Laser Plasma-Accelerated Ultra-Intense Electron Beam for Efficiently Exciting Nuclear Isomers

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Utilizing a laser plasma wakefield to accelerate ultrahigh charge and current electron beams is critical for many pioneering applications, for example, to efficiently produce nuclear isomers with short lifetimes that may be widely used. However, because of the beam loading effect, the electron charge in a single plasma bubble is limited to hundreds of picocoulombs. Herein, via a tightly focused intense laser pulse self-guiding in dense gas, a twenty-nanocoulomb, tens-of-MeV, hundred-kiloampere collimated electron beam is produced from a chain of plasma bubbles in the saturated injection regime. This intense electron beam is utilized to excite inductium nuclear isomers with an ultrahigh peak rate of $1.12 \times 10^{15}$ particles s$^{-1}$ via photonuclear reaction. This efficient isomer production method can be widely used for pumping isotopes with excited state lifetimes on the picosecond scale, which is beneficial for deeply understanding nuclear transition mechanisms or stimulating $\gamma$-ray lasers.

1. Introduction

Laser plasma wakefield accelerators (LWFA) have attracted significant interest in recent years[1–4] due to their high acceleration gradients (hundreds of GV m$^{-1}$) and beam currents (tens of kiloamperes), thus not only enabling the reduction of GeV electron accelerator lengths to mere centimeters[5] but also driving secondary radiation/particle sources with ultrahigh brightness/flux.[6–8] In the bubble[9] or blowout[10] regimes of LWFA, an ultrashort intense laser pulse excites the wakefield in underdense plasma, and the laser ponderomotive force expels electrons, forming ion cavities (called bubbles) in which electrons can be accelerated. In the last decade, some breakthroughs in LWFA research have been achieved, such as fine stability,[11] multi-GeV energy,[12] femtosecond (fs) beam duration,[13] ultralow energy spread,[7,14] etc., but the beam charge is still limited below hundreds of picocoulombs (pC).[15–17] This beam charge limitation is caused by the beam loading effect in a single bubble: injected electrons reduce the electrostatic potential of the plasma bubble and then block the injection process.[10,18,19]

Recently, several hundred-pC electron beams, reaching the beam loading effect, have been observed in the process of double self-injection,[15,20] which occurs during the combination of two plasma bubbles to form a large single bubble, and in the process of self-truncated ionization injection,[27] which is continuous injection in a single bubble, but it is still difficult to approach the nanocoulomb (nC) limit. There have been some efforts to improve the beam charge to reach the nC limit via ionization injection in multiple plasma bubbles,[21,22] but the total beam charge is still one order of magnitude lower than the theoretical maximum load of plasma bubbles[23] due to unsustainable injection[21] or unsaturated injection.[22] While tens of nC and tens of MeV electron beams can be realized by laser interactions with near-critical-density plasma,[24,25] the beam divergence angle is usually large ($\approx 15^\circ$) due to electron interactions with laser fields, and they also scatter in dense plasma after rapid laser pulse depletion. Moreover, tens of nC directional electron beams can be produced from vacuum laser acceleration[26–28] or target surface electron acceleration[29] by laser–solid interactions, but an important limiting factor is the short acceleration distance resulting in electron energies that are usually less than 10 MeV. There
Figure 1. Experimental setup. The femtosecond intense laser pulse is focused onto the gas jet to drive the plasma wakefield electron acceleration, then the electron beam bombarded on the converter to induce photonuclear reactions. The illustrations in the upper right corner are schematics of photofission, neutron capture and the level scheme of the $^{115}$In nucleus, respectively. The illustrations in the left lower corner are the simulated plasma wake and the Top-view plasma channel, respectively.

is another method called self-modulated laser wakefield acceleration (SM-LWFA)\cite{30-33} in which long laser pulses overlap with several tens of plasma waves, thereby trapping a large number of electrons and accelerating them in every wakefield, and the beam charge can be increased tremendously to hundreds of nC\cite{34,35} but this method requires a kilojoule-class picosecond (ps) laser facility with a size and cost that are at least ten times that of a fs laser facility. In general, for laser plasma electron acceleration using a small laser, it is a great challenge to realize an electron beam with a large charge, tight collimation and high energy.

In this work, electron beams with a charge of $\approx 20$ nC and a small divergence angle of $\approx 6^\circ$ were generated experimentally from a tens of TW tabletop tightly focused fs laser pulse interacting with dense nitrogen gas. Particle in cell (PIC) simulation ascribed this high-charge collimated electron beam to an efficient electron injection scenario and the self-generated fields in the plasma channel. The electron beams had appropriate energies (tens of MeV) for driving photonuclear reactions\cite{36} and exciting nuclear isomers\cite{37}. By utilizing these intense and energetic electron beams to hit the indium target, a significant amount of isomers were produced within an ultrashort time duration.

2. Experimental Section

2.1. Experimental Setup

The experiment was carried out using a Ti:sapphire laser system at the Laboratory of Laser Plasmas of Shanghai Jiao Tong University. The experimental setup is shown in Figure 1. A linearly polarized laser pulse with wavelength of 800 nm, duration of 45 fs (FWHM), energy of 3.2 J was focused by an f-number 4 off-axis parabolic (OAP) mirror onto a supersonic gas jet with a width of 1.2 mm, a length of 10 mm and a well-defined uniform density distribution.\cite{38} The nozzle was placed horizontally relative
to the laser propagation direction. The laser pulse was focused to a main spot with radius \( w_0 = 3.8 \, \mu m \), containing approximately 38% of the laser power, i.e., \( \approx 27 \, \text{TW} \), resulting in an intensity of \( \approx 5.8 \times 10^{19} \, \text{Wcm}^{-2} \) and the normalized vector potential \( a_0 \approx 4.9 \). By regulating the gas back pressure, the outflow nitrogen gas density ranged from \( 3 \times 10^{17} \, \text{cm}^{-3} \) to \( 2 \times 10^{19} \, \text{cm}^{-3} \). The plasma channel was monitored by a Top-view system. After the laser interaction with pure nitrogen gas (purity 99.999%), the electron beam from the laser plasma wakefield acceleration bombarded the DRZ fluorescent screen (covered with 14 \( \mu m \) aluminum film to block stray light) emitting fluorescence, which was detected by an EM-CCD to obtain an electron beam spot image. When moving in the magnet, the electron beam angular distribution was recorded by a 20 cm \( \times \) 20 cm SR-type image plate (IP), which was covered with 200 \( \mu m \) copper to block low-energy electrons. The electron beam charge was also calculated according to this IP,[39] which was scanned by a Typhoon-7000 IP reader. The electron beam energy distribution was recorded by another SR-type IP. When moving in the converter, the accelerated electron beam bombarded a 1 mm-thick tungsten (W) (purity 99.99%)) converter located downstream of the nozzle to generate bremsstrahlung radiation. Then, the collimated \( \gamma \)-rays induced the \( (\gamma, \gamma') \) reaction, photofission \((\gamma, n)\), \((\gamma, 2n)\), \((\gamma, 3n)\) reactions and neutron capture \((n, \gamma)\) reaction in a 3 mm-thick indium (In) (purity 99.995%) converter. These products of the above reactions were in nuclear isomeric states or became radionuclides, which were identified by measuring their characteristic decay radiations with a high purity germanium detector. Because the converter (In) had two natural isotopes\(^{115}\text{In} (95.71\%) \) and \(^{115}\text{In} (4.29\%)\), the experimental layout only shows the schematics of photofission, neutron capture and the excited states of the \(^{115}\text{In} \) nucleus.

2.2. Particle in Cell Simulation Setup

The 2D particle in cell simulations were carried out using the EPOCH code.[40] The simulation box size was 120 \( \times \) 120 \( \mu \text{m}^2 \) with 4800 \( \times \) 1200 cells in the x and y directions. The simulation box propagated along the x-axis at the speed of light. Ionization was modeled with the ADK rates,[41] and one macroparticle per cell was used as the nitrogen atom. The neutral nitrogen longitudinal profile had a 100 \( \mu \text{m} \) upramp followed by a 1.2 \( \mu \text{m} \) long plateau with a uniform density, then followed by a 100 \( \mu \text{m} \) downramp. The p-polarized laser pulse propagated along the x-direction, and the laser-focusing plane was located at the middle of the upramp. The laser pulse had a Gaussian transversal profile with \( w_0 = 3.8 \, \mu \text{m} \) and a Gaussian longitudinal envelope with a pulse duration of 45 fs. The normalized vector potential was \( a_0 = 4.9 \).

3. Results and Discussion

3.1. Efficient Electron Acceleration

The laser pulse focused by the small f-number OAP mirror has an intense power density but a shorter Rayleigh length \( l_0 \approx 60 \mu \text{m} \) for a radius value of \( w_0 = 3.8 \, \mu \text{m} \). Due to the plasma bubble radius \( R = 2c\sqrt{\omega_0/\omega_{pe}} \) and the laser self-focusing power \( P_c \approx 17n_e/n_i \) \([\text{GW}]\), where \( \omega_{pe} \approx \sqrt{4\pi n_e e^2/m_i} \) is the plasma frequency and \( n_i \) is the critical density, a higher plasma density is usually required to match the small laser focal spot \( (w_0 \approx R) \) for maintaining laser intensity and overcoming quick defocus.[42] Because the 27-TW focused laser pulse was hard to self-guide in the plasma at a density of approximately 5.6 \( \times \) 10\(^{18} \) \( \text{cm}^{-3} \), corresponding to \( P_c = 52 \, \text{TW} \), an electron beam with multiple spots and low energy was generated, as shown in Figure 2a.e. When the plasma density increased to 1.12 \( \times \) 10\(^{19} \) \( \text{cm}^{-3} \) \( (P_c = 26 \, \text{TW}) \), the matched laser \( w_0 \approx R \approx 6 \, \mu \text{m} \) during self-focusing and \( a_0 \approx 3 \) was higher than the ionization threshold of the inner-shell electrons of the nitrogen atom \((6^6, 7^6, \alpha_0 \approx 1.7, 1.9)\).[43,44] The electron beam charge increased prominently to approximately 5 nC, far exceeding the charge scaling law of the nonlinear bubble regime[23,45,46] (the detailed mechanism to be discussed later). With an increase in plasma density to 3.68 \( \times \) 10\(^{19} \) \( \text{cm}^{-3} \) \( (P_c = 8 \, \text{TW}) \), the electron beam charge reached 20 nC, and the divergence angle was just \( \approx 6^\circ \) (Figure 2c,f). We have also fired 70 consecutive shots at a repetition rate of 0.025 Hz under this parameter condition, the results were shown in ref. [39]. The average divergence angles in the x and y directions were 7.0° \( \pm \) 1.2° and 6.1° \( \pm \) 1.3° respectively, and the average charge was 15.59 \( \pm \) 1.68 nC, in which the error represented the standard deviation. However, upon further increasing the plasma density, all of the electron beam parameters deteriorated (Figure 2d–f) due to the aggravated dephasing of the electrons and the faster depletion of the laser pulse in the higher-density plasma. It is worth mentioning that the optimal energy conversion efficiency from the laser to the collimated electron beam could be up to 12.4% for \( E \approx 1 \, \text{MeV} \) (Figure 2g).

3.2. Density Matching for Tightly Focused LWFA

To better understand the density matching condition for the generation of large-charge, energetic, and collimated electron beams, we carried out PIC simulations to study the effects of the gas density on the tightly focused LWFA. The drive laser pulse with a power \( \approx 30 \, \text{TW} \) did not self-focus in nitrogen gas with a density of \( 4 \times 10^{17} \, \text{cm}^{-3} \), corresponding to a fully ionized plasma density of \( 3.6 \times 10^{18} \, \text{cm}^{-3} \) and \( P_c = 52 \, \text{TW} \). However, because the initial laser \( \alpha_0 \approx 4.9 \) was much higher than the ionization threshold of the inner-shell electrons, they experienced ionization injection (Figure 3a) in the process of laser defocusing until \( \alpha_0 \) was less than the threshold of the 6th electron; then, the injection stopped, as shown in Figure 3e. Notably, many inner-shell electrons were injected successively into multiple bubbles. When the gas density increased to \( 6 \times 10^{17} \, \text{cm}^{-3} \), the corresponding \( P_c \approx 35 \, \text{TW} \) was still higher than the laser power. The laser pulse did not self-focus but attained a slower defocus speed and maintained \( \alpha_0 \geq 1.7 \) at the same distance (Figure 3e,f), which resulted in a greater level of inner-shell electron ionization injection into bubbles (Figure 3b). If the gas densities continuously increased to \( 1 \times 10^{18} \, \text{cm}^{-3} \) and \( 2.2 \times 10^{18} \, \text{cm}^{-3} \) corresponding to \( P_c \geq 21 \, \text{TW} \) and 10 TW, respectively, a tightly focused laser pulse can be matched to the plasma bubble during the self-focusing process, resulting in the laser intensity being easily maintained above the ionization threshold of the 7th electron (Figure 3g,h), and a large amount of inner-shell electrons injecting into over ten bubbles (Figure 3c,d).
Figure 2. Experimental electron beam results for different density plasmas. a–d) Electron beam angular distributions at plasma densities of 0.56, 1.12, 3.68, and \(4.47 \times 10^{19}\) cm\(^{-3}\), respectively. The white lines represent the axis distributions. e) Electron beam energy distributions at different plasma densities. f,g) are the corresponding total charge (\(E_k \geq 1\) MeV) and energy conversion efficiency of the laser to the electron beam, respectively.

Figure 3. Small focal spot laser wakefield acceleration in pure nitrogen gas with different densities. a–d) Plasma electron density distributions, where the upper/lower regions represent the outer/inner-shell electrons of the nitrogen atom, the red line (normalized) is the electron number longitudinal distribution for \(E_k \geq 1\) MeV, and the number is the corresponding total number of microparticles. e–h) Electric-field distributions, where the upper/lower regions represent Ex/Ey, the scattering points represent the electron phase-space (x, \(E_k\)) distribution, and the color represents the electron divergence angle. a,e) correspond to a nitrogen gas density of \(4 \times 10^{17}\) cm\(^{-3}\); b,f) \(6 \times 10^{17}\) cm\(^{-3}\); c,g) \(1 \times 10^{18}\) cm\(^{-3}\); d,h) \(2 \times 10^{18}\) cm\(^{-3}\).
In LWFA, the number of electrons that can be loaded into a bubble is limited by the beam loading effect. According to M. Tzoufras’s theory, the maximum charge of loaded electrons \( Q_s \approx 0.047 \sqrt{a_0} \) per for 4.9, \( n_e \approx 3.68 \times 10^{19} \) cm\(^{-3}\) and the acceleration field after reduction due to the beam loading effect is \( E_s \approx 0.1 \) TVm\(^{-1}\) (Figure 3h), thus a single bubble can sustain a charge of \( Q_s \approx 1.8 \) nC. Therefore, the total beam charge \( \approx 20 \) nC \( (\approx 5.4 \) nC, \( E_s \geq 7 \) MeV) in the experiment reached injection saturation in approximately ten bubbles. Although ref. [22] utilized a laser energy of \( \approx 1.1 \) J \( (a_0 \approx 2) \) and realized electron injection in several bubbles, the process was limited by the injection length and injection speed, and the presented beam charge of \( \approx 270 \) pC \( (E_s \geq 7 \) MeV) still has much room for improvement by increasing the laser intensity.

### 3.3. Multiple Ionization Injections

The electrostatic potential \( \Psi \) of the plasma wake can be used to analyze the injection mechanisms. Electrons born at the peak of the wake potential \( (E_z = 0) \) will experience the largest \( |\Delta \Psi_{\text{max}}| \); if \( |\Delta \Psi_{\text{max}}| < -1 \), then electrons will be ionized earlier in the wake and can be trapped. The maximum wake potential can be approximated as \( \Psi_{\text{max}} \approx \frac{\psi_0 R}{x} \approx a_0 \), so a higher \( \psi_0 \) is beneficial for trapping electrons and realizing the saturation of the plasma wake. In contrast, electrons born on the pulse rising edge, as shown by the blue arrow in Figure 4a, do not experience a sufficient potential to be trapped in the first bubble. Fortunately, the potential in the subsequent wakefield is lower than that in the previous wakefield due to the nonlinear blowout regime and beam loading effect, resulting in these electrons being trapped by the subsequent bubbles. In practice, if the inner-shell electrons are ionized transversely far away from the axis of \( y = 0 \), they will be longitudinally located close to the peak of the pulse envelope, and then they would experience sufficient potential to be trapped by the first bubble, as shown in “1st” in Figure 4b. However, when the electrons are ionized close to the axis of \( y = 0 \), they will be born at the front edge of the laser pulse. These electrons would be trapped by more backward bubbles, for example, as shown in “3rd” and “4th” of Figure 4b[c].

At the beginning of the LWFA process, the electron beam longitudinal distribution has many modulation peaks with an interval \( \approx 23 \) fs and a duration \( \approx 4 \) fs (Figure 4d). As electrons are continuously injected into plasma bubbles, these electron bunches begin to combine after \( \approx 3 \) ps, and injection tends to be saturated at \( \approx 4 \) ps; then, the outgoing electron beam has a duration of \( \approx 150 \) fs (FWHM). Hence, the estimated beam peak current is \( \approx 100 \) kiloamperes (kA), considering the beam average charge \( \approx 15 \) nC in our experiment. Because of the limit of the dephasing length...
L_{d} \approx \frac{2na_{0}^{3}R}{4\pi} \approx 120 \mu m$, the electron maximum energy gain$^{[45]}$ is defined as $\Delta E \approx \frac{1}{2}m_{e}c^{2}(\frac{a_{0}}{R})^{2} a_{0} \approx 100$ MeV (Figure 3h). However, because the beam loading effect reduces the acceleration electric field, the energies of most electrons are located in the range of 3 to 30 MeV (Figure 4e), which agrees well with the experimental results. Moreover, because of the existence of self-generated confinement electromagnetic fields in the plasma bubbles,$^{[47]}$ the electron beam has a small source size ($\approx 4.3 \mu m$) and divergence angle ($\approx 7.8^\circ$) (Figure 4f), which also agrees with the experimental results.

3.4. Nuclear Isomer Excitation

A nuclear isomer is a metastable state of an atomic nucleus, whose excited states have lifetimes usually longer than $10^{-3}$ s. They have a broad range of potential applications,$^{[48,49]}$ including new energy-storage materials,$^{[50,51]}$ medical isotopes,$^{[52]}$ nuclear clocks,$^{[53,54]}$ and nuclear $\gamma$-ray lasers.$^{[55,56]}$ Especially for nuclear $\gamma$-ray lasers, one of the key bottlenecks for this application appears to be how to pump excited states efficiently. Because the total linewidth of the upper energy levels is mainly contributed by Doppler broadening, their lifetimes need to be as short as possible, and the total linewidth of the upper energy levels is mainly $\approx 8.2$% after the electron beam bombarding the attenuation loss $\approx 5.0$% due to the decay radiation (336 keV) transmission in the In sheet, the collection loss $\approx 50$% due to the decay radiation emission from the two sides of In sheet, and the absorption coefficient $\approx 100$% at 300 keV, the total yield of $^{115m}$In (336 keV) is $\approx 1.12 \times 10^4$. As for the production paths of the isomer $^{115m}$In (336 keV), Excitation directly from the ground state (0 keV 9/2+) to the isomeric state (336 keV 12/2+) is negligible, due to its transition mode M4(E5) has a tiny cross-section. Other transition mode which are M1(E2) etc. generally, the transition probability of En is approximately $2-3$ orders of magnitude higher than that of E(n+1) or M(n). The main paths to generate isomer $^{115m}$In (336 keV) are, as shown in the level scheme of $^{115}$In in Figure 1 (detailed see...
the peak excitation rate for the three isomeric states nuclei via $(\gamma, \gamma')$ reaction mainly through E1, or M1(E2) transition; second, some of these excited states may cascade decay to the states including (597 keV 0.25 ns 3/2-), (828 keV 5.78 ns 3/2+), and (864 keV 0.91 ns 1/2+); finally, these three states may decay to the state (336 keV 4.486 h 1/2-).

In this way, a very high rate of isomer generation can be achieved. Because the bremsstrahlung $\gamma$-ray beam duration is as short as the electron beam duration $\approx 150$ fs, the time required for $\gamma$-rays to pass through the 3 mm-thick In target is approximately 10 ps, which can be regarded as the excitation time duration for the generation of the above three isomeric states. Thus, the peak excitation rate for the three isomeric states nuclei via $(\gamma, \gamma')$ reaction is estimated to be $1.12 \times 10^{15}$ ps$^{-1}$. However, using a commercial electron accelerator ($E = 5$ MeV, $I_{peak} = 2$ mA, pulse duration of 15 $\mu$s) to bombard the same converter (W+In) as we did, the peak excitation rate can be estimated to be $\approx 10^6$ ps$^{-1}$. Even for a traditional high-energy electron accelerator (e.g., BEPC in China, $E = 1.5$ GeV, $I_{peak} = 600$ mA, pulse duration of 1.5 ns), the peak excitation rate could be only $\approx 10^{11}$ ps$^{-1}$. In addition, limited by the vacuum pump efficiency and laser repetition rate in this experiment, the average excitation efficiency of the isomer is less than $10^3$ ps$^{-1}$. However, an average efficiency of $10^6$ ps$^{-1}$ can be realized by utilizing a more powerful pump set and a 100 Hz hundred-TW laser facility, which will soon be available for example the laser “DUHA” in ELI-Beamlines.[62] This exciting method will also be greatly beneficial for the tabletop investigations of medical isotopes and nuclear batteries.[63]

4. Conclusion

In summary, we have presented a novel method for pumping nuclear isomers effectively via an ultrahigh current electron beam accelerated by a tightly focused intense laser self-guiding in dense plasma. When the laser pulse is matched with a gas with a suitable density, the inner-shell electrons of the nitrogen atom can be continuously injected into multiple plasma bubbles to reach the limit of saturation of the plasma wake. A hundred-kilomperare electron beam was generated with 12.6% energy conversion efficiency from the driving laser. The electron beam has a maximum charge of $\approx 20$ nC, divergence angle of $\approx 6^\circ$ and suitable energies for photonuclear reactions. By utilizing this intense electron beam to excite nuclear isomers via the $(\gamma, \gamma')$ reaction, an ultrahigh peak excitation rate of $\approx 1.12 \times 10^{15}$ ps$^{-1}$ was realized, which is at least four orders of magnitude higher than that obtained using traditional electron accelerators. This efficient and easily accessible production method of exciting nuclear isomers could be greatly beneficial for the study of nuclear transition mechanisms or nuclear $\gamma$-ray lasers.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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