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Tomographic Imaging of Molecular Orbitals Using Femtosecond Lasers

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I will show how we can experimentally reconstruct a single-electron orbital wave function from a simple molecule, dinitrogen. We use a seemingly unlikely technique, high harmonic generation (HHG). HHG occurs when a gas is irradiated with an intense femtosecond laser pulse. The atoms or molecules are tunnel-ionized by the laser field, and then are driven back to the parent ion within the same optical cycle. Some of these electrons recombine with the parent, and release their kinetic energy as xuv photons. The resulting spectrum covers the photon range of 10–100 eV. The radiation results from a transition from a continuum electron wave function, described as a plane wave, and the orbital from which the electron was removed, the HOMO. Thus the experiment is a measure of the transition dipole matrix elements between the single-electron orbital and a set of plane waves. By rotating the molecule in the gas phase using a second laser pulse, we can map out the matrix elements for different projections of the molecule. This data can then be inverted using a tomographic algorithm to yield an image of the orbital wave function. We will show the recovered $3\sigma_g$ orbital of N_2. Beyond the single active electron approximation, there are other transitions that are allowed between the HOMO and inner orbitals. This is due to the indistinguishability of the electrons. Including these exchange terms gives even better agreement between theory and experiment. Because this measurement is made with a 25 fsec laser pulse, it is now possible to perform pump-probe experiments to observe dynamic changes in the electronic structure of molecules. It may even be possible to observe *electronic* wave packet motion within an atom with attosecond resolution.