

Muonic Molecules in Superintense Laser Fields

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We study theoretically the ionization and dissociation of muonic molecular ions (e.g., $dd\mu$) in superintense laser fields. We predict that the bond breaks by tunneling of the lightest ion through a bond-softened barrier at intensity $I \geq 10^{21}$ W/cm². Ionization of the muonic atomic fragment occurs at much higher intensity $I \geq 6 \times 10^{22}$ W/cm². Since the field controls the ion trajectory after dissociation, it forces recollision of a $\sim 10^5$ – 10^6 eV ion with the muonic atom. Recollision can trigger a nuclear reaction with sub-laser-cycle precision. In general, molecules can serve as precursors for laser control of nuclear processes.

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Nuclear fusion occurs during the collision of selected isotopes of hydrogen ions with relative energy $\sim 10^4$ – 10^5 eV. High-energy ions occur naturally in high-power laser clusters or laser-plasma interactions [1–3]. However, all processes demonstrated so far are the result of random collisions. We show that the coherent oscillation of an ion in an intense laser field can be harnessed to stimulate nuclear reactions, allowing nuclear decay to be timed and optically controlled. We use muonic molecules to establish favorable initial conditions. Since their internuclear distance is very small, a muonic molecule can survive much stronger fields than traditional molecules.

Our work is related to laser control of electrons in atomic media. At $I > 10^{14}$ W/cm² electron dynamics is also dominated by the recollision between a newly ionized electron and its parent ion [4,5]. The recollision electron can collide with its parent ion with energy as high as $3.17 U_p$ ($U_p = e^2 E_0^2 / 4 m_e \omega^2$) where e and m_e are the electronic charge and mass, E_0 is the laser field strength, and ω is the angular frequency of the optical beam. During this recollision the electron can elastically or inelastically scatter from its parent atom, or recombine [4]. Controlling the electron by the amplitude, phase, and frequency of the laser field controls all of these fundamental processes [6] leading to, for example, tuning of high harmonic radiation [6,7] and to attosecond pulses [6,8].

To extend recollision from electrons colliding with their parent ion to ions recolliding with an adjacent nucleus and initiating nuclear reaction, two conditions must be met. First, we must fix the positions of nucleons in space before the process is initiated. This is achieved by using a molecule as a precursor. Second, we must use very high laser intensities to control the trajectories of heavy (charged) particles and to achieve kinetic energies in the range of 1 MeV. This requires that the molecule be stable in the strong field and/or that the field be applied more rapidly than the molecule dissociates. We achieve these

aims by using muonic molecules. We show that high-order multiphoton processes that occur at $\sim 10^{14}$ W/cm² in $D_2^+(dde^-)$ (such as ionization and bond breaking [9,10]) occur at $\sim 10^{22}$ W/cm² in $dd\mu^-$. When a bond breaks at these extremely high intensities, the deuteron can be accelerated to $\sim 10^5$ – 10^6 eV and recollides with a $d\mu^-$ atom with enough energy to stimulate fusion.

The physics of muonic hydrogen molecules has been widely studied for muon-catalyzed fusion [11,12]. Simple scaling laws relate the properties of muonic hydrogen to traditional atomic and molecular physics. Since the muon mass is ~ 200 times larger than the electron mass, the muonic Bohr radius a_μ is ~ 200 times smaller than the atomic Bohr radius $a_{at} = \hbar^2 / (m_e e^2)$ (\hbar is the Planck constant) and the muonic atom ionization potential is ~ 200 times larger. Similarly, by replacing the electron by a muon in and its isotopomers, we get muonic molecules with ~ 200 times deeper potential well and 200 times smaller equilibrium internuclear separation. At such small internuclear distances, fusion occurs with significant probability [11,12]. The lifetime of the muon is $\tau_\mu = 2.197 \times 10^{-6}$ s, which is a much longer time than the fusion time in muonic molecule [11], and is also much longer than the duration of currently available intense laser pulses, $\tau_p = 5$ – 50 fs.

The dynamics of the muonic atom in intense laser fields is described by the time-dependent Schrödinger equation. In scaled form,

$$i \frac{\partial}{\partial t'} \psi(z', t') = \left(-\frac{1}{2} \Delta - \frac{1}{r'} + z' E'(t') \right) \psi(z', t'). \quad (1)$$

In Eq. (1) the length, energy, time, field units are

$$\begin{aligned} a'_\mu &= \frac{\hbar^2}{m'_\mu e^2} = a_{at} / \alpha E_\mu = \frac{e^4 m'_\mu}{\hbar^2} = E_{at} \alpha, \\ \alpha &= \frac{m'_\mu}{m_e} t_\mu = \frac{\hbar}{E_\mu} = t_{at} / \alpha, \quad E_\mu^{\text{field}} = E_{at}^{\text{field}} \alpha^2, \end{aligned} \quad (2)$$

where E_μ and E_{at} are muonic and atomic energy units and

E_{μ}^{field} , $E_{\text{at}}^{\text{field}}$ are muonic and atomic field units. In Eq. (2) the electron mass present in atomic units is replaced by the muon reduced mass $m'_{\mu} = m'_{\mu} m_N / (m'_{\mu} + m_N)$. Knowing the atomic electronic ionization rates R_I^{at} at laser intensity I and laser frequency ω , we can predict the rate for muonic systems, at intensity I and frequency ω , using the following rules: $R_I^{\mu} = R_I^{\text{at}} \alpha$, $\omega' = \omega \alpha$, $I' = I \alpha^4$, where $\alpha = m_{\mu} / m_e = 185.8$ for a μp atom, 195.74 for μd , and 199.274 for μt . At 800 nm (1.55 eV), the muonic atom is described by quasistatic models [13] since the photon energy is well below the atom's ionization potential $I_p = \alpha \times 13.6$ eV. As shown in Fig. 1, tunnel ionization in muonic atoms only becomes important for $I \sim 5 \times 10^{22}$ W/cm². At $I = 6 \times 10^{22}$ W/cm² the ionization rate for μp is 10^{13} s⁻¹, and for μd it is 10^{12} s⁻¹.

These scaling rules can also be used for molecular ions at fixed internuclear distance $R' = R/\alpha$. Consequently, all processes familiar from strong field molecular physics [14] should occur in muonic molecules. Just as in electronic molecules, for muonic molecules at their equilibrium position, the ionization rate is much smaller than atomic rates since the ionization potential is much larger. Therefore, dissociation via bond softening and tunneling occurs before ionization. As the molecule dissociates, it passes through a critical internuclear separation where the ionization rate peaks [15,16]. In [16], the enhancement is about an order of magnitude larger than atomic rates. Thus Fig. 1 implies that the ionization rate reaches $\sim 2 \times 10^6$ s⁻¹ at intensities of 1.7×10^{22} W/cm². We will now show that dissociation via bond softening dominates.

Figure 2(a) illustrates bond softening of the ground state potential (Σ_g) for $dd\mu$ in a dc electric field corresponding to the indicated laser intensity $I = cE_0^2/(8\pi)$. The potentials were calculated by diagonalizing the Σ_g

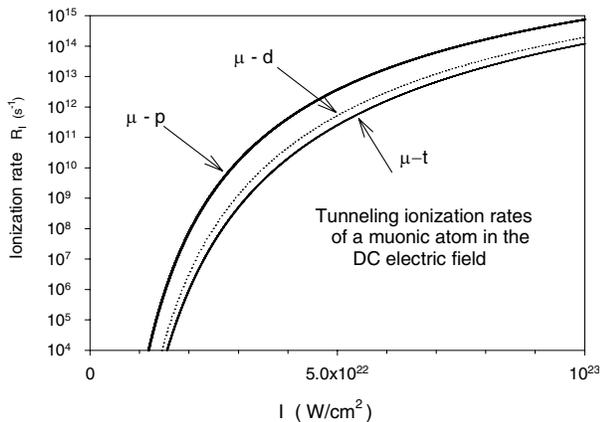


FIG. 1. Ionization rates of muonic atoms, obtained from tunneling electronic rates [18] by replacing the electron mass by the muon reduced mass m' , plotted as a function of the peak laser field. Since laser photon energies are low compared to I_p , the rate is independent of the laser frequency.

and Σ_u potentials coupled via $eR/2$ [17]. The threshold for overbarrier dissociation for the $v=0, J=0$ state of $dd\mu$ occurs at $I_{\text{thr}} = 2.3 \times 10^{22}$ W/cm² whereas for a $pp\mu^-$ molecule (not shown), this threshold occurs at $I_{\text{thr}} = 10^{22}$ W/cm². However, because the barrier is ~ 200 narrower, dissociation occurs via tunneling at much lower intensity than I_{thr} as we now show by solving the one-dimensional coupled time-dependent Schrödinger equations for $dd\mu$:

$$i\hbar \frac{\partial \psi_g}{\partial t} = -\frac{\hbar^2}{2m} \frac{\partial^2}{\partial R^2} \psi_g + V_g(R) \psi_g + \kappa |e| E(t) \frac{R}{2} \psi_u,$$

$$i\hbar \frac{\partial \psi_u}{\partial t} = -\frac{\hbar^2}{2m} \frac{\partial^2}{\partial R^2} \psi_u + V_u(R) \psi_u + |e| \kappa E(t) \frac{R}{2} \psi_g, \quad (3)$$

$$\kappa = 1 + \frac{m_{\mu}}{m_{\mu} + m_N}.$$

In Eq. (3) $\psi_g \equiv \psi_g(R, t)$ and $\psi_u \equiv \psi_u(R, t)$ are nuclear wave functions corresponding to the gerade and ungerade

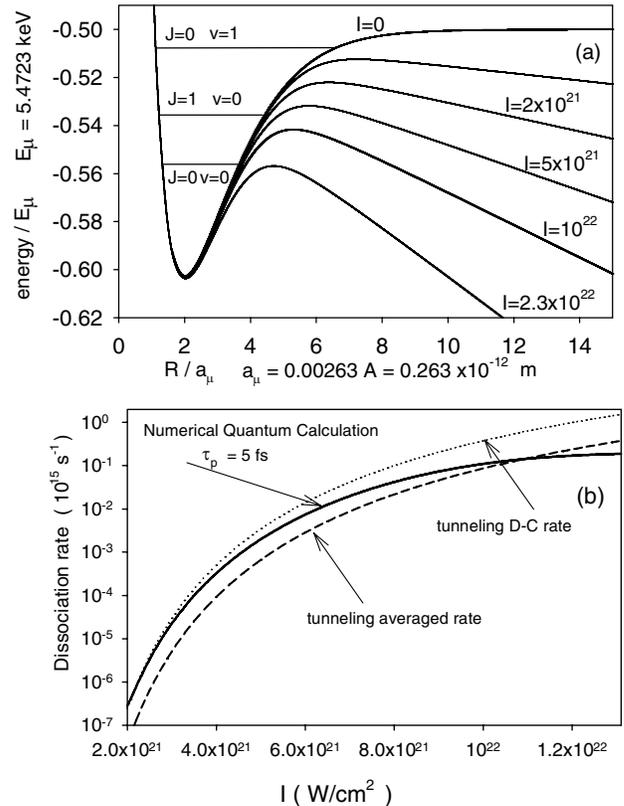


FIG. 2. (a) The potential energy surfaces of a $dd\mu$ molecular ion in a range of static electric fields plotted as a function of the same electric field amplitude. (b) The dissociation rates (solid line), defined as P_{diss}/τ_p ($\tau_p = 5$ fs, $\lambda = 800$ nm), where P_{diss} is the dissociation probability at the end of the laser pulse, calculated from the time-dependent Schrödinger equation, are compared with tunneling rates (dotted line) in the dc field rate, and with the cycle averaged tunneling rate [18] (dashed line).

potentials V_g and V_u , and m is the dd reduced mass. The solid curve in Fig. 2(b) plots the 1D dissociation rate as a function of the peak laser intensity calculated using Eq. (3) for a 5 fs (full width at half maximum) 800 nm pulse. The dissociation energies $E_{\text{diss}}(J=0, \nu=0)$ of $dd\mu$ are 325 eV [11] and the deuterium ponderomotive energy in the center of mass system $U_p = e^2 E_0^2 / (16m^2 \omega) = 81.4 \text{ keV}$ at $I = 5 \times 10^{21} \text{ W/cm}^2$. Therefore, the corresponding Keldysh parameter for dissociation is $\gamma = \sqrt{I_p / 2U_p} \ll 1$. Thus, we can adapt the three dimensional tunnel ionization rates [18] to dissociation by replacing the atom ionization potential I by E_{diss} and the electron mass by molecular reduced mass yielding the tunneling dissociation rates:

$$w_{\text{diss}} = \frac{eE_{\text{diss}}E_{\text{unit}}^{\text{field}}}{\hbar E(t)} \exp\left(-\frac{2E_{\text{unit}}^{\text{field}}}{3E(t)}\right), \quad (4)$$

$$E_{\text{unit}}^{\text{field}} = \frac{2\sqrt{m}}{e\kappa\hbar} [2E_{\text{diss}}]^{3/2},$$

The dotted curve in Fig. 2(b) is obtained from Eq. (4) where the dc electric field is associated with the peak laser field. The broken line corresponds to the dc rates averaged over one cycle. While tunneling plays a small role in dissociation via bond softening in electronic molecules, it is essential in the dynamics of a muonic molecule in a laser field due to the much narrower tunneling barrier. Because of such a strong tunneling effect, the dissociation channel dominates; one needs much higher intensity to break the $dd\mu$ molecule into three fragments, e.g., at $I = 10^{22} \text{ W/cm}^2$ the dissociation rate is 10^{14} s^{-1} whereas the $d\mu$ atom ionization rate is 10^4 s^{-1} .

We also use Eq. (3) to estimate the 1D probability $P(t)$ of finding both nuclei within the radius $R < R_0 = 66 \times 10^{-15} \text{ m}$:

$$P(t) = \int_0^{R_0} dR [|\psi_g(R, t)|^2 + |\psi_u(R, t)|^2]. \quad (5)$$

We plot $P(t)$ and the electric field $E(t)$ in Fig. 3 for a 200 nm pulse (period, $T = 0.66 \text{ fs}$), intensity $I = 2 \times 10^{22} \text{ W/cm}^2$, and for a 400 nm, $I = 10^{22} \text{ W/cm}^2$ pulse. The figure shows that the laser pulse enhances the probability of finding the deuterium ions close together. The enhancement occurs at well defined times delayed following the peaks of the laser field by $\sim 0.7 T$, lasting $\sim 1/4$ laser period and separated by $1/2$ laser period. This is similar to electron-ion recollision during atomic and molecular ionization [4,5]. Since we can control nuclear position, we can stimulate nuclear processes with attosecond precision. We now estimate the 3D probability of fusion in $dd\mu$.

Since tunneling formula can be adapted from ionization to dissociation, the wave packet spread that tunneling imposes in the recollision problem [4,19] can also be adapted. In the electronic ionization of an atom, the

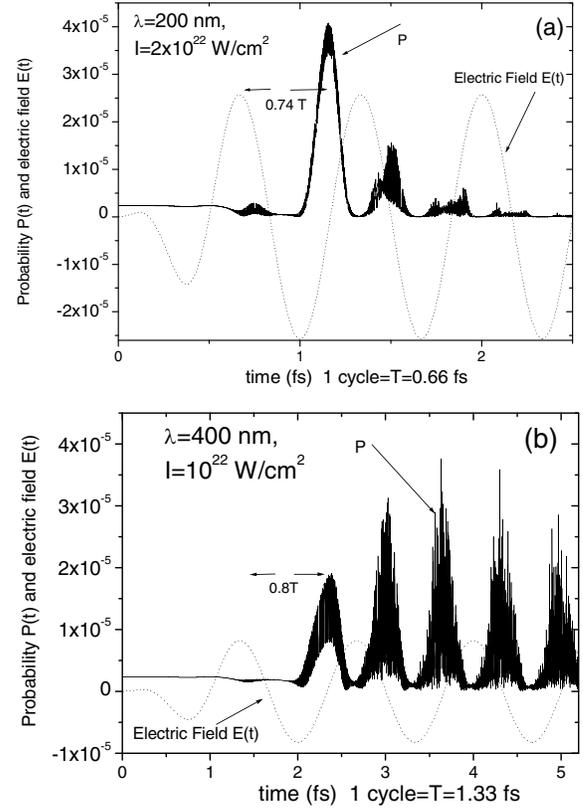


FIG. 3. The return probability $P(t)$ defined by Eq. (5) is plotted together with the shape of the laser electric field for a 200 nm ($I = 2 \times 10^{22} \text{ W/cm}^2$), and for an 400 nm pulse ($I = 10^{22} \text{ W/cm}^2$). The laser field is plotted in arbitrary units.

lateral momentum p_T distribution is [19]

$$w_{\text{el}}(p_T) = w(0) \exp(-p_T^2 / p_{0-\text{el}}^2), \quad (6)$$

$$p_{0-\text{el}}^2 = e\hbar E_0 \sqrt{\frac{m_e}{2I_p}},$$

where I_p is the atoms ionization potential and E_0 is the maximum value of the laser electric field. We obtain the lateral momentum width p_0 in muonic molecule by replacing I_p in Eq. (6) by the dissociation energy E_{diss} , and E_0 by $\kappa E_0 / 2$ (since molecular coupling is asymptotically $\kappa ER / 2$ in a $dd\mu$ molecule, $\kappa \sim 1$), and the electron mass m_e by the molecular reduced mass m . The momentum width p_0 in a $dd\mu$ molecule is $p_0^2 = e\hbar E_0 \sqrt{m / (2E_{\text{diss}})}$, which leads to spatial lateral spreading of the wave packet $x_T = p_0 t / m\hbar$. For $I = 10^{22} \text{ W/cm}^2$, $t = 0.7 T$ (T is the cycle of a 800 nm laser), $m = 1835m_e$, $E_{\text{diss}} = 325.074 \text{ eV}$, we get $x_T = 1.08 \text{ \AA}$. This spread is thus 205 larger than the size of the muonic molecule. By contrast, in atomic ionization the spread of an electronic wave packet is $x_T \sim 10\text{--}20 \text{ \AA}$ at $I = 10^{14} \text{ W/cm}^2$, or 20–40 times the atomic orbital dimensions.

We now estimate the upper limit for (3D) laser-induced fusion probability and rate. Assuming $dd\mu$ dissociates almost completely in one half laser cycle, about 50% of the fragments recollide in a duration of $\tau = T/21.3$ fs. Thus the laser-induced fusion probability is $P_{\text{laser}} = \sigma_F/(\pi x_T^2)$. For 800 nm light, $I \sim 10^{22}$ W/cm², peak deuteron energy in the fixed $dd\mu$ frame is ~ 500 keV, and the corresponding fusion cross section $\sigma_F = 1.2 \times 10^{-25}$ cm² [20]. This gives us the laser-induced probability $P_{\text{laser}} = 1.8 \times 10^{-10}$ and the corresponding rate $R_{\text{laser}} = 1.2 \times 10^5$ s⁻¹. The experimental background muon catalyzed fusion rate is 1.5×10^9 s⁻¹ which means that during the muon life time fusion will occur. Thus at 800 nm, laser-triggered fusion in 3D contributes on a much lower scale than the background fusion rate. However, this need not limit our ability to observe tunnel dissociation via bond softening or laser triggered fusion.

First, our prediction of bond softening and tunnel dissociation can be tested on any muonic molecule including those with low fusion rates such as $pt\mu$. Second, laser stimulated experiments seem most accessible with $dd\mu$ or $dt\mu$. These experiments must therefore deal with a large background fusion signal because $dd\mu$, for example, decays in 10^{-9} s⁻¹ while we predict that the laser-induced fusion probability is only 1.8×10^{-10} per molecule/laser shot and $\sim 10^{-8}$ in $dt\mu$. However, laser stimulated fusion is labeled by the presence of the strong laser field, as is the dynamics of any process that is stimulated. For example, in the absence of the field, the fragments from the fusion reaction $d + d \rightarrow n + {}^3\text{He}$ have very well defined energies, i.e., neutron energy $E_n = 2.45$ MeV [11] and helium energy $E_{\text{He}} = 0.82$ MeV. By contrast, the laser creates d^+ with initial energy $\sim 3U_p \sim 0.5$ MeV which will yield significantly more energetic fragments. Thus the measurement of neutrons having energy higher than $E_n = 2.45$ MeV is a clear signature of the laser triggered action. In addition, the field accelerates charged laser-fusion fragments. Their energy and direction is a measure of their time of birth.

In conclusion, we stress the implications of our calculations. Intense laser pulses can initiate nuclear process with subfemtosecond precision provided we use molecules to fix the initial relative positions of the nuclei. The nuclear fragments should be clearly distinguishable from the background events. Just as charged particles

released into the laser field by attosecond pulses can be used to measure the pulse duration [21], so also the energy and direction of the nuclear fragments arising from the laser-induced process will allow time resolution of nuclear dynamics. Bringing such seemingly divergent areas as molecular physics and nuclear physics together offers great potential.

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