IEEE Photonics Journal
An IEEE Photonics Society Publication

Invited Paper

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Volume 3, Number 2, April 2011

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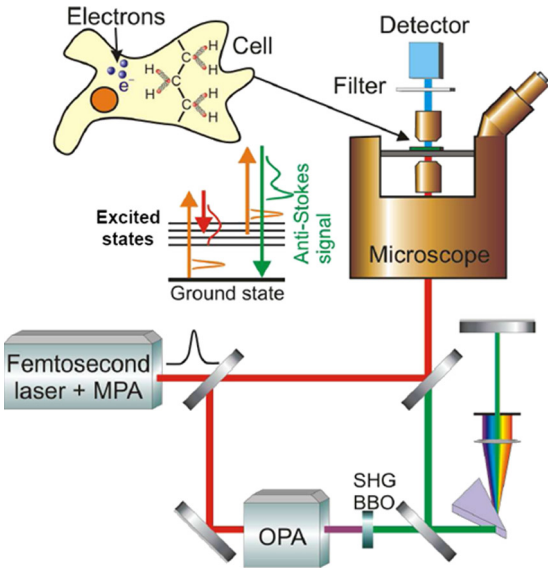

DOI: 10.1109/JPHOT.2011.2142180
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Nonlinear-Optical Probe for Ultrafast Electron Dynamics: From Quantum Physics to Biosciences

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(Invited Paper)

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DOI: 10.1109/JPHOT.2011.2142180
1943-0655/$26.00 ©2011 IEEE

Manuscript received February 18, 2011; revised March 24, 2011; accepted March 26, 2011. Date of current version April 26, 2011. Corresponding author: A. Zheltikov (e-mail: zheltikov@physics.tamu.edu).

Abstract: Recent discoveries in ultrafast optical science and advances in laser technologies offer unique tools for probing the extremely fast dynamics of bound and free electrons, giving the key to identifying the key scenarios and understanding the decisive early episodes of light-induced processes in physics, chemistry, and biology.

Index Terms: Ultrafast photonics, nonlinear optics, light–matter interactions.

The interaction of electromagnetic radiation with electrons is the key mechanism whereby the laser field acts upon matter. Laser fields with a sufficiently high intensity, which is typical of modern short-pulse laser sources, give rise to field-induced ionization of atoms and molecules, generating free electrons. Tunneling of an electron through a potential barrier distorted by a high-intensity light field is a key elementary event that launches a broad diversity of light-induced effects in physics, chemistry, and biology, turning on the clock in an ultrafast light–matter interaction. Since such an ionization occurs on an extremely short time scale, i.e., often faster than 1 fs, long before any other laser–matter interaction processes become effective, it defines time zero for cascades of complicated physical, chemical, and biological transformations in matter [see Fig. 1(a)]. While these later phases of light–matter interactions can be accessed with a number of advanced laser techniques using ultrashort light pulses [1], methods enabling a clear identification of the decisive early episodes in light–matter interactions related to extremely fast electron dynamics are still under development.

The cutting-edge technologies of attosecond spectroscopy developed in recent years are primarily based on the detection of the yield of charged particles [2]–[4]. These methods have resulted in revolutionary breakthroughs in our understanding of fundamental electronic dynamics in the gas-phase media and on solid surfaces. However, the detection of ultrafast electronic dynamics in the bulk of condensed-phase systems, including real-life chemical and biological systems, calls for further developments [5]–[7]. Recent discoveries in ultrafast nonlinear optics suggest new ways of confronting these challenges. Experiments performed at the Vienna University of Technology show [7] that, upon an accurate discrimination against the signal related to atomic nonlinear-optical susceptibilities and harmonics related to electron rescattering on parent ions, the spectra of optical

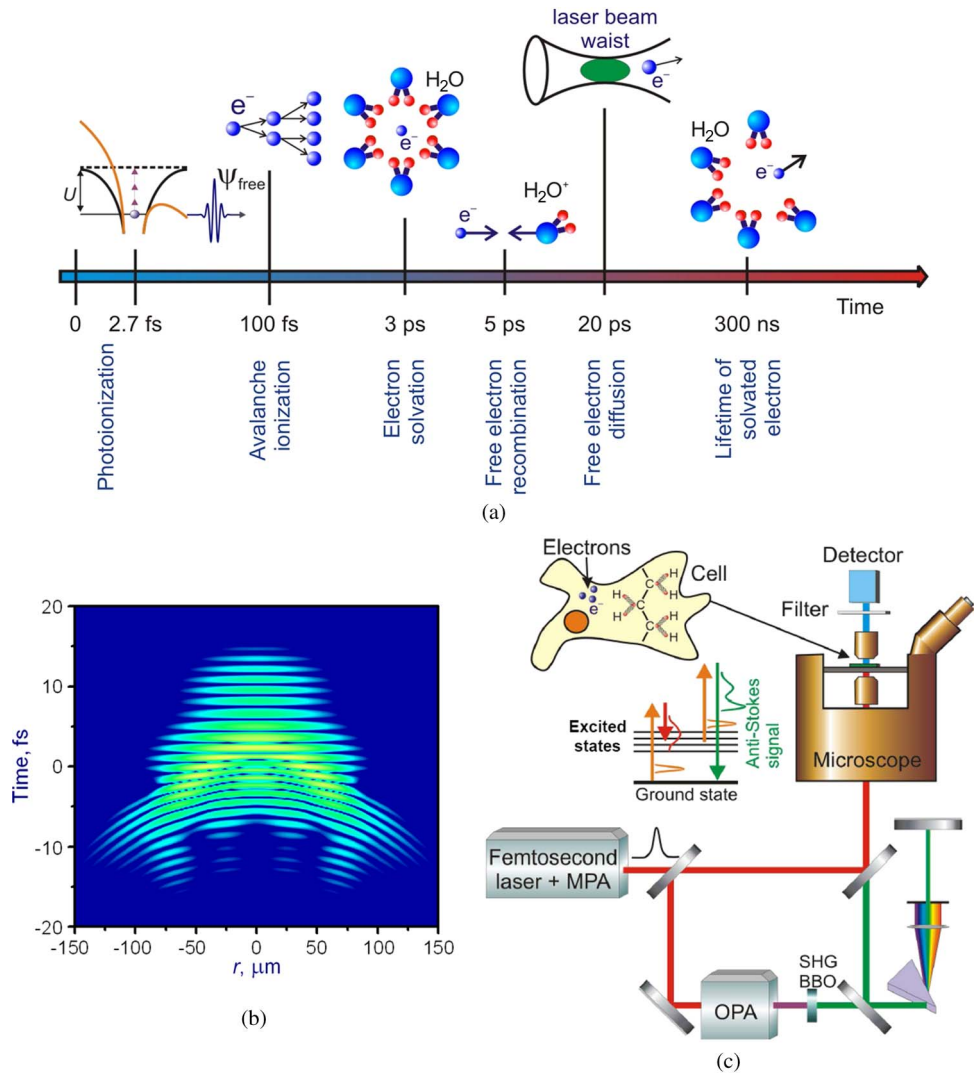

Fig. 1. (a) Timeline of ultrafast electron dynamics and electron-initiated processes in the field of ultrashort laser pulses, including electron photoionization through tunneling or a multiphoton process, electron salvation, capture of electrons into antibonding molecular orbitals, electron autodetachment, dissociation of molecules, recombination of free electrons, and dynamics of vibrational wave packets. (b) Spatiotemporal map of a few-cycle laser pulse propagating through an ionizing medium. (c) Schematic of nonlinear-optical bioimaging using coherent Raman scattering. MPA: multipass amplifier; OPA: optical parametric amplifier; SHG: second-harmonic generation.

harmonics and wave mixing can provide a wealth of information on electron tunneling dynamics. This approach allows subfemtosecond electron ionization dynamics to be detected without attosecond pulses but using a fraction of the field cycle of an ultrashort laser pulse as a subfemtosecond probe. More generally, oscillations of electromagnetic field in a few-cycle pulse are ideally suited to probe ultrafast dynamics of ionization [8], [9]. This argument is illustrated by the spatiotemporal map of a few-cycle pulse in Fig. 1(b). Compression of field half-cycles, which is clearly visible in this map, visualizes ionization-induced blue shift, while the defocusing dynamics, which are seen on the trailing edge of the pulse, maps both the spatial and temporal profiles of the electron-density buildup within the laser pulse.

In a standard beam-focusing geometry, laser-induced ionization is accompanied by the absorption of laser radiation along the entire beam-propagation path. A transverse profile of free electrons generated under these conditions displays a maximum on the beam axis, giving rise to a

negative lens, which defocuses the laser beam. These phenomena limit the efficiency of the interaction of laser radiation; maximum field intensity and the maximum electron density in the focus of the laser beam; prevent laser radiation energy from being efficiently deposited in a gas, liquid, or solid target; and impose serious restrictions on material micromachining, as well as on spectral and temporal transformation of high-power ultrashort laser pulses. In particular, in the filamentation regime, beam defocusing induced by free electrons limits the field intensity and the density of free electrons at levels dictated by the balance of beam focusing due to the positive lens related to the third-order nonlinear susceptibility of the medium (Kerr effect) and defocusing caused by the negative lens induced by the transverse profile of free-electron density [10]. Recent studies have shown [8] that a multibeam ionization of gas and condensed-phase media with interfering ultrashort laser pulses can help to substantially increase the maximum field intensity and the density of free electrons attainable in the focus of the laser field relative to the regime of single-beam ionization. Multibeam ionization schemes have been shown to offer new solutions for laser micro- and nanomachining, micro- and nanosurgery, spectral and temporal transformation of ultrashort light pulses, as well as remote sensing of the atmosphere. Subfemtosecond changes in the local refractive index induced in the regime of multibeam tunneling ionization enable the high-speed switching of optical signals.

In silicon photonics, which offers an advanced platform for the creation of broadband on-chip components and integrated networks for emerging optical information technologies [11], ultrafast electron dynamics can help to implement ultrafast switches and finely tunable frequency comb generators [12]. In recent experiments [13], a photonic platform integrating a silicon nanowaveguide ring resonator and a photonic-crystal fiber (PCF) frequency shifter has been developed. These studies demonstrate that the ringdown response of a silicon nanowaveguide ring resonator can be efficiently controlled through ultrafast light-induced free-carrier generation by a femtosecond laser pulse. Recent theoretical studies, on the other hand, show [14] that the enhancement of multiphonon tunneling recombination of free carriers in strong laser fields offers a channel whereby ultrafast carrier-density dynamics in a semiconductor can be controlled by properly shaped laser pulses. This regime of laser–solid interaction enables an ultrafast switching of optical and electric properties of semiconductor materials, suggesting new strategies for laser micro- and nanomachining, optical data processing, and ultrafast plasmonics.

Modern optical technologies offer a broad variety of powerful methods and tools for chemically selective high-resolution microspectroscopy and imaging [see Fig. 1(c)] of biochemical processes and biological objects [15]–[18]. Because of the generic I^N scaling of an N-photon response of a material to a laser field with intensity I, high laser intensities are needed to provide a high sensitivity, a high signal-to-noise ratio, and a high image-acquisition speed in nonlinear-optical neuroimaging. The flip side of bioimaging with high laser intensities is an increased risk of irreversible light-induced modifications and damage of biotissues [19]. Accumulation of free electrons generated by ultrashort laser pulses with intensities below the single-pulse laser damage threshold 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 [20]. These issues raise concerns regarding the noninvasiveness of nonlinear-optical imaging techniques, calling for in-depth quantitative studies of ultrafast ionization phenomena accompanying nonlinear-optical interactions of laser pulses with brain tissue. Recent experiments at M. V. Lomonosov Moscow State University demonstrate [21] that free-electron generation that accompanies coherent anti-Stokes Raman scattering (CARS) of ultrashort laser pulses in brain tissue [see Fig. 1(c)] manifests itself in a detectable blue shift of the anti-Stokes signal. Experimental studies suggest that this blue shift can be used to quantify the ionization penalty of CARS-based neuroimaging.

The advent of laser systems capable of routinely generating fully controlled few-cycle light pulses leads us to rethink and redefine the concepts of fast and slow in natural sciences. In this new era of ultrafast science, methods for detecting and understanding the early phases of light–matter interactions are in great demand. Nonlinear optics is among the most promising approaches, as it opens routes to unexplored territories and otherwise inaccessible fundamental events and

processes, giving the key to identifying and understanding the decisive early episodes of light-induced processes in physics, chemistry, and biology.

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