Temporal Coherence in the Sliding Charge-Density-Wave Condensate

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The narrow-band noise (NBN) in the charge-density-wave conductor $NbSe_3$ is found to fluctuate dynamically in frequency, amplitude, and harmonic content. The NBN width is dominated by slow frequency fluctuations rather than a spatial velocity distribution. Mode locking quenches these fluctuations, yielding a dramatic narrowing of the NBN; but amplitude fluctuations remain. The NBN width also scales with the broad-band noise amplitude. These results demonstrate the crucial importance of temporal incoherence in the sliding charge-density-wave condensate and particularly on mode locking.

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An incommensurate charge-density-wave (CDW) condensate pinned by random impurities in quasi one-dimensional conductors such as $NbSe_3$ or TaS_3 possesses an internal washboard frequency in the sliding state given by $\omega_0 = \mathbf{q}\cdot\mathbf{v}$ where $\mathbf{q}$ and $\mathbf{v}$ are, respectively, the wave vector and velocity of the sliding CDW.[1] Since the discovery[2] of the narrow-band noise (NBN)—sharp peaks in the noise spectrum at ω_0 and its higher harmonics—considerable work has gone into the understanding of its origin, whether in the bulk of the sample[3] or at contacts.[4] Relatively little is known about the finite width of the NBN; it is generally viewed to reflect a distribution of velocities within the sample. The other kind of noise, generally called the broad-band noise (BBN), with an $\omega^{-\alpha}$-type spectrum, is also sparingly studied. A phenomenological pinning-force fluctuation[5] model has been used to describe the BBN, but its microscopic origin and any connections it may have with the NBN are still missing. Yet another recent controversy surrounds the observation of interference features, the so-called Shapiro steps, in the differential resistance under a combined ac-dc field ($V = V_{dc} + V_{ac}\cos\omega t$) whenever the condition $\omega_0/\omega = p/q$ (p and q integers) is satisfied.[6] Specifically, whether mode locking of the internal frequency ω_0 can be understood in terms of a single-particle picture[7] or an extended deformable-medium picture[8] of the CDW is a subject of considerable debate. Once again, the spatial coherence of the CDW, as reflected in the width of the NBN, was suggested[7,9] as the central parameter governing the nature of mode locking.

In order to gain insight into the mode-locking behavior, we have performed a direct frequency-domain study of the phenomenon of mode locking. It has been shown[10] that an amplitude-modulated drive produces frequency modulation (FM) of the NBN that can be evaluated quantitatively in the regime of small-amplitude, low-frequency ac drive (i.e., $V_{ac} \ll V_T$, the threshold voltage, and $\omega \ll \omega_0$). We follow this procedure for the mode-locking case where $V_{ac} \simeq V_T$ and $\omega \simeq \omega_0$.

Measurements were made in high-purity $NbSe_3$ samples (typical $E_T \lesssim 1$ mV/cm at 42 K). The samples have reasonably sharp NBN ($Q \simeq 100$ at $\omega_0 = 1$ MHz). Figure 1 shows the noise spectrum averaged over many scans, around 1 MHz, where the $p/q = \frac{1}{2}$ subharmonic locking occurs for the external drive frequency $\omega = 2$ MHz. I_{dc} is increased from Fig. 1(a) through 1(e). Two peaks correspond to the NBN at ω_0 and the frequency-modulated sideband at $\omega - \omega_0$. As I_{dc} is increased, the two peaks approach each other [as in Fig. 1(b)]; in Fig. 1(c) they merge producing a mode-locked NBN. For a range of I_{dc}, they remain locked. Subsequently, they unlock; a barely unlocked state where the sideband cannot be resolved from the NBN is shown in Fig. 1(d). Upon further increase of I_{dc}, they move apart in Fig. 1(e).

The fascinating result in Fig. 1(c) is the drastic reduction in the width of the mode-locked NBN; also, the background noise level returns to the ambient value [less easily discernible in Fig. 1(c)]. In Fig. 1(c) the width of NBN is resolution limited, and a high-resolution scan is shown in the inset. The width is less than a few hertz; even in this case, the intrinsic width could not be measured since we are limited by the width of the drive oscillator. The mode-locked NBN is thus immeasurably sharp ($Q > 10^5$ at $\omega_0 = 1$ MHz)—an enhancement of the sharpness by at least three orders of magnitude!

Figure 2 shows a direct demonstration of mode locking of NBN; the measured peak frequency of the time-averaged NBN spectrum is plotted versus I_{dc}. Plateaus show the ranges of I_{dc} over which the NBN frequency does not shift—corresponding to various mode-locked subharmonics forming the "staircase" structure. The harmonic step ($p/q = 1$) is included for completeness; this is determined by our monitoring the disappearance and the reappearance of the NBN from underneath the

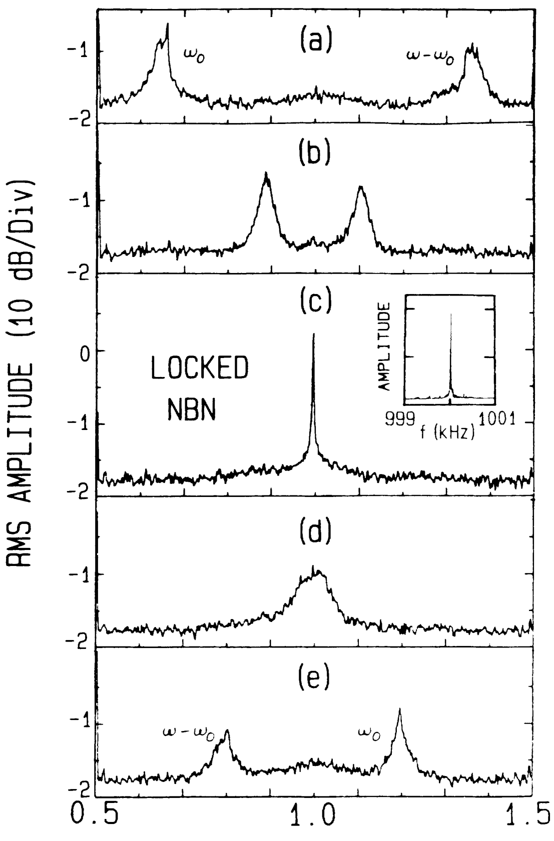

FIG. 1. Evolution of the noise spectrum as dc current is swept through the $p/q=\frac{1}{2}$ subharmonic locking with external ac drive at $\omega=2$ MHz. Current increases monotonically from (a) through (e). Inset in (c): High-resolution scan of the mode-locked NBN. See text for discussions.

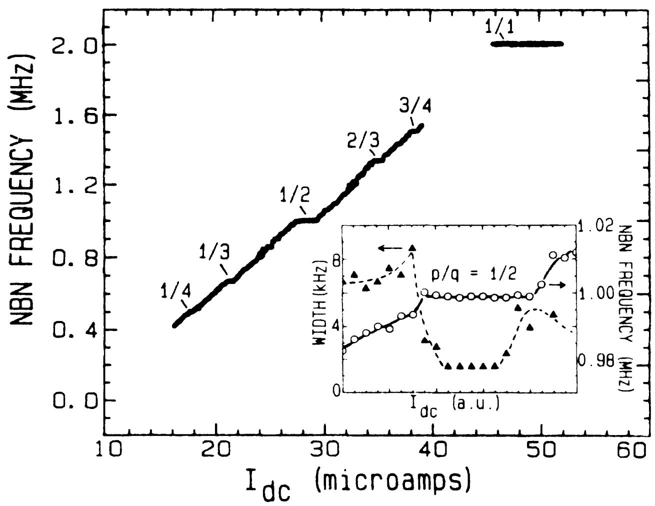

FIG. 2. Variation of the NBN peak frequency with I_{dc}. Plateaus correspond to mode-locked steps. Inset: Region spanning $p/q=\frac{1}{2}$ step; NBN peak frequency and the width (HWHM). The resolution-limited width corresponds to the truly locked region.

ac drive in the spectrum. The inset shows an isolated region around the $p/q=\frac{1}{2}$ subharmonic, where both the position and the width of the time-averaged NBN are plotted. Locking as given by the resolution-limited width (in a low-resolution measurement as in Fig. 1) occurs over a smaller range of I_{dc} than what is given by the NBN frequency itself. We find that the former correctly identifies situations such as in Fig. 1(d) as unlocked but the latter may not, and is thus a better measure of the step width.

Direct comparison with standard dV/dI measurements demonstrates that flat-top features correspond precisely with the resolution-limited NBN width. However, peaks in dV/dI do not. Therefore, as we stated elsewhere,[8,11] peaks should not be considered locked; nor should the area under the peak in a dV/dI vs I plot be considered a pseudowidth of an unlocked step, as is commonly done. It is possible that these situations represent a temporal intermittency in locking.

It has been argued elsewhere[11] that mode locking must imply "frequency pulling." Figures 1 and 2 represent the first direct evidence of this phenomenon. Figure 2 shows that the NBN peak frequency is pulled to the drive frequency in the plateaus. But more than that, the width of the NBN is pulled to the single frequency, i.e., there is no longer a distribution of frequencies in a mode-locked NBN. It is tempting to suggest that locking proceeds via a sudden pulling of the entire velocity distribution to the drive. This process may yield a velocity and phase homogenization over the entire sample, as has been suggested frequently.

These interpretations depend crucially on the assumption that the NBN width is a measure of a time-invariant velocity distribution. We have therefore investigated the NBN width in detail and find, surprisingly, this not to be the case. Unlike what is commonly believed,[12] the NBN width is dominated by slow temporal velocity fluctuations rather than a time-invariant velocity distribution.

We find that the NBN (without V_{ac}) has large temporal fluctuations in both frequency and amplitude. Amplitude fluctuations of NBN have been reported earlier.[9] But the fluctuations in frequency have not been reported nor studied in detail although they appear to be a common occurrence.[13] We find that the NBN width is dominated by the temporal fluctuations over time scales much larger than the washboard period and even comparable to the sampling time in a typical scan. In other words, the fluctuations are easily discernible from one scan to another. Morover, we find that the harmonic content, too, of the NBN fluctuates from one scan to the other![14]

Figure 3 shows the fluctuations in frequency and am-

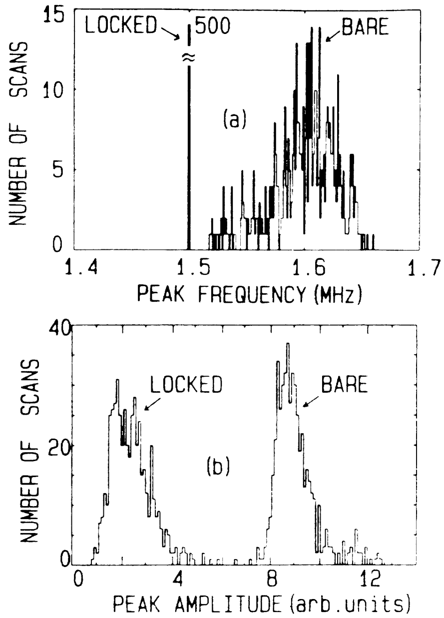

FIG. 3. Histogram of the temporal fluctuations in (a) frequency and (b) amplitude of a "bare" NBN ($V_{ac}=0$) and a locked NBN ($p/q=\frac{1}{2}$ at $\omega=3$ MHz). Results represent 500 scans of each case. In (b), the two histograms are shifted arbitrarily for clarity.

plitude. In each scan, the "bare" NBN fundamental (with no V_{ac}) shows a spread in frequency. The peak frequency and amplitude are measured. The histogram in Fig. 3(a) shows the distribution of peak frequency. For the locked case, the peak always occurs at 1.5 MHz, corresponding to the $p/q=\frac{1}{2}$ subharmonic, with no spread in frequency in individual scans. Remarkably, however, both cases show large fluctuations in amplitude with roughly equivalent Q (somewhat lower in the locked case) of the distributions as shown in Fig. 3(b). The width of the distribution is comparable to the most probable value. The unlocked case with finite V_{ac} represents an intermediate but very complex situation. Both frequency and amplitude fluctuations are present; but the histogram in frequency space shows sharp spikes at subharmonics that are covered in the frequency spread in each scan. The peak commonly occurs in individual scans at these commensurate frequencies which suggests that frequency fluctuations are not totally random. This raises possibilities of noise-induced intermittency as mentioned earlier. We are currently investigating these situations in greater detail.

We find that the broad-band noise (BBN) provides a clue to the resolution of these phenomena. At locking, the BBN amplitude decreases drastically together with the reduction of the NBN width. This is consistent with the fact that BBN amplitude is reduced, coincident with a flat-top feature in dV/dI, observed independently by us[15] and Sherwin and Zettl.[16] Even in the absence of V_{ac}, the field dependences of the NBN width and the BBN amplitude are the same. A detailed report of the tracking of the NBN width by the BBN amplitude with and without V_{ac} will be reported elsewhere.[17]

At mode locking, the NBN frequency is pulled to the drive and all temporal fluctuations of the frequency are quenched, yielding a drastic reduction of not only the NBN width but also the amplitude of the BNN. These results suggest that the NBN width may not be a measure of a stationary velocity distribution within the sample as is commonly believed but of the temporal fluctuations of the instantaneous velocity. We do not know what the intrinsic spatial distribution of velocity is, but it is considerably narrower than what is implied by the width.[18] In the event that at locking *only* the temporal velocity fluctuations are quenched, then the mode-locked NBN provides the intrinsic width which suggests that the CDW velocity is spatially uniform within the sample.

In a phenomenological model[5] of BBN where the total dc current is held fixed, since $I_{CDW}=I-V/R_N$, one trivially obtains that the noise is given by $\langle\delta V^2\rangle I=\langle\delta I_{CDW}^2\rangle R_N^2$, where R_N is the resistance of normal electrons. Thus, within this model, the temporal fluctuations in CDW velocity are proportional to the amplitude of the BBN.

Upon locking, the amplitude of the NBN is found to fluctuate in time. Since the velocity degree of freedom is quenched, these fluctuations may be attributed to a phase fluctuation of the CDW (with the assumption, of course, that amplitude fluctuations of the CDW order parameter are prohibitively expensive). This would imply that a mode-locked NBN is temporally phase coherent as is commonly believed. Moreover, if locking of the entire velocity distribution to the drive imposed phase coherence, a great enhancement of the NBN strength would be expected which is not seen experimentally. We do not know whether this behavior can be understood within a rigid model, since velocity coherence implies a time-invariant phase coherence for rigid objects. Phase fluctuations can occur in a deformable CDW even though the time-averaged velocity is spatially uniform, but we do not know what these fluctuations correspond to. We are currently investigating the temporal behavior of the rms amplitude of a mode-locked NBN.[19]

To conclude, we have demonstrated mode locking directly in the frequency domain. The results clearly show that temporal incoherence plays a very important role in the dynamics of the sliding CDW condensate. Barring a few attempts,[5,9,16,20] experimental studies of the sliding state have generally ignored this aspect. Theoretical studies have also not considered this feature adequately in contrast with the pinned-state behavior where metastability, hysteresis, i.e., generic glassy behavior has been studied extensively both theoretically and experimentally. We believe that the mode-locking behavior, in particular, depends crucially on the temporal coherence of the condensate. Any detailed quantitative analysis and determination of what model is appropriate must await a resolution of these issues. We also believe that earlier studies of the NBN relied heavily on certain notions of what the NBN measures which may be questionable. One needs to reconsider several of these issues; e.g., can a temperature gradient broaden the NBN, or does it merely split it even in a bulk model with the number of splittings increasing with increasing T gradient? We believe that these kinds of experiments performed on a mode-locked NBN will be particularly instructive.

We also note that the identification of the amplitude of the BBN with the width of the NBN provides the long-sought correlation between the two as has been noticed empirically not only between different samples but also between different CDW conductors.[21] The pinning-force fluctuation model[5] provides a phenomenological description of the behavior of both.[17] Moreover, temporal fluctuations of the harmonic content suggest that the effective pinning force fluctuates not only in magnitude but also in shape. Whether this can be understood without recourse to dynamic phase deformations of the sliding CDW is not clear. We emphasize that, even though these results pose a more unified question, an answer based on a microscopic origin of the temporal fluctuations (e.g., mobile strong-pinning centers, normal electrons producing screening of CDW distortions, etc.) is still missing and awaits further theoretical and experimental developments.

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[1]For a recent review, see *Charge Density Waves in Solids*, edited by Gy. Hutiray and J. Sólyom, Lecture Notes in Physics, Vol. 217 (Springer-Verlag, Berlin, 1985).

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

[3]G. Mozurkewich and G. Grüner, Phys. Rev. Lett. **51**, 2206 (1983).

[4]N. P. Ong, G. Verma, and K. Maki, Phys. Rev. Lett. **52**, 663 (1984).

[5]S. Bhattacharya, J. P. Stokes, M. O. Robbins, and R. A. Klemm, Phys. Rev. Lett. **54**, 2453 (1985).

[6]See, for example, S. E. Brown, G. Mozurkewich, and G. Grüner, Phys. Rev. Lett. **52**, 2277 (1984), and references therein.

[7]R. E. Thorne, J. R. Tucker, and J. Bardeen, Phys. Rev. Lett. **58**, 828 (1987), and references therein.

[8]S. Coppersmith and P. B. Littlewood, Phys. Rev. Lett. **57**, 1927 (1986).

[9]S. E. Brown, G. Mozurkewich, and G. Grüner, Ref. 1, p. 318. Recently a real-time wave-form analysis (G. Lee Link and G. Mozurkewich, to be published) has also shown that the NBN amplitude obeys the statistics of Gaussian random noise and not those of a coherent signal.

[10]J. P. Stokes, S. Bhattacharya, and A. N. Bloch, Phys. Rev. B **34**, 8944 (1986).

[11]S. Bhattacharya, J. P. Stokes, and M. J. Higgins, to be published.

[12]Measurements reported here refer to samples with a single well-defined NBN. Samples with multiple NBN frequencies (presumably showing spatially distinct velocity coherent regions) show separate locking. If they are closely spaced, the merging of two NBN peaks is seen as reported in Ref. 9. Details of such behavior will be reported elsewhere.

[13]R. M. Fleming, private communications.

[14]In these studies, the amplitude and frequency of the NBN fundamental are measured. For the unlocked NBN, since the harmonic content fluctuates in time, we do not yet know if the total strength (summed over all harmonics) fluctuates as well.

[15]M. J. Higgins, S. Bhattacharya, and J. P. Stokes, to be published.

[16]M. S. Sherwin and A. Zettl, Phys. Rev. B **32**, 5536 (1985).

[17]S. Bhattacharya, M. J. Higgins, and J. P. Stokes, to be published.

[18]This also implies that the NBN width cannot be used to infer the velocity correlation length directly. Since the BBN amplitude correlates with the NBN width, it is likely that the velocity correlation length can be obtained from it the same way as in the work of M. O. Robbins, J. P. Stokes, and S. Bhattacharya, Phys. Rev. Lett. **55**, 2822 (1985).

[19]In the experiments described here, V_{ac} is large enough so that the system spends time below threshold. We do not know if these results pertain to situations described in Ref. 7. Measurements of amplitude fluctuations of the mode-locked NBN in such samples (with anomalous dV/dI curves) would be particularly instructive.

[20]Robbins, Stokes, and Bhattacharya, Ref. 18.

[21]G. Grüner, private communication, and in Ref. 1, p. 263.