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Mixing in charge-density-wave conductors

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Harmonic and direct ac mixing properties of the Fukuyama-Lee-Rice model and the incommensurate chain are determined and compared with experimental data. Field and frequency dependences of both the amplitudes and phases of both of these responses are examined. Agreement with experiment is generally good. For example, classical models can yield low harmonic-mixing quadrature components simultaneously with substantial frequency dependence in both components of the linear response.

An interesting series of experiments, involving the nonlinear mixing of ac signals in charge-density-wave (CDW) conductors, was initiated by Seeger, Mayer, and Philipp[1] and thoroughly extended by Miller and coworkers.[2-5] These experiments merit theoretical study in their own right as probes of the unique properties of CDW conductors.

Further, the suggestion has been made that the results "may prove difficult to reconcile with *any* classical theory."[4] It is thus important to determine the ac mixing properties of classical models of sliding CDW's to see whether there is indeed a failure of the classical picture of bulk CDW motion, and whether these experiments give us evidence for Bardeen's fascinating proposal[6] that CDW conductors are exhibiting macroscopic quantum tunneling.

The principal experiments[2-5] are direct mixing (or rectification) and harmonic mixing. The sample is driven by a voltage of the form

$$E(t)=E_0+E_1\cos(\omega_1 t+\phi)+E_2\cos(\omega_2 t) \ , \qquad (1)$$

where E_1 and E_2 are generally small compared to the threshold voltage E_T. The two measurements consist of detecting the component of the current with frequency ω_0, where $\omega_0=\omega_1-\omega_2$ for direct mixing, and $\omega_0=2\omega_1-\omega_2$ for harmonic mixing. The frequencies are chosen so that ω_0 is much smaller than ω_1 and ω_2.

The properties of two different models are reported here: the Fukuyama-Lee-Rice (FLR) model[7] of random pinning and the incommensurate chain.[8] Perturbation[9] theory was used to obtain analytic information for the FLR model at large bias fields E_0. The incommensurate chain was simulated numerically to obtain solutions in the strong-coupling region closer to the sliding threshold field E_T. The equation of motion for the incommensurate chain with infinite-range interactions[10,11] is

$$\frac{du_j}{dt}=\langle u_j\rangle-u_j+P\sin(Hj+u_j)+E(t) \ , \qquad (2)$$

where u_j is the displacement of the jth particle, $\langle u_j\rangle$ is the center-of-mass displacement, P is the pinning strength, and $H/(2\pi)$ is chosen to be $(\sqrt{5}-1)/2$.

By simulating Eq. (2) in systems up to 377 particles in size we were able to restrict finite-size effects to the region very close to threshold and ensure that the results obtained at all other fields reliably reflected the thermodynamic limit. The applied frequencies were chosen to have a rational ratio so that the response (in the thermodynamic limit) is periodic. Transients were allowed to decay for a time $t_0=T$ sufficiently long that $t_0=T$ and $t_0=2T$ give results that are indistinguishable within numerical error. The sampling rate of the time series was chosen high enough that increasing it further produced no noticeable changes.

We now compare the theoretical results with those of experiments, examining each feature of the data in turn.

I. HARMONIC-MIXING PHASE SHIFT

The experimental result which has been most emphasized[2,4,5] is the failure of some experiments to observe an "internal" phase shift in the small difference-frequency harmonic-mixing response. Classical models were conjectured[2] to yield nonzero phase shifts at high applied frequencies ω_1. Wonneberger[12] then showed that the classical single-particle model exhibits a zero phase shift for all ω_1 at large dc bias fields. He proved this result to leading order in perturbation theory and argued in an appendix that the result is valid everywhere above threshold. Nonetheless, it was claimed that the nonobservation of a phase shift above threshold, where *in addition* both components of the *linear* ac response have substantial frequency dependence, may be difficult to reconcile with any classical theory.[4] It was further claimed that this experimental observation provided particularly significant evidence for CDW tunneling.[5]

We determined the harmonic-mixing properties of the FLR model using perturbation theory at large dc bias fields. The principal result is[13] that the response component at frequency ω_0 is proportional to $\cos(\omega_0 t+2\phi)$ in the limit of small ω_0 for large dc bias fields E_0 and all ω_1. The sine component vanishes linearly with ω_0. Thus the classical FLR model also exhibits a zero internal phase shift at large bias fields.

In that region, however, the linear ac response has no strong frequency dependence. It is therefore necessary to probe the region of lower dc bias closer to threshold. For this purpose, we turn to the results for the incommensurate chain.

Figure 1 shows some typical field and ω_0 dependences of both components of the harmonic-mixing signal. It is seen that the out-of-phase component is considerably smaller than the in-phase component, but appears to be approaching a nonzero limit as ω_0 approaches zero. As ω_0 decreases through 0.05, 0.025, and 0.0125 the out-of-phase component shows practically no change on the scale of the plot.

This limit is generally small. For the data shown in Fig. 1, ω_0/ω_1 is about 0.01, it corresponds to a phase shift of about 10°, which is the reported resolution of the experiments.[4,5] Experimentally ω_0/ω_1 was chosen to be about 0.001. We found that this phase shift decreases as E_1 and E_2 are increased with the other parameter fixed. The value of ω_1 in Fig. 1 was chosen approximately to *maximize* the phase shift.

Earlier work[11] has established substantial frequency dependence in both components of the linear ac response in this region. One may thus conclude that classical theories can indeed account for the absence of strong quadrature in the harmonic mixing and simultaneous presence of strong frequency dependence in the linear response.[14]

The above classical results do appear to show a nonzero quadrature, however, and if a classical picture were appropriate, one would expect to see a phase shift under some experimental conditions. A harmonic-mixing phase shift has, in fact, been reported.[15] The ratios E_1/E_T and E_2/E_T were smaller than in the experiments[2,3] that saw no phase shift. This is consistent with our observation above that the phase shift decreases as E_1 and E_2 increase.

Turning to the region below threshold, the calculated out-of-phase component is found to be much larger than the in-phase one for small ω_0. This is consistent with experimental reports[4] of an internal phase shift of about $\pi/2$.

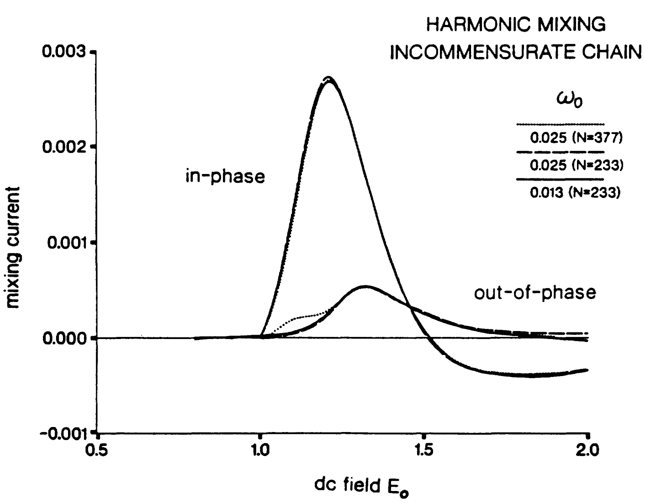

FIG. 1. dc field (E_0) and difference-frequency (ω_0) dependencies of both components of the harmonic-mixing response current ($N^{-1}\sum_j du_j/dt$) of the incommensurate chain ($\omega_1 = 1.0$, $P = 3.0$, and $E_1 = E_2 = 0.2$). All quantities are dimensionless [see Eq. (2)]. The curves for $N = 377$ indicate the size of truncation error. Both components have finite $\omega_0 \rightarrow 0$ limits, with the out-phase response considerably smaller than the in-phase response.

II. HARMONIC-MIXING MAGNITUDE

Since the harmonic-mixing phase shift is generally small, we focus now on the magnitude. Figure 2(a) shows the field (E_0) and frequency (ω_1) dependencies of the magnitude of the harmonic-mixing response, as calculated

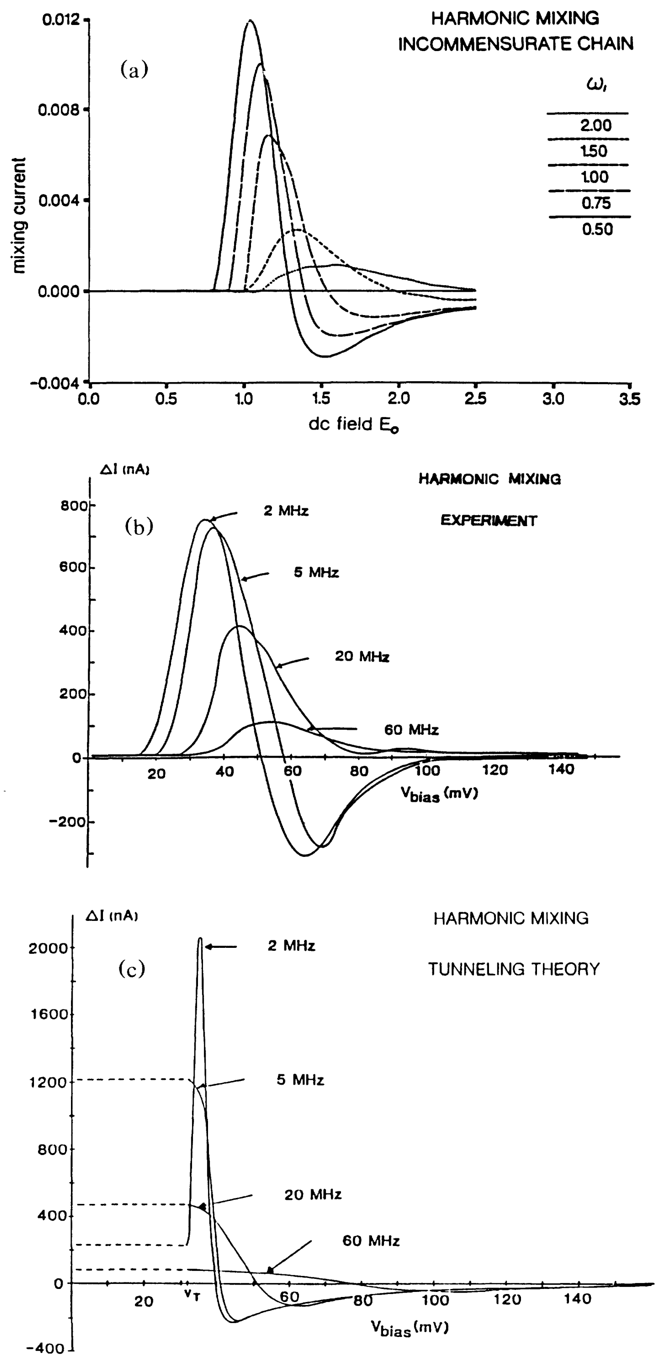

FIG. 2. dc field (E_0) and frequency (ω_1) dependencies of the magnitude of the harmonic-mixing response current ($N^{-1}\sum_j \times du_j/dt$): (a) the incommensurate chain, $N = 233$, $P = 3.0$, and $E_1 = E_2 = 0.30$; (b) experiment (Ref. 2); (c) results of a tunneling analysis (Ref. 2).

for the incommensurate chain. Figure 2(b) shows the experimental data,[2,4] and Fig. 2(c) shows the results[2,4] which have been presented based on quantum tunneling arguments.

(a) The peaks. In Fig. 2(a), the positions (fields) and the heights of the peaks change in the same sense with frequency as seen experimentally. The present results for the incommensurate chain change somewhat more rapidly with frequency than do the experimental data.

(b) The threshold. The incommensurate chain is seen in Figs. 2(a) and 2(b) to give the experimentally observed qualitative forms in the vicinity of threshold. The threshold-lowering effect of an applied ac field is also apparent in the incommensurate chain results, the effect increasing as the ac frequency decreases, as observed experimentally.[16]

A comparison with Fig. 2(c) then shows that these threshold features reveal substantial differences between the properties of the classical chain and the results of the quantum tunneling analysis.

(c) The negative dips. The experimental data [see Fig. 2(b)] show that for lower frequencies ω_1 the harmonic-mixing response goes negative above a (frequency-dependent) field (the phase switches rapidly from a value at or near zero to one at or near 180°). Thereafter the response approaches zero from below. Above a certain frequency, however, this no longer happens and the harmonic-mixing response is always positive.

This points up another qualitative difference between the two sets of theoretical results: While the incommensurate chain, like the experimental results, shows an ω_1 above which the response no longer changes sign, the quantum tunneling analysis appears to show a sign change for all ω_1.

(d) Below threshold. The calculated amplitude of the harmonic-mixing signal is about two or three orders of magnitude less than the peak above threshold, which is consistent with the experimental data [see Fig. 2(b)].

III. DIRECT-MIXING PHASE

Experimentally,[2-4] the response at $\omega_0 = \omega_1 - \omega_2$ to the cosine input signal of Eq. (1) is found to be proportional to $\cos(\omega_0 t + \phi)$ for small ω_0. A leading-order perturbative solution of the FLR model[13] showed this same feature. This conclusion can be seen to be true to all orders as follows. Consider the case $\phi = 0$. Equation (1) is then symmetric with respect to interchanging ω_1 and ω_2. The total response current at frequency plus or minus ω_0, J_{DM}, then also has this symmetry. Writing $j_{\text{DM}} = I_s \sin(\omega_0 t) + I_c \cos(\omega_0 t)$ then requires $I_s(\omega_1, \omega_2) = -I_s(\omega_2, \omega_1)$, so that $I_s \to 0$ as $\omega_1 \to \omega_2$. Even if the input fields are changed to sines by shifting the origin of time, the low-frequency direct-mixing response is still cosine.[17]

IV. DIRECT-MIXING MAGNITUDE

Figure 3(a) shows field and frequency dependence of the magnitude of the direct-mixing response for the incommensurate chain. Figure 3(b) shows the corresponding experimental data. The principal features of the experimental data are seen to be reasonably well reproduced by the incommensurate chain.

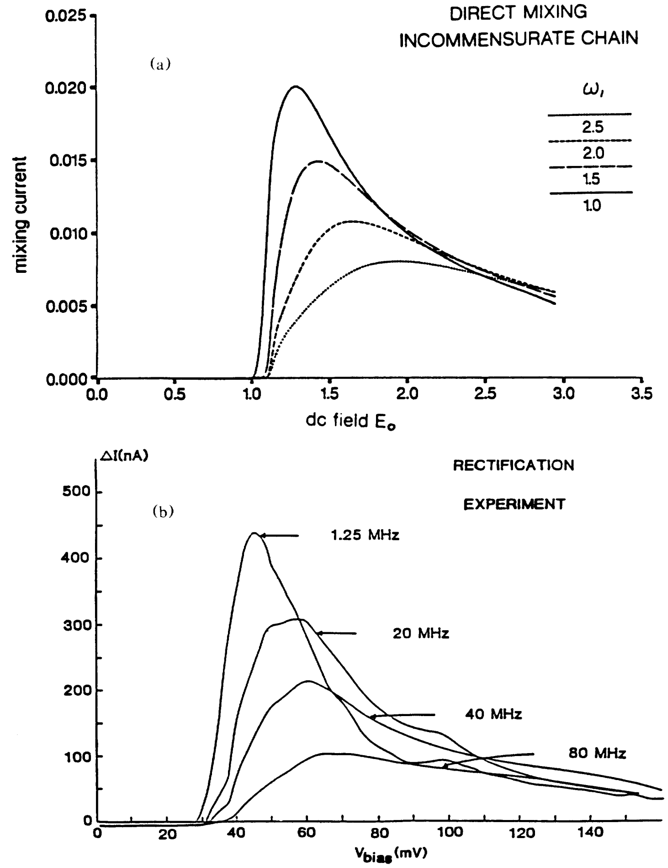

FIG. 3. dc field (E_0) and frequency (ω_1) dependencies of the direct-mixing current ($N^{-1}\sum_j du_j/dt$) magnitude: (a) the incommensurate chain, $N = 233$, $P = 3.0$, and $E_1 = E_2 = 0.30$; (b) experiment (Ref. 2).

V. CONCLUSION

The classical incommensurate chain has been seen to provide a fairly complete account of the field and frequency dependencies of both amplitude and phase components of the direct and harmonic-mixing responses of sliding CDW's.

For example, the challenge to classical theories posed[4] by some nonobservations of the harmonic-mixing phase shift has been resolved by the observations that (a) phase shifts have been seen experimentally, and (b) classical theories can account for phase shifts as small as the experimental uncertainties. (It may be interesting now to have both experimental and theoretical phase shifts determined more precisely to provide more telling comparisons of theories and experiment.)

The pinning of CDW's is believed to be due to random impurities,[18] and not a periodic incommensurate potential. To end on a question, then, we note that the issue of why incommensurate systems reproduce such a variety of observed CDW properties is still not properly understood.

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