Dynamics of the electro-optic response of blue bronze $K_{0.3}MoO_3$

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We have studied the charge density wave (CDW) repolarization dynamics in blue bronze by applying square-wave voltages of different frequencies to the sample and measuring the changes in infrared transmittance, proportional to CDW strain. The frequency dependence of the electrotransmittance was fit to a modified harmonic oscillator response and the evolution of the parameters as functions of voltage, position, and temperature are discussed. Resonant frequencies decrease with distance from the current contacts, indicating that the resulting delays are intrinsic to the CDW with the strain effectively flowing from the contact. For a fixed position, the average relaxation time has a voltage dependence given by $\tau_v \sim V^p$, with $1 < p < 2$. The temperature dependence of the fitting parameters shows that the dynamics are governed by both the force on the CDW and the CDW current: for a given force and position, both the relaxation and delay times are inversely proportional to the CDW current as temperature is varied. The long relaxation and delay times ($\sim 1 \text{ ms}$) suggest that the strain response involves the motion of macroscopic objects, presumably CDW phase dislocations.

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I. INTRODUCTION

Interest in quasi-one dimensional conductors with sliding charge-density-waves (CDW’s) has continued for three decades because of the variety of unusual properties they exhibit. In the CDW ground state, a periodic lattice distortion is accompanied by a modulated electron density: $n = n_0 + n_1 \cos[Qz + \varphi(z,t)]$, where $z$ is the direction of the conducting chains and $Q$ is the CDW wave vector, $n_1$ its amplitude, and $\varphi$ its local phase. Because CDW pinning results from the deformation of the CDW (i.e., variations of $n_1$ and $\varphi$) due to interaction energy with impurities, CDW’s are also model systems for studying the effects of quenched disorder on a deformable periodic medium. This competition results in the CDW responding to different stimuli at a very large array of time scales.

When a voltage greater than its depinning threshold is applied, the CDW can slide through the sample, carrying current. At the same time, it becomes elastically strained, i.e., its phase varies throughout the sample so that the CDW is compressed near one current contact and rarefied at the other. The strain ($Q^{-1} \partial \varphi/ \partial t$) near a current contact is required to drive the phase-slip process needed for current conversion, i.e., to allow electrons to enter and leave the CDW condensate at the contacts. Strains near the center of the sample reflect shifts in the chemical potential due to imperfect screening by “normal” quasiparticles, i.e., those from noncondensing bands and/or those thermally excited out of the CDW. The spatial-temporal dependence of the strain in turn affects that of the CDW current.

Consequently, the illumination of the spatial dependence of the CDW current and strain, through tunneling, x-ray and infrared measurements, has been a major thrust area for the last several years. The Cornell group addressed this in an elegant series of conductivity experiments on NbSe$_3$ by closely spacing nonperturbative contacts along the sample, they measured local electric fields and solved model transport equations to deduce the local strain as functions of position and time after reversing the direction of current flow. Their results indicated that strains near ($\sim 100 \text{ } \mu m$) the current contacts reversed quickly ($\sim 10 \text{ } \mu s$) whereas the smaller strains in the center took several times longer to change. Subsequently, spatially resolved x-ray diffraction measurements at Grenoble (Ref. 7) directly measured the CDW strains in NbSe$_3$ and observed much larger contact deformations than those deduced by the Cornell group. The difference was attributed to the fact that in the Cornell model the quasiparticle conductivity was treated as a constant, whereas in the model of Ref. 7 the uncondensed and thermally excited quasiparticles respond differently to CDW strains, resulting in local changes in their density, larger strains near the contacts, and more extended regions of current conversion. The strains near the center of the sample persisted even when the applied voltage was removed. These nonequilibrating strains were associated with pinning of CDW phase dislocations, discussed further below.

Whereas NbSe$_3$ remains metallic in the CDW state, in most other sliding CDW materials all conduction bands are gapped in the CDW state. The CDW’s in these semiconducting materials are much less coherent than in NbSe$_3$, preventing x-ray measurements of the spatial dependence of the strain. Analysis of position dependent transport properties of these materials has also been complicated by the difficulty in preparing Ohmic but nonperturbative contacts. The absence of uncondensed electrons furthermore complicates the analysis of transport properties because as the density of thermally excited quasiparticles falls at low temperatures, solitonlike defects can dominate even the low field, Ohmic, conductance.

Instead, our group has used measurements of infrared transmittance and reflectance to measure position and time dependent changes in the semiconducting CDW phase in TaS$_3$ (Ref. 4) and $K_{0.3}MoO_3$ (blue bronze). The infrared changes occur primarily because, even at relatively high temperatures, the quasiparticle density is low enough that CDW strains can cause relatively large changes in their density, resulting in changes in intraband absorption. In addi-
... contacts. As in Fig. 1, most of these measurements were... of the sample for small applied voltages. The onset voltage for the... the sample bulk whereas \( V_T \) is associated with... measurements of the spatial/... frequency dependence of the electrotransmittance of blue bronze as functions of voltage, position, and temperature. We... the fragile samples which were mounted over... the incident IR light and joule heating from the applied currents. Infrared light from a tunable infrared diode laser was focused on the sample using an infrared microscope. The light, polarized along the sample width and with multimode power \( \sim 0.1 \text{ mW} \), was incident (on the face of the sample containing the contacts) through a rectangular aperture into a spot 50 \( \mu \text{m} \) along the length of the sample and 80 \( \mu \text{m} \) perpendicular. The light position was measured between the closest edges of the light spot and a metal film contact \( (x=0) \), but the contact edges were not always sharp, so the absolute positions are uncertain by \( \sim 20 \mu \text{m} \); in addition, the aperture image typically drifted with respect to the sample during a measurement by \( \sim 10 \mu \text{m} \). For relative changes in transmittance or reflectance, the infrared beam was chopped (at 390 Hz) and a square-wave voltage applied to the sample. The modulated transmitted or reflected signal was measured simultaneously at the square-wave \( (\omega) \) and chopping frequencies; the ratio of the signals precisely gives \( \Delta \theta/\theta \) or \( \Delta R/R \), even though the transmittance or reflectance were not separately measured...

precisely.\textsuperscript{17,18} Most measurements were made with symmetric bipolar square-waves, for which the changes in transmittance and reflectance are $\Delta \theta = \theta(+V) - \theta(-V)$ and $\Delta R = R(+V) - R(-V)$. Some unipolar transmittance measurements, for which $\Delta \theta = \theta(V) - \theta(0)$, were also made. In both cases, the responses both in-phase and in quadrature with the square wave were measured; the frequency-dependent phase shift of the microscope detector electronics was determined with a precision of $2^\circ$, possibly slightly affecting results at the lowest frequencies, where the quadrature component becomes much smaller than the in-phase component.\textsuperscript{4}

Thin samples were used to help ensure uniform current flow through the sample cross section. This was checked by comparing the dependence of the electrotransmittance and electroreflectance on position, voltage, and frequency, as shown in Fig. 2. As seen, only the absolute magnitudes of $\Delta \theta/\theta$ and $\Delta R/R$ differ, with $\Delta \theta/\theta \approx 3 \Delta R/R$, but they had the same spatial, voltage, and frequency dependences, indicating that the CDW phase gradient and current were uniform across the sample cross section, at least on the scale of the penetration depth of the light, typically a few times smaller than the sample thickness.\textsuperscript{16,17} Detailed measurements were then made primarily on the transmittance, for which the microscope was less susceptible to mechanical noise than reflectance. For each sample, a wavelength was chosen for which the laser power and electrotransmittance were large. (For a sample with $ad > 1$, where $a$ is the absorption coefficient and $d$ the sample thickness, the transmittance is given by $\theta = (1-R)^a \exp(-ad)$, making the electrotransmittance spectrum sample thickness dependent.) The photon energies used were between 775 and 890 cm$^{-1}$, on a broad plateau in the $\Delta \theta/\theta$ spectrum associated with quasiparticle absorption.\textsuperscript{17}

Samples were also chosen for having fairly regular, linear variations of $\Delta \theta/\theta$ with position when measured with bipolar square waves. An example is shown in Fig. 3. Samples with irregular spatial dependences presumably contained macroscopic defects (e.g., cracks, grain boundaries) that affected the CDW current flow but were not apparent from visible inspection. An interesting feature of Fig. 3, that we observed in most samples, is that the zero crossing of $\Delta \theta/\theta$, i.e., the position of zero CDW strain, varies with voltage. As discussed further in Sec. VI, this suggests that the relative “quality” of the two contacts varies somewhat with voltage.

Also shown in Fig. 3 is the spatial dependence when a unipolar square wave is applied. (Note that the magnitude of the unipolar response is roughly consistent with the deviation from linear spatial dependence near the contact observed for the bipolar response at the same voltage.) As discussed above, the transmittance will only oscillate for applied unipolar voltages for strains which decay when the voltage is removed, and that these “nonpinned” strains were previously only observed to occur adjacent to the contact.\textsuperscript{14,17} Now, we observe that there are also small unipolar variations in the center of the sample; similar observations were made for other (but not all,---see Fig. 12, below) samples. The unipolar spatial dependence shown in Fig. 3 suggests that the nonpinned strain may “overshoot,” flowing into adjacent regions and leaving them with strains of opposite sign during the “on” half cycle and/or strains of the same sign in the “off” half cycle.

All the present measurements are at temperatures (a) well above the “CDW-glass” transition (23 K) where the dielec-

![Image](https://example.com/image.png)

FIG. 2. Comparison of the (a) spatial, (b) voltage, and (c) frequency dependences of the electrotransmittance (solid symbols, $\nu = 820$ cm$^{-1}$) and electroreflectance (open symbols, $\nu = 850$ cm$^{-1}$) for sample 1 at $T = 80$ K at the frequencies, voltages, and positions indicated. Both the response in-phase and in quadrature with the driving bipolar square-waves are shown.

![Image](https://example.com/image.png)

FIG. 3. The spatial dependence of the electrotransmittance ($\nu = 820$ cm$^{-1}$) of sample 1 at $T = 80$ K in-phase with bipolar square waves at several voltages at 25 Hz, for which the quadrature response is negligible. The sample was 550 $\mu$m long and the light spot was 50 $\mu$m wide. Each data set is vertically offset by 0.2%; the dashed zero line for each data set is shown, with the voltage given on the right. The solid lines through the data points are for reference only. The open symbols show the response for a positive unipolar square wave (multiplied by 10).
tric time constant diverges\textsuperscript{22} and high enough that (b) the low-field resistance has activation energy approximately half the gap, indicating that the Ohmic transport is dominated by thermally activated quasiparticles\textsuperscript{11} and (c) the non-Ohmic conductivity is dominated by thermal activation over pinning barriers and only exhibits a single threshold voltage without hysteretic switching\textsuperscript{23}.

III. FREQUENCY DEPENDENCE OF BIPOLAR RESPONSE AT T=80 K

We have measured in detail the voltage and position dependence of the frequency dependence (2 Hz< ω/2π < 4 kHz) of the electrotransmittance at T≈80 K for three samples, of lengths L=550 μm (sample 1), L=810 μm (sample 2), and L=990 μm (sample 3), using bipolar square waves. For bipolar square waves, the response contains both the bulk, nonequilibrating portion of the strain which would stay pinned at V=0 and the nonpinned, contact strain. For samples 1 and 2, however, the nonpinned component piece was extremely small (e.g., see Fig. 3). It was relatively larger for sample 3, but as discussed in Sec. IV, still did not affect the fits significantly. We therefore assume that the dynamics of the bipolar response is always essentially that of the non-equlibrating, bulk strain.

Representative data sets at x=0 (adjacent to a contact) and x=200 μm for the three samples are shown in Figs. 4–6. The following features, all qualitatively similar to TaS\textsubscript{3},\textsuperscript{4} can be seen: (a) At x=0, the response is essentially relaxational. There is a peak in the quadrature response at the same frequency at which the in-phase response falls; the peak height is roughly half the amplitude of the low frequency in-phase response. (b) The relaxation peak moves to lower frequencies with decreasing voltage. (c) At x=200 μm, the response is smaller and slower than at x=0 for each voltage. (d) At high frequencies, the in-phase response becomes inverted, corresponding to a delay in the electro-optical response. This occurs at much lower frequencies for x=200 μm than at x=0. (It is most noticeable at x=0 for sample 1; for sample 3 it is generally out of our frequency window.) (e) For some voltages and positions, the quadrature signal becomes inverted at low frequencies. It is clearest for sample 1, where it occurs in some cases for frequencies >50 Hz. For the other two samples, it only occurs for ω/2π<10 Hz, where noise and the difficulty in determining the electronics phase shift makes this inversion less certain.

The relaxation and delay, and its strong position dependence, can also be seen in time-averaged oscilloscope traces of the transmitted signal (with the chopper turned off), as shown in Fig. 4(c), where for sample 1 a delay of ~70 μs is observed at x=100 μm and V=3.6 V\textsubscript{on}, but the delay is only ~20 μs for the same voltage at x=0. As for TaS\textsubscript{3},\textsuperscript{4} the delayed reversal of transmittance seems to begin “abruptly,” especially at x=0. We do not observe the striking polarity dependence of the delay observed for TaS\textsubscript{3}, however. Also, the delay time increases much more rapidly as one moves away from the contacts for blue bronze than for TaS\textsubscript{3}, indicating that the delay is not, as we suggested in Ref. 4, a contact effect, e.g., due to the formation of Schottky barriers at the contacts,\textsuperscript{11} but is intrinsic to the pinning and repolarization of the CDW.

As shown in Figs. 4–6 we have characterized the relaxation and delay by fitting the complex response to the modified harmonic-oscillator equation\textsuperscript{4}

$$\Delta \theta/\theta_0 = \Delta \theta/\theta_0' [1 - (\omega_0/\omega)^2 + (-i\omega \tau_0)^\gamma]. \quad (1)$$

Here the inertial term is used to parametrize the delays. The exponent γ allows for distributions in ω\textsubscript{0} and τ\textsubscript{0}; in particu-
for overdamped cases $\gamma < 1$ corresponds to a distribution of relaxation times. The low-frequency inverted quadrature signal for sample 1, discussed further below, is not included in these fits.

The voltage and position dependence of the fitting parameters are shown in Fig. 7. For each sample, the relaxation times increase slightly as one moves away from the contact and for each sample/position, the relaxation time is seen to vary as $\tau_0 \sim V^{-p}$, with $p$ between 1 and 2, as for TaS$_3$. We note that while a similar value of $\tau_0 \sim V$ was deduced for the rate of polarization from an unpolarized state, it had a much stronger, exponential, voltage dependence. Note that the onset of CDW current at $V_T$ appears to have no effect on the relaxation time. The resonant frequencies tend to weakly increase with increasing voltage, but much more striking is the rapid increase in resonant frequency, i.e., decrease in inertia as one approaches the contact, with the resonant frequency in some cases exceeding our frequency window. The amplitudes decrease rapidly as one approaches $V_T$; away from the contacts, the amplitudes start decreasing again at high voltage whereas they approximately saturate at $x=0$. Finally, for samples 2 and 3 the exponent $\gamma$ decreases at small voltages as the resonance becomes increasingly overdamped, indicating that the distribution of relaxation times is becoming broad. For example, $\gamma \sim 0.9$ corresponds to a distribution of $\tau$'s approximately a half decade wide.
The dynamics of many properties associated with CDW pinning/depinning are governed by the density of quasiparticles, and hence time constants are thermally activated for semiconducting materials. The dielectric constant, associated with small oscillations of the CDW about its pinning configuration, is a very prominent example. The magnitude of strain for a given voltage, since it depends on screening by quasiparticles, is also expected to be activated, and indeed the magnitude of the electrottransmittance was observed to be activated at low temperatures. On the other hand, in the Cornell model, the rate of change of strain, when reversing the current, is governed not only by the magnitude of the strain (near the contacts) but by the CDW current. It was therefore interesting to study the dynamics of the electrottransmittance at different temperatures to see how these different factors are manifested.

Measurements were made on sample 3 for temperatures between 45 K and 100 K. (The decreasing resistance prohibited measurements at voltages much above threshold at higher temperatures, while the decreasing magnitude of the electrottransmittance prevented frequency dependent measurements at lower temperatures.) To make comparisons of the response at different temperatures, we had to decide on an appropriate voltage criterion. Both the onset and threshold voltages decrease with decreasing temperature, so temperature-dependent comparisons at a constant voltage are not appropriate. (\( V_I/V_{on} \) increases with decreasing temperature) so that most of the pinning becomes associated with the contacts, e.g., see Fig. 1. However, \( V_I/V_{on} \) actually decreases from 8 mV to 3 mV with decreasing temperature for this sample, in striking contrast with suggested thermal activation of the phase-slip voltage.

Figure 7 also shows that the relaxation time does not change as the voltage passes through \( V_T \) (and hence must also have a weak dependence on CDW current here). Therefore, we chose as our criterion fixed values of \( V-I/V_{on} \) approximately proportional to the average driving force on the strain and CDW. (The pinning force, in fact, depends on the voltage and CDW current for small currents.)

Figure 8 shows the temperature dependence of the parameters of Eq. (1) for two driving voltages, \( V=V_{on}+10 \) mV and \( V=V_{on}+20 \) mV, and two positions, \( x=0 \) and \( x=200 \) \( \mu \)m. Also shown is the temperature dependence of the low field, quasiparticle resistance and conductance. For each position/voltage, the magnitude of the electrottransmittance varies roughly as the conductance for \( T<70 \) K and saturates at higher temperature, as we previously observed. In contrast, for each data set the relaxation times and resonant frequencies have a weaker temperature dependence, with activation energies approximately half that of the conductance. However, the CDW current for each voltage, defined as \( I_{CDW} = I_{total} - V/R_0 \), was observed to also have this smaller ac-
tivation energy. We therefore plotted the relaxation times and frequencies vs $I_{CDW}$, as shown in Fig. 9. $I_{CDW}$ is plotted vs voltage in the inset to Fig. 9; for this sample, $I_{CDW} \sim (V-V_T)^{3/2}$. For each voltage/position, $\tau_0$ and $1/\omega_0$ scale roughly as $1/I_{CDW}$; that is, for a given driving force and position, the dynamical rates are proportional to the CDW current.

We should emphasize that this dependence on CDW current only occurs for a given driving voltage; the relaxation rates themselves are not simply functions of $I_{CDW}(T)$, even for large CDW currents. This is shown in Fig. 10, where we plot the fit parameters of Eq. (1) for fixed $I_{CDW}=100 \mu$A. While the temperature dependence of the rates are weaker than for fixed driving voltage, they are still strongly temperature dependent.

To check this dependence of the dynamical rates on driving voltage and CDW current, we did temperature dependent measurements on an additional sample. For sample 4 ($L \sim 960 \mu$m), the fits to Eq. (1) were not as good as for the other samples, especially at the highest and lowest frequencies, but the fits at intermediate frequencies were sufficient to determine relaxation times. As shown in the inset to Fig. 11, the relaxation times for this sample tended to saturate at low temperatures, very different behavior from sample 3. Nonetheless, as shown in Fig. 11, there is still a linear dependence, for each voltage/position, between the relaxation rate and CDW current.

V. FREQUENCY DEPENDENCE OF CONTACT STRAINS

For most samples, the contact strain measured with unipolar square waves above threshold has been too small for frequency dependent measurements. An exception was sample 3, for which the spatial dependence of the unipolar ($V=2V_T$) and bipolar ($V=1.5V_T$) responses at $T=101$ K, $\omega/2\pi=25$ Hz are shown in the inset to Fig. 12. For this sample, the unipolar response disappears away (>200 $\mu$m) from the contacts.

Figure 12 shows the frequency dependence of the bipolar response and both the positive and negative unipolar re-
responses at 101 K, 2V_T, and x=0. The parameters for the fits to Eq. (1) are listed in Table I. Note the following features: (i) The negative unipolar response is larger than the positive, as we previously observed.18 Polarity dependent strains have also been observed in NbSe_3 and are not understood.5,12 (ii) Pronounced relaxation peaks in the quadrature response are not observed for unipolar excitation. In fact, the unipolar response can be fit to Cole-Cole relaxation,29 i.e., Eq. (1) without the resonance term, as shown in the table, but with a very small $\gamma=0.65$, implying a decade wide distribution in relaxation times,24 which effectively washes out the relaxation peak. (iii) Given this wide distribution in $\tau$'s, the differences in relaxation times between the bipolar and unipolar responses is probably not significant. Indeed, it is striking that the “nonpinned,” contact unipolar response is not much faster than the (mostly nonequilibrating) bipolar response. This suggests that changes in the contact strains, as the CDW current is turned on and off, occur through a similar mechanism as oscillations in the bulk strain, as discussed below.

Also shown in Fig. 12 is the bipolar response at x =200 $\mu$m, where the unipolar response is zero. As usual, the striking difference between the response here and at the contact is that the resonant frequency has moved well into our window; in fact the resonance at x=200 $\mu$m is underdamped even though the relaxation time (see Table I) has also increased considerably. In Ref. 8, we speculated that the faster bipolar response at the contact vs the interior might be due to the fact that at the contact the bipolar response comes from both nonpinned and nonequilibrating strains, whereas the response in the interior only comes from nonequilibrating strains. However, the difference between the bipolar and two unipolar responses, also shown in Fig. 12, gives the x=0 nonequilibrating response only. This difference was also fit to Eq. (1) and its parameters listed in Table I. The difference response is only slightly slower (i.e., relaxation time is essentially the same and resonant frequency only slightly smaller) than that of the full bipolar response, showing that the dynamics of the bipolar response is essentially determined by that of the nonequilibrating part of the strain, even at points adjacent to the contact.

VI. SUMMARY AND DISCUSSION

We have used measurements of the changes in infrared transmittance when square-wave voltages are applied to the sample to determine the position, voltage, and frequency dependence of the electrotransmittance (v=820 cm⁻¹) of sample 3 at V=2V_T=3.1V_{on} and T=101 K. Shown are the measured in-phase and quadrature responses for bipolar square-waves at x=0 and x=200 $\mu$m and positive and negative unipolar square-waves at x=0. Also shown is the difference between the x=0 bipolar and (difference of the) unipolar responses. The curves are fits to Eq. (1). Inset: The spatial dependence of the (in-phase) bipolar (V=1.5 V_T) and negative unipolar (V=2 V_T) responses at 25 Hz, for which the quadrature responses are negligible.

TABLE I. Comparison of Eq. (1) fit parameters (see Fig. 12) for different square waves with V=2V_T=3.1V_{on} for sample 3 at T =101 K.

<table>
<thead>
<tr>
<th>Square-wave/position</th>
<th>$\Delta(\theta/2\pi)$</th>
<th>$\tau_0$ (ms)</th>
<th>$\omega_0/2\pi$ (kHz)</th>
<th>$\gamma$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bipolar, x=0</td>
<td>1.05%</td>
<td>0.15</td>
<td>4.70</td>
<td>0.98</td>
</tr>
<tr>
<td>+unipolar, x=0</td>
<td>0.15%</td>
<td>0.053</td>
<td>$\infty$</td>
<td>0.65</td>
</tr>
<tr>
<td>-unipolar, x=0</td>
<td>-0.23%</td>
<td>0.13</td>
<td>$\infty$</td>
<td>0.65</td>
</tr>
<tr>
<td>Bipolar, x=200 $\mu$m</td>
<td>0.49%</td>
<td>0.46</td>
<td>0.55</td>
<td>0.94</td>
</tr>
<tr>
<td>Bipolar, x=200 $\mu$m, -(+uni.)</td>
<td>0.70%</td>
<td>0.16</td>
<td>2.92</td>
<td>1.06</td>
</tr>
</tbody>
</table>

pendence of CDW strains (i.e., phase gradients) in blue bronze. This technique has the advantage of being able to probe the interior of the sample without placing multiple probes, which can perturb the CDW, on the sample, which has hindered transport measurements of the CDW strain in semiconducting CDW materials.11

Of course, it is still necessary to place contacts on the ends of the sample, and the largest changes in dynamical properties occur near (~100 μm) these contacts. Important questions, therefore, are to what extent these contacts are equipotentials with minimal band bending, e.g., due to formation of Schottky barriers in the CDW state,11 and to what extent current enters the sample from the edges of the contacts and quickly distributes through the cross section. The very small contact strains observed for some samples and the fact that the bulk strain, while varying approximately linearly with position in the sample, is not symmetric on the two sides of the sample with a voltage dependent asymmetry (e.g., see Fig. 3) certainly suggest that our contacts are “imperfect.” Indeed, the longitudinal length scale with which current is expected to spread through the sample cross section, calculated from the contact resistances, is11 λ ~ dη, where η is the ratio of longitudinal and transverse conductivities. For blue bronze, η ~ 1000,20 making λ comparable to the length scale of measured changes at the contacts.

However, the fact that the electrotransmittance (probing the whole sample cross section) and the electroreflectance (probing only the <2 μm penetration depth16,17) have the same spatial and frequency dependence (Fig. 2) suggests that current spreading is not a significant problem for our contacts. (Note that the spatial dependence of the electroreflectance is different for thicker samples.18) Two possible reasons are that sample defects effectively distribute the current through the cross section in a distance shorter than λ or that, because the contacts are over 100 μm long, the current actually spreads below them. The rapid variation of our measured relaxation times and resonant frequencies near the contacts suggests that the first effect is dominant. In fact, the expected length scale for the contact strain is determined by the single particle diffusion length and is expected to be ~100 μm,7 consistent with our measurements. We therefore assume that CDW current is approximately injected from the edges of our contacts so that the relatively small contact strains we measure are not artifacts of poor contacts but intrinsic, e.g., due to the incoherence of the CDW and/or strong pinning of dislocations, discussed below.

Most of our measurements were for the oscillating response to symmetric bipolar square-wave voltages, so that the CDW strain is oscillating between two opposite configurations. In this case, the oscillating strain has contributions from both the bulk polarization, which does not decay in zero field, and a nonpinned strain associated with current conversion, but the latter is small and does not significantly affect the overall frequency dependence.

For large voltages, the electro-optic response can be fit as a damped harmonic oscillator, with the resonant frequency corresponding to a delay with respect to the applied square-wave voltage. The delay times increase rapidly between x =0 (the contact) and x=100 μm and then continue increasing (by 50–100 %) between x=100 and 200 μm. This spatial variation indicates that the delays are intrinsic to the CDW (i.e., not associated with contact barriers14) and suggests that the signal driving the strain relaxation effectively flows out of the contacts. In this case, the delays we observe near the contacts may be a consequence of our finite spatial resolution. Similarly, in their measurements on NbSe3, the Cornell group found that there was a delay ~10 μs for changes in the electric field, and therefore the strain, in the center of the sample, but no delay at the contacts, and simulated these effects in terms of a strongly strain (and therefore position and voltage) dependent phase-slip rate, presumably reflecting the pinning and motion of dislocation lines.21 In our case, the resulting typical (T~80 K) propagation velocity of ~100 μm/100 μs is comparable to that observed for voltage pulses30 but orders of magnitude larger than the drift velocity of the CDW,21 whose motion is limited by scattering with quasiparticles.1 If we model the CDW strain propagation as a wave on a stretched wire with tension (per electron) ~e(V-V_{cdw})/L~10 eV/m (consistent with the observed ω0 ~ V^1/2 behavior at large voltages), then a velocity of 1 m/s corresponds to a reasonable effective mass density for the strain wave of ~1 QM_F, where Q is the CDW wave vector and M_F is the Fröhlich mass [~300m_e Ref. 31] associated with CDW motion.1 Of course, this simple result should only be considered order of magnitude and needs to be qualified to account for the strong sample and temperature dependence of the delays.

The relaxation time also increases as one moves away from the current contact. For any position in the sample, the average relaxation time τ_0 ~ V^p, with p between 1 and 2. No divergence or other structure is observed in τ_0 near V_T, where the CDW is depinned at the contacts and dc CDW current can flow. The dynamics are governed both by the force on the CDW (e.g., V-V_{cdw}) and the CDW current. For a given force and position in the sample, both the relaxation and delay times are inversely proportional to the CDW current as temperature is varied. The temperature dependent screening of the quasiparticles directly affects the amplitude of the electro-optic response,15,16 but it only appears to affect the dynamics through its influence on the CDW current, so that while the response slows with decreasing temperature, it does not slow as much as expected from the quasiparticle density.

Near V_T at T ~80 K, the typical relaxation time is 1–10 ms, more than three orders of magnitude greater than the dielectric response time governing small amplitude oscillations of the pinned CDW,22,23 indicating that repolarization requires large scale rearrangements of the CDW and suggesting that the strain relaxation involves the motion of extended defects in the CDW. In the model of Ref. 6, CDW polarization occurs essentially through continuous and gradual changes in CDW phase. However, this may be a coarse-grained average of a more complicated phase landscape in which regions in which CDW has its equilibrium wave vector are separated by localized, solitonlike defects.32,33 These presumably accumulate on neighboring chains to form extended defects, such as CDW phase dislocation loops.32,34 As mentioned above, in Ref. 7 the pinning of the dislocations was considered to be the cause of persistence of the bulk strains when V=0. If so, polarization current would require
the lateral motion of these defects along the conducting
chains, e.g., glide of the dislocations, in contrast to the
growth of the loops by climb perpendicular to the chains,
responsible for phase slip. As mentioned above, a simi-
lar mechanism would hold for the changes in the contact
strains, but the wider distribution of relaxation times might
indicate a broader distribution of dislocation loop sizes near
the contacts where phase slip is also occurring.

For low voltages, the simple damped harmonica fits break
down (for most samples), and the response requires a distrib-
tion of time constants, which we have parametrized with
the exponent $\gamma$. This may reflect inhomogeneous CDW
pinning (and current, when above $V_T$) on a length scale much
smaller than our typical 50 $\mu$m light spot. As mentioned
above, this broadening may mask any possible dynamic criti-
cal slowing down at $V_{on}$. However, sample 1 did not exhibit
this broadening, at least for $V \sim V_T$, and it showed some
evidence of critical behavior at lower voltages; unfortunately,
the sample broke before the low voltage range could be in-
vestigated well. Since the presence of dynamic critical be-
havior at depinning has long been an open question that has
generated much interest, we will continue measurements
on crystals at low voltages; our electro-optic probe may al-
low us to avoid some of the problems encountered with
transport measurements.

In conclusion, we have used infrared electrotransmittance
to probe the dynamics of CDW repolarization. Long (milli-
second) relaxation and delay times suggest that the response
involves the motion of macroscopic objects, presumably
CDW phase dislocation lines; this appears to be true for the
contact strains driving phase slip as well as the bulk polar-
ization. Most striking is the growth in the delay time with
position away from the contacts. The temperature depend-
ence of the time constants suggests that quasiparticles in-
fluence the dynamics of polarization only through their effect
on the CDW current. In future work, we will attempt to
generalize the models of Refs. 6 and 7 to accommodate
seemingly dependent quasiparticle densities, to more quantitatively
account for some of these observations.

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