High Harmonic Generation from Multiple Orbitals in N_2

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Synopsis: Contribution from the HOMO and HOMO-1 orbitals are observed in high harmonics from N_2. We discuss the harmonic modulations in the rotational revival structure due to the HOMO and HOMO-1.

For molecules, the highest occupied molecular orbital (HOMO) is generally thought to be responsible for ionization and recombination during HHG [1]. We report experimental evidence that the more deeply bound HOMO-1 with its π_u symmetry also contributes to HHG [2]. This opens the route to imaging coherent superpositions of electronic orbitals.

In the experiment, an ultrashort laser pulse (alignment pulse) creates a rotational wave packet resulting in molecular alignment of N_2. A second delayed laser pulse with higher intensity generates high harmonics in the aligned molecules. The harmonic spectrum is detected as a function of delay between alignment and harmonic generating pulses (Fig. 1A).

The molecular axes are preferentially perpendicular with respect to the harmonic generating polarization at 4.1 ps. This appears as a reduction in the signal at harmonic 15 compared to the unaligned case prior to 3.6 ps. At 4.4 ps the molecular axes are partially parallel to the harmonic generating polarization and an increase of the signal at low harmonics is observed compared to the unaligned case. However, as the harmonic number increases above 25, the temporal modulation compared to harmonic 15 becomes inverted.

The first step of HHG, tunnel ionization, is sensitive to the molecular wave function far away from the nuclei in direction of the generation polarization. Thus, the σ_g HOMO orbital ionizes more easily if the generation polarization is parallel to the internuclear axis. Also, the recombination dipole is larger if the molecular axis is aligned with the generation polarization, since the recombination dipole has larger amplitude in the long direction of the orbital. For the HOMO, the HHG signal is maximized for molecules standing parallel to the HHG polarization compared to the perpendicular polarization, consistent with the observed experimental trend in harmonics 15-25 in Fig. 1A.

The peak at 4.1 ps at harmonics 25-39 can be explained by the HOMO-1. Tunnel ionization perpendicular to the molecular axis is higher for the HOMO-1 compared to the HOMO because it has a larger spatial extent in this direction (Fig. 1C). Ionization and recombination parallel to the internuclear axis give rise to a vanishing dipole for the HOMO-1, because of the dipole cancellation due to the opposite sign of the wave function on either side of the axis. In contrast, the recombination dipole perpendicular to the axis is strong.

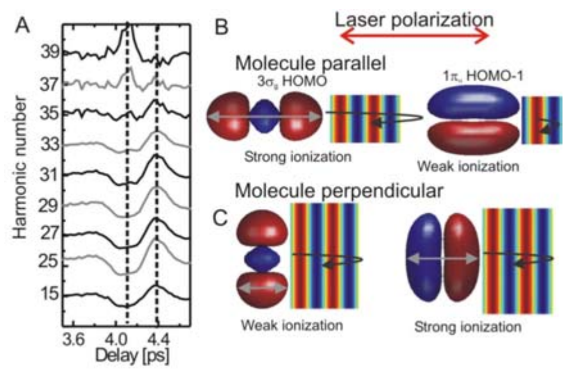

Fig. 1. A) Harmonic signal as a function of alignment-generation pulse delay. Scheme for HHG from HOMO and HOMO-1 for parallel (B) and perpendicular (C) polarization of molecules and harmonic generating laser.

We support our interpretation of the spectral modulations by simple model calculations of the recombination dipole. We thank the Deparment of Energy, BES for funding.

References

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