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Multi-Orbital contributions in High Harmonic Generation[1]

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The high harmonic spectrum generated from atoms or molecules in a strong laser field contains information about the electronic structure of the generation medium. In the high harmonic generation (HHG) process, a free electron wave packet tunnel-ionizes from the molecular orbital in a strong laser field. After being accelerated by the laser electric field, the free electron wave packet coherently recombines to the orbital from which is was initially ionized, thereby emitting the harmonic spectrum. Interferences between the free electron wave packet and the molecular orbital will shape the spectrum in a characteristic way. These interferences have been used to tomographically image the highest occupied molecular orbital (HOMO) of N_2 [1]. Molecular electronic states energetically below the HOMO should contribute to laser-driven high harmonic generation (HHG), but this behavior has not been observed previously. We have observed evidence of HHG from multiple orbitals in aligned N_2 [2]. The tunneling ionization (and therefore the harmonic generation) is most efficient if the orbital has a large extension in the direction of the harmonic generation polarization. The HOMO with its σ_g symmetry therefore dominates the harmonic spectrum if the molecular axis is parallel to the harmonic generation polarization, the lower bound π_u HOMO-1 dominates in the perpendicular case. The HOMO contributions appear as a regular plateau with a cutoff in the HHG spectrum. In contrast, the HOMO-1 signal is strongly peaked in the cutoff region. We explain this by semi-classical simulations of the recombination process that show constructive interferences between the HOMO-1 and the recombining wave packet in the cutoff region. The ability to monitor several orbitals opens the route to imaging coherent superpositions of electronic orbitals.

[1] J. Itatani *et al.*, Nature **432**, 867 (2004)
[2] B. K. McFarland, J. P. Farrell, P. H. Bucksbaum and M. Gühr, Science **322**, 1232 (2008)

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