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NON-EQUILIBRIUM TRANSPORT IN $NbSe_3$: EFFECTS OF A TEMPERATURE GRADIENT

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We have measured the dc response characteristics of $NbSe_3$ in the charge density wave (CDW) state, in the presence of an applied temperature gradient. The threshold field E_T for the onset of nonlinear conduction remains sharp, and is determined by the average temperature of the specimen. No change is observed in the amplitude or quality factor of the coherent current oscillations in the nonlinear conductivity region, other than that expected from a change in average temperature. We interpret our results as evidence for macroscopic dynamical coherence throughout the specimen.

The spectacular transport properties associated with the charge density wave (CDW) in $NbSe_3$ and related compounds has been the subject of recent activity. Both the electric field dependent dc response and the frequency dependent conductivity have been interpreted in terms of collective dynamics of the CDW condensate.[1] For low applied dc electric fields, the CDW is pinned to the lattice by impurities. Nonlinear conduction occurs, however, when a critical field E_T is exceeded.[2]

A remarkable phenomenon[2] is that current oscillations are associated with the nonlinear conductivity region. For applied dc fields $E > E_T$, the excess current I_{CDW} carried by the condensate contains an ac component whose frequency is directly proportional to I_{CDW}.[3] The magnitude of the ac component is significant, and in pure specimens near threshold the oscillations constitute nearly 100% of the excess current.[4,5] Fourier analysis of the oscillations leads to a spectrum rich in harmonics and with extremely sharp frequency peaks ($Q \sim 10^3 - 10^4$) termed narrow-band noise. Experiments on $NbSe_3$[4] and TaS_3[6] have shown that the ratio I_{CDW}/f, where f is the fundamental frequency peak, is temperature dependent and reflects directly $n_c(T)$, the number of carriers condensed in the CDW state.

The origin of the current oscillations is not well understood at present. It has been suggested that they are due to the motion of the CDW condensate over the hills and valleys of the impurity potential.[4] In a simple description[7] the impurity potential may be treated as a washboard potential and the internal degrees of freedom of the condensate are neglected. However, since the source of the pinning potential is assumed to be impurities randomly distributed throughout the crystal, the CDW itself must be deformable for pinning to occur.[8] Such deformations lead to finite phase coherence in the pinned state. Recent experiments[9] demonstrated that a large fraction of the specimens acts as a coherent domain in the current carrying state, but the amplitude of the current oscillations decreases with increasing sample volume Ω, and vanishes in the thermodynamic, $\Omega \to \infty$ limit. These experiments suggest that the current oscillations are the consequence of impurity potentials and are a bulk phenomenon.

It has also been suggested that current oscillations may originate from the contacts placed for measurement purposes on the ends of the sample.[10] The normal metal - CDW interface near the contact may in principle create phase vortices, leading to the observed current oscillations. Although in this model CDW dynamics still play an important role in the generation of the oscillations, the oscillation phenomenon itself is not a true bulk effect.

To address the important question of local versus bulk nature of the current oscillations, we have carried out transport measurements on $NbSe_3$ samples in the CDW state, but in the presence of thermal gradients. We find that E_T and the current oscillations are affected by a temperature gradient, but only in the sense that the average temperature of the crystal is altered.

Figure 1 shows the sample mounting configuration. The ends of the sample were thermally

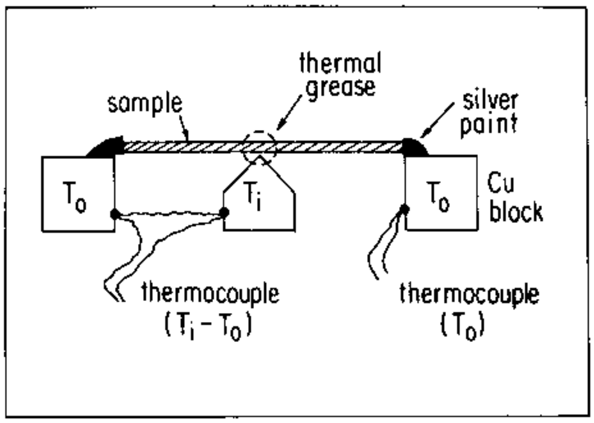

Fig. 1. Sample mounting configuration for temperature gradient experiments.

anchored to copper blocks, both at temperature T_o, while the center of the sample was thermally anchored with non-electrically conducting thermal grease to a sapphire wedge at temperature T_1. For all experiments $T_1 \geq T_o$. Temperatures were determined with high sensitivity thermocouples. A variety of sample mounting configurations with applied temperature gradients were tried, and we found the most reliable configuration to be that in which the ends of the sample were held always at the same temperature. In this configuration thermoelectric effects and temperature dependent reactification problems due to the silver paint contacts are eliminated.

Later we shall discuss an average temperature T_{avg} for the entire $NbSe_3$ crystal. This was determined by comparing the low-field (normal) resistance of the sample in the presence of a temperature gradient with the temperature dependence of the low-field resistance in the absence of a temperature gradient. The average temperature thus determined agreed well with that obtained assuming a triangular temperature distribution across the sample, where T_{avg} is given by $(T_o+T_1)/2$.

Figure 2 shows the dc threshold field E_T for $NbSe_3$ as a function of temperature gradient $dT = T_1 - T_o$, with T_o fixed at 48 K. E_T was determined by lock-in detection of the differential resistance dV/dI as a function of dc bias voltage. It is clear that a temperature gradient changes E_T even with the ends of the sample always at constant temperature. The average sample temperature T_{avg} is given on the upper horizontal axis of the figure. Figure 2 also shows E_T measured as a function of temperature in the absence of a temperature gradient. From Fig. 2 we conclude that the threshold characteristics with an applied temperature gradient are equivalent to those obtained for a uniform temperature sample at temperature $T = T_{avg}$.

In Fig. 3 we show, as a function of temperature gradient, the narrow-band noise spectrum for a different $NbSe_3$ crystal. T_o is again 48 K. For each value of dT in Fig. 3a, the dc bias has been adjusted to keep the fundamental oscillation frequency f at approximately 5 MHz. Figure 3b shows the narrow-band noise spectrum for the same sample under similar conditions, except that here dT = 0 and the entire sample temperature has been

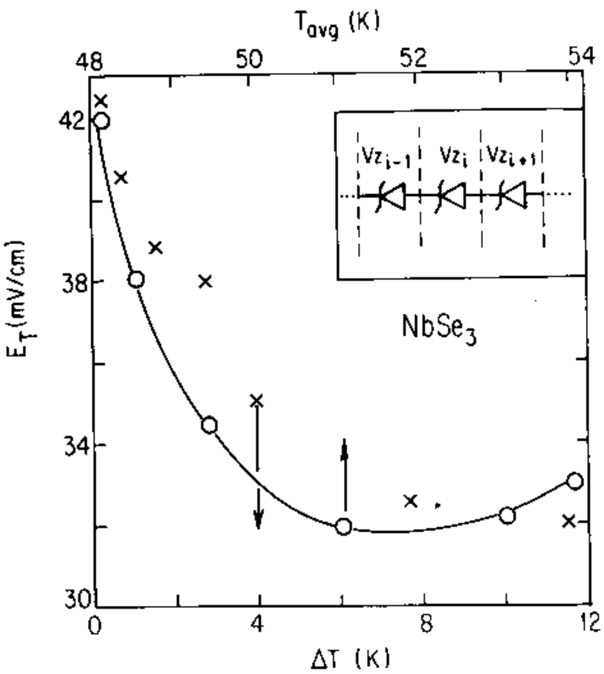

Fig. 2. Threshold dc electric field E_T (crosses) in $NbSe_3$ as a function of temperature gradient dT. T_o = 48 K (see Fig. 1). The upper scale gives the average sample temperature. Also plotted (open circles) is E_T, for dT = 0 and T = T_{avg}. The inset shows a simple electric analog for the threshold behavior with a distribution in pinning strengths (see text).

readjusted to match T_{avg} of the corresponding trace in Fig. 3a. It is apparent that a temperature gradient does not drastically alter the coherent current oscillation characteristics. With increasing dT, the amplitude and quality factor of the peaks remains relatively high, and the spectrum is entirely consistent with that found for the sample at dT = 0, T = T_{avg}. An interesting and important point is that, for the bottom trace in Fig. 3a, dT is such that T_1, the temperature at the center of the sample, is greater than Tp, the CDW transition temperature of the lower CDW state. Thus the $NbSe_3$ crystal is "normal" near its center, and in effect an artificial "contact" has been introduced. We have observed no additional structure in the narrow-band noise spectrum as T_1 is swept through Tp, in contrast to what might be expected for phase vortex generation[10] at this CDW-normal interface.

We define the excess CDW current as

$$I_{CDW} \equiv I - \frac{V}{R_o} \quad (1)$$

with I the total sample current, V the voltage across the sample, and R_o the low field (ohmic) resistance of the sample. In a simple model, I_{CDW} is related to the CDW drift velocity v_d by

$$I_{CDW} = n_c(T)ev_d \quad . \quad (2)$$

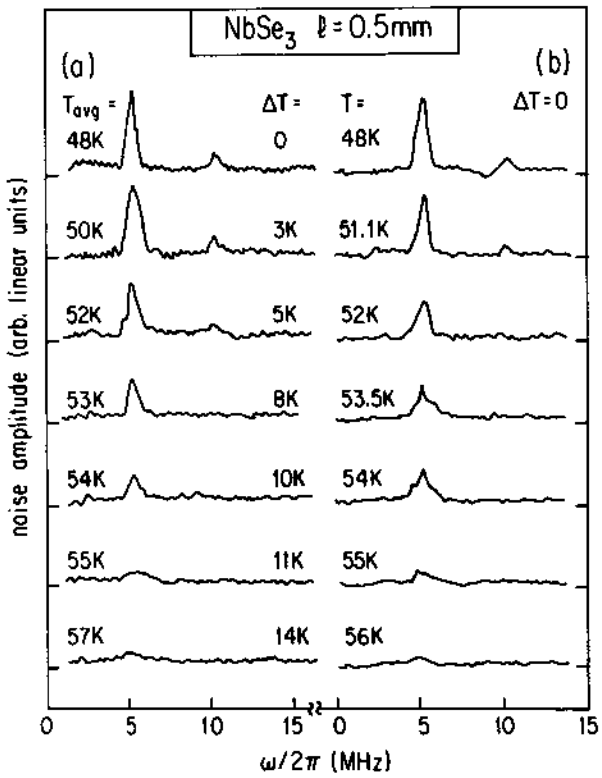

Fig. 3. (a) Narrow-band noise spectrum for $NbSe_3$ in the presence of a temperature gradient. The average sample temperature is indicated for each trace. T_0 = 48 K (see Fig. 1). Note that the last trace corresponds to $T_1 > T_P$ (see text).

(b) Narrow-band noise spectrum for $NbSe_3$ at selected temperatures with no temperature gradient. Comparing (a) and (b) shows tha a temperature gradient does not degrade the noise spectrum.

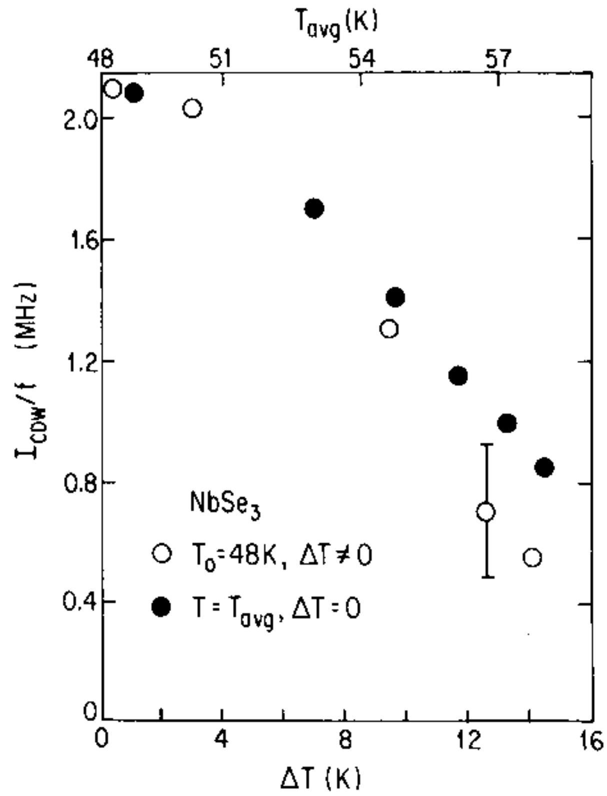

Fig. 4. Ratio I_{CDW}/f for $NbSe_3$ as a function of temperature gradient, with T_0 = 48 K (see Fig. 1). Note that for dT > 11 K the center of the sample is at $T_1 > T_P$. Also shown is I_{CDW}/f for dT = 0, T = T_{avg}. The effective carrier density in the presence of a temperature gradient is given by the average carrier density.

Thus, under isothermal conditions, the ratio I_{CDW}/v_d allows a direct evaluation of the carrier concentration n_c. Assuming $v_d = f\lambda$, with f the fundamental noise frequency and λ a constant, n_c may also be represented by I_{CDW}/f. Fig. 4 shows the ratio I_{CDW}/f measured isothermally for various sample temperatures T_{avg}. The results are consistent with earlier studies on $NbSe_3$.[4]

Under conditions of a finite temperature gradient, Eqs. (1) and (2) must be interpreted carefully, as they represent now local equations and do not in general apply to the sample as a whole. However, even with dT ≠ 0, I, R_o, and V remain well defined for the sample as a whole, and we may thus define, in analogy to Eq. (1), an "average" CDW current for the entire sample,

$$\langle I_{CDW} \rangle = I - \frac{V}{R_o} \quad . \tag{3}$$

In terms of local (temperature dependent, and hence position dependent) parameters, Eq. (3) becomes

$$\langle I_{CDW} \rangle = I - \frac{\frac{1}{\ell}\int_o^\ell V(x)\,dx}{\frac{1}{\ell}\int_o^\ell R_o(x)\,dx} \quad , \tag{4}$$

with ℓ the sample length. To a good approximation both normal (uncondensed) electrons and CDW electrons experience the same local electric field. Hence, $V(x) = [I - I_{CDW}(x)]R_o(x)$, and Eq. (4) becomes

$$\langle I_{CDW} \rangle = I - \frac{\int_o^\ell [I - n_c(x)ev_d(x)]R_o(x)\,dx}{\int_o^\ell R_o(x)\,dx} \quad . \tag{5}$$

We have explicitly assumed that the total current through the sample is constant, independent of local temperature. The local CDW order parameter, and hence local CDW carrier concentration $n_c(T)$, are dictated by the local temperature; thus n_c and R_o are position (x) dependent.

Although Eq. (5) allows for a position-dependent CDW drift velocity, the experimental results summarized in Fig. 3 suggest that for a moving CDW, the CDW drift velocity is single-valued. A position-dependent drift velocity would lead to a distribution in noise frequencies, and hence an increased smearing of the noise spectrum with increasing dT. This is in strong contrast to what is observed experimentally. What we have observed is that v_d, and hence the associated noise spectrum, is determined uniquely by the average sample temperature, and not by any particular local temperature, for example the temperature of the contacts.

The average value of the CDW carrier concentration for the whole sample is given by, $\overline{n_c} = \overline{I_{CDW}}/v_d e$, where

$$\overline{I_{CDW}} = \frac{1}{\ell}\int_0^\ell n_c(x) e v_d dx = \frac{ev_d}{\ell}\int_0^\ell n_c(x) dx \ . \quad (6)$$

In the limit $R_o(x) \rightarrow$ const., Eq. (5) reduces to Eq. (6). Hence, for small values of dT, the ratio $\langle I_{CDW}\rangle/f$ is expected to reflect the average carrier concentration $\overline{n_c}$. Fig. 4 shows $\langle I_{CDW}\rangle/f$ measured at various values of dT. It is evident that, for low values of dT, the observed average carrier density $\overline{n_c} \sim \langle I_{CDW}\rangle/f$ is equivalent to the carrier density obtained under isothermal conditions, with $T = T_{avg}$. At larger values of dT, $\langle I_{CDW}\rangle/f$ deviates from the isothermal n_c, as expected from the definition of $\langle I_{CDW}\rangle$ and the finite position dependence of R_o.

Our experiments suggest that the average temperature and carrier concentration in $NbSe_3$ determine uniquely the threshold field and "noise" characteristics in the presence of a temperature gradient.

We now further discuss our results. Several length scales are important in the statistics and dynamics of CDW condensates. The amplitude correlation length

$$\xi_A = \varepsilon_F/kT_P \quad (7)$$

where ε_F is the Fermi energy and T_p is the transition temperature, is approximately 100 Å and determines the length scale over which a local temperature (and local order parameter and n_c) can be established. This length scale is orders of magnitude smaller than all other relevant length scales of the problem. The Fukuyama-Lee-Rice[8] phase phase correlation length ξ_{FLR} is determined by the decay of phase correlation in the pinned mode, and

$$\langle\phi(0)\phi(x)\rangle \sim \exp(-x/\xi_{FLR}) \quad (8)$$

where $\xi_{FLR} = \kappa/V_o c$ where κ is the elastic constant associated with the condensate, V_o is the pinning potential associated with impurities of concentration c. ξ_{FLR} is the order of 1 µm- 1 mm[1] and is comparable although smaller than the size of the specimens. ξ_{FLR} determines the threshold field, which experimentally is found to be temperature dependent. Consequently a distribution of local E_T values is expected in the presence of a thermal gradient. The effective E_T which is measured in the presence of finite at dT may be simply understood using an electrical analog consisting of a series of reverse biased Zener diodes, as shown in the inset of Fig. 2. The breakdown voltage for the i-th diode is V_{Zi} . If we consider each diode unit to be of "length" ℓ_i, the breakdown field for the circuit becomes

$$E_T = \sum_i V_{Zi} \ / \ \sum_i \ell_i \ . \quad (9)$$

It is possible that $E_T < E_i$, where E_i is the breakdown field for the i-th element. As a concrete example, consider three diodes, each 1 cm in "length," and each with $V_Z = 2V$. The local threshold field is 2 V/cm, and is equivalent to E_T for the entire circuit. If we assume a parameter gradient such that $V_{Z1} = 1$ V, $V_{Z2} = 2$ V, and $V_{Z3} = 3$ V, E_T becomes 6V/3cm = 2V/cm, less than ^3E_T = 3V/cm for element 3. Thus our experimental observation of a sharp threshold field in $NbSe_3$ may be adequately explained by a simple distribution of local threshold fields. It is important to note that this distribution need not lead to a smearing of the actual threshold field. This is clear from the diode analogy, where the threshold E_T is equally sharp, at 2V/cm, with or without a distribution in parameters.

A third correlation length, which we call ξ_D, the dynamic coherence, determines the length scale over which the time dependence of the phase $d\phi/dt = \dot{\phi}$ is coherent. As the drift velocity is given by

$$v_d(x) = \frac{1}{\pi}\dot{\phi}(x) \quad , \quad (10)$$

ξ_d also establishes the length scale over which v_d is uniform. Our experiments thus indicate that $\xi_D > \ell$, the length of the sample, and $v_d = v_d(T_{avg})$, i.e., v_d is not determined by the local, but by an average, temperature. Similar coherent response was observed recently in specimens of various dimensions,[9] where it was shown that while velocity correlations are maintained over the specimens, phase correlations are not. While it is apparent that ξ_{FLR} and ξ_D are different quantities, we are not aware of any microscopic description in which they both are evaluated. The question, however, has recently been addressed.[11] We also note that our experiments demonstrate that neither the threshold electric field nor the coherent current oscillation characteristics depend on the temperature of the contacts, suggesting strongly that current oscillations observed at these frequencies are a bulk phenomenon. We finally remark that the experimental data presented here are appropriate for sample lengths approximately 0.5 – 1 mm. Whether a distribution of phase velocities can be induced by a temperature gradient for significantly longer samples, beyond the implied macroscopic dynamic phase coherence length, is presently under investigation.

We have learned recently that similar experiments were performed by Ong after our results were communicated to him.

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