Increasing the degrees of orientation of state-selected OCS molecules with relative-delay-adjusted nonresonant two-color laser fields

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Orientation of linear asymmetric molecules such as carbonyl sulfide (OCS) is both theoretically and experimentally demonstrated by our group [1,2]. The asymmetric molecules interact with the nonresonant two-color laser fields through their anisotropic polarizability and hyperpolarizability. The anisotropic polarizability interaction is responsible for creating the symmetric alignment potential, whereas the anisotropic hyperpolarizability interaction is responsible for forming the asymmetric orientation potential [1]. The polarizability interaction of general molecules such as OCS is stronger than the hyperpolarizability interaction by orders of magnitude [3]. In addition, the second harmonic pulse always follows the rising part of the fundamental pulse. This delay between the two wavelengths leads to the creation of a deep symmetric alignment potential before the relatively weak asymmetric orientation potential can be formed [4]. Then, although stronger alignment is observed, the degrees of orientation do not increase. It is theoretically shown that delaying the fundamental pulse is useful in avoiding such an unfavorable situation [5]. This time, we delayed the fundamental pulse by 1.75 ns with a Michelson-type delay line. By using two-color laser fields from an injection-seeded Nd:YAG laser and OCS molecules buffered with 60-bar He as a sample, we have observed significantly higher degrees of orientation ($<\cos\theta>\approx\pm0.3$) as shown in Fig. 1 compared to those in the proof-of-principle experiment ($<\cos\theta>\approx\pm0.04$) [2]. The observed degrees of orientation are the highest ever achieved with an all-optical technique and characterized with the Coulomb explosion imaging.

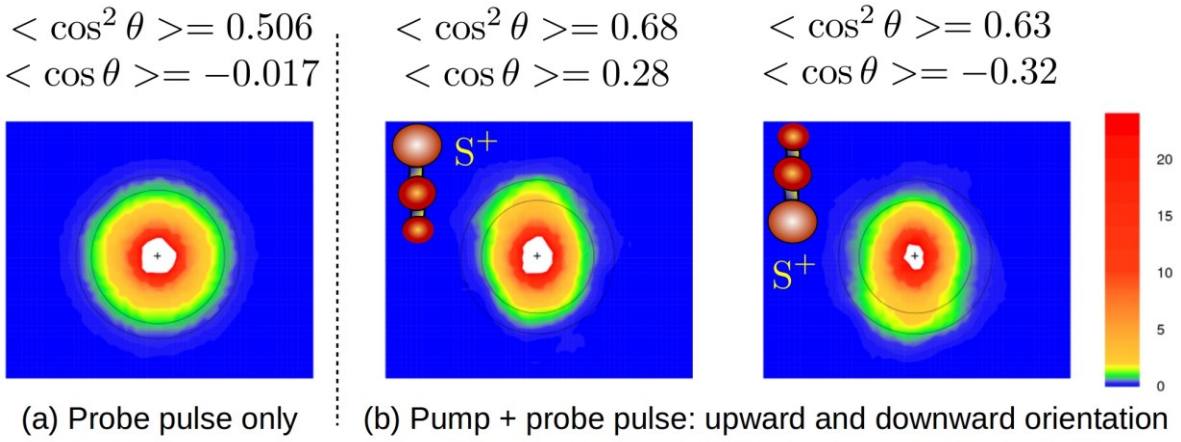

Fig. 1: Observation of the increased degrees of orientation of state-selected OCS molecules with an all-optical technique. Shown are the images for S^+ fragment ions.

References: [1] T. Kanai and H. Sakai, J. Chem. Phys. **115**, 5492 (2001), [2] K. Oda *et al*., Phys. Rev. Lett. **104**, 213901 (2010), [3] G. Maroulis and M. Menadakis, Chem. Phys. Lett. **494**, 144 (2010), [4] M. Muramatsu *et al*., Phys. Rev. A **79**, 011403(R) (2008), [5] J. H. Mun, and H. Sakai, to appear in Phys. Rev. A Vol. 98, Issue 1.