the  $H_{c2//}$  and  $H_{c2\perp}$  behaviors, respectively.

of pure crystals should be considered to be preliminary because of the broad width of  $T_c$  and the narrow temperature region in the  $^3\text{He}$  cryostat used. It is emphasized that  $H_{c2//}/H_{c2\perp}$  is quite large and the behavior of  $H_{c2//}(T)$  near  $T_c$  is characteristic of positive curvature for  $2\text{H-TaS}_2(\text{py})_{1/2}$ . The characteristics are similar to pure  $2\text{H-NbSe}_2$  /1/ to some extent, and much more remarkable both in magnitude and in temperature dependence. Though not shown in figure 2,  $H_{c2//}/H_{c2\perp}$  of  $2\text{H-TaS}_2$  decreases with increasing purity, but the anisotropy is still pretty large even in the purest sample.

In order to study the superconducting coupling strength between layers, the Klemm theory on the basis of the Josephson phase-coupling model /4/ is used for these quasi-two-dimensional layer superconductors. The temperature dependence of  $H_{c2//}$  for the low-field limit (1) and high-field one (2) is given as follows, assuming the dirty case within

the layer,

$$H_{c2//}(T) = (M/m)^{1/2} (4\phi_0/\pi^2\hbar D)k_B(T_c - T), \quad (1)$$

and

$$H_{c2\perp}(T) = (m/M) (\phi_0/4s^2\pi) (\pi\hbar D)^{1/2} (T - T_c^*)^{-1/2} \quad (2)$$

where  $T_c^* = T_c(1 - \pi\gamma/8)$ ,  $M/m = (H_{c2//}/H_{c2\perp})^2$  near  $T_c$  and  $D$  is the intralayer diffusion constant.

$\gamma = (2m\hbar D/Ms^2k_B T_c)$  is the coupling constant, where  $s$  is the separation between layers.

From our experimental results, we estimate  $\gamma$  as follows ;

for $2\text{H-TaS}_2(\text{py})_{1/2}$ ,	$\gamma \approx 2$ ,
for $2\text{H-TaS}_2$ ,	$\gamma \approx 2500$ ,
for $2\text{H-NbSe}_2$ ,	$\gamma \approx 100/1,2/$ .

For  $2\text{H-TaS}_2(\text{py})_{1/2}$ ,  $\xi_{\perp}$  is estimated to be  $7.6 \text{ \AA}$ . This is smaller than the layer spacing of  $12 \text{ \AA}$ .

We conclude that the Josephson coupling model in which the order parameter in adjacent layers is weakly coupled by Josephson tunneling can be applied to the intercalated superconductors such as  $2\text{H-TaS}_2(\text{py})_{1/2}$  because of the small  $\gamma$  and  $\xi_{\perp}$  values. On the other hand, the  $\gamma$  value of pure  $2\text{H-TaS}_2$  is so large that the Josephson coupling model cannot be applied to this material. The situation is similar to the  $2\text{H-NbSe}_2$  case. For nonintercalated layer compounds, the theory on anisotropic superconductors such as reference /5/ is newly necessary.

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