Temperature dependence of the pulse-duration memory effect in $NbSe_3$

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The temperature dependence of the oscillatory response of the 59 K charge-density wave in $NbSe_3$ to a sequence of repetitive current pulses was investigated. For 52 $K > T > 45$ K the learned behavior commonly referred to as the pulse-duration memory effect (PDME) is very evident; after training the voltage oscillation always finishes the pulse at a minimum. At lower temperatures the PDME changes qualitatively. In nonswitching samples the voltage oscillation always finishes the pulse increasing. In switching samples there is a conduction delay which becomes fixed after training, but no learning of the duration of the pulse.

Quasi-one-dimensional conductors exhibit a rich variety of response to an applied current or voltage due to the many internal degrees of freedom. Above threshold an incommensurate charge-density wave (CDW) depins from the impurities and other crystalline defects, and effectively ''slides'' through the crystal creating a new conduction channel.[1] There are a large number of metastable pinned states accessible to the CDW below threshold. This large number leads to numerous memory effects,[2–6] including the pulse-duration memory effect (PDME).[7–9] Upon application of a series of identical unipolar current pulses, the voltage response learns when the current pulse is going to end. The learning is due to the special metastable state into which the CDW relaxes between pulses. This metastable state effectively encodes the length of the previous pulse. After training, if the same current pulse is applied again then the voltage response, which oscillates in time, always finishes the pulse at the same phase in the oscillation. A multiple pulse-duration memory effect has also been observed.[10]

Experimental studies of the PDME have focused on the materials $NbSe_3$ (Ref. 8) and $K_{0.3}MoO_3$.[7,9] The main emphasis of these studies has been the dynamics of the learning. Here we report the temperature dependence of the PDME in $NbSe_3$. We find that there is a qualitative change in the PDME upon cooling, and at low temperatures there is a difference between switching and nonswitching samples.

For this study we used high quality single crystals of undoped $NbSe_3$ with residual resistance ratios $[\rho(300\ \mathrm{K})/\rho(4.2\ \mathrm{K})]$ of over 300. Crystals grow as long needles with typical dimensions of 1 cm$\times$10 μm$\times$1 μm. Data were taken in a two-probe configuration after determining that contact resistance was less than 5% of the total sample resistance. Current pulses were created using a Stanford DS345 function generator and a large resistor (typically 100 kΩ) in series with the sample. Voltage wave forms were captured with a Tektronix TDS744 digitizing oscilloscope.

$NbSe_3$ undergoes two CDW transitions. For this study we investigated only the 59 K CDW transition because the amplitude of the voltage oscillation, hereafter referred to as the narrow-band noise (NBN), is much larger than for the 143 K CDW. Most studies of CDW dynamics concentrate on the temperature range near 50 K. This is where the threshold voltage for CDW depinning (V_T) is a minimum,[1] and ac-dc mode locking and pulsed mode locking are most likely complete.[11]

We investigated the temperature dependence of the PDME in seven samples. Figures 1 and 2 show data for two of these samples, which are typical for the temperature range from 52 K to 45 K for all seven samples. For this range the PDME is very pronounced; the learned response to a repetitive unipolar current pulse sequence results in the NBN oscillation finishing the pulse at a minimum (maximum CDW velocity). Okajima and Ido previously reported similar results for this temperature range.[8]

Characterizing the PDME below 35 K is difficult for a number of reasons. The amplitude of the NBN oscillation in the pulsed-mode-locked state diminishes, and the mode locking tends to degrade and become more intermittent.[12] Also

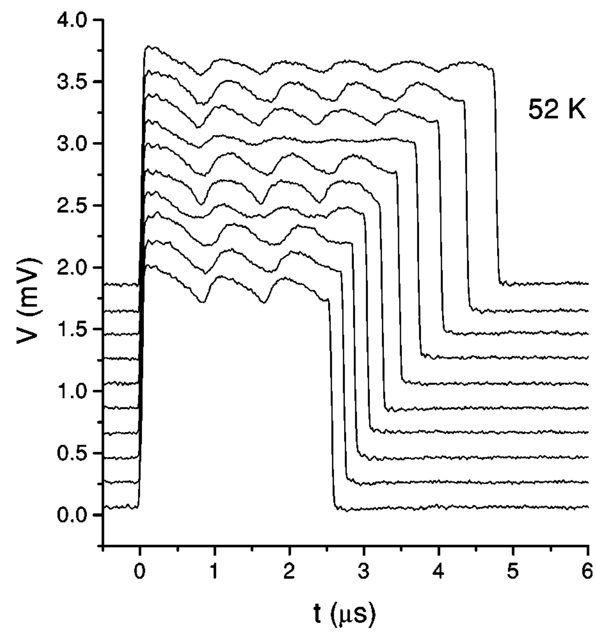

FIG. 1. Wave forms of the voltage response to a sequence of repetitive unipolar current pulses (see inset Fig. 2). The ten wave forms are each for a sequence with a different pulse duration. $I_{high}=1.5I_T$ and $t_{low}=100$ μs for all ten sequences. Wave forms are averaged 20 times to reduce noise. Curves are offset in steps of 0.2 mV for clarity. At this temperature $V_T=1.5$ mV and $I_T=8.0$ μA. Sample length, width, and thickness are 820 μm, 5 $\pm$1 μm, and 1.0$\pm$0.2 μm, respectively. Note that the number of oscillations increases stepwise with increasing pulse duration, and that the oscillation always ends near an oscillation minimum.

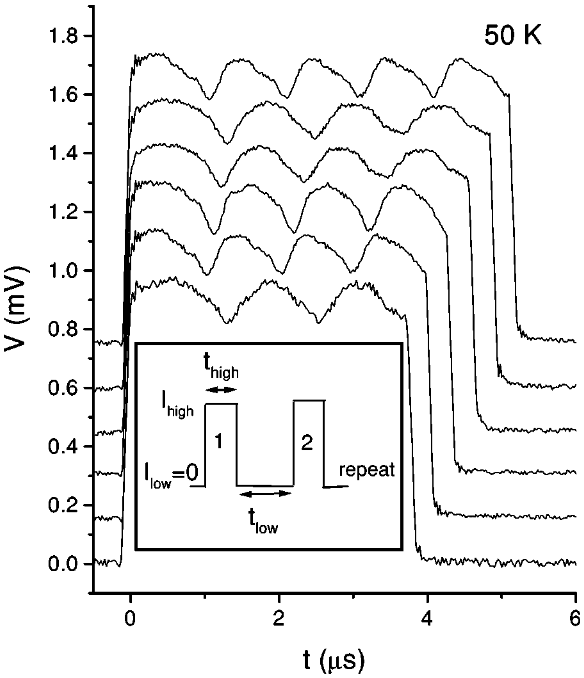

FIG. 2. Wave forms of the voltage response to a sequence of repetitive unipolar current pulses. The six wave forms are each for a sequence with a different pulse duration. $I_{high}=1.5I_T$ and $t_{low}=100$ μs for all six sequences. Wave forms are averaged 25 times to reduce noise. Curves are offset in steps of 0.15 mV for clarity. At this temperature $V_T=0.8$ mV and $I_T=6.5$ μA. Sample length, width, and thickness are 390 μm, 3 ± 1 μm, and 1.3 ± 0.4 μm, respectively. Note that the number of oscillations increases stepwise with increasing pulse duration, and that the oscillation always ends near an oscillation minimum. Inset shows schematic of the repetitive current pulse sequence. For all data reported here at least 10^6 pulses in the sequence were applied before capturing the wave form.

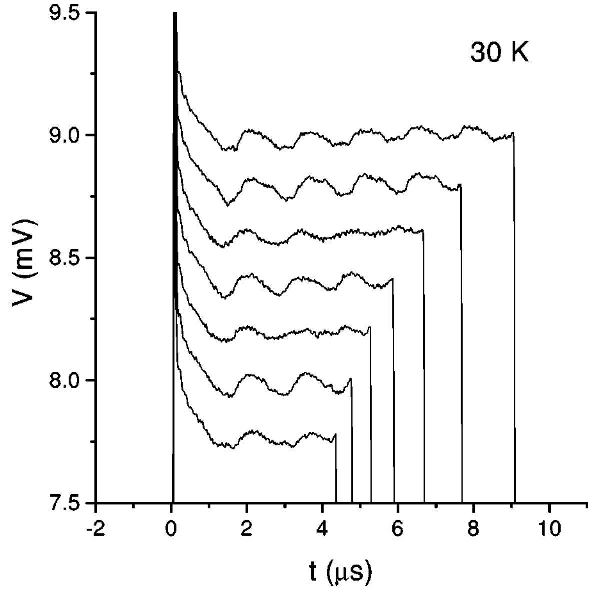

FIG. 3. Wave forms of the voltage response to a sequence of repetitive unipolar current pulses. The seven wave forms are each for a sequence of different pulse duration. Same sample as in Fig. 1. These data illustrate what we refer to as type A behavior. $I_{high}=1.5I_T$ and $t_{low}=100$ μs for all seven sequences. Wave forms are averaged 20 times to reduce noise. Curves are offset in steps of 0.2 mV for clarity. At this temperature $V_T=5.1$ mV and $I_T=41.2$ μA. This is a nonswitching sample.

the rapid increase of V_T with decreasing temperature[1] complicates measurements because the NBN oscillation is a smaller fraction of the total dc voltage. Despite these difficulties we were able to resolve the real time NBN oscillation down to 25 K in four of the samples. A qualitative change in PDME behavior is clearly apparent. This crossover occurs between 35 and 45 K.

The main point of this paper is the low temperature behavior. We observe two qualitatively different behaviors at 30 K. What we refer to as type A behavior was observed in two of these four samples, and is illustrated in Fig. 3. Unlike the higher temperature data, the NBN oscillation always finishes the pulse increasing.

Type B behavior is illustrated in Fig. 4. For the first 0.5 μs of the pulse the CDW remains pinned. After a delay it depins, but the CDW does not learn the length of the pulse; for an arbitrary pulse length the NBN oscillation can finish the pulse at any position. Type B behavior was observed in both of the samples that exhibited switching at 30 K. Switching refers to a sudden jump in the dc I-V characteristic near threshold. Conduction delays are a common phenomenon in switching samples. Extensive studies characterizing conduction delays have been reported by Levy and Sherwin.[13] They find that for this temperature regime the delay time is on the order of 1 ms to 1 s for pulse drive amplitudes just above threshold. The delay time rapidly decreases with increasing pulse height, and is on the order of 1 μs for pulse amplitudes above $1.1V_T$.[13] Data from the two switching samples are consistent with their results. Levy and Sherwin find a

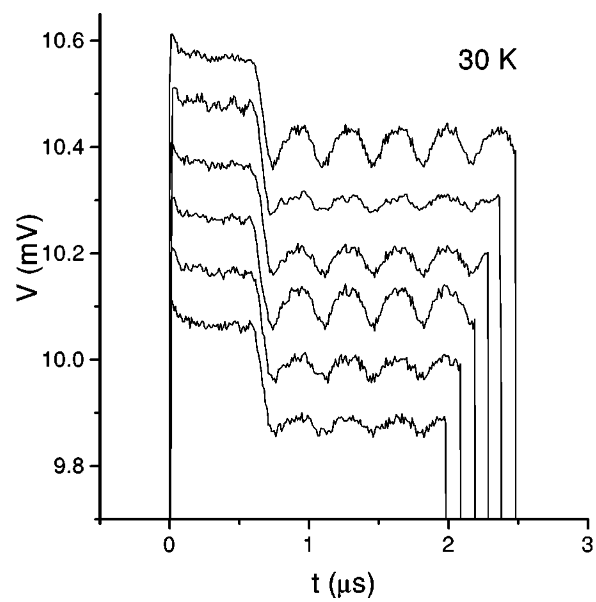

FIG. 4. Wave forms of the voltage response to a sequence of repetitive unipolar current pulses. The six wave forms are each for a sequence of different pulse duration. Same sample as in Fig. 2. These data illustrate what we refer to as type B behavior. $I_{high}=1.2I_T$ and $t_{low}=100$ μs for all six sequences. Wave forms are averaged 25 times to reduce noise. Curves are offset in steps of 0.15 mV for clarity. At this temperature $V_T=9.2$ mV and $I_T=121$ μA. This sample switches at this temperature.

distribution of conduction delay times for a virgin cooled CDW. In contrast, *we find that for a repetitive pulse sequence the conduction delay is the same from pulse to pulse.* In other words, we observe a pulse delay memory effect. We attempted to recover type A behavior in the two type B samples by increasing the pulse height in order to reduce the delay time. We were unsuccessful in this attempt.[14]

First we will discuss the high temperature data. Here comparison with the phenomenological single coordinate model (SCM) is useful.[1] In this model the phase ϕ of the CDW superlattice changes in time according to

$$\gamma \frac{d\phi}{dt} = V - V_T \sin(\phi). \tag{1}$$

Here γ is a phenomenological damping constant, $d\phi/dt$ is proportional to the CDW current, and $-V_T \sin(\phi)$ is the pinning force. The CDW conduction channel is assumed in parallel with a conduction channel of normal carriers which obeys Ohm's law. The experimental observation that the voltage oscillation finishes the pulse near an oscillation minimum implies that $\phi_{end} = 3\pi/2 + (2\pi \times \text{integer})$ where ϕ_{end} is the phase of the CDW at the end of the pulse. However, for a driving pulse of arbitrary height and width this model cannot reproduce the learned behavior. Assuming the CDW fully relaxes between pulses, $\phi_{initial}$ is equal to $2\pi \times \text{integer}$. For an arbitrary current pulse ϕ_{end} can take on any value, and therefore the induced *V* oscillation for the SCM can finish the pulse at any point along the oscillation, not necessarily at a minimum. This indicates that the internal degrees of freedom of the CDW are responsible for the learned behavior.

The theory of phase organization, which takes into account the internal degrees of freedom of the CDW, provides a possible explanation of the PDME.[15,16] This theory describes the learned dynamics of the Fukuyama-Lee-Rice model (FLR),[17] and provides a good description of the PDME in $K_{0.3}MoO_3$.[7] The FLR model simply includes the elastically coupled internal degrees of freedom of the CDW into the SCM. The name ''phase organization'' derives from the novel pattern formation associated with the learned dynamics. For a sequence of repetitive driving pulses the internal phase degrees of freedom of the FLR model relax into a special metastable state such that if the same driving pulse is again applied the pinning potential energy for each phase degree of freedom is near a maximum $[\phi_{end} = \pi + (2\pi \times \text{integer})]$ at the end of the pulse. However, as first noted by Okajima and Ido[8] the experimental data imply that $\phi_{end} = 3\pi/2 + (2\pi \times \text{integer})$. Ito has also questioned the theory of phase organization as an explanation of the PDME in CDW systems.[18] The major point here is that a phase-only model appears inconsistent with the high temperature data.

The low temperature data are even more surprising. In type A samples, comparison with the SCM implies $\phi_{end} = 2\pi \times \text{integer}$. This is the phase at which the pinning potential energy is a minimum, which is π rad away from the standard phase organization picture.

In type B samples, the training of the conduction delay is unexpected, but not surprising given the richness of CDW transport in $NbSe_3$. Previous investigations by various researchers have shown that additional physics must be incorporated into the FLR model to produce conduction delays, such as the inclusion of normal carrier screening or amplitude collapse of the CDW near contacts.[19,20] Future research needs to address the issue of whether the additional physics necessary to describe the data below 45 K implies that the phase-only description is inadequate above 45 K.

Above 45 K, to account for the PDME in $NbSe_3$ Okajima and Ido speculate that phase slip events near strong pinning centers organize their timing upon application of a repetitive drive sequence, although they provide no numerical evidence for this mechanism.[8] We speculate as follows. At low temperatures the conversion of normal carriers to CDW carriers near the contacts controls the dynamics. This process involves the formation of dislocations in the CDW superlattice. Pulsed mode locking of the CDW to a repetitive pulse sequence forces the dislocations to organize. The difference in type A and type B behavior is a contact effect; that is, a change in boundary conditions for dislocation formation. At higher temperatures, the dislocations that are pinned to the nucleation sites at the contacts ''melt,'' and the pinning due to the impurities in the bulk controls the dynamics. This is why the dynamics of type A and type B samples is similar at higher temperatures. We have recently observed that the energy required to shear the CDW abruptly changes at 44 K, supporting this hypothesis.[21] This is all speculative, and requires further investigation, both experimentally and theoretically.

Understanding the systematics of this phenomenon will provide a new direction in the study of learned behavior in nonlinear systems.[22] Further studies will require better control of contact fabrication and a better understanding of the current conversion process.

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