

Chapter 9 Interaction of light with lattice vibrations

We have now looked at oscillations of bound and free electrons driven by light. The remaining optical effect to consider is the interaction of light with phonons. In a classical treatment, we can re-use the Lorentz oscillator model, applied now to the charged ions. We consider:

- Phonon revision
- Coupling of light to optical phonons
- Infrared reflectivity of insulators and semiconductors

9.1 Types of phonon

A phonon is a coherent collective motion of all the atoms in a solid; it is a quantum of lattice vibration. The typical energy scale of phonons is 0.01 to 0.1 eV (corresponding to the energies of far infrared photons).

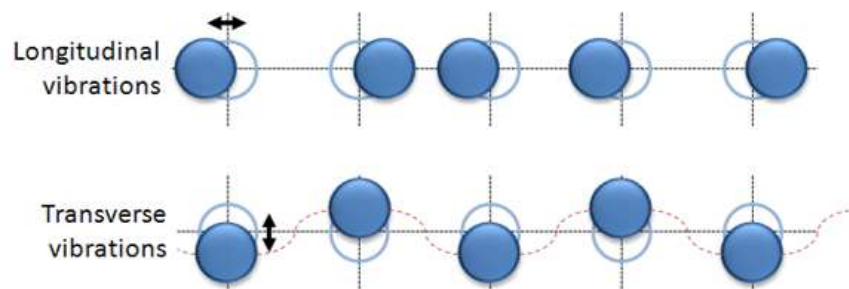


Figure 9.1: Atomic displacements in longitudinal and transverse vibrations. •

Phonons may involve either longitudinal (L) or transverse (T) displacements. Note that phonons of type L and T with the same wavevector need not have the same frequency; in general, the restoring forces are different for L and T modes.

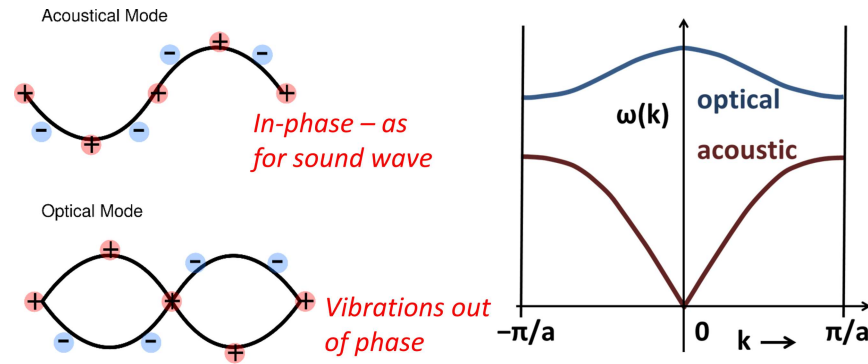


Figure 9.2: Left: atomic displacements in acoustic and optic phonons; right: schematic phonon dispersion for a diatomic linear chain of atoms. •

9.1.1 Phonon dispersion curves

We have seen that the direction of motion of the atoms compared to the wavevector can be longitudinal or transverse and atoms within a unit cell can vibrate in phase (acoustic mode) or out of phase (optical mode) with each other. This gives four types of mode, usually denoted as LA, TA, LO, TO.

In diamond the LO and TO phonons have the same energy at $q=0$: this is because, although the two carbon atoms in each unit cell are distinct (that is, not connected by any symmetry operation), they are chemically identical and their bonds are non-polar; polar bonds would be required to lift the LO-TO degeneracy. For the acoustic branch at very small wavevectors (and very small frequencies) the dispersion curve is linear: $\omega = v_S q$ where v_S is the speed of sound in direction q in the reciprocal space of the material.

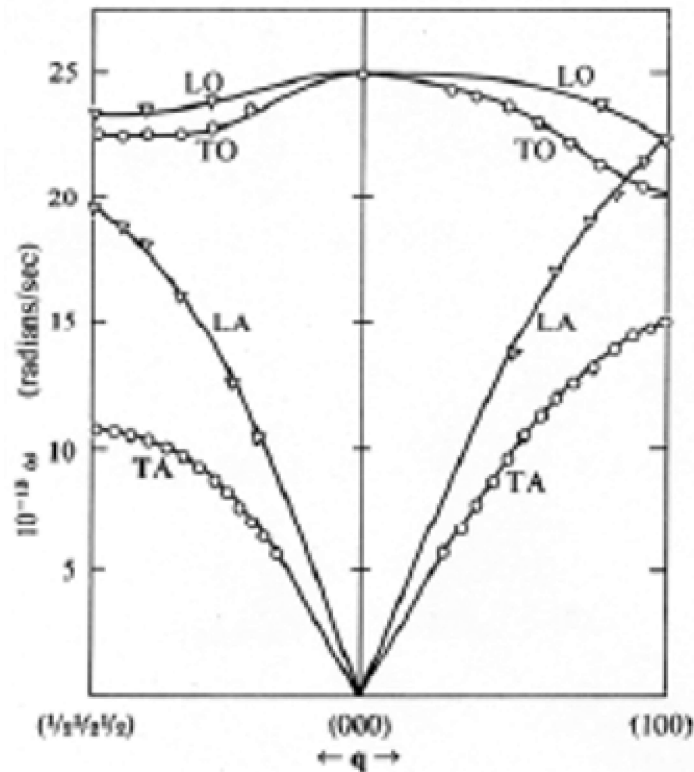


Figure 9.3: Phonon dispersion of diamond measured by inelastic neutron scattering in two directions in reciprocal space, showing LA, TA, LO and TO modes. ●

9.2 Infrared active phonons

In an ionic solid positively charged atoms move in opposite direction to negatively charged ones in an optical mode, i.e. there is an oscillating dipole that can couple to an EM wave if the phonon is transverse. Polar TO phonons are said to be “IR active”

The EM wave and the vibrational wave must have the same frequency and momentum to couple. The dispersion curves shows that this is only possible directly for the optical branch at wavevectors $q \sim 0$ (because $2\pi/\lambda \ll \pi/a$).

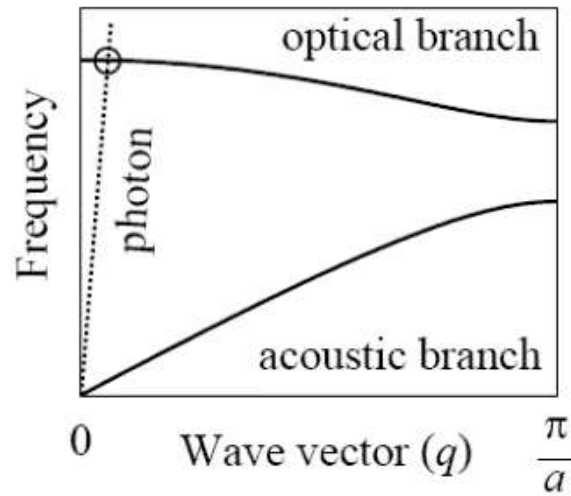


Figure 9.4: Dispersion of photon mode (exaggerated slope) compared to vibrational dispersion curves. From Fox, Fig. 10.2. Note, this is the same as the figure we used much earlier to illustrate polariton formation. •

9.3 Lorentz oscillator model of driven lattice vibrations

We start in a similar way to the calculation of the dispersion curve for the phonons of a linear diatomic chain of atoms. The equations of motion for positive and negative ions in an optical field $E(t)$ are:

$$\begin{aligned} m_+ \frac{d^2 x_+}{dt^2} &= -K(x_+ - x_-) + QE(t) \\ m_- \frac{d^2 x_-}{dt^2} &= -K(x_- - x_+) - QE(t) \end{aligned} \quad (9.1)$$

where Q is the effective charge on an ion and K is the restoring force constant for the ionic motion.

Dividing each equation by the relevant mass and subtracting them from each other, we get

$$\frac{d^2}{dt^2}(x_+ - x_-) = -\frac{K}{\mu}(x_+ - x_-) + \frac{Q}{\mu}E(t) \quad (9.2)$$

where we use the reduced mass for the ions:

$$\mu = \frac{m_+ m_-}{m_+ + m_-}. \quad (9.3)$$

We now use the coordinate $x = x_+ - x_-$ and introduce a phonon damping constant γ , the inverse of the phonon lifetime of ~ 1 -10 ps. The finite phonon lifetime is due to phonon-phonon scattering associated with the anharmonicity of lattice vibrations. The equation of motion becomes:

$$\frac{d^2 x}{dt^2} + \gamma \frac{dx}{dt} + \Omega_{TO}^2 x = \frac{Q}{\mu} E(t) \quad (9.4)$$

where

$$\Omega_{TO}^2 = \frac{K}{\mu}. \quad (9.5)$$

Hence,

$$\varepsilon_r(\omega) = 1 + \chi_b + \frac{NQ^2}{\varepsilon_0 \mu} \frac{1}{(\Omega_{TO}^2 - \omega^2 - i\gamma\omega)} \quad (9.6)$$

using the results of the atomic oscillator model. Now Eq.(9.6) can be re-written as:

$$\varepsilon_r(\omega) = \varepsilon_\infty + (\varepsilon_{st} - \varepsilon_\infty) \frac{\Omega_{TO}^2}{(\Omega_{TO}^2 - \omega^2 - i\gamma\omega)} \quad (9.7)$$

with high frequency dielectric constant ($\omega \gg \Omega_{TO}$):

$$\varepsilon_\infty = 1 + \chi_b \quad (9.8)$$

and static dielectric constant ε_{st} for $\omega = 0$. Note that for light damping,

$$\varepsilon_r(\omega) \approx \varepsilon_\infty + (\varepsilon_{st} - \varepsilon_\infty) \frac{\Omega_{TO}^2}{(\Omega_{TO}^2 - \omega^2)} \quad (9.9)$$

which has a solution $\varepsilon_r(\omega') = 0$ when

$$\omega' = \left(\frac{\varepsilon_{st}}{\varepsilon_\infty} \right)^{1/2} \Omega_{TO}. \quad (9.10)$$

From lecture 8, $\varepsilon_r(\omega') = 0$ is the condition under which longitudinal EM waves can exist in materials, so that ω' can be identified with Ω_{LO} . The ratio of LO to TO frequencies is thus given by the **Lyddane-Sachs-Teller** relation:

$$\frac{\Omega_{LO}^2}{\Omega_{TO}^2} = \frac{\varepsilon_{st}}{\varepsilon_{\infty}}. \quad (9.11)$$

9.4 IR reflectivity of polar crystals for light damping

We now inspect the predictions of Eq.(9.7), Fig.9.5 for example parameters $\nu_{TO} = 10.0$ THz, $\nu_{LO} = 11.0$ THz, $\varepsilon_{st} = 12.1$ and $\varepsilon_{\infty} = 10.0$ (these are close to real values for GaAs).

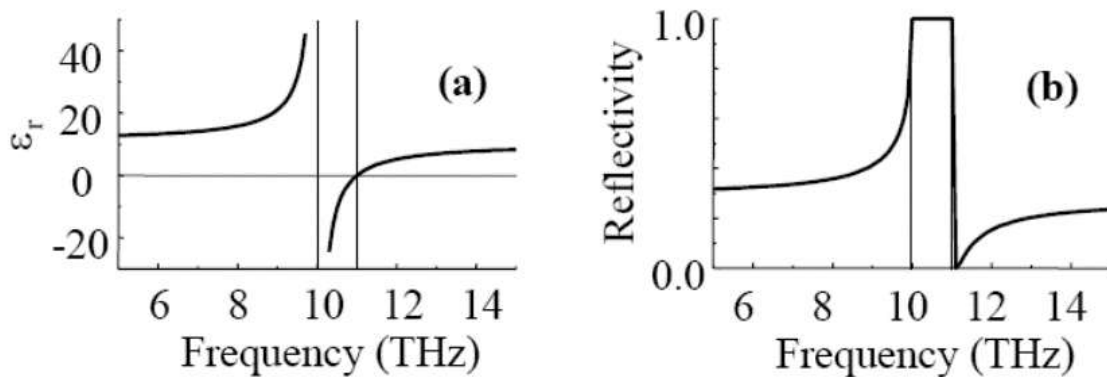


Figure 9.5: (a) dielectric constant and (b) reflectivity for a material with very light damping. •

A key feature to note is the perfect reflectivity between ν_{TO} and ν_{LO} , which is the region where $\varepsilon_r < 0$, giving a purely imaginary n . This frequency region is called the **Reststrahlen** ('residual ray') band.

9.5 Infrared reflectivity of some real polar crystals

Experimental data, Fig.9.6 shows that the reflectivity is high between the LO and TO frequencies but is less than 100% because there is some damping of the resonance. The damping also softens the edges of the resonance.

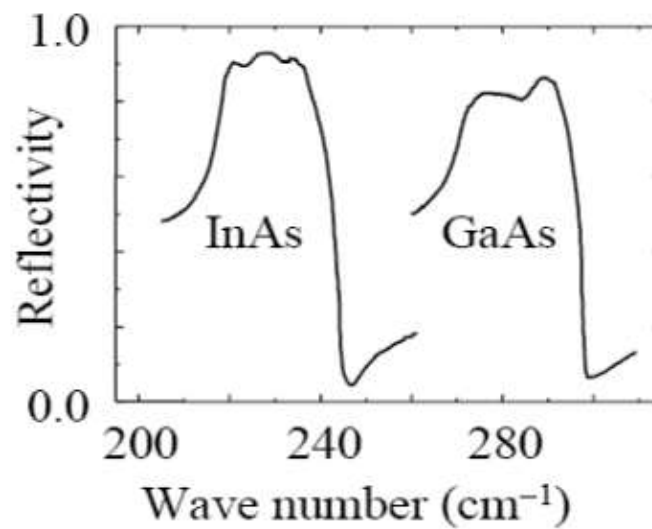


Figure 9.6: IR reflectivity of InAs and GaAs at 4.2 K. ●

Note, the infrared transmission dip due to TO phonon absorption can also be seen in the spectra for sapphire and CdSe shown earlier. Fig. 4.1.