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Role of current oscillations in ac-dc interference effects in $NbSe_3$

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We investigate the relation between rf-induced Shapiro steps and current oscillations in the charge-density-wave (CDW) state of $NbSe_3$. The Shapiro step magnitude is independent of the oscillation amplitude, and a rich spectrum of harmonic and subharmonic steps is observed. These results provide evidence for phase homogenization or excitation of internal modes of the CDW condensate.

Since its synthesis in 1975, the linear chain metal $NbSe_3$ has been the prototype material for studies of dynamical charge-density-wave (CDW) response.[1] The highly nonlinear dc conductivity, together with x-ray[2] and Hall effect[3] studies in the nonlinear conductivity region, provide clear evidence for a mobile collective-mode condensate. A highly controversial feature of dc CDW conduction remains the generation of coherent current oscillations or narrow-band "noise" in the nonlinear state.[4] Studies of other dynamical CDW systems, however, suggest that narrow-band noise is not restricted to $NbSe_3$, but is a general signature of CDW conduction.

Monceau, Richard, and Renard[5] were the first to demonstrate that interference effects could be achieved in $NbSe_3$ when ac and dc driving fields are combined. Subsequent studies[6] have shown that the interference effects, manifested as steps in the dc current-voltage (I-V) characteristics, are entirely analogous to Shapiro steps first observed in Josephson junctions. The steps are thus interpreted as a direct interference between the dc-induced current oscillations and the externally applied ac signal.

We have carefully examined the relation between Shapiro steps and narrow-band noise in $NbSe_3$. It is shown that, for moderate ac drive levels, the Shapiro step magnitude is independent of the narrow-band noise amplitude, and also independent of sample volume. We also observe an unusually rich spectrum of harmonic and subharmonic steps in the dc I-V characteristics. At high ac drive levels, sharp interference effects still occur, but the Shapiro step magnitude becomes random. In this limit the dc conduction is associated with a dramatic increase in low-frequency noise.

We have prepared single high-purity crystals of $NbSe_3$ by direct reaction of the elements, and have employed a two-probe sample mounting configuration with silver paint contacts. ac and dc currents were combined by high-frequency adder circuits, and detection was either by low-frequency lock-in (dc) or spectrum analyzer (ac) methods. Figure 1 shows a series of differential resistance measurements as a function of dc bias current and applied rf for a $NbSe_3$ crystal in the lower CDW state. These data were obtained by direct lock-in detection of dV/dI in the presence of superposed dc and rf currents. The rf amplitude V_{rf} indicated on the figure corresponds to that obtained for zero dc bias. For finite values of V_{rf} the sharp peaks in the nonlinear region of dV/dI correspond to Shapiro steps. In addition to inducing steps, the rf field also gradually reduces the dc threshold I_T,[7] and tends to flatten out dV/dI beyond I_T. In fact, for applied ac fields greater than approximately $5V_T$, with V_T the dc threshold voltage, dV/dI becomes absolutely independent of dc bias beyond I_T, neglecting the sharp Shapiro step structure. This constant value of dV/dI corresponds to the high field saturated limit of dV/dI, measured in the absence of rf. At moderate values of applied rf (corresponding to $I_{rf} \geqslant 0.1 I_T$) the Shapiro step structure is extraordinarily sharp and regular. At high rf levels, the interference peak amplitudes become apparently random. Consequently there is no obvious relation between the amplitudes of successive peaks, and also no correlation between the amplitude of a given peak at positive current bias and the amplitude of the corresponding peak at negative current bias. In addition, a large increase in low-frequency noise is observed in the bottom trace of Fig. 1. This effect is not due to sample heating as dV/dI retains its saturated value, and the positions of the interference peaks remain regular in current.

The major peaks in Fig. 1 correspond to $n\omega_{ex} = \omega_N$, where ω_{ex} is the external ac driving frequency, ω_N is the dc induced noise frequency, and n is an integer. Also evident is a rich spectrum of subharmonic peaks, corresponding to nonintegral values of n. This is shown more clearly in Fig.

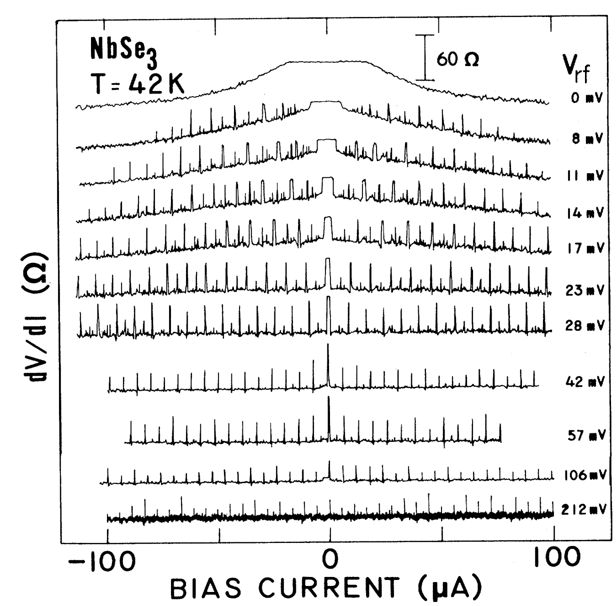

FIG. 1. dV/dI vs dc bias current for $NbSe_3$ in the presence of rf radiation at 5 MHz. The sharp peaks correspond to Shapiro steps.

2, which indicates numerous subharmonic peaks occurring for $n = m/p$, with m and p integers.[8]

The magnitude δV of the Shapiro steps represented in Figs. 1 and 2 is determined directly by the area bounded by the respective interference peaks in the dV/dI plots. To correlate δV to the dc-induced noise oscillations, we have measured during the same experimental run (in the absence of applied rf) the amplitude V_N of the fundamental peak in the narrow-band-noise spectrum. This was obtained by Fourier analysis of the voltage oscillations, and employed a digital average and integration technique. The measured absolute values of δV and V_N were normalized by V_T, the dc threshold voltage in the absence of rf. This normalization is appropriate for a current driven resistively shunted CDW condensate, where the shunt represents normal electrons.

Figure 3 shows, for various $NbSe_3$ samples, the relation between the narrow-band-noise amplitude and the $n = 1$ Shapiro step magnitude, for two selected temperatures. The rf amplitude used for each particular sample was that which yielded the maximum Shapiro step.[9] In all cases, this (variable) ac amplitude corresponded approximately to an ac current $I_{rf} = 3I_T$, for $\omega/2\pi = 5$ MHz. The astonishing feature of Fig. 3 is that, while V_N varies more than one order of magnitude between samples, δV *is constant for all samples,* at fixed temperature. Thus in this rf parameter range *the magnitude of the Shapiro steps is independent of the original narrow-band noise spectrum!* Although we expect δV and V_N to scale directly at very low values of rf amplitude, the independence of δV and V_N was verified to $I_{rf} = 0.1 I_T$. Below this driving field we were not able to accurately determine the Shapiro step magnitude. We note that the samples represented in Fig. 3 had geometric volumes differing by as much as two orders of magnitude; hence δV is also, within experimental error, *independent of sample volume.*

We now turn to an interpretation of these results. We write for the average CDW phase $\langle\phi\rangle$ an equation of motion

$$\frac{\delta^2\phi}{\delta t^2} + \frac{1}{\tau}\frac{\delta\phi}{\delta t} + \omega_P^2 \sin\phi = \frac{2k_F eE}{m^*} , \quad (1)$$

where τ is a phenomenological damping constant, m^* the

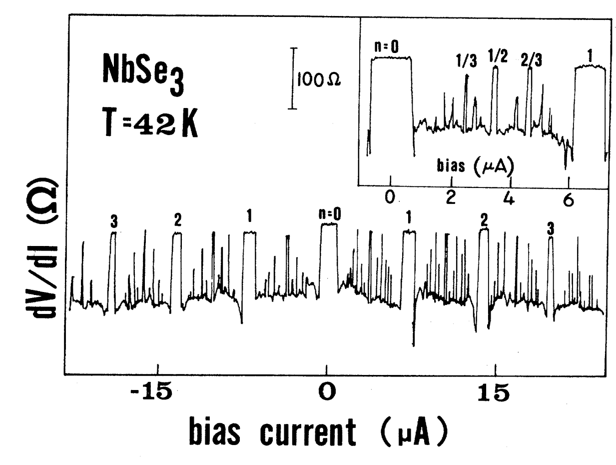

FIG. 2. dV/dI vs dc bias current for $NbSe_3$. The rf frequency is 5 MHz and the rf amplitude is 7 mV. A rich spectrum of harmonic and subharmonic steps is observed. The inset shows the subharmonics in greater detail.

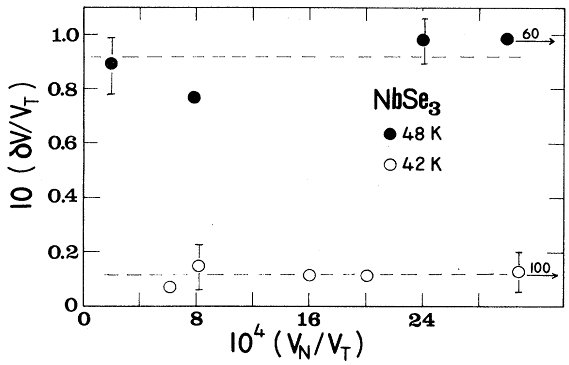

FIG. 3. Normalized Shapiro step magnitude δV and noise amplitude V_N for various $NbSe_3$ samples at two selected temperatures. δV is independent of both V_N and sample volume. The two right-hand-most points are off the horizontal scale.

effective mass of condensed electrons, ω_P an effective CDW plasma frequency, and k_F the Fermi wave vector. Replacing $\langle\phi\rangle$ with a displacement x results in the rigid particle classical model of Gruner, Zawadowski, and Chaikin.[10] This model has provided an excellent quantitative description of Shapiro step phenomena in $NbSe_3$.[6] The predicted step magnitude for the nth step is[6]

$$\delta V = \alpha_S V_T 2 |J_n(V_{rf}\Omega/\omega_{ex} V_T)| , \quad (2)$$

where J_n is the Bessel function of order n, Ω is a scaling constant, and α_S is the volume fraction of the sample which responds collectively to the external field. Equation (1) also predicts voltage oscillations associated with a nonlinear dc current. For a resistively shunted current driven condensate, the narrow-band-noise voltage oscillation amplitude is

$$V_N = \alpha_N V_T 2\Gamma(1+\Gamma)(I'^2 - 1)^{1/2}[I' - (I'^2 - 1)^{1/2}] , \quad (3)$$

where $I' = I_{dc}/I_T$, $\Gamma = (R_0/R_{sat}) - 1$ with R_0 and R_{sat}, respectively, the Ohmic and high-field limits of the sample resistance, and α_N describes the degree of phase coherence within the macroscopic crystal. The data of Fig. 3 correspond to fixed values of n, I', and ω_{ex}. Hence the normalized δV and V_N represent directly the parameters α_S and α_N. It is apparent that for fixed ω_{ex} and temperature, α_S is constant, whereas α_N is sample dependent.

We thus find that although the Shapiro steps phenomenon appears to represent an interaction between the narrow-band noise and the externally applied rf signal, the magnitude of the interference does not scale with the noise amplitude. The strong temperature dependence of α_N suggests a loss of volume phase coherence with lowering temperature below 48 K. This effect may in part contribute to the increase in dc threshold field with lowering temperature in $NbSe_3$.

Recently it was suggested[9] that the narrow-band noise associated with CDW conduction is a finite-size effect, where various domains within the sample oscillate at the same frequency but randomly in phase. In this description the noise frequency is fixed by the CDW drift velocity, but the noise amplitude is determined by the phase correlation function $\langle\phi(0)\phi(x)\rangle$ between various domains. For a macroscopic sample with dimensions exceeding the intrinsic phase-phase correlation length, not all domains oscillate in phase, and hence the noise amplitude is reduced from the ideal "single domain" value. If we assume the Shapiro steps reflect direct interaction between the external rf and the narrow-band noise, then an expected result is a direct scaling between the step magnitude and the noise amplitude. However, as demonstrated in Fig. 3, this is clearly not the case. In terms of domain structure, our experiments suggest that application of an rf field enhances $\langle \phi(0)\phi(x) \rangle$, providing increased phase coupling between domains. We identify this coupling process as "phase homogenization." Thus, independent of sample volume, phase coherence is induced by the external rf, leading to strong Shapiro step interference.

Our experimental results clearly suggest that Shapiro steps should persist in the infinite volume limit, although the noise oscillations may not.[11] CDW dynamics in the infinite volume limit has been addressed by Sneddon, Cross, and Fisher,[12] who consider a perturbation solution to the hydrodynamic problem. The CDW is treated as a deformable medium, where internal degrees of freedom are included. The solution for the CDW displacement predicts no current oscillations in the nonlinear conductivity region, but interference steps *are* predicted for combined ac and dc driving fields. The interference effects correspond to excitations of internal modes of the CDW condensate. The predicted differential conductivity near the interference steps is[12]

$$\frac{dI}{dE} \sim \frac{1}{v_d} J_n^2(gV_{\text{rf}}/\omega_{\text{ex}}) P[(gv_d - n\omega_{\text{ex}})/v_d^2] \quad , \tag{4}$$

where v_d is the dc velocity of the condensate, g represents a reciprocal lattice vector, and $P(z)$ is a function peaked at $z=0$. The behavior predicted by Eq. (4) in the infinite volume limit is in general qualitative agreement with that observed in Fig. 1. The Shapiro steps phenomenon at moderate rf drive levels may thus provide evidence for excitations of internal modes of the deformable CDW condensate.

In attempting to distinguish between phase homogenization and excitation of internal modes, it is instructive to consider more carefully the general shape of the Shapiro step structure shown in Fig. 2. It is apparent that the onset and loss of the interference is exceptionally sharp.[13] In addition, each step is associated with a fairly constant differential resistance, roughly equal to dV/dI at zero dc bias. In terms of Eq. (1), such sharp features are expected. The constant value of dV/dI on each step indicates a constant CDW current (and hence constant CDW velocity) on each step, consistent with *frequency* locking between narrow-band noise and the external rf.

The model of Sneddon *et al.*[12] does not require frequency locking for interference; rather, interference occurs whenever the intrinsic and applied frequencies are relatively close. Interference is thus expected over a finite range of CDW current, leading to continuous (but still reasonably sharp) "steps" in the I-V characteristics. The important point here, however, is that interference in the deformable medium theory represents internal mode excitations, the effects of which are quite independent of sample volume or narrow-band noise spectrum, as observed.

We conclude that a "rigid particle" treatment[10] of CDW dynamics is only applicable to $NbSe_3$ if domain structure, with the possibility of phase homogenization, is taken into account, whereas a deformable medium theory, without further assumptions, is in general agreement with our experimental results. At this point it is, however, difficult to clearly distinguish between these possible effects.

It is evident that, at sufficiently high rf drive levels, Eqs. (2) and (4) are no longer applicable to the CDW response. The asymmetry and apparent randomness of the interference phenomenon, together with the increase in low-frequency noise shown in Fig. 1, suggests system instability characterized by turbulent response. This may be related to the rf-induced period doubling route to chaos[14] and self-induced intermittent chaos,[15] both recently observed in $NbSe_3$. The rich spectrum of subharmonic steps observed for moderate rf driving fields is also suggestive of complex system response. For example, experiments subsequent to those reported here have investigated the possibility of a complete devil's staircase for the interference phenomenon.[16]

Finally, we note that preliminary experiments on Shapiro steps in TaS_3 suggest behavior similar to that reported here for $NbSe_3$.

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