All-optical orientation of state-selected OCS molecules

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A sample of aligned or oriented molecules can serve as a quantum system for investigating the anisotropic dependence of different phenomena such as stereodynamics in chemical reactions, electronic stereodynamics in molecules, high-order harmonic generation, etc. Both one-dimensional and three-dimensional molecular orientations in the adiabatic regime have been achieved by our group with combined electrostatic and intense nonresonant laser fields [1-3]. In these techniques, the permanent dipole interaction with an electrostatic field is enhanced by the anisotropic polarizability interaction with an intense nonresonant laser field. On the other hand, all-optical molecular orientation utilizes both anisotropic polarizability and hyperpolarizability interactions with an intense nonresonant two-color laser field. All-optical molecular orientation has also been proposed and demonstrated by our group [4-6]. The direction of molecular orientation depends on the polarization direction and the relative phase between the two wavelengths. In this study, we aim to increase the degrees of molecular orientation. We use carbonyl sulfide (OCS) molecules diluted with He buffer gas as sample molecules. For the two-color laser field, we use the fundamental pulse and its second harmonic pulse from an injection seeded Nd:YAG laser. In order to increase the degrees of orientation, we employ the following two strategies. (1) We use lower-lying rotational states of OCS molecules, which can be selected by a home-built molecular deflector (Fig. 1). (2) Furthermore, in order for the hyperpolarizability interaction to work more effectively than in the proof-of-principle experiment [6], we adjust the rising parts of the two wavelengths by introducing a Michelson-type delay line in the optical path of the pump pulse (Fig. 2). The relative phase between the two wavelengths is controlled by rotating a fused silica plate.

Fig. 1: Vertical profiles of OCS molecular beams.

Fig. 2: Adjustment of the rising parts of the two wavelengths.