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Reprint of: The role of delay times in subcycle-resolved probe retardation measurements

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HIGHLIGHTS

- The retardation of a probe pulse by a strong pump pulse is investigated.
- Subcycle resolution in the pulse delay is achieved for low crossing angles and small spot-sizes.
- The measurement is sensitive to a delay in the electronic Kerr response and/or ionization.

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ABSTRACT

The delay in the nonlinear response of matter to intense laser pulses has been studied since a long time regarding its nuclear contribution. In contrast, the electronic part of the nonlinear response in wide-band-gap dielectrics, which is usually dominant, is not well explored regarding its delay, and previous studies have revealed that the timescale is below 1 fs. Here, the influence of delay times on the recently introduced method of subcycle-resolved probe retardation measurements is investigated using a simulation. In the model assumed, the electronic nonlinearity is divided into the third order Kerr effect and the plasma contribution due to conduction band population in the strong laser field. In the regime of close-to-collinear pump-probe geometries, the probe retardation shows both π- and 2π-oscillations in the pump-probe delay. Sub-femtosecond delay times influence the phase of the oscillations significantly, but it remains difficult to distinguish the influence of the Kerr response from the plasma contribution.

1. Introduction

The use of delay times in the description of quantum mechanical processes, such as for example tunneling, has become a useful concept in the area of strong-field and attosecond physics [1]. It is not an indispensable concept for the description of the processes, and often the delay times have no clear definition in the form of quantum mechanical observables. This leads usually to a lack of a clear prescription how the delay times should experimentally be measured. Also, the expressions *delay time*, *tunneling time*, *traversal time* etc. are used in manifold ways in the literature [2].

One approach, stimulated by a classical view, is to trace some characteristic feature of a wave packet, often its peak, as it transverses a barrier. This approach is not very rigorous because, as Büttiker and Landauer pointed out [3], an incoming peak does not, in any causative sense, turn into an outgoing peak. A more versatile approach is to introduce a clock, established by some degree of freedom of the process. For example, the spin precession of a tunneling particle can be measured when a magnetic field is confined to the barrier [4–6]. Another way to establish a clock in a tunneling process is to make the barrier oscillate in time. The Büttiker-Landauer traversal time for tunneling [3] is defined such that particles with a traversal time shorter than the period time of the barrier do not pick up energy quanta from the barrier and pass the barrier adiabatically, whereas particles with a traversal time longer than the period time of the barrier see the barrier long enough to exchange energy quanta with the barrier. The situation of an oscillating barrier is naturally encountered in the length-gauge when the potential of a strong laser pulse deforms the Coulomb potential of an atom and a bound electron is separated from the continuum by a barrier that oscillates with the laser frequency. Keldysh analyzed this situation already in 1964 and introduced the Keldysh parameter γ [7], defined by

$$\gamma = \frac{\omega_0\sqrt{2E_B}}{E_0}, \tag{1}$$

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where ω_0 and E_0 are the angular frequency and the amplitude of the laser field (atomic units are used throughout the paper) and E_B is the binding energy. The regime $\gamma > 1$ designates the regime of multiphoton ionization and $\gamma < 1$ designates the regime of adiabatic tunneling. The interpretation that the interaction time must be long compared to a laser cycle for multiphoton ionization (so multiple photons can be absorbed) and short compared to the laser cycle for adiabatic tunneling (so there is no time for energy exchange) leads to an expression for a traversal time

$$\Delta t_T = \gamma/\omega_0, \tag{2}$$

which formally agrees with the Büttiker-Landauer traversal time for tunneling as explained by Yücel and Andrei [8]. Care must be taken when the Büttiker-Landauer traversal time is used in simplified models. It was pointed out later by Landauer and Martin [2] that "the expression *traversal time*, introduced by Büttiker and Landauer, may not have been an optimum choice; it is too easily interpreted as a time delay in transmission, rather than an interaction time." Therefore, when used in simplified models, the Büttiker-Landauer traversal time should not be used as a delay in transmission. Instead, the most direct implication of the Büttiker-Landauer traversal time concerns the transmission rate as a function of barrier frequency, such as the transition from multiphoton ionization to tunneling.

In cases when the actual delay should be accessed, beyond the concept of spin precession times or interaction times, it is often necessary to define a delay time on the basis of a measurement prescription. Eckle et al. [9] have used close-to-elliptically polarized few-cycle laser pulses to deflect the electrons after tunneling ionization. The angle in between the maximum laser field and the maximum ionization current was used to define a tunneling delay time and subsequently measure it. The result for strong field ionization of helium for $1.17 < \gamma < 1.45$ was zero delay with an upper limit of 34 as, given by the uncertainty of the experiment. The existence of a significant delay time, which would cause the ionization current to lack behind the electric field, would have far-reaching consequences. Much of the interpretation in strong-field physics relies on a semi-classical model, where electrons appear in the continuum following an ionization rate that is assumed to follow the laser field instantaneously.

Here, in this special issue paper in memory of Markus Büttiker, the role of potential delay times in strong-field driven transparent solids is investigated. The delay times are introduced in a simplified model that is used to reproduce the data of a measurement. Specifically, the influence of such delay times is investigated on the recently introduced experimental method to measure the retardation of a probe pulse in a strong-field pumped sample with subcycle resolution [10]. The method is all optical and therefore applicable not only for gas-phase samples, but also for transparent solid samples, where the strong-field effects are not yet thoroughly explored. First, the influence of a delay in the electronic nonlinearity of the polarization response is investigated. Second, the influence of a delay in the ionization (i.e. promotion of electrons from the valence band to the conduction band) is investigated. This is especially interesting because it was found recently that ionization in a dielectric solid does not follow the envelope of the laser pulse in the strong-field regime, but shows fast dynamics within the cycles of the optical field [11–14]. In contrast to atoms, where the ionization yield is usually assumed to increase monotonically in time except for small dips around the field strength minima [15,16], the ionization yield in dielectric solids is believed to significantly decrease around field strength minima [12,14].

2. Theory and calculation

A non-collinear pump-probe geometry is considered where the nonlinear dipole response of a sample (in this case a thin dielectric sample) to a strong pump pulse is read out by a weak probe pulse. The basic influence of the pump on the probe in a nonlinear medium was already described in 1966 as "weak-wave retardation" [17]. Whereas this phenomenon is usually described using the envelopes of the pulses, the phenomenon is studied here regarding its subcycle dependence. That means that features of the probe pulse are analyzed that depend on the pump-probe delay on a timescale shorter than the optical period time of the pulses (in this example shorter than 2.7 fs). As explained below, a time resolution better than the optical cycle can only be achieved for very small crossing angles between pump and probe. The limiting case of a collinear geometry would require non-degenerate polarizations and/or wavelengths in order to separate pump and probe after the interaction, but this is experimentally challenging in the strong-field regime [10]. Here, the pump and probe beams are close-to-collinear, and the pump is spatially separated after sufficient free-space propagation following the interaction in the sample. The remaining probe pulse is refocused using a thin lens and the temporal retardation is analyzed. The beam path, visualized in Fig. 1, is similar but not identical to a recent experiment [10].

The pump and probe pulses are initialized as Gaussian pulses with linear polarization in x-direction, such that the electric field in x-direction of each pulse is initially given by

$$E_{\text{pump,probe}}(t, \rho, \zeta = 0) = E_0 \exp\left(-\frac{\rho^2}{w_0^2}\right)\exp\left(-\frac{t^2}{2\sigma}\right)\cos(\omega_0 t). \tag{3}$$

A 5-fs envelope of the intensity and a central wavelength of 800 nm are assumed for both pulses ($\sigma = 124$ au, $\omega_0 = 0.057$ au). The peak intensity is 5×10^{10} W/cm^2 ($E_0 = 0.0012$ au) for the probe pulse and 10^{13} W/cm^2 ($E_0 = 0.0169$ au) for the pump pulse. For the probe pulse, the transversal coordinate ρ coincides with the y-axis and the propagation coordinate ζ coincides with the z-axis. For the pump pulse, the field is rotated by an angle of 0.3° around the x-axis and a pump probe delay τ is introduced through $t \rightarrow t - \tau$. The

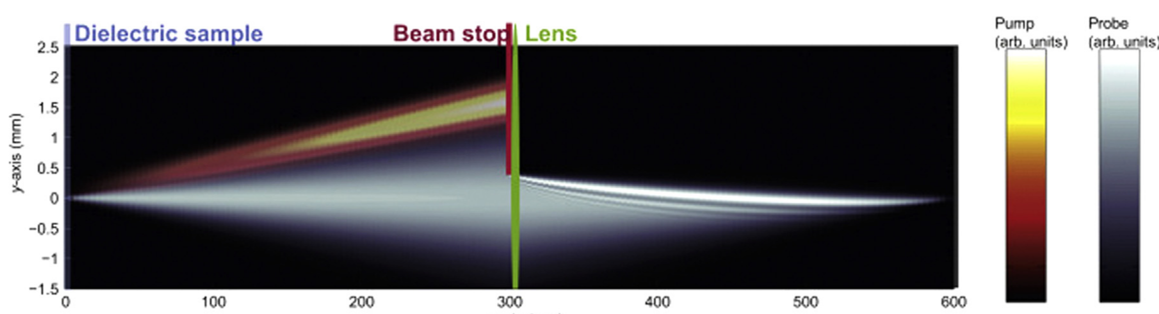

Fig. 1. The beam paths of the pump and the probe fields are visualized following the respective color bars. A pump pulse with a waist of 300 μm interacts with a probe pulse with a waist of 50 μm in a 5-μm-thick dielectric sample. After 0.3 m of free-space propagation, the pump pulse is blocked. A thin lens refocuses the remaining probe pulse. To enhance visibility throughout the beam path, the color scales do not have a direct relation to the intensities, but are rescaled at each position along the z-axis.