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Multiphoton Resonance in Attosecond Transient Absorption

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Abstract: We present the theory and simulation of attosecond transient absorption in helium atoms under the single-active-electron approximation. This study investigates the attosecond dynamics of intrinsic atomic states that interact with a field comprising vacuum ultraviolet (VUV) and extreme ultraviolet (XUV) fields. The absorption spectrum of the helium atom is obtained from the response function, which is constructed by numerically solving the three-dimensional time-dependent Schrödinger equation. We observe a fine structure near the intrinsic atomic level, which is modulated with a 0.2 fs period. Based on high-order time-dependent perturbation theory, the frequency-dependent phase of the dipole response induced by the VUV and XUV fields is analytically obtained, and the fine structure is well explained by the phase difference. In addition, the absorption fringes are dependent on the chirp of the VUV field. This study investigates the features of the attosecond transient absorption in the VUV region, which may have valuable applications in the study of ultrafast phenomena in atoms, molecules, and solids.

Keywords: attosecond pulse; attosecond transient absorption; high order harmonics



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1. Introduction

Attosecond transient absorption spectroscopy is a newly developed method that can probe the ultrafast dynamics of electrons in atoms [1–14], molecules [15–19], or solids [20–27]. This spectroscopy uses the pump-probe scheme, where an infrared (IR) or extreme ultraviolet (XUV) field is used to probe or pump the attosecond dynamics. The transmitted spectrum of the XUV field is recorded while scanning the time delay between the IR and XUV fields.

The first attosecond transient absorption experiment was performed using an IR pump field that was strong enough to initiate dynamics by directly ionizing krypton atoms [1]. The valence electron dynamics were measured by monitoring the absorption spectrum of the XUV field. Subsequently, several studies have been experimentally and/or theoretically performed based on the scenario in which the XUV field (pump beam) initiates the dynamics and the IR field probes the dynamics by coupling to nearby dark or continuum states. Several new phenomena, such as the light-induced-state [28–34], sub-cycle AC Stark shift [28], Autler–Townes splitting [35], hyperbolic sidebands [29,32,33,36], and quantum beats [8,37] have been experimentally observed and theoretically investigated. Recent studies used probe fields in the IR range, and the dynamics were modulated with a half- or quarter-cycle period of an 800 nm IR field. However, the couplings from the excited states to the ground state have been ignored, which may build faster dynamics of electrons that cannot be probed by such an IR field.

Several methods, such as the two-level model [29,31–33] and adiabatic three-level model [38], have been used extensively to study the attosecond dynamics of atoms dressed by IR field. An N-surface model [15], based on the first-order time-dependent perturbation theory, is applied to include the effect of nuclei motion on the attosecond transient

absorption spectrograms (ATAS). However, these models are intended for calculating the dynamics initiated by an XUV field then disturbed by an IR field. For the XUV+VUV circumstances, the VUV field can only weakly affect the dynamic process induced by the XUV field (due to the larger detuning of the VUV field in the transitions between the bright states and nearby dark states), but it can strongly excite electrons from the ground state to the excited state through multi-photon absorption. The models mentioned previously are no longer suitable for describing this process; therefore, a high-order time-dependent perturbation theory is used to conveniently calculate the VUV absorption process in ATAS.

In this study, we theoretically and numerically investigate the ATAS of a helium atom as it interacts with a combined field composed of a VUV and an XUV field. The ATAS is obtained by calculating the response function that is constructed using the wave function solution of the three-dimensional (3D) time-dependent Schrödinger equation (3D-TDSE) under the single-active-electron (SAE) approximation [39,40]. All dynamic processes are included in the 3D-TDSE; therefore, we introduce the time-dependent perturbation theory to capture the essential process. From the ATAS of the helium atom, the absorption probability is modulated with a period of 0.2 fs, and a fine structure near the intrinsic atomic level is also observed. The frequency-resolved phase of the dipole response induced by the VUV and XUV fields was analytically obtained based on the time-dependent perturbation theory, which successfully explained the emergence of fine structures. We believe that time-dependent perturbation theory can be expanded to include the effects of re-absorption and emission in the attosecond absorption process, and the results of this study extend the research scope of attosecond physics. In addition, the relatively large (compared with the IR field) photon energy of the VUV field produced unique absorption features in the ATAS, which cannot be observed in the ATAS driven by the IR field. Furthermore, the chirp-dependent absorption fringes suggest a new method to characterize the VUV field.

2. Theoretical Model

2.1. 3D-TDSE

Numerically solving the fully 3D-TDSE has been widely used to investigate the ultrafast dynamics of electrons and nuclei in atoms and molecules. The ATAS of helium atoms was obtained under the SAE approximation by constructing the response function [41] $S(\omega, t_d) = -2\text{Im}[d(\omega, t_d)\varepsilon^*(\omega, t_d)]$, where t_d is the time delay between the IR (VUV) and XUV fields, $\varepsilon(\omega, t_d)$ is the Fourier spectrum of the total electric field of the IR (VUV) and XUV fields, and $d(\omega, t_d)$ is the frequency-dependent dipole response of the helium atom, which was constructed by $d(\omega, t_d) = \tilde{F}[d(t, t_d)]$, $d(t, t_d) = \langle \Psi(t) | r | \Psi(t) \rangle$. $\tilde{F}$ represents Fourier transformation, and the wave function $|\Psi(t)\rangle$ was obtained by numerically solving the 3D-TDSE under the SAE approximation. Briefly, the time-dependent wave function is first expanded into a series of spherical harmonics. As a result, the 3D-TDSE is reduced to a set of coupled equations between the different angular quantum numbers for the radial wave function, which is further discretized by the finite-element discrete variable representation (FE-DVR) method with the advantage of providing block-diagonal sparse matrix representation of kinetic operator and the diagonal matrix representation of the effective Coulomb potential. The temporal evolution of the wave function is carried out by the Arnoldi–Lanczos algorithm. A more detailed description of the 3D-TDSE solution can be found in reference [42]. We have used this 3D-TDSE to investigate the attosecond chirp effect [43] and wavelength effect [44] on the ATAS of the helium atom. The response function $S(\omega, t_d)$ was derived from the energy exchange between the helium atom and the total electric field; thus, the positive (negative) values represent the absorption (emission) at frequency ω . The pulses used in the 3D-TDSE simulation have a chirped formula, and the vector potential is expressed as follows:

$$A_j(t - t_d) = \text{Re} \left\{ -i \frac{1}{\omega_j} \sqrt{\frac{I_j}{1 - i\zeta_j}} \exp \left\{ -i \left[\omega_j(t - t_d) - 2 \ln 2 \frac{(t - t_d)^2}{\tau_j^2(1 - i\zeta_j)} \right] \right\} \right\}, \quad (1)$$

where $j = \text{IR, VUV, or XUV}$. The total electric field is expressed as $E(t) = -\partial A(t)/\partial t$, $A(t) = A_{\text{XUV}}(t - t_d) + A_{\text{IR(VUV)}}(t)$. t_d is the time delay between the XUV and IR (VUV) pulses, and ξ_j is a dimensionless parameter for the chirp. I_j , ω_j , and τ_j are the intensity, angular frequency, and time-domain duration of the corresponding electric field, respectively. In the numerical simulation, $I_{\text{XUV}} = 1 \times 10^{11} \text{ W/cm}^2$, $\omega_{\text{XUV}} = 21 \text{ eV}$, and $\tau_{\text{XUV}} = 300 \text{ as}$. For the XUV+IR case, the IR field used in the simulation has the following parameters: $I_{\text{IR}} = 1 \times 10^{13} \text{ W/cm}^2$, $\lambda_{\text{IR}} = 2\pi c/\omega_{\text{IR}} = 800 \text{ nm}$, and $\tau_{\text{IR}} = 5 \text{ fs}$; c is the speed of light in a vacuum. We chose parameters of the IR pulse for better absorption signals. A weaker intensity (such as 10^{11} W/cm^2) may lead to weak modulation of the absorption spectrum, whereas a stronger intensity (such as 10^{14} W/cm^2) may blur the absorption signal due to ionization. For the XUV+VUV case, the VUV field has the following parameters: $I_{\text{VUV}} = 3 \times 10^{13} \text{ W/cm}^2$, $\omega_{\text{VUV}} = 7 \text{ eV}$, and $\tau_{\text{VUV}} = 3 \text{ fs}$. For weaker VUV intensity, the transition from ground state to excited state may be too weak to be observed, whereas for stronger intensity, the ionization can no longer be ignored. Calculated through the ADK theory [45,46], the tunnel ionization probability of a helium atom illuminated by such VUV field is in the order of 10^{-11} , which can be ignored safely.

Under the SAE approximation, the helium atom has an ionization potential of 24.58 eV, and the first dipole-allowed excited state is 1s2p with an energy level of 21.056 eV. Thus, the XUV pulse could resonantly excite the electrons from the ground state 1s² to the excited state 1s2p with a single photon. The VUV field could pump the ground electron to the excited 1s2p state via three-photon resonance because parity is conserved in the excitation process.

2.2. Time-Dependent Perturbation Theory

The motion of a quantum system can be described by the Schrödinger equation: $H\Psi(r, t) = i\hbar\partial\Psi(r, t)/\partial t$, where $H(t)$ is the Hamiltonian, which is typically written as the sum of a time-independent H_0 and a time-dependent perturbation, $H(t) = H_0 + V(t)$. If the interaction is switched on at $t = 0$, the transition amplitude according to 1st-order perturbation theory is:

$$c_b^{(1)}(t) = -\frac{i}{\hbar} \int_0^t U_{ba}(t') \exp(i\omega_{ba}t') dt', \quad (2)$$

where $U_{ba}(t) = \langle \Psi_b(t) | V(t) | \Psi_a(t) \rangle$ is the time-dependent perturbation matrix element, $c_b^{(1)}(t)$ is the probability amplitude of the final state $|\Psi_b(t)\rangle$, and $\omega_{ba} = (W_b - W_a)/\hbar$ is the transition frequency. The system is assumed to be at initial state $|a\rangle$. We may iterate the procedure to obtain the 2nd solution, and so on. We are only interested in the evolution of the probability amplitude after the end of the external electric field, that is, for $t \gg 1/\omega_{ba}$. For the 3rd-order perturbation, the probability amplitude is:

$$c_b^{(3)}(t) = \frac{(-i)^3}{\hbar^3} \sum_{\zeta, \nu} \int_0^t \int_0^{t'} \int_0^{t''} U_{\zeta a}(t''') e^{i\omega_{\zeta a}t'''} dt''' U_{\nu \zeta}(t'') e^{i\omega_{\nu \zeta}t''} dt'' U_{b\nu}(t') e^{i\omega_{b\nu}t'} dt', \quad (3)$$

where $U_{\zeta a}$, $U_{\nu \zeta}$, $U_{b\nu}$ and $\omega_{\zeta a}$, $\omega_{\nu \zeta}$, $\omega_{b\nu}$ have meanings similar to those of U_{ba} and ω_{ba} in Equation (2), respectively. ζ and ν are the immediate states. The time-dependent perturbation theory described here is valid under the conditions of weak couplings between related states and low external electric field intensity, which is satisfied by the simulation parameters used in this study.

3. Results and Discussion

Figure 1 shows the ATAS of helium atoms, which was calculated using the response function obtained with the laser parameters specified in Section 2.1 for two cases: (a) XUV+IR and (b) XUV+VUV. Figure 1c is the level scheme of the relevant states in a helium atom. Figure 1a shows the typical ATAS of a helium atom under the SAE approximation, which has been extensively studied. The half-cycle modulation of the

absorption probability, the Aulter-Townes splitting of the $1s2p$ state, and the light-induced virtual states ($LISs$, $2s^-$, $2s^+$) can be accurately reproduced by the 3D-TDSE model. The notation used for the $LISs$ follows that of reference [36]. For example, $2s^-$ indicates that the LIS has a one-IR-photon separation from the $1s2s$ state. In Figure 1b, the ATAS is significantly different from the XUV+IR case in Figure 1a; the absorption only occurs near the $1snp$ excited states, and the modulation period along the time delay is not a half-cycle of the VUV field, but the oscillation period of the $1s2p$ state, i.e., 0.2 fs. $1snp$ indicating that one of the two electrons in the helium atom is in the ground state $1s$ while the other is the excited state np , $n \geq 2$. In addition, near the $1s2p$ state, the absorption fringes are inclined for photon energies in the range of $W \approx W_{1s2p}$. At a specific time delay, the response function is negative; hence, the XUV spectrum is emitted. For $W > W_{1s2p}$ or $W < W_{1s2p}$, the absorption fringes have no inclination, and the absorption strength is weaker than that of $W \approx W_{1s2p}$. It is worth noting that the time delay between the two pulses is scanned with a step of 0.1 fs and 0.01 fs in Figure 1a,b, respectively, which is critical in sampling the modulation period. Besides, the decay of the excited state ($1s2p$) can also be controlled by the time delay between the VUV and XUV fields. This is because the electron in the excited state experiences different field strengths of the VUV field at different time delays, thus the coupling strength from the excited state to the ground state is delay-dependent. Therefore, the population (decay) of the excited electron can be controlled by varying the time delay.

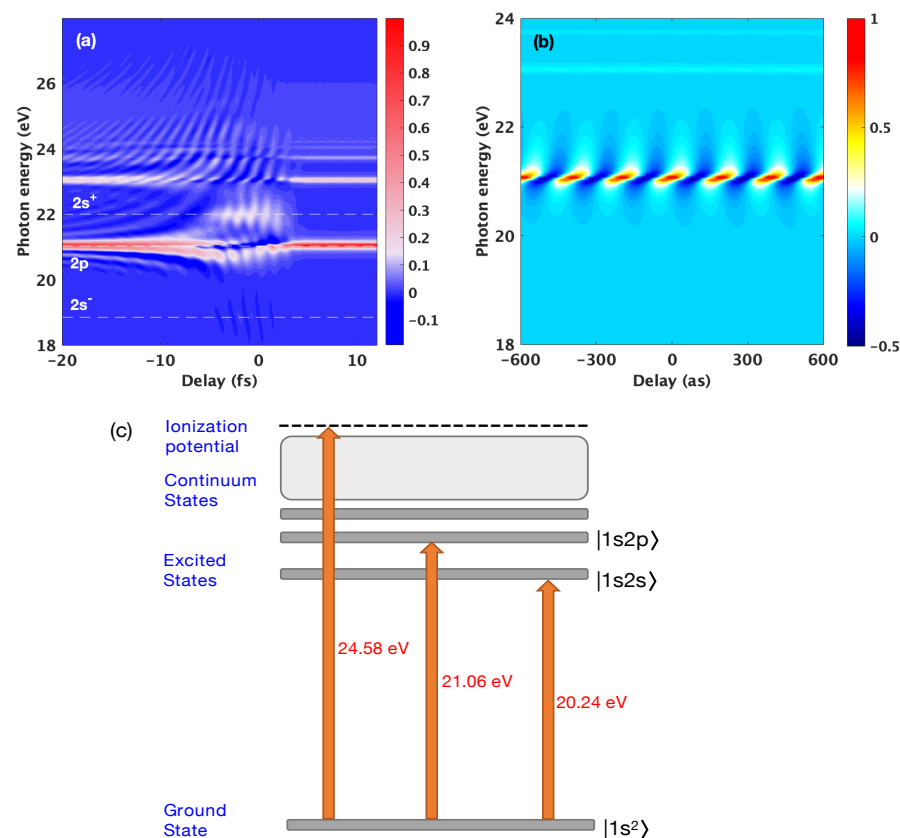


Figure 1. The ATAS (color-coded) of a helium atom obtained using the response function $S(\omega, t_d)$. (a) ATAS for XUV+IR, (b) ATAS for XUV+VUV. (c) The relevant level scheme of a helium atom.

The time-dependent perturbation theory described in Section 2.2 was applied to investigate the absorption features quantitatively shown in Figure 1b. We assumed that the single-photon excitation process does not affect the three-photon excitation because the XUV field is relatively weak, and the change in the ground-state amplitude can be ignored. Thus, the absorption process is simplified to the interference between the absorption of

the single XUV photon and the three VUV photons. The XUV and VUV fields are written as follows:

$$E_j(t) = E_j \exp(-i\omega_j t), j = 1, 2, \quad (4)$$

where E_1, E_2 and ω_1, ω_2 are the field strength and angular frequency of XUV and VUV field, respectively. This expression is suitable for describing the resonant absorption, and the integral appearing in Equation (2) can be solved analytically. As demonstrated previously, the XUV field can pump an electron in the ground state to the excited 1s2p state with a single photon; thus, the probability amplitude can be obtained from Equation (2):

$$c_1(t) = -i \frac{e d_{ba}}{\hbar} \int_0^t E_1(t') e^{i\omega_{ba} t'} dt', \quad (5)$$

where d_{ba} is the dipole transition element between the initial state $|a\rangle$ and the final state $|b\rangle$. Here, the initial and final states $|a\rangle$ and $|b\rangle$ are the ground state 1s² and excited state 1s2p, respectively. The transition angular frequency ω_{ba} is replaced by $\omega_{ba} - iA/2$, $A = 1/\tau$ to account for the lifetime τ of the 1s2p state. In the limit $t \rightarrow \infty$, the 1st-order probability amplitude is

$$c_1(\infty) = i \frac{e E_1 d_{ba}}{\hbar} \frac{1}{\omega_{ba} - \omega_1 - iA/2}. \quad (6)$$

This equation gives the frequency response of the targeted atom from the initial state to the final state excited by an XUV field. Therefore, the final probability $|c_1(t \rightarrow \infty)|^2$ represents the absorption spectrum because it is the final population of the excited state $|b\rangle$. Explicitly, the probability $|c_1(t \rightarrow \infty)|^2$ has a Lorentz profile, which describes an absorption line with a natural line width. The complex probability amplitude in Equation (6) has a phase of

$$\phi_1 = \arctan \frac{\omega_{ba} - \omega_1}{A/2}. \quad (7)$$

This phase is critical for analyzing the absorption process of the XUV field because the phase carries the information of the probability amplitude excited by the XUV field via single-photon absorption.

For the VUV field, three photons can be absorbed without violating any selection rules. By inserting the VUV field of Equation (4) into Equation (3) and integrating analytically, we obtain the probability amplitude for three-photon absorption:

$$c_2(\infty) = \frac{e^3 E_2^3}{\hbar^3} \sum_{\zeta, \nu} \frac{d_{\zeta a} d_{\nu \zeta} d_{b \nu} (X_1 + iX_2)}{[(\omega_{\zeta a} - \omega_2)^2 + \frac{A^2}{4}][(\omega_{\nu a} - 2\omega_2)^2 + \frac{A^2}{4}][(\omega_{ba} - 3\omega_2)^2 + \frac{A^2}{4}]}, \quad (8)$$

where

$$X_1 = [(\omega_{\zeta a} - \omega_2)(\omega_{\nu a} - 2\omega_2) - \frac{A^2}{4}](\omega_{ba} - 3\omega_2) - (\omega_{\zeta a} + \omega_{\nu a} - 3\omega_2) \frac{A^2}{4}, \quad (9)$$

$$X_2 = \frac{A}{2} [(\omega_{\zeta a} - \omega_2)(\omega_{\nu a} - 2\omega_2) - \frac{A^2}{4} + (\omega_{\zeta a} + \omega_{\nu a} - 3\omega_2)(\omega_{ba} - 3\omega_2)]. \quad (10)$$

The finite lifetime of the final state has been included in the expression, and Equations (8)–(10) were obtained in the limit of $t \rightarrow \infty$. A phase similar to that in Equation (7) can be obtained based on the complex probability amplitude in Equation (8):

$$\phi_2 = \arctan \left\{ \frac{\frac{A}{2} [(\omega_{\zeta a} - \omega_2)(\omega_{\nu a} - 2\omega_2) - \frac{A^2}{4} + (\omega_{\zeta a} + \omega_{\nu a} - 3\omega_2)(\omega_{ba} - 3\omega_2)]}{[(\omega_{\zeta a} - \omega_2)(\omega_{\nu a} - 2\omega_2) - \frac{A^2}{4}](\omega_{ba} - 3\omega_2) - (\omega_{\zeta a} + \omega_{\nu a} - 3\omega_2) \frac{A^2}{4}} \right\}. \quad (11)$$

The difference between ϕ_1 and ϕ_2 determines the ATAS interference. At present, we only consider the ground state 1s² ($|a\rangle$) and excited state 1s2p ($|b\rangle$); therefore, the time-dependent dipole is expressed as $d(t) = 2\text{Re}\{c_a^*(t)[c_1(t + t_d) + c_2(t)]\}$. The amplitude of

the ground state assumed to be a constant, i.e., $c_a(t) = 1$; therefore, $d(t) = 2\text{Re}[c_1(t + t_d) + c_2(t)]$ and $d(\omega) = 2\text{Re}[c_1(\infty)e^{i\omega_{ba}t_d} + c_2(\infty)]$. We replaced $c_{1,2}(\omega)$ by $c_{1,2}(\infty)$ because it represents the frequency response of the final state. This procedure has been used in the investigation of Lorentz, Gaussian, and Voigt absorption profiles [47]. Thus, the response function $S(\omega, t_d)$ can be rewritten as:

$$S(\omega, t_d) = 4\text{Im}[c_1(\infty)e^{i\omega_{ba}t_d} + c_2(\infty)]. \quad (12)$$

The frequency spectrum of the XUV electric field is treated as a constant value and is factored out in Equation (12).

We now calculate the response function using Equation (12) with the same parameters as in Figure 1b, as shown in Figure 2. The response function in Figure 2 generally recaptures the essential features of Figure 1b: near the resonance energy $W \approx W_{1s2p}$, the absorption is positively tilted along the delay axis, whereas for $W > W_{1s2p}$ or $W < W_{1s2p}$, the absorption is not inclined. However, there is some discrepancy between the figures, that is, the absorption extends to a much larger energy range, as shown in Figure 2. This discrepancy may be attributed to the exclusion of the 1s2s state as well the 3rd-order perturbation used to calculate the frequency response.

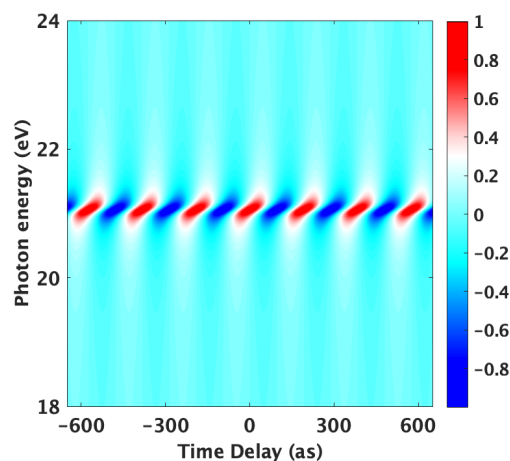


Figure 2. ATAS of a helium atom calculated using Equation (12).

It could be envisioned that at negative delays, the transient signal is absent. This can be proved by calculating the response function by scanning the time delay from -17 fs to 29 fs (not shown here). We fixed the time delay between the VUV and XUV fields to quantitatively analyze the tilted absorption spectrum near resonance. The phases ϕ_1 and ϕ_2 and their differences are plotted in Figure 3a,b, respectively. The pink and red curves represent ϕ_1 and ϕ_2 , respectively. For single-photon resonance, the phase ϕ_1 first increased slowly with an increase in energy ($W < W_{1s2p}$), and it then rapidly increased from approximately 0 to π near resonance ($W \approx W_{1s2p}$).

For three-photon resonance absorption, the phase behavior is different from that of ϕ_1 . As shown by the red curve in Figure 3a, phase ϕ_2 first decreases as the energy increases, and it then reaches its minimum value of $\pi/2$ near resonance ($W \approx W_{1s2p}$). For higher energy, phase ϕ_2 slowly increases to 0 . These behaviors significantly affect the absorption process. A more intuitive way to show the phase evolution is through the difference between ϕ_1 and ϕ_2 , which is plotted in Figure 3b. Three characteristic regions are indicated by vertical dashed lines: I ($W < W_{1s2p}$), II ($W \approx W_{1s2p}$), and III ($W > W_{1s2p}$). In resonance region II ($W \approx W_{1s2p}$), the phase difference decreases rapidly, whereas in the non-resonance region, the phase difference changes slowly. Therefore, it is straightforward to write down the condition for constructive interference as a function of frequency and time delay:

$$\omega_{ba}t_d + \phi_2(\omega) - \phi_1(\omega) = 2k\pi, \quad (13)$$

where k is a positive integer. Near resonance region II, the phase difference $\phi_2 - \phi_1$ decreases rapidly; thus, to maintain a constant phase, the time delay t_d must be increased, which results in a positive absorption tilt. For the non-resonance region, the phase difference $\phi_2 - \phi_1$ changes very slowly; therefore, the absorption exhibits no inclination. We noticed that an expression similar to that in Equation (13) was used to investigate the quantum path interference in the ATAS (XUV+IR scenario) and photon-electron spectroscopy of helium atoms [48]. For a clear comparison, we rewrite the equation from reference [48] as follows:

$$(W_{1\text{snp}} - W_{1\text{s2p}})|t_d - t_0| + \Delta\phi = 2k\pi, \quad (14)$$

where $W_{1\text{snp}}$ and $W_{1\text{s2p}}$ are the energies of the excited states 1snp and 1s2p, respectively. t_d and t_0 are the time delay and the time at which the two-photon transition (XUV+IR) peaks, respectively, and $\Delta\phi$ is the phase owing to the two-IR-photon transition. This equation successfully explains the slope of the interference fringes around 1snp or the LIS. The phase $\Delta\phi$ owing to the two-IR-photon transition is treated as a constant; however, this phase is dependent on frequency. In contrast, from Equation (13), the phase difference $\phi_2 - \phi_1$ is frequency-resolved, and the fine structure near the excited 1s2p state can be observed, as shown in Figures 1b and 2.

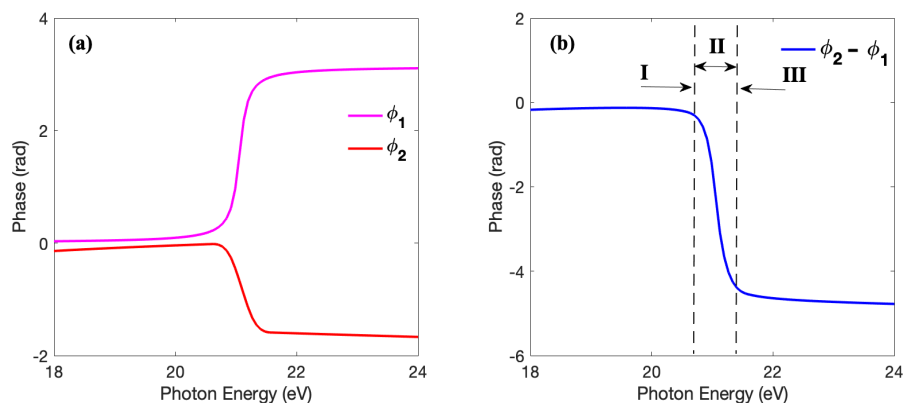


Figure 3. (a) Phase of ϕ_1 and ϕ_2 and (b) their difference $\phi_2 - \phi_1$.

The XUV or VUV pulses generated from high harmonics are naturally chirped. Therefore, investigating the chirp effect on the ATAS may provide some calibration for the experiments. Based on this consideration, we calculated the ATAS of the helium atom under the action of XUV+VUV, where the chirp of the VUV field is varied, and the other parameters are the same as in Figure 1b. As shown in Figure 4a,b, the ATAS was calculated for two chirps: $\zeta = -4$ and $\zeta = +4$, respectively. Near the resonance region, similar to the results for the chirp-free scenario in Figure 1b, the absorption fringes in Figure 4a,b exhibit a positive slope, whereas for the non-resonance region ($W > W_{1\text{s2p}}$ or $W < W_{1\text{s2p}}$), the absorption fringes become complicated. In Figure 4a, the VUV field is negatively chirped, and the absorption has a negative (positive) slope for $W > W_{1\text{s2p}}$ ($W < W_{1\text{s2p}}$). However, in Figure 4b, the VUV field is positively chirped, and the slope of the fringes is positive (negative) for $W > W_{1\text{s2p}}$ ($W < W_{1\text{s2p}}$).

The time-dependent perturbation theory presented in Section 2.2 was applied to quantitatively demonstrate the effect of chirp on the absorption fringes in Figure 4. Another formula for the chirped VUV field is applied to analytically integrate Equation (5):

$$E_2(t) = E_0 \exp[-i(\omega_2 t + \beta t^2)], \quad (15)$$

where β is the chirp rate that characterizes the chirped VUV pulse. Inserting Equation (15) into Equation (3) and taking the limit $t \rightarrow \infty$, we obtain the probability amplitude for a chirped VUV field:

$$c_2(\infty) = -\frac{8e^3\pi^{\frac{3}{2}}E_0^3}{\sqrt{i}\hbar^3\beta^{\frac{3}{2}}}\sum_{\zeta,v}d_{b\zeta}d_{\zeta v}d_{va}\exp\left\{\frac{1}{4\beta}\left[(\omega_{b\zeta}-\omega_2-i\frac{A}{2})^2+(\omega_{\zeta v}-\omega_2-i\frac{A}{2})^2+(\omega_{va}-\omega_2-i\frac{A}{2})^2\right]\right\}, \quad (16)$$

Similar to Equation (11), its phase is

$$\phi_2 = \arctan \frac{A}{4\beta}(\omega_{ba} - 3\omega_2), \quad (17)$$

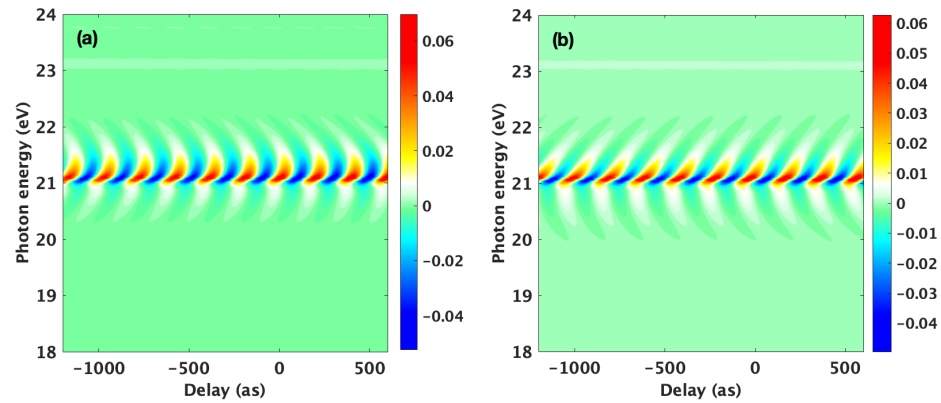


Figure 4. ATAS of a helium atom calculated using the 3D-TDSE for variable chirps: (a) $\xi = -4$ and (b) $\xi = +4$.

This is the frequency-dependent phase of the probability amplitude excited by the chirped VUV field, which significantly alters the absorption fringes in Figure 4a,b. The phase in Equation (17) is plotted for variable chirp rates in Figure 5: the light blue, brown, and magenta curves are the phases for chirp rates $\beta = 0$, $+3 \times 10^{-3}$ and -3×10^{-3} , respectively. For the negative chirp $\beta = -3 \times 10^{-3}$ (magenta curve), the phase is larger (smaller) for $W > W_{1s2p}$ ($W < W_{1s2p}$) compared with $\beta = 0$. This increases (decreases) in the phase difference $\phi_2 - \phi_1$; thus, the absorption fringes are shifted to smaller (larger) delay positions. A similar procedure may be applied to explore the effect of a positive chirp on the absorption fringes. The phase for $\beta = +3 \times 10^{-3}$ is delayed by 2π compared with $\beta = -3$; thus, the slope of the absorption fringes is enhanced, as shown in Figure 4b.

The ATAS features observed in Figures 1b and 4 may be experimentally confirmed in the future. The isolated XUV field can be obtained from high harmonic generation, and in theory, the VUV field can be generated from a KBBF ($\text{KBe}_2\text{BO}_3\text{F}_2$) crystal [49–52] or ultra-violet dispersive wave radiation [53–56]. For now, such a VUV field is unavailable in the experiment, which is a limitation of this study. The delay step used in our simulation is very small; however, a time resolution of 2.5 attoseconds can be experimentally implemented through a co-axis setup [57], which is accurate enough to sample the 0.2 fs modulation period in Figure 1b. With the development of attosecond science, the available power of XUV pulse is now in the order of 10^{13} W/cm^2 [58]. Interacting with matters, nonlinear absorption of the XUV field may occur, thus the couplings from the excited state back to the ground state can no longer be ignored. The VUV+XUV scheme applied in this study may be useful in probing these couplings.

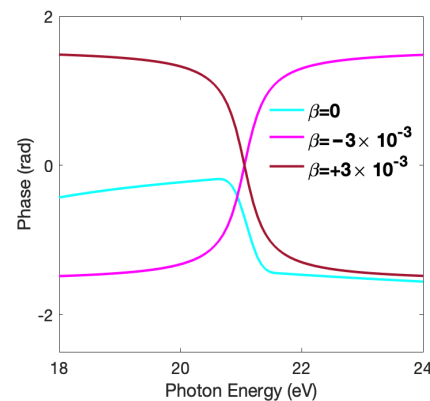


Figure 5. Phase of ϕ_2 for variable chirp rate: the brown, light blue, and magenta curves represent the phase for $\beta = 0$, $+3 \times 10^{-3}$ and -3×10^{-3} , respectively.

4. Conclusions

In this study, the ATAS of helium atoms was investigated by numerically solving the 3D-TDSE. In contrast to previous work, where synchronized IR and XUV fields interacted with a helium atom, the XUV+VUV was used to study the absorption process in the coupling between the excited states and the ground state. The central photon energy of the XUV field is 21 eV, which is a single-photon resonant with the 1s2p state. The VUV field is three-photon resonant with the 1s2p state. By interacting with these XUV+VUV fields, the absorption of the helium atom was modulated with a period that was equal to the oscillation cycle of the 1s2p state. The interference of the single- and three-photon resonant absorption results in a fine structure near the resonant state. For near-resonant energy $W \approx W_{1s2p}$, the absorption fringes are positively sloped; however, for non-resonant energy, $W > W_{1s2p}$ or $W < W_{1s2p}$, the absorption fringes exhibit no slope. Furthermore, the slope of the absorption fringes can be tailored by the chirp of the VUV field. High-order time-dependent perturbation theory was used to provide a qualitative and quantitative picture of the fine structures. The frequency-dependent phases of the single- and three-photon absorption were obtained by analytically integrating the probability amplitude induced by the VUV and XUV fields. The characteristics of the phase difference successfully explained the fine structures observed in the helium atom ATAS.

This study expands the application scope of ATAS by investigating the absorption process in the presence of XUV+VUV fields. High-order time-dependent perturbation theory is suitable for describing the resonant multi-photon absorption (not limited to one color field) and the re-absorption or emission process, in which negative frequency components $\exp(i\omega t)$ should be included in Equation (4).

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Abbreviations

The following abbreviations are used in this manuscript:

VUV	vacuum ultraviolet
XUV	extreme ultraviolet
IR	infrared
ATAS	attosecond transient absorption spectrograms
3D-TDSE	3-dimensional time-dependent Schrödinger equation
ADK	Ammosov M. V., Delone N. B. and Krainov V. P.
KBBF	KBe ₂ BO ₃ F ₂ crystal

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