Improving molecular orientation by optimizing relative delay and intensities of two-color laser pulses

Je Hoi Mun[1, 2] and ○Hirofumi Sakai[1]
[1]Department of Physics, Graduate School of Science, The University of Tokyo
[2]Center for Relativistic Laser Science, Institute for Basic Science
E-mail: hsakai@phys.s.u-tokyo.ac.jp

Techniques for controlling rotational degrees of freedom of gas-phase molecules have provided opportunities to observe phenomena related to anisotropic structures of molecules, such as optimal control of multiphoton ionization [1] and molecular imaging based on high-order harmonic generation [2-5]. Molecules are aligned if the angular confinement along the laboratory-fixed axis is symmetric upon the inversion of the molecular axis, while they are oriented if the head-versus-tail order confinement is also created. The techniques for molecular orientation differ from those for molecular alignment in that the adiabatic control becomes much more important for achieving higher degrees of orientation. The importance of adiabatic orientation in two-color laser pulse technique [6] and in the combined electrostatic and laser field technique [7] has been numerically examined. It has been shown that while adiabatic alignment of small linear molecules is achievable with relatively short, several hundred picosecond laser pulse, the adiabatic orientation requires the pulse width much longer than one nanosecond [6]. Recent studies have revisited the importance of adiabatic molecular orientation from the viewpoint of pendular states of sample molecules. For rotational state-selected OCS molecules, nonadiabatic transitions between 'pendular doublet', which is a pair of quantum states oriented oppositely, prevent the strong orientation in the combined electrostatic and laser field technique [8]. Adiabatic orientation cannot be achieved as simply as adiabatic alignment.

In this work, we show that, in the two-color several nanosecond laser pulse technique, the relative delay between ω and 2ω laser pulses and the intensities of the two wavelengths determine the adiabaticity of the orientation dynamics. The degrees of molecular alignment increase more and more with the total intensity of the two-color laser pulse. Thus, strong molecular alignment can be achieved when the two laser pulses are temporally overlapped. For the case of molecular orientation, however, a two-color pulse with an appropriate delay between the two wavelengths can give rise to much higher degrees of orientation. This orientation dynamics seriously deviates from the dynamics of adiabatic approximation. We have solved the TDSE in order to examine time evolutions of OCS molecules in the rotational ground state, which are irradiated with both ω and 2ω laser pulses whose pulse widths (FWHM) are 12 ns and 8 ns, respectively. With this fixed pulse width condition, the intensities of and the delays between the two wavelengths are varied. In Fig 1, the achieved degrees of alignment $<\cos^2\theta>$ and orientation $<\cos\theta>$ are 2D-mapped by the delay between the two wavelengths and the peak intensity of the ω pulse. For the orientation, one can see that the TDSE result seriously deviates from the adiabatic approximation result. We report how the orientation dynamics is influenced by intensities of the two wavelengths and the delay between them and provide recipe for achieving higher degrees of orientation.

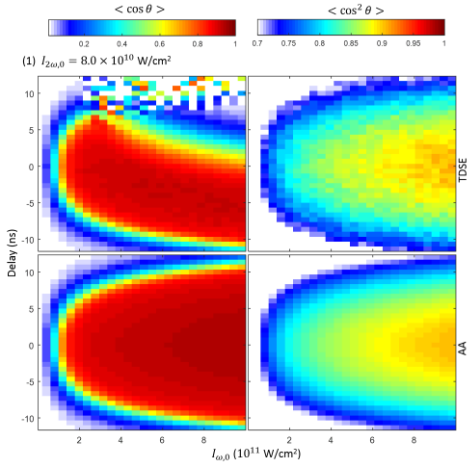

Fig. 1. Degrees of orientation and alignment: Degrees of orientation $<\cos\theta>$ and alignment $<\cos^2\theta>$ of OCS molecules in the rotational ground state that are achieved at the peak of the 2ω pulse. The horizontal axis is the peak intensity of the ω pulse while the vertical axis is the delay between the ω and 2ω pulses. The peak intensity of the 2ω pulse is fixed at 8.0×10^{10} W/cm^2. The degrees of orientation and alignment are calculated for TDSE and adiabatic approximation (AA).

[1] T. Suzuki, S. Minemoto, T. Kanai, and H. Sakai, Phys. Rev. Lett. **92**, 133005 (2004).
[2] J. Itatani *et al*., Nature (London) **432**, 867 (2004).
[3] T. Kanai, S. Minemoto, and H. Sakai, Nature (London) **435**, 470 (2005).
[4] T. Kanai, S. Minemoto, and H. Sakai, Phys. Rev. Lett. **98**, 053002 (2007).
[5] C. Vozzi *et al*., Nat. Phys. **7**, 822 (2011).
[6] M. Muramatsu, M. Hita, S. Minemoto, and H. Sakai, Phys. Rev. A **79**, 011403(R) (2009).
[7] Y. Sugawara, A. Goban, S. Minemoto, and H. Sakai, Phys. Rev. A **77**, 031403(R) (2008).
[8] J. H. Nielsen *et al*., Phys. Rev. Lett. **108**, 193001 (2012).