Three-dimensional Imaging of Nanoplasma by Ion Nanoscopy

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Abstract

The remarkable abilities of laser-irradiated nanostructures to emit X-ray photons, accelerate charged particles, and launch shock waves for multiple applications are primarily governed by a localized plasma near the surface. However, the full-spatially resolving the localized plasma is still challenging due to the difficulty of measuring the three-dimensional (3D) momentum of ions generated from partially or fully ablated nanostructures in high-intensity laser fields. Here, we bridge this gap by introducing ion nanoscopy based on the tomographic reconstruction of the 3D momentum of ions emitted from constantly refreshed aerosolized TiO₂ nanospheres. The measured 3D ion momenta map the localized plasma charge well through an electrostatic repulsion model. A pronounced variation in localized plasma charge across the nanosphere surface is obtained, which also extends the idea of the combined action of external and internal fields on plasma formation. This work represents an advance in understanding plasma generation from nanostructures and in the probing, visualization, and manipulation of localized nanoplasmas.

Introduction

Plasma is known as the fourth state of matter. When its size shrinks to the nanoscale, it is possible to utilize the arising exotic physical effects such as ultrahigh-energy densities and extreme radiation¹⁻³. While being a dream of theoreticians for decades, nanoplasma generation has only recently become an affordable and reliable experimental reality⁴. This advance is underpinned by the advent of table-top femtosecond laser sources capable of increasing the energy density delivered to the interface of a nanostructure. Nanostructured targets are of particular interest because they improve the coupling of laser energy to the target. On the other hand, they feature size-, composition-, orientation-, and shape-dependent effects in disassembling a solid lattice for plasma generation⁵⁻⁸. These effects provide more degrees of freedom to introduce nanometer site sensitivity to plasma and control the momentum of ejected charged species. It has been shown that the direction-controlled ion shock wave produced by a spatially localized nanoplasma can destroy a tumor with minimal damage to nearby tissues⁹⁻¹¹. Similar opportunities exist to further enhance plasma-assisted catalytic activity and etching^{8,12}. Highly spatially varying nanoplasmas, with optical control possibilities, also serve as an efficient ion or X-ray source for imaging applications^{13,14}. Therefore, the 3D visualization of spatially varying nanoplasma charge density is essential to understand, control, and optimize the performance of nanoplasma-based applications.

At very low radiation intensities, much progress has been made experimentally in understanding how the nanostructure affects localized plasmons¹⁵⁻¹⁷, owing to the development of detection solutions for the plasmonic spatial distribution from two-dimensional (2D) planar (such as photoemission electron microscopy and near-field scanning optical microscopy) to three-dimensional (3D) planes¹⁸⁻²³. However, we cannot apply these technologies to image nanoplasmas generated at high radiation intensities because the underlying physics for plasmons is different from that for plasma generated at high radiation intensities as discussed here. At low radiation intensities, plasmons increase the strength of the

localized surface field, while the nanostructure remains in the solid state despite significant electron delocalization. In contrast, for the case of high radiation intensities discussed here, the ion-ion bonds are weakened in the phase transition from plasmonic excitations to nanoplasma states, which leads to a partially or entirely damaged nanostructure^{4,6,24}.

By introducing nanoparticle aerosol generation technology, a fresh sample can be provided for each laser shot^{25,26}. Therefore, the applicable laser intensity can be dramatically increased without worrying about sample deterioration. Further combining traditional 2D velocity map imaging (VMI) metrology, a size-dependent forward-backward asymmetry for ion emission from NaCl nanoparticles ionized by intense near-infrared laser pulses was observed and explained by an enhanced internal electric field inside the nanoparticles in the forward direction of the propagating laser pulses⁵. Although the external field is also believed to contribute to nanoplasma formation, previous studies have shown that it is difficult to separate the role of the external field from the role of the internal field^{6,27}. This ambiguity stems from both internal and external factors being simultaneously enhanced in the forward propagation direction of the laser pulses. Thus, the asymmetric analysis along the propagation direction obtained in a reduced-dimension measurement is not enough to distinguish between the two.

This study merges tomography technology into aerosolized 2D VMI for the reconstruction of the 3D ion momentum distribution generated from laser-irradiated titanium dioxide (TiO₂) nanospheres. By employing the intensity binning method, we can ensure that each 2D image is recorded at the same intensity for 3D reconstruction. The obtained anisotropic 3D ion momentum distribution maps outline the localized plasma charge density on the surface of the nanosphere, as well as the enhanced internal and external fields near the surface. The mapping relationships are modeled and reproduced by electrostatic calculations and Mie scattering theory with finite difference time domain (FDTD) calculations. These results provide a new understanding of plasma generation in nanospheres and clarify the different roles of the internal and external fields. In addition, the possibility of all-optical control of localized plasma density on the surface of nanospheres is also demonstrated, the validity of which is verified by our 3D visualization method.

Results

Three-dimensional velocity-map-imaging results

We combined the tomographic reconstruction method and aerosol VMI technique. Therefore, the information lost along the axis perpendicular to the detector is added for ions emitted from laser-irradiated TiO₂ nanospheres. A schematic illustration of the experiment is shown in Fig. 1, and the detailed experimental setup and steps are described in the Methods section. Briefly, a stream of isolated 100-nm TiO₂ nanospheres is first delivered to interact with a 400-nm linearly polarized (LP) or circularly polarized (CP) laser field by aerodynamic lens focusing. Then, the emitted ions are focused onto a microchannel plate (MCP) and phosphor screen detector through an integrated electrostatic lens to obtain a 2D ion momentum distribution, as shown in Fig. 1a. Second, the polarization of the incident

laser is rotated over a 180-degree range, and the 2D VMI images are recorded for a step size of 1 degree, as depicted in Fig. 1b. It is worth mentioning that the intensity binning technique avoids focal volume averaging. In this technique, a 'bin' is defined as a portion of the hit histogram that corresponds to similar ion yields (see Fig. 1c). As the predicted ion yields are proportional to the laser intensity, the number of emitted ions per laser shot serves as a guide for near-single intensity sampling of the nanosphere interaction²⁸. This data discrimination provided us with each ion image used for reconstruction under the same experimental conditions. Finally, an inverse radon transform is performed to integrate those 2D images with the ion emission number of 1400 and obtain a reconstructed 3D ion momentum distribution²⁹ (see Methods for details).

We project the reconstructed 3D ion momentum spectra for the LP and CP lasers onto two different planes, the detector plane (x-z plane) and the plane perpendicular to the laser propagation direction (y-z plane), as shown in Figs. 2a-d. These projections allow us to represent 3D distribution in a 2D environment for further analysis purposes. For the LP laser whose polarization is perpendicular to the detector plane, the ion emission is dominant along the laser polarization direction (z-axis), as shown in Fig. 2a. This asymmetric distribution in the y-z plane is also verified in the angular resolved ion yield shown in Fig. 2e, where the polar angle α (0° - 360°) is the counterclockwise angle from the y-axis. We emphasize that this emission asymmetry in the y-z plane cannot be observed in conventional 2D VMI detection due to the lack of data on the y-axis. To quantify these asymmetric ion distributions in the y-z plane, we introduce a vertical to horizontal ratio $\gamma_{y-z} = I_z/I_y$, where I_y and I_z are the integrated ion yields along the y-axis (0°) and the z-axis (90°), respectively. For the LP laser, the ratio γ_{y-z} in Fig. 2e is 1.41. We would expect that the ratio γ_{y-z} goes to 1 in the isotropic limit. When the nanosphere is exposed to the CP laser, the ion distribution becomes more symmetric, as shown in Figs. 2b and 2f, and the ratio γ_{y-z} is reduced to 0.99, which is expected due to a more uniform excitation. It is also noticed that the ion distributions for both the LP and CP lasers in the x-z plane are asymmetric along the laser propagation direction (x-axis). Although Hickstein and Antonsson^{5,6} have previously reported similar forward-backward asymmetry, whether the internal field or the external field is the decisive factor remains unclear due to the previous lack of direct mechanistic insight into the internal and external field excitation processes.

Plasma imaging results

One would expect that this anisotropic ion momentum distribution measured in our experiment correlates with the charge distribution on the surface of nanospheres during plasma formation²⁷. Two reasons support this speculation: First, when small aerosol nanospheres are irradiated with femtosecond lasers, the appearance of ion emission itself is considered a significant sign of laser-induced plasma formation⁴. Second, continuous ionization removes electrons from plasma to infinity, which leads to a gradual buildup of positive charges on the surface of the nanosphere³⁰. Thus, the Coulomb explosion sets in, converting the energy deposited by the laser in the nanosphere into the ion kinetic energy through the electrostatic repulsion². Since the removal of nanoplasma electrons occurs before the ion expansion

sets in notably, we can easily separate the time scales for ionization and Coulomb explosion. The release of electrons from the nanosphere is highly nonlinear, so this process is temporally confined to the laser pulse duration, usually on the femtosecond time scale. The emitted electrons mainly gain energy from direct acceleration or rescattering on the nanosphere surface in the presence of local near-fields^{19,31-33}. In contrast, the much heavier ions are almost undisturbed by the enhanced field around the nanosphere. Their final energy mainly arises from long-range electrostatic repulsion^{22,34}, which occurs over the time scale of picoseconds. It is also worth mentioning that the Coulomb attraction between the fast escaping electrons and the slow ions created on the nanosphere is negligible and has no significant effect on the final ion momenta²². Since the trajectories of the ions are also hardly affected by the laser electric field, a relatively simple and straightforward static electrostatic model³⁴ is then applicable to correlate the ion momentum distribution with the charge distribution on the surface of the nanosphere.

This model simplifies the interaction between the radially emitted **monovalent cation** and the plasma charge accumulated on the surface with the electrostatic repulsion between two point charges. Thus, a repulsive model along the radial direction can be employed to capture the essence of the ion dynamics. As shown in Fig. 3a, a fixed point charge Q situated in the center of the nanosphere is set to represent the buildup of positive charges on the surface. An ion is launched from the surface on the lines emanating radially from the charge Q . Then, the subsequent motion of emitted ions is described by Newton equations. With the free parameter charge Q , we can reproduce the measured maximal ion momentum (cutoff) integrated along a specific radial direction defined by elevation φ and azimuthal angles θ and obtain the corresponding surface charge via the learning inversion algorithm (see Methods for details). As shown in Figs. 3b and 3c, the points of maximum enhancement (hot spot) for both ion yield and surface charge distributions manifest a dipole-like character along the laser polarization direction in the LP laser field. We also notice that the hot spots for the two distributions bend forward in the laser propagation direction. We next consider the ion yield and surface charge distributions in the CP laser field. In contrast to the LP laser field, a ring-like plasma charge distribution is observed for the CP laser field, as shown in Fig. 3d. This ring-like structure is also perfectly reproduced in the ion yield distribution shown in Fig. 3e. In both cases of LP and CP laser fields, the similar distributions between the ion yield and the surface charge suggest a close relationship between the location of plasma charge and the emitted ion in momentum space.

Discussion

By detecting the ion momentum distribution generated from a laser-irradiated nanoparticle, earlier work has concentrated on providing information about localized light absorption, Hickstein and Antonsson connected forward-backward asymmetric ion emission to an enhanced inhomogeneous internal electric field^{5,6}. However, the ion momentum distribution and internal field distribution are only qualitatively consistent because the connection between these two distributions is indirect. The field distribution directly determines the plasma charge distribution rather than the ion momentum distribution. Moreover, as the wavelength of the driven laser becomes similar to or smaller than the size of the nanospheres, Mie

scattering theory predicts a shift of the maximal field enhancement in the direction of the light propagation for the external field. Thus, the forward-backward asymmetric ion momentum distribution may also stem from the inhomogeneous external electric field. This result indicates that the internal and external contributions and fields for plasma formation are indistinguishable from a simple analysis of the forward-backward asymmetric 2D ion momentum distribution. In contrast, we have well established the mapping relationship between the measured ion momentum distribution and plasma charge distribution in the present study. Therefore, the internal and external fields can directly associate with the plasma charge distribution to reveal their roles. It should be noted that the measurement of the 3D ion momentum distribution is a prerequisite for the reconstruction of the plasma charge distribution with the electrostatic repulsive model.

For further insight into the roles of internal and external fields, we employed FDTD Solutions software to simulate the 3D electric field distributions for 100-nm TiO_2 spheres exposed to 400-nm LP and CP laser fields, as shown in Fig. 4. The results show some clear differences between the internal and external fields. When the nanospheres are exposed to the LP laser field, the central slice of the external field distribution in the x-z plane shows a dipole-like character along the polarization direction with a shift of the region for maximal field enhancement in the direction of light propagation (Fig. 4a). Nevertheless, for the internal field, the 100-nm TiO_2 spheres act like a lens that focuses the light and forms one hotspot in the forward propagation direction in the x-z plane (Fig. 4c). For the CP laser field, similar differences between external and internal field distributions are also observed in the x-z plane (Fig. 4e and 4g). In the y-z plane, the dipole-like distribution is absent. Thus, both the external and internal field distributions for the nanospheres are governed by only the polarization states of the driving light. The field distributions are dipole-like in the LP laser field (Figs. 4b and 4d) and uniform ring-like in the CP laser field (Figs. 4f and 4h).

Therefore, the differences in internal and external field distributions make it possible to identify their roles in plasma generation by comparing the plasma charge density and the field distribution. We find good agreements between the obtained plasma charge density (Fig. 3) and the distributions of the external fields, whose 3D distributions in the LP and CP lasers are shown in Fig. 4i and 4j. Such good agreement is a striking demonstration that the external field dominates the confinement of nanoplasma on the nanosphere surface. In our experiment, the incident laser intensity is $3 \times 10^{13} \text{ W/cm}^2$, which ensures that the strength of external field hotspots exceeds the damage threshold for nanospheres and forms a plasma on the surface. In these field hotspots, field-driven electrons are accelerated to gain enough energy, resulting in a weakening of the surrounding ion bonding to form a higher plasma density on the surface locally³⁵. Due to the surface plasmon, the external field (enhanced field) of a nanosphere is approximately three times stronger than the internal field (shielded field), as shown in Fig. 4. In this case, the strength of the external field is already above the damage threshold for the nanosphere, while the internal field is too weak to loosen the ion-ion bonds of the lattice inside the nanosphere. The situation changes as one increases the incident laser intensity. In that case, the internal laser field is strong enough to destroy the irradiated nanosphere and form hot plasma wholly. This process is typically described by

the three steps of ionization, heating, and explosion^{6,7,36,37}. First, a small number of electrons are liberated by direct optical multiphoton or tunneling ionization locally. Second, the energetically freed electrons inside the nanosphere frequently collide with ions near the electron's birthplace and weaken the ion-ion bonds, leading to rapid heating of the nanosphere to a superheated state. Finally, the ions inside the nanosphere transfer to the surface and then leave the surface, undergoing an electrostatic repulsion process. For particles with a scale of hundreds of nanometers, the number of atoms inside the particle is greater than the number of atoms on the surface. Thus, once the internal field becomes strong enough to open the ion-ion bonds, it will soon replace the external field as the main factor in producing plasma.

To show that the nanoplasma formation at relatively low laser intensity primarily originates from the external laser field and at relatively high intensity primarily originates from the internal field, we explore the ion momentum distributions and their angular integration as a function of laser intensity, as shown in Supplementary Figs. 2 and 3. To guide the relative laser intensities, the measured results are cataloged by ion count. As the laser intensity increases, the y_{y-z} ratio increases to a maximum value of 1.4 and gradually decreases to 1.1, as shown in Supplementary Fig. 4. The increase in asymmetry in the initial stage is due to enhancement of both the external and internal fields along the forward propagation direction. The subsequent decrease in asymmetry indicates that the internal field begins to damage the nanosphere interior, accompanied by intense collision averaging to form hot dense plasma. When we use the highest relative laser intensity with the ion count of 1700, the ion yield imaging is shown in Supplementary Fig. 5. Unlike the dipole-like ion yield distribution in Fig. 3c, the yield distribution shows only a single hotspot of in the forward propagation direction, agrees well with the focusing internal field instead of the dipole-like external field. It therefore confirms that the internal field plays the main role in producing plasma when the laser intensity is high enough, the resulted plasma has changed from surface-localized plasma to hot dense plasma. From the above discussion, we can conclude that the external field plays a dominant role in surface-localized plasma generation, and the internal field plays a dominant role in hot dense plasma generation inside the nanosphere.

Moreover, the well-established relationship between the field distribution and the plasma charge density distribution also motivates the possibility of all-optical control of the nanoplasma location. In previous studies, spatial confinement of the nanoplasma has been associated with control over near-field localization in optimized nanostructure geometries⁶. For example, nanoplasmas will be formed only in the vicinity of gold nanospheres for gold nanosphere-dielectric nanostructures or on the tips for metal nanostars. However, when an aerodynamic lens system delivers unsupported nanostructures with optimized geometries, their orientations are utterly random in the laboratory frame due to the complex hyperdynamics (fluid interaction) involved. Unlike relying on the geometry and orientation of nanostructures, we show an all-optical controllable way to manipulate the location of nanoplasmas on isotropic nanospheres without uncertainty in the laboratory frame. Three different ways can be applied to achieve all-optical manipulation of 3D plasma charge distribution: changing (1) the direction of laser polarization, (2) the state of laser polarization and (3) the size-dependent laser-nanosphere coupling.

First, we consider the effects of the directions of laser polarization for rotation of the polarization direction of the incident laser by 90 degrees around the direction of propagation (x-axis), here from the z-direction to the y-direction. As shown in Fig. 5a, the dipole hotspots for the external field is rotated on the polarization axis accordingly, and, finally, along the y-direction. Correspondingly, the result of plasma imaging is rotated by 90 degrees in Fig. 5b. Next, we consider the effects of different states of laser polarization, as already shown in Fig. 3. The plasma charge density distribution for the LP and CP driving laser fields is different and consistent with their external field distributions, indicating that all-optical manipulation can also be achieved by adjusting the laser polarization state. Finally, we consider the size effects in laser-nanosphere coupling by probing the forward-backward asymmetries along the propagation direction in the 2D ion momentum distributions. By simulating the field distributions for the different sphere sizes and laser wavelengths, we find some differences in the forward-backward asymmetries along the direction of propagation between the internal field and external field. Based on these differences, we define three typical field distributions, shown in Fig. 4 and Supplementary Fig. 1. For a smaller nanosphere (40-nm) with the 800-nm laser, propagation effects do not occur, and both the internal field and the external field show no forward-backward asymmetry along the propagation direction (Supplementary Figs. 1a and 1b), as obtained for the first typical field distribution. For the second typical field distribution of a 100-nm nanosphere with the 800-nm laser (Supplementary Figs. 1c and 1d), the external field distribution is still front-to-back symmetrical. However, the internal field shows a focusing hotspot along the propagation direction. Third, as shown in Figs. 4a and 4c, when we expose the bigger diameter (100 nm) nanosphere to the shorter wavelength (400 nm) laser, the internal and external fields are both enhanced along the forward propagation direction due to the propagation effects. For those three typical field distributions, we probe the corresponding ion momentum distributions in the x-z plane (Figs. 5c-e). Here, an asymmetrical factor I_{fb} is defined to describe the forward-backward asymmetry along the propagation direction of the ion momentum distribution, and the factor I_{fb} is calculated from the ratio of the integrated results in the forward and backward hemispheres of a whole ion momentum distribution. As shown in Figs. 5c and 5d, the factor I_{fb} value is calculated to be approximately 1, indicating that the ion distribution is symmetric along the propagation direction, corresponding to the first and second typical fields. However, the value of I_{fb} is less than 1 in the ion momentum distribution of the third typical field, which means that the ion emission is dominant along the forward propagation direction (Fig. 5e). We revealed the relationship between the field distribution and the detected results for the ion momentum in the three typical field distributions, which verifies that the size effect of the laser field still affects the localized plasma distribution. Furthermore, the forward-backward asymmetry of the ion momentum distributions only agrees with the external field for all three typical field distributions, confirming that the plasma is localized on the surface through the interaction with the external field in our experiment.

In this work, by combining a nanosphere aerosol source with a 3D velocity map imaging spectrometer, we reconstruct the 3D information for the plasma charge density and the ion yield for nanospheres. This is achieved by radial integration and learning inversion of the 3D ion momentum distributions. The capability of the 3D imaging provides a direct way to distinguish the infield and outfield contributions

during the plasma generation. The consistency between the plasma distribution and the external field distribution proves that the external field plays a dominant role in surface-localized plasma generation, contrary to the internal field playing a dominant role in hot dense plasma generation. Moreover, we also demonstrate all-optical 3D manipulation of the localized plasma density by changing the laser wavelength, laser polarization, and nanosphere size through a corresponding variation in the external field. Plasma with controllable 3D localized density on the surface of nontoxic TiO₂ spheres can produce directional ion emission, and, therefore, has excellent potential for minimizing toxicity to surrounding living cells in cancer therapy applications. Our 3D ion nanoscopy method can also be used to extend the image plasma charge density on the surface of a wide range of isolated nanostructures, including nanospheres, clusters, and even droplets with different sizes and materials. This study will open up the door for spatially resolved studies and deepen understanding of the mechanisms of plasma generation from other functionally isolated nanostructures.

Methods

Experimental apparatus

We aerosolized a beam of aerosols consisting of the isolated TiO₂ nanospheres suspended in CO₂ carrier gas by a commercial atomizer (TSI model 3076) from the suspension liquid including the TiO₂ nanospheres dispersed in water (1 g/L). A silica gel diffusion dryer was used to dry the TiO₂ nanosphere aerosols, and then an aerodynamic lens system was used to collimate the aerosol beam into the center of the velocity-map imaging spectrometer³⁸. The pressure in the chamber of the spectrometer was approximately 10⁻⁵ mbar. The nanospheres were then irradiated by thy intense femtosecond laser pulses centered at 400-nm, which were obtained by doubling the frequency of a 1 kHz Ti:sapphire laser system with a BBO crystal. Ions derived from the interaction region are accelerated within an electrostatic field and recorded on a microchannel plate (MCP)/phosphor screen detector. The higher ion yields easily distinguish the laser hitting nanosphere signal images from the background noise. Then the resulting images were recorded by a CCD camera with an average readout frequency of 250 Hz and a 0.1–1% hit rate to balance the recording efficiency and signal-to-noise ratio.

Binning technique and tomographic reconstruction

The ion count per 2D VMI image can be used as a coarse guide for the near-single intensity sampling of the laser interacting on the nanospheres, called the intensity binning technique²⁸. By a LabVIEW script, we firstly recorded the 2D VMI image and counted the ion numbers of every image. And then those images which were recorded in the same condition of the laser intensity were identified by their similar ion count. By a MATLAB script, those 2D images with a similar ion count were automatically added and averaged into one slice image, which ensured the near-single intensity sampled imaging. At the same time, on a few the nanospheres, some surface defects or protrusions may exist. The random locations of the defects or protrusions will raise the uneven distributions of the enhanced field hotspots on the surface, which will also affect the charge density distributions of the surface-localized plasma, leads to the

random ion momentum distributions. However, this random effect with a low probability will be effectively weakened in the slice images averaged from a number of 2D VMI images. This binning method provides us with each the near-single intensity sampled 2D slice image used for 3D reconstruction under the same experimental conditions, eliminating the laser focusing volume effect and the influence of the defects or protrusions.

Then, with an electric half-wave plate, the polarization of the laser pulses is rotated about the propagation axis. At the same time, we measured a series of the near-single intensity sampling 2D slice images of the ion momentum distribution at different angles θ of 0-180° for a step size of 1 degree, as shown in Fig. 1b. Then, the 3D ion momentum spectrum was reconstructed by a Fourier transform-based tomographic technique with our MATLAB script.

Ion dynamics model

The Coulomb repulsive process between the ions and the plasma surface charges is the main source of the acquisition of ion kinetic energy. Thus, here, we used a simplified classical model of ion emission to simulate the repulsive motion of the Ti^+ ions emitted from the surface, neglecting the electron-ion interaction and the space charge effect between the emitted ions. In addition, the femtosecond laser field will not affect this repulsion process at the picosecond time scale. As shown in Fig. 3a, the Coulomb repulsive dynamics of the repulsion process is described by a one-dimensional (1D) model between two point charges, where the fixed charge Q (position $r = 0$) in the center of the sphere represents the buildup of positive charges on the surface and the motional charge q represents the Ti^+ ion. And ion q launches outward along the axis defined by Q and q , where the initial distance between Q and q is the radius of the nanosphere (50-nm). Then the final momentum of the Ti^+ ion q can be calculated numerically by integrating the differential Newton equations.

Finite-difference time-domain (FDTD) calculations

We simulated the field distributions by finite-difference time-domain (FDTD) calculations (Lumerical, Solutions Inc.), and the refractive index of TiO_2 at different wavelengths is given by previous work³⁹. The TiO_2 nanoparticles were simulated as 40-nm or 100-nm dielectric spheres with $n = 2.52$ (the refractive index of TiO_2 at 800 nm) and $n = 2.99$ (the refractive index of TiO_2 at 400 nm).

Learning inversion algorithm

The learning inversion algorithm is written to retrieve the plasma charge density on the surface. In our learning inversion algorithm, a library including the numerical relationships between the plasma charge density Q and the final ion momentum is firstly established. We set Q as a free parameter and then calculate the corresponding values of the final ion momentums by the ion dynamics model, those projective relationships are then saved to the library. By the inversion from the library, the charge density distributions of the surface localized plasma can be extrapolated from the measured 3D ion momentum distributions. In detail, by a MATLAB script, the values of the maximal momentum (cutoff) along the

different spatial angles (φ, θ) will be orderly recorded from the measured 3D ion momentum. Then those cutoff values are firstly brought into the above library, and the corresponding density values are then indexed by the Q in the library and exported to a 2D mapping image. This two-dimensional plasma charge density of the spatial angles (φ, θ) is then spanned over a unit sphere to yield the 3D plasma density distribution on the nanosphere surface shown in Fig. 3. By this inversion algorithm, we can directly retrieve the plasma charge density from any measured ion momentum distribution, which demonstrates a wide range of measurements for unknown plasma charge distributions.

Declarations

Data Availability

The data that support the findings of this study are available from the corresponding authors upon reasonable request.

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Author contributions

X. Han and X. Huang contributed equally to this work. Q.B.Z. and P.X.L. conceived and designed the experiment. X. Han, X. Huang, X.L.F., and Q.B.Z. performed the measurements. X. Han, Q.B.Z. developed the experimental setup and the ion dynamics model. X. Huang performed tomographic reconstruction and FDTD simulations. X.L.F. and Q.B.Z. conducted the design of the aerosol source. P.F.L., Y.M.Z., M.L. and W.C. participated in the discussions and contributed to the final manuscript drafted by X. Han, K.L.L. and Q.B.Z.

Competing interests

The authors declare no competing interests.

Additional information

Supplementary information is available for this paper in...

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Figures

Figure 1

Laser-induced ion velocity mapping imaging. a The experimental apparatus used for nanoplasma imaging. A femtosecond laser pulse hits gas-carried aerosol nanoparticles transported by an aerodynamic lens to form a plasma in an ultrahigh vacuum environment. The MCP and phosphor screen detector are used to record typical two-dimensional ion emission images. b All 2D images are measured at different angles θ ranging from 0-180° with a revolving half-wave plate, and then the 3D reconstruction of the emitted ion distribution is obtained by inverse Radon transformation. c We count the ion numbers for each ion image and obtain a histogram for the ion-signal numbers. The ion yield per laser shot is used as a relative measure of local laser intensities, called single intensity approximations, and the maximum number of ions is approximately 1700.

Figure 2

Display of 3D velocity distribution in different directions by projection. a Linearly polarized and b circularly polarized laser-induced ion distribution in the plane (y-z) perpendicular to the direction of propagation (x). c Linearly polarized and d circularly polarized laser-induced ion distribution in the plane (x-z) formed by the polarization (z) and propagation (x) directions. e Linearly polarized and f circularly polarized normalized angular integral yields for the ion distribution in the y-z plane. The vertical to horizontal ratio γ_{y-z} is calculated from the ratio of the ion yields parallel to and perpendicular to the z-axis.

Figure 3

Spatial plasma density and ion yield imaging. a Schematic diagram of ion radial repulsion dynamics. b The normalized plasma charge distribution obtained by the learning inversion algorithm in the LP laser field. c Ion yields over a unit sphere retrieved by integrating the measured ion momentum distributions along the radial coordinate in the LP laser field. d, e Same as c, d but for the CP laser field.

Figure 4

The internal and external electric field distribution for the 100 nm nanosphere calculated by FDTD simulations. a The external and b internal linearly polarized electric field distribution in the x-z plane. c External and d internal linearly polarized electric field distribution in the y-z plane. e External and f internal circularly polarized electric field distribution in the x-z plane. g External and h internal circularly polarized electric field distribution in the y-z plane. Both the LP and CP laser fields propagate along the x-direction, and the scattering field intensities are normalized to the incident field intensity. The black circle in each figure represents the nanosphere surface. i The 3D projection of the external field distribution in the LP laser. j The 3D projection of the external field distribution for the CP laser.

Figure 5

All-optical modulation and size-effect-based modulation of nanoplasma. a Rotate the polarization of the linearly polarized laser by 90 degrees around the propagation direction (x) to the y-axis direction (along the trajectory of the dotted line). The simulated 3D external field distribution hotspots are rotated by 90 degrees accordingly with respected to the hotspots shown in Fig. 4i. b The distribution of ion yields after the laser field is roatated by 90 degrees, demonstrating the change of surface plasma density distribution. c-e The measured ion momentum distributions in the x-z plane for three combinations of laser wavelength and nanosphere diameter: 800-nm wavelength and 40-nm diameter, 800-nm wavelength and 100-nm diameter, and 400-nm wavelength and 100-nm diameter. The left-to-right signal ratio I_{fb} represents the forward-backward asymmetry of ion emission. The laser propagates from left to right, with polarization along the z-direction.

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