Field propagation-induced directionality of carrier-envelope phase-controlled photoemission from nanospheres

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Near-fields of non-resonantly laser-excited nanostructures enable strong localization of ultrashort light fields and have opened novel routes to fundamentally modify and control electronic strong-field processes. Harnessing spatiotemporally tunable near-fields for the steering of sub-cycle electron dynamics may enable ultrafast optoelectronic devices and unprecedented control in the generation of attosecond electron and photon pulses. Here we utilize unsupported sub-wavelength dielectric nanospheres to generate near-fields with adjustable structure and study the resulting strong-field dynamics via photoelectron imaging. We demonstrate field propagation-induced tunability of the emission direction of fast recollision electrons up to a regime, where nonlinear charge interaction effects become dominant in the acceleration process. Our analysis supports that the timing of the recollision process remains controllable with attosecond resolution by the carrier-envelope phase, indicating the possibility to expand near-field-mediated control far into the realm of high-field phenomena.
nanostructures enable the concentration of laser light in highly localized near-fields with dimensions far below the incident wavelength. Utilizing optical near-fields for the control of electron motion in nanostructures with attosecond resolution is a major prospect and challenge in ultrashort light-wave driven nanoelectronics. Enhanced strong-field photoemission in near-fields has been demonstrated for metal nanotips, dielectric nanospheres and surface-assembled nanoantennas. In analogy to atomic above-threshold ionization, electron backscattering dominates the high-energy electron emission if the electron quiver amplitude is small compared with the near-field extension into free space. Many-particle charge interaction effects can increase the electron cutoff energy far beyond the values expected from the linear field enhancement, as demonstrated in previous experiments on small nanospheres. The coherent nature of the near-field driven acceleration has been revealed by the fact that photoelectron spectra depend on the laser’s electric field waveform, controlled by the carrier-envelope phase (CEP). Key prerequisite for near-field-mediated tailoring of the underlying electronic field dynamics is knowledge about its dependence on and feedback to the spatiotemporal near-field evolution. A detailed exploration of near-field control up to high field intensities is offered by studying unsupported reproducible nanosystems in the gas phase. Theoretical studies of the electron acceleration from droplets support that propagation-induced near-field effects have a strong impact on the electron dynamics, including the directionality of the emission, and can be utilized for the generation of attosecond electron bunches up to relativistic intensities. Control of photoelectron angular distributions has also been demonstrated for atoms and chiral molecules via polarization-shaped femtosecond laser pulses. In the latter cases, however, the directionality results from selection rules and the chirality of the electronic initial or/and continuum states, respectively, and can be described in dipole approximation. The physics is therefore fundamentally different from control of electronic motion via field propagation-induced near-fields as considered in the present work.

Here we employ isolated 50–550 nm SiO$_2$ nanospheres and demonstrate the size-dependent effect of field propagation-induced near-field deformation on the directionality of the strong-field photoemission. We observe systematic directional tunability of the electron emission with respect to the propagation direction via the sphere size and find evidence for the persistence of robust attosecond control of the dominant surface backscattering process via the CEP. Our combined experimental and theoretical analysis shows that dynamical many-particle charge interaction results in substantial quenching of the electron emission and becomes dominant for the electron acceleration for the largest investigated sphere sizes. A systematic trajectory analysis based on semiclassical transport simulations enables for a clear discrimination of the impact of near-field enhancement, vectorial field properties and self-consistent collective electron dynamics on the strong-field photoemission process.

**Results**

**Propagation-induced near field deformation.** Exposing nanospheres to few-cycle pulses with known CEP allows the generation of well-defined near-fields, whose linear response structure is accurately described by the Mie solution. For a given refractive index, the latter depends mainly on the dimensionless Mie size parameter $\rho = nd/\lambda$ (where $d$ is the sphere diameter and $\lambda$ is the wavelength) and resembles Rayleigh’s quasi-static dipole solution for small spheres ($\rho \ll 1$). Propagation-induced near-field deformation arises and becomes significant for $\rho \gtrsim 1$ because of substantial excitation of higher order multipole modes, resulting in a gradual shift of the region of maximal field enhancement in propagation direction. Increasing the size parameter to the range $\rho \approx 1$ leads first to nanojet-type focusing employed in superlenses followed by the regime of geometric optics. Such systematic modifications can be achieved by changing the excitation wavelength or the sphere size. We varied the sphere size to realize scale parameters between $\rho \approx 0.2$ and 2.4, ensuring peak field enhancement to occur at the surface; the employed wide-bandgap dielectric material ensures minimal pulse broadening such that the few-cycle character of the near-field is preserved. Nevertheless, field propagation induces a nontrivial elliptic local field (see Fig. 1d,e and the Methods).

**Size- and carrier-envelope phase-dependent directionality.** We measured the angle-resolved photoemission from SiO$_2$ nanospheres ($d \approx 50–550$ nm, cf. Fig. 1a) via velocity-map imaging (VMI) using a setup similar to Zherebtsov et al. The photoelectron dynamics was controlled by the CEP, $\varphi_{\text{CE}}$, of 4 fs few-cycle laser fields at 720 nm central wavelength (see Methods). Near-field induced symmetry breaking of the photoemission for $\rho \gtrsim 1$ is revealed by the asymmetry of CEP-averaged photoelectron momentum projections with respect to the laser propagation direction (left-to-right in Fig. 1f). Below the 10$^4$ U$_p$ backscattering cutoff (see shaded areas in Fig. 1fg), the momentum maps may contain spurious photoemission signal from residual background gas. The CEP-dependent signal from nanospheres shows that the electron emission can be effectively switched upwards or downwards while the left-right asymmetry remains (Fig. 1g). From the modulus of the projected momentum up to which phase-dependent VMI signal is observed we determined the global cutoff momentum $p_c$ (solid circles) to define the cutoff energy $E_c = p_c^2/2m$, where $m$ is the electron mass. This cutoff is attributed to surface backscattering, see Fig. 1c.

Most importantly, we observed a size-dependent directionality of the phase-controlled photoemission. Maps of the high-energy electron yield show strong phase dynamics and signal concentration in a narrow angular range for the upward and downward direction for all investigated sphere diameters, see examples in Fig. 2a,b. From each map we extracted the critical final emission angle, $\theta_{\text{crit}}^F$, and critical CEP, $\varphi_{\text{CE},\theta_{\text{crit}}}^F$, to characterize the directionality and CEP dependence (see circular symbols). Comparison of results for 95 and 550 nm spheres reveals similar phase dynamics but a significant shift of the critical emission directions from nearly $90^\circ$ to almost $45^\circ$.

**Simple man’s model.** To explore the impact of the linear near-field we describe the strong-field photoemission dynamics classically with the simple man’s model (SMM). Electron trajectories launched at rest at the nanosphere surface are integrated under the local field obtained from Mie’s solution and assuming elastic specular reflection at the surface. The resulting cutoff momenta as function of the asymptotic emission angle, $p_c(\theta)$, indicate maximal energies for single recollision trajectories ($n = 1$) and substantially lower energies for direct ($n = 0$) and higher order ($n > 1$) recollision electrons (Fig. 3). The critical birth angle of most energetic electrons ($n = 1$, small circle) coincides with the angle of peak radial near-field enhancement and deviates only weakly from the critical final angle (large circle), underlining the dominance of the radial field in the recollision-based process. Comparison with experiment shows that the SMM reasonably explains the critical emission angles but
Figure 1 | Photoemission from large nanospheres. (a) Typical electron microscopy images of the employed silica nanospheres. The indicated diameters (top) reflect typical sizes and are used as a reference throughout the manuscript. Measured mean diameters and their s.d. of representative samples studied in this work are indicated in boxes, respectively. The scale bar applies to all images and corresponds to a length of 500 nm. (b) Maximum enhancement of the radial electric field in the propagation plane ($z = 0$) predicted by Mie’s solution for $d = 400$ nm (Mie size parameter $\rho$ as indicated). The incident laser field $E_r(t) = E_0 f(t) \cos(\omega t + \varphi_{E_0})$ with 4 fs (intensity full-width at half-maximum) Gaussian envelope $f(t)$ at centre wavelength $\lambda = 720$ nm propagates along the x axis. (c) Schematic illustration of the electron recollision process. (d) Vectorial representation of the field evolution in the x – y plane normalized to incident peak amplitude $E_0$, sampled at the point with peak radial near-field enhancement ($d = 61.0^\circ$, CEP as indicated). Coloured arrows indicate the local reference frame for radial (red) and tangential (blue) fields to illustrate the evolution of the field ellipticity. (e) Evolution of radial and tangential electric field components $E_r$ (blue) and $E_t$ (red). (f) CEP-averaged VMI electron momentum projection (momenta in atomic units) measured for $d = 400$ nm at $2.7 \times 10^{13} \text{Wcm}^{-2}$. (g) Phase-resolved VMI images (CEP as indicated) after subtraction of the CEP-averaged spectra. Solid circles in (f) and (g) indicate the extracted cutoff momentum and dashed circles the uncertainty estimated from the deviation of the results for the upper and lower half of the momentum distribution. The shaded circular areas in (f, g) indicate the momentum range that corresponds to electron energies below the 10 $U_p$ classical rescattering cutoff and may contain residual signal from background gas.

Self-consistent simulation model. For a realistic description we employed quasi-classical Mean-field Mie Monte-Carlo (M$^3$C) simulations (see Methods) and compared the results with the measured data (Fig. 2). Both the CEP-dependent switching of the photoemission and the size-dependent critical emission angles are well reproduced. M$^3$C energy spectra (Fig. 4a) show that single recollision electrons are dominant over direct and higher order recollisions for all energies and the cutoff energy by far exceeds the SMM prediction. For classifying M$^3$C trajectories, we counted each electron re-entry as one recollision event even if several microscopic collisions are involved. Both the quenching of direct electrons (because of the local trapping potential) and the enhanced acceleration reflect the importance of charge interaction, which is analysed in more detail below. The resulting global M$^3$C cutoff momentum (red filled circle in Fig. 3) is close to the experimental result regarding both magnitude and direction, indicating that all major effects contributing to the electron acceleration have been captured by the model.

Attosecond recollision dynamics. To characterize the attosecond dynamics, we analysed CEP-dependent M$^3$C data for emission into the upper half space. Energy spectra for single recollision electrons (Fig. 4b) visualize the cutoff energy modulation underlying the switching effect. By timing analysis (Fig. 4e) we find that, irrespective of CEP, essentially all fast electrons are born into the upper half space. Energy spectra for single recollision electrons (Fig. 4b) visualize the cutoff energy modulation underlying the switching effect. By timing analysis (Fig. 4e) we find that, irrespective of CEP, essentially all fast electrons are born by tunnelling within a narrow time interval ($< 300$ as) in a single...
Figure 2 | Directionality and phase-dependent switching. (a,b) Measured angle and CEP-resolved electron yields $Y(\theta, \varphi_{\text{CE}})$ of near-cutoff electrons (projected momenta $p > \sqrt{2m_eeE_0}$ with $E_0 = 0.5E_c$) discriminating electrons below the threshold energy $E_0$; sphere sizes and laser intensities as indicated. (c) Amplitudes $A(\theta)$ of harmonic fits (black) of the data in (a,b) with $Y = Y_0(\theta) + A(\theta) \cos[\varphi_{\text{CE}} - \Delta \varphi(\theta)]$ and critical emission angles (vertical dashed lines) as determined from the peaks of Gaussian fits of $A(\theta)$ (green and red curves) for upward and downward emission, respectively. Horizontal black curves in (a,b) show the fitted phase offsets $\Delta \varphi(\theta)$; white dots indicate the critical CEP values $\varphi_{\text{CE}}^\text{crit}$ and critical emission angles $\theta^\text{crit}$ for maximal upward emission. (d-f) same as (a-c) as predicted from M$^3$C simulations for the experimental parameters.

Systematic comparison of experiment and theory. The size-dependent impact of propagation-induced near-field deformation on the electron emission is analysed in Fig. 5. First, the evolution of measured critical angles (Fig. 5a) agrees well with both the SMM and M$^3$C simulations, substantiating a systematically tunable directionality; the remaining offset between experiment and theory is attributed to a systematic error (for example, inhomogeneous VMI detector response). Second, the measured weakly size-dependent critical phase (Fig. 5b) shows a notable offset from the SMM result but is well described by M$^3$C theory, supporting persistence of robust attosecond control and a proper description of the electron dynamics up to strongly deformed near-fields. Third, the measured cutoff energies are reasonably captured by the full M$^3$C simulations but exceed the predictions dominant half-cycle. The nonlinear near-field contribution (see short-term evolution in Fig. 4d) reveals strong dynamics and has a significant effect on the local radial field (Fig. 4c). In particular, the resulting trapping field quenches tunnel ionization, as discussed in more detail later. However, although the ionization dynamics and energetics are substantially changed in the M$^3$C treatment, the excursion length (Fig. 4d) and average timing (Fig. 4e) of recollision trajectories remain similar to the SMM prediction, substantiating the applicability of the recollision picture. Altogether, the half-cycle selectivity, the smooth systematic shift of birth and recollision times with CEP, and the resulting pronounced high-energy signal modulation give evidence for robust attosecond control.

Figure 3 | Photoemission dynamics predicted by simple man’s model. Predicted electron cutoff momenta as function of emission angle, $p_\theta(\theta)$, for $n = 0, 1, 2$ recollisions at optimal CEP for upward emission; $p_0 = E_0/c$ is the free space quiver momentum. Small/large circular symbols denote the critical birth/emission angles associated with respective highest cutoff momenta (see inset for corresponding radial trajectories); blue/red filled circles show critical emission angles and cutoff momenta from experiment and M$^3$C simulations; shaded rings are guides to the eye.
of SMM and $M^3C$ theory with charge interaction switched off by up to a factor of two (Fig. 5c).

Selective analysis of acceleration mechanisms. Finally, we disentangle and quantify the different many-particle contributions to the acceleration process using a selective energy gain analysis, see Methods for technical details. The resulting size-dependent analysis is depicted in Fig. 5c. The relative cutoff energy enhancement because of the local trapping potential is only weakly size-dependent and results mainly from the radial electron motion (compare black and blue lines). This behaviour supports that the trapping potential is only determined by the local electron density (surface charge density) and thus insensitive to the system size. The tangential field effect is notable only for large spheres (dashed versus solid blue line) but remains small even if tangential and radial field amplitudes become comparably strong, for example at $d = 500$ nm (see Methods). This supports that the tangential field is neither crucial for the emission angles nor for the acceleration process. The relative enhancement due to the space-charge repulsion increases strongly with size and becomes dominant for large spheres. This trend can be explained with stronger Coulomb repulsion due to an increasing number of electrons in the escaping bunch, being an effect that is sensitive to the full (non-local) electron distribution.

The time-resolved analysis of the energy gain contributions in full $M^3C$ simulations shows the following dynamics, see examples in Fig. 5d,e. Compared with the SMM results (solid black curves), the enhancement of the gain associated with the radial Mie field (dotted blue curves) develops shortly after the moment of birth within the recollision process. The fact that no substantial change of the relative energy is found in later stages of the pulse supports that the impact of the trapping potential unfolds close to the surface. The gains calculated from the full Mie field (dashed blue curves) show that the tangential field effect also develops on short time-scales, that is, during the pulse. Finally, the energy gains with the full $M^3C$ field (red curves) reveal that the additional acceleration due to Coulomb explosion develops on a substantially longer timescale as it results solely from electron repulsion within the emitted bunch.

Mean-field-induced quenching of the electron emission. In the investigated range of laser intensities and particle sizes we find a strong impact of the Coulomb field on the electron emission. The yield predicted by simulations without the Coulomb interaction scales roughly exponentially with sphere diameter and laser intensity, reflecting the highly nonlinear tunnelling rate. Inclusion of Coulomb effects decreases the yield by up to two orders of magnitude (red versus black dashed curves in the insets of Fig. 6a,b). The reduction is a direct consequence of the trapping field, which quenches tunnel ionization at the surface and limits the number of electrons that can escape with a given initial kinetic energy. This pivotal influence of the Coulomb field on the actual ionization dynamics also explains why the $M^3C$ results are only weakly affected from changes of the (certainly approximate) tunnelling rate (for example, by slight variations of the effective ionization potential) as soon as a substantial trapping field...
develops. On the other hand, the reasonable description of the Coulomb effects in the model then requires quantitative agreement of experiment and theory. However, a quantitative comparison of the total yield is difficult because of the spurious signal contribution from residual gas in our experiment and the imprint of focal averaging. To define a photoemission yield...
specific to nanoparticles and free from background gas contributions, we counted only electrons with momenta beyond the threshold $p_\text{c}/\sqrt{2}$, which includes only signal well above the gas signal cutoff (see Fig. 1g). Further, focal averaging is circumvented by assigning most intense single-shot VMI images to the peak laser intensity. Because of the low particle density in the beam, such images reflect the emission from a single nanoparticle. Considering the estimated experimental electron detection efficiency of $(30 \pm 20\%)$, the resulting measured near-cutoff electron yield as function of particle size and laser intensity is compatible with the corresponding $\text{M}^3\text{C}$ predictions with mean-field, see symbols and solid red lines in Fig. 6a,b. The remaining discrepancies for small particles and at low intensities are attributed to residual background signal. The agreement strongly supports the $\text{M}^3\text{C}$ prediction that the number of emitted electrons evolves nearly linearly with size and intensity in the presence of substantial Coulomb interaction.

### Discussion

The present results suggest that the size-dependent directionality of the strong-field photoemission from nanospheres directly relates to the field propagation-induced near-field deformation. The analysis reveals the dominance of the radial field driven recollision dynamics up to large sizes with substantial near-field ellipticity and gives evidence for the persistence of attosecond control of the electron dynamics. As the near-field deformation could in principle be manipulated directly via the excitation wavelength, our findings indicate feasibility of near-field-induced photoemission with optically tailored directionality with respect to the beam propagation axis.

Furthermore, we identify the transition from local-field-dominated dynamics\textsuperscript{8,9} to a regime where charge interaction effects due to the non-local structure of the emitted electron bunches become equally important or even dominant for the energetics of the electron acceleration. These results are of general relevance for strong-field electron dynamics in nanosystems (nanoparticles, -jets, -solids and -tips) and indicate the extension of near-field-mediated waveform control into the extremely nonlinear regime\textsuperscript{21,22}. We anticipate that this enables steering of attosecond electron bunch emission from droplets\textsuperscript{1,12} via phase control and near-field enhancement of surface high-harmonic generation\textsuperscript{23} from nanostructured targets. Eventually, correlating the near-field driven electron dynamics with ion spectra\textsuperscript{24} and imaging the resulting particle damage via single-particle X-ray scattering promises unprecedented insights into the poorly understood processes of sub-wavelength laser machining in dielectrics\textsuperscript{25}.

### Methods

#### Sample preparation

Silica nanoparticles with diameters between 50 and 550 nm and a narrow size distribution were prepared by wet chemistry approaches. All chemicals (ethanol (Berkel AKL ultrapure, 100%), tetraethoxysilane (TES, Fluka, purum, 98%), ammonia solution (Merck, p.a., 28–30%)) were used as received without further purification. The reaction flasks were cleaned by hydrofluoric acid and a narrow size distribution were prepared by wet chemistry methods. Silica nanoparticles with diameters between 50 and 550 nm have been stored in ultrapure ethanol or an ethanol ultrapure water mixture (80:20) after cleaning. Characterization by transmission electron microscopy and scanning electron microscopy yielded a polydispersity of $\sim 15\%$ for small particles around 50 nm and decreases substantially to $3\%$ for the large particles under study. The surface of silica nanoparticles prepared by the Stöber method\textsuperscript{26} is negatively charged and slightly hydrophilic\textsuperscript{29}.

#### Experimental approach

The few-cycle laser pulses were generated by spectral broadening of a Ti:Sa amplifier output (25 fs, 790 nm) using a Ne filled hollow-core fibre and chirped mirror compression. A fraction of the laser beam was split off and sent to a stereo atomic above-threshold ionization phasemeter\textsuperscript{20} for single-shot CEP measurement. The remaining laser beam was intersected with the nanoparticle beam in the focus of the VMI apparatus\textsuperscript{51}. The jet of isolated nanospheres was generated from a dispersion of SiO$_2$ particles in ethanol by...
Figure 7 | Size-dependent near-field enhancement. (a) Schematics of the polarization and propagation directions of the impinging few-cycle pulses and spatial trace at which the field is analysed (white arc). Radial and tangential fields (Mie solution) are evaluated by projection on the unit vectors \( e_\theta \) and \( e_\phi \) (as indicated). (b) Profiles of the local radial field enhancement, \( E_r(d, \theta) / E_0 \), and evolution of the characteristic angle, \( \theta(d) \), with highest local radial field enhancement (black line). (c) Radial and tangential peak field amplitudes, \( E_r(d, \theta) \) and \( E_\theta(d) \), sampled under the characteristic angle. (d) Field ellipticity (solid), defined via the ratio \( b/a \) of the field amplitudes along the minor and major field axis (see inset), and tilt angle \( \Delta \theta \) (dashed) of the major field axis with respect to the local surface normal (see inset). The unit vectors \( e_\theta \) and \( e_\phi \) in the inset are defined as in (a). (e) Effective pulse length (solid) and CEP shift \( \Delta \phi_{\text{CEP}} \) (dashed) of the local radial field. Black arrows in (d) and (e) indicate the ordinate that correspond to each of the curves.

Linear few-cycle near-fields of nanospheres. For the time-domain description of the near-fields at the SiO\(_2\) nanospheres we employ Mie’s continuous wave solution of the medium Maxwell equations combined with a spectral field decomposition. In linear response and spectral complex field representation, the spatiotemporal electric field evolution can be expressed as

\[
\mathbf{E}(t, \mathbf{r}) = \frac{1}{2} \int f(\omega) \mathbf{E}(\mathbf{r}, \omega) e^{-i\omega t} e^{-i\omega \mathbf{r} \cdot \mathbf{k}_0} d\omega + c.c.,
\]

with \( E_0 \) the field peak amplitude, \( f(\omega) \) the normalized amplitude spectrum, \( \mathbf{E}(\mathbf{r}, \omega) \) the spatial mode structure as function of angular frequency, \( \omega \), and the CEP \( \phi_{\text{CEP}} \).

For a bandwidth-limited few-cycle laser pulse (polarized along \( y \) axis, propagating along \( x \) axis) with Gaussian temporal field envelope, the spectrum and mode structure read

\[
f(\omega) = \frac{1}{\sqrt{2\pi\Delta\Omega^2}} e^{-\omega^2/2\Delta\Omega^2},
\]

and \( E^{\text{sp}}(\mathbf{r}, \omega) \) provide the boundary conditions for the Helmholtz equation of a dielectric sphere and lead to the well-known Mie solutions

\[
E^{\text{Mie}}(\mathbf{r}, \omega, \phi) = \sum_j E_j^{\text{sp}}(\mathbf{r}, \omega) Y_j(\phi),
\]

where the summation over \( j \) runs over the multipole orders of the expansion into spherical vector wave functions and \( d \) and \( c(\omega) \) describe the diameter and relative permittivity of the sphere. We evaluate the integral in equation (1) numerically by a discrete sum over 25 spectral components and by including multipole order up to \( l = 5 \) in the Mie modes of equation (4). We checked that the dispersion of SiO\(_2\) is sufficiently small to justify the use of a fixed relative permittivity \( \varepsilon = 2.12 \) sampled at the angular frequency \( \omega_0 \) corresponding to the laser central wavelength.

Near-field structure. To characterize the size-dependent near-field we consider 4 fs pulses at 720 nm central wavelength and analyse the radial and tangential fields \( E_r(t) \) and \( E_\theta(t) \) in the \( z=0 \) plane at (outside) the nanosphere surface for \( y>0 \), see white arc in Fig. 7a, where the field component along the \( z \) axis vanishes for symmetry reasons. In the relevant size range, highest field enhancement occurs on this axis with characteristic angle \( \theta(d) \), see Fig. 7b. With increasing sphere size and under this characteristic angle, the radial and tangential peak amplitudes show a modest and rapid increase, respectively (Fig. 7c). The resulting local fields become increasingly elliptic with size and exhibit a tilt of the major field axis with respect to the local surface normal (Fig. 7d). The pulse duration (intensity envelope) grows by \( \sim >4 \% \) in the investigated size range and can thus be considered as essentially constant (Fig. 7e). Finally, the propagation effect introduces a small shift of the effective CEP of the radial field.

Simulation model. In the quasi-classical M3C model, electron trajectories are generated via Monte-Carlo sampling of the surface tunnel ionization. In each time step, a tunnelling path is determined for randomly chosen surface atoms using an effective ionization potential of 9 eV to describe the band gap of SiO\(_2\) and following the local field direction. The ionization probability is determined from Ammosov–Delone–Krainov atomic tunnel ionization rates\(^{35} \) using the field gradient averaged over the tunnelling path. Trajectories are launched at the classical tunnel exit and integrated classically in the local electric fields. The latter contain the time-dependent Mie solution and the instantaneous, self-consistent mean-field. The mean-field describes the Coulomb interaction with free charges (electrons and residual ions) in the presence of the dielectric sphere via high-order multipole expansion up to multipole order \( l = 10 \). In the mean-field solver, the sphere polarization is described by the same relative permittivity as in the Mie solver. Elastic electron–atom collisions are described by isotropic scattering events using an energy-dependent mean free path derived from quantum mechanical scattering cross-sections for the atomic potentials of Si and O atoms. Inelastic collisions are modelled with Lotz’s electron impact ionization cross-sections\(^{36} \). Electron spectra for escaped electrons are calculated from trajectories with positive single-particle energy and positive normal velocity. A typical single high-resolution simulation run contains 2.5 \( \times 10^6 \) trajectories and takes \( \sim 1 \) week on a single CPU core. The systematic scans over CEP and particle size performed in the current study required several tens of CPU years on the HLRN supercomputer, taking maximum advantage of massive parallel computation techniques. For the investigated laser intensities (I \( \lesssim 10^{15} \) W cm\(^{-2} \)) adiabatic metalization\(^{37} \) breakdown effects\(^{38} \) and the nonlinear optical response of the dielectric sphere material\(^{39} \) can be neglected.

Energy gain analysis. The contributions from the local trapping potential and space charge repulsion to the acceleration process are mediated by the self-consistent mean-field potential in the M3C simulations. To separate and quantify these effects we employ a selective energy gain analysis. Therefore, we consider the kinetic energy gain \( \Delta E(\mathbf{r}) = \int E^2(\mathbf{r}'(t)) - E^2(\mathbf{r}(t)) d\mathbf{r}' \) resulting from different field contributions \( E \). Considering the results of the SMM, evaluation of this integral for a given electron trajectory (and using only the Mie fields) is equal to the instantaneous kinetic energy. Likewise, integration over the full M3C trajectories and using the full near-field (Mie and dynamic mean-field) converges to the final asymptotic kinetic energy. For the analysis we select trajectories with final energy equal to the cutoff energy. To separate the effect of the local trapping potential, we...
use the full MC trajectories but include only the Mie field in the energy gain integration. This includes the modification of the trajectories due to the presence of the mean-field, but excludes the acceleration mediated by its dynamical evolution, for example, due to Coulomb explosion of the escaping electron bunch.

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