

Mapping time-dependent quasi-energies of laser dressed helium

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Abstract: Extreme ultraviolet (EUV) transient absorption spectrum of helium dressed by a moderately intense infrared laser pulse is investigated. Strategies for correct retrieval of the time-dependent quasi-energies of helium with excitation energies covering both singly and doubly excited states are proposed. For long-lived singly excited states, the profound hyperbolic structures due to long lasting dipole can be diminished by convoluting the transient absorption spectrogram with a spectral window, allowing the time-dependent quasi-energies close to Is2p resonance to be correctly mapped out. For short-lived doubly excited states near 2s2p resonance, the radiation dipole decays rapidly due to autoionization and the transient absorption spectrogram already recovers the main structure of quasi-energies without the convolution operation. The quantum simulation indicates that the convolution operation controls the effective decay speed of the dipole moment, which effectively builds up an instant probe that is essential for mapping time dependent quasi-energies of laser dressed systems.

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1. Introduction

When atoms, molecules and solids are exposed to a moderately intense electromagnetic laser field, the eigenstates of the field-free Hamiltonian become laser dressed, forming "artificial" matter with energy structure optically tunable. Assuming the bare quantum system is initially prepared in state $|n\rangle$, the laser couples the eigenstates of the bare system and the dressed system evolves as $\sum_n C_n(t) |n\rangle$ with time-dependent complex amplitudes $C_n(t)$. Expanding the time varying amplitudes into Fourier series directly redefines the new stationary energies of the dressed system, which are equivalent to the so called quasi-energies derived from the Floquet theorem [1,2]. Related phenomena including AC Stark effect [3], Autler-Townes splitting [4], electromagnetically induced transparency (EIT) [5] emerge, leading to enormous applications in fields of physics [6,7], chemistry [8] and quantum information [9]. Exploring further applications of such optically dressed system normally requires the accurate determination of its energy structure. In case of a monochromatic laser field, detection of such quasi-energies can be easily implemented using static absorption and photoionization spectroscopies with high resolution [10-12]. However, in a more general case when the dressing field has finite pulse duration, the quasi-energies are no longer well-defined due to the property of the Fourier transformation and the energy landscape of the "laser+matter" system becomes time-dependent. A proper way to depict such energy structure evolution is using the windowed Fourier analysis of the associated complex amplitudes, and experimentally this relies on time-resolved approaches.

Recently, time-resolved spectroscopies with femtosecond and attosecond time resolution have been utilized to investigate the energy structure of quantum systems dressed by short pulses. Žitnik *et al.* [13] used high order harmonic to photoionize laser coupled autoionizing states of

helium and the energy structure of short-lived dressed states were successfully revealed from the ion yield map. In photoelectron/ion spectroscopies, since the ionization event occurs almost instantaneously the ultrashort EUV pulse can be treated as an instant probe to successfully interrogate the ongoing dynamics. As compared to the aforementioned spectroscopies that require sophisticated particle detection technique, EUV transient absorption spectroscopy (TAS) [14–16] is an important complementary all-optical approach that offers both simplicity and superb time resolution. Attosecond TAS has been used to trace ultrafast processes with high temporal resolution. Applications include the lifetime measurement of highly excited states [17], population transfer dynamics between different electronic states in atoms and molecules [18–20], and bandgap dynamics in solids [21]. In addition, EUV TAS is also used for study time-dependent energy structure of laser dressed atom. Michael Chini et al. [22] used isolated attosecond pulse to illuminate singly excited states of laser dressed helium. Oscillations with period of twice the frequency of the dressing laser are observed in the transmitted spectrum near 3p and 4p resonances and are attributed to the sub-cycle AC Stark effect. Recently, Kobayashi et al. [23] studied the core excited states of Xe dressed by a strong laser field with an intensity well into the tunneling regime, and higher order Floquet band with half-cycle delay dependent modulation in the absorption spectrum was observed. Stooßet al. [24] reported on the successful time-domain reconstruction of laser perturbed dipole response using a single absorption spectrum. They demonstrated the real-time mapping of the dipole response, which determines the final observable in the EUV TAS. However, the light induced dipole results from the combined action of the EUV and dressing laser fields and generally contains a long tail due to the free induction decay (FID) [25]. Thus, decoupling the effect of the EUV pulse from that of the laser field in the dipole response is key for quantitative evaluation of the transient energy structure of the laser-dressed system.

In this work, we examine the energy structure of laser dressed atom using EUV TAS, and propose a simple strategy to correctly track the time-dependent quasi-energies. A prototypical atom, helium, dressed by a few cycle near infrared (NIR) laser pulse is inspected using ultrashort EUV pulse with photon energies covering both singly and doubly excitation. When the resonant state is long-lived such as the singly excited states, a convolution between the absorption spectrogram and a spectral window function is needed to effectively shorten the time window of the radiation dipole, allowing the time-dependent quasi-energies close to 1s2p resonance to be correctly mapped out. When the resonant state is short-lived such as the doubly excited autoionizing states, the corresponding dipole is sufficiently short due to autoionization and enables directly probing time-dependent quasi-energies near 2s2p resonance. This work established the quantitative correspondence between the measured spectrogram and the predicted time-dependent quasi-energies, and proposed a simple strategy to recover the quasi-energies in a complex spectrogram, thus allows one to directly observe the time-dependent energies of virtual states. The validity of the method has also been verified by the numerical simulations.

2. Experimental method

The experimental setup is shown in Fig. 1. A 1mJ 25 fs laser pulse centered around 800 nm from a Ti: sapphire laser amplifier (Coherent Legend Elite HE+) is focused into a neon-filled hollow core fiber for spectral broadening. The broadened NIR pulse is then compressed using chirp mirrors and the pulse duration is characterized using a second-harmonic FROG. The NIR pulse is split into two arms by a beam splitter. The transmitted arm is focused into a static cell filled with 30 torr argon gas for high harmonic generation (HHG). The position of the gas cell is adjusted with respect to the driving laser focus to phase-match the short trajectory of high harmonics, resulting in EUV attosecond pulses that serve as a fast probe. Assuming the intrinsic chirp is the main cause of pulse broadening, the pulse duration of individual attosecond burst is estimated to be 300 as around 60 eV and 400 as around 21 eV. The residual laser pulse after

HHG cell is blocked by a 200 nm (Lebow company) aluminum film. Downstream the aluminum film a toroidal mirror focuses the EUV beam into a second gas cell for absorption experiment. The reflected arm from the beam splitter is acting as a pump to dress the helium atom. Its spectral phase can be tuned by inserting fused silica (FS) with variable thicknesses, leading to an adjustable waveform of the laser pulse. The reflected arm passes through a delay stage controlled by a piezoelectric transducer and recombines with the EUV pulse using an annular mirror with a 3 mm hole in the center. When the first aluminum film is unblocked, laser beams from both arms can interfere and leads to interference pattern monitored by a NIR CCD camera, from which the zero delay can be determined. Alternatively, zero delay can also be accurately determined using higher order nonlinear signals in ATAS [26]. The transmitted spectrum of the EUV beam is diagnosed with a high resolution EUV spectrometer consisting of a flat-field grating and a X-ray CCD camera. In order to block the NIR laser from reaching the X-ray CCD camera, a second aluminum film is inserted before the grating. A 300 lines/mm grating (300-006) and a 1200 lines/mm grating (300-002) are used for studying singly excited and doubly excited states of helium, respectively. The corresponding resolution of the spectrometer is calibrated to be 25 meV@20 eV and 45 meV@60 eV.



Fig. 1. Schematic of the experimental setup for EUV transient absorption spectroscopy in helium. BS: beam splitter, FS: fused silica, FM: focusing mirror, AF: aluminum filter, AM: annular mirror, TM: toroidal mirror, GC1: gas cell for high-order harmonic generation (HHG), GC2: gas cell for transient absorption, G: grating. Insets show the typical image of transmitted EUV beam of singly excited states (SES) and doubly excited states (DES) in helium from the x-ray CCD camera as well as the absorption lineout.

3. Experimental results

Figure 2 shows the absorption spectrum of DES in helium dressed by a near Fourier transform limited 7 fs NIR pulse with an intensity of about 2×10^{12} W/cm². Negative delay means the NIR pulse precedes the EUV pulse. Zero delay is determined by unblock the aluminum film right after the HHG cell. For large positive and negative delays ($\tau < -10$ fs or $\tau > 10$ fs) where the EUV and NIR pulse are well separated in time, a single absorption line is observed. It corresponds to the EUV induced $1s^2-2s2p$ transition of helium without the dressing field. The asymmetric Fano line-shape is due to configuration interaction of helium and is a typical spectroscopic fingerprint of electron correlation [27,28]. When the EUV pulse is temporally overlapping with the NIR pulse (Fig. 2(a)), the absorption profile splits into two components. This doublet structure is known as the Autler-Townes splitting when the NIR laser sets in and mainly couples the 2s2pand $2p^2$ excited states. As compared to traditional Autler-Townes splitting experiments using

much longer laser pulse, the NIR laser used here is carefully compressed and only lasts for a few optical cycle, thus the doublet feature shows profound time dependence. The bifurcation of the $1s^2$ -2s2p absorption line emerges at around -5 fs and disappears at around 10 fs, corresponding to a short-lived laser dressed atom interrogated by an ultrafast EUV pulse.



Fig. 2. Transient absorption spectra near the $1s^2$ to 2s2p resonance of helium when the NIR dressing field is carefully compressed (a); overcompensated by 1mm FS(b) and undercompensated by 1mm FS(c).

The energy structure of the dressed atom sensitively depends on the temporal waveform of the laser field. When the dressing fields with different waveforms are applied, the transient absorption spectrum which carries the fingerprint of the energy land scape of the dressed atom changes dramatically. Figure 2(b) and (c) show the transient absorption spectra of helium when the laser field is overcompensated and undercompensated by 1 mm of fused silica, corresponding to about 36 fs^2 and $-36 fs^2$ group delay dispersion introduced to the original pulse, respectively. For both cases, the laser pulse is stretched and the NIR pulse duration is estimated to be 16 fs, so the doublet structure in the absorption spectrum lasts for longer delay ranges. The relatively weaker sub-peak $(2p^{2-} \text{ in Fig. 2(a)})$ appearing in the higher photon energy is similar to previous studies when the laser frequency is slightly detuned from the atomic transition energy [29]. The fringes in Fig. 2 are present in the overlap region and are due to the interference between two quantum path leading to the same final virtual state. Due to the dispersion introduced by the fused silica, the stretched laser pulse carries a frequency sweep which results in the monotonic increase/decrease of the energy of the light induced state with respect to the delay (see the black dashed lines in Fig. 2). Therefore, the absorption profile is a direct manifestation of the time-dependent quasi-energy of doubly excited helium dressed by a laser field.

EUV transient absorption spectroscopy for SES of helium is also performed by using the 300 lines/mm grating. In order to generate harmonics below the first ionization threshold of helium with sufficient flux, a NIR pulse with a pulse duration of 12 fs is used as the driver, and its replica is used as the dressing field. The intensity of the NIR pulse is kept below $1 \times 10^{12} \text{ W/cm}^2$ to avoid significant depletion of the excited states. The measured spectra at the vicinity of the $1s^2$ -1s2ptransition are shown in Fig. 3. As compared to the case of DES in Fig. 2, the absorption lines are narrower since SES have much longer lifetimes [30]. In addition, more absorption features are observed. The profound absorption features around 21.7 eV and 22.1 eV can be identified as light induced states $2s^+$ and $3d^-$ involving the dark states 1s2s and 1s3d, respectively [31,32]. These are the quasi-energies of a dressed atom that we are after. However, prominent hyperbolic lines (white lines in Fig. 3(a), (b) and (c)) near the corresponding transitions emerge and overlay with the quasi-energy features. Such complex absorption profiles hinder the direct visualization of the time-dependent laser-dressed states. These hyperbolic lines are attributed to perturbed free induction decay. The EUV pulse initiates a long lasting ringing dipole, which is then perturbed by a delayed laser field via coupling the excited *np* states to nearby *ns* or *nd* states. The abrupt temporal perturbation on the dipole leads to sideband structure in the frequency domain.

In order to suppress the hyperbolic sidebands and emphasize the absorption features corresponding to laser-dressed states, we convolute the EUV absorption spectrum with a spectral



Fig. 3. Transient absorption spectra for singly excited states of helium. (a), (b) and (c) are same as Fig. 2 except that the absorption spectrum near the $1s^2$ to 1s2p resonance is inspected, white lines represent the prominent hyperbolic lines. (d), (e) and (f) are the corresponding convoluted spectra by convoluting the TAS with a 70 meV spectral window function for each delay.

window at each delay, and the convoluted spectrogram is shown in Fig. 3(d), (e) and (f). It shows that the convolution operation significantly diminishes the sideband structure, and a clean spectrum mainly consisting of the laser-dressed states is obtained. Although the spectral width of each absorption line is broadened, the mean position of the transient quasi-energy can be readily recovered.

4. Discussion

To confirm the correspondence between the observed absorption lines and the quasi-energies of a laser dressed atom, we numerically solve the time-dependent Schrödinger equation using a multi-level model. To mimic the experimental results for DES (Fig. 2), we include the $1s^2$ ground state, 2s2p, $2p^2$ excited states in the model. 30 continuum states are also included to consider the autoionizing effect [15]. The time-dependent Schrödinger equation is:

$$i\partial_t \begin{pmatrix} c_g \\ c_a \\ c_b \\ c_\varepsilon \end{pmatrix} = \begin{pmatrix} E_g & d_{ga}F(t) & 0 & d_{g\varepsilon}F(t) \\ d_{ga}^*F^*(t) & E_a & d_{ab}F(t) & V_{\varepsilon,a} \\ 0 & d_{ab}^*F^*(t) & E_b & 0 \\ d_{g\varepsilon}^*F^*(t) & V_{\varepsilon,a}^* & 0 & C \end{pmatrix} \begin{pmatrix} c_g \\ c_a \\ c_b \\ c_\varepsilon \end{pmatrix}$$
(1)

where $c_i(i = g, a, b, \varepsilon)$ is the time dependent coefficients of states $|g\rangle \equiv |1s^2\rangle$, $|a\rangle \equiv |2s2p\rangle$, $|b\rangle \equiv |2s^2\rangle$ and the continuum states. d_{ij} is the dipole moment. F(t) is the external electromagnetic field. The configuration interaction matrix elements $V_{\varepsilon,a} = -0.0025$ a.u. and the decaying rate parameter $\gamma = 0.025$ are adjusted to reproduce the absorption line profile of the 2s2p state in the weak field limit. The simulation results are shown in Fig. 4. The frequency sweep of carrier of the dressing field can be clearly observed. It should be noted that higher-order dispersion should also contribute to the observed absorption profiles. For 1 mm glass, the transient frequency variation of the dressing field caused by the higher-order dispersion is small, thus the observed curvature of the time-dependent quasi-energy is dominated by the intensity profile of the dressing field in our measurements. These results show rather similar pattern to that of the experimental ones in Fig. 2.



Fig. 4. Quantum simulation results for doubly excited states of helium when the NIR dressing field is Fourier transform limited (a); overcompensated by 1mm FS(b) and undercompensated by 1mm FS(c). The white dashed and solid lines represent the predicted quasi-energies. The 760 nm NIR pulse has a Fourier transform limited pulse duration of 7 fs and the peak intensity of 2×10^{12} W/cm².

Time delay τ (fs)

In case of SES, we include the ground state, 1s2p, 1s2s and 1s3d excited states in the simulation. We numerically solve the TDSE in a time window of ~ 200 fs to account for the instrumental resolution. The simulation results are shown in Fig. 5. The main light-induced absorption structures $2s^+$ and $3d^-$ around zero delay and their delay dependent evolution are well reproduced in the calculated TAS. (Fig. 5(a)-(c)). In addition, clear hyperbolic sidebands for positive delays appear in case of SES, which are greatly suppressed after convoluting the absorption spectrum with a 120 meV spectral window function (Fig. 5(d)-(f)).



Fig. 5. Quantum simulations for singly excited states of helium. (a), (b), (c) are same as Fig. 4 except that the absorption spectrum near the $1s^2$ to 1s2p resonance is inspected. (d), (e) and (f) are the corresponding convoluted spectra by convoluting the TAS in (a) (b)(c) with a 120 meV spectral window function for each delay. The NIR pulse has a pulse duration of 12 fs with a central wavelength of 800 nm, the peak intensity is 8×10^{11} W/cm².

The quasi-energies of the laser-dressed helium can be evaluated by analyzing the interaction Hamiltonian in the adiabatic basis. The Hamiltonian of helium atom in the presence of an external non-ionizing field $\varepsilon_{NIR}(t)$ is expressed as $\hat{H}(t) = H_0 + \varepsilon_{NIR}(t) \cdot \vec{r}$ under the dipole approximation. Here H_0 is the field-free Hamiltonian and \vec{r} is the position operator. Considering the Rotating Wave Approximation (RWA) [33], and viewing in a proper rotating frame, the time-dependent Hamiltonian $\hat{H}(t)$ in the interaction picture is:

$$\widehat{H}(t) = \begin{pmatrix} 0 & \Omega_{ab} \\ \Omega_{ba} & -(\dot{\Delta}t + \Delta) \end{pmatrix}$$
(2)

where $\Omega_{ab} = \Omega_{ba}^* = \frac{1}{2}\hbar d_{ab}A(t)$ is the time-dependent Rabi frequency, d_{ab} is the dipole matrix element between state $|a\rangle$ and state $|b\rangle$. A(t) is the envelope of the NIR laser pulse. $\Delta = \omega(t) - E_{ba}$ is the detuning of the laser frequency $\omega(t)$ from the transition energy $E_{ba} = E_b - E_a$. Equation (2) is derived for a two level atom and can be easily extended to a multi-level system. The quasienergies are evaluated by diagonalizing the Hamiltonian $\hat{H}(t)$. The calculated quasi-energies are depicted using white lines overlaid on the calculated TAS in Fig. 4 and Fig. 5. The positions of the quasi-energies for both DES and SES cases show excellent agreement with that of the absorption features, indicating that the time dependent quasi-energies of helium can indeed be precisely probed using the TAS.

In order to interpret the physical mechanism and extract the quasi-energies from the absorption profile, we carried out an analytical analysis of the delay dependent absorption spectrum. In case of a dilute gas sample, the delay dependent optical density is proportional to the imaginary part of the microscopic dipole moment [34]:

$$OD(\omega, \tau) \propto \operatorname{Im}\{d(\omega, \tau)\} \propto \operatorname{Im}\left\{\sum_{n} C_{n}(\omega, \tau)d_{gn}\right\}$$
(3)

Here $C_n(\omega, \tau)$ is the Fourier transform of the complex-valued amplitude $C_n(t, \tau)$ and depicts the contribution of state $|n\rangle$ to the dipole moment $d(\omega, \tau)$, here τ represents the arrival time of the EUV pulse. Assuming the EUV pumping process is instantaneous, the amplitude of the excited state $|n\rangle$ can be formulated as:

$$C_n(t,\tau) = \begin{cases} A e^{-i\left(\int_{\tau}^{t} \omega_1 dt' + \varphi_0\right)} \times e^{-\kappa(t-\tau)} & t \ge \tau \\ 0 & t < \tau \end{cases}$$
(4)

where A is determined by the intensity of the attosecond EUV pulse, ω_1 is the time dependent quasi-energy, φ_0 is the initial phase of the EUV induced polarization dipole, and κ is associated with the lifetime of state $|n\rangle$. The decaying factor $e^{-\kappa t}$ is effectively setting a time window $[\tau, \tau + 1/\kappa]$ on the phase evolution $e^{-i(\int_{\tau}^{t} \omega_1 dt' + \varphi_0)}$, therefore the absorption spectrum depicted by Eq. (3) is essentially a windowed Fourier analysis of the associated complex amplitudes. If the time window is sufficiently narrow, the time evolution of the quasi-energy can be estimated as:

$$OD(\omega,\tau) \propto \frac{Ad_{ng}\kappa \cdot \cos\varphi_0}{(\omega-\omega_1(\tau))^2 + \kappa^2} + \frac{Ad_{ng}(\omega-\omega_1(\tau)) \cdot \sin\varphi_0}{(\omega-\omega_1(\tau))^2 + \kappa^2}$$
(5)

The corresponding energy where the local maxima of Eq. (5) occurs is:

$$\omega = \begin{cases} \omega_1(\tau) & \varphi_0 = 0\\ \omega_1(\tau) + \frac{1 - \cos \varphi_0}{\sin \varphi_0} \kappa & \varphi_0 \neq 0 \end{cases}$$
(6)

Equation (6) indicates that the time dependent quasi energy ω_1 can be directly retrieved from the measured absorption spectrum by finding the local maxima in the OD. A prerequisite is that the time window $[\tau, \tau + 1/\kappa]$ needs to be sufficiently narrow. The width of the time window is determined by the lifetime of the exited states. In case of DES in helium which has a short lifetime (the lifetime of the 2s2p state is measured to be 17 fs [35]), the quasi-energy in time domain is directly trackable from the absorption spectrum as demonstrated in Fig. 4. Figure 6(a), (b), and (c) show the retrieved quasi-energy using Eq. (6). The initial dipole φ_0 can be fitted as -0.25π from the fano absorption line profile [36]. The quasi-energies in Fig. 6 is extracted directly from the calculated OD in Fig. 4. For each delay, the local maxima of the OD are

directly read out from the convoluted absorption spectrum from 59.5 eV to 61 eV. Since the dressing field is compressed to 7 fs in Fig. 4(a), the quasi-energy of the laser dressed atom only lasts for a few femtoseconds. Therefore, we only consider a time window of about 7 fs for the retrieved quasi-energies in Fig. 6(a). Absorption structures beyond this time window show hyperbolic lines caused by the residual tail of the decaying EUV induced dipole, and can be eliminated if convoluted with extra spectral window with sufficient width. However, even without the convolution, the quasi-energies for DES in helium can still be recovered reasonably well as shown in Fig. 6(a), (b), and (c).



Fig. 6. Retrieval of time dependent quasi-energies from the calculated transient absorption spectrum. (a), (b), (c) are the retrieved results (solid lines) from Fig. 4 as compared with the predicted ones (dashed lines) for DES in helium. (d), (e), (f) represent retrieved results (solid lines) from Fig. 5 as compared with the predicted ones (dashed lines) for SES in helium.

In case of a long-lived excited state, e.g. the laser dressed *Is2p* state, Eq. (4) indicates that the absorption spectrum does not provide time-resolved information of the quasi-energy but rather shows a weighted average of all quasi-energies. In order to create a fast decaying time window on the dipole moment for accurate retrieval of the laser dressed states, the original absorption spectrum $OD(\omega, \tau)$ is convoluted with a Lorentzian function $\sigma(\Delta)$ with a spectral width of Δ , leading to a modified spectrum,

$$OD_{conv}(\omega, \tau) = OD(\omega, \tau) \otimes \sigma(\Delta)$$
 (7)

 \otimes is the convolution operation. According to the convolution theorem, the modified amplitude is $C_M(t,\tau) = C(t,\tau) \cdot e^{-\Delta t} = A e^{-i \int_{\tau}^{t} \omega_1 dt'} \cdot e^{-(\kappa + \Delta)(t-\tau)}$. Thus the convolution operator is equivalent to multiplying a fast decaying factor in the time domain dipole moment and controls the effective

decay speed of the dipole. With Δ sufficiently large, the time dependent energies of the laserdressed states can be properly recovered again using Eq. (6). In case of SES $\varphi_0 = 0$, the quasi-energy corresponds to the energy where OD maximizes. The retrieved results are shown in Fig. 6(d), (e), and (f). Note that following Eq. (6), the local maximum in OD coincides with the predicted quasi-energy when the decaying factor $e^{-\kappa t}$ has a width approaching zero for SES in principle. Therefore, when a finite width is considered for the convolution process, small discrepancy exists as shown in Fig. 5, the maximal discrepancy is ~ 0.03 eV in Fig. 6(d) close to zero delay. However, the overall agreement between the retrieved results and the predicted ones calculated from the Hamiltonian is quite satisfactory, verifying the validity of the current method. Using the same method, the time-dependent quasi-energies of the DES (SES) of helium atoms have been extracted from the measured absorption spectrogram in Fig. 2 (Fig. 3) and the results are shown in Fig. 7.



Fig. 7. Retrieval of time dependent quasi-energies from the measured transient absorption spectrum. (a), (b), (c) are the retrieved results from Fig. 2 for DES in helium. (d), (e), (f) represent retrieved results from Fig. 3 for SES in helium.

5. Conclusions

In conclusion, the energy structure of laser-dressed helium atom is investigated using EUV attosecond transient absorption spectroscopy. Complex absorption features appear when both EUV probe and NIR dressing field overlap in time. Theoretical analysis shows that the absorption spectrum is equivalent to a windowed Fourier transform of the oscillating dipole, indicating an effective means for tracing the time-dependent quasi-energies. We have demonstrated that the quasi-energies of DES can be directly observed thanks to their short lifetimes. The quasi-energies

associated with long-lived SES is also trackable when a convolution operation is applied to the absorption spectrogram. The current study demonstrated the possibility to accurately map time-dependent quasi-energies in atoms using transient absorption spectroscopy, and it holds the promise for detecting quasi-energies for complex systems such as molecules. Molecules have more congested energy levels and the convoluted absorption peaks may merge together. To extend this method to molecules, additional fitting procedures are needed to pinpoint the individual quasi-energy. Such interesting and important aspect will be subject to future studies.

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