

Molecular Structures Revealed by Angle-Resolved High-Harmonic Spectroscopy from Aligned N₂ Molecules

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we report the results of high-order harmonic generation (HHG) from field-free aligned N₂ molecules were investigated by rotating the molecules with respect to the polarization of harmonic generation pulses. The results indicated that high harmonic generation and molecular structure were closely linked.

High-order harmonic generation from nonadiabatically aligned molecules presents a rich set of new physical phenomena^[1]. Both strong field ionization and acceleration of the ionized electron are governed by the symmetry of the highest occupied molecular orbital (HOMO), or HOMO-1 etc., and it has become a new avenue for retrieve molecular structures and dynamics^{[2][3]}.

The experiments were carried out in a pump-probe technique, where the pump pulse performs field-free alignment of molecules, and the delayed probe pulse generated high-order harmonic radiation with the information of its dynamic alignment. The linearly polarized 35fs, 800nm pump and probe pulses with a time delay Δt are produced from the output of a Ti:Sapphire laser system. The polarization of the pump laser beam is fixed and rotated the half wave plate to acquire the alignment-dependent HHG. Harmonic spectra were spatially dispersed by a home-made flat-field grating spectrometer and detected by micro-channel plate equipped with a phosphor screen. A CCD camera was used to image the phosphor screen.

In experiments, the harmonic intensity was normalized to the one obtained without the aligning pulse and recorded at different time delays of N₂ molecules as shown in Fig.1(a). The wave packet of N₂ has been sketched in Fig.1(a) at alignment and anti-alignment moments. Measuring the harmonic intensity as a function of the angle Θ between pump laser polarization and

probe laser polarization in laboratory frame is as show in Fig.1(b).

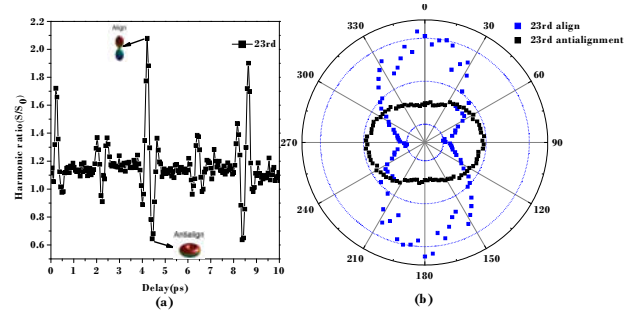


Fig. 1(a) The modulations of the relative yields of 23rd harmonic; (b) Measured harmonic spectra from aligned N₂ molecules obtained at the time delay of align anti-alignment with respect to angle of molecular alignment to the polarization of harmonic generation pulses.

The Θ dependent modulation demonstrates that the 23rd harmonic radiation from aligned N₂ is peaked at $\Theta \sim 0^\circ$ and minimized at $\Theta \sim 90^\circ$ while HHG from anti-alignment is peaked at $\Theta \sim 90^\circ$ minimized at $\Theta \sim 0^\circ$. The characteristic behaviors of angle-dependent harmonic signal are due most likely to the different orbital symmetry or the molecular structures. And more contributions from multi-orbitals will be further analyzed.

References

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