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Transient charge-density-wave dynamics in $NbSe_3$

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The voltage response to rectangular current pulses has been investigated in the charge-density-wave (CDW) state of $NbSe_3$. For a variety of dc bias current levels and current pulse amplitudes, transient "ringing" is observed in the real-time response, which we associate with CDW domain dephasing. Using the ringing phenomenon as a probe, we determine explicitly the time scale for fast relaxation of internal deformations of the pinned CDW condensate.

Over the past decade, there has been considerable interest in the unusual charge-density-wave (CDW) transport properties of $NbSe_3$ and related conductors.[1] A variety of recent experiments[1-4] have demonstrated that the macroscopic CDW condensate is not always well described by only a single-phase coordinate, and thus multiple degrees of freedom, corresponding to internal CDW deformations or macroscopic domain structure, must be included in a complete theoretical description.

One method of probing CDW dynamics and internal CDW phase structure is through investigations of transient electrical response. Gill[5] has used transient response to demonstrate long-time memory effects resulting from metastable states (i.e., particular configurations of local CDW phase) in $NbSe_3$. Related experiments by Fleming[6] have revealed that the coherent voltage oscillations or "narrow-band noise" in $NbSe_3$ can be "synchronized" to the current pulses, and thus observed directly on an oscilloscope. Transient oscillations in $NbSe_3$ with a frequency corresponding to the internal (narrow-band noise) frequency were first reported by Zettl,[7] and subsequent work[8] has demonstrated similar "ringing" in the blue bronze $K_{0.3}MoO_3$.

In this Rapid Communication we report measurements of transient voltage response resulting from relatively short (nsec to μsec) current pulses in the CDW state of $NbSe_3$. For current pulse amplitudes exceeding the threshold value for CDW conduction, we observe dramatic ringing during the pulse, associated with initial CDW motion. We identify the transient ringing behavior as reflecting time dephasing of CDW domains. In a different experiment, we use the ringing phenomenon as a probe to explore the temporal evolution (i.e., relaxation of internal phase deformations) of a pinned but initially "polarized" CDW state, in the absence of an applied electric field.

Our experiments employed single $NbSe_3$ crystals in two-probe sample mounting configurations with silver paint contacts. Although our setup allowed complex waveforms (such as multiple pulses, pulses of alternating polarity, and pulses of variable rise and fall times, etc.) to be applied, we here report only on the response due to unipolar rectangular current pulses superposed on a variable dc bias current. The minimum pulse rise and fall times obtainable were limited by pulse generator characteristics to approximately 7 nsec. The voltage across the sample was first amplified with a low-noise high-frequency preamplifier (bandwidth ~100 MHz) and subsequently recorded with a high-speed digitizer. Our digitizing system (Tektronix 7912AD) allowed single-shot events to be recorded with an exceptionally fast real-time digitizing rate of 100 GHz (10 psec per address), and no interpolation or fill techniques were necessary. Signal averaging of successive single-shot events was used on occasion to either improve signal to noise or, more importantly, to compare the average of multiple traces (each triggered by the pulse generator) to a single-shot trace.

Figure 1(a) shows the voltage response of $NbSe_3$ at $T = 48$ K for three different values of current pulse amplitude, all with zero dc bias. Each trace represents the average of 64 nearly identical traces. For the two traces where the average voltage V_f during the pulse exceeds the threshold voltage V_T for CDW conduction, transient oscillation behavior, or ringing, is clearly observed. The frequency of the oscillations corresponds directly to the steady-state narrow-band noise frequency, as determined by spectrum analyzer observation of the noise with an applied sample voltage $\langle V \rangle = V_f$.

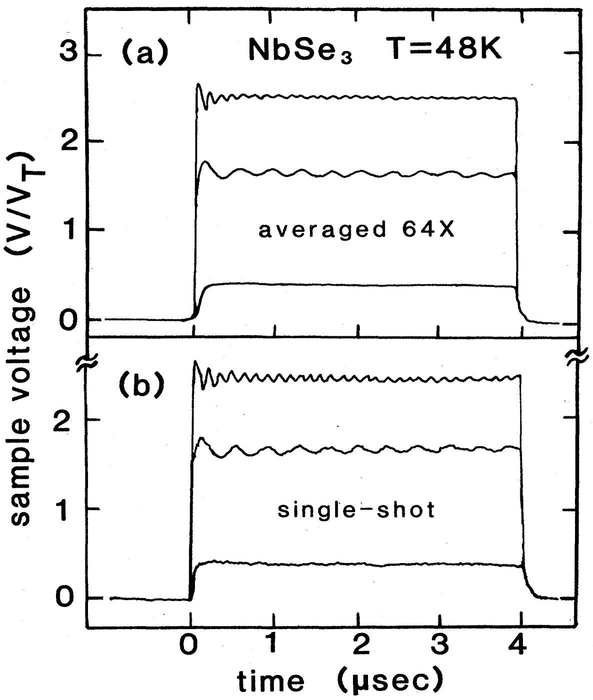

FIG. 1. Voltage response waveforms resulting from rectangular current pulses of various magnitude in $NbSe_3$. (a) Signal-averaged traces, (b) single-shot traces.

From the data of Fig. 1(a) alone it is difficult to determine if the ringing represents true transient behavior or is simply a manifestation of the signal-averaging process. For example, if different waveforms used in the averaging all had uniform amplitudes and the same phases at the start of the pulse, but slightly different phases near the end of the pulse, then the normalized sum of the waveforms would erroneously resemble ringing.[9] To rule out such a possibility, single-shot recordings were performed of the voltage response due to single isolated current pulses, again with zero dc bias. The results, displayed in Fig. 1(b), confirm that in $\mathrm{NbSe_3}$ transient ringing is indeed a real effect. A careful comparison of the data of Figs. 1(a) and 1(b) shows a nearly identical amplitude of the transient oscillations near the start of the pulse for signal-averaged and single-shot traces, indicating that the oscillations always start with a unique phase. The decay of the oscillations is, however, more severe in the signal-averaged traces, and in the signal-averaged representation the oscillations eventually die out completely. In the single-shot cases, the oscillation amplitude never falls completely to zero, but rather approaches a steady-state value, equivalent to the narrow-band noise amplitude. Comparing the decay of the signal-averaged traces to that of the single-shot traces allows a determination of the phase correlation time for the steady-state oscillations.[9]

For the moment, we consider only the ringing characteristics of single-shot traces. h_1 is defined as the amplitude of the first transient oscillation (i.e., the maximum height of the first overshoot above the time-averaged steady-state voltage V_f during the pulse[10]) and V_i is the dc current bias level just prior to and after the pulse. We find that, for $V_i=0$, h_1 is independent of the pulse amplitude (and hence ringing frequency), as long as V_f exceeds V_T. This independence was verified up to $V_f/V_T=5$; for higher amplitude pulses we were not able to accurately determine the amplitude of the oscillations due to bandwidth limitations. We find, however, the oscillation amplitude h_1 to be extremely sensitive to dc bias V_i, *even for* $V_i < V_T$. Figure 2 shows the results of an experiment where V_f was *fixed* at $2V_T$, and V_i was varied from $-3V_T$ to over $20V_T$. Results from two different $\mathrm{NbSe_3}$ crystals are shown, and all voltages have been normalized to V_T. The frequency of the ringing was found to be determined strictly by V_f and independent of V_i. However, as Fig. 2 shows, the amplitude of the transient oscillations is strongest if the CDW is "started" from the pinned regime, i.e., if $-V_T < V_i < V_T$. As V_i approaches V_T from below or $-V_T$ from above, h_1 decreases rapidly. Of particular interest in Fig. 2 is the position of the maximum value of h_1, which occurs not at $V_i=0$, but at a slightly positive current bias value, where $V_i/V_T=0.25V_T$. Thus ringing is somewhat enhanced if the CDW is pulsed from a slightly polarized configuration. Careful tests were performed to insure that the "shift" of the h_1 peak from $V_i=0$ was not due to spurious thermal emf offsets or a "built-in" directionality in the sample or the contacts. For example, the entire experiment was repeated with all applied currents reversed in sign; for this case the peak in h_1 was found to be at $V_i=-0.25V_T$.

In the range $V_T < V_i < V_f$, no ringing or transient "overshoot" was observed in single-shot voltage recordings. Upon signal averaging, however, transient oscillations of small magnitude were resolved, and h_1 corresponding to these oscillations has been plotted in Fig. 2 for the range $V_T < |V_i| < V_f$. For $V_i > V_f$, *no* transient oscillations or overshoot effects were observed, in either single-shot recordings or signal-averaged responses.

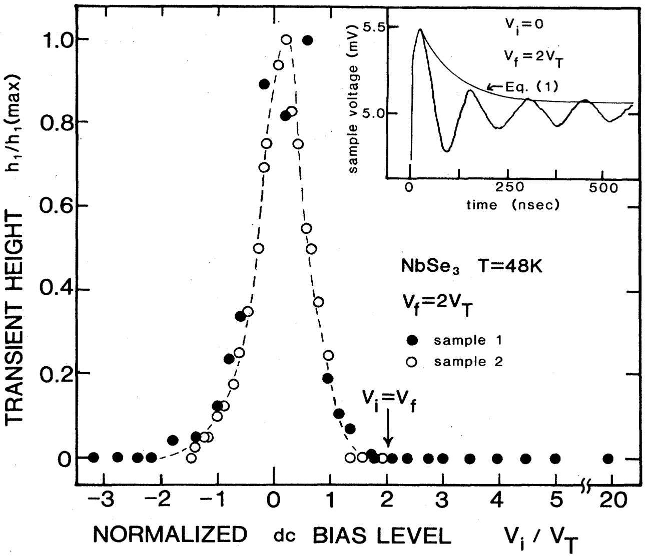

FIG. 2. Transient height h_1 vs dc bias current level V_i in $\mathrm{NbSe_3}$. The current magnitude during the pulse is fixed at $V_f=2V_T$. The arrow indicates the position of zero relative amplitude for the current pulse. The inset shows Eq. (1) fit to a typical ringing waveform, with fitting parameters given in the text.

The transient ringing behavior displayed in Fig. 1 is reminiscent of inductive response in simple electronic circuits, and one might associate with the ringing "inertial" CDW response. This is, however, in apparent disagreement with the overdamped response observed in low-field ac conductivity studies,[11] and with the lack of observed ringing following the end of the current pulse. On the other hand, in a recent study of the response of sliding CDW's from nonlinear mixing, Coppersmith and Littlewood[12] suggest a mechanism whereby the internal modes of the CDW system lead to enhanced dissipation at the characteristic "washboard" (or narrow-band noise) frequency. A consequence of the dissipation is pseudoinductive ringing in the voltage response resulting from sudden changes in the applied field above threshold. This model has been applied to transient voltage oscillations observed in the CDW state of $\mathrm{K_{0.3}MoO_3}$,[8] and has been suggested as a possible explanation for the low-frequency inductive loops observed in $\mathrm{NbSe_3}$ driven by large amplitude ac electric fields.[13]

An important feature of the dissipation mechanism suggested by Coppersmith and Littlewood is that the ringing is a consequence only of the rapid change in applied field; the sign of the change is not critical. Hence, a simple test of the theory as it might apply to $\mathrm{NbSe_3}$ is to compare the voltage response from current pulses which originate from below V_f to the voltage response from pulses which originate from above V_f. As was discussed previously, and is apparent from Fig. 2, *no ringing is observed* in $\mathrm{NbSe_3}$ if $V_i > V_f$. Hence, the dissipation mechanism of Coppersmith and Littlewood[12] does not appear to give a self-consistent account of the transient response here observed, in particular, in the regime where V_i is close to V_f, and perturbation theory expansions apply.

We suggest an alternative mechanism for the transient oscillations in NbSe$_3$ which again assumes additional degrees of freedom for the CDW condensate, but in the form of phase domain structure. For a moving (i.e., depinned) CDW condensate, the static phase coherence length ξ_s (Lee-Rice length[14]) is supplanted by ξ_D, the dynamic phase coherence length. Typically $\xi_S < \xi_D < l$, where l is the dimension (length) of the specimen. Thus "domain" structure results for which the (time-dependent) CDW phase is correlated only over a volume significantly smaller than the sample volume.[4,15] Internal domain structure allows for a natural distribution of phases associated with different regions of the sliding CDW condensate, and under steady-state conditions a moving CDW will not in general be fully phase coherent. On the other hand, it has been demonstrated[16] that ξ_D may be dramatically enhanced by the application of moderate amplitude rf electric fields, even to the point where $\xi_D >> l$ and the entire sample volume becomes 100% phase coherent.

We suggest that, in the experiments discussed here, a sharp current pulse originating from below threshold serves to temporarily homogenize the CDW phase throughout the sample. Thus all regions of the sample start oscillating with a unique phase, and a large oscillation amplitude is obtained at the start of the pulse. The subsequent decay of the oscillations, as displayed in Fig. 1(b), then corresponds to a time dephasing of domains. Following the start of the pulse, the dephasing process does not continue indefinitely, but rather approaches a limiting value, dictated by the intrinsic dynamic phase coherence length ξ_D in the crystal. We note that for large sample volumes, the limiting amplitude value of the oscillations may be so small that narrow-band noise is no longer observable, as is often the case in K$_{0.3}$MoO$_3$.[8] The CDW domain dephasing process is entirely consistent with no transient response being observed when the NbSe$_3$ sample is pulsed from a relatively high dc bias level to a lower (conducting) state. In this case, the CDW domains are, just prior to the pulse, already fully dephased, and no improvement in phase correlation is possible by a simple change in electric field.[17] The (very small) ringing observed in the range $V_T < V_i < V_f$ in Fig. 2 suggests there exists a finite number of pinned domains near threshold in the sliding CDW state.

As a first approximation to the dephasing mechanism, we write for the voltage oscillation amplitude during the current pulse

$$h = A + B \exp(-v_d t/\Gamma) \quad , \tag{1}$$

where A is the steady-state narrow-band noise amplitude, v_d is the CDW drift velocity, and B and Γ are adjustable parameters. We associate with the dephasing process a relaxation time $t_D = \Gamma/v_d = \Gamma'/f$, with f the narrow-band noise frequency. The inset of Fig. 2 shows Eq. (1) fit to a typical transient oscillation in NbSe$_3$ with the fitting parameters $A = 0.06$ mV, $B = 0.44$ mV, $\Gamma' = 0.65$, and $f = 7$ MHz. Similar fits indicate that Γ' is relatively independent of current pulse amplitude (or dc bias level), as long as f exceeds approximately 3 MHz. This was verified up to $f = 20$ MHz. For lower f, Eq. (1) still yields excellent fits to the transient oscillations, but Γ' increases with decreasing f. The strong current bias dependence of the ringing amplitude is contained in the parameter B.

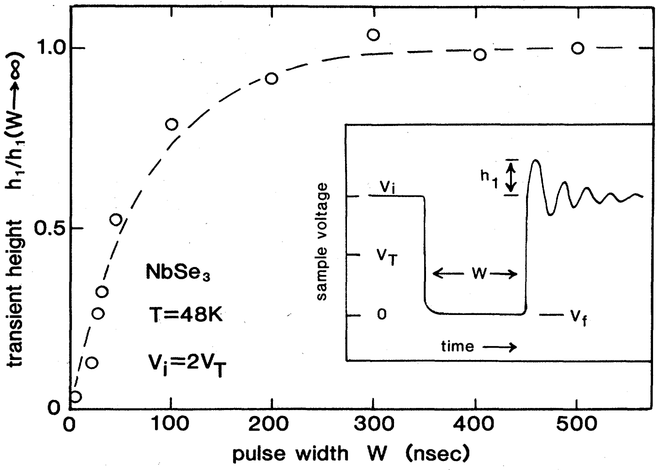

FIG. 3. Transient height h_1 vs pulse width W for NbSe$_3$, using the pulse configuration shown in the inset. The dashed line is Eq. (2), with fitting parameters given in the text.

In a second experiment, we have used the ringing phenomenon as a probe to directly determine the time dependence of relaxation to the equilibrium configuration of a CDW condensate rapidly quenched from the sliding state to the zero-field state. As shown schematically in the inset of Fig. 3, the experiment consists of setting the dc bias level to $V_i > V_T$, and then applying a (negative polarity) current pulse of width W. During the pulse, $V_f = 0$. Of interest is the transient response which occurs immediately *following* the end of the pulse, as the sample voltage returns to V_i. For sufficiently long pulse widths, the transient response following the end of the pulse is found to be identical to that observed at the beginning of pulses such as described in Fig. 1, with zero dc bias and a similar pulse amplitude. As the pulse width W is shortened, the transient oscillation amplitude h_1 decreases markedly, although the decay time t_D and frequency of the ringing remain unchanged. Figure 3 shows h_1 as a function of W for NbSe$_3$ at $T = 48$ K, with V_i/V_T fixed at 2. The data fit well an exponential time dependence of the form

$$h = B[1 - \exp(-W/t_r)] \quad , \tag{2}$$

with a characteristic relaxation time $t_r = 77$ nsec. This relaxation time corresponds closely to that determined from the classical "crossover" frequency 45 MHz $= (22 \text{ nsec})^{-1}$ for NbSe$_3$ at the slightly lower temperature 45 K, in low-field ac conductivity studies.[11] We associate t_r with the relaxation of internal phase deformations. In the present experiments, these deformations set in as the external electric field is removed from the sliding CDW state, and the CDW phase is repinned in a strongly polarized configuration. We note that the fast CDW relaxation observed here is clearly distinct from that previously determined for NbSe$_3$ and TaS$_3$ from pulsed memory[5] or thermal quenching[2] experiments. In effect, those experiments probe only the very slow relaxation processes associated with phase deformations, with characteristic time scales orders of magnitude larger than t_r evaluated here.

In conclusion, we have demonstrated transient ringing in NbSe$_3$, which we identify with time dephasing of CDW domains. We have also determined explicitly the time constant for fast relaxation of induced CDW polarization in the pinned regime. It appears that a significant enhancement in the dynamic phase coherence length, induced through the application of individual current pulses, is possible only if the CDW condensate is initially in a pinned and only weakly polarized configuration.

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