Beyond Fermi’s golden rule in free-electron quantum electrodynamics: acceleration/radiation correspondence

Yiming Pan 1,2,∗ and Avraham Gover 3

1 Department of Physics of Complex Systems, Weizmann Institute of Science, Rehovot 76100, Israel
2 Physics Department and Solid State Institute, Technion, Haifa 32000, Israel
3 Department of Electrical Engineering Physical Electronics, Center for Light–Matter Interaction (LMI), Tel Aviv University, Ramat Aviv 69978, Israel

∗ Author to whom any correspondence should be addressed.
E-mail: yiming.pan@weizmann.ac.il and gover@eng.tau.ac.il

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Abstract

In this article, we present a unified reciprocal quantum electrodynamics (QED) formulation of free-electron and quantum–light interaction. For electron–light interactions, we bridge the underlying theories of photon-induced near-field electron microscopy, laser-induced particle accelerators, and radiation sources, such as quantum free electron laser, transition radiation and Smith–Purcell effect. We demonstrate an electron–photon spectral reciprocity relation between the electron energy loss/gain and the radiation spectra. This ‘acceleration/radiation correspondence’ (ARC) conserves the electron energy, and photon number exchanged, that is,

\[ \Delta E/\hbar \omega + \Delta \nu_q = 0, \]

and in the representation of a quantum electron wavepacket, displays explicit dependence on the history-dependent phase and shape of the wavepacket configuration. It originates from an interaction-induced quantum interference term that is usually ignored in Fermi’s golden rule analyses, but is kept in our combined quantum free electron–photon state formulation. We apply this formulation to both stimulated interaction and spontaneous emission of classical and quantum light by the quantum-featured electrons. The ‘spontaneous’ emissions of coherent states (‘classical’ light) are remarked and squeezed states of quantum light is shown to be enhanced with squeezing. This reciprocal QED formulation has promise for extensions to other fundamental research issues in quantum light and quantum matter interactions.

The particle-wave duality of electron and its interaction with light has been a matter of debate and interest since the early days of quantum theory [1–4]. The consistency bridge between the quantum-mechanical wave-like picture of electron–light interaction (e.g. [5–12] and the classical point-particle picture can be established by using a finite size quantum wavepacket presentation of the free electron [13–20]. In classical theory, the momentum transfer to a point-particle electron, propagating in direction-\( z \), due to interaction with a longitudinal classical electric field component \( E = Ez \cos(\omega t - qz + \phi_0) \) of frequency \( \omega \), wavenumber \( q_z \), and phase \( \phi_0 \), is

\[ \Delta p_{\text{point}} = -eEz \left[ \text{sinc} \left( \theta \right) \cos \left( \theta + \phi_0 \right) \right] . \]

(1)

with \( \theta = (\omega/\nu_0 - q_z) L \) defined as the synchronism detuning parameter, and \( L \) is the interaction length, and \( \nu_0 \) is the electron velocity. The classical picture of point-particle acceleration in a configuration of ‘dielectric laser accelerator’ (DLA) [21, 22], is shown in figures 1(a) and (b). However, the stimulated linear...
interaction of a coherent radiation wave with an electron in a similar configuration, results in a quite different result in the quantum mechanical description of the electron as a wavefunction [5–12].

Recent development in electron microscopy has brought a renewed interest in the quantum analysis of electron–light interaction. Of particular interest is the development of photon-induced near-field electron microscopy (PINEM) techniques in ultrafast transmission electron microscopy (UTEM) by Zewail and co-workers [7–9] and consequent recent experiments of multiphoton emission/absorption quantum sideband spectra and wavefunction density attosecond-bunching of free electron pulses [7–12, 23–30]. The underlying theory of PINEM-kind interactions is based on plane-wave-like quantum wavefunction modeling of single electrons. For the same configuration of the classical single electron interaction with light (figures 1(c) and (d)), this model results in a discrete electron energy spectrum of sidebands, spaced apart by the photon energy $h\omega$, having no net acceleration [8, 10]. This spectrum is radically different from the phase-dependent net acceleration energy spectrum of (1), and it cannot be explained in terms of point-particle trajectory theory. The results of this PINEM model are therefore very different from the standard formulations of classical free-electron interaction schemes with light, such as free electron laser (FEL) [31–35] and other radiation schemes [36–47] and also the recently developing classical schemes of DLA [21, 22, 28, 29].

The bridge between the single electron point-particle acceleration/deceleration model and the PINEM-kind interaction model of electron stimulated interaction with light, is the quantum electron wavepacket (QEW) representation, as shown in figure 1(e). Based on the solution of the time-dependent Schrödinger equation with a classical radiation field, it was shown in the framework of semiclassical theory [13], that the linear acceleration (1) of a point particle is modified in the limit of an electron wavepacket modeled by a Gaussian distribution of standard deviation width $\sigma_{z}$ to:

$$\Delta E_{\text{QEW}} = \Delta E_{\text{point}} e^{-\frac{z^2}{\sigma_{z}^2}},$$
where $\Gamma = 2\pi \sigma_z(t) / \beta \lambda$, $\beta = v_0/c$ is the QEW group velocity, and $\lambda$ is the wavelength of the field. Namely, the QEW acceleration is still phase-dependent like for a point-particle (1), but it is reduced relative to a point particle by a universal factor $e^{-\Gamma^2/2}$ (we will derive it explicitly in the following sections). Therefore, the classical point-particle limit of an accelerated electron wavepacket is [13, 14]:

$$\Gamma \ll 1 \quad \text{or} \quad \sigma_z(t) \ll \beta \lambda. \quad (2)$$

The wavepacket size $\sigma_z(t)$ depends on pre-interaction transport history ($t$). It can never be null (ideal ‘point-particle’, i.e., $\sigma_z(t) = 0$), because of the Heisenberg uncertainty principle, but in the limit (2), the QEW interacts with light, similar to a charged point particle (1). In the opposite limit ($\Gamma \gg 1$), the QEW acceleration is diminished, and the wavepacket-dependent theory predicts the PINEM spectrum with perfectly symmetric spectral sidebands. As a result, this satisfactorily bridges the two regimes of point-particle and plane-wave interaction pictures in the case of stimulated radiative interaction. Such transition was demonstrated earlier by Talebi in a semiclassical model by numerical solution of Schrodinger point-particle and plane-wave interaction pictures in the case of stimulated radiative interaction. Several earlier publications took into consideration the case of radiative interaction by a quantum wavepacket [e.g.15–17, 19, 20, 53], but and therefore, they could not produce the point-particle acceleration limit. Several earlier publications took into consideration the case of radiative interaction by a quantum wavepacket [e.g. 15–17, 19, 20, 53], but did not investigate the transition of QED theory of stimulated radiative interaction of QEWs into the classical point-particle acceleration/deceleration limit. The spontaneous and stimulated photon emission/absorption rates were derived there from first-order perturbation theory, using Fermi golden rule (FGR) [6, 15, 16]. In this model, the net photon emission rate of a radiation mode ($q, \nu$) at frequency $\omega$, is the difference of the single-photon emission and absorption rates: $d\nu_q(t)/dt = W_{q \nu = \nu + 1} - W_{q \nu = \nu - 1}$. For a large class of free-electron interaction schemes (e.g., FEL, Smith–Purcell, Cerenkov, and transition radiations) [6, 43–46, 49–52, 54–57], as depicted in figures 1(c) and (d), the FGR–QED model general expression for spontaneous/stimulated photon emission is given by [5]:

$$\Delta \nu_q = \Gamma_{sp} \left( \left( \nu_q^{(0)} + 1 \right) F_a(\omega) - \nu_q^{(0)} F_a(\omega) \right), \quad (3)$$

where the spontaneous emission coefficient $\Gamma_{sp}$ and emission/absorption spectral line functions $F_{em} (\omega)$ depend on the specific light-electron interaction configuration (see appendix C). Here, the index of polarization ($\sigma$) is suppressed. The terms with the coefficients of initial photon number $\nu_q^{(0)} \neq 0$ describe the stimulated emission, and the spontaneous emission is described by the term left when $\nu_q^{(0)} = 0$. This expression is manifestly independent of the electron phase and shape of the QEW in both spontaneous and stimulated parts, in contrast to the classical and semiclassical expressions (1), (2). Since the FGR analysis that leads to (3) does not include the electron wavepacket dynamics, it cannot reduce to the point particle acceleration limit [13, 63], and cannot bridge the QED and classical theories of electron–light interaction.

Only if one considers the electron–light interaction in a model of a QEW, and carries out a self-consistent QED analysis [14] or semiclassical analysis [35] beyond the FGR approximation, the resulting expressions for stimulated radiation emission, reduce to the classical limit of point-particle radiation. Here we review these results, and derive, in a self-consistent QED formulation, the expressions for both radiation emission and electron acceleration, as shown in figure 1(e). We show that the expressions for the post-interaction electron energy spectrum of a QEW reduce to the point particle acceleration in the limit (2), and to the PINEM-kind sidebands spectrum in the opposite limit. Furthermore, based on the quantum analysis of electron deceleration/acceleration and photon emission/absorption, we derive and prove in the following comprehensive analysis, a universal electron–photon spectral reciprocity relation of acceleration-radiation correspondence (ARC):

$$\Delta \nu_q + \Delta E / \hbar \omega = 0. \quad (4)$$

This is the fundamental energy-conserving relation between the exchanged photon number ($\Delta \nu_q$) of the radiation field of a single monochromatic mode ($\omega, q$) and the particle energy transfer (gain/loss, $\Delta E$) of the interacting single electron wavefunction (figure 1(e)).

The analysis in this work and the fundamental electron–light interaction relations are general in the sense that they are applicable to a wide variety of electron–photon interaction schemes. For instance, the analysis model is of extended interaction of an electron with a radiation wave in a slow-wave structure, such as Smith–Purcell interaction [50, 56, 57] or Cerenkov interaction [43], including short interaction schemes like transition radiation [44–46]. However, the conclusions would apply also to other kinds of interaction.
schemes that are based on pondermotive interaction, such as FEL and Compton scattering [5, 6], where essentially the axial electric field $E_z$ in (1) and in (3) (implicit in $\Gamma_{\nu p}$) should be replaced by the pondermotive potential of the interaction [5, 33]. However, this observation is asserted only within the limits of a one-dimensional (1D) interaction model.

The outline of this manuscript is as follows. Based on QEW representation of the electron, we solve Schrödinger equation in first-order perturbation theory for the combined electron and photon states. Going beyond the FGR formulation [5, 6] (that describes only the photon emission part (3)), we first present explicit expressions for spontaneous and stimulated radiative emission and post-interaction energy spectrum of a single QEW interacting with a coherent (Glauber) state of the radiation field. These reduce respectively both to the classical electrodynamics 'point-particle' limit (1) and to the conventional QED 'plane-wave' limit (3). We then further extend this robust QED analysis of wavepacket-size-dependent electron–light interaction to cases of electron wavefunction interaction with quantum light sources such as Fock states, coherent states, and squeezed states [64–69], and further on, to cases of entanglement between photons (or optical excitations) and electrons [70, 71].

The ARC relation beyond FGR. Let us consider the light–matter interaction of a single electron wavepacket in a general interaction scheme, as shown in figure 1(e). Single free-electron wavefunction emission is attainable and well-controlled in electron microscopy (TEM or UTEM) [72–75], and its interaction with light can be presented in terms of well-defined photon states. The initial combined electron–photon state can be presented as a superposition of combined electron–photon states in terms of explicit expressions for spontaneous and stimulated radiative emission and post-interaction energy beyond the FGR formulation [5, 6] (that describes only the photon emission part (3)), we first present

\[ |i\rangle = \sum_{p, \nu} c_{p, \nu}^{(0)}(t) |p, \nu\rangle, \tag{5} \]

where $c_{p, \nu}^{(0)}(t)$ is the initial component of the combined electron and photon state basis $|p, \nu = |p\otimes |\nu\rangle$. In our quantum electron–photon state formulation, the entire electron–photon wavefunction transforms under interaction, exchanging energy and momentum by processes of photon emission/absorption and corresponding electron recoils. This pertains to any of the free electron and light interaction schemes discussed previously. The final electron–photon state after interaction is also formally given by

\[ |f\rangle = \sum_{p, \nu} c_{p, \nu}^{(f)}(t) |p, \nu\rangle. \tag{6} \]

The light–electron coupling redistributes the components of the electron–photon state from $c_{p, \nu}^{(0)}$ to $c_{p, \nu}^{(f)}$, within the same representation of electron–photon basis. For this purpose, we describe the coupling of an electromagnetic field with the electron, using the Schrödinger equation or a 'modified' Schrödinger equation (derived from Klein–Gordon (KG) equation, suppressed the spin index of the electron, see appendix B) and the interaction Hamiltonian $H_{\text{int}} = -e \left( \hat{A} \cdot \hat{p} \right) / 2\gamma m + \hat{A}^2 / 2\gamma m$ [14, 44, 47, 50, 76], with

\[ \gamma = 1 / \sqrt{1 - \beta^2} \]

the Lorentz factor. $\hat{A}$ is the quantized field operator of the classical field $A_0$ of a single radiation mode $q_i$ quantized in a general quantization cavity in free space or in a slow wave structure [14, 44]. Note that the first term $(\hat{A} \cdot \hat{p})$ is applicable to 'slow-wave' light–matter interaction schemes, like Smith–Purcell, Cherenkov, and transition radiations. The second term in the Hamiltonian describes radiative schemes like FEL and Kapitza–Dirac effect [33, 77, 78], in which a longitudinal force interaction is exerted through the pondermotive potential term ($\hat{A}^2$) [5, 27, 28, 77, 78].

Via second quantization of the vector potential in terms of photon creation and annihilation operators $A = A(\hat{a}_p, \hat{a}_\nu)$, we can unitarily split the interaction Hamiltonian into two parts:

$H_{\text{int}} = H^{(e)}_{\text{int}} + H^{(a)}_{\text{int}}$, in which $H^{(e)}_{\text{int}}$ correspond to the reciprocal photon emission $(e)$ and absorption $(a)$ respectively. In the weak-coupling perturbation approximation to the order of single photon process, we describe these processes in terms of reciprocal scattering ladders representing the electron acceleration and photon radiation, as depicted in figure 2.

In a very general universal description of free-electron interaction schemes with quantum light, these diagrams manifest the ARC (4). Explicitly, the scattering diagram of figure 2(a) indicates, that by emitting a photon, the final coefficient of state $|p, \nu + 1\rangle$ is given by $c_{p, \nu+1}^{(f)} = c_{p, \nu+1}^{(0)} + c_{p, \nu+1}^{(1)(e)}$, where $c_{p, \nu+1}^{(0)}$ is the initial coefficient of the electron in momentum state $p$ and the radiation in Fock state $\nu + 1$, and $c_{p, \nu+1}^{(1)(e)}$ represents a reciprocal electron momentum (or energy) conserving process of photon emission and momentum backward recoil: $|p, \nu \rightarrow |p - \delta p, \nu + 1\rangle$ with quantized momentum spacing $\delta p = h\omega / 2\nu$. Likewise, by absorbing a photon, the final coefficient of state $|p, \nu - 1\rangle$ is given by $c_{p, \nu-1}^{(f)} = c_{p, \nu-1}^{(0)} + c_{p, \nu-1}^{(1)(a)}$, corresponding
to the absorption of a photon and an electron momentum forward recoil: $|p, \nu \rangle \rightarrow |p - \delta p, \nu + 1 \rangle$.

Therefore, describing the physical consequence of the stimulated photon emission as the imbalance between contributions of emission and absorption processes results in (figure 2(a)):

$$\Delta \nu_q = \sum_{p, \nu} \left| \nu_{p, \nu}^{(0)} + \nu_{p, \nu}^{(1)(e)} \right|^2 - \left| \nu_{p, \nu}^{(0)} + \nu_{p, \nu}^{(1)(a)} \right|^2.$$  \hspace{1cm} (7)

The first term counts the final emitted photon number by integrating over any electronic and photon state which might participate in the interaction, while the second term counts the absorbed photon number. The scattered components $\nu_{p, \nu}^{(1)(e)}$ represent the reciprocal momentum conserving processes through emission or absorption of a single-photon and momentum recoils, relating to the incoming component $\nu_{p, \nu}^{(0)}$, and also the specific scattering matrix elements $\left\langle \nu | H^{|e\nu | f} | \nu \right\rangle$.

On the other hand, minding the final electron spectral energy (momentum) transfer, also as shown in figure 2(a), the particle acceleration of electron wavefunction in the single-photon emission and absorption processes is given by

$$\Delta E = \sum_{p, \nu} \left| C_{p, \nu}^{(0)} + C_{p, \nu-1}^{(1)(e)} + C_{p, \nu+1}^{(1)(a)} \right|^2 \left( E_p - E_0 \right),$$  \hspace{1cm} (8)

where

$$E_p = \hbar c \sqrt{p^2 + m^2 c^2} = E_0 + \nu_0 (p - p_0) + \frac{(p - p_0)^2}{2m^*},$$

is the electron energy–momentum dispersion relation expanded to second order, where the longitudinal effective mass $m^* = \gamma^3 m$, and $E_0 = \sum_{p, \nu} | \nu_{p, \nu}^{(0)} |^2$ is the initial electron energy and $p_0$ is the average momentum of the QEW [13, 14]. Note that the final component $C_{p, \nu}^{(f)} = C_{p, \nu}^{(0)} + C_{p, \nu-1}^{(1)(e)} + C_{p, \nu+1}^{(1)(a)}$ is not normalized, because of the additional photon-emitted ($e$) and photon-absorbed ($a$) contributions in terms of the photon-scattering redistributions $|p, \nu \rangle \rightarrow |p + \delta p, \nu \rangle$. Thus, we normalized the energy transfer (8) by the corresponding term $\sum_{p, \nu} | \nu_{p, \nu}^{(f)} |^2 E_0$.

To prove the general correspondence of radiation and acceleration (ARC) (4), we expand the expressions for the photon emission and electron acceleration $\Delta \nu_q$, $\Delta E$, respectively, cancel the initial terms, and rewrite separately the phase-dependent interference terms of the squared binomials $\Delta \nu_q^{(1)}$, $\Delta E^{(1)}$ and the

![Figure 2](image-url)
phase-independent scattering terms \( \Delta \nu_q^{(2)}, \Delta E^{(2)} \):

\[
\Delta \nu_q^{(1)} = 2 \sum_{p, p'} \Re \left\{ \left( \epsilon_{p, p'}^{(1)(e)} \right)^* \left( \epsilon_{p, p'}^{(1)(a)} \right) \right\},
\]

\[
\Delta E^{(1)} = 2 \sum_{p, p'} \Re \left\{ \left( \epsilon_{p, p'}^{(1)(e)} \epsilon_{p, p'}^{(1)(a)} + \epsilon_{p, p'}^{(1)(a)} \right) (E_p - E_0) \right\},
\]

and

\[
\Delta \nu_q^{(2)} = \sum_{p, p'} \left| \epsilon_{p, p'}^{(1)(e)} \right|^2 - | \epsilon_{p, p'}^{(1)(a)} |^2,
\]

\[
\Delta E^{(2)} = \sum_{p, p'} \left| \epsilon_{p, p'}^{(1)(e)} \epsilon_{p, p'}^{(1)(a)} + \epsilon_{p, p'}^{(1)(a)} \right|^2 (E_p - E_0).
\]

We expand the electron energy dispersion relation in (9) and (10) only to first order:

\[
E_p = E_0 + v_0 (p - p_0).
\]

The second order expansion has a negligible effect, and also falls off in the integration over \( p \) because the integrand is nearly antisymmetric. One substitutes separately in the summations (9), (10) \( E_p - E_0 = \hbar \omega \) for the absorption terms and \( E_p - E_0 = -\hbar \omega \) for the emission terms (neglecting here the small quantum correction to the recoils). This substitution reverses the signs between the terms in the expressions \( \Delta E^{(1,2)} \). Shifting the dummy index \( \nu \) in the summations to \( \nu \pm 1 \) and neglecting the emission–absorption mixing term in \( \Delta E^{(2)} \) (see appendix D), these elementary algebraic steps can result in the ARC relation for each order

\[
\Delta \nu_q^{(1,2)} + \Delta E^{(1,2)} / \hbar \omega = 0,
\]

and their sum establishes the general reciprocity relation (4) which is valid independently of any assumption on the wavepacket distribution or the nature of the light.

We call the emission/absorption and acceleration/deceleration terms (9) ‘phase-dependent terms’. They are dependent on the phase of the radiation light relative to the electron wavepacket (figure 1(e)). Likewise, we call the second order terms in (1) ‘phase-independent terms’ because they are proportional to the squared absolute value of the radiation field. In conventional QED analysis, the photon emission rate is calculated in perturbation theory based on FGR [5, 6, 15–17, 19, 20] (see appendix C), keeping only the phase-independent terms (10) and neglecting the mixture terms (9), thus missing the interesting physics of interference between overlapping initial and scattered terms. Keeping the contribution of this interference, beyond the FGR model, is a pivotal methodological step in our formulation in the present article, and also an inevitable necessity for bridging the quantum wavefunction and classical point-particle descriptions of the electron, which is the essence of wave-particle duality [13, 14, 63, 79].

**Electron–photon interaction in a slow-wave structure.** The formulation, so far, is valid for the broad class of electron–photon interaction schemes reviewed in [5, 33], including schemes of extended interaction length like Smith–Purcell interaction through a grating, dielectric waveguide, and Cerenkov radiation, but also schemes of ‘transition radiation’ with a short interaction length, such as a tip, nanowire or a foil (figure 1). They only differ in the derivation of the interaction matrix elements. Following [14], we exemplify here the explicit derivation for interaction schemes, in which a slow-wave axial field component of a radiation mode \( (\omega, q_z) \) interacts synchronously with a co-propagating electron wavefunction of velocity \( v_0 \) along an extended interaction length \( \omega / q_z = v_0 \). With neglect of the ponderomotive potential term \( A^2 \) and scalar potential term \( \phi \) [18], the interaction Hamiltonian in minimal coupling is taken to be

\[
H_I (t) = -\frac{e}{2\gamma m} \left( \vec{A} \cdot (-ih \nabla) + (-ih \nabla) \cdot \vec{A} \right).
\]

For the case of our concern, \( \vec{A} = -\frac{i}{2\hbar c} \left( \vec{E} (r) e^{-i\omega t} - \vec{E}^\dagger (r) e^{-i\omega t} \right) \), where \( \vec{A}, \vec{E} \) are vector potential and electrical field operators, respectively. In our one-dimensional analysis, we assume for simplicity that the light–electron coupling takes place through an axial slow-wave field component of one of the modes \( q \) of a quantized field radiation mode expansion [44]: \( \vec{E} (r) = \sum_q \vec{E}_q e^{i\omega z - i q_z z} \), where \( \vec{a}_q \) is the annihilation (creation) operator of photon number state \( \nu \). For the example of Cerenkov radiation [40, 43], \( E_{zq} = E_{zq} \sin \Theta \) and \( q_z = n (\omega) (\sin \Theta) \cos \Theta \), where \( \Theta \) is the wave propagation angle relative to the electron propagation axis \( z \), \( E_{zq} \) is the transverse component of the wave, and \( n(\omega) \) is the index of refraction of the medium. For a Smith–Purcell setup, the near-field is the axial component of one of the space-harmonics of a Floquet-mode radiation wave incident on a grating [35]: \( E_q (r) = \sum_m \vec{E}_{qem} e^{i\omega m z} \), such that one of the space-harmonics, with \( q_z = q_{zm} \) satisfies near synchronism condition selectively: \( \omega / q_z = v_0 \) [14]. \( E_q (r) \) is a classical radiation field solution of a radiation-quantization
cavity [5, 14, 44] that incorporates a conducting surface grating at its boundary. The grating is assumed to be long enough with multiple periods to allow the approximation of a Floquet mode.

We note, in passing, that the formulation of our problem here is different from the formulation of cavity quantum electrodynamics [80, 81]. The radiation modes in most many problems of free electron interaction with light are free space quantized plasmonic and photonic modes [82], but they also can be applied also to cavity modes (e.g. interaction with whispering-gallery modes confined in a concave surface or a ring resonator [83]). However, in our case, the radiation mode couples to a free electron of continuous motional degrees of freedom.

First-order time-dependent perturbation solution of Schrödinger equation with the interaction Hamiltonian (12) results in a first order differential equation for the coefficients of the combined quantum states (6): \( i \hbar \dot{\psi}^{(1)}_{p', \nu'} = \int \frac{dp}{2\pi v_0} \sum_{\theta} \psi_\theta^{(0)} (p', \nu') |H_\theta (t)| \psi_\theta (p, \nu) e^{i \left( \nu_\theta - \nu \right) / \hbar} \). By integrating in time from minus infinity to infinity (see appendix C) [14], the emission and absorption terms of the perturbation coefficients \( \psi^{(1)}_{p', \nu'} = \psi^{(1)(e)}_{p', \nu'} + \psi^{(1)(a)}_{p', \nu'} \) are given by

\[
\psi^{(1)(e)}_{p', \nu'} = \left( \frac{\nu' - \nu_\theta \pm \hbar q_{zm}}{p_0} \right) \frac{\tilde{T} \sqrt{\nu' + 1} e^{(0)}_{\nu'} \nu'_{\nu' - 1} \nu'_{\nu' + 1} \sin \left( \frac{\phi_{\nu'_{\nu' + 1}}}{2} \right) e^{\left( \phi_{\nu'_{\nu' - 1}} / 2 \right)}}{\sin \left( \frac{\phi_{\nu'_{\nu' - 1}}}{2} \right)}.
\]

Here we defined the normalized photon exchange coefficient \( \tilde{T} = \frac{E_{\text{gen}} L}{4 \hbar \omega} \), and the emission/absorption quantum recoiled detuning parameters \( \tilde{\phi}_{\nu_{\nu'_{\nu' - 1}}} = \left( \nu_{\nu'_{\nu' - 1}} \pm \hbar q_{zm} \right) / L = \tilde{\phi}_m \pm \epsilon / 2 \), where \( \tilde{\phi}_m = \left( \tilde{\phi}_L - q_{zm} \right) L \) is the classical interaction ‘detuning parameter’ with \( \epsilon = \delta (\omega / v_0) L \) is the interaction quantum recoil parameter with \( \delta = \hbar / 2 m^* v_0^2 \) [5, 13]. From here on, we suppress the index \( m \), considering synchronism with a single Floquet space-harmonic or Cherenkov radiation mode.

The photon emission and electron energy transfer of ARC (4) can be calculated for a Fock state initial condition by just substituting \( \psi^{(0)}_{p', \nu'} = \psi^{(0)}_{\nu} \delta_{\nu_\theta} \) in (11), and using it in equations (9) and (10) for the photon and electron energy increments. With the plausible approximations \( \hbar \omega / E_0 \ll 1, \hbar q_{zm} / p_0 \ll 1 \), this results in [14] for the phase-independent term (see also equation (C8) in appendix C):

\[
\Delta \nu_{q_{\nu_{\nu'_{\nu' + 1}}}} = \tilde{T}^2 \cos \left( \frac{\phi_{\nu_{\nu'_{\nu' + 1}}}}{2} \right),
\]

where \( \Delta \nu_{q_{\nu_{\nu'_{\nu' + 1}}}} = \Delta \nu_{q_{\nu_{\nu'_{\nu' - 1}}}} = \Gamma_{\nu_{\nu'_{\nu' + 1}}} = \Gamma_{\nu_{\nu'_{\nu' - 1}}} = \left( \frac{E_{\text{gen}} L}{4 \hbar \omega} \right)^2 \). However, the phase-dependent terms in (9) vanish in this case: \( \Delta \nu_{q_{\nu_{\nu'_{\nu' + 1}}}} = \Delta \nu_{q_{\nu_{\nu'_{\nu' - 1}}}} = 0 \). Namely, there is no phase-dependent contribution of a single Fock state \( \xi_{\nu_0} \), neither for \( \nu_0 \neq 0 \), nor for \( \nu_0 = 0 \). A consequent significant conclusion is that there is no phase-dependent spontaneous emission in vacuum from a single electron wavepacket, and the only QED contribution to spontaneous emission is phase-independent resulting from the ‘zero-point vibration’ [5, 14], see figure 2(b):

\[
\Delta \nu_{q_{\nu_{\nu'_{\nu' + 1}}}} = \tilde{T}^2 \cos \left( \frac{\phi_{\nu_{\nu'_{\nu' + 1}}}}{2} \right).
\]

These results, valid for any electron wavepacket distribution, are identical with the stimulated and spontaneous emission expressions derived before only for a plane-wave intrinsic quantum wavefunction model of the electron (3) [5, 6]. Thus, even though we start with an electron wavepacket formulation, the Fock state analysis of light cannot provide the transition to the classical point particle picture (1) of electron–light interaction.

**Interaction of QEW with coherent state light and its quantum to classical transition.** Consider the case of an initial state of electron wavepacket, modeled as a chirped Gaussian distribution combined with a coherent photon state \( \xi^{(0)}_{\nu_0} \)

\[
\psi^{(0)}_{p_0, \nu_0} = \psi^{(0)}_{\nu_0} \xi^{(0)}_{\nu_0},
\]

and

\[
\psi^{(0)}_{\nu_0} = \left( 2 \pi \sigma_{\nu_0}^2 \right)^{-1} \exp \left( - \frac{(p - p_{\nu_0})^2}{4 \sigma_{\nu_0}^2} \right) e^{i \phi_{\nu_0}} e^{i \phi_{\nu_0}},
\]

where \( \sigma_{\nu_0}^2 (t_0) = \sigma_{\nu_0}^2 (1 + i \eta t_0)^{-1} \), \( \eta = \frac{\omega^2}{c^2} \) is an intrinsic chirp parameter of the QEW. This chirp effect of the electron wavefunction, accompanied with ‘history dependent’ wavepacket size expansion, develops in
free space due to the nonlinearity of the electron energy dispersion equation as derived in [13, 14, 23] by second-order expansion of the dispersion equation in terms of \((p - p_0)\) (equation after (8)). Thus, this general complex Gaussian wavepacket represents a wavepacket that evolves from its waist point at time \(t_0\) and distance \(L_0 = v_0t_0\) before the interaction region. In the interaction region, further time-dependent chirp and expansion of the QEW is neglected in first order perturbation analysis. \(v_0\) is the expectation value of the photon number of mode \(q\). Substituting (16), (17), (13) into the formulae (9), (10), one can calculate the electron acceleration and photon radiation terms (11) independently, and obtain after some detailed derivation steps (see appendix D), the explicit ARC relation for coherent light and Gaussian wavepacket.

\[
\Delta \nu_{q}^{(1)} = -\Delta E^{(1)}/\hbar \omega = \left(\frac{\epsilon E_d L}{\hbar \omega}\right) e^{-\xi^2/2} \frac{1}{\sqrt{2}} \frac{\cos \left(\frac{\pi}{2} + \phi_0\right)}{\sin \phi_0},
\]

\[
\Delta \nu_{q}^{(2)} = -\Delta E^{(2)}/\hbar \omega = \hat{T}^2 \left\{ (v_0 + 1) \sin^2 \left(\frac{\theta_0}{2}\right) - v_0 \sin^2 \left(\frac{\theta_0}{2}\right) \right\}.
\]

Note that \(v_0\) in this equation is the photon number expectation value of the coherent state (17) replacing the Fock state number in (14). Here we used the following relations of a coherent state:

\[
\langle \sqrt{v_0} | \hat{a}_q \rangle | \sqrt{v_0} \rangle = \sqrt{v_0} \langle \sqrt{v_0} | \hat{a}_q^\dagger \hat{a}_q \rangle | \sqrt{v_0} \rangle = v_0,
\]

\[
\langle \sqrt{v_0} | \hat{a}_q \hat{a}_q^\dagger | \sqrt{v_0} \rangle = v_0 + 1.
\]

In (18), \(E_d = \sqrt{v_0} E_{sym}\) is the axial slow wave field component of the incident single mode interacting radiation wave.

The phase-dependent interference term (18), results in the semiclassical expression for stimulated interaction with a QEW [13, 14], including the significant extinction coefficient exp \((-\Gamma^2/2)\), where

\[
\Gamma = \left(\frac{\omega}{v_0}\right) \frac{\sigma_z(t_0)}{\hbar \omega} = \left(\frac{\hbar \omega}{v_0}\right) \frac{\sqrt{1 + \xi^2 t_0^2}}{2 \sigma_{pi}} = \Gamma_0 \sqrt{1 + \xi^2 t_0^2},
\]

with \(\sigma_{pi} = 2 \pi \sigma_{\alpha} / \beta \lambda\). This ‘beyond FGR’ relation for wavepacket-dependent acceleration (18a) results out of the commonly neglected interference terms (9) after integration with the center-shifted Gaussian distributions (17a) (see appendix D). It confirms the ARC relation (4), and demonstrates the transition from the quantum electron plane-wave limit \(\Gamma \gg 1\), where only phase-independent emission/acceleration exists, to the point-particle picture of classical acceleration (1) in the opposite limit (2). Moreover, it also demonstrates the emission/acceleration dependence on the history-dependent wavepacket size (20) in the quantum to classical transition regime.

It is interesting to examine also the phase-independent component (18b). In the limit of small interaction recoil parameter \((\epsilon \ll 1)\), the phase-independent ARC expression (19b) reduces to the famous low-gain expression of FEL [5, 13, 26, 27]; \(\Delta \nu_{q}^{(2)} = \hat{T}^2 \left\{ \sin^2 \left(\frac{\theta_0}{2}\right) + v_0 \frac{\epsilon}{\hbar \omega} \sin^2 \left(\frac{\pi}{2}\right) \right\}\right\}\). The ever-existing spontaneous emission contribution (15) of the zero-point quantum vibration of Fock state \(\nu_0 = 0\) is kept in (18b), and phase-independent second order emission–absorption and acceleration–deceleration are still possible in second order at nonzero detuning condition.

In practice, confirmation of the ARC relation and observation of the predicted radiation phase and wavepacket-size dependent term (18a), would be hampered by the phase-independent term (18b). Even in the case \(\theta = 0\), when the second-order stimulated emission part vanishes, the spontaneous emission part (15) ever persists. Thus, it acts as background quantum noise, such that the detection of the phase-dependent term requires exceeding a signal to noise limit condition

\[
\frac{S}{N} \equiv \frac{\Delta \nu_{q}^{(1)}}{\Delta \nu_{q,vac}} \biggr|_{\text{max}} = \frac{4\sqrt{\nu_0}}{\sqrt{\Gamma}}
\]

Thus, the signal to noise condition \((S/N > 1)\) for the detection of the phase-dependent signal requires sufficiently intense incident laser pulse \(v_0 > \hat{T}^2/16\). The explicit determination of the quantum spontaneous emission per mode depends on the configuration of the radiative interaction scheme. In a model of a radiation plane wave incident on a slow-wave structure of length \(L\) (a Cerenkov medium or a Smith–Purcell structure), one may quantize the traveling wave radiation mode by the single-mode energy quantization requirement \([\sqrt{\nu_0/\hbar \omega} \hat{E}_{q,L}^2]) \approx A_{eff} \Delta t/2 = \hbar \omega\), where \(\Delta t = L/v_0 = L/\beta c\) is the interaction time along the structure and \(A_{eff}\) is the effective cross-section of a diffraction-limited incident radiation mode.

Our result for the expression of wavepacket-independent spontaneous emission per mode in Smith–Purcell radiation, is consistent with independent semiclassical and QED derivations [5, 62, 76]. To
be specific, in an open grating structure (see figure 1), the general spectral radiant energy \( W \) per unit solid angle d\( \Omega \) per unit interval frequency d\( \omega \) is given by [5] \( \left( \frac{d^2 W}{d\Omega d\omega} \right)_{SP} \approx \frac{\hbar}{\omega} \rho(\omega) \Delta \nu_{\text{vac}}, \) where

\[
\Delta \nu_{\text{vac}} = \tilde{T} \sin^2 \left( \frac{\pi}{2} \frac{2}{\Gamma} \right), \quad \tilde{T} = \tilde{E}_q L / 4 \hbar \omega, \]

the single-mode spontaneous photon emission per mode is recast to the known expression for Smith–Purcell spontaneous photon energy emission per steradian and unit frequency by a single electron [5, 14]:

\[
\left( \frac{d^2 W}{d\Omega d\omega} \right)_{SP} = \frac{e^2 L^2}{64 \pi^2 c^2} \frac{\tilde{E}_q^2}{\epsilon_0} \left| \frac{\mu_0}{\epsilon_0} \right| |\tilde{E}_q|^2 \sin^2 \left( \frac{\theta}{2} \right),
\]

where \( \tilde{E}_q = E_q / E_{q,0} \) is the fraction of the synchronous Floquet space harmonic amplitude relative to the fundamental Floquet radiation mode of the grating structure.

Summarizing this section of QEW interaction with classical light, we determined that the transition from the quantum plane-wave limit of the electron to the classical ‘point-particle’ limit requires that the radiation wave is a classical coherent state of light, and that the extinction coefficient in (19a) \( \Delta E^{(1)} = \Delta E e^{-\Gamma^2/2} \) tends to \( e^{-\Gamma^2/2} \to 1 \). This requires that the electron wavefunction spatial distribution is point-like, with the wavepacket-size satisfying condition: \( \Gamma \ll 1 \). In the opposite limit \( \Gamma \gg 1 \) the phase-dependent term (18a) diminishes: \( \Delta E^{(1)} \to 0, \Delta \nu_{\text{vac}}^{(1)} \to 0, \) and only the phase-independent term (18b) remains, which defines the earlier known quantum plane-wave limit [5]. Figure 3(a) depicts the classical to quantum wavepacket transition of the new ‘beyond FGR’ first-order term (18a). Besides the wavepacket-dependent Gaussian decay, it shows in broken line also the non-vanishing wavepacket-independent spontaneous emission contribution of the second-order term (15) that acts as noise, and it must be exceeded in order to observe the first order stimulated emission. Figure 3(b) is a color code presentation of the extinction coefficient \( e^{-\Gamma^2/2} \) as a function of the ratio between the intrinsic wavepacket size (at its waist) \( \sigma_{\text{z}} \) and the radiation wavelength at the electron rest frame: \( \Gamma_0 = 2 \pi \sigma_{\text{z}} / \beta \lambda \) and of the drift length of the wavepacket \( L_D = \nu t_D \) from its waist point to the interaction point. It demonstrates that the phase-dependent interaction of the quantum wavepacket depends, curiously enough, on its history before interaction.

Experimental confirmation of the ARC relation and the wavepacket dependence prediction of (18a), would require simultaneous measurement of photon emission and electron energy spectra of single-electron radiative interaction events. This is a challenging experiment that requires accumulating multiple interaction events data of electron wavepacket radiative interaction and preselection of the wavepackets, using wavepacket shape-formation laser streaking schemes [23–25] and perhaps using weak value measurement, as suggested in reference [79, 84, 85]. The detection of the radiation, certainly requires also the satisfaction of the signal-to-noise ratio condition \( S/N > 1 \), considering the ever-present wavepacket-independent noise due to spontaneous emission (quantum noise).

The photon 'size' effect of quantum light—squeezed coherent state. Coherent and squeezed states of photons are two basic elements of quantum optics. Coherent state is the closest analog to a classical
electromagnetic field, but the squeezed state has no such classical counterpart. [66, 86]. The phase squeezing of photon coherent state in quadrature representation, is analogous to controlling the size of the Gaussian electron wavepacket in phase-space. It is thus interesting to examine the 'photon size' effect of squeezed light when interacting with a 'finite-sized' quantum electron. Following the standard definition for the case of squeezed vacuum state (squeezing of photon coherent state in quadrature representation, is analogous to controlling the size of the wavepacket phase-dependent interaction, and certainly not on its point particle limit. In second-order, we did find such dependence on the absolute squeezing factor $|\xi|$ where $|\xi| \equiv S(\xi) D(\alpha) |0\rangle$.

\[ |\xi, \alpha\rangle = S(\xi) D(\alpha) |0\rangle, \quad \text{(23)} \]

where $|\alpha|^2 = \nu_0$, and $\xi$ is the squeezing parameter of the unitary squeezing operator $S(\xi)$.

We can now derive the radiation/acceleration correspondence (11) for this case, starting from (9) and (10), following the derivation of (18) in the coherent state case, but instead of (19), using now in the calculation of the matrix elements, the expectation values of a squeezed coherent photon state [86]:

\[ \langle \xi, \alpha | \hat{a}^\dagger \hat{a} | \xi, \alpha \rangle = \sqrt{\nu_0} \langle \xi, \alpha | \hat{a}^\dagger \hat{a} | \xi, \alpha \rangle = \sqrt{\nu_0}, \]

\[ \langle \xi, \alpha | \hat{a}^\dagger \hat{a} | \xi, \alpha \rangle = \nu_0 + \sinh^2 |\xi|. \quad \text{(24)} \]

The first two expressions for the expectation value of field strength are not different from the first two expressions in (19) for the case of coherent state of light, and thus do not change the result for the phase-dependent ARC expression (18a). However, the third expression in (24) makes a difference in the calculation of the matrix element of the phase-independent term (18b). Therefore, the ARC expressions for the case of squeezed state and a Gaussian QEW are modified to

\[ \Delta \nu_\text{q}^{(1)} = -\Delta E^{(1)}/\hbar \omega = 2 \Upsilon \sqrt{\nu_0} \tau^2/2 \sin \left( \frac{q}{2} \cos \left( \frac{q}{2} + \phi_0 \right) \right), \]

\[ \Delta \nu_\text{q}^{(2)} = -\Delta E^{(1)}/\hbar \omega = \Upsilon^2 \left( (\nu_0 + \sinh^2 |\xi| + 1) \sin^2 \left( \frac{q}{2} \right) - (\nu_0 + \sinh^2 |\xi|) \sin^2 \left( \frac{q}{2} \right) \right). \quad \text{(25)} \]

The coherent state is position-squeezed at $\xi > 0$, and momentum squeezed at $\xi < 0$, where the relative photon 'size' is defined as $e^{-|\xi|}$. We have found that the squeezing has no effect on the ARC expression of the wavepacket phase-dependent interaction, and certainly not on its point particle limit. In second-order, we find such dependence on the absolute squeezing factor $|\xi|$, increasing the effective input photon number to $\nu_0 + \sinh^2 |\xi|$. In particular, the conventional spontaneous emission expression (15) is modified for the case of squeezed vacuum state ($\nu_0 = 0$), and is given in the limit of small interaction recoil $\varepsilon \ll 1$, by

\[ \Delta \nu_\text{q} = \Delta \nu_\text{vac} + \Upsilon^2 \sinh^2 |\xi| \left( \frac{d}{d\theta} \sin^2 \left( \frac{q}{2} \right) \right), \quad \text{(26)} \]

which can be interpreted (compare to (18)) as FEL-kind stimulated amplification of the squeezed vacuum state.

In passing, it is worth noting that this additional term is similar to the Hawking–Unruh effect for accelerating observers (photon detection), widely studied in the context of black holes and optics [67, 87–89]. The Unruh-like Bose–Einstein factor is analogous to our squeezing photon number $\sinh^2 |\xi| = \left( e^{\frac{q}{2} + \phi_0} - 1 \right)^{-1}$, with $T_U$ being the Unruh temperature [67] and $k_B$ being the Boltzman constant. The connection may be attributed to the common mathematical formulation of the squeezing operator $S(\xi)$ (23), for the case of squeezed vacuum states of quantum light and the Bogoliubov transformation from the Kinder space-time of non-inertial observer to Minkowski space-time [88].

Summarizing the results of this section on QEW interaction with squeezed quantum light, we find no quantum photon 'size' effect in connection to the wavepacket size and phase-dependent interaction term. However, we find that the phase-independent acceleration/emission term (25b) of a QEW with squeezed light state of finite photon number ($e^{-|\xi|}$) is modified, and exhibits non-classical optical properties of non-Poissonian distribution. Thus, the general theoretical framework of ARC spectral reciprocity presented here lays ground for studying super- or sub-Poissonian photon distributions in numerous quantum sources beyond the Fock state and coherent state of light [64–67].

Remarks on 'classical' spontaneous emission. Here, we address some queries regarding spontaneous emissions in classical and quantum theories. In a semiclassical model in which the expectation value of the squared absolute value of the wavefunction is interpreted as the density of the QEW [35], the solution of Maxwell equations results in wavepacket size and shape dependent spontaneous radiation, and in the case of density modulation—superradiant harmonic frequency emission. This results in the classically derived expression for coherent radiation emission from a bunched point-particle beam of electrons in a variety of
interaction schemes (figure 1), such as Cherenkov, Smith–Purcell and Undulator–Synchrotron radiations [32, 33, 54]. However, our QED formulation of the same radiative interaction scheme, that originates from ‘zero-point quantum vibration’, does not produce this wavepacket-dependent ‘spontaneous emission’. It produces only the expressions for spontaneous emission per mode (15) and for angular-spectral energy emission of spontaneous Smith–Purcell radiation (22), both manifestly independent of finite-size or modulation features of the QEW [14], not adding anything to the spontaneous emission expression of point-particle or plane-wave models [5]. Similar lack of dependence of spontaneous Smith–Purcell emission on the transverse dimensions of the QEW was derived in [61] in the framework of QED theory. Thus, contrary to the stimulated interaction case, QED analyses of spontaneous radiation emission from a single QEW fail to produce the wavepacket and ‘point particle’ ‘classical spontaneous emission’ limit [35, 37]. We should point out that transition of quantum theory of spontaneous radiation emission of a free electron to the classical point-particle trajectories limit has been studied in several publications in a quasi-classical trajectory-coherent states (TCS) formulation [58–60, 90]. This included also derivation of first order (in \(\hbar\)) quantum correction to the classical expression. However, this still does not show QEW dependence of spontaneous emission.

The transition of QED-derived expressions of spontaneous emission to the classical or semiclassical limits, and its dependence on the interaction scheme are still a disputable and raising theoretical and experimental research problems [62]. We conjecture that the generalized QEW formulation presented here, and particularly the interesting electron–photon energy reciprocity relation (ARC) (4), (11), that originates from not neglecting the interference terms in (7), (8) would be instrumental in bridging the QED to classical theories of radiation from charged particles, perhaps in the context of non-projective measurement of single particle wavefunctions using weak or protective measurement schemes [79, 85]. The ARC relations have wide validity in a variety of quantum light and electron interaction cases, including the case of entangled electron–photon states (on which we did not expand in the present work). We also conjecture that further development of this methodology may provide a new QEW approach in the investigation of the long-standing problem of single-particle Lorentz–Abraham self-force and radiation reaction effect in second quantization treatment [54].

Conclusion

The main result of this work is the acceleration/radiation correspondence (ARC) (4), (11) that intimately connects the physics of particle acceleration and single mode photon radiation at the fundamental level. In the framework of QED theory, these fundamental relations can only be derived through a methodologically new analysis of electron–wave interaction as a scattering process, keeping the interference terms between the initial and scattered states. These terms have been previously ignored in QED scattering analyses based on FGR. Beyond showing consistency of our QED analysis with classical and semiclassical theories of phase- and shape-dependent stimulated interaction with light in the case of coherent state, we presented a new extension of the QED interaction theory to the case of interactions with quantum light–Fock state and squeezed state.

Our reciprocal QED formalism beyond FGR allows straightforward extension to other quantum light interactions, such as interaction of entangled electron–light states that we did not explore in the present work. We conjectured that further development of this formulation can be useful in the study of interesting fundamental quantum theory problems [53, 91, 92], such as an electron wavefunction Schrödinger’s cat states, the Lorentz–Abraham self-force, and QED derivation of classical limit spontaneous emission.

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Appendix A. Classical point-particle acceleration theory

In classical mechanics, the electron interaction with radiation field is described in terms of the Hamiltonian:

\[
H = \frac{(p + eA)^2}{2m_0} - e\phi, \tag{A1}
\]
where $A$ is the vector potential, $\phi$ is the scalar potentials, $e$ and $m_0$ are the charge and mass of electron, respectively. This is the same Hamiltonian from which the Schrödinger equation interaction Hamiltonian (equation (12)) is derived. The Lorentz force equation, which is derived from Hamilton’s equation, gives the forces that govern the dynamics of a charged point-particle in response to electric and magnetic fields:

$$\frac{d\mathbf{p}}{dt} = e(\mathbf{E} + \mathbf{v} \times \mathbf{B}), \quad \frac{d\mathbf{E}}{dt} = -e\mathbf{E} \cdot \mathbf{v},$$

(A2)

where the relativistic momentum $\mathbf{p} = m_0\gamma \mathbf{v}$, $\gamma$ is the Lorentz factor. The electric and magnetic field components are given by $\mathbf{E} = -\partial\mathbf{A}/\partial t - \nabla\phi$, $\mathbf{B} = \nabla \times \mathbf{A}$, respectively. The kinetic energy increment of a free electron is given by the work done:

$$\Delta E = -e \int E \cdot v \, dt,$$

(A3)

For our concerns, suppose that a free electron passes through an electromagnetic field with longitudinal electric component $E = E_z \cos(\omega t - q_z z + \phi_0)$, of frequency $\omega$, wavenumber $q_z$, and phase $\phi_0$, and with a constant electron velocity ($v_0$), the classical point-particle energy gain from equation (A3) is,

$$\Delta E = -ev_0 \int_0^{\frac{t}{\omega}} E_z \cos \left(\frac{\omega t'}{v_0} - q_z \left(t' + \phi_0\right)\right) \, dt'$$

$$= -eE_zL \sin \left(\frac{\theta}{2}\right) \cos \left(\frac{\theta}{2} + \phi_0\right).$$

(A4)

This is the expression of linear particle acceleration (equation (1)), with $\theta = (\omega/v_0 - q_z) L$, defined as the synchronism detuning parameter, $L$ the interaction length and $v_0$ the electron axial velocity. This formula is widely used in the context of traditional rf cavity and laser-driven particle accelerators, as well as in free-electron lasers.

Appendix B. The ‘modified’ Schrödinger equation from the Klein–Gordon equation

Some of the electron–radiation interaction schemes referred to in the paper (Smith-Purcell radiation, PINEM, FEL, etc) operate with a relativistic beam; therefore, the use of Schrödinger equation would not be satisfactory for all cases of interest. Since spin effects are not relevant for the present problem, we do not need to use Dirac equation, but rather can base our analysis on the Klein–Gordon (KG) equation. Therefore, we reiterate the derivations in references [5, 13] of a Schrödinger-like equation out of the KG equation, using a second-order iterative expansion of the free electron energy around its center energy $\varepsilon_0 = \sqrt{p_0^2 c^2 + m_0^2 c^4}$. This expansion reduces the quadratic KG equation into the parabolic Schrödinger equation under the well-satisfied approximation that the initial average momentum and the momentum transfer due to the interaction are within the range $\Delta p \ll p_0$. In typical electron microscope PINEM-kind experiments, the light quantum is $\hbar \omega = 1.6$ eV, much smaller than the incoming electron energy $\varepsilon_0 = 100–200$ keV.

The KG equation originates from the relativistic energy-momentum dispersion: $E_p^2 = p^2 c^2 + m^2 c^4$, where $m$ is the electron rest mass, and $c$ is the speed of light. To obtain the KG equation, we make the replacements $E \rightarrow i\hbar \frac{\partial}{\partial t}, p \rightarrow -i\hbar \nabla - eA$ (minimal coupling with electromagnetic radiation) and apply the differential operator on a wavefunction:

$$\left(i\hbar \frac{\partial}{\partial t}\right)^2 \psi(r,t) = c^2(-i\hbar \nabla - eA)^2 \psi(r,t) + m^2 c^4 \psi(r,t),$$

(B1)

where $e$ is an electron charge. The KG equation can describe the relativistic electrons in most of the considered radiation schemes if spin effects are negligible. If the radiation field is weak, $eA/mc \ll 1$, then excitation of the negative (positron) energy branch of the dispersion equation is negligible and one can approximate the wavefunction $\psi(r,t)$ with a single quasi-harmonic positive energy wave

$$\psi(r,t) = u(r,t) e^{-i\varepsilon_0 t/\hbar},$$

(B2)

where $\varepsilon_0 = \sqrt{p_0^2 c^2 + m^2 c^4} = \gamma_0 mc^2$, $p_0$ the center momentum and $u(r,t)$ is a slowly varying function of time. Then substitution of equation (B2) in (B1) and canceling the fast-varying coefficient $e^{-i\varepsilon_0 t/\hbar}$, result in the form of a modified Schrödinger equation,

$$i\hbar \frac{\partial \psi(r,t)}{\partial t} = -i\hbar \frac{\partial u(r,t)}{\partial t} e^{-i\varepsilon_0 t/\hbar} + \varepsilon_0 u(r,t) e^{-i\varepsilon_0 t/\hbar} = H\psi(r,t),$$

(B3)
where the effective Hamiltonian is

$$H = \varepsilon_0 + \frac{c^2(-i\hbar \nabla - eA)^2 + (m^2 c^4 - \varepsilon^2_0)}{2\varepsilon_0} - \frac{1}{2\varepsilon_0} \left( \frac{c^2(-i\hbar \nabla - eA)^2 + (m^2 c^4 - \varepsilon^2_0)}{2\varepsilon_0} \right)^2. \tag{B4}$$

The Hamiltonian can be split into an unperturbed electronic part and a radiative perturbation part 

$$H = H_0 + H_1(t),$$

where

$$H_0 \simeq \varepsilon_0 + \nu_0 \left( -i\hbar \nabla - p_0 \right) + \frac{1}{2\gamma_0 m} \left( -i\hbar \nabla - p_0 \right)^2, \tag{B5}$$

and to first order in the vector potential $A$,

$$H_1(t) = -\frac{e(A \cdot (-i\hbar \nabla) + (-i\hbar \nabla) \cdot A)}{2\gamma_0 m}, \tag{B6}$$

where $\nu_0 = p_0/\gamma_0 m = \beta_0 e$ and $\gamma_0 = 1/\sqrt{1 - \beta^2}$ is the Lorentz factor, $\beta = \nu_0/c$ and $m$ is the free electron mass. For specification in our one-dimensional electron–photon interaction model in a slow-wave structure (such as a grating), we consider a monochromatic laser field with frequency $\omega$, $\tilde{A} = -\frac{1}{\gamma_0 \omega}(\tilde{E}(r) e^{-i\nu t} - \tilde{E}^*(r) e^{i\nu t})$, where $\tilde{A}, \tilde{E}$ are electromagnetic field operators. In our one-dimensional analysis, we assume that the light–electron coupling takes place through an axial slow-wave field component of one of the traveling modes $(q)$: $\tilde{E}(r) = \sum_q \tilde{E}_q e^{ikz - i\omega t} a_q e_z$, where $a_q (\tilde{a}_q^\dagger)$ is the annihilation (creation) operator of the photon’s Fock state $|\nu\rangle$ in this quantized mode.

$$H_1(t) = \frac{-\hbar \nabla^2}{2\gamma_0 m \omega} \sum_q \left( \tilde{E}_q e^{ikz - i\omega t} \tilde{a}_q - \tilde{E}_q^* e^{-ikz + i\omega t} \tilde{a}_q^\dagger \right)$$

$$= V_{int}^{(d)} \tilde{a}_q e^{-i\nu t} + V_{int}^{(c)} \tilde{a}_q^\dagger e^{i\nu t}, \tag{B7}$$

in which $V_{int}^{(d)} = -\frac{\hbar \nabla^2}{2\gamma_0 m \omega} \tilde{E}_q e^{ikz - i\omega t} \tilde{a}_q$, $V_{int}^{(c)} = \frac{\hbar \nabla^2}{2\gamma_0 m \omega} \tilde{E}_q^* e^{-ikz + i\omega t}$, with $q$ satisfying near synchronism condition selectively: $\omega / \tilde{q} = \nu_0$, so that the integration over the mode index $q$ is suppressed. Note that the Coulomb gauge is chosen, i.e., $\nabla \cdot A = 0$.

**Appendix C. Quantum theory of free-electron spontaneous and stimulated radiation interaction schemes and FGR**

Here we derive from first principles the QED equation for spontaneous and stimulated emission by free electrons (3) that was previously analyzed in [5]. Using time-dependent perturbation theory, we express the development of the initial state time dependent coefficients in (5) into the final state (6) by expansion in terms of powers of the interaction strength $|\langle H_1|\rangle$,

$$c_{p,\nu}^{(f)}(t) = \sum_{p',\nu'} c_{p,\nu}^{(0)}(t) + \sum_{p',\nu'} c_{p,\nu}^{(1)}(t) + \sum_{p',\nu'} c_{p,\nu}^{(2)}(t) + \cdots, \tag{C1}$$

where $c_{p,\nu}^{(m)} \sim O(|H_1|^m)$. In the interaction presentation, the final state $|f\rangle$ can be related to the initial state $|i\rangle$ through the time-evolution operator, i.e., $|f\rangle = U_1(t, t_0)|i\rangle$, where

$$U_1(t, t_0) = T e^{-i\hbar \int_{t_0}^{t} H_1^{(i)}(t') dt'}, \tag{C2}$$

where $H_1^{(i)}(t) = e^{iH_0/\hbar} H_1(t) e^{-iH_0/\hbar}$. We take the Taylor expansion and consider only the first and second-order terms:

$$U_1(t, t_0) = 1 - \frac{i}{\hbar} \int_{t_0}^{t} H_1^{(i)}(t') dt' + \left( \frac{i}{\hbar} \right)^2 \int_{t_0}^{t} H_1^{(i)}(t') \int_{t_0}^{t'} H_1^{(i)}(t'') dt'' + \cdots, \tag{C3}$$

Making use of the definitions from equations (5) and (6), and the identity of basis, $\sum_{p,\nu} |p, \nu\rangle \langle p, \nu| = 1$, $H_0 |p, \nu\rangle = \tilde{E}_p |p, \nu\rangle$, and we can get

$$c_{p,\nu}^{(f)}(t) = c_{p,\nu}^{(0)}(t) - \frac{i}{\hbar} \sum_{p',\nu'} c_{p',\nu'}^{(0)} \int_{t_0}^{t} \langle p, \nu | H_1^{(i)}(t') | p', \nu' \rangle \langle p', \nu' | \int_{t_0}^{t'} H_1^{(i)}(t'') dt'' + \cdots$$

$$- \frac{1}{\hbar^2} \int_{t_0}^{t} dt' \int_{t_0}^{t} dt'' \sum_{p'',\nu''} c_{p'',\nu''}^{(0)} \langle p, \nu | H_1^{(i)}(t') | p', \nu' \rangle \langle p', \nu' | \int_{t_0}^{t'} H_1^{(i)}(t'') | p'', \nu'' \rangle + \cdots$$

13
Therefore, we find that

\[ c^{(1)}_{p',\nu'}(t) = -\frac{i}{\hbar} \sum_{p''\nu''} c^{(0)}_{p''\nu''} \int_0^t dt' e^{-i(E_{p''\nu''} - E_{p'\nu'})/\hbar} \langle p, \nu | H(t') | p', \nu' \rangle, \]

\[ c^{(2)}_{p',\nu'}(t) = -\frac{1}{\hbar^2} \int_0^t dt' e^{-i(E_{p''\nu''} - E_{p'\nu'}/n} \int_0^t dt'' e^{i(E_{p''\nu''} - E_{p'\nu'}/n} \times \sum_{p''\nu''} c^{(0)}_{p''\nu''} \langle p, \nu | H(t') | p', \nu' \rangle \langle p', \nu' | H(t'') | p'', \nu'' \rangle. \]  

(C4)

In particular, the probability of effecting a photon-assisted transition from an initial state \[ |\tilde{\psi}(t)\rangle = \sum_{p',\nu'} c^{(0)}_{p',\nu'} \langle p', \nu' | \] to a final state \[ |\tilde{\psi}(t)\rangle \], for instance, i.e., \[ c^{(0)}_{p',\nu'} = \delta_{p',p} \delta_{\nu',\nu_0} \], neglecting interference effect between the initial and final states, is given by

\[ P_{p,\nu_0\rightarrow p',\nu} = \left| c^{(1)}_{p',\nu'}(t) + c^{(2)}_{p',\nu'}(t) + \cdots \right|^2, \quad \text{for} \quad p \neq p', \quad \nu \neq \nu_0. \]

(C5)

In order to finish the time integration in (C4), we use the electron–photon coupling Hamiltonian (equation (12)) that includes photon emission (+) and absorption processes (+), \[ H_{int}(t) = V^{(e)}_{\nu_0} a_{\nu_0} e^{-i\omega t} + V^{(a)}_{\nu_0} a_{\nu_0} e^{i\omega t} \] and by scaling of delta function \[ \int_0^t dt' e^{-i(E_{p''\nu''} - E_{p'\nu'}/h)} = e^{i(E_{p''\nu''} - E_{p'\nu'}/h)} \delta(t - t_0) \].

Therefore, the probability of single-photon transition after a duration \[ \Delta t = t - t_0 \] is given by

\[ P_{p,\nu_0\rightarrow p',\nu} = \left| c^{(1)(\text{calc})}_{p',\nu'}(t) \right|^2 = \frac{1}{\hbar^2} \left| \langle p, \nu | V^{(\text{calc})}_{\nu_0} \hat{a}_{\nu_0} (\hat{a}_{\nu_0}^\dagger) | p_0, \nu_0 \rangle \right|^2 \frac{\sin \left( \frac{(E_{p''\nu''} - E_{p'\nu'}) \Delta t}{\hbar} \right)}{\Delta t}. \]

(C7)

Using the formula \[ \lim_{\Delta t \to \infty} \frac{1}{\Delta t} \left[ \sin(\omega \Delta t) / \omega \right]^2 = 2\pi \delta(2\alpha) \] leads to the following expression:

\[ W_{p,\nu_0\rightarrow p',\nu} = \lim_{\Delta t \to \infty} \frac{P_{p,\nu_0\rightarrow p',\nu}}{\Delta t} = \frac{2\pi}{\hbar^2} \left| \langle p, \nu | V^{(\text{calc})}_{\nu_0} \hat{a}_{\nu_0} (\hat{a}_{\nu_0}^\dagger) | p_0, \nu_0 \rangle \right|^2 \delta \left( \frac{E_{p''\nu''} - E_{p'\nu'}}{\hbar \omega} \right) \]

and by scaling of delta function \[ \delta(\chi/a) = |a| \delta(x/a) \], we obtain the expression for transition rate:

\[ W_{p,\nu_0\rightarrow p',\nu} = \frac{2\pi}{\hbar} \left| \langle p, \nu | V^{(\text{calc})}_{\nu_0} \hat{a}_{\nu_0} (\hat{a}_{\nu_0}^\dagger) | p_0, \nu_0 \rangle \right|^2 \delta \left( E_{p''\nu''} - E_{p'\nu'} + \hbar \omega \right). \]

(C8)

This expression is also known as Fermi’s golden rule (FGR). In the second-order perturbation analysis, the calculation involves two-photon processes, such as absorption–absorption, emission–emission, absorption–emission and emission–absorption, and requires the detailed expressions.

To obtain expressions for stimulated radiation, we can use the transition rate \[ W_{p,\nu_0\rightarrow p',\nu} \] in the first-order perturbation approximation we obtain the scattering probability rate of the electron from state \[ |p_0\rangle \] to state \[ |p\rangle \] associated with emission (or absorption) of a photon having energy quanta \( \hbar \omega \). Note that the final photon state is determined by generating or annihilating a photon in the matrix elements,

\[ \langle \nu | \hat{a}_{\nu} | \nu_0 \rangle = \sqrt{\nu_0 \delta_{\nu_0,\nu-1}}, \quad \langle \nu | \hat{a}_{\nu}^\dagger | \nu_0 \rangle = \sqrt{\nu_0 + 1} \delta_{\nu_0,\nu+1}, \]

(C9)

where the additional ‘1’ in the emission term leads to spontaneous emission. Therefore, we can rewrite the transition rate,

\[ W_{p,\nu_0\rightarrow p,\nu_0+1} = \frac{2\pi (\nu_0 + 1)}{\hbar} \left| \langle p | V^{(\text{calc})}_{\nu_0} | p_0 \rangle \right|^2 \delta \left( E_{p''\nu''} - E_p + \hbar \omega \right), \]

\[ W_{p,\nu_0\rightarrow p,\nu_0-1} = \frac{2\pi \nu_0}{\hbar} \left| \langle p | V^{(\text{calc})}_{\nu_0} | p_0 \rangle \right|^2 \delta \left( E_{p''\nu''} - E_p - \hbar \omega \right). \]
Finally, we integrate over all the electronic degrees of freedom \((p_0, p)\), taking into account the initial component. We obtain the net stimulated photon transition rate \([5]\)

\[
\Delta \nu_q = W_{\nu_q=\nu_{q+1}} - W_{\nu_q=\nu_{q-1}} = \Gamma_{\nu_q} ((\nu_0 + 1) F_\nu (\omega) - \nu_0 F_\nu (\omega)),
\]

and for the case \(\nu_0 = 0\), the spontaneous photon transition

\[
\Delta \nu_{qsp} = \Gamma_{\nu_q} F_\nu (\omega),
\]

where the spontaneous coefficient \(\Gamma_{\nu_q}\) and the emission/absorption spectral lines functions \(F_{\nu_\omega} (\omega)\) are found from the calculation of the emission matrix element for the specific light–electron interaction scheme \((C10)\). We refer the reader to more literature on the quantum theory of photon radiations \([5, 41, 74]\). For our Smith–Purcell setup, we find that

\[
\Gamma_{\nu_q} = \tilde{T}^2 = \left( e \tilde{E}_{z_{qm}} L / 4\hbar\omega \right)^2,
\]

\[
F_\nu (\omega) = \sin^2 \left( \frac{\tilde{\theta}_\nu}{2} \right), \quad F_{\nu_\omega} (\omega) = \sin^2 \left( \frac{\tilde{\theta}_{\nu_\omega}}{2} \right).
\]

The crucial departure point of our beyond-FGR formulation from the conventional FGR \((C8)\) is in using the relations \((7)\) and \((8)\) for the transition probability instead of \((C5)\). This difference matters in the case of a finite size QEW, for which the interference between the zero-order and first-order scattered terms cannot be neglected.

**Appendix D. Some relevant integrations involving Gaussians**

We note that in the present analysis \((10)\) we neglect the contribution of possible interference between the emission and absorption scattering amplitudes

\[
2 \sum_{p\nu} \Re \left\{ \left( \tilde{c}_{p,\nu}^{(0)} \right) \left( \tilde{c}_{p,\nu+1}^{(0)} \right)^* \right\} \left( F_p - E_0 \right).
\]

This emission–absorption interference term is small in the first-order perturbation analysis when

\[
\left| \tilde{c}_{p,\nu}^{(0)} \right|^2 \gg \left| \tilde{c}_{p,\nu+1}^{(0)} \right|^2.
\]

We thus neglect, in first-order perturbation approximation, the higher-order interference between emission and absorption components, that are second order terms, and concentrate in the paper only on the interference terms between the initial and the scattered components, which are first order in perturbation approximation. However, we point out that the interference term \((D1)\) may be important in nonperturbative shaping of the electron wavefunction in the opposite case \(\left| \tilde{c}_{p,\nu}^{(0)} \right|^2 \ll \left| \tilde{c}_{p,\nu+1}^{(0)} \right|^2\), which may relate to the case discussed by Murdia and coauthors \([90]\) in the context of Bremsstrahlung.

Second, to derive the photon emission and electron acceleration expressions \((18), (25)\), the integration over \(p\) in \((9), (10)\) should be carried out with the Gaussian distribution function of the amplitude in momentum space of the drifted electron. For the phase-independent second-order photon emission \((e)\) terms \(\Delta \nu^{(2)}_q, \Delta E^{(2)}_q\), this involves the following integrations

\[
\sum_p \left\{ \left( \frac{p + p^{(e)}_{rec} - \hbar q_{zm}/2}{p_{0}} \right)^2 \left| \tilde{c}_{p,\nu}^{(0)} \right|^2 \left| \tilde{c}_{p,\nu+1}^{(0)} \right|^2 \right\}
\]

\[
= (2\pi\sigma_{p_{0}}^2)^{-1/2} \int dp \left( \frac{p + p^{(e)}_{rec} - \hbar q_{zm}/2}{p_{0}} \right)^2 \exp \left( - \frac{(p + p^{(e)}_{rec} - p_{0})^2}{2\sigma_{p_{0}}^2} \right)
\]

\[
= \left( 1 - \frac{\hbar q_{zm}}{2p_{0}} \right)^2 \left( \frac{\sigma_{p_{0}}}{p_{0}} \right)^2 \approx 1,
\]

\[
\sum_p \left\{ \left( \frac{p + p^{(e)}_{rec} - \hbar q_{zm}/2}{p_{0}} \right)^2 \left| \tilde{c}_{p,\nu}^{(0)} \right|^2 \left| \tilde{c}_{p,\nu+1}^{(0)} \right|^2 \right\}
\]

\[
= (2\pi\sigma_{p_{0}}^2)^{-1/2} \int dp \left( \frac{p + p^{(e)}_{rec} - \hbar q_{zm}/2}{p_{0}} \right)^2 \exp \left( - \frac{(p + p^{(e)}_{rec} - p_{0})^2}{2\sigma_{p_{0}}^2} \right)
\]

\[
\approx p_{0}.
\]
Similarly, for the absorption \((a)\) term
\[
\sum_p \left\{ \left( p - \left( p_{\text{rec}}^{(a)} - \hbar q_{\text{zm}}/2 \right) / p_0 \right)^2 \left| c_p^{(0)} \right|^2 \right\} = \left( 1 + \hbar q_{\text{zm}} / 2 p_0 \right)^2 + \left( \sigma_p / p_0 \right)^2 \approx 1, \tag{D3} \]

For the phase-dependent first-order photon emission \((e)\) part \((\Delta \nu_1^{(1)}, \Delta E^{(1)})\)
\[
\sum_p \left\{ \left( p + \left( p_{\text{rec}}^{(e)} - \hbar q_{\text{zm}}/2 \right) / p_0 \right)^2 \left| c_p^{(0)} e^{i \omega / \hbar} \right|^2 \right\} = \left( 2\pi \sigma_p^2 \right)^{-1/2} \int dp \left( p + \left( p_{\text{rec}}^{(e)} - \hbar q_{\text{zm}}/2 \right) / p_0 \right)^2 \right\} \mathcal{R} \left\{ \left( p + \left( p_{\text{rec}}^{(e)} - \hbar q_{\text{zm}}/2 \right) / p_0 \right)^2 \left| c_p^{(0)} e^{i \omega / \hbar} \right|^2 \right\} e^{-\Gamma^2 / 2} \tag{D4} \]

Similarly, for the absorption \((a)\) term
\[
\sum_p \left\{ \left( p - \left( p_{\text{rec}}^{(a)} - \hbar q_{\text{zm}}/2 \right) / p_0 \right)^2 \left| c_p^{(0)} e^{i \omega / \hbar} \right|^2 \right\} = e^{-\Gamma^2 / 2} \left( 1 - p_{\text{rec}}^{(a)} - \hbar q_{\text{zm}} + ip_{\text{rec}}^{(a)} \xi_0 \right) \approx e^{-\Gamma^2 / 2}, \tag{D5} \]

where we define the decay parameter \(\Gamma = \left( \sigma \right)^{-1} / \hbar \) \(\sigma \approx \left( \hbar / \omega \right) \left( \sqrt{1 + \xi^2 \xi_0^2} - 1 \right) = \Gamma_0 \sqrt{1 + \xi^2 \xi_0^2} \), and \(\xi = \sqrt{\sqrt{\sigma} / \hbar m} \approx \sqrt{\sigma / \hbar m} \). Similarly, in all cases, we took the approximations \(\left( \xi_0 / \hbar \right) \approx 1, \hbar q_{\text{zm}} / p_0 \ll 1, \sigma_p / p_0 \ll 1. \)

**ORCID iDs**

Yiming Pan [https://orcid.org/0000-0003-4391-0226]
Avraham Gover [https://orcid.org/0000-0002-6132-9462]

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