Current Oscillations and Stability of Charge-Density-Wave Motion in $NbSe_3$

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Observations are reported of current oscillations with frequency proportional to current in the nonlinear conduction region in $NbSe_3$. At high electric field strengths the amplitude of the oscillations decreases with increasing charge-density-wave current. It is suggested that this behavior is due to the periodic motion of the charge-density wave as it slides through pinning potential, and good agreement is found when the potential is evaluated from the frequency-dependent conductivity. The same theory is used to estimate the stability of the pinned charge-density wave.

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A nonlinear current is observed in the quasi one-dimensional transition metal trichalcogenides $NbSe_3$[1] and TaS_3[2] when the applied field is above a threshold field, E_T. The current results from depinning of the charge-density waves (CDW's) to give Fröhlich sliding-mode conduction.[3] The excess current, $I(t)$, has, in addition to broad-band noise, current oscillations giving narrow-band noise at a fundamental frequency, ν, and various harmonics with amplitude decreasing with the order of the harmonic.[1,4,5] The relative magnitude of the harmonics is independent of frequency.

We report direct measurements of the amplitude and frequency of the current oscillations at various applied fields E above E_T in the lower CDW state ($T_c < 59$ K) of $NbSe_3$. It is suggested that this noise arises from sliding of the CDW over the peaks and valleys of the potential that pins the phase of the wave.

We have used two methods to observe the oscillating response of the $NbSe_3$ sample. In the first method a current pulse was applied through the sample, and the resulting voltage drop across the sample was displayed on an oscilloscope. The second method involved application of a voltage pulse to the sample and observation of the sample current on the oscilloscope. Both above methods employed unidirectional, rectangular pulses.

For applied currents I or voltages V less than the threshold value, V_T, for the onset of nonlinear conduction, a time-independent response with only broad noise was observed. A typical wave form obtained for a constant-current pulse in the nonlinear conductivity region above V_T is shown in Fig. 1.

The response is characterized by a time-dependent voltage, which is the sum of a constant and a periodic oscillating component: $V(t) = V + \Delta V g(t/\tau)$, where V is the time-average component of the voltage, and g is a periodic function of time t, of

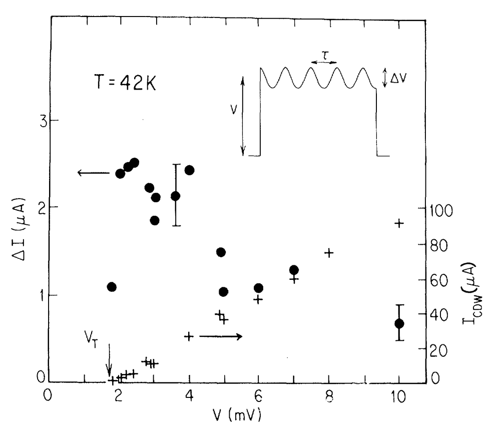

FIG. 1. Amplitude of the current oscillation ΔI and CDW current I_{CDW} vs voltage in $NbSe_3$. The threshold voltage $V_T = 1.75$ mV. The upper portion of the figure is a schematic representation of the response wave form from a current pulse with I well above I_T (see Ref. 5).

unit amplitude and period τ. For currents just above threshold, the wave form is a sensitive function of V, but for voltages V larger than approximately $1.5V_T$ the form of the oscillating component is independent of V up to highest voltages applied, $\sim 6V_T$. At higher voltages this has been confirmed earlier by measurements of the harmonic content.[5] The applied current I and the time-average voltage drop across the specimen was also measured by a dc technique. The threshold voltage V_T, the sample resistance R, and the excess current due to the moving CDW, $I_{CDW} = I(V) - I(\text{ohmic})$, where $I(\text{ohmic})$ is the current which corresponds to the ohmic conductivity measured below threshold, were evaluated from the measured I and V values.

The amplitude of the current oscillations ΔI together with I_{CDW} is shown in Fig. 1. These data were obtained using the constant-current pulse method, and we have used the relation $\Delta I = \Delta V/R$, where R is field-dependent sample resistance, to convert the measured voltage oscillation to an effective current oscillation. The arrow indicates the onset voltage of the oscillations and it also corresponds to the onset of nonlinear conduction V_T as measured by dc technique.

The relation between I_{CDW} and the frequency of the current oscillations $\nu = \tau^{-1}$ is shown in Fig. 2. As established by previous studies where the frequency of the current oscillations was measured at various applied fields,[4-6] ν is proportional to the CDW current. Experimental data similar to

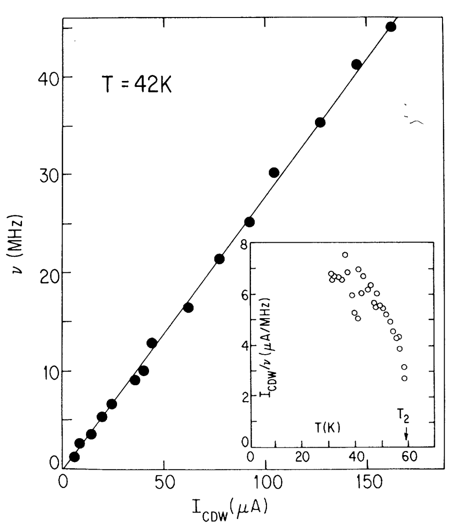

FIG. 2. Noise frequency $\nu = \tau^{-1}$ vs CDW current I_{CDW}. The inset shows I_{CDW}/ν measured at various temperatures.

those shown in the figure were taken at various temperatures, and the temperature dependence of I_{CDW}/ν is shown in the inset of Fig. 2. The linear relation between I_{CDW} and ν has been interpreted by Monceau, Richard, and Renard[4] as due to the motion of the CDW in a periodic potential. The CDW current is given by $I_{CDW} = n_c e V_d A$, where V_d is the drift velocity of the CDW, n_c is the density of electrons condensed in the CDW mode, and A is the cross section of the specimen. The oscillation frequency is $\nu = V_d/\lambda$, where λ is the wavelength of the CDW, leading to $I_{CDW}/\nu A = n_c e \lambda$.

With λ independent of the temperature, I_{CDW}/ν is a direct measure of the number of electrons participating in the CDW state. The temperature dependence of this ratio, as shown in the inset of Fig. 2, closely follows the order parameter as established by x-ray studies,[7] and can also be fitted well with the BCS expression for the temperature dependence of the order parameter Δ. The condensed fraction[8] $\rho_c = n_c/n$ varies as $\Delta(T)$ near the Peierls temperature, T_P. Such a fit gives $I_{CDW}/\nu = 8\ \mu A/\text{MHz}$ at $T = 0$. From the published room-temperature resistivity $\rho = 250\ \mu\Omega \cdot \text{cm}$ of similar specimens, the cross section A of the specimen can be evaluated and we obtain $I_{CDW}/\nu A = 38\ \text{A}/(\text{MHz} \cdot \text{cm}^2)$, in good agreement with previous experimental results[4-6] that suggest that λ corresponds to the wavelength of the CDW, $\lambda = 14$ Å. However, the uncertainties are such that if, as suggested by Bak[9] and by Weger and Horowitz,[10] the CDW is commensurate over most of the volume, the period possibly could be the lattice constant $a \sim \lambda/4$ rather than λ.

The amplitude of the oscillating current just above threshold is a large fraction of the time-average current. Figure 3 shows $\Delta I/I_{CDW}$ as a function of V/V_T obtained from both the current-pulse and voltage-pulse methods. At $V = 1.05V_T$ the oscillating component has the same magnitude as the time-average CDW current. We were not able to evaluate ΔI and I_{CDW} closer to V_T, but we expect $\Delta I/I_{CDW}$ to increase further as V approaches V_T from above. We conclude that in this specimen the active volume must be acting as a single domain with a single degree of freedom for the motion of the CDW. A similar conclusion has been reached by Fleming[11] recently.

While the oscillating component of the CDW current is a large fraction of the time-average current just above threshold, ΔI decreases in the high-field ($V \gg V_T$) limit. In Fig. 3 we also show $\Delta I \times I_{CDW}$ which is constant within experimental

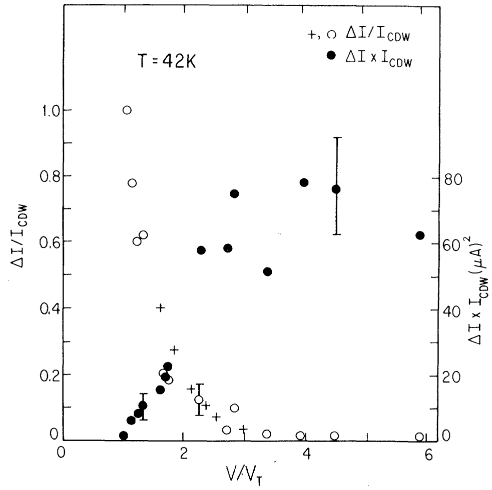

FIG. 3. $\Delta I/I_{\rm CDW}$ and $\Delta I \times I_{\rm CDW}$ vs V/V_T. Open and solid circles are constant-current pulses; crosses are constant-voltage pulses.

error at applied voltages between about $2V_T$ and $6V_T$. We account for this behavior by an extension of a model proposed recently[12-14] to account for the nonlinear and frequency-dependent conductivity and for ac-dc coupling experiments.[14-16] The theory does not depend on the details of the model but requires only that the CDW slides without additional dissipation over the peaks and valleys of the pinning potential.

Pinning may be described by a pinning potential $V(\varphi)$, where φ is the phase of the wave relative to that of the potential minimum $V(0)=0$. The potential $V(\varphi)$ is defined as the energy per electron condensed in the CDW without including the associated kinetic energy of the ion motion. The total energy is then $V(\varphi)$ multiplied by the Fröhlich mass ratio, $\alpha=(M_{\rm F}+m)/m$, to include the ion motion, and by N_c, the number of electrons condensed in the wave.

The coefficient of φ^2 in the expansion of $V(\varphi)$ in powers of φ, expressed in terms of the pinning frequency, ν_p, is

$$V(\varphi) \to \tfrac{1}{2} m^* \lambda^2 \nu_p^2 \varphi^2 = \tfrac{1}{2} m^* V_p^2 \varphi^2, \quad O(\varphi^4), \tag{1}$$

where m^* is the band mass, $\lambda=\pi/k_{\rm F}$ is the wavelength of the CDW, and V_p is the velocity for a frequency ν_p.

The maximum height of the barrier depends on its shape. If tunneling of electrons is described as Zener tunneling in a semiconductor model with an energy gap $h\nu_p$, the effective height of the tunneling barrier is[13]

$$V_{\rm max} = h^2 \nu_p^2/16E_{\rm F} = \tfrac{1}{2} m^* V_p^2, \tag{2}$$

which happens to be equal to the coefficient of φ^2 in (1). The peak of the sine-Gordon function $N^{-2}(1-\cos N\varphi)$ is $2/N\varphi$ which gives (2) for $N=2$ and is four times larger for $N=1$.[13,14] Equation (2) is also consistent with the impurity-pinning theory of Lee and Rice[8] which relates pinning energy with pinning frequency.

The current oscillations may result from the CDW moving adiabatically over the varying pinning potential in such a way that the total energy of the wave, kinetic plus potential, is constant. If the velocity of the wave is $V_d + V_1(\varphi)$, where V_d is the average velocity and V_1 is the periodic part that gives the oscillating current, then

$$\tfrac{1}{2} m^*(V_d + V_1)^2 + V(\varphi) = \text{const} = \tfrac{1}{2} m^* V_d^2. \tag{3}$$

Thus if $V_1 \ll V_d$,

$$V(\varphi) = -m^* V_d V_1(\varphi). \tag{4}$$

This implies that the current oscillations, given by V_1, should be inversely proportional to the drift velocity, V_d. As $I_{\rm CDW}=n_c e V_d A$ and $\Delta I = n_c e V_1 A$, Eq. (4) leads to $\Delta I \times I_{\rm CDW} = -V(\varphi)/m^* n_c^2 e^2 A^{-2}=$const, which is consistent with the values plotted in Fig. 3. We can combine Eq. (4) with the relation $\nu = I_{\rm CDW}/n_c \lambda A$ to obtain

$$V_1 V_d = (\Delta I/I_{\rm CDW})(\nu\lambda)^2, \tag{5}$$

where the right-hand side of Eq. (5) contains only parameters which are measured during the experiment.

For $V > 2V_T$, $\Delta I \times I_{\rm CDW} = 7\times10^{-11}$ A^2. With $\nu/I_{\rm CDW}=2.8\times10^{11}$ Hz·A^{-1} and $\lambda=14$ Å, Eq. (5) leads to $V_1 V_d = 7.7\times10^{-2}$ (cm/sec)^2. According to (2) this equals $\frac{1}{2}V_P^2$; thus $V_P=0.39$ cm/sec corresponding to a threshold frequency $\nu_P = V_P/\lambda = 2.8$ MHz. The threshold frequency can also be evaluated by using the scaling relation[12] between the field- and frequency-dependent conductivity predicted by the tunneling model. We have measured $\sigma(\nu)$ and $\sigma(E)$ on specimens from the same preparation batch, and the scaling of the ν and E dependence leads to $\nu_T=3.5$ MHz, in excellent agreement with the above estimate.

The same theory may be used to estimate the stability of a pinned CDW in a domain containing N_c electrons against thermal fluctuations. It is necessary that the maximum of the pinning barrier be larger than thermal energy, $\frac{1}{2}k_{\rm B}T$, or

$$\tfrac{1}{2}\alpha N_c m^* V_P^2 > \tfrac{1}{2} k_{\rm B} T. \tag{6}$$

For $\nu_p=3$ MHz, $V_P=0.4$ cm/sec, $\alpha=10^3$, and T

=40 K,

$$N_c > 3\times 10^{10}/(m^*/m), \qquad (7)$$

or the order of 4×10^{10}. There are about $2\times 10^{21} e/\mathrm{cm}^3$ in the chain, so that this corresponds to a volume greater than $4\times 10^{10}/2\times 10^{21} = 2\times 10^{-12}$ cm^3. The cross-sectional area for a domain of length 100 μm is then about 2×10^{-9} cm^2.

Thus to be stable, a coherent domain must have a large volume if the pinning frequency is small. In specimens for which the narrow-band noise is a large fraction of the time-average current, the entire volume must be acting as a single domain in the sliding CDW region with a single degree of freedom for motion of the CDW. The most likely explanation for this observation is that if there are CDW segments they are strongly coupled to lead to a highly coherent response.

Although the explanation of current oscillations given here is rather general and model independent, the agreement between the pinning frequency derived from the current oscillations and that derived from the scaling relation between field- and frequency-dependent conductivity lends strong support to the tunneling model of CDW depinning.

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[9] Per Bak, Phys. Rev. Lett. 48, 672 (1982). Bak suggested a different explanation of narrow-band noise in $NbSe_3$ based on motion of a soliton lattice. He suggested that the CDW is commensurate over most of the volume ($\lambda_{CDW} = 4a$) with the extra charges corresponding to the difference from commensurability going into solitons or discommensurations that form a regular lattice with spacing l. In both his model and ours the current per chain may be written $I = e^*v/l = e^*\nu$, where ν is the frequency, v is the velocity of motion, and e^* is the charge in length l. For the soliton lattice $e^* = e/2$ while for transport by incommensurate CDW's $e^* = 2e\rho_c$ and $l = \lambda_{CDW}$. Earlier values quoted by Bak that favor $e^* = e/2$ were based on measurements near T_P where ρ_c is small. We feel that the temperature dependence, which follows that of ρ_c, gives strong support for transport by CDW's. It is possible that a soliton lattice could exist if the entire CDW system, including solitons, moves under the influence of the field.

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