Attempt to form a molecular ensemble with macroscopic threefold symmetry

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If gas molecules that are normally oriented randomly can be aligned or oriented, the alignment/orientation dependence of various physical phenomena caused by the interaction between laser pulses and molecules can be directly clarified. (The molecules are considered to be “aligned” when the molecular fixed axis is synchronized with respect to a laboratory fixed axis with the directional plus-minus inversion symmetry. If the aligned molecules are under the additional head-versus-tail order confinement, they are considered to be “oriented”). Unidirectional molecular ensembles cannot be realized in normal surroundings, it can be regarded as "a new quantum phase of molecules" or as a "gaseous crystal". The researches about molecule alignment/orientation using the interaction of intense laser fields with dipole moments induced in molecules continue to progress remarkably over the last two decades. Almost all milestone techniques related to orientation control have been developed by Sakai group, starting with one-dimensional orientation control. Recently, Sakai group has achieved a "completed form" of molecular orientation control that satisfies the three conditions of three-dimensional orientation control with higher degrees of orientation in the laser-field-free condition [1]. Furthermore, all-optical orientation control using only nonresonant two-color laser fields is also progressing. In this method, the anisotropy in the interaction potential created by the anisotropic hyperpolarizability interaction between the two-color laser fields and the molecules is utilized for orientation control. The proposal of the method itself came from Sakai group [2], and the principle has already been successfully demonstrated [3]. We are aiming at realizing the “completed form” of all-optical orientation control.

With a threefold symmetric electric field formed by the superposition of counterrotating circularly polarized two-color laser fields (fundamental pulse and its second-harmonic pulse), it is possible to align sample molecules with threefold symmetry such as BCl_3 with their three arms along (or in between) the laser electric fields with threefold symmetry as shown in Figure 1 [4]. Therefore, it is expected to form a molecular ensemble with macroscopic threefold symmetry (a gaseous crystal with threefold symmetry). We developed an ion imaging spectrometer which observes the angular distributions of fragment ions within the polarization plane of circularly-polarized laser pulses in order to investigate the actual state of “the gaseous crystal with threefold symmetry". The spectrometer projects the angular distributions of fragment ions in the polarization plane onto the two-dimensional detector plane by applying a pulsed high voltage perpendicular to the TOF axis at an appropriate timing. The operation of the spectrometer has been checked by OCS molecules as sample molecules. The proof-of-principle experiment is now in progress in our lab.

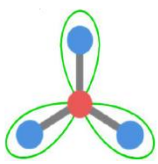

Figure 1: Principle of forming a molecular ensemble with macroscopic threefold symmetry using an electric field with threefold symmetry created by the superposition of counterrotating circularly polarized fundamental and its second-harmonic pulses.

References

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