Femtosecond plasmon and photon wave packets excited by a high-energy electron on a metal or dielectric surface

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Swift electrons generate coherent transition radiation (TR) when crossing a material surface, as well as surface plasmon polaritons (SPPs) when the material is metallic. We present analytical and numerical calculations that describe the time- and space-dependent electric fields of TR and SPPs induced by 30–300 keV electrons on a Drude metal surface. The generated SPPs form wave packets a few-hundred femtoseconds in duration, depending on the material permittivity. High-frequency components close to the plasmon resonance are strongly damped, causing the wave packets to shift to lower frequencies as they propagate further. TR is emitted to the far field as ultrashort wave packets consisting of just a few optical cycles, with an intensity and angle dependence that is determined by the material permittivity. The excitation reaches its peak amplitude within a few femtoseconds and then drops off strongly for longer times. From a correlation between material permittivity and the calculated emission behavior, we determine qualitative predictions of the TR evolution for any given material. The results presented here provide key insights into the mechanisms enabling swift electrons to serve as nanoscale optical excitation sources.

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I. INTRODUCTION

Electron-beam spectroscopies such as cathodoluminescence (CL) spectroscopy and electron energy-loss spectroscopy (EELS) have gained much attention in nanophotonics research because of their ability to resolve optical excitations with nanometer precision. Just like photons, electrons carry electromagnetic fields, allowing them to optically excite polarizable matter. In fact, due to their short de Broglie wavelengths and correspondingly high momenta, electrons can be used as a highly localized optical excitation source with a spatial resolution unattainable using optical excitation techniques [1–7]. Many advances have been made in the theoretical descriptions of the interactions between electrons and matter that generate radiation [1,8–13]. These electron-light-matter interactions occur on the femtosecond time scale and have only recently been studied in the time domain, in an indirect way, using adapted electron microscopes that combine EELS with synchronized ultrafast optical excitations [14,15]. Most work thus far has focused on exploring the frequency and momentum domains, measuring and simulating spectral responses. Accordingly, analytical theory and simulation techniques have not been applied to investigate the time evolution of CL processes in detail, although time-domain simulation methods have been gaining traction [16,17].

In this article, we develop a theoretical framework to study the radiation excited by swift electrons impinging on a metallic or dielectric surface. We investigate in detail the time evolution of coherent excitation and emission of transition radiation (TR) and surface plasmon polaritons (SPPs). We examine the space, time, and frequency dependence of the electric fields of the moving electron and how they interact with matter to generate radiation. We start by deriving the external fields of an electron in a homogeneous medium, determining the time- and position-dependent behavior of the electric-field components. We then examine an electron impinging on a planar surface, interacting with the medium to induce fields that produce radiation emitted into the far field. The general formalism is derived in frequency and momentum space, after which the time and space dependence is obtained by Fourier transforming the fields. First, we investigate SPPs and study the generation and propagation of these excitations on the femtosecond time scale, for metals described by a Drude dielectric response. We determine the evolution of the SPP wave packet as it propagates away from the point of excitation. We then explore TR, which is composed of far-field emission characterized by ultrashort wave packets that strongly depend on the emission direction. We study both the time and frequency dependence of TR and elucidate the interrelation between the material permittivity and TR emission. This allows us to formulate qualitative predictions of TR emission behavior.

II. ELECTRON EXTERNAL FIELD

A point charge moving with constant velocity in vacuum possesses an electromagnetic field that represents an evanescent source of radiation. Inside a homogeneous medium, the field can be described quite simply, allowing us to study both the time and spectral dependence of the electron field components. We focus on an electron traveling in a straight-line trajectory along the z axis, with a constant velocity vector $\mathbf{v} = v\hat{\mathbf{z}}$, passing by the origin $R = z = 0$ at time $t = 0$. The direction perpendicular to the trajectory is denoted as $\mathbf{R} = R\hat{\mathbf{R}}$, with the position vector defined as $\mathbf{r} = (\mathbf{R},z)$. The electron charge density is given by $\rho(\mathbf{r},t) = -e\delta(\mathbf{r} - \mathbf{v}t)$. This can be Fourier transformed to ($\mathbf{q}$-ω) space as $\rho(\mathbf{q},\omega) =$

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$-2\pi e\delta(\omega - \mathbf{q}\cdot\mathbf{v})$. In our derivations, we use Gaussian units and follow the notation used in Ref. [1], but focus only on the electric fields $\mathbf{E}$ (the magnetic field $\mathbf{H}$ can be obtained using the Maxwell-Faraday equation). The equations take into account retardation effects (c is finite and the electron velocity can reach a sizable fraction of c). We also use linear response theory, which assumes that the induced field is linear with the external field of the electron and, consequently, the photon emission probability scales as the square of the external charge ($-e$ for the electron). Maxwell's equations can be solved in momentum-frequency space, leading to the following expression for the electric field of the moving electron:

$$\mathbf{E}(\mathbf{q},\omega) = -\frac{8\pi^2 ie}{q^2 - k^2\epsilon}\left(\frac{k}{c}\mathbf{v} - \frac{\mathbf{q}}{\epsilon}\right)\delta(\omega - \mathbf{q}\cdot\mathbf{v}), \quad (1)$$

where ϵ is the permittivity of the homogeneous medium and $k = \omega/c$ is the free-space wave number. The momentum $\mathbf{q}$ of the electron field can be decomposed as $\mathbf{q} = (\mathbf{Q},q_z)$, in components perpendicular ($\mathbf{Q}$) and parallel (q_z) to the trajectory, with $q_z = \omega/v$. The latter expression, which expresses energy conservation for transfers of frequency and wave vector from the electron to the material, is the nonrecoil approximation [1]. It holds for $\hbar q^2/m_e \ll \omega$, which is valid for photon-energy exchanges $\hbar\omega \ll 1$ MeV, as is usually the case in the study of photonic nanostructures. Integrating $\mathbf{E}(\mathbf{q},\omega)$ over the z component of $\mathbf{q}$ results in

$$\mathbf{E}(\mathbf{Q},z,\omega) = \frac{4\pi ie}{v\epsilon}\frac{\mathbf{q} - \mathbf{v}k\epsilon/c}{q^2 - k^2\epsilon}e^{i\omega z/v}. \quad (2)$$

The electron dispersion $\omega = qv$ lies outside the light cone in free space $\omega = kc$, so the electron does not radiate and the electric field decays exponentially away from the trajectory. In contrast, the electron velocity can exceed the speed of light inside a material, so the electron can couple to excitations in the medium, leading to the emission of Cherenkov radiation; we will not study this here. Performing the Fourier transform over $\mathbf{Q}$ to obtain the electric field in real space, we obtain [1]

$$\mathbf{E}(\mathbf{r},\omega) = \frac{2e\omega}{v^2\gamma_\epsilon\epsilon}e^{i\omega z/v}\left[\frac{i}{\gamma_\epsilon}K_0\left(\frac{\omega R}{v\gamma_\epsilon}\right)\hat{\mathbf{z}} - K_1\left(\frac{\omega R}{v\gamma_\epsilon}\right)\hat{\mathbf{R}}\right], \quad (3)$$

where $\gamma_\epsilon = 1/\sqrt{1 - \epsilon v^2/c^2}$ is the Lorentz contraction factor and K_m are modified Bessel functions of the second kind [18] (see also Chap. 14 of Ref. [19]). The fields diverge at the origin, so close to the trajectory there is a large contrast in the field strength. Further away from the trajectory, the field amplitude decays with the Bohr cutoff ($v\gamma_\epsilon/\omega$) as a characteristic decay length [1,4]. Using Eq. (3), we can determine the spectral components of the electric field at different points in space. To determine the electric field as a function of time, we Fourier transform Eq. (3) as

$$\mathbf{E}(\mathbf{r},t) = \int \frac{d\omega}{2\pi}\mathbf{E}(\mathbf{r},\omega)e^{-i\omega t}. \quad (4)$$

Direct numerical integration is used below for the fields produced when the electron crosses an interface. For the swift electron moving in vacuum ($\gamma_\epsilon \to \gamma$), the $E(\mathbf{r},t)$ fields can be determined directly from $\rho(\mathbf{r},t)$ and the Liénard-Wiechert potentials (see pp. 661–665 in Ref. [19]), leading to

$$\mathbf{E}(\mathbf{r},t) = -\frac{\gamma}{[R^2 + \gamma^2(z - vt)^2]^{3/2}}[R\hat{\mathbf{R}} + (z - vt)\hat{\mathbf{z}}]. \quad (5)$$

For illustration, we study an electron traveling through vacuum, with an energy of 30 keV, corresponding to a velocity of $v = 0.328c = 98.45$ nm fs^{-1}. The results are displayed in Fig. 1, with a schematic of the electron, the coordinate system, and the field orientations as an inset in Fig. 1(a). We present the evanescent decay of the field away from the trajectory of the electron in Fig. 1(a), calculating the total electric-field intensity (time integrated over the entire pulse) as a function of radial distance R, for the two field components E_R and E_z. Notice that the electric field produced by the moving electron diverges at its position, thus reflecting the divergence of the expected value of the electric force that it produces on a test point charge at an arbitrarily small separation from it. Incidentally, this divergence of the real part of the self-interaction is known to be removed by renormalization in the calculation of the Lamb shift [20] or in a proper derivation of the polarizability of a point particle [21]. In CL and EELS, it also disappears because the transition probability is limited to a value given by the imaginary part of the self-interaction [1]. This asymptotic behavior of the fields is clearly visible in Fig. 1(a), with a difference of eight orders of magnitude in the intensity between $R = 1$ nm and $R = 100$ nm. The E_R field component, perpendicular to the trajectory, has a higher intensity than the E_z component. The strong gradient in the field intensity observed here is responsible for the very high excitation resolution of electron-beam spectroscopies.

Figure 1(b) displays the electric-field components as a function of time, for distances $R = 10$ nm and $R = 100$ nm away from the electron trajectory. The E_z component is asymmetric, while the E_R component is symmetric, both displaying a single oscillation. For a distance of $R = 10$ nm, the field transient occurs within $\sim$2 fs, highlighting the extremely short pulse felt by an observer close to the moving electron. At $R = 100$ nm, the field amplitudes have decayed by a factor 100 and the transient spreads out in time by a factor 10. This is due to the fact that further away from the trajectory, the distance between moving electron and observer varies more slowly with time.

Figure 1(c) displays the electric field in the frequency domain. Spectra for the two field components are presented for $R = 10$ nm and the total intensity for $R = 10$ nm and $R = 100$ nm. The different field components display characteristic spectral shapes, with E_z vanishing for small frequencies while E_R has a maximum. At $R = 10$ nm, the spectral range extends well beyond 10 eV; at $R = 100$ nm, the intensity has decayed by a factor 100 and the spectrum is confined to $\sim$1 eV ($\sim$242 THz).

These calculations show that an electron moving through vacuum possesses an electric field that is characterized (at a certain position) by ultrashort pulses, with energies extending beyond 10 eV close to its trajectory. The field strength and spectral content decay when moving further away. Since these fields are evanescent, they cannot couple directly to far-field

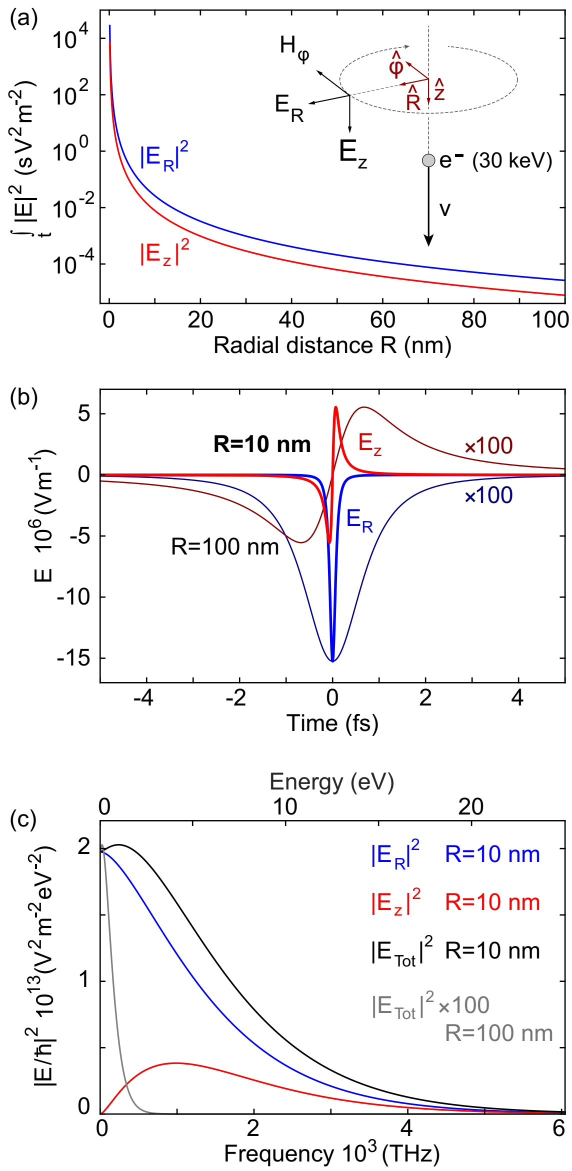

FIG. 1. Electric field produced by a 30 keV electron in vacuum. (a) Total (time-integrated) electric-field intensity of the radial (E_R, in blue) and vertical (E_z, in red) components, as a function of radial distance to the electron trajectory R, displaying the diverging intensity close to the electron trajectory at $R = 0$. The inset depicts a schematic of the moving electron with the coordinate system and orientation of the field components. We evaluate the electric fields for a given radial distance R, height z, and time t or frequency ω. (b) Electric-field amplitudes of the E_R (blue) and E_z (red) components as a function of time, for distances of $R = 10$ nm (thick lines) and $R = 100$ nm (thin lines, amplitudes multiplied by 100), and with the minimum electron-observer distance at $t = 0$. (c) Electric-field intensity in the frequency domain, as a function of frequency and corresponding energy, for the E_R (blue), E_z (red), and $E_{\rm tot}$ (black) components at a distance of $R = 10$ nm, compared to the total intensity for $R = 100$ nm (gray, multiplied by a factor of 100).

radiation. Next, we study an electron impinging on a dielectric medium, leading to processes that can emit radiation.

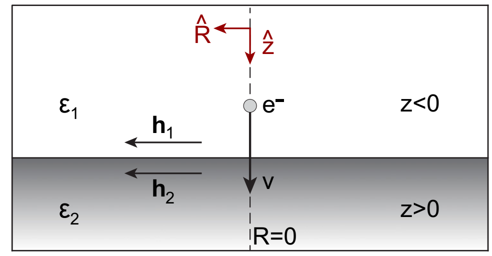

FIG. 2. Schematic of the electron traversing the interface between two media, presenting the coordinate system, as well as the permittivities ϵ_j and surface currents $\mathbf{h}_j$ on either side of the interface.

III. AN ELECTRON IMPINGING ON A PLANAR SURFACE

When an electron reaches and traverses the interface between two media, it generates transition radiation (TR), as well as surface plasmon polaritons (SPPs) in the case of a metal. In general, the electric field can be described by separating the contributions of the external field of the electron in each medium (as if it were an infinite homogeneous medium) and the field that is induced at the surface. The induced field is created by surface charges and currents induced by the approaching electron,

$$\mathbf{E} = \mathbf{E}_j^{\rm ext} + \mathbf{E}_j^{\rm ind}.$$

Figure 2 depicts a schematic of the geometry studied here. The induced field can be expressed in terms of the surface currents $\mathbf{h}_j$ as [1,10,22]

$$\mathbf{E}^{\rm ind}(\mathbf{Q},z,\omega) = \frac{-2\pi k}{q_{zj}} e^{iq_{zj}|z|}\bigg(\mathbf{h}_j - \frac{1}{k^2\epsilon_j}[\mathbf{Q},{\rm sign}(z)q_{zj}] \times \{[\mathbf{Q},{\rm sign}(z)q_{zj}]\cdot\mathbf{h}_j\}\bigg). \qquad (6)$$

Here, $q_{zj} = \sqrt{k^2\epsilon_j - Q^2 + i0^+}$, where the square is chosen to yield a positive real part. From the boundary conditions (i.e., the continuity of the $\mathbf{E}$ and $\mathbf{H}$ components parallel to the surface), it follows that the currents only have components of parallel momentum $\mathbf{Q}$ and can be written as $\mathbf{h}_j = D\mu_j\hat{\mathbf{Q}}$. The induced electric field takes the form

$$\mathbf{E}_j^{\rm ind}(\mathbf{Q},z,\omega) = \frac{2\pi}{k\epsilon_j} D\mu_j e^{{\rm sign}(z)iq_{zj}z}[-q_{zj}\hat{\mathbf{Q}} + {\rm sign}(z)Q\hat{\mathbf{z}}], \qquad (7)$$

where

$$D = \frac{2ieQ/c}{\epsilon_1 q_{z2} + \epsilon_2 q_{z1}}, \qquad (8)$$

$$\mu_1 = \frac{\epsilon_1 q_{z2} - \epsilon_1\omega/v}{q^2 - k^2\epsilon_2} - \frac{\epsilon_1 q_{z2} - \epsilon_2\omega/v}{q^2 - k^2\epsilon_1}, \qquad (9)$$

and

$$\mu_2 = \frac{\epsilon_2 q_{z1} + \epsilon_1\omega/v}{q^2 - k^2\epsilon_2} - \frac{\epsilon_2 q_{z1} + \epsilon_2\omega/v}{q^2 - k^2\epsilon_1}. \qquad (10)$$

We can now combine the external and reflected fields together to obtain the complete expressions for the electron electric fields on both sides of the interface, for both field components. Recall from Eq. (2) that the external field contains components $\mathbf{q} - \mathbf{v}k\epsilon/c$, with $\mathbf{q} = (\mathbf{Q},\omega/v)$. We obtain the following expressions for $\mathbf{E}(\mathbf{Q},z,\omega) = \mathbf{E}_R + \mathbf{E}_z = E_R\hat{\mathbf{Q}} + E_z\hat{\mathbf{z}}$:

$$E_R = \frac{4\pi i e}{v\epsilon_1}\frac{Q}{q^2 - k^2\epsilon_1}e^{i\omega z/v} - \frac{2\pi}{k\epsilon_1}q_{z1}D\mu_1 e^{-iq_{z1}z}, \quad z < 0 \tag{11a}$$

$$E_z = \frac{4\pi i e}{v\epsilon_1}\frac{\frac{\omega}{v}\left(1 - \frac{v^2}{c^2}\epsilon_1\right)}{q^2 - k^2\epsilon_1}e^{i\omega z/v} - \frac{2\pi}{k\epsilon_1}QD\mu_1 e^{-iq_{z1}z}, \quad z < 0 \tag{11b}$$

$$E_R = \frac{4\pi i e}{v\epsilon_2}\frac{Q}{q^2 - k^2\epsilon_2}e^{i\omega z/v} - \frac{2\pi}{k\epsilon_2}q_{z2}D\mu_2 e^{iq_{z2}z}, \quad z > 0 \tag{11c}$$

$$E_z = \frac{4\pi i e}{v\epsilon_2}\frac{\frac{\omega}{v}\left(1 - \frac{v^2}{c^2}\epsilon_2\right)}{q^2 - k^2\epsilon_2}e^{i\omega z/v} + \frac{2\pi}{k\epsilon_2}QD\mu_2 e^{iq_{z2}z}, \quad z > 0. \tag{11d}$$

In order to obtain the fields as a function of space and time, we Fourier transform back from ($\mathbf{Q}$-ω) space:

$$\mathbf{E}(\mathbf{r},t) = \int \frac{d^2\mathbf{Q}}{(2\pi)^2}e^{i\mathbf{Q}\cdot\mathbf{R}}\int\frac{d\omega}{2\pi}e^{-i\omega t}\mathbf{E}(\mathbf{Q},z,\omega). \tag{12}$$

The integral over $\mathbf{Q}$ can be partially simplified by removing the azimuthal component of $\mathbf{Q}$ to leave only the radial part, so that we obtain a single integral over Q. This approach differs between the two field components, since $\mathbf{E}_z$ contains no vectorial component of $\mathbf{Q}$, but $\mathbf{E}_R$ does. We find for $\mathbf{E}_z$ that

$$\int \frac{d^2\mathbf{Q}}{(2\pi)^2}e^{i\mathbf{Q}\cdot\mathbf{R}} = \int_0^\infty \frac{Q}{2\pi}dQ J_0(QR),$$

while for $\mathbf{E}_R$,

$$\int \frac{d^2\mathbf{Q}}{(2\pi)^2}e^{i\mathbf{Q}\cdot\mathbf{R}}\hat{\mathbf{Q}} = \int_0^\infty \frac{Q}{2\pi}dQ i J_1(QR)\hat{\mathbf{R}}.$$

We apply these identities to Eq. (12), and further use causality $[E(\omega) = E^*(-\omega)]$, to reduce the fields to

$$\mathbf{E}_z(\mathbf{r},t) = \int_0^\infty \frac{Q}{2\pi}dQ J_0(QR)\int_0^\infty\frac{d\omega}{\pi}\mathrm{Re}\{e^{-i\omega t}\mathrm{E}_z(Q,z,\omega)\}\hat{\mathbf{z}}, \tag{13a}$$

$$\mathbf{E}_R(\mathbf{r},t) = \int_0^\infty \frac{Q}{2\pi}dQ J_1(QR) \times \int_0^\infty \frac{d\omega}{\pi}\mathrm{Im}\{-e^{-i\omega t}\mathrm{E}_R(Q,z,\omega)\}\hat{\mathbf{R}}, \tag{13b}$$

where $E_z(Q,z,\omega)$ and $E_R(Q,z,\omega)$ are given by Eq. (11). Now that we have derived this general formalism, these integrals can be solved numerically to study SPP and TR generation. From these equations, we can derive analytical expressions for the electric-field components in the far field. We demonstrate this in the following sections.

IV. SURFACE PLASMON POLARITONS

The excitation of surface plasmon polaritons by fast electrons was discovered several decades ago [23–25] and has since been demonstrated in a broad variety of experiments [2,26–28]. In studying the excitation of SPPs, we can ignore the external electric field as it decays evanescently and thus does not generate radiation. In our formalism, plasmons are revealed by the induced electric fields [Eq. (7)] and, more specifically, they originate in the pole of the denominator of D: $\epsilon_1 q_{z2} + \epsilon_2 q_{z1} = 0$. This equation leads to the plasmon dispersion relation,

$$Q_{\rm SPP} = k\sqrt{\frac{\epsilon_1\epsilon_2}{\epsilon_1 + \epsilon_2}}. \tag{14}$$

We now use the plasmon-pole approximation [1,10,29], retaining only the contribution of the plasmon pole at $Q = Q_{\rm SPP}$. For large distances ($R \gg \lambda$), one can perform the integral over Q from Eq. (13), allowing us to derive analytical expressions for the fields of the SPPs. Using this method, D becomes

$$D \approx \frac{C}{Q - Q_{\rm SPP}}, \tag{15}$$

where the denominator arises from a Taylor expansion around the plasmon pole,

$$\epsilon_1 q_{z2} + \epsilon_2 q_{z1} \approx -Q_{\rm SPP}\left(\frac{\epsilon_1}{q_{z2}} + \frac{\epsilon_2}{q_{z1}}\right)(Q - Q_{\rm SPP}) = \frac{(Q - Q_{\rm SPP})}{A},$$

leading to

$$C = \frac{2eiQA}{c}.$$

To determine the SPP electric fields in space as a function of frequency, we can rewrite the frequency-domain part of Eq. (13), making the plasmon pole explicit, and perform the integral over Q,

$$\mathbf{E}_{\rm SPP}(\mathbf{r},\omega) \propto \int_0^\infty dQ J_m(QR)\frac{f(Q,z,\omega)}{Q - Q_{\rm SPP}} \approx 2\pi\frac{i}{2}f(Q_{\rm SPP},z,\omega)H_m^{(1)}(Q_{\rm SPP}R), \tag{16}$$

where $H_m^{(1)}$ is a Hankel function of the first kind. By combining Eq. (16) with Eq. (11), we can consolidate an expression for the frequency-dependent SPP fields in a relatively compact way, and find

$$\mathbf{E}_{\rm SPP}(\mathbf{r},\omega) = \frac{2\pi e i A Q_{\rm SPP}^2}{ck\epsilon_j}\mu_j e^{\mathrm{sign}(z)iq_{zj}z}\big[\mathrm{sign}(z)iQ_{\rm SPP} \times H_0^{(1)}(Q_{\rm SPP}R)\hat{\mathbf{z}} + q_{zj}H_1^{(1)}(Q_{\rm SPP}R)\hat{\mathbf{R}}\big]. \tag{17}$$

This equation is applicable on both sides of the interface that the electron traverses. As in the previous section, the final step to obtain the time-dependent electric fields is to Fourier transform over the frequency domain,

$$\mathbf{E}_{\rm SPP}(\mathbf{r},t) = \int_0^\infty \frac{d\omega}{\pi}\mathrm{Re}\{\mathbf{E}_{\rm SPP}(\mathbf{r},\omega)e^{-i\omega t}\}. \tag{18}$$

We calculate the time- and frequency-dependent SPP electric fields for a 30 keV electron impinging on a Drude metal under normal incidence, with a permittivity described by

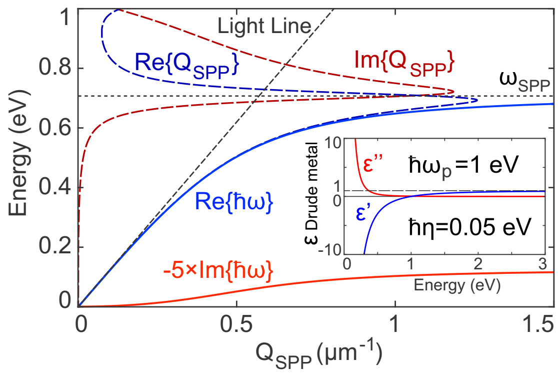

FIG. 3. Dispersion relation of surface plasmon polaritons (SPPs) for a Drude metal with $\hbar\omega_p = 1$ eV and $\hbar\eta = 0.05$ eV, displaying the energy as a function of the SPP wave vector $Q_{\rm SPP}$. Real (imaginary) parts are represented as blue (red) curves for both complex $Q_{\rm SPP}$ (dashed curves for Re$\{Q_{\rm SPP}\}$ and Im$\{Q_{\rm SPP}\}$, as a function of real energy in the vertical axis) and complex ω (solid curves for Re$\{\hbar\omega\}$ and $-5 \times$ Im$\{\hbar\omega\}$, as a function of real wave vector in the horizontal axis). The diagonal dashed line denotes the light cone, while the horizontal one indicates the energy of the SPP resonance frequency $\omega_{\rm SPP}$. The inset depicts the permittivity ϵ of the Drude metal as a function of energy.

$\epsilon(\omega) = \epsilon_{\rm D}(\omega) = \epsilon_0 - \omega_{\rm p}^2/(\omega^2 + i\eta\omega)$. The Fourier transform is performed numerically by summing over energies in the range $\hbar\omega = 10^{-6} - 100$ eV, divided into 10^6 steps. We choose $\hbar\omega_{\rm p} = 1$ eV and $\hbar\eta = 0.05$ eV. The corresponding plasmon-dispersion relation is depicted in Fig. 3, the SPPs being excited for frequencies $\omega \leqslant \omega_{\rm SPP} = \omega_{\rm p}/\sqrt{2}$. We show representations of both complex frequency and complex wave vector in the dispersion relation.

The representation with real wave vector and complex frequencies is well suited for pulsed excitations (i.e., such that a single frequency is not well defined), in which the imaginary part of the complex frequency describes the decay in time [30]. In contrast, a quasimonochromatic, spatially delocalized excitation is better represented by the real-frequency description, in which the imaginary part of the complex wave vector accounts for propagation losses. These two representations are very similar along most of the dispersion curve, except near the SPP horizontal asymptote, where the complex wave-vector picture describes a band bending followed by negative dispersion, while the complex frequency picture fully retains the asymptotic behavior. For the electron incident on a planar surface, both representations are equivalent, depending on which of the integrals (ω or Q) is performed earlier in Eq. (12). In the expression of Eq. (18), the frequency remains real, as we have chosen to carry out the wave-vector integral first. The Im$\{\omega\}$ term is negative (as expected for $e^{-i\omega t}$ to decay for $t \to \infty$), so we multiply it by -5 to show it on the same scale as the other quantities.

Figure 4(a) shows the time evolution of an SPP wave packet, as observed for a height $z = -10$ nm above the metal surface, at a radial distance $R = 10$ μm from the electron trajectory ($R \gg \lambda$). After ~30 fs, we clearly observe an increase in amplitude and an oscillating wave packet that then decays, for both field components. As expected for the transverse-magnetic (TM) polarized SPPs, E_z oscillates with larger amplitude than E_R. The wave packet has a duration of a few-hundred femtoseconds, and considering the distance R and the time of the onset, is propagating close to the

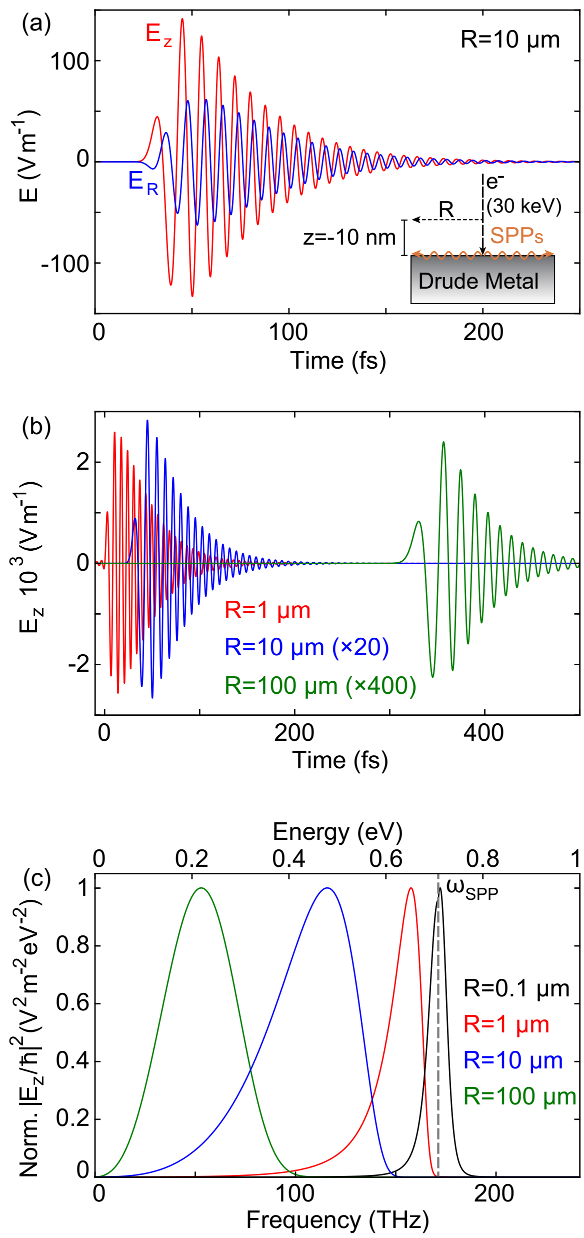

FIG. 4. Electric field of surface plasmon polaritons (SPPs) excited by a 30 keV electron, propagating along the surface of a Drude metal (same parameters as in Fig. 3). (a) Electric-field amplitudes of the SPP wave packet, showing the E_R (blue) and E_z (red) components as a function of time, evaluated for a height 10 nm above the metal surface and a distance $R = 10$ μm away from the electron impact position (see inset). The time $t = 0$ corresponds to the electron crossing the interface. (b) Comparison of the E_z SPP amplitude as a function of time for a fixed height $z = -10$ nm at different distances of 1 μm (red), 10 μm (blue, multiplied by 20), and 100 μm (green, multiplied by 400). (c) Comparison of the intensities of the E_z SPP field component in the frequency domain, each normalized to the maximum value at the same distances as in (c). Data for $R = 0.1$ μm (in black) are also presented. The SPP resonance frequency $\omega_{\rm SPP}$ is indicated.

speed of light. This is to be expected at large distances and times, as the lowest-frequency components close to the light line propagate with the lowest loss and the slower, high-frequency components are strongly damped; see both $\mathrm{Im}\{\hbar\omega\}$ and $\mathrm{Im}\{Q_{\mathrm{SPP}}\}$ in Fig. 3. In Fig. S1 of the Supplemental Material [31], we present the full time evolution and spectral behavior over a 60 μm range.

In Fig. 4(b), we compare the time evolution of the SPP wave packet at $R = 1$, 10, and 100 μm. The period of the oscillations in the wave packet increases for further distances, corresponding to the shift to lower frequencies. The wave packet reaches its maximum amplitude within a single optical cycle, as expected for the single-cycle excitation; see Fig. 1(b). It then decays within ~200 fs after the initial onset, the duration determined by the damping rate η. In Fig. S2 of the Supplemental Material [31], we compare the SPP wave-packet evolution for different material parameters and electron energies. Essentially, the electromagnetic field oscillates in time with a characteristic period $\sim 1/\omega_p$, and therefore all the results reported here can be scaled appropriately for other values of the plasma frequency. The attenuation introduced through η affects the temporal extension of the field (increasing $|\mathrm{Im}\{\omega\}|$) and, consequently, also the number of oscillations that can be resolved.

To compare the time evolution to the spectral behavior, we calculate the frequency-dependent fields in Fig. 4(c), where data for a distance of $R = 100$ nm are also used. All spectra have been normalized to their maximum: 5.1×10^{12}, 5.7×10^9, 7.4×10^6, and 2.3×10^4 V^2 m^{-2} eV^{-2} for $R = 0.1$, 1, 10, and 100 μm, respectively. Close to the electron trajectory, for $R = 100$ nm, the SPP spectrum is very sharp and peaks at ω_{SPP}. For larger distances, the spectrum broadens and shifts to lower frequencies, as expected from the frequency-dependent damping discussed above.

V. TRANSITION RADIATION

Transition radiation (TR) is a common form of radiation excited by an electron interacting with matter, as it occurs for any swift electron (or charged particle) crossing the boundary between two different media [32–36]. A simple and intuitive way to view TR is that the electromagnetic fields of the electron are "in equilibrium" with their environment, obeying the equations for a homogeneous medium outlined above. If the two media are different, however, the fields of the electron must have adjusted to the new electromagnetic properties of the second material. This modification of the fields as the electron transitions between the media is then accompanied by radiation. Jackson describes TR as portions of the electromagnetic field that must shake it off as radiation (see pp. 646–654 in Ref. [19]). An alternate intuitive explanation is provided by the method of image charges. The negatively charged electron produces a positive image charge below the surface, inducing an effective dipole normal to the surface which vanishes and radiates when the electron passes through the interface. This problem can be treated by describing the field lines of two moving charges of opposite sign that instantaneously stop (at the interface) or start (moving away from the interface) [37–39]. More generally, the approaching electron induces surface charges and currents which polarize the atoms in the material close to the trajectory. These polarization charges react and create an induced field which can radiate out to the far field as transition radiation.

The emission actually must originate from the induced field, just as for SPPs, since the homogeneous fields decay evanescently away from the trajectory of the moving electron and do not couple directly to far-field radiation, so we can discount them in the derivation of the TR fields. Using the general formalism derived for the single interface, we can write the $\mathbf{E}(\mathbf{r},\omega)$ fields, taking only the integral over Q from Eq. (12) applied to the components of Eq. (11):

$$\mathbf{E}(\mathbf{r},\omega) = \int_0^\infty \frac{Q}{2\pi} dQ \frac{2\pi}{k\epsilon_j} D\mu_j e^{iq_{z1}|z|} \times [J_0(QR)\mathrm{sign}(z)Q\hat{\mathbf{z}} - iJ_1(QR)q_{zj}\hat{\mathbf{R}}]. \quad (19)$$

TR is emitted to the far field, so taking the limit for large distances, $kr \longrightarrow \infty$, we can evaluate the integral over Q for the two different field components. Here we examine only the upper hemisphere ($z < 0$, see Fig. 2), taken to be vacuum:

$$\int_0^\infty dQ J_0(QR) e^{iq_{z1}|z|} f(Q) \approx -i\frac{q_{z1}}{Q}\frac{e^{ikr}}{r} f(Q), \quad (20a)$$

$$\int_0^\infty dQ J_1(QR) e^{iq_{z1}|z|} f(Q) \approx -\frac{q_{z1}}{Q}\frac{e^{ikr}}{r} f(Q), \quad (20b)$$

where the value of Q in the right-hand side of the equation is determined by the emission direction (see below). Applying this approximation to Eq. (19) leads to a compact expression for the spectral components of the TR electric field:

$$\mathbf{E}_{\mathrm{TR}}(\mathbf{r},\omega) = i\frac{q_{z1}}{k} D\mu_1 \frac{e^{ikr}}{r}(Q\hat{\mathbf{z}} + q_{zj}\hat{\mathbf{R}}). \quad (21)$$

The fields for the lower hemisphere can be obtained by analogy, using the corresponding components from Eq. (11). Making use of the geometrical relations between R, z, and r as well as Q, q_{z1}, and k, we can define an angle θ that determines the emission direction with respect to the surface normal: $\cos\theta = q_{z1}/k = z/r$ and $\sin\theta = Q/k = R/r$. Making use of these relations, we can rewrite the vectorial part of Eq. (21) as

$$(Q\hat{\mathbf{z}} + q_{zj}\hat{\mathbf{R}}) = k\left(\frac{R}{r}\hat{\mathbf{z}} + \frac{z}{r}\hat{\mathbf{R}}\right) = k\hat{\theta}.$$

This helps to further simplify Eq. (21), which reduces to

$$\mathbf{E}_{\mathrm{TR}}(\mathbf{r},\omega) = iq_{z1} D\mu_1 \frac{e^{ikr}}{r}\hat{\theta}. \quad (22)$$

We can rewrite this with a more explicit dependence on θ, finding a result very similar to that for the magnetic field as derived in Ref. [1]:

$$\mathbf{E}_{\mathrm{TR}}(r,\theta,\omega) = ik\cos\theta D\mu_1 \frac{e^{ikr}}{r}\hat{\theta}. \quad (23)$$

We can evaluate this expression for $Q = k\sin\theta$. The asymptotic part $f(Q)$ of Eq. (20) can be used to obtain the TR emission probability by integrating over the upper hemisphere, leading to

$$\Gamma_{\mathrm{TR}}(\omega) = \frac{1}{2\pi\hbar k}\int_0^{\pi/2} \sin\theta d\theta |k\cos\theta D\mu_1|^2. \quad (24)$$

Finally, Eq. (23) can also be used to determine the TR electric field as a function of time by performing the Fourier transform over frequency,

$$\mathbf{E}_{\mathrm{TR}}(r,\theta,t) = \int_0^{\infty} \frac{d\omega}{\pi} \mathrm{Re}\{\mathrm{E}_{\mathrm{TR}}(r,\theta,\omega)e^{-i\omega t}\}\hat{\theta}. \tag{25}$$

For a nondispersive medium, this can be simplified even further, since there are no longer any frequency-dependent components except for the $e^{-i\omega t}$ term. The integral over ω then results in a $\delta(r-ct)$ term. In particular, for a perfect electric conductor, the TR electric field in space and time reduces to [38]

$$\mathbf{E}_{\mathrm{TR}}(r,\theta,t) = \frac{2ev\sin\theta}{rc}\frac{\delta(r-ct)}{1-\left(\frac{v}{c}\right)^2\cos^2\theta}\hat{\theta}. \tag{26}$$

We evaluate Eq. (25) numerically, since it is generally applicable for any material. We use a fixed distance $r_0 = \sqrt{2}$ μm from the origin (so that $-z = R = 1$ μm for $\theta = 45^\circ$) for different combinations of R and z to obtain a suitable distribution of angles θ. We use a 30 keV electron impinging on the same Drude metal as for the SPP calculation, with $\hbar\omega_p = 1$ eV and $\hbar\eta = 0.05$ eV, summing over energies in the range $\hbar\omega = 10^{-6} - 100$ eV, divided into 10^6 steps. We find that a large range of energies is necessary to obtain good convergence ($\hbar\omega \sim 100$ eV), although the main features are resolved even with a smaller range ($\hbar\omega \sim 10$ eV). These values are, however, related to the energy/frequency dependence of the material permittivity. The larger the range in frequencies over which ϵ displays features, the larger the range needed in the calculation for good convergence.

Figure 5 displays the results of these time-resolved calculations. Figure 5(a) presents the TR electric-field intensity (on a log scale) as a function of the emission angle θ and normalized time $T = t - r_0/c$. Radiation emitted at the instant the electron traverses the interface will reach the position of the observer (at a distance r_0) at the time $T = 0$, at which a sharp increase in intensity is observed for all angles. Just as for the SPPs, TR forms a very short electromagnetic wave packet that travels through space. Comparing different angles, we clearly observe different regimes. For angles up to $\sim 60^\circ$, there are distinct oscillations in the intensity, which propagate longer at lower angles. For angles above $\sim 60^\circ$, we no longer see these oscillations, but instead see a uniform decay in intensity.

To study the time evolution in more detail, we directly compare the time trace for different angles (denoted by the horizontal white dashed lines), showing the TR electric-field amplitude on a linear scale, in Fig. 5(b). The inset depicts a schematic of the process, with the simple model of the negatively charged electron inducing a positive mirror charge that creates a vertical dipole. Indeed, the angular emission pattern of TR displays two lobes (representing a cross cut through a toroidal three-dimensional pattern), similar to the emission from a vertical point dipole above the interface. Examining the TR wave packets, it is obvious that for all angles there is no discernible emission before $T = 0$. The radiation only begins at $T = 0$, rising rapidly to a maximum within a few femtoseconds, before decaying within ~ 30 fs (for the material parameters used here). TR thus consists of extremely short-lived wave packets. Comparing the different

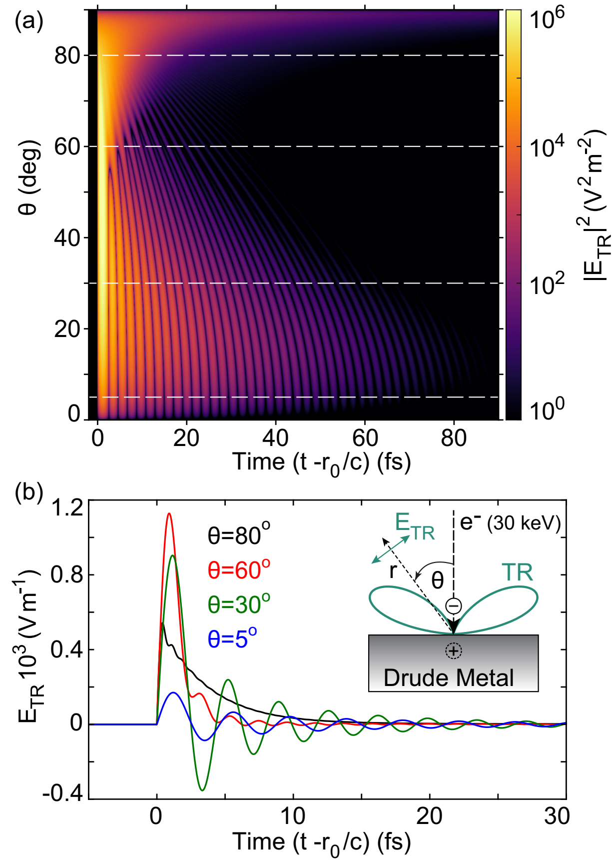

FIG. 5. Electric field of transition radiation (TR) excited by a 30 keV electron, propagating away from the same Drude metal as in Figs. 3 and 4, as a function of time. (a) The TR electric-field intensity, shown on a logarithmic scale, as a function of the emission angle θ and of the normalized time $T = t - r_0/c$, with $r_0 = \sqrt{2}$ μm the position at which the field is calculated for each angle [see inset schematic in (b)]. (b) Comparison of the TR electric-field amplitude as a function of the normalized time for four different emission angles, corresponding to the dashed lines in (a). The inset depicts a schematic of the TR excitation and emission process.

emission angles, we notice a variation in the maximum amplitude, with the smallest and largest angles exhibiting a lower intensity than those in between (by a factor $\sim$2–5). This agrees with the expected dipolar emission pattern that also exhibits low intensity for small and large angles with a maximum in between. At small angles of emission, $\theta = 5^\circ$ and 30°, we observe clear oscillations in time, which have a similar initial period before becoming out of phase after the second optical cycle. For higher emission angles, $\theta = 60^\circ$ and 80°, the oscillations are strongly damped and the time evolution displays a single slow decay from the initial amplitude maximum.

In order to gain insight into this strong angle-dependent, short-lived behavior, we calculate the TR electric field as a function of frequency. Figure 6(a) shows the TR intensity as a function of frequency and emission angle. The intensity is limited to certain sections of θ-ω space. For the Drude metal with $\hbar\omega_p = 1$ eV, the permittivity approaches unity above an energy of several eV, so the electron will not perceive a

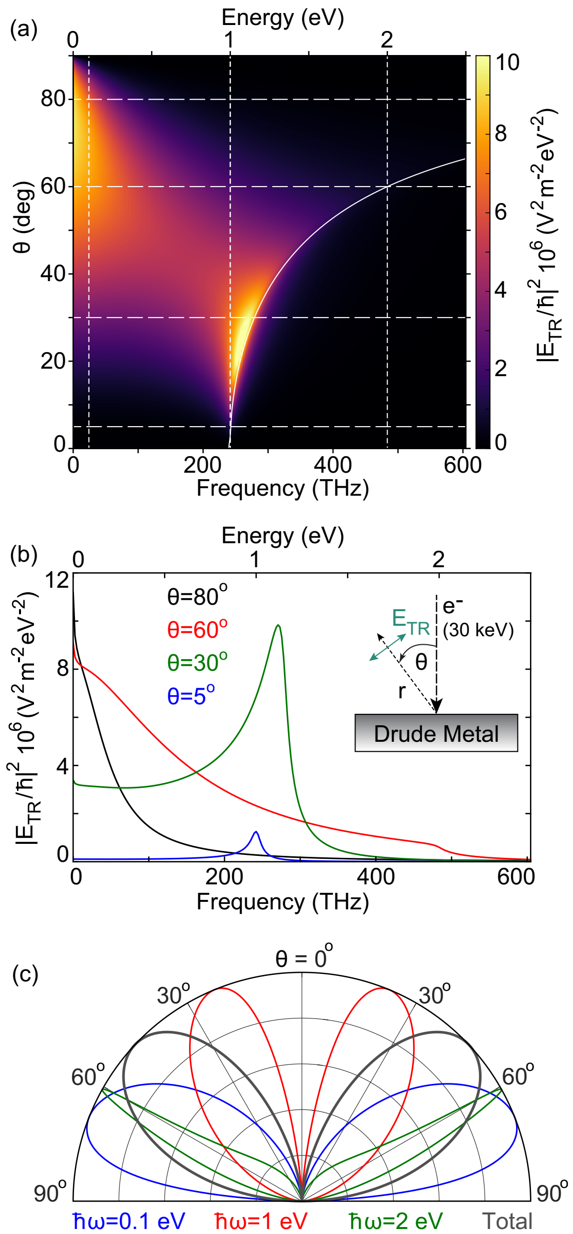

FIG. 6. Electric field of TR in the frequency domain, excited by a 30 keV electron impinging on the same Drude metal as in Figs. 3–5, evaluated at the same distance $r_0 = \sqrt{2}$ μm as in Fig. 5. (a) TR electric-field intensity as a function of emission angle θ and frequency/energy. The curved white line corresponds to $\theta = \arcsin(\sqrt{\epsilon_D})$. (b) Comparison of the TR electric-field spectra for different emission angles, denoted by the horizontal dashed lines in (a). The inset depicts a schematic of the TR excitation and emission geometry. (c) Normalized TR electric-field intensity as a function of emission angle, for the total integrated intensity (gray) and for different frequencies, denoted by the vertical dashed lines in (a).

transition in that energy range. The intensity is also depleted at the smallest and largest angles for most of the frequency range. Examining Eq. (23), we find that it contains a $\cos\theta$ term and a $\sin\theta$ term (due to Q), so $E_{\rm TR}$ is expected to go to 0 for angles close to 0 and $\pi/2$. Physically, these angles correspond to either very small Q or q_{zj}, indicating that the light modes are more delocalized (in space) and the coupling between the electrons and the impact region is smaller, leading to lower TR intensity.

At low frequencies and large angles (grazing to the surface), there is a broad and intense feature that occurs when the real and imaginary parts of ϵ_2 reach very large magnitudes ($\epsilon_2 = \epsilon_D$ of the Drude metal). At small angles (close to the surface normal), there is a sharp, bright peak that starts at $\omega = \omega_p$ and then bends off to higher frequencies and larger angles before disappearing around $\theta = 40^\circ$. This feature occurs when $|\epsilon_D|$ displays a sharp kink and can be a result of an accumulation effect comparable to van Hove singularities [40,41]. The kink in $|\epsilon_D|$ bears resemblance to divergences in the density of states that lead to anomalies in optical absorption spectra. An additional explanation, described by Ferrell [42] and Stern [43] for metal foils, is that the incoming electron drives plasma oscillations of electrons at the surface, which radiate at the plasma frequency ω_p. This emission is only expected when Im$\{\epsilon\}$ is much smaller than 1, which is the case here. In the Supplemental Material [31], we confirm that the effect indeed disappears for increasing loss.

We attribute the sudden depletion of the TR intensity of this feature for $\omega > \omega_p$ to the coupling strength between the electron and the material becoming focused in the forward direction, into the metal. The dispersion relation of the light inside the metal is given by $q = k\sqrt{\epsilon_D}$, while the parallel wave vector of the emitted radiation is $Q \leqslant q$. The boundary for coupling the energy into the metal instead of the far field of the upper hemisphere should thus be determined by $\sin\theta = Q/k = \sqrt{\epsilon_D}$, so $\theta = \arcsin(\sqrt{\epsilon_D})$. We find excellent agreement with the data, as confirmed by the curved solid white line in Fig. 6(a), as well as later on in Fig. 7 and in Fig. S5 of the Supplemental Material [31]. This explanation is consistent with the behavior of the μ_1 term [Eq. (9)] that appears in Eq. (23): the first denominator $q^2 - k^2\epsilon_D$ goes to 0 for $q = k\sqrt{\epsilon_D}$, leading to the divergent behavior that we observe.

These results indicate that the frequency-dependent permittivity of the Drude metal leaves a strong imprint on the TR emission. We will study this dependence on permittivity in more detail in Fig. 7. Additionally, we study the effect of different material parameters and electron energies on the TR emission explicitly in both the time and frequency domains in Figs. S3–S5 of the Supplemental Material [31].

While we present extensive results for a Drude metal with a characteristic bulk plasmon energy of 1 eV, the duration of the emitted wave packets of both SPPs and TR depends on η and ω_p. For a constant ratio between these quantities the pulse duration will be inversely proportional to ω_p, so that it becomes ∼15 times shorter for a prototypical Drude metal such as aluminum for example.

In Fig. 6(b), we examine the corresponding spectra for the same angles as shown in Fig. 5(b) (denoted by the horizontal white dashed lines). For $\theta = 5^\circ$, we note a low intensity but a clear peak in the spectrum at the plasma frequency ω_p. For $\theta = 30^\circ$, the spectrum is still dominated by this peak that has moved to slightly higher frequencies, but there is a noticeable increase at lower frequencies as well. We can now say that the oscillations in time from Fig. 5 for small angles correspond roughly to the plasma frequency. For larger angles, the frequency of oscillations increases. For $\theta = 60^\circ$, a broad, decreasing spectral band is observed up to ∼2 eV (∼484 THz).

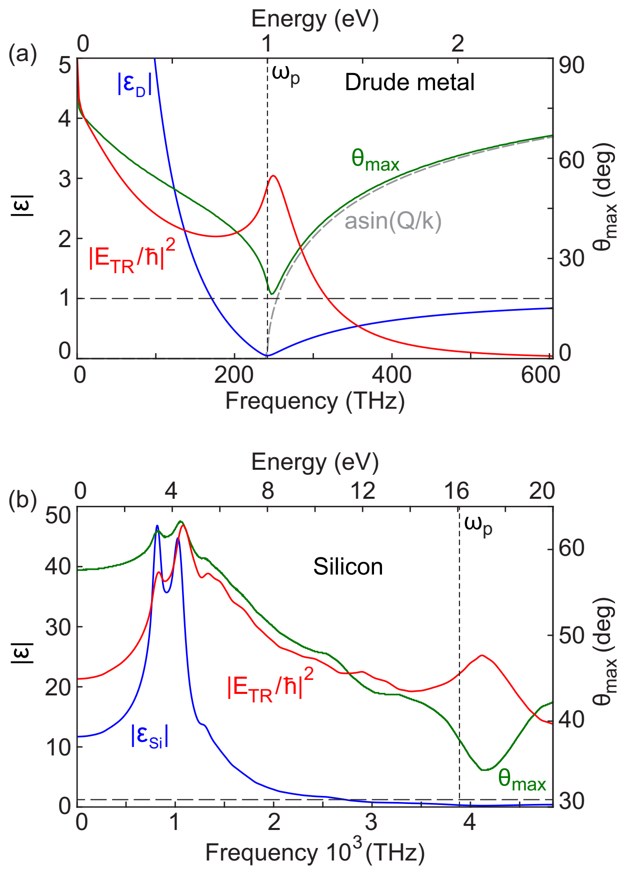

FIG. 7. Dependence of TR on the material permittivity ϵ. (a) Absolute value of the permittivity of the Drude metal ($\hbar\omega_p = 1$ eV and $\hbar\eta = 0.05$ eV), as a function of frequency/energy (in blue) compared to the normalized total TR electric-field intensity (integrated over all angles, in red) and the angle of maximum emission intensity $\theta_{\max}$ (in green). The TR intensity spectrum is normalized to the same maximum as $|\epsilon|$, the vertical dashed line denotes the plasma frequency, and the horizontal dashed line indicates $|\epsilon| = 1$. The gray dashed line corresponds to $\theta = \arcsin(Q/k) \sim \arcsin(\sqrt{\epsilon})$. (b) Same as for (a), but now for silicon, with a permittivity that displays typical features of a dielectric but also contains a metallic region.

At $\theta = 80^\circ$, a narrower spectral band is distinguished, falling off well below 1 eV. These data correspond to the time evolution that is observed at large angles, which does not exhibit clear oscillations but instead a strongly decaying signal.

Examining the frequency extent of the spectra more closely, they are most confined for the largest and smallest angles [Figs. 6(a) and 6(b)], where the fields are more delocalized. Correspondingly, the TR is more spread out in time. For angles around $\theta = 50$–70°, the fields extend over a much wider range of frequencies and are very localized in time, corresponding to stronger coupling between the electrons and the impact region. Combining the fields over all angles and determining the far-field component, as described in Eq. (24), allows one to calculate the emission intensity that can be measured experimentally. This approach has shown excellent agreement with multiple experiments on a variety of materials, including metals and semiconductors [44–46].

In addition to studying the spectra for given angles, we can determine the angular profile at different frequencies. This is experimentally accessible, for example, by using angle-resolved detection in combination with spectral filters. As for spectral measurements, good agreement with theory has been found for a variety of materials [46,47]. Figure 6(c) presents the normalized intensity as a function of θ for the frequencies corresponding to the vertical white dashed lines in Fig. 6(a), as well as the total intensity summed over all frequencies. We clearly observe the characteristic dipolar lobes, which vary in orientation and width for different frequencies. As expected, there are broader lobes at grazing angles for low frequencies and narrower lobes close to the surface normal at $\omega = \omega_p$. For higher frequencies, the distribution moves back to higher angles and becomes sharper.

Figure 7 examines the relation between TR emission and material permittivity by comparing the absolute value of ϵ to the total intensity (summed over all angles) and to the angle of maximum intensity $\theta_{\max}$, as a function of frequency/energy. The incoming medium is vacuum. We first study the Drude metal with $\hbar\omega_p = 1$ eV and $\hbar\eta = 0.05$ eV for a 30 keV electron [Fig. 7(a)]. For low frequencies, $|\epsilon_D|$ and the total intensity exhibit the same decreasing trend when approaching ω_p; the two then show opposite behavior in the frequency range where $|\epsilon_D| < 1$. $\theta_{\max}$, meanwhile, displays the same trend as ϵ_D across the whole frequency range, decreasing and increasing in lockstep. We examine the angle $\theta = \arcsin(\sqrt{\epsilon_D})$ for frequencies $\omega > \omega_p$, indicated by the gray dashed line. As discussed above, this should indicate the boundary between regions where the radiation is coupled in the backward (upper hemisphere) or forward (into the metal) directions. The resulting angle exhibits excellent agreement with $\theta_{\max}$, indicating that the sudden depletion of TR can indeed be attributed to coupling into the metal. This is dependent on the specific material permittivity; we confirm that there is good agreement for different parameters in Fig. S5 of the Supplemental Material [31].

In order to further explore these trends, we examine the same variables for silicon in Fig. 7(b), again using a 30 keV electron. In the $\hbar\omega = 0$–20 eV range, the permittivity ϵ_{Si} of silicon displays a strong double peak around 4 eV (~967 THz) and $\mathrm{Re}\{\epsilon_{\mathrm{Si}}\} < 0$ from 4 to 16 eV (~967–3869 THz). $|\epsilon_{\mathrm{Si}}|$ and $\theta_{\max}$ follow the same trend over the entire frequency range, while the total TR intensity again exhibits the same trend as $|\epsilon_{\mathrm{Si}}|$ for frequencies where $|\epsilon_{\mathrm{Si}}| > 1$ and an opposite trend for frequencies where $|\epsilon_{\mathrm{Si}}| < 1$. As for the Drude model, the minimum of $|\epsilon_{\mathrm{Si}}|$ and $\theta_{\max}$ corresponds to a peak in the total intensity.

Determining a direct relation between ϵ and all aspects of the TR emission is difficult because ϵ appears multiple times in the equations, but it is possible to get an intuitive understanding of the behavior. Looking back at the definition of TR, it is essentially a "reflected" field induced by the approaching electron that has to adapt to its new electromagnetic environment. The term μ_j from the equations even bears some resemblance to the Fresnel equations. Just as for reflection of light, one can understand that the higher the contrast between the two media, the more the field of the electron will have to "shake off" components and induce a strong response [48]. The ratio between the permittivities of the two materials can go both ways, however. What we observe in Fig. 7 is that for both $|\epsilon| \gg 1$ and $|\epsilon| \ll 1$, the TR intensity is high due to the large contrast from 1. The angle of maximum emission $\theta_{\max}$ consistently follows the same trend as $|\epsilon|$. In Figs. S6 and S7 of the Supplemental Material [31], we study this in more detail.

VI. CONCLUSIONS

We have determined that transition radiation and surface plasmon polaritons excited by swift electrons are composed of ultrashort, femtosecond time-scale wave packets. We have studied the time, space, and frequency dependence of the electric fields induced by the electron, both in vacuum and when traversing a metallic or dielectric surface, providing intuitive physical insight into these ultrafast processes. The external field of the swift electron in vacuum comprises a single oscillation, similar to a single optical cycle, and thus represents an ultrabroadband optical excitation spectrum. In vacuum, the fields evanescently decay away from the electron trajectory and cannot directly couple to radiation. When impinging on a polarizable material, however, the electron induces fields at the surface that can radiate out to the far field.

We first studied the surface plasmon polaritons propagating along the surface of a Drude metal, finding that the plasmon wave packets are several-hundred femtoseconds in duration (depending on the material parameters). The SPPs decay rapidly and redshift in frequency as they propagate away from the point of excitation. This redshift is due to the fact that high-frequency components close to the plasmon resonance are strongly damped.

We have also examined transition radiation, the far-field emission which occurs when a charged particle traverses the interface between two different media. Using the Drude metal as an example, we find that TR wave packets are strongly dependent on the emission angle and ultrashort in duration, lasting only a few tens of femtoseconds (again depending on the material parameters). The TR intensity and emission-angle dependence is correlated with well-defined trends in the permittivity of the material. Given the frequency dependence of ϵ for a certain dielectric, the TR emission can be qualitatively predicted.

The theoretical insights and predictions presented here further our knowledge and understanding of electron-light-matter interactions at the nanoscale and can be applied to study fundamental physical processes occurring at the femtosecond time scale.

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