DAMOP19-2019-020148

Abstract for an Invited Paper
for the DAMOP19 Meeting of
the American Physical Society

Wave mixing with attosecond pulses: Multidimensional and transient grating spectroscopies

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By combining weak attosecond pulses with strong few-cycle NIR pulses, the emission of four- wave mixing signal fields in the extreme ultraviolet are observed in the laboratory. This development allows multiple avenues of atomic and molecular dynamics investigations to be pursued with background free sensitivity.

A multidimensional technique is developed to retrieve single state dynamics of Rydberg states of atomic argon, whereas complex superposition states are formed by the large bandwidth attosecond pulses. This four-wave mixing based technique utilizes a pulse shaper that imposes a tunable narrow band phase and amplitude modulation on the NIR spectrum. The new NIR energy axis permits the separation of the various field emissions, as made evident by the elimination of quantum beats in the time evolution.

Five orders of XUV wave mixing emission, corresponding to four- through twelve-wave mixing pathways, are observed within a noncollinear geometry in helium gas. In this excitation regime, wave-mixing emission is observed at the energies of light-induced states as well as the 1snp resonant states. Systematic few-femtosecond delays are observed between the various orders of emission signals. The emission delays and the signal scaling are successfully interpreted with the accumulation of a phase grating arising from the AC stark shift.

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