Thermal Lens Detection of One and Two Photon Absorption of Benzene in Cryogenic Liquid Solutions

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In thermal lens a molecule is excited with a laser (pump) and releases the energy to return to the ground state. The heat produced through non-radiative relaxation processes increases the temperature along the excitation region. A localized non-uniform distribution of the refractive index is created and acts like a concave lens dispersing a beam of a second laser (probe) which passes through that region. The decrease of the probe beam intensity is proportional to the absorption process. The pump and probe thermal lens technique has been found to be very sensitive for detecting samples of low concentration in transparent liquefied gas solvents at low temperatures. The C-H fifth vibrational ($\Delta\upsilon = 6$) overtone of benzene in liquefied ethane, argon, and krypton have been recorded at concentrations between 10 and 58 ppm in a range of temperatures between 87 and 150 K. Thermal lens experiments with two photon excitation are presented to show that the limit of detection could be increased. Figure 1 shows the two photon excitation scheme. The excitation laser promotes the C-H bond from the ground state ($\tilde{X}\ \ {}^1A_{1g}(\upsilon = 0)$) to the fifth vibrational overtone ($\tilde{X}\ \ {}^1A_{1g}(\upsilon = 6)$) of the ground electronic state. A second laser of a different wavelength is used to promote the vibrationally excited molecule to an excited electronic state ($\tilde{A}\ \ {}^1B_{2u}$). Non-radiative relaxation to the ground state is detected using the thermal lens method. The two photon signal enhancement with respect to single photon excitation under the same experimental conditions will be discussed.

Figure 1