Elliptically polarized high-order harmonics from aligned molecules within the strong-field approximation

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Synopsis A correction term is introduced in the stationary-point analysis on high-order harmonic generation (HHG) from aligned molecules. Arising from a multi-centre expansion of the electron wave function, this term brings our numerical calculations of the Lewenstein model into qualitative agreement with recent experiments [1].

Within the field of HHG there is currently a large deal of interest in the ellipticity of high-order harmonics from aligned molecules. Experimental results, such as those presented in [1], have challenged the validity of standard theoretical models. These models are extensions of the Lewenstein model to molecules and have the appeal of allowing qualitative predictions of experiment based on a few well-known approximations.

The process of HHG is typically split into three steps: *i)* Ionization of the outer electron *ii)* Propagation of the free electron in the laser field *iii)* Recombination of the electron to the ground state followed by emission of radiation. A standard treatment of step two is to perform a stationary-point analysis on the fast varying phase factors yielding the instantaneous free-electron momentum

$$\mathbf{k}_{st}(t) = -\frac{1}{t-t'}\int_{t'}^{t}\mathbf{A}(t'')\mathrm{d}t'',$$

where t' is the ionization time and $\mathbf{A}$ the vector potential of the laser field. However, if the molecular orbital is written explicitly in a multi-centre expansion it can easily be seen that the momentum is

$$\mathbf{k}_{st}(t) = -\frac{1}{t-t'}\int_{t'}^{t}\mathbf{A}(t'')\mathrm{d}t'' + \frac{1}{t-t'}\left(\mathbf{R}_f - \mathbf{R}_i\right),$$

where $\mathbf{R}_i$ ($\mathbf{R}_f$) is the ionization (recombination) site [2]. Depending on the orientation of the molecule, the extra term allows a non-zero component of the emitted harmonics polarized perpendicular to the driving laser.

The spectrum in Fig. 1 was calculated according to the Lewenstein model for molecules using the laser parameters from [1] and incorporating the additional term in the electron momentum. This term is responsible for the perpendicular component, which becomes comparable to the parallel component near the 21st harmonic.

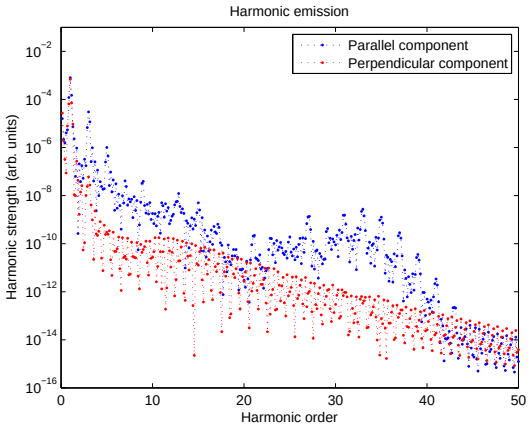

Fig. 1. Spectrum of N_2 HOMO at Euler angle $\phi = 55, \theta = 90, \chi = 0$.

References

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