Ab-Initio Modeling of Attosecond Streaking Measurements

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Synopsis We model attosecond streaking measurement beyond the single-active-electron approximation. Our model takes advantage of the "state-specific-expansion approach", where a many-electron wave function is described within a basis of relevant atomic and ionic states.

The attosecond streak camera is currently one of the main tools of attosecond metrology. Its theoretical foundations were laid in [1], where a simple quantum-mechanical model was proposed. Within the single-active-electron approximation, this approach has successfully been used for the interpretation of attosecond streaking measurements. Recently, the accuracy and reproducibility of such measurements have dramatically improved [2]; also, angle-resolved streaking measurements are becoming accessible [3]. This demands more accurate and detailed modeling. To provide a realistic theoretical description of attosecond streaking measurements, and to clarify the limits of single-electron simulations, we have developed a model based on accurate atomic structure calculations. The electron correlation and the interaction of the photoelectron with the ion enter this model via matrix elements $D_{\mathrm{i,f}}(\mathbf{p}) = \langle\psi_{\mathrm{f}}(\mathbf{p})|\hat{D}|\psi_{\mathrm{i}}\rangle$ for dipole transitions from an initial atomic state $|\psi_{\mathrm{i}}\rangle$ to a final state $|\psi_{\mathrm{f}}(\mathbf{p})\rangle$. In this final state, the photoelectron is described by a scattering wave, while the state of the remaining ion is described by a set of quantum number (such as L_{ion} and M_{ion}), which we collectively denote by the index f. These matrix elements, evaluated with the aid the multi-configurational Hartree-Fock method [4], enter the conventional formalism describing laser-assisted photoionization by a short pulse of extreme ultraviolet (XUV) radiation. The probability density for observing a photoelectron with a momentum $\mathbf{p}$ is represented as an incoherent sum $S(\mathbf{p}) = \sum_{\mathrm{i,f}} S_{\mathrm{i,f}}(\mathbf{p})$ of contributions from individual channels, which we evaluate within the Coulomb-Volkov approximation:

$$S_{\mathrm{i,f}}(\mathbf{p}) = \left| \int_{-\infty}^{\infty} dt\, \mathcal{E}_{\mathrm{X}}(t) D_{\mathrm{i,f}}(\mathbf{p} + \mathbf{A}(t)) e^{i\Phi_{\mathrm{i,f}}(\mathbf{p},t)} \right|^2 . \quad (1)$$

Here, $\mathcal{E}_{\mathrm{X}}(t)$ is the envelope of the XUV pulse, $\mathbf{A}(t)$ is the vector potential of the streaking field, and the phase $\Phi_{\mathrm{i,f}}$ is defined as $\Phi_{\mathrm{i,f}}(\mathbf{p},t) = \left(\frac{p^2}{2} - W_{\mathrm{i,f}}\right) t - \int_t^{\infty} \left(\mathbf{p}\mathbf{A}(t') + \frac{1}{2}A^2(t')\right) dt'$ with $W_{\mathrm{i,f}}$ specifying the centers of photoemission lines for each of the channels.

While simpler models correctly describe the shift and broadening of photoelectron spectra induced by the streaking field, the knowledge of accurate angle-dependent transition matrix elements $D_{\mathrm{i,f}}(\mathbf{p})$ is required for a more detailed analysis. In particular, we study the role of the momentum-dependent phase of these matrix elements. Also, we investigate how the probability of observing a photoelectron is modulated by the streaking field and show that this modulation has a non-trivial angular dependence.

References

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