A molecular rotation assisted non-sequential double ionization (MR-NSDI) mechanism is identified in the breakup of rotational \( \text{H}_2 \) molecules in a few-cycle intense laser pulse using a semi-classical trajectory Monte Carlo method. Applying a molecular source in an appropriate rotational state could intensively boost NSDI probability, and conclude with an additional small anti-correlated electron momentum distribution. It reveals the critical role of the continuous dynamics of molecular rotation, which assists the recollision process and subsequently releases the excited electron from the potential portal resulted from molecular rotation. Two underlying exit channels are found to contribute to MR-NSDI, i.e. tunneling ionization (dominated by enhanced ionization) and direct ionization. The two channels are confirmed to have different critical breakup internuclear distances. A prominent nuclear emission in \((-30^\circ, 30^\circ)\) along the laser polarized direction is identified to be a signature of MR-NSDI.

The study of atoms and small molecules in ultra-fast intense laser pulse has achieved great advances in the past decades \([1,3]\). For intense laser around \(10^{14-10^{15}} \text{W}/\text{cm}^2\), the strong interplay of Coulomb correlation effects and laser-induced effects leads to particularly complex ionizations, especially the well-known non-sequential double ionization (NSDI) \([4,11]\) and the frustrated ionization \([12,14]\). The underlying mechanisms of these processes, beyond the ability of single-active-electron approach, have been well addressed in the past two decades with the development of a large number of numerical methods accounting for the electron correlation \([2,15]\). In particular, the semi-classical strong field approximations \([16,19]\) and the semi-classical trajectory Monte Carlo (STMC) \([20,22]\) have provided the most straightforward interpolations and predictions of the attosecond dynamics in these phenomena, given the difficulties in performing a full quantum mechanical calculation \([15]\).

The study of NSDI reveals its fundamental mechanism to be a recollision process \([20]\), namely, the first ionized electron could, when the laser field reverses its direction, return to the parent ion and collide with the bound electron, leading to complex subsequent ionization. The dominant mechanisms for NSDI at different laser intensities and durations have been established to be the direct electron impact (EI) ionization, the slingshot NSDI \([27]\), and the recollision excitation with subsequent ionization (RESI) \([6,7,28,30]\). These understandings are mostly obtained in atoms considering a fixed nucleus.

For NSDI in molecules, the effects of multi-center nuclear potential gets involved. As the most simple two-electron molecule, \( \text{H}_2 \) is considered one of the best candidates for the insight into the fundamental mechanisms of laser-molecule interaction both on theory and experiment \([31]\). One of the well-known mechanisms in \( \text{H}_2 \) is the enhanced ionization \([1,8,32,34]\) induced by the electron tunneling from the upper potential well \([34]\) during the breakup of the molecule. In a perspective of the potential profile, the electron tunneling prefers an orientation away from the laser polarized direction. Meanwhile, ionization has been established to be very sensitive to molecular alignment \([35,38]\). An alignment away from the laser polarized direction could significantly reduce the NSDI probability in diatomic molecules, when the recollision process is highly depressed. It leads to a well-known contradiction in generating NDSI in \( \text{H}_2 \), namely, the recollision process prefers a molecular alignment along the laser polarized direction, while the electron is easier to be released away from the laser polarized direction, either by direct ionization or enhanced ionization (generally tunneling ionization \([34]\)). All these discussions, to date, consider the \( \text{H}_2 \) nuclear motion to be a mild dynamical contributor in NSDI, in particular the rotation being in a timescale of \(10^{-12}\) \([24,31,39]\).

In this letter, we show that the continuous rotational dynamics of \( \text{H}_2 \) molecule could intensively promote NSDI in a specific mechanism, termed molecular rotation assisted non-sequential double ionization (MR-NSDI). It is found that a proper molecular rotation could assist the recollision process and subsequently release the electron away from the laser polarized direction. In the experiment, using a strong field, molecules could be driven up to \( J \) (rotational quantum number) around a hundred \([40]\). Rotational molecule at this level could be also found in photochemical reactions and inelastic collisions \([41]\). Moreover, using an optical centrifuge, we could obtain a molecular source with \( J \) up to a few hundreds \([42,44]\). Therefore, we inspect the interaction of a linearly polarized few-cycle laser pulse with a rotational \( \text{H}_2 \) molecular source up to \( J=100 \).

We employ an STMC method \([45]\) in the tunneling regime \([20,39]\) following the well-established model in the simulation of NSDI and frustrated ionization \([21,22,30,46,47]\). We consider a linear polarized laser pulse in a Gaussian envelope along the lab \( z \)-axis as

\[
E(t) = E_0 e^{-2 \ln 2 \left( \frac{t}{\text{FWHM}} \right)^2} \cos(\omega t) z, \tag{1}
\]
where $t_{\text{FWHM}}$ is the full-width-half-maximum of the pulse duration. $\omega$ is the angular frequency. The atomic unit is used all through the paper unless indicated otherwise. To account for the focal volume effect (FVE) \cite{1, 48}, we consider the weight of a shell with an intensity $I$ in the laser spot as \cite{1, 42}

$$w_{\text{FVE}}(I) = \frac{1}{\lambda} \left( \frac{\pi w_{\text{FWHM}}^2}{2 \ln 2} \right)^2 \left( I_0 + 2I \right),$$

(2)

where $w_{\text{FWHM}}$ is the spot full-width-half-maximum. $\lambda$ is the wavelength. $I_0$ is the maximum intensity at the laser spot center.

For the initial conditions of the two electrons \cite{21, 22}, one ($e_1$) is started with an instantaneous tunneling ionization opposite to the field direction, the other ($e_2$) is assumed to be in a bound state described by a microcanonical distribution. For the tunneled $e_1$, the transverse velocity is described by a Gaussian distribution \cite{50}, while the parallel component is assumed to be 0. A rigid rotor approach is applied for the initial conditions of nuclei on the $z$-$\hat{z}$ plane with the angular frequency $\omega_{H_2} = \sqrt{J(J+1)/I_{H_2}}$, where $I_{H_2}$ is the moment of inertia. Assume $\theta^o \in (-180^o, 180^o)$ to be the angle of the internuclear axis with respect to $\hat{z}$-axis. The initial nuclear orientation $\theta^o_n$ is randomly and uniformly chosen in between. This assumption holds particularly for a short pulse as the laser-induced alignment before the tunneling ionization of $e_1$ is weak for $H_2$. It is worth noting that, even for $J=100$, the molecular rotation is in a timescale of $10^{-14}$s, much slower than that of vibration ($10^{-15}$s).

An adiabatic approach for the initial conditions of the two electrons is valid.

These initial conditions are integrated into an importance sampling process with a biased weight of each trajectory as $w^* = \Gamma(E(t_0), -\theta^o_n, I_1) \cdot w_{\text{FVE}}(I_0)$, where $\Gamma$ is the instantaneous semi-classical ionization rate \cite{51} for the initial tunneling of $e_1$. $E(t_0)$ is the field strength at initial time $t_0$. The importance sampling is performed for $t_0$, $\theta^o_n$, and $I_1$ by a Monte Carlo process \cite{49} using the maximum weight $w_{\text{max}} = \Gamma^{\text{max}} \cdot w_{\text{FVE}}(I_0)$, where $\Gamma^{\text{max}}$ is the maximum ionization rate obtained at $\theta^o_n=0$ and $I=I_0$.

The propagation of classical equations of motion is performed without approximation by a combination of the global regularization \cite{52, 53} and high precision step adaptive integration algorithms \cite{53}, e.g. the Bullish-store method and five-order Runge Kutta method, to take care of the Coulomb singularity and ensure high accuracy. Meanwhile, we account for the electron tunneling during the propagation (ETP) opposite to the laser field direction by the JWKB approximation with the tunneling probability \cite{21, 22, 54}

$$P_{\text{tun}} \approx \exp \left( -2 \int_{r_1}^{r_2} \sqrt{2(V(r)-V(r_1))} \, dr \right),$$

(3)

where $V(r)$ is the potential for the tunneling electron. $r_1$ and $r_2$ are the classical tunneling and exit points, respectively.

The NSDI events are collected according to the energy convergence of ionized electrons. The ionization time is decided by the time the electron energy keeps positive afterward. The NSDI probability could be calculated by $P_{\text{NSDI}} = N_{\text{NSDI}}/N_{\text{all}}$, where $N_{\text{NSDI}}$ and $N_{\text{all}}$ indicate the number of NSDI events and all events obtained, respectively.

As a verification of the method, we present a comparison of the simulation with a benchmark experiment for the NSDI of $H_2$ molecules in a 790nm ($\omega=0.05768$), 8fs laser pulse with an intensity $I_0=1.5 \times 10^{14}$ W/cm$^2$ (2000 fs). Panel (a) shows the KER results, while panels (b) and (c) present the internal relative momentum of the fragments $H^+$ obtained in experiment \cite{55} and simulation, respectively.

![FIG. 1. A comparison of the simulation with the benchmark experimental results redrawn from Ref. \cite{55} for the NSDI of $H_2$ in an 8fs laser pulse with an intensity $1.5 \times 10^{14}$ W/cm$^2$. Panel (a) shows the KER results, while panels (b) and (c) present the internal relative momentum of the fragments $H^+$ obtained in experiment \cite{55} and simulation, respectively.](Image)
The dashed lines are guides of eye. Panels (b-d) present the CEMS along $\hat{z}$-axis at $J=1.60$ as indicated in panel (a). The same axis ticks and labels are employed for (b-d). The large black dashed rectangles indicate the positions of $\pm 2 \sqrt{U_p}$. The small red dashed rectangles at $\pm \sqrt{U_p}$ emphasize the anti-correlated momentum pattern formed by MR-NSDI. Panel (e) illustrates the two critical instants in MR-NSDI. See the main text for more details.

FIG. 2. Panel (a) shows the NSDI probability as a function of $J$ with (●) or without ETP (■). The × indicates the normal NSDI probability as discussed in the main text. The dashed lines are guides of eyes. Panels (b-d) present the CEMS along $\hat{z}$-axis at $J=1.60$ as indicated in panel (a). The same axis ticks and labels are employed for (b-d). The large black dashed rectangles indicate the positions of $\pm 2 \sqrt{U_p}$. The small red dashed rectangles at $\pm \sqrt{U_p}$ emphasize the anti-correlated momentum pattern formed by MR-NSDI. Panel (e) illustrates the two critical instants in MR-NSDI. See the main text for more details.

much higher than the case when $H_2$ is pre-aligned along $\hat{z}$-axis ($2.4 \times 10^{-3}$) [49]. This high peak around $J=50 \sim 60$ contradicts the intuition that the rotation energy of nuclei leads to the increase of NSDI. It suggests the existence of a new mechanism that intensively promotes NSDI, beyond the effect of molecular alignment. An insight into the underlying mechanism reveals the essential role of the continuous dynamics of molecular rotation, thus termed MR-NSDI.

The mechanism of MR-NSDI is illustrated in Fig. 2(e) with two critical instants. After the tunneling ionization of $e_1$, the nuclei could approach $\hat{z}$-axis with the rotation of the molecule when $e_1$ is returning. It promotes the recollision probability, leading to the excitation of the bound electron $e_2$ as shown in the first instant. The subsequent molecular rotation and internuclear separation open an additional potential portal for the excited electron to escape as shown in the second instant. The molecular rotation at an appropriate speed could promote this process, leading to the peak profile in the probability distribution. Specifically, we consider the criteria (i) MR-NSDI: $\theta_\alpha^a \cdot \theta_\beta^a < 0$; (ii) the normal NSDI, namely, the molecular rotation does not across $\hat{z}$-axis: $\theta_\alpha^a \cdot \theta_\beta^a > 0$, where $\theta_\alpha^a$ is the $\theta^a$ at the final time. The proportions of the two cases gives (i) 6.3% (ii) 93.7% for $J=1$, and (i) 80.7% (ii) 19.3% for $J=60$. The probability of case (ii) as a function of $J$ is shown (×) in Fig. 3(a), exhibits a stable distribution. When ETP is turned off, the basic profile of the NSDI probability distribution (■) remains the same, while the maximum probability is reduced to $3.4 \times 10^{-3}$.

It comes from the existence of two different exit channels in MR-NSDI, namely, tunneling ionization ($\alpha$) and direct ionization ($\beta$), as indicated in Fig. 2(a). The $\alpha$ channel is dominated by enhanced ionization.

The potential profile resulted from molecular rotation in MR-NSDI allows an $\alpha$ or $\beta$ channel ionization of low-excited electrons from a relatively weak recollision. It leads to an additional small anti-correlated momentum pattern in $(-\sqrt{U_p}, \sqrt{U_p})$ as observed at the center (marked as red rectangles) of the CEMS in Fig. 2(b)(c). $U_p$ is the ponderomotive energy. It is distinct from the pattern of general NSDI (see Fig. 2(d)) for the CEMS of $J=1$ that mainly contributed by a mixture of EI and RESI. This anti-correlated momentum tends to be smaller at higher $J$, even beyond the generally allowed region of RESI, see the supplement for a transition of CEMS as a function of $J$. By restricting the $\theta^\alpha$ in a range away from $\hat{z}$-axis in case (ii), we could observe a normal correlated momentum pattern mainly formed by EI [49]. These NSDI events are generated by the recollision of the drifted $e_1$ with a large initial perpendicular momentum on $\hat{z}$-axis, thus in a low probability.

FIG. 3. Panels (a)(c) present two typical MR-NSDI events as a function of time (in laser period) for exit channel $\alpha$ and $\beta$, respectively. The $E_{e_2}$, $E_{e_2} - E_{e_2,f}$ (purple thin lines), $R_{ee}$, and $\theta_\beta^a$ (black dashed lines) are shown together with the laser pulse (grey bold lines) as indicated. The $R_{ee}$ and laser pulse are scaled 1/80 and 10 times, respectively, to fit the figure. Panels (b)(d) illustrate two refined exit pathways with a potential contour for channel $\alpha$ and $\beta$, respectively.

We show in Fig. 3(a)(c) two typical MR-NSDI events for the exit channel $\alpha$ and $\beta$, respectively. The $e_2$ energy $E_{e_2}$ (red lines), $E_{e_2} - E_{e_2,f}$ (purple thin lines), the electron distance $R_{ee}$ (blue lines), and $\theta_\beta^a$ (black dashed lines) are shown together with laser pulse (grey bold lines) as a function of time in laser period. $E_{e_2,f}$ indicates the $e_2$ energy in laser field. Two related refined exit pathways are
The time of tunneling is indicated as a thin arrow in panel (a), at which the $\theta^i$ is closed to 0 as the black arrows indicated, followed with the further increase of $E_{c2}$ until ionization. The recollision strength in channel $\beta$ is relatively stronger than that in $\alpha$, where the electron is mainly escaped by an exciton-tunneling process from the upper potential well resulted from the molecular rotation. The time of tunneling is indicated as a thin arrow in panel (a), at which the $E_{c2}$-$E_{c2f}$ line has a jump. The breakup internuclear distances of the two exit channels also exhibits a clear difference due to the different nature of the two exit pathways as shown in panels (b)/(d). Further analysis of the exit channels suggests the existence of a critical internuclear distance $R_c$ and a critical nuclear emission angle $\theta^i_n$ at the ionization time of MR-NSDI, as indicated in Fig. 2(e).

We present in Fig. 4(a) the internuclear distance $R_{nn}$ at ionization time as a function of $\theta^i_n$ at $J=60$, where $\theta^i_n$ is the $\theta^n$ at ionization time. $\theta^i_n$ is closed to $\theta^n$ (see the black dashed lines in Fig. 3(a)/(c) and the supplement for an example of $\theta^i_f$). The waves-like structure indicates the electron ionization around the peaks of different laser cycles. MR-NSDI events are concentrated around a rotation from -75° to 15° for clockwise molecular rotation, or 75° to -15° for anti-clockwise rotation [40]. This effect is even more pronounced for longer pulses (see supplement for a test of 16fs laser pulse [49]). At $J=60$, $\alpha$ channel mainly takes place around $R_{nn}$=8,13 and $\theta^i_n\in(30°,30°)$, while $\beta$ channel dominates $R_{nn}$=4,6 and $\theta^i_n\in(-30°,30°)$. Furthermore, we present in Fig. 4(b) and (c), for clockwise molecular rotation, the $\theta^i_n$ as a function of $J$ with or without ETP, respectively. The result for each $J$ is normalized to $P_e^{NSDI}$. Both cases show similar properties. A dominant nuclear emission window is observed for MR-NSDI as $\theta^i_e\in(0,30°)$ for clockwise molecular rotation. Panel (b) shows a more pronounced concentration around $\theta^i_n$ than panel (c). It is consistent with the dominant exit channel $\alpha$ in panel (b), where the enhanced ionization is more sensitive to the nuclear geometry. The most probable $\theta^i_n$ is increased from 0 at $J=1$ to roughly 15° around $J=50$, where the maximum NSDI probability takes place. Afterward, it gradually decreases. The increase of $J$ does not induce a continues increasing of $\theta^i_n$. It thus contradicts the intuitive hypothesis that the $\theta^i_n$ simply comes from the molecular rotation during the ionization time delay after the recollision. The further increase of $J$ beyond 60 relatively reduces the time window for recollision and subsequent exit channels, given the short duration of the pulse. Therefore, we obtain the overall dominant nuclear emission angle for MR-NSDI to be $\theta^i_e\in(-30°,30°)$. A molecular source with two imbalanced rotational components would lead to an asymmetric nuclear angular distribution. The $R_{nn}$ at ionization time as a function of $J$ with or without ETP are shown in Fig. 4(d) and (e), respectively. Electrons in $\alpha$ and $\beta$ channel tend to ionize at different critical internuclear distances around $R_{nn}^0\in(8,12)$ and $R_{nn}^0\in(4,7)$, respectively. They are consistent with the known results of enhanced ionization in H$_2^+$ [57, 58].

These MR-NSDI properties could be understood by a simple physical picture. We indicate in Fig. 4(a) (red solid lines) the region of $R_{nn}$ and $\theta^n$ with an enhanced ionization preferred potential profile (A) [34, 41] as the examples in panel (f) for $R_{nn}=12$, or a direct ionization preferred potential profile (B) as the examples in panel (g) for $R_{nn}=6$, at field strength $E=0.036$. $E=0.036$ is one of the laser peak strengths the $\alpha$ channel mainly takes place at $J=60$. The regions for $E=0.065$ are also shown with dashed red lines. The A and B regions are accord with the regions dominated by $\alpha$ or $\beta$ channels, respectively. See the supplement for a full analysis of five different types of potential profiles [40]. The potential profile, after the recollision, sweeps through the parameter space of $\theta^n$ and $R_{nn}$ during the molecular rotation and internuclear separation. The comparison of potential profiles at $\theta^n=0°$ and 15° in panels (f) and (g) shows how the ionization is more likely to take place when $\theta^n$ is away from 0°. It is also apparently easier for an electron...
to ionize from $\alpha$ channel than $\beta$, which is accord with the higher increased probabilities observed for $\alpha$ channel than $\beta$ in Fig.2(a). In the mean time, the $A$ and $B$ regions, although changed with laser field, is overall stable around each laser peak. They slightly tend to larger $R_{\alpha \beta}$ and $\theta^\circ$ with the decrease of field strength as the two examples for $E$=0.065 and 0.036 shown in Fig.4(a). As a result, the molecule with a proper rotational speed could, after the recollision, sweep through the “best” region for enhanced ionization or direct ionization, which explains the substantial increase of NSDI probability, the nuclear emission angle in $(-30^\circ, 30^\circ)$, and the different breakup $R_{\alpha \beta}$ in $\alpha$ and $\beta$ channels. It is worth noting that, for initially aligned $\text{H}_2$ molecules, the NSDI probability will fast decay with the increase of $\theta^\circ$, since the recollision is depressed. Therefore, a prominent nuclear emission in $(-30^\circ, 30^\circ)$ could be a signature of MR-NSDI, see the supplement for a comparison of the final nuclear momentum distribution at $J$=1 and 60 [49].

In conclusion, with an inspection of the rotational $\text{H}_2$ molecules in a few-cycle intense laser pulse, we have identified a new NSDI mechanism, MR-NSDI. In MR-NSDI, the continuous molecular rotation assists the electron recollision and subsequently release electrons from two ionization channels away from the laser polarized direction. MR-NSDI could intensively promote the NSDI probability and concludes with a small anti-correlated pattern at the center of CEMS. Moreover, the nuclear emission in MR-NSDI forms a prominent distribution in $(-30^\circ, 30^\circ)$, while two different critical breakup internuclear distances are confirmed for the two channels. This study reveals the important role of continuous nuclear dynamics in ultrafast laser-driven ionization, which was normally underestimated. It is fundamentally essential for the further study of ultrafast laser-excited molecule interaction and laser control of molecular fragmentation and chemical reactions [52, 60].

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