Routes to Control of H₂ Coulomb Explosion in Few-Cycle Laser Pulses

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We have measured coincident ion pairs produced in the Coulomb explosion of H_2 by 8–30 fs laser pulses at different laser intensities. We show how the Coulomb explosion of H_2 can be experimentally controlled by tuning the appropriate pulse duration and laser intensity. For laser pulses less than 15 fs, we found that the rescattering-induced Coulomb explosion is dominated by first-return recollisions, while for longer pulses and at the proper laser intensity, the third return can be made to be the major one. Additionally, by choosing suitable pulse duration and laser intensity, we show H_2 Coulomb explosion proceeding through three distinct processes that are simultaneously observable, each exhibiting different characteristics and revealing distinctive time information about the H_2 evolution in the laser pulse.

An ultimate goal of investigating the interaction of molecules with short laser pulses is to provide insight into the molecular dynamics of molecules exposed to strong external fields. Pump-probe experiments have been widely employed to study the motion of vibrational and rotational wave packets in the presence of laser fields [1–5]. In these experiments, two pulses of tens or hundreds of femtoseconds durations are typically used, the first as a pump to initiate a certain reaction and later, (after tens or hundreds of femtoseconds) the second one to probe the reaction of the target to the first pulse. For H_2, the simplest of all molecules, most of the dynamics involving vibrational motion are expected to occur on a time scale of about 10 fs. This short time has made it hard to probe its dynamics with existing techniques. Pump-probe experiments on H_2 (D_2) are still scarce [6]. New efforts have been developed to produce attosecond pulses in order to probe dynamics on the subfemtosecond time scale [7–9]. An alternative approach has been to use a *molecular clock* [10,11] which ticks with the period of the optical cycle and which is "started" by a first ionization event and "stopped" by a second one. In this approach, the measured fragments' kinetic energies are used to read the internuclear distance of the wave packet as a function of time. This approach has been recently demonstrated by investigating the rescattering process for the dissociation and double ionization of H_2 (D_2) [10–12] and in the sequential ionization of D_2 by intense few-cycle laser pulses [13].

For the rescattering double ionization of H_2 (D_2) and its utilization as a molecular clock, the following scheme is used [12,14,15] (see Fig. 1): the clock is triggered when H_2 is first singly ionized near the peak field of an optical cycle. At this time, a vibrational wave packet as well as a correlated electron wave packet are launched. The electron released in the first ionization returns to the parent ionic core and excites the H_2^+ from its ground electronic state $1\sigma_g$ to the $1\sigma_u$ or $1\pi_u$ excited electronic states. Once

the H_2^+ is excited, the laser field readily ionizes it when it reaches the next peak. The return times are about 2/3 of an optical period for the first return and an additional half-integer multiple of the optical period for later returns [14,15]. The clock is read when the second electron is ionized. The internuclear separation of the wave packet is read through the internal kinetic energy release (KER) of the two released protons. From previous studies of this process, it has been shown that the first and the later returns may all contribute importantly to the dissociation and double ionization of H_2 [10–12], and in certain situations may produce comparable KER values which might blur the molecular clock's reading. However, it has been recently predicted [14] that the use of few-cycle laser pulses will reduce or even terminate the role of the
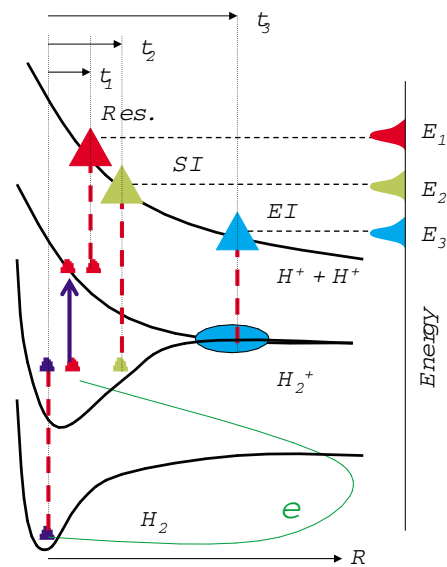

FIG. 1 (color online). Schematic diagram showing the different steps involved in the rescattering (Res) sequential ionization (SI), and enhanced ionization (EI) of H_2.

later returns and will give rise to better time resolution in reading the molecular clock.

In this Letter we show that the rescattering double ionization can be experimentally controlled by tuning the proper pulse duration and laser intensity. We present *return-selective* KER spectra for the rescattering-induced Coulomb explosion of H_2 at different pulse durations and laser intensities. For laser intensities $\leq 1.5 \times 10^{14}$ W/cm^2 and pulse durations less than 15 fs, we show that the first return dominates the production of the high-energy protons when H_2 is Coulomb exploded through a rescattering mechanism, with minor contributions from the later returns, while using longer pulses (30 fs) at relatively low intensities ($\leq 1 \times 10^{14}$ W/cm^2) will result in prominent contribution from the third return. Additionally, we find that we can adjust the pulse duration and the laser intensity in such a way that we see, in a single spectrum, H_2 Coulomb explosions proceeding through three different processes: enhanced ionization [16,17], sequential ionization [13,18], and rescattering-double ionization. The coexistence of these three processes with their individual features delivers a complete picture of the progression of H_2 in the laser field.

We used a mode-locked Ti:sapphire laser with a 790 nm center wavelength amplified to pulse energies up to 4 mJ at 2 kHz repetition rate with 30 fs pulse duration. The amplified pulse was spectrally broadened in a gas-filled hollow fiber and compressed down to 8 fs by means of several bounces on chirped mirrors after the hollow fiber [19]. Intermediate pulse lengths were controlled by inserting silica plates of various thicknesses in the optical path. An achromatic quarter wave plate placed in the way of a laser beam was used to switch from linear to circular polarization. The laser beam was focused with a 10-cm-focal-length on-axis spherical mirror onto a collimated supersonic gas jet. The laser peak intensity was determined by measuring the momentum distribution of the recoil ions in circularly polarized light [5,20]. To analyze the interaction products, we used a standard COLTRIMS arrangement [21,22]. The ions were extracted onto a time- and position-sensitive channel plate detector by a 20 V/cm electric field. The channel plate detector was followed by a multihit anode to measure the position of the extracted ions. From the times of flight and the positions, the full vector momenta of these ions were obtained.

Figure 2 shows a sliced density plot of the internal relative momenta P_{xz} of the two coincident H^+ ions in the center of mass frame produced when H_2 is Coulomb exploded by linearly polarized light of 8 fs duration and 1.5×10^{14} W/cm^2 peak intensity. The momentum image shown in Fig. 2 lies in the xz plane, where the laser propagates in the z direction, the jet moves in the y direction, and the collection electric field and laser polarization are in the x direction. By repeating the measurements using circularly polarized light at twice the
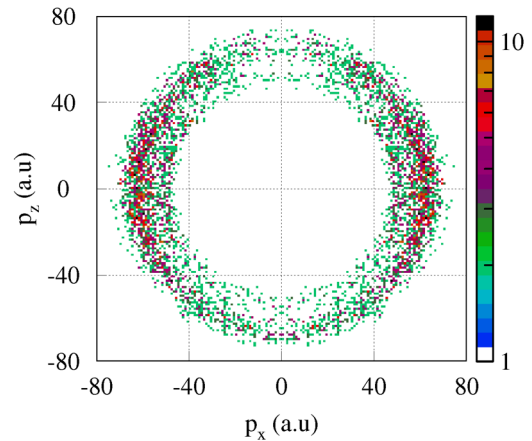

FIG. 2 (color online). Momentum image of the proton pairs produced in the double ionization of H_2 by 8 fs, 1.5×10^{14} W/cm^2 laser pulses.

peak intensity (to keep the peak field constant), we found that these ions disappear. This confirms that the origin of these ions is a rescattering process. The angular distribution shown in Fig. 2 displays how the rescattering mechanism depends much more weakly on the angle between molecular axis and the laser polarization vector (horizontal in this figure) than do the sequential-ionization and enhanced-ionization features (Fig. 4). This feature results from the fact that neither the first step (single ionization) nor the electron-impact excitation of the molecular ion is very sensitive to the molecular alignment with respect to the laser field. The radial distance in the plot represents the KER from the Coulomb explosion which mainly peaks around 15 eV. This energy value corresponds to a Coulomb explosion originating at an internuclear distance of 1.8 a.u.

Within the picture of the rescattering model (see Fig. 1), the electron released in the single ionization will first revisit the H_2^+ after 2/3 of an optical period (1.9 fs) from the moment it was born [14,15]. At this return, it may excite the H_2^+ to one of its excited electronic states from which it will readily be further ionized by the laser field. This sequence of steps will result in producing a pair of protons with KER ~ 15 eV, consistent with the findings in Fig. 2. The observation of such a nearly monoenergetic peak is attributed to the present experimental conditions. The few-cycle pulse has eliminated the fragmentation of the molecule through the enhanced-ionization process [16,17], and has clearly diminished the contribution of the later returns seen in Refs. [12,23].

To show the manipulation of the different rescattering returns and how the rescattering-induced Coulomb explosion can be controlled, we compare the KER spectra produced in 8 and 30 fs laser pulses with similar peak intensities. Figure 3 displays this comparison as measured (left column) and as predicted (right column) by the
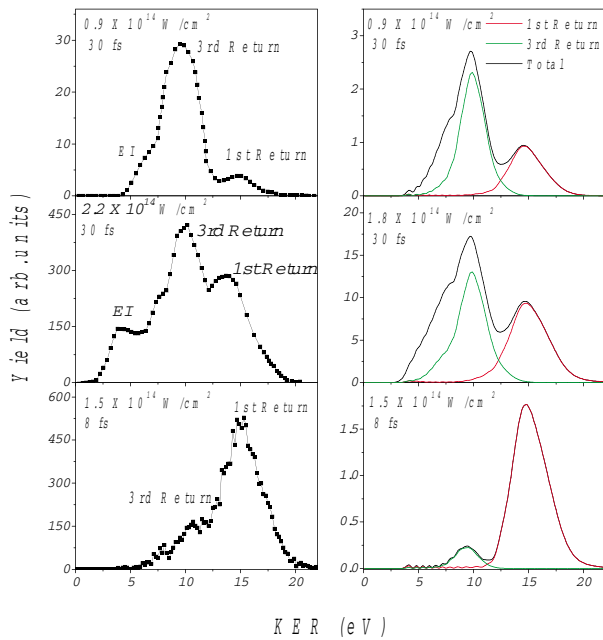
FIG. 3 (color online). Comparison of the measured rescattering KER spectra of H_2 in 30 and 8 fs laser pulses. The right column shows this comparison as predicted by the rescattering model [14,15].

rescattering model [14,15]. For 30 fs pulses and low peak intensity ($< 10^{14}$ W/cm^2), the KER spectrum has a major peak around 9.5 eV with no noticeable peaks elsewhere, as seen by Ref. [23]. As the laser intensity increases, the KER spectrum exhibits a broader energy distribution that extends to higher values while maintaining a relatively strong peak around 9.5 eV. In the few-cycle laser pulses (8 fs), the KER spectrum is prominently peaked around 15 eV with negligible yields at lower energies. These features can be explained as follows. For many-cycle laser pulses ($\sim$ 30 fs), the electron revisits the parent ion many times. The resulting collisional excitation may occur many cycles after the first ionization, and the removal of the second electron will take place at progressively larger internuclear separations. The excitation probability is determined by the returning electron energy (which is determined by the laser intensity) compared to the excitation energy of the molecular ion at certain internuclear separation. If the electron energy is lower than the excitation energy of the molecular ion at a certain nuclear separation, the rescattering-induced ionization will be suppressed until the vibrational wave packet moves to a large separation where the electron energy exceeds the excitation energy. This accounts for the single peak at the lowest intensity. For higher intensity, the electron will have sufficient energy to overcome the excitation energy at smaller internuclear distance as well. This will result in the broadening of the KER spectra toward higher KER (smaller internuclear distance), and, in particular, a feature due

the first return becomes strong. The peak at 9.5 eV corresponds to an internuclear distance of 2.8 a.u., close to the outer turning point of 3.2 a.u. where the vibrational wave packet spends a long time and many returns "pile up" (see Fig. 3 and Ref. [12]). This peak was previously observed [23] and has been recently recognized to be dominated by the third-return recollisions [12,14,15]. Meanwhile, in the few-cycle pulses (8 fs), the contribution of the later returns is turned off such that only the first return is effective, as is seen also in Fig. 2. It is important to mention here that for the longer laser pulses (30 fs), the contribution of the rescattering events was isolated by subtracting the yields obtained in circularly polarized light (no rescattering) at the same peak intensity [10–12] or by choosing large angles [12] to separate these events from those produced by enhanced ionization. In the case of the few-cycle laser pulses (8 fs), the rescattering events were directly measured with no further need for specific selection, since at the present laser intensity H_2 can be solely doubly ionized through the rescattering process.

It has been recently shown [13], that when the pulse rise time is sufficiently short ($\tau \le 15$ fs), Coulomb explosion of H_2 (D_2) may occur at small internuclear distance via sequential ionization, where both electrons are removed with high probability by the laser field within $\sim$3–6 femtoseconds. The measured KER spectrum produced by the sequential ionization can also be used to read the internuclear distance at different times in units of half optical cycle (see Fig. 1).

In Fig. 4 we show the momentum image (Fig. 4) generated when H_2 Coulomb is exploded by 14 fs, 3×10^{14} W/cm^2 laser pulses. This particular choice of the pulse duration and laser intensity was chosen to make the contributions of the three processes, rescattering, sequential ionization, and enhanced-ionization, comparable (the identification of these processes was done using their ellipticity sensitivity). Having these three processes present in the same spectrum allows the reading of the molecular clock to subfemtosecond accuracy over several optical cycles. The three distinct rings shown in Fig. 4 (the geometry is the same as Fig. 2) correspond to three different "reading times" or "ticks of the clock" at approximately 1.9, 4, and greater than 10 fs, each releasing ion pairs with a different KER. The three processes responsible for the Coulomb explosion are enabled by the first ionization of H_2 (see Fig. 1). Singly ionizing the neutral hydrogen will launch a wave packet on the ground state $1\sigma_g$ of H_2^+, starting the clock. The fastest double ionization process occurs when the molecular ion is ionized by the laser field after the electron released in the single ionization makes its first revisit and excites the molecular ion to one of its excited states. This rescattering process occurs after 1.9 fs from the first ionization and gives rise to the outermost ring (KER $\sim$ 15 eV) shown in
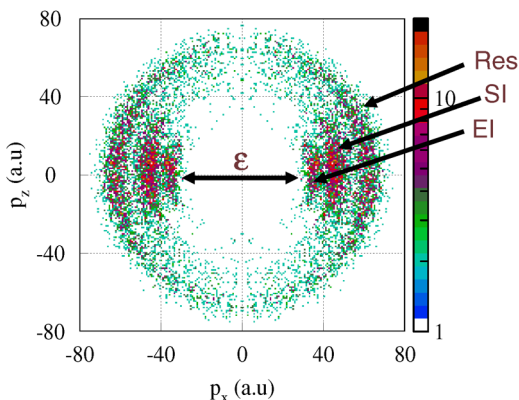
FIG. 4 (color online). Momentum image of the proton pairs produced in the double ionization of H_2 by 14 fs, 3×10^{14} W/cm^2 laser pulses. The coexistence of the rescattering (Res), sequential ionization (SI), and enhanced ionization (EI) delivers a complete picture about the evolution of H_2 in the laser field.

Fig. 4. If the H_2^+ is not ionized, its nuclear wave packet will continue to move to a larger internuclear separation where the ionization energy is smaller and where it faces the possibility of sequential ionization (see Fig. 1). When the initial ionization occurs via tunneling ionization on the rising side of the pulse envelope, at subsequent optical cycles the field strength may have increased to a level capable of tunnel ionizing the second electron as well. For the short pulse used here, this second ionization occurs ~4 fs after the first ionization, giving rise to the middle ring shown in Fig. 4 (KER ~ 10 eV). As discussed in Refs [13,18], this time is actually an average time resulting from sequential ionization at three, four, and five optical half-optical cycles after the first ionization, which occur at times of 4.0, 5.4, and 6.7 fs, respectively. A wave packet that survives both the above processes can finally escape the $1\sigma_g$ potential well via "bond softening," where it faces enhanced ionization when the molecular ion reaches a critical internuclear distance around 5.4 a.u. at a time of 10–12 fs.

In summary, we have shown that the rescattering-induced double ionization can be controlled experimentally by tuning the proper laser parameters. Choosing low laser intensities with longer pulses results in ionization only at the outer turning point, where the molecular ion requires the least energy to excite. Using higher intensities at the same pulse length shows that this peak

remains strong because of the pileup of contributions from many returns, but that other returns also contribute. However, the use of few-cycle (~ 8 fs) pulses eliminates all but the first recollision return and provides better resolution in the molecular clock reading. Moreover, we showed that by appropriately tuning the laser pulse length and intensity, it is possible to see in a single spectrum all three Coulomb explosion mechanisms, these being rescattering, sequential ionization, and enhanced ionization. The separation of these mechanisms is clearly made by nature, not by the experimentalist, and rather good understanding of all three is in hand.

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