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Shapiro-step spectrum and phase-velocity coherence in $NbSe_3$ in a uniform temperature gradient

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We have measured the Shapiro-step spectrum of the charge-density-wave conductor $NbSe_3$, under conditions of combined ac and dc electric fields and an applied uniform temperature gradient. Increasing the temperature gradient from zero results in a well-defined breakup of the Shapiro-step spectrum. Under fixed temperature gradient, the isothermal Shapiro-step spectrum can be effectively recovered by application of a sufficiently-large-amplitude ac driving field. We identify different interference peaks of the broken spectrum with different macroscopically phase-velocity-coherent regions of the crystal, and we explicitly determine the volumes of these dynamically coherent domains.

There has been considerable recent experimental and theoretical interest in the unusual charge-density-wave (CDW) transport properties of the linear-chain compound $NbSe_3$ and related materials.[1] For applied dc electric fields as low as 5 mV/cm in $NbSe_3$, the incommensurate CDW may be depinned from the underlying crystal lattice and impurity structure, thus carrying with it an excess current I_{CDW}. In a "current-controlled" experiment, the total current I through the crystal is regulated, and for I exceeding a threshold current I_T, the CDW moves and, in so doing, generates coherent voltage oscillations (narrow-band noise) which appear across the ends of the sample.[2] Although the origin of the noise is highly controversial, it has been clearly demonstrated that the fundamental noise frequency f is directly proportional to I_{CDW} and that the ratio I_{CDW}/f is highly temperature dependent and reflects directly the CDW order parameter.[3-5]

In the presence of combined dc and ac electric driving fields (or currents), interference[3] occurs between intrinsic oscillations of the crystal and the applied ac field, leading to "Shapiro" steps[6] in the dc current-voltage (I-V) characteristics of the specimen. The Shapiro-step phenomenon in $NbSe_3$ has a rich harmonic[6] and subharmonic[7] structure, and both incomplete and complete mode locking have been observed.[8]

Recently it was demonstrated[7] that the Shapiro-step magnitude in $NbSe_3$ is in fact independent of the quality of the underlying narrow-band noise spectrum, suggesting that Shapiro-step interference reflects the true "internal" oscillations of the system at frequency ω_I, independent of potential boundary effects. Experimentally, under isothermal conditions, $\omega_I = f$.

An interesting experimental method for the study of noise phenomena in $NbSe_3$ has been the external application of temperature gradients, where the temperature dependences of the local order parameter, CDW depinning field, and low-field resistivity may be exploited. Original studies[9] of the noise spectrum in $NbSe_3$ in a temperature gradient demonstrated that (for relatively short samples) the quality factor and amplitude of the noise are not degraded by the gradient, but rather reflect the average temperature of the specimen. Subsequent experiments[10-12] by Ong and coworkers have indicated that application of a uniform temperature gradient can result in a splitting of the noise spectrum. Multiple splittings induced by temperature gradients have also been observed.[13] The above results on the noise have been interpreted in terms of a contact-induced vortex phase slip at the ends of the samples,[10] and in terms of a breakup of the sample into macroscopic "domains."[9,13]

We have applied a temperature-gradient technique to the study of Shapiro-step interference in $NbSe_3$. We find that under uniform temperature-gradient conditions sharp Shapiro-step interference still occurs, although there results a well-defined breakup of the isothermal Shapiro-step spectrum. Analysis of the Shapiro-step structure allows us to identify, for a given set of electric field and temperature-gradient parameters, the volume fraction of the sample synchronized to, and hence oscillating at, the frequency of the applied ac field. We determine critical values for the temperature gradient and ac amplitude for which the sample breaks from a single, macroscopically phase-velocity-coherent domain, to two distinct domains, each comprising approximately one-half the sample volume.

Our experimental setup employed single crystals of $NbSe_3$ suspended (in vacuum) between two large copper mounting posts anchored to Peltier heater chips. Electrical contact to the sample was made by conductive silver paint. Our two-probe mounting configuration allowed the temperature of the sample ends to be independently varied, and thermometry was accomplished through a diode sensor and several miniature differential thermocouples. Shapiro steps were induced by driving the sample with a dc current and a superposed radio frequency (rf) current. An additional low-frequency (280 Hz) low-amplitude current modulation provided a suitable signal for lock-in detection of the (dc) differential resistance dV/dI.

Figure 1 shows a series of dV/dI traces for a $NbSe_3$ sample of length $l = 0.5$ mm, with successively increasing temperature gradient. Here the temperature of the "cold" end of the sample, T_0, was fixed at 47 K, while the temperature of the "hot" end, T_1, was varied between 47 and a maximum of 52.2 K. We define $\Delta T = T_1 - T_0$. The top trace in Fig. 1 corresponds to isothermal conditions, and a rich Shapiro-step spectrum, consistent with previous studies,[7] is observed. We note that in this high-quality sample the fundamental ($n = 1$) interference peak (at 20 μA bias) displays complete mode locking, as do several subharmonic interference peaks. n is the step index, to be discussed below. With increasing ΔT, the Shapiro-step spectrum is observed to remain well-defined (i.e., no smearing), but a general breakup or splitting of both harmonic and subharmonic interference peaks occurs. The bottom trace of Fig. 1, for ex-

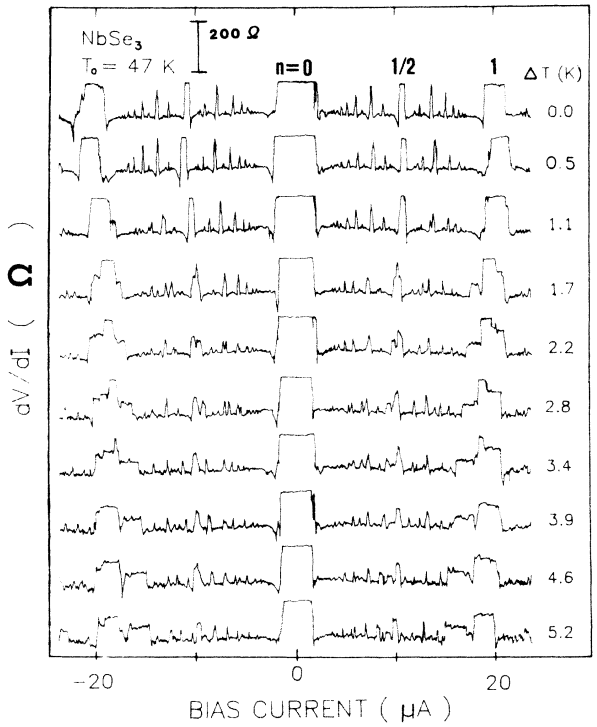

FIG. 1. Shapiro-step spectrum of $NbSe_3$ for various values of an applied uniform temperature gradient. ΔT is the total temperature difference across the sample of length 0.5 mm. T_0 is the (fixed) temperature of the cold end of the sample. $\omega/2\pi = 10$ MHz, $V_{rf} = 111$ mV. n is the step index (see text).

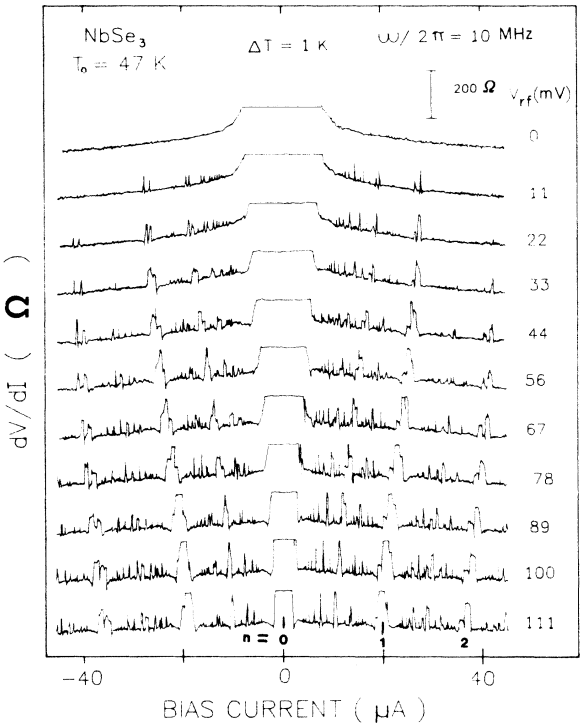

FIG. 2. Shapiro-step spectrum of $NbSe_3$ for various values of the rf electric field drive. The temperature gradient is fixed at 20 K/cm. The splitting of the Shapiro-step spectrum induced by ΔT may be reversed by a strong amplitude ac field. n is the step index (see text).

ample, is best described as a superposition of two distinct Shapiro-step spectra, although neither of the two "subspectra" displays complete mode locking (see below). By direct spectrum-analyzer detection of the narrow-band noise spectrum (no rf current), it was confirmed that the breakup of the Shapiro-step spectrum is associated with a corresponding splitting of the narrow-band noise spectrum. No minimum ΔT was observed for the onset of splitting of the noise spectrum.

In the presence of a temperature gradient, the breakup of the Shapiro-step spectrum can be reversed by increasing substantially the rf current amplitude. This is shown in Fig. 2, which displays several dV/dI traces of the same $NbSe_3$ crystal with a constant $\Delta T = 1$ K. V_{rf} is the rf voltage amplitude across the sample measured at zero dc bias. At low rf amplitude, a dual or split Shapiro-step spectrum is observed. With increasing rf field amplitude the Shapiro-step magnitude increases (see Ref. 6), and a smooth merging of the main interference peaks occurs. The merging is, however, a sensitive function of the order of the harmonic or subharmonic interference peak. For example, in the bottom trace of Fig. 2, the $n = 1$ interference peaks (at 20 μA bias) have almost fully coalesced, as have the $n = \frac{1}{2}$ (at 10 μA bias) subharmonic peaks, whereas higher-order subharmonic and harmonic peaks (e.g., $n = \frac{3}{2}$ at 25 μA bias); $n = 2$ at 35 μA bias) remain distinct. These higher-order peaks can, however, be forced to merge by further increasing the rf drive amplitude.

We now analyze these results. In Shapiro-step experiments, interference occurs whenever $\omega_J = n\omega_{ex}$, where the step index $n = p/q$ with p and q integers.[6,7] Two parameters are of particular importance in the discussion of Shapiro-step interference. The first is the actual height h of the interference peak recorded on a dV/dI plot, measured from the effective base line, or saturated dV/dL value. h determines what portion (volume fraction) of the CDW is locked to the external rf drive. If dV/dI on the interference steps attains the Ohmic differential resistance value R_0, then $h = h_{max}$ and locking is complete; the entire CDW condensate assumes a unique and fixed phase velocity, and only normal (uncondensed) electrons contribute to the differential resistance. It has been demonstrated that in the complete mode-locked state, all internal phase velocity fluctuations (sources of broad-band noise) are absent.[8] In incomplete locking, $h < h_{max}$, and only portions of the CDW condensate are synchronized or mode locked to the external rf drive.[14] Here slight increases in dc bias field results in an increase in I_{CDW}, due to the unlocked regions. The second parameter is the actual Shapiro-step magnitude δV which would be observed on a dc I-V plot. In a current-driven differential resistance experiment, δV is given by the integrated area of the interference peak, $\delta V = h\Delta I$, where ΔI is the current bias range over which locking persists.

We interpret the breakup of the Shapiro-step spectrum in Fig. 1 as reflecting a breakup of the sample into macroscopic domains each with a different intrinsic frequency for a particular value of dc bias current. The difference in intrinsic frequency arises mainly from the strong temperature dependence of the ratio I_{CDW}/ω_J, which decreases sharply with increasing temperature above 45 K.[4,5] As we discuss below,

the temperature gradient appears to break the sample into two distinct macroscopic domains, which we identify as the "hot" and "cold" domains. As dc bias current is swept through the sample with an applied temperature gradient, one expects first locking with the hot domain (with a higher intrinsic frequency for a given I_{CDW}), and then locking with the cold domain. The effect is clearly demonstrated in Fig. 1, where the gradient induces new interference structure at lower bias currents. The interference peaks identified with the cold domain also shift to slightly lower bias currents with increasing temperature gradient, due to an increase in average temperature of the cold domain.[15] We focus for the moment on the detailed structure of the dominant ($n=1$) interference peak associated with the $\Delta T=2.8$ K trace of Fig. 1. The external rf frequency is fixed at $\omega_{ex}/2\pi=10$ MHz. As the bias current approaches 16.9 μA, the intrinsic oscillation frequency of the hot domain, $\omega_{i,h}$, becomes sufficiently close to ω_{ex} such that mode locking takes place between this domain and the external rf drive. From $h/h_{max}=0.33$ in this mode-locked region, we find that the hot domain comprises 33% of the sample volume. As the dc bias current I is increased to 18.4 μA, $\omega_{i,h}$ remains locked to ω_{ex}. At this current bias, the intrinsic frequency of the cold domain, $\omega_{i,c}$, is sufficiently close to ω_{ex} that the cold domain also becomes locked to ω_{ex}. With both the hot and cold domains mode locked to ω_{ex}, $h=h_{max}$ and complete mode locking results. At a bias current $I=19.4$ μA, the hot domain becomes unlocked from ω_{ex} (it apparently no longer remains energetically favorable for ω_{ex} to "pull" $\omega_{i,h}$ so far from its unperturbed value), and between 19.4 and 20.6 μA, only the cold domain remains locked. In the later mode-locked region, h/h_{max} determines the volume of the cold domain; 67% of the sample volume. As an independent check, we note that the volumes of the hot and cold domains sum to 100% of the total sample volume.

The mode-locking process described above for $\Delta T=2.8$ K is essentially unaltered for different values of the temperature gradient, except that the locking range of the hot domain moves successively to lower (higher) bias currents with increasing (decreasing) ΔT, and the locking ranges of the hot and cold domains no longer overlap if ΔT exceeds 3.9 K. The respective volumes of the hot and cold domains, however, remain at 33% and 67% of the sample volume, independent of the magnitude of the temperature gradient (up to $dT/dl=10.4$ K/cm). The slight asymmetry may be due to enhanced domain-boundary formation at a grain boundary or other relatively weak defect within the crystal structure.

The Shapiro-step magnitude associated with oscillations in different domains is given from classical mixing theory as[5]

$$\delta V=\alpha V_T 2|J_n(V_{rf}\Omega/\omega_{ex}V_T)|\ , \tag{1}$$

where J_n is the Bessel function of order n, V_T is the threshold voltage for the onset of CDW conduction (no rf), Ω is a scaling parameter, and α describes both completeness and range of mode lock of the interference phenomenon. Figure 3(a) shows δV associated with the $n=1$ interference peaks of Fig. 1, as a function of ΔT. The step magnitudes have been decomposed into contributions from the cold domain, hot domain, and mixed domain (appropriate when the two domains are locked together). The breakup of the sample into two distinct domains is demonstrated clearly in Fig. 3(a). Also shown in this figure is the sum δV_{tot} of all

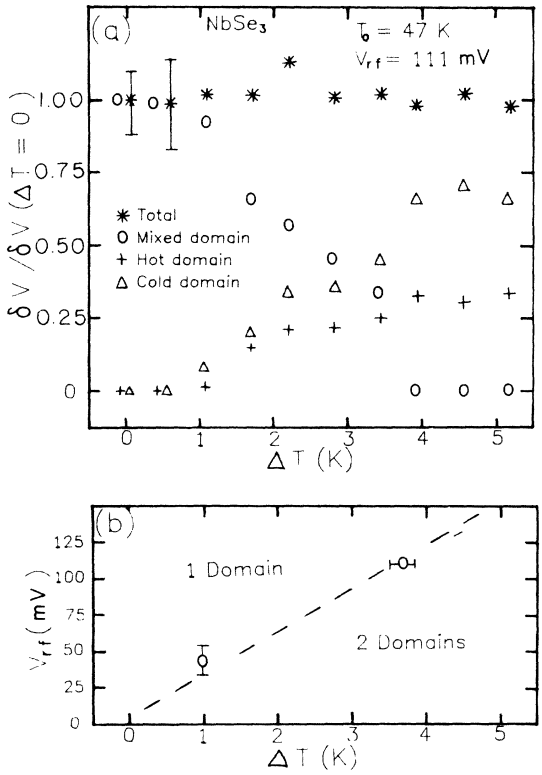

FIG. 3. (a) Shapiro-step magnitude δV associated with different macroscopic domains in the CDW crystal. (b) Phase portrait of critical temperature gradient and rf field amplitude which divide the sample from consisting of a single macroscopic domain to two independently coherent domains. The sample length is 0.5 mm; the total sample volume is 1.02×10^{-9} cm^3.

three contributions to δV. In the temperature range of interest, the parameters V_T and Ω are only weakly temperature dependent, and hence δV_{tot} reflects directly the parameter α. δV_{tot} is seen to remain approximately constant as ΔT is increased from zero to 5.2 K, and hence the data of Fig. 3(a) consistently account for the entire CDW condensate volume throughout the range of applied temperature gradient. We note that, at the boundary between the hot and cold domains, a CDW velocity discontinuity is implied, with a corresponding fluctuation in the CDW order parameter at the boundary. Within each domain, however, the CDW amplitude is dictated by the local temperature,[9] and hence CDW to normal-carrier conversion takes place everywhere along the length of the crystal, as well as at the boundary.

The "merging" effect shown in Fig. 2 appears to be a manifestation of rf-induced phase homogenization[7,8,16] within the sample volume. Increasing the rf amplitude greatly increases the coupling between domains, making a temperature gradient less effective in separating hot and cold domains in the Shapiro-step spectrum. The sensitivity of the dependence of the Shapiro-step splitting on the harmonic (or subharmonic) order n reflects the relatively weak coupling of the external rf to the CDW condensate at high p and q values, consistent with the limited range of mode lock (under isothermal conditions) for the higher-order interference.[7,17]

From Figs. 2 and 3(a) we may extract critical values of ΔT and the rf amplitude V_{rf} for which the sample breaks up from one coherent domain into two independently coherent domains. For fixed V_{rf}, we define ΔT_{crit} as the temperature difference above which no common locking (complete mode lock) can be achieved, while for fixed ΔT, $V_{rf,crit}$ is that value of rf amplitude above which a common locking of the hot and cold domains is obtained. The resulting phase diagram is displayed in Fig. 3(b). It is apparent that the dynamic coherence length ξ_D is a strong function of both ΔT and V_{rf}. For very short samples, we expect the phase plot of Fig. 3(b) to be modified; in particular, a finite x-axis (ΔT) intercept is expected when the intrinsic dynamic phase-coherence length greatly exceeds the sample length. This effect would be consistent with temperature gradient experiments on the behavior of the narrow-band noise spectrum for short samples.[9,12]

Finally, it is interesting to speculate on the relation of our work to the origin of narrow-band noise in $NbSe_3$ and related CDW conductors. In the vortex model of Ong, Verma, and Maki,[10] the noise originates from phase slip at the metallic contacts at the ends of the sample, while impurity models[4] suggest a modulation of the CDW current due to bulk impurities. Our Shapiro-step experiments clearly demonstrate that under uniform temperature gradient conditions macroscopic domains are formed with different intrinsic frequency. While this would seem to favor coherent domain models for the narrow-band oscillations,[13] we stress that while the intrinsic frequency ω_I is usually associated with the narrow-band noise frequency, ω_I may be deduced even if the specimen displays *no* narrow-band noise.[18,19] As we have mentioned earlier, it has been clearly demonstrated that the Shapiro-step magnitude is independent of details (such as absolute amplitude or quality factor) of the noise oscillations in $NbSe_3$. Thus, although in many cases the noise may reflect directly the intrinsic domain frequency, it should not be necessarily assumed that the two oscillation effects have common origins.

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[1]For a review, see G. Grüner and A. Zettl, Phys. Rep. **119**, 117 (1985); see also *Proceedings of the International Conference on Charge Density Waves in Solids, Budapest, Hungary, 1984,* edited by Gy. Hutiray and J. Solyom, Springer-Verlag Lecture Notes in Physics, Vol. 217 (Springer-Verlag, New York, 1985).

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

[3]P. Monceau, J. Richard, and M. Renard, Phys. Rev. Lett. **45**, 43 (1980).

[4]J. Bardeen, E. Ben-Jacob, A. Zettl, and G. Grüner, Phys. Rev. Lett. **49**, 493 (1982).

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

[6]A. Zettl and G. Grüner, Solid State Commun. **46**, 501 (1983).

[7]R. P. Hall and A. Zettl, Phys. Rev. B **30**, 2279 (1984).

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

[9]A. Zettl, M. Kaiser, and G. Grüner, Solid State Commun. **53**, 649 (1985).

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

[11]G. Verma and N. P. Ong, Phys. Rev. B **30**, 2928 (1984).

[12]X. J. Zhang and N. P. Ong, Phys. Rev. B **30**, 7343 (1984).

[13]S. E. Brown, A. Janossy, and G. Grüner, Phys. Rev. B **31**, 6869 (1985).

[14]The relationship between h/h_{max} during mode lock and the volume fraction of the corresponding velocity coherent domain is linear. Consider, for example, a simple one-dimensional CDW crystal with N independent domains in series; each domain is comprised of a normal electron resistance R_N in parallel with a CDW differential resistance R_{CDW}. $R_{CDW}=\infty$ for all locked domains, and $R_{CDW}=R_c$ for all unlocked (and conduction-saturated) domains. If j of the N domains are locked, $h=jR_N^2/(R_c+R_N)$. Hence $h/h_{max}=j/N$, the locked volume fraction.

[15]In this temperature range, the (relatively weak) temperature dependences of the depinning field and Ohmic resistance contribute in second order to the frequency shift. See Ref. 9.

[16]R. P. Hall, M. S. Sherwin, M. Hundley, and A. Zettl (unpublished).

[17]S. E. Brown, G. Mozurkewich, and G. Grüner, Phys. Rev. Lett. **54**, 2272 (1984).

[18]L. Sneddon, M. C. Cross, and D. S. Fisher, Phys. Rev. Lett. **49**, 292 (1982).

[19]R. M. Fleming, L. F. Schneemeyer, and R. J. Cava, Phys. Rev. B **31**, 1181 (1985).