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Attosecond counter rotating wave effect in xenon driven by strong fields

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We investigate the sub-femtosecond dynamics of a highly excited xenon atom coherently driven by a strong control field at which the Rabi frequency of the system is comparable to the frequency of a driving laser. The widely used rotating wave approximation (RWA) breaks down at such fields, resulting in new features such as counter rotating wave (CRW) effect. We present the first time-resolved observation of the CRW effect motion in the highly excited $4d^{-1}np$ xenon using using attosecond transient absorption spectroscopy (ATAS). Time-dependent many-body theory confirms the observation and explains the various features of the absorption spectrum seen in experiment.

The manipulation of atoms, molecules and solids and interest in electron dynamics in strong fields has been long standing. Much attention has been recently paid to radically new optical and electrical properties such as electromagnetically induced transparency (EIT) [1,2], red shifted optical absorption [3], high-order harmonics generation (HHG) [4-7] and semi-metallization [8,9]. We have just begun to understand the strong-field response of various forms of materials. In analysis of light-matter interaction, the rotating wave approximation (RWA) has been widely used, which holds as long as the Rabi frequency, which is proportional to the driving field, is much smaller than the driving frequency. For strong light-matter interaction, this approximation breaks down and new effects appear, originating from the counter-rotating terms in the Hamiltonian [10]. With ultrashort pulses and the development of attosecond metrology [11], it is now possible to study CR effects on a sub-femtosecond time scale. These new CR effects have important consequences on the dynamics of the system and can lead to interesting applications as shown in circuit quantum electrodynamics [12], quantum computation (QC) using SQUIDS [13] and in the creation of entanglement between two atoms [14].

In this work, we report, for the first time, the real time observation of ultrafast sub-cycle CRW feature with optical frequencies. We study the CRW effect in highly excited $4d^{-1}np$ states in atomic xenon, which are dressed by an intense IR pulse. We probe the field-driven dressing dynamics in the $4d^{-1}np$ manifolds using an attosecond pulse delayed with respect to the IR pulse. All $4d^{-1}np$ states are autoionizing states with a lifetime of about 6.6fs, due to the Auger decay of the unstable 4d hole. Consequently, the CRW has to be faster than the lifetime to be observed. For an 800nm pulse, the optical period is 2.6fs and the CRW motion occurs within 1.3fs, beating several times within the core lifetime. To observe the ultrafast electron dynamics in the real time, we applied ATAS [15-20], which extends transient absorption spectroscopy to attosecond time scales. The appearing of CRW effects in ATAS has been theoretically studied [21,22] but has not been clearly seen in experiment.

ATAS experiments have focused so far on ionizing or excited outer-valence electrons [15-19]. Here, we study for the first time highly excited metastable states with a core-hole in the 4d shell. These states have lifetimes of only a few femtoseconds. Studying their dynamical properties with common spectroscopic approaches using quasi-monochromatic pulses has not been possible. ATAS overcomes this obstacle where the attosecond extreme ultraviolet (XUV) pulses accesses the highly excited states and at the same time probes their dynamical response on a sub-cycle time scale using an intense IR pulse [16,17,20].

The experimental setup used in our experiments is shown in Fig.1. A carrier-envelope phase stabilized

Ti:Sapphire, few-cycle laser pulse (4 fs FWHM, 800nm, 300 μ J) is focused on to a static neon gas cell to produce HHG radiation in the XUV region. The XUV attosecond pulses generated by the process of HHG in Ne gas co-propagates along with the residual infrared (IR) pulse to the main experimental chamber and is used for pump probe studies. The details of the experimental setup is given in the Appendix .

The attosecond XUV and IR pulse are then focused on to a target gas cell. After the gas cell, the IR pulse is cut off using a 0.15 μ m aluminium filter and only the attosecond XUV pulse is steered and focused at the entrance slit of an XUV spectrometer (see Appendix) using a spherical mirror.

The ATAS is constructed by measuring the transmitted spectrum (I_{gas}) through the gas target (Xe) as a function of delay (τ) between IR and XUV pulses from delays of -10fs to 6fs (negative delays correspond to XUV pulse arriving first) with a step of 200as. Each measurement corresponds to an average taken over \sim 9000 shots. The absorption spectrum is calculated using the reference XUV spectrum (I_0) taken in the absence of the xenon gas target. The optical density (OD) is calculated by using the following expression

$$OD(E, \tau) = \ln \left(\frac{I_{\text{gas}}(E, \tau)}{I_0} \right)$$

for each delay. The raw transient absorption spectrum obtained from this raw data is then filtered and processed for greater fidelity (see Appendix) to obtain the ATAS shown in Fig 2a.

Figure 2a shows the optical density measured from xenon in the spectral region of 64 to 70eV as a function of delay between the NIR and XUV pulse. The intensity of the NIR field for these measurements was about 2×10^{13} W/cm². The absorption lines correspond to the core-electron excited states $4d_{5/2}^{-1}6p$, $4d_{3/2}^{-1}6p$ and $4d_{3/2}^{-1}7p$ as shown in Fig 2c. The three main features noted in the region of overlap between the pulses are (1) the splitting of the absorption lines, (2) the asymmetry in the splitting (dotted yellow line) and (3) the oscillating substructures within them. The oscillating sub-structures have a frequency of about twice the NIR frequency. This is seen more clearly in Fig 2d (blue line), where the optical density integrated over a bandwidth of 200 meV centered at the 65eV (area between the black vertical lines in Fig 2a) absorption line ($4d_{5/2}^{-1}6p$) is shown as a function of delay between the IR and XUV pulse.

We have used a time-dependent many-body approach based on configuration-interaction singles (TDCIS) [23,24] to understand the features seen in the absorption spectrum. TDCIS describes successfully many-electron atoms in the presence of strong-field pulses and has been used to understand various atomic and many body effects at ultrashort time scales [25,26]. The absorption spectrum from the calculations was convoluted with the spectrometer resolution to compare with the experimental results. The calculations were performed assuming a Gaussian IR pulse of about 4fs, centered at 800 nm with a peak intensity of 2×10^{13} W/cm² and a 200as XUV pulse centered at 66 eV.

The calculated absorption spectrum for xenon (Fig 2b) agrees well with experiment (Fig 2a) showing both the sub-cycle oscillating features and the asymmetric Autler-Townes splitting (dotted line). The calculated absorption spectrum also reproduces the slopes for the oscillating structures (shown by red lines in Fig 2a) for the $4d_{5/2}^{-1}6p$, $4d_{3/2}^{-1}6p$ absorption lines. The differences in the slopes are a direct consequence of the phase difference between the different pathways involved in the interactions [27].

The most important feature seen in our experiments are the $2\omega_{\text{IR}}$ oscillations of the absorption line when the IR and XUV pulse overlap. The $2\omega_{\text{IR}}$ oscillation are seen clearly in Fig 2d, where the $\cos(2\omega\tau)$ fit to the experimental data shown in red. This oscillation should not exist according to the RWA but constitute the CRW effect. The $2\omega_{\text{IR}}$ oscillation has been observed in earlier experiments [28,29] and have been typically understood in terms of which-way (two path) interference where the absorption of a XUV photon represents the direct pathway and the indirect pathway consists of a third order process involving the absorption of a XUV photon plus the absorption/emission of two IR photons. In our case, the possibility of two-path interference contributing to the sub-cycle oscillations is much smaller than that of CRW effect. This can be inferred from Fig 2c, where the accessible states from $4d_{3/2}^{-1}6p$ and $4d_{5/2}^{-1}6p$ states by single IR photon are shown. A two-photon process would involve continuum energy states above the $4d_{3/2,5/2}^{-1}$ ionization limit rendering the process very unlikely. Also the fact that we do not see $2\omega_{\text{IR}}$ oscillations for negative delays (XUV pulse comes first) is another strong indication that which-way interference is negligible here [27,28]. Including only bound states in

the calculations, so that which-way interferences are basically prohibited (see Fig 3a), the simulated result reproduces the experimental observation and is another confirmation for the CR nature of the $2\omega_{\text{IR}}$ oscillations.

The origin of the counter-rotating wave terms can be understood in a simple field-driven 2-level system. The Hamiltonian in the rotating frame [30] reads

$$\hat{H} = \begin{pmatrix} \Delta & 0 \\ 0 & 0 \end{pmatrix} + \frac{\Omega}{2} \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} + \frac{\Omega}{2} \begin{pmatrix} 0 & e^{2i\omega t} \\ e^{-2i\omega t} & 0 \end{pmatrix}$$

where $\Omega = \mu E$ is the Rabi frequency, μ is the transition dipole moment, E is the amplitude of the electric field, ω is the transition frequency, and Δ is the detuning. In the RWA, only the first two terms are kept, and the rapidly oscillating third term, which is the CRW term, is dropped. In the first order perturbation theory for the RWA, one finds that the strength of the CRW correction is proportional to Ω/ω and oscillates with frequency of 2ω [16,19]. Note that the strength of this term does not depend on the pulse duration [30]. Conventionally, in the optical regime, the Rabi frequency is commonly much smaller than the driving frequency $\Omega/\omega \ll 1$. Even for weak field, the CRW term may have a significant influence by modifying the resonance condition known as Bloch-Siegert shift [31,32] and rendering the Rabi frequency time-dependent [30]. The CRW term gives rise to a second (time-dependent) pathway to excite/deexcite the system, which depending on the time t interferes constructively or destructively with the RWA pathway. This interference happens twice per cycle resulting in the 2ω modulation of the population. As the Rabi frequency Ω increases comparable to ω , the probability of excitation (de-excitation) within a quarter of an optical cycle increases as well making CRW modulation more visible. With the emergence of intense femtosecond pulses reaching intensities above 10^{13} W/cm², it is nowadays possible to induce these Rabi splittings of ~ 1 eV [33,34], breaking down RWA. Generally, large noble gas atoms possess larger dipole transition moments than light atoms favoring CRW effects. Therefore, xenon is an ideal choice for studying CRW effects. In xenon, the Rabi splitting is 1.4 eV, for resonantly driven the $4d_{5/2}^{-1}6p - 4d_{5/2}^{-1}6s$ transition of the intensity of 2×10^{13} W/cm² of laser light (photon energy of ~ 1.3 eV), and exceeds the resonant driving frequency. CRW effects should be consequently very prominent as Figs. 2a and 2d confirm.

To demonstrate the influence of the CRW, we focus on the $4d_{5/2}^{-1}6p$ line and performed two series of calculations: (1) varying laser intensities (Fig 3a) and (2) changing driving frequency and the energy spacing between the excited states (Fig 3b). We limit ourselves to the 3-level system and resonant driving frequency ω between the states $4d_{5/2}^{-1}6p$ and $4d_{5/2}^{-1}6s$ to demonstrate how the CRW depends on these variables as the ratio Ω/ω varies. By increasing the intensity, the Rabi frequency increases but the driving frequency stays the same. The visibility of the 2ω oscillations improves as the intensity increases as shown in Fig 3a. Also the Autler-Townes splitting move further apart and becomes spectrally broader. In xenon this is a problem, as the neighboring $4d_{3/2}^{-1}6p$ states is only 2eV away and starts to overlap with feature of the $4d_{5/2}^{-1}6p$ line. This also limits the range of useful IR pulse intensities to observe the CRW effect.

The series in Fig 3b shows the absorption spectra keeping the $4d_{5/2}^{-1}6p$ level fixed but scaling all the transition energies to the $4d^{-1}nd$ and $4d^{-1}ns$ energies by a common factor. In this way, the Rabi frequency Ω is kept constant and the driving frequency ω is varied. The energy levels and the driving frequency is scaled by factors of 0.5, 1 and 1.5 respectively keeping the intensity constant at 2×10^{13} W/cm². With increasing driving frequency, the CRW oscillations become faster and the visibility goes down due to the decreasing ratio of Ω/ω . Both trends shown in Fig. 3 are beneficial when changing the atom from neon to xenon. The ratio Ω/ω increases because Ω increases due to the larger dipole strengths between states and the energy spacing between the neighboring ns and np states slightly decreases.

Even though CRW effect have been often discussed in the connection with short femtosecond pulses, it is not the shortness of the pulse (or broad spectrum) that leads to an enhanced CRW effect but only the large peak intensities that are possible with these short pulses. These can be seen in Fig 4 where the ATAS is shown for a few-cycle (Fig. 4a) and many-cycle (Fig. 4b) pulses, respectively. The cut along a Autler-Townes branch (black lines in Fig4a and 4b) is made and shown in Fig. 4c. The 2ω oscillations are clearly seen and the strengths of the oscillations are very similar for both pulses in the region where pump and probe pulses overlap, which is highlighted for the few-cycle pulse. As expected, in the non-overlapping region no 2ω oscillations exist. The overlap region for the multi-cycle pulse lies between -10.7 fs and 10.7 fs making the CRW

oscillations visible over a larger range of delays.

To address the asymmetry of Autler-Townes splitting observe in the experiments (Fig 2a), the deformation of the $4d_{5/2}^{-1}6p$ line was calculated, including only $4d_{5/2}^{-1}6p$, $4d_{5/2}^{-1}6s$ and the neutral ground state, and we find that the Autler-Townes splitting is not very asymmetric. Including additionally $4d_{5/2}^{-1}nd$ ($n=5,6,7$) and $4d_{5/2}^{-1}ns$ ($n=7,8$) configurations totaling to an overall of 14 energy levels, the calculations resemble the asymmetric Autler-Townes splitting quite well. This strong asymmetry, where almost exclusively the upper branch of Autler-Townes splitting is seen, is due to the contributions of all the excited states.

In conclusion, we have shown that an intense IR field with an intensity of 10^{13} W/cm² leads to counter-rotating wave dynamics, which for optical pulses are on a sub-femtosecond time. With attosecond transient absorption spectroscopy we time-resolve this fast dynamics, which has not been seen so far in the optical regime. The shortness of the IR and the attosecond pulse allows for characterizing highly excited electronic states that are challenging to access with direct spectroscopic approaches due to their short lifetimes. Theoretical calculations match well with our experimental measurements and explain why the line deformation is highly asymmetric even though the excited electron is still bound by several eV. We could also confirm that the sub-cycle oscillation in the asymmetric Autler-Townes doublet is due to the CRW effects, which has been theoretical predicted but has not been seen clearly experimentally so far. This work demonstrates the power of ATAS to observe complex electronic motion like the electronic motion beyond the RWA approximation, which has been shown to be fundamentally important in areas like circuit QED [12] and QC [12,13].

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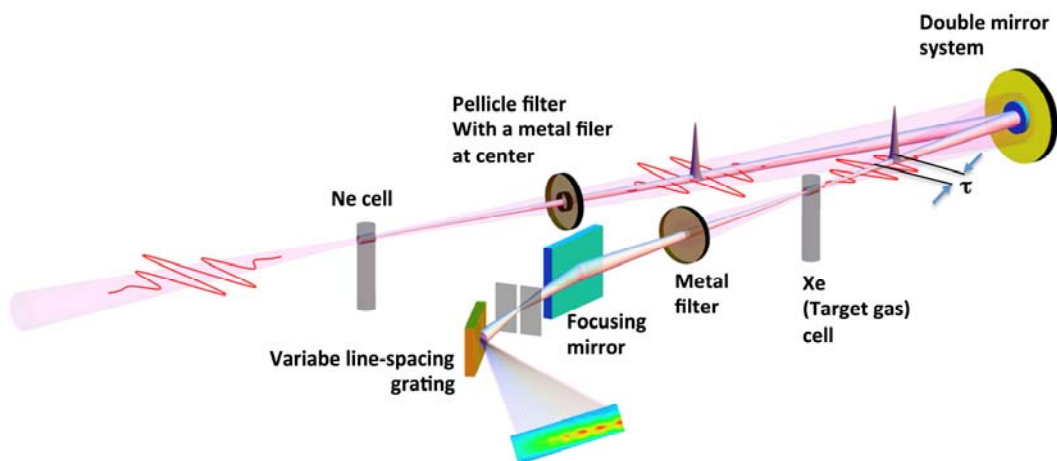


Fig. 1. Experimental setup for attosecond transient absorption spectroscopy

Attosecond XUV pulses are generated from Neon gas, using few-cycle femtosecond NIR pulses (4 fs, 800nm, 0.3 mJ). A concentric two segment curved mirror along with a special metal filter (see appendix), separates the attosecond XUV and the IR pulses and provides a precision delay between them. The two beams are then focused collinearly onto a quasi-static gas cell containing xenon as a target gas. The absorption spectrum is obtained by measuring the transmitted spectrum (I_{gas}) from gas target and also a spectrum in the absence of the gas target (I_0) as a function of delay between NIR and XUV pulses.

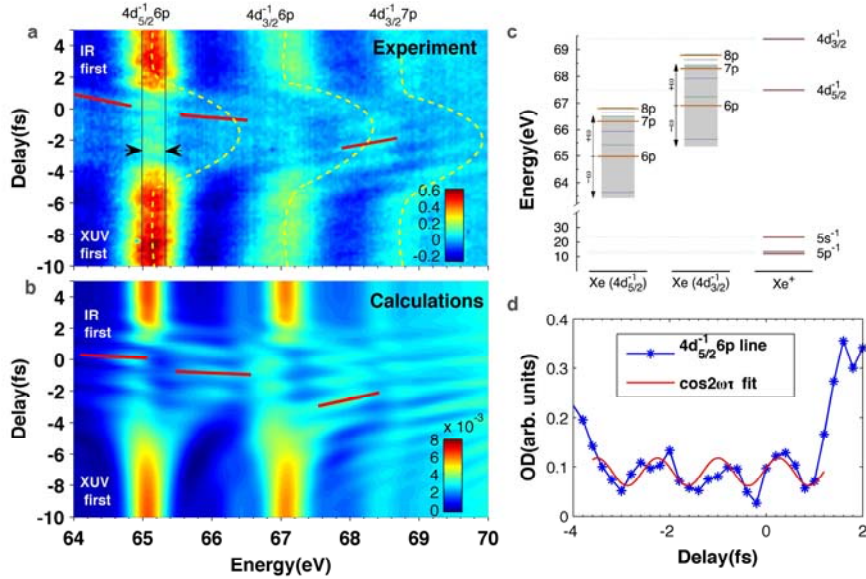


Fig. 2. Attosecond Transient absorption spectrum of Xe. (a) Experimental absorption spectra at different delays between XUV and IR pulse at IR intensity of about $2 \times 10^{13} \text{ W/cm}^2$ (b) TDCIS calculations under similar conditions. (c) inner shell excited states of xenon with a hole in $4d_{5/2}$ and $4d_{3/2}$ shell. The ionization thresholds of the various shells are shown in the third column. (d) Sub-cycle oscillation seen at 65eV line ($4d_{5/2}^{-1}6p$, blue curve) in the experimental spectrum, averaged over 200 meV bandwidth shown using two black lines in figure (a). The $\cos(2\omega\tau)$ fit to the experimental data is shown in red. Negative delays correspond to the arrival of an XUV pulse before an NIR pulse.

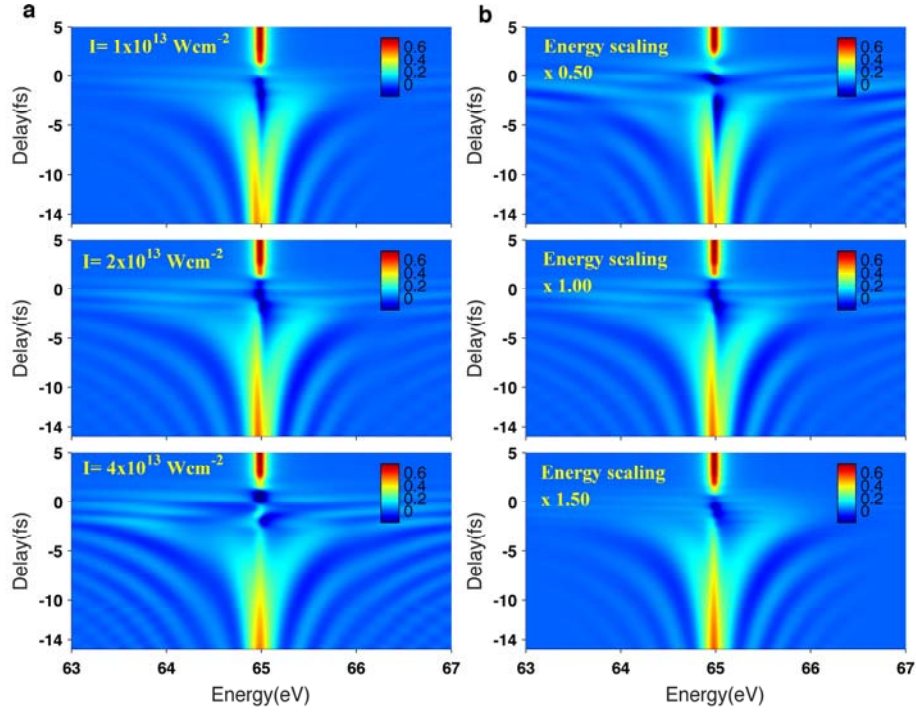


Fig. 3. Change of absorption line of the $4d_{5/2}^{-1}6p$ state for different conditions of the control field.
 (a) Calculations are done for different intensities ($1, 2, 4 \times 10^{13} \text{W/cm}^2$). As the ratio of Rabi frequency to the control field frequency increases, the oscillating features become more prominent. (b) Calculations are done at an fixed intensity of $2 \times 10^{13} \text{W/cm}^2$ with scaling the separation of all the $4d_{5/2}^{-1}nd$, $4d_{5/2}^{-1}ns$ states with respect to $4d_{5/2}^{-1}6p$ state by a factor of 0.5, 1 and 1.5. As the scaling increases, the oscillating effect diminishes considerably.

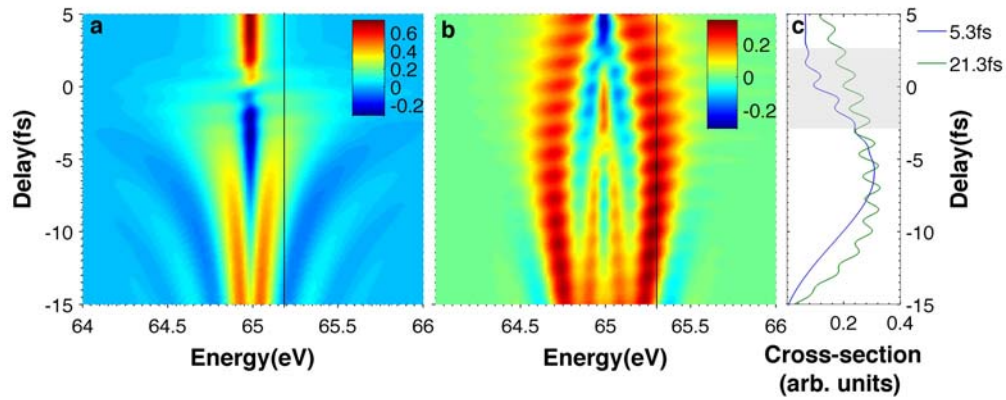


Fig. 4 CRW oscillation in the $4d_{5/2}^{-1} 6p$ absorption line for different IR pulse durations.

Calculations are done for (a) 5.3 fs (1.7 cycles) and (b) 21.3 fs (6.9 cycles) pulses. For both cases, the field strength is kept the same; namely, Ω/ω is the same. (c). The cut along a Aulter-Townes branch (indicated by the black lines in 4a and 4b) is shown. The amplitudes are re-scaled so that they are in the same range. The 2ω oscillation (period is ~ 1.6 fs) is clearly seen for both pulses in the overlapping region, which is highlighted in grey for the 5.3 fs pulse in Fig. 4c.

Appendix

EXPERIMENTAL SETUP

The atomic physics beam line at the Max Planck Center for Attosecond Science (MPC-AS) was used to carry out the measurements of Attosecond Transient Absorption spectroscopy (ATAS) in xenon. We use a carrier envelope (CE) phase stabilized Ti:Sa laser system to produce ultra-short pulses of about 28 fs and 0.9 mJ per pulse. This pulse is first broadened using super continuum generation and finally compressed to a pulse width of about 4 fs using chirped mirrors. The schematic of the beam line and absorption spectroscopy setup used in our measurements is shown in Fig 1. The few cycle pulse produced by the laser (4.2 fs, 800nm, 300 μ J) is focused on to neon gas source in a vacuum chamber to produce radiation in the XUV region by the process of higher harmonics generation (HHG).

The XUV beam along with the residual IR then co-propagates to the main experimental chamber. The IR and the XUV pulse are then separated using a special filter consisting of a plastic pellicle (15 μ m thickness) with a round metal filter (Al film) at the center. The annular IR beam and the XUV beam at the center then fall on a two segment curved mirror assembly. The inner segment of the mirror is a multilayered XUV mirror centered around 75 eV, having a bandwidth of about 15 eV and having a reflectivity up to 13 %. The annular shaped outer mirror is used to reflect the IR beam. The inner mirror can be moved relative to the outer mirror precisely using piezo electric stages for providing the delay between the pulses. The annular outer mirror is used to reflect and focus the IR pulse, which can be used for streaking measurements or transient absorption experiments as pump or probe pulse. The IR intensity at the focus can be controlled using an iris placed between the special filter and the two-segment mirror assembly. The intensity of the IR pulse is estimated using the focal spot, pulse width and the energy of the pulse, and accurate measurement of the intensity is measured by comparison of the experimental AC stark shift and the calculated shifts.

The two beams are then focused on to a quasi-static gas (xenon) cell, which is a Nickel tube of 3 mm diameter with a pinhole of \sim 100 μ m diameter providing an interaction length is about 2.4 mm. The xenon gas in the gas cell is controlled using a flow controller to maintain a pressure of about 10^{-3} mbar in the target chamber. The beams are then steered using a XUV mirror and focused at the entrance slit of XUV spectrometer using a spherical mirror. The IR part of the beam is cut off using a 0.15 μ m Aluminium filter, allowing only the XUV part (64-73eV) to the spectrometer. The XUV spectrometer consists of a slit, a spherical reflective grating (1200 lines/mm) and a X-ray CCD camera for imaging the spectrum. The spectrometer was used with XUV energies from 64-73eV corresponding to the energy range of absorption for xenon. The spectrometer resolution was measured to be about 500 meV by measuring the FWHM of the $4d_{5/2}^{-1} 6p$ absorption line at 65.1 eV.

EXPERIMENTAL DATA ACQUISITION

All the measurements were obtained by measuring the transmitted spectrum (I_{gas}) through the gas target (Xe) as a function of delay (τ) between IR and XUV pulses from delays of -10fs to 6fs (negative delays corresponds to XUV pulse arriving first) with a step of 200as. Each measurement corresponds to an average taken over \sim 9000 shots. The absorption spectrum is calculated using the reference XUV spectrum (I_0) taken in the absence of the xenon gas target. The optical density (OD) is calculated by using the following expression

$$OD(E, \tau) = \ln\left(\frac{I_{gas}(E, \tau)}{I_0}\right)$$

for each delay. The raw transient absorption spectrum is obtained by using the above expression. Such raw data contain the noises due to the fact that (1) transmitted and reference spectra were not taken simultaneously, and (2) there are always the fluctuations in transmitted and reference spectra. For the quantitative analysis and the extraction of the meaningful changes, these noises were filtered out by using Fourier method similar to one used by C. Ott et al. [28]. For each recorded XUV spectrum (containing absorption lines) at a given delay time, the spectrum was Fourier transformed and the components corresponding to the ‘slowly’ modulating ($\sim 3\text{eV}$ period) high-harmonic XUV spectrum were filtered out. This filtered data were then inverse Fourier transformed to get $I_{gas\ F}(E, t)$. This filtering procedure is done for all the delays for both the transmitted and reference spectrum. The transient absorption spectrum is reconstructed using these filtered spectra, according to the equation mentioned. As a result of this filtering procedure, the statistical noise of the two-dimensional absorption spectrum is significantly reduced (see Fig.A1).

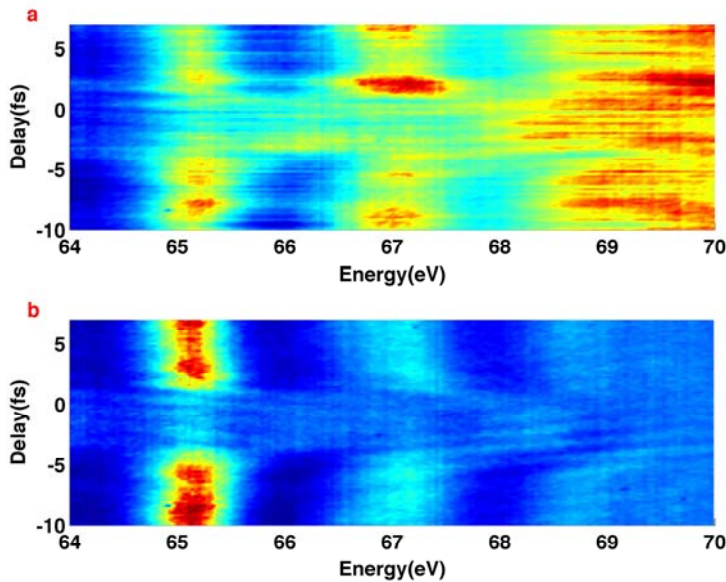


Fig. A1. (a). Raw transient absorption spectrum obtained for xenon and various delays between IR and xuv pulse
 (b) The spectrum obtained after smoothing and filtering the original spectrum shown in (a)