Ultrafast Electron Diffraction from Aligned Molecules in the Gas Phase

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Abstract:

Ultrafast electron diffraction can provide information about the structure of a molecule as it undergoes a photoreaction. Ideally, one would capture images of the initial, intermediate and final states with sub-Angstrom spatial resolution, but there are challenges that need to be overcome. The first challenge is to retrieve the structure directly from the diffraction pattern for molecules that are in the gas phase, and thus have random orientation, and the second is to achieve sufficient temporal resolution to observe the relevant dynamics. The random orientation of molecules can be overcome by diffracting from aligned molecules.

We have shown that by using femtosecond laser pulses to impulsively align the molecules, it is possible to capture a field-free diffraction pattern while the molecules are transiently aligned. First experiment were done on a static molecular structure, and we have recently demonstrated imaging of a short lived excited state, with a resolution of 1 ps. The second challenge is to improve the temporal resolution to the regime of 100 fs in order to map the structural changes as the molecule transitions from the ground to the excited states. This means not only delivering sufficiently short pulses on the target, but also compensating the effect of the velocity mismatch of electrons and laser as they traverse the sample.

We are pursuing two approaches for this, one is to use RF compression of 100 keV electron pulses in combination with a tilted laser pulse excitation to overcome the velocity mismatch, the second is to use relativistic (MeV) electron pulses where the velocity mismatch is negligible. Recent progress on both of these efforts will be reported.