

## 論文の内容の要旨

# Experimental and theoretical studies toward stronger molecular orientation with an all-optical technique

(全光学的手法による気体分子の配向度向上を目指した実験的及び理論的研究)

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A sample of aligned or oriented gaseous molecules can be used as an ideal quantum mechanical system for investigating the anisotropic dependence in many light-molecule interactions. For example, by orienting molecules to a specific direction, it is possible to control the stereodynamics of chemical reactions and selectively produce the desired product. High-harmonics generated from a sample of aligned or oriented molecules can be analyzed to probe the instantaneous structure of sample molecules. On the other hand, electromagnetic field intensity dependence of the degree of alignment or orientation can be used to estimate the anisotropies of polarizability and hyperpolarizability of sample molecules experimentally. Due to these multifaceted and compelling prospects, controlling the rotational motion of asymmetric molecules is an active area of research.

We can realize molecular alignment or orientation by utilizing the nonlinear interaction of asymmetric molecules with an intense nonresonant laser field. However, a single wavelength laser pulse can only realize molecular alignment because the interaction potential is symmetric upon the inversion of the laser electric field vector. For achieving molecular orientation, one needs to induce asymmetry in the interaction potential along the polarization axis. This asymmetric potential can be created either by introducing a weak electrostatic field or by using a phase-controlled two-color laser field. In the first approach, the permanent dipole interaction

with an electrostatic field is enhanced by the anisotropic polarizability interaction with the intense nonresonant laser field. However, the presence of a weak electrostatic field may induce Stark effect in the rotationally controlled sample of asymmetric molecules, limiting their usefulness for various applications. Moreover, the orientation direction is limited by the arrangement of the electrodes used to generate the electrostatic field.

In contrast, laser fields can be rapidly turned off by using the plasma-shutter technique, and the orientation direction can be arbitrarily controlled by changing the polarization direction of the two-color laser field and the relative phase difference between the two wavelengths. Hence, all-optical molecular orientation utilizing both anisotropic polarizability and hyperpolarizability interactions with an intense nonresonant two-color laser field is the way toward the realization of completely field-free molecular orientation. However, since the second harmonic is generated by passing the intense fundamental pulse through a second-harmonic generation crystal, the rising part of the second harmonic pulse in a two-color laser field always follows the fundamental pulse. As a result, a deep symmetric potential due to polarizability interaction is created before the relatively weak asymmetric orientation potential can be formed, resulting in an enhanced degree of alignment, but a suppressed degree of orientation. Furthermore, since the orientation directions for states with even and odd quantum number  $J$ 's are opposite, the average degree of orientation of a thermal ensemble becomes weak. Despite these challenges, the proof-of-principle experiment performed in our laboratory has confirmed the validity of all-optical method of molecular orientation using a two-color laser field. The goal of this research is to experimentally and theoretically investigate different ways of further increasing the degree of molecular orientation with an all-optical method.

To realize stronger molecular orientation with two-color laser fields, we have adjusted the relative delay between the two wavelengths by introducing a Michelson-type delay line in the optical path. By delaying the fundamental pulse in this way, we effectively allow the asymmetric potential to be created at the same time when the dominant symmetric potential due to polarizability interaction is formed. We have also used a home-build molecular deflector to select lower-lying rotational states of OCS molecules as a sample and avoid the weakening of the average degree of orientation due to opposite contributions from even and odd numbered states. By applying the fundamental pulse and its second harmonic pulse from an injection-seeded Nd:YAG laser as the two-color laser field, we have observed stronger degrees of molecular orientation:  $\langle\langle\cos\theta\rangle\rangle \sim \pm 0.3$ . The observed degree of orientation is an order of

magnitude stronger than the degree of orientation obtained in the proof-of-principle experiment. It is also the strongest molecular orientation with an all-optical method, which was observed appropriately with the Coulomb explosion imaging by keeping the probe polarization perpendicular to the detection plane.

Although the applied laser pulse (FWHM  $\sim 12$  ns) is orders of magnitude longer than the free rotational period of the sample molecule (82.24 ps), we theoretically and experimentally prove that all-optical molecular orientation dynamics is still nonadiabatic. Due to the nonadiabaticity, merely increasing the laser intensity will not necessarily improve the degree of orientation. Since the polarizability interaction usually is orders of magnitude larger than the hyperpolarizability interaction, the asymmetry in the potential may increase by increasing the laser intensity. However, at the same time, symmetric potential due to polarizability interaction will get deeper faster, making the tunneling to a deeper potential well more difficult. Here we propose a new all-optical molecular orientation approach which combines a linearly polarized fundamental pulse and an elliptically polarized second harmonic pulse to suppress the symmetric effect created by the polarizability interaction while accelerating the tunneling to a deeper potential well, leading to the efficient creation of molecular orientation. By solving the time-dependent Schrödinger equation numerically, we show that it is possible to increase the degree of orientation by tuning the peak intensity along the minor axis of the elliptically polarized second harmonic laser pulse. This approach is especially useful for general molecules with smaller hyperpolarizability anisotropy and larger polarizability anisotropy. Finally, numerical results are presented to prove that the same combination of linearly and elliptically polarized two-color laser field is capable of realizing three-dimensional molecular orientation, which corresponds to an all-optical method of controlling all the spatial directions of an asymmetric top molecule.