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Measuring the Electronic Structure of Molecules Using High Harmonic Emission

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Electronic structure is commonly measured by photoelectron spectroscopy, in which short wavelength radiation detaches an electron from a molecule. The kinetic energy of the electron labels the orbital from which it came. The angle of the electron relative to the polarization direction gives further information about the orbital through a partial wave expansion.

When a molecule is exposed to an intense femtosecond laser field, selective ionization of the highest orbital occurs, but the electron contains no molecular information because its subsequent motion is completely dominated by the laser field. However, there is a very small probability that the electron will recombine with the parent ion, and liberate its kinetic energy into an xuv photon. This process is known as high harmonic generation, and produces a coherent, collimated xuv beam that is composed of a train of attosecond pulses. It can be viewed as time-reversed photoelectron spectroscopy. The xuv spectrum contains both amplitude and phase information about the ionized orbital.

I will show how we can reconstruct the shape of a single orbital from nitrogen molecules. This technique can also be used to observe dynamical changes to the electronic structure during chemical reactions using pump-probe techniques with a time resolution of 5-30 fs. Because the high harmonic process occurs within a fraction of an optical cycle, and because the attosecond bursts are chirped in frequency, it is possible to map attosecond electron wave packet motion onto the spectrum. Thus attosecond electron motion may be visible to this process.