Interference Phenomena and Mode Locking in the Model of Deformable Sliding Charge-Density Waves

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We study how a deformable charge-density wave in the presence of random impurities responds to driving fields with both ac and dc components. Features in the differential resistivity are found when an internal frequency proportional to the velocity is either a harmonic or subharmonic of the driving frequency. Even infinitely large systems can exhibit true mode locking to the driving frequency. The behavior cannot be described by an equation of motion for a single effective degree of freedom.

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We study the competition of two frequencies in a system with many interacting degrees of freedom and compare the results to experiments on the charge-density-wave (CDW) system $NbSe_3$. A CDW with wave vector $\mathbf{Q}$ sliding at velocity $\mathbf{v}$ possesses an internal "washboard" frequency $\omega_0 = \mathbf{Q}\cdot\mathbf{v}$.[1] When an electric field E with both dc and ac components $[E(t) = E_0 + E_1 \cos\omega_{ac} t]$ is applied to $NbSe_3$, strong interference features (peaks) appear in the differential resistance dV/dI when ω_0 is a harmonic or subharmonic of the driving frequency ω_{ac}. Mode locking of these frequencies occurs for small ω_{ac} ($\lesssim 10$ MHz at 42 K),[2] so that $\mathbf{Q}\cdot\mathbf{v} = (p/q)\omega_{ac}$ for a *range* of E_0, with p and q integers.

Here we show that the classical model of deformable CDW's, which has successfully accounted for many other experiments,[3] is consistent with observations of interference phenomena and mode locking. The theory predicts peaks in dV/dI (but *not* locking) at large ac driving amplitudes and frequencies, and true mode locking when ω_{ac} is small and $E_1 \sim E_0$ so that significant relaxation takes place while the field is below the threshold field E_T. We find that subharmonic mode locking arises from the presence of many pinned metastable states. We show explicitly that the subharmonic mode locking of the model overdamped deformable CDW to a nonsinusoidal driving cannot be described by use of a single "effective" degree of freedom.[2,4] These results apply to the general question of when macroscopic systems are accurately described by equations of motion with a few degrees of freedom.

We use the model of a deformable CDW interacting with random impurities introduced by Fukuyama, Lee, and Rice (FLR)[5] and extended to include dynamics by Sneddon, Cross, and Fisher.[6] The equation of motion for $u(\mathbf{r},t)$, the CDW distortion at position $\mathbf{r}$ at time t, is

$$\lambda\, du(\mathbf{r},t)/dt - K\nabla^2 u(\mathbf{r},t) = \rho_c E(t) + \rho(\mathbf{r})\, dU(\mathbf{r}+u(\mathbf{r},t))/dz. \tag{1}$$

Here K is the elastic constant of the CDW, coupled to the electric field $E(t)$ through the collective charge density ρ_c of the CDW, and to the random impurity potential $U(\mathbf{r})$ through the periodic CDW amplitude $\rho(\mathbf{r})$.

First we discuss the harmonic peaks in the differential resistance dV/dI when $\mathbf{Q}\cdot\mathbf{v} = n\omega_{ac}$ in the limit of large ac frequency and amplitude. Sneddon, Cross, and Fisher[6] calculated harmonic interference features in the FLR model for the case of the total field $E(t)$ *always* much greater than E_T, but in this regime the peaks are too small to be experimentally resolvable. We extend their theory to the case of large ac driving and compare the results to experiment. The impurity potential is effectively weak if $E \gg E_T$. If the ac amplitude E_1 and frequency ω_{ac} are large enough, then the total field is extremely large except for intervals of duration $\sim E_T/E_1\omega_{ac}$. The CDW lacks time to deform before the field once again becomes large, and so remains nearly undistorted. Expanding about the uniform state, we write

$$u(\mathbf{r},t) = v_0 t + v_{ac}\sin\omega_{ac}t/\omega_{ac} + \bar{u}(\mathbf{r},t),$$

and determine v_0 and v_{ac} self-consistently to second order in $Q\bar{u} \ll 1$.[7] In Fig. 1 our calculated dV/dI (dotted line) is fitted to data on $NbSe_3$ for two different ac amplitudes at 25 MHz (solid line)[8]; the dashed line shows a fit to the single-particle model of Grüner, Zawadowski, and Chaikin (GZC).[2,9] While the GZC model exhibits true locking (plateaus where $dV/dI = \rho_n$, the normal resistivity) as well as "wings" (regions where dV/dI is less than in the absence of pinning), the perturbation-theory calculation of the FLR model shows only peaks (increased dissipation) when $\mathbf{Q}\cdot\mathbf{v} \approx \omega_{ac}$. (Mode locking does occur for other experimental conditions, as discussed below.) The peak *locations* as well as the *shape* of the features agrees much better with the FLR result than that of GZC. The FLR fit uses the measured values of ρ_n and E_1/E_0 as well as one parameter, of order $E_T \rho_c Q/\lambda \omega_{ac}$,[10] which is fixed for the two curves.

The data of Fig. 1 were chosen to be within the regime of validity of the second-order pertubation theory, which applies for small $E_T/\omega_{ac}E_1$, when the subharmonic peaks (which are higher-order perturbative effects) are small and there is no mode locking. Higher-order perturbation theory yields subharmonic interference features[7,11] but not mode locking for the FLR model.[12] The deformable-CDW model yields peak widths and amplitudes that exhibit an oscillatory dependence on E_1 (not evident in Fig. 1), as obtained for single-degree-of-freedom models.[2] However, the detailed field and frequency dependence is different.[7]

Experimentally,[2] mode locking is enhanced either by a decrease in ω_{ac} *or* by choice of $E_0 \approx E_1$ so that the total field is below threshold long enough for significant relaxation toward a pinned state to occur. To investigate this nonperturbative regime theoretically, we have simulated numerically a simplified 1D version of Eq. (1).

Our equation of motion for the local phase $\phi_i = Qu_i$ at the impurity site i is

$$\dot{\phi}_i = (\phi_{i+1} - \phi_i)/l_i - (\phi_i - \phi_{i-1})/l_{i-1} + U \sin(\theta_i + \phi_i) + E(t)(l_i + l_{i-1})/2, \tag{2}$$

where $\theta_i = QR_i$, and $l_i = R_{i+1} - R_i$. The impurity positions R_i are randomly distributed. The approximations involved in obtaining Eq. (2) from Eq. (1) and the numerical methods of solution are standard.[13,14] Pinned states have a characteristic correlation length L_0,[5] and a sample of length $L >> L_0$ is expected to have $\approx \exp(aL/L_0)$ metastable pinned states, with $a \sim 1$. In our simulations, typically $U = 4$, $L_0 \sim 1$, and we expect of order 10^{10} distinct metastable states in a system with fifty impurities.[14]

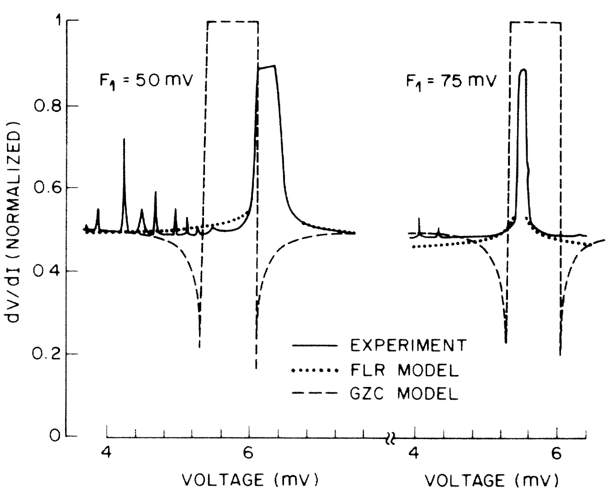

FIG. 1. Differential resistance dV/dI vs dc voltage V_0 near the first harmonic feature $\mathbf{Q} \cdot \mathbf{v} \sim \omega_{ac}$ for $NbSe_3$ with ac frequency $\omega_{ac} = 25$ MHz and ac voltage amplitudes $V_1 \sim 50$ and 75 mV (Ref. 8), for a sample with $V_T \sim 2$ mV. Also shown are theoretical fits using the FLR deformable-CDW model (dotted line) and the GZC one-degree-of-freedom result (dashed line). For FLR, both curves are fitted by use of a single parameter of order unity, as discussed in the text. The tops of the peaks are not calculated because the perturbation theory breaks down when the change in dV/dI is large.

Rather than apply sinusoidal driving, we apply a train of field pulses (from zero to above threshold) to the system.[15] This alteration greatly simplifies the numerical analysis and highlights the importance of relaxations below threshold. We vary the pulse height E_{on} and duration t_{on}, but allow a long time t_{off} between pulses so that the CDW can approach a stationary configuration before the next pulse. The pulses drive the CDW into a sequence of metastable states; the nth pulse maps the metastable state with configuration $\{\phi_i\}_n$ onto the configuration $\{\phi_i\}_{n+1}$. Since the equations of motion are deterministic, if a metastable configuration repeats once, the sequence repeats indefinitely. The time-averaged current is $\bar{I} = \langle \Delta\phi \rangle / (t_{on} + t_{off})$, where $\langle \Delta\phi \rangle = \lim_{n \to \infty} (\langle \phi \rangle_n - \langle \phi \rangle_1)/n$, with $\langle \phi \rangle_n = N^{-1} \sum_{i=1}^{N} \phi_i^{(n)}$, the spatially averaged phase before the nth pulse.

For large t_{off}, the GZC model with *any* potential with one minimum per period (even with inertia) exhibits only harmonic locking ($\langle \Delta\phi \rangle / 2\pi$ is always an integer). The FLR model does not suffer from this limitation. In fact, it has so many metastable states that one might think that enormously many pulses are needed to have a configuration repeat. Remarkably, following a short transient, the CDW *always locks into a finite sequence of states.* The configurations repeat periodically; after q pulses the CDW has moved everywhere by *precisely* p wavelengths, with p and q integers. An example of this behavior is shown in Fig. 2, where plots of the metastable phase configurations $\phi(j)/2\pi$ versus position j for successive pulses are shown. The curves are offset vertically to allow several pulses to be plotted on the same graph. After a short transient, every third pulse yields the same metastable configuration. The current traces (similarly offset) exhibit the same period-3 behavior. In order to repeat the same states, the CDW must move an integral number of wavelengths every three pulses, and direct calculation

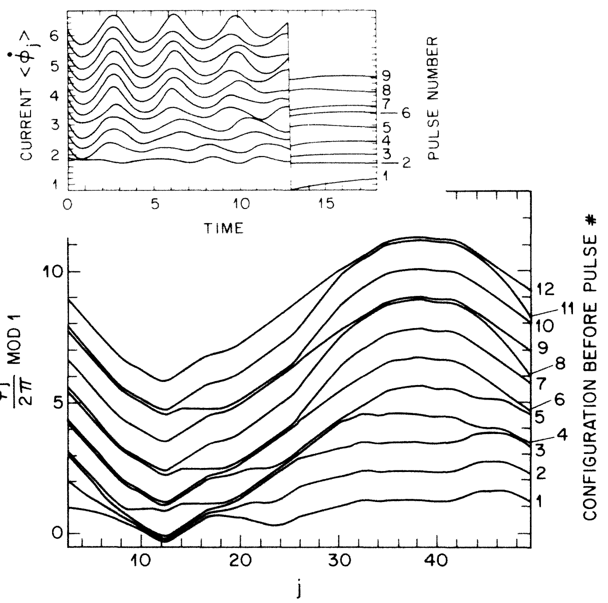

FIG. 2. Plots of metastable phase configurations $\phi(j)/2\pi$ vs position index j for a system with fifty degrees of freedom, with configurations before successive pulses offset vertically. After a short transient, the configurations repeat every third pulse. Inset: Current traces for successive pulses (again offset vertically), which display the same period-3 behavior after a short transient.

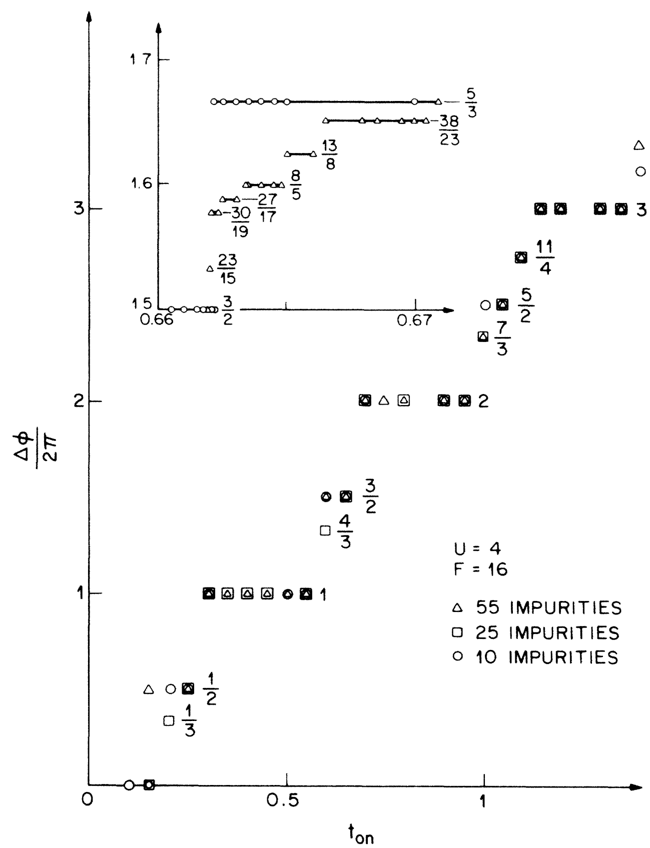

FIG. 3. Number of wavelengths moved per pulse $\langle\Delta\phi\rangle/2\pi$ vs the pulse duration t_{on} with a fixed $E_{on}=16$ for two systems with $U=4$, one with 55 degrees of freedom and the other with 10. Locking is demonstrated because $\langle\Delta\phi\rangle/2\pi$ is always a rational fraction. Inset: Magnified portion of the plot, demonstrating that increasing the number of degrees of freedom causes the appearance of high-order subharmonics.

indeed yields $\langle\Delta\phi\rangle/2\pi=\frac{11}{3}$.

When either the pulse amplitude E_{on} or pulse length t_{on} increases, the CDW moves farther per pulse. Figure 3 shows $\langle\Delta\phi\rangle/2\pi$ as a function of the pulse duration t_{on} for fixed pulse height E_{on}.[16] The CDW is always locked; both harmonics ($\langle\Delta\phi\rangle=2p\pi$) and subharmonics ($\langle\Delta\phi\rangle=2p\pi/q$) are observed. Subharmonics appear as a consequence of the many degrees of freedom; the inset of Fig. 3 compares a portion of the plots for systems with 10 and 55 impurities. High-order subharmonics are only observed for the larger system.[17] The natural description of the locking process is a cycle among a finite sequence of pinned states. Locking of $\Delta\phi$ occurs because a state can repeat only after a displacement of an integral number of wavelengths.

We now examine if the locking can be described with an equation of motion for a single degree of freedom, the most natural being the spatially averaged phase $\langle\phi\rangle$.[18] For an effective equation of motion for $\langle\phi\rangle$ to exist, knowing *only* $\langle\phi\rangle_n$ for the nth pulse in the stable periodic sequence must be sufficient information to determine $\langle\phi\rangle_{n+1}$ for the $(n+1)$th pulse. We have found a stable nineteen-cycle with $E_{on}=8$, $t_{on}=1.5347$, and $t_{off}=10$. In this sequence there are two similar states with values of $\langle\phi\rangle/2\pi$ differing by less than 0.0035. The metastable configurations for the pulses *following* these two are substantially different, having $\langle\phi\rangle/2\pi$'s differing by >0.055. Any function of $\langle\phi\rangle$ alone that describes our data must vary extremely rapidly and have regions with large *negative* slope, and so the utility of a one-degree-of-freedom description is doubtful.

The mode locking in our simulations does not merely reflect finite-size limitations. We have studied mode locking in systems ranging from 10 to 55 degrees of freedom and found no tendency for the lockings to disappear as the system size increases (see Fig. 3). The number of transient pulses remains small (<10) for the low-order subharmonics even in the largest systems. Also, one can show rigorously that true harmonic and mode locking occurs even in infinitely large systems if $E_{on}\gg U\gg 1$.[19] This special case is useful to show that true mode locking is possible even in an infinite system, although real CDW's are described by the weak-pinning limit $U\ll 1$.

In summary, we have shown that the model of extended deformable charge-density waves is consistent with experimental observations of harmonic and subharmonic interference effects in $NbSe_3$. We systematically predict harmonic peak shapes and the relative sizes of the subharmonics in dV/dI curves for large frequency and amplitude sinusoidal driving, and we demonstrate mode locking for pulsed driving when the CDW relaxes substantially between pulses. Mode locking is expected to be enhanced when there is significant relaxation while the total field is below threshold, a point stressed by Brown, Grüner, and Mihaly[15] from their study of pulsed forcing. Very small samples with few metastable states are expected to lack high-order subharmonic structure. A simple description of the model's mode-locking behavior by an equation with one degree of freedom does not appear to be possible.

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[1]See, e.g., *Charge Density Waves in Solids,* edited by G. Hutiray and J. Solyom, Lecture Notes in Physics Vol. 217 (Springer, Berlin, 1985).

[2]G. Grüner, A. Zawadowski, and P. M. Chaikin, Phys. Rev. Lett. **46**, 511 (1981); J. Richard, P. Monceau, and M. Renard, Phys. Rev. B **25**, 948 (1982); A. Zettle and G. Grüner, Phys. Rev. B **29**, 755 (1984); S. E. Brown, G. Mozurkewich, and G. Grüner, Phys. Rev. Lett. **52**, 2277 (1984); R. P. Hall and A. Zettl, Phys. Rev. B **30**, 2279 (1984); M. F. Hundley and A. Zettl, Phys. Rev. B **33**, 2883 (1986).

[3]Recent theoretical comparisons of the deformable-CDW model to experiment include P. B. Littlewood, in Ref. 1, p. 369, and Phys. Rev. B **33**, 6694 (1986); D. S. Fisher, Phys. Rev. Lett. **50**, 1486 (1983), and Phys. Rev. B **31**, 1396 (1985); L. Sneddon, Phys. Rev. Lett. **56**, 1194 (1986), and references therein.

[4]R. E. Thorne *et al.*, Phys. Rev. B **33**, 7342 (1986).

[5]H. Fukuyama and P. A. Lee, Phys. Rev. B **17**, 535 (1977); P. A. Lee and T. M. Rice, Phys. Rev. B **19**, 3970 (1979).

[6]L. Sneddon, M. C. Cross, and D. S. Fisher, Phys. Rev. Lett. **49**, 292 (1982); L. Sneddon, Phys. Rev. B **29**, 719, 725 (1984), and Phys. Rev. Lett. **52**, 65 (1984).

[7]P. B. Littlewood and S. N. Coppersmith, to be published. For $E_1 > E_0$, one needs $Q|\bar{u}| \approx \rho_c E_T^2/\lambda\omega E_1 << 1$. When the perturbation theory is valid, $1-\lambda v_{ac}/E_1$ and $\lambda\, dv_0/dE_0$ are both small. Increasing E_1 makes the impurity potential effectively weaker and suppresses the apparent threshold field, as experimentally observed.

[8]S. Brown and G. Grüner, unpublished. Large sample-to-sample variations occur (possibly arising from current inhomogeneities), but trends for a given sample are clear. We have allowed for a parallel free-carrier conductivity σ_n (which is measured experimentally) for the theoretical curves.

[9]The GZC curves are calculated following J. R. Waldram and P. H. Wu, J. Low Temp. Phys. **47**, 363 (1982); they are rather insensitive to the choice of parameters. A nonsinusoidal potential in the GZC equation induces subharmonic lockings but does not alter the dV/dI line shape significantly.

[10]This parameter is the ratio of the threshold field E_T (when $E_1=0$) to the dc field when $\mathbf{Q}\cdot\mathbf{v}=\omega_{ac}$ (for $E_1\rightarrow\infty$). Its order of magnitude is known but appears as a fitting parameter because E_T is not accurately determined by perturbation theory. Although some experiments are performed in a current-driven configuration, the normal conductivity is high enough that the CDW motion can be regarded as voltage driven.

[11]S. N. Coppersmith and P. B. Littlewood, Phys. Rev. B **31**, 4049 (1985).

[12]However, singularities in high-order perturbation theory may be a clue that mode locking occurs. P. F. Tua and J. Ruvalds, Solid State Commun. **54**, 471 (1985), have obtained harmonic and subharmonic lockings by use of a different model, but their step widths vanish in the infinite-volume limit.

[13]L. Pietronero and S. Strässler, Phys. Rev. B **28**, 5863 (1983).

[14]Littlewood, Ref. 3.

[15]Experiments using pulsed driving are reported by S. E. Brown, G. Grüner, and L. Mihaly, Solid State Commun. **57**, 165 (1986), and unpublished.

[16]To save computer time, Fig. 3 was constructed with a modified version of Eq. (2), where the l_i's are all 1 but the θ_i's are random. We find no essential differences between the mode locking for this case and for Eq. (2).

[17]If t_{off} is long enough so that almost complete relaxation to a metastable state occurs, the particular harmonic or subharmonic seen depends only on the pulse duration t_{on} rather than the overall period $t_+ t_{off}$. Changing the initial configuration can lead to different configurations in the locked cycle. Decreasing $t_{off} << 1/U$ suppresses the locking (Ref. 15) as expected.

[18]The experiments of Brown, Mozurkewich, and Grüner (Ref. 2) have been interpreted in this way; see, e.g., P. Bak, in Ref 1; M. H. Jensen, P. Bak, and T. Bohr, Phys. Rev. A **30**, 1960, 1970 (1984).

[19]S. N. Coppersmith, unpublished.