Volume Dependence of Current Oscillations in $NbSe_3$: A Finite-Size Effect

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Experiments performed in the linear chain compound $NbSe_3$ demonstrate that the oscillations associated with the current-carrying charge-density-wave condensate decrease with increasing volume of the specimens. The observed dependence of the oscillating current density Δj_1 on volume Ω, $\Delta j_1 \propto \Omega^{-1/2}$, is attributed to interference between independent charge-density-wave domains which oscillate at the same frequency but randomly in phase.

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The highly unusual transport properties[1] of inorganic linear chain compounds with an incommensurate charge-density-wave (CDW) ground state, such as nonlinear and frequency-dependent conductivity, $\sigma(E)$ and $\sigma(\omega)$, are due to the collective response of the electrons condensed in the CDW mode. A remarkable phenomenon, the observation of coherent current oscillations[2] with frequency proportional to the current carried by the condensate,[3] is regarded as the most compelling evidence of translational CDW motion, a mechanism originally postulated by Fröhlich.

The reason for the observation of oscillation phenomena in macroscopic samples is not clear at present. Two phenomenological models, one classical,[4] the other quantum mechanical,[5] neglect the internal degrees of freedom of the condensate, and discuss the motion of the condensate in a periodic potential $V(\varphi)$, where $\varphi = 2k_F x$ is the phase of the condensate and k_F is the Fermi wave vector in the chain direction. The period of the potential is determined by the CDW wavelength, $\lambda = \pi/k_F$. While these models account for a broad variety of observations, they do not answer the question of how randomly positioned impurities can lead to a periodic potential in the thermodynamic limit.

Another approach[6] considers the local deformations of the CDW around impurities. The pinning of the collective mode is due to the energy gain associated with these deformations. While this model and various extensions using computer simulations,[7] diagram techniques,[8] hydrodynamic approach,[9] and mean-field treatment of the dynamics[10] account for certain features of $\sigma(E)$ and $\sigma(\omega)$, they all suggest that long-range phase coherence is absent, and that coherent current oscillations cannot be observed in the infinite-volume limit.

A different model, proposed recently by Ong, Verma, and Maki[11] and independently by Gorkov[11] suggests that the oscillations originate at the contacts. The difficulty associated with rough contact surfaces is avoided by postulating generation of vortices at the contacts. Models in terms of discommensuration or soliton lattices have also been advanced.[12]

In this communication we report our experiments on the dependence of the oscillation amplitude on the length l and cross section A of the specimens. The oscillation amplitude normalized to current carried by one conducting chain decreases with increasing volume $\Omega = lA$ and can be represented as $\Delta j_1 = \text{const} \times \Omega^{-1/2}$.

The oscillation amplitude in the $NbSe_3$ samples was measured below the second phase transition with a spectrum analyzer using a two-probe configuration and constant current bias. An analyzer bandwidth of 100 kHz was used in order to measure all the power in the oscillation peak. Figure

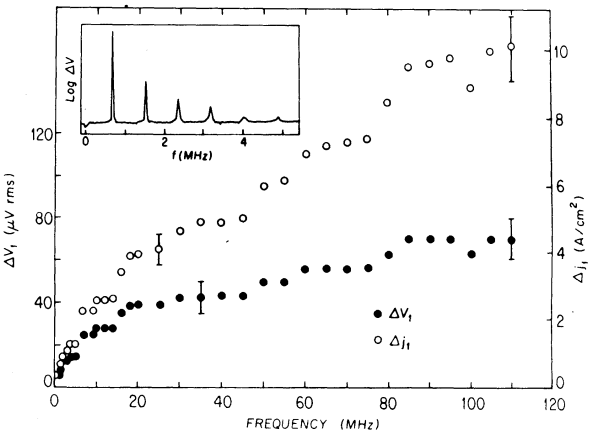

FIG. 1. Amplitude of the fundamental oscillation in $NbSe_3$ at 42.5 K, vs oscillation frequency. The amplitude is plotted both as a voltage ΔV_1 (solid symbols, left-hand scale) and as an equivalent current density Δj_1 (open symbols, right-hand scale). The inset shows a typical spectrum.

1 shows the rms oscillation voltage at 42.5 K versus oscillation frequency for a specimen of length $l = 1.03$ mm and cross section $A = 16\ \mu m^2$. l and A were determined, respectively, from direct microscope examination and from room-temperature resistance (with use of $\rho_{RT} = 250\ \mu\Omega$ cm)[13] and were corroborated by the slope of the excess current versus frequency relationship at 42.5 K ($j_{CDW}/f = 30$ A/MHz cm^2).[13] Only the voltage corresponding to the fundamental peak is plotted in Fig. 1; the harmonics have a similar dependence. The voltage ΔV_1 in the fundamental increases rapidly at low frequency, then more slowly, but continues to rise even by 110 MHz.[14,15]

The amplitude may also be characterized by an oscillating current density, $\Delta j_1 = \Delta V_1/RA$, where R is the measured (nonlinear) dc resistance under the applied bias field. Defined in this way, Δj_1 is the current oscillation which *would* be observed in a constant-voltage experiment. In the attained frequency range, Δj_1 rises more rapidly than ΔV_1, as shown in Fig. 1. Comparisons between samples of different dimensions will be made at fixed time-averaged current density, corresponding to fixed oscillation frequency; we have arbitrarily chosen to compare the mean oscillation amplitude in the range 10 to 20 MHz.

Each sample used in this investigation was cut into three or more different lengths and remeasured. The length dependence of the voltage oscillations is shown in the log-log plot of Fig. 2(a) for samples of three different cross sections. The voltage amplitude increases with increasing length at a rate approximately equal to, or slightly weaker than, $\Delta V_1 \propto l^{1/2}$. The cross-section dependence of ΔV_1 for samples of fixed length $l = 0.3$ mm, determined from plots like Fig. 2(a), is shown in Fig. 2(b). A good square-root dependence $\Delta V_1 \propto A^{-1/2}$ is obtained for all samples but the one of smallest A. Thus, $\Delta V_1 \propto (l/A)^{1/2}$, suggesting that intrinsic parameters are determined by the volume alone: $\Delta j_1 = \Delta V_1/\rho l \propto \Omega^{-1/2}$. Figure 3 shows this $\Omega^{-1/2}$ dependence of Δj_1, extracted from ΔV_1 with use of the experimental nonlinear resistance R.

We have also measured the threshold field E_T for the onset of nonlinear conduction, but found no systematic variation with the sample volume, in agreement with earlier studies.[16]

Our experiments clearly establish that the amplitude of the current oscillations decreases with increasing volume, suggesting that the oscillations are finite-size effects which would not be observed in the thermodynamic limit. It is evident, however, that the observation of the oscillating current in macroscopic specimens is associated with extremely large length scales. While the amplitude correlations are short in

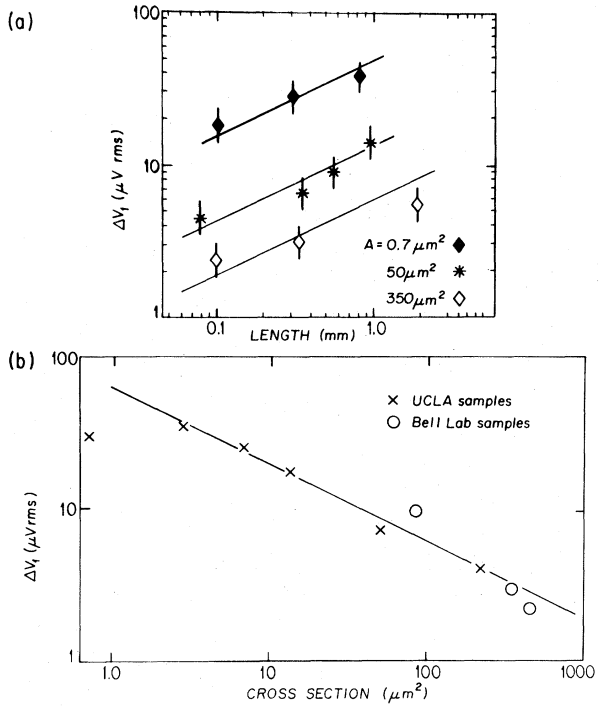

FIG. 2. Oscillation amplitude ΔV_1 in $NbSe_3$ at 42.5 K, measured between 10 and 20 MHz, for samples of different dimensions. (a) ΔV_1 vs length for samples of three cross sections A. The lines have slope $\frac{1}{2}$. (b) ΔV_1 vs A for samples of fixed length $l = 0.3$ mm. The line has slope $-\frac{1}{2}$.

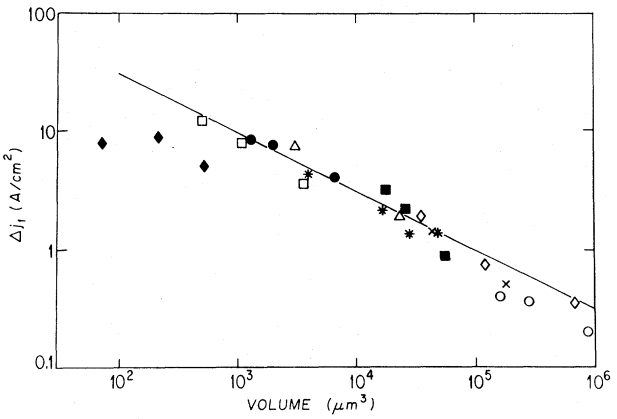

FIG. 3. Oscillation amplitude Δj_1 in $NbSe_3$ at 42.5 K vs sample volume Ω for all samples. The line represents $\Delta j_1 = (3 \times 10^{-4}\ \text{A cm}^{-1/2})\Omega^{-1/2}$.

CDW systems, the important length scale is that associated with the phase correlations $\langle \varphi(0)\varphi(x)\rangle$. The length L over which the phase fluctuation $\langle \delta\varphi^2\rangle^{1/2}$ is less than 2π has been calculated by Fukuyama and Lee[6] and by Lee and Rice[6] who obtain

$$L = 2\pi/n_i V_0^2, \quad (1)$$

where V_0 is the strength of the impurity potential and n_i is the impurity concentration. Although the phase coherence decreases smoothly with increasing distance, the phase of the condensate is usually assumed to be constant within a "domain" of size L^3, and the specimens are usually[8] treated as composed of domains with weak correlations between the phases of different domains. Within this framework, two important size regimes can be distinguished.

For a small sample where $\Omega \lesssim L^3$, the entire CDW acts as one coherent "domain" and may be considered rigid throughout. In the pertinent phenomenological theories,[4,5] the oscillations at high frequencies are related to the strength of the periodic potential which is assumed to have the form

$$V(\varphi) = (m\omega_0^2/2k_F)\sin(2k_F x) \quad (2)$$

per condensed electron, where $m\omega_0^2$ is the restoring force and m is the mass of the condensate. In the limit of small damping, the total (kinetic and potential) energy of the moving condensate is constant,[13] and one obtains for large velocity ($j \gg \Delta j_1$)

$$\Delta j_1 = (\omega_0/2\pi f)^2 j. \quad (3)$$

For overdamped motion[4] one obtains, again for $j \gg \Delta j_1$,

$$\Delta j_1 = (\omega_0^2 \tau/2\pi f) j. \quad (4)$$

When both damping and inertial effects are included, it is expected that Eq. (4) is appropriate at moderately large velocities, while inertial effects dominate at extremely large velocities. In both cases $j \propto f$, and the limiting frequency dependences are $\Delta j_1 \propto f^{-1}$ for Eq. (3) and $\Delta j_1 \to$ const for Eq. (4). While the two cases predict different frequency dependences, Δj_1 is proportional to $V(\varphi)$ in both. If the CDW is rigid throughout the sample, the periodic potential per electron may be expected to decrease like $V(\varphi) \propto \Omega^{-1/2}$ because of the random distribution of impurities. This expectation does not, however, provide a viable interpretation of the volume dependence of Δj_1 because, within the rigid-CDW models, the threshold field E_T has the same dependence[4] on $V(\varphi)$:

$$E_T = (\lambda/2\pi) m\omega_0^2/e.$$

Therefore the lack of a corresponding $\Omega^{-1/2}$ dependence of E_T rules out this interpretation.

As E_T is independent of l and A, it is natural to assume[6] that the dimensions of the coherent CDW "domains" are much smaller than the sample dimensions. In a large sample, $\Omega \gg L^3$, each of the $N \approx \Omega/L^3$ independent domains oscillates with random phase, leading to a $\Omega^{-1/2}$ behavior of Δj_1.

An estimate of the domain volume may be obtained by comparing the observed current oscillations with the rigid-CDW predictions. As we do not observe in these specimens a decreasing Δj_1 at the highest frequencies investigated, the estimate will be made using Eq. (4), according to which $\Delta j_1 \to$ const for $\Delta j_1 \gg j$. It is clear from Fig. 1 that if our Δj_1 data ever attain a constant value, it will be at least 4 times as large as Δj_1 between 10 and 20 MHz. Thus a lower limit for Δj_1 in the high-velocity overdamped case is $\Delta j_1 = (1.2\times10^{-3}\ \mathrm{A\ cm^{-1/2}})\Omega^{-1/2}$. The overdamped model has been fitted[17] to the frequency-dependent conductivity at 42 K with use of $\omega_0^2\tau/2\pi \approx 100$ MHz, whence Eq. (4) becomes $\Delta j_1 = 3\times10^3\ \mathrm{A/cm^2}$. Equating these two expressions gives $\Omega = 0.2\ \mu\mathrm{m}^3$ as an approximate lower limit for the domain volume. An estimate based on the underdamped model, Eq. (3), is problematic because the high-velocity limit, in which $\Delta j_1 \propto f^{-1}$, is clearly not attained in our measurements. We regard the above result, however, as a good order of magnitude estimate. The domain size evaluated here is consistent with a domain of dimensions given by Eq. (1), and with x-ray results of Fleming,[18] and it is also not much less than the minimum stable domain size evaluated on the basis of the tunneling model.[13] We also note that specimens with higher purity should have large domains. This, we believe, may explain the highly coherent response observed earlier[13] in high-quality specimens.

In conclusion, this investigation has found an $\Omega^{-1/2}$ dependence of the oscillating current density over a wide range of volume, strongly suggesting that the current oscillations in $NbSe_3$ vanish in the thermodynamic limit. The observed volume dependence is interpreted in terms of independent CDW "domains" which oscillate at the same frequency, but randomly in phase. The domain size is estimated to be of order $0.2\ \mu\mathrm{m}$.[3]

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