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Two photon absorption in anthracene crystal with a nitrogen laser pumped dye laser

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Abstract. Experimental studies are reported on the blue fluorescence signal ($^1B_2u \rightarrow {}^1Ag$) of anthracene single crystal excited by Nitrogen Laser and Nitrogen laser pumped dye laser (NLPD Laser). The experimental method was based on the fluorescence signal intensity measurements induced by single and double photon absorption. The crystal was excited by Nitrogen laser pumped dye laser having 4·5 ns (FWHM) pulse duration and approximately 10 KW peak power. Two photon absorption cross section is calculated by varying the incident photon energy. Observations were taken at room temperature and at liquid nitrogen temperature.

1. Introduction

With the advent of high power laser sources, non linear interaction of radiation with matter became amenable to the experimental observations The two photon spectroscopy provides new information concerning the properties of system levels which are not assessable by single photon transistions. The present work reports the two photon absorption phenomena by studying the induced fluorescence in anthracene single crystal. In the previous studies on two photon absorption in anthracene crystal, high power lasers such as Q-switched Ruby laser (Singh 1965, Webman 1969) and Neodymium laser (Fröhlich 1967) as well as nitrogen laser pumped dye laser (Bergman 1972) have been used. The present report summarizes our observations in both single and double photon absorption in anthracene single crystal when excited by nitrogen laser and nitrogen laser pumped dye laser.

The dependence of two photon induced fluorescence intensity on incident two photon energy was carried out in the range 3·8 eV to 4·6 eV for crystal at 300°K and 77°K. The relative two photon absorption cross section has been calculated from the relation $F = \eta n V \sigma I^2$, (Bergman 1972) where η is the fluorescence quantum yield for emission from the crystal, n is the molecular number density, V is the irradiated volume, I is the intensity of the incident radiation and F is the fluorescence intensity.

2. Experimental set up

Figure 1 gives the complete experimental set up for recording the fluorescence signal from the anthracene single crystal when irradiated by nitrogen laser pumped dye laser. The nitrogen laser, (Blumlein type design) with characteristic power output of 200 KW and a pulse width of 8 ns (FWHM) has been used in this present set up.

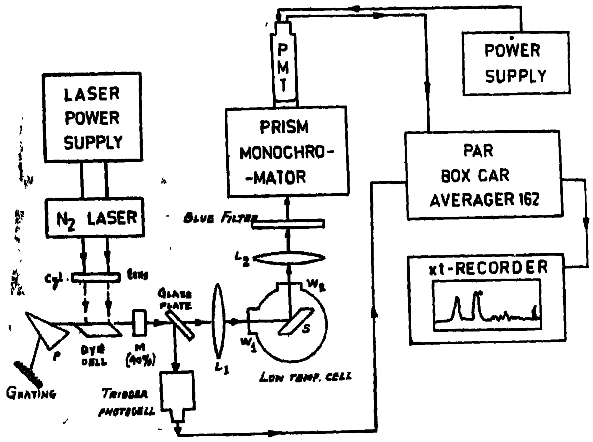

Figure 1. Experimental set-up for recording the two-photon induced fluorescence signal.

This nitrogen laser is used to pump the dye laser. The dye cavity is a Hänsch type (Hänsch 1972) In our experiments we have used (i) Rhodamine 6G, (ii) Rhodamine B, (iii) Sodium Fluorescein dyes. Table 1 shows the charac-

Table 1. Dyes used with their respective concentrations and wavelength ranges

No.	Dyes used	Concentration	Solvant	Peak wave length (nm)	Wavelength range covered (nm)
1.	Rh 6G	7×10^{-3} M/litre	Ethanol	594	570–610
2.	Rh B	$7{\cdot}5 \times 10^{-3}$ M/litre	Ethanol	622·5	600–642
3.	Na F	5×10^{-3} M/litre	Ethanol	547	532–560

teristics of these dyes. 40% reflectance mirror and a plane reflectance grating with 1800 lines/mm (obtained from Bausch and Lomb) forms the cavity with prism as a beam expander. The grating is rotated by a calibrated cosec drive. The output power of the dye laser was estimated with a PIN-Photodiode (hp5082-4204) and a Tektronix 585A oscilloscope. The pulse height being directly proportional to the intensity of the laser, pulse height was taken for the estimation of two photon absorption cross section.

The crystal was mounted on a goniometer such that the laser beam is always perpendicular to its ab-plane so as to get the maximum two photon induced fluorescence signal (Hernandez 1967). The single crystalline property of the crystal was confirmed by taking a Laue pattern. The purity of the crystal was checked by analytical methods. The presence of only blue fluorescence from the crystal itself is a good test for the purity for the same (Windsor 1965). In order to carry out the observations at 77°K the crystal was mounted in contact with a cold finger in a low temperature cell. The laser beam was focussed on to the crystal with a convex lens ($f = 10$ cm). The fluorescence signal was collected by a lens of focal length 5 cm and focussed on to the entrance slit of the prism monochromator. A corning blue filter (peak transmission at 4,390 Å) was inserted to cut off all the scattered laser light from the fluorescence signal. The fluorescence signal was detected with EMI 6255 fast photomultiplier in conjunction with a signal averager PAR model 162 main frame and a gated integrator model 164 along with a strip chart recorder. A similar experimental set up was used to study the blue fluorescence in Anthracene crystal excited with N_2 laser.

3. Results

Figure 2 shows the fluorescence spectra obtained when the crystal was irradiated by nitrogen laser both at room temperature and liquid nitrogen temperature (77°K). We observe an intense peak at 23249 cm^{-1} at room temperature and in addition to this, at 77°K, we observe a peak at 24490 cm^{-1} as reported earlier (Singh 1965). Plots of two photon absorption cross section against two photon energies at room temperature and 77°K is given in Figure 3. Our results are in agreement with the previous results, as far as the positions of the peaks and the general trend of the curve is concerned.

As we have estimated the relative values of two-photon absorption cross section, the comparison between our values and those reported by Bergman (1972) is not possible. However the two photon absorption cross section has its maximum value at 3·93 eV in the present report whereas Bergman observed it at 4·1 eV. The two photon absorption cross section is a function of laser line width. Theoretical prediction of dependence of two photon absorption cross section on line width is reported by Wong *et al* (1977). However, the effect of line width of exciting radiation was not considered in two photon absorption studies in anthracene by previous workers. In the present set up, different dyes were used in different regions. Hence there are variations in the laser line width, and only after making the necessary correction for line width the proper correction for the two photon absorption cross section is made.

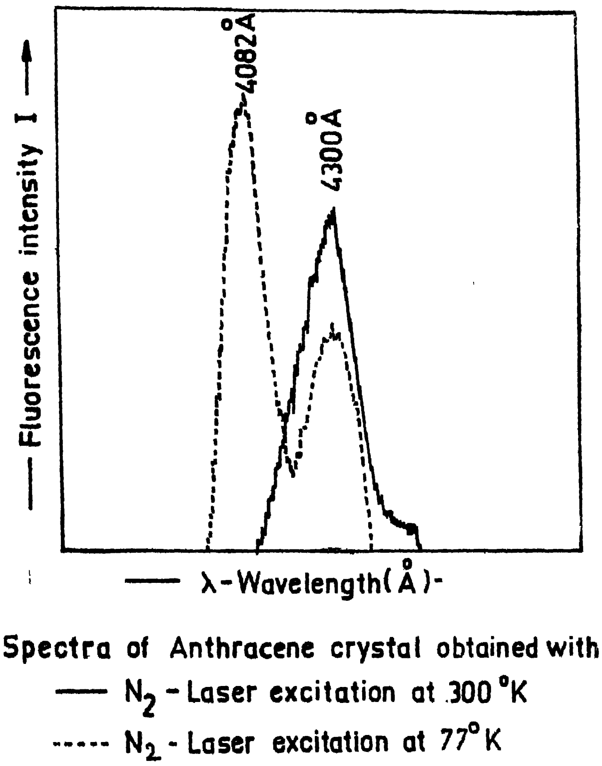

Figure 2. Spectra of anthracene crystal obtained with nitrogen laser excitation at room temperature (300°K) and at liquid nitrogen temperature (77°K).

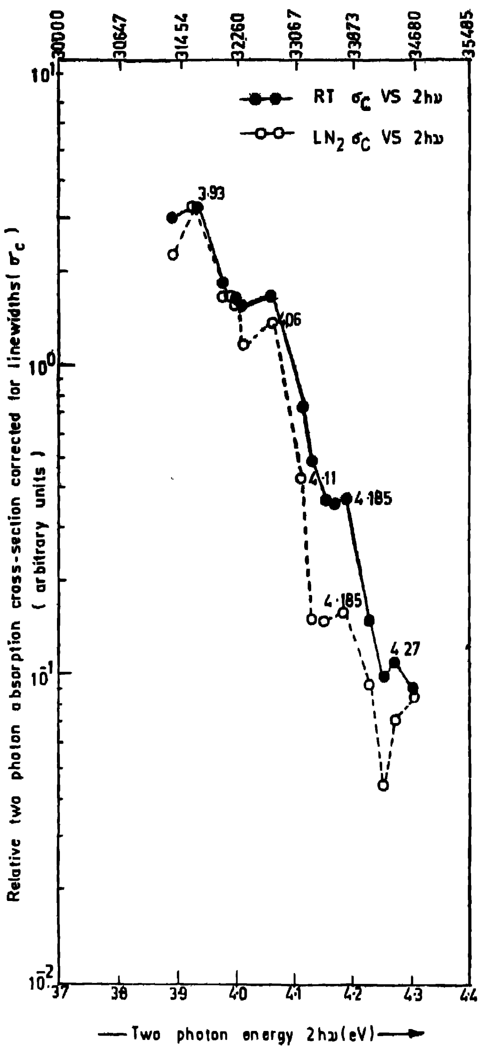

Figure 3. Plot of relative two-photon absorption cross-section (arbitrary Units) against incident two-photon energy (eV) at 300°K and 77°K.

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