Contribution of a moving charge-density wave to the Hall effect in TaS_3

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(Submitted 25 January 1984)
Pis'ma Zh. Eksp. Teor. Fiz. **39**, No. 6, 258–261 (25 March 1984)

A contribution to the Hall voltage from the motion of a charge-density wave has been detected experimentally for the first time. A theory is derived for the effect.

It is widely believed that the motion of a charge-density wave does not contribute to the Hall voltage. This opinion is supported by measurements[1,2] in $NbSe_3$. In contrast, the results reported in the present letter show that this effect is observed in TaS_3. We offer a corresponding theoretical analysis.

The test samples were single crystals of orthorhombic TaS_3 of high quality ($E_c = 0.2$–0.4 V/cm, $T_P = 220$ K), which were synthesized in a high-temperature ($\sim 650\,^\circ$C) heterogeneous reaction of Ta with S, followed by crystallization from the gas phase in a temperature field with a slight gradient. The Hall voltage was measured along the crystallographic **b** axis of the crystal with a Keithley 180 nanovoltmeter. The Hall voltage was symmetric with respect to a reversal of the magnetic and electric fields in the sample (**H**||**a**, **E**||**c**), and it was found to be linear in the magnitude of H over the entire ranges of the temperature and the magnetic field studied.

Figure 1 shows curves of $V_H(E)$ obtained from sample No. 12 (with dimensions of $2.4 \times 0.038 \times 0.008$ mm) at various temperatures over the range 100–300 K. In weak fields E the curves are linear, as are the corresponding curves recorded previously[1,2] for $NbSe_3$. As the field is raised further, however, there is a characteristic deviation

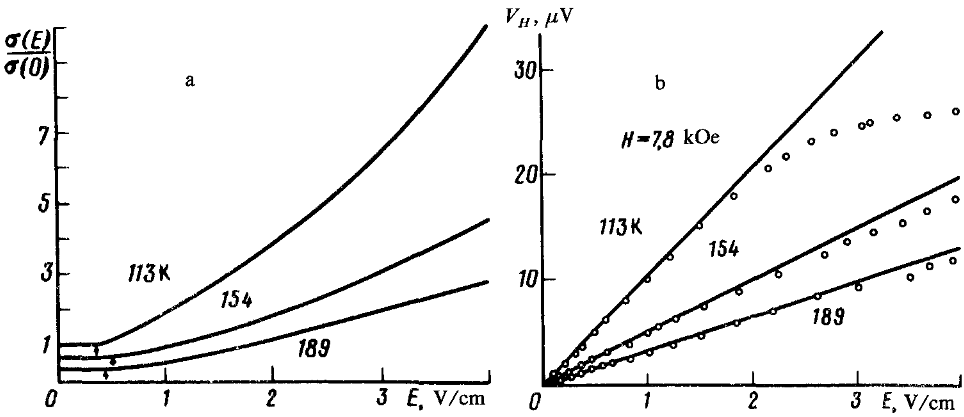

FIG. 1. The conductivity and the Hall voltage V_H of sample No. 12 vs the field E. The values of $\sigma(0)/\sigma_{300}$ at 113, 154, and 189 K are 0.013, 0.077, and 0.182, respectively. The arrows show the positions of E_c. The error of the V_H measurements is 4%.

from linearity on all the $V_H(E)$ curves at temperatures below T_P; this nonlinearity was the subject of our investigation. The deviation of the $V_H(E)$ curves from linearity was reproducibly observed in all three samples. The circumstance that this deviation becomes more pronounced under the same conditions (lower temperatures and higher values of E) which increase the nonlinear conductivity (Fig. 1a) suggests that this deviation is also associated with the motion of a charge-density wave.

To rule out Joule heating of the sample in strong fields, we carried out some special comparison measurements, in which a given sample (No. 13) was held at a given temperature in various coolants: gaseous helium (curve 1 in Fig. 2) and liquid methane (curve2). The heat removal in the liquid methane was two orders of magnitude more rapid, according to our data. The results of this comparison show that there exists a region in the manifestation of the effect (to ≈ 4 V/cm in the gaseous coolant) in which the effect cannot be attributed to Joule heating. This is the region which corresponds to the coincidence of curves 1 and 2 in Fig. 2.

Further experiments were carried out in liquid methane (no effect of heating up to ≈ 8 V/cm) with samples with various values of E_c (curves 2 and 3 in Fig. 2). We see from this figure that at fields $\lesssim 4$ V/cm, at which a significant effect is observed for sample No. 13, with $E_c = 0.4$ V/cm, no effect of any sort is observed for sample No. 6, with a higher value of E_c, for which the charge-density wave is fixed at such fields. These experiments also confirm that the nature of the effect is determined by the motion of a charge-density wave.

Let us summarize the characteristics of this effect. 1) The contribution of the charge-density wave to the Hall voltage is first seen in fields slightly above E_c. It is possible that only at such fields does the charge-density wave begin to more as a whole.

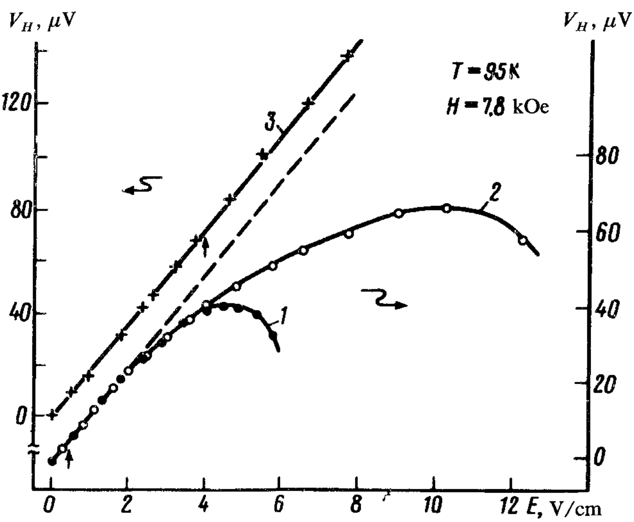

FIG. 2. Comparison of the $V_H(E)$ curves in different coolants. 1—Gaseous helium; 2, 3—liquid methane. The arrows show the positions of E_c. Curves 1, 2—Sample No. 13 (3.5×0.047×0.007 mm); 3—sample No. 6 (4.4×0.062×0.012 mm).

Furthermore, the field E is weaker under the Hall junctions than far from them,[3] so that voltages across the sample must exceed $E_c L$ (L is the length of the sample) in order to disrupt the charge-density wave in this region. 2) The motion of the charge-density wave reduces the Hall voltage, i.e., makes a contribution opposite in sign to the Hall effect in the case of a fixed charge-density wave. 3) The contribution of the charge-density wave decreases with increasing temperature, as the temperature approaches the point of the Peierls transition, T_P.

To interpret the results of these measurements, we take the microscopic approach developed in Refs. 4 and 5. Despite its simplicity, this approach explains the basic features of the critical behavior of the resistance in $NbSe_3$ (Ref. 4). To analyze the Hall effect we work from a semiclassical kinetic equation which we derive by the Keldysh method from the equations for the Green's functions of the electrons in the Peierls phase. The spectrum of the two types of quasiparticles, $\epsilon^{1,2}{}_p = \eta \pm \sqrt{(\xi + \dot{\chi}/2)^2 + \Delta^2}$, and the collision integral in the kinetic equation incorporate the motion of a charge–density wave [here $\xi = v_F(p - p_F)$, χ and Δ are the phase and modulus of the order parameter, $\dot{\chi} = 2mv_F s$, s is the velocity of the charge-density wave, and $\eta(p_\perp)$ represents the transverse dispersion law]. We assume that the Fermi surface is only slightly curved (i.e., $\eta \ll \Delta$), in accordance with TaS_3.

If we ignore the effect of the field E on the distribution function, and if we ignore the scattering of electrons, then the quasiparticles would be at equilibrium with the superlattice and would move along with it during the motion of a charge-density wave. This equilibrium distribution of quasiparticles, however, would not contribute to the Hall effect, according to the calculations. In first order a deviation of the distribution function from equilibrium arises from, first, the drift of quasiparticles under the influence of the field E along the direction in which the charge-density wave is moving (the charge-density wave is being driven by the same field E) and, second, the collisions of impurities with quasiparticles moving with the charge-density wave. Correspondingly, the terms in the distribution function which describe these two types of deviations from equilibrium stem from different parts of the equation. The first term, which results from the field term, is proportional to E; the second comes from the collision integral, is proportional to $\dot{\chi}$, and results from the change in the energy of the quasiparticles during the scattering by $\dot{\chi} = 2p_F s$.

For the longitudinal current we find

$$j = \left(\sigma_1 E - a\frac{\sigma_1}{l}\dot{\chi}\right) + \frac{\sigma_N}{l}\dot{\chi}, \tag{1}$$

where σ_N is the longitudinal conductivity in the normal state, and l is the mean free path. At low temperatures we have $T \ll \eta, a \sim \sqrt{T\Delta}/\eta$, and $\sigma_1 \sim \sigma_N e^{-\Delta/T}$. The terms in parentheses in (1) result from the deviation of the quasiparticle distribution from equilibrium. These terms differ in sign according to the discussion above. The last term in (1) describes the direct contribution of the motion of the charge-density wave to the longitudinal current. At low temperatures the second term in parentheses is small in comparison with the last term. For this reason, we may ignore that contribution of the quasiparticles to the longitudinal current which is proportional to $\dot{\chi}$. On the other hand, we note that precisely this quasiparticle contribution is important in the

Hall effect, since the motion of a charge-density wave along the filaments does not directly contribute to the Hall voltage.

The expression for the Hall voltage takes different forms, depending on the relationships among the parameters. For $T \ll \eta \ll \Delta$ we have

$$V_H \sim j_H/\sigma_{tr}, \quad j_H = j_1 + j_2,$$
$$j_1 \sim \sigma_{tr} l EH \frac{1}{a\Delta}, \quad j_2 \sim -\sigma_{tr} \dot{\chi} H \frac{1}{\Delta}, \tag{2}$$

where σ_{tr} is the transverse conductivity in the Peierls phase.

The currents j_1 and j_2 in (2) result from these two mechanisms which cause a deviation of the quasiparticle distribution from equilibrium. They differ in sign, as in the case of the longitudinal current. The motion of a charge-density wave can thus reduce the Hall voltage.

Furthermore, estimates of $j_2/j_1 \sim \dot{\chi}/E$ show that the ratio $\dot{\chi}/E$ in TaS_3 increases with decreasing temperature, again in qualitative agreement with experiment. At low temperatures in strong fields E, the contribution of the charge-density wave can change the sign of the Hall voltage.

Calculations show that a charge-density wave should also contribute to the Hall voltage in $NbSe_3$. The contribution should be significant near T_P, where its magnitude is such that the motion of a charge-density wave can cancel out the anomalous features in $R_H(E)$.

We wish to thank A. F. Volkov and L. P. Gor'kov for discussions.

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Translated by Dave Parsons
Edited by S. J. Amoretty