High Resolution Metrology of Autoionizing States Through Raman Interferences

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Photoelectron interferometry with ultrashort light pulses is a powerful probe of the fast electron wavepackets; however, it has limitations on the energy resolution. In this study, we advanced ultrafast photoelectron interferometry to show that one can simultaneously obtain both high temporal and spectral resolution by stimulating Raman interferences with a laser pulse and monitoring the modification of the electron yield in a separate step. As a proof of the principle, we applied this approach to the autoionizing nf' wavepacket between the spin-orbit split ionization thresholds in argon. The experimental and theoretical results are shown in the figure 1. We demonstrate our ability to resolve the electronic composition and time evolution of the wave packet in exquisite detail. This highly sensitive probing and control of electron dynamics opens the path for study of molecular excited states.

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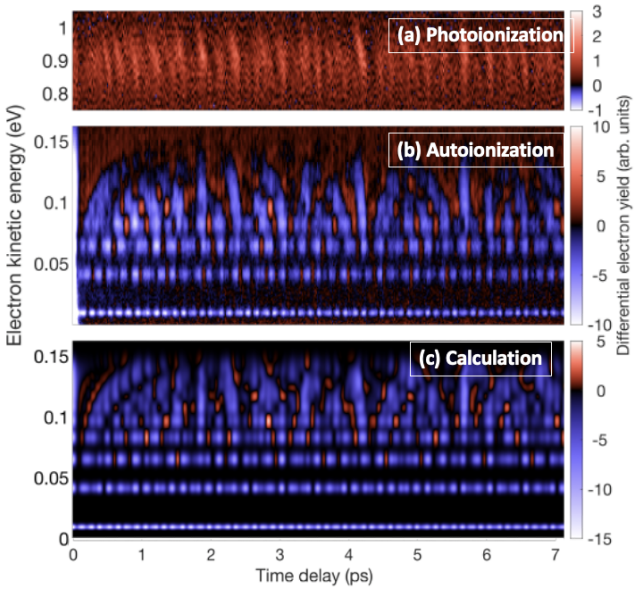

Figure 1: (a) Transient electron spectrograms corresponding to the photoionization (a) and autoionization (b) channels. (c) Theoretical results for the autoionization channel. The photoionization channel shows beating, but there is loss of kinetic energy resolution due to the probe bandwidth. The autoionization yield is modified by the coherent Raman redistribution caused by the probe. The delayed detection in autoionization channel provides excellent kinetic resolution, allowing high fidelity monitoring of the beats and wavepacket evolution

References

[1] A Plunkett, M Alarcón, J Wood, C H Greene and A Sandhu, Phys. Rev. Lett. **128**, 083001 (2022)