Observing the attosecond dynamics of nuclear wavepackets in molecules by using high harmonic generation in mixed gases

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Abstract. We probe the attosecond dynamics of nuclear wavepackets in H₂ and D₂ molecules by measuring the relative phase of high harmonics generated in each molecule by using a novel method with a mixed gas of H₂ and D₂. We find that not only the single molecule responses but also the propagation effects of harmonics differ between the two isotopes and we conclude that in order to discuss the dynamics of molecules in the single molecule responses, the propagation effects need to be excluded from the raw harmonic signals. The measured relative phase as well as the intensity ratio are found to be monotonic functions of the harmonic order and are successfully reproduced by applying Feynman’s path integral method fully to the dynamics of the nuclei and electrons in the molecules.

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High harmonic generation (HHG) in atomic and/or molecular gases using ultrashort lasers is widely accepted as a robust method to obtain coherent attosecond pulses in the extreme ultraviolet and soft x-ray regions [1]–[8]. Recently, we introduced mixed gases as the nonlinear media for HHG for the first time and demonstrated destructive and constructive interference (DI and CI) of harmonics in a mixed gas of He and Ne [9] and dramatic enhancement (DE) of harmonics in a mixed gas of He and Xe [10]–[12]. Among these effects, DI and CI are the most fundamental processes of HHG in mixed gases; where the harmonic spectra from each gas overlap, they always take place according to the relative phase of the harmonics. By using this fundamental process, we demonstrated ultrabroadband measurement of the chirped harmonic phase and succeeded in observing its dependence on ionization potential [9].

The basic ansatz of the above measurement method is that harmonic phases as well as harmonic intensities should be determined by the underlying electron dynamics in the atoms and molecules. The electron dynamics are composed of three quantum transitions [13]–[15]: first, a part of the bound-state electron wavefunction tunnels through the potential barrier modified by the intense laser field, and appears in the continuum (step 1). The freed electron wavepacket is then driven by the laser field and, after the field reverses its direction, it has a probability of returning to the parent ion (step 2). A harmonic photon is emitted by the coherent oscillation between the returning electron wavepacket and the bound-state electron wavefunction (step 3). In the case of atoms with simple potentials, the electron dynamics are mainly determined by their ionization potentials \( I_p \) [9], [13]–[15], while in the case of molecules the structures of them should give additional effects [16]–[18]. Accordingly, if we measure the harmonic intensities and phases from a pair of atoms and/or molecules with the same \( I_p \), the net effects due to the structures of the molecules and their dynamics should be extracted [9].

As for harmonic intensities, Baker et al [19] compared the intensities of harmonics from jetted gases of H\(_2\) and D\(_2\) with the same \( I_p \), and concluded that the observed larger intensities of harmonics from D\(_2\) gases are due to the slower nuclear motion of each D\(_2\) molecule during the single molecule responses of HHG (figure 1(a)) [19]. In measuring the intensities, however, they ignored the differences in phase-matching effects of harmonics during their propagation, which can give measurable contributions to the observed harmonics [20]–[24]. Since it is a hard task to control the conditions of jetted gases and determine the phase-matching effects, it is experimentally important to investigate the phenomenon by using a gas cell, with which the conditions of gases can be controlled precisely. Furthermore, to investigate the phenomenon by measuring harmonic phases is a remaining goal. Since harmonic phases are directly connected to the action of the electrons and nuclei that determine their dynamics, the phase should encode the dynamics of nuclei in an essential way.

In this paper, we probe attosecond dynamics of nuclear wavepackets in H\(_2\) and D\(_2\) molecules by measuring the relative phase of high harmonics generated in each molecule by using a novel method with a mixed gas of H\(_2\) and D\(_2\). We find that not only the single molecule responses but also the propagation effects of harmonics differ between the two isotopes and conclude that in order to discuss dynamics of molecules in the single molecule responses, the propagation effects need to be excluded from the raw harmonic signals. In order to control H\(_2\) and D\(_2\) gases with different hydrodynamic properties under the same conditions precisely, we adopted a short gas cell (\( L = 1.5 \text{ cm} \)) rather than a gas jet for providing nonlinear media for HHG (SEM of pressure measurement \( \sim 0.2\% \)). The measured relative phase as well as intensity ratio are found to be monotonic functions of the harmonic order and are successfully reproduced.
Figure 1. (a) A schematic diagram illustrating the attosecond dynamics of the strongly correlated nuclear and electron wavepackets that lead to HHG in H₂ and/or D₂. When the internuclear distance is large, HHG is suppressed. (b) The integrated signals of harmonics from an H₂ gas (10.0 Torr, red squares), a D₂ gas (10.0 Torr, blue squares) and a mixture of the two gases (10.0 Torr, green squares) measured as a function of the harmonic order q. The signals are normalized to those from the H₂ gas and vertical errors represent SEM for 800 laser shots.

by applying Feynman’s path integral method [25, 26] fully to the dynamics of the nuclei and electrons in the molecules [9].

The experiment was carried out with a Ti:sapphire laser having a center wavelength of \(\sim 800 \text{ nm}\), a maximum energy \(\sim 200 \text{ mJ}\) and a pulse duration \(\sim 30 \text{ fs}\). The fundamental pulse was loosely focused with a lens (\(f = 2400 \text{ mm}\)) into the gas cell filled with a pure gas and/or a mixed gas, and phase-matched harmonics could be generated in the cell by balancing the Gouy phase and the negative dispersion of the gas medium. The generated harmonics were spectrally resolved with a flat-field grating and detected by a microchannel plate and a CCD camera [8]. The intensity of the pulses was estimated to be \(I_R \sim 2 \times 10^{14} \text{ W cm}^{-1}\), which is below the saturation of ionization of the molecules, ensuring that HHs were generated mostly at the peak of the laser pulse.

Figure 1(b) shows the integrated signals of harmonics from an H₂ gas (10.0 Torr), a D₂ gas (10.0 Torr) and a mixture of them (10.0 Torr; the mixing ratio = 1:1) measured as a function of the harmonic order q. As in the results of Baker et al [19], the observed harmonic intensity from the D₂ gas is significantly larger than that from the H₂ gas and the information on the relative phase of harmonics is encoded in that from the mixed gas [9]. The observed harmonic signals are, however, the integration of the single molecule response and the propagation effects of harmonics and the latter effects must be excluded to discuss the nuclear dynamics in the single molecule responses.

The propagation effects of harmonics, which are due to the two processes of the photoabsorption by each gas and the phase (mis-)matching between the harmonics and the fundamental pulse, can be determined by observing harmonic intensities as functions of the gas

\[\text{http://physics.nist.gov/PhysRefData/}\]
Figure 2. The measured harmonic intensities from (a) H$_2$ and (b) D$_2$ gases as functions of harmonic order $q$ and gas pressure $p$. The solid lines indicate the phase matching pressures, which maximize the harmonic intensities.

pressure [9]. Figure 2 shows the harmonic intensities from (a) H$_2$ and (b) D$_2$ gases as functions of harmonic order $q$ and gas pressure $p$. A significant difference can be seen between them, i.e. the responses in the propagation processes differ between H$_2$ and D$_2$ gases. In particular, the difference in the phase matching effects can be seen directly in the ridges in figure 2, which indicate the phase matching pressures $p_{PM}^{H_2}$ for example, for the 23rd harmonics, $p_{PM}^{H_2} \sim 10$ Torr and $p_{PM}^{D_2} \sim 12$ Torr, which indicate that the dispersion of H$_2$ is 1.2 times as large as that of D$_2$. Considering the accuracy of the pressure measurement (SEM $\sim 0.02$ Torr), this is the first clear observation indicating the differing phase matching effects between the two isotopes. Furthermore, this observation also indicates the other factor of different photoabsorption cross sections indirectly via the Kramers–Kronig relation and, in fact, the photoabsorption cross section of H$_2$ molecules is significantly larger than that of D$_2$ molecules in this spectral region [20]. Since the larger photoabsorption cross section of H$_2$ molecules by itself can explain the observed smaller harmonic signals from them qualitatively, one should be careful to evaluate the net magnitude of the enhancement.

Disentangling the single molecule responses and the propagation effects of harmonics is crucial, especially for measuring the relative phase $\Delta \phi(q)$ of harmonics. Ignoring the propagation effects of the harmonics leads to inconsistent solutions of imaginary $\Delta \phi(q)$. For intensity ratio $r(q)$, ignoring the propagation effects leads to significant overestimation of $r(q)$ (figure 1(b)). Figure 3 shows (a) the intensity ratio $r(q)$ in the single molecule responses and (b) the relative phase $\Delta \phi(q)$ as functions of harmonic order $q$. As demonstrated by the experiment in [9], the difference in excursion times between successive harmonic orders corresponds to $\sim$100 attoseconds and correspondingly the information on the nuclear dynamics is encoded in the harmonic order (superior axis in figure 3(b)). Considering that $r(q)$ and $\Delta \phi(q)$ are accumulated during the excursion of the electron, it is natural that they are monotonically increasing functions of the excursion time and thus the harmonic order. These results are well reproduced by our model described in the following except for the cutoff region, where the signals are weak and HHG are dominated by the interference between a pair of nearly equal excursion times [27].

This is confirmed directly by measuring the harmonic intensities as functions of the length of the media.

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Figure 3. (a) The measured (squares) and predicted (thick lines) intensity ratio \( r(q) \) between harmonics from \( \text{H}_2 \) and that from \( \text{D}_2 \) molecules as functions of harmonic order \( q \). \( r_{\text{QM}} \) and \( r_{\text{CM}} \) are the predictions with and without quantum effects for nuclear dynamics; see text. (b) The measured (squares) and predicted (thick line) relative phases between harmonics from \( \text{H}_2 \) and those from \( \text{D}_2 \) molecules as functions of harmonic order \( q \). Measuring the relative phases directly corresponds to observing the nuclear motions, i.e. the nuclear displacement of \( \text{H}_2 \) \( \Delta R_{\text{H}_2} \) (right axis) as a function of excursion time \( \tau \) (superior axis). For (a) and (b), vertical and horizontal errors represent SEM for 800 laser shots and and those from quantum mechanical uncertainty [19].

The best way to extract physics from the present experiment quantitatively is to apply Feynman’s path integral method [25, 26] fully to the dynamics of the nuclei and the electron. The action that determines HHG in a molecule \( i \) reads

\[
S_{\text{tot}}^i = S_{\text{free}}^i + S_{\text{e}^{-n}}^i + S_n^i + S_{\text{rad}} + S_{\text{int}}.
\]

Here, \( S_{\text{free}}^i + S_{\text{e}^{-n}}^i \equiv S_{\text{e}}^i \) is the action of the active electron used in Lewenstein’s model, which indicates that a few trajectories are responsible for HHG. Among them, the so-called short trajectory, which starts at time \( t_s' \) and ends at time \( t_s = t_s' + \tau_s \), can be selected by using propagation effects\(^4\). \( S_{\text{e}}^i \sim \int_{t_s'}^{t_s} d\tau I_p(R(\tau)) \) determines the interaction between the molecular ion and the electron, where \( I_p(R) \) is the ionization potential of the molecules as a function of internuclear distance \( R \) and was assumed to be constant in [27]. \( S_n^i = \int_{t_s'}^{t_s} d\tau [P^2/(2M') - V(R(\tau))] \) is the action of the nuclei with the reduced mass \( M' \), the canonical momentum \( P \) and the potential energy \( V(R') \). Finally, \( S_{\text{rad}} \) and \( S_{\text{int}} \) are the actions of the radiation oscillator and that of the interaction between matter and radiation field, respectively [25, 26].

When a harmonic photon is observed, the relevant quantum path of the active electron and nuclei in the molecules is determined. The nuclei can move after ionization due to their exceptionally light masses (figure 1(a)). Lein determines the potential \( V(R) \) for their dynamics, which can be approximated by the lowest potential of \( \text{H}_2^+ \) with a genetic algorithm (GA) to fit to the full numerical results [27]. In order to use GA, however, the solution space must be assumed by hand, and one of the advantages of our method is that such assumptions can be excluded.

\(^4\) For complex time, the integrand is extended to a complex function using analytic continuation.
Considering that the duration $\tau_s$ is short enough, the Feynman propagator $Z^i(\mathcal{R}, t_s; R, t'_s) := \int D\mathcal{P}D\mathcal{R} e^{i(S_{\mathcal{E}} + S_{\text{pot}})} \sim \int D\mathcal{P}D\mathcal{R} e^{iS_{\mathcal{E}}}$, which determines the quantum dynamics of nuclei, is given by [25, 26]

$$Z^i(\mathcal{R}, t_s; R, t'_s) = \left(\frac{M'}{2\pi i t_s}\right)^{1/2} \exp\left[i\left[\frac{M'(\mathcal{R} - R)^2}{2\tau_s} + f_0 \tau(R + R) + \frac{f_0^2 \tau_s^3}{24M'}\right]\right], \quad (2)$$

where $f_0 := -V'(R_0)$ with the equilibrium internuclear distance $R_0 := R(0)$. The initial state can be well approximated by $w^i(R, t_s) = N \exp[-(R - R_0)^2/(\sqrt{2} \delta R_0^2)]$, where $R_0 = 7.4 \times 10^{-2}$ nm $\equiv R_0$ and $\delta R_0^{D_1} = 7.8 \times 10^{-3}$ and $7.5 \times 10^{-3}$ nm express quantum fluctuations of the initial states. Then the probability density in the recombination is given by

$$|w^i(R, t_s)|^2 = N(1 + \tau_s^2/(\delta R_0^2)^4(M')^2)^{-1/2} \exp\left[-\frac{(R - R_0)^2/(\delta R_0^2)}{1 + \tau_s^2/(\delta R_0^2)^4(M')^2}\right], \quad (3)$$

where $R_{\text{cl}}(t_s) := R_0 + f_0 \tau_s^2/(2M') \equiv R_0 + \Delta R'(t_s)$ corresponds to the classical trajectory of uniformly accelerated nuclear motion. This evolution of $R_{\text{cl}}(t_s)$ results in the variation of electron dynamics that leads to HHG. The quantum fluctuation is given by $\delta R'(t_s) := \delta R_0^i(1 + \tau_s^2/(\delta R_0^2)^4(M')^2)^{1/2}$, which is due to the fluctuation at the initial state and quantum diffusion during the dynamics on the potential curve. This quantum treatment of the nuclear dynamics is crucial to the present phenomena as discussed later.

First, we treat the nuclear dynamics classically to clarify the quantum effects. For the $q$th harmonic generation, by using the steepest descent method, the induced dipole moment in a linear molecule whose axis is oriented at an angle of $\theta$ with respect to the $x$-axis of the laboratory system is given by

$$x_0^i(q) = \frac{2\pi i}{\sqrt{\det(S'_v)}} \left[\frac{2\pi}{it_s}\right]^{3/2} e^{-is_{\text{cl}}^i q \cdot \mathbf{d}'[\mathbf{p}', R_{\text{cl}}^i(t_s), \theta]} E(t_s') \cdot d[\mathbf{p} - A(t_s'), R_0, \theta]. \quad (4)$$

Here, $\mathbf{p}_s = \int_0^t dt' A(t')/\tau_s$ with vector potential $A(t')$ and $d[\mathbf{p}, R, \theta] := \langle \mathbf{p} | \hat{\mathbf{x}} | \psi_0(\mathbf{R}) \rangle$ is the bound-free dipole transition matrix element between the ground state $|\psi_0(\mathbf{R})\rangle$ and the continuum state $|\mathbf{p}\rangle$. We assume that the active electron in step 2 feels the homogeneous but time-dependent potential, $S'_v = S'_v e^{-n} + [\tau_s - 2(1 - \cos \tau_s)/\tau_s - C(\tau) \cos(2\tau_s - \tau_s)] U_p$ with the ponderomotive potential $U_p$ and $C(\tau) := \sin(\tau) - 4\sin^2(\tau/2)/\tau$. The determinant of the 2 × 2 matrix of the second derivatives of $S'_v$ with respect to $t_s$ and $t'_s$. Here, the parameter of electron acceleration [17, 18] is set to be $\xi = 0$, following the numerical model by Lein et al [28, 29]; $\mathbf{p}_s' = \sqrt{2q} (\mathbf{p}_s - A(t_s)) / |\mathbf{p}_s - A(t_s)|$.

For H$_2$ and D$_2$, the valence orbital 1s$_g$ is simply given by $\langle \mathbf{x} | \psi_{1g}(\mathbf{R}) \rangle = N' [\phi_{1s}(x + \mathbf{R}/2) + \phi_{1s}(x - \mathbf{R}/2)]$, where $\phi_{1s}(x)$ is the atomic 1s orbital in the configuration space, $N'$ is the normalization factor and $\pm \mathbf{R}/2$ are the positions of the nuclei. Then the dipole transition moments for H$_2$ and D$_2$ are given by

$$d_{1s_g}(\mathbf{p}, R, \theta) = N' \left[2i \cos(\mathbf{p} \cdot \mathbf{R}/2) d_{1s}(\mathbf{p}) - \sin(\mathbf{p} \cdot \mathbf{R}/2) \phi_{1s}(\mathbf{p}) \right], \quad (5)$$

where $d_{1s}(\mathbf{p})$ is the atomic dipole moment from the 1s orbital and $\phi_{1s}(\mathbf{p})$ is the 1s wavefunction in the momentum space. The first term in equation (5) is the product of two parts and its physical meaning can be understood by the simple two-point emitter model with $\mathbf{p} = \mathbf{p}_s'$. In fact, this term is zero when $\mathbf{p}_s' \cdot \mathbf{R}/2 = (n + 1/2) \pi$ $\Leftrightarrow R \cos \theta = (n + 1/2) \lambda_s$, where $n$ is an integer and $\lambda_s = 2\pi / \mathbf{p}_s'$ is the wavelength of the recombining electron with $\mathbf{p}_s'$. The physical meaning can be understood by the simple two-point emitter model with $\mathbf{p} = \mathbf{p}_s'$.
\[ \Theta := \arccos(p'_i \cdot R/p'_c R). \] The second term in equation (5), which is oriented along the molecular axis, corrects the error of the two-point emitter model [17, 18].

Considering that \( I^H_2 \sim I^D_2 \) and that the ionization rates at \( R = R_0 \) have little \( \theta \) dependence [30], the dominant difference should be due to the electron and nuclear dynamics in steps 2 and 3. The observed single molecule responses are obtained by superposing the radiation from all the molecules with random orientation coherently. For a linearly polarized laser field, whose direction is defined as the \( x \)-axis, the observed dipole moment is given by

\[ |\langle x_0(q) \rangle| \propto |\langle d^x_s(p'_s, R^i_{cl}(\tau_s), \theta) \rangle| = 2N' |\text{sinc}(p'_s R^i_{cl}(\tau_s)/2) d_{cl,1s}(p'_s)|. \]

Here, \( \langle \cdot \cdot \cdot \rangle \) shows the average over all directions and \( \langle \text{sinc}(p'_s R^i_{cl}(\tau_s)/2) \rangle \) can be neglected for small \( R \). Considering that \( |\text{sinc}(p'_s R^i_{cl}(\tau_s)/2)| \) is a decreasing function of \( R^i_{cl}(\tau_s) \) in \( p'_s R^i_{cl}(\tau_s)/2 \in [0, \pi] \), the intensity ratio \( r_{CM}(q) := |\langle x'^{D_2} \rangle/\langle x'^{H_2} \rangle|^2 \) from classical treatment of nuclei satisfies

\[ r_{CM}(q) \sim \left| \frac{\text{sinc}(p'_s R^{D_2}_{cl}(\tau_s)/2)}{\text{sinc}(p'_s R^{H_2}_{cl}(\tau_s)/2)} \right|^2 > 1. \]

The ratio explains, however, only \( \lesssim 30\% \) of the enhancement of HHG in \( D_2 \) \( (r_{CM}(q)) \) in figure 3(a): the remaining part is due to the quantum effects of nuclear dynamics.

In the full quantum mechanical frame, the valence orbital itself fluctuates due to the fluctuation of the nuclei, which forbids us to write \( R \) as arguments of states. Accordingly, the initial and final kets are given by \( |\psi_{iQ}\rangle|w_i^{(t_i)}\rangle \) and \( |\psi_{fQ}\rangle|w_f^{(t_f)}\rangle \) and after inserting the identity operator of \( I = \int_0^\infty dR |R\rangle\langle R| \) into equation (4), we obtain the final expression for the intensity ratio

\[ r_{QM}(q) = |s^{D_2}(q)/s^{H_2}(q)|^2, \]

where \( s^i(q) := \int_0^\infty dR w^i(R, \tau_s)^* w^i(R, 0)\text{sinc}(p'_s R/2) [19, 27] \) is the overlap integral. As can be seen in figure 3(a), the experimental results are successfully reproduced with \( r_{QM}(q) \), which is reasonably determined by \( R^i_{cl}(\tau_s) \) and the quantum fluctuation \( \delta R^i(\tau) \). However, \( r_{QM}(q) \) takes a rather complicated form and some assumptions such as GA [19] are needed to retrieve the nuclear motions.

On the other hand, as stressed in the introduction, the information on the dynamics of the nuclei is naturally encoded in the harmonic phases. In fact, measuring the relative phase of the nuclei is naturally encoded in the harmonic phases. In fact, measuring the harmonic phases [31–33], all of the attempts to observe molecular structures by using HHG in molecules were performed by measuring the harmonic intensities [16–19], [34]. Here, by measuring harmonic phases, we
presented a novel approach to observe the attosecond dynamics of molecular structures. The robustness and versatility of the present method will be demonstrated by applying it to other mixture systems with the same $I_p$ such as those of CO$_2$ and Kr [35].

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