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Inelastic light scattering in crystals

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In 1988, the year of the Raman birth centenary and the diamond jubilee of the discovery of the Raman Effect[1], it seems appropriate to recall Lord Rutherford's statement in his presidential address to the Royal Society[2]: "'The Raman Effect' must rank among the best three or four discoveries in experimental physics of the last decade. It has proved, and will prove an instrument of great power in the study of the theory of solids."

The nature of the one-phonon and multiphonon-Raman spectra (Rasetti & Fermi[3]; R. S. Krishnan[4]; Raman-Born controversy[5]); the LO-TO splitting of polar modes (Mathieu & Couture[6]); Brillouin scattering and elastic/elasto-optic constants (Krishnan[7]; Krishnan & Chandrasekharan[8]; Chandrasekharan[9]); symmetry (Bhagavantam & Venkatarayudu[10]); and soft modes and phase transitions (Raman & Nedungadi[11]) represent some of the major contributions to inelastic light scattering in crystals during the pre-laser period.

The invention of the laser in 1960 and its application to Raman spectroscopy in 1962 have made inelastic light scattering a powerful tool in condensed-matter physics. With major innovations in techniques (photoelectric detection; optical multi-channel detector systems; holographic gratings; piezo-electrically scanned Fabry-Perot interferometers; tunable dye lasers, etc.), the scope of the field is now vastly extended. Collective and localized excitations of vibrational, electronic or magnetic nature have become accessible with Raman and Brillouin spectroscopy. Extreme physical conditions—high-magnetic fields, ultrahigh pressures—are experimentally tractable. Time-resolved Raman spectroscopy has profound applications for systems evolving with-time scales as small as a Femto-second.

Personally I have thoroughly enjoyed the applications of Raman and Brillouin spectroscopy in semiconductor physics. Since the first report on the 'Raman scattering in silicon' by Russell[12], Raman scattering by polar phonons in compound semiconductors as well as their coupling with plasmons have been observed. Raman scattering from local modes; polaritons; donors and acceptors; free carriers in a magnetic field—all of these have extended and deepened our knowledge of semiconductor physics. Raman spectra of novel semiconductors like 'diluted magnetic semiconductors' and novel hetero-

structures (superlattices and multiple quantum wells) provide some of the most spectacular and delicate illustrations of the power of Raman spectroscopy.

In my talk I will focus on:

(1) **First and second-order Raman spectrum of diamond as well as the Brillouin components**[13,14]: Interpretation in terms of the dispersion curves for the lattice vibrations determined by Warren *et al*[15] using inelastic neutron scattering; absolute cross section of the Raman scattering; elastic and elasto-optic constants—these are the significant aspects of the phonon spectrum of diamond, a crystal of fundamental importance in condensed-matter physics.

(2) **Piezo-spectroscopy**[16] **and the unusual aspects**[17] **of the zone center optical phonons in α-quartz**: α-quartz has a rich Raman spectrum exhibiting one of the sharpest lines in Raman spectroscopy along with linear-q effects characteristic of polar phonons in crystals free of improper symmetry. Uniaxial stress effects—piezospectroscopy—is a powerful tool in the study of Raman spectra of crystals; this is illustrated with examples from the Raman spectrum of α-quartz.

(3) **Phase transition, mode softening, and zone-folding in crystalline benzil**[18]: Crystalline benzil ($C_6H_5COCOC_6H_5$) is an isomorph of α-quartz at room temperature. It undergoes a phase transition to a monoclinic (C_2^3) symmetry below T_c = 84 K. Associated with this phase transition the unit cell enlarges. The external modes of benzil show striking effects in the Raman spectrum.

(4) **Magnetic excitations in diluted magnetic semiconductors**[19]: Raman: electronic paramagnetic resonance and Raman: anti-ferromagnetic resonance are exhibited by the novel, tetrahedrally co-ordinated diluted magnetic semiconductors (*e.g.* $Cd_{1-x}Mn_xTe$). Mutual exclusion of Stokes and anti-Stokes spectrum; magnetically—and temperature-tuned resonance Raman Effect—such unusual, striking phenomena are displayed by these semiconductors.

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