Temperature-Gradient–Induced Open-Circuit Electric Currents in Charge-Density-Wave Condensates

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We have induced nonlinear conduction and current oscillations in the charge-density-wave state of o-TaS_3 in an open-circuit ($I=0$) configuration using a temperature gradient to generate thermal voltages exceeding a threshold. The above features are associated with a current carried by the condensate which is compensated by a normal current in the opposite direction. Our experiments provide the first clear observation of decoupled motion of the condensed and uncondensed electrons in solids with a charge-density-wave ground state.

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Electric and heat currents induced by electric and thermal gradients are described by well-known transport equations. Aside from the electrical conductivity σ, the most commonly measured quantity is the thermoelectric power S, which relates the temperature gradient ∇T to the thermal voltage ΔV_T measured in an open-circuit ($I=0$) configuration. If carriers of different types contribute to the transport, the most widely used description is in terms of a two-band (or many-band) model; and σ and S are given by[1]

$$\sigma=\sigma_a+\sigma_b, \text{ and } S=\frac{\sigma_a S_a+\sigma_b S_b}{\sigma_a+\sigma_b}, \tag{1}$$

where the subscripts a and b refer to the different bands. With two types of carriers present and providing different channels for electric and heat conduction, electric currents in the two bands could in principle be induced by a temperature gradient in an open-circuit ($I_t=I_a+I_b=0$) configuration. The flow of electrons in one band is compensated by a flow in the opposite direction in the other band with the conversion between the two bands due to interband scattering processes. While two-band descriptions of transport have been widely used for specimens where both types of carriers are single-particle-like, the situation is much clearer when part of the carriers are condensed into a collective mode that can support an electric current; and the other part is uncondensed, providing single-particle transport. This occurs in superconductors, and the effect has been clearly observed.[2,3] Two-fluid models have also been used to describe the conductivity and thermoelectric power in materials where a different type of condensate, called a charge-density wave (CDW), develops.[4,5] Because the conductivity due to both the normal and the condensed electrons is finite, a finite voltage can be generated by a temperature difference between the two ends of the specimen, ΔT, in an open-circuit configuration.

In this paper we give unambiguous evidence for decoupled normal and charge-density-wave–supported electric currents provided by condensed and uncondensed electrons in an open-circuit configuration. We have induced both nonlinear conduction and coherent current oscillations with a temperature gradient in the CDW state of orthorhombic TaS_3 (o-TaS_3). The above features are regarded as trademarks of collective CDW conduction,[4] and consequently our experiments demonstrate the existence of both normal and CDW currents which persist in the presence of a temperature gradient with the total current across the sample $I_t=I_N+I_{CDW}=0$.

The low-field thermoelectric power S has been studied in o-TaS_3 before.[5–7] It rises sharply below the phase transition at $T=220$ K and reflects the progressive development of the single-particle gap. Around 100 K, S is approximately 1 mV/K; and at this temperature the threshold electric field, where in pure specimens the onset of nonlinear conduction occurs, is around 200 mV/cm. Earlier studies[5] concentrated on the effect of electric fields on the thermoelectric power by keeping ΔT small. The values quoted above indicate that in pure, short specimens the thermal voltage ΔV_T induced by a substantial temperature gradient may exceed the threshold voltage for nonlinear conduction V_t.

In order to study the various nonlinear effects which occur under such circumstances, we used short ($l=0.3$ mm), high-purity ($E_t=180$ mV/cm at $T=100$ K) specimens. One end of the crystal was attached to a short gold wire by silver paint, and the gold wire was thermally anchored to a Peltier cell capable of generating temperature differences as large as 50 K. The other end was silver painted to a thick copper wire serving as both a heat shunt to the base and a current lead. A differential thermocouple measured the temperature difference across the sample. The system was sealed and evacuated in order to keep the thermal leaks as low as possible. A liquid N_2 bath cooled the device, and a controlled heater stabilized the temperature of the base at 100 K.

We first describe the joint effect of thermal gradients and dc currents. In order to measure the thermal voltage, the external circuit connected to the sample has to have a much greater impedance than the resistance of the specimen. We used a 1-MΩ output-impedance current generator and a 100-MΩ input-impedance lockin amplifier for the resistance measurements, while the typical sample resistance was approximately 40 kΩ.

In Fig. 1 the gradient-induced thermal voltage obtained in an open-circuit configuration is displayed together with the differential resistance measured with utilization of small ac electric fields [Figs. 1(a) and 1(b), respectively]. The slope $\Delta V/\Delta T$ in Fig. 1(a) gives the thermoelectric power directly. Notice that because the ΔV vs ΔT curve is nonlinear, both chordal and differential thermopowers can be defined in a manner analogous to the chordal and differential conductivities widely used to characterize the current-voltage characteristics in various materials which display CDW transport. For small gradients, ΔV is proportional to ΔT giving values for S in agreement with that observed before. With an increasing temperature gradient, a substantial nonlinearity sets in, indicated by a decreased ΔV induced by the temperature gradient when compared with the slope at small ΔT.

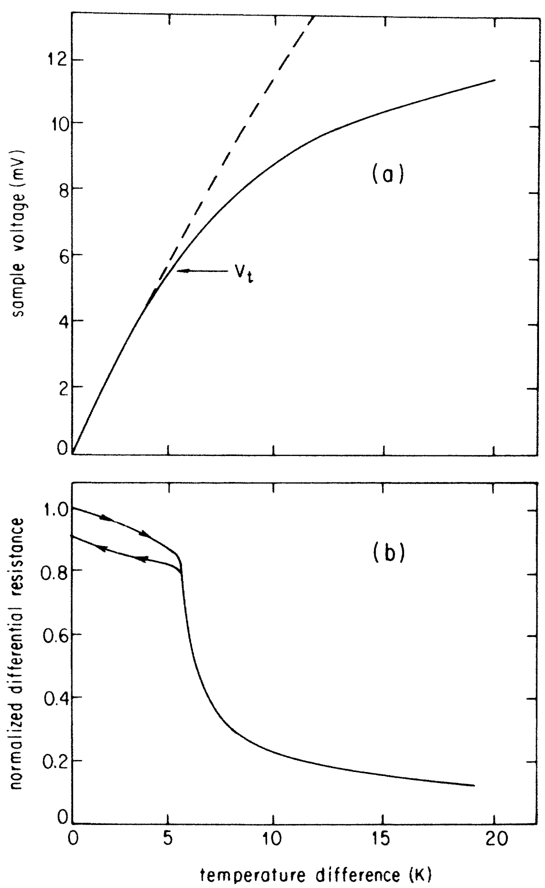

FIG. 1. (a) Thermal voltage ΔV_T vs temperature difference ΔT and (b) low-field differential resistance vs ΔT for sample No. 1 (length $l = 0.3$ mm, room-temperature resistance $R_{RT} = 90$ Ω). The dashed line corresponds to the thermal voltage obtained by integration of the temperature-dependent thermopower (see text). The arrow denotes the threshold voltage for nonlinear conduction measured at 100 K in the absence of a temperature gradient.

For a sample having a large temperature difference between the ends, the total thermal voltage ΔV_T should be calculated by taking into account the temperature dependence of the thermopower $S(T)$. This leads to

$$\Delta V_T = \int_0^l S(T(x))(dT/dx)\,dx = \int_{T_0}^{T_0+\Delta T} S(T)\,dT, \qquad (2)$$

where $T_0 = 100$ K is the base temperature. We assumed that the temperature gradient is constant along the sample length l, $dT/dx = \Delta T/l$. The thermal voltage calculated with the above assumptions is indicated by the dashed line in Fig. 1(a). The agreement between the measured and calculated ΔV_T is within the experimental accuracy for small ΔT, but a considerable deviation becomes apparent at large gradients. The threshold voltage for nonlinear conduction, determined in an independent measurement by recording of direct I-V curves at $T = 100$ K, is also indicated in the figure. The coincidence of voltages obtained by applying an electric field or a temperature gradient suggests that the breakdown of the thermal voltage is associated with the onset of nonlinear conduction due to a CDW current.

To investigate this possibility further, we recorded the differential resistance of the sample, dV/dI, versus the temperature difference, ΔT [Fig. 1(b)]. dV/dI was calculated from the voltage response at low-amplitude ac currents. At low thermal gradients, dV/dI changes smoothly, and a hysteresis is observed in qualitative accordance with the earlier measurements performed with homogeneous temperature cycling.[8,9] A sharp drop is observed in the resistance at the temperature difference where the anomaly in the thermopower is observed, again suggesting that CDW conduction is induced by the temperature gradient. Differential resistance measurements were extended by the simultaneous application of a temperature gradient and a dc bias current (Fig. 2). It is apparent that the anomaly in the differential resistance observed in the zero-current measurement [Fig. 1(b)] is indeed associated with the onset of nonlinear conduction in the presence of a temperature gradient.

It is well known that the CDW conduction is associated with coherent current oscillations generally studied by the recording of the Fourier-transformed vol-

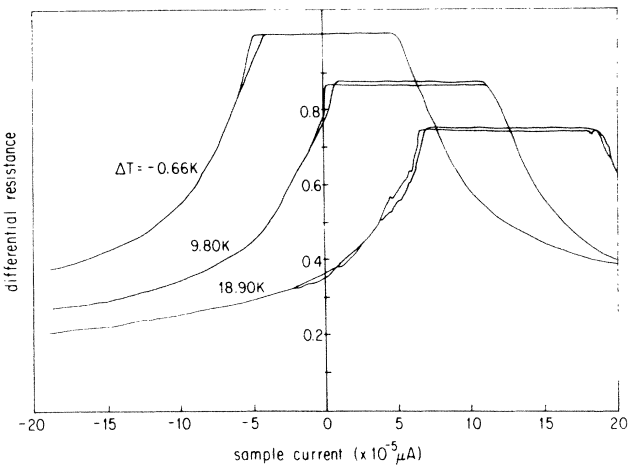

FIG. 2. Differential resistance, normalized to the 100-K low-field value, vs sample current at three different temperature gradients for sample No. 2 ($l = 0.3$ mm; $R_{RT} = 170\ \Omega$). The curves shift as ΔT is increased. No similar shift is observed in the R-vs-V curve.

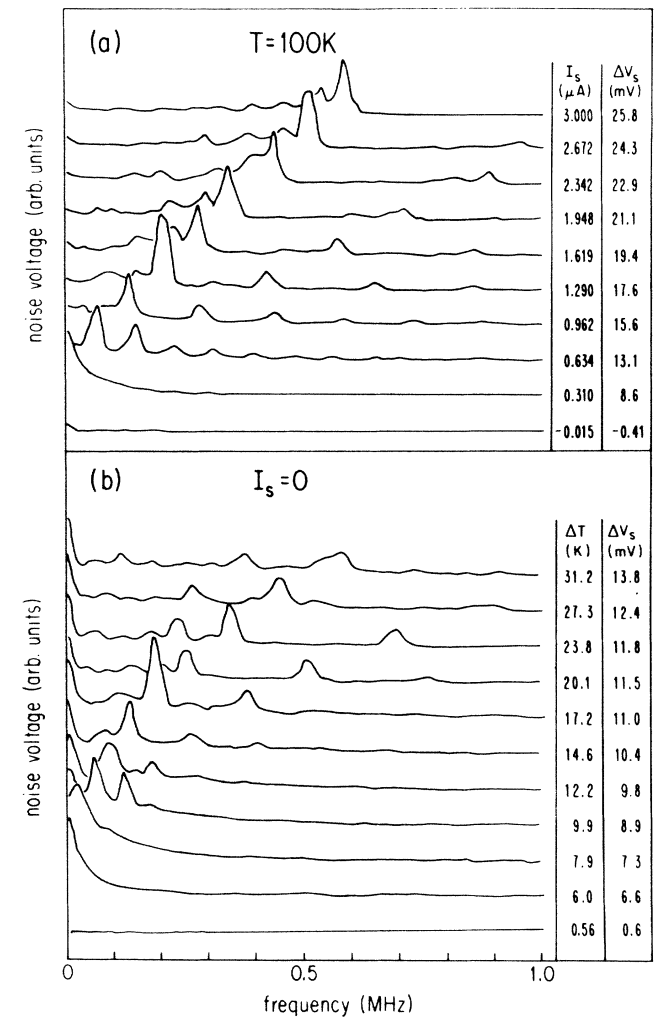

FIG. 3. Noise spectra taken on sample No. 1 at (a) zero temperature gradient with various currents and (b) at zero current with several temperature gradients. The spectra are shifted vertically for a better view. The temperture difference ΔT, the sample current I_S, and the dc voltage ΔV are also denoted.

tage or current. The spectrum displays a fundamental and several harmonics with decreasing intensity.[4] Figure 3 shows the Fourier-transformed voltage spectra recorded on our sample in the absence of a temperature gradient for different applied dc currents. The behavior exhibited in Fig. 3(a), where the frequency of the fundamental peak increases linearly with the excess current associated with the nonlinear conduction, is regarded as clear evidence for electric-field–induced CDW motion. We detected voltage oscillations in the presence of a thermal gradient with no current flowing through the sample [Fig. 3(b)]. Oscillations appear when the thermally induced voltage exceeds approximately 8 mV, and the main peaks increase with increasing ΔT and consequently with increasing ΔV. The conclusion which can be reached on the basis of Fig. 3 is clear: Temperature gradients of sufficient magnitude lead to CDW transport in the absence of a current in the specimen. We note that the spectrum also becomes progressively more complex with increasing ΔT, indicating a progressive breakdown of coherence, a phenomenon studied in $NbSe_3$ in detail before.[10]

We believe that the experimental results presented above provide clear evidence for thermal-gradient–induced electric currents in materials displaying charge-density-wave conduction. For a small applied temperature gradient, the resulting thermal voltage is small, and a CDW current is not induced. Consequently, after an initial transient normal current flow, both I_N and I_{CDW} are zero. With an increased gradient, the thermal voltage exceeds the threshold voltage required to produce a CDW current. Under such circumstances, current carried by the condensate starts to flow with I_{CDW} determined by the thermal voltage ΔV_T. This current is balanced by a normal current flowing in the opposite direction with $I_N = -I_{CDW}$. The phenomenon requires a continuous conversion of carriers between the condensate and the sea of uncondensed electrons. Such a conversion may occur at the ends of the specimen[11] and also in the bulk[12]; the experiments reported here do not address this question. The observations also provide strong support for the two-band model used before to describe the electric-field and frequency-dependent conductivity and the thermoelectric power.[4]

While the implications and the qualitative explanation of our experiments are straightforward, a detailed description is most likely complicated for several reasons. The electric current in the presence of an electric field E and a temperature gradient ∇T is given by

$$j(E)=\frac{L_{11}(E)E}{T}+\frac{L_{12}(E)\nabla T}{T^2}$$
$$=\sigma(E)E+S(E)\sigma(E)\nabla T, \qquad (3)$$

where L_{11} and L_{12} are the Onsager coefficients, and σ and S refer to quantities that are the standard conductivity and thermoelectric power when the electric and thermal gradients are small. For $j(E)=0$, $E=S(E)\nabla T$ which for small applied temperature gradients reduces to the conventional expression for the thermoelectric voltage. The analysis can be extended to a situation where the current-voltage characteristic is nonlinear. For a small ∇T, the expression for the current in terms of the temperature gradient is similar to Eq. (3) with differential conductivities replacing the chordal conductivities.[5] Such analyses and experiments employing a small temperature gradient demonstrate that $S(\nabla T=\text{small})$ decreases when nonlinear conduction is induced.[5] Figure 1(a) clearly shows that the total thermoelectric voltage induced by a large temperature gradient exhibits anomalies when the thermal voltage exceeds the threshold voltage. However, because of the nonlinearity in the problem, an analysis similar to that performed in Ref. 5 cannot be carried out. Similar comments apply for our experiments where both the temperature gradient and the applied dc current are large, and neither of these can be treated as a small perturbation.

Our experiments demonstrate that decoupled motion of normal and condensed electrons can occur in materials displaying CDW transport, as in superconductors,[2,3] with quasiparticle current compensating the CDW current in an open-circuit configuration.

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[1]J. M. Ziman, *Electrons and Phonons* (Oxford Univ. Press, New York, 1960), p. 400.

[2]D. J. Van Harlingen, Physica (Amsterdam) **109&110B**, 1710 (1982).

[3]H. J. Mamin, J. Clarke, and D. J. Van Harlingen, Phys. Rev. Lett. **51**, 1480 (1983).

[4]For a review, see G. Grüner and A. Zettl, Phys. Rep. **119**, (1985).

[5]J. P. Stokes, A. N. Bloch, A. Jánossy, and G. Grüner, Phys. Rev. Lett. **52**, 372 (1984).

[6]R. Allgeyer, B. H. Suits, and F. C. Brown, Solid State Commun. **43**, 207 (1982).

[7]B. Fisher, Solid State Commun. **46**, 227 (1983).

[8]Gy. Hutiray, G. Mihály, and L. Mihály, Solid State Commun. **47**, 121 (1983).

[9]J. C. Gill and A. W. Higgs, Solid State Commun. **48**, 709 (1983).

[10]A. Zettl, M. Kaiser, and G. Grüner, Solid State Commun. **53**, 649 (1985).

[11]N. P. Ong, G. Verma, and K. Maki, Phys. Rev. Lett. **52**, 663 (1984).

[12]S. E. Brown and L. Mihály, Phys. Rev. Lett. **55**, 742 (1985).