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Complete charge-density-wave mode locking and freeze-out of fluctuations in $NbSe_3$

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We report the observation of complete rf-induced mode locking in the charge-density-wave (CDW) state of $NbSe_3$. Over each mode-locked region, the CDW phase velocity is uniquely determined by the frequency of the rf field, and all broadband noise normally associated with CDW conduction is absent. These observations demonstrate that broadband CDW conduction noise is not a necessary consequence of CDW motion. We suggest that, in the mode-locked state, random-phase velocity fluctuations are frozen out through a macroscopic synchronization of CDW domains.

Numerous transport and structural studies have by now clearly established that the spectacular nonlinear electrical properties of $NbSe_3$ and related compounds are the consequence of collective motion of the charge-density-wave (CDW) condensate.[1] In $NbSe_3$, the CDW for low electric field strengths is pinned by impurities, but a phase mode can be excited by application of a dc electric field exceeding a well-defined threshold field E_T. The "sliding" CDW condensate carries with it an excess current I_{CDW}.

In a current-regulated experiment, the CDW depins if the applied total current I exceeds a threshold current I_T, where $I_T = E_T l/R_0$ with l the sample length and R_0 the low-field (Ohmic) sample resistance representing normal (uncondensed) electrons. As first demonstrated by Fleming and Grimes,[2] for $I > I_T$ the sample voltage contains both coherent and incoherent frequency structure. The coherent structure, or "narrow-band noise," has a fundamental frequency directly proportional to I_{CDW},[3] while the incoherent structure, or "broadband noise," often follows a $1/f^\alpha$ power spectrum for fixed current, with α near 1.[4] Similar CDW conduction noise has been observed in the sliding CDW states of TaS_3, $K_{0.3}MoO_3$, $(TaSe_4)_2I$, and $(NbSe_4)_{3.33}I$, and it appears to be a general trademark of CDW motion.[1] However, the origin of the noise is not well understood, and is presently the subject of intense study.

Early experiments[3] on $NbSe_3$ identified interference between the intrinsic narrow-band noise and externally applied rf signals through sharp spikes in the differential resistance, and subsequent studies[5] have demonstrated that the associated "steps" in the current-voltage (I-V) curves are analogous to Shapiro steps first observed in Josephson junctions. Strong interference occurs for $\omega_N = n\omega_{rf}$, with ω_N the intrinsic narrow-band noise frequency, ω_{rf} the frequency of the externally applied rf, and n an integer. More recent studies[6,7] of the Shapiro step phenomenon in $NbSe_3$ have investigated the relationship between the narrow-band noise amplitude and the magnitude of the Shapiro steps, and also subharmonic interference structure corresponding to nonintegral values of n.

We have investigated interference phenomena in $NbSe_3$. Under appropriate conditions of applied dc and rf currents, we find that the phase of the *entire* CDW can be completely mode locked to the frequency of the external rf drive. The locking persists over well-defined ranges of dc bias current (or dc bias field), and complete harmonic and complete subharmonic locking occurs. In the mode-locked states (and only there), all broadband noise normally associated with CDW conduction vanishes identically. These observations indicate that, during complete mode locking, all random internal phase velocity fluctuations of the CDW condensate are frozen-out.

We have prepared nominally pure single crystals of $NbSe_3$ by direct reaction of the elements. A two-probe sample mounting configuration was used exclusively, with conductive silver paint contacts. The perturbation applied to the sample was a superposition of a dc current and two ac currents. One ac current was of very low frequency (200 Hz) and amplitude, and provided a suitable signal for lock-in detection of the differential resistance of the sample. A bridge circuit was used for this purpose. The second rf current, typically in the MHz frequency range, was the source of the Shapiro step interference. Broadband noise measurements were obtained by first amplifying the voltage across the sample with a low-noise preamplifier (bandwidth 0.03 Hz–10 kHz, gain 10^4), followed by detection with either a sensitive rms voltmeter or a low-frequency spectrum analyzer.

Figure 1(a) shows the differential resistance dV/dI and broadband noise of $NbSe_3$ at $T = 48$ K, plotted versus dc bias current I. The threshold current I_T identified by the sharp bend in dV/dI also corresponds to the threshold for the onset of broadband noise. It is evident that the broadband noise amplitude is a strong function of excess CDW current I_{CDW} and there exists a broad maximum in the noise amplitude for bias currents slightly above I_T. The measurements of Fig. 1(a) are consistent with earlier studies of differential resistance and broadband noise in $NbSe_3$.[2,8] Figure 1(b) shows the effect of an externally applied rf current on the differential resistance and broadband noise of the same $NbSe_3$ crystal. The addition of rf current reduces I_T, although the threshold for CDW conduction (break in dV/dI) still coincides with the onset of broadband noise. The sharp step structure in the upper curve in Fig. 1(b) corresponds to Shapiro step interference, and both harmonic and subharmonic structure is observed. The step labeled $n = 1$ corresponds to a region where the fundamental narrow-band noise frequency ω_N equals the externally applied rf frequency, ω_{rf}. The important features of the $n = 1$ step structure are that the top of the step appears flat over a finite range of dc bias current, and that over this range dV/dI corresponds *exactly* to the low-field (Ohmic) differential resistance. Over the finite range of bias field comprising a step, the CDW velocity (and hence the internal period corresponding to the time derivative of the CDW phase)

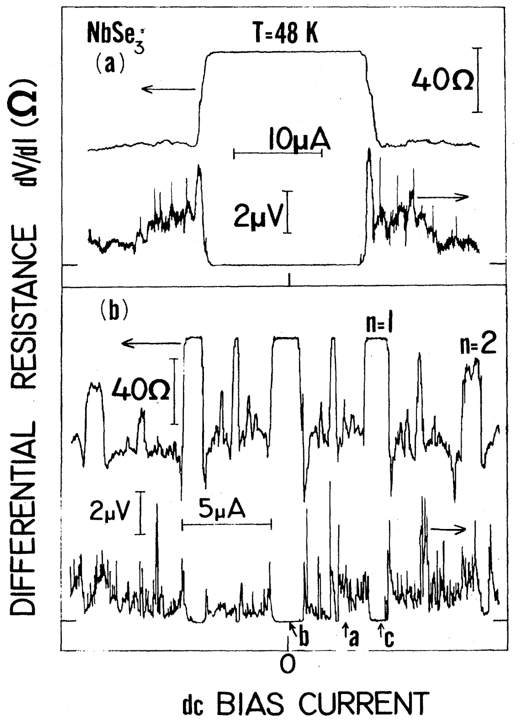

FIG. 1. (a) Differential resistance dV/dI and broadband noise amplitude vs bias current in $NbSe_3$; (b) same as (a), except that an rf current ($\omega_{rf}/2\pi = 2$ MHz, $I_{rf} = 40$ μA) has been added to the sample. Complete mode locking is evident on the step labeled $n = 1$. All broadband noise vanishes in the mode-locked regime. The lettered arrows refer to where the corresponding frequency spectra of Fig. 2 were taken.

does not change, but remains "locked" to the frequency of the applied rf field. The observation that dV/dI attains its Ohmic value on the step confirms that the mode locking is *complete,* and involves the entire CDW condensate. This is the first observation of complete mode locking for an rf current driven CDW condensate. We note that not all interference steps in Fig. 1(b) display complete locking. For example, only one of the subharmonics locks completely (at $n = \frac{1}{2}$), and although harmonics equal to or greater than $n = 2$ show some evidence of locking, they fail to achieve the Ohmic value for dV/dI. Similar incomplete locking was first reported by Hall and Zettl.[6]

The fact that the CDW velocity is "fixed" in the mode-locked states suggests that related "plateau" structure might occur in the broadband CDW conduction noise response. One expectation might be that the broadband noise amplitude should remain constant over the whole range of mode lock, with a value dictated by the (fixed) CDW velocity. This value could be extracted from Fig. 1(a). The lower trace of in Fig. 1(b) shows, however, dramatically different results. The observed behavior of $NbSe_3$ is that, on a mode-locked step, *all broadband noise vanishes identically.* The complete suppression of noise occurs only for those (harmonic or subharmonic) steps which display *complete* mode lock; on the $n = 2$ step in Fig. 1(b), for example, which shows incomplete locking, the broadband noise is significantly reduced, but not identically zero.

To investigate further the broadband noise response in the presence of mode locking, we have measured the frequency spectrum of the noise of the range of 0–100 Hz. Figure 2(a) shows the response for applied dc and rf currents yielding a sliding CDW with incomplete mode locking. The noise power level here follows a $1/f^{\alpha}$ law, with $\alpha = 1.1 \pm 0.1$, consistent with other studies of broadband noise in $NbSe_3$ (Ref. 4) and the related compound TaS_3.[9] Figure 2(b) shows the spectral response for zero applied dc current. In this pinned regime, no CDW conduction noise is generated, and only instrumental noise is observed. Figure 2(c) demonstrates that when the $NbSe_3$ sample is dc and rf driven to a complete mode-locked step, the spectral response again becomes identical to that of a pinned CDW condensate. Thus, in the mode-locked state, all fluctuations leading to broadband noise are completely frozen-out.

A number of models have been proposed to describe the dynamics of a depinned CDW. Bardeen[10] has suggested that quantum tunneling plays an important role in CDW transport, although in this model the generation of broadband noise has not been explicitly addressed. Pietronero and Strassler[11] have performed computer simulations on a finite-size one-dimensional classical model originally proposed by Fukuyama and Lee.[12] Here both narrow-band and broadband noise are observed, but no power-law behavior is obtained in the low-frequency limit for the broadband noise. The classical hydrodynamical model of Sneddon, Cross, and Fisher[13] predicts no narrow-band noise in the infinite-volume limit, although interference does obtain for combined dc and ac driving fields.

A highly simplified model of CDW transport is that of Grüner, Zawadowski, and Chaikin,[14] where the center of mass of the CDW obeys an equation of motion

$$\frac{d^2\theta}{dt^2} + \frac{1}{\tau}\frac{d\theta}{dt} + \omega_p^2 \sin\theta = \frac{q_0 eE}{m^*} , \tag{1}$$

where θ is the CDW phase, τ is a phenomenological damp-

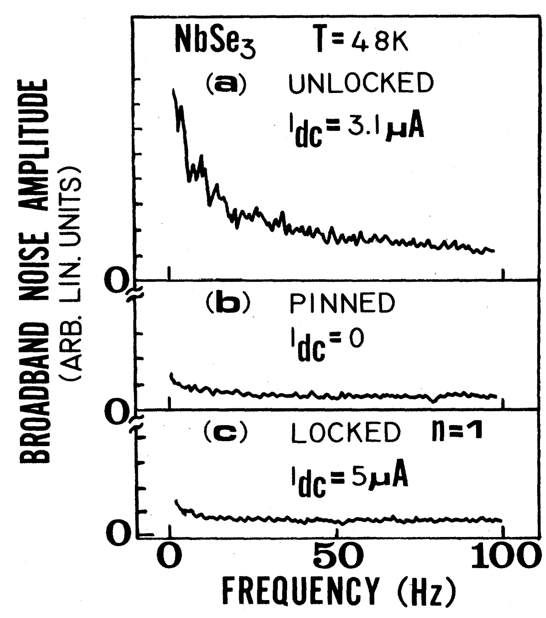

FIG. 2. Broadband noise spectrum in $NbSe_3$ (a) depinned CDW with rf current but without mode locking; (b) pinned CDW; (c) in a mode-locked regime. $\omega_{rf}/2\pi = 2$ MHz, $I_{rf} = 40$ μA.

ing constant, ω_p is the CDW plasma frequency, q_0 is the CDW wave vector, E is the applied field, and m^* is the mass of a condensed electron. This "rigid particle" model has given excellent quantitative accounts of Shapiro step interference phenomena in $NbSe_3$.[5] Equation (1) also predicts complete mode locking for the interference.[15] This simple model has, however, severe deficiencies in that it does not yield the proper I-V characteristics for $I > I_T$ (with no rf), and fails to predict the observed detailed form of the low-field frequency-dependent ac conductivity. More importantly, Eq. (1) cannot account for broadband noise generation by a moving CDW condensate.

On the other hand, Richard, Monceau, Papoular, and Renard[4] have suggested that in $NbSe_3$ Eq. (1) may correspond to the dynamics of a single domain within the macroscopic crystal, and that broadband noise arises from an interference between different domains. Stokes, Robbins, Bhattacharya, and Klemm[16] have used a related argument in describing broadband noise generation in TaS_3. In the model of Stokes *et al.*, fluctuations in the local threshold field E_T within a coherent volume or domain of size λ^3 cause local voltage fluctuations which add incoherently to produce broadband noise, whose magnitude is given by

$$\langle \delta V^2(\omega)\rangle = I^2(\partial R/\partial V_T)^2 E_T^2 \lambda^3 (l/A) S(\omega,T) \quad . \qquad (2)$$

Here $\langle \delta V^2(\omega)\rangle$ is the average noise power at frequency ω, I is the total current, R is the total resistance of the sample, V_T is the threshold voltage, V is the voltage across the sample, l is the length of the sample, A is its cross-sectional area, and $S(\omega,T)$ is a temperature-dependent spectral weight function. The predictions of Eq. (2) are in excellent agreement with the experimentally determined bias dependence of broadband noise in TaS_3.[16]

We here adopt a similar view that the broadband noise observed in $NbSe_3$ is due to internal CDW phase fluctuations arising from coupled CDW domains. The phase fluctuations lead to a distribution in CDW velocities, and hence to internal resistive fluctuations with a noise voltage similar to that of Eq. (2). The spectral response of the noise may be $1/f$, in analogy to $1/f$ noise observed in metals due to resistive fluctuations.[17] In the absence of an external rf field, the macroscopic CDW dynamics represent the CDW phase $\langle\theta\rangle$ spatially averaged over many coupled domains, but incoherent noise still exists due to the finite distribution in CDW phase velocity, arising from relatively weak domain coupling.

We suggest that the addition of an external rf current (or field) modifies the coupling between domains. If the external rf frequency closely approaches the characteristic internal frequency of the domains, ω_N, then mode locking may take place throughout the *entire sample.* The electromagnetic interaction between the applied rf field and the condensed electrons effectively "synchronizes" the domains, thereby causing an effective divergence of the domain coupling. This macroscopic and complete locking forces all incoherent internal fluctuations to be frozen out. With the entire CDW condensate assuming a unique phase velocity, which remains constant over the whole region of mode lock, the differential resistance remains flat, equal in magnitude to the Ohmic limit, as demonstrated in Fig. 1(b).

Our model suggests that, even in the absence of mode locking, an rf field of sufficient magnitude might still serve to enhance domain coupling, resulting in a uniform CDW phase velocity throughout the sample. We term this effect rf-induced phase homogenization.[6,18] Figure 3 shows the total broadband noise across a $NbSe_3$ sample (current biased past I_T), as a function of the magnitude of external rf current. ω_{rf} was chosen sufficiently far away from ω_N as to prevent any obvious mode locking. From Fig. 3 it is apparent that the total broadband noise amplitude in $NbSe_3$ can be smoothly driven to zero by an rf field of sufficient magnitude, confirming our expectations. The magnitude of the rf field required to completely suppress the noise is nearly an order of magnitude larger than that typically needed to achieve complete mode lock as in Fig. 1(b).

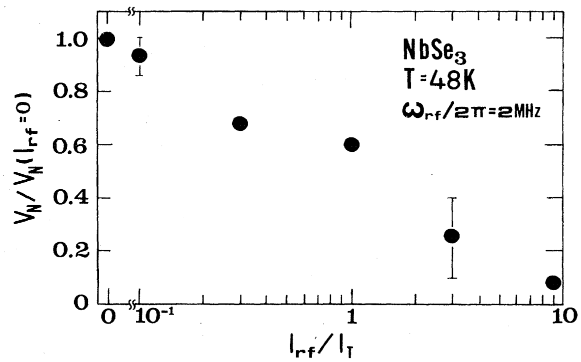

FIG. 3. Total broadband noise amplitude vs amplitude of an applied rf current in $NbSe_3$. Intense rf fields suppress the noise by homogenizing the CDW phase.

In the limit of strong coupling and a reduction in the effective number of internal degrees of freedom, one might expect Eq. (1) to asume substantial validity in describing the dynamics of the entire CDW condensate. Indeed, the best quantitative agreement between Eq. (1) and experimental results for $NbSe_3$ has been obtained in the limit of large rf driving fields.[5] We point out, however, that our observations show significant deviations from the behavior predicted by Eq. (1), even in the strong coupling limit. For example, the return map appropriate to Eq. (1), the circle map,[15] predicts complete mode locking for *all* subharmonic and harmonic interference, independent of the rf driving field amplitude or the order of the harmonic or subharmonic. From Fig. 1 it is, however, apparent that while low-order harmonics and subharmonics may lock completely in $NbSe_3$, higher-order interference peaks fail to display complete locking. The degree of mode lock for the high-order interference regions appears saturated; simply increasing the amplitude of the rf field does *not* result in a complete mode-locked state for these regions. Hence the details of the mode locking we have observed are more complex than the simple interaction of resonances described by the circle map. The circle map provides a first-order account of the interference and mode-locking phenomena.

In the regime of absent or very low amplitude rf driving fields, domain structure renders a simple single-coordinate description such as Eq. (1) entirely inadequate, and models[11,13,19] which explicitly include additional degrees of freedom of the CDW condensate must be employed. Some of these models have, in fact, explicitly demonstrated interference phenomena for combined dc and ac driving fields (for example, the hydrodynamical model of Sneddon, Cross, and Fisher[13] predicts Shapiro step interference), and it is most likely that a detailed quantitative analysis of our results (for example, the broadband noise behavior in the incomplete mode-locked regions) will only be possible with a many degrees of freedom model. Our results have nevertheless, demonstrated that rf-induced complete subharmonic and harmonic mode locking occurs in $NbSe_3$, along with an elimination of broadband noise generation, as predicted by a simple single-coordinate model. Our results also quite naturally explain the apparent contradiction in parameter values (such as effective inertia leading to subharmonic Shapiro steps) extracted from fits of Eq. (1) to experimental results of $NbSe_3$ in the low amplitude and high ampitude rf driving regimes.

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