DAMOP05-2005-000034

Abstract for an Invited Paper
for the DAMOP05 Meeting of
the American Physical Society

Imaging a Molecular Orbital Wave Function Using High Harmonic Emission

DAVID VILLENEUVE, National Research Council of Canada

Single-electron molecular orbital wave functions are mathematical constructs that are used to describe the multi-electron wave function of molecules. The highest lying orbitals are of particular interest since they are responsible for the chemical properties of molecules. To observe them change as molecular bonds are formed and broken is to observe the essence of chemistry. Yet single orbitals are difficult to observe experimentally — until now impossible on the time scale of chemical reactions. We show that the full 3-dimensional structure of a single orbital can be imaged using a seemingly unlikely technique — high harmonic generation from aligned molecules using intense femtosecond laser pulses. We show how the broadband harmonic spectra, measured for a series of molecular alignments, lead to a tomographic reconstruction of the single electron orbital wave function of dinitrogen. This leads to ontological discussions about the meaning of a wave function, particularly in a multielectron system. A non-ionizing femtosecond laser pulse creates a rotational wavepacket that causes periodic molecular alignment. A more intense pulse induces high harmonic emission from the aligned molecules. The recollision electron current pulse is characterized as it returns to a reference argon atom. Assuming that we know the shape of the 3p orbital of argon, we can determine the spectral phase and amplitude of the recollision current. The phase of the harmonic emission from nitrogen is referenced to the phase of argon by measuring the interference in a mixed target gas. The polarization of the emission is also recorded by polarimetry. All of these measurements lead to the reconstruction of the nitrogen sigma-g orbital shape. We also show that attosecond dynamics of an electron wave packet can be measured in the high harmonic spectrum.