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Transient excited singlet state absorption in Rhodamine 6G*

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Abstract. Transient excited singlet state absorption (ESSA) has been studied in Rhodamine 6G in ethanol using a nitrogen laser and nitrogen laser-pumped dye laser. Broad absorption with several submaxima and possible shoulders, which represent the vibrational structure, has been observed in Rhodamine 6G in the region, 4175–4640 Å. The position of the lowest vibrational level of the first excited singlet state S_1 has been determined from the crossing point of the long and short wavelength spectral wings of absorption and fluorescence respectively. The energy level scheme of the molecule has been obtained with the help of the absorption and fluorescence spectra recorded. The observed structure in ESSA has been tentatively interpreted to be due to transitions from the different vibrational levels of S_1 to one or more vibrational levels of the upper singlet electronic state S_4.

Keywords. Rhodamine 6G; excited singlet state absorption; absorption spectra; fluorescence spectra.

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1. Introduction

Laser dyes like Rhodamine 6G are a class of organic molecules which show lasing action under appropriate conditions of excitation. Dye lasers are among the most useful types of lasers because of their easy tunability, wavelength coverage and simplicity (Schafer 1983; Pavlopoulos and Hammond 1974; Sorokin *et al* 1967; Sorokin *et al* 1968; Shank 1975; Peterson 1979). Dyes can be used in solid, liquid or gaseous phase and their concentration and hence their absorption and gain can be readily controlled. Solutions of dyes are especially convenient for experimental studies. Different scintillators, coumarins, xanthines, oxazines and polymethines can be conveniently chosen to cover a wide range of the spectrum from 3200 Å to 13000 Å.

The ground electronic state of a dye molecule is in general a singlet state (Schafer 1983). The first excited singlet state is generally higher than the lowest triplet state. Dye molecules, when excited to the higher singlet states, relax by nonradiative interactions to the lowest excited singlet state S_1 in a short time of the order of picoseconds. This is the only state from which dye molecules have been observed to fluoresce, which, in most cases, have lifetimes of a few nanoseconds. Also this is the state from which the radiative transition that is directly responsible for laser action takes place under appropriate conditions of stimulation in all laser dye molecules.

Though singlet to triplet state inter-system transitions are generally forbidden, relaxation of molecules in the S_1 to the lowest triplet state T_1 is possible through