High order harmonic generation in fullerenes

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Synopsis Within the Strong Field Approximation we have found that the high harmonic spectra for icosahedral fullerenes at midinfrared wavelengths show modulations in the plateau region. These modulations are due to the coherent superposition of the contributions to the dipole moment from different atomic centers of the molecule and are present in spectra for ensembles of aligned fullerenes as well as randomly oriented ones. In the case of the larger fullerenes the radius of the fullerene can be determined from the positions of the interference minima in the high harmonic spectra using a spherical model.

One of the exciting goals in intense laser science is that of imaging dynamical changes in molecular structure and molecular reactions on an ultrafast time scale. A promising tool for such investigations is high-order harmonic generation (for a review, see [1]), since an initially ionized electron is driven back by the field to the parent ion and recombines under the emission of a high-energy photon. The harmonic spectrum is, in general, sensitive to the orientation and structure of the molecule and the symmetry of the molecular orbital involved.

In order to evaluate the potential of molecular imaging using harmonic generation it is important to study the process in large polyatomic molecules. We have therefore analyzed high harmonic generation in fullerenes of icosahedral fullerenes [2, 3] using the strong-field approximation. Our results for the high harmonic spectra generated in C_{20},C_{60},C_{80}, and C_{180} show modulations in the plateau at midinfrared but not at the near-infrared wavelengths. We have shown that these modulations are due to a multislit interference effect arising from the coherent superposition of the contributions to the dipole moment, from different atomic centers of the molecule (see Figure). Due to the highly symmetric geometric structure of the fullerenes the modulations are present in spectra for ensembles of aligned fullerenes as well as randomly oriented ones. The Figure shows that the positions of these minima agree with minima of the recombination matrix element. We further used a simple spherical-shell model of a fullerene to reproduce the minima of the recombination matrix element and determine the radius of the fullerene from the positions of the interference minima. The observation of high harmonic spectra may be therefore useful to identify structural changes in such complex molecules induced by an intense laser pulse.

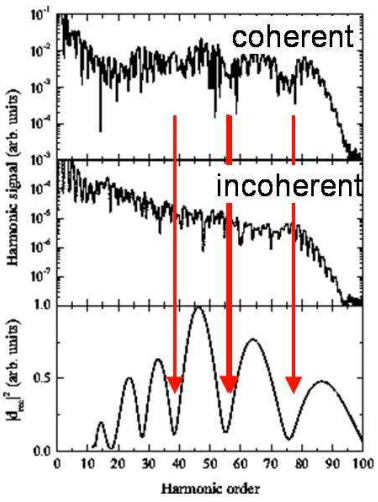

Fig. 1. Theoretical predictions for high-order harmonic spectra generated in C_{180} at $\lambda = 1800$ nm (upper panel) and an intensity of $I = 5 \times 10^{13}$ W/cm^2 [3]. The characteristic minima in the high harmonic spectra are induced by destructive interference effects between the partial electron waves emanating from the different atomic centers of the molecule (middle panel) and correspond to zeros in the recombination matrix element (lower panel).

References

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