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(G*) (POS-2) High-Harmonic Sidebands for Time-Resolved Spectroscopy

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High-Harmonic Sidebands for Time-Resolved Spectroscopy

In the semi-classical picture of high-harmonic generation (HHG), a strong ($10^{18} - 10^{20} \text{ W/m}^2$) laser field is applied to an atom, repeatedly accelerating one of its valence electrons into the continuum and back to the parent atom. Upon recollision with the parent atom, the electron emits photons of odd-integer harmonics of the driving field [1]. When a second weaker field is applied to the system, the trajectory of the electron is perturbed, causing sidebands to occur in the harmonic spectrum that are characteristic of the perturbing field energy [2],[3]. Consequently, HHG can be used as a method of upconverting mid-infrared light for sensitive spectroscopy, as high-harmonics of infrared (IR) light lie in the visible regime and beyond.

We perform numerical simulations of HHG in order to inform future experimental work that will use HHG cross-correlation to measure time-resolved fields in the mid-IR. HHG cross-correlation involves the mixing of the strong femtosecond driving field with a weak mid-IR field in a nonlinear material, leading to the sidebands that appear in the harmonic spectrum. The position of the sidebands in the spectrum indicates the frequency of the optical free-induction decay [2].

This is a particularly pragmatic approach to IR spectroscopy because it precludes the need for expensive detectors that need to be cooled. Furthermore, the emitted bursts of radiation are on the order of attoseconds (10-18s) in duration, opening the possibility of ultrafast spectroscopy extending into the mid- and far-IR.

References

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- [2] T.J. Hammond et al.. Femtosecond time-domain observation of atmospheric absorption in the near-infrared spectrum. *Physical Review A* (2016).
- [3] G. Vampa et al.. Linking high harmonics from gases and solids. *Nature* 522, 462–464 Springer Science and Business Media LLC, 2015.

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