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Nonlinear Fourier-transform spectroscopy revealing wave-packet dynamics of D_2^+ with multicolor harmonic field

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Abstract. We report on a series of studies concerning D_2 molecules irradiated by highharmonic pulses generated from intense femtosecond laser pulses. The kinetic energy (KE) spectrum of dissociated D^+ ion fragments with a scanning delay between two replica harmonic pulses exhibits specific characteristics that are completely different from a conventional interferometric autocorrelation signal. We have successfully determined and separated three distinct ionization/dissociation processes by analyzing KE-resolved interferometric fringes by Fourier transform. We call this method for analyzing the KE spectrum of ion fragments "nonlinear Fourier-transform spectroscopy (NFTS)." NFTS provides us molecular information in stationary states because it is intrinsically a frequency domain analysis. Nevertheless, we have resolved the real-time evolution of the vibrational wavepacket of D^+ ions with a period of 22 fs by shortening the pulse duration of the fundamental laser pulse to 12 fs and extending the scanning delay range of two harmonic pulses to 150 fs. The probing process of the wavepacket can be described as a model with one-photon absorption of a multicolor harmonic field. We discuss a possible method of reconstructing the phase and magnitude of the wavepacket from the measured delay-KE spectrogram.

1. Introduction

New light sources always attract interest from researchers in the community of atomic/molecular physics because they are expected to reveal novel features of atoms and molecules. X-ray free electron laser (XFEL) facilities at present provide such novel light pulses and are providing many fruitful outcomes in physics, chemistry, and biology. This is due to the fact that XFEL light is basically coherent in the time and spatial domains, and it combines an extremely high photon energy (>keV) with a high pulse energy (> μ J) to induce or observe novel phenomena. We acknowledge the superiority of XFEL light in terms of photon and pulse energies. Nevertheless, high-order harmonic fields generated from an intense femtosecond laser pulse exhibit outstanding characteristics compared with XFEL light, such as spectral (or temporal) coherence between different harmonic fields. In other words, phase locking always holds among all harmonic fields.

We already proved this fact several years ago by demonstrating interferometric autocorrelation (IAC) measurement of an attosecond pulse train (APT) formed by the Fourier

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synthesis of high-order harmonic fields [1, 2, 3], as shown in Fig.1. We recorded the time-of-flight



Figure 1. Top: sequence of time-of-flight (TOF) mass spectra of N^+/N_2^{2+} with scanning delay between two replicas of multiple highorder harmonic fields. Bottom: averaged line profile of the far-side peak regions bounded with violet dashed lines (trace with crosses) and that of the near-side regions bounded with green dashed lines in the top panel image (trace with circles).

(TOF) mass spectra of N^+/N_2^{2+} ion fragments with a scanning delay between two replicas of multiple high-order harmonic fields, resulting in the figure shown in the upper panel of Fig.1. We can clearly see that the line profiles of TOF mass spectra, shown in the bottom panel of this figure, exhibit a train of humps with a period of 1.33 fs as a correlation envelope of the APT. Fine modulations superposed on the correlation envelope are the interference fringes of the high-harmonic fields, revealing the alternate constructive and destructive interferences at the tops of humps. We could not have observed these characteristics of the correlation trace without the multicolor coherence of high-order harmonic fields.

Another important feature was found in the line profiles at different TOF regions. The principal frequency of the interference fringes of the trace, depicted as a violet line with crosses in the bottom panel of Fig.1, was the carrier frequency of the 11th-order harmonic field, while that of the trace depicted as a green line with circles was the carrier frequency of the 9th-order harmonic field. We expected from this study that the IAC measurement might be useful for exploring the ionization/dissociation processes of molecules interacting with an APT field. This idea was examined by changing the target molecule from N₂ to $CO_2[4]$, then, we found that each IAC trace, obtained from the molecular ions of O⁺, CO⁺, C⁺, and $CO_2^{2^+}$, certainly exhibited distinct characteristics in the frequency components. We call this spectroscopic method utilizing the Fourier transform of IAC traces of an APT "nonlinear Fourier-transform spectroscopy (NFTS)." The NFTS of CO₂ was beneficial for qualitatively distinguishing the ionization/dissociation processes of each fragment ion. We could not, however, exactly specify relevant pathways of fragmentation only in stationary states because it is intrinsically a frequency domain analysis.

In this paper, we report on the feasibility of NFTS applied to the D₂ molecule[5]. We have succeeded in finding the three distinct ionization/dissociation pathways from D₂ to D⁺, owing to the fact that we can approximately consider only the excitation from the ground $(1s\sigma_g)$ electronic state to the first excited $(2p\sigma_u)$ states of D₂⁺ for the dissociation process following the ionization of the D₂ molecule with the one-photon absorption of XUV harmonic fields. This simplicity of the D₂ molecule and NFTS enable us to decompose a velocity map image (VMI)[6] of D⁺ into three VMIs representing three pathways. Afterwards, we demonstrate another study of D₂ molecules that follows an improved NFTS experiment with an increased delay scanning range and a reduced pulse duration of the fundamental laser pulse[7]. The dynamical evolution of a vibrational wavepacket of D₂⁺ is resolved into a two-dimensional spectrogram of the D⁺ yield with delay and kinetic energy coordinates. This result cannot be obtained without the complete synchronization or phaselocking of a multicolor harmonic field. We also discuss a possible method for reconstructing the phase and magnitude of vibrational states forming the wavepacket from the measured two-dimensional spectrogram.

2. Nonlinear Fourier-transform spectroscopy of D_2

2.1. Experimental setup

The experiment was implemented with a harmonic beam line consisting of a conventional chirped pulse amplification (CPA) system of a Ti:sapphire laser and a harmonic generator with a static gas cell. The CPA laser system delivered the fundamental laser pulse with a duration of 40 fs, an energy of 15 mJ, and a repetition rate of 10 Hz. The fundamental laser pulse was focused through a pinhole on the entrance side of a 10-cm long gas cell filled with Xe. In order to increase the pulse energy of the harmonic fields to the μ J range, the fundamental laser pulse was loosely focused with a 5m focusing lens[8]. The generated harmonic fields left via a pinhole on the exit side of the gas cell, then co-propagated with the fundamental laser pulse. A pair of Si harmonic separator mirrors set 4m away from the gas cell reflected the harmonic fields, while only a $\sim 10^{-4}$ fraction of the fundamental laser pulse were injected to the boundaries of these two mirrors, they were spatially split into two replicas after reflection. We could control the delay between the two replica fields by adjusting a translation stage on which one of the Si mirrors was mounted[10, 11].



Figure 2. Intensity distribution of harmonic fields at the focal point. Figure 3. Momentum distribution of D^+ .

The two replica beams consisting of the phase-locked harmonic fields from the 1st (fundamental) to 19th orders were introduced in a VMI chamber, and then focused into a molecular beam of D_2 using a concave mirror. The intensity of each harmonic field at the focal point of the concave mirror was estimated by measuring the 5th-19th harmonic spectra with an XUV spectrograph and evaluating the pulse energies of the fundamental, 3rd harmonic, and 5th harmonic fields with an energy meter. The resultant relative intensities of the harmonic fields are shown in Fig.2[4].

The ions yielded at the intersection of the focused beams and the molecular beam were accelerated with holed acceleration plates such that the radial position of a detected fragment ion at the microchannel plate (MCP) should be proportional to its velocity. The fluorescence from a phosphor screen set behind the MCP was relay-imaged onto a CCD camera. We extracted the VMI of D⁺ by subtracting the background image of the parent ions.

The momentum distribution of D^+ fragment ions was numerically retrieved from the measured image with a conventional inversion method[12]. A typical distribution of D^+ exhibited

three distinct crescent-shaped parts at peak KEs of 0.4, 1.4, and 2.6 eV, as shown in Fig.3. We scanned the delay between the two replicas of the harmonic beam and recorded the image of D^+ fragment ions at each delay with 1000-shot accumulation of the laser pulse.

2.2. Results

If the process for yielding D^+ fragment ions had mainly involved nonlinear interactions of highorder harmonic fields, the delay-KE spectrogram should have presented the characteristics of an IAC of an APT. Hence, we arranged the KE spectra, which were derived from angular integration of the retrieved momentum distributions, in accordance with the delay. The delay-KE spectrogram is shown in Fig.4. Contrary to our expectation, the KE-delay spectrogram of



Figure 4. Top: sequence of KE spectra of D⁺ with scanning delay between two replicas of high-order harmonic fields. Bottom: red, blue, and green curves exhibit averaged line profiles in region (A), region (B), and region (C), respectively.

 D^+ did not show repeated humps with a period of 1.33 fs originating from an APT envelope, but revealed modulations with the cycle periods of the fundamental field in the KE region of (A), the 3rd harmonic field in (B), and the 5th harmonic field in (C). It is reasonable to consider these modulations to be the interference fringes of the fundamental, 3rd harmonic, and 5th harmonic fields, respectively.

These features are completely different from those on the IAC image and the traces depicted in Fig.1, and the result seemed inconsistent with the basic principle of the autocorrelation measurement with two spatially split replica beams, because we could not observe the interference fringes without the correlation envelope of an APT under the condition that the correlation signal was yielded directly via two or more photon processes[3]. Otherwise, we could not find any delay-dependent signals due to the one-photon process. The only solution for this issue was that the D⁺ fragment ions had to emerge with a sequential process involving two photons of the harmonic beams focused into the D₂ molecular beam. We could expect that one of the two photons would be associated with the fundamental, 3rd-, or 5th-order harmonic field for each KE region because of the observed fringe period. We could also expect that the other photon might originate from the 11th- or a higher-order harmonic field since we noticed the difference (the 6th harmonic) frequency component between the 5th- and 11th-order harmonic fields by Fourier transform of the green trace in the bottom panel of Fig.4. The 8th harmonic peak, corresponding to the 11th minus 3rd harmonic frequency, also appeared in the Fouriertransform spectrum of the blue curve[5].

We note that the effective contribution of the 11th order harmonic pulse to the ionization originates from the large ionization crosssection of D_2 at the photon energy of the 11th order harmonic field as discussed in ref.[5]. This is also the reason the advanced pulse acts as a pump to create the vibrational wavepacket of D_2^+ and the delayed pulse acts as a three-color probe in the experiment mentioned in the next section, although the two replica pulses should be equivalent[7].

Consequently, we specified the most probable ionization and dissociation processes consistent with the experimental data. Please refer to Fig.6 in the next section showing the relevant potential energy curves of the electronic states of D_2 and D_2^+ related to this process. First, a D_2 molecule was ionized with one-photon absorption of the 11th- or higher-order harmonic field and promoted to the ground electronic state $(1s\sigma_g)$ of D_2^+ . Then, the electron in the $1s\sigma_g$ state was made to undergo a transition from the $1s\sigma_g$ state to the first excited state $(2p\sigma_u)$ by one-photon absorption of the 5th-order harmonic field. Since the potential energy curve of $2p\sigma_u$ against the nuclear coordinate is repulsive, the D_2^+ molecule dissociated. The transition from the $1s\sigma_q$ state to the $2p\sigma_u$ state is most likely to have occurred at the nuclear distance where the energy difference between the two states coincided with the photon energy of the 5th-order harmonic field. Thus, the peak of the kinetic energy release (=2KE) spectrum of D+D⁺ should have been located at the corresponding energy at that nuclear distance. The second excitation process was also induced with one-photon absorption of the 3rd-order harmonic and fundamental fields at different nuclear distances, resulting in different kinetic energy releases. The interference fringes of the correlation data originated from those appearing on the spatial profile of the focused beam. The detailed analysis of the fringes was reported in refs. [3] and [5].



Figure 5. Images of momentum distributions of D^+ with dissociation pathways with one-photon absorption of the fundamental (a), 3rd-order (b), and 5th-order (c) harmonic fields.

Since we specified the three dissociation pathways with one-photon absorption of three different laser fields, we could utilize the interference fringes to decompose the momentum image into three images in accordance with the dissociation pathways. We performed the Fourier transform of the correlation trace at each pixel on the measured image, and separately extracted the frequency component at the fundamental field or 3rd- or 5th-order harmonic field by applying a bandpass filter. Each image composed of filtered pixels was converted to a momentum distribution. We can clearly see decomposed images of momentum distributions in Figs.5(a), (b), and (c).

3. Real-time observation of vibrational wavepacket of D_2^+

Thanks to the NFTS analysis, we could elucidate the ionization and dissociation processes of D_2 molecules irradiated by APT harmonic and fundamental laser fields, as shown in Fig.6. We noticed from this energy diagram that a vibrational wavepacket in the ground state $(1s\sigma_g)$ of the D_2^+ ion should be created if the pulse duration of the high-order harmonic field ionizing the D_2 molecule is significantly shorter than the vibrational period of the wavepacket. Signals of the time evolution of the wavepacket might also be embedded in the KE spectrum of D^+ fragments by the scanning delay between the ionizing pulse (high-order harmonic field) and the



Figure 6. Energy diagram of the relevant electronic states of D_2 and D_2^+ . D_2 molecules are ionized with one-photon absorption of the high-order harmonic field and promoted into the ground electronic state $(1s\sigma_g)$ in the D_2^+ ion. The 5th- and 3rd-order harmonic and fundamental laser fields excite an electron to the repulsive state $(2p\sigma_u)$, resulting in dissociation.

excitation pulse (three-color field with a superposition of the fundamental, 3rd-order harmonic, and 5th-order harmonic fields) if the pulse duration of the excitation pulse is also sufficiently short.

The pulse duration of the fundamental laser field used in the experiment mentioned in the previous section was, however, 40 fs, which is much longer than the fastest vibrational beat period (21 fs) between v = 0 and v=1 in the $1s\sigma_g$ state. Therefore, we developed an advanced Ti:sapphire laser system that delivered 12-fs pulses with an energy of 40 mJ at a repetition rate of 100 Hz[13]. We also developed a high-order harmonic beam line in accordance with the loosely focused geometry of the fundamental laser pulse. The pulse durations of any order harmonic fields were expected to be less than 10 fs. The equipment other than the light source was the same as that used in the study described in the previous section.

As a consequence, we derived a delay-KE spectrogram with an extended delay range from 0 to 150 fs, as shown in the top panel of Fig.7[7]. We can clearly see intensity modulations in both the regions labeled (B) and (C) in this figure, the periods of which are much longer than the periods of the interference fringes of any light fields relevant to the D^+ yield. We also found that the KE spectrum in region (A) modulated with a period of 37 fs because a clear peak appeared at 36.5 THz in the trace of the Fourier transform (not shown) of the line profile in region (A), depicted as a red solid curve in the bottom panel of Fig.7. The periods (frequencies)



Figure 7. Top: Delay-KE spectrogram of D^+ fragment ion obtained by using sub-10-fs harmonic pulses. Bottom: solid lines are line profiles in the region (A) (red), region (B) (blue), and region (C) (green) of the spectrogram in the top panel. We also show traces calculated from a theoretical model as dashed curves.

of the modulations in the line profiles in regions (B) and (C) are 27 fs (37.6 THz) and 22 fs (44.5 THz), respectively. We conclude that these modulations express the real-time motion of the

vibrational wavepacket. The probed vibrational levels are selected with the photon energy and bandwidth of the excitation laser pulses, which are estimated to be v = 9-13 in region (A), v = 3-7in region (B), and v = 0-4 in region (C) by comparing the difference frequencies of these levels with the experimental results[7]. We do not find any notable vibrational frequency components in the KE region lower than 0.3 eV. The most probable candidate for this fragments yield is the direct two-photon ionization (harmonic photon + fundamental photon) to make a transition to the $1s\sigma_q$ state at the dissociation limit.

We can also find that the relative height of the peak in the region (B) compared with other peak heights in the regions (A) and (C) is much smaller than that shown in Fig. 4 (or Fig. 2(a) in ref.[5]). We suppose that the highest peak at region (B) in the previous experiment has been due to the high spectral density at a resonant vibrational level (v=3 or 4) of narrow spectral width of the 3rd-order harmonic field with a long pulse duration, while the simultaneous excitation of multiple vibrational levels at a moment (< 10fs) should reduce the transition amplitude to the $2p\sigma_u$ state.

4. Discussion: probing process of wavepacket

In the previous section, we have concluded, only from the reason that the modulation frequencies agree with the beat frequencies of the vibrational levels, that the delay-KE spectrogram in Fig.7 is deduced to be manifestation of the real time motion of the wavepacket. We discuss a physical model to simulate the delay-KE spectrogram. We modeled the excitation process from the $1s\sigma_g$ to $2p\sigma_u$ states with a dipole interaction of the three-color field as the following Schrödinger equation[14, 15, 16]:

$$i\hbar\frac{\partial}{\partial t}\begin{pmatrix}\psi_u(R,t)\\\psi_g(R,t)\end{pmatrix} = \begin{pmatrix}H_u(R) & \mu(R)E(t-\tau)\\\mu^*(R)E^*(t-\tau) & H_g(R)\end{pmatrix}\begin{pmatrix}\psi_u(R,t)\\\psi_g(R,t)\end{pmatrix},\tag{1}$$

where we define the Hamiltonians describing the nuclear motion in the $1s\sigma_g$ and $2p\sigma_u$ states as $H_{g/u}(R) \equiv -M^{-1}\partial^2/\partial R^2 + V_{g/u}(R)$, respectively, for the deuteron mass of M and the potential curves of $V_{g/u}(R)$. The nuclear coordinate is notated as R. The transition between the $1s\sigma_g$ and $2p\sigma_u$ states is governed by the off-diagonal elements of the Hamiltonian matrix on the right-hand side of the equation, which are formed as the product of the dipole moment, $\mu(R)$, and the three-color field, $E(t-\tau)$, with delay, τ , and its conjugate. The wave functions $\psi_g(R,t)$ and $\psi_u(R,t)$ give us the probability amplitude for nuclear motion in the $1s\sigma_g$ and $2p\sigma_u$ states, respectively. We assume that $\psi_u(R,t)$ should be equal to 0 at the initial time t = 0, whilst $\psi_g(R, t = 0)$ should be a projection of the vibrational ground state on the potential of the $X^1\Sigma_g^+$ electronic ground state of D₂. The dipole moment is assumed to be proportional to R for the simplicity [17]. The 3-color laser field is assumed to have Gaussian spectral shapes in the spectral regions of the fundamental, 3rd-, and 5th-order harmonic components with their pulse durations of 14 fs, 6.8 fs, and 5.5 fs, and the central photon energies of 1.54 eV, 4.62 eV, and 7.70 eV, under the Fourier limit condition, respectively.

The estimated intensity of the probe field is sufficiently low to apply the perturbative expansion for the solution of Eq. (1); thus, we adopted the first-order perturbative solution to simulate the experimental data. The dashed traces in the bottom panel of Fig.7 were calculated from this model. We are convinced that the experimental traces agree well enough with the calculated traces to express the real-time motion of the wavepacket.

Nevertheless, we are not satisfied with this analysis because we impose the initial wavepacket form (and the characteristices of the probe field) by an assumption. Even though the Franck— Condon principle is appropriate for the ionizing process from D_2 to D_2^+ , the wavepacket form, namely, the magnitude and phase of each vibrational eigenstate, should be uniquely determined by experimental data if we state that the wavepacket is measured. Such measurement for reconstructing quantum states from experimental data is significant for quantum physics and

its applications including quantum computing[18]. We can find many studies demonstrating quantum states with a tomographic method[19] or wavepacket interferometry[20, 21]. In this sense, we aim to specify the magnitude and phase of the vibrational wavepacket of D_2^+ from the measured delay-KE spectrogram. This is basically possible because the transition amplitude from the $1s\sigma_g$ state to the $2p\sigma_u$ state can be expressed as a simple analytical formula, which is similar to the formula describing an optical field amplitude measured in the frequency-resolved optical gating (FROG)[22] method. The FROG measurement is nowadays a versatile tool for characterizing the magnitude and phase of an ultrashort optical pulse from the picosecond to attosecond regime[23].

However, we have to solve some issues to apply the FROG method to the measured delay-KE spectrogram. We need a sufficiently wide scanning range of the delay to resolve each beat frequency between adjacent vibrational states. The energy resolution of KE should also be improved. We hope we will be able to retrieve an unknown D_2^+ wavepacket from a measured delay-KE spectrogram by fixing these issues in the near future.

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