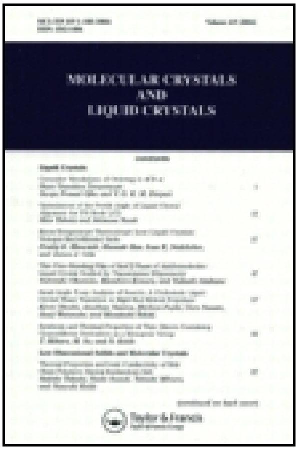

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Electrical Transport Properties Of $2H\text{-}TaS_2$ Intercalation Compounds With Variable Charge Transfer

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ELECTRICAL TRANSPORT PROPERTIES OF 2H-TaS_2 INTERCALATION COMPOUNDS WITH VARIABLE CHARGE TRANSFER

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Abstract We report on low temperature measurements of the electrical resistivity, the Hall coefficient and the magnetoresistance in hydrated potassium intercalation compounds of 2H-TaS_2, where the stoichiometry and hence the charge transfer can be varied.

INTRODUCTION

The electrical transport properties of 2H-MCh_2 layered compounds (M = Nb, Ta, Ch = S, Se) have been studied in detail[1], but only little is known about these properties in intercalation compounds. In the case of 2H-TaS_2 at $T \sim 78$ K a phase transition to a charge-density-wave (CDW) state occurs which is indicated by a small rise in the electrical resistivity ρ and a faster decrease of ρ below the onset temperature, in a change of the Hall coefficient from positive to negative sign and a cusp in the magnetic susceptibility. It is known that these anomalies are partially or totally suppressed by intercalation of organic molecules[2]. In most of these samples the amount of charge transfer is not known and its influence is therefore not discussed.

The hydrated potassium intercalation compounds of the formula $K_x(H_2O)_yTaS_2$ are comparable to the phases with organic molecules as far as the anisotropy of physical properties is concerned[3]. A complete charge transfer of the potassium 4s-electron to the conduction band of the TaS_2 is guaranteed by the relatively high layer separation (Δd = 3 Å), which is mainly caused by the water molecules. The electrochemical preparation technique allows to determine the stoichiometry x of the sample (and hereby the charge transfer) and to adjust it between x = 0.27 and 0.45 [3].

Here we report for the first time on measurements of the electrical resistivity ρ, the Hall coefficient R_H and the magnetoresistance of these samples as a function of charge transfer.

EXPERIMENTAL

Thin single crystals of 2H-TaS_2, prepared by the vapour phase transport reaction and a subsequent annealing procedure[3], were cut into a rectangular form (1.5 x 4 mm^2) with a razor blade. The electrointercalation of the hydrated potassium ions was carried out at room temperature in an aqueous solution of K_2SO_4 (0.5 m) under galvanostatic conditions. We used a four probe configuration and a dc-method for the measurements of ρ and R_H. All data reported here were taken in a transverse configuration, i.e. the current I parallel to the layers and the magnetic field perpendicular to the layers. In order to correct for misalignments of the contacts for the measurements of the magnetoresistance and Hall coefficient, the magnetic field was applied in both directions.

RESULTS AND DISCUSSION

The intercalation of K^+/H_2O into 2H-TaS_2 has only a small effect on the electrical resistivity ρ at room temperature, if the volume increase is neglected (2H-TaS_2: $\rho = 120\ \mu\Omega m$; $K_{0.33}(H_2O)_yTaS_2$: $\rho \sim 150\mu\Omega m$). On the contrary the temperature dependence of ρ is changed drastically, as is shown in fig. 1a for a sample with x = 0.33. The anomaly in ρ at CDW onset temperature in 2H-TaS_2 is completely wiped out in $K_{0.33}(H_2O)_yTaS_2$ (see insert of fig. 1a) and there is no obvious other anomaly to be seen. The residual resistance ratios RRR of the intercalated samples ranged from 20 to 35 (2H-TaS_2: RRR = 70 - 80) indicating a high quality of these compounds. Furthermore we analysed the resistivity at low temperatures ($T < 25$ K) and tried to fit the data by the usual formula $R = R_o + AT^p$. The ideal resistance $R-R_o$ for a sample with x = 0.33 is shown in fig. 1b together with a theoretical curve $R-R_o = AT^{2.9}$. The dependence of the exponent p on the stoichiometry x is given in the following

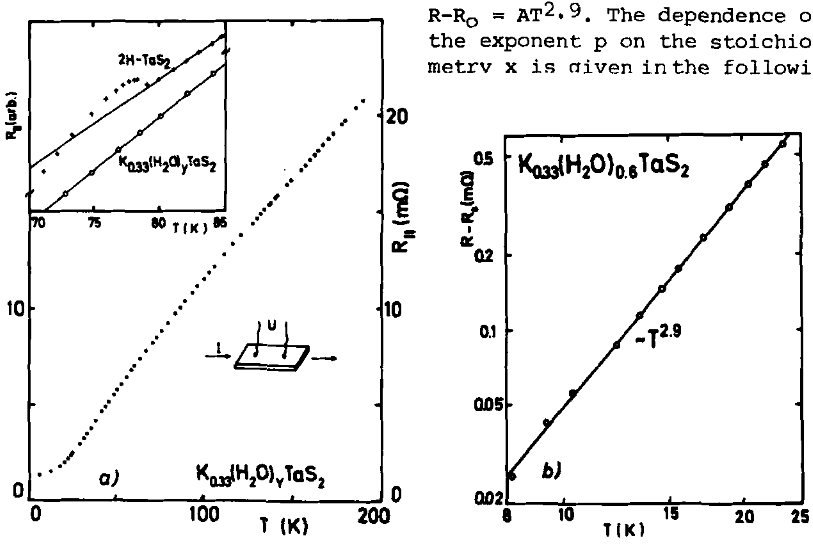

FIGURE 1 a) Electrical resistance as a function of temperature for a sample $K_{0.33}(H_2O)_yTaS_2$ ($y \sim 0.6$); insert: comparison with 2H-TaS_2 around 78 K; b) log-log plot of the ideal resistance (see text) table:

x	0.28	0.305	0.33	0.365	0.42	2H-TaS_2
p	3.0	3.0	2.9	3.4	3.4	4.0

A qualitative understanding of these results is possible in a model proposed by Naito and Tanaka[1] for the binary 2H-compounds. These authors assume two bands with very different mobility, e.g. s- and d-band. (Band structure calculations[4] for the undistorted 2H-phases show, that the conduction band is a narrow d-band, with the Fermi energy E_F near a maximum of the density of states N(E). An overlap with a s-like band is not proposed by theoretical calculations, but there is some experimental evidence for this in 2H-TaS_2 [5].) In this model the electrical resistivity caused by the electron-phonon scattering has two main parts: i) ρ_{ss}, which describes scattering within the s-band and which shows the usual T^5-dependence; ii) ρ_{sd}, which describes s-d scattering; the temperature dependence of this part is complex. After Naito and Tanaka the observed T^3 dependence of the ideal resistivity in 2H-NbS_2 and 2H-$NbSe_2$[1] is caused by strong s-d scattering in these compounds, which show no or only weak CDW effects, respectively, and therefore have a high $N(E_F)$ in the d-band. In 2H-TaS_2 and 2H-$TaSe_2$, which show strong CDW effects, specific heat measurements indicate a considerable reduction of $N(E_F)$ in the distorted phase. Therefore the influence of s-d scattering is reduced and one measures a T^4 or T^5 dependence, respectively, in these compounds[1].

Our experimental data confirm the T^4 law in 2H-TaS_2; for the intercalated compounds we find a T^3 dependence of $\rho(T)$ as was found for 2H-NbS_2 and 2H-$NbSe_2$. In the above-mentioned model this result implies a considerable contribution of s-d scattering and therefore a rise of the density of states at E_F compared to 2H-TaS_2. This is expected if a suppression of the CDW occurs. The analysis of low temperature behavior of $\rho(T)$ gives therefore a further confirmation for the absence of strong CDW effects in $K_x(H_2O)_yTaS_2$. In a rigid band model an increasing charge transfer to the TaS_2 d-band leads to a continuous decrease of $N(E_F)$. The higher values of the exponent p for x = 0.365 and 0.42 may be caused by this fact.

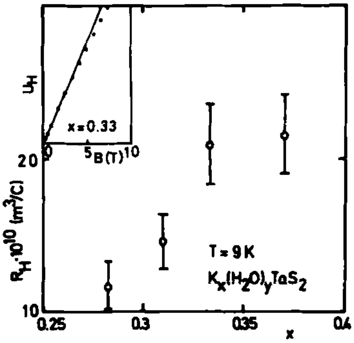

FIGURE 2 Hall coefficient of $K_x(H_2O)_yTaS_2$ at about 9 K as a function of potassium content x (≙ charge transfer); insert: Hall voltage U_H as function of magnetic field for a sample with x = 0.33

Naito and Tanaka[1] pointed out that the CDW phase in 2H-$TaSe_2$ and 2H-$NbSe_2$ is not only reflected by a change in the Hall coefficient at the onset temperature but also by an anomalous field dependence of R_H even at low fields in the CDW state. These effects were also found in our binary 2H-TaS_2 samples[3]. In contrast to this the intercalated compounds don't show this anomalous behavior: as it is demonstrated in the insert of fig. 2 for $K_{0.33}(H_2O)_yTaS_2$, there is only a small deviation from a linear relationship between the Hall voltage U_H and the applied field for high fields. The $U_H(B)$-curves for compounds with other stoichiometries x are very similar. In fig. 2 we present the deduced low field Hall coefficient as a function of x at T = 9 K. All intercalated samples show a positive sign of R_H at low temperature in contrast to the binary compounds. These results further indicate the suppression of the CDW phase and the strong influence of the charge transfer.

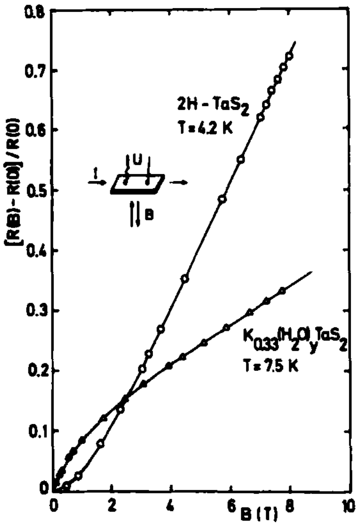

FIGURE 3 Comparison of the magnetoresistance for 2H-TaS_2 and $K_{0.33}(H_2O)_yTaS_2$.

In fig. 3 the magnetoresistance of 2H-TaS_2 and $K_{0.33}(H_2O)_yTaS_2$ is plotted versus the magnetic field at low temperatures. The observed curve for 2H-TaS_2 is very similar to the results of Naito and Tanaka for 2H-$TaSe_2$ and 2H-$NbSe_2$[1]. These authors try to explain the almost linear dependence for higher fields by magnetic breakdown in the CDW phase. The magnetoresistance for the intercalated compound shows a different and also anomalous behavior. In the sample with $x = 0.33$ we could not find a B^2 dependence of the magnetoresistance, which is to be expected theoretically at low fields, even for $B < 0.1$ T. The measurements were done in the temperature range between 6 and 15 K ($K_{0.33}(H_2O)_yTaS_2$ becomes superconducting at 5.5 K). The samples with a lower or higher potassium content x show the same behavior of the magnetoresistance, but there seems to be a B^2 dependence for $B < 0.1$ T. An explanation of these data is not possible at present. For a better understanding more theoretical research (e.g. bandstructure calculations) and experimental work are required.

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