

Evaluation hyperpolarizability by hyper-Rayleigh scattering method under photo excitation.

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Optical enhancement or decrease of the nonlinear optical (NLO) properties of organic compounds is expected at the excited state due to changes in dipole moments and oscillator strength caused by electron density distribution different from that at the ground state. It is necessary to evaluate NLO properties at the excited state for development and practical application of highly functional organic molecules.

In Chapter 2, hyper-Rayleigh scattering (HRS) measurement system was constructed to evaluate hyperpolarizability β of molecules in solutions both at the ground and excited states.

In Chapter 3, the increase of HRS intensities was observed under photoexcitation of p-nitroaniline (pNA) with argon-ion CW laser. It was attributed to the excited state molecule.

In Chapter 4, the dual-HRS measurement system was constructed for evaluation of the β -value at the excited state with high sensitivity and accuracy. This system made measurements at low power probe light intensities.

In Chapter 5, a ns tunable pump laser was employed for excitation. The HRS intensity for pNA in methanol solutions increased upon pulsed laser excitation at 464 nm.

The HRS intensity depended on the delay time between the pump and the probe pulses. The β value for pNA at the excited triplet state was evaluated in Chapter 6.

In conclusion, we constructed several new HRS measurement systems to evaluate hyperpolarizability at the excited state with CW and pulsed laser excitation. We clarified the β value of pNA actually increased upon laser excitation. The present excited state HRS experiments will open a new field of excited second order nonlinear optics and will contribute a great deal to develop novel highly functional materials at the excited state.