Tailoring near-field-mediated photon electron interactions with light polarization

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Abstract

Inelastic interaction of free-electrons with optical near fields has recently attracted attention for manipulating and shaping free-electron wavepackets. Understanding the nature and the dependence of the inelastic cross section on the polarization of the optical near-field is important for both fundamental aspects and the development of new applications in quantum-sensitive measurements. Here, we investigate the effect of the polarization and the spatial profile of plasmonic near-field distributions on shaping free-electrons and controlling the energy transfer mechanisms, but also tailoring the electron recoil. We particularly show that polarization of the exciting light can be used as a control knob for disseminating the acceleration and deceleration pathways via the experienced electron recoil. We also demonstrate the possibility of tailoring the shape of the localized plasmons by incorporating specific arrangements of nanorods to enhance or hamper the transversal and longitudinal recoils of free-electrons. Our findings open up a route towards plasmonic near-fields-engineering for the coherent manipulation and control of slow electron beams for creating desired shapes of electron wavepackets.

1. Introduction

By the early 21st century, research into photon-induced near-field electron microscopy (PINEM) [1] opens exciting new techniques in manipulating and shaping [2, 3] electron wavepackets. In PINEM, electron wavepackets interact with laser-induced near-field excitations, allowing for spatially, energetically, and temporally resolving the optical modes of nanostructures and materials excitations, such as phonon polaritons [4]. Coherent control of the shape of quantum wavepackets has applications in bond-selective chemistry [5], quantum computing [6, 7], and ultrafast control of plasmonic systems [8]. By developing the pump-probe electron spectroscopy and its theoretical quantum description [9–11], investigating the ultrafast dynamics of quantum systems with energetic free electrons and femtosecond light pulses have experienced an impressive boost toward shaping the electron beams [12, 13]. Modulating the continuous energy spectrum of free-electron wavepackets [8, 14–16] with the evanescent field [17] and bunching the free-electron wavepacket with light [17, 18] has promises in quantum technology and sensing. This quantized energy-momentum exchange in electron-photon coupling offers a new degree of freedom for nano-scale spectroscopy, attosecond control of free-electron quantum wavepackets and for temporal electron pulse manipulation [19, 20]. In addition to the longitudinal spread of the electron momentum distribution [21], the wiggling motion of electron wavepackets in the electromagnetic field leads to a transversal Lorentz force as well and, consequently, time-dependent vertical elastic deflection of the electron [20, 22–25]. In addition, quantum mechanical diffraction that leads to distinct diffraction orders are observed. This effect paves the way toward ultrafast electron diffraction experiments and manipulating the spatial profile of the electron beam using near-field light as well.
The near-field zone that is responsible for inelastic interactions is tightly bound to the nanoparticle surface for slow electrons [26]. Consequently, electron wavepackets are effectively shaped according to the geometry of the nanoparticle [26]. For slow electrons, the inelastic interaction of optical near fields with electrons take advantages from the long interaction time and thus the experienced electron recoil is enhanced. Therefore, when the momentum matching is achieved over a long interaction region and duration, the electron recoil can be boosted and tailored. Furthermore, designing shapes [27], sizes [1], and geometric configurations [28] of metallic (plasmonic) nanostructures, but also laser beam properties such as wavelength and incident polarization angle [21] can provide a machinery for generating intense and highly localized plasmonic confined near-field modes [17] to foster synchronous motions between near-field oscillations and electron wavepacket—thus acting as a designing platform for shaping the free-electron wavepackets in both longitudinal and transversal directions.

Here we numerically investigate the interaction of slow electron wavepackets with localized plasmons. Particularly, we intend to explore the experienced transverse recoil of the electron beam and its dependence to the polarization of the incident laser beam interacting with plasmonic nanoparticles. We define the polarization-dependent optical near-field excitations as a quantity for populating selective momentum orders. We also demonstrate that we can pattern the final spatial distribution of the electron beam by tuning the gap configuration of plasmonic nanostructures and thus shaping the plasmonic near-field modes. We further show that the spacing between longitudinal attosecond bunches and transversal momentum exchange orders can be controlled by shaped plasmonic nanogaps.

2. Results and discussions

The interaction of electron wavepackets with laser-induced plasmon excitations results in an ultrafast amplitude and phase modulation and consequently, the energy modulation and also diffraction of electron wavepackets. The relevant quantity to calculate is thus the angle (momentum)-resolved inelastic cross-section, as we outline later. First, we investigate such effects in two-dimensional space when specific plasmonic modes of a gold nanorod are considered. Moreover, we analyse the effect of the polarization of the incident laser beam on the experienced energy modulation and recoil of the electron wavepacket. Here, we employ our recently developed self-consistent Maxwell–Schrödinger numerical toolbox [9, 22] to investigate dynamics of the spatial and spectral evolution of the electron pulse by the laser-induced near-field light. In this semi-classical approach, we solve the Maxwell equations numerically based on the finite-difference time-domain method, where the permittivity of the gold was modelled using a Drude model in addition to this semi-classical approach, we solve the Maxwell equations numerically based on the finite-difference time-domain method, where the permittivity of the gold was modelled using a Drude model in addition to two critical point function [9–11], then, Maxwell’s and Schrödinger equations are combined in a time-dependent loop, using the minimal coupling Hamiltonian [26]. The minimum gap size considered in our studies is 20 nm, beyond the range where quantum effects and Landau damping play a prominent role [29–32]. As a result, after the interaction, the final electron wavepacket (ψ_f(t, x)) is analysed to visualize the dynamics of the electron recoil and energy transfer from the electromagnetic field to the electron wavepacket.

The energy modulation of the electron wavepacket is given by solving the expectation value of the electron kinetic-energy operator as:

\[
\langle \psi(x, y; t \to \infty) \mid H \mid \psi(x, y; t \to \infty) \rangle = \frac{\hbar^2}{2m_0} \int dk_x dk_y \left( k_x^2 + k_y^2 \right) \left| \tilde{\psi}(k_x, k_y; t \to \infty) \right|^2,
\]

where (x, y) and (k_x, k_y) in equation (1) denote the real and reciprocal space coordinates, and \( \tilde{\psi}(k_x, k_y; t \to \infty) \) is the Fourier transform of the wave function after the interaction.

In the one-dimensional space, when the electron recoil is neglected, an analytical formalism is obtained to describe the energy transfer along the longitudinal direction. The discrete probability amplitude for the exchange of n quanta of energy between the electron wavepacket and the near-field light are given by expanding the wave function versus a Bessel series using Jacobi–Anger relation [26], as \( \tilde{\psi}_n(x, t)^2 \propto F_n^2(2|g|) \), where \( F_n \) is the nth Bessel function of the first kind, and g is the PINEM coupling strength specified by

\[
g = \left( \epsilon/\hbar \omega_{ph} \right) \int_{-\infty}^{\infty} dx' \tilde{E}_x(x', y, \omega) e^{-i\omega t'}/\sqrt{\epsilon}. \tag{2}
\]

For an electron propagating in the x-direction, and within the nonrecoil approximation [1] \( \tilde{E}_x \) is the Fourier transform of the x component of the scattered field. In general, the energy and momentum conservation along the electron propagation direction implies a selection rule for energy transfer from the near-field light to the electron wavepacket. Where the plasmonic near-field couples to the initial momentum state of the electron at \( p_e = \hbar k_e \), the wavefunction evolves into a superposition of momentum distribution of
$p_e = \hbar \left( k_e + n \left( \frac{\omega_{ph}}{c} \right) \right)$, where $n$ is an integer. Therefore, it forms an energy comb with the spacing between the peaks ascertained by the exciting photon energy ($\hbar \omega_{ph}$).

We first simulate the interaction of a slow electron wavepacket with the plasmonic mode around a single nanorod when the nanorod is excited with a linearly polarized laser field, with different excitation angles, using the Maxwell–Schrödinger numerical scheme. To conduct the simulation in two-dimensional space, we have assumed a plasmonic nanorod with infinite height. As schematically depicted in figure 1, an essential parameter for controlling the coupling and the interaction strength is the direction of the polarization of the excitation field with respect to the electron-beam trajectory. The electron wavepacket has an initial Gaussian distribution at the centre kinetic energy of 1 keV, and is excited by a linearly polarized laser pulse with the centre wavelength of $\lambda = 800$ nm and temporal FWHM broadening of 5 fs. The polarization and inclination angles are shown in the figure 1(a). The gold nanorod used as the nanoparticle here has a radius of 15 nm.

By assigning the kinetic energy of the electron as $E = \hbar^2 (k^2_x + k^2_y) / 2m_e$, and the scattering angle as $\varphi = \tan^{-1}(k_x / k_y)$, the inelastic scattering cross section $\sigma(E, \varphi) = (m_e / \hbar^2) \left| \tilde{\psi}(E, \varphi; t \to \infty) \right|^2$ for different inclination angles is obtained and demonstrated in figure 1(b). In general, a spreading of the electron wavepacket in the momentum space along both longitudinal and transverse directions is observed. Thus, the near-field zone acts as a mediator to overcome the phase mismatch between the light and the electron wavepacket and allows for energy and momentum exchange between them. Due to the small size of the nanoparticle, it can only hosts dipolar plasmonic fields. Thus, retardation and also higher-order multipole excitations are negligible. As we have already shown, higher order multipolar localized plasmons excited in larger nanoparticles and the race between them affect the final electron modulation spectra in a rather complicated scenario [26]. Therefore, we focus on only dipolar excitations. Here, $\theta$ is defined as the polarization angle with respect to the $y$-axis. By changing $\theta$, the induced dipolar field (figure 1(a)) align itself along the incident electric field polarization and hence the wiggling motion of the electron beam is oriented also along the same direction. This effect is particularly highlighted by demonstrating the dynamics of the electron wavepacket during the interaction (figure 2). Therefore, the different polarization of the electric field of the incident light can excite plasmonic dipoles with an angle $\theta$ relative to the electron propagation direction and causes a rotation of the populated momentum orders. The PINEM spectra as a function of laser incident angle are depicted in figure 1(c). Altering the incident angle from $\theta = 0$ to $\theta = 90^\circ$, a gradual decrease of the PINEM coupling strength (equation (2)) is observed; thus, the extend of energy gain and loss orders is reduced. In contrast, the experienced recoil along the transverse direction is enhanced, and distinct high-angle scattering lobes are observed. For $\theta = 60^\circ$, three regions in the inelastic scattering cross section are distinguished, namely, (a) the domain within the range $-2^\circ < \varphi < 2^\circ$, where the gain and loss peaks are rather symmetrically distributed, (b) the domain $\varphi < -2^\circ$, where the energy transfer is centred at $-15$ eV and within the range $-20$ eV $-10$ eV, and (c) vice versa for $\varphi > 2^\circ$, where the energy gain peaks within the

![Figure 1](New J. Phys. 25 (2023) 013033 F Chahshouri and N Talebi)
range 10 eV–20 eV are observed. This obvious asymmetry could allow for selective acceleration and deceleration of the electron beams and their detection using a mechanical slit, controlled via the near-field polarization.

Thus, concomitant electron acceleration and deceleration caused by the rotational wiggling motion of the electron beam in the dipolar near-field distribution, and quantum mechanical gain and loss peaks could be tailored to effectively shape the electron beam and engineer the recoil. For example, by detecting the energy distribution of the electron beam only for $\phi > 2^\circ$ or $\phi < -2^\circ$, via tuning the position of a filter like a mechanical slit, a uniform platform for acceleration and deceleration of a bunched electron wavepacket is obtained.

To better understand the wiggling motion of the electron beam in the optical near-field distribution, we show the dynamics of the electron wavepacket in both momentum and spatial domains, at selected times during the interaction (figure 2). An electron wavepacket at the group velocity of 1 keV interacts with a gold nanorod with the radius of 15 nm, excited by an obliquely polarized light at the centre wavelength of $\lambda = 800$ nm and the incidence angle of $\theta = 45^\circ$. Obviously, the extent of the electron wavepacket in the momentum distribution is mostly determined by the spread of the wiggling motion, which is by itself controlled by the classical Lorentz force. The experienced energy gain and loss peaks is semiclassically understood as a phase-modulation phenomena, described with the Volkov representation [9, 33]. Thus, the interference between phase-modulated and amplitude modulated waves, appears as distinct energy gain and loss peaks in the momentum distribution of the electron wavepacket.

During the interaction time, the electron experiences either a repelling or attracting force towards the nanostructure, resulting in circular wiggling motions observed in the momentum representation.
(figure 2(a)). As shown in figure 2(b), the gradual dynamics of the energy modulations in the scattering of the electron wavepacket off the near-field light can be described as the outcomes of a quantum walk [9, 23] in the discrete momentum states spaced by the classical electromagnetic waves. This behaviour leads to a gradual occupation of momentum states of the electron wavepacket by the interaction with the near field light. The ability to dynamically control the outcome of the random walk by a few parameters, such as the polarization, and incident angle of the incident optical beams, makes the proposed system as a promising candidate for boson-sampling schemes [34].

It has been shown elsewhere that an enhanced PINEM coupling strength is achieved by incorporating a so-called phase-matched configuration, where the aforementioned interference paths are constructively accumulated. This could be for example enabled by geometrical considerations, where the oscillation period of the dipole field and the interaction time of the electron beam within the effective interaction length are made to be synchronous [26]. In a system where the phase-matching condition is obtained, the overall recoil that the electron experiences can be additively accumulated as well. We now employ a nanosystem that enables both an enhanced optical near-field by incorporating gap plasmons, as well as a phase-matched configuration, to explore the effect of both on the experienced longitudinal and lateral modulations of the electron wavepacket.

In the following, we investigate the interaction of an electron wavepacket at a kinetic energy of \( U_e = 200 \text{ eV} \) with a dimer gold nanorod with a radius of 10 nm and gap spacing of 25 nm. This nanosystem is excited by an x-polarized light (figure 3, I) and a linearly polarized oblique light \( \theta = 65^\circ \) (figure 3, II), respectively, both with a centre carrier wavelength of \( \lambda = 700 \text{ nm} \) and a temporal broadening of 34 fs FWHM, with 2 GV m\(^{-1}\) laser field amplitude. Exciting the nanorods with linearly polarized light results in the manipulation of the linear momentum of the electron wavepacket, whereas circularly polarized light could be used to transfer angular momentum to the electron as well [13]. The electromagnetic near-field confinement can produce an enhanced field in the middle of the hotspot region in a plasmonic dimer nanostructure with a gap, where the localization of the field and its spatial profile can be controlled by the polarization of the incident field. Thus, owing to the coupling of the dipoles in the nanorods, the coupled plasmonic cavity system can lead to an enhanced near-field localization, which may be more beneficial in shaping electron pulses, as optical field distributions with higher momentum distributions are enabled. This configuration does not allow for a phase-matched interaction. Thus, when only a single nanorod is used, transfer of energy from the near-field to the electron wavepacket, at the extent discussed below are not observed (see supplementary figure 1 for more information).

During the interaction of the electron beam with a plasmonic dimer excited with light, momentum modulation of the electron beam can be controlled by the spatial profile of the electric field (figure 3(a)). In addition to the formation of an attosecond pulse train in real space (figure 3(a)), a quantized modulation of the electron momentum distribution (figure 3(b)) along the longitudinal and transverse directions is observed. Indeed, the gap between the nanorods supports a standing-wave-like pattern within the gap, which can cause the diffraction of the electron wavepacket in a similar way to the Kaptitza–Dirac effect [9, 23], while the longitudinal phase modulation results from the already mentioned PINEM effect. As shown in figure 3, for the inelastic scattering cross section for \( \theta = 0 \), an eye-like pattern is observed in the momentum space (figure 3(b), I). However, for the obliquely excited plasmonic dimer, the populated transverse states are more intensely occupied around the higher order gain and loss peaks (figure 3(b), II). This behaviour happens due to the symmetric and asymmetric spatial profile of the localized plasmonic modes with respect to the electron trajectory (figure 3(a)).

The transversal modulation of the electron wavepacket is better visualized by the line profiles shown in figure 3(c). The transverse electron distribution after the interaction, integrated within a short energy range (within the coloured boxes depicted in figure 3(b)), shows distinguishable momentum peaks. The incident light with \( \theta = 0 \) causes a rather symmetric momentum distribution. Because of the overall dipolar pattern of the near-field distribution, the induced near-field at one side of the cavity is against the restoring force acting on the electron at the other side. Therefore, having a symmetric dipolar configuration sets a unified electron transverse recoil in all energetically distributed photon orders. The obliquely excited plasmonic dimer causes an asymmetric recoil distribution over the electron wavepacket. This behaviour is much more pronounced for the occupied higher and lower longitudinal momentum states. The summation of the transverse probability distribution within the range of \(-4 \text{ eV} \leq E \leq +4 \text{ eV} \) shows a rather symmetric distribution, whereas for the accelerated electrons within the range of \( 14 \text{ eV} \leq E \leq 20 \text{ eV} \), or decelerated electron in the range of \(-20 \text{ eV} \leq E \leq -14 \text{ eV} \) the interspacing between the maxima of the diffraction orders is roughly equal to \( \delta k_x = 30k_{ph} = 2\pi d^{-1} \) (figure 3(c)).

A more complicated electron-bunching effect is observed by keeping the parameters of the x-polarized laser light constant but changing the configuration of the nanoparticles (figure 4(a)). The electron kinetic energy here is chosen based on the synchronicity condition \( (\lambda_{ph}v_e)/c = 2\pi \) [26] to achieve an enhanced...
coupling constant for each individual nanorod. Therefore, here, we have considered an electron pulse with the kinetic energy of 600 eV interacting with a gold nanorod (15 nm and 10 nm radius, respectively) cavity, illuminated by a laser pulse at the centre wavelength of 700 nm and the temporal FWHM broadening of 34 fs.

Therefore, here we have employed three strategies to explore the effect of the plasmonic modes on the electron modulation. In the first arrangement (figure 4, I), where there is an inversion symmetry along the \( x \)-direction, both nanorods experience the same in-phase strongly-coupled dipolar field (figure 4(a)). In the other configurations, one nanorod is rotated 30° (figure 4, II), and 60° (figure 4, III) around the \( x \)-axis, in a way that the projected spacing of all configurations along the \( y \)-axis is kept constant. For the first case, only a slight asymmetry is observed in the overall inelastic scattering cross section (supplementary figure 2 shows more detail) with respect to the origin, while the electron wavepacket receives a total transverse recoil toward the negative \( y \) axis. In the latter two cases, the delay between the wiggling motions induced by each of the dipolar near-field distributions of the individual nanorods (supplementary figure 3) results in an asymmetry in the overall inelastic scattering cross section (figure 4(b)). Particularly, higher-order energy and momentums states are more intensely populated, in contrast with the situation discussed beforehand. For the second configuration, the most intense populated states are centred around \( E = 20 \text{eV} \) and \( \varphi = -1^\circ \), as well as \( E = -20 \text{eV} \) and \( \varphi = +1^\circ \), whereas for the third case, they are located around \( E = -20 \text{eV} \) and \( \varphi = -1^\circ \), as well as \( E = +20 \text{eV} \) and \( \varphi = +1^\circ \) (figure 4(c)).

The distribution of the electron wavepacket along the longitudinal direction spaced by \( n \) integers of \( \hbar \omega_{\text{ph}} \), depicts the critical role of highly confined coupled plasmonic modes for achieving the momentum matching requirement and fostering synchronous motions between near-field oscillations and electron wavepackets. The overall extent of the PINEM spectrum (figure 4(d)), are similar for all systems, reaching up to \( \pm 40 \hbar \omega_{\text{ph}} \), meaning that quantum interference paths originating from various modes lead to a constructive interference pattern and a strong electron-photon coupling. In addition, since the extent of the gain and loss is comparable with the initial kinetic energy of the electron (13% of the initial kinetic energy), the coupling strength and therefore, the probability amplitude for each loss/gain photon order does not satisfy the nonrecoil approximation \([22]\). In addition, due to the ultrafast energy modulation in the near-field zone, the
Figure 4. Controlling the electron recoil with the near-field distribution of localized plasmons. (I) Both nanorods positioned along the y-axis, (II) second nanorod is rotated 30° around the x-axis, and (III) second nanorod is rotated 60° around the x-axis. In all three cases, the project spacing of the nanorods along the y-axis is the same. The amplitude of the electron wavepacket (a) in the real space and (b) its inelastic scattering cross section, after the interaction with the near-field zone excited by the x-polarized laser light. The laser pulse has the centre wavelength of 700 nm, the electric field amplitude of $E_0 = 2 \text{ GVm}^{-1}$, and the temporal FWHM broadening of 34 fs, respectively. (c) Transversal momentum distribution of the electron wavepacket after the interaction with the plasmonic near-field in the specific $k_x$ ranges highlighted in panel b, and (d) PINEM spectra in the overall $k_y$ range. The electron kinetic energy is 600 eV ($\beta = 0.0484 c$), and its initial longitudinal and transversal broadenings are 45 nm and 3 nm, respectively.

Figure 5. Maximizing the transverse recoil with the polarization of the incident light. (a) Inelastic scattering cross section, after the interaction with the localized plasmons in a dimer excited by the laser light with the incident angle of 60°, and (b) the PINEM spectrum, real-space distribution of the electron wavepacket and the snapshot of the spatial profile of the induced plasmonic near-field at a given time, and (c) transverse recoil of the electron beam along the elastic path $E = 0$. The kinetic energy of the electron beam is 600 eV, and its FWHM longitudinal and transversal broadenings are 45 nm and 3 nm, respectively. The centre wavelength of the laser pulse is 700 nm, and its peak field amplitude is 2 GVm$^{-1}$.

accelerated and decelerated parts of the wavepacket experience a different coupling constant with the near-field distribution. The latter phenomenon underpins the observed asymmetry patterns as well.

When exciting the dimer structure with an inclined laser pulse (figure 5) with its wave vector aligned along the symmetry axis of the dimer, the transverse recoil of the electron beam is also maximized along the elastic path $E = 0$ (figure 5(c)). Interestingly, the overall extent of the wavepacket along the longitudinal direction (figure 5(b)) is still within $-40 \hbar \omega_{ph}$ to $+40 \hbar \omega_{ph}$, ascertaining a coupling strength approximately the same as the structures investigated before, regardless of the excitation configuration of the laser beam. Comparing the inelastic scattering cross sections for all the configuration studied here, allows to conclude that while the size of the nanoparticles, the synchronicity condition, and the intensity of the laser field have a
large impact on the strength of electron–light interactions, the polarization of the light and the shape of the nanoparticles offer additional degrees of freedoms for tailoring the overall shape of the electron wavepacket along both longitudinal and transverse directions.

3. Conclusion

In conclusion, we propose a scheme to shape the electron beam by controlling the spatial distribution of localized plasmonic fields. By controlling the polarization and the geometry of the highly localized optical fields and varying the phase delay between plasmonic fields, the experienced recoil by the electrons is controlled in arbitrary angular deflections. Particularly we investigated the role of electric-field polarization on controlling the wiggling motion of the electron beam interacting with near-field distributions, and thus controlling the overall shape of the electron wavepacket. Flavouring from the merits of the plasmonic cavity for enhanced near-field localization and intensity, we expect our approach may offer a promising platform to study the electron–light interactions on the nanoscale, and it also can be applied to a wide range of applications including ultrafast phase manipulation and free-electron wavepacket shaping for applications in quantum-sensitive measurements.

Data availability statement

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

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