Decoupling of ac dynamics from dc polarization below threshold for charge-density waves in $NbSe_3$

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Experiments on nonlinear charge-density wave motion in $NbSe_3$ demonstrate that the small-signal ac dynamics for megahertz frequencies is effectively decoupled from the dc response for bias voltages $|V_0| < V_T$ below threshold. Mixing signals generated at megahertz frequencies indicate an effective dc voltage $V_0' = 0$ for all $|V_0| < V_T$ in this region. This ac-dc decoupling is further illustrated by rectification measurements made in the presence of a large-amplitude ac signal. The results are consistent with a revised tunneling theory, but inconsistent with the simplest classical model.

Collective charge transport due to motion of charge-density waves (CDW's) is observed in $NbSe_3$ (Ref. 1) and TaS_3 (Ref. 2) below their Peierls transitions. The dc conductivity I/E increases due to CDW depinning for applied fields greater than a threshold field $E_T \sim 0.01$ to 1 V/cm, and eventually saturates at about the same conductivity that would be expected in the absence of a CDW condensation.[3] Similarly, the nonlinear ac conductivity due to CDW motion increases with applied frequency in the megahertz region, and at high frequencies approaches the same value as the high-field dc conductivity.[4] Periodic oscillations with frequencies proportional to CDW current are present at dc biases above threshold,[3,5] and this effect can produce interference phenomena with applied ac signals.[6,7] A number of experiments have shown that the CDW can store energy in metastable polarization states with extremely long relaxation times.[8] Experiments reported here demonstrate that the small-signal ac dynamics in the megahertz region is decoupled from the dc polarization below threshold—a result which is inconsistent with the simplest classical model, but consistent with a revised tunneling theory.

In the simple classical oscillator model,[9] the response of a rigid CDW is characterized by an overdamped motion of its position in the periodic pinning potential created by either spatial fluctuations in impurity density or lattice commensurability. Applied dc fields tilt the pinning potential in space as illustrated in Fig. 1, so that dc current accompanied by periodic oscillations occurs above the threshold field E_T. This picture is qualitatively inconsistent, however, with

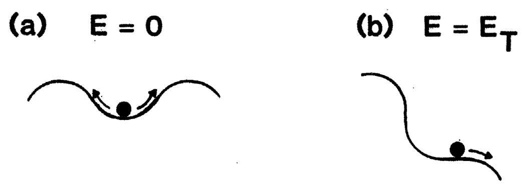

FIG. 1. Classical overdamped oscillator model: (a) at zero applied dc field, and (b) at threshold.

several pieces of experimental evidence. First, the differential conductivity dV/dI is predicted to diverge at E_T, while good quality crystals show a smooth onset of CDW conduction. Also, since the restoring force goes continuously to zero at threshold, the ac conductivity should depend dramatically on applied dc bias in this region. To the contrary, the observed ac conductivity is essentially independent of dc bias below threshold.[7,10] This fact could be explained if each well were taken to have a parabolic shape in a piecewise continuous periodic pinning potential. Such a picture would predict no harmonic mixing below threshold, however, in contrast to the experiments reported here.

The quantum theory of CDW conduction[11] is based on Zener-type tunneling of condensed electrons through a small pinning gap. This theory yields an expression for the dc current of the form

$$I(E) = I_N(E) + I_{\mathrm{CDW}}(E) = G_a(E) + G_b(E - E_T)e^{-E_0/E} \quad , \qquad (1)$$

when $E > E_T$. Here G_a and G_b represent the normal and CDW conductances, and E_0 is the activation field for Zener tunneling. Equation (1) has been found to fit the dc current accurately in both $NbSe_3$ (Ref. 12) and TaS_3.[13]

The theory of photon-assisted tunneling (PAT)[14] has been adapted to interpret the small-signal ac dynamics using only a fit of Eq. (1) to the measured dc I-V characteristic. The PAT theory predicts a scaling relationship between the voltage-dependent dc conductance and the real part of the frequency-dependent ac admittance taken at zero bias. This scaling behavior is observed in both $NbSe_3$ (Ref. 12) and TaS_3.[13] The nonlinear ac response was probed in mixing experiments[15] using an applied voltage of the form

$$V(t) = V_0 + V_1\cos(2\pi\nu_1 t) + V_2\cos(2\pi\nu_2 t + \phi) \quad . \qquad (2)$$

The generated current components $\Delta I\cos(2\pi\nu_0 t + \phi)$ for direct mixing ($\nu_0 = \nu_2 - \nu_1$) and harmonic mixing ($\nu_0 = \nu_2 - 2\nu_1$) were measured and compared with the tunneling theory. The amplitude of the mixing signal is given by $\Delta I = \frac{1}{2}V_1V_2 d^2I/dV_0^2$ for direct mixing or $\frac{1}{8}V_1^2V_2 d^3I/dV_0^3$

for harmonic mixing when ν_1 and ν_2 are small. According to the PAT theory, the derivatives are replaced by corresponding finite difference forms with a quantum step size $\Delta V = \alpha\nu$ proportional to the frequency. For example, d^2I/dV_0^2 becomes

$$[I(V_0+\alpha\nu_1)-2I(V_0)+I(V_0-\alpha\nu_1)]/(\alpha\nu_1)^2$$

when $\nu_0 \ll \nu_1$. A value of the parameter $\alpha \sim 1$ mV/MHz ($\sim 10^5$ h/e) is inferred from the scaling relation between ac and dc conductances, consistent with the theory for CDW tunneling over a distance ~ 10 μm with V_0 applied accross an $\sim$ 1-mm sample length.

In these previous experiments on ac conductance, direct mixing, and harmonic mixing in TaS_3,[15] the tunneling theory was shown to provide a complete and semiquantitative account of the observed small-signal ac dynamics for frequencies 1 to 1000 MHz. This agreement with experiment depends, however, on regarding all dc bias voltages below threshold as equivalent to zero bias in calculating the ac response, yielding the collapsed effective dc I-V curve illustrated in Fig. 2(a). The amplitude of the direct mixing signal, for example, is calculated as a second difference using the values of CDW current shown in the figure. At dc voltages above threshold, the total dc bias V_0 is replaced with an effective bias $V_0' = \mathrm{sgn}(V_0)(|V_0| - V_T)$. Using this revised interpretation, the PAT theory predicts a scaling between $I_{\mathrm{CDW}}(V)/(V-V_T)$ and the ac CDW conductance $G_{\mathrm{ac}}(\nu) - G_a$ measured at zero bias. This revised scaling is accurately obeyed over the entire experimental range of frequencies and dc fields in TaS_3. An applied dc field below threshold apparently distorts the CDW into a different metastable configuration,[8] without affecting its high-frequency behavior.

This revised interpretation of the tunneling theory predicts that there should be a bias-independent harmonic mixing signal in the region $V_0' = 0$ below threshold, since d^3I/dV^3 and its finite difference generalization are even functions of the effective dc bias. No harmonic mixing signal was observed in this region in the previous experiments on TaS_3 using a quasi-dc output frequency $\nu_0 = 1$ kHz. Figure 3 (solid lines) confirms that a measurable current is indeed generated in $NbSe_3$ when the output frequency

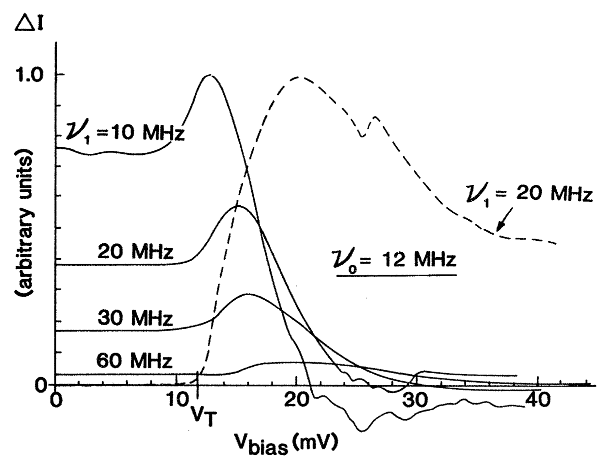

FIG. 3. Bias-dependent harmonic mixing (solid lines, $\nu_2 = 2\nu_1 + \nu_0$) and direct mixing (dashed line, $\nu_2 = \nu_1 + \nu_0$) for a sample of $NbSe_3$ at $T = 125$ K. Applied rf amplitudes were $V_1 = V_2 = 4.4$ mV. The relative scale for the direct mixing signal at $\nu_1 = 20$ MHz is approximately a factor of 4 higher than for the harmonic mixing curves.

($\nu_0 = 12$ MHz) becomes sufficiently large. Experiments in which ν_0 is varied while holding ν_1 constant show a monotonic increase of the zero-bias harmonic mixing signal with output frequency, until saturation takes place for $\nu_0 \gtrsim 12$ MHz. The same qualitative behavior is also observed in TaS_3 for ν_0 in the megahertz region.

In contrast with harmonic mixing, the tunneling theory predicts that direct mixing should vanish for $V_0' = 0$ below threshold at all applied frequencies. This has been verified at all frequencies observed in the ranges 1 MHz $\leq \nu_1 \leq 500$ MHz and $0 < \nu_0 \leq 100$ MHz. Figure 3 (dotted line) shows a representative bias-dependent direct mixing curve with $\nu_1 = 20$ MHz and $\nu_0 = 12$ MHz. It should be noted that a classical CDW oscillating in an anharmonic potential well would also yield a finite harmonic mixing signal but no direct mixing at zero bias. However, such a picture would require that the effective well does not tip with an applied dc potential below threshold, in contrast to the interpretation of the threshold field illustrated in Fig. 1(b).

A revealing aspect of CDW dynamics is the nature of the response in the presence of large-amplitude ac signals. Early measurements found that the low-field dc conductance increases with the amplitude of an applied ac field for $V_{\mathrm{ac}} > V_T$,[16] and this result was interpreted in terms of both the classical overdamped oscillator model[16] and the unmodified tunneling theory.[17] More recently, Zettl and Grüner[7] have measured dV/dI versus bias voltage in the presence of a large-amplitude ac signal. Their results show that the dc threshold decreases with increasing ac amplitude, but that a threshold voltage $V_T' < V_T$ remains sharply defined until it reaches zero and is completely suppressed.

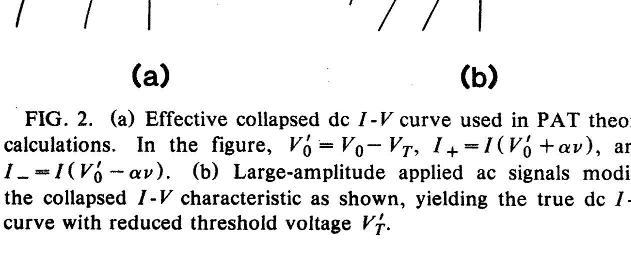

FIG. 2. (a) Effective collapsed dc I-V curve used in PAT theory calculations. In the figure, $V_0' = V_0 - V_T$, $I_+ = I(V_0' + \alpha\nu)$, and $I_- = I(V_0' - \alpha\nu)$. (b) Large-amplitude applied ac signals modify the collapsed I-V characteristic as shown, yielding the true dc I-V curve with reduced threshold voltage V_T'.

Our model predicts that a sufficiently large applied ac field should cause the effective collapsed I-V curve to approach a straight line with slope G_b as shown in Fig. 2(b). However, so long as the actual dc threshold V_T' has not been completely suppressed, a very low-frequency probe should detect a sharp peak in d^2I/dV^2 as a result of the rapid change in differential conductance at $V_0 = V_T'$. This expecta-

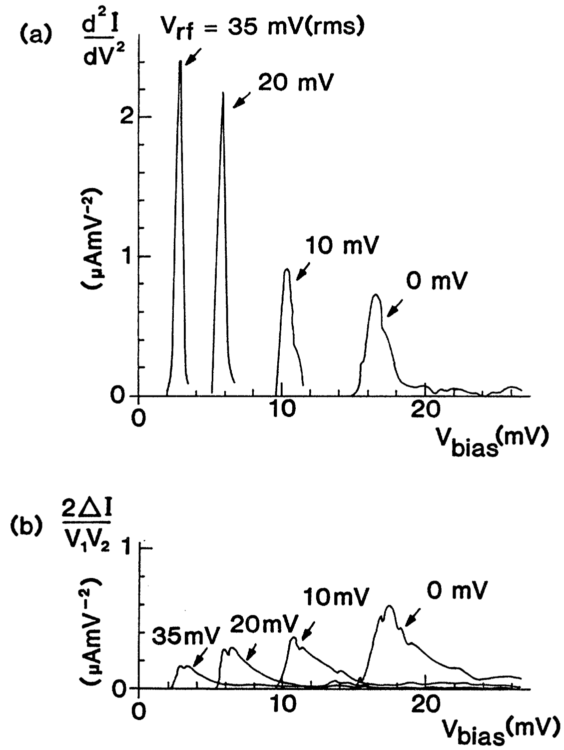

FIG. 4. (a) d^2I/dV^2 vs bias voltage for a sample of $NbSe_3$ at $T=125$ K in the presence of a 10-MHz signal with rms amplitudes indicated in the figure. (b) Direct mixing ($\nu_1=\nu_2=5$ MHz, $\nu_0=1$ kHz, $V_1=V_2=0.37$ mV) vs bias voltage in the presence of the same 10-MHz signals.

tion is confirmed in Fig. 4(a), which shows d^2I/dV^2 versus bias voltage for several amplitudes of an applied 10-MHz signal. The traces beyond the initial peaks at $V=V_T'$ are suppressed for clarity, since this region exhibits interference effects associated with the coherent oscillations. These measurements were performed by applying a 1-kHz, 0.3-mV (rms) signal to the sample and probing the 2-kHz component of the induced current with a lock-in amplifier.

If the small-signal ac dynamics is decoupled from the dc response at bias voltages below threshold, then the singularity at V_T' should not be present when the rectification is measured by small-signal direct mixing at megahertz applied frequencies. In this case, the effective I-V curve will be the *collapsed* curve of Fig. 2(b). Figure 4(b) shows that the peak value of the direct mixing signal for $\nu_1 \cong \nu_2 \simeq 5$ MHz and $\nu_0=1$ kHz is indeed successively reduced as the amplitude of the 10-MHz signal is increased. This remarkable difference in the rectification properties measured at 1 kHz and at 5 MHz, which are nearly identical in the absence of the large ac signal, provides strong evidence for the interpretation of ac-dc decoupling illustrated in Fig. 2.

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[1]P. Monceau, N. P. Ong, A. M. Portis, A. Meerschaut, and J. Rouxel, Phys. Rev. Lett. 37, 602 (1976).

[2]T. Takoshima, M. Ido, K. Tsutsumi, and T. Sambogi, Solid State Commun. 35, 911 (1980).

[3]R. M. Fleming and C. C. Grimes, Phys. Rev. Lett. 42, 1423 (1979).

[4]N. P. Ong and P. Monceau, Phys. Rev. B 16, 3443 (1977); G. Grüner, L. C. Tippie, J. Sanny, W. G. Clark, and N. P. Ong, Phys. Rev. Lett. 45, 935 (1980).

[5]P. Monceau, J. Richard, and M. Renard, Phys. Rev. Lett. 45, 43 (1980); J. Bardeen, E. Ben-Jacob, A. Zettl, and G. Grüner, *ibid.* 49, 493 (1982).

[6]J. Richard, P. Monceau, and M. Renard, Phys. Rev. B 25, 948 (1982).

[7]A. Zettl and G. Grüner, Phys. Rev. B 29, 755 (1984).

[8]J. C. Gill, Solid State Commun. 39, 1203 (1981); R. M. Fleming, *ibid.* 43, 167 (1982).

[9]G. Grüner, A. Zawadowski, and P. M. Chaikin, Phys. Rev. Lett. 46, 511 (1981); P. Monceau, J. Richard, and M. Renard, Phys. Rev. B 25, 931 (1982).

[10]R. E. Thorne, W. G. Lyons, J. H. Miller, Jr., J. Richard, J. R. Tucker, and J. Bardeen (unpublished).

[11]J. Bardeen, Phys. Rev. Lett. 42, 1498 (1979); 45, 1978 (1980).

[12]G. Grüner, A. Zettl, W. G. Clark, and J. Bardeen, Phys. Rev. B 24, 7247 (1981).

[13]A. Zettl, G. Grüner, and A. H. Thompson, Phys. Rev. B 26, 5760 (1982); A. Zettl, C. M. Jackson, and G. Grüner, *ibid.* 26, 5773 (1982).

[14]J. R. Tucker, IEEE J. Quantum Electron. 15, 1234 (1979).

[15]J. H. Miller, Jr., J. Richard, J. R. Tucker, and J. Bardeen, Phys. Rev. Lett. 51, 1592 (1983).

[16]G. Grüner, W. G. Clark, and A. M. Portis, Phys. Rev. B 24, 3641 (1981).

[17]J. R. Tucker, J. H. Miller, Jr., K. Seeger, and J. Bardeen, Phys. Rev. B 25, 2979 (1982).