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Observation of a pulse-duration memory effect in $K_{0.30}MoO_3$

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We have observed a new memory effect related to metastable states of a moving charge-density wave (CDW) in $K_{0.30}MoO_3$. At 45 K we find that the transient oscillations seen in the leading edge of current or voltage pulses are influenced by the duration of the preceding pulse. For repetitive pulses this results in an unusual waveform where the phase of the last oscillation in a pulse is anomalously large and phase locked to the trailing edge of the pulse. The phase of the initial oscillation remains phase locked to the leading edge of the pulse. If one changes the pulse width in a string of repetitive pulses, the anomalous shape of the waveform is attained by the second pulse at the new width. These results suggest that information about the width of a pulse is contained in the phase configuration of the pinned charge-density-wave state present between pulses. The effect of a pulse, on average, is to advance the CDW phase by integral multiples of 2π.

One of the more remarkable manifestations of moving charge-density waves (CDW's)[1] is the pulse-sign memory effect.[2-4] Pulse-sign memory occurs when one applies current pulses of alternating sign to a material with a moving charge-density wave and observes the resulting voltage response. At temperatures well below the CDW onset (e.g., 77 K in $K_{0.30}MoO_3$), the voltage response will be fast (<2 μs) if the current pulse is the same sign as the preceding pulse, but slow (~ 1 ms) if the pulse is of opposite sign. A hysteretic CDW response, such as the pulse-sign memory effect, occurs because of metastable states of the CDW. Metastable states are local variations in the phase of the CDW which occur because the pinning on the CDW is not uniform on a microscopic scale.[5] They can be populated by changing the field or by changing the temperature in the presence of a field.[4,6] Metastable states occur as a result of local motion of the CDW in electric fields which are too small to induce bulk depinning. This has been shown to lead to a loss of CDW order transverse to the CDW wave vector.[7] In order for the CDW to evolve from its ground state to a moving state, the phase must progress through a large number of metastable configurations, a process which occurs over time intervals on the order of milliseconds. When the field is turned off, the CDW relaxes to a pinned metastable state with a high elastic energy, rather than the ground state. Consequently, if a current pulse has the same sign as the previous pulse, the response is fast because the CDW does not have to progress through as many metastable phase configurations.

In this study, we report a highly unusual manifestation of metastable behavior in which $K_{0.30}MoO_3$ shows a "memory" of the duration of the previous pulse in addition to the pulse-sign memory. The pulse-duration memory is seen in the transient voltage oscillations which occur in response to a current or voltage pulse.[8] We find that for repetitive pulses, a large amplitude oscillation, which is phase locked to the trailing edge of the pulse, is present. Our results imply that information about the width of a pulse is contained in the phase configuration of the pinned CDW attained after a pulse. After the current is turned off, the CDW returns to a unique metastable state, or perhaps a set of states, determined by the duration of the pulse. Our results further imply that the phase configuration of the pinned CDW state influences the transient oscillations seen subsequently in the moving state. In this manner a "memory" of the duration of the previous pulse is achieved.

We applied constant current pulses to a $K_{0.30}MoO_3$ sample mounted in a two-lead configuration with ultrasonically soldered indium contacts. The quality of the contacts was checked as described previously.[9] The voltage response was recorded with a Textronix model No. 7854 digital storage oscilloscope, and repetitive waveforms were averaged to improve the signal-to-noise ratio.[8] (The anomalous waveforms discussed below can also be seen in single-shot waveforms.) The sample was mounted on a sapphire substrate in a He-gas-filled capsule attached to a closed-cycle He refrigerator. Most data were taken at 45 K, where the transient oscillations are most pronounced. The results shown are from a current-biased sample; however, we also measured the current response of a voltage-biased sample. The results in the two cases are identical with the only change being the expected 180° phase shift of the oscillations in the measured curve.

A demonstration of the pulse-duration memory is shown in Fig. 1, where the voltage response is shown for rectangular current pulses from zero to a fixed final level, but of different pulse widths. The end points of the various pulses map the slow response of the CDW reported earlier.[4] The period of the pulses is 10 msec and the transient voltage oscillations seen previously[8] are prominent. In all curves the voltage response shows an initial large peak followed by smaller oscillations. As the pulse width is increased, note that the phase of the final oscillation remains fixed relative to the end of the pulse. For a pulse of arbitrary width, the observed oscillations are not truly periodic. The phase of the final oscillations is determined by the end point of the pulse rather than the phase of initial oscillations. Paradoxically, the material appears to have prescience and "know" when the end of the pulse will occur. What actually happens is a "learned" response where the system acquires a preferred phase after one repetition of a pulse of a given width. The phase of the final oscillations is only phase locked to the end of the pulse in the case of repeated pulses.

In Fig. 2 we show the oscillatory response observed if the

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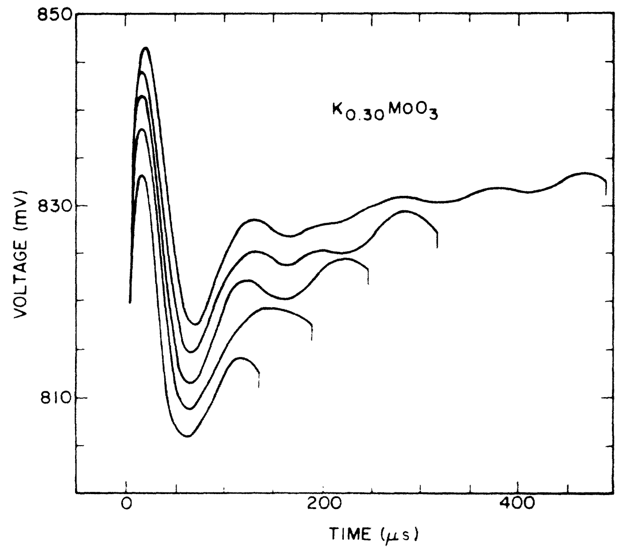

FIG. 1. The transient, oscillatory response of $K_{0.30}MoO_3$ at 45 K to a series of repetitive pulses with a period of 10 ms and varying widths. The phase of the last oscillation is fixed relative to the end of the pulse.

current is not turned off at the usual point. The solid line is the signal-averaged response of a pulse 1350 μs wide which had been preceded by 20 pulses 600 μs wide. Just before 600 μs, the point where the previous pulse ended, notice the anomalous large oscillation. The end point of the previous pulse corresponds to a zero in the oscillatory response. The dotted line in Fig. 2 shows the averaged response of a 900-μs pulse which was preceded by 20 pulses of random width. The vertical scales have been shifted to overlay the two responses. Not surprisingly, the large anomalous oscillations are not present if the measured pulse is preceded by pulses of random width. Also, for a pulse preceded by random pulses, the response is truly periodic with all oscillations in phase with the first.

The time needed to acquire a "learned" response is shown in Fig. 3. Each counted pulse in Fig. 3 is preceded by 50 "training" pulses with a width of 600 μs and then N pulses with a width of 1000-μs. Only the Nth wide pulse 1000 μs wide is recorded and averaged. The vertical scales in Fig. 3 have been shifted for clarity, and the initial large oscillation is not shown as most changes begin with the third period. For $N=1$ pulse, the anomalous large oscillation at 600 μs is prominent as described above. If one records the second wide pulse ($N=2$), the curve is quite different. By $N=2$, the large oscillation at 600 μs has disappeared, and a new anomalous oscillation can be seen just before the end of the 1000-μs pulse. Little change between the second wide pulse and the fiftieth is evident. Thus at 45 K, only one pulse is required to "train" the sample and establish an anomalous oscillation in the next pulse.

For pulse periods between about 1 ms and 1 s (the longest period used), the transient oscillatory response is independent of the period at 44 K. However, if the off time is less than about 1 ms, the amplitude of all oscillations decreases. At 77 K the amplitude of the oscillatory response is diminished for periods less than about 15 ms. This suggests that CDW needs time periods on the order of the transient response time to become pinned. If a pulse begins before the CDW has stopped, the transient oscillatory response is diminished. Furthermore, this establishes that the anomalous oscillations are not a simple beat phenomena

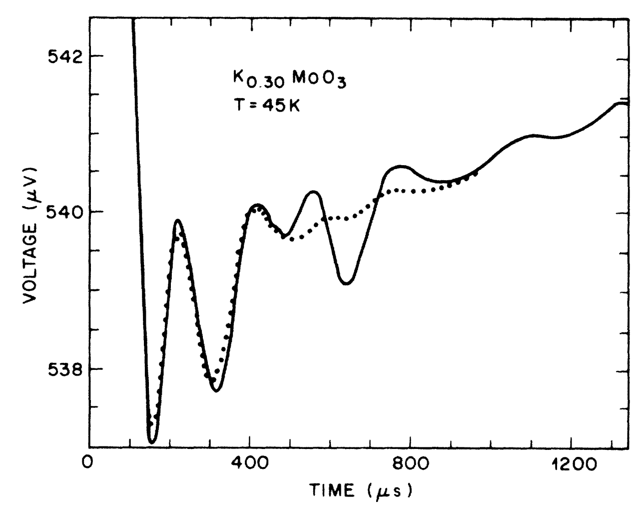

FIG. 2. Solid line: the response measured with a pulse 1350 μs wide, which was preceded by 20 pulses 600 μs wide. Note the large oscillation at 600 μs, the end point of the previous pulses. Dotted line: the response measured with a pulse 900 μs wide which was preceded by 20 pulses of random widths. For a random pulse sequence, the anomalous oscillatory effects are not present.

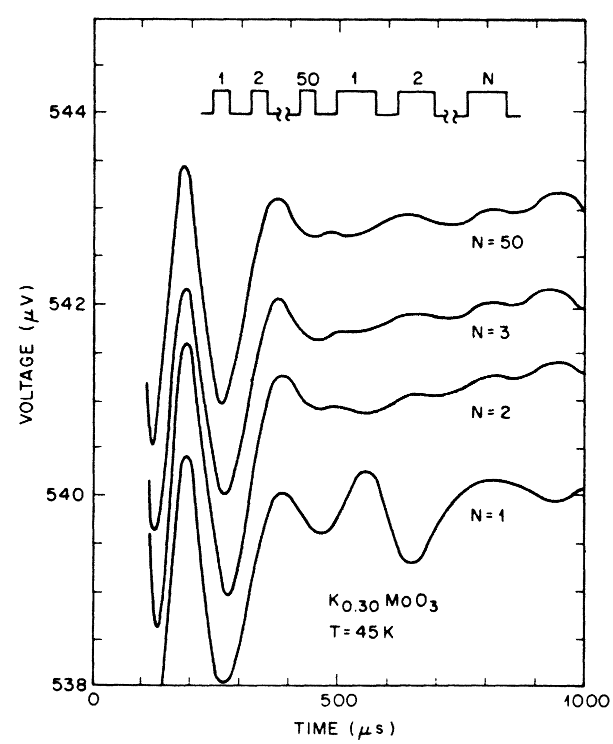

FIG. 3. The response measured with a pulse 1000 μs wide which was preceded by 50 pulses 600 μs wide and $N-1$ pulses 1000 μs wide. After one repetition the large oscillation at 600 μs (marking the end of the narrow pulses) vanishes. Note also the development of an anomalous oscillation at 1000 μs, the end point of the new pulses, after one repetition.

related to the pulse period.

We presently have no explanation of the large final oscillation of a pulse seen after repeated pulses. The fact that the final oscillation is phase locked to the end of the pulse may be a feature resulting from mode locking of the charge-density wave, a phenomenon which has been studied extensively in $NbSe_3$. Mode locking occurs when one applied both an ac and a dc in bias to a sample. For large values of the ac amplitude, the sample can become mode locked in the sense that the washboard frequency of the moving CDW (a measure of the CDW velocity) is constant and equal to the driving frequency for a range of dc biases.[10] A pulse can be thought of as a combined ac-dc bias, so mode locking may play a role in pulse experiments such as the present one. In $NbSe_3$, the oscillatory response to a current pulse is dominated by steady-state dc oscillations, the so-called "narrow-band noise." The steady-state dc oscillations are phase locked to the leading edge of the pulse only if the pulse width is equal to an integral number of oscillations,[3] i.e., only when the CDW phase is advanced by an integral multiple of 2π. The effects of mode locking on the pulse response are apparently quite different in the case of $K_{0.30}MoO_3$. First, no steady-state oscillations are seen in our samples, so locking of the phase of the pulse and that of the steady-state oscillations as in $NbSe_3$ is not seen. Instead, a transient oscillatory response which is influenced by the duration of the previous pulse is present. The information needed to couple the oscillatory response in one pulse to that of the next must be contained in the phase configuration of the pinned metastable state present during the off time. This implies that the phase configuration of the pinned state is a function of the pulse duration. Furthermore, at least on average, the phase of the pinned CDW at each point in the sample is advanced by integral multiples of 2π after each pulse, so that the same pinned state is attained. Conversely, the nature of the transient oscillatory response is influenced by the phase configuration of the starting state. The present experiments suggest that mode locking occurs in $K_{0.30}MoO_3$ in the sense that the net CDW motion is constant for a range of pulse widths; however, this is not demonstrated explicitly. Therefore, we suggest that the CDW may only have a tendency to advance by 2π after a pulse.

Coppersmith and Littlewood have performed numerical simulations of charge-density-wave systems using both a periodic pinning potential[11] and a random-pinning potential.[12] Their models exhibit metastable behavior, the pulse memory effect, and transient oscillations in response to a pulse. Within these models, the system will eventually reach an equilibrium moving state which is unique for a given field. Therefore, at long times, one would not expect the phase configuration of the moving CDW to be influenced by the starting configuration. However, for short times, as is the case in our experiments, the phase in the moving state at a given time may be a function of the starting configuration. This supposition is confirmed by numerical simulations, on a system with a periodic pinning potential, which show that the spectral content of the transient oscillations is dramatically changed if one starts from a uniform, rather than a pinned state.[13] In the case of the random-pinning model, mode locking, as discussed above, is evident. After repetitive pulses, the phase of the pinned CDW, at each point of the sample, is advanced by precisely integral multiples of 2π (Ref. 14).

Prior to this experiment it was thought that metastable states were only a property of the pinned charge-density wave, and that the moving state is unique for a given field level. This experiment demonstrates that this assumption is not valid for short-time scales. For times less than the CDW relaxation time, interpreted here as the time required for the oscillatory response to vanish, the phase configuration of the moving CDW is a function of the history of the sample, just as the phase of the pinned CDW is a function of the sample history. As a result, the oscillatory, transient oscillations in a given pulse contain information about the duration of the previous pulse, and the sample has a "memory" of the duration of the previous pulse as well as the sign. It appears that for repetitive pulses shorter than the CDW relaxation time (the time required for the transient oscillations to decay to zero), the CDW has a tendency to advance by integral multiples of 2π at each point in the sample, a manifestation of a mode-locked response. Although a physical model of anomalous oscillations is presently lacking, it is encouraging to note that numerical simulations of CDW transport contain many of these same features.[10–14]

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