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Electromagnetic field growth triggering super-ponderomotive electron acceleration during multi-picosecond laser-plasma interaction

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The dependence of the mean kinetic energy of laser-accelerated relativistic electrons (REs) on the laser intensity, so-called ponderomotive scaling, explains well the experimental results to date; however, this scaling is no longer applicable to multi-picosecond (multi-ps) laser experiments. Here, the production of REs was experimentally investigated via multi-ps relativistic laser-plasma-interaction (LPI). The lower slope temperature shows little dependence on the pulse duration and is close to the ponderomotive scaling value, while the higher slope temperature appears to be affected by the pulse duration. The higher slope temperature is far beyond the ponderomotive scaling value, which indicates super-ponderomotive REs (SP-REs). Simulation and experimental evidence are provided to indicate that the SP-REs are produced by LPI in an under-critical plasma, where a large quasi-static electromagnetic field grows rapidly after a threshold timing during multi-ps LPI.

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When a material is irradiated with a high-intensity laser pulse, its surface is instantaneously ionized, and the electrons in the ionized material, i.e., plasma, are then accelerated close to the speed of light by the ponderomotive force of the laser light. These energetic electrons are often called relativistic electrons (REs). The energy distribution of REs can be approximated by a Maxwell–Boltzmann distribution function with slope temperature T_{RE} . The scaling laws of T_{RE} with respect to laser intensity have been investigated experimentally^{1–3}, theoretically, and computationally^{4–7}. The effect of pulse duration on T_{RE} is not considered explicitly in the reported scaling laws; however, recent computational and theoretical studies have revealed that T_{RE} generated by multi-picosecond (multi-ps) laser pulses could be several times higher than that predicted by the reported scaling laws.

Kemp et al.⁸ investigated multi-ps relativistic laser–plasma-interaction (LPI) using a two-dimensional (2D) particle-in-cell (PIC) simulation. They clarified that ripples grow at the critical density surface during the interaction and the plasma then expands into the ripple valley from the side wall. This mechanism allows underdense plasma to expand gradually from the critical density surface over several tens of microns during the multi-ps relativistic LPI, even though the ponderomotive pressure in the laser propagation direction is larger than the plasma pressure. Electron acceleration is enhanced as the density scale length increases in the multi-ps timescale.

Sorokovikova et al.⁹ investigated the temporal evolution of a quasi-static electric field in multi-ps relativistic LPI. They reported that enhancement of electron acceleration in multi-ps relativistic LPI is caused by a combined effect of both the laser field and quasi-static electric field.

The quasi-static magnetic field generated at the critical density surface starts to re-inject electrons into the region where the laser field and quasi-static electric field coexist. The RE acceleration mechanism with re-injection has been investigated^{10,11} as loop-injected direct acceleration (LIDA). In particular, Krygier et al.¹⁰ predicted that the super-ponderomotive REs (SP-REs) can be accelerated by multi-ps LPI via the LIDA mechanism.

These results of the large timescale simulation revealed that electron acceleration by multi-ps laser pulses is not a simple extension of the electron acceleration mechanism of conventional sub-ps laser pulses. With the development of kilojoule-class high-power lasers such as LFEX¹², NIF-ARC¹³, LMJ-PETAL¹⁴, and OMEGA-EP¹⁵, it has become possible to irradiate relativistic laser pulses continuously over several picoseconds. These lasers open a new regime of LPI studies, i.e., multi-ps relativistic LPI.

In this study, we report an experimental investigation of energy conversion from laser to REs through multi-ps relativistic LPI using ultra-high-contrast LFEX laser pulses. Energy distributions of the measured REs are approximated well with two-temperature Maxwell–Boltzmann functions. The lower slope temperature shows little dependence on the pulse duration, and the lower temperature is close to the ponderomotive scaling value. On the other hand, the higher slope temperature is increased more than twofold by extending the laser pulse duration from 1.2 to 4.0 ps. The following acceleration mechanism is identified as essential for the generation of SP-REs in a multi-ps LPI with the help of PIC simulations. We observe a sudden growth of quasi-static electric and magnetic fields by an order of magnitude in an expanding plasma during multi-ps LPI due to positive feedback between the growth of the fields and the $\mathbf{E} \times \mathbf{B}$ drift current. In such a situation, a quasi-static magnetic field of tens of megagauss (MG) triggers the LIDA as the dominant acceleration mechanism. Previous studies assumed a magnetic field generated by the Biermann battery, which grows gradually with time and produces the LIDA. In our proposed mechanism, the LIDA process is

triggered by this sudden growth of the quasi-static fields after the transition of the relativistic LPI state from the hole-boring phase to the blowout phase in a laser-heated plasma¹⁶. Thus, LIDA becomes the dominant mechanism of SP-RE acceleration in a threshold manner. Here, we also obtain a theoretical model to calculate the transition time of the relativistic LPI state from the hole-boring phase to the blowout phase for an arbitrary temporal intensity profile of the incident laser.

Results

Observation of super-ponderomotive relativistic electrons. We have experimentally investigated the dependence of RE energy distributions on the pulse durations under conditions free from pre-plasma formation. The experiment was conducted using the LFEX laser system at the Institute of Laser Engineering, Osaka University¹². The LFEX laser consists of four beams, where the spot diameter of the spatially overlapped LFEX beams on a target was 70 μm of the full width at half maximum (FWHM), and 30% of the laser energy was contained in this spot. One LFEX beam delivered 300 J of 1.053 μm wavelength laser light with a 1.2 ps duration (FWHM), and the peak intensity of one beam was $2.5 \times 10^{18} \text{ W cm}^{-2}$.

SP-REs can be accelerated in a long-scale-length pre-plasma; therefore, a plasma mirror (PM)¹⁷ was implemented to realize the pre-plasma-free condition to exclude the other known mechanisms from this experiment (as shown in Fig. 1a). These clean pulses were focused on a 1 mm³ gold cube.

LFEX laser pulses can be stacked temporally with arbitrary delays between the beams, as shown in Fig. 1b. In this study, a single beam (case A (#38834): 1.2 ps FWHM pulse duration and peak intensity of $2.5 \times 10^{18} \text{ W cm}^{-2}$) was used and two types of four-stacked beams (case B (#38753): 4.0 ps FWHM pulse envelope and peak intensity of $3.0 \times 10^{18} \text{ W cm}^{-2}$, and case C (#38752): 1.2 ps FWHM pulse duration and peak intensity of $1.0 \times 10^{19} \text{ W cm}^{-2}$). We emphasize here that the leading edge of the stacked pulse remains similar to that of the single beam. If the pulse duration is extended by adjusting the pulse compressor of the laser system, then the leading edge would inevitably be modified into a more gradual shape.

Absolute energy distributions of REs were obtained with two diagnostics, a vacuum electron spectrometer (ESM) and a high-energy X-ray spectrometer (HEXS). The ESM captures REs emanated from the rear side of the gold cube to the vacuum region in the direction normal to the target. Less energetic electrons of typically <1 MeV are more easily trapped by a sheath electric field generated at the target surface by charge separation. Therefore, the ESM can be used to measure only high-energy REs with >1 MeV. The HEXS was used to evaluate the energy distribution of low-energy REs with <1 MeV from the X-ray spectrum emitted from the gold cube. HEXS consists of differential X-ray filters and dosimeters that are alternately stacked, as shown in Fig. 1c. The layer response of the HEXS is shown in Fig. 1d.

Figure 2a, b show the experimental results of the time-integrated energy distribution of REs obtained with the ESM¹⁸. The slope temperatures were 0.65 MeV for case A (red circles) and 1.7 MeV for case B (green triangles). The slope temperature for case B was more than twice that for case A, even though the peak intensities were very close. The energy distributions of REs obtained for case B (green triangles) and case C (blue squares) were almost identical, even though the peak intensities were different by a factor of four. These slope temperatures cannot be explained using the reported scaling laws, whereby the dependence of the slope temperature on the pulse duration is not considered.

Fig. 1 Experimental conditions and diagnostics. **a** Experimental set-up with geometrical positions of the target, plasma mirror, and the diagnostics instruments, and the ray trace. **b** Temporal intensity profiles of LFEX laser pulses. Pulses temporally stacked to generate various pulse shapes. **c** Schematic view of the high-energy X-ray spectrometer (HEXS) showing the configuration of metallic filters and their thickness. **d** Layer response of the HEXS. Each of the 12 lines corresponds to the response of the image plate behind the different filters

Fig. 2 Relativistic electron (RE) energy distributions measured experimentally and computationally with various intensities and duration of the laser pulses. **a** Comparison of experimental data for cases A (red circles) and B (green triangles), and computational data for cases A (gray dashed line) and B (black solid line). **b** Comparison of experimental data for cases A (red circles) and C (blue squares), and computational data for cases A (gray dashed line) and C (black solid line). Errors of the RE numbers are obtained by convolution of statistical errors and errors ($\pm 5\%$) of uncertainty of the detector response (image plate) for electrons. The error of the electron energy is $\pm 4.5\%$ due to the uncertainty of the calibration

The energy distribution of the low-energy REs was evaluated from HEXS signals by coupling with a Monte Carlo simulation that handles radiation-particle-matter interactions, such as the GEANT4 code¹⁹. Two HEXSs were allocated at 20.9° and 41.8° from the normal direction of the target rear surface. The REs are converted into Bremsstrahlung X-rays in the gold cube. A spectrum of the Bremsstrahlung X-rays reflects the energy distribution of REs moving in the gold cube. The energy

distribution of REs ($f(E)$) was approximated using Maxwell-Boltzmann distribution functions with two different slope temperatures (T_1 , T_2 , and $T_1 < T_2$) and a relative coefficient (A), i.e., $f(E) = A \exp(-E/T_1) + (1 - A) \exp(-E/T_2)$. T_2 was obtained from the ESM in the range of $E > 3$ MeV to reduce the diversity of the estimation process²⁰. A single divergence angle ($\theta_{\text{div}} = 41^\circ$) was used in the GEANT4 computation.

Fig. 3 Number of matching image plates for **a** case A, **b** case B, and **c** case C. In case A, the maximum was obtained as $A = 0.90 \pm 0.10$, $T_1 = 0.3 \pm 0.1$ MeV, and $T_2 = 0.7$ MeV, and 22 of the 24 calculated points matched with the experimental points in this range. In case B, the maximum was obtained as $A = 0.85 \pm 0.05$, $T_1 = 0.35 \pm 0.1$ MeV, and $T_2 = 1.7$ MeV, and 20 of the 24 calculated points matched with the experimental points in this range. In case C, the maximum was obtained as $A = 0.64 \pm 0.11$, $T_1 = 0.92 \pm 0.58$ MeV, and $T_2 = 1.7$ MeV, and all of the 24 calculated points matched with the experimental points in this range

Table 1 Summary of relativistic electron (RE) energy distributions and energy conversion efficiency										
Case	Experiment					Simulation				
	A	T_1 [MeV]	T_2 [MeV]	CE_{low} [%]	CE_{all} [%]	A	T_1 [MeV]	T_2 [MeV]	CE_{low} [%]	CE_{all} [%]
A	0.9 ± 0.10	0.3 ± 0.1	0.7	10.4 ± 1.6	12.5 ± 2.5	0.95	0.3	0.8	7.1	9.6
B	0.85 ± 0.05	0.35 ± 0.1	1.7	15 ± 2.5	27.5 ± 2.5	0.97	0.4	2.5	17	22
C	0.64 ± 0.11	0.92 ± 0.58	1.7	14.5 ± 0.75	17.5 ± 2.5	0.97	0.8	2.5	24	34

The RE energy distribution was approximated using Maxwell-Boltzmann distribution functions with two different slope temperatures (T_1 , T_2 , and $T_1 < T_2$) and a relative coefficient (A). T_1 was obtained from the high-energy X-ray spectrometer (HEXS) and T_2 was obtained from the vacuum electron spectrometer in the range of $E > 3$ MeV. From an absolute comparison between the experimental X-ray signal of HEXS and the calculated signal, the energy conversion efficiency from the laser to the low-energy ($E < 3$ MeV) REs (CE_{low}) and all REs (CE_{all}) can be evaluated

The numbers of the matched points are color coded in Fig. 3a–c. In case A, the maximum was obtained as $A = 0.90 \pm 0.10$, $T_1 = 0.3 \pm 0.1$ MeV, and $T_2 = 0.7$ MeV, and 22 of the 24 calculated points matched with the experimental points in this range. In case B, the maximum was obtained as $A = 0.85 \pm 0.05$, $T_1 = 0.35 \pm 0.1$ MeV, and $T_2 = 1.7$ MeV, and 20 of the 24 calculated points matched with the experimental points in this range. In case C, the maximum was obtained as $A = 0.64 \pm 0.11$, $T_1 = 0.92 \pm 0.58$ MeV, and $T_2 = 1.7$ MeV, and all of the 24 calculated points matched with the experimental points in this range. The lower slope temperature shows little dependence on the pulse duration, and they are in good agreement with the ponderomotive scaling value (0.4 MeV for cases A and B or 1.0 MeV for case C)⁴.

From an absolute comparison between the experimental X-ray signal and the calculated signal, the energy conversion efficiency from the laser to the low-energy ($E < 3$ MeV) REs (CE_{low}), and all REs (CE_{all}) can be evaluated. Details of the analysis are described in the Methods section (Analysis of high-energy X-ray spectrometer). 10.4 ± 1.6 , 15 ± 2.5 , and $14.5 \pm 0.75\%$ of the laser energy is converted to the low-energy REs, and 12.5 ± 2.5 , 27.5 ± 2.5 , and $17.5 \pm 2.5\%$ of the energy is converted to all REs for cases A, B, and C, respectively. CE_{all} increases by a factor of 2.2 by extension of the pulse duration. These results are summarized in Table 1.

Two-dimensional particle-in-cell simulation. The experimental results were compared with those computed using the 2D PIC simulation code (PICLS-2D²¹). Calculations were performed with temporal and spatial scales that were comparable to

the experimental scales. The gold cube was replaced with a $20 \mu\text{m}$ planar plasma with a peak density of $40n_c$, where $n_c = 1.0 \times 10^{21} \text{ cm}^{-3}$ is the critical electron density for $1.053 \mu\text{m}$ wavelength light. The bulk plasma has an exponential density profile from 0.1 to $40n_c$ and a scale length of $1 \mu\text{m}$.

The lower slope temperatures of the REs in the simulation were 0.3, 0.4, and 0.8 MeV for cases A, B, and C, respectively. The higher slope temperatures of the REs in the simulation were 0.8, 2.5, and 2.5 MeV for cases A, B, and C, respectively. 7.1, 17, and 24% of the laser energy was converted to the lower energy component ($E < 3$ MeV) of the REs, and 9.6, 22, and 34% of the energy was converted to all REs for cases A, B, and C, respectively. CE_{all} increases by 2.3 times by extension of the pulse duration. These results are also summarized in Table 1.

Thus, the experimental findings, i.e., that (i) the lower slope temperature is close to the ponderomotive scaling value and is not dependent on the pulse duration; (ii) the higher slope temperature is dependent on the pulse duration; and (iii) the increment of the energy conversion efficiency by extension of the pulse duration, were completely reproduced by the PICLS-2D code.

The PIC simulation also revealed that RE generation in the multi-ps relativistic LPI has two isolated interaction regions, which are essential to explain the characteristics of the absolute energy distributions of REs generated in the multi-ps relativistic LPI.

The lower temperature REs are produced near the relativistic critical density surface. The bulk electron density profile around the relativistic critical density surface is steepened by the ponderomotive pressure of the incident laser, and the lower

Fig. 4 Three examples of relativistic electron (RE) trajectories at two different periods ($t = 3.0\text{--}3.5$ and $5.0\text{--}5.5$ ps) overlaid on the electron densities ((a) and (b)), self-generated azimuthal magnetic fields ((c) and (d)), and self-generated electric fields ((e) and (f)). **g, h** Kinetic energies of REs along the longitudinal position for the two different periods. **i–k** Energy distributions (solid lines) of REs accelerated in case A ($2.0\text{--}2.5$ ps), case B ($5.0\text{--}5.5$ ps), and case C ($2.5\text{--}3.0$ ps)

temperature REs are produced by the relativistic LPI in the steepened interaction zone. Such density steepening remains over the multi-ps timescale, which is why the lower temperature component is independent of the pulse duration.

The majority of the more energetic REs are produced in an underdense plasma. Figure 4a–f show three selected RE trajectories at two different periods ($t = 3.0\text{--}3.5$ and $5.0\text{--}5.5$ ps) overlaid on the electron densities (Fig. 4a, b), self-generated azimuthal magnetic fields (Fig. 4c, d), and self-generated electric fields (Fig. 4e, f). Figure 4a, b are colored using the lookup table of electron density logarithm normalized with respect to the critical density (n_c).

In the earlier period (Fig. 4a, c, e, g), the REs move around the near-critical density region. The electron travels outwardly (the opposite direction of incident laser propagation) through the magnetic and electric fields that are generated by the Biermann battery effect and charge separation. The self-generated electric field decelerates the outwardly moving RE; the RE eventually stops and is then accelerated again inwardly. The effect of the quasi-static electric field not only directly imparts additional energy to the electrons, but also reduces the dephasing rate of the

RE from the acceleration phase of the laser field^{19,22–25}. The RE continues to ride on the acceleration phase, whereby the RE gains energy from the laser field. In this simulation, the RE is accelerated up to 15 MeV by the combination of the quasi-static electric field and the laser field, as shown in Fig. 4g.

In the later period (Fig. 4b, d, f, h), the self-generated magnetic field is sufficiently strong that some of the REs (blue and green trajectories) are reflected outwardly by the $\mathbf{v} \times \mathbf{B}$ force, and they are re-injected to the region where both the self-generated electric field and laser field coexist (Fig. 4b, loop(ii)). In loop (ii), the turning point of the RE is farther from the near-critical density region than that in loop (i) because the RE receives additional kinetic energy in loop (i). After loop (ii), the kinetic energy of the REs reaches beyond 15 MeV, as shown in Fig. 4h. This re-injection mechanism is the LIDA¹⁰.

The solid lines in Fig. 4i–k show energy distributions of REs accelerated for case A ($2.0\text{--}2.5$ ps), case B ($5.0\text{--}5.5$ ps), and case C ($2.5\text{--}3.0$ ps). The histograms show the ratio of the RE numbers between the two groups: one group (red bars) consists of REs that experienced single loop-injection and the other (green bars) consists of REs that experienced multiple loop-injection. The

correlation between multiple loop-injection and energetic RE generation is clearly evident, i.e., the highest energy component of REs in Fig. 4j above 20 MeV is generated predominantly by multiple loop-injection. On the other hand, in case C, multiple loop-injection does not account for energetic RE generation (as shown in Fig. 4k).

Sudden growth of quasi-static magnetic field. The PIC simulation shows that the quasi-static magnetic field is generated by three different mechanisms in case B, which are dependent on the time during the multi-ps relativistic LPI. At the leading edge of the 4 ps flat-top pulse (<2 ps), the ponderomotive force of the incident laser pushes the relativistic critical density surface (γn_c) into the overdense region, and the heated underdense plasma expands into the vacuum. Here $\gamma = \sqrt{1 + (1 + R)a_0^2/2}$, where R is the reflectivity. An electric field is generated at the outer boundary of the expanding plasma (which is referred to as the first electric field.). An azimuthal magnetic field is generated in the overdense plasma due to the $\nabla n \times \nabla I$ effect^{4,26,27}, where n and I are the plasma electron density and laser intensity, respectively. When the laser intensity reaches the plateau at 2.0 ps, plasma evacuation by the laser field is eventually halted by the charge separation due to depletion of the local electron density. The $\nabla n \times \nabla I$ mechanism becomes relatively small, whereas the $\nabla T \times \nabla n$ (Biermann battery) effect^{28,29} becomes the dominant mechanism for generation of the magnetic field. Here, T is the plasma electron temperature. The strongest magnetic field is generated at the edge of the laser spot in the underdense plasma. This magnetic field influences the motion of REs around the near-critical density region. Some of the REs are moved transversely from the laser spot by the $\mathbf{E} \times \mathbf{B}$ drift. The drift current heats the surface of the bulk plasma via the two-stream instability³⁰. The electric field that contributes to the $\mathbf{E} \times \mathbf{B}$ drift is a weak electric field generated in a limited region near the critical density surface. The heated bulk plasma begins to expand at the edge of the laser spot, while the expansion is suppressed at the inside of the laser spot by the laser ponderomotive pressure. The heated bulk plasma surface, which has been flat so far, deforms into a bow shape (which is referred to as a bow-shaped bulk plasma surface). The first electric field is carried out by the plasma expansion, far away from the critical density surface and no longer contributes to the drift.

When the thermal pressure of the heated bulk plasma exceeds the ponderomotive pressure of the incident laser at 3.8 ps, the bulk plasma begins to expand at the inside of the laser spot, and the strong quasi-static electric field (the second electric field) is then generated at the near-critical density region. Figure 5a shows the electric fields in the longitudinal direction (E_x) of the two regions. The second electric field is generated at the expansion front of the heated bulk plasma in the inside of the laser spot. The newly generated strong electric field contributes to the $\mathbf{E} \times \mathbf{B}$ drift by combination with the magnetic field (Fig. 5b). When the REs flow in the plasma, the return-current is driven to maintain current neutrality in the plasma. Figure 5c shows the RE drift current in the lower density region and the return-current flow in the higher density region. In the plasma region where RE current terminates, the electric field is enhanced by the inflow of electrons (Fig. 5d). The current loop produced by the spatial separation between the RE drift current and the return-current generates a magnetic field along the outer edge of the bow-shaped bulk plasma surface (Fig. 5e).

The positive feedback between the growth of the fields and the field-driven drift current results in the rapid growth of the quasi-static electric and magnetic fields with time (Fig. 5a–h). This third magnetic field (30–50 MG) is stronger than the magnetic field generated by the $\nabla T \times \nabla n$ effect (<10 MG).

Figure 6a–f show a comparison of the pulse shapes (black lines), the temporal evolution of the maximum energy of the REs (lines between circles) for cases A (red), B (green), and C (blue), and the temporal evolution of the average strength of the quasi-static electric (Fig. 6a–c) and magnetic (Fig. 6d–f) fields in the underdense plasma (gray lines between squares). The temporal evolution of the average strength of the quasi-static electric and magnetic fields in the underdense plasma is similar to the laser pulse shapes for case A (red). Sudden onset of the quasi-static electric and magnetic fields appears in cases B (green) and C (blue). In case C, the onset appears just after the laser intensity reaches its peak at 2.0 ps because the sudden growth is driven by plasma expansion during the trailing edge of the laser pulse. The laser intensity rapidly decreases before the growth of the quasi-static magnetic field necessary for LIDA. Thus, LIDA does not account for the energetic RE generation. On the other hand, the laser intensity is maintained after the sudden onset of growth in case B; therefore, strong quasi-static electric and magnetic fields are generated by the positive feedback between the growth of the fields and the $\mathbf{E} \times \mathbf{B}$ drift current. In such a situation, the SP-REs are generated predominantly by LIDA. Thus, SP-RE acceleration in case B not only has a process that gradually progresses with time, but also has a process that progresses in a threshold manner. This has not been pointed out in previous studies on RE acceleration by multi-ps laser pulses^{8,9,16,31,32}.

The transition of the acceleration mechanism to LIDA is essential in boosting SP-RE acceleration. There are other competing mechanisms to accelerate SP-REs; however, in the multi-ps relativistic LPI, SP-REs are predominantly produced by the two mechanisms discussed above, as detailed in Supplementary Note 1.

Transition of acceleration mechanism. SP-RE acceleration is started when the plasma thermal pressure exceeds the laser ponderomotive pressure. Figure 7a shows the evolution of an initially exponential plasma profile during interaction with a high-intensity laser pulse. The color map shows the electron density ($\log_{10}(n_e/n_c)$) and the red solid line shows the temporal intensity profile of the laser. At $t = 3.8$ ps, the motion of the relativistic critical interface stops even though the laser pulse is still irradiated, and the state of the relativistic LPI transits from the hole-boring phase to the blowout phase.

The position of the interface that interacts with the laser pulse having an arbitrary intensity temporal profile is obtained by integrating the velocity of the interface with respect to time³³:

$$x_i(t) = x_c(0) + 2l_s \ln \left[1 + \frac{c}{2l_s} \sqrt{\frac{R \cos \theta}{(1+R)} \frac{Zm_e}{M_i}} \int_{t_0}^t \left(\frac{\gamma(t)^2 - 1}{\gamma(t)} \right)^{1/2} dt \right], \quad (1)$$

where we assume an initial plasma density profile as $n_e(x) = \gamma n_c \exp[(x - x_c)/l_s]$ with a scale length l_s , x_c is the position of the critical density, R is the reflectivity, θ is the laser incident angle, Z is the ion charge, M_i is the ion mass, m_e is the mass of an electron at rest, and γ is the relativistic factor of an electron. Note that x_c should vary with time. However, here we use the initial position of the critical density, i.e., $x_c = x_c(0) = \text{const}$, considering that the temporal profiles of realistic lasers increase from 0 to the peak intensity.

The transition time of the relativistic LPI state from the hole-boring phase to the blowout phase can be obtained by coupling Eq. (1) with the hole-boring limit density, which is derived from the momentum transfer equation for the stationary state of the

Fig. 5 Mechanism for the sudden growth of quasi-static electric and magnetic fields. When the thermal pressure exceeds the ponderomotive pressure, the bulk plasma begins to expand, and the strong quasi-static electric field (a) is then generated at the near-critical density region. The newly generated strong electric field contributes to the $\mathbf{E} \times \mathbf{B}$ drift by combination with the magnetic field (b). When the relativistic electrons (REs) flow in the plasma, the return-current is driven to maintain current neutrality in the plasma (c). In the plasma region where RE current terminates, the electric field is enhanced by the inflow of electrons (d). The current loop produced by the spatial separation between the RE drift current and the return-current generates a magnetic field along the outer edge of the bow-shaped bulk plasma surface (e). The positive feedback between the growth of the fields and the field-driven drift current results in the rapid growth of the quasi-static electric and magnetic fields with time ((c)–(h))

Fig. 6 Comparison of the pulse shapes (black lines), the temporal evolution of the maximum energy of the relativistic electrons (lines between circles) for cases A (red), B (green), and C (blue), and the temporal evolution of the average strength of the quasi-static electric ((a)–(c)) and magnetic ((d)–(f)) fields in the underdense plasma (gray lines between squares)

Fig. 7 Transition time to super-ponderomotive electron acceleration. **a** Evolution of the initially exponential plasma profile during interaction with a laser pulse having a normalized laser intensity of $a_0 = 1.79$ ($I\lambda^2 = 4.0 \times 10^{18} \text{ W } \mu\text{m}^2 \text{ cm}^{-2}$). The motion of the interface calculated from Eq. (1) (black dashed line) is in good agreement with the motion obtained by the particle-in-cell simulation (black solid line) until $t = 3.8$ ps. **b** Transition time for constant intensity temporal profile calculated from Eq. (3) for a gold target with a preformed plasma scale length of $l_s = 1 \mu\text{m}$ in the charge state of $Z = 40$. Correction factor of the pulse temporal profile F_c for cases of **c** low reflectivity $R = 0.3$ and **d** high reflectivity $R = 0.7$. The normalized laser intensity a_0 or half width at half maximum (HWHM) of the Gaussian leading edge increases; therefore, a larger correction factor is required

interface³⁴:

$$\frac{n_s}{n_c} = 8a_0^2 \left[\frac{1+R-(1-R)\beta_h^{-1}\alpha^{-1}}{2} \right], \quad (2)$$

where β_h is the ratio of the drift velocity of REs (v_h) to the speed of light, c . $\alpha \equiv ir/2$ is the geometrical factor, where $r = 1$ and 2 for non-relativistic and relativistic Maxwell momentum distributions, respectively, and $i = 1, 2$, or 3 represents the dimension of the momentum distribution. When we assume a 1D condition ($\alpha = 1$) and the relativistic limit $\beta_h = 1$, Eq. (2) is reduced to $n_s/n_c = 8Ra_0^2$.

By substituting $a_0 = 1.79$ and $R = 0.7$, the electron density threshold n_s for the experimental condition of case B in Fig. 7 is obtained as $n_s = 17.9n_c$. This density is almost identical to the electron density threshold in which the plasma compression terminates in the PIC simulation. Substituting Eq. (2) and the initial electron density profile ($n_e(x) = n_c \exp[(x - x_c)/l_s]$) into

Eq. (1), the transition time is obtained as

$$t_{s0} = \left[\frac{2l_s}{c} \left\{ \sqrt{4\epsilon^2 a_0^2 [1 + R - (1 - R)\beta_h^{-1}\alpha^{-1}]} - 1 \right\} \sqrt{\frac{(1+R)am_e Z^*}{R \cos \theta}} \frac{y}{Zm_e} \frac{y}{y^2 - 1} \right] + t_0, \quad (3)$$

for the case where the laser intensity is constant in time. Here, t_0 is the time when the normalized laser amplitude a_0 reaches 1, $M_i = \alpha^* m_p Z^*$ represents the ion mass, m_p is the proton mass, Z^* is the ion charge number for the fully ionized state, and $\alpha^* = 1$ for hydrogen and $\alpha^* = 2$ for other species.

Here, we introduce the correction factor F_c to take the laser pulse profile into account as $t_s = (t_{s0} - t_0)F_c + t_0$, where t_s is the transition time for the time-dependent laser intensity. For cases where the laser intensity is constant in time, $F_c = 1$. The lines in Fig. 7b show the transition time calculated from Eq. (3) with $F_c = 1$ for various reflectivities. Here, we assumed that a gold plasma ($Z^* = 197$) with the preformed plasma scale length

$l_s = 1 \mu\text{m}$ in the charge state of $Z = 40$ interacts with a linearly polarized laser ($\varepsilon = 1$). The spot size of the LFEX laser is large; therefore, it is assumed that electron acceleration occurs one-dimensionally ($\alpha = 1$). As an approximate trend, when the normalized laser intensity a_0 or reflectivity R increases, the transition time is delayed. Low reflectivity reduces the effective laser intensity at the interface and reduces the hole-boring limit density. When the normalized laser intensity is $a_0 = 1.79$ (intensity is $I\lambda^2 = 4.0 \times 10^{18} \text{ W } \mu\text{m}^2 \text{ cm}^{-2}$), the transition time is estimated to be $t_s = 2.8 \text{ ps}$, which is in good agreement with the simulation result.

In an experimental case, the laser intensity increases in time with the Gaussian profile, so that the transition time is delayed compared with that obtained for $F_c = 1$. The color maps in Fig. 7c, d show the dependence of the correction factor F_c on the normalized laser intensity a_0 and the half width at half maximum (HWHM) of the Gaussian leading edge for the low reflectivity ($R = 0.3$) and high-reflectivity ($R = 0.7$) cases. A larger correction factor is required as the normalized laser intensity or HWHM increases. In the present range of $1 \leq a_0 \leq 6$ and $0 \text{ ps} \leq \text{HWHM} \leq 1.8 \text{ ps}$, the correction factor increases only ~ 1.2 times at the maximum; therefore, it is sufficient to use Eq. (3) with $F_c = 1$ for a rough estimation of the transition time.

Discussion

A new regime of LPI studies, i.e., multi-ps relativistic LPI, has been realized due to the development of kilojoule-class high-power lasers. Although electron acceleration using a conventional sub-picosecond laser pulse has been explained theoretically as the interaction of a single electron with a laser field, it is necessary to consider the collective effect of electrons when the pulse duration reaches the multi-ps range. Energetic RE generation was experimentally clarified with an average energy far beyond the ponderomotive scaling using the prepulse-free LFEX laser. During the multi-ps interaction, a quasi-static electric field is generated by plasma expansion. In addition, a quasi-static magnetic field is gradually generated due to three different mechanisms of the $\nabla n \times \nabla I$ effect, the $\nabla T \times \nabla n$ effect, and a loop current driven by the $\mathbf{E} \times \mathbf{B}$ drift. The third mechanism of the current loop rapidly amplifies the magnetic field strength by positive feedback between the electric and magnetic fields and the field-driven drift current. Under the quasi-static electric field, REs are accelerated efficiently above ponderomotive scaling by the laser field because the dephasing rate of the REs from the laser field is reduced. Furthermore, when the quasi-static magnetic field becomes sufficiently strong to reflect REs back to the relativistic LPI region, the reflected REs gain additional energy. The boosting timing of electron acceleration by the LIDA mechanism is related to the transition time of the relativistic LPI state from the hole-boring phase to the blowout phase. The equation for the transition time can be derived from the equations for the motion of a relativistic critical density interface and the equations for the electron density where the hole-boring terminates. Finally, the multi-ps LPI shows two positive aspects. One is the efficient production of more energetic REs, which is required for ion acceleration³⁵ and electron-positron pair generation³⁶. The other is that the efficient production of less energetic REs ($< 3 \text{ MeV}$) is maintained, which is critical for fast ignition³⁷. Extension of the heating laser pulse duration seems to work for the efficient deposition of a few tens of kilojoules of energy in a pre-compressed fusion fuel core with low-temperature REs produced by the optimal intensity heating laser (ca. $10^{19} \text{ W cm}^{-2}$).

Methods

Selection of target size. The thickness of the gold cube is an important parameter to investigate RE acceleration by multi-ps relativistic LPI. The REs generate a

sheath electric field at the rear surface of the target. This sheath field refluxes especially low-energy REs and the refluxed REs are re-injected to the acceleration region. This recirculation process also generates SP-REs, which has been investigated by Yogo et al.¹⁶ and Iwata et al.³². One cycle of the recirculation process takes at least 6.7 ps in the 1 mm thick gold cube, which is longer than the pulse durations (1.2 or 4.0 ps) in this experiment; therefore, the recirculation process can be eliminated from the SP-RE mechanisms in this study.

Plasma mirror implementation. The LFEX parabola cannot be focused at an offset position far from the target chamber center due to its mechanical limitations; therefore, a spherical PM was used that allows the original focal pattern to be relayed at an offset position with respect to the target chamber center. A spherical concave mirror (50.8 mm diameter and 202 mm curvature length) with a $1.053 \mu\text{m}$ anti-reflection coating on both surfaces was placed after the focus point, as shown in Fig. 1a. The LFEX was focused at 3 mm above the target chamber center (offset position). The image at the offset position is relayed to the target chamber center with an image magnification of 1 by the spherical mirror. According to ray-trace code calculations, the deterioration of the image due to spherical aberration and astigmatism of the spherical mirror is negligible compared to the $70 \mu\text{m}$ diameter of the LFEX spot. The laser energy fluence on the PM surface was optimized to be 90 J cm^{-2} to obtain acceptable reflectivity (50%) and spatial uniformity of the reflected pulse³⁸. The contrast ratio of the LFEX laser pulse was improved by two orders of magnitude down to 10^{11} by implementation of the PM³⁹ at 150 ps before the main pulse. These clean intense laser pulses create an ideal situation where the REs are accelerated predominantly in the inherent plasma formed by the main laser pulse itself within the picosecond time range. The density scale length of the preformed plasma was calculated to be $1.5 \mu\text{m}$ at 10 ps before the intensity peak using a 2D radiation hydrodynamics simulation with the PINOCO-2D code⁴⁰.

Analysis of high-energy X-ray spectrometer. A high-energy X-ray spectrometer (HEXS) consists of differential X-ray filters and dosimeters that are alternatively stacked in a radiation shield case (Fig. 1c)^{41,42}. The dosimeters are image plates (IPs), which were scanned with an absolutely calibrated IP scanner (Typhoon FLA-7000)⁴³. An IP signal can be converted to an X-ray energy absorbed in the IP layer with a calibration coefficient $(3.28 \pm 0.31) \times 10^{-13} \text{ J PSL}^{-1}$. PSL is a local unit used in the IP scanner for photostimulated luminescence intensity. An experimentally measured X-ray dose of the M -th IP in the N -th HEXS is denoted as $D_{N,M}^{\text{expt}}$, where M is the order number of the dosimeter ($M = 1, 2, \dots, m = 12$) and N is label number of the HEXS ($N = 1, n = 2$). Red solid circles in Fig. 8a–f in the main article are X-ray doses normalized with respect to that of the first dosimeter as $D_{N,M}^{\text{expt}}/D_{N,1}^{\text{expt}}$. The uncertainty of the measured X-ray dose was ± 0.131 of the median value.

X-ray doses on the M -th IP in the N -th HEXS $D_{N,M}^{\text{calc}}$ were calculated with changing values of A and T_1 of the energy distribution function, $f(E)$. The relative coefficient A is changed from 0 to 1 with an interval of 0.01, and the lower slope temperature T_1 is changed from 0.3 MeV to T_2 with an interval of 0.1 MeV. We attempted to find combinations of A and T_1 to maximize the number of matched points, at which the calculated dose ($D_{N,M}^{\text{calc}}/D_{N,1}^{\text{calc}}$) is in agreement with the experimental values ($D_{N,M}^{\text{expt}}/D_{N,1}^{\text{expt}}$) within the measurement uncertainty ($\eta = 0.131$).

From an absolute comparison between the experimental $D_{N,M}^{\text{expt}}$ and calculated $D_{N,M}^{\text{calc}}$ data, the total number of REs $N^{\text{expt}}(A, T_1)$ can be obtained with

$$N^{\text{expt}}(A, T_1) = \left[\frac{1}{n} \sum_{N=1}^n \left\{ \frac{1}{m} \sum_{M=1}^m \frac{D_{N,M}^{\text{expt}}}{D_{N,1}^{\text{expt}}} \right\} \right] \cdot N^{\text{calc}}. \quad (4)$$

Here, $N^{\text{calc}} = 10^9$ in our GEANT4 simulation. $N^{\text{expt}}(A, T_1)$ is derived by taking an average for m -th IPs ($m = 12$) equipped in n -th HEXSs ($n = 2$).

The energy conversion efficiency from laser to the lower energy component REs (CE_{low}) and both energy components of REs (CE_{all}) are given by

$$CE_{\text{low}} = \int_0^{3 \text{ MeV}} Ef(E)dE \cdot \frac{N^{\text{expt}}(A, T_1)}{E_L}, \quad (5)$$

and

$$CE_{\text{all}} = \left\{ \frac{A}{T_{\text{comb}}} \cdot T_1 + \frac{(1-A)}{T_{\text{comb}}} \cdot T_2 \right\} \frac{N(A, T_1)}{E_L}. \quad (6)$$

Here, E_L is the laser energy and $T_{\text{comb}} = A \cdot T_1 + (1-A) \cdot T_2$. The curves shown in Fig. 8a, d were calculated for three combinations of A , T_1 , and T_2 : $A = 0.91$, $T_1 = 0.3 \text{ MeV}$, and $T_2 = 0.7 \text{ MeV}$ (blue solid line); $A = 1$ and $T_1 = 0.3 \text{ MeV}$ (orange solid line); and $A = 0$ and $T_2 = 0.7 \text{ MeV}$ (green dashed line). The curves shown in Fig. 8b, e were calculated for three combinations of A , T_1 , and T_2 : $A = 0.85$, $T_1 = 0.35 \text{ MeV}$, and $T_2 = 1.7 \text{ MeV}$ (blue solid line); $A = 1$ and $T_1 = 0.35 \text{ MeV}$ (orange solid line); and $A = 0$ and $T_2 = 1.7 \text{ MeV}$ (green dashed line). The curves shown in Fig. 8c, f were calculated for three combinations of A , T_1 , and T_2 : $A = 0.64$, $T_1 = 0.92 \text{ MeV}$, and $T_2 = 1.7 \text{ MeV}$ (blue solid line); $A = 1$ and $T_1 = 0.92 \text{ MeV}$ (orange solid line); and $A = 0$ and $T_2 = 1.7 \text{ MeV}$ (green dashed line).

Fig. 8 Comparison between relative X-ray doses experimentally recorded on image plates in high-energy X-ray spectrometers located at 20.9° and 41.8° from the normal direction of the target rear surface for cases A ((a), (b)), B ((c), (d)), and C ((e), (f)). Three lines (blue, green, and orange lines) were calculated for three cases of relativistic electron energy distribution. Errors in the measured doses are caused by the uncertainty of the image plate response calibration for high-energy X-rays

As shown in Fig. 8a–f, the experimentally measured doses, especially at the 9th to 12th points, cannot be reproduced with the single slope higher temperature (T_2) obtained with the ESM (green dashed lines). This discrepancy indicates the existence of a lower temperature component that cannot be measured with the ESM. A combination of ESM and HEXS would thus be a useful technique to quantitatively evaluate energy distributions of REs produced by relativistic LPI. The two-temperature Maxwell–Boltzmann function is thus a better solution to reproduce experimental results obtained with both ESM and HEXS.

Detailed conditions of particle-in-cell simulation. Artificial boundary conditions were applied to the rear surface of a 20 μm thick layer to prevent electron recirculation in the simulation. The rear surface reflects electrons with kinetic energies of <511 keV. The remaining electrons with kinetic energies of >511 keV escape from the rear surface. These boundary conditions enable appropriate simulation of the electron transport in the massive target that was used in the experiment, using a 20 μm thick layer within an acceptable computational time. In addition, the escape condition was applied to the boundary in the transverse direction. Under this condition, gyrating electrons escape from the computation system when the gyration length exceeds half of the system size. Therefore, simulations with such a boundary condition underestimate the maximum energy of the SP-REs. In the present simulation, the transverse system size is three times larger than the laser spot size. Therefore, the effect of electron escape is not expected to be significant in the simulation.

Owing to computational limitations, the ionization degree was fixed to be +40 in the PICLS-2D simulation, which was determined based on the result of a one-dimensional PICLS simulation with the dynamic ionization model of gold described by field ionization⁴⁴ and fast electron collisional ionization⁴⁵. The ionization degree rose from +10 (given by the radiation hydrodynamic code PINOCO-2D) to around +40 for the first several picoseconds.

Dynamics of the interaction surface. We have previously derived the transition time from the hole-boring phase to the blowout phase t_b , under constant laser irradiation (Eq. (8) in ref.³⁴). Here, the equation of transition time is extended to a laser pulse with an arbitrary intensity temporal profile and the theoretical result is compared with that obtained by PIC simulation.

At the interface, plasma is pushed into a high-density region by the laser ponderomotive pressure caused by hole boring. The hole-boring velocity is

conventionally derived with assumption of the initial and terminal states of the plasma components (ions, bulk electrons) and REs^{33,46–49}. It is assumed that the laser is reflected at the interface, which is moving with velocity v_p , and that electrons and ions are initially immobile. The flow velocity of REs is assumed to be v_h .

In a frame moving with the interface at velocity v_p , ions and bulk electrons drift at $-v_p$ toward the interface, where they are reflected. $1 - f_e$ is the fraction of electrons reflected elastically to the velocity $+v_p$ at the interface. The remaining fraction f_e of electrons are heated by the laser and accelerated to relativistic velocities $p_h/\gamma m_e$. All ions are assumed to be reflected elastically to $+v_p$, which is the same assumption as that made by Vincenti et al.⁴⁸. The equations of the momentum flux conservation and the energy flux conservation are given by $(1 + R)I(t)/c \cos \theta \approx 2M_i n_i(t) v_p^2 + f_e n_e(t) v_h(t) p_h(t)$ and $(1 - R)I(t) \cos \theta = f_e n_e(t) v_h(t) p_h(t) c$, respectively. Here, R is the reflectivity of the incident laser on the plasma and θ is the laser incident angle. M_i and n_i (m_e and n_e) are the ion (electron) mass and number density, respectively. Reflection and density steepening occur at the relativistic critical electron density $\gamma(t) n_c$ with $\gamma(t) = \sqrt{1 + (1 + R)a_0^2(t)/2}$, where a_0 is the normalized laser field amplitude. The initial electron density profile is assumed to be $n_e(x) = (\gamma n_c) \exp[-(x - x_c(0))/l_e]$ with scale length l_e . Note that in ref.³⁴, $\cos \theta = 1$ and $f_e = 1$ are assumed.

The dashed lines in Fig. 7a indicate the motion of the interface calculated by Eq. (1) with various reflectivities. The black dashed line ($R = 0.7$) reproduces the motion obtained by the PIC simulation until $t = 3.8$ ps. This result shows that the velocity of the interface at each time can be determined by the momentum and energy balance among the laser, ions, and hot electrons, and that the integration of the interface velocity, Eq. (1), explains the motion of the interaction surface.

Data availability

The datasets analyzed during the current study are available from the corresponding author upon reasonable request. In accordance with the guideline for research data storage at the Institute of Laser Engineering, Osaka University, all data are properly stored in the SEDNA database system.

Code availability

The computer codes used in the current study are accessible from the corresponding author upon reasonable request.

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Author contributions

S.K. and S.F. are the principal investigators who proposed and organized the experiment. M.H. performed the PIC simulations in collaboration with T.J. and H.S., N.I., S.K., M.H., and Y.S. developed the theoretical model. Y.A. and A.M. designed and constructed the large size plasma mirror. H.M. and Y.O. conducted the theoretical analysis. H.N. and A.S. performed the radiation hydrodynamic simulations. K.M., S.S., S.L., K.F.F.L., and Y.A. measured electron energy distributions of high-energy components with assistance from T.O.S.T. measured electron energy distributions of low-energy components with assistance from Z.Z. and A.Y.S.T. and J.K. were in charge of the LFEX laser facility development in ILE. M.N., H.N., H.S., and H.A. supervised the project and provided overall

guidance. All authors participated in the discussions and contributed to the preparation of the manuscript.

Additional information

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