Interference between quantum paths in coherent Kapitza–Dirac effect

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Abstract

In the Kapitza–Dirac effect, atoms, molecules, or swift electrons are diffracted off a standing wave grating of the light intensity created by two counter-propagating laser fields. In ultrafast electron optics, such a coherent beam splitter offers interesting perspectives for ultrafast beam shaping. Here, we study, both analytically and numerically, the effect of the inclination angle between two laser fields on the diffraction of pulsed, low-energy electron beams. For sufficiently high light intensities, we observe a rich variety of complex diffraction patterns. These do not only reflect interferences between electrons scattered off intensity gratings that are formed by different vector components of the laser field. They may also result, for certain light intensities and electron velocities, from interferences between these ponderomotive scattering and direct light absorption and stimulated emission processes, usually forbidden for far-field light. Our findings may open up perspectives for the coherent manipulation and control of ultrafast electron beams by free-space light.

Introduction

Coherent control of the shape of quantum wave functions has set the way towards bond-selective chemistry [1], quantum computing [2, 3], and ultrafast control of plasmons [4]. Quantum coherent control originates from the ability to manipulate the interference between quantum paths towards realizing a desired shape of a target wave function by means of shaped, coherent, and/or strong laser excitation. Theoretically, such quantum paths may intuitively be studied using Feynman’s path integral approach [5]. This has been instrumental in interpreting observations of above-threshold ionization [6, 7] and high-harmonic generation [8], but also for selective control of the quantum paths using circular and elliptical polarizations [9]. The success of path integrals in understanding coherent control lies in the offered ease of selecting few, physically significant paths from a wealth of mathematically available options. Moreover, important concepts such as the action are easily related to classical quantities.

So far, coherent control has mostly been used to shape the wave function of bound-electron states [10], while applications in controlling free electron waves have emerged only recently [11–17]. Specifically, the inelastic interaction of electrons with optical near-fields can cause attosecond longitudinal electron bunching. Population amplitudes of certain electron states can be controlled precisely by the laser phase in a Ramsey-type experiment [18]. It has been learned that the inelastic processes involved in electron-near-field scattering require moderate laser intensities and are well understood in a minimal coupling Hamiltonian, neglecting ponderomotive forces [19].

For future progress in ultrafast electron microscopy, it appears desirable to avoid the need for matter-based near-field interactions and to simply use light waves in free space to control and shape pulsed electron beams. As a step in this direction, we show here how to use the elastic interaction of free-electron waves with focused, freely-propagating laser fields to coherently control the transversal distribution of electron wave packets.
achieved by a generalization of the Kapitza–Dirac (KD) effect [20–22] to the concomitant utilization of standing-wave and traveling-wave light patterns. In the normal KD effect, electron waves, traveling through a standing-wave pattern of light, are diffracted to transversely populated momentum states at multiples of twice the momentum of free space light. We show that the KD effect can be generalized to the realization of arbitrary momentum states of the electron wavepacket by controlling the interference between quantum pathways originating from distinctly different parts, absorptive/emissive and ponderomotive, of the interaction Hamiltonian. This offers fundamentally new degrees of freedom for designing light-controlled phase masks for free-space electron pulses.

**Results**

In a normal KD effect, electrons are propagating through a standing–wave pattern of the light intensity and in a direction perpendicular to the momentum of the light. We assume here that the standing wave pattern of light is formed by two counter-propagating light waves with the wave vectors \( \vec{k}_1 = +k_{ph,y} \) and \( \vec{k}_2 = -k_{ph,y} \), respectively. Consequently, the electrons are transversely diffracted into distinct orders which are displaced by \( \delta k_{el} = |\vec{k}_2 - \vec{k}_1| = 2k_{ph} \), where \( k_{el} \) and \( k_{ph} \) are electron and light wavenumbers, respectively (figures 1(a) and (b)). The probability distribution of the diffraction orders has been shown to be dominated by Bessel functions as \( J_n(k_{el}t) \), where \( t \) is the time of propagation of electrons through the standing–wave pattern and the coupling strength \( \kappa = e^2E_0^2/(8\hbar m_0\omega^2) \) scales with the square of the electric field amplitude \( E_0 \) [21, 23]. Here, \( e \) and \( m_0 \) are the elementary charge and mass respectively, \( \hbar \) the Planck constant, and \( \omega \) the angular frequency of the light wave. Essentially, the spatially varying light intensity introduces a periodic ponderomotive potential along the y-axis which results in a bunching of electron density in regions of low light intensity. After leaving the interaction zone, this electron wave packet transforms into a coherent superposition of plane waves propagating in direction \( k_{el}^f = k_{el} \pm n\hbar \delta k_{el} \).

Theoretically, the dynamics of such free-space electron wave packets in KD gratings are commonly described in terms of the Wolkow representation of the electron wavefunction propagating through a vector potential \( \vec{A}(\vec{r}, t) \). This representation is a special case of the Eikonal approximation where the wavefunction amplitude is taken as constant and spatial gradients of the vector potential are neglected.

Then, the wavefunction can be represented as [24]

\[
\psi(\vec{r}, t) = \psi_0(\vec{r}, t \to 0) \exp \left( -i \frac{e^2}{2\hbar m_0} \int_0^t A^{-1}(\vec{r}, \tau) \, d\tau \right) \\
\times \exp \left( -i \frac{ek_{el}^f}{m_0} \int_0^t A_x(\vec{r}, \tau) \, d\tau \right)
\]

(1)
where \( \psi_0(R, t \to 0) \) is the initial state of the electron wave packet, which for a plane wave electron is given by \( \psi_0(R, t \to 0) = (2\pi)^{-1.5}\exp(i k_x x - i\Omega t) \). Here, \( \hbar \Omega = 0.5m_0v_0^2 \), is the kinetic energy of the electron, propagating with velocity \( v_0 \) along the \( x \)-axis. As usual, the vector potential \( \hat{A} \) is related to the electric field, by neglecting the scalar potential, as \( \hat{E} = -\hat{\nabla} \cdot \hat{A} \). Furthermore, \( \hat{A}^2 = \hat{A} \cdot \hat{A} \). To describe the KD effect, it is sufficient to write the vector potential as \( \hat{A} = A_0 \hat{k} \cos(\omega t) \cos(k_{ph} y) \), which by the direct substitution in equation (1) can be cast in the form

\[
\psi(R, t) = \frac{1}{(2\pi)^{3/2}} \exp(i k_x x - i\Omega t) \exp\left\{-i \frac{2E_0^2}{2k_x \hbar^2 \omega^2} \right\} \sum_{n} \nu_n \exp\left(2i n(k_{ph} y)\right) \exp\left(i2 n(k_{ph} y)\right),
\]

For this, we have used \( E_0 = \omega A_0 \) and \( k_{ph} = \omega / c \) and have related the delay \( \tau \) to the propagation length \( x \) as \( x = v_0 \tau, \) with \( v_0 \) being the phase velocity of the electrons [25, 26]. To derive equation (2), the temporal oscillation of \( A_x^2 \cos^2 (\omega t) \), giving highly oscillatory phase variations at twice the light frequency, has been replaced by its average value \( \bar{A}_x^2 / 2 \). Then, the ponderomotive phase modulation, (the first exponential in equation (1)) shows a phase variation \( \varphi \propto \cos(k_{ph} y) = \frac{1}{2}(1 + \cos(2 k_{ph} y)) \) along the transverse direction. Using the Jacobi–Anger transform [27], this can be expressed as a series of Bessel functions corresponding to the different diffraction orders of the electron beam. This is the last term in equation (2). The probability for populating the \( n \)th diffraction order then is given as \( P_n^2 = J_n^2 \left( \frac{k_{ph}}{2m_e c} \right) \). Moreover, the argument of the exponential inside the summation in equation (2) can be written as \( i2 n k_{ph} y = \ln(k_2^\pm k_2) \cdot R \). This specifies that the final electron wave function can be regarded as a superposition of plane wave electrons occupying the momentum states \( |n, n \rangle \) in the complete basis specified by the momentum states of the two incident light beams, leading to the final momentum of \( \hbar k_2^\pm = \hbar k_2 + m h k_0 - nh k_0 \). Importantly, the phase modulation introduced by the second exponential in equation (1) is usually discarded in the description of the KD effect. As shown in more detail in the supporting information is available online at stacks.iop.org/NJP/21/093016/mmmedia, this term causes an ultrafast oscillation of the phase, which for plane wave electrons averages to zero. Hence, the second exponential in equation (1) is simply replaced by a multiplicative factor of unity in equation (2).

This shows, that in conventional treatments of the KD effect, only the ponderomotive forces on the electrons, expressed by the Hamiltonian \( H_e = e^2 A^2 / 2 \hbar m_0 \), are responsible for the phase modulations and the resulting electron diffraction. In contrast, the absorptive/emissive part \( H_0 = e k_A x / m_0 \) of the Hamiltonian in equation (1) induces only highly oscillatory phase terms, which—in a phase-cycling approach—are neglected.

The above-mentioned approximations lead to the general assumption that free-electrons and light waves cannot interact inelastically in free space. In contrast, for well-controlled electron-light interactions, achieved, e.g. by ultrafast laser excitations and/or employing slow-electron pulses, none of the previous assumptions necessarily holds true. This has recently been evidenced, e.g., by the direct acceleration and bunching of electrons with laser pulses in free space [28, 29]. In this case, \( H_0 \) may not only contribute to the elastic but also to the inelastic interaction, which will be demonstrated below. As a result, one might observe, even in free space, interferences between quantum paths arising from both parts of the Hamiltonian. We show that, for light gratings formed by optical beams with finite inclination angles, different paths can reach the momentum recoil of \( k_{ph} = 2n k_{ph} \) provided by the \( H_e \) and \( H_0 \) parts of the Hamiltonian, respectively (see figures 1(c) and (d)). These quantum path interferences appear as modulations in the diffraction pattern of the electron waves, different to the diffraction orders observed in the normal KD effect, and hence can—in principle—be observed by a regular position-sensitive detector. To verify these conclusions, we present comprehensive analytical and numerical studies, directly from first principles.

For the sake of simplicity of our analytical model, we first consider the interaction of an electron wave packet with two plane waves of light propagating at angles \( \varphi \) and \( -\varphi \) with respect to the direction of propagation of the electron (see figure 1(c)). In this way, we construct a standing wave pattern along the \( y \)-axis and perpendicular to the initial propagation direction of the electron, whereas along the \( x \)-axis, the superposition of the light beams imposes a traveling wave. The \( x \)- and \( y \)-components of the vector potential will be given by

\[
A_x = -2A_0 \sin \varphi \cos (k_{ph} \sin \varphi y) \cos (\omega t + k_{ph} \cos \varphi x) \quad \text{and} \quad A_y = -2A_0 \cos \varphi \sin (k_{ph} \sin \varphi y) \sin (\omega t + k_{ph} \cos \varphi x),
\]

respectively. Inserting them into the Wolkow propagator (equation (1)), including only the \( H_0 \) (ponderomotive) Hamiltonian, we obtain
\[ \psi(r, t) = \frac{1}{3} \exp \left( i k_ex - i\Omega t \right) \exp \left( -\frac{1}{2} \frac{e^2 E_0^2}{2\epsilon_0 \epsilon_r \omega^2} (1 + 2 \cos^2(\varphi)) \right) \]

\[ \times \sum_n p_n \left( \cos(2\varphi) \frac{e^2 E_0^2}{2\epsilon_0 \epsilon_r \omega^2} x \right) \exp \left( 2 i n \varphi \sin(\varphi) \right). \]

Evidently, for \( \varphi = 90^\circ \), equation (3) will recast equation (2). Interestingly we observe that the probabilities for coupling to the nth diffraction order, with transverse momentum \( n\varphi = n(\vec{k} - \vec{k}_0) \), now depend sensitively on the inclination angle \( \varphi \). Of particular interest is that for \( \varphi = 45^\circ \), only the 0th—diffraction order is populated. In other words, for \( \varphi = 45^\circ \), the plane wave electrons will be not diffracted by the light beams. We explain this by the competition between the \( \sin^2 \varphi \) and \( \cos^2 \varphi \) terms, respectively. Hence, at exactly \( \varphi = 45^\circ \), destructive interference between the two paths of the Hamiltonian cancels the diffraction. This cancellation at \( \varphi = 45^\circ \), will also occur for incoherent electron beams since their interaction with the pondermotive potential is phase insensitive.

To further investigate the formation of KD diffraction orders under the pure effect of ponderomotive forces, we simulated the interaction of a pulsed electron wave packet with two inclined continuous optical beams with \( E_0 = 60\ \text{GV}\ \text{m}^{-1}, \omega = 62.79\ \text{Prad}\ \text{s}^{-1} (\lambda_{ph} = 30\ \text{nm}) \), and \( \varphi = 50^\circ \). Such short-wave and intense fields, difficult to realize in the laboratory with present state-of-the-art technology, have been chosen for reducing the numerical complexity of the simulations. A scaling to less intense, longer wavelength pulses will be discussed in more detail below. In these simulations, the interaction Hamiltonian is restricted to \( H_{int} = e^2 \vec{A}^2 / 2m_0 \); i.e. we neglect the part linearly related to the vector potential. The initial longitudinal and transverse broadenings of the incident electron wave packet are \( W_L = 2.4\ \text{nm} \) and \( W_T = 20\ \text{nm} \), respectively, and its initial carrier velocity is \( v_0 = 0.03c \). At this velocity, electron experiences many ponderomotive diffraction orders up to \( n = 3 \) (see figure 2(a) for lower order diffraction probabilities). This electron wave packet, which has a pulse duration of about 2.67 fs, propagates along the x-axis, and its overall interaction time with the focused light field is roughly 6 fs, corresponding to about 26.7 oscillation cycles of the light field (figure 2(b)). In the simulations we assume a fixed relative phase between light field and electron wavepacket, as commonly is the case in ultrafast laser-driven electron generation schemes. At the exit of the interaction zone, we notice a gradual formation of symmetric and transversal electron bunching effects with up to 5 distinct lobes (figure 2(c)). This generation of KD diffraction orders is better visualized by the momentum representation of the electron wave function (figure 2(d)).

Longitudinal electron bunching along the direction of the propagation of the electron is not observed, which confirms that inelastic scattering processes are not effective. The generation of up to 3 KD diffraction orders, however, is nicely seen. Considering that the detector is not energy selective, the integration of the probability amplitude along the \( k_x \)-axis as \( P(k_x) = \int dk_x |\psi(k_x, k_y)|^2 \) gives the probability distribution of finding the electron in the transversal momentum state \( k_y \) (figure 2(e)). The peaks of the probability distribution are found at \( k_{m,0} = nk_{ph} \) and \( n\varphi k_{ph} = nk_{ph} \sin \varphi \), in agreement with the predictions of our previous Eikonal approximation (equation (3)).

Noticeable changes in the diffraction spectrum are observed when considering the full interaction Hamiltonian as \( H_{int} = -i \hbar m_0^{-1} \vec{A} \cdot \vec{\nabla} + 0.5 e^2 m_0^{-1} \vec{A}^2 \). In the Wolkow approximation, the complete wave function is then formulated as the product of two terms as \( \psi(\vec{r}, t) = \psi_\perp(\vec{r}, t) \psi_\parallel(\vec{r}, t) \), where \( \psi_\perp \) is given by equation (3) and \( \psi_\parallel \) is obtained as

\[ \psi_\parallel = \exp \left( i \frac{e k_{ph}}{m_0} 2A_0 \sin(\varphi) \sin(k_{ph} \sin(\varphi)) \right) \int_0^{t_\parallel} \cos(\omega \tau + k_{ph} \cos(\varphi) x) d\tau. \]

This can be further recast into

\[ \psi_\parallel = \sum_n \sum_{l,m} \sum_{-\infty}^{\infty} \left[ \delta(\tau - \omega \delta t) \right] \exp \left( i(\vec{k}_l - \vec{k}_0) \cdot \vec{r} \right) \]

\[ \times \int_{-\infty}^{\infty} \exp \left( i\alpha \right) F_m(\alpha) F_n(\alpha) d\alpha, \]

(5)

where \( \alpha = (e k_{ph} E_0 / \omega^2 m_0) \sin(\varphi)(1 - \cos(\omega \delta t)) \) and \( \alpha = (e k_{ph} E_0 / \omega^2 m_0) \sin(\varphi)(1 - \cos(2\omega \delta t)) \) (see supplementary information for details of the derivation). Here, \( \delta t = x / v_0 \) is the interaction time. More precisely, it is the ratio of the overall interaction time to the temporal period of the laser oscillations, i.e. \( \delta t / T \), where \( T = 2\pi / \omega \) which cause oscillations in the coupling strengths; i.e. \( \alpha \propto \sin^2(\pi \delta t / T) \) and \( \alpha \propto \sin(2\pi \delta t / T) \). This cycling behavior is akin to the Rabi oscillation of the electronic population densities between the photonic states of the momentum ladder set by the free-space electromagnetic excitations [112]. Please note that changing the interaction time \( \delta t \), which is proportional to a change in the interaction length, can be achieved by tuning the waist of the laser beam. In this sense, the interaction length is specified by \( x = v_0 \delta t \), which is used later on to derive the complete probability amplitude considering both \( H_1 \) and \( H_2 \) parts of the Hamiltonian. Note also that
indeed it is not \( \omega t \), but \( \sin(\omega t) \) and \( \cos(\omega t) \), which are appearing in the arguments of the probability amplitudes. In this sense, one can freely choose \( \omega t = nT + \theta t \) and therefore arbitrary tune the interaction length by considering as much oscillation cycles as required for achieving a realistic focused beam, where the probability amplitudes will be demonstrated later on versus modulus \( \omega t \).

The arguments of the Bessel functions, denoted as \( A_{sc} \), are functions of the parameters of the laser excitation (\( E_0 \) and \( \omega \)) as well as the parameters of the electron such as its mass, charge and initial momentum. They scale linearly with the product of initial momentum and field amplitude, \( kE \), in contrast to the arguments obtained for the ponderomotive Hamiltonian, \( H_2 \), scaling quadratically with the field amplitude. Also, the arguments are well related to the harmonics of the interaction time \( \omega t \) relative to the period of the oscillations of the laser excitation, and the angle \( \varphi \). Obviously for \( \varphi = 0 \), \( \alpha_x = \alpha_y = 0 \) and \( \psi_{ik} \equiv 1 \), regardless of the laser intensity and the interaction time. This is happening because for \( \varphi = 0 \), there exists no projection of the vector potential over the momentum of the moving electron. Moreover, for interaction times \( \omega t = nT \) with \( n \) being an integer, no diffraction which can be correlated to the \( H_2 \) part of the Hamiltonian will occur.

It is of particular interest to notice, that, in contrast to the action of the ponderomotive potential, causing the generation of diffraction orders only at harmonics of \( \delta k = k_n - k_0 \), the action of the absorptive/emissive part \( H_2 = -i\hbar m_0^{-1} \mathbf{A} \cdot \mathbf{\nabla} \) will result in harmonics of the form \( l k_n + \phi_0 \), where \( l \) and \( \phi \) can now be any arbitrary integer—note that within the Eikonal approximation \( H_1 \) can be simplified to \( H_2 = \hbar m_0^{-1} \mathbf{A} \cdot \mathbf{k}_0 \), as introduced indirectly in the Wolkow solution (equation (1)). Thus, this interaction can, in principle, prepare the electron wavepacket in states \( |l, \phi \rangle \) with momentum \( \hbar k_{d,l}^f = \hbar k_{d,l} + l \hbar k_{1} + \phi \hbar k_{2} \). In contrast, only states \( |l, -l \rangle \) can be accessed by ponderomotive scattering. The probability of finding the electron after the interaction in state

![Figure 2](image-url)

**Figure 2.** Diffraction of electron pulses by the ponderomotive potential of a standing wave light pattern at the wavelength of \( \lambda_0 = 30 \text{ nm} \). (a) Population densities in the lowest diffraction orders as a function of electron velocity and electric field amplitude of the light beams. (b) Probability of finding the electron in one of the three lowest diffraction orders, calculated for a plane wave electron at a velocity of \( u_0 = 0.03c \). Amplitude of the electron wave function for an electron moving at \( v_c = 0.03c \) in (c) real space and (d) momentum space, at selected times during the interaction with the standing-wave light field. The electric field amplitude of each optical beam is \( E_0 = 60 \text{ GV m}^{-1} \) and the wavelength is 30 nm. (e) Transverse momentum distribution of the electron pulse before and after the interaction with the standing-wave field, demonstrating the diffraction of the electron wave into orders \( k_{d,l}(n) = n\hbar k_{\phi,l} \).
\[ |l, o\rangle \text{ can be written as} \]
\[ P_{l}^{\delta t, \varphi} = \sum_{m, n} i^{m+n} J_{m}(-f(\alpha), f(\alpha) J_{m}(-\alpha) J_{n} J_{m} J_{n}) \]
\[ (6) \]

Interestingly, for fixed values of \( \delta t \) and \( \varphi \), we notice a different dependence of the population densities \( P_{l}^{\delta t, \varphi} \) on electron velocity and electric field amplitude than for \( P_{l}^{\delta t} \) in the case of purely ponderomotive scattering (compare figure 3 with 2(a)). Even at very high field amplitudes, it is still possible to observe the 0th diffraction order, albeit only for electrons slower than 0.03c. The locations of the maxima in general follow a hyperbolic curve in both \( (E_{0}, v_{el}) \) and \( (E_{0}, \lambda_{ph}) \) spaces. This is different for the curves formed under the action of the \( H_{1} \) Hamiltonian, which follow a parabolic shape. For fast electrons, the electron wave function populates higher momentum states already for much lower field amplitude since now the coupling scales linearly with the field amplitude and electron momentum. This behavior is in distinct contrast with the action of the ponderomotive potential on the electron, for which larger field amplitudes will be necessary to observe higher order diffractions of the electron by the light. At a specific photon energy and electron velocity, two other parameters, i.e. \( \delta t \) and \( \varphi \) might be used to control the diffraction orders (figure 3, bottom panels). For interaction times given by \( \delta t = n T \), the Rabi oscillations between the photonic states will cause the population densities to average back to the initial \( |0, 0\rangle \) state. For this reason, controlling the interaction time appears as a precise way of tuning the population densities. Additionally, the excitation angle \( \varphi \) of the photon beams with respect to the direction of the propagation of the electron affects the diffraction orders correlated with both the \( H_{1} \) and the \( H_{2} \) parts of the Hamiltonian, however in a quite different way. At a given interaction time, being set by the longitudinal broadening of the electron wave packet, the angle \( \varphi \) can still be used to suppress or release certain diffraction orders.
Indeed, the ability to reach the momentum states \( |l, o \rangle \) is intrinsic to the \( H_2 \) part of the Hamiltonian. In the case of an interaction with standing wave patterns of plasmons, this interaction results in energy loss and gain effects, as reported elsewhere [30]. However, in contrast to our case, \( |l, o \rangle \) states are only temporally populated and are averaged out after the interaction (see figure 5 and discussions therein). Nevertheless, by means of an appropriate tuning of the coupling strengths specified by both \( H_1 \) and \( H_2 \) parts of the Hamiltonian, arbitrary momentum states are occupied as will be discussed in the following.

Importantly, in the presence of large field amplitudes and slow electrons, both parts of the Hamiltonian, i.e. \( H_1 \) and \( H_2 \) parts may contribute to the diffraction of the electron beam. This offers a new degree of freedom for manipulating the temporal and spatial structure of the electron pulse by controlling quantum path interferences resulting from the action of both, \( H_1 \) and \( H_2 \), on the transition of the electron beam from \( |0, 0 \rangle \) initial state to the final state \( |l, o \rangle \). To better understand this, we derive the final electron wave packet using equations (1), (3), and (4) as

\[
\psi(r, t) = \frac{1}{\sqrt{(2\pi)^2}} \exp(-i\hat{r}\hat{\tau}) \times \exp \left( \left( k_{\text{cl}} - \frac{e^2E_0^2}{2\hbar^2k_{\text{cl}}^2} \cos^2(\varphi) \right) x \right) \times \sum_p \sum_n \sum_m \left( \begin{array}{c} i^{l+2n+m} f_{p}(\alpha_z) I_m(\alpha_{c}) \times \exp(i [\hat{k}_1 + \hat{k}_2] \cdot \hat{r}) \end{array} \right).
\]

As a result the probability of finding the electron at the final \( |l, o \rangle \) momentum state will be given as

\[
P_{l,o} = \left| \sum_p \sum_n \sum_m \left( \begin{array}{c} i^{l+2n+m} f_{p}(\alpha_z) I_m(\alpha_{c}) \times \exp(i [\hat{k}_1 + \hat{k}_2] \cdot \hat{r}) \end{array} \right) \right|^2.
\]

This establishes an interesting interference pattern for each given final \( |l, o \rangle \) momentum state (figure 4). One might observe, at specific ranges of field amplitude and electron velocities, quantum coherent interference paths in the diffraction orders. In contrast to previously reported Rabi oscillations in photon-induced near-field electron microscopy [12, 18], the quantum interferences discussed here will occur in free space, and may be utilized as a neat way of controlling the transversal distribution of the electron wave packets, without changing their energy. Interestingly, even in the presence of the \( H_2 \) interaction, the probability of finding the electrons in the final \( |l, -l \rangle \) states is still dominant over \( |l, 0 \rangle \) states. More specifically, the interferences between \( H_1 \) and \( H_2 \) interations are only observed at specific electron velocities. At higher velocities, it is the \( H_2 \) interaction which is dominating the final diffraction orders of the electron wavepacket, whereas at lower electron velocities, the pondermotive interaction is more pronounced (compare figure 4 with figures 2(a) and 3). This behavior implies that there is a range of electron velocities which allows for the above mentioned interference phenomenon to take place between the \( H_1 \) and \( H_2 \) interaction paths. This range of electron velocities strongly depends on applied light frequencies and interaction time as well.

For example, for \( \lambda_{\text{ph}} = 30 \) nm, \( \nu_{l} = 0.03c \), and at proper laser field amplitudes, it should be possible to observe an interference between \( |0, 0 \rangle \) \xrightarrow{H_1} \( |l, -l \rangle \) and \( |0, 0 \rangle \) \xrightarrow{H_2} \( |l, -l \rangle \) paths, and furthermore to coherently control it by means of the incidence angle \( \varphi \) and the laser intensity.

The above-mentioned theory is further benchmarked by directly employing a time-dependent Schrédinger solver based on first principles to analyze the electron-light interaction in the regime where we expect interferences between both terms of the Hamiltonian (see Materials and Methods section for more details). The electron wave packet has initial longitudinal and transverse broadenings of \( W_{\mu} = 20 \) nm and \( W_{\nu} = 25 \) nm, respectively, and its kinetic velocity is \( \nu_{l} = 0.03c \). The laser carrier wavelength and electric-field amplitude are set to \( \lambda_{\text{ph}} = 30 \) nm and \( E_0 = 200 \) GV m\(^{-1}\), respectively and \( \varphi = 50^\circ \). Including the complete Hamiltonian, the interaction of the electron wave packet with the laser beams (figure 5(a)) is very different from the case of purely pondermotive scattering (figure 2(c)). The center of the wave packet is soon depopulated and the wavepacket breaks up into a sequence of sub-peaks along the transverse direction. These subpeaks are becoming increasingly narrower as the wavepacket continues to propagate through the interaction region. The formed bunches of ultrathin wave packets move further away from the interaction region along a direction preassigned by the wave vector of the light beams, hence building up the coherent diffraction orders. In the momentum space, we notice the population of the electron beams prominently into the \( |l, -l \rangle \) states, evident by the 5 distinguished bright spots along the transverse direction. However, fainter probability maxima located at \( \vec{k}_{\text{cl}} \pm \hat{k}_i \) and \( \vec{k}_{\text{cl}} \pm \hat{k}_j \) unravel from the non-equilibrium population distribution at \( |\pm 1, 0 \rangle \) and \( |0, \pm 1 \rangle \) states, as well as higher order \( |0, \pm 2 \rangle \) and \( |\pm 2, 0 \rangle \) states, which only last for ultrashort times during the interaction (figure 5(b)). Moreover, these states provide means to form the desired interference paths between various quantum levels (see...
resulting in diffraction orders which are not located at \( \pm \ell \), states (figures 5(d) and (e)). These diffraction orders appear as ultrathin electron bunches which are caused by the interference between the above-mentioned quantum paths (figure 5(c)). Interestingly, all the diffraction levels will form along the circumference of an Ewald-like sphere (marked in figure 5(d)). The radius of the Ewald sphere is equal to the wavenumber of the electron \( k_{el} \), as expected. This further strengthens the fact that the inelastic light–matter interaction, though temporarily happening during the interaction of an electron wave packet with continuous-wave light in vacuum, cannot give rise to a steady-state outcome. This is due to the fact that (i) both light waves specified by wave vectors \( \vec{k}_1 \) and \( \vec{k}_2 \) have exactly similar field strengths, polarizations, and wave numbers, while the only difference between them is the propagation direction, and (ii) the probability amplitude for the transition of the electron wave function from \( |0, 0, 0\rangle \) to the final states \( |\pm \ell, \pm \ell, 0\rangle \), under the effect of \( H_2 \) interaction and for a given \( \ell \), are all equal. Nevertheless, the transverse (elastic) scattering to the two \( |\ell, -\ell\rangle \) and \( |-\ell, \ell\rangle \) states will be caused by both \( H_1 \) and \( H_2 \) parts of the Hamiltonian, and the different paths giving rise to the transverse interaction will interfere with each other, causing the appearance of subpeaks in the detected diffraction pattern, which are not displaced by \( n k_{el} \) (figure 5(e)).

The above mentioned quantum coherent interferences can be controlled by several parameters, including \( \varphi \) and \( E_0 \). Of particular interest is the role of the laser electric-field amplitude. For slow electrons and low laser
intensities, the role of the pondermotive force will become dominant. This is to be understood particularly by comparing the figure 4(d) with figures 3 and 2(a). As a result one expects to observe diffraction peaks at the order of only $d\frac{\rho}{k}n$, which is further confirmed by our first-principle calculations (figure 6). This ponderomotive scattering remains dominant at electric field amplitudes $E \leq 100$ GV m$^{-1}$. However for $E > 100$ GV m$^{-1}$, the diffraction orders correlated with the $H_2$ part of the Hamiltonian become active as well, and the resulting diffraction pattern is an outcome of the interference between the quantum paths associated with each part of the Hamiltonian. By increasing the field amplitude, the overall number of diffraction orders will be also increased. Interestingly, the highest order diffraction patterns are all at the orders of only $d\frac{\rho}{k}n\hat{k}$, which demonstrates that the ponderomotive potential still dominates the overall response. Hence the distribution of the diffraction orders due to the $H_2$ part of the Hamiltonian is within a smaller angular range, when compared to the total angular distribution of the diffraction patterns.

**Discussions**

The short wavelength choice of $\lambda_{ph} = 30$ nm and ultrahigh field amplitudes employed above, were only introduced to meet our simulation resources. These parameters are far beyond what practically can currently be realized in the normal laboratories, but a free-electron laser might be used for a commensurate light wavelength and intensity. It is to be noticed that the coherent phase manipulations suggested here appear to be indeed achievable by the current state-of-the-art ultrafast electron sources, which in interaction with laser-induced near-field polarizations, now even lead to the generation of a train of attosecond electron pulses [12]. The effects that we describe here, however, are neither restricted to isolated or bunches of attosecond pulses. They mainly require ultrafast driving lasers with sufficiently high field amplitudes to enable a strong ponderomotive acceleration of the electron pulses passing through the interaction region. Now, we briefly outline laboratory-
based practical approaches towards the realization of quantum path interferences with field strengths and laser wavelengths routinely employed in light–matter interaction experiments. The initial condition for the formation of interferences in quantum paths is to have similar levels of interaction strengths associated with $H_1$ and $H_2$ parts of Hamiltonian. Since $H_1$ is quadratic and $H_2$ is linear in $\vec{A}$, there will be always two values for vector potential amplitudes where $H_1$ and $H_2$ meet each other—one point is $A_0 = 0$ and the other point is related to the electron velocity as $A_0 = \frac{2m_0v_0}{\epsilon}$. Thus, the lower the electron velocity, the lower becomes the required field amplitude (figure 7(b)). Additionally, the electric field amplitude is related to the vector potential as $E_0 = \omega A_0$. In other words, it will be possible to employ lower field amplitudes by using oscillating fields at lower frequencies (figure 7(b)). For example, by assuming a laser wavelength of $\lambda_{ph} = 1250$ nm, the quantum path interferences between $H_1$ and $H_2$ will be observed at electron velocities within $0.01 \leq v_0 < 0.1 \leq c$ and electric field amplitudes as low as $0.2 \text{ GV m}^{-1} < E_0 < 1.0 \text{ GV m}^{-1}$ (figures 7(c) and (d)). Such conditions can certainly be created by currently available light sources, or by including near-field field-enhancement effects [31]. Additionally, we have performed numerical simulations using $\lambda_{ph} = 800$ nm and $E_0 = 10 \text{ GV m}^{-1}$, and an electron wavepacket at the kinetic energy of $U_e = 600 \text{ eV} (v_0 = 0.0484c) W_e = 60 \text{ nm}$, and $W_p = 250 \text{ nm}$ to consider another situation with realistic lab-based parameters (see supporting information for more details).

The appearance of new diffraction orders due to combined action of the ponderomotive part $H_1$ and the absorptive/emissive part $H_2$ of the interaction Hamiltonian inevitably requires energy- and momentum conservation in the light–electron interaction. Commonly, for electrons in free space that are driven by sufficiently weak light pulses, this is not fulfilled since the slope of the free-electron dispersion relation is always smaller than the speed of light. This weak-field limit is indeed well-reproduced in our simulations (see figure 2). Modifications in the diffraction pattern appear in a regime of rather strong light intensities, i.e. strong ponderomotive acceleration of the light-driven electrons. In this regime, in a classical picture, the momentum mismatch between electrons and far field light can be overcome by transiently gaining momentum by the light-driven ponderomotive acceleration of the electrons. This additional contribution to the momentum change is essential to explain the absorption and/or emission of photons during the presence of the driving laser pulse. The momentum change that is needed to transiently match the energy- and momentum conservation is not overly large. Even for slow electrons, where the momentum mismatch is largest, and photon energies in the visible spectral range (1200 nm), a ponderomotive change in electron velocity of about $0.001c$ is needed to fulfill momentum matching. For electrons with a velocity of $0.1c$, this change in velocity decreases further to $10^{-4}c$, requiring electric field amplitudes in the GV m$^{-1}$ range. For lower photon energies, the required field

Figure 6. Dependence of the diffraction orders on the electric field amplitude. (a) Spatial distribution of the amplitude of the electron wave function and (b) integrated probability distributions after the interaction versus the amplitude of the electric-field component of the laser excitation. $\varphi = 50^\circ$, $W_e = 20 \text{ nm}$, $W_p = 25 \text{ nm}$, and $v_0 = 0.03c$. See the text for description of the parameters.
amplitudes reduce even further, supporting again that the effects that are discussed in the present manuscript can indeed be verified experimentally with present day technology.

Dynamics of sideband modulations in the scattering of single-particle electron wave packets with free-space light can be described as the outcomes of a quantum walk \[32\] in the discrete momentum states specified by the classical electromagnetic waves. This behavior is akin to the random walks of photons in classical reliable interferometers with low losses and high stability performances \[33\]. As a result of the coherent action of the unitary operator specified in equation (1) on the single electron wave packet, at each given time the electron will be left entangled between different momentum states. As expected, features of this quantum walk are interferences and boson sampling \[33\] from a sea of many possible states, as for example selectively selecting few elastic or inelastic scatterings, as a result of such inferences. The ability to dynamically control the outcome of the random walk by few parameters as the polarization, wavelength, intensity, and the inclination angle of the incident Gaussian beams, and particularly by avoiding matter and hence electron-electron and electron-core interactions, make the proposed system a possible promising candidate for bosonic-fermionic random walks and new generation of boson-sampling devices \[32, 34, 35\].

As a summary, we have generalized the KD effect to the inclusion of two laser fields which propagate at an inclination angle with respect to the electron trajectory. The spatio-temporal behavior of the introduced light waves appears as a standing wave pattern transverse to the electron momentum—similar to the normal KD effect. In addition, a traveling wave pattern forms longitudinal to the initial electron velocity—in contrast with the normal KD effect. We have shown, both theoretically and numerically, that the interaction of the electron wave packet with the introduced light beams, results in an exotic momentum distribution in the final electron wave packet, which can be described as the quantum path interferences between two parts of the Hamiltonian, namely ponderomotive and absorptive/ emissive channels. These interference paths and their coupling strengths can be further controlled in general by the shape of the light waves and in particular by the inclination

Figure 7. Criterion for observation of quantum path interferences between ponderomotive $H_1$ and absorptive/ emissive $H_2$ parts of the Hamiltonian. (a) Interaction strengths versus the amplitude of the magnetic vector potential, for different electron velocities. (b) $|H_1 - H_2|$ as a function of frequency and electric field amplitude, at the electron velocity of $v_{el} = 0.1c$. Population densities for (c) $|1, -1\rangle$ and $|1, 1\rangle$ and (d) $|2, -2\rangle$ and $|2, 2\rangle$ diffraction orders resulting from the interaction of an electron beam with a propagating light field created by two inclined Gaussian light beams. The electrons interact with both the ponderomotive $\hat{A}$ and the absorptive/ emissive $\hat{A} \cdot \hat{k}$ parts of the Hamiltonian. Here $\varphi = 50^\circ$, $\delta t = 0.3T$, and $\lambda_{0} = 1250$ nm. Much less electric field amplitudes are required to observe the interference effects as compared with figure 6.
angles and intensity of the two Gaussian beams, but also by tuning the interaction time and the electron velocity. We anticipate that the aforementioned interference effects can be proposed as new boson sampling device for tailoring the random walk of a single quantum wavepacket in the discrete levels imposed by the momentum of the light.

Materials and methods

For our first principle calculations, we have used a time-dependent propagator combined with a pseudospectral Fourier method [36, 37], which conserves the norm of the wave function altogether, and propose a convergent numerical scheme. The accuracy and stability of the numerical method can be controlled by an appropriate choice of the time steps. All the simulations were performed in two-dimensional space. This is particularly rationalized by the choice of polarizations for the fields (magnetic field normal to the simulation domain, electric field lying in the plane). In this way, the classical trajectory of the electron and its experienced quantum mechanical recoils all remain in the simulation plane. Moreover, only a single electron wavepacket is considered. For calculating the optical Gaussian beams, we have used the analytical solutions based on the paraxial approximations, which perfectly model the laser excitations, and is valid for focus regions larger than 1.5 \( \lambda \), where \( \lambda \) is the optical wavelength. Here the waist of the introduced optical beams are 2\( \lambda \). We however have benchmarked this approximation by comparing our results with those obtained using a self-consistent Maxwell–Schrödinger numerical toolbox, and the same diffraction orders and probability amplitudes have been noticed [38].

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Notes

The authors declare that they have no conflict of interest.

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