Coherent Phonons and Coherent Control in Semiconductors

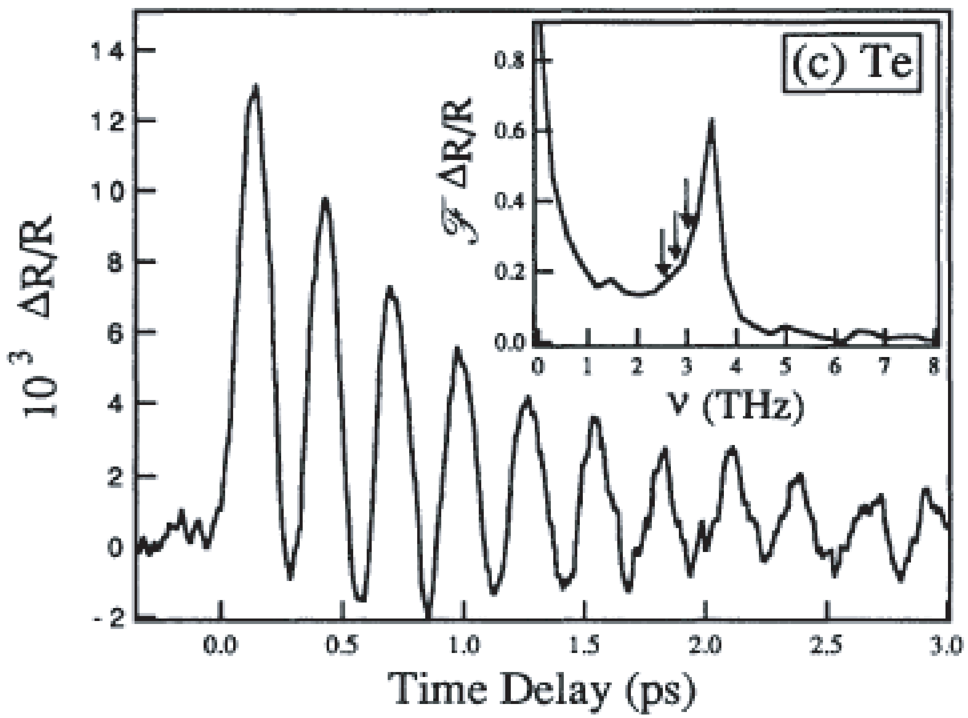
M. Kandyla, C.A.D. Roeser, A.M.-T. Kim and Eric Mazur
Department of Physics and Division of Engineering and Applied Sciences

Harvard University

Outline

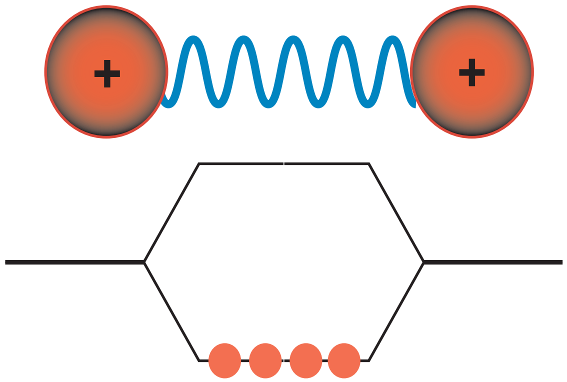
1. Displacive Excitation of Coherent Phonons
2. Experimental Results
3. Coherent Control

Only one phonon mode excited in certain materials such as Te, Sb, Ti_2O_3, Bi.

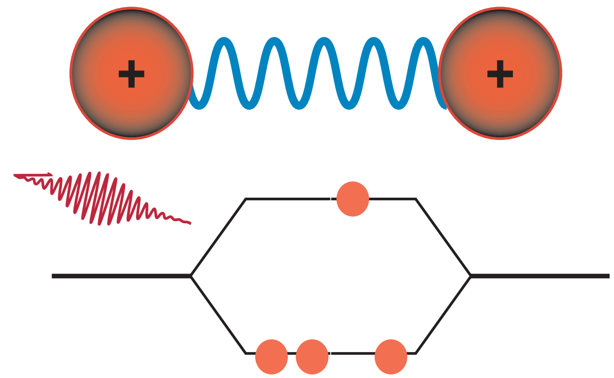


Zeiger at al., Phys. Rev. B 45, 768 (1992).

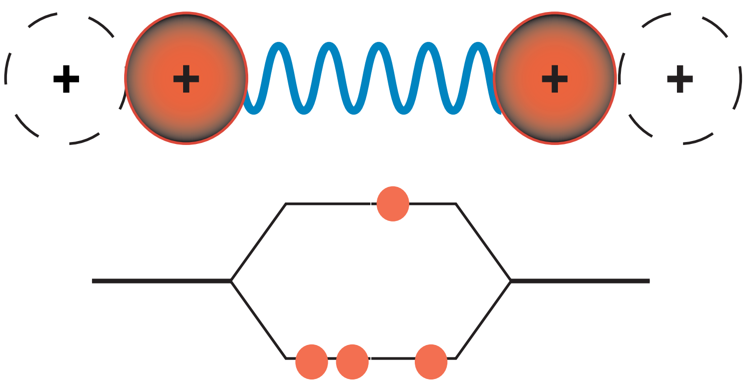
Displacive Excitation of Coherent Phonons (DECP)



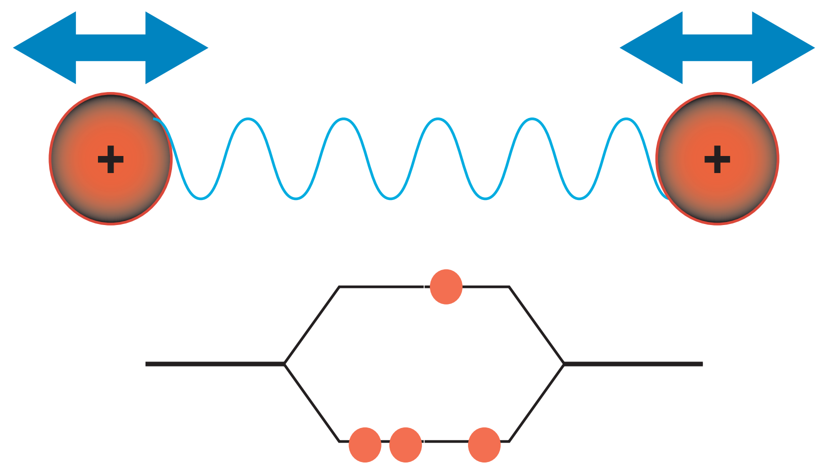
Equilibrium position



Laser excites an electron



New equilibrium separation is established for the ions



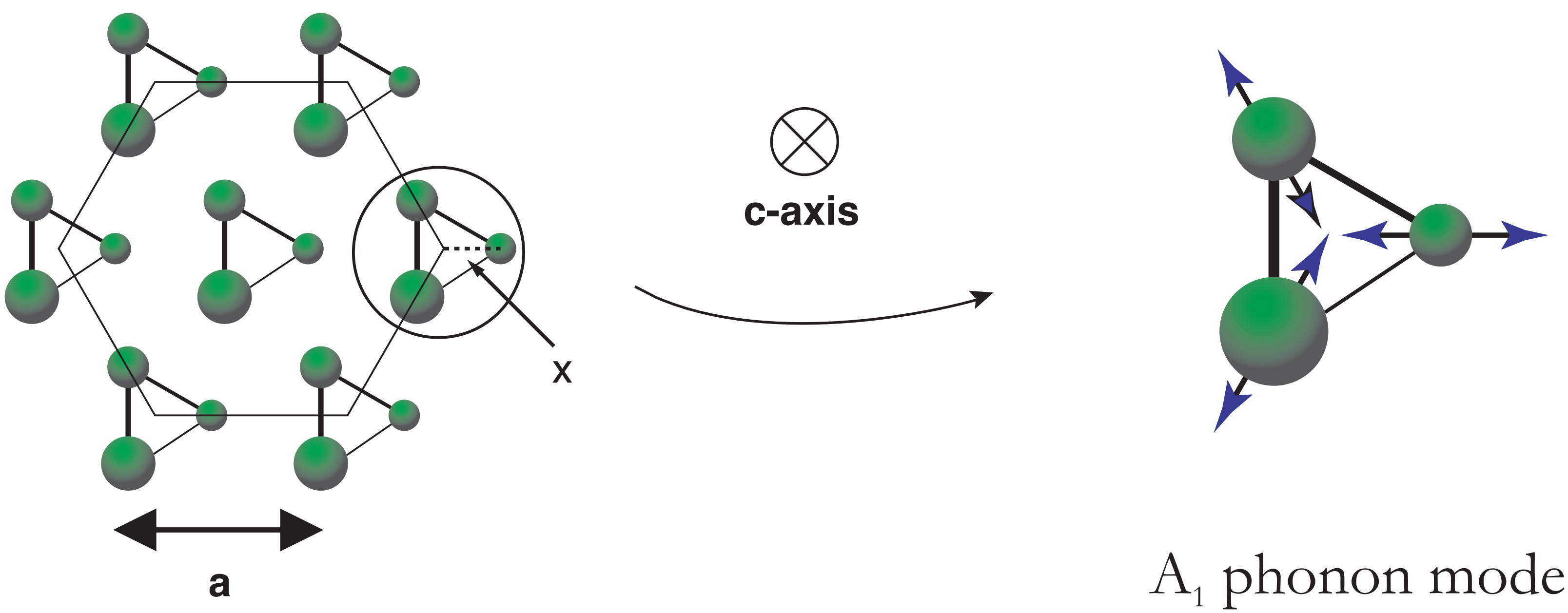
Ions oscillate around the new position before relaxing

DECP in Te

DECP does not impose any symmetry breaking direction onto the crystal.

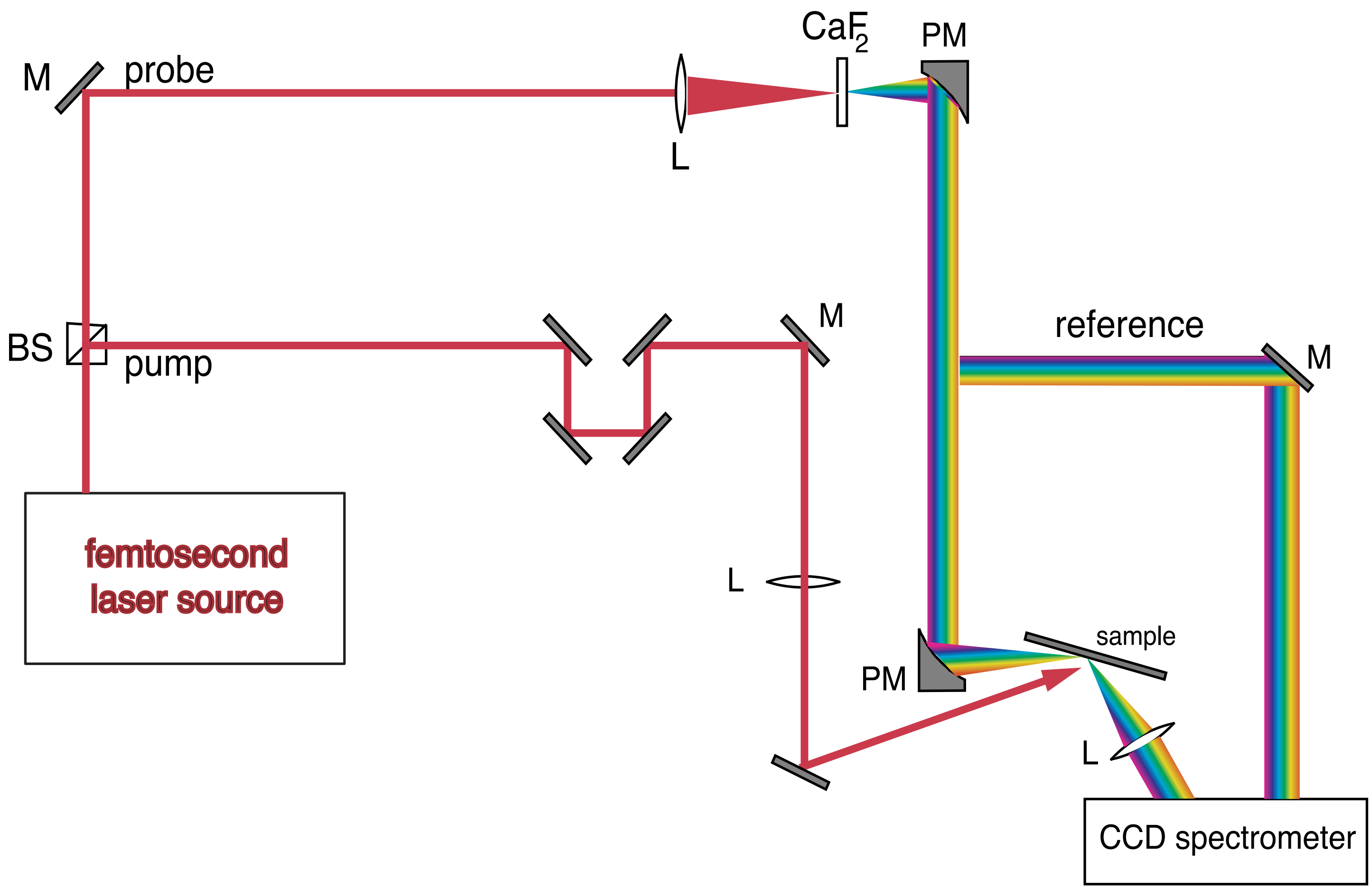
Only phonon modes which fully preserve the symmetry of the crystal are excited via DECP.

Te possesses a a fully symmetric phonon mode, the A_1 mode.



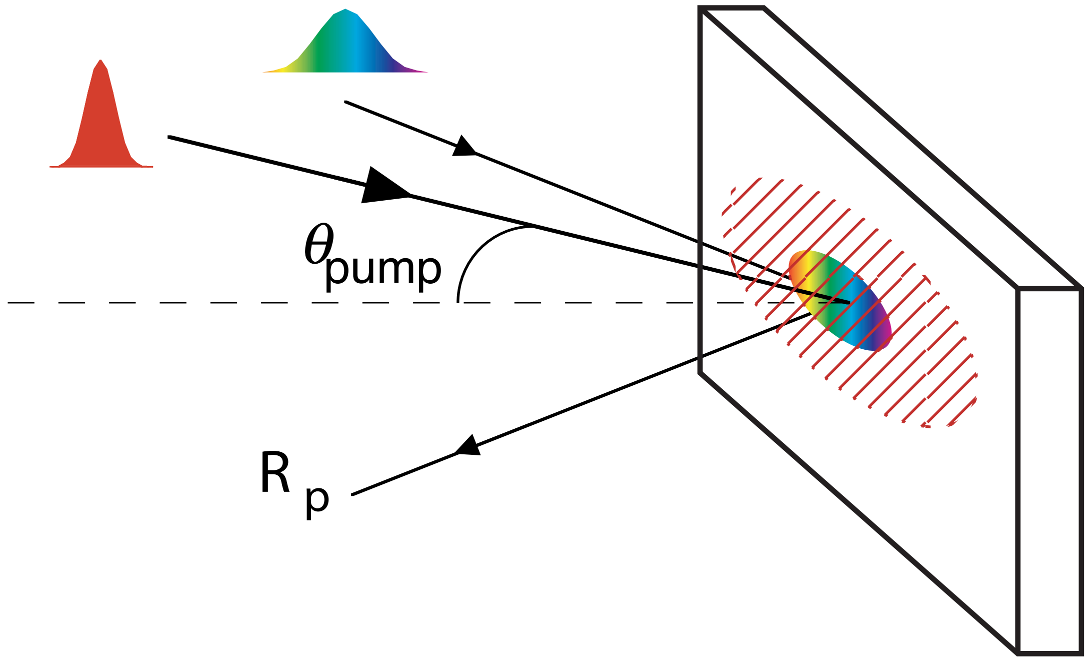
A_1 phonon mode

White-light pump-probe setup



Experimental Setup

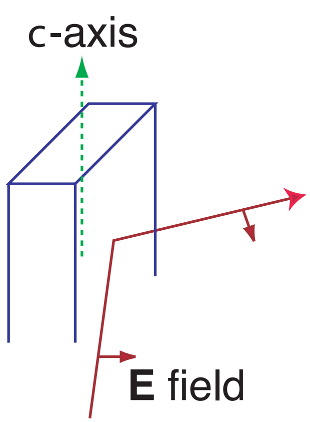
Pump-probe details



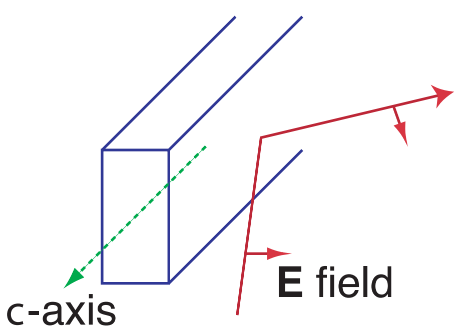
pump pulse:
1.5 eV (800 nm)
up to 250 mJ

probe pulse:
1.7 - 3.5 eV (350nm-750nm)
< 0.1 mJ

Time-resolved reflectivity measurements have been performed and from them the dielectric constant of Te has been calculated using the Fresnel formulae:

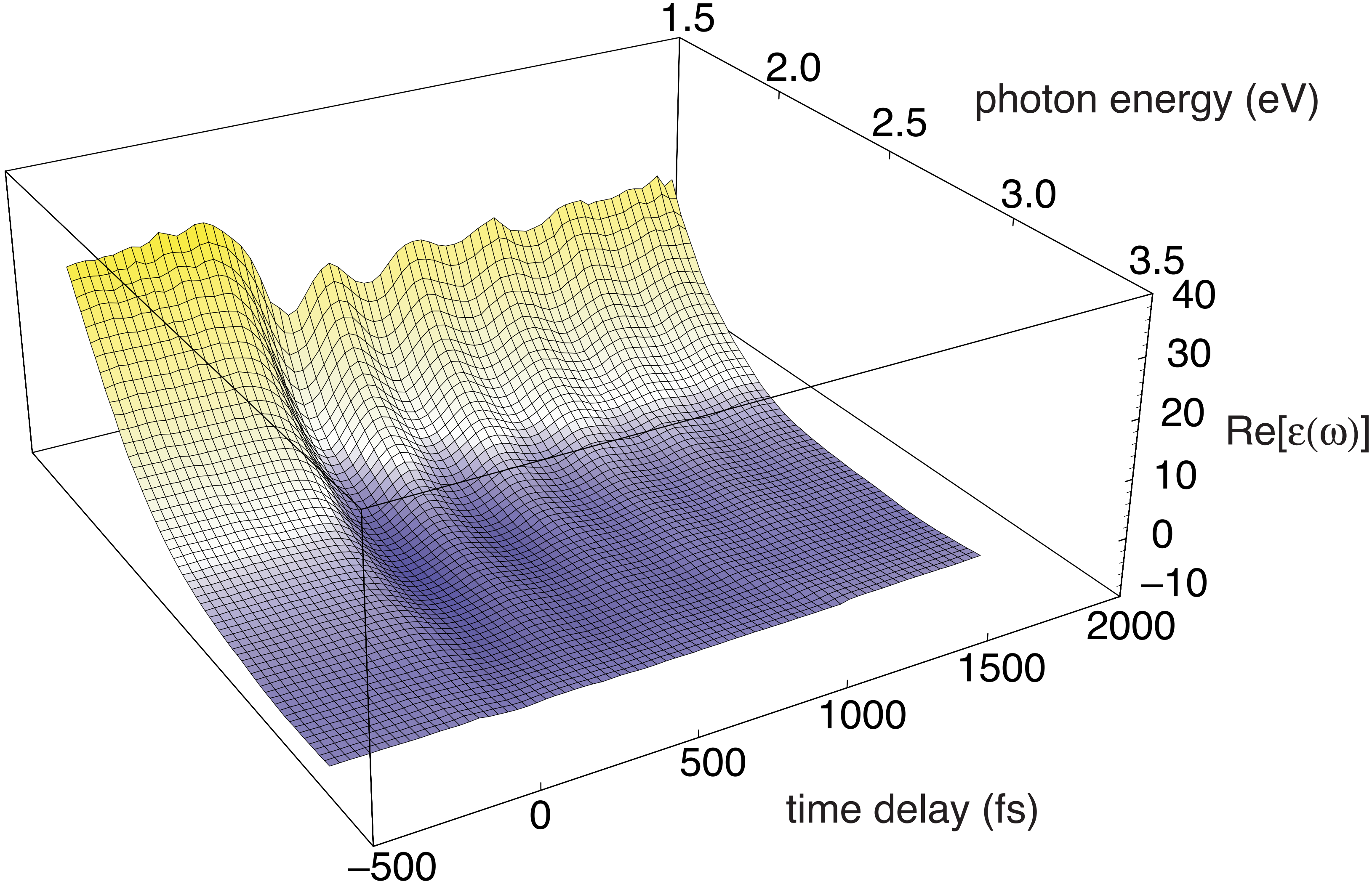


$$r_p = \frac{\varepsilon_{\text{ord}} \cos\theta - \sqrt{\varepsilon_{\text{ord}} - \sin^2\theta}}{\varepsilon_{\text{ord}} \cos\theta + \sqrt{\varepsilon_{\text{ord}} - \sin^2\theta}}$$



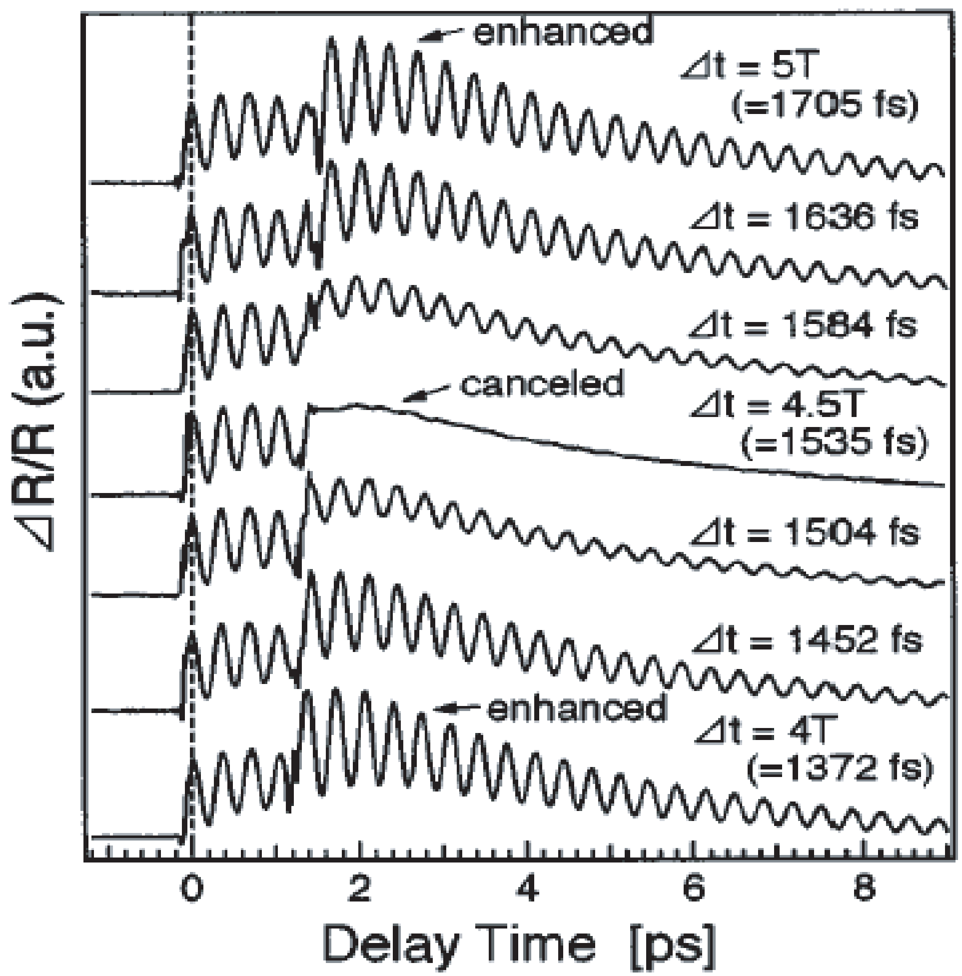
$$r_p = \frac{\sqrt{\varepsilon_{\text{ord}}\varepsilon_{\text{ext}}} \cos\theta - \sqrt{\varepsilon_{\text{ord}} - \sin^2\theta}}{\sqrt{\varepsilon_{\text{ord}}\varepsilon_{\text{ext}}} \cos\theta + \sqrt{\varepsilon_{\text{ord}} - \sin^2\theta}}$$

Past Results



Double-pump experiments on Te: control of coherent phonons.

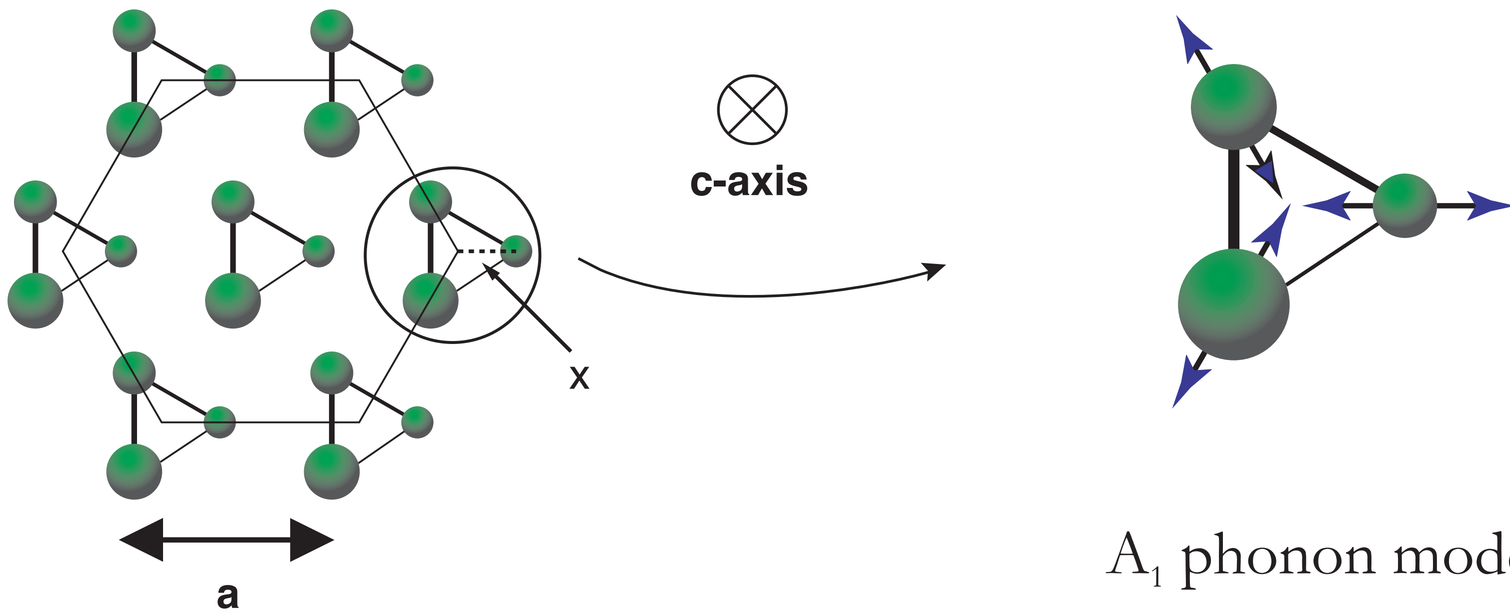
It has been demonstrated in Bi



Hase et al., Appl. Phys. Lett. 69, 2474 (1996)

Why Is Phonon Cancellation Interesting?

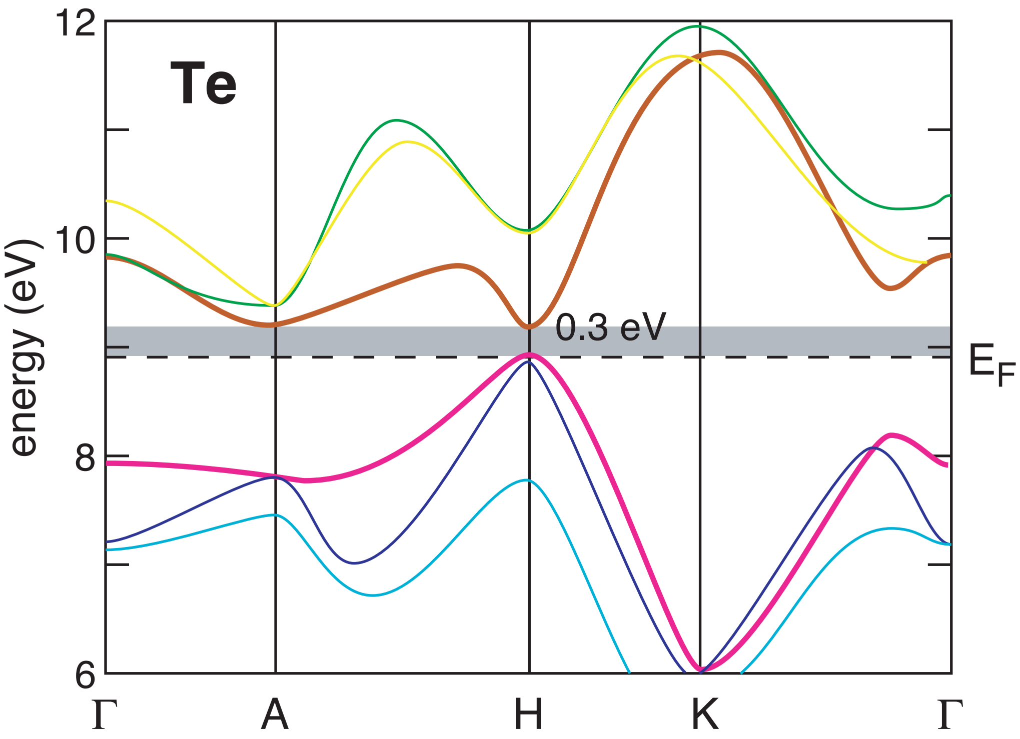
The motion of the ions according to the A_1 mode results in a variation of the radius of the helices, x.



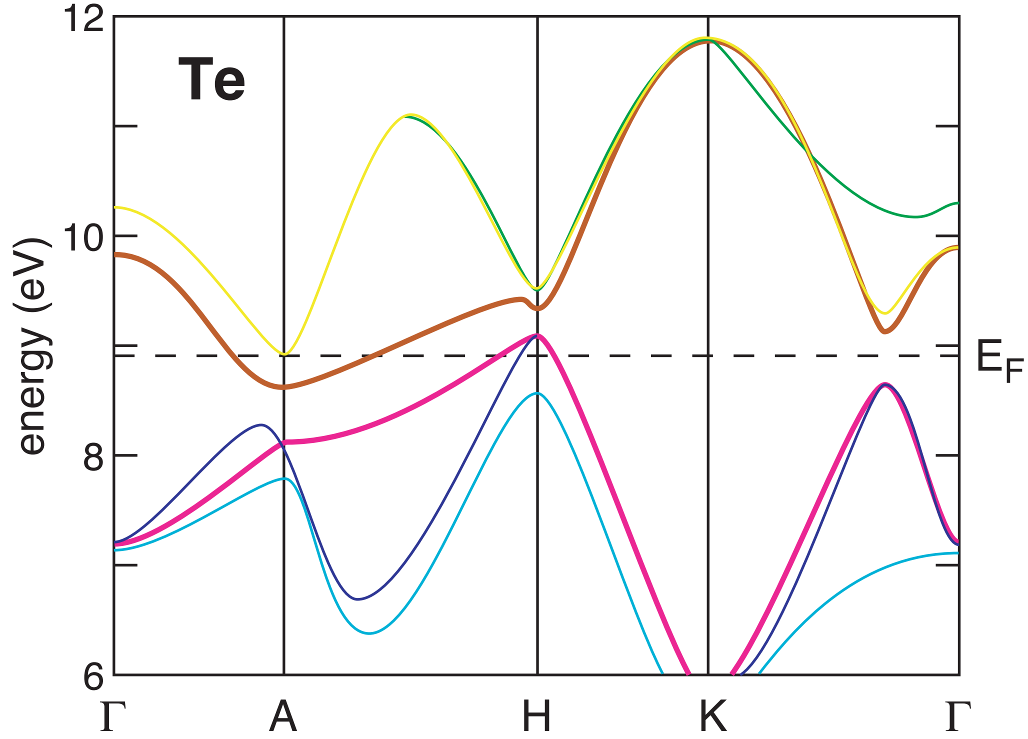
A_1 phonon mode

Why Is Phonon Cancellation Interesting?

Theoretical studies predict that when x changes sufficiently (x=0.286a) the 0.3 eV indirect gap of Te closes:



equilibrium
x = 0.2686a

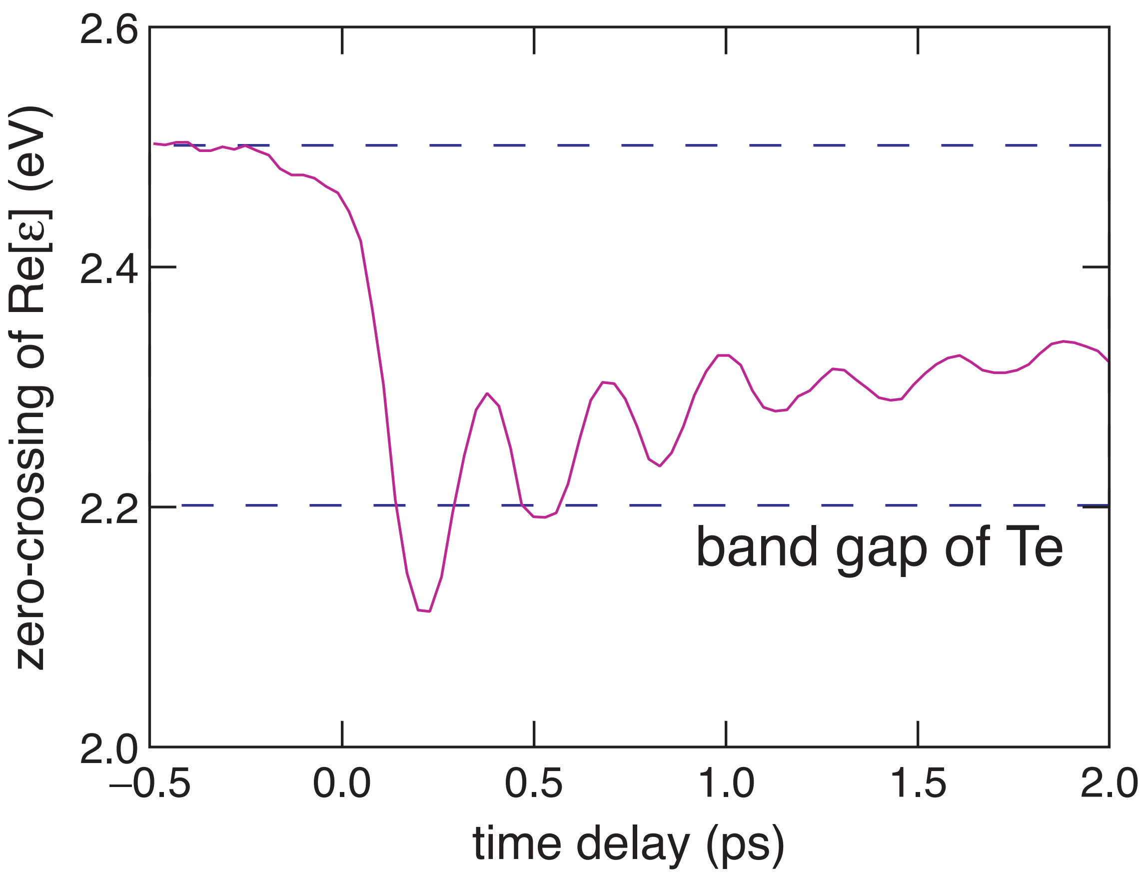


x = 0.31a

P. Tangney, Master's thesis, University College Cork, Ireland (1998)

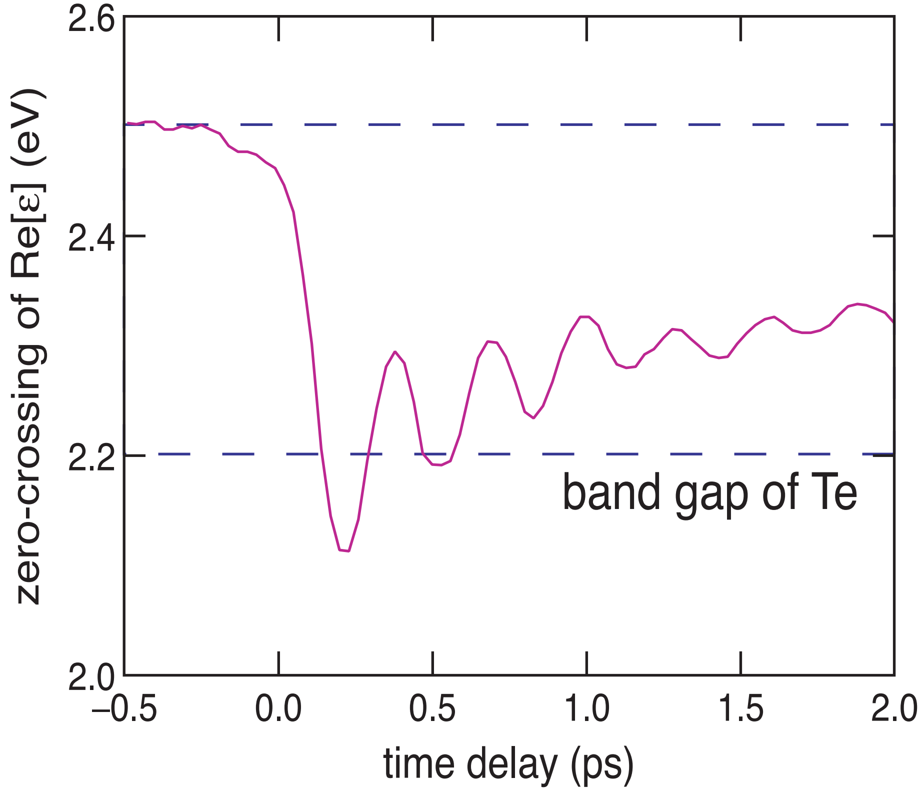
Why Is Phonon Cancellation Interesting?

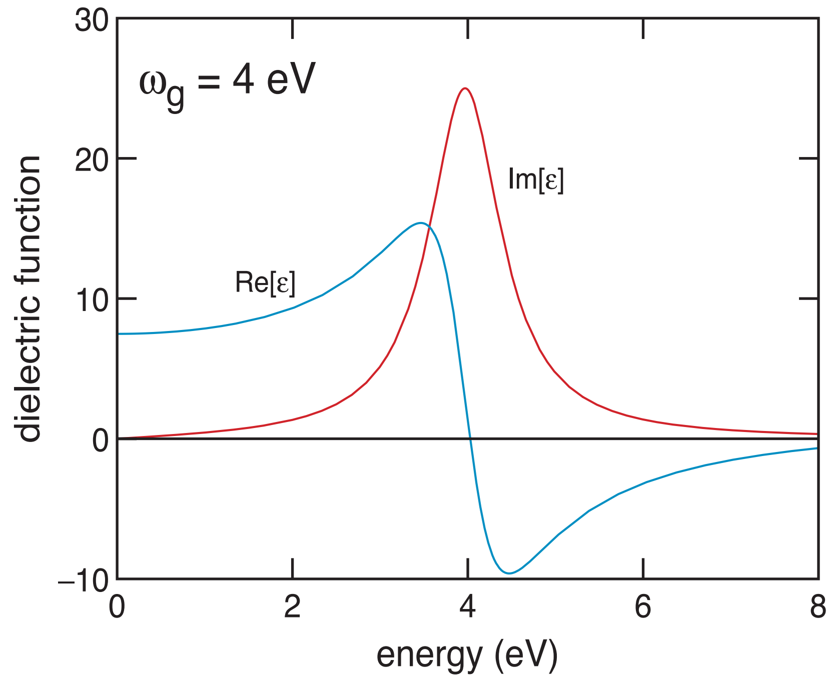
The experimental data presented before also indicate closing of the 0.3 eV indirect gap:

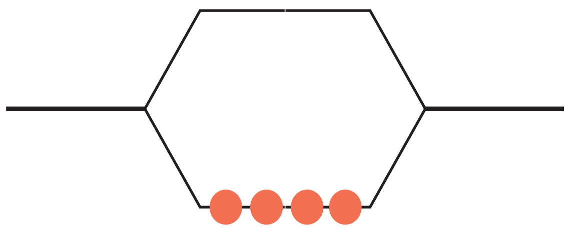


Bandgap Closes

In the oscillator model of the dielectric function the zero crossing frequency indicates the bonding-antibonding splitting.







Oscillator Model

Electrons are tied to ions like classical oscillators.

$$m\ddot{x}_j = -m\omega_j^2 x_j - m\Gamma_j \dot{x}_j - eE_0 e^{-i\omega t}$$

Solving for $x(t) = x_0 e^{-iwt}$ we can find:

$$P_j(t) = f_j N\left[-ex_j(t)\right] = \varepsilon_0 \chi_j E_0 e^{-i\omega t}$$

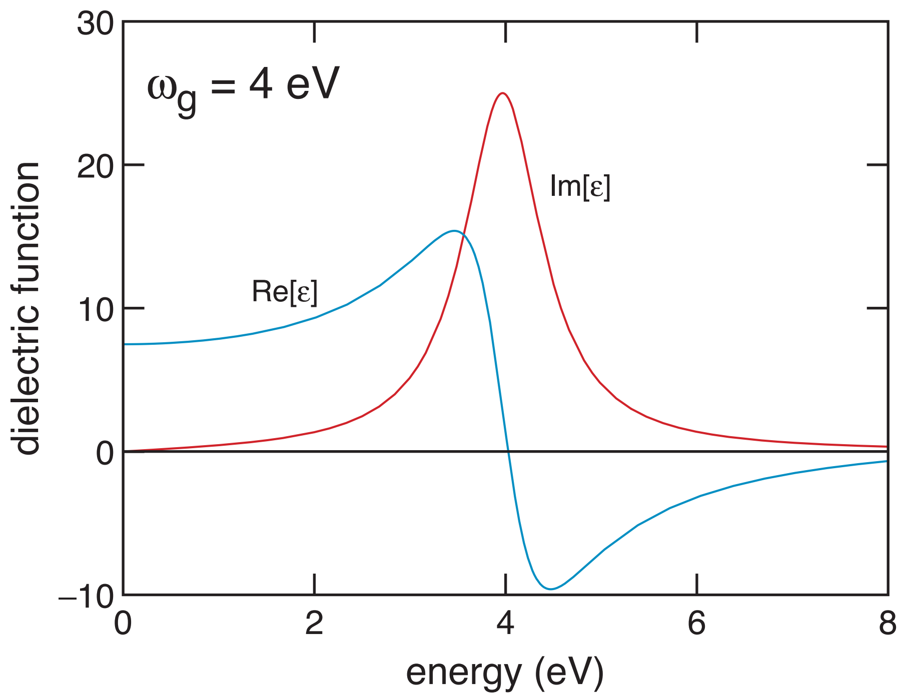
and from here the dielectric function:

$$\varepsilon(\omega) = 1 + \Sigma\chi_j$$

Oscillator Model

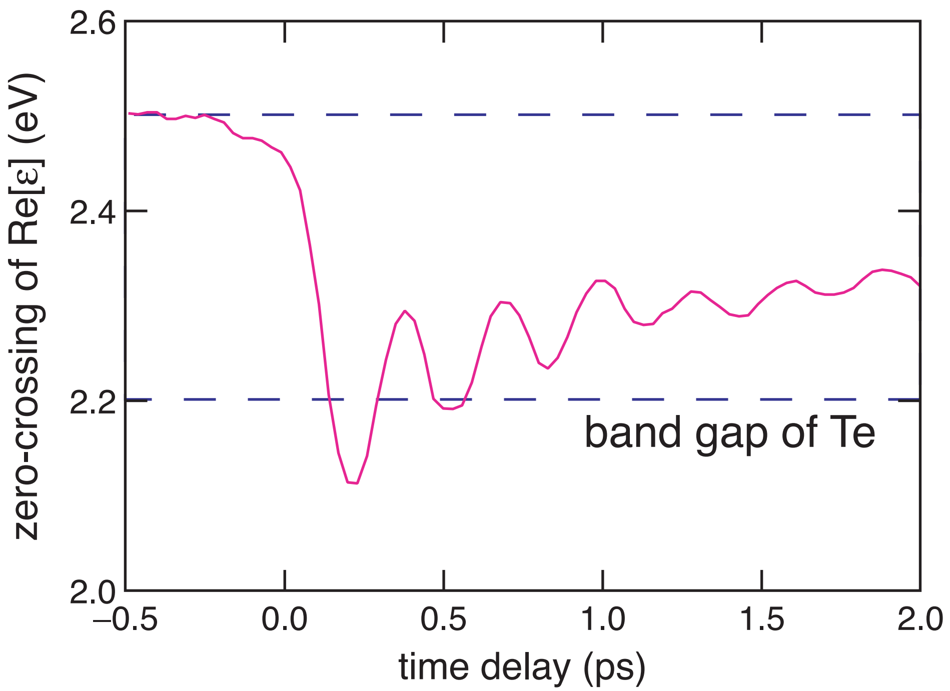
In a system modeled with one oscillator:

$$\varepsilon(\omega) = 1 + \left(\frac{Ne^2}{\varepsilon_0 m}\right)\frac{1}{\omega_g^2 - \omega^2 - i\omega\Gamma}$$



From microscopic theory of dielectric function we know that the peak of its imaginary part approximately gives the average splitting between bonding and antibonding states of the material.

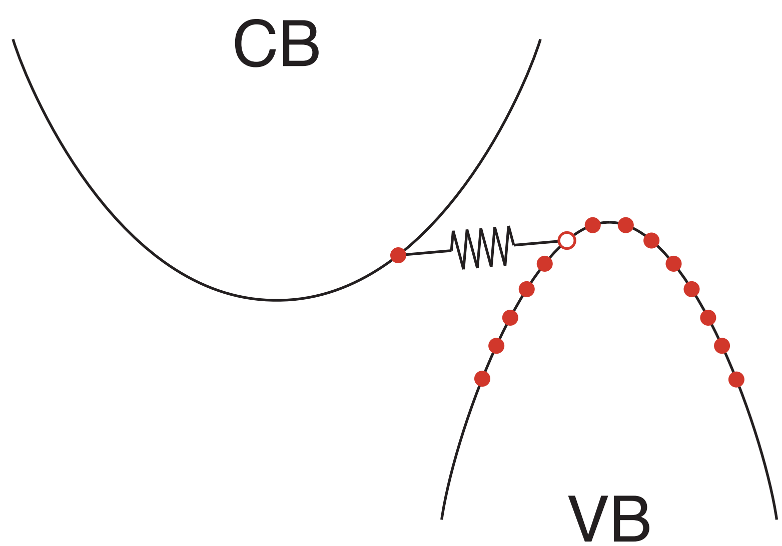
Te



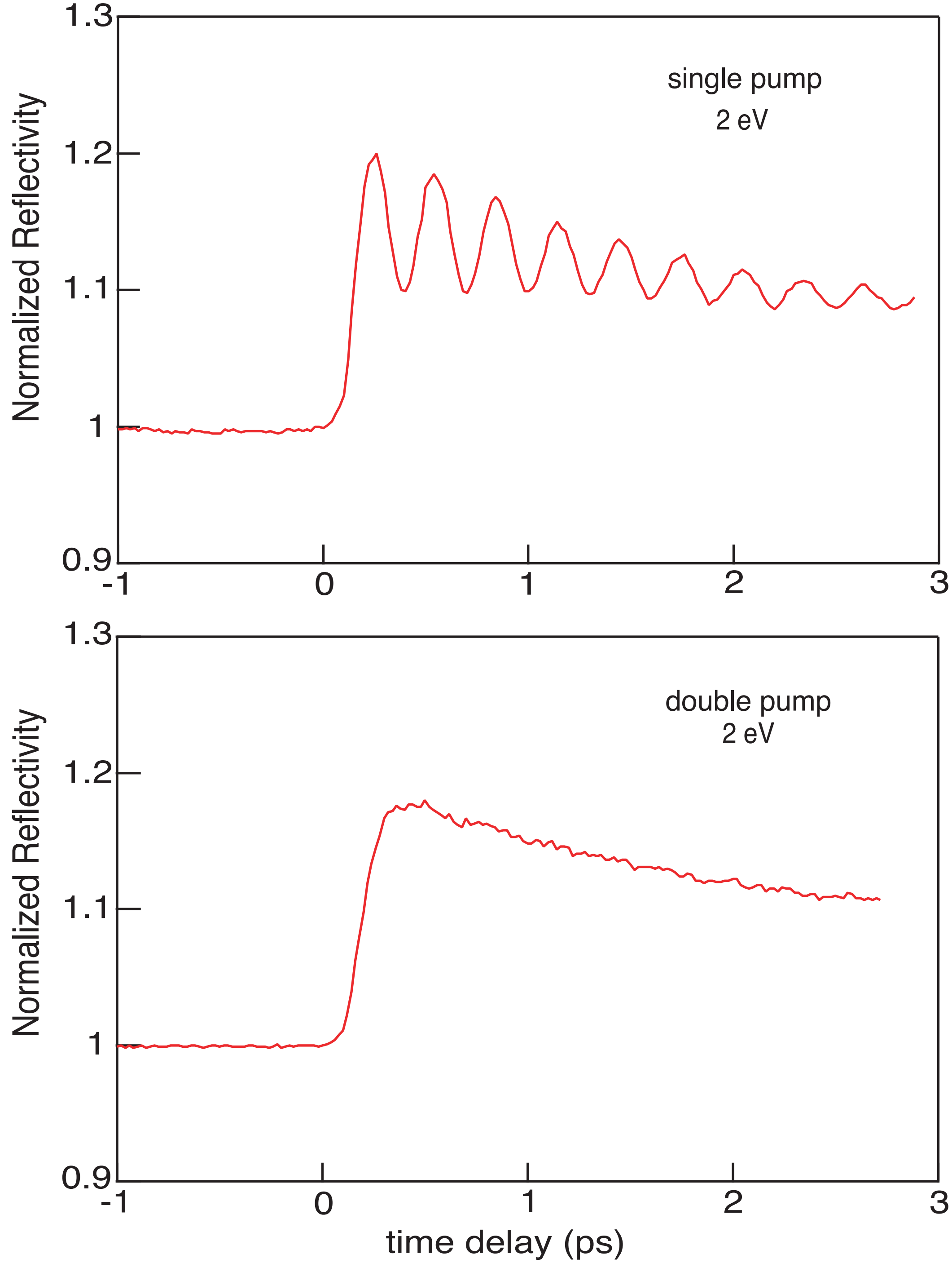
The bands seem to cross for approximately 100 fs.

Phonon Cancellation

Cancelling the phonons will enable us to study carrier scattering events.



Current Results



Conclusions

A_1 phonons can be excited coherently in certain class of materials via DECP

Our technique enables us to observe time-resolved dynamics in a broad frequency range

Te exhibits a band crossing transition

Double pump experiments will investigate the material behaviour in the crossed bands state

Acknowledgements

Thanks to the Mazur group

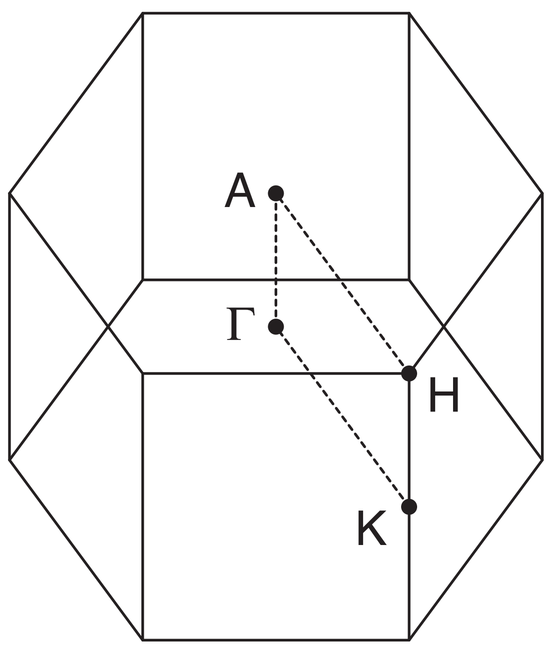
Te

Te has 3 atoms and 18 valence electrons in the unit cell.
The structure consists of infinite helices with three atoms per turn parallel to the c(z) axis. Each atom has 2 nearest neighbours displaced +-1/3c from it and rotated from it +-120 degrees about the c axis and 4 second nearest neighbours in adjacent helices.

x is the radius of the helices. Equilibrium value (experimental) x=0.2686 interhelical distance. A1 mode only varies x.x is not determined by lattice symmetry but by electron distribution. Excited electrons are placed in the center of the helices pushing the helices apart.

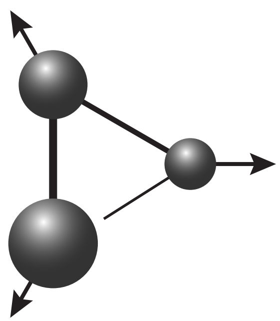
As x increases the band gap decreases. Indirect band gap=0.3 eV at equilibrium. Indirect crossing at x=0.286. direct crossing at x>1/3.

Brillouin zone of Te



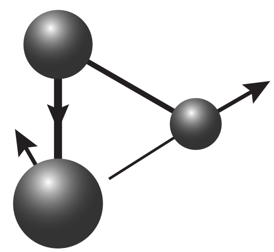
Other optical phonon modes in Te

A_1



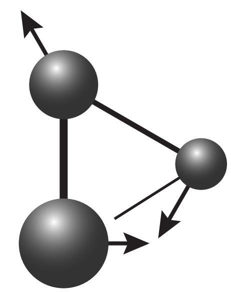
Raman
3.6 THz

E'_{LO}



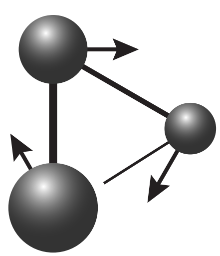
Raman/infra-red
3.09 THz

E''_{LO}



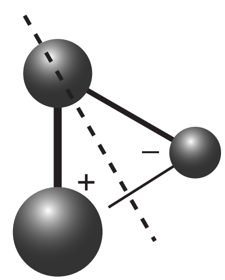
Raman/infra-red
4.26 THz

A_2



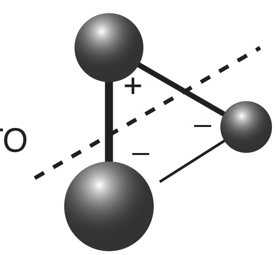
infra-red
2.6 THz

E'_{TO}



Raman/infra-red
2.76 THz

E''_{TO}



Raman/infra-red
4.22 THz