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The work presented in this thesis can be divided into two main parts: time resolved spectroscopy and laser induced photoconductivity with particular emphasis to the nonlinear processes. The whole thesis has been divided into five Chapters. The first two chapters deal with the development of a laser fluorimetric system using pulsed N₂-laser and its application to the study of deactivation process and energy transfer mechanism. The last three chapters deal with the photoelectronic properties of CdI₂ and ZnS crystals using different lasers.

CHAPTER I deals with the development of a laser fluorimetric system based on single photon counting technique. This unique set up has got many advantages outlined in the thesis over the conventional phase shift and sampled single photon detection methods. It consists of a pulsed N₂-laser, a reference signal fast discriminator, a fluorescence signal fast discriminator, two fast photo-multipliers, a nanosecond delay unit, a time-to-pulse height converter, a multichannel analyser (as a pulse height analyser) and a printer. The working of the system has been tested by recording the intensity-time profile of the excitation source and the fluorescent decay profile of different laser grade dyes. The adaptation of the system to lifetime measurements has been examined by studying the fluorescence time behaviour of 8 x 10⁻⁷ M
solution of anthracene in benzene. The accuracy of measurement has been tested by a quenching experiment with rhodamine 6G as the fluorescent medium and potassium iodide in its aqueous solution as the quencher.

CHAPTER II emphasises mainly on the applications of the laser fluorimetric system. The solvent deactivation of riboflavin in the environment of polar and nonpolar solvents has been studied using this system. The deactivation of riboflavin excited state has been understood in terms of an internal conversion process. The system has further been employed to investigate the energy transfer mechanism in (i) Rhodamine 6G — Rhodamine B and (ii) Coumarin 30 — Rhodamine 6G dye mixtures. In the former case the study shows the energy transfer to be due to dipole-dipole resonance type interaction; while in the latter case it was found to be radiative (emission-reabsorption) type. The accuracy of the detection system which was found to be in the subnanosecond range has been very helpful in ascertaining the above mentioned properties in comparison to some of the studies on these mixtures made by earlier investigators.

CHAPTER III includes the photoelectronic properties of CdI$_2$ single crystals carried out using N$_2$-laser pumped dye lasers. The spectral distribution of photocurrent has been recorded and the same has been explained by proposing an
energy level scheme for CdI$_2$. The nonlinearity of the lux-ampere characteristics has been interpreted in terms of the Rose model. The current-voltage characteristics suggest the contribution of space charge limited current in the investigated crystal. A combined investigation of the intensity dependence of carrier lifetime and of photocurrent has been used to understand the trap distribution in the band gap of the crystal. The CdI$_2$ crystal exhibited "thermal quenching" behaviour at about $390\,\text{K}$ and the same has been explained on the basis of the well known Rose model.

CHAPTER IV concerns with the study of two photon photoconductivity in CdI$_2$ using two dye laser beams. The main aim behind this study is to gain information about the additional states/bands, if any, in the crystal. The study indicates the enhancement of photocurrent when the sum energy of the laser photons ($\hbar\omega_1 + \hbar\omega_2$) becomes $0.66\,\text{eV}$ above the band edge. The results reveal the presence of a valence band at a separation of $0.66\,\text{eV}$ below the upper most valence band, wherefrom one photon transition is allowed. The intensity dependent measurements suggest the linearity of photocurrent when intensity of one laser beam is fixed and that of the other is varied.

CHAPTER V describes the optoelectronic properties of ZnS polycrystal under pulsed laser excitation. The
photoconductivity spectrum has been recorded using dye lasers and its correlation to the luminescence spectrum reported by earlier workers reveals that the donor levels associated with donor-acceptor pairs are responsible for the observed photoconductivity bands in the long wavelength range. The nonlinearity of the lux-ampere characteristics indicates the presence of space charge and the same has been confirmed by studying the current-voltage characteristics. This chapter also describes:

(i) four photon photoconductivity in ZnS crystal using Nd : YAG laser;

(ii) one photon and two photon photoconductivity using frequency doubled Q switched Nd : YAG laser, and

(iii) one photon photoconductivity using XeCl excimer laser.

The relaxation curves of photocurrent of ZnS exhibited hyperbolic type decay with two distinct regions. The fast response of the relaxation curve suggests the absence of trapping levels close to the equilibrium Fermi level and a comparatively low trap densities.