Light generation via quantum interaction of electrons with periodic nanostructures

The MIT Faculty has made this article openly available. Please share how this access benefits you. Your story matters.

<table>
<thead>
<tr>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>As Published</td>
<td><a href="http://dx.doi.org/10.1103/PhysRevA.95.013832">http://dx.doi.org/10.1103/PhysRevA.95.013832</a></td>
</tr>
<tr>
<td>Publisher</td>
<td>American Physical Society</td>
</tr>
<tr>
<td>Version</td>
<td>Final published version</td>
</tr>
<tr>
<td>Citable link</td>
<td><a href="http://hdl.handle.net/1721.1/107954">http://hdl.handle.net/1721.1/107954</a></td>
</tr>
<tr>
<td>Terms of Use</td>
<td>Article is made available in accordance with the publisher’s policy and may be subject to US copyright law. Please refer to the publisher’s site for terms of use.</td>
</tr>
</tbody>
</table>
Light generation via quantum interaction of electrons with periodic nanostructures

Shai Tsesses,1 Guy Bartal,1 and Ido Kaminer1,2,*

1Andrew and Erna Viterbi Department of Electrical Engineering, Technion–Israel Institute of Technology, 32000 Haifa, Israel
2Department of Physics, Massachusetts Institute of Technology, 77 Massachusetts Avenue, Cambridge, Massachusetts 02139, USA

(Received 25 July 2016; published 23 January 2017)

The Smith-Purcell effect is a hallmark of light-matter interactions in periodic structures, resulting in light emission with distinct spectral and angular distribution. We find yet undiscovered effects in Smith-Purcell radiation that arise due to the quantum nature of light and matter, through an approach based on exact energy and momentum conservation. The effects include emission cutoff, convergence of emission orders, and a possible second photoemission process, appearing predominantly in structures with nanoscale periodicities (a few tens of nanometers or less), accessible by recent nanofabrication advances. We further present ways to manipulate the effects by varying the geometry or by accounting for a refractive index. Our derivation emphasizes the fundamental relation between Smith-Purcell radiation and Čerenkov radiation, and paves the way to alternative kinds of light sources wherein nonrelativistic electrons create Smith-Purcell radiation in nanoscale, on-chip devices. Finally, the path towards experimental realizations of these effects is discussed.

DOI: 10.1103/PhysRevA.95.013832

I. INTRODUCTION

Interaction of light with periodic media has fueled many scientific and technological advances over the last few decades. Bragg reflectors, photonic crystals, metallic gratings, and other periodic structures have significantly improved the creation and efficiency of light sources [1–3], light manipulation and shaping [4], the transfer and storage of optical information [5,6], and even particle acceleration [7–10]. Periodic photonic structures also provide fascinating mechanisms to observe fundamental light-matter interactions in man-made [11–13] or natural [14–16] materials. One such interaction, inherently stemming from a medium’s periodicity, is the Smith-Purcell effect.

The Smith-Purcell effect is a diffraction radiation effect, induced by a charged particle moving at a constant velocity near a periodic structure (a grating, for example) or through it (e.g., an array of slits), thereby emitting electromagnetic waves. The possibility of such an effect was first predicted by Frank [17], and was later observed by Smith and Purcell [18]. Constructive interference of waves diffracting from the periodic structure results in the wavelength of the radiation satisfying the following equation [19]:

$$\lambda = \frac{\alpha}{g} \left[ \frac{1}{\beta} - \cos(\theta) \right],$$  \hspace{1cm} (1)

where $\alpha$ is the periodicity of the surface; $g$ is the diffraction order (here, a negative integer); $\beta$ is the initial particle velocity, normalized to the light’s vacuum velocity; and $\theta$ is the angle at which the emitted light propagates relative to the initial direction of the electron’s motion, which is parallel to the grating periodicity. Note that the effect above and our work below can be directly extended to any charged particle, even though we refer explicitly to electrons.

We shall call this result the conventional Smith-Purcell effect (CSP), and note that it can be explained in many other ways: the diffraction of a charged particle self-field from a grating or other periodicity [20,21]; as a product of induced currents [22], or an oscillating dipole moment, created by a mirror charge [17]; as resonant diffraction radiation [23]; in the Green’s function formalism [24]; and in a quantum mechanical formalism [25]. All of these approaches match the findings of the Smith-Purcell experiment, as well as many experiments that followed (e.g., [26–30]), ever confirming the conventional theory represented by Eq. (1).

Here, we present a simple approach to the Smith-Purcell effect and reveal significant corrections to the conventional Smith-Purcell theory that arise due to the quantum nature of the radiation emission and the wave nature of the charged particle causing it. Our approach is based on exact conservation of energy and momentum during the interaction of the charged particle with the grating and with a photon. In certain regimes, the corrections shift the frequency spectra of the photon emission, and surprisingly, can even occur in visible wavelengths. Through the quantum corrections, we can predict a resonance and cutoff of the emitted wavelength at low electron kinetic energies; a convergence of all effect orders at high electron kinetic energies; and even show the possibility for another, yet undiscovered, photoemission process. We further lay out possible ways to control these effects using a more complex structure (such as a two-dimensional grating) or by introducing a material’s refractive index. Our results can shed light on understanding and using the Smith-Purcell effect in nanometric systems, and suggest exciting avenues to explore light-matter interactions in periodic media.

II. DERIVING QUANTUM CORRECTIONS

We derive the general form of the Smith-Purcell effect by assuming a grating is placed in vacuum, while an electron of a certain kinetic energy is moving in the close vicinity of the grating, in the direction of its periodicity (Fig. 1). Note that this picture is just for the purpose of illustration, and an equivalent effect appears for a particle moving through a hole in a periodically layered structure. We require conservation of energy and momentum in the process of a single-photon emission, and consider the electron to be relativistic...
(to keep the derivation general). Thus, these conditions correspond to three equations:

\[ E_f + \hbar \omega_c = E_i, \]  
(2)  
\[ \hbar q \cos (\theta) + E_f \beta_f \cos (\alpha) = E_i \beta_i + \hbar \omega, \]  
(3)  
\[ \hbar q \sin (\theta) + E_f \beta_f \sin (\alpha) = 0, \]  
(4)

where \( E_i, E_f \) are the initial and final energies of the electron; \( q \) is the emitted photon wave vector; \( c \) is the light’s vacuum velocity; \( 2\pi \hbar \) is Planck’s constant; \( \kappa \) is the grating vector, assumed for now to be parallel to the electron’s initial motion; \( \beta_i, \beta_f \) are the initial and final electron velocities, normalized to light’s vacuum velocity; and \( \alpha, \theta \) are the scattering angles of the electron and the photon, respectively, relative to the initial direction of the electron’s motion. Using simple algebra and the relation between momentum and energy in a relativistic regime, we come to a single equation:

\[
(E_i - \hbar \omega c)^2 = E_i^2 + h^2 c^2 (q^2 + \kappa^2) - 2 \hbar c [\hbar \omega c \cos (\theta) + E_i \beta_i q \cos (\theta) - \kappa].
\]  
(5)

From Eq. (5) the emitted photon’s momentum can be derived, and subsequently the emitted photon’s wavelength (note that this is an exact expression):

\[
\lambda = \frac{2\pi \hbar c \cos (\theta) - 2\pi \frac{1}{\kappa} \left[ \frac{1}{\kappa} - \cos (\theta) \right]}{\hbar c \kappa/\beta_i + 1} = \frac{\Delta DB \cos (\theta) - 2\pi \frac{1}{\kappa} \left[ \frac{1}{\kappa} - \cos (\theta) \right]}{\Delta DB \kappa/\beta_i + 1},
\]  
(6)

where \( \kappa = g[2\pi/a] \), \( a \) being the grating’s periodicity and \( g \) the diffraction order (here, a nonzero integer); \( \lambda = 2\pi/q \), the emitted photon’s wavelength; and \( \Delta DB = 2\pi \hbar c/E_i \beta_i \)—the de Broglie wavelength of the electron. Our description of the interaction in Eqs. (2)–(6) matches previous derivations [25,31], even some concerning other similar effects (like coherent bremsstrahlung [32]). However, all previous work neglected the implications of quantum corrections on the Smith-Purcell effect since the de Broglie wavelength was negligible relative to all other length scales (equivalent to neglecting the electron recoil, which removes all \( \hbar \) dependence). The implications of Eq. (6)—namely, the changes to the predicted wavelength due to the quantum corrections—are presented in Fig. 2 (with additional examples presented in Figs. 3 and 4).

It is noteworthy that electrons moving in close proximity to the grating and at low energies may start to resemble bound electrons that adhere to Bloch’s theory. Even then, our approach is still valid by substituting the electron dispersion relation (Bloch band structure) into \( E_i, E_f \) in Eqs. (2)–(4) [25] (such a procedure has been shown to work for the Čerenkov effect [33]). This treatment is true for any case where the electron dispersion relation is well defined and its motion is as depicted, while the same can be said for the photon dispersion relation. For example, Eqs. (2)–(4) can be changed to depict emission into confined photonic modes (such as surface plasmon polaritons, which exist at the interface of metallic gratings and air). These confined photonic modes can also significantly enhance the emitted intensity via their high density of states, as explained in [34].
Grating periodicity is the same as Fig. 2.

respectively. Smooth (dashed) lines represent the MSSP (CSP) effect.

first, second, and third negative diffraction orders (dark gray), orange (gray), and gold (light gray) lines represent the low-energy limit as a function of the electron kinetic energy. The blue (dark gray), orange (gray), and gold (light gray) lines represent the first, second, and third negative diffraction orders \((g = -1, -2, -3)\), respectively. Smooth (dashed) lines represent the MSSP (CSP) effect. Grating periodicity is the same as Fig. 2.

It is also important to note that Eqs. (2)–(6) apply for a grating wide and thin enough, in otherwise homogeneous space (so the grating is virtually two dimensional and infinite), such that the momentum is preserved in both directions transverse to the electron motion. However, they are valid even for the inhomogeneous case (i.e., different media on both sides of the grating). Effectively, the inhomogeneity will cause line broadening of the solutions of Eqs. (2)–(6), as the momentum perpendicular to the surface of the grating cannot be exactly preserved (as described by [34], and in the supplemental material of [33] for the Čerenkov effect). This indicates that, for emission parallel to the grating surface (as in Fig. 2), there is very little to no broadening.

In the classical limit \((h \to 0)\), as expected, Eq. (6) degenerates to the CSP effect [Eq. (1)]. This suggests an interesting physical interpretation to the CSP effect: In order to allow photon emission, the grating takes a part of the electron’s momentum, since the diffraction order is ever negative for any positive wavelength in Eq. (1). However, our description of the Smith-Purcell effect [Eq. (6)] suggests that the opposite may become possible as well, i.e., having the grating contribute momentum to the process. Generally, two separate phenomena become possible simultaneously: a momentum-subtraction Smith-Purcell effect (MSSP), analogous to the CSP, where the quantized grating moment contributions (diffraction orders) are negative \((g, \kappa < 0)\); and a momentum-addition Smith-Purcell effect (MASP), where they are positive \((g, \kappa > 0)\). Intriguingly, the latter (MASP) has no prior known analog in the CSP effect. However, it requires an additional mechanism to satisfy Eqs. (2)–(4), such as the material’s index of refraction influencing the momentum of the emitted photon [see Eq. (8) below].

Figure 2(a) indicates that, while for a wide range of electron energies the MSSP effect is indistinguishable from the CSP effect, at very low and very high initial electron kinetic energies we find significant corrections. In the low-energy limit, we find a substantial discrepancy between the MSSP and the CSP where the period becomes comparable to the de Broglie wavelength of the electron. There, we predict a resonant increase in the wavelength, followed by a cutoff effect. The cutoff occurs at higher energies for larger grating diffraction orders, and causes a significant change in the emitted radiation for low-energy electrons, as portrayed in Fig. 2(b). This change is also apparent in optical wavelengths, yet to a lesser extent (around 6% at the third diffraction order, as portrayed in Fig. 3).

This phenomenon, and the cutoff effect in particular, can be explained by energy conservation: The grating takes a quanta of the electron’s momentum, so its kinetic energy is, in turn, reduced by a corresponding amount. If the initial electron kinetic energy is small enough, losing the required momentum will cause the electron energy to be lower than its rest energy, preventing photon emission.

In the high-energy limit, the electron velocity becomes comparable to light’s vacuum velocity, and we can predict that the wavelength of emitted photons of all orders will converge to the electron’s de Broglie wavelength. This convergence, present at extremely short wavelengths, can only occur at very small angles, as seen in Fig. 2(c).

An easily apparent conclusion of Fig. 2(c) is that the emitted wavelengths in the high-energy regime are gamma rays, which means that no matter what material the grating is made of, it is completely transparent and thus Smith-Purcell radiation is not expected to occur. However, certain core-shell atomic transitions make specific atoms exhibit resonances at narrow bandwidths, displaying nontrivial permittivity for extremely short wavelengths. These might allow our predictions to be observed far beyond the visible spectrum, as with x-ray Čerenkov radiation [35]. Furthermore, a recent paper has presented measurements hinting to the speculative possibility of an index of refraction larger than 1 for gamma rays [36], which may allow the predictions in Fig. 2(c) to be observed for photon emission that falls at these specific gamma-ray spectral windows. Consequentially, to alleviate some experimental
limitations, it is crucial to understand how one can control the high- and low-energy regimes, and shift them to more convenient electron kinetic energies or photon wavelengths.

III. CONTROLLING THE QUANTUM CORRECTIONS

Our approach towards the Smith-Purcell effect predicts an even more complex scenario by using a two-dimensional (2D) grating—a condition akin to Smith-Purcell radiation achieved in 2D photonic crystals or plasmonic crystals [37]. Conventionally, it is expected that a periodicity perpendicular to the electron’s motion (grating lines parallel to that motion) would have no effect on the emitted photon spectrum [38], yet inserting this condition into the equations for momentum and energy conservation [Eqs. (2)–(4)], with an additional momentum conservation condition, yields a surprising result:

\[
\lambda = \frac{\Delta \text{DB}}{2} \left[ \cos(\theta) + \frac{\tilde{g}_1}{\tilde{a}_1} \sin(\theta, \phi) \right] - \frac{\tilde{a}_1}{\tilde{g}_1} \left[ 1 - \cos(\theta) \right] - \frac{\Delta \text{nP}}{2} \left[ 1 + \left( \frac{\tilde{g}_2}{\tilde{a}_2} \right)^2 \right] + 1,
\]

\[\text{Eq. (7)}\]

where \(\tilde{a}_2\) is the perpendicular periodicity and \(\tilde{g}_2\) is its corresponding diffraction order; \(\tilde{a}_1, \tilde{g}_1\) represent the parallel periodicity and diffraction order, and \(\theta, \phi\) are the polar and azimuthal scattering angles of the photon, relative to the initial direction of the electron’s motion. For the case \(g_2 = 0\) (no perpendicular periodicity) we return, as expected, to Eq. (6) (where \(g_1 = g\) and \(a_1 = a\)). Assuming \(g_1 = 0\) (only a perpendicular grating), the photon can never be emitted into free space due to energy conservation, in consistence with the CSP effect.

An intriguing feature of using a 2D grating, as can be seen in Fig. 5, seems to be changes to the MSSP effect’s resonance, and grants effective control over the low-energy regime. The resonance will occur at higher electron energies, similar to a regular grating with periodicity that is decreased by the sum of 1 and the squared ratio between the two periodicities [see Eq. (7)]. Moreover, it seems that in the low-energy regime noticeable effects occur in a wider range of wavelengths and for a wider range of electron energies, particularly at larger angles \((\theta, \phi \to 90^\circ)\). However, the perpendicular periodicity will have little influence in the high-energy regime—its effect is minimal in very small angles, and in larger angles it lacks the ability to compensate for the shortening of the de Broglie wavelength.

Finally, we generalize our approach towards the Smith-Purcell effect to cases where the photon is emitted into an environment with a nontrivial index of refraction \(n\). This occurs when the grating is covered by a dielectric; enveloped in a liquid; or even in the simple scenario described by Fig. 1, where the substrate is transparent and some of the photons are emitted through it instead of into free space. We solve Eqs. (2)–(4) for emission into an environment with an index \(n\) by replacing the term \(h\tilde{q}\) in Eqs. (3) and (4) by \(h\tilde{q}n\), while also considering an additional grating. The momentum of the emitted free-space photon is then generally given by the following formula:

\[
q = \frac{n}{n^2 - 1} \left( \frac{E_i \beta_i}{\hbar c} \left[ \cos(\theta) - \frac{1}{\beta_i n} \right] + \kappa \cos(\theta) + \xi \sin(\theta) \sin(\phi) \right) \pm \sqrt{\left( \frac{E_i \beta_i}{\hbar c} \left[ \cos(\theta) - \frac{1}{\beta_i n} \right] + \kappa \cos(\theta) + \xi \sin(\theta) \sin(\phi) \right)^2 - \left( \kappa^2 + \xi^2 + 2 \tilde{c} \left( \frac{E_i \beta_i}{\hbar c} \right) \frac{n^2 - 1}{n^2} \right)},
\]

\[\text{Eq. (8)}\]

where \(\xi = g_2[2\pi/\tilde{a}_2]\) represents the perpendicular grating vector, and we must remember that the angles \(\theta, \phi\) are now in the material as well, so when coupled to free space the propagating photon will obey Snell’s law.

Essentially, changing the refractive index will push the effects of the high-energy regime to lower electron velocities, allowing us to bring them to the optical spectrum (instead of gamma radiation). In this respect, the refractive index will also allow the MASP effect to propagate (possibly at visible wavelengths as well), since the free-space wavelength of the MASP will now be larger than the de Broglie wavelength of the electron. In the low-energy regime, increasing the refractive index will not change the critical velocity value for which the resonance and cutoff occur (as they are dependent on the electron’s de Broglie wavelength alone) but will push the emission spectrum towards smaller wavelengths. If we assume the grating vectors are zero \((\kappa = \xi = 0)\), then Eq. (8) degenerates to the known quantum Čerenkov effect [39–42].

An interesting connection between Smith-Purcell radiation and Čerenkov radiation arises from this equation. On the one hand, the quantum Čerenkov effect can be described as the Smith-Purcell zero-order effect in a medium; on the other hand, this analogy allows us to derive the Smith-Purcell effect from the quantum Čerenkov equation by inserting the grating as an effective refractive index. In this manner, many interesting traits explored in the field of Čerenkov radiation may apply to Smith-Purcell radiation, with examples such as the reversed Čerenkov cone in materials of negative refraction and the plasmonic Čerenkov effect in metasurfaces and 2D materials [34,43–45]. These suggest analogous Smith-Purcell effects that would occur when such modern nanophotonic structures are fabricated with periodic structures in or near them.

IV. MEASURING THE QUANTUM CORRECTIONS

Let us now address an important question: How is it that the quantum corrections to the CSP effect have never been observed, given the wide range of parameters showing these corrections, and the wide range of experiments observing Smith-Purcell-type effects? The immediate reason, which also highlights a requirement important to our prediction, is the use of gratings of very small periodicities, that have been, until recently, outside the reach of our fabrication tools. However, recent years have shown significant advances in the
FIG. 5. Momentum-subtraction Smith-Purcell effect with a 2D grating. Emission wavelength of the first parallel order \( (g_1 = -1) \), at an azimuthal angle of 90° and a polar angle of 0° (a) and 90° (b), as a function of the electron kinetic energy in the low-energy regime. Panel (c) portrays the emission wavelength of the first parallel order \( (g_1 = -1) \) as a function of the angle, for electron kinetic energy of 200 keV. The blue (dark gray), orange (gray), and gold (light gray) lines represent the first, second, and third perpendicular orders. The black line represents the CSP effect, which is virtually inseparable in the chosen parameters from the MSSP effect for \( g_2 \neq 0 \). Parallel periodicity is taken as \( a_1 = 5.16 \) μm for all graphs, while the perpendicular periodicity is at \( a_2 = 10 \) nm. The values of change from conventional results noted on the panels emphasize that the quantum correction in this regime significantly influences the emitted wavelength, observable by using standard electron sources and IR detectors.

resolution of nanofabrication techniques such as electron beam lithography and ion beam milling, that now reach features of a few nanometers and gratings with periodicities on the order of 10 nm (see e.g., [46,47]).

Another reason that the quantum corrections have been missed is evident by observing Fig. 2(a): The quantum features emerge at the two extremes of low and high energies, while most systems working with charged particles, such as electron microscopes and electron energy loss spectroscopy, electron lithography, most synchrotrons, and RF accelerator guns, use electron beams with energies that lie in the “dead region” in between the two regimes. This becomes even more apparent when noting that grating periodicities higher than 10 nm broaden this “dead region” even further. Therefore, there would be no noticeable change from the conventional theory unless one deliberately looks for it in nonconventional parameters.

Fortunately, a grating with such periodicity is well within reach of current nanofabrication techniques; thus observing the quantum corrections should be possible with existing setups. Let us present several alternatives (assuming 10-nm periodicity): the first setup requires a slow, directed electron beam (with a range of about 0.1–10 eV), together with detection capabilities in the visible or IR [as shown in Figs. 2(b) and 3]. In this respect, existing devices such as the high-resolution electron energy loss spectrometer (HREELS) or certain kinds of low-energy electron microscopes (LEEM) can be utilized as electron beam sources. Another alternative is using microwave guiding of electrons [48], a method that has shown very high quality control over electron trajectories in the relevant energy range.

We can also ease the requirements of the electron beam source by working with a 2D grating [Eq. (7)], which will push the low-energy regime to higher electron velocities. This way, it will be possible to witness the low-energy wavelength resonance even with modestly relativistic electrons—Figs. 5(c)
and 6 present examples with 200-keV electrons, easily available in transmission electron microscopes (TEMs). As another example, Fig. 5 shows how a parallel periodicity of 5.16 μm combined with a perpendicular periodicity of 10 nm will cause a spectral resonance and cutoff of the orders $g_1 = 1$, $g_2 = \pm 1$ to occur at electron kinetic energies of about 1 keV, which are achievable in many standard scanning electron microscopes (SEMs).

V. CONCLUSIONS AND DISCUSSION

In conclusion, by applying a simple approach to the known Smith-Purcell effect, we predict noticeable corrections that occur due to the quantized nature of the radiation and the wave nature of matter. Further investigation of these quantum corrections also revealed effects for different configurations (2D grating, emission into a material) that have not been, as of yet, predicted or measured by any prior work. These include an emission cutoff at low electron energies, a convergence of effect orders at high electron energies, and the possibility for an undiscovered photoemission process. The quantum corrections appear predominantly for a system with sub-100-nm periodicity, where nonrelativistic electrons suffice for the generation of Smith-Purcell radiation, bypassing the need for significant acceleration and vacuum chambers. The possibility of such slow electrons producing Smith-Purcell radiation suggests the use of this effect for light generation in nanoscale, on-chip applications.

Therefore, understanding the quantum effects is a pivotal step towards nanoscale devices using the Smith-Purcell effect for practical applications. Such applications benefit from the diverse and tunable spectrum the Smith-Purcell effect can potentially produce (including microwave and terahertz radiation [49]). Our results may also contribute to the general understanding of light-matter interactions in scenarios that are traditionally considered purely classical but now are shown to contain quantum effects. Finally, we have established a direct connection between the quantum Čerenkov effect and the Smith-Purcell effect, and there might yet be a way to tie even more similar effects into a single equation.


