Title

Attosecond Noncollinear Four Wave Mixing

ISBN

9781665418768

Author

Leone, Stephen R

Publication Date

2021-06-25

DOI

10.1109/cleo/europe-eqec52157.2021.9542497

Peer reviewed

2021 Conference on Lasers and Electro-Optics Europe & European Quantum Electronics Conference (CLEO/Europe-EQEC) | 978-1-6654-1876-8/21/$31.00 ©2021 IEEE | DOI: 10.1109/CLEO/Europe-EQEC52157.2021.9542497

Attosecond Noncollinear Four Wave Mixing

Stephen R. Leone[1,2,3]

1. Department of Chemistry, 2. Department of Physics, and 3. Lawrence Berkeley National Laboratory, University of California, Berkeley, CA 94720 USA

Attosecond pulses produced by high harmonic generation are excellent sources of broadband excitation in the extreme ultraviolet. Such pulses can be used to produce coherent superpositions of electronic and vibrational states in atoms and small molecules. The resultant coherent superpositions, or wave packets, can then probed by a time-delayed optical pulse, which modifies the polarization of the medium by coupling to neighboring states. The time dependent spectral interferences in this method, called attosecond transient absorption in the collinear configuration, are recorded with outstanding temporal and spectral resolution to reveal electronic and vibrational structure and dynamics.

Through a recently successful method of attosecond four wave mixing that combines two optical/near infrared pulses with one attosecond extreme ultraviolet pulse [1], several noncollinear geometries are now used (Fig. 1) to generate new, directed emission beams that are wave-vector phase-matched at various angles and provide background-free measurements of wave packets, used to investigate short time dynamics. In nitrogen, the dynamics of specific high-lying electronic states are observed through interferences in individual vibrational levels. The results are measured with excellent spectroscopic accuracy. With the noncollinear four wave mixing geometry of two time-delayed near infrared pulses together with the extreme ultraviolet pulse, the dynamics of a vibrational wave packet on the inner and outer turning points of an electronic state is directly accessed [2]. In atoms (Fig. 1), the decays of autoionizing states are observed in the four wave mixing emission signals and lifetimes are obtained. In hydrogen, vibrational superpositions in two different electronic states in a spectrally congested region are selectively revealed. Using a spatial light modulator to remove or alter the phase of specific frequencies of one of the optical pulses, a new coherent multidimensional attosecond spectroscopy in the extreme ultraviolet is obtained [3]. Self-heterodyned signal amplification is also achieved, resulting in many orders of magnitude improvement in sensitivity [4]. In a solid such as sodium chloride, attosecond four wave mixing is used to disentangle features underlying the line broadening of core level excitons, to identify lifetimes and spectral features of localized core level exciton states on the sodium cations in the solid lattice.

In atoms, molecules, and solids, coherent electronic quantum beats are observed, and the results are compared to theoretical simulations of coherent dynamics though calculation of interrogated state structures and the optical-pulse-induced couplings. Future work will address attosecond core level dynamics by four wave mixing of extreme ultraviolet pulses targeting selected sites in molecules and solids.

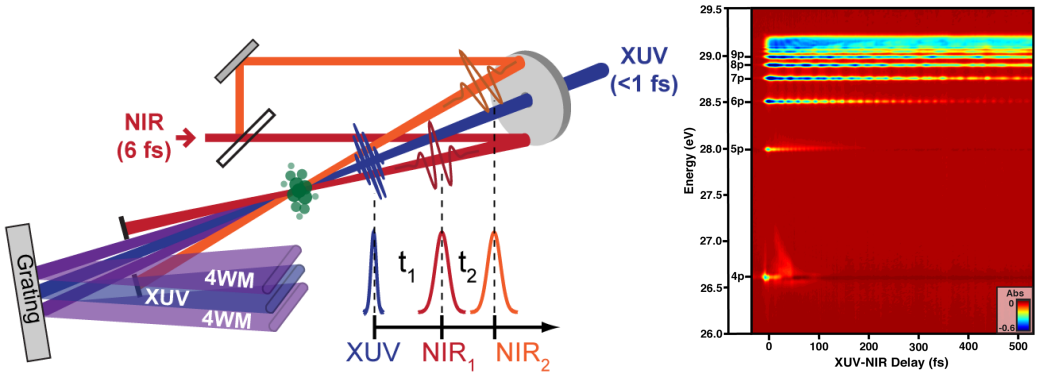

Fig. 1. (a) Attosecond four wave mixing arrangement (left). (b) background free quantum beats in Ar (right).

The speaker gratefully acknowledges the support of the Department of Energy AMOS program, collaborator Daniel Neumark, and co-workers Wei Cao, Erika Warrick, Ashley Fidler, Hugo Marroux, Etienne Bloch, James Gaynor, and Yen-Cheng Lin.

References

[1] W. Cao, E. R. Warrick, A. Fidler, D. M. Neumark, and S. R. Leone, "Noncollinear wave mixing of attosecond XUV and few-cycle optical laser pulses in gas-phase atoms: toward multidimensional spectroscopy involving XUV excitations," Phys. Rev. A **94**, 053846 (2016).
[2] E. R. Warrick, A. P. Fidler, W. Cao, E. Bloch, D. M. Neumark, and S. R. Leone, "Multiple pulse coherent dynamics and wave packet control of the N_2 a″ $^1\Sigma_g^+$ dark state by attosecond four wave mixing," Faraday Discuss **212**, 157 (2018).
[3] H. J. B. Marroux, A. P. Fidler, D. M. Neumark, and S. R. Leone, "Multidimensional spectroscopy with attosecond extreme ultraviolet and shaped near-infrared pulses," Sci. Adv. **4**, 1-7 (2018).
[4] A. P. Fidler, E. R. Warrick, H. J. B. Marroux, E. Bloch, D. M. Neumark, and S. R. Leone, "Self-heterodyned detection of dressed state coherences in helium by noncollinear extreme ultraviolet wave mixing with attosecond pulses," J. Phys. Photonics **2**, 034003 (2020).