MULTI-PHONON RAMAN SCATTERING

IN

SEMICONDUCTORS

by

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CERTIFICATE

This is to certify that the thesis entitled "MULTI-PHONON RAMAN SCATTERING IN SEMICONDUCTORS", which is being submitted by Miss C.S. Jayanthi for the award of degree of Doctor of Philosophy, to the Indian Institute of Technology, Delhi, is a bonafide record of research work. She has worked for the last four years and four months under my guidance and supervision.

The thesis has reached the standard, fulfilling the requirements of the regulations relating to the degree. The results obtained in this thesis have not been submitted to any other University or Institute for the award of any degree or diploma.

KPJain

(K.P. JAIN)

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C.S Jayanthi
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PREFACE

This thesis is devoted to a study on Raman line-shapes and oscillator strengths of a wide-variety of semi-conductors. Some of the recent resonance experiments have been analysed with this in view. This sort of spectroscopic study can be generally used as a tool to probe into the nature of electronic states and to understand interactions present in solids. It is the purpose of this thesis to understand electron-phonon interactions and to investigate the nature of resonant intermediate state in various Raman experiments.

Raman spectroscopy uptil now has usually investigated phonons, since the scattered photons in this case lie within observable energy range. In the present work, two-phonon and multi-phonon Raman scattering have been studied and accordingly there are two parts to the current investigation. The first part of the thesis deals with line-shapes of 2-phonon Raman spectra in the vicinity of an indirect gap, while the latter half focusses attention on anomalous intensities of multi-phonon lines.

The entire work of this thesis is presented in five chapters and the chapterwise summary of each of these is given below:

The first chapter is the introduction. This we begin by giving an account of the current-status of the field. After this review, a Kubo-type formula for light scattering is derived. It is meaningful to present this formal theory, as the focus in this thesis is on understanding the line-shape of Raman spectra. In a later section of this chapter, this formalism has been used to calculate Raman cross-section for a specific case of Wannier excitons, as the resonant intermediate state.

Line-shape studies of 2-phonon Raman spectra in the vicinity of an indirect gap show some unusual effects. It is seen that select pairs of phonons are enhanced more than rest of the 2-phonon spectra and this results in distortion of its line-shape. In Chapter-II, a theory is formulated which accounts for this feature in a phonon-assisted transition. Studies of this kind are particularly significant in Si and GaAs for which a comparison of theory and experiment is made. The results of this chapter are useful in obtaining information about electron-two-phonon interactions.

The origin of multi-phonon Raman spectra (MRS) is discussed at length in Chapter-III. The observed intensities of multi-phonon Raman lines are incompatiable with predictions of perturbation theory and cannot also be understood completely with the other existing models on MRS.

It is suggested that the results of MRS experiments can be understood if the resonant intermediate state is considered as a quasi-bound exciton-phonon system (EPQBS). MRS is then explained on the basis of spontaneous decay of EPQBS into continuum states of the free exciton, which may be accompanied by emission of one or more phonons. The Raman matrix-element for such a process is calculated and the relative intensities of the various harmonics are then compared with the experimental results of CdS.

Chapter-IV discusses the effects due to coupling of two phonon channels in a multi-phonon Raman experiment. Such mode-coupling might become important in a multi-channel optical response of the system. A many-body calculation is given here to describe this phenomenon. In this calculation Raman cross-section is expressed in terms of a generalized dynamic form factor, which is related to a composite Green's function. The composite Green's function so defined, takes into account interference of the two discrete channels and one then solves for different terms of this by summing the various relevant diagrams. It is shown that such an interference results in an oscillator strength transfer from one mode to the other. A general condition for comparable intensities of the two interfering channels has been obtained and from this the exciton-phonon coupling strength can be estimated.

In the last chapter we have summarized and discussed the main results of this thesis.

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