NRC Publications Archive Archives des publications du CNRC

Demonstration of attosecond ionization dynamics inside transparent solids

Gertsvolf, M.; Spanner, M.; Rayner, D. M.; Corkum, P. B.

This publication could be one of several versions: author's original, accepted manuscript or the publisher's version. / La version de cette publication peut être l'une des suivantes : la version prépublication de l'auteur, la version acceptée du manuscrit ou la version de l'éditeur.

For the publisher's version, please access the DOI link below. / Pour consulter la version de l'éditeur, utilisez le lien DOI ci-dessous.

Publisher's version / Version de l'éditeur:

<https://doi.org/10.1088/0953-4075/43/13/131002>

Journal Of Physics. B, Atomic, Molecular And Optical Physics, 43, 13, p. 131002, 2010-07-23

NRC Publications Record / Notice d'Archives des publications de CNRC:

<https://nrc-publications.canada.ca/eng/view/object/?id=85928698-68be-4eca-ae35-ac55d8ce57ff>

<https://publications-cnrc.canada.ca/fra/voir/objet/?id=85928698-68be-4eca-ae35-ac55d8ce57ff>

Access and use of this website and the material on it are subject to the Terms and Conditions set forth at

<https://nrc-publications.canada.ca/eng/copyright>

READ THESE TERMS AND CONDITIONS CAREFULLY BEFORE USING THIS WEBSITE.

L'accès à ce site Web et l'utilisation de son contenu sont assujettis aux conditions présentées dans le site

<https://publications-cnrc.canada.ca/fra/droits>

LISEZ CES CONDITIONS ATTENTIVEMENT AVANT D'UTILISER CE SITE WEB.

Questions? Contact the NRC Publications Archive team at

PublicationsArchive-ArchivesPublications@nrc-cnrc.gc.ca. If you wish to email the authors directly, please see the first page of the publication for their contact information.

Vous avez des questions? Nous pouvons vous aider. Pour communiquer directement avec un auteur, consultez la première page de la revue dans laquelle son article a été publié afin de trouver ses coordonnées. Si vous n'arrivez pas à les repérer, communiquez avec nous à PublicationsArchive-ArchivesPublications@nrc-cnrc.gc.ca.

Demonstration of attosecond ionization dynamics inside transparent solids

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

2010 J. Phys. B: At. Mol. Opt. Phys. 43 131002

(<http://iopscience.iop.org/0953-4075/43/13/131002>)

View [the table of contents for this issue](#), or go to the [journal homepage](#) for more

Download details:

IP Address: 132.246.118.152

The article was downloaded on 24/02/2011 at 19:29

Please note that [terms and conditions apply](#).

FAST TRACK COMMUNICATION

Demonstration of attosecond ionization dynamics inside transparent solids

M Gertsvolf^{1,2}, M Spanner², D M Rayner² and P B Corkum^{1,2}

¹ University of Ottawa, Ottawa, Ontario, K1N 6N5, Canada

² National Research Council of Canada, Ottawa, Ontario, K1A 0R6, Canada

E-mail: David.Rayner@nrc.gc.ca and Paul.Corkum@nrc.gc.ca

Received 7 January 2010, in final form 3 May 2010

Published 23 June 2010

Online at stacks.iop.org/JPhysB/43/131002

Abstract

Attosecond science has arisen from intense light pulses interacting with low density gases. We show that the initiating process—sub-cycle ionization—also survives in large band gap condensed media. Using fused SiO₂ as an example, we measure the differential nonlinear absorption between the major and minor axis of elliptically polarized light. Through simulations that include ionization and light propagation, we confirm that changes in the ellipticity between the incident beam and the transmitted beam encode sub-cycle absorption dynamics. As the pulse duration is increased, we observe that sub-cycle ionization is masked by collisional processes. We propose a general class of methods for measuring attosecond dynamics in condensed media.

(Some figures in this article are in colour only in the electronic version)

The natural time scale of electronic processes in solids must encompass the sub-femtosecond or attosecond range. Despite this, the recent major advances in attosecond science are almost exclusively based on atoms and molecules in the gas phase [1]. In atoms and molecules it is established that attosecond dynamics are accessible through intense laser fields because tunnel ionization localizes electron release on a sub-cycle basis. This is due to the exponential dependence of the tunnelling process on the instantaneous electric field. As ionization, leading to electron–hole pair generation in wide band-gap dielectrics, is analogous to ionization to the continuum in atoms and molecules [2], dielectrics are a natural place to begin to explore attosecond science in solids. Many questions are open: (i) Molecular structure influences attosecond dynamics [3]—does crystal symmetry [4] also influence dynamics? (ii) Tunnelling creates bound-state electronic wave packets in molecules [5]—are electronic wave packets being created by tunnelling in solids? (iii) Does the solid-state equivalent of enhanced ionization in molecules [6] influence tunnelling dynamics? (iv) Electron scattering following ionization is the essence of attosecond science [7]—can we directly measure characteristics of electron scattering (the first step in avalanche ionization) in highly disordered

materials? (v) Finally, in gases the electron always responds with the real electron mass. In solids the band structure gives an electron an effective mass. Is there a time scale for the electron to assume its effective mass?

To answer these and other related questions requires a new metrology. The ion, electron and high-harmonic spectroscopies that have enabled attosecond science in the gas phase [1] are not available inside solids. Fortunately, the high density in solids, the very property that precludes these approaches, provides an alternative method that we exploit. This new diagnostic is based on modification of the laser beam itself during the interaction.

Dielectrics, band gap $\Delta E \sim 10$ eV, are transparent for low intensity near-infrared light. Until now, in solids, only the pulse envelope has been analysed [8–10], ignoring any sub-cycle information it carries. Using experiment and simulation, we show that sub-cycle information is carried in changes in laser ellipticity. Figure 1(a) shows schematically the expected change in polarization due to Kerr nonlinearity and other delayed nonlinear propagation effects that depend on the laser intensity envelope. Figure 1(b) shows the change caused by differential absorption during the laser cycle. We use fused

Figure 1. Schematics of polarization changes due to Kerr nonlinearity (ellipse rotation [11]) (a) and differential absorption (b). The incident and the transmitted laser polarization ellipsoids are shown with solid and dashed lines correspondingly. In (a) the ellipticity, ϵ , remains the same, in (b) ϵ increases.

SiO_2 , as a typical wide band gap dielectric. In the case of fused silica the efficient generation of third or higher harmonic is not present due to high dispersion. We find that multiphoton absorption maximizes near the peak of the field—the major axis of the ellipse—and is minimal when the field points towards the minor axis (figure 1(b)).

For our experiment we use a microscope objective to focus femtosecond laser pulses inside solid. This allows us to achieve intensities required for multi-photon ionization at pulse powers below the self-focusing power. Furthermore, the beam is depleted when the free carrier density reaches only about 0.1% of the atomic density of SiO_2 , because of the low fluence of ultrashort pulses. The resulting intensity clamping further suppresses the influence of the low order nonlinearities on the beam. Finally, the relatively low integrated free carrier density makes plasma refraction over the interaction length negligible. Thus, our measurements are robust—insensitive to propagation effects. Depletion, in our experiment, is directly related to the ionization probability [12].

Theoretically, we concentrate on the idea of laser depletion in strong field ionization. We approximate a dense, large band gap material by a dense gas of non-interacting atoms. This allows us to leverage our knowledge of the attosecond response of atomic media with enough atoms for sub-cycle absorption.

The total absorption includes contributions from (i) direct multiphoton free carrier generation, (ii) generation of odd-harmonics and (iii) inverse Bremsstrahlung absorption (by the generated free carriers) leading to avalanche ionization. The model allows us to test the first two contributions. In addition, we minimize, experimentally, any contribution from harmonic generation through the very short phase matching length of SiO_2 . Inverse Bremsstrahlung and avalanche ionization are not in our model. We evaluate their impact experimentally by exploiting the fact that both require a build-up in the free carrier concentration. We measure sub-cycle absorption as a function of the pulse duration. We find the ellipticity changes to be less important for longer pulses (where avalanche ionization plays a larger role). We evaluate the impact of collisions theoretically by arranging absorbing boundary conditions to absorb the electrons in about one mean free path.

Our experiment was performed with 800 nm, 43 fs (FWHM) pulses which could be stretched by tuning the grating compressor to a longer pulse. We used a microscope objective

Figure 2. Polarization state analysis of the transmitted light. Peak normalized transmission through a polarization analyser as a function of its angle. Input pulse energies are shown in the legend. The horizontal shift is due to the ellipse rotation ($\rightarrow$). The change of the minimal value is due to the ellipticity increase ($\uparrow$).

with a numerical aperture of 0.25 to focus the laser beam $\sim 150 \mu\text{m}$ inside a fused silica sample. Our pulse energies would have ranged from 20 to 300 nJ corresponding to intensities between 0.5×10^{13} and $7 \times 10^{13} \text{ W cm}^{-2}$ if the focus were in vacuum. The transmitted beam was collimated using an identical microscope objective. By translating the sample, we ensured fresh material for each laser shot.

We controlled the polarization of the input beam with a quarter-wave plate before the focusing objective. We analysed the transmitted polarization using a rotating polarizing prism, followed by a photodiode. The intensity measured by the photodiode is $I = I_0(\cos^2(\phi) + \epsilon^2 \sin^2(\phi))$, where ϕ , is the polarizer rotation angle relative to the major axis of the polarization ellipsoid of the incident light and the ellipticity, ϵ , $= \sqrt{I_{\min}/I_{\max}}$.

Figure 2 shows the polarization of the transmitted laser light for four different incident pulse energies. As the laser pulse energy increases, the ellipsoid rotates and ϵ increases. The position of the curve with respect to the analyser angle, ϕ , shows the orientation of the polarization axis. We can see that the 35 and 100 nJ curves are progressively shifted with respect to the lowest 13 nJ case (as shown with the horizontal arrow). Ellipse rotation is a $\chi^{(3)}$ effect [11]. The rotation direction follows the polarization handedness of the incident light and the amount of rotation, $\Delta\phi$, initially grows linearly with laser intensity (figure 3(a)) and then saturates after the onset of absorption that clamps the light intensity at the level determined by the ionization threshold [12] (the vertical line located at the inflection point of the dotted transmission curve).

The second energy-dependent effect in figure 2 is the increase in the minimal value of the curves indicated by the vertical arrow. For all measurements we selected $\epsilon \approx 0.58$ for the incident laser field, chosen to give the highest signal to noise level in ellipticity changes, $\Delta\epsilon$. With the peak of the curves normalized to 1, the minimal value is a direct measurement of the square of the field ellipticity.

Figure 3(b) shows $\Delta\epsilon$ as a function of the input pulse energy for a 43 fs pulse and compares it with the transmission curve. For pulse energies below the ionization threshold, the ellipticity remains constant, i.e. $\Delta\epsilon = 0$. At the onset of

Figure 3. (a,b,c) Experimental results. Transmission (small solid $\circ$, green online) and (a) ellipse rotation, $\Delta\phi$ ($\square$), (b,c) ellipticity change, $\Delta\epsilon$ (large $\circ$, red online (b) and $\triangle$ (c)), as a function of the incident pulse energy. Pulse duration 43 fs (a,b) and 115 fs (c). Input ellipticity $\epsilon = 0.58$ in all cases.

absorption, the ellipticity starts to increase, reaching $\epsilon \approx 0.64$ at three times the threshold pulse energy.

For longer pulses, inverse Bremsstrahlung and avalanche ionization become important [13]. Here we expect less reshaping of the laser electric field and, therefore, less change in the ellipticity of the transmitted light. With the pulse energy adjusted to ensure the same total absorption of 20%, figures 3(b) and (c) show that the ellipticity change for the 43 fs pulse is 7% while for the 115 fs pulse it is only 3%. The ellipticity change falls below our experimental noise level for a 180 fs pulse (not shown).

We now introduce a dense atomic gas model. Our aim is to illuminate the underlying concept of our experiment and to explore a wider range of parameters that would be relevant to more complex experiments. For this we need enough atoms to make absorption important. We also need to be sure that we understand the attosecond material dynamics. The non-interacting ideal gas model satisfies both criteria.

We do not expect quantitative agreement between the model and experiment. For this we would need to use the correct density of atoms; the correct atomic potentials with correct bound states; 3D electromagnetic wave propagation and collision modelling. Aside from this being a major theoretical undertaking, the added complexity would obscure the simplicity of the underlying concept of sub-cycle absorption.

Figure 4. Numerical modelling results for the laser intensity temporal profile as a function of the propagation depth.

We solve numerically the Maxwell propagation equation

$$\left[-\frac{\partial^2}{\partial z^2} + \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right] \vec{E}(z, t) = -\frac{4\pi}{c^2} \frac{\partial^2}{\partial t^2} \vec{P}(z, t)$$

and the Schrödinger equation for a one-electron atom

$$\left[i\frac{\partial}{\partial t} + \frac{1}{2} \frac{\partial^2}{\partial \vec{r}_\perp^2} - V(r_\perp) + \vec{r}_\perp \cdot \vec{E}(z, t) \right] \Psi(\vec{r}_\perp, t; z) = 0,$$

where $\vec{r}_\perp$ is coupled through the atomic polarization $\vec{P}(z, t) = N \langle \Psi(\vec{r}_\perp, t; z) | \vec{r}_\perp | \Psi(\vec{r}_\perp, t; z) \rangle$, N is the atomic density. All equations are in atomic units ($m_e = \hbar = e = 1$).

We first verify, with linearly polarized light, that the model describes the expected depletion of the laser energy by the solid [12]. For this we use a 1D approximation, where $r_\perp = x$ is the electronic coordinate and z is the laser propagation direction. The atomic binding potential, $V(r_\perp) = -0.6 \exp[-4 \ln 2 (r_\perp/1.7)^4]$, chosen to give a single bound state with ionization potential $I_p = 9$ eV equal to the SiO₂ band gap. Before the pulse arrives, all atomic population is in the ground state. Absorbing boundary conditions are used to model the loss of coherence of the electron due to electron-lattice scattering [14]. The boundaries smoothly attenuate the wavefunction over the range of 17–30 au.

For the calculation we used an input light intensity of $I_0 = 1.5 \times 10^{14}$ W cm⁻², a laser frequency of $\omega = 2.35 \times 10^{15}$ rad s⁻¹ (800 nm), a pulse duration (FWHM) of 20 fs, and a density of $N = 5 \times 10^{-4}$ au. This density corresponds to an average atomic separation of 6 Å; thus, our model is less dense than typical solids but still dense enough to support macroscopic absorption. In the calculation we filter the field to only pass frequencies up to 4.5 times the fundamental frequency. We do this because, in solids, the dispersion and the absorption of laser higher harmonics are very strong.

Figure 4 shows the changes in the pulse envelope that occur due to absorption at different propagation depths, L . The input pulse at $L = 0$ μ m is Gaussian. Following the strongest absorption between $L = 0$ and 4 μ m, the pulse intensity is reduced to the level where there is little ionization. Intensity

Figure 5. Numerical modelling results for the transmission (○, green online) and ellipticity (solid line, red online) as a function of the input pulse intensity.

clamping is so efficient that there is a very little difference between the 8 and the 12 μm curves. This basic behaviour is captured in an approximate model [12] that allows for an analytical description of laser absorption in solids.

Next we approximate the propagation and the absorption for elliptically polarized light by extending the model to 2D. Now $\vec{r}_\perp = (x, y)$, $r_\perp = \sqrt{x^2 + y^2}$ and the binding potential $V(r_\perp) = -0.945 \exp[-4 \ln 2 (r_\perp/2)^4]$, again chosen such that $I_p = 9$ eV. Figure 5 shows the change in ellipticity between the incident and the transmitted beams as a function of the pulse intensity for 20 fs pulse. The ellipticity remains constant for the low input pulse intensities. At higher intensities, beginning at the nonlinear absorption threshold, the $\Delta\epsilon$ (solid line) increases, indicating the change in polarization towards circular. This behaviour qualitatively agrees with experimental results shown in figures 3(a)–(c), confirming that sub-cycle ionization alone can produce a change in ellipticity.

Elliptical polarization has an inherent pump–probe structure with the major axis component of the field acting as the pump and the minor axis as the probe [15]. With a single beam, the pump and the probe are phase locked providing the equivalent of a fixed delay, limiting the resolution to $\sim 1/4$ laser cycle. The use of a second field, whose phase can be moved relative to the fundamental, can serve as a moving probe and improve the resolution. We illustrate the opportunity by studying the fundamental and its second harmonic. In centrosymmetric media, where even harmonics are not produced, the two colours, $\omega + 2\omega$, provide the most intuitive way to probe the sub-cycle absorption [16, 17].

Figure 6 demonstrates the idea. It shows the pulse profile prior to (a) and after (b, c) propagating through absorbing high density gas. The initial pulse, at $L = 0$ μm , is composed of equal intensity ω and 2ω beams with phase delay, $\delta\phi = 0$. The square of the instantaneous laser field amplitude consists of a large peak which appears at each period interspersed with two side peaks, initially of equal magnitude.

Figure 6(d) shows that the field reshapes after propagation through 3 μm of absorbing material. Absorption starts at the highest points of the field and extends for $\sim \pi/2$ radians that follow. This results in the asymmetric shape of the pulse. The two frequencies of the laser field, ω and 2ω , are attenuated and delayed differently due to the nonlinear polarization of the media. The attenuation and delay depend on the relative

Figure 6. Relative intensity of $\omega + 2\omega$ field before and after absorption. (a) Input field at $L = 0$ μm . (b, c) Pulse shape after absorption in 3 μm and 6 μm of dense gas. (d) Pulse reshaping, zoom-in on (b).

phase of the input beams. In our model there are no excited states. The polarization field can only be generated by the ground-to-continuum coupling (i.e. the tunnelling process). Only the changes in the tunnelling electron wavefunction that are defined by its attosecond ionization dynamics and electron–lattice scattering (boundary position and width in our model) are responsible for the attenuation and the delay that produce the waveform in figure 6(d).

Figure 6(c) shows the field profile at the depth of 6 μm inside the gas. Between 3 and 6 μm in the sample, the light intensity is reduced and the field shape remains nearly unchanged. In other words, the magnitude and the phase of polarization arising from the ground-to-continuum coupling depend nonlinearly on the laser intensity.

These calculations indicate how studying the attenuation and phase delay as a function of the input phase of the ω and 2ω beams over a range of intensities can provide a basis for characterizing attosecond dynamics in dielectrics. In the model, the low atomic density and lack of bound states effectively eliminates linear dispersion. Experimentally, the wavelength of the fundamental and its second harmonic can be chosen to have the same dispersion in the sample. Then, without phase walk-off in the interaction region, two-colour experiments provide a route to high temporal resolution measurements in solids.

The ultimate impact of attosecond technologies will be much wider if they can be realized in solid-state devices, rather than gas phase environments. We have demonstrated an approach to attosecond metrology in solids. Polarization-based nonlinear optical probes can form the basis for attosecond photonic devices. An initial device could target carrier envelope phase measurements of a few cycle laser pulses [18]. Since only nJ pulse energies, a fraction of typical fundamental laser pulse energies, are needed, this approach offers the potential for minimally invasive shot-by-shot phase measurement.

Acknowledgments

The authors acknowledge financial support from NSERC and from the Ontario Centers of Excellence as well as valuable discussions with G Yudin, M Y Ivanov, P P Rajeev, D Grojo and S Golin.

References

- [1] Corkum P B and Krausz F 2007 Attosecond science *Nature Phys.* **3** 381
- [2] Keldysh L V 1965 Ionization in the field of a strong electromagnetic wave *Sov. Phys. JETP* **20** 1307
- [3] Boutu W *et al* 2008 Coherent control of attosecond emission from aligned molecules *Nature Phys.* **4** 545
- [4] Gertsvolf M, Jean-Ruel H, Rajeev P P, Klug D, Rayner D M and Corkum P B 2008 Orientation-dependent multiphoton ionization in wide band gap crystals *Phys. Rev. Lett.* **101** 243001
- [5] Smirnova O, Mairesse Y, Patchkovskii S, Dudovich N, Villeneuve D M, Corkum P B and Ivanov M Yu 2009 High harmonic interferometry of multi-electron dynamics in molecules *Nature* **460** 972
- [6] Siedeman T, Ivanov M Yu and Corkum P B 1995 Role of electron localization in intense-field molecular ionization *Phys. Rev. Lett.* **75** 2819–22
- [7] Corkum P B 1993 Plasma perspective on strong-field multiphoton ionization *Phys. Rev. Lett.* **71** 1994–7
- [8] Couairon A, Sudrie L, Franco M, Prade B and Mysyrowicz A 2005 Filamentation and damage in fused silica induced by tightly focused femtosecond laser pulses *Phys. Rev. B* **71** 125435
- [9] Winkler S W, Burakov I M, Stoian R, Bulgakova N M, Husakou A, Mermillod-Blondin A, Rosenfeld A, Ashkenasi D and Hertel I V 2006 Transient response of dielectric materials exposed to ultrafast laser radiation *Appl. Phys. A* **84** 413
- [10] Rajeev P P, Gertsvolf M, Corkum P B and Rayner D M 2009 Field dependent avalanche ionization rates in dielectrics *Phys. Rev. Lett.* **102** 08300
- [11] Boyd R W 1992 *Nonlinear Optics* (New York: Academic)
- [12] Rayner D M, Naumov A and Corkum P B 2005 Ultrashort pulse non-linear optical absorption in transparent media *Opt. Exp.* **13** 3208–17
- [13] Stuart B C, Feit M D, Rubenchik A M, Shore B W and Perry M D 1995 Laser-induced damage in dielectrics with nanosecond to subpicosecond pulses *Phys. Rev. Lett.* **74** 2248–51
- [14] Manolopoulos D E 2002 *J. Chem. Phys.* **117** 9552
- [15] Dudovich N, Levesque J, Smirnova O, Zeidler D, Comtois D, Ivanov M Yu, Villeneuve D M and Corkum P B 2006 Attosecond temporal gating with elliptically polarized light *Phys. Rev. Lett.* **97** 253903
- [16] Dudovich N, Smirnova O, Levesque J, Mairesse Y, Ivanov M Yu, Villeneuve D M and Corkum P B 2006 Measuring and controlling the birth of attosecond xuv pulses *Nature Phys.* **2** 781
- [17] Shafir D, Mairesse Y, Villeneuve D M, Corkum P B and Dudovich N 2009 Atomic wavefunctions probed through strong-field light-matter interaction *Nature Phys.* **5** 412
- [18] Paulus G G, Lindner F, Walther H, Baltuška A, Goulielmakis E, Lezius M and Krausz F 2003 Measurement of the phase of few-cycle laser pulses *Phys. Rev. Lett.* **91** 253004