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X-ray microprobe of optical strong-field processes

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Abstract

A time-resolved X-ray microprobe to study optical strong-field processes has been developed. Individual atoms or molecules located within the strong-field environment created by a focused ultrafast laser are probed by undulator-produced X-ray pulses to achieve spatial, temporal, spectral and polarization selectivity. Approximately 10^6 monochromatic X-rays per 100-ps pulse are focused into a $\sim$10 μm spot to selectively probe atoms in focal volumes where intensities up to 10^{15} W/cm^2 can be present. In this paper, we describe the time-resolved X-ray microprobe and provide some illustrative examples from our work studying strong-field phenomena such as laser-modified absorption spectra, Coulomb explosion, transient laser-produced plasmas and molecular alignment.

Keywords: X-ray; Pump-probe; Strong-field effects; Synchrotron radiation; Ultrafast X-rays; Microfocus X-rays

1. Introduction

The study of structural dynamics on an ultrafast time scale is a prime motivation for the construction of large-scale next-generation X-ray sources, i.e. free electron lasers (XFELs), such as the Linac Coherent Light Source (Arthur et al., 2002) and the Tesla Test Facility (Tschentscher, 2004). These new sources will access the sub-picosecond timescale with very high single pulse fluence ($\sim 10^{13}$ X-rays/200 fs pulse) in comparison with current synchrotron sources ($\sim 10^8$ X-rays/100 ps pulse) and laser-produced plasma sources ($\sim 4 \times 10^6$ X-rays/sr/100 fs pulse) (Bargheer et al., 2004). X-rays are particularly valuable in ultrafast dynamics research, since they provide complementary information to that available from optical and charged particle probes. X-rays yield direct information on atomic positions and chemical bond lengths, unlike ultrafast optical probes (Zewail, 2003). X-rays penetrate to probe isolated molecules in solution, buried interfaces and bulk material response, unlike recently developed ultrafast electron sources (Zewail, 2005; King et al., 2005; Siwick et al., 2003). In addition, tunable X-rays from synchrotron sources provide chemical, elemental and polarization specificity (Stöhr, 1996). To this suite of synchrotron basics, one can add "ultrafast" temporal *and* "ultrasmall" spatial information using the laser-pump/X-ray microprobe method described here, thus enabling new studies of the response of atoms and molecules to strong-optical fields.

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Recently, there has been an explosion of activity at third-generation light sources using pump-probe techniques to track structural dynamics with ~100 ps X-ray pulses. These studies have been of solid-state materials (~10^{22} atoms/cm^3) and solvated species (~10^{19} molecules/cm^3). In ordered solid-state materials, phase-transitions and phonon propagation have been studied extensively, primarily using X-ray diffraction techniques (Sondhauss et al., 2005; DeCamp et al., 2003; Reis et al., 2001; Lindenberg et al., 2000). In some of these studies, the time resolution was extended below the X-ray pulse duration to the few ps level through the use of X-ray streak cameras. Particularly noteworthy is the recent demonstration using ~100 femtosecond pulses from a laser-slicing source (Schoenlein et al., 2000) to study metal–insulator phase-transition dynamics with extended X-ray absorption techniques (Cavalleri et al., 2005). Studies of photoinduced structural dynamics of solute molecules have been done using both X-ray absorption spectroscopy (Chen et al., 2001; Saes et al., 2003) and X-ray diffraction (Neutze et al., 2001; Plech et al., 2004). Some recent achievements include the time-resolved study of chemical reactions and the isolation of a molecular transition state in solution by X-ray diffraction (Davidsson et al., 2005; Ihee et al., 2005). Access to the subpicosecond regime with hard X-rays has already started with experiments on crystal melting at the Sub-Picosecond Pulse Source (SPPS), the precursor to the LCLS (Lindenberg et al., 2005).

Our experiments, in contrast, focus on gas phase samples ($\leqslant 10^{14}$ cm^3) with no long-range order. Also in contrast to other pump-probe studies, we use strongly focused X-ray beams (~10 μm spot diameter). The combination of strongly focused tunable X-rays with a strongly focused ultrafast laser enables both the study of atomic and molecular response to strong-optical fields and an exploration of laser-induced modification of standard X-ray processes. In addition, gas phase studies are special in that X-ray interactions with isolated atoms and molecules are amenable to first-principles theory and will provide a solid foundation for pump-probe experiments with XFELs, where strong-optical fields are expected.

The response of atoms and molecules to strong-electromagnetic fields is of significant intrinsic interest. Within the past two decades, developments in ultrafast laser technology have powered revolutionary advances in the study of light-matter interactions at high intensities (Brabec and Krausz, 2000). The diffraction-limited quality of the ultrafast laser pulses permits efficient focusing and thus with modest mJ pulse energies one can easily reach intensities of 10^{15} W/cm^2, corresponding to electric field strengths of ~10 V/Å. These external field strengths are comparable to those binding outer-shell electrons in an atom and manipulation of these electrons with strong-laser fields has led to the discovery of many fascinating phenomena, e.g. above threshold ionization (Agostini et al., 1987), high-harmonic generation (Salières et al., 1999) and molecular alignment (Stapelfeldt and Seideman, 2003). At higher intensities in the relativistic regime ($>10^{16}$ W/cm^2), there have been observations of laser-induced X-ray generation in clusters (McPherson et al., 1994; Ditmire et al., 1995) and even nuclear fusion (Ditmire et al., 1999).

These experiments typically use a single intense laser pulse to irradiate a gas-phase sample and analyze the products (electrons, ions, photons) to gain information on the interaction mechanisms. An independent, tunable ultrafast probe would add a valuable dimension to these studies, permitting the interrogation of state-specific atomic dynamics induced by the strong field. Earlier studies which use pump-probe techniques to study strong-field light-matter interactions have generally derived pump and probe pulses from the same laser, thus limiting both spectral tunability and pump-probe time delay range. In a pioneering experiment, Schins et al. (1994) made the first observation of laser-assisted Auger decay by using output from a single Ti:sapphire laser to provide both the "pump" IR dressing beam of $<10^{12}$ W/cm^2 and the broadband "probe" X-rays generated from laser-irradiated liquid Ga. Subsequently, Glover et al. (1996) made the first observation of the laser-assisted photoelectric effect using an IR dressing beam combined with a family of high-harmonic probe beams within an energy range of 33–42 eV. In both cases, the Ti:sapphire lasers were multistage-amplified, and had low repetition rate and high pulse energy (10 Hz, ~40–65 mJ). In these two experiments, effect of the laser-dressing field is to cause free–free transitions in the continuum adding sidebands to the photoelectric or Auger peak in the electron energy spectrum. More recently, high harmonics have been used as probes of transient molecular alignment, (Kanai et al., 2005) again deriving the pump and probe from the same laser.

With the X-ray microprobe, there are many advantages relative to pump-probe experiments derived from a single laser. First, spectral tunability: a probe X-ray beam derived from synchrotron radiation provides wide tunability (chemical and elemental sensitivity) and independent polarization control. Second, temporal tunability: independent control of the pump laser beam and the probe X-ray beam allows delay ranges from picoseconds to milliseconds. This enables the probing of properties of individual atoms and molecules at $t = 0$ and probing of collective dynamics of the gas-phase ensemble, e.g. Coulomb explosion, at later times. Third, microfocus advantages: the micron-sized X-ray beam allows both easy target replenishment with a flowing gas or liquid source to avoid sample damage, and, efficient use of laser pulse energy. For X-ray pulses of spot diameter d separated by time Δt a flow speed of only