

## Nuclear Reaction Induced by Carrier-Envelope-Phase Controlled Proton Recollision in a Laser-Driven Molecule

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Nuclear reactions induced by proton recollision with a nearby nucleus are studied in a setup where a neutral molecule is exposed to an extremely intense, few-cycle laser pulse. At the rising edge of the laser pulse, all electrons in the molecule are first ejected by field ionization, resulting in a molecule consisting of the bare nuclei only. A proton in the bare molecule is subsequently accelerated by the laser field in such a way that it recollides with a nearby, heavier nucleus, with a kinetic energy high enough to induce a nuclear reaction. As a specific example, the probability of triggering the  $^{15}\text{N}(p, \alpha)^{12}\text{C}$  reaction by exposing either a  $^{15}\text{NH}$  molecule or a  $^{15}\text{NH}_3$  molecule to an intense laser pulse is calculated using the classical trajectory Monte Carlo method. We show that the proton recollision process can be controlled both by varying the carrier-envelope phase of the laser field and by the degree of molecular orientation. We also find that the magnetic field of the laser pulse plays a crucial role in the recollision dynamics.

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*Introduction.*—Electron recollision, where an electron is first liberated from an atom or molecule by an intense laser field, and then accelerated by the same field so that it returns close to the core with a high kinetic energy, is a central concept in strong-field physics [1,2]. If the kinetic energy of the recolliding electron is radiated away as a photon, the process is called high-harmonic generation, and can be employed to create attosecond pulses of light [3–6], or to gain information on the valence electrons [7,8]. The recolliding electron can also scatter, in which case the high-energy part of the photoelectron spectrum can be used to retrieve the molecular structure [9], or it can knock out a second electron, yielding highly correlated double-electron spectra [10].

Given that a lot of interesting atomic and molecular physics has been investigated with recolliding electrons, it is natural to try to extend the recollision scheme to heavier particles such as protons. If we could make a proton recollide with another nucleus at a kinetic energy of around 1 MeV or above (in the rest frame of the target nucleus), various nuclear reactions could be triggered, with attosecond temporal control of the collision event. The laser intensity required to accelerate a proton directly to MeV energies can be estimated from the ponderomotive potential (the average kinetic energy of a charged particle in an oscillating electric field)  $U_p = q^2 E_0^2 / (4M\omega_0^2)$ , where  $q$  is the particle charge,  $M$  is the particle mass,  $E_0$  is the peak electric field, and  $\omega_0$  is the angular frequency. For a proton,  $U_p = 1$  MeV requires  $E_0 = 5 \times 10^{14}$  V/m, or a laser intensity of  $I_0 = 3 \times 10^{22}$  W/cm<sup>2</sup>, at a laser wavelength of  $\lambda_0 = 800$  nm. Laser pulses with this intensity have already been demonstrated [11], and are expected to be routinely available at planned facilities such as the Petawatt

Field Synthesizer in Garching, Germany [12], the European Extreme Light Infrastructure [13], the Russian Exawatt Center for Extreme Light Studies [14], or the Japanese GEKKO EXA laser [15,16].

We should note here that fusion of deuterons has already been experimentally realized in laser-heated plasmas [17,18] and laser-driven clusters of deuterated molecules [19–21]. However, in this case the timing of the  $dd$  collisions is essentially random, and the field accelerating the deuterons to high energy is the Coulomb field of the surrounding ions in the plasma. The major advantage of driving the proton directly with the laser field is that the instant when the nuclear collision takes place can be precisely controlled. This might allow for time-dependent probing of short-lived excited nuclear states which is difficult to study with conventional accelerators [22].

Except for the requirement of high laser intensity, a major problem that has to be overcome is how to set up the initial conditions for the proton to be accelerated by the laser field. The proton first has to be separated from the nucleus with which it later recollides. Direct ejection of a proton bound to a stable nucleus by the absorption of laser photons can be ruled out, since a typical proton separation energy for stable nuclei is several MeV [23]. One solution previously proposed is to use a muonic molecule consisting of two deuterons and one muon [24–27], where after dissociation, the free deuteron is accelerated to recollide with the neutral muonic atom. Recently, another approach was put forward, involving the recollision of an alpha particle, resulting from alpha decay, with the daughter nucleus [28]. This scheme requires nuclear isotopes with short half-lives, such that an appreciable fraction decays during the duration of the laser pulse.

In this Letter, we investigate an alternative way of implementing proton recollision, which does not require exotic targets. Instead, we start with a molecule containing protons and one stable nuclear isotope. The scheme is general, and can be applied to any nuclear isotope that can form a molecule with hydrogen. If a small molecule is placed in an intense laser pulse with an intensity of the order of  $10^{22}$  W/cm<sup>2</sup>, all electrons, including the core electrons, are ejected immediately already at the leading edge of the pulse. Using the critical, over-the-barrier field strength  $E_c = 3 \times 10^{10} Z^3$  V/m [29], where  $Z$  is the charge number (the number of electrons), to estimate the number of electrons that are promptly ejected, we find that at  $I_0 = 1.4 \times 10^{20}$  W/cm<sup>2</sup> ( $E_0 = 3 \times 10^{13}$  V/m), 10 electrons can be removed. To a good approximation, we may therefore assume that we start with a molecule stripped of all electrons, consisting solely of bare nuclei. In addition, since the initial ejection of the electrons occurs at a time scale much faster than the motion of the nuclei, we assume that the nuclei start out at the positions given by the molecular equilibrium structure. We note that in principle, the proposed scheme could also be applied to other molecules containing heavier nuclei, provided that the difference in charge-to-mass ratio of two ions created after (partial) ionization is large enough.

The idea of nuclear recollision within a molecule was first mentioned in [30,31], and later theoretically implemented for HT molecules in [32,33]. Here, we point out two ways of controlling the recollision process, not exploited in [32,33], which lead to a dramatically enhanced recollision probability: One way is to align, or orient, the target molecule with a weaker prepulse [34,35] before the interaction with the main laser pulse; the second way is to change the carrier-envelope phase (CEP) of the driving laser pulse. CEP control of laser-driven plasma processes at intensities of  $10^{18}$  W/cm<sup>2</sup> has already been demonstrated [36], and CEP-stabilized, 5-fs pulses at focused intensities of  $10^{22}$  W/cm<sup>2</sup> are proposed in [12]. We also mention that the CEP of laser pulses with intensity  $> 10^{20}$  W/cm<sup>2</sup> can be measured via the multiphoton Compton scattering spectrum [37,38], which might allow for an approach where the CEP is measured at each laser shot instead of controlled.

*Theoretical model and simulation.*—In order to theoretically simulate the above-mentioned scenario, we first fix the target molecule. In order to keep things simple, we select the diatomic molecule <sup>15</sup>NH (imidogen with <sup>14</sup>N substituted for <sup>15</sup>N). This molecule can be obtained from plasma sources [39,40]. Nitrogen-15 is a stable isotope which is frequently used as a label in biological research [41]. We additionally give a few results for the commercially available molecule <sup>15</sup>NH<sub>3</sub> (ammonia-<sup>15</sup>N).

We also fix the nuclear reaction induced by the proton recollision. In the following, we focus on the <sup>15</sup>N( $p, \alpha$ )<sup>12</sup>C reaction, alpha particle emission by proton impact, which is

a reaction of importance for nuclear astrophysics [42]. The total cross section is of the order 0.1 b for a proton impact energy  $\epsilon$  in the interval  $0.4 \leq \epsilon \leq 0.8$  MeV [43] (in the rest frame of the <sup>15</sup>N nucleus).

The total probability for alpha particle emission by proton recollision is calculated with the classical trajectory Monte Carlo method. This method is well established for treating laser-induced recollision processes [44–46]. First,  $n$  different initial conditions for the velocities  $\mathbf{v}_p, \mathbf{v}_N$  and positions  $\mathbf{r}_p, \mathbf{r}_N$  of the proton and nitrogen nucleus are sampled from a distribution given by the molecular structure of <sup>15</sup>NH and the degree of alignment (details are discussed below). For each initial value, the classical equations of motion ( $X = p, N$ ),

$$M_X \frac{d^2 \mathbf{r}_X}{dt^2} = e^2 Z_N Z_p \frac{a_X \mathbf{r}}{|\mathbf{r}|^3} + e Z_X [\mathbf{E}(t) + \frac{\mathbf{v}_X}{c} \times \mathbf{B}(t)] \quad (1)$$

are solved numerically. In Eq. (1),  $e = |e|$  is the elementary charge,  $Z_p = 1, Z_N = 7, a_p = 1, a_N = -1, \mathbf{r} = \mathbf{r}_p - \mathbf{r}_N$  is the relative coordinate,  $\mathbf{E}(t)$  and  $\mathbf{B}(t)$  are the electric and magnetic field, respectively, of the laser pulse, and  $c$  is the speed of light. The spatial dependence of  $\mathbf{E}, \mathbf{B}$  in Eq. (1) is ignored due to the long wavelength employed. The explicit form adopted for the laser field is  $\mathbf{E}(t) = (\hat{\mathbf{z}} \cos \psi + \hat{\mathbf{x}} \sin \psi) f(t), \mathbf{B}(t) = -\hat{\mathbf{y}} f(t)$ , where the angle  $\psi$  defines the polarization direction,  $f(t) = E_0 \sin^2(\omega_0 t / 2N_c) \sin(\omega_0 t + \phi_0)$  if  $0 \leq t \leq 2\pi N_c / \omega_0$ , and 0 otherwise,  $N_c$  is the total number of cycles and  $\phi_0$  is the CEP. The propagation direction of the laser pulse is denoted by  $\hat{\mathbf{k}}$  and is given by  $\hat{\mathbf{k}} = \hat{\mathbf{x}} \cos \psi - \hat{\mathbf{z}} \sin \psi$ . We note that since, at the laser intensities used here ( $\sim 10^{22}$  W/cm<sup>2</sup>),  $|\mathbf{v}_{p,N}| \ll c$ , so that a nonrelativistic treatment is permissible. However, as we shall see, the inclusion of the magnetic field of the laser pulse is important to obtain correct results. In fact, the drift  $\delta$  of a charged particle during one cycle in the laser propagation direction due to the magnetic field can be estimated as  $\delta = 2\pi U_p / (cM\omega_0)$  [47]. For a proton in a  $\lambda_0 = 800$  nm,  $I_0 = 10^{22}$  W/cm<sup>2</sup> laser field,  $\delta \approx 3$  Å, which is comparable to the N-H internuclear distance. For comparison, the maximal displacement  $\eta$  of the proton in the electric field direction is approximately  $\eta = eE_0 / (M_p \omega_0^2) \approx 47$  Å.

Another thing worth mentioning, which is different compared to electron recollision, is that the quantum mechanical spread of a proton wave packet during one laser cycle is negligible due to the large mass. The width  $\Delta$  of a Gaussian wave packet of a free particle (in a laser pulse) increases with time as  $\Delta(t) = \sqrt{\Delta_0^2 + \hbar^2 t^2 / (\Delta_0 M)^2}$ , where  $\Delta_0$  is the initial width [48]. After a laser cycle  $\tau = 2\pi / \omega_0$ , we have  $\Delta(\tau) \approx \Delta_0$  for a proton, assuming an initial width of  $\Delta_0 = 0.5$  Å. Classically, the above argument implies that the initial velocity of the proton can be ignored. The negligible wave packet spreading is the reason why proton recollision is possible despite the long excursion of the proton before the recollision event.

After running  $n$  classical trajectories  $\mathbf{r}_{p,N}^j(t)$ ,  $\mathbf{v}_{p,N}^j(t)$ ,  $j = 1, \dots, n$ , we estimate the probability  $P$  of alpha particle emission as

$$P = \frac{1}{n} \sum_{j=1}^n \frac{\sigma(\epsilon_j)}{\pi b_0^2} \Theta(b_0 - b_j), \quad b_j = \frac{|\mathbf{v}_j(t_j) \times \mathbf{r}_j(t_j)|}{|\mathbf{v}_j(t_j)|}, \quad (2)$$

where  $\sigma(\epsilon)$  is the total cross section for the  $^{15}\text{N}(p, \alpha)^{12}\text{C}$  reaction taken from [43],  $\Theta(\cdot)$  is the step function,  $\mathbf{v}_j = \mathbf{v}_p^j - \mathbf{v}_N^j$  is the relative velocity,  $\epsilon_j = M_p |\mathbf{v}_j(t_j)|^2 / 2$  is the kinetic energy, and  $t_j$  is the instant during the trajectory where the N-H distance  $|\mathbf{r}_j| = |\mathbf{r}_p^j - \mathbf{r}_N^j|$  takes its smallest value.  $b_0$  is a parameter which we set to  $b_0 = 0.05 \text{ \AA}$ . In other words, we sum over all trajectories where the proton comes close enough to the  $^{15}\text{N}$  nucleus, and, in addition, has sufficiently high relative velocity for the cross section to be non-negligible; each such trajectory  $j$  contributes  $\sigma(\epsilon_j) / (\pi n b_0^2)$  to the total probability  $P$ . Multiple rescattering, i.e., a proton returning close to the  $^{15}\text{N}$  nucleus more than once during the laser pulse, can be neglected. We have checked that the results presented are not affected by variations of  $b_0$  in the interval  $0.01 \text{ \AA} < b_0 < 0.05 \text{ \AA}$ .

The initial conditions are sampled as follows. The  $^{15}\text{N}$  nucleus is always placed at rest at the origin. The initial position  $\mathbf{r}_p(0) = R(\sin \theta \cos \varphi, \sin \theta \sin \varphi, \cos \theta)$  of the proton is sampled according to a probability distribution

$$\frac{df(R, \theta, \varphi)}{d\theta d\varphi dR} = C \sin \theta \cos^{2k}(\theta/2) R^2 e^{-(R-R_0)^2/\rho^2}, \quad (3)$$

where  $C$  is a normalization constant. The employed values of the parameters  $\rho = 0.1 \text{ \AA}$  and  $R_0 = 1 \text{ \AA}$  for the radial distribution were derived from a fit to the square of the ground state wave function for the internuclear distance in the NH molecule (potential energy curve calculated with GAMESS [49]). The degree of orientation of the molecule, supposed to be realized by the interaction with a weaker, orienting prepulse, depends on the parameter  $k$ . For the distribution (3), we have  $\langle \cos \theta \rangle = k / (k + 2)$  for the average of  $\cos \theta$ , a commonly used measure of orientation [35]. Experimentally,  $\langle \cos \theta \rangle = 0.74$  (corresponding to  $k \approx 6$ ) has been achieved [34], and values up to  $\langle \cos \theta \rangle = 0.96$  ( $k = 48$ ) are speculated to be possible [34]. This last case would imply that more than 99% of the molecules are oriented in the same direction [34]. The initial velocity of the proton is set to zero, which is a good approximation in view of the small wave packet spreading mentioned above. We have checked that our results are not changed if we allow for an initial momentum of the proton distributed according to the square of the Fourier transform of the corresponding spatial wave function.

**Results and discussion.**—Representative results of our simulations are shown in Fig. 1, obtained running  $n = 10^6$  trajectories for each data point. The laser intensity

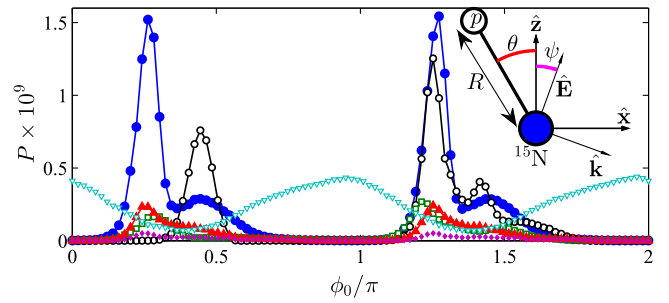


FIG. 1 (color online). Total probability  $P$  for alpha particle emission as a function of the CEP  $\phi_0$ , for different alignment parameters  $\psi$ ,  $k$ , and number of optical cycles  $N_c$ . Blue solid circles:  $^{15}\text{NH}$ ,  $\psi = \pi/2$ ,  $k = 100$ ,  $N_c = 3$ ; cyan open triangles:  $^{15}\text{NH}$ ,  $\psi = \pi/2$ ,  $k = 100$ ,  $N_c = 5$ ; black open circles:  $^{15}\text{NH}$ ,  $\psi = \pi/4$ ,  $k = 100$ ,  $N_c = 3$ ; red solid triangles:  $^{15}\text{NH}$ ,  $\psi = \pi/2$ ,  $k = 5$ ,  $N_c = 3$ ; magenta diamonds:  $^{15}\text{NH}$ , unaligned ( $k = 0$ ),  $N_c = 3$ ; green squares:  $^{15}\text{NH}_3$  (ammonia molecule),  $\psi = 0$ ,  $k = 100$ ,  $N_c = 3$ . The inset displays the coordinate system and the relevant vectors and angles:  $\theta$  is the angle between the molecular axis and the  $z$  axis,  $\hat{\mathbf{E}}$  is the direction of the electric field of the laser pulse, and  $\hat{\mathbf{k}}$  is the propagation direction of the laser pulse.

employed is  $I_0 = 2.5 \times 10^{22} \text{ W/cm}^2$ , at wavelength  $\lambda_0 = 800 \text{ nm}$ ,  $N_c = 3$  or 5 optical cycles (corresponding to pulse widths of 3 and 5 fs FWHM, respectively), different polarization directions  $\psi$  and varying value of the CEP  $\phi_0$ . One curve calculated with  $^{15}\text{NH}_3$  as the target is also shown. In this case, we allow for all three protons to recollide, and sum up the contributions to obtain the total probability. In practice, however, the trajectories where two (or three) protons recollide make a negligible contribution. Before the interaction with the laser field, the  $C_3$  symmetry axis of  $^{15}\text{NH}_3$  is assumed to be aligned with the  $z$  axis with the same  $\theta$  distribution as in Eq. (3), and  $^{15}\text{N}$  is placed at the origin. The proton initial position vectors  $\mathbf{r}_{p,j}$ ,  $j = 1, 2, 3$ , have positive  $z$  coordinates  $\hat{\mathbf{z}} \cdot \mathbf{r}_{p,j} > 0$ , and fixed angle  $\angle(\mathbf{r}_{p,i}, \mathbf{r}_{p,j}) = 107.8^\circ$  [50] between two proton position vectors, but are randomly rotated around the  $C_3$  symmetry axis. The N-H bond length is sampled from the same kind of distribution as in Eq. (3), with  $R_0 = 1 \text{ \AA}$  and  $\rho = 0.1 \text{ \AA}$ .

Figure 1 clearly reveals that the proton recollision process can be controlled in several ways. As in electron recollision processes [10,51,52], the recollision probability depends crucially on the CEP, and is most efficient for short pulses. The curve for the five-cycle pulse has a lower maximum probability, and is less dependent on the CEP compared to the three-cycle results. In the case of  $N_c = 3$ , the most efficient value of the CEP (around  $\phi_0/\pi = 0.25$  and 1.25) is when many initial positions lead to small proton impact parameters, while values of  $\phi_0$  in the range  $0.7 < \phi_0/\pi < 1$  hardly lead to any recollision events at all. Another control parameter is the alignment of the molecule. 1 order of magnitude higher probability is obtained by strongly aligning the  $^{15}\text{NH}$  molecule in the right direction

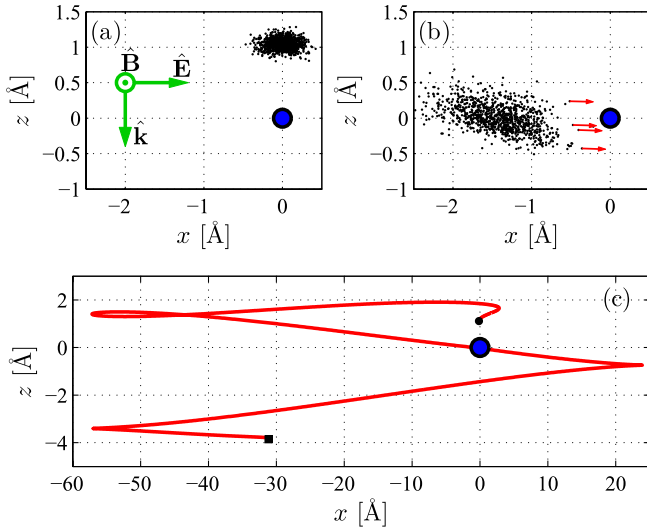


FIG. 2 (color online). Trajectories calculated at  $I_0 = 2.5 \times 10^{22}$  W/cm<sup>2</sup>, with CEP  $\phi_0/\pi = 0.26$ ,  $N_c = 3$ , and  $\psi = \pi/2$ . (a) Distribution of the initial relative positions  $\mathbf{r}_j(0) = \mathbf{r}_p^j(0) - \mathbf{r}_N^j(0)$  for  $10^3$  proton trajectories, projected onto the  $xz$  plane. Initial positions of the proton are shown with black dots, the  $^{15}\text{N}$  nucleus is shown with a larger, blue circle. The directions of the laser electric field  $\mathbf{E}(t) = \hat{\mathbf{E}}f(t)$ , the laser magnetic field  $\mathbf{B}(t) = \hat{\mathbf{B}}f(t)$ , and the propagation direction  $\hat{\mathbf{k}}$  of the laser pulse are indicated in the inset. (b) Distribution of the relative proton positions  $\mathbf{r}_j(t)$  at the instant  $t = 3.8$  fs (1.4 laser cycles). The directions of the relative velocity  $\mathbf{v}_j(t)$  are shown with red arrows for a few trajectories. All trajectories shown have relative kinetic energy  $\epsilon_j(t) = 0.4 \pm 0.002$  MeV. Note that the scales of the  $x$  and  $z$  axes are the same in (a) and (b). (c) Example of a trajectory  $\mathbf{r}(t)$  (small solid circle is the starting position, and the small solid square is the position at the end of the laser pulse) which results in a recollision with small impact parameter.

(compare the unaligned [magenta diamonds] and aligned [blue circles; alignment parameter  $k = 100$ , corresponding to  $\langle \cos\theta \rangle = 0.98$ ] in Fig. 1). The reason why  $\psi = \pi/2$  results in slightly higher values of  $P$  than  $\psi = \pi/4$  is due to the magnetic part of the Lorentz force, which accelerates the proton in the laser propagation direction. It is therefore advantageous to orient the molecule in such a way that  $\hat{\mathbf{k}}$  points from the initial proton position toward the position of the  $^{15}\text{N}$  nucleus. We have also verified that the alpha particle production does not occur at lower laser intensity. A calculation performed at  $I_0 = 1.3 \times 10^{22}$  W/cm<sup>2</sup> (and otherwise the same parameters as for the solid circle curve in Fig. 1) resulted in negligible recollision probabilities. This means that the alpha particles are produced only in the laser focus.

In order to obtain a more direct picture of the recollision process, we show in Fig. 2 the relative positions  $\mathbf{r}_j(t)$  of the proton at  $t = 0$  and  $t = 3.8$  fs, just before recollision. The laser field parameters are the same as for Fig. 1, with  $\psi = \pi/2$ , orientation parameter  $k = 100$ , and CEP  $\phi_0/\pi = 0.26$  (the position of one of the peaks of the curve shown

with solid circles in Fig. 1). It can be seen in Fig. 2(b) that the spatial distribution of the trajectory ensemble has broadened both in the  $z$  and  $x$  directions. This broadening is due to the strong Coulomb repulsion between the proton and the  $^{15}\text{N}$  nucleus. We can also better understand from Fig. 2(b) why the  $\psi = \pi/2$  orientation of the electric field together with  $\phi_0/\pi = 0.26$  yield the highest recollision probability (see Fig. 1). In this configuration, the impact parameter distribution at recollision is essentially given by the initial  $R$  distribution (with additional Coulomb-field broadening), which is rather narrow [see Eq. (3) and Fig. 2(a)].

**Conclusions.**—We have shown that proton recollisions at MeV energies can be induced in the interaction of a small molecule and an extremely intense laser pulse, and that the recollision process can be controlled by varying the CEP of the laser pulse. In contrast to previous proposals [24,28,33], exotic or unstable targets are not required. An additional advantage of using a molecular target is that the recollision probability can be enhanced by orienting the molecules with a weak prepulse. The molecular structure naturally provides a starting configuration such that the proton and the heavy nucleus are spatially separated by one chemical bond length. By selecting the polarization direction of the laser field in a suitable way, this initial separation can be exploited to overcome the inevitable drift in the polarization direction caused by the magnetic field.

Finally, we comment on the experimental feasibility of our proposal. A laser intensity of  $2.5 \times 10^{22}$  W/cm<sup>2</sup> can be obtained by focusing a PW laser pulse [12] to an area of  $4 \mu\text{m}^2$ , which implies an effective focal volume of  $40 \mu\text{m}^3$ . A gas pressure of  $10^{-3}$  atm (to avoid plasma effects [53]), a repetition rate of 10 Hz [12], and the reaction probability  $P = 1.5 \times 10^{-9}$  per molecule and shot (see Fig. 1) then result in a total of about 50 alpha particle ejection events/h.

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- [1] P. B. Corkum, *Phys. Rev. Lett.* **71**, 1994 (1993).
- [2] F. Krausz and M. Ivanov, *Rev. Mod. Phys.* **81**, 163 (2009).
- [3] M. Hentschel, R. Kienberger, Ch. Spielmann, G. A. Reider, N. Milosevic, T. Brabec, P. Corkum, U. Heinzmann, M. Drescher, and F. Krausz, *Nature (London)* **414**, 509 (2001).
- [4] P. M. Paul, E. S. Toma, P. Breger, G. Mullot, F. Augé, P. Balcou, H. G. Muller, and P. Agostini, *Science* **292**, 1689 (2001).
- [5] E. J. Takahashi, P. Lan, O. D. Mücke, Y. Nabekawa, and K. Midorikawa, *Phys. Rev. Lett.* **104**, 233901 (2010).
- [6] C. Hernández-García, J. A. Pérez-Hernández, T. Popmintchev, M. M. Murnane, H. C. Kapteyn, A. Jaron-Becker, A. Becker, and L. Plaja, *Phys. Rev. Lett.* **111**, 033002 (2013).

- [7] J. Itatani, J. Levesque, D. Zeidler, H. Niikura, H. Pépin, J. C. Kieffer, P. B. Corkum, and D. M. Villeneuve, *Nature (London)* **432**, 867 (2004).
- [8] P. Salières, A. Maquet, S. Haessler, J. Caillat, and R. Taïeb, *Rep. Prog. Phys.* **75**, 062401 (2012).
- [9] C. D. Lin, A.-T. Le, Z. Chen, T. Morishita, and R. Lucchese, *J. Phys. B* **43**, 122001 (2010).
- [10] C. Figueira de Morisson Faria and X. Liu, *J. Mod. Opt.* **58**, 1076 (2011).
- [11] V. Yanovsky *et al.*, *Opt. Express* **16**, 2109 (2008).
- [12] Z. Major *et al.*, *Review of Laser Engineering* **37**, 431 (2009).
- [13] Extreme Light Infrastructure (ELI), <http://www.extreme-light-infrastructure.eu/>.
- [14] Exawatt Center for Extreme Light Studies (XCELS), <http://www.xcels.iapras.ru/>.
- [15] GEKKO EXA project, <http://www.ile.osaka-u.ac.jp/Gekko-EXA/>.
- [16] J. Kawanaka, HEC-DPSSL workshop, Lake Tahoe, California on Sept. 12 2012; presentation material available at [https://lasers.llnl.gov/workshops/hec\\_dpssl\\_2012/pdf/9-12-12/J.Kawanaka.pdf](https://lasers.llnl.gov/workshops/hec_dpssl_2012/pdf/9-12-12/J.Kawanaka.pdf) (unpublished).
- [17] H. Daido, M. Yamanaka, K. Mima, K. Nishihara, S. Nakai, Y. Kitagawa, E. Miura, C. Yamanaka, and A. Hasegawa, *Appl. Phys. Lett.* **51**, 2195 (1987).
- [18] T. R. Dittrich, B. A. Hammel, C. J. Keane, R. McEachern, R. E. Turner, S. W. Haan, and L. J. Suter, *Phys. Rev. Lett.* **73**, 2324 (1994).
- [19] T. Ditmire, J. Zweiback, V. P. Yanovsky, T. E. Cowan, G. Hays, and K. B. Wharton, *Nature (London)* **398**, 489 (1999).
- [20] G. Grillon *et al.*, *Phys. Rev. Lett.* **89**, 065005 (2002).
- [21] Z. Zhou *et al.*, *J. Phys. B* **43**, 135603 (2010).
- [22] X. Mougeot *et al.*, *Phys. Lett. B* **718**, 441 (2012).
- [23] International Atomic Energy Agency, Nuclear Data Services, <http://www-nds.iaea.org/>.
- [24] S. Chelkowski, A. D. Bandrauk, and P. B. Corkum, *Phys. Rev. Lett.* **93**, 083602 (2004).
- [25] S. Chelkowski, A. D. Bandrauk, and P. B. Corkum, *Laser Phys.* **14**, 473 (2004).
- [26] G. K. Paramonov, *Chem. Phys.* **338**, 329 (2007).
- [27] A. D. Bandrauk and G. K. Paramonov, *AIP Conf. Proc.* **1209**, 7 (2010).
- [28] H. M. Castañeda Cortés, C. Müller, C. H. Keitel, and A. Pálffy, *Phys. Lett. B* **723**, 401 (2013).
- [29] R. Shakeshaft, R. M. Potvliege, M. Dörr, and W. E. Cooke, *Phys. Rev. A* **42**, 1656 (1990).
- [30] O. Smirnova, M. Spanner, and M. Y. Ivanov, *Phys. Rev. Lett.* **90**, 243001 (2003).
- [31] H. Niikura, F. Légaré, R. Hasbani, M. Yu. Ivanov, D. M. Villeneuve and P. B. Corkum, *Nature (London)* **421**, 826 (2003).
- [32] A. V. Sokolov and M. Zhi, *J. Mod. Opt.* **51**, 2607 (2004).
- [33] M. Zhi and A. V. Sokolov, *Phys. Rev. A* **80**, 023415 (2009).
- [34] O. Ghafur, A. Rouzée, A. Gijsbertsen, W. Kiu Siu, S. Stolte, and M. J. J. Vrakking, *Nat. Phys.* **5**, 289 (2009).
- [35] K. Oda, M. Hita, S. Minemoto, and H. Sakai, *Phys. Rev. Lett.* **104**, 213901 (2010).
- [36] A. Borot, A. Malvache, X. Chen, A. Jullien, J.-P. Geindre, P. Audebert, G. Mourou, F. Quéré, and R. Lopez-Martens, *Nat. Phys.* **8**, 416 (2012).
- [37] F. Mackenroth, A. Di Piazza, and C. H. Keitel, *Phys. Rev. Lett.* **105**, 063903 (2010).
- [38] M. Wen, L. L. Jin, H. Y. Wang, Z. Wang, B. F. Shen, Y. R. Lu, J. E. Chen, and X. Q. Yan, *Phys. Rev. E* **85**, 035401(R) (2012).
- [39] P. Chollet, G. Guelachvili, M. Morillon-Chapey, P. Gressier, and J. P. M. Schmitt, *J. Opt. Soc. Am. B* **3**, 687 (1986).
- [40] E. Tsikata, W. C. Campbell, M. T. Hummon, H.-I. Lu, and J. M. Doyle, *New J. Phys.* **12**, 065028 (2010).
- [41] M. Meselson and F. W. Stahl, *Proc. Natl. Acad. Sci. U.S.A.* **44**, 671 (1958).
- [42] M. La Cognata *et al.*, *Phys. Rev. C* **76**, 065804 (2007).
- [43] A. Redder, H. W. Becker, H. Lorenz-Wirzba, C. Rolfs, P. Schmalbrock, and H. P. Trautvetter, *Z. Phys. A* **305**, 325 (1982).
- [44] P. J. Ho, R. Panfili, S. L. Haan, and J. H. Eberly, *Phys. Rev. Lett.* **94**, 093002 (2005).
- [45] H. Bauke, H. G. Hetzheim, G. R. Mocken, M. Ruf, and C. H. Keitel, *Phys. Rev. A* **83**, 063414 (2011).
- [46] E. Lötstedt and K. Midorikawa, *Phys. Rev. A* **87**, 013426 (2013).
- [47] A. Di Piazza, C. Müller, K. Z. Hatsagortsyan, and C. H. Keitel, *Rev. Mod. Phys.* **84**, 1177 (2012).
- [48] D. J. Tannor, *Introduction to Quantum Mechanics: A Time-Dependent Perspective* (University Science Books, Herndon, VA, 2007).
- [49] M. W. Schmidt *et al.*, *J. Comput. Chem.* **14**, 1347 (1993).
- [50] N. N. Greenwood and A. Earnshaw, *Chemistry of the Elements* (Butterworth Heinemann, Oxford, 1998).
- [51] G. G. Paulus, F. Grasbon, H. Walther, P. Villoresi, M. Nisoli, S. Stagira, E. Priori, and S. De Silvestri, *Nature (London)* **414**, 182 (2001).
- [52] B. Bergues *et al.*, *Nat. Commun.* **3**, 813 (2012).
- [53] D. Umstadter, *J. Phys. D* **36**, R151 (2003).