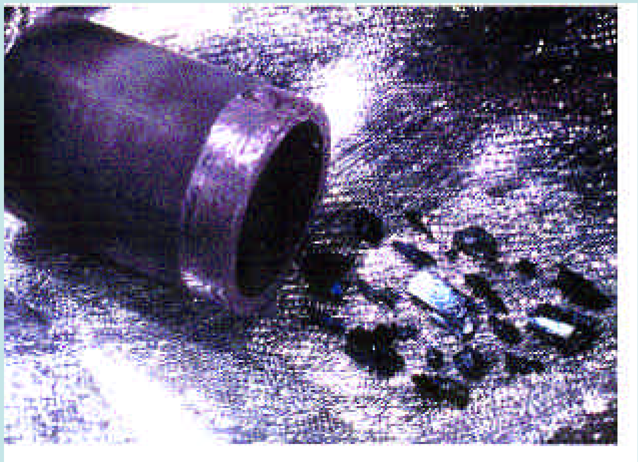
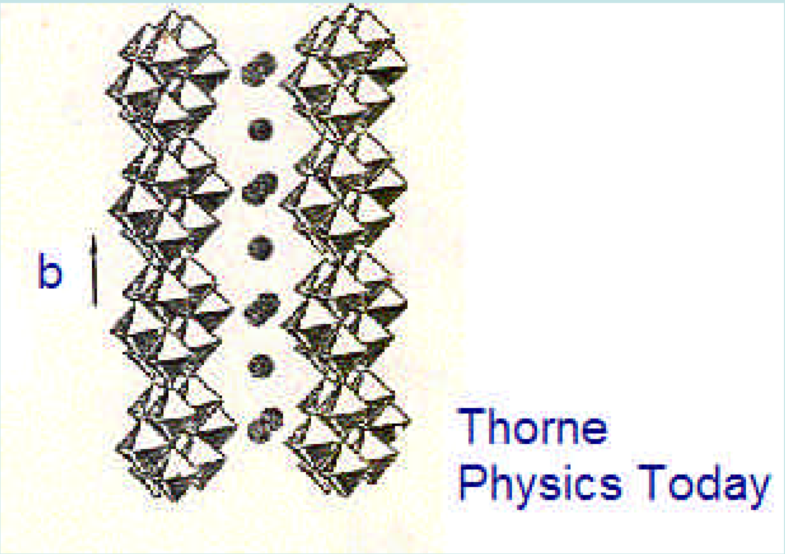
Microscopic Electro-Optical Studies on Blue Bronze, a Charge-Density-Wave Conductor

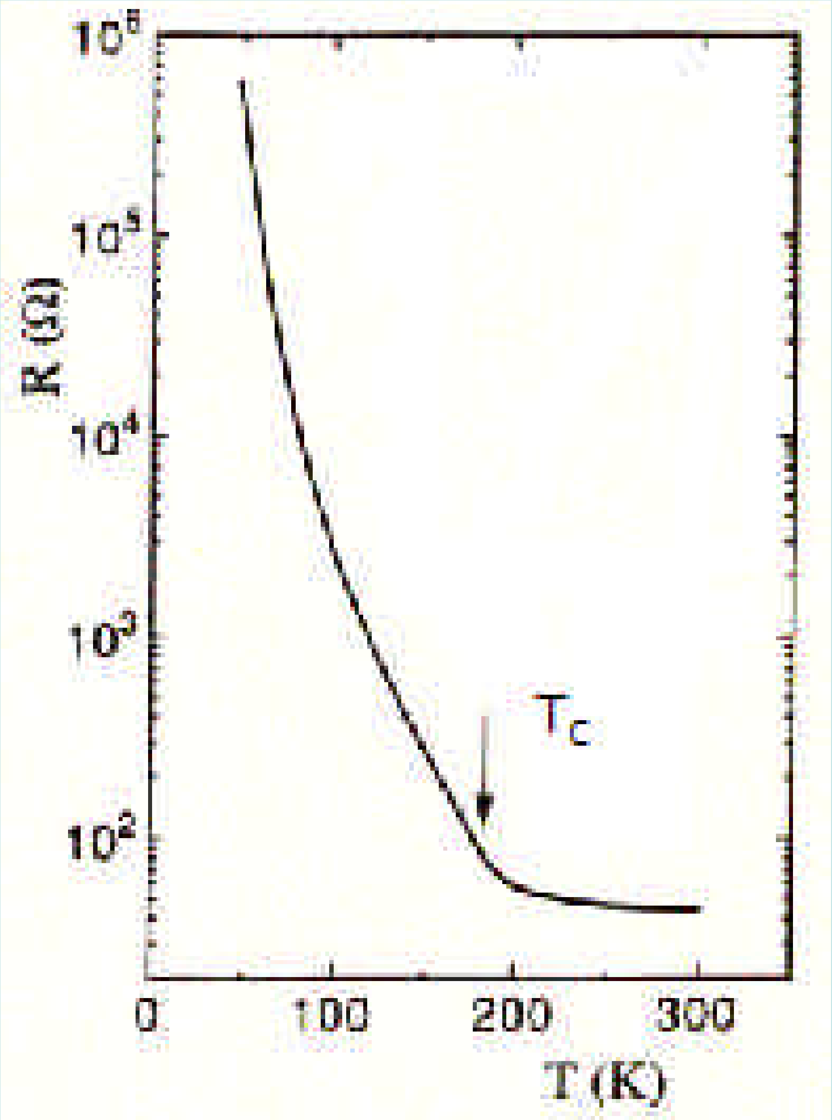
J.W. Brill, R.C. Rai, B.M. Emerling,
M.E. Itkis, and V.A. Bondarenko
University of Kentucky

Crystals from R.E. Thorne, Cornell University

Blue Bronze ($K_{0.3}MoO_3$), a quasi-one dimensional conductor undergoing T_c = 180 K metal-insulator transition







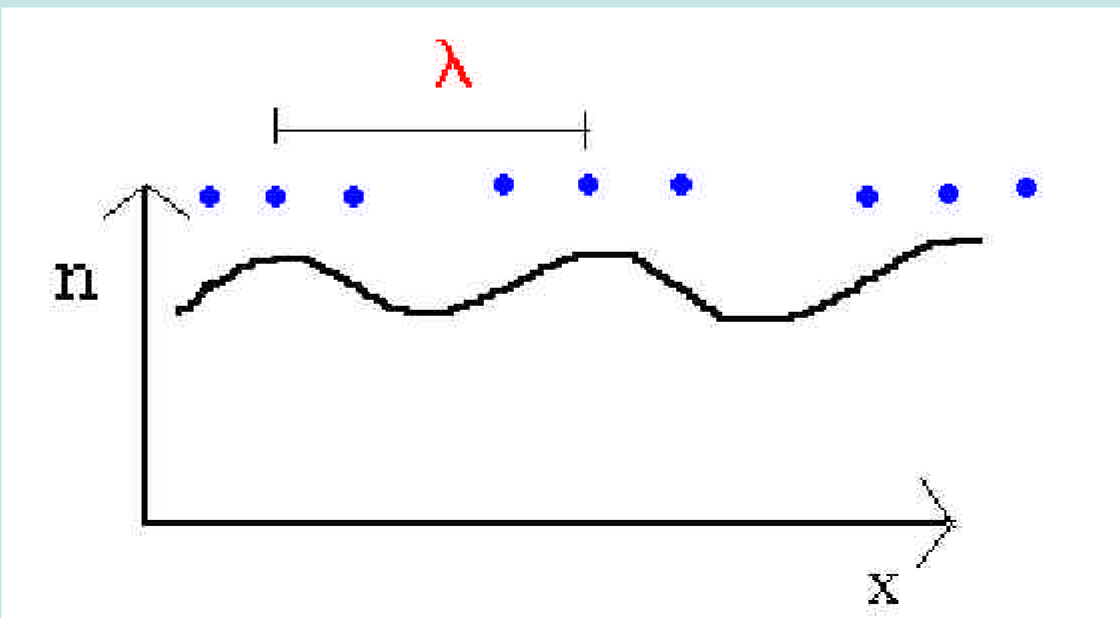
Charge Density Wave (CDW)

Periodic Lattice (Peierls) Distortion: x_j = ja + dx cos(2$k_F$ja+ ö)

modulation of electron density (CDW): n = n_0 + dn sin(2k_Fx + ö)

and gap on Fermi surface (~ 1200 cm^{-1} in blue bronze)

semiconducting behavior.



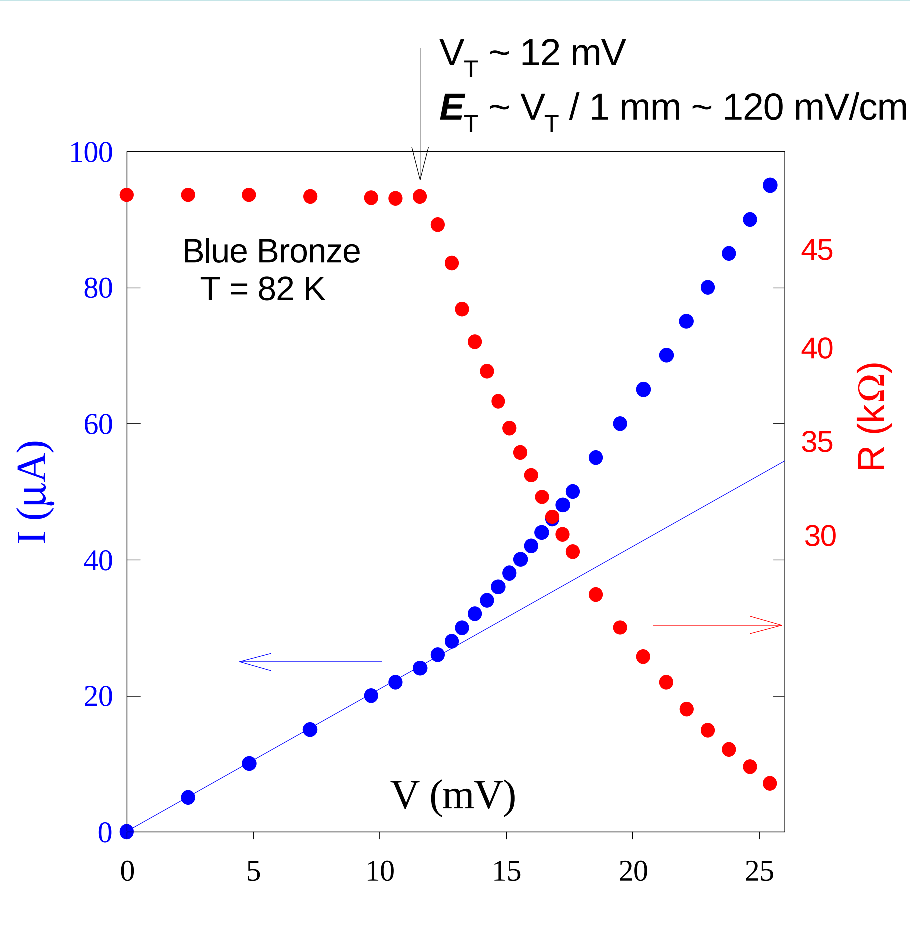
T_c=180 K,

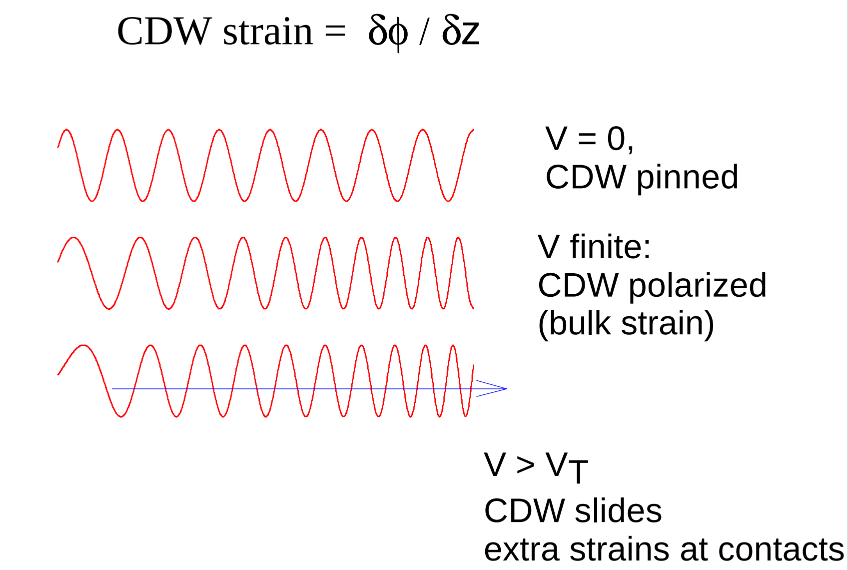
l = 2p/2k_F ~ 10

Typical: dn/n < 1%,

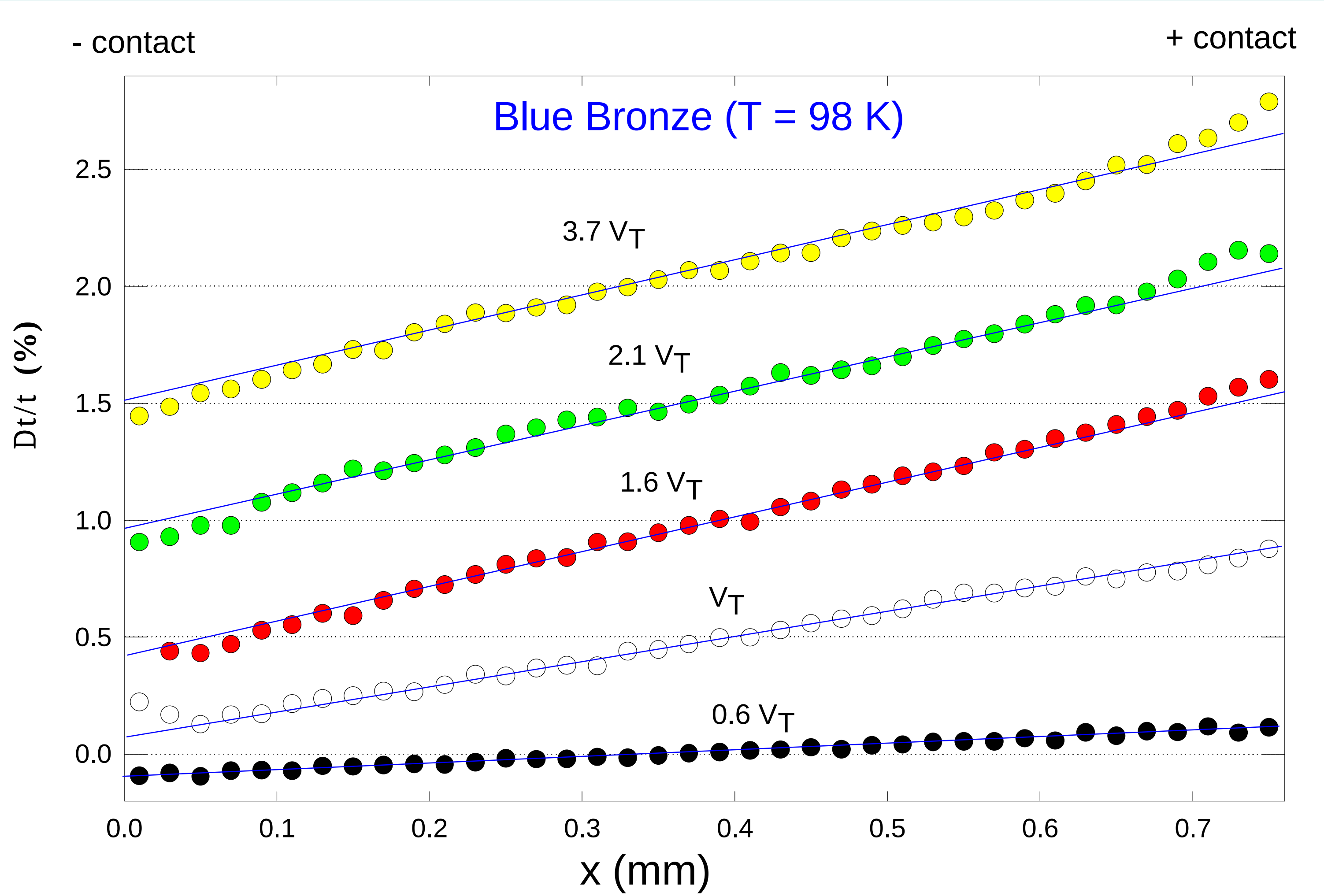
dx < 0.1

Incommensurate CDW is pinned to lattice by impurities.
Application of V > V_T (threshold voltage)
Polarization and Depinning (sliding) of CDW:
Non-Ohmic Conductivity





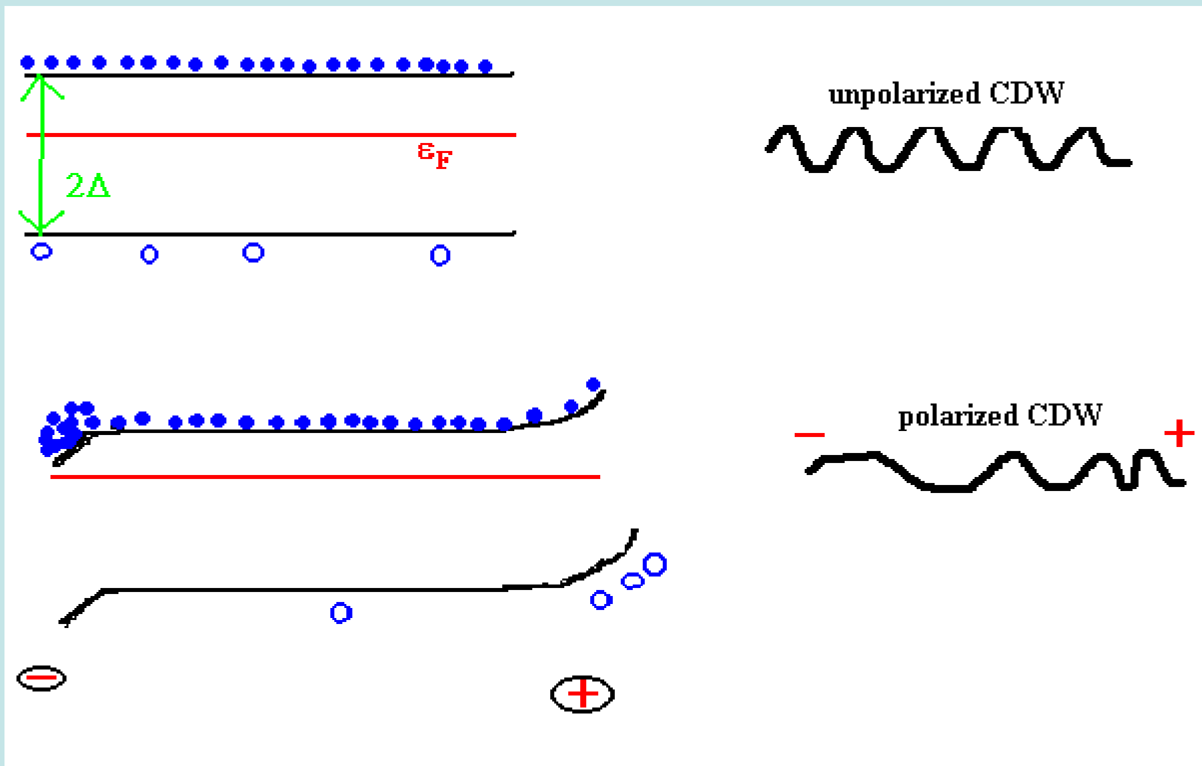
Spatial variation of infrared ($\nu < 2\Delta \sim 1200$ cm^{-1}) transmission *(polarized ^ chains, t ~ 1%)* with applied voltage near/above threshold



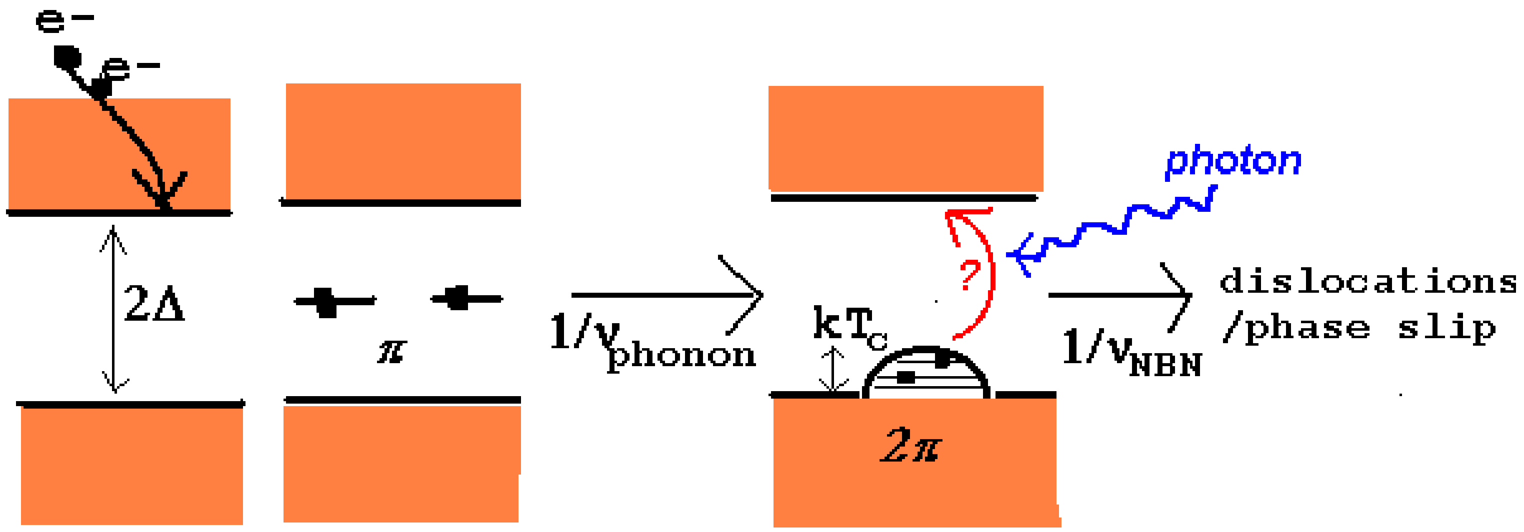
Model: Changes in transmission due to intraband absorption of thermally excited electrons, which screen CDW deformation

($\Delta\tau/\tau \sim \Delta n \sim$ äö/äx)

Extra strains at contacts needed to drive current conversion into CDW



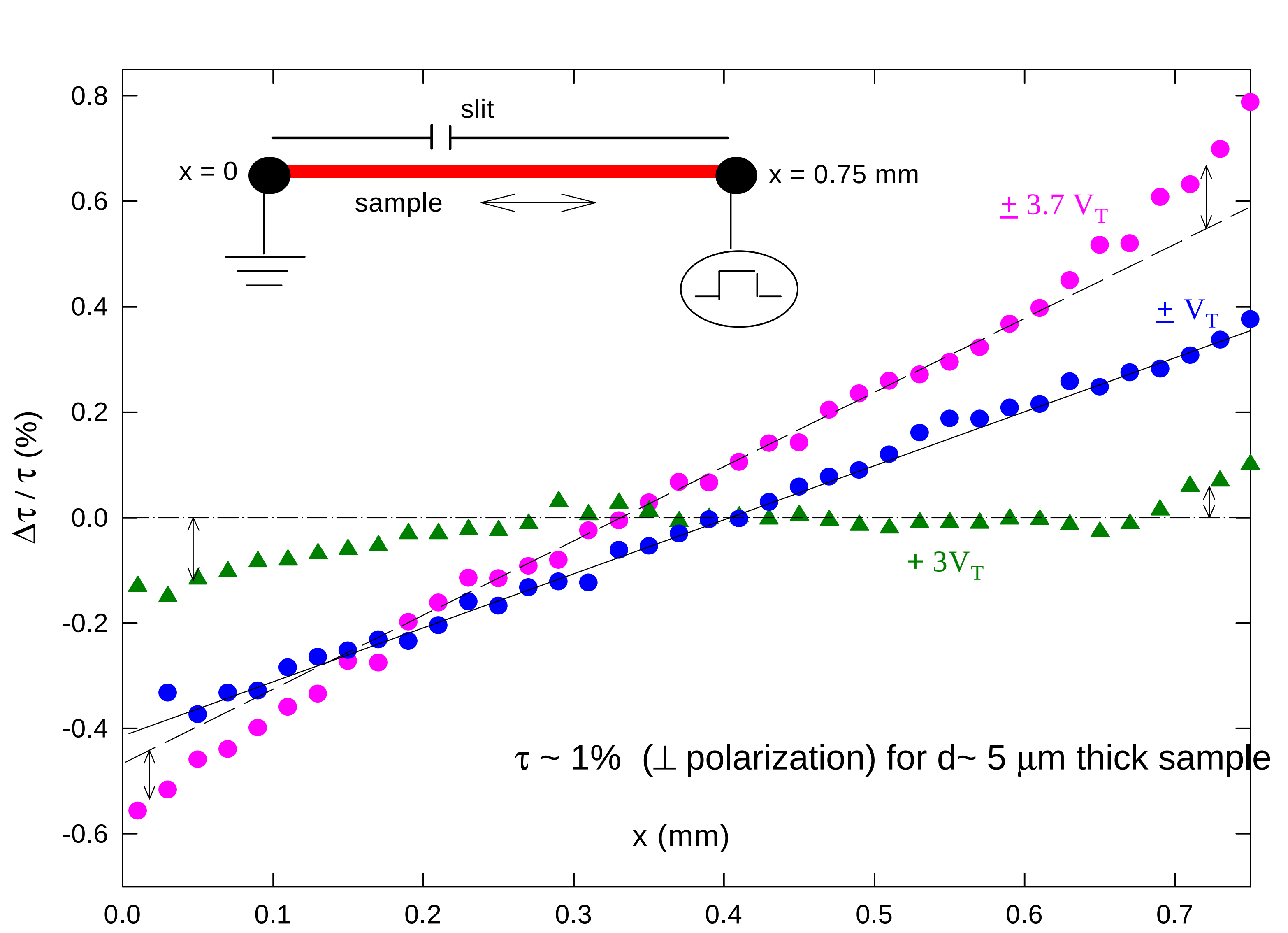
Lifestory of charge injected into CDW (*Brazovski*)



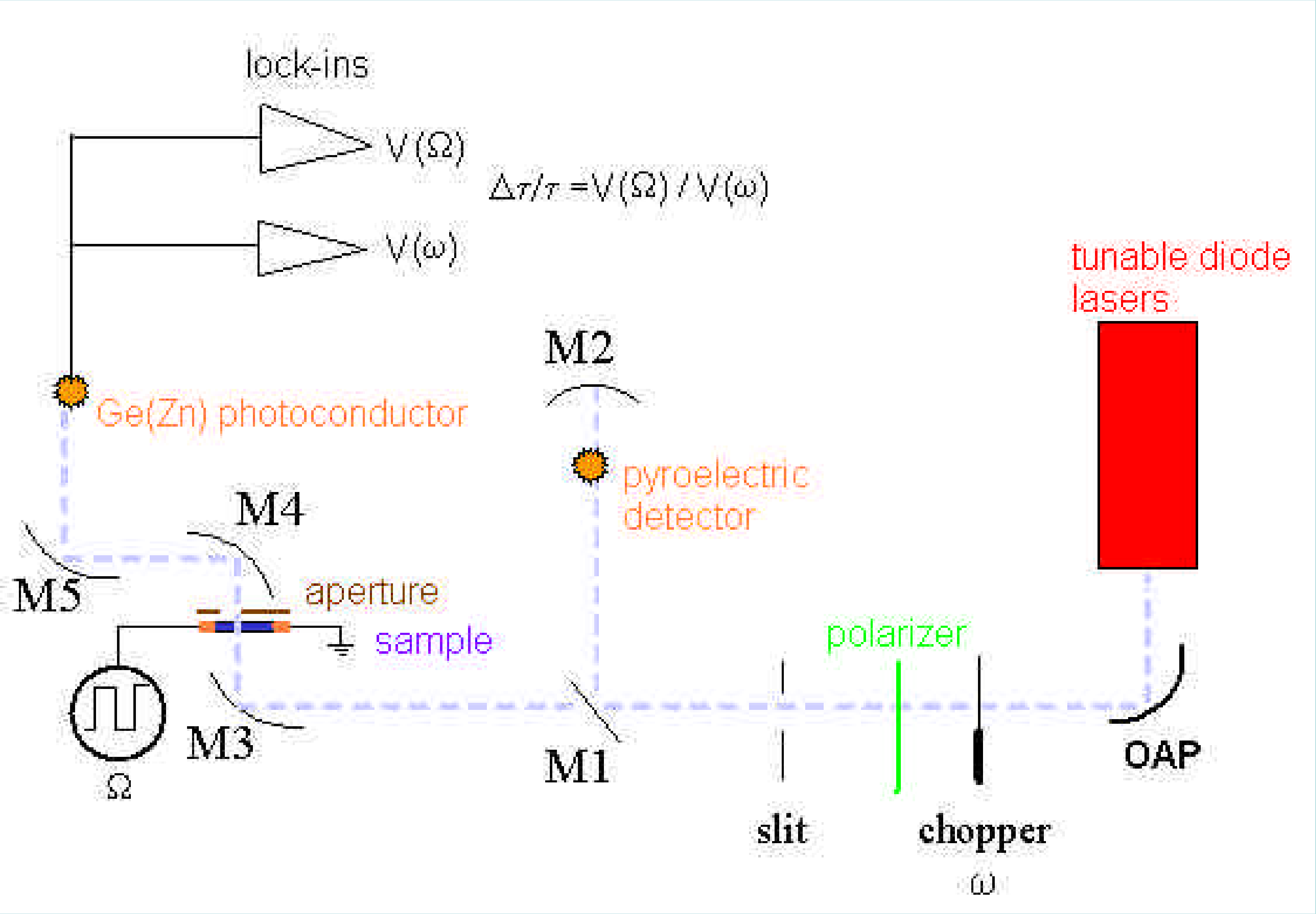
?? Can energy of phase (2π) soliton be measured with optical spectroscopy??

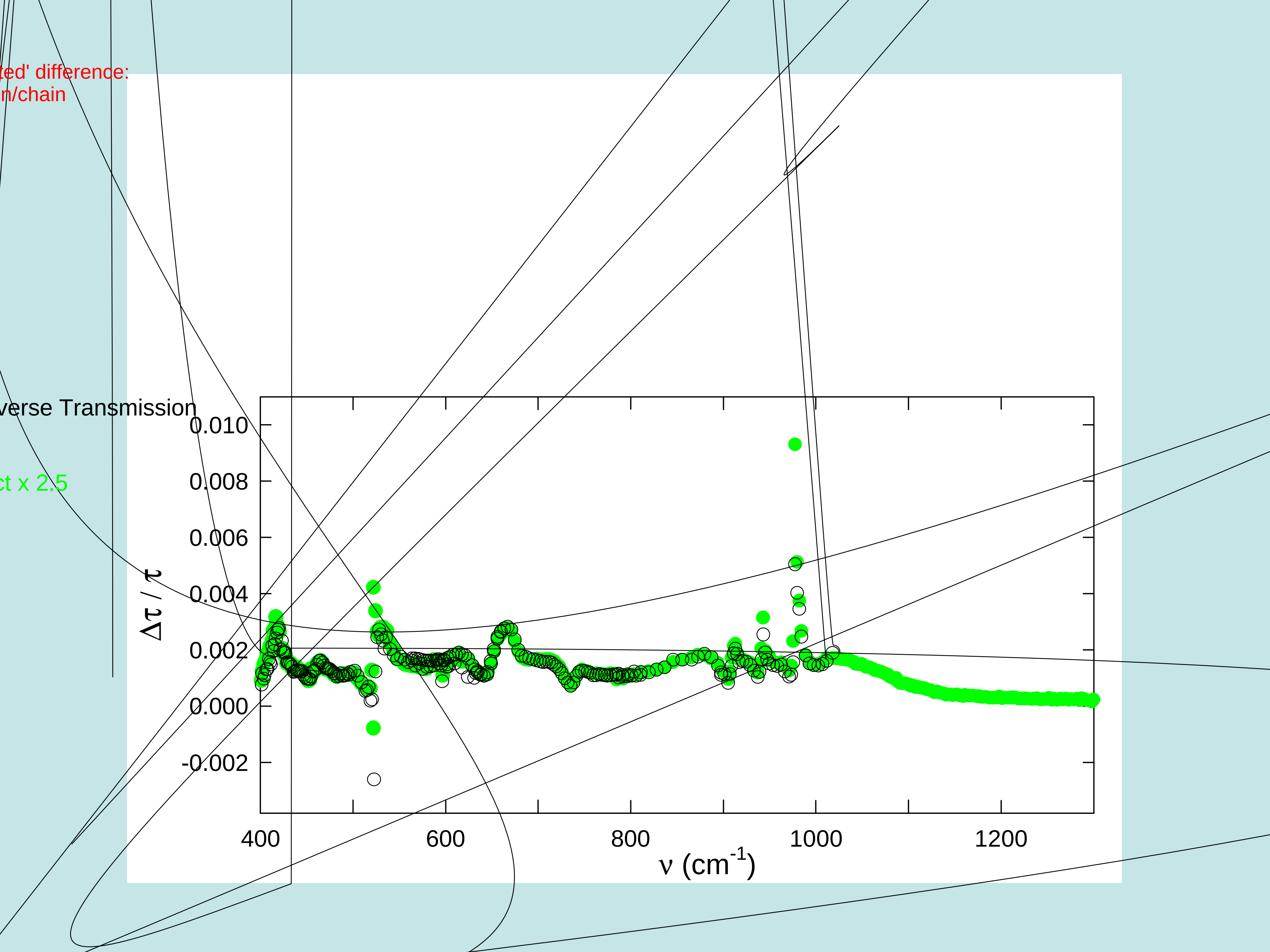
Tool: Electromodulated transmission spectroscopy

Compare Bulk and Contact $\Delta\tau/\tau$ spectra.
Bulk: Bipolar square wave.
Contact: unipolar square wave: CDW does not depolarize when V = 0



Electro-transmission spectra measurements





ted' difference:
n/chain
verse Transmission
ct x 2.5
Δτ / τ
0.010
0.008
0.006
0.004
0.002
0.000
-0.002
400
600
800
1000
1200
ν (cm-1)

Discussion

- Phonons are modulated by CDW strain, with linewidths and frequencies decreasing at positive contact

  $\Delta\Gamma \sim \Delta\nu \sim 0.01\ cm^{-1}$.

  Mechanisms:

  $\Delta\Gamma$: changes in screening by thermally excited electrons.

  $\Delta\nu$: ??

- Difference between contact and bulk: $\delta(\Delta\tau/\tau) < 4 \times 10^{-5}$

  (vibrations from helium refrigerator cooling lasers)

  $n\sigma/\Gamma < 4 \times 10^{-10}$ / (A cm^{-1}).

  Expect: soliton linewidth $\Gamma \sim kT_c \sim 125\ cm^{-1}$,

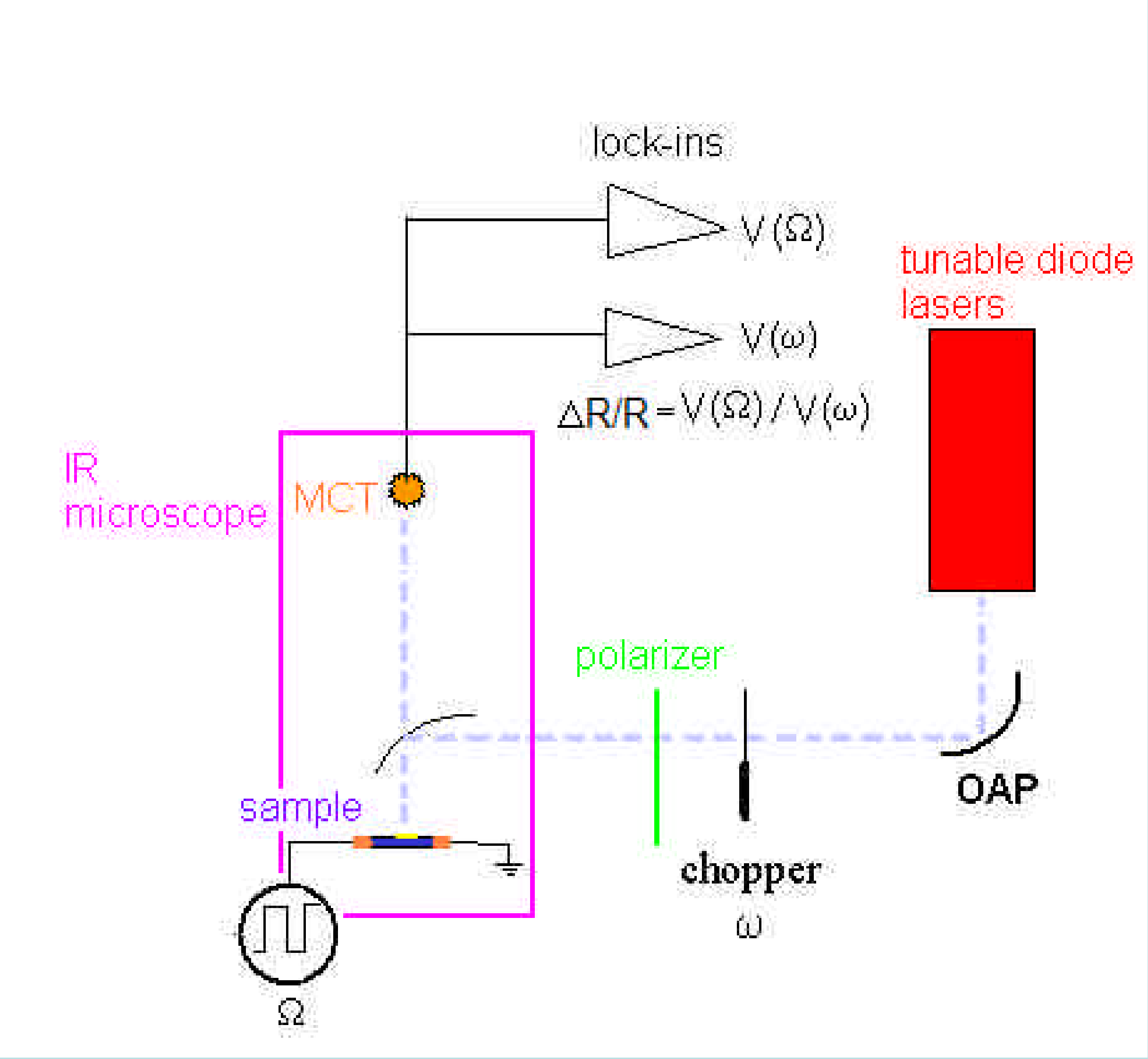
  cross-section $\sigma \sim$ î$_{//}$î ~100 $Å^2$ (!?polarization?!).

Results: solitons on < 4% of chains.

Expect: n ~ 1/chain for $V > V_T$

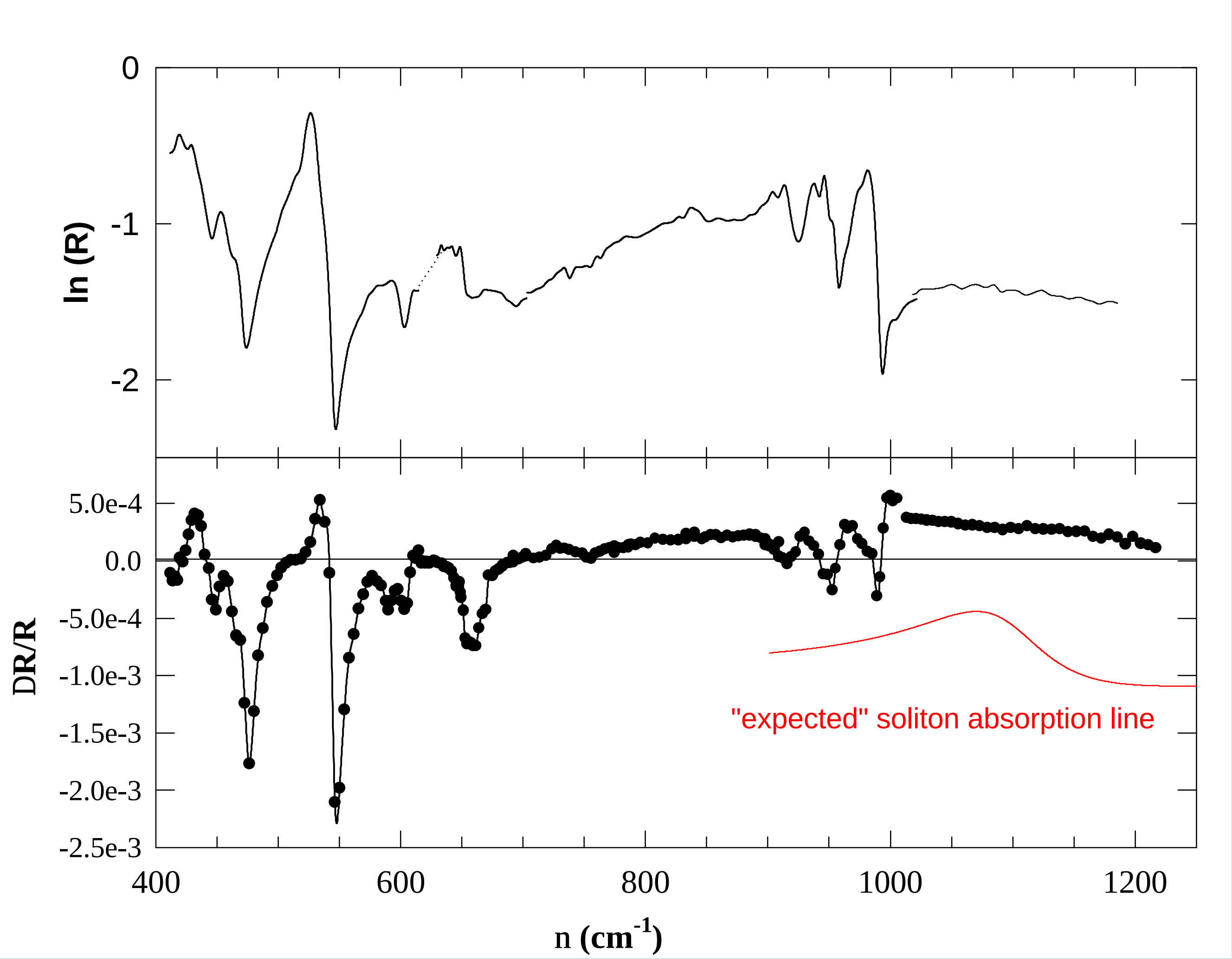
For parallel polarization, absorption too strong, must study reflectance

Electro-reflectance measurements

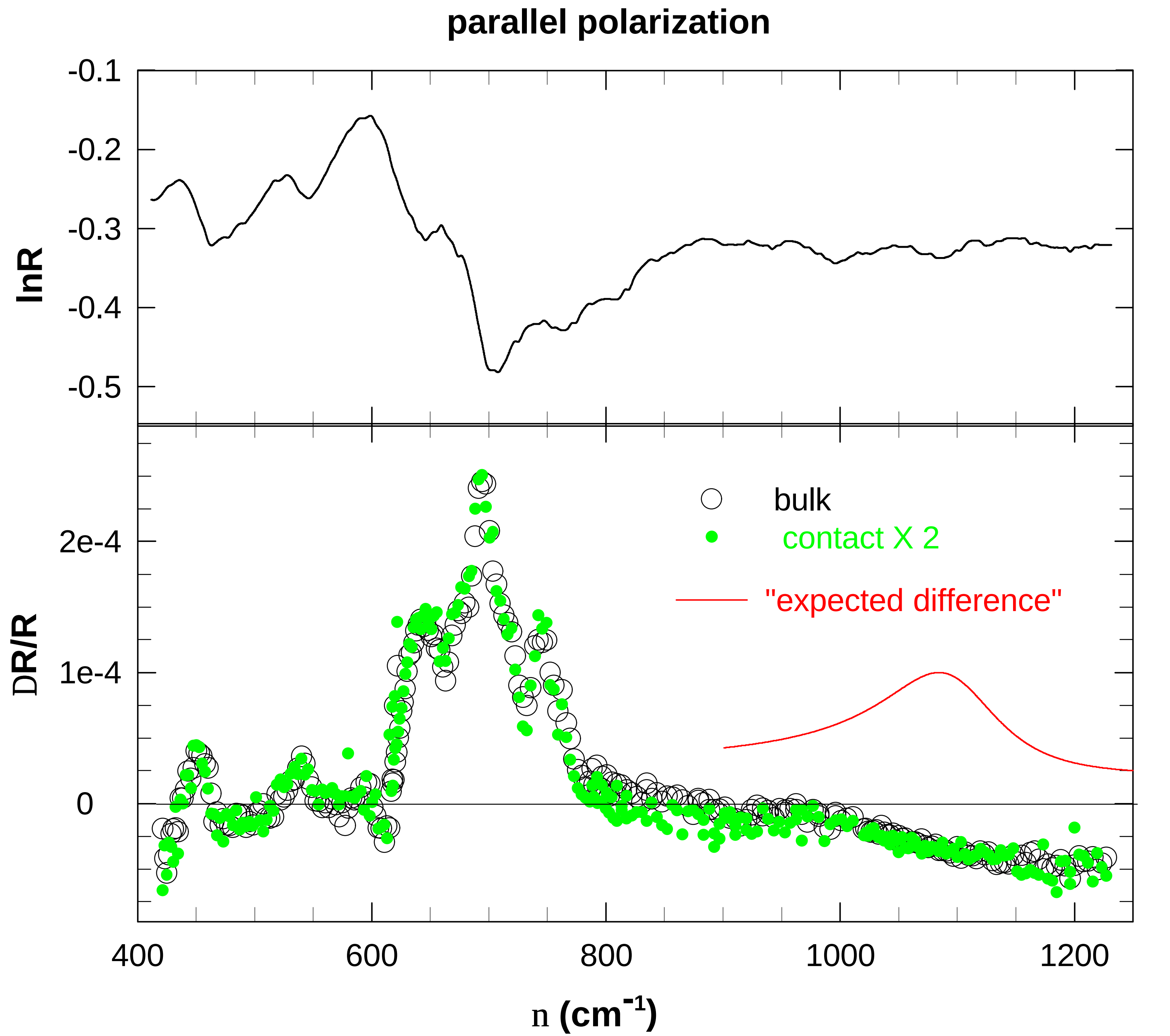


Electromodulated Reflectance, **transverse** polarization

$\Delta R/R = [R(+V) – R(-V)] / R$



- Phonon modulations similar to what's observed in transmission


parallel polarization
lnR
-0.1
-0.2
-0.3
-0.4
-0.5
DR/R
2e-4
1e-4
0
bulk
contact X 2
"expected difference"
400
600
800
1000
1200
n (cm-1)

Discussion

- Phonon modulations similar (some larger) than for transverse transmission but more difficult to analyze changes in Fano lineshapes.

- Contact (unipolar) and bulk (bipolar) spectra identical:

  $\delta(\Delta R/R) < 5 \times 10^{-6}$ $n\sigma/\Gamma < 10^{-9}$ / (A cm^{-1}).

  Solitons on < 15% of chains.

CDW materials exhibit a unique electro-optic effect:

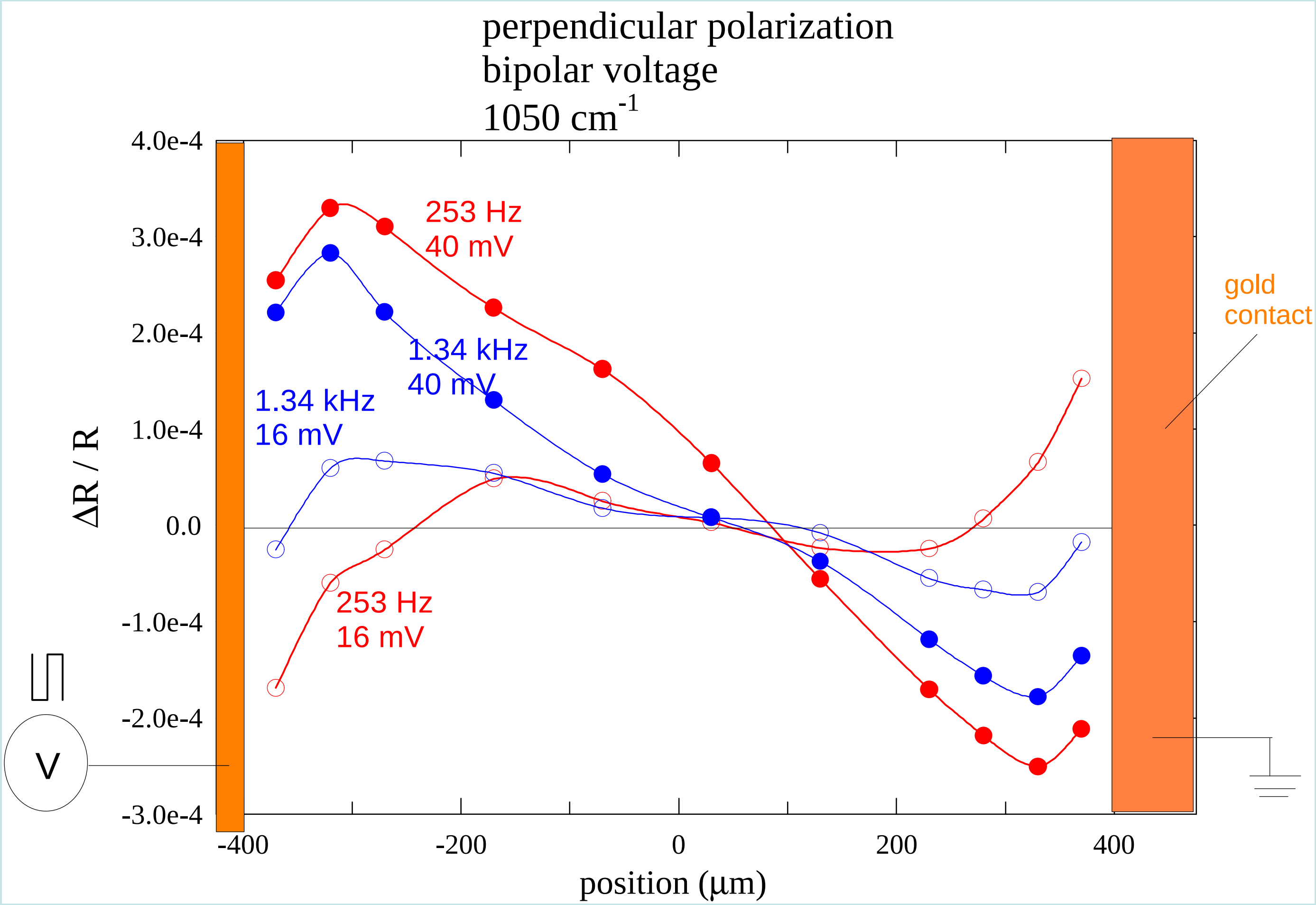
1) Varies spatially throughout sample
2) Occurs at extremely small electric field (~100 mV/cm)
3) Occurs over very wide (infrared) spectral range

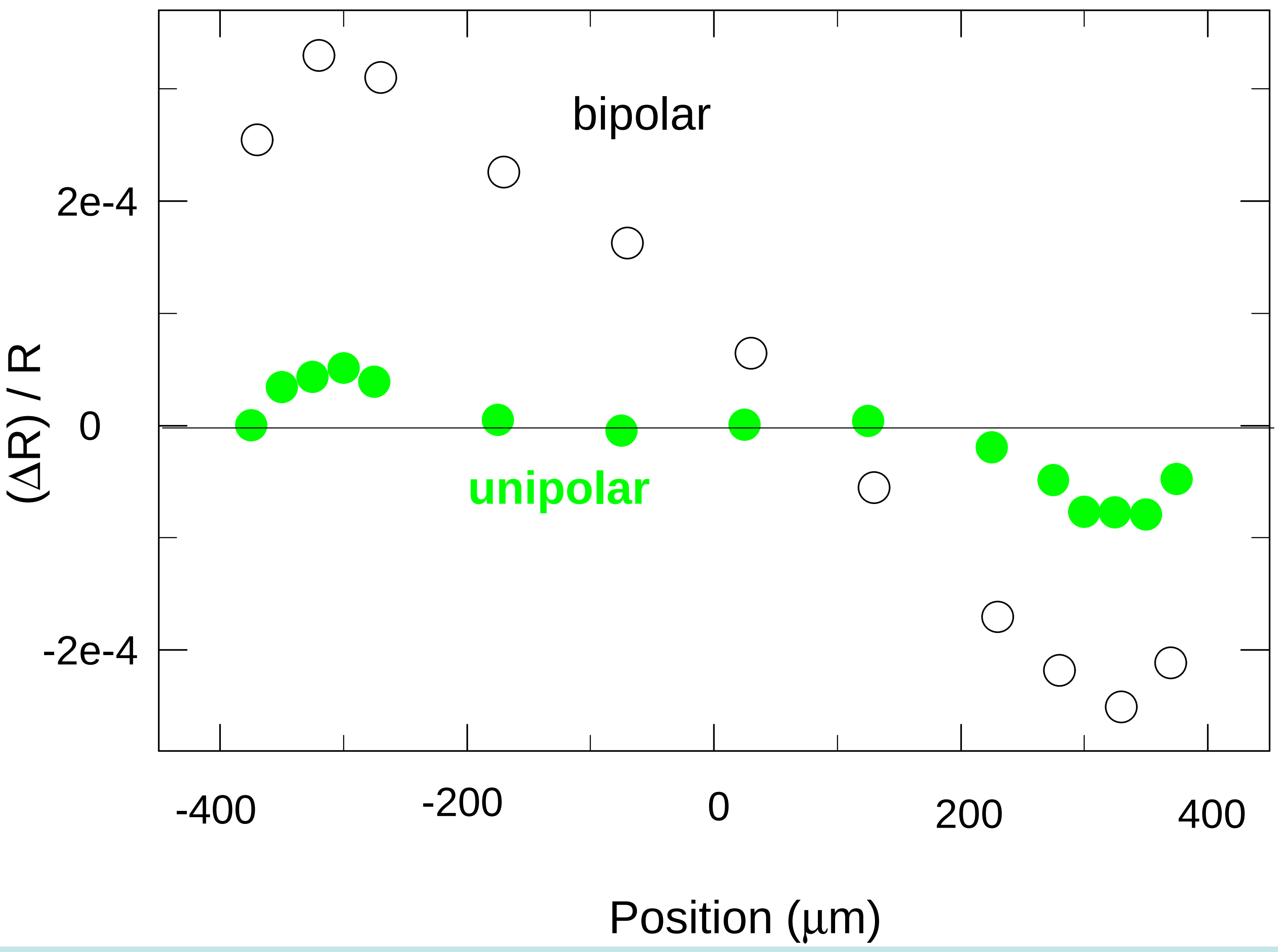
Hopes for New Experiments at ALS (Mike Martin)

- Better sensitivity/resolution in spectra: search for intragap states, phonon changes, etc.

- Study the frequency, voltage, (wavelength, temperature) dependence of the spatial variation of $\Delta R/R$, $\Delta\tau/\tau$.

Electro-reflectance signal inverted near contacts for voltages near threshold (V_T = 15 mV)





bipolar
unipolar
2e-4
0
-2e-4
(ΔR) / R
-400
-200
0
200
400
Position (μm)

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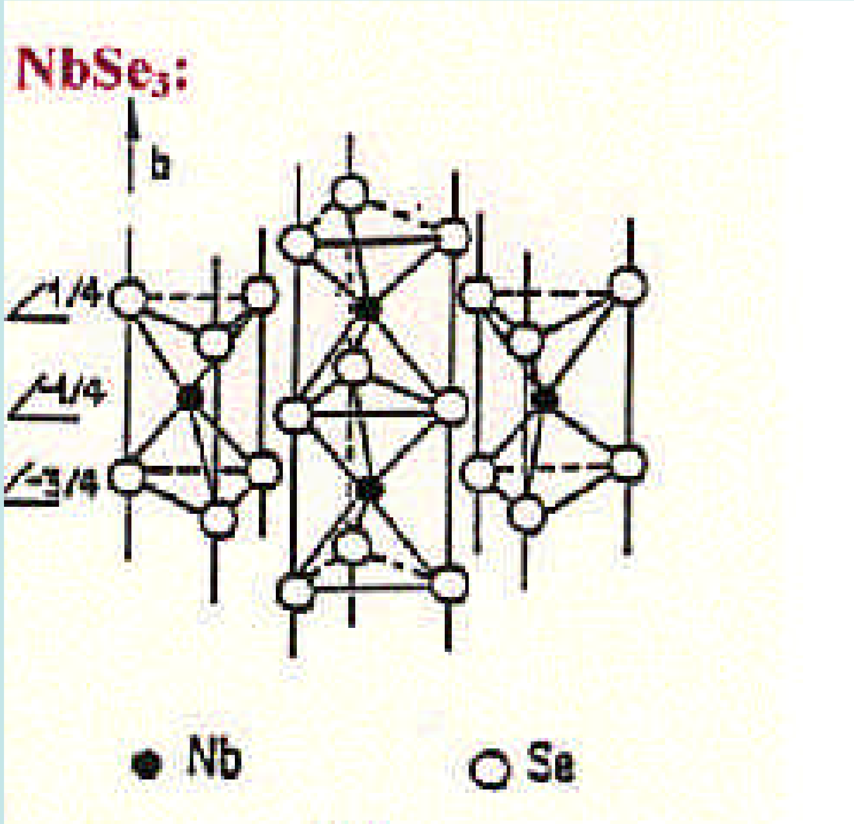
Hopes for New Experiments at ALS (Mike Martin)

- Better sensitivity/resolution in spectra: search for intragap states, phonon changes, etc.
- Study the frequency, voltage, (wavelength, temperature) dependence of the spatial variation of $\Delta R/R$, $\Delta\tau/\tau$.
- Study more materials:
  - Organic SDW materials with $2\Delta < 100$ cm^{-1}
  - Other whisker-size CDW crystals ($NbSe_3$, TaS_3)

$NbSe_3$, TaS_3 crystals typically < 20 μm wide



Thorne
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