Spontaneous and stimulated emissions of a preformed quantum free-electron wave function

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Do the prior history and the wave-packet size and form of a free electron have a physical effect in its interaction with light? Here we answer these fundamental questions on the interpretation of the electron quantum wave function by analyzing spontaneous and stimulated emissions of a quantum electron wave packet, interacting with a general, quantized radiation field. For coherent radiation (Glauber state), we confirm that stimulated emission and absorption of photons depends on the preinteraction-history-dependent size, exhibiting spectral cutoff when it exceeds the interacting radiation wavelength. Furthermore, stimulated emission of an optically modulated electron wave packet has a characteristic harmonic emission spectrum beyond the cutoff, which depends on the modulation features. In either case, there is no wave-packet-dependent radiation of the Fock state, and particularly the vacuum state spontaneous emission is wave-packet independent. The classical-to-quantum transition of radiation from the point-particle to the plane-wave limits, and the effects of wave-packet modulation indicate a way of measuring the wave-packet size of a single electron wave function, and suggest an alternative direction for exploring light-matter interaction.

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I. INTRODUCTION

Accelerated free electrons emit electromagnetic radiation when subjected to an external force (e.g., synchrotron radiation [\[1\]](#page-11-0), undulator radiation [\[2\]](#page-11-0), and Compton scattering [\[3\]](#page-11-0)). Radiation can also be emitted by currents that are induced by free electrons in polarizable structures and materials, such as in Cherenkov radiation [\[4\]](#page-11-0), transition radiation [\[5\]](#page-11-0), and Smith-Purcell radiation [\[6\]](#page-11-0). Some of these schemes were demonstrated to operate as coherent stimulated radiative emission sources, such as free-electron lasers (FELs) [\[7–11\]](#page-11-0), as well as accelerating (stimulated absorption) devices, such as the dielectric laser accelerator (DLA) and the inverse Smith-Purcell effect [\[12](#page-11-0)[–14\]](#page-12-0).

Most of the free-electron radiation schemes of emission or acceleration operate in the classical theoretical regime of electrodynamics, where the electrons can be considered point particles, and the radiation field is described by Maxwell equations (no field quantization). However, a variety of freeelectron radiation schemes [\[15,16\]](#page-12-0), and particularly FELs (e.g., Refs. [\[17–19\]](#page-12-0)) have been analyzed in the framework of a quantum model in which the electron is described in the inherent quantum limit, given as a plane-wave quantum wave function—the opposite limit of the point-particle classical presentation. Quantum description of the electron wave function is also used in another recently developed research field of electron interaction with radiation: photoinduced nearfield electron microscopy (PINEM) [\[20,21\]](#page-12-0). In this scheme, a single electron quantum wave function interacts with the near field of a nanometric structure illuminated by a coherent laser beam. Of special relevance for the present discussion is a recent PINEM-type experiment of Echternkamp *et al.* [\[22\]](#page-12-0), in

which it was demonstrated that optical frequency modulation of the energy and the density expectation values of a single electron wave packet are physically possible.

The extremely different presentations of the radiative interaction of free electrons in the classical and quantum limits raise interest in the theoretical understanding of the transition from the quantum to the classical limit of the radiative interaction process. In the classical description, the point-particle dynamics is governed by the Lorentz force equation, and its radiation—by Maxwell equations. The radiation field emitted spontaneously by a single free electron and its stimulated emission and absorption have phase dependence on the radiation field. However, in the "classical spontaneous emission" of free electrons (e.g., undulator radiation, Čerenkov radiation) the phase dependence of the individual electron washes out after averaging over an ensemble of electrons entering the interaction region at random phase. On the other hand, in the quantum description of spontaneous and stimulated radiation by a free electron there is no phase dependence already at the level of a single electron, because the electron is described by an infinitely extended plane wave [\[15\]](#page-12-0). The spontaneous emission is described as a consequence of "zero-field vibration" in a quantized field model. The stimulated radiation emission (absorption) is explained in terms of multiphoton emission (absorption) processes in which the electron makes a transition to lower (higher) energy states of its continuous energy dispersion curve.

The way to settle these two diverse points of view of the electron-radiation interaction, and understand the classicalto-quantum transition, is to describe the free electron as a quantum wave packet $[23]$, which would tend to resemble a plane wave when the wave packet is long relative to the radiation wavelength, and a point particle in the opposite limit. In this article, we analyze the spontaneous and stimulated emission problem of a quantum electron wave function in a

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FIG. 1. Smith-Purcell radiation experimental setup for measuring electron wave-packet spontaneous and stimulated emission and corresponding electron energy spectrum and wave-packet-dependent acceleration. (a) An expanding electron wave packet. (b) An energy-modulated wave packet, turning into a density-modulated bunched electron wave packet, and emitting stimulated-superradiant Smith-Purcell radiation and corresponding acceleration at harmonic frequencies.

quantum-electrodynamics (QED) formulation. We will show that in the case of stimulated interaction with a coherent (Glauber state) radiation field, the more general QED model is consistent with the semiclassical analyses [\[17,24,25\]](#page-12-0), but as expected, it has different predictions in the case of spontaneous emission [\[26–28\]](#page-12-0). Since recent experimental progress [\[29,30\]](#page-12-0) makes it feasible to generate, accelerate, control the shape and size, and modulate a single electron wave packet [\[22,31–34\]](#page-12-0), we assert, based on the presented theory, that radiative interaction experiments in the transition range between classical-to-quantum electron wave-packet limits, provide a viable way for measuring the dimension and structure of the electron quantum wave packet. It can help to resolve the difference in description of spontaneous emission in the classical and quantum formulations, and offers an alternative way to study fundamental aspects of radiation-matter interaction in the quantum limits. In particular, it can provide better insight into the fundamentally disputed problem of the physical interpretation of the quantum electron wave function and the particle-wave duality nature of the electron [\[35,36\]](#page-12-0).

II. MODELING AND METHODS

In the present work, we exemplify the general analysis for the case of radiative interaction of a free electron with a slow-wave axial field component of a radiation mode of frequency and wave number (ω, q_z) , such that it can interact synchronously with a copropagating electron of velocity $v_0 \simeq$ ω/q_z . Such a slow-wave component can be a near-field evanescent diffraction order (Floquet space harmonic) propagating along a periodic grating (Smith-Purcell structure), or it can be a TM wave in a dielectric structure or in a hollow dielectric waveguide (Cerenkov radiation structure) $[4,16,9]$ $[4,16,9]$ $[4,16,9]$ $[4,16,9]$.

In Fig. $1(a)$ we show a Smith-Purcell experimental setup for measuring the spontaneous and stimulated emission from such structures as well as the electron energy spectrum after interaction. In these experimental setups, it is possible to measure the radiation emission and the dependence of the electron energy spectrum on the size of the expanding wave packet $\sigma_z(t)$, which depends in this case on the drift time of the electron wave packet from its virtual emission point. Figure 1(b) shows an elaborate version of the Smith-Purcell experiment, in which the electron quantum wave packet is optically modulated (as demonstrated by Echternkamp *et al.* [\[22\]](#page-12-0)) before it drifts and enters into the radiative interaction section. We point out that even though the present analysis demonstrates the interaction for the case of a slow-wave structure, it can be straightforwardly extended to other radiative interaction schemes of a free electron, such as transition radiation and FELs.

Our QED analysis is based on first-order perturbation solution of the relativistically modified Schrödinger equation [\[15,24\]](#page-12-0) for a free-electron wave function and a quantized radiation field. The unperturbed Hamiltonian is similar to the one used in conventional quantum analysis of free-electron interaction $[15]$, but, as in $[24]$, the equation is solved here with initial conditions of a finite size electron quantum wave packet instead of a plane wave. The interaction Hamiltonian is taken to be

$$
H_I(t) = -\frac{e[\hat{\mathbf{A}} \cdot (-i\hbar \nabla) + (-i\hbar \nabla) \cdot \hat{\mathbf{A}}]}{2\gamma m},
$$
 (1)

where $\gamma = 1/\sqrt{1 - \beta^2}$ is the Lorentz factor, $\beta = v_0/c$, and *m* is the free-electron mass. For the case of our concern,

$$
\hat{\mathbf{A}} = -\frac{1}{2i\omega} [\hat{\mathbf{E}}(\mathbf{r})e^{-i\omega t} - \hat{\mathbf{E}}^{\dagger}(\mathbf{r})e^{i\omega t}], \tag{2}
$$

where \hat{A} , \hat{E} are 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 modes *q*:

$$
\hat{\mathbf{E}}(\mathbf{r}) = \sum_{q} \tilde{E}_{qz} e^{iq_z z - i\phi_0} \hat{a}_q \mathbf{e}_z, \tag{3}
$$

where $\hat{a}_q(\hat{a}_q^{\dagger})$ is the annihilation (creation) operator of photon number state $|v\rangle$ in this mode *q*. For Smith-Purcell interaction, the field (3) is the axial component of one of the space harmonics of a classical Floquet-mode radiation wave incident on a grating in a Smith-Purcell structure [\[24,25\]](#page-12-0), $\hat{\mathbf{E}}_q(\mathbf{r}) =$ $\sum_{m} \tilde{E}_{qm}e^{iq_{zm}z}$, where $q_{zm} = q_{z0} + m2\pi/\lambda_G$, and one of the space harmonics, *m*, satisfies the near-synchronism condition with the electron velocity $\omega/q_{zm} \simeq v_0$. The ratio between the interacting (synchronous) axial wave component at the electron path and the amplitude of the incident radiation wave (the fundamental space harmonic), $\eta_{qm} = \tilde{E}_{qzm}/\tilde{E}_{q\perp 0}$, is a function of the specific grating structure that can be calculated by direct solution of classical Maxwell equations. Here \tilde{E}_{qzm} and $\tilde{E}_{q\perp 0}$ are the axial component and transverse component of the *m*th-order and zeroth-order space harmonics, respectively. We assume that the fundamental space harmonic (that radiates in free space) is the dominant space harmonic and is box in tree space) is the dominant
quantized as $\tilde{E}_{q\perp 0} = \sqrt{\frac{2\hbar\omega}{\varepsilon_0 V}}$ [\[37\]](#page-12-0).

Following the standard quantum-electrodynamics theory, we expand the initial wave function in terms of the quantum numbers *p* of the electron state and the Fock photon

occupation state of mode q, which is given by $|i\rangle =$ $\sum_{p,\nu} c_{p,\nu}^{(i)}(t) |p,\nu\rangle$. For the case of an electron wave packet in our one-dimensional model, the initial wave function is given by

$$
|i\rangle = \int \frac{dp}{\sqrt{2\pi\hbar}} \sum_{\nu} c_{p,\nu}^{(0)} e^{-iE_p t/\hbar} |p,\nu\rangle, \tag{4}
$$

where the energy dispersion relation of relativistic free electrons is $E_p = c\sqrt{m^2c^2 + p^2}$.

First-order time-dependent perturbation theory of the Schrödinger equation [\[37\]](#page-12-0) (see derivation details in Appendix [A\)](#page-7-0) results in

$$
i\hbar \dot{c}_{p',\nu'}^{(1)} = \int \frac{dp}{\sqrt{2\pi\hbar}} \sum_{\nu} c_{p,\nu}^{(0)} \langle p',\nu'|H_I(t)|p,\nu\rangle e^{-i(E_p - E_{p'}x)t/\hbar}.
$$
\n(5)

Integrating (5) in time *t* from 0 to infinity, the emission and absorption process terms of the first-order perturbation coefficient $c_{p',v'}^{(1)} = c_{p',v'}^{(1)(e)} + c_{p',v'}^{(1)(a)}$ are given by, respectively,

$$
c_{p',\nu'}^{(1)(e,a)} = \frac{\pi}{2i\hbar} \int \frac{dp}{\sqrt{2\pi\hbar}} \sum_{\nu} c_{p,\nu}^{(0)} \langle p', \nu' | H_I^{(e,a)}(0) | p, \nu \rangle
$$

$$
\times \delta \left(\frac{E_p - E_{p'} + \hbar \omega}{2\hbar} \right), \tag{6}
$$

where $H_I^{(e,a)}$ correspond, respectively, to the second and first terms in the interaction Hamiltonian [\(1\)](#page-1-0).

The momentum quantum recoil of the electron is found from substituting in (6) the energy dispersion relation, expanded to second order: $E_p = c\sqrt{m^2c^2 + p^2} \approx \varepsilon_0 +$ $v_0(p - p_0) + \frac{(p - p_0)^2}{2m^*}$. Determined by the delta functions, it is $p_{\text{rec}}^{(e,a)} = p'^{(e,a)} - p_0 = p_{\text{rec}}^{(0)}(1 \pm \delta), p_0 = \gamma_0 m v_0$, where $p_{\text{rec}}^{(0)} =$ $\hbar \omega/v_0$, $\delta = \hbar \omega/2m^*v_0^2$, $m^* = \gamma_0^3 m$. Then the first-order perturbation coefficient is given by

$$
c_{p',v'}^{(1)(e,a)} = \begin{cases} +\left(\frac{p' + p_{\text{rec}}^{(e)} - \hbar q_{zm}/2}{p_0}\right) \tilde{\Upsilon} \sqrt{\nu'} c_{p' + p_{\text{rec},v'-1}}^{(0)} \text{sinc}\left(\overline{\theta}_m^{(e)}/2\right) e^{i(\overline{\theta}_m^{(e)}/2 + \phi_0)} \\ -\left(\frac{p' - p_{\text{rec}}^{(a)} + \hbar q_{zm}/2}{p_0}\right) \tilde{\Upsilon} \sqrt{\nu' + 1} c_{p' - p_{\text{rec},v'+1}}^{(0)} \text{sinc}\left(\overline{\theta}_m^{(a)}/2\right) e^{-i(\overline{\theta}_m^{(a)}/2 + \phi_0)}, \end{cases} \tag{7}
$$

$$
\frac{d(u,v)}{dt} = \frac{(p_{\text{rec}}^{(e,a)} \pm hq_{\text{zm}})L}{\hbar} = \left(\frac{\omega}{v_0} - q_{\text{zm}}\right)L \pm \left(\frac{\omega}{v_0}\right)L\delta = \overline{\theta}_m \pm \frac{\varepsilon}{2},\tag{8}
$$

where $\overline{\theta}_m = (\frac{\omega}{v_0} - q_{zm})L$ is the classical interaction "detuning parameter"; $\varepsilon = \delta(\frac{\omega}{v_0})L \ll 1$ is the interaction quantum recoil parameter [\[15\]](#page-12-0). The normalized photon exchange coefficient is $\tilde{\Upsilon} = \frac{e\tilde{E}_{qzm}L}{4\hbar\omega}$.

θ

The schematic diagram in Fig. 2 shows the light-matter scattering processes of emitting and absorbing a photon. Explicitly, for emitting a photon, the final coefficient of state $|p, v + 1\rangle$ is given by $c_{p, v+1}^{(0)} + c_{p, v+1}^{(1)(e)}$. This represents a reciprocal electron momentum and energy conserving process through emission of photon and momentum backward recoil: $|p + p_{\text{rec}}^{(e)}, v\rangle \Rightarrow |p, v + 1\rangle$ [\[38\]](#page-12-0). On the other hand, for absorbing a photon, the final coefficient of state $|p, v - 1\rangle$ is given by $c_{p,v-1}^{(0)} + c_{p,v-1}^{(1)(a)}$, which corresponds to the process of absorbing a photon and electron momentum forward recoil:

FIG. 2. A schematic diagram showing the light-matter scattering processes of emitting and absorbing a photon from an initial electronphoton distribution $({|c_{p,\nu}^{(0)}|}^2)$ to a final distribution.

 $|p - p_{\text{rec}}^{(a)}, v\rangle \Rightarrow |p, v - 1\rangle$. Finally, the net photon emission and absorption is obtained from

$$
\Delta v_q = \sum_{p,\nu} \left(\left| c_{p,\nu+1}^{(0)} + c_{p,\nu+1}^{(1)(e)} \right|^2 - \left| c_{p,\nu-1}^{(0)} + c_{p,\nu-1}^{(1)(a)} \right|^2 \right). \tag{9}
$$

This can be expressed as the sum of two terms, $\Delta v_a =$ $\Delta v_q^{(1)} + \Delta v_q^{(2)}$, i.e.,

$$
\Delta v_q^{(1)} = 2 \sum_{p,\nu} \text{Re}\left\{ \left(c_{p,\nu+1}^{(0)*} c_{p,\nu+1}^{(1)(e)} \right) - \left(c_{p,\nu-1}^{(0)*} c_{p,\nu-1}^{(1)(a)} \right) \right\}, \quad (10a)
$$

$$
\Delta v_q^{(2)} = \sum_{p,v} \left(\left| c_{p,v+1}^{(1)(e)} \right|^2 - \left| c_{p,v-1}^{(1)(a)} \right|^2 \right). \tag{10b}
$$

Note that we replaced the index $p' \rightarrow p$, $v' \rightarrow v$ for the final momentum and photon distributions. The second photon emission term $\Delta v_q^{(2)}$ is the same as the expression that has been derived in previous QED formulations by free electrons in the plane-wave limit using Fermi's "golden rule" [\[15\]](#page-12-0). The first term $\Delta v_q^{(1)}$ is the contribution from the interference between the initial and scattered states that depends on the features of the initial wave-function distribution. This phasedependent term, that is beyond the conventional Fermi's golden rule [\[38\]](#page-12-0), has not been considered in previous analyses, and is a pivotal *observation* of the present formulation.

Additionally, at this point, it is proper to explain the neglect of the phase-dependent second-order emission and absorption terms $2\text{Re}\{c^{(2)*}(p')c^{(0)}(p')+c^{(1)(e)*}(p')c^{(1)(a)}(p')\}\propto e^{-4\Gamma^2/2}$ and other second-order processes of interference between emission and absorption terms. Their inclusion requires second-order perturbation analysis $(c_{p,\nu}(t)) = c_{p,\nu}^{(0)} + c_{p,\nu}^{(1)} + c_{p,\nu}^{(2)}$

 $c_{p,\nu}^{(2)}$) beyond the present first-order perturbation analysis, which would not affect the main results of the derived firstorder momentum density expressions. They would add small wave-packet-dependent contributions to the second-order momentum density expression, and will produce second-order sidebands of two-photon emission and two-photon absorption processes [\[17,24\]](#page-12-0).

III. RESULTS

In the present analysis, we consider the case where the electron wave function and the radiation field are initially disentangled, $c_{p,\nu}^{(0)} = c_p^{(0)} c_{\nu}^{(0)}$, where the component c_p is only for the electron state and c_v is only for the photon state. Substitution of (7) in (10) then results in

$$
\Delta v_q^{(1)} = \sum_{p,v} \left(\sqrt{\nu + 1} \rho_{p,v}^{(1)(e)} + \sqrt{\nu} \rho_{p,v}^{(1)(a)} \right),
$$

$$
\Delta v_q^{(2)} = \sum_{p,v} \left[(\nu + 1) \rho_{p,v}^{(2)(e)} - \nu \rho_{p,v}^{(2)(a)} \right],
$$
 (11)

where

$$
\rho_{p,\nu}^{(1)(e,a)} = 2\tilde{\Upsilon} \left[\frac{p \pm \left(p_{\text{rec}}^{(e,a)} \mp \hbar q_{zm}/2 \right)}{p_0} \right] \text{sinc}(\overline{\theta}_m^{(e,a)}/2) \times \text{Re}\left\{ \left(c_p^{(0)*} c_{p \pm p_{\text{rec}}^{(e,a)}}^{(0)} \right) \left(c_{\nu \pm 1}^{(0)*} c_{\nu}^{(0)} \right) e^{\pm i(\overline{\theta}_m^{(e,a)}/2 + \phi_0)} \right\},
$$
\n(12a)

$$
\rho_{p,\nu}^{(2)(e,a)} = \tilde{\Upsilon}^2 \left[\frac{p \pm \left(p_{\text{rec}}^{(e,a)} \mp \hbar q_{\text{zm}}/2 \right)}{p_0} \right]^2
$$

$$
\times \text{sinc}^2 (\overline{\theta}_m^{(e,a)}/2) |c_{p \pm p_{\text{rec}}^{(e,a)}}^{(0)}|^2 |c_{\nu}^{(0)}|^2. \tag{12b}
$$

We are set now to examine various cases of interest: (A) spontaneous emission; (B) stimulated emission with quantum light, and particularly with a single Fock state— $c_{\nu}^{(0)} = \delta_{\nu,\nu_0}$; and (C) stimulated emission from a coherent Glauber state.

A. Spontaneous emission

In this case,

$$
c_{\nu}^{(0)} = \delta_{\nu,0},\tag{13}
$$

and we get from the second-order perturbation of Eq. [\(10b\)](#page-2-0) that the only nonzero quantum transition term is the first (emission) term (see Fig. 3), giving the single photon emission from the vacuum state:

$$
\Delta v_{q,SP} = \Delta v_q^{(2)}\big|_{v=0} = \sum_p \left|c_{p,1}^{(1)(e)}\right|^2 = \tilde{\Upsilon}^2 \text{sinc}^2(\overline{\theta}_m^{(e)}/2),\tag{14}
$$

where we approximated at $p_{\text{rec}}^{(e)}/p_0$, $\hbar q_{zm}/p_0 \ll 1$.

Remarkably, in this case, (11) and (12) produce the null result $\Delta v_q^{(1)} = 0$ for the spontaneous emission contribution of the first-order perturbation term. Equation 14 is then the only source of spontaneous photon emission, and therefore, to first-order approximation, there is no wave-packet size or shape dependence of spontaneous emission! In the case of an open structure, such as the Smith-Purcell setup [Fig. $1(a)$],

FIG. 3. Spontaneous emission occurs due to the radiation field of the vacuum state. In the absence of any light sources, there is no process of absorption.

there is a continuum of modes, and the useful parameter is the spontaneous spectral radiant energy emission per unit solid angle per unit frequency:

$$
\left(\frac{dW_q}{d\omega d\Omega}\right)_{SP} = \hbar \omega \rho_{ph}(\omega) \Delta v_{q,SP}
$$

$$
= \frac{e^2 L^2}{64\pi^2} \frac{\omega^2}{c^2} \sqrt{\frac{\mu_0}{\varepsilon_0}} |\eta_{qm}|^2 \sin c^2 \left(\frac{\overline{\theta}_m}{2}\right), \qquad (15)
$$

where we used for the free-space density of modes $\rho_{ph}(\omega)$ = $\omega^2 V / 8\pi^2 c^3$ (suppressed the photon polarization index) [\[15\]](#page-12-0). This expression for the spontaneous emission is consistent with both classical and QED expressions, previously derived in [\[25,15\]](#page-12-0).

B. Stimulated emission—Fock photon state

In this case,

$$
c_{\nu}^{(0)} = \delta_{\nu,\nu_0}.
$$
 (16)

Inspecting (11) and (12) , it appears straightforward that, similarly to the case of spontaneous emission (which is simply the Fock state $v_0 = 0$, there is in general no Fock-statestimulated emission due to the first-order terms, namely, $\Delta v_q^{(1)}|_{v_0} = 0$, because substituting (16) in (12a) results in null terms: $\sqrt{v_0 + 1} (c_{v_0+1}^{(0)*} c_{v_0}^{(0)}) = \sqrt{v_0} (c_{v_0-1}^{(0)*} c_{v_0}^{(0)}) = 0$. There is therefore no linear field stimulated radiative interaction with a Fock-state radiation wave. This is hardly surprising, since a Fock-state wave has no phase.

The second-order terms in (11) do produce spontaneous and stimulated emission that is also wave-packet independent. With the approximation $p_{\text{rec}}^{(e,a)}/p_0$, $\hbar q_{\text{zm}}/p_0 \ll 1$, and the limit of an infinite (plane-wave) electron wave packet $c_p^{(0)}$ = $\delta(p - p_0)$ the momentum integration of (12b) results in

$$
\Delta v_q^{(2)} = \tilde{\Upsilon}^2 \left[(\nu_0 + 1) \text{sinc}^2 (\overline{\theta}_m^{(e)}/2) - \nu_0 \text{sinc}^2 (\overline{\theta}_m^{(a)}/2) \right]. (17)
$$

This result is fully consistent with the previously derived expressions for spontaneous and stimulated emission of FELs and other free-electron radiation schemes in the infinite electron quantum wave-function limit [\[15\]](#page-12-0).

C. Stimulated emission—coherent photon state

A coherent Glauber state represents the classical multiphoton radiation field of a laser beam. In this case the photon state coefficient presentation in terms of Fock states is given by [\[37\]](#page-12-0)

$$
|\sqrt{\nu_0}\rangle = e^{-\nu_0/2} \sum_{\nu=0}^{\infty} \frac{(\nu_0)^{\nu/2}}{\sqrt{\nu!}} |\nu\rangle, \tag{18}
$$

and here v_0 is the expectation value of the photon distribution of the Floquet mode *q* incident on the grating:

$$
\sum_{\nu} \nu \left| c_{\nu}^{(0)} \right|^2 = \nu_0. \tag{19}
$$

In this case, contrary to the Fock case, substitution of Σ (18) into $(12a)$ includes a nonvanishing sum of terms, s) into (12a) includes a nonvanishing sum of
 $\sqrt{v} + 1(c_{v+1}^{(0)}c_v^{(0)}) = \sum_{v} \sqrt{v}(c_{v-1}^{(0)}c_v^{(0)}) = \sqrt{v_0} \neq 0.$

Therefore, the phase-dependent stimulated photon emission of a coherent state $\Delta v_q^{(1)}$ is nonzero. This is consistent with the conclusion of our earlier semiclassical analysis of this problem [\[24\]](#page-12-0), and is fully expected, since the coherent state represents a classical radiation field. The substitution of (18) into [\(11\)](#page-3-0) results in, for this case, the stimulated photon emission contributions:

$$
\Delta v_q^{(1)} = \sum_p \left(\sqrt{\nu_0} \rho_p^{(1)(e)} + \sqrt{\nu_0} \rho_p^{(1)(a)} \right),
$$

\n
$$
\Delta v_q^{(2)} = \sum_p \left[(\nu_0 + 1) \rho_p^{(2)(e)} - \nu_0 \rho_p^{(2)(a)} \right],
$$
 (20)

where

$$
\rho_p^{(1)(e,a)} = 2\tilde{\Upsilon} \left[\frac{p \pm (p_{\text{rec}}^{(e,a)} \mp \hbar q_{zm}/2)}{p_0} \right] \text{sinc}(\overline{\theta}_m^{(e,a)}/2) \times \text{Re}\{(c_p^{(0)*}c_{p \pm p_{\text{rec}}^{(e,a)}}^{\text{(0)}})e^{\pm i(\overline{\theta}_m^{(e,a)}/2 + \phi_0)}\}, \n\rho_p^{(2)(e,a)} = \tilde{\Upsilon}^2 \left[\frac{p \pm (p_{\text{rec}}^{(e,a)} \mp \hbar q_{zm}/2)}{p_0} \right]^2 |c_{p \pm p_{\text{rec}}^{(e,a)}}^{\text{(0)}}|^2. (21)
$$

Noted that the second expression for the stimulated emission is essentially the same for the coherent state and the Fock state, and is given, in the limit of a plane-wave quantum wave function, by the same "FEL gain" [\[15\]](#page-12-0) phase-independent expression [Eq. [\(17\)](#page-3-0)]. The wave-packet case differs only concerning the first-order contribution that is null for a single Fock state but is finite for a coherent state. Our formulas can be extended into more general photon-electron interactions with quantum light, such as the squeezed state or the cat state [\[38,39\]](#page-12-0). Note, though, that the radiation or acceleration with quantum light is still out of experimental capability, since the quantum light sources are too weak at the present state of the art.

IV. DISCUSSIONS

We now apply the formulation to two specific examples of quantum electron wave packets: (a) a single finite size electron wave packet represented by a Gaussian envelope function, and (b) an optically modulated Gaussian envelope wave packet [\[22\]](#page-12-0)

A. Gaussian electron wave packet

We consider stimulated emission with a fixed photon coherent state, interacting with an electron wave packet of Gaussian distribution, chirped after drift length *LD*:

$$
c_p^{(0)} = \left(2\pi\sigma_{p_0}^2\right)^{-1/4} \exp\left[-\frac{(p-p_0)^2}{4\tilde{\sigma}_p^2(t_D)}\right] e^{i(p_0L_D - \varepsilon_0t_D)/\hbar}, \quad (22)
$$

where $\rho_p^2(t_D) = \sigma_{p_0}^2 (1 + i \xi t_D)^{-1}, \xi = 2\sigma_{p_0}^2 / m^* \hbar, L_D =$ v_0t_D . We perform the momentum integration in (20) for this case, using (21) , under the same approximation, $p_{\text{rec}}^{(e,a)}$, $\hbar q_{zm}$, $\sigma_{p_0} \ll p_0$ (see Appendix B and Ref. [\[24\]](#page-12-0)), which results in

$$
\Delta v_q^{(1)} = 2\tilde{\Upsilon}\sqrt{v_0}e^{-\Gamma^2/2}\left\{\operatorname{sinc}\left(\overline{\theta}_m^{(e)}/2\right)\cos\left(\overline{\theta}_m^{(e)}/2+\phi_0\right) + \operatorname{sinc}\left(\overline{\theta}_m^{(a)}/2\right)\cos\left(\overline{\theta}_m^{(a)}/2+\phi_0\right)\right\},\tag{23a}
$$

$$
\Delta v_q^{(2)} = \tilde{\Upsilon}^2 \left\{(v_0 + 1)\operatorname{sinc}^2(\overline{\theta}_m^{(e)}/2) - v_0 \operatorname{sinc}^2(\overline{\theta}_m^{(a)}/2)\right\},\
$$

(23b)

where we defined the extinction parameter,

$$
\Gamma = \left(\frac{\omega}{v_0}\right) \sigma_z(t_D) = \left(\frac{\hbar \omega}{v_0}\right) \frac{\sqrt{1 + \xi^2 t_D^2}}{2\sigma_{p_0}} = \Gamma_0 \sqrt{1 + \xi^2 t_D^2},\tag{24}
$$

with $\Gamma_0 = \frac{2\pi}{\beta}(\frac{\sigma_{z_0}}{\lambda})$. This expression (23) is the main result of this paper. While the phase-independent expression $\Delta v_q^{(2)}$ can be found in the earlier publications $[15,17-19]$, the phasedependent term $(23a)$ is new. In the limit of negligible interaction recoil, $\varepsilon = \delta(\frac{\omega}{v_0})L \ll 1$, $\overline{\theta}_m^{(e)} = \overline{\theta}_m^{(a)} = \overline{\theta}_m$, Eq. (22) reduces to $\Delta v_q^{(1)} = 4 \tilde{\Upsilon} \sqrt{v_0} e^{-\Gamma^2/2} \text{sinc}(\bar{\theta}_m/2) \cos(\bar{\theta}_m/2 + \phi_0)$. Substituting $\sqrt{v_0}E_{qzm} = E_{m,cl}$, where $E_{m,cl}$ is the classical *m*th-order axial slow-wave field component, then one obtains the phase-dependent radiation increment:

$$
\hbar\omega\Delta v_q^{(1)} = (eE_{m,cl}L)e^{-\Gamma^2/2}\text{sinc}\left(\frac{\bar{\theta}_m}{2}\right)\cos\left(\frac{\bar{\theta}_m}{2} + \phi_0\right). \tag{25}
$$

This result restores the semiclassical expression for electron wave-packet acceleration and deceleration that was derived from solution of the Schrödinger equation for the electron [\[24\]](#page-12-0), and confirms the electron-wave energy conservation spectral reciprocity relation $\Delta v_q^{(1)} + \Delta W_e^{acc} / \hbar \omega = 0$ [\[38\]](#page-12-0). It is suggested that measurement of the dependence of Eq. (25) on Γ can be used for evaluating the wave-packet size $\sigma_z(t_D)$ at the entrance to the interaction region.

Einstein relations between the wave-packet-independent stimulated emission and spontaneous emission are directly derived from $(23b)$, as in $[15]$. However, in the case of the wave-packet-dependent expression (23a), the stimulated emission is proportional to $\tilde{\Upsilon}\sqrt{\nu_0}$, while the spontaneous emission expression [\(14\)](#page-3-0) is proportional to $\tilde{\Upsilon}^2$. This suggests another form of "Einstein relation" [\[25\]](#page-12-0) between spontaneous emission and wave-packet-dependent stimulated emission:

$$
\frac{(\Delta v_q^{(1)})^2}{\Delta v_{q,SP}} = 16v_0 e^{-\Gamma^2} \cos^2\left(\frac{\bar{\theta}_m}{2} + \phi_0\right).
$$
 (26)

This universal relation is independent of the interaction structure, because the structure-dependent coefficient Υ cancels out. It can be useful for estimating the wave-packetdependent (and the point particle, $\Gamma \ll 1$) stimulated emissions based on measurement of the spontaneous emission.

Comparison of $(23a)$ to (14) also reveals that while the wave-packet-dependent stimulated emission vanishes in the range $\Gamma(L_D) \gg 1$ (and absolutely so at any drift distance from its source $L_D < z_G = \frac{\beta_0^3 \gamma_0^3}{\pi} \frac{\lambda^2}{\lambda_F}$ [\[24\]](#page-12-0)), the quantum spontaneous emission always exists, independently of Γ . Therefore, observation of classical single point-particle emission and recognizing the transition of wave-packet-dependent stimulated emission from the classical to the quantum limit in the regime $\Gamma \sim 1$, require overcoming a signal-to-noise ratio condition $S/N > 1$, where

$$
S/N \equiv \left(\frac{\Delta v_q^{(1)}}{\Delta v_{q,SP}}\right)\Big|_{\text{max}} = \frac{4\sqrt{v_0}}{\tilde{\Upsilon}}.\tag{27}
$$

Note that the coefficient $\tilde{\Upsilon} = \frac{e\tilde{E}_{qzm}L}{4\hbar\omega}$ is grating-structure dependent. Derivation of explicit expressions for the measurable S/N value is given in Appendix [C.](#page-10-0)

B. Modulated quantum electron wave packet

Now we consider the case where the initial electron state is an optically modulated Gaussian wave packet. Such an electron wave function can be generated by multiphoton emission and absorption from a laser beam of frequency ω_b . After a drift length L_D in dispersive free space, its multiharmonic momentum distribution is chirped [\[22\]](#page-12-0):

$$
c_p^{(0)} = \left(2\pi\sigma_{p_0}^2\right)^{-1/4} \sum_{n=-\infty}^{\infty} J_n(2|g|) \exp\left[-\frac{(p-p_0 - n\delta_p)^2}{4\sigma_{p_0}^2}\right]
$$

$$
\times e^{in\phi_b} e^{-i(p-p_0)^2 t_D/2m^* \hbar} e^{i(p_0 L_D - \varepsilon_0 t_D)/\hbar}, \qquad (28)
$$

 $\int eE_b(z)dz/\hbar\omega$ is the photon exchange energy modulation where ϕ_b is the phase of the modulating laser, $2|g| =$ coefficient at the optical modulation point, and $\delta_p = \hbar \omega_b/v_0$ is the multiphoton emission and absorption electron momentum recoil quantum at this modulation point. The detailed derivation of (28) can be found in Appendix [B](#page-8-0) and $[22,25]$, where it is shown that the expectation value $\rho^{(0)}(t_D) = |C_p^{(0)}(t_D)|^2$ (*t*_D)² is the electron free drift time after the modulation point) represents, for $t_D > 0$, a density-modulated wave packet. After the integration over momentum space in (20) and (21) with (28) (see Appendix \bf{B}), the photon emission is obtained as

$$
\Delta v_q^{(1)} = 2\tilde{\Upsilon}\sqrt{\nu_0} \left\{ \mathrm{sinc}\left(\overline{\theta}_m^{(e)}/2\right) B^{(e)}(\omega) + \mathrm{sinc}\left(\overline{\theta}_m^{(a)}/2\right) B^{(a)}(\omega) \right\},\tag{29a}
$$
\n
$$
\Delta v_q^{(2)} = \tilde{\Upsilon}^2 \left\{ (\nu_0 + 1) \mathrm{sinc}^2 \left(\overline{\theta}_m^{(e)}/2\right) - \nu_0 \mathrm{sinc}^2 \left(\overline{\theta}_m^{(a)}/2\right) \right\}.\tag{29b}
$$

The second-order phase-independent term (29b) turned out to be the same as the case of an unmodulated Gaussian wave packet $(23b)$, where the following sum rule was used, $\sum_{n,k=-\infty}^{\infty} J_n(2|g|)J_k(2|g|) \exp(-\frac{(n-k)^2\delta_p^2}{8\sigma_{p_0}^2})e^{i(n-k)\phi_b}$ 1, which indicates independence of the second-order emission

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of the electron wave-function envelope and its internal distribution. On the other hand, the first term is dependent on both the wave-packet dimension and modulation parameters through the bunching parameter:

 $B^{(e,a)}$

$$
= e^{-\Gamma^{2}/2} \sum_{n,k=-\infty}^{\infty} J_{n}(2|g|)J_{k}(2|g|) e^{-\frac{(n-k)^{2}\delta_{p}^{2}}{8\sigma_{p_{0}}^{2}} + \frac{(n-k)\delta_{p}p_{\text{rec}}^{(0)}}{4\sigma_{p_{0}}^{2}}}
$$

$$
\times \cos\left[\frac{\overline{\theta}_{m}^{(e,a)}}{2} + \phi_{0} - (n-k)\phi_{b} + \frac{(n+k)\delta_{p}p_{\text{rec}}^{(e,a)}t_{D}}{2m^{*}\hbar}\right],
$$
(30)

where we approximate $p_{\text{rec}}^{(e,a)} = p_{\text{rec}}^{(0)} = \hbar \omega / v_0$. In the limit of negligible interaction recoil, $\varepsilon = \delta(\frac{\omega}{v_0})L \ll 1$, $\overline{\theta}_m^{(e)} = \overline{\theta}_m^{(a)} =$ $\overline{\theta}_m$, we can express the phase-dependent term (29a) as

$$
\Delta v_{q,\text{mod}}^{(1)} = \left(\frac{eE_{m,cl}L}{\hbar\omega}\right) \text{sinc}\left(\frac{\bar{\theta}_m}{2}\right)B(\omega),\tag{31}
$$

with

$$
B(\omega) = \frac{B^{(e)} + B^{(a)}}{2} = \sum_{l=-\infty}^{\infty} B_l \exp\left(-\frac{(\omega - l\omega_b)^2 \sigma_l^2(t_D)}{2}\right),\tag{32}
$$

where we changed the summation indices (n, k) to $(n, \ell =$ *n* − *k*), and substituted in the exponents $\delta_p = \hbar \omega_b/v_0$, $p_{\text{rec}}^{(0)} =$ $(n - k)\delta_p \Rightarrow \omega = l\omega_b$. The *l*th-order bunching coefficient is then given by (see Appendix [B\)](#page-8-0)

$$
B_l \approx \sum_{n=-\infty}^{\infty} J_n(2|g|) J_{n-l}(2|g|) \exp\left[-\frac{l^2(\delta_p \xi t_D)^2}{8\sigma_{p_0}^2}\right]
$$

$$
\times \cos\left[\frac{\bar{\theta}_m}{2} + \phi_0 - l\phi_b + \frac{(2n-l)l\delta_p^2 t_D}{2m^* \hbar}\right].
$$
 (33)

It is instructive to affirm that the zero-order harmonic $(l = 0)$ has the same functional dependence as the wave-packet-dependent expression of the unmodu-lated wave packet [\(25\)](#page-4-0) with the same cutoff, $B(\omega)|_{l=0} =$

FIG. 4. The wave-packet-dependent photon emission rate as a function of wave-packet size for unmodulated electron wave packet interacting with a coherent laser beam. Inset: the schematic diagram of a Gaussian wave packet.

FIG. 5. The high-order-harmonics spectrum of stimulated emission by a periodically bunched wave packet, interacting after a drift length corresponding to maximal density bunching. The harmonics are separable, $\omega_b \sigma_t(L_D) = 4 > 1$. Inset: the schematic diagram of a periodically bunched Gaussian wave packet.

 $e^{-\Gamma^2/2}$ sinc $(\frac{\bar{\theta}_m}{2})$ cos($\frac{\bar{\theta}_m}{2} + \phi_0$), but because of the modulation, higher-order-harmonic emission is spectrally possible beyond the cutoff condition $\Gamma > 1$. Also notice that in the absence of modulation ($g = 0$), but also in the absence of drift ($t_D = 0$), Eq. [\(31\)](#page-5-0) reduces to the expression of the unmodulated wavepacket case (25) , which leads to the significant conclusion that *in the absence of density modulation there is no modulationdependent emission.*

The multiharmonic spectrum of the spectral parameter $B(\omega)$ is shown in Fig. 5 for the case of $\omega \sigma_t(t_D) = 4 > 1$, that corresponds to a long wave packet with internal modulation and separable harmonics. The harmonic amplitudes were calculated for a modulation coefficient value $2|g| = 11.4$ and a drift length for which the density modulation ampli-tude is maximal [\[22,25,32–34\]](#page-12-0), $t_{D,\text{max}} = (\pi/\omega_b)/(\Delta p_{\text{max}}/p_0)$, where $\Delta p_{\text{max}} \simeq 2|g|\hbar\omega_b/v_0$. The emission spectrum is shown for maximum acceleration phase for each harmonic. Comparative measurement of the emission spectrum with modulation (Fig. 5) and without (Fig. [4\)](#page-5-0) may be helpful in the measurement of the wave-packet size.

V. CONCLUSIONS

The main results of the present analysis are summarized in Table I for cases of finite size, both unmodulated and modulated quantum electron wave packets. Solving for the interaction of an electron wave packet with quantized radiation, we identified two additive contributions to the photon emission: wave-packet dependent $(\Delta v_q^{(1)})$ and wave-packet independent $\Delta v_q^{(2)}$. The second-order term is consistent with the conventional quantum theory for spontaneous and stimulated emission of free electrons in the infinite (intrinsic) quantum wave-function regime [\[15\]](#page-12-0). The first-order term $\Delta v_q^{(1)}$ is innovative. It predicts wave-packet dependence of stimulated emission when the interacting radiation state is coherent (Glauber) state, consistently and complementarily with earlier predictions of electron wave-packet-dependent acceleration and deceleration, based on semiclassical (electron Schrödinger equation) analysis [\[24\]](#page-12-0). A Fock state interacting wave has null contribution to the first-order term $\Delta v_q^{(1)}$. This includes also the case of the quantum vacuum fluctuation, indicating that the QED-modeled spontaneous emission of a free electron is wave-packet independent in all regimes independently, whether the wave packet is modulated or not.

The main result of this work is the affirmation that the first-order stimulated emission spectrum of a wave packet depends on its size at the entrance to the interaction region, $\sigma_z(t)$. It exhibits an exponential decay scaling [\(25\)](#page-4-0) with a short-wavelength cutoff when $\sigma_z(t_D) \sim \beta_0 \lambda$, corresponding to the transition from the point-particle classical interaction limit to the quantum electron wave-function limit. An even more intricate characteristic of the stimulated emission spectrum takes place when the wave packet is optically

TABLE I. A gallery of phase-dependent and phase-independent photon emission rates of unmodulated and modulated quantum electron wave packets.

PHOTON EMISSIONS $\Delta\nu_q = \Delta\nu_q^{(1)} + \Delta\nu_q^{(2)}$		Gaussian Electron Wavepacket	Modulated Electron Wavepacket
Spontaneous Emission	Vacuum State of Light	$\Delta\nu_a^{(1)}=0$	
		$\Delta\nu_q^{(2)} = \tilde{\Upsilon}^2 \operatorname{sinc}^2\left(\overline{\theta}^{(e)}/2\right)$	
Stimulated Emission	Fock State of Light	$\Delta\nu_a^{(1)}=0$	
		$\Delta\nu_q^{(2)} = \tilde{\Upsilon}^2 \left\{ (\nu_0 + 1) \operatorname{sinc}^2 \left(\overline{\theta}^{(e)} / 2 \right) - \nu_0 \operatorname{sinc}^2 \left(\overline{\theta}^{(a)} / 2 \right) \right\}$	
	State of Light	Coherent $\Delta \nu_q^{(1)} = \left(\frac{eE_{el}L}{\hbar \omega}\right) e^{-\Gamma^2/2} \operatorname{sinc}(\bar{\theta}/2) \cos(\bar{\theta}/2 + \phi_0)$	$\Delta\nu_q^{(1)} = \left(\frac{eE_{cl}L}{\hbar\omega}\right)B(\omega)\,\mathrm{sinc}(\bar{\theta}/2)$
		$\Delta\nu_a^{(2)} = \tilde{\Upsilon}^2 \left\{ (\nu_0 + 1) \operatorname{sinc}^2 \left(\overline{\theta}^{(e)}/2 \right) - \nu_0 \operatorname{sinc}^2 \left(\overline{\theta}^{(a)}/2 \right) \right\}$	

modulated, displaying a wave-packet-dependent harmonic frequencies spectrum beyond the cutoff wavelength (Fig. [4\)](#page-5-0). This observation presents alternative understanding in light-matter interaction: the well-known klystron-type stimulated-superradiant interaction schemes of radiation emission from density-bunched point-particle beams [\[40\]](#page-12-0) are physically possible also with density modulation on the level of the quantum wave function of a single electron. The experiment reported by Priebe *et al.* [\[32\]](#page-12-0) may be a demonstration of such phase-dependent stimulated interaction at second-harmonic bunching frequency.

We assert that measurement of the characteristic stimulated emission spectra of modulated and unmodulated quantum electron wave packets provides a way for evaluating their size and internal features. Such measurement can be done by changing the interaction wavelength λ or the drift length *L_D* = *v*₀*t*_D in the range $\sigma_z(t_D) = \sigma_{z_0} \sqrt{1 + \xi^2 t_D^2} \sim \beta_0 \lambda$, which is attainable at short enough drift lengths away from the source: $L_D < z_G = \frac{\beta_0^3 \gamma_0^3}{\pi} \frac{\lambda^2}{\lambda_c}$ [\[24\]](#page-12-0). We stress, however, that only for simplicity we assumed that the wave-packet size at the entrance to the interaction region is determined by the drift time. Advancement of optical streaking techniques [\[31\]](#page-12-0) would provide full control over the phase, size, and chirp characteristics of the quantum electron wave packet. Hence, the more general conclusion of this work is that the stimulated interaction of a free-electron wave packet is dependent on the history of the electron transport prior to the radiative interaction.

Practical measurement of photon emission and electron energy spectra of single electron radiative interaction is a challenging experiment. It requires accumulating multiple interaction events data with wave-packet preselection of Aharonov's weak measurement scheme [\[41\]](#page-12-0) and using wavepacket shape formation schemes as in [\[31\]](#page-12-0). The detection of the radiation certainly also requires satisfaction of a signalto-noise ratio condition $S/N > 1$, considering the ever-present wave-packet-independent noise due to spontaneous emission.

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APPENDIX A: FIRST-ORDER PERTURBATION THEORY ANALYSIS

Following the standard quantum-electrodynamics theory, we expand the initial wave function in terms of the quantum numbers *p* of the electron state and the Fock photon occupation state of mode *q*, which is given by $|i\rangle = \sum_{p,\nu} c_{p,\nu}^{(0)}(t) |p,\nu\rangle$. For the case of an electron wave packet in our one-dimensional model, the initial wave function is given by

$$
|i\rangle = \int \frac{dp}{\sqrt{2\pi\hbar}} \sum_{\nu} c_{p,\nu}^{(0)} e^{-iE_p t/\hbar} |p,\nu\rangle,\tag{A1}
$$

where the energy dispersion relation of relativistic free electrons is $E_p = c\sqrt{m^2c^2 + p^2}$.

First-order time-dependent perturbation theory of the Schrödinger equation [\[42,43\]](#page-12-0) results in

$$
i\hbar \dot{c}_{p',\nu'}^{(1)} = \int \frac{dp}{\sqrt{2\pi\hbar}} \sum_{\nu} c_{p,\nu}^{(0)} \langle p', \nu'| H_I(t) | p, \nu \rangle e^{-i(E_p - E_{p'})t/\hbar}.
$$
 (A2)

Integrating (A2) in time *t* from 0 to infinity, the emission and absorption process terms of the first-order perturbation coefficient $c_{p',v'}^{(1)} = c_{p',v'}^{(1)(e)} + c_{p',v'}^{(1)(a)}$ are, respectively,

$$
c_{p',\nu'}^{(1)(e,a)} = \frac{\pi}{2i\hbar} \int \frac{dp}{\sqrt{2\pi\hbar}} \sum_{\nu} c_{p,\nu}^{(0)} \langle p', \nu' | H_l^{(e,a)}(0) | p, \nu \rangle \delta\left(\frac{E_p - E_{p'} + \hbar \omega}{2\hbar}\right),\tag{A3}
$$

where $H_I^{(e,a)}$ correspond, respectively, to the second and first terms of the field operator $\hat{\mathbf{A}} = -\frac{1}{2i\omega} [\hat{\mathbf{E}}(\mathbf{r})e^{-i\omega t} - \hat{\mathbf{E}}^{\dagger}(\mathbf{r})e^{i\omega t}]$ used in the interaction Hamiltonian [Eq. (1) in the main text].

The momentum quantum recoil of the electron is found from substitution in $(A3)$ of the energy dispersion relation expanded to second order: $E_p = c\sqrt{m^2c^2 + p^2} \approx \varepsilon_0 + v_0(p - p_0) + \frac{(p - p_0)^2}{2m^*}$. Determined by the delta functions, the emission and absorption recoils are found to be $p_{\text{rec}}^{(e,a)} = p'^{(e,a)} - p_0 = p_{\text{rec}}^{(0)}(1 \pm \delta)$, where $p_0 = \gamma_0 m v_0$, $p_{\text{rec}}^{(0)} = \hbar \omega / v_0$, $\delta = \hbar \omega / 2m^* v_0^2$, and $m^* = \gamma_0^3 m$. Then the first-order perturbation coefficient is given by

$$
c_{p',v'}^{(1)(e,a)} = \frac{\pi}{iv_0} \sum_{v} c_{p' \pm p_{\text{rec}}^{(e,a)},v}^{(0)} \langle p', v'| H_I^{(e,a)}(0) | p' \pm p_{\text{rec}}^{(e,a)}, v \rangle, \tag{A4}
$$

and the general matrix element is given explicitly by

$$
\langle p',\nu'|H_I^{(e,a)}(0)|p,\nu\rangle = \begin{cases} +\frac{e\hbar\tilde{E}_{qsm}}{2\gamma_0 m\omega}\langle\nu'| \hat{a}_q^\dagger|\nu\rangle \int \frac{dz}{2\pi\hbar}e^{-i(q_{zm}z-\phi_0)}e^{-ip'z/\hbar}(\frac{\partial}{\partial z}-iq_{zm}/2)e^{ipz/\hbar} \\ -\frac{e\hbar\tilde{E}_{qsm}}{2\gamma_0 m\omega}\langle\nu'| \hat{a}_q|\nu\rangle \int \frac{dz}{2\pi\hbar}e^{i(q_{zm}z-\phi_0)}e^{-ip'z/\hbar}(\frac{\partial}{\partial z}+iq_{zm}/2)e^{ipz/\hbar} \end{cases} (A5)
$$

After integrating over the spatial space (*z*) and using $\langle v' | \hat{a}^{\dagger}_{q} | v \rangle = \sqrt{v' + 1} \delta_{v', v+1}$, $\langle v' | \hat{a}_{q} | v \rangle = \sqrt{v'} \delta_{v', v-1}$, one obtains

$$
c_{p',\nu'}^{(1)(e,a)} = \begin{cases} +\left(\frac{p' + p_{\text{rec}}^{(e)} - \hbar q_{\text{zm}}/2}{p_0}\right) \tilde{\Upsilon} \sqrt{\nu'} c_{p' + p_{\text{rec}}^{(e)},\nu'-1}^{(0)} \text{sinc}\left(\overline{\theta}_m^{(e)}/2\right) e^{i(\overline{\theta}_m^{(e)}/2 + \phi_0)} \\ -\left(\frac{p' - p_{\text{rec}}^{(a)} + \hbar q_{\text{zm}}/2}{p_0}\right) \tilde{\Upsilon} \sqrt{\nu'} + 1 c_{p' - p_{\text{rec}}^{(a)},\nu'+1}^{(0)} \text{sinc}\left(\overline{\theta}_m^{(a)}/2\right) e^{-i(\overline{\theta}_m^{(a)}/2 + \phi_0)}, \end{cases} \tag{A6}
$$

with the normalized photon exchange coefficient $\tilde{\Upsilon} = \frac{e\tilde{E}_{qzm}I}{4\hbar\omega}$, and

$$
\overline{\theta}_{m}^{(e,a)} = \frac{\left(p_{\text{rec}}^{(e,a)} \pm \hbar q_{\text{zm}}\right)L}{\hbar} = \left(\frac{\omega}{v_0} - q_{\text{zm}}\right)L \pm \left(\frac{\omega}{v_0}\right)L\delta = \overline{\theta}_m \pm \frac{\varepsilon}{2},\tag{A7}
$$

where $\bar{\theta}_m = (\frac{\omega}{v_0} - q_{zm})L$ is the classical interaction detuning parameter, and $\varepsilon = \delta(\frac{\omega}{v_0})L \ll 1$ is the interaction quantum recoil parameter [\[44\]](#page-12-0). Note that we replaced the index $p' \to p$, $v' \to v$ for the final momentum and photon distributions.

APPENDIX B: DERIVATION OF PHOTON EMISSION BY A GAUSSIAN ELECTRON WAVE PACKET AND A MODULATED GAUSSIAN WAVE PACKET

To derive the photon emission expressions for a single quantum electron wave packet [Eq. [\(23\)](#page-4-0) in the main text)], the integration over p in Eqs. [\(11\)](#page-3-0) and [\(12\)](#page-3-0) should be carried out with the Gaussian distribution function of the drifted electron amplitude in momentum space [Eq. [\(22\)](#page-4-0) in the main text]: $c_p^{(0)} = (2\pi\sigma_{p_0}^2)^{-1/4} \exp[-\frac{(p-p_0)^2}{4\tilde{\sigma}_p^2(t_p)}]$ $\frac{(p-p_0)^2}{4\tilde{\sigma}_p^2(t_D)}$]*e*^{*i*(*p*₀*L_D*-ε₀*t_D*)/ \hbar . For the} *phase-independent second-order photon emission* $\Delta v_q^{(2)}$, this involves the following integrations:

$$
\sum_{p} \left\{ \left(\frac{p + p_{\text{rec}}^{(e)} - \hbar q_{\text{zm}}/2}{p_0} \right)^2 |c_{p + p_{\text{rec}}^{(e)}}^{(0)}|^2 \right\}
$$

= $(2\pi \sigma_{p_0}^2)^{-1/2} \int dp \left(\frac{p + p_{\text{rec}}^{(e)} - \hbar q_{\text{zm}}/2}{p_0} \right)^2 \exp \left[-\frac{\left(p + p_{\text{rec}}^{(e)} - p_0 \right)^2}{2\sigma_{p_0}^2} \right] = \left(1 - \frac{\hbar q_{\text{zm}}}{2p_0} \right)^2 + \left(\frac{\sigma_{p_0}}{p_0} \right)^2 \approx 1,$ (B1)

and similarly, for the absorption term,

$$
\sum_{p} \left\{ \left[\frac{p - (p_{\text{rec}}^{(a)} - \hbar q_{\text{zm}}/2)}{p_0} \right]^2 |c_{p - p_{\text{rec}}^{(a)}}^{(0)}|^2 \right\} = \left(1 + \frac{\hbar q_{\text{zm}}}{2p_0} \right)^2 + \left(\frac{\sigma_{p_0}}{p_0} \right)^2 \approx 1. \tag{B2}
$$

For the *phase-dependent first-order photon emission part* $(\Delta v_q^{(1)})$,

$$
\sum_{p} \left\{ \left[\frac{p + (p_{\text{rec}}^{(e)} - \hbar q_{\text{zm}}/2)}{p_0} \right] \text{Re}\left\{ c_p^{(0)*} c_{p + p_{\text{rec}}^{(e)}} \right\} \right\}
$$
\n
$$
= (2\pi \sigma_{p_0}^2)^{-1/2} \int dp \left(\frac{p + p_{\text{rec}}^{(e)} - \hbar q_{\text{zm}}/2}{p_0} \right) \text{Re}\left\{ \exp \left[-\frac{(p - p_0)^2}{4\sigma_{p_0}^2 (1 - i\xi t_D)^{-1}} \right] \exp \left[-\frac{\left(p + p_{\text{rec}}^{(e)} - p_0 \right)^2}{4\sigma_{p_0}^2 (1 + i\xi t_D)^{-1}} \right] \right\}
$$
\n
$$
= e^{-\Gamma^2/2} \left(1 + \frac{p_{\text{rec}}^{(e)} - \hbar q_{\text{zm}}}{2p_0} \right) \approx e^{-\Gamma^2/2},
$$
\n(B3)

and similarly, for the absorption term,

$$
\sum_{p} \left\{ \left[\frac{p - (p_{\text{rec}}^{(a)} - \hbar q_{\text{zm}}/2)}{p_0} \right] (c_p^{(0)*} c_{p - p_{\text{rec}}^{(a)}}^0) \right\} = e^{-\Gamma^2/2} \left(1 - \frac{p_{\text{rec}}^{(e)} - \hbar q_{\text{zm}} + i p_{\text{rec}}^{(e)} \xi t_D}{2p_0} \right) \approx e^{-\Gamma^2/2},\tag{B4}
$$

where we define the decay parameter,

$$
\Gamma = \left(\frac{\omega}{v_0}\right) \sigma_z(t_D) = \left(\frac{\hbar \omega}{v_0}\right) \frac{\sqrt{1 + \xi^2 t_D^2}}{2\sigma_{p_0}} = \Gamma_0 \sqrt{1 + \xi^2 t_D^2} \quad \text{and} \quad \Gamma_0 = \frac{2\pi}{\beta_0} \left(\frac{\sigma_z}{\lambda}\right), \quad \xi = \frac{2\sigma_{p_0}^2}{\gamma_0^3 m \hbar}. \tag{B5}
$$

These result in Eq. [\(23\)](#page-4-0) of the main text. Note that in all cases we took the approximation $p_{\text{rec}}^{(e,a)}$, $\hbar q_z$, $\sigma_{p_0} \ll p_0$ in the last steps of calculation. Also, note that the imaginary part may contribute to an additional phase to the cosine function in the case of very long drift time *tD*.

Similarly, to derive the photon emission of the modulated quantum electron wave packet [Eq. [\(29\)](#page-5-0) in the main text], the integration over *p* in Eqs. [\(11\)](#page-3-0) and [\(12\)](#page-3-0) should be carried out with the modulated Gaussian distribution function of the drifted electron in momentum space:

$$
c_p^{(0)} = \left(2\pi\sigma_{p_0}^2\right)^{-1/4} \sum_{n=-\infty}^{\infty} J_n(2|g|) \exp\left[-\frac{(p-p_0 - n\delta_p)^2}{4\sigma_{p_0}^2}\right] e^{in\phi_b} e^{-i(p-p_0)^2 t_D/2m^* \hbar} e^{i(p_0 L_D - \varepsilon_0 t_D)/\hbar}.\tag{28'}
$$

For the *phase-independent second-order photon emission* $\Delta v_q^{(2)}$, this involves the following integrations:

$$
\sum_{p} \left\{ \left(\frac{p + p_{\text{rec}}^{(e)} - \hbar q_{\text{zm}}/2}{p_0} \right)^2 |c_{p+p_{\text{rec}}^{(e)}}|^2 \right\} \n= \left(2\pi \sigma_{p_0}^2 \right)^{-1/2} \sum_{n,k=-\infty}^{\infty} J_n(2|g|) J_k(2|g|) e^{-i(n-k)\phi_b} \int dp \left(\frac{p + p_{\text{rec}}^{(e)} - \hbar q_{\text{zm}}/2}{p_0} \right)^2 \n\times \exp \left[-\frac{\left(p + p_{\text{rec}}^{(e)} - p_0 - n \delta_p \right)^2}{4\sigma_{p_0}^2} \right] \exp \left[-\frac{\left(p + p_{\text{rec}}^{(e)} - p_0 - k \delta_p \right)^2}{4\sigma_{p_0}^2} \right] \n= \left\{ \left[1 - \frac{(n+k)\delta_p + \hbar q_{\text{zm}}}{2p_0} \right]^2 + \left(\frac{\sigma_{p_0}}{p_0} \right)^2 \right\} \sum_{n,k=-\infty}^{\infty} J_n(2|g|) J_k(2|g|) \exp \left[-\frac{(n-k)^2 \delta_p^2}{8\sigma_{p_0}^2} \right] e^{-i(n-k)\phi_b} \n\approx \sum_{n,k=-\infty}^{\infty} J_n(2|g|) J_k(2|g|) \exp \left[-\frac{(n-k)^2 \delta_p^2}{8\sigma_{p_0}^2} \right] e^{-i(k-k)\phi_b} = 1,
$$
\n(B6)

and similarly,

$$
\sum_{p} \left\{ \left[\frac{p - (p_{\text{rec}}^{(a)} - \hbar q_{\text{zm}}/2)}{p_0} \right]^2 |c_{p - p_{\text{rec}}^{(a)}}^{(0)}|^2 \right\} \approx \sum_{n, k = -\infty}^{\infty} J_n(2|g|) J_k(2|g|) \exp \left[-\frac{(n - k)^2 \delta_p^2}{8\sigma_{p_0}^2} \right] e^{-i(n - k)\phi_b} = 1. \tag{B7}
$$

Here we took the approximation $p_{\text{rec}}^{(e,a)}$, $|n+k|\delta_p$, $\hbar q_{zm}$, $\sigma_{p_0} \ll p_0$ in the last steps of calculation and adopted an identity relation of Bessel functions.

For the derivation of the *phase-dependent first-order photon emission term* of the modulated quantum wave packet $(\Delta v_q^{(1)})$ [Eq. [\(29a\)](#page-5-0) in the main text] we went through the following derivation steps of integration [\(11\)](#page-3-0) and [\(12\)](#page-3-0) with [\(28\)](#page-5-0), using the same approximations:

$$
\sum_{p} \left\{ \left[\frac{p + (p_{\text{rec}}^{(e)} - \hbar q_{\text{zm}}/2)}{p_0} \right] (c_p^{(0)*} c_{p + p_{\text{rec}}^{(e)}}) \right\}
$$
\n
$$
= (2\pi \sigma_{p_0}^2)^{-1/2} \sum_{n,k=-\infty}^{\infty} J_n(2|g|) J_k(2|g|) e^{-i(n-k)\phi_b} \int dp \left(\frac{p + p_{\text{rec}}^{(e)} - \hbar q_{\text{zm}}/2}{p_0} \right)
$$
\n
$$
\times \exp \left[-\frac{(p - p_0 - n\delta_p)^2}{4\sigma_{p_0}^2} - \frac{(p + p_{\text{rec}}^{(e)} - p_0 - k\delta_p)^2}{4\sigma_{p_0}^2} \right] e^{\frac{i(p - p_0 - n\delta_p)^2 p_0}{2m^2 \hbar} - \frac{i(p + p_{\text{rec}}^{(e)} - p_0 - m\delta_p)^2 p_0}{2m^2 \hbar}} \right\}
$$
\n
$$
= \left[1 + \frac{(n + k)\delta_p + p_{\text{rec}}^{(e)} - \hbar q_{\text{zm}} + i p_{\text{rec}}^{(e)} \xi t_D}{2p_0} \right] e^{-\frac{(1 + k^2 t_D^2) p_{\text{rec}}^{(e)}}{8\sigma_{p_0}^2}}
$$
\n
$$
\times \sum_{n,k=-\infty}^{\infty} J_n(2|g|) J_k(2|g|) \exp \left[-\frac{(n - k)^2 \delta_p^2}{8\sigma_{p_0}^2} + \frac{(n - k)\delta_p p_{\text{rec}}^{(e)}}{4\sigma_{p_0}^2} + \frac{i(n + k)\delta_p p_{\text{rec}}^{(e)} t_D}{2m^* \hbar} \right] e^{-i(n - k)\phi_b}
$$
\n
$$
\approx e^{-\frac{(1 + k^2 t_D^2) p_{\text{rec}}^{(e)}}{8\sigma_{p_0}^2}} \sum_{n,k=-\infty}^{\infty} J_n(2|g|) J_k(2|g|) \exp \left[-\frac{(n - k)^2 \delta_p^2}{8\sigma_{p_0}^2} + \frac{(n - k)\delta_p p_{\text{rec}}^{(e)}}{4\sigma_{p_0}^2} + \frac{i(n
$$

and similarly, for the absorption term,

$$
\sum_{p} \left\{ \left[\frac{p - (p_{\text{rec}}^{(a)} - \hbar q_{zm}/2)}{p_0} \right] (c_p^{(0)*} c_{p-p_{\text{rec}}^{(a)}}^{(0)}) \right\} \approx e^{-\frac{(1 + \xi^2 r_B^2) p_{\text{rec}}^{(a)}}{8\sigma_{p_0}^2}} \sum_{n,k=-\infty}^{\infty} J_n(2|g|) J_k(2|g|)
$$

$$
\times \exp \left[-\frac{(n-k)^2 \delta_p^2}{8\sigma_{p_0}^2} - \frac{(n-k) \delta_p p_{\text{rec}}^{(a)}}{4\sigma_{p_0}^2} - \frac{i(n+k) \delta_p p_{\text{rec}}^{(e)} t_p}{2m^* \hbar} \right] e^{-i(n-k)\phi_b}.
$$
 (B9)

We note that the expression for the *second-order photon emission* $\Delta v_q^{(2)}$ [Eq. [\(29b\)](#page-5-0) in the main text], including the expression for spontaneous emission ($v_0 = 0$), comes out identical to the expression for the unmodulated wave packet [\(23b\)](#page-4-0), namely—the modulation, as well as the wave-packet dimension, does not affect the spontaneous emission spectrum at all. Also, note that the expression for the *first-order photon emission* $\Delta v_q^{(1)}$ [Eq. [\(29a\)](#page-5-0)] reduces to the expression of the wave-packet-dependent first-order term of the unmodulated wave packet [Eq. [\(23a\)](#page-4-0)] in the limit of the diminished modulation parameter $2|g| \to 0$, where the identity $e^{-(1+\xi^2 t_D^2)p_{\text{rec}}^{(e)2} / 8\sigma_{p_0}^2} = e^{-\Gamma^2/2}$ recovers the spectral cutoff factor in Eq. [\(23a\)](#page-4-0).

Notice that besides the momentum integration, Eqs. [\(11\)](#page-3-0) and [\(12\)](#page-3-0) involve also summation over the photon occupation states. In the derivation of the *first-order photon emission expression* (that is dependent on both wave-packet size and the modulation parameters), we used the sum-rule expression [Eq. [\(18\)](#page-4-0)], that is valid only for a coherent (Glauber) state of the radiation state (and not valid for quantum light). This results in

$$
\Delta v_q^{(1)} = 2\tilde{\Upsilon}\sqrt{v_0} \left\{ \mathrm{sinc}(\overline{\theta}_m^{(e)})B^{(e)}(\omega) + \mathrm{sinc}(\overline{\theta}_m^{(a)}/2)B^{(a)}(\omega) \right\}, \nB^{(e,a)} = \exp\left[-\frac{\left(1 + \xi^2 t_D^2\right)P_{\text{rec}}^{(0)2}}{8\sigma_{p_0}^2} \right] \sum_{n,k=-\infty}^{\infty} J_n(2|g|)J_k(2|g|) \exp\left[-\frac{(n-k)^2\delta_p^2}{8\sigma_{p_0}^2} + \frac{(n-k)\delta_p p_{\text{rec}}^{(0)}}{4\sigma_{p_0}^2} \right] \times \cos\left[\frac{\overline{\theta}_m^{(e,a)}}{2} + \phi_0 + \frac{(n+k)\delta_p p_{\text{rec}}^{(0)}t_D}{2m^* \hbar} - (n-k)\phi_b \right].
$$
\n(B10)

Using the approximations $p_{\text{rec}}^{(e,a)} = p_{\text{rec}}^{(0)} = \hbar \omega / v_0$, $\bar{\theta}_e = \bar{\theta}_a = \bar{\theta}$ results in Eqs. [\(31\)](#page-5-0) and [\(32\)](#page-5-0) in the main text:

$$
\Delta v_{q, \text{ mod}}^{(1)} = \left(\frac{eE_{m,cl}L}{\hbar\omega}\right) \text{sinc}\left(\frac{\bar{\theta}_{m}}{2}\right)B(\omega), \quad B(\omega) = \frac{B^{(e)} + B^{(a)}}{2}
$$

\n
$$
= e^{-\frac{(1+\xi^{2}i_{D}^{2})p_{\text{rec}}^{(0)}\xi}{8\sigma_{p_{0}}^{2}}} \sum_{n,k=-\infty}^{\infty} J_{n}(2|g|)J_{k}(2|g|) \exp\left[-\frac{(n-k)^{2}\delta_{p}^{2}}{8\sigma_{p_{0}}^{2}} + \frac{(n-k)\delta_{p}p_{\text{rec}}^{(0)}}{4\sigma_{p_{0}}^{2}}\right]
$$

\n
$$
\times \cos\left[\frac{\bar{\theta}}{2} + \phi_{0} + \frac{(n+k)\delta_{p}p_{\text{rec}}^{(e,a)}t_{D}}{2m^{*}\hbar} - (n-k)\phi_{b}\right]
$$

\n
$$
= \sum_{l=-\infty}^{\infty} \left\{\sum_{n=-\infty}^{\infty} J_{n}(2|g|)J_{n-l}(2|g|) \exp\left[-\frac{(p_{\text{rec}}^{(0)} - l\delta_{p} + l\delta_{p})^{2}\xi^{2}t_{D}^{2}}{8\sigma_{p_{0}}^{2}}\right]
$$

\n
$$
\times \exp\left[-\frac{(l\delta_{p} - p_{\text{rec}}^{(0)})^{2}}{8\sigma_{p_{0}}^{2}}\right] \cos\left[\frac{\bar{\theta}}{2} + \phi_{0} + \frac{(2n-l)\delta_{p}p_{\text{rec}}^{(0)}t_{D}}{2m^{*}\hbar} - l\phi_{b}\right]\right\}
$$

\n
$$
= \sum_{l=-\infty}^{\infty} B_{l} \exp\left[-\frac{(p_{\text{rec}}^{(0)} - l\delta_{p})^{2}}{8\sigma_{p_{0}}^{2}(1 + \xi^{2}t_{D}^{2})^{-1}}\right] = \sum_{l=-\infty}^{\infty} B_{l} \exp\left[-\frac{(\omega - l\omega_{b})^{2}\sigma_{t}^{2}(t_{D})}{2}\right], \tag{B11}
$$

with $p_{\text{rec}}^{(0)} = (n - k)\delta_p \Rightarrow \omega = l\omega_b$ and $l = n - k$ being the microbunching harmonic order. The *l*th-order bunching parameter is given by

$$
B_{l} = \sum_{n=-\infty}^{\infty} J_{n}(2|g|) J_{n-l}(2|g|) \exp\left[\frac{-l\delta_{p}(2p_{\text{rec}}^{(0)} - l\delta_{p})\xi^{2}t_{D}^{2}}{8\sigma_{p_{0}}^{2}}\right] \cos\left[\frac{\bar{\theta}_{m}^{(e,a)}}{2} + \phi_{0} + \frac{(2n-l)\delta_{p}p_{\text{rec}}^{(e,a)}t_{D}}{2m^{*}\hbar} - l\phi_{b}\right]
$$

$$
\approx \sum_{n=-\infty}^{\infty} J_{n}(2|g|) J_{n-l}(2|g|) \exp\left[-\frac{l^{2}(\delta_{p}\xi t_{D})^{2}}{8\sigma_{p_{0}}^{2}}\right] \cos\left[\frac{\bar{\theta}_{m}^{(e,a)}}{2} + \phi_{0} + \frac{(2n-l)l\delta_{p}^{2}t_{D}}{2m^{*}\hbar} - l\phi_{b}\right],
$$
(B12)

where $\delta_p = \hbar \omega_b/v_0$. This frequency dependence of the bunching parameter factor in the first-order stimulated emission explains the remarkable resonant radiative "spots" at $\omega = l\omega_b$ in the stimulated emission spectrum (Fig. [5](#page-6-0) in the main text) beyond the frequency cutoff of $l = 0$, reflecting the interior microstructure of the electron wave packet.

APPENDIX C: SIGNAL-TO-NOISE RATIO

A necessary condition for measuring the wave-packet-dependent stimulated emission per electron is addressed here. The phase-dependent term (the 'signal') is given by $[Eq. (25)$ $[Eq. (25)$ in the main text]:

$$
\Delta v_q^{(1)} = 4\tilde{\Upsilon}\sqrt{v_0}e^{-\Gamma^2/2}\text{sinc}\left(\frac{\bar{\theta}_m}{2}\right)\cos\left(\frac{\bar{\theta}_m}{2} + \phi_0\right),\tag{C1}
$$

should be detected under the background of the ever-existing quantum spontaneous emission (the 'noise'), which is the phaseindependent term [Eq. [\(15\)](#page-3-0) in the main text],

$$
\left(\frac{d\nu_q}{d\omega d\Omega}\right)_{\rm SP} = \rho_{ph}(\omega)\Delta\nu_{q,\rm SP} = \frac{e^2 L^2}{64\pi^2} \frac{\omega^2}{c^2} \sqrt{\frac{\mu_0}{\epsilon_0}} |\eta_{qm}|^2 \text{sinc}^2 \left(\frac{\overline{\theta}_m}{2}\right). \tag{C2}
$$

Then, the satisfication of a signal-to-noise ratio condition $S/N > 1$ requires:

$$
S/N = \Delta v_q^{(1)} / \left(\frac{d v_q}{d \omega d \Omega}\right)_{SP} \Delta \Omega_{\text{det}} \Delta \omega_{\text{det}},\tag{C3}
$$

where $\Delta\Omega_{\text{det}}$, $\Delta\omega_{\text{det}}$ are, respectively, the solid angle acceptance and the frequency bandwidth of the detection system.

Alternatively, we will present here a derivation of the S/N parameter by calculating from first principles the spontaneous emission per single mode corresponding to the Floquet radiation mode incident on the grating in the stimulated emission problem. In this case, we generalize the quantization procedure, such that

$$
\frac{1}{2}\sqrt{\frac{\varepsilon_0}{\mu_0}}|\tilde{E}_{q\perp 0}|^2 t_r = \hbar\omega,\tag{C4}
$$

where $t_r = L/v_0$ is the interaction time along the grating. For this model we get

$$
\tilde{\Upsilon} = \frac{\tilde{E}_{qzm}L}{4\hbar\omega} = \left\{ \frac{1}{8} \sqrt{\frac{\mu_0}{\varepsilon_0}} |\tilde{\eta}_{qm}|^2 \frac{e^2 L^2}{A_{\text{eff},q}t_r} / \hbar\omega \right\}^{1/2},\tag{C5}
$$

with $t_r = L/\beta_0 c$, and substituting for the mode *q* (e.g., a Gaussian mode incident on the grating at angle Θ), $A_{\text{eff},q}$ $L_w w_{\text{eff},q} \cos(\bar{\Theta}/2)$, this simplifies into

$$
\tilde{\Upsilon} = \left\{ \frac{1}{8} |\tilde{\eta}_{qm}|^2 \frac{e^2 \beta_0}{\varepsilon_0 w_{\text{eff},q}} / \hbar \omega \right\}^{1/2}.
$$
 (C6)

This expression can be used to calculate the spontaneous emission per mode [Eq. [\(14\)](#page-3-0) in the main text],

$$
\Delta v_{q,SP} = \tilde{\Upsilon}^2 \text{sinc}^2(\overline{\theta}_m/2),\tag{C7}
$$

and then, with the expression for stimulated emission [\(C1\)](#page-10-0), evaluate explicitly the maximal signal-to-noise ratio for an isolated single mode detection [Eq. (27) in the main text]:

$$
S/N \equiv \left(\frac{\Delta v_q^{(1)}}{\Delta v_{q,SP}}\right)\Big|_{\text{max}} = \frac{4\sqrt{v_0}}{\tilde{\Upsilon}}.\tag{C8}
$$

This equation indicates that the wave-packet-dependent stimulated emission can be detected $(S/N > 1)$ in a Smith-Purcell experiment with a sufficiently intense incident laser pulse: $v_0 > \tilde{\Upsilon}^2/16$.

Note that the same result can be obtained also by substituting $\Delta \omega_{\text{det}} = 2\pi/t_r$, $\Delta \Omega_{\text{det}} = (\lambda/2A_{\text{eff},q})^2$ in Eq. (C3) for an optical detection system optimally matched to measure the signal and spontaneous emission from a diffraction limited single mode.

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