Nonlinear optics of fast-ionizing media:  
From the nanosecond to attosecond time scale

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Ionization is one of the key processes accompanying the interaction of high-intensity laser radiation with matter. It generates charged particles, resulting in a dynamic modification of linear and nonlinear optical properties of the medium. Methods of nonlinear-optical spectroscopy, powered by modern laser technologies, offer unique tools for all-optical studies of complex physical processes involved in ultrafast ionization dynamics and the evolution of light fields in fast-ionizing media on a time scale from nanoseconds to attoseconds. A broad assortment of four-wave mixing techniques (Figs. 1a – 1c), including coherent anti-Stokes Raman scattering (CARS), has been used over the past decades to understand the dynamics of charged-particle generation and relaxation in gas and condensed media [1, 2]. In recent experiments, the nonlinear-optical methodology has been extended to enable time-resolved studies of an ultrafast build-up of the electron density in the ionized gas [3, 4], quantum-pathway-selective CARS spectroscopy of autoionizing states [5], and an all-optical mapping of attosecond electron tunneling dynamics [6, 7]. In nonlinear-optical bioimaging, an accumulation of free electrons generated by high-repetition-rate ultrashort laser pulses tends to initiate cascades of unwanted processes in biotissues, including the formation of reactive oxygen species, causing the death of cells, as well as DNA-strand breaking by low-energy electrons due to the rapid decay of transient molecular resonances localized on DNA constituents. These issues raise concerns regarding the noninvasiveness of nonlinear-optical neuroimaging techniques, calling for in-depth quantitative studies of ultrafast ionization phenomena accompanying nonlinear-optical interactions of laser pulses with brain tissues. An ionization-induced blue shift of the CARS signal can serve as an indicator of this ionization penalty in nonlinear-optical bioimaging [8]. This talk will give an overview of new techniques and approaches in the nonlinear-optical spectroscopy of fast-ionizing media.

References