

# Electro-Optical Studies of Charge-Density-Wave Conductors

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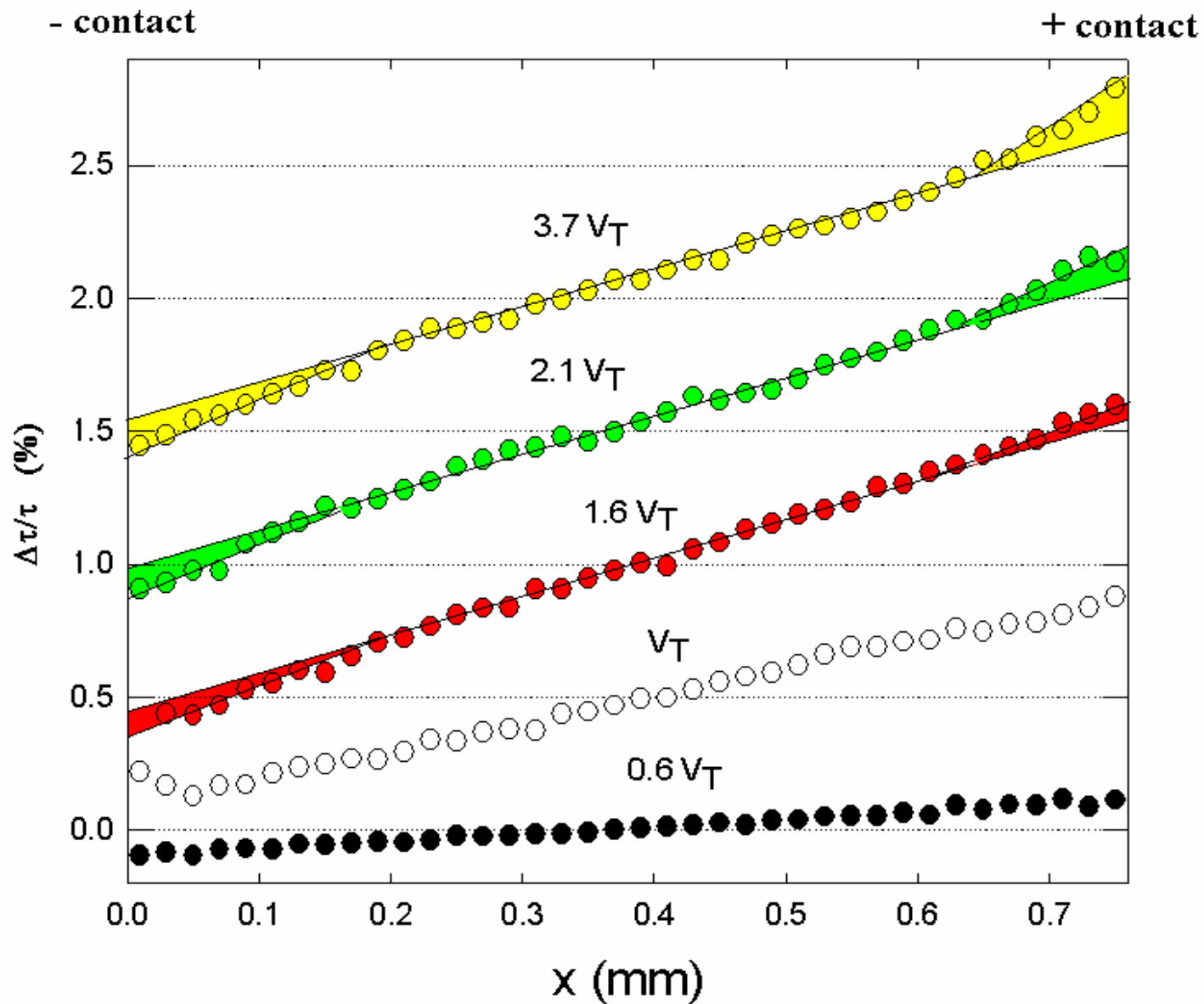
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In 1994, we reported that, for energies below the charge-density-wave (CDW) gap ( $2\Delta \sim 1300 \text{ cm}^{-1}$ ), the IR transmission ( $\tau$ ) of the quasi-one-dimensional conductor “blue bronze” ( $\text{K}_{0.3}\text{MoO}_3$ ) changes in an applied electric field [1]. The changes are thermally activated and vary with position in the sample: the transmission increases on the positive side of the sample and decreases on the negative. We associated these changes with changes in the intraband absorption of quasiparticles, whose density changes to screen deformations of the CDW in the applied field:

$$\Delta\tau/\tau \propto \Delta n_{\text{qp}} \propto \partial\phi/\partial x \equiv \Delta Q, \text{ where } \phi \text{ and } Q \text{ are the CDW phase and wavevector .}$$

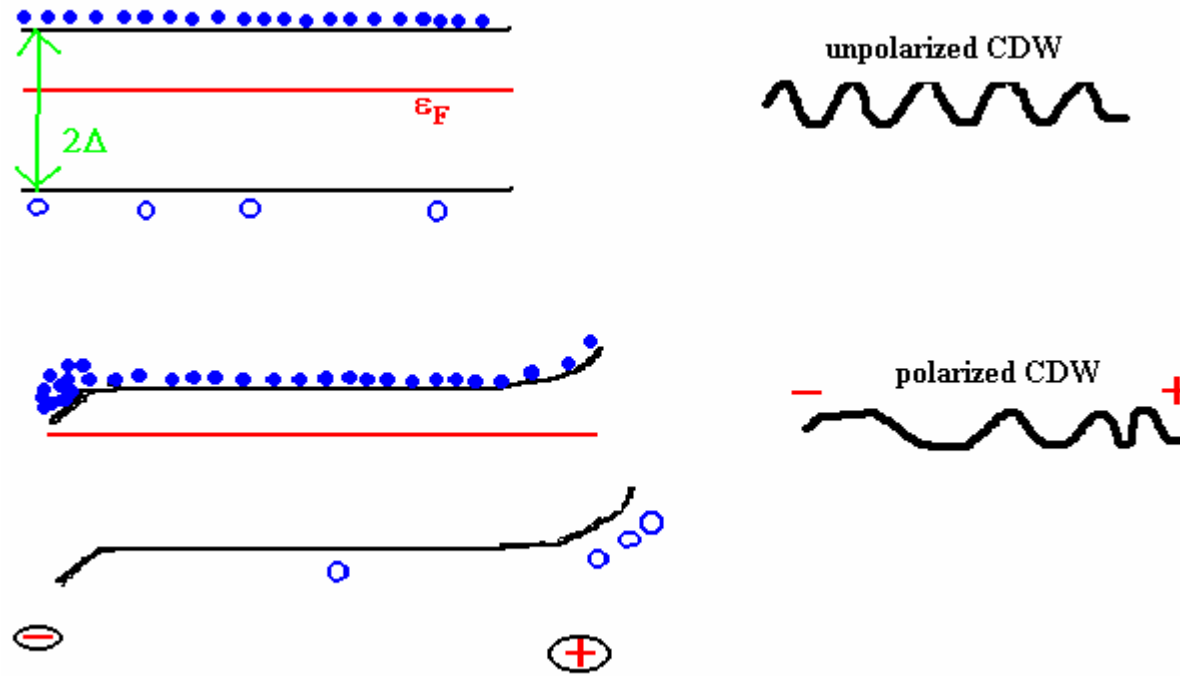
*Figure 1* shows the spatial dependence of the relative change in transmission ( $\Delta\tau/\tau = -\Delta \ln \tau$ , where  $-\ln \tau$  is the optical density) for a crystal of blue bronze at different voltages, referred to  $V_T$ , the threshold for nonlinear current caused by CDW depinning and sliding [2]. Samples were typically 1 mm x 0.3 mm x 10  $\mu\text{m}$  in dimensions, and were measured at  $T \sim 100$  K with a broadband IR source. Bipolar square waves ( $\pm V$ ) were applied to the sample and the local changes in the transmitted intensity at the square-wave frequency ( $\Delta\tau$ ) were measured. The spatial dependence of  $\Delta\tau/\tau$  is very similar to that of the CDW phase gradient determined in  $\text{NbSe}_3$  by transport [3] and x-ray [4] measurements, and support our identification of the modulation with the phase gradient. Typically:  $\Delta Q \sim Q / 3000 \sim 10^{-4} \text{ A}^{-1}$ . Note that for  $V \leq V_T$ ,  $\Delta\tau/\tau$  varies  $\sim$  linearly with position; this corresponds to bulk polarization of the CDW. For  $V > V_T$ , there are extra deformations near ( $\sim 100 \mu\text{m}$ ) the contacts, shown by the shading. These grow logarithmically with the CDW current [2]:

$$\Delta\tau/\tau)_{\text{contact}} \propto \ln I_{\text{CDW}} \Rightarrow I_{\text{CDW}} \propto \exp(\Delta Q/\Delta Q_0)_{\text{contact}}.$$



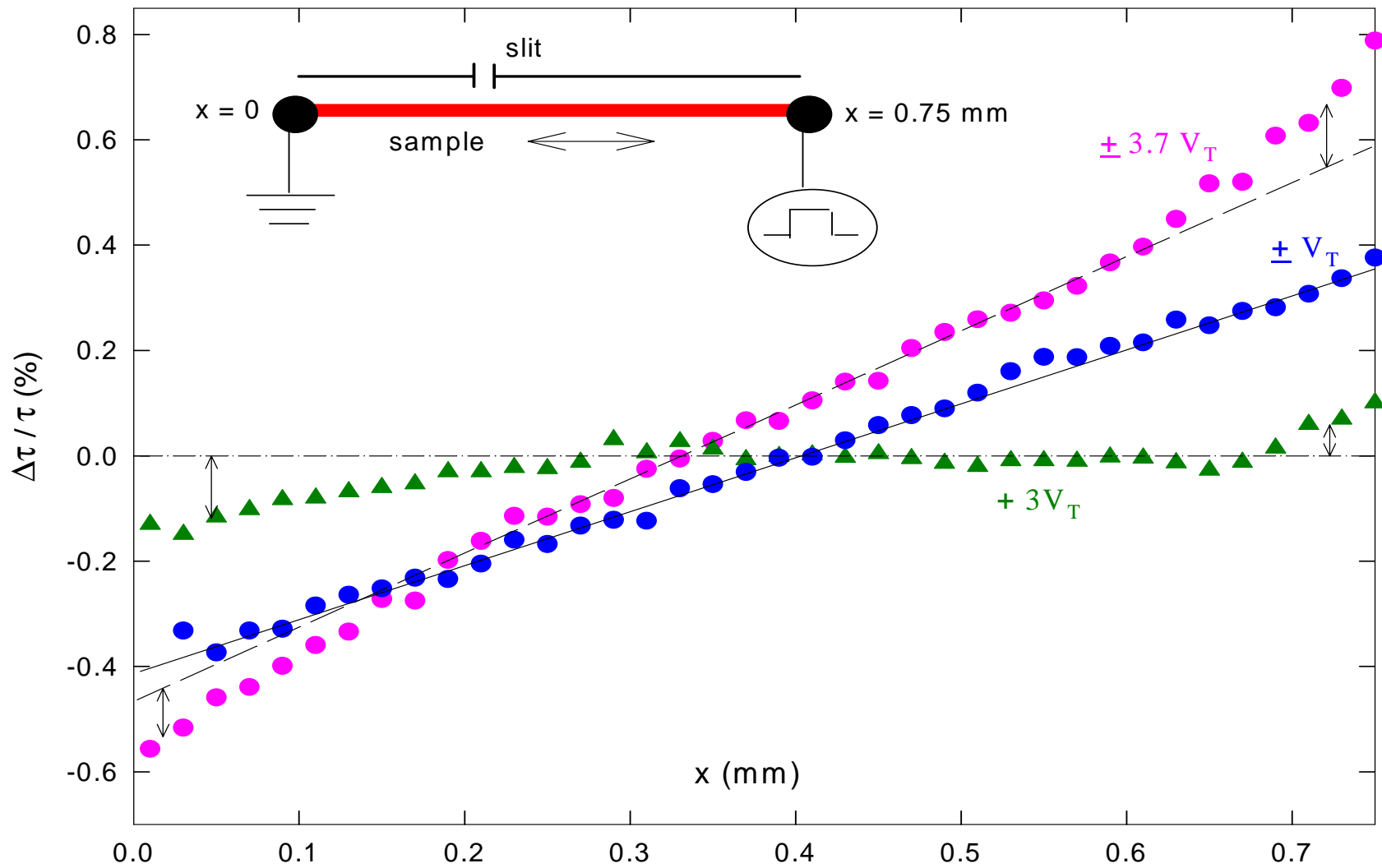
**Figure 1.** Spatial dependence of relative change in transmission at different voltages; successive curves are offset by 0.5%.

**Figure 2** shows a cartoon of how a CDW phase gradient will bend the bands around the CDW gap, causing changes in quasiparticle density. Note that from the Hall effect of blue bronze, we know that the majority of mobile quasiparticles are electrons; their compression (*rarefaction*) at the negative (*positive*) contact is consistent with the decrease (*increase*) in transmission there.



**Figure 2.**

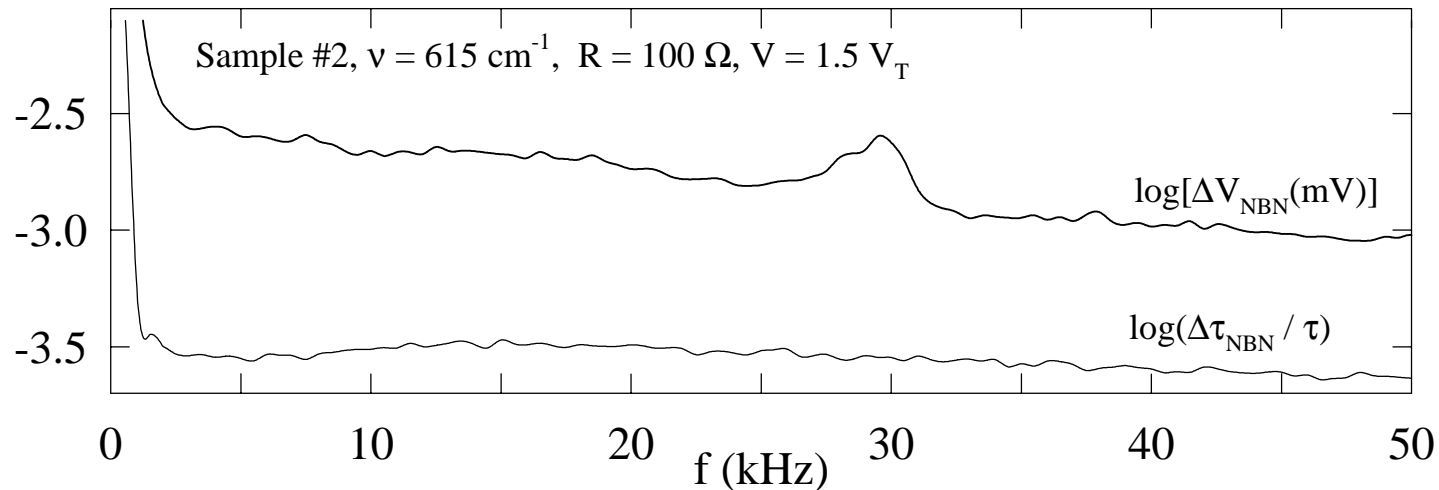
We used this electro-optic effect to study the dynamics of the CDW phase response to applied voltages. We found [2] that the **bulk CDW polarization** (i.e. linear deformation in center) is very sluggish, with a response time  $\sim 1$  ms when the CDW current is reversed. Furthermore, if the applied voltage is only turned off, and not reversed, the bulk polarization remains. However, the **contact deformations**, which are responsible for driving the non-linear currents at  $V > V_T$  [3], change “instantaneously” with changes in voltage. These features are illustrated in *Figure 3*, comparing the spatial dependence of changes in IR transmission (at the square wave frequency) for applied bipolar and unipolar square waves.



**Figure 3.** Spatial dependence of the oscillating change in transmission for bipolar and unipolar square wave voltages.

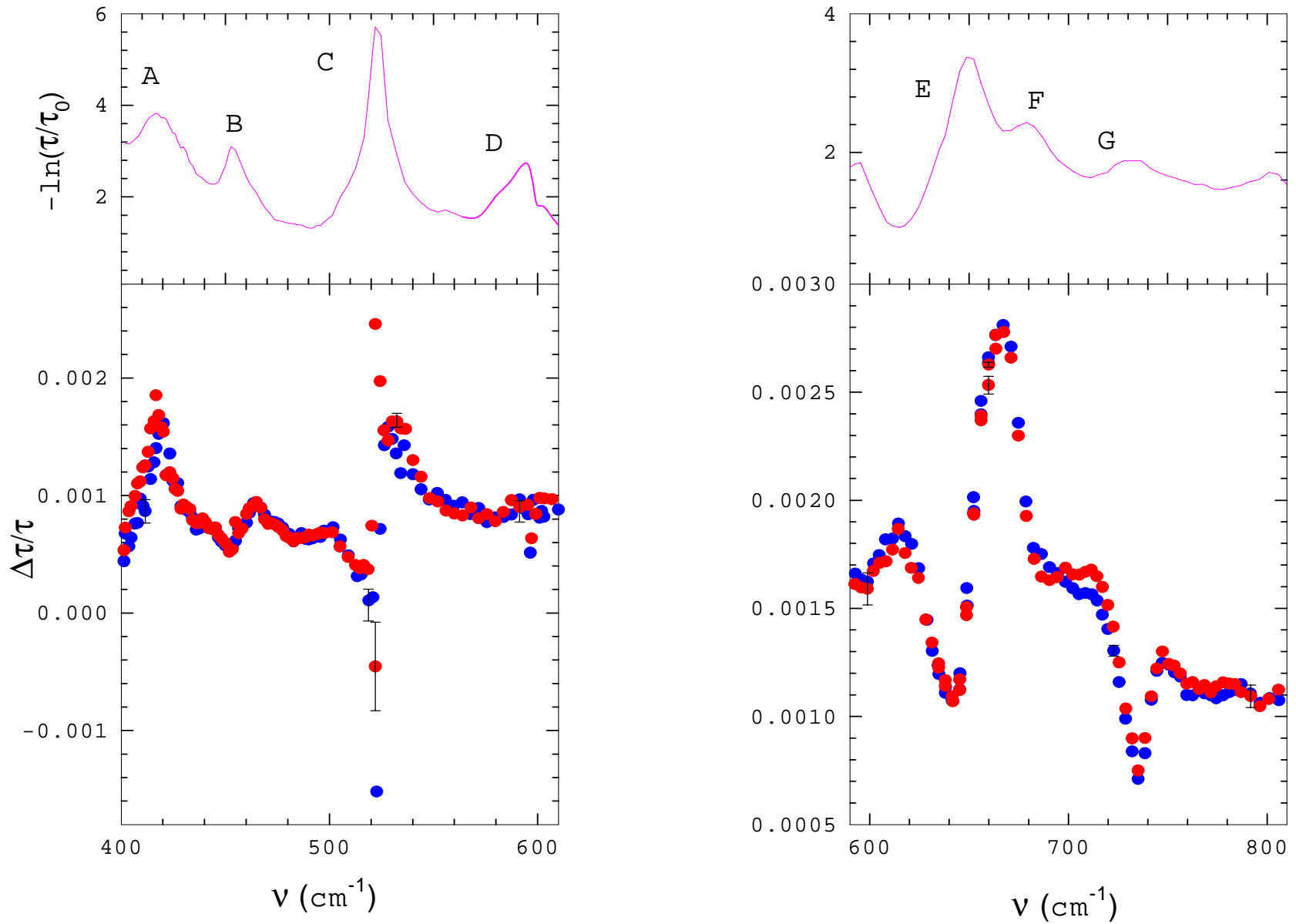
One of the most prominent attributes of a sliding CDW is the presence of “narrow-band-noise”: voltage oscillations at a frequency proportional to the CDW velocity:  $f = Qv_{\text{CDW}}/2\pi$ . These are thought to be generated inhomogeneously in the sample, in which case the condensate and quasiparticle densities would also locally oscillate at  $f$ . Hence we looked, *with negative results*, for oscillations in the IR transmission, as shown in **Figure 4**. From these negative results, we find that the CDW velocity coherence length  $> 1 \mu\text{m}$ , comparable to the measured phase coherence length [5].

**Figure 4.** Fourier transforms of the voltage across a blue bronze sample and its IR transmission at  $T = 80 \text{ K}$ .

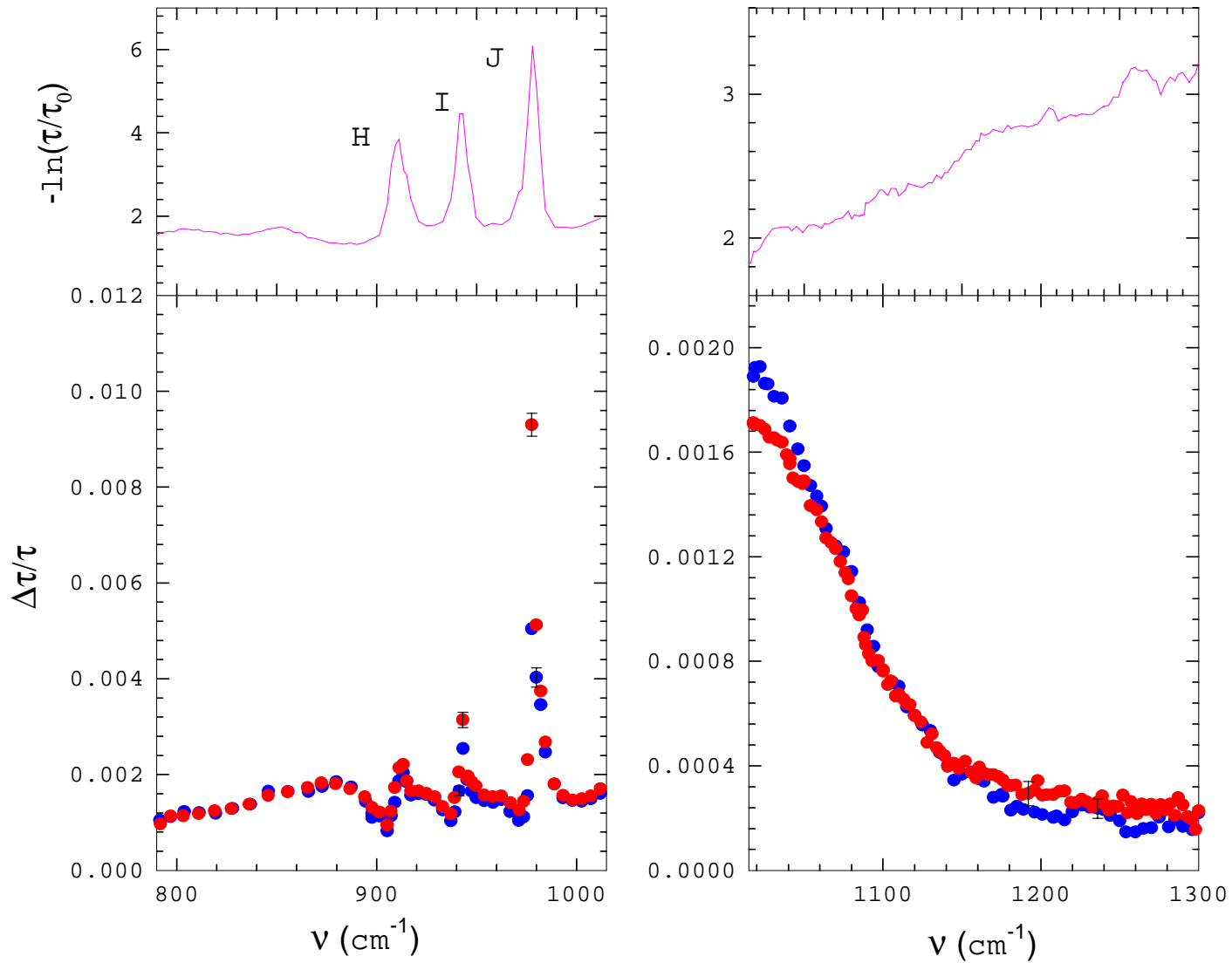


We have investigated the spectra associated with this electro-optic effect [6], with the initial goal of trying to observe intragap electronic states associated with CDW current conversion; short-lived “ $\pi$ -solitons” near the center of the gap and longer lived “ $2\pi$ -solitons” near ( $\sim 100 \text{ cm}^{-1}$ ) the gap edge, which condense into CDW dislocation loops [7]. For such measurements, we wanted to compare the  $\Delta\tau/\tau$  spectra associated with current conversion (e.g. measured with a unipolar square wave voltage  $V > V_T$ , as shown in *Figure 3*) with the bulk polarization (e.g. bipolar,  $\pm V_T$ ) spectra. For sources, we used **tunable infrared diode lasers**. Typical results, taken on a  $\sim 3 \mu\text{m}$  thick crystal at  $T = 82 \text{ K}$ , are shown in *Figure 5* and *Figure 6*; **blue** symbols are bulk spectra ( $V = \pm V_T$ ) while **red** symbols show the unipolar spectra ( $V = +3.2 V_T$ ) (times a normalization factor = 2.45). Also shown in **pink** are the optical density ( $-\ln \tau$ ) spectra at the same temperature. (*Note that even for this thin sample, only transmission for light polarized **transverse** to the conducting chains could be measured.*)

**Figure 5.** Optical density (pink) and relative changes in transmission for energies between 400 and 800  $\text{cm}^{-1}$  at  $T = 82$  K. **Blue:**  $V = \pm V_T$  (corresponding to bulk spectra); **red:**  $V = + 3.2 V_T$ , (contact spectra).



**Figure 6.** Optical density (pink) and relative changes in transmission for energies between 800 and 1300  $\text{cm}^{-1}$  at  $T = 82$  K. **Blue:**  $V = \pm V_T$  (corresponding to bulk spectra); **red:**  $V = + 3.2 V_T$ , (contact spectra).



*No new states* are observed in the contact spectra, implying that  $n\sigma/\Gamma < 10^{-10} (\text{Å cm}^{-1})^{-1}$  [6], where  $\sigma$ ,  $\Gamma$ , and  $n$  are the cross-section, linewidth, and density of soliton states. Assuming that  $\Gamma \sim k_B T_c \sim 120 \text{ cm}^{-1}$  and  $\sigma(\text{transverse polarization}) \sim \sigma(\text{parallel polarization}) \sim 100 \text{ Å}^2$ , this implies that soliton excitations exist on  $< 1\%$  of the chains at any time. This, in turn, has the surprising implication that the  $2\pi$ -soliton/dislocation states have optical lifetimes  $< 1\%$  of the narrow-band-noise period.

However, *striking changes are observed in the phonon* spectra; these changes are identical in the **bulk** and **contact** spectra, and indicate that the changes are proportional to the CDW phase gradient and independent of its source. Changes include oscillator strength changes (phonon A: strength 0.07% larger at the negative contact than at the positive), frequency shifts (phonons C, E, H, I, J; typical shifts of  $0.01 \text{ cm}^{-1}$ , with the frequency decreasing at the positive contact and increasing at the negative), and linewidth changes (phonons B, G, D: typical change of  $0.01 \text{ cm}^{-1}$ , with the line broadening at the negative contact and sharpening at the positive). The linewidth changes can be understood in terms of changes in damping by quasiparticles (and a similar change was observed in a Raman line [8]), but the mechanisms for changes in oscillator strength and frequency are unclear.

In present measurements, we are using an IR microscope, in conjunction with the lasers, to better combine imaging and spectroscopic measurements. The lasers will also allow us to study other CDW conductors, e.g. TaS<sub>3</sub> and NbSe<sub>3</sub>, for which available crystals are much narrower (e.g. ~ 20 μm) than those of blue bronze. Preliminary results on TaS<sub>3</sub>, which has a prominent 500 cm<sup>-1</sup> absorption line that has been associated with a midgap soliton [9], indicate that its transmission varies with voltage in a manner similar to that of blue bronze. Use of the microscope will also allow us to search for electromodulated reflectivity, and thereby study changes for light polarized **parallel** to the conducting chains in these quasi-one-dimensional conductors.

## References

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