

# Raman, Brillouin, Infrared and Modulation Spectroscopy of Collective and Localized Excitations in Tetrahedrally Coordinated Semiconductors and their Heterostructures

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*Do electronic energy gaps of semiconductors depend on isotopic mass?*

It is well known that semiconductors are characterized by the onset of strong absorption when  $\hbar\omega$ , the incident photon energy, exceeds  $E_g$ , the energy gap separating the highest filled (i.e. valence band, VB) from the lowest, empty conduction band (CB). These band extrema in Si occur with wave vectors differing by, say,  $\mathbf{q}$ . In such a semiconductor, optical transitions between these extrema cannot occur without the assistance of collective vibrations of the constituent atoms (phonons) having wave vector  $\mathbf{q}$ . At **low temperatures** sharp, derivative signatures are observed in the wave-length modulated transmission of Si at  $E_g + \hbar\omega_{\mathbf{q}}$ , with the *creation* of phonons.

In photoluminescence (PL) observed at low temperatures also with phonon creation, corresponding signatures occur at  $E_g - \hbar\omega_{\mathbf{q}}$ . Thus wavelength modulated transmission and the PL spectra together yield  $E_g$  and  $\hbar\omega_{\mathbf{q}}$  separately, with phonons of different polarizations (Transverse Optic (TO), Longitudinal Optic (LO), Transverse Acoustic (TA) and Longitudinal Acoustic (LA)) but the same  $\mathbf{q}$ . Figure 1 shows such spectra for naturally occurring Si whose isotopic composition is  $^{28}\text{Si}$ (92.23%),  $^{29}\text{Si}$ (4.67%), and  $^{30}\text{Si}$ (3.1%).

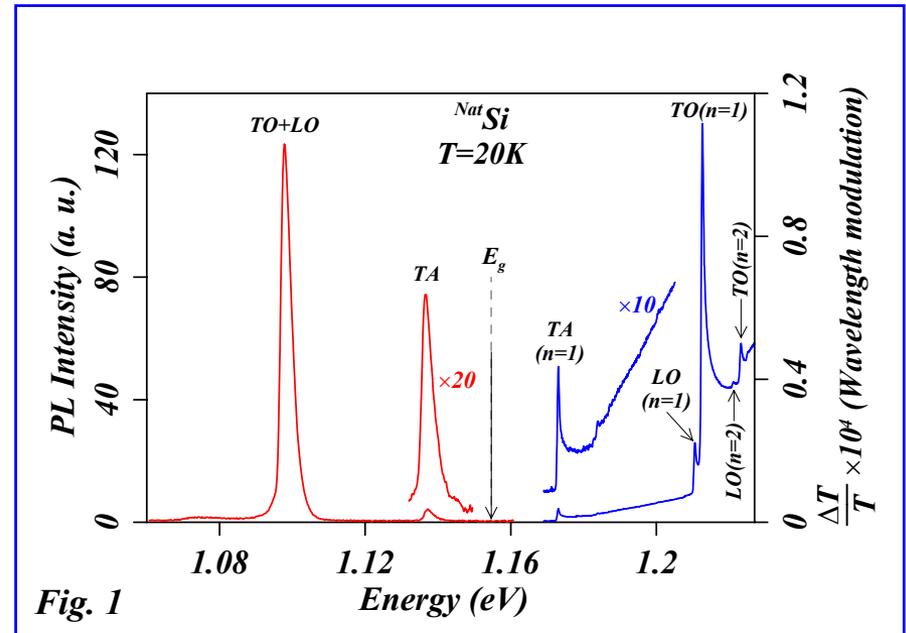


Fig. 1

# “Raman, Brillouin, Infrared and Modulation Spectroscopy of Collective and Localized Excitations in Tetrahedrally Coordinated Semiconductors and their Heterostructures”

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**Does the band gap  $E_g$  of Si depend on its isotopic composition?** This intriguing question was addressed by us, in collaboration with Prof. E. E. Haller (UCB), with the experiment in Fig 1, but carried out with single crystals of  $^{28}\text{Si}$ ,  $^{29}\text{Si}$ , and  $^{30}\text{Si}$ . In Fig 2 we display the signatures at  $E_g - \hbar\omega_q$  in PL and at  $E_g + \hbar\omega_q$  in modulated transmissions. The  $E_g$ 's deduced from these experiments follow  $E_g(M) = E_g(M = \infty) + CM^{-1/2}$  ( $E_g(M = \infty) = 1.212$  eV,  $C = -0.304$  eV,  $M$  in amu.). While the decrease in  $\hbar\omega_q$  as  $M^{-1/2}$  follows from the essential independence of spring constants from  $M$  – they originate in the strength of the chemical bonds entirely electronic in character – the dependence of  $E_g$  on  $M$  is a quantum mechanical effect. Even at absolute zero temperature, atoms do not cease to vibrate but rather experience the so-called “zero point motion”. The resulting electron-phonon interaction – it can be shown – depresses the CB minimum and lifts the valence band maximum. This then accounts for the mass dependence of  $E_g$ , decreasing in the order  $E_g(^{30}\text{Si}) > E_g(^{29}\text{Si}) > E_g(^{28}\text{Si})$ .

The graduate students participating in the program have benefited enormously from the stimulating national and international collaboration involving unique materials and state-of-the art spectroscopic techniques.

