Hall effect accompanying a Peierls transition in TaS_3

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The positive sign and activation nature of the temperature dependence of Hall's constant in TaS_3 below the Peierls transition temperature T_p are established. The anomalous decrease in the Hall carrier mobility in the region $T \gtrsim T_p$ is observed.

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The first investigations of the Hall effect in $NbSe_3$ (Refs. 1 and 2) demonstrated the fruitfulness of this technique for studying the Peierls state in quasi-one-dimensional conductors. Thus, for example, in Ref. 3, based on Hall measurements, it was proved that electron-hole pockets exist in $NbSe_3$ in the temperature range below the second Peierls transition.

In the other intensively investigated quasi-one-dimensional compound of the class MX_3, orthorhombic TaS_3, Hall measurements have been performed only at room temperature.[4] The sign of the carriers in TaS_3 was not established accurately until recently, since the measurements in Ref. 4 were conducted using ac current, while available data on the thermoemf[5,6] were contradictory. The main point is, however, that the Peierls transition ($T_p = 210$ K) and the Peierls state itself were not yet investigated with the help of the Hall effect in TaS_3. This is what determined the problem addressed in this paper.

We obtained specimens of orthorhombic TaS_3 by crystallization out of the gas phase.[7] The Hall emf was measured with dc current in the direction of the crystallographic *b* axis, the measuring current was passed along the specimen in the direction of the *c* axis, and the field **H** was oriented along the *a* axis. In this case, the electric field along the specimen did not exceed 0.3 V/cm, which corresponded to the active nature of the conductivity. Transverse contacts were prepared using the procedure described in detail in Ref. 7. The temperature of the specimen was monitored by a calibrated silicon diode and was maintained constant with the help of an automatic bridge to within 40 mK. The Hall emf was measured with a F-138 photocompensation nanovoltmeter.

Measurements were performed on six specimens at room temperature, and in all of them the Hall constant R_x was positive. The average value of R_x at room temperature was $3.5 \pm 0.7 \times 10^{-3}$ cm^3/C, five times greater than in Ref. 4. The dependence of the Hall emf on the magnetic field was linear at room temperature (Fig. 1) and retained its linear form at temperatures below the Peierls transition in the entire temperature range investigated.

The typical temperature dependence of R_x is shown in Fig. 2. As is evident from this figure, in the range 400–260 K, R_x is essentially independent of temperature and, then, in the region of the Peierls transition, it sharply increases approximately by an order of magnitude while remaining positive and, finally, below 160 K, it assumes the

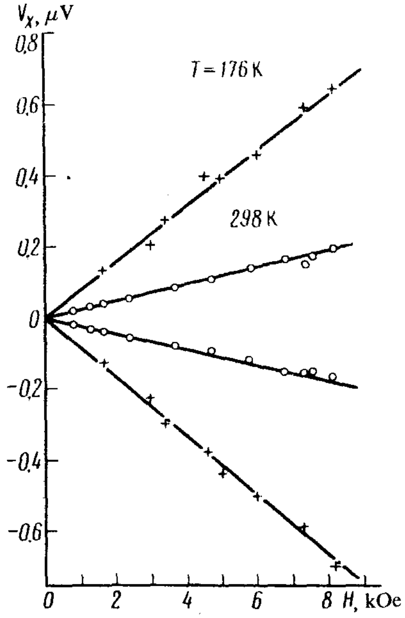

FIG. 1. Dependence of the Hall emf v_x on the field H (specimen No. 3). The two branches correspond to opposite orientations of the magnetic field. The current through the specimen was 0.36 mA at $T = 176$ K and 2.07 mA at 298 K. The dimensions of the specimens were $4.6 \times 0.06 \times 0.033$ mm.

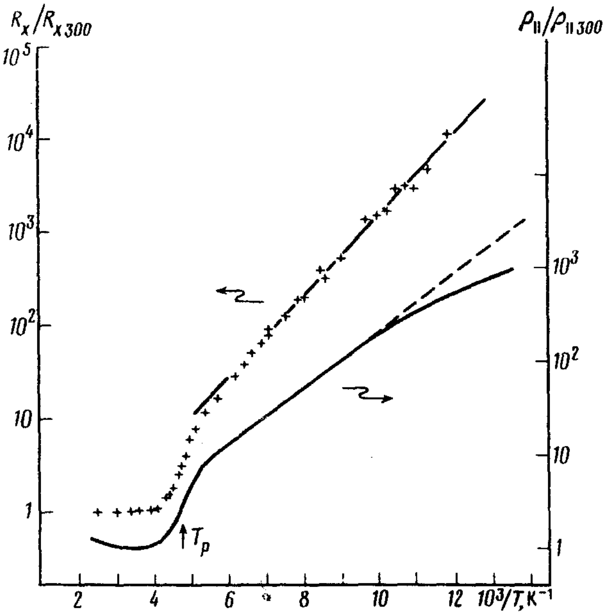

FIG. 2. Temperature dependences of the Hall constants and of the resistivity (in the longitudinal direction) of specimen No. 3, normalized to the characteristic values at 300 K, $R_{x300} = 4.4 \times 10^{-3}$ cm^3/C and $\rho_{\parallel 300} = 3.5 \times 10^{-4}$ Ω·cm.

activation dependence $R_x \sim \exp \Delta / T$ with activation energy $\Delta = 1000$ K. No deviations of the dependence $\lg R_x(1/T)$ from a straight line were observed down to 80 K. As the temperature decreased from 260 to 80 K, R_x increased by a factor of 2×10^4, without changing sign.

The results obtained by us (assuming that the band model is valid) indicate that only one type of carriers, the hole carriers, contribute in the temperature range investigated (see, also Ref. 6). In this case, the transverse magnetoresistance should vanish.[3] The measurements performed showed that $[\rho_{\|}(H) - \rho_{\|}(0)]/[\rho_{\|}(0)]$ is indeed equal to 0 with experimental accuracy (10^{-3}) in the entire range $400 \text{ K} > T > 80$ K and in fields up to 8 kOe. In this case, R_x is expressed only in terms of the carrier concentration p

$$R_x = (pec)^{-1} \tag{1}$$

while the anisotropy of the conductivity is determined by the anisotropy of the mobility:

$$\mu_{\|} = (\rho_{\|} pe)^{-1} , \tag{2}$$

$$\mu_{\perp} = (\rho_{\perp} pe)^{-1} . \tag{3}$$

From measurements of R_x and $\rho_{\|}$, $\rho_{\perp}$ at room temperature, we obtain $p_{300} = 1.8 \cdot 10^{21}$ cm^{-3}, $\mu_{\|300} = 10$ cm^2/V·s, $\mu_{\perp 300} = 0.06$ cm^2/V·s. As the temperature is decreased, the carrier density drops $p_{80} = 1.1 \times 10^{17}$ cm^{-3}, while their mobility increases $\mu_{\|80} = 250$ cm^2/V·s, $\mu_{\perp 80} = 0.38$ cm^2/V·s. The decrease in the hole density indicates that an energy gap appears in TaS_3, as in the case of $NbSe_3$,[3] on the hole side of the Fermi surface, but, in contrast to $NbSe_3$, in TaS_3 pockets of free carriers apparently do not remain on the Fermi surface below the temperature of the Peierls transition. This is indicated by the fact that the increase in R_x up to 80 K follows an activation law without any signs of saturation (Fig. 2) and the well-known fact that $\rho(T)$ in TaS_3 continues to increase up to liquid helium temperatures.[8] In any case, at 80 K the carrier density in TaS_3 is already an order of magnitude lower than the corresponding carrier density in the "pockets" in $NbSe_3$ at 2 K.

Comparison of the temperature dependences $R_x(T)$ and $\rho(T)$ in Fig. 2 shows that the increase in R_x with a decrease in temperature at $T < 160$ K occurs with a higher activation energy than the increase in ρ. This is evidently related to the fact that the growth in R_x is determined only by the freezing-in of carriers, while the growth of ρ is partially cancelled by the increase in their mobility as the temperature is decreased.

It follows from a comparison of $R_x(T)$ and $\rho(T)$ in the temperature range below 110 K that the deviation of $\rho(T)$ from the activation dependence in this region[8,9] is not related to the change in the magnitude of the Peierls gap, since the dependence $R_x(T)$ in this region proceeds with a constant activation energy. We did not observe any singularities in the dependence $R_x(T)$ in the region 140–160 K, corresponding to an incommensurate–commensurate transition in orthorhombic TaS_3.[10]

In the range 400–260 K, the carrier density is constant and does not depend on temperature. The decrease in p begins at temperatures 50 K higher than the temperature of the Peierls transition and at $T = T_p$ (T_p was determined from the position of

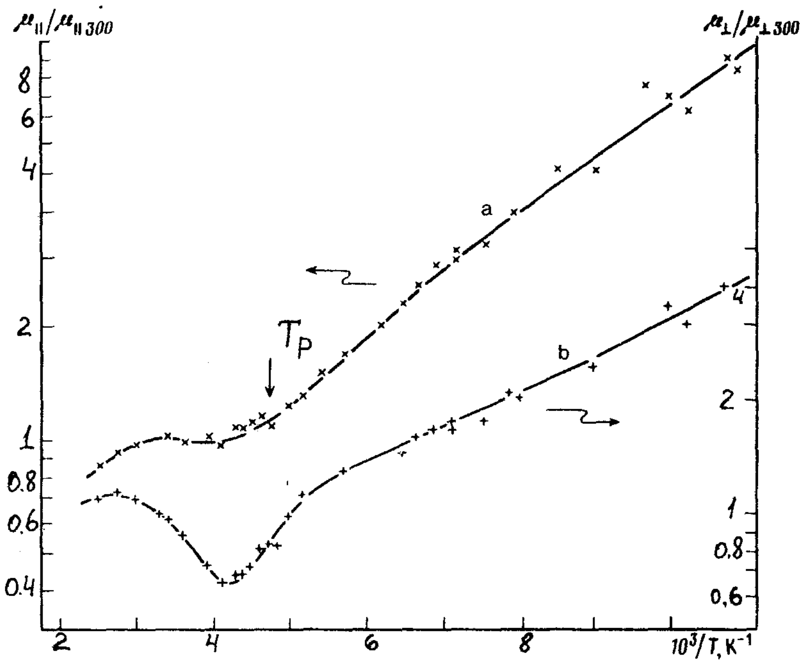

FIG. 3. Temperature dependence of the Hall mobility in the longitudinal direction, along the *c* axis (curve a) and in the transverse direction, along the *b* axis (curve b). Specimen No. 3.

the sharp peak in $(d\lg\rho/d\,1/T)$ already decreases by a factor of 3, indicating the presence of a gap in the region $T \gtrsim T_p$.

In this temperature region, we observe an anomalous decrease in the Hall mobility of carriers (Fig. 3). The values of the mobility at different temperatures were determined from the one-dimensional measurements of $R_x(T)$ and $\rho(T)$ in accordance with (2). As is evident from Fig. 3 (curve a), against the background of a general increase in $\mu(T)$, the mobility is observed to decrease with decreasing temperature in the region immediately preceeding T_p. The maximum drop in $\mu(T)$ is reached at 220–210 K.

We assumed that the anomaly in the temperature dependence of the carrier mobility is related to the softening of the phonon spectrum in the region preceding the Peierls transition, since the softening of the phonon frequency (see, for example, Ref. 11) increases the amplitude of scattering by a soft phonon as the temperature decreases $T \to T_p$. This additional scattering is manifested as a decrease in the carrier mobility.

In view of the three-dimensionality of the phonon spectrum, such a decrease in the mobility was expected in the transverse direction. Special measurements of $\rho_{\perp}(T)$, together with $R_x(T)$, show that this is indeed observed experimentally (Fig. 3, curve b). In this case, the relative decrease in the longitudinal and transverse mobilities turns out to be of the same order of magnitude $\delta\mu_{\parallel}/\mu_{\parallel} \approx \delta\mu_{\perp}/\mu_{\perp}$

Analogous anomalies in the mobility, in all probability, should also be observed in other quasi-one-dimensional compounds in the limit $T \to T_p$.

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