Quantum interference in attosecond transient absorption of laser-dressed helium atoms

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Attosecond transient absorption (ATA) studies provide a way to push our understanding of the energy transfer between electromagnetic fields and matter to the sub-femtosecond time scale [1, 2]. They are an all-optical attosecond metrology that complements methods based on the measurement of charged particles, such as attosecond streaking [3, 4] and electron interferometry [5, 6]. Like those methods, high time-resolution is gained in an ATA spectrometer by using attosecond extreme ultraviolet (XUV) pulses that are synchronized to the field oscillations of an infrared (IR) laser pulse. The first ATA experiments used an attosecond pulse to probe a valence electron wave packet (EWP) created by ionizing an atom with a strong IR laser pulse [1, 2]. Recent ATA experiments have used attosecond pulses as a pump that creates an EWP which is then probed by a moderately strong IR field. In this XUV-pump/IR-probe configuration the frequency-resolved transient absorption signal varies as a function of the IR intensity, duration, and the sub-cycle timing between the two fields [3, 10]. These experiments raise the possibility of studying time-dependent absorption down to the attosecond time scale.

In this theoretical study, similar to recent experiments [10, 11], we consider helium atoms that are excited by an isolated attosecond pulse (IAP) with a central frequency near the ionization threshold, together with a delayed few-cycle IR pulse. We elucidate the key features of the resulting delay-dependent attosecond absorption (DDAA) spectra. These features derive from the fact that the IAP “starts the clock” by exciting the system at a well-defined time, and that it makes an EWP that is essentially independent of the pump-probe delay. The DDAA measurement is a spectrogram that records the interference between different excitation pathways which lead to the same absorption or emission processes. Because the attosecond pulse is locked to the IR field oscillations, it is suggestive that many of the features in the spectrum are modulated at half the IR laser period \(T_{\text{IR}}\), about 1.3 fs. However, the absorption spectrum results from the time-integrated response of everything that happens after the initial XUV excitation, which complicates the analysis. Our main concern in this paper is the connection between these oscillations and the real-time attosecond dynamics.

We discuss two distinct manifestations of attosecond dynamics in the DDAA spectrum. First, near the ionization threshold the delay-dependent absorption exhibits fully modulated interference fringes at half the IR period. We show these are due to the interference between two pathways, separated in time, which give rise to the same dipole response. This “which way” quantum path interference [12] can be used to time-resolve two photon transitions between excited states, and shows how the absorption at XUV frequencies is altered on a sub-cycle time scale. We find \(T_{\text{IR}}/2\) oscillations with a similar origin in absorption features associated with non-linear XUV+IR processes, which appear as light-induced structures in the ATA spectrum. These fringes illustrate how energy can be exchanged between excitation modes (dressed states) that exist only when the XUV and IR fields overlap in time. The second feature results from the IR driven sub-cycle AC Stark shift of the lower-lying 2p and 3p resonances [11]. The time-dependent Stark shift leads to a dispersive line-shape, which results from interference between the time-dependent dipole induced before, during, and after the IR pulse. We show that, due to the integrated nature of the absorption spectrum, a measurement of the instantaneous energy shift is only possible when ionization limits the interaction time.

The essential features of DDAA spectra can be understood at the single atom level. The energy lost or gained by a light field in the interaction with an atom can be described by a frequency-dependent response function \(\tilde{S}(\omega, t_d) = 2 \text{Im}[d(\omega)\tilde{E}^*(\omega)]\), where \(t_d\) is the pump-probe delay (we use atomic units unless otherwise indicated). \(d(\omega)\) and \(\tilde{E}(\omega)\) are the Fourier transforms of the time-dependent dipole moment \(d(t)\) and the total driving field \(E(t)\). The dipole moment is obtained by solving the time-dependent Schrödinger equation (TDSE) in the
For large positive delays, \( t_d \) strongly modifies the dipole, which induces sidebands on pulse, at one half the laser period. Of particular interest are pulse undergoes free decay until the \( s \) from the ground state to non-dipole coupled (“dark”) \( ir \) and \( xuv \) induced structures (LISs) appear, for example, between the ground state and a group of \( np \) states simultaneously in a manner that is independent of the delay. The indirect pathway is a 3rd-order process in which amplitude in the 2\( p \) state is transferred to an \( np \) state by a two-\( ir \)- photon process. The role of the indirect pathway via the 2\( p \) state is confirmed by a test calculation which dynamically eliminates the 2\( p \) state during the time-propagation of the TDSE, in which these fringes disappear. The two interfering processes, since they are driven by the \( xuv \) field and the \( ir \) field respectively, happen at different times depending on the value of \( t_d \). The resulting interference fringes are analogous to those observed in photoelectron spectra \([6]\), however, in DDAA the multiphoton process need not result in ionization.

Having identified the interfering pathways it is straightforward to write down the constructive interference condition as a function of delay:

\[
(\omega_{np} - \omega_{2p})|t_d - t_0| + \Delta \phi = 2\pi k,
\]

where \( \omega_{np} \) and \( \omega_{2p} \) are the energies of the \( np \) and 2\( p \) states respectively, and \( k \) is a positive integer \([7]\). Two additional parameters enter: \( \Delta \phi \) is a phase due to the two-\( ir \)-photon transition and \( t_0 \) is the time when the two photon transition probability peaks. It is reasonable to choose \( t_0 = -T_{IR}/4 \) due to the sine-like \( ir \) carrier wave used in the calculation. This formula correctly predicts a decreasing slope for the fringes as \( |t_d - t_0| \) increases, corresponding to increasing \( k \) in Eq. (1). The fit to the interference fringes using \( \Delta \phi = 0 \), one set of integers \( k \) that start with \( k = 1 \) at \( t_0 \), and no other free parameters, is excellent for \( t_d < 0 \), as shown in Fig. (2)(b). For \( t_d > 0 \) the biggest contribution to the indirect pathway comes from whatever \( ir \) field maximum follows the XUV pulse (never more than one half cycle away). The slope of the fringes should therefore saturate for \( t_d > 0 \), which is indeed what we observe in Fig. 1. We can thus conclude that the strength of the two photon transition peaks at the local intensity maxima of the \( ir \) field. This supports a time-dependent picture of multi-photon absorption below threshold as a process that follows the sub-cycle oscillations of the \( ir \) electric field.

One application of this 1st vs. 3rd order interference process is illustrated in Fig. (2)(a). Realizing that the indirect pathway is enhanced due to a near one-\( ir \)-photon resonance with the intermediate 3\( s \) state, it seems possible that we can use this to determine \( \Delta \phi \) in Eq. (1). For an 800 nm pulse we can safely assume that \( \Delta \phi = 0 \) because the intermediate 3\( s \) state lies above the energy reached by absorbing one \( ir \) photon from the 2\( p \) state.

FIG. 1: Single atom response function \( \tilde{S}(\omega, t_d) \) in helium where \( t_d \) is the time delay in \( ir \) cycles between the \( ir \) laser pulse (800 nm, \( 3 \times 10^{12} \) W/cm\(^2\), 4 cycles, \( \cos^2 \) envelope, sine-like carrier envelope) and the attosecond pulse (330 as, centered at 25 eV). The \( ir \) intensity oscillations are shown in black in the top panel.

The single-active-electron approximation \([13]\).

Fig. 1 shows a typical response function vs pump-probe delay. The 25 eV, 330 as \( iap \) pump pulse has a bandwidth of 5.5 eV, which means that it overlaps all the singly excited and low-energy continuum states of the He atom. The FWHM of the 800 nm probe pulse is 11 fs, corresponding to 4 optical cycles \((T_{IR} = 2.7 \) fs\). The \( ir \) field is of moderate intensity, so that it can not itself excite the atom in its ground state. The pump and probe fields have parallel linear polarizations. We include a dipole dephasing time \( T_2 = 65 \) fs in the calculations by smoothly windowing the dipole moment. We have verified that using a longer \( T_2 \) does not change any of our conclusions. Positive (negative) values of \( S(\omega, t_d) \) mean that a dilute gas will absorb (emit) energy at frequency \( \omega \) as the dipole-driven source term in the wave equation will be out of (in) phase with \( \tilde{E}(\omega) \) \([13,14]\).

We begin by briefly describing the main features in Fig. 1. For large positive delays, \( t_d \geq 4T_{IR} \), the \( iap \) arrives after the end of \( ir \) pulse and the absorption spectrum exhibits only one-XUV-photon transitions from the ground state to the \( np \) states. When the two pulses overlap, \(-4T_{IR} < t_d < 4T_{IR} \), these absorption features are strongly modified. In this same delay range, light-induced structures (LISs) appear, for example, between the 2\( p \) and 3\( p \) resonant lines. These are associated with two-photon (XUV±\( ir \)) processes that transfer population from the ground state to non-dipole coupled (“dark”) \( s \) and \( d \) states, and have recently been observed experimentally \([11]\). Finally, when the \( iap \) arrives before the \( ir \) pulse, \( t_d \leq -4T_{IR} \), the dipole established by the XUV pulse undergoes free decay until the \( ir \) pulse arrives and strongly modified the dipole, which induces sidebands on the main resonance features.

Many of the features in \( S(\omega, t_d) \) show a modulation at one half the laser period. Of particular interest are fringes that are present both when the pulses overlap and when the \( iap \) arrives before the \( ir \). An example is seen in Fig. 1 between 24.2 and 24.8 eV near the label “2\( p \) + 2\( \omega \)”, also shown in more detail in Fig. (2)(b). These fringes are caused by quantum interference between two distinct pathways for establishing the same coherence between the ground state and a group of \( np \) states \((n > 5)\), near threshold. The process is diagrammed in Fig. (2)(a). The direct pathway is an XUV-driven 1st-order process that populates all of the \( np \) states simultaneously in a manner that is independent of the delay. The indirect pathway is a 3rd-order process in which amplitude in the 2\( p \) state is transferred to an \( np \) state by a two-\( ir \)- photon process. The role of the indirect pathway via the 2\( p \) state is confirmed by a test calculation which dynamically eliminates the 2\( p \) state during the time-propagation of the TDSE, in which these fringes disappear. The two interfering processes, since they are driven by the XUV field and the \( ir \) field respectively, happen at different times depending on the value of \( t_d \). The resulting interference fringes are analogous to those observed in photoelectron spectra \([6]\), however, in DDAA the multiphoton process need not result in ionization.
predicts that the fringes of the 3s state are exactly out of phase with the 2p state. LISs associated with coupling of the 3s state and nearby dark states is in a regime where the rotating wave approximation breaks down, and that the short duration of the IAP yields a well-defined phase of the IR field at excitation. In such situations it has been demonstrated that the final state populations are sensitive to the phase of the laser at the time of excitation [16]. Here we have shown that the DDAA spectrum shows the same sensitivity.

Another source of fast dynamics in the transient absorption spectrum is the sub-cycle AC Stark shift of the bound state energies [17, 18]. Figs. 3(a) and (b) show the evolution of the 2p and 3p line shapes with delay, which include both emission and absorption. Qualitatively, this results from the redistribution of XUV energy across the resonances due to the perturbation of the dipole by the IR field. Similar dispersive line shapes have been discussed previously in connection with the control of exciton polarizations of a quantum dot using picosecond laser pulses [17, 18].

In a time-domain picture, the dispersive shape of the np absorption lines is caused by multiple contributions to the dipole moment: (i) a perturbed response during the IR pulse, and (ii) free decay of the coherence after the IR pulse ends [19, 20]. The relative importance of these terms is determined by the IR field intensity and delay, as well as $T_d$. If the state amplitude is unchanged by the IR field then the main contribution to the dipole moment comes from the free decay which, however, acquires a phase shift, $\theta_S$, equal to the integrated optical Stark shift during the time when the IR pulse acts. In this approximation the response is given by

$$\tilde{S}(\omega, t_d) \approx L(\omega, T_d) [\cos(\theta_S) + (\Delta \cdot T_d) \sin(\theta_S)]$$  \hspace{1cm} (2)

where $\Delta$ is the difference $\omega - \omega_{np}$ and $L(\omega, T_d)$ is the Lorentzian line shape in the absence of the IR field. For positive delays $1 < t_d < 4T_d$ we find that the line shape is well described by this simple form. We see that the shift of the absorption is in the direction of the Stark shift, but the magnitude of the shift is proportional to $\theta_S/T_d$ and is therefore constrained by the natural linewidth. Thus, for these delays, the 2p and 3p lines have Stark shifts of opposite sign, as shown in Fig. 3(c), but the magnitude of these shifts does not generally equal the instantaneous Stark shift.

At delays near zero ($-4T_d < t_d < 1T_d$) the line shape becomes much wider than $1/T_d$ and more complex than the simple dispersive shape discussed above. Here depopulation of the excited states by the IR field plays a larger role (see Fig. 3(d-e)), and the response is dominated by the perturbed dipole decay. For the 3p state the loss is predominantly by one photon ionization to the continuum and the dipole response exists for only a few laser cycles after it is excited. This rapid ionization-induced dephasing allows us to probe the real-time subcycle dynamics using a delay-dependent method: during

\[ \Delta \]
the dipole established by the IAP oscillates freely until the IR pulse strongly perturbs it, greatly altering the dipole amplitude and phase in a few IR cycles. This perturbed free polarization decay has been observed previously at optical frequencies \[13, 21\]. It yields sidebands, seen clearly above the 3p and below the 2p lines, with a characteristic hyperbolic shape that depends on the delay as \(1/t_d\) and shows no attosecond time scale dynamics.

To conclude, we have identified two configurations where attosecond dynamics can be observed in attosecond transient absorption spectra: which-way interference and the sub-cycle AC Stark shift. Since experimental observations always result from propagation in a macroscopic gas, we also solve the Maxwell wave equation (MWE) for the time propagation of light fields through the atomic helium gas medium, in which the polarization and ionization source terms are obtained by solving the single-atom TDSE \[13\]. The absorption/emission probability after propagation of a dilute gas is almost identical to the single atom result in Fig. 1, exhibiting both the half-cycle modulations, the LISs, and the emission features discussed above. This indicates that our DDAA predictions can be observed using current attosecond technology, especially as it regards half-IR-cycle oscillations.

We have demonstrated that interference fringes in DDAA spectra result from the coherent addition of two quantum paths that lead to the same dipole excitation. They reveal the time delay between the initial excitation and a later IR-field driven multiphoton transition. This was found to be true both for transitions between bound states as well as between excitation modes (the LISs) that are observable only when the pulses overlap. We expect therefore that they will be a general feature in more complex systems \[22\], and could be observed between resonant states embedded in a continuum, as long as the lifetimes are longer than the IR period. Using this argument in reverse, the interference fringes visible in the DDAA spectrum could be used as a precise timing device, a probe of decoherence, or a phase meter when an intermediate resonance is involved.

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