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FRONTIERS ARTICLE

Probing ultrafast dynamics with attosecond transient absorption

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ABSTRACT

The broad bandwidth of an isolated attosecond pulse excites a vast number of states simultaneously, and the corresponding absorption features can be monitored with exceptional temporal resolution. Novel transient absorption experiments in gases using isolated attosecond pulses are performed in two regimes, one in which the attosecond pulse is overlapped in time with a near-infrared (NIR) pulse and one in which the NIR pulse follows the attosecond pulse. In the latter regime, the attosecond pulse first interacts with a sample, then the observed absorption features are modified by a NIR pulse, which interacts with the sample well after the attosecond pulse has passed. In these experiments, which seem counterintuitive when compared to conventional transient absorption spectroscopy, the weak attosecond pulse induces a polarization of the medium, which is then perturbed by the time-delayed NIR pulse. Recent measurements demonstrate the rich variety of information that can be extracted in this regime.

1. Introduction

Electronic dynamics in atoms and molecules typically occur on a few-femtosecond or sub-femtosecond timescale. These processes are too fast to be resolved by ordinary femtosecond optical laser pulses. However, in the past ten years, techniques to generate isolated pulses with durations of a few hundred attoseconds, or even shorter, have been established. These isolated attosecond pulses provide unprecedented opportunities to study ultrafast electron dynamics on a few-femtosecond timescale.

Isolated attosecond pulses are generated using high harmonic generation, in which a femtosecond laser pulse is upconverted to produce photon energies in the extreme ultraviolet (XUV) [1], in conjunction with optical gating techniques. The short duration of the pulse necessarily results in a broad and continuous spectrum, with bandwidths ranging from a few electron volts (eV) to tens of eV and central photon energies typically between 10 eV and 100 eV. These ultrashort pulses have been used in many successful experiments incorporating photoelectron and ion detection methods. For example, attosecond pulses have been used to measure the few-femtosecond timescale of Auger decay after core excitation of krypton [2] and to measure the time delay between the emission of an electron from two different atomic orbitals in neon [3].

The broad and continuous bandwidth of an isolated attosecond pulse, with photon energies in the extreme ultraviolet (XUV), is well suited to absorption measurements. The XUV photon energies can access highly excited states such as Rydberg states approaching the ionization limit or autoionizing states embedded in the ionization continuum. Electronic dynamics can be tracked in real time by monitoring the resulting changes in the absorption spectrum.

The majority of attosecond transient absorption experiments have been performed on rare gas atoms, which are relatively simple systems that still manifest complex behavior. However, the recent application in our group of attosecond transient absorption to observe the transfer of electrons from the valence band to conduction band in solid silicon [4] illustrates the generality of the technique. Attosecond transient absorption will soon be extended to study gas phase molecular systems, but the focus of this article is the experiments performed on rare gas atoms, in which a variety of novel phenomena have been identified.

Isolated attosecond pulses can be utilized in novel transient absorption experiments, in which the XUV attosecond pulse first passes through the sample, then a near-infrared (NIR) laser pulse follows at a time delay. It may seem counterintuitive that a change in the absorption spectrum of the XUV pulse can be observed when the NIR pulse follows the XUV pulse. This unconventional pulse order is possible because the XUV pulse induces a polarization of the medium. This polarization persists long after the XUV pulse passes through the sample, and it dephases on a timescale that depends on the lifetime of the states excited, which can range from tens of

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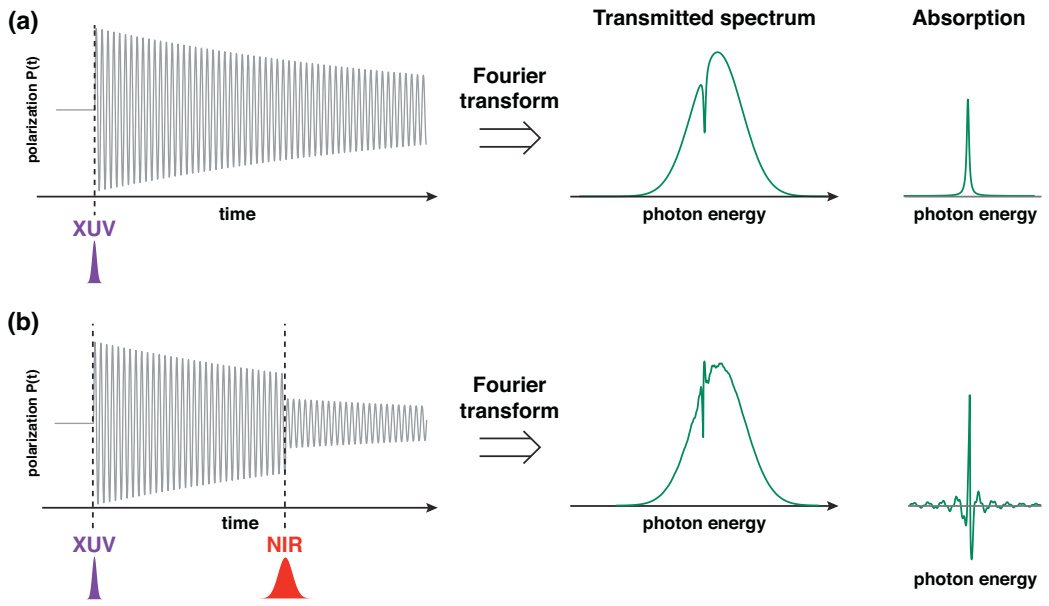

Figure 1. Simplified diagram of transient absorption. (a) XUV-only absorption measurement. The XUV pulse induces a polarization in the sample (left). The transmitted XUV spectrum (middle) shows a clear absorption feature and the calculated absorption (right) displays a single peak. (b) The NIR pulse can perturb the polarization at some delay (left), altering the transmitted spectrum (middle) and creating sidebands or altered absorption lineshapes in the expected absorption (right).

femtoseconds for a short-lived autoionizing state to nanoseconds for an atomic Rydberg level. The subsequent NIR pulse perturbs this polarization when it interacts with the sample. The macroscopic polarization is formed by a collection of oscillating dipole moments, which emit light. This emission is then dispersed in a spectrometer, essentially measuring the Fourier transform of the time-dependent signal. Absorption features are observed due to destructive interference between the emission from the excited sample and the transmitted broadband pulse. Because the induced polarization has been perturbed by the NIR pulse, the measured absorption features change. This is shown schematically in Figure 1.

The temporal resolution of a transient absorption experiment depends on the duration of the laser pulses used in the experiment while the resolution in the frequency domain depends only on the energy resolution of the spectrometer, as described by Pollard and Mathies [5]. This is because the final absorption measurement, which is the signal at the detector integrated over all times, is not time-dependent. Therefore, attosecond transient absorption measurements can be performed with both excellent temporal resolution and good spectral resolution.

In conventional transient absorption spectroscopy, a weak broadband pulse is used to measure the absorption spectrum of a sample. A pump pulse initiates dynamics, and changes in absorption features are observed when the broadband pulse follows the pump pulse. These methods have been successfully extended to XUV photon energies. In the femtosecond experiments of Loh et al. [6], a femtosecond near-infrared (NIR) laser pulse is used to induce strong-field ionization in a sample. Afterwards, the absorption of the XUV pulse is measured, and changes in the absorption are observed as the sample becomes ionized. Analogous experiments using attosecond pulses instead of an XUV pulse train were initially proposed by Pfeifer et al. [7]. Later experiments using isolated attosecond pulses by Goulielmakis et al. [8] detected quantum beating in a wavepacket created in Kr^+ after strong-field ionization.

Attosecond transient absorption experiments can thus be divided into three distinct categories based on the relative time delay between the NIR and attosecond pulse: relative time delays when the NIR pulse precedes the attosecond pulse, when the NIR and attosecond pulses are overlapped in time, and when the NIR pulse follows the attosecond pulse. In conventional transient absorption experiments, the range of time delays when the NIR pulse follows the XUV pulse might be deemed uninteresting, as no dynamics can occur before the pump pulse has interacted with the system. However, important recent results in attosecond science have capitalized on transient absorption experiments in which the NIR pulse overlaps with or follows after the attosecond pulse. This Frontiers article focuses on the rich variety of dynamics that can be observed and characterized in these two regions of relative NIR/attosecond pulse time delay.

2. Experimental techniques of attosecond transient absorption

A generalized experimental apparatus for attosecond transient absorption is shown in Figure 2. Similar apparatuses used by other groups are described in detail by Chini et al. [9] and Ott et al. [10]. A NIR pulse (duration of approximately 7 fs) is first split by a beam splitter in an interferometer. One arm of the interferometer generates the attosecond pulse, while the other arm acts as the NIR probe pulse. The XUV pulse generating arm may pass through optics to implement an attosecond optical gating technique, such as Double Optical Gating [11], which manipulates the laser field in order to produce a single attosecond pulse. The pulse then enters the vacuum system and is focused into a gas cell, in which the high harmonic generation process occurs. After this gas cell, an XUV attosecond pulse is produced and propagates collinearly with the residual femtosecond laser field. This residual light is subsequently blocked by a metal foil, which also acts as a filter for the attosecond pulse spectral bandwidth. The attosecond pulse is then focused by a gold-coated grazing incidence toroidal mirror.

The NIR probe arm propagates outside the vacuum system. A relative delay can be introduced in this arm, and fused silica can be added for optimal pulse compression. In the apparatus shown here, the NIR pulse is first focused by a spherical focusing mirror and then is recombined with the XUV pulse by a mirror with a small hole drilled in the center. The XUV pulse passes through the hole in the center of the mirror, while the NIR pulse reflects off the mirror, resulting in an annular beam.