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"Key and important developments in inelastic light scattering from semiconductors"

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INELASTIC LIGHT SCATTERING FROM SEMICONDUCTORS

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INTRODUCTION

As mentioned in the Preface of these Proceedings, one important aspect of the Workshop was to review recent advances in the field of inelastic light scattering from elementary excitations in semiconductors. To that end, the workshop organizers requested an introductory lecture from A. Pinczuk that would cover developments in what could be called the artificially structured materials era, which has occupied the last decade or so. But rather than just review recent work for this introductory chapter, the organizers felt a more comprehensive history of research developments would be appropriate, particularly as this had not been attempted before. E. Burstein's lecture on early developments in light scattering spectroscopy of semiconductors, together with M. Cardona's encyclopaedic knowledge of the field, naturally lead to their co-option in such a task. What follows is a general account of key and other important developments in the subject to date, biased to some extent by the knowledge and preferences of the contributors. We apologize in advance for any inadvertent omission of other major relevant research work.

EARLY DEVELOPMENTS

The subject of inelastic light scattering was first investigated theoretically by Brillouin,[1] who in 1922 published a study of light scattering by density waves in liquids, followed a year later by Smekal's fundamental work on two-level atoms.[2] As a result of his prediction that elementary excitations would produce shifts in the frequency of the scattered light, the inelastic scattering observed experimentally by Raman[3] in 1928 was often called the Smekal-Raman effect, but is now more commonly known as Raman scattering. In 1925, Kramers and Heisenberg[4] developed the old-style quantum theory of Smekal to derive a scattering formula from classical wave theory by means of the correspondence principle. The Kramers-Heisenberg intensity relation was of fundamental importance in the development of quantum mechanics. Then, in 1927, Dirac[5] rederived these results using his quantum theory of radiation.

Light Scattering in Semiconductor Structures and Superlattices
Edited by D.J. Lockwood and J.F. Young, Plenum Press, New York, 1991

The announcement of Raman's discovery in 1928 led to an intense flurry of activity in light scattering spectroscopy, as well as the Nobel Prize in Physics for Raman in 1930. Raman had investigated inelastic light scattering from vibrations in liquids and solids, and Landsberg and Mandelstam[6] almost simultaneously announced the same effect in quartz. As a result of this latter work, Raman scattering is referred to as combination scattering in Russian literature. Other notable advances at this time were: an interpretation by Rocard[7] of the Raman effect as a modulation of the electric dipole moment by vibrational modes of the oscillating molecule; Wood[8] re-examined luminescence spectra dating from 1906 and found features due to Raman scattering in quartz, and he also introduced the term anti-Stokes scattering; in 1930 Robertson and Fox, and Ramaswamy independently reported the first-order Raman spectrum of diamond,[9] which could also be considered as the first Raman measurement on a "semiconductor"; the second-order Raman scattering from NaCl was published by Rasetti[10] and interpreted in terms of a second-order polarizability; in 1933 Born and Blackman[11] introduced an anharmonicity mechanism for second-order infrared and Raman processes. The work of this period culminated in the seminal paper by Placzek,[12] published in 1934, wherein he presented a phenomenological theory for vibrational and electronic Raman scattering in terms of the first- and higher-order polarizabilities. The first experimental observation of electronic Raman scattering (in gaseous NO) had been reported earlier in 1930 by Rasetti.[13]

Not long after the experimental discovery of the Raman effect, Gross[14] found a triplet light-scattering spectrum in liquids. The frequency shift of the outer components was much smaller than in previous vibrational scattering, consistent with the expected acoustic wave scattering predicted earlier by Brillouin and independently by Mandelstam.[15] These two lines comprise what is now commonly called the Brillouin (or Mandelstam-Brillouin) spectrum. The central elastic component was explained by Landau and Placzek[16] as scattering from nonpropagating density fluctuations.

Other key stepping stones and milestones along the way in this early pre-laser period include: the discovery by Raman and Negungadi[17] of Raman scattering from a soft mode associated with the α-β phase transformation in quartz; the Born-Bradburn theory[18] of second-order Raman scattering in alkali halides couched in terms of the polarizability of atoms and the dependence on the change in distance between pairs of atoms; Krishnan's detailed study[19] of the first- and second-order spectrum of diamond; the analysis by Couture and Mathieu[20] of the "polarization" character of the scattering for different orientations of crystals and for different directions of the incident and scattered light; the observation by Couture-Mathieu and Mathieu[21] of Raman scattering from longitudinal optical (LO) phonons as well as transverse optical (TO) phonons in ZnS; and Poulet's explanation[22] of the I_{LO}/I_{TO} intensity ratio in ZnS (or other piezoelectric crystals) in terms of atomic displacements and electro-optic contributions to the Raman tensor (later referred to as the Faust-Henry coefficient). There was also much fine experimental work on the second-order Raman spectrum of alkali halides (see, for example, Refs. 19, 23-25). At the end of this pre-laser period, the seminal theoretical work of Loudon[26] on light scattering from solids was published. Loudon developed a microscopic model for Raman scattering in non-polar and polar crystals, summarized the Raman tensors for all crystal classes, discussed the electro-optic tensor in terms of local electric fields, demonstrated the cancelation of electron and hole contributions to Raman scattering in the zero wave vector limit, etc. Like Placzek's earlier polarizability theory, this work had far-reaching consequences.

POST-LASER DEVELOPMENTS

The introduction of the laser light source in the 1960s revolutionized the experimental methods of light scattering spectroscopy. New developments in grating spectrometry and Fabry-Perot interferometry soon followed. These new devices coupled with extremely sensitive photomultiplier pulse-counting and (later) multichannel detector

systems allowed many new discoveries to be made, including, for example, the very weak scattering of light by magnons.[27] In this part of the review we consider developments mainly in the semiconductor field, but key milestones in related areas are also noted. The particularly recent subjects of quantum wells, superlattices and time-resolved studies are discussed separately.

1962—1971

The importance of the laser as a light scattering source was vividly demonstrated in the early 1960s. Stimulated Raman,[28] Brillouin,[29] and Rayleigh[30] scattering were soon discovered, and a historical account of the early theoretical and experimental research into these stimulated effects has been given by Bloembergen.[31] In 1964, Hobden and Russell[32] reported on the first application of a laser as a source to excite the Raman spectrum of a semiconductor (GaP), and Jones and Stoicheff[33] observed the inverse Raman effect. Then in 1965, Maker and Terhune[34] pioneered the technique of what is now called coherent anti-Stokes Raman spectroscopy (CARS); Terhune et al.[35] observed the hyper-Raman effect (a three-photon process) in water; Russell[36] observed photographically the He-Ne laser excited first- and second-order spectrum of CaF_2 and GaP and, significantly, the third-order spectrum of GaP; Krishnan and Krishnamurthy[37] analyzed the GaP spectrum; Burstein and Ganesan[38] developed a method for determining the Raman matrix element from the electric-field-induced infrared absorption in diamond; Russell[39] recorded the laser Raman spectrum of an opaque material, Si, for the first time in backscattering; Birman[40] derived selection rules for two-phonon Raman scattering; Kleinman[41] made use of polarization effects to determine which Raman-active irreducible representations are present in two-phonon spectra; and Henry and Hopfield[42] observed laser Raman scattering from lower-branch polaritons in GaP.

In 1966, Damen et al.[43] developed the now standard polarization notation used to describe Raman spectra when investigating the phonons in ZnO, in which they also observed polariton Raman scattering;[44] Anastassakis et al.,[45] from a measurement of electric-field induced infrared absorption in diamond, deduced the magnitude of the atomic-displacement Raman tensor to be $|a| = 4 \times 10^{-16}$ cm^2 (later, Grimsditch and Ramdas[46] found from direct measurement $|a| = 4.4 \pm 0.3 \times 10^{-16}$ cm^2); Wolff[47] studied theoretically Thomson and Raman scattering by mobile electrons in crystals and suggested the possibility of Raman scattering involving Landau levels; Wolff's theory was extended by Yafet[48] to predict a spin-flip Raman transition involving virtual transitions between the conduction and valence bands of InSb; Mooradian and Wright[49] observed phonon Raman scattering in the III-V compounds GaAs, InP, AlSb, and GaP, and also observed LO phonon-plasmon coupled modes in GaAs,[50] the first of many such studies; Leite and Porto[51] measured resonant phonon Raman scattering in CdS; Ganguly and Birman[52] analyzed the exciton enhancement of Raman scattering by optical phonons; and Giordmaine and Kaiser[53] observed Raman scattering from intense coherently-driven lattice vibrations in calcite (a forerunner of the pump-probe technique applied later to time-resolved studies).

Developments in 1967 included measurement of the Raman spectrum of Ge[54] and electronic light scattering from impurity levels in Ge;[55] the spontaneous scattering from magnetic levels in InSb[56] including transitions between Landau levels and the spin-flip transition; phonons in ZnSe, ZnTe and InSb;[57] electric-field induced scattering by odd-parity soft optical phonons in $KTaO_3$ and $SrTiO_3$;[58] and soft phonons in $BaTiO_3$ including the first use of backscattering in a transparent crystal.[59] There followed in 1968 an experimental and theoretical analysis of Raman scattering in SiC polytypes,[60] which involved the first considerations of zone folded modes and can be considered a natural precursor to the artificial superlattice work of the 1980s; a study of light scattering from single-particle electronic excitations;[61] the observation of extremely strong spin-flip Raman scattering by donor and acceptor impurities in CdS;[62] and the observation of space-charge

electric-field induced Raman scattering by LO phonons in the surface depletion region of InSb.[63] The first of a new series of international conferences on light scattering in solids was held in New York in September 1968, and many important developments in the light scattering field were reported in the proceedings of that meeting.[64]

In 1969 there were reports on phonons in Se;[65] the polariton formulation of exciton enhanced Raman scattering by LO phonons;[66] multiple LO phonon peaks due to resonant Raman scattering in CdS and the relationship to hot luminescence;[67] followed, in 1970, by reports on multiple LO phonon peaks in various zincblende and wurtzite semiconductors;[68] the Franz-Keldysh mechanism for space-charge electric-field induced scattering by LO phonons;[69] inelastic light scattering from semiconductor plasmas in a magnetic field;[70] strain-induced shifts and splittings of the first-order Raman line in Si;[71] stimulated spin-flip Raman scattering in InSb;[72] and electric-field induced Raman scattering in diamond.[73]

The second international conference on light scattering in solids was held in Paris in July 1971 and again many new advances were reported.[74] Other significant advances reported elsewhere include measurement of the Raman spectrum of graphite;[75] examination of morphic effects due to external forces on the infrared and Raman spectra of optical phonons;[76] a theory of wave-vector dependent exciton resonant-Raman scattering mediated by the Fröhlich interaction;[77] the observation of resonant light scattering by single particle spin-flip and collective charge-density excitations in GaAs;[78] a theory of interband electronic Raman scattering in semimetals and semiconductors;[79] a report on spin-flip Raman scattering from conduction electrons in CdSe and ZnSe in a magnetic field;[80] an investigation of wave-vector dependent (Fröhlich) Raman scattering by odd-parity LO phonons in Mg_2Si, Mg_2Ge, and Mg_2Sn;[81] and measurement of the Raman spectrum of α-Sn.[82]

1972—1990

After ten years of post-laser Raman spectroscopy, applications in solids had become routine and the number of studies in semiconductor systems had mushroomed. Because so much work has been done, only highlights of the later developments can be given here. In-depth reviews of this more recent period of research can be found in the series of Springer-Verlag volumes on "Light Scattering in Solids" edited by M. Cardona and G. Güntherodt.[83]

In 1972, Brillson and Burstein,[84] in a calculation of electric-field induced Raman scattering by LO phonons in an external magnetic field, showed that the intraband-Fröhlich-interaction Franz-Keldysh matrix element is proportional to the average separation of the electron and hole in the intermediate state; Geurts and Richter[85] observed interband electronic Raman scattering in InSb; and Yu and Shen[86] used a tunable dye laser to obtain the excitation profile for electric-field induced Raman scattering by LO phonons in InSb. Also observed were electric-field induced resonant Raman scattering by LO phonons in CdS,[87] resonant Raman scattering in Ge at the E_1 and ΔE_1 gaps,[88] and near-resonance spin-flip electronic Raman scattering in InSb.[89] Brenig et al.[90] investigated spatial dispersion effects in resonant Brillouin scattering from polaritons. In 1973, Compaan and Cummins[91] observed resonant Raman scattering by odd-parity LO phonons at the 1s yellow exciton in CdS; Doehler et al.[92] carried out measurements at 2 μm (i.e., in transmission) of Raman scattering by phonons and electronic impurity levels in Ge; Cardona[93] elaborated on Raman scattering as a form of modulation spectroscopy; and Evans et al.[94] observed Raman scattering from surface polaritons in a thin GaAs film on Al_2O_3, which lead to a subsequent theoretical analysis by Chen et al.[95] in 1975. Scattering by coupled phonon-electron excitations was observed in p- and n-type Si and quantitatively interpreted.[96,97] The line shapes exhibited Fano profiles with asymmetry parameters dependent on laser frequency. This work lead to the determination of the sign of the

Raman tensor of Si.[98] Vogt, in 1974, applied the hyper-Raman technique to solids, investigating first CsI[99] and later, odd-parity and "silent" phonons in $SrTiO_3$.[100]

The third international conference on light scattering in solids was held in Campinas in July 1975 and the maturity of the bulk-semiconductor light scattering field was evident from papers presented at that meeting.[101] In 1975, a definitive study of Raman scattering from phonons in the chalcopyrite archetype $AgGaS_2$ was reported along with an interpretation of chalcopyrite optic modes based on the Bettini lattice dynamical model,[102] and the first observation of Raman scattering from the upper-branch polariton in a semiconductor (ZnO) was also reported.[103] Zone folding in GaSe polytypes was studied in 1976,[104] as was resonant two-photon electronic Raman scattering in CuCl,[105] the use of resonant Raman scattering by LO phonons as a probe of the surface space-charge field at n- and p-type InAs surfaces,[106] and the lattice dynamics of the ordered vacancy compound $HgIn_2\square Te_4$, where the Brewster-angle Raman scattering technique was fully exploited for an opaque semiconductor.[107] Scattering by intervalley density fluctuations was observed in n-type Si in 1977.[108]

Light scattering by guided wave polaritons in GaSe was investigated theoretically[109] and experimentally[110] in 1978, and conditions were established for observing inelastic light scattering by intersubband charge-carrier excitations at inversion and accumulation layers.[111] Then in 1980 there were investigations of the coupling of intersubband charge-density excitations with LO phonons and the microscopic mechanisms for Raman scattering by intersubband single-particle non-spin-flip excitations and by intersubband charge-density excitations,[112] resonant Raman scattering at InAs surfaces in MOS junctions (observation of coupled LO phonon - intersubband charge density mode),[113] and electronic Raman scattering by shallow donors in CdTe and GaAs and its formulation in terms of exciton-mediated light scattering.[114] Very recently, double and triple resonances have been observed in the scattering by phonons in semiconductors[115] and absolute resonant cross sections have been interpreted using excitons as intermediate states.[116] Also, in the past couple of years, considerable experimental information has been obtained by measuring resonant Raman scattering by phonons in a strong magnetic field.[117]

SEMICONDUCTOR SUPERLATTICES

Phonons

The mini-Brillouin-zone (MBZ) of a superlattice can always be approximately considered as a folded version of that of the bulk material. The concept of folding, however, is particularly useful for branches whose bulk counterparts overlap and differ relatively little from each other. This is usually the case for most of the acoustic branches of the bulk materials. We can then describe the corresponding dispersion relations of superlattices as an average of the bulk ones, folded back to the MBZ, and with minigaps opening at the zone center and edges. The concept of "folding", however, is not very useful for the bulk optical modes, which are usually well separated in frequency in the two components (e.g., Ge-Si, GaAs-AlAs). In this case the concept of "confinement" becomes more useful: optical vibrations are localized to either one of the components and are non-dispersive throughout the MBZ (except for the so-called interface modes, see below). The distinction between folded and confined modes is not merely academic. Surely one can "fold" confined modes to the first MBZ but this folding is of no use since the modes are non-dispersive. Note that for superlattices composed of a thickness d_1 (d_2) of material 1 (2) the equivalent q-vector of folded acoustic modes at the center of the MBZ is

$$q = \frac{2\pi}{d_1 + d_2} \cdot \quad m, \quad m = 1,2,3,... \qquad (1)$$

while that of confined ones is

$$q = \frac{\pi}{d_1} \cdot m \quad \text{in medium 1} \qquad (2)$$

$$q = \frac{\pi}{d_2} \cdot m \quad \text{in medium 2}$$

These wavevectors are different unless $d_1 = d_2$.

The concepts of folding and confinement were used as being more or less equivalent since the first attempts at observing confined optical modes were made in 1977.[118] We have already mentioned above that the concept of folding has been used earlier[60] to describe light scattering in SiC polytypes, materials for which folding is appropriate to either optical or acoustic phonons.

The first conclusive observation of folded acoustic modes in superlattices was probably made by Colvard et al.[119] Their theory has been worked out at an early stage by Rytov,[120] in connection with the propagation of seismic waves through stratified media. Minigaps had been observed earlier by Narayanamurti et al.[121] by acoustic transmission spectroscopy and, more recently in amorphous superlattices by Santos et al. with the same techniques.[122]

Shifts in optical modes versus layer thickness, resulting from confinement, were reported in Ref. 118 and later by several Japanese groups.[123,124] Strong evidence for the formation of good quality superlattices, however, was first obtained through the observation of several optic modes under resonant conditions.[125,126] Even stronger evidence for these "confined" optic modes was obtained under non-resonant conditions for the $GaAs/Ga_{1-x}Al_xAs$ system by Jusserand et al.[127] In order to avoid the difficulties involved in the theoretical treatment of "phonons" in mixed crystals such as $GaAl_xAs_{1-x}$ Sood et al.[128] investigated $(GaAs)_l(AlAs)_m$ systems. These authors clearly spelled out the difference between "confinement" and "folding" which has been unequivocally used since then. They also investigated the polarization selection rules for these superlattices on and off resonance. In the latter case they found that for parallel polarizations (in backscattering) modes with m even (Eq. (2)) are seen. They identified the relevant electron-phonon coupling as the Fröhlich interaction (we would like to recall with respect and admiration the recent death of H. Fröhlich on January 23, 1991). For crossed polarizations parallel to cubic axes they observed modes with m odd and were able to prove that they were induced by the deformation potential interaction, equivalent to that at the top of the bulk valence bands. These selection rules settled unambiguously the question of boundary conditions for confined phonons in polar materials: like in the covalent counterparts (e.g., Ge-Si) the vibrational amplitude, not the electrostatic potential, must vanish at the interfaces.

Modes which have now become known as electrostatic interface modes were observed by Merlin et al.[126] who assigned them to long wavelength LO and TO modes propagating with in-plane $\vec{q}$ vectors, with frequencies given by the solution of $\langle \varepsilon \rangle = 0$ and $\langle \varepsilon^{-1} \rangle = 0$ ($\langle\ \rangle$ represents the arithmetic average over the two media). More detailed observations were performed by Sood et al.[129] who attributed them to "electrostatic interface modes". In the in-plane propagation limit and for $q \rightarrow 0$, these modes, which had been calculated by several authors,[130] coincide with those of Merlin et al.[126]

We note that the interface modes observed in Ref. 129 correspond to large in-plane wavevectors which cannot be produced in a near-backscattering process. Hence the contribution of interface roughness or other defects was invoked. This conjecture received

support from the observation that the scattering by interface modes was screened out for strong laser power densities.[131] Observations of similar modes have been also made by electron energy loss spectroscopy.[132] Only recently the dispersion relations of interface modes with a well-defined $\vec{q}$ has been measured by scattering on a bevel cut at the edge of the superlattice[133] and also on superlattices with an in-plane periodicity generated by a second, MBE-grown, vertical superlattice.[134]

The detailed nature of the interface modes has been the source of considerable confusion since their original observation in Refs. 126, 129. Its understanding is important for the treatment of the electron-phonon interaction. It has more recently become clear that interface modes are not "additional" vibrational modes but conventional modes of the superlattice considered as a crystal with a large primitive cell, affected by long-range electrostatic fields. This understanding has been reached through simple lattice dynamical models[135] and also through detailed lattice dynamical calculations based on force constants and effective charges carried over from the bulk.[136] The interface phenomena correspond to long-range electrostatic field effects for propagation with an in-plane component of q. Hence only infrared-active modes (for the space group of the superlattice) exhibit such effects. Such are the confined modes with m odd, in particular that with m = 1 (interface effects for m = 3 and larger are usually negligible). The confined m = 1 LO and TO modes for $\vec{q}$ along the growth axis evolve into interface modes when $\vec{q}$ is tipped towards the layer planes.

Most of the experiments on "confined" and "folded" phonons in superlattices have been performed for systems grown along the [001] axis. In these systems only LO modes are allowed for backscattering on the planes. TO modes can, however, be seen if the laser is incident on the superlattice edge.[137] More recently, experiments on superlattices grown along lower symmetry axes, such as [111], [110], and [012] have been performed.[136,138,139] In many of these cases LO, TO, LA, and TA modes can be observed. In all cases they map well onto bulk dispersion relations using Eqs. (1) and (2) or, in the case of confined phonons, a slight modification obtained by adding to m a number δ ($0 < \delta < 1$) to allow for a slight penetration into the forbidden layer.[140] These low-symmetry growth directions yield a large variety of possible space groups, especially in the case of the Ge/Si system.[141]

Multiphonon peaks are also observed in superlattices.[142] They usually correspond to overtones and combinations of modes near $\vec{q} = 0$ (confined modes) with m even, although in the case of short period superlattices m = 1 (interface) modes are also seen.[143] These peaks are particularly easy to observe near outgoing resonance, i.e., when the scattered frequency equals that of an edge exciton. Such resonances, including those for one-phonon scattering, were first observed and treated theoretically by P. Manuel et al.[144] However, no detailed understanding of the nature of these resonance and the resulting scattering cross sections is available to date. This is also the case for the folded acoustic modes whose Raman polarizability is known to be related to the photoelastic constants of both constituents.[145]

Among recent important developments in the field of Raman scattering by phonons in superlattices we mention the observation of confined modes with a fairly sharp bulk $\vec{q}$ vector in the mixed crystal layers of $GaAs/Ga_xAl_{1-x}$ superlattices.[146] These experiments prove that, in spite of the disorder, optical modes with a well defined $\vec{q}$ vector exist in bulk alloys of the Ga_xAl_{1-x} type.

Because of the available knowledge on the effect of strain on Raman phonons of bulk materials[147,148] it is possible to obtain the degree of strain in lattice mismatched superlattices using Raman spectroscopy.[149] The technique is particularly appropriate for Ge/Si superlattices because of the large mismatch (~4%) between the lattice constants of both constituents. Nevertheless, not only Ge-Si but also III-V and II-VI systems have been investigated.

Infrared investigations of superlattices have not been as productive as their Raman counterparts. However interesting results are beginning to appear.[150,151] Infrared spectroscopy enables one to obtain information about TO modes in [001]-oriented superlattices,[151] which is hard to obtain by Raman spectroscopy. The quantitative analysis of the data, however, requires cumbersome fits with the optical response functions of multilayer systems.[150]

We conclude by mentioning a number of review articles on phonons in superlattices which the reader may find useful.[152]

Electronic Excitations

Interest in light scattering by free carriers confined at semiconductor surfaces and interfaces was stimulated by a communication of Burstein et al. presented at the 14th International Conference on the Physics of Semiconductors.[111] This work considered the mechanisms of inelastic light scattering in semiconductors and pointed out that with resonant enhancements of the scattering cross sections, as demonstrated in experiments carried out with conventional lasers,[79,153-155] the method has the sensitivity required to observe elementary excitations of two-dimensional electron systems in semiconductors. This proposal led to the first observations of resonant light scattering by free electron systems in modulation doped GaAs-AlGaAs heterostructures.[156,157] During the last ten years light scattering research of free carriers in semiconductor quantum wells and superlattices has been extensive. Much of this work has been reviewed in Refs. 158-160.

Within the framework of the effective-mass approximation the light scattering mechanisms of free carriers at semiconductor interfaces are similar to those in the parent 3D systems. The basic light scattering processes that apply in bulk semiconductors were considered in Refs. 47, 48, and 161-164; and the first experiments, in III-V compounds, were reported in Refs. 50, 56, 61, and 165. This pioneering work stimulated many light scattering studies of free carriers in bulk semiconductors that have been reviewed in Refs. 158 and 166-168. From these studies light scattering emerged as a very flexible experimental method to study the single particle and collective excitations of semiconductors. The collective excitations are plasmons or coupled plasmon-LO phonon modes. In the 3D systems the single particle excitations are spin-density modes and in the case of multivalley semiconductors, like n-type silicon, there are also intervalley density fluctuations.

The large resonant enhancements of the light-scattering cross sections required to observe the excitations of low density (10^{10} - 10^{12} cm^{-2}), 2D electron systems are predicted for photon energies near the optical transitions between the valence and conduction states that contribute to the effective mass of the free carriers.[111,112] In the case of direct gap III-V semiconductors free electrons occupy states of zone-center conduction band minima. For these carriers and for shallow donor levels the relevant optical resonances occur at the fundamental E_0 and spin-orbit split $E_0 + \Delta_0$ gaps. For free holes in states of the valence band maxima and for shallow acceptor levels the most important resonance is at the fundamental gap E_0. In the case of GaAs-AlGaAs heterojunctions and quantum wells the optical gap energies are $E_0 \simeq 1.5$ eV and $E_0 + \Delta_0 \simeq 1.90$ eV, easily accessible with tunable visible and infrared lasers. In the case of free holes in Si the relevant resonance is $E_0 \simeq 3.4$ eV.[111,169] Light scattering by free electrons at Si interfaces should show resonant enhancements near the zone-boundary optical gap at $E_2 = 4$ eV. The intensities of collective modes display resonant enhancements at all the optical gaps.[111-113]

Most of the light scattering studies of free electron systems with reduced dimensionality have been carried out in the semiconductor multilayer structures grown by molecular beam epitaxy. Several successful experiments have also been reported in the case of space-charge layers at metal-insulator-semiconductor interfaces.[113,169,170] The systems investigated most extensively are in the n-type modulation doped GaAs-AlGaAs

heterostructures.[158-160] Studies of 2D hole gases have revealed some of the complexities of the energy levels and interactions of these systems.[171,172] Resonant inelastic light scattering has been a very useful spectroscopic tool in studies of periodic doping superlattices (nipi structures),[173-175] of shallow donor and acceptor states in quantum wells,[176,177] and of δ-doping layers.[178,179]

In recent work the light scattering method has also been applied in the investigation of the elementary excitations of one-dimensional electron gases.[180-183] We believe that light scattering will continue to have a broad impact in studies of free carrier systems with reduced dimensionality in artificial semiconductor microstructures and nanostructures. The papers on electronic light scattering presented in these proceedings cover some of the most exciting current research in the field.

TIME-RESOLVED STUDIES

The majority of Raman scattering experiments make use of continuous wave (CW) laser sources and are designed to elucidate the elementary excitation spectrum of a system of interest that is in equilibrium. As most of the contributions to this volume attest, this conventional light scattering technique is an extremely powerful means by which to study the influence of varying material parameters on a system's lattice and electronic excitation spectrum. Experimentally, it is usually the case that measures are taken to ensure the laser light used as a source for the scattering does not disturb the system of interest away from its equilibrium state (i.e., care is taken to ensure that the spectra are independent of the laser intensity).

By either increasing the intensity of the CW laser source, or by using pulsed laser excitation, it is possible to induce changes in the elementary excitation spectra, which then represent the non-equilibrium, laser-excited system.

High-intensity CW laser excitation was used by Pinczuk et al.[184] to study the Raman spectra of electron-hole plasmas in GaAs. By fitting the spectra with a finite-temperature Lindhard-Mermin dielectric response function they were able to identify both optic coupled plasmon-LO phonon modes, and perhaps more significantly, they also observed the acoustic plasma mode characteristic of a multi-component plasma.

Nather and Quagliano[185] performed an extensive Raman study of CW photo-excited plasmas in confined, bulk GaAs layers. They succeeded in obtaining both single particle and collective mode spectra over a large range of plasma densities and temperatures. They also extracted density, temperature and plasma drift velocities from high-quality fits of their spectra using a multi-component dielectric function calculation which (necessarily) included intervalence band transitions.

Q-switched YAG laser pulses were used by Vasconcellos et al.[186] to both excite and probe the electron-hole plasma resulting from two-photon absorption in the bulk of GaAs crystals as early as 1977. By studying the shape of the single particle spectra they were able to deduce the intensity dependent carrier temperature, averaged over the laser pulse width (a few nanoseconds). In 1979, Kardontchik and Cohen[187] employed nanosecond dye laser pulses to both excite and probe electron-hole plasmas in GaP at 2 K. They measured the excitation frequency dependence of both branches of the coupled electron-hole-plasmon-LO phonon modes, but were unable to quantitatively explain the spectra. They also commented on the narrow linewidth of these modes as compared to those observed in comparable density n-type doped GaP samples. Interestingly, in 1986, Yugami et al.[188] reported similar nanosecond studies of coupled mode spectra in photo-excited GaP at 80 K, and concluded that the photo-excited plasma was heavily overdamped.

The examples above demonstrate the utility of pulsed lasers as a means of producing and probing non-equilibrium semiconductor plasmas. However, the full potential of pulsed lasers for Raman scattering is realized only when a relatively intense "pump" pulse is used to induce the non-equilibrium state, and a second "probe" pulse, synchronized and delayed with respect to the pump, is used to scatter from the non-equilibrium system as it returns towards equilibrium. Owing to the coherent nature of the Raman process, the temporal resolution of such experiments is limited (except under resonant conditions) only by the laser pulse duration.

Depending upon the laser pulse duration and intensity, this time-resolved Raman technique can and has been used to study a number of interesting non-equilibrium processes in semiconductors and semiconductor structures. There is an extensive literature[189,190] on the use of nanosecond probe pulses to time-resolve the lattice temperature rise of Si irradiated by high intensity nanosecond pulses which ultimately cause melting of the Si surface. This was done by measuring the temporal evolution of the optical phonon anti-Stokes/Stokes scattering ratio, and making a number of complicated corrections for the temperature dependent parameters which influenced it.

Von de Linde et al.[191] were the first to use picosecond probe pulses to study the dynamics of individual LO phonon mode populations following the injection of an electron-hole plasma in bulk GaAs by means of a picosecond pump pulse. The basic technique introduced in their 1980 paper has been subsequently used by many others in different material systems and on different timescales. In their experiment, the pump pulse injects electrons into the central valley of the GaAs with an excess energy above the conduction band edge of ~ 500 meV. These electrons relax towards the band edge primarily by the emission of small wavevector LO phonons via the Fröhlich interaction. By monitoring the strength of the anti-Stokes component of the LO phonon Raman signal produced by a delayed probe beam (proportional to the mode occupation number), von der Linde et al.[191] were able to observe the buildup of LO phonons produced via this electronic relaxation process, as well as the subsequent decay of the non-equilibrium LO phonon population. The decay time represented a direct measure of the population lifetime of the LO phonons as determined by anharmonic decay processes into lower energy phonons. It was noted that this lifetime determined directly in the time domain, ~ 7 ps at 77 K, was consistent with the lifetime deduced previously from CW Raman linewidth measurements of the LO phonon mode in GaAs.

Kash and coworkers[192] subsequently used subpicosecond laser pulses to extend von der Linde et al.'s measurements to room temperature, at which the LO phonon lifetime was found to be only 3.5 ps. The improved temporal resolution also allowed them to estimate the electron-LO phonon emission time, ~ 165 fs, from the risetime of the non-equilibrium LO phonon signal. More recently Kash et al.[193] used picosecond Raman scattering to study the nature of the Fröhlich interaction in group III-V semiconductor alloys. By monitoring the peak non-equilibrium populations of both GaAs-like and AlAs-like LO phonon modes in AlGaAs, they were able to deduce the relative Fröhlich coupling strengths of the electrons with the two types of LO phonons. They concluded that the overall coupling of the electrons to the LO phonons was unaffected by the alloying and that the relative coupling strength could be accounted for using spectroscopically determined parameters in a calculation of the Fröhlich interaction strengths.

In a separate experiment, Kash et al.[194] compared the wavevector dependence of non-equilibrium LO phonon generation in bulk GaAs, $Al_{0.11}Ga_{0.89}As$, and a 50 nm thick slab of GaAs by time-resolving the anti-Stokes LO phonon signal observed both in forward and back scattering geometries. The absence of a non-equilibrium signal in the forward scattering geometry for bulk GaAs and $Al_{0.11}Ga_{0.89}As$ samples, together with a significant forward scattered signal in the thin GaAs slab, led Kash and coworkers to conclude that the wave vectors of Raman active LO phonons in AlGaAs are well-defined, and the corresponding modes are not spatially localized.

Tsen and Morkoç[195,196] extended the same technique to study the GaAs-like mode in GaAs/AlGaAs multiple quantum well structures. They deduced that, within

experimental error, the LO phonon lifetime was the same in 20-nm thick GaAs layers as it was in the bulk. They also concluded that the electron-LO phonon emission time was also very similar to that in the bulk.

Collins and Yu[197] devised an elaborate experiment in which the LO phonon generation rate, as deduced using von der Linde et al.'s technique, was studied as a function of the pump beam frequency. Discontinuities in the generation rate occurred at incident photon energies corresponding to the excitation of electrons into the conduction band with sufficient energy to scatter into the L and X (separate discontinuities) satellite valleys. Theoretical fits to their data were used to extract intervalley deformation potentials.

Young et al.[198] and Genack et al.[199] independently reported the results of optical phonon dynamic studies in the group IV material, Ge. Genack et al.[199] pointed out the fact that the lifetimes obtained in the time-domain were consistently longer than the corresponding inverse CW linewidths, over a range of temperatures up to 300 K, and postulated that this might be due to isotopic disorder in Ge. However, very recent results on an isotopically-pure Ge crystal[200] indicate that this disorder mechanism does not contribute to the optical phonon linewidth in naturally occurring Ge. By modeling the microscopic plasma and phonon dynamics in picosecond-laser-excited Ge, Othonos et al.[201] showed that the temporal evolution of the optical phonon population deduced from the Raman experiments[198] could be quantitatively understood using deformation potentials obtained from the literature.

All of the picosecond experiments described above basically involved monitoring the changing population of a well-defined optical phonon mode through its anti-Stokes Raman efficiency. There are also a number of examples in which the evolution of an entire portion of the spectrum (anti-Stokes and/or Stokes) is time-resolved. In the case of the single-particle spectrum from photo-excited plasmas in GaAs[202] and Si,[203] this information has been used to infer the temporal evolution of the plasma temperature,[202] and relative changes in its density.[203]

Another example of solid state plasma diagnosis using temporally and spectrally resolved Raman signals makes use of the coupling of photo-excited plasmons and LO phonons in zincblende materials. By using a 3 ps probe pulse to monitor the Stokes spectrum near the bare LO phonon line in InP and GaAs as an 80 ps pump pulse injected an electron-hole plasma, Young et al.[204,205] showed that the LO phonon spectrum broadened and reduced in intensity by a factor of ~ 5 times, and shifted up in energy by ~ 20 cm^{-1} for injected plasma densities of $< 5 \times 10^{17}$ cm^{-3}. By fitting these coupled plasmon-LO phonon spectra using a multi-component dielectric function calculation, the temporal evolution of the surface plasma density was deduced and described by a simple diffusion model. These results also explicitly point out a limitation on phonon dynamic studies in GaAs and InP; namely that the photo-excited plasma density must be kept well below 5×10^{16} cm^{-3} in order that the anti-Stokes signal intensity reflects only the occupation number of the mode, and not its renormalization via interaction with plasmons.

Time-resolved Raman scattering from intersubband electronic transitions in GaAs/AlGaAs multiple quantum wells was reported by Oberli et al.[206] They used pump and probe pulses of different frequencies to selectively inject electrons into different conduction subbands and to resonantly enhance the signal from different intersubband transitions. By monitoring the decay of the Stokes signal from the second to third subband transition when injecting carriers into both the first and second subbands, they deduced a second-to-first intersubband scattering time of ~ 325 ps when the subbands were separated by less than an LO phonon energy. They could not resolve the intersubband scattering rate with their apparatus (resolution ~ 8 ps) in thinner GaAs quantum wells in which the subbands were separated by more than an LO phonon energy.

More recently, Tatham et al.[207] used sub-picosecond pulses and different resonance conditions to time-resolve the anti-Stokes, second-to-first intersubband signal in a pump probe experiment similar to that of Oberli et al.[206] Using this technique they successfully resolved the intersubband relaxation rate in samples where the subband spacing exceeded

the LO phonon energy. It was found it to be ~ 1 ps, in good agreement with model calculations of LO-phonon assisted intersubband scattering rates, including multiple subband occupancies.

A different type of quantum well experiment was recently reported by Tsen et al.[208] They studied the lateral diffusion dynamics of electron-hole plasmas in GaAs quantum wells by spatially and temporally monitoring the intersubband Raman signal following local excitation. They found that the lateral ambipolar diffusion coefficient was ~ 120 cm^2/s at an ambient temperature of ~ 105 K, and that this diffusion coefficient was not sensitive to the GaAs quantum well width from 10 to 30 nm.

This brief review of time-resolved Raman studies of semiconductors and semiconductor structures is meant only to highlight some of the main techniques and the types of information that can be deduced using them. Much more detail of these techniques and new methods of extracting important dynamical information using time-resolved Raman scattering can be found in the five articles on this topic in these proceedings.

FUTURE PROSPECTS

The above portions of this chapter are intended to serve as background material, laying the foundation for the remaining chapters of this volume that summarize the current state-of-the-art in the field of light scattering in semiconductor structures. In comparing the articles that follow with examples from the historical review in this chapter, it should be clear that this field is now being driven to a large extent by the rapidly advancing semiconductor growth and fabrication technologies. As these technologies continue to expand at an increasing rate, their role in determining the directions of the light scattering field will continue to grow in the coming decade. To keep pace with these technological developments, advances in our fundamental understanding of the light scattering process are essential in order to take full advantage of the information provided by this powerful technique.

From the materials point of view, there will definitely be developments in smaller bandgap compound semiconductor materials compatible with opto-electronic/fibre optic applications. Silicon will surely remain the dominant electronic material, and there will be increasing demands to better understand the interfaces of Si with insulators, metals, and other semiconducting materials. For any of these materials, nanofabrication techniques will be applied to produce reduced-dimensional structures from the basic 2-D quantum well structures studied so extensively during the 1980s. In order to use light scattering to effectively study the nature of structures with multiple dimensions less than 100 nm in a variety of material hosts, new experimental methods and a better theoretical framework will have to be developed in parallel. Resonance enhancement will become increasingly important, requiring the development of more tunable light sources (at shorter and longer wavelengths) and associated detectors, as well as a quantitative understanding of resonant mechanisms in these structures. Also of practical importance is a theoretical understanding of selection rules (and the breakdown thereof), the fundamental overlap of Raman scattering with the general field of non-linear optics, and the development of electronic Raman theory to the same level as our current knowledge of phonon Raman scattering.

All of the above relate to the use of Raman scattering to elucidate the equilibrium properties of exotic new structures and materials. As the ultimate application of these structures is in novel electronic and opto-electronic devices, there will also be increasing applications of Raman scattering to help further our understanding of non-equilibrium transport processes in these devices.

Of course, these are all predictions based on extrapolation of past experience, and the most exciting developments to be reported in the year 2000 at a Workshop on Light Scattering in Semiconductors will undoubtedly have to do with developments that cannot be foreseen. What can be foreseen is that the future will be more exciting than we can predict!

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