Structure determination of molecules in an alignment laser field by femtosecond photoelectron diffraction using an X-ray free electron laser

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Synopsis We have successfully measured the X-ray photoelectron diffraction (XPD) of laser-aligned iodine molecules by using X-ray free electron laser pulses. Thanks to a higher degree of alignment of sample molecules, we have obtained the XPD image having structural information. The experimental XPD image is analyzed with the help of the XPD theory to obtain the structure of molecules in an intense laser field.

In the last two decades, laser-induced alignment and orientation techniques of molecules are well developed and successfully applied to high-order harmonic spectroscopy [1], photochemical reaction controls [2], and so on. However, a fundamental aspect of the technique was not scrutinized so far; the intense field of the alignment pulse may modify the ground-state structure of the molecules, especially in the case of the adiabatic alignment technique. Here we report our recent results on the structure determination of iodine molecules, I_2, in an intense laser field by the X-ray photoelectron diffraction (XPD) using ultrafast X-ray Free Electron Laser (XFEL) pulses [3-5].

Figure 1(a) shows the schematic diagram of the XPD method of aligned molecules. The sample I_2 molecules are aligned parallel to the polarization direction of XFEL by the Nd:YAG laser pulses. The momentum image of photoelectrons produced by the XFEL pulses with the photon energy of 4.7 keV have been measured with a velocity-map imaging spectrometer [3,4]. The angular distribution of I 2*p* photoelectrons thus measured corresponds to the XPD pattern as shown in Fig. 1(b). The experiment was performed at BL3 of the SACLA facility.

To determine the molecular structure, i.e., the bond length of the I_2 molecules in the Nd:YAG laser field of ~1×10^{12} W/cm^2, we have applied our molecular-structure-determination methodology [5] to the measured XPD data. Our analyses reveals that the internuclear distance of I-I is most likely to be 0.18–0.3 Å longer than the equilibrium distance of the ground state molecule. The XPD patterns calculated for the most probable and the ground state molecules are shown by red solid and blue dotted curves, respectively, in Fig. 1(b). The details of the present work will be discussed in the presentation.

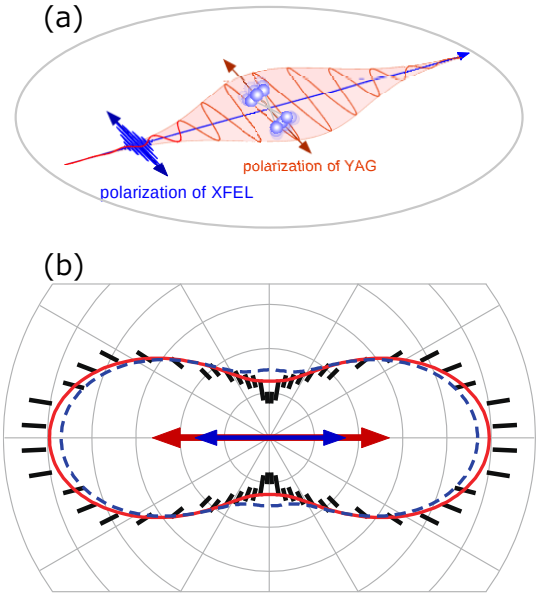

Figure 1. (a) Schematic diagram of X-ray photoelectron diffraction of aligned molecules. (b) The I 2*p* photoelectron diffraction pattern of aligned I_2 molecule.

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