Measurement of Negative Nonlinear Refractions on Rhodamine B Dye Solutions by Nonlinear Ellipse Rotation Signals

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Most of $\pi$-conjugated organic materials widely studied for nonlinear optical applications present positive real component of the refractive third-order polarizability, $n_2$. However, due to important benefits for nonlinear optical applications, negative nonlinearity is also important to be found and studied. There are only a few organic systems studied in the past that present negative nonlinearity and then new organic molecular architectures are proposed [1]. Surprising, here we observed that well-known rhodamine laser dyes also can present negative contribution for the refractive nonlinearity. Preliminary measurements at 800 nm with femtosecond pulses using nonlinear ellipse rotation have shown this negative contribution on rhodamine B. We note a linear change as a function of dye concentration and, for very high concentration; we have measured also a negative effective $n_2$ for a methanol solution, when the negative nonlinearity of the dye overtake the positive contribution of the methanol. We could observe this effect by means of nonlinear ellipse rotation (NER) measurements for thick samples [2] where we are able to measure locally the nonlinear refraction of the solution without the interference of the cuvette. It is well known that laser dyes need to be dissolved in liquid solvents and held in a cuvette in order to be characterized, and then, the nonlinearities of the solvents and the cuvette walls need to be taken into account. For instance, most of measurements methods have difficult to remove the contribution of the cuvette except for the case of solutions with very high nonlinearities, where the contribution of the cuvette can be disregarded, and NER measurements using tightly focused laser beam since the nonlinearity of the solution can be directly measured. New measurements of $n_2$ in other laser dyes as a function of wavelength are been conducted to understand the origin of this negative contribution of rhodamine B. Tunable femtosecond pulses from an optical parametric amplifier pumped by amplified laser system (Dragon, KM Labs) at 1 kHz repetition rate are been used for these experiments. Theoretical nonlinear Kramers-Kroning and three-level energy model are been used to try to explain our results. [1] R.L.Gieseking, T.R.Ensley, H.Hu, D.J.Hagan, C. Tisko, E.W.VanStriland, and J.L.Bredas, “Nonlinear optical properties of X(C6H5)4(X=B-,C,N+,P+): A new class of molecules with a negative third-order polarizability.” JACS 137, 9635-9642 (2015).