Transient absorption and nonlinear spectroscopy with attosecond light pulses

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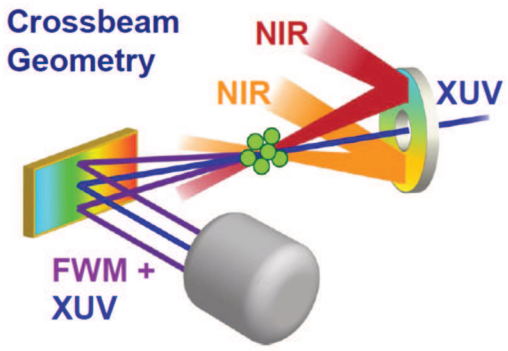
The development of attosecond light sources in the extreme ultraviolet (XUV) frequency regime opens up new opportunities in the general area of ultrafast dynamics.[1,2] Attosecond transient absorption enables the study of the photodissociation of CH_3Br and C_2H_3Br on a few femtosecond time scale. This species is excited with a few fs near infrared (NIR) pump pulse. Its subsequent absorption spectrum is probed with a broadband attosecond XUV pulse centered around 70 eV, where Br electronic transitions originating from the 3d core orbital occur. One can then use these time-evolving transition energies to follow the dissociation and ionization dynamics inducted by the pump pulse. In another effort, four-wave mixing (FWM) experiments have been developed using one XUV pulse and two NIR pulses in non-collinear geometries.[3] This experiment enables one to probe and spatially separate FWM signals resulting from couplings between excited electronic states in the XUV that are induced by the NIR pulses. This method is used to extract lifetimes of autoionizing states of Kr and to characterize the inner and outer wells of the *a*" dark state of N_2.



References

[1] K. Ramasesha, S. R. Leone, and D. M. Neumark, *Annu. Rev. Phys. Chem.* **67**, 41 (2016).

[2] S. R. Leone and D. M. Neumark, *Faraday Discuss.* **194**, 15 (2016).

[3] W. Cao, E. R. Warrick, A. Fidler, D. M. Neumark and S. R. Leone, *Phys. Rev. A* **94**, 053846 (2016).