Control of atom-photon interactions with shaped quantum electron wavepackets

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Abstract Photon emission from atoms and free electrons underlie a wealth of fundamental science and technological innovations. We present a regime where atom-photon and electron-photon interactions interfere with each other, resulting in substantial changes in the spontaneous emission rate compared to the sum of each interaction considered in isolation. We highlight the critical role played by quantum electron wavepackets, and how the emission can be tailored via the electron wave shape, as well as the atomic population and coherence. Our findings reveal that atom-photon and electron-photon interactions cannot be considered in isolation even when higher-order contributions involving all three bodies (atom, photon and free electron) are negligible. Our findings pave the way to more precise control over photon emission processes and related diagnostics.

Introduction Atom-photon interactions encompass many fundamental phenomena including stimulated and spontaneous emission [113], Rabi oscillations [4] [5], Dicke superradiance [6,9], and high-harmonic generation [10–12]. Such effects form the basis of many fields including quantum metrology [13–17], quantum information technologies [18–20], quantum integrated circuits [21–31], optical switching [32–35], and photon generation and manipulation [28–39] [45]. Innovative platforms for atom-photon interactions continue to arise in the form of quantum dots [46–51], 2D materials [52–55], superconducting qubits [56], crystal vacancy centers and defects [57–60], and cathodoluminescence [61]. These effects have fueled the development of light-matter interactions.

At the same time, electron-photon interactions comprise many intriguing effects including the Kapitza-Dirac effect [65–68], Smith-Purcell radiation [69–71], Cerenkov radiation [72–75], transition radiation [76–77], Bremsstrahlung [78–80], and Compton scattering [81,82]. These effects have fueled the development of light sources [83–88], particle accelerators [89–93] and detectors [94–99], and medical devices. Electron-photon interactions form the basis of innovative diagnostic tools including electron energy-loss spectroscopy (EELS) [100–109] and its variants [101–109], cathodoluminescence (CL) [101–109,110], and photon-induced near-field electron microscopy (PINEM) [107–109,112–120]. These tools have enabled the study of the most fleeting and minute excitations in both light and matter. Recent works have shown that photons can shape quantum electron wavefunctions [111–114,116–117,121–135], and that shaped electrons can in turn tailor free electron stimulated and spontaneous emission [111,136,147].

While atom-photon and electron-photon interactions are individually well studied, they are typically treated as independent processes whose cross-sections can be added incoherently.

Contrary to this notion, we present here a regime where electron-photon and atom-photon interactions interfere significantly with each other, resulting in strong modulation of the total spontaneous emission (SE) rate with interaction length (Figs. 1(a),(b)). We show that this interference is facilitated, and can be engineered, by shaped quantum electron wavepackets (QEWs). The SE rate can be significantly enhanced or suppressed compared to the sum of atomic SE and free electron SE considered in isolation – an effect absent when unshaped (Gaussian) QEWs are used (Fig. 1(c),(d)). Importantly, the shape of the QEW can be used to control the total SE rate. Our results show that maximum SE enhancement or suppression can be achieved over a wide range of electron kinetic energies (e.g., 100 eV to 1 MeV) and emission frequencies (e.g., optical to terahertz), within interaction lengths of ~10^{-7} m to ~10^{-4} m. Our findings pave the way to more precise control over photon emission processes, and motivate the development of advanced QEW shaping techniques for on-demand tailoring of light-matter interactions.

Results Consider a QEW of velocity \( \mathbf{v} = v_0 \hat{z} \), modulation frequency \( \omega_{\text{mod}} \), and bunching factor \( \langle b \rangle = \langle |b| e^{i\phi_b} \rangle \) passing through an electromagnetic (EM) environment (e.g., cavity, waveguide) of length \( L \), which is also the interaction length. The EM environment contains an atom, modeled as a two-level system. To first order in perturbation theory (i.e., weak coupling regime), we find that the single-photon SE rate arising from the interference between the atom-photon and electron-photon interactions is given by (details in Supplemental Material (SM) Section I)

\[
\Gamma_{\text{al/el}} = \frac{\tau}{\hbar} \frac{e \epsilon_0 \hbar V}{\epsilon_0} |d| \rho_{\text{eg}} \langle b \rangle |\cos(\xi)\sin\left(\frac{\omega_{\text{cav}} - \omega_a}{2}\right) \times \left[\sin\left(\frac{\delta \omega_{\text{cav}} - \omega_{\text{mod}}}{2}\right) \sin\left(\frac{\omega_{\text{cav}} - \omega_{\text{mod}}}{2}\right)\right]\right],
\]

where the atom located at \( \mathbf{r} = (0, 0, z_a) \) with a bandgap corresponding to angular frequency \( \omega_a \). The EM environment supports a single dominant longitudinal field
mode of angular frequency \( \omega_{\text{cav}} \) and wavevector \( q = (0,0,\omega_{\text{cav}}/c) \), where \( c \) is the free-space speed of light. Such a mode is realizable, for instance, using a racetrack system within an electromagnetic (EM) environment separately emit photons at rates of \( \Gamma_{\text{al}} \) and \( \Gamma_{\text{el}} \) respectively. Interference between these distinct quantum processes (dotted line) results in a third emission process at rate \( \Gamma_{\text{al/el}} \), which can significantly enhance or suppress the total SE rate. (b) For a 30 keV modulated QEW of bunching factor \( b = 0.99 \), the total SE rate is strongly modulated as a function of interaction length \( L \). In contrast, for an unmodulated Gaussian QEW (c), quantum interference is absent, resulting in the unmodulated emission rate profile (d). The two-level system here (Sn-N vacancy) has emission angular frequency \( \omega_a = 2\pi c/(620 \text{ nm}) \approx 3 \times 10^{15} \text{ rad/s} \) and transition dipole moment of \( d = (4.33 \times 10^{-29}) \hat{z} \text{ Cm} \) aligned parallel to the field, with equal population initially in the excited and ground states. Here, \( \Psi_b = 0 \), \( z_a = 0 \), and \( \phi_{ea} = (\pi/2) + (\omega_{\text{cav}} z_a/c) \).

Figure 1(b) shows that a modulated QEW can lead to significant enhancement or suppression of SE, depending on interaction length \( L \) and cavity frequency \( \omega_{\text{cav}} \), compared to the incoherent sum of isolated SE events \( \Gamma_{\text{al}} + \Gamma_{\text{el}} \). The latter scenario is shown in Fig. 1(d), which also corresponds to the total SE for an incoming unmodulated Gaussian QEW. In Fig. 1(b) we consider a modulated QEW of central kinetic energy \( E_K = 30 \text{ keV} \) and bunching factor \( b \approx 0.99 \), which was recently shown to be feasible using multiple electron-lensing stages [122]. The two-level system we consider is a tin-vacancy (SnV) center [148] of emission frequency \( \omega_b \approx 3 \times 10^{15} \text{ rad/s} \) (corresponding to wavelength 620 nm) and transition dipole moment \( d = 2.33 \times 10^{-29} \text{ Cm} \) (aligned parallel to the field). The initial excited and ground state populations are equal, corresponding to atomic coherence magnitude \( |\rho_{eg}^a| = 1/2 \). We set \( \phi_a = (\omega_{\text{cav}} z_a/c) \) as a result of the sinc term in the first line of Eq. 1, which approaches a delta function (when multiplied by \( \tau \)) that enforces energy conservation at long interaction times. We consider the resonant case for the rest of this work.

![FIG. 1. Tailoring the spontaneous emission (SE) rate through quantum interference between atom-photon and electron-photon interactions, facilitated by shaped quantum electron wavepackets (QEW). (a) Modulated QEWs and two-level systems within an electromagnetic (EM) environment separately emit photons at rates of \( \Gamma_{\text{al}} \) and \( \Gamma_{\text{el}} \) respectively. Interference between these distinct quantum processes (dotted line) results in a third emission process at rate \( \Gamma_{\text{al/el}} \), which can significantly enhance or suppress the total SE rate. (b) For a 30 keV modulated QEW of bunching factor \( b = 0.99 \), the total SE rate is strongly modulated as a function of interaction length \( L \). In contrast, for an unmodulated Gaussian QEW (c), quantum interference is absent, resulting in the unmodulated emission rate profile (d). The two-level system here (Sn-N vacancy) has emission angular frequency \( \omega_a = 2\pi c/(620 \text{ nm}) \approx 3 \times 10^{15} \text{ rad/s} \) and transition dipole moment of \( d = (4.33 \times 10^{-29}) \hat{z} \text{ Cm} \) aligned parallel to the field, with equal population initially in the excited and ground states. Here, \( \Psi_b = 0 \), \( z_a = 0 \), and \( \phi_{ea} = (\pi/2) + (\omega_{\text{cav}} z_a/c) \).](image)

To quantify the relative contribution of the interference term \( \Gamma_{\text{al/el}} \), we define the modulation factor

\[
\gamma = \frac{\Gamma_{\text{al/el}}}{\Gamma_{\text{al}} + \Gamma_{\text{el}}} = \frac{2|\rho_{eg}^a||\langle b\rangle|\cos(\xi)\text{sinc}(\delta\beta\omega_a\tau/2)}{4\Lambda \frac{1-\text{sinc}(\delta\beta\omega_a\tau)}{(\delta\beta\omega_a\tau)^2} + (\rho_{ee}^a/\Lambda)},
\]

where \( \delta_\beta = |1 - \beta_0| \), \( \Lambda = e\nu_0/(\omega_0 |d|) \) measures the strength of the electron-photon interaction relative to that of the atom-photon interaction, and \( \rho_{ee}^a \) is the initial excited atomic state population. Figure 2 presents the dependence of \( |\gamma_{\text{max}}| = \text{the maximum possible } \gamma \text{ across all } \tau \) – on the QEW shape as determined by the bunching factor \( b \). We see that \( |\gamma_{\text{max}}| \) grows linearly with \(|\langle b\rangle|\).
Our results also indicate that strong SE enhancement and suppression can be attained for atomic emission frequencies from the optical to the terahertz regime. Figure 3(a) shows that the corresponding $L_{\text{opt}}$ ranging from tens of nm to hundreds of $\mu$m, which correspond to experimentally realizable waveguide/cavity dimensions in the optical to terahertz regimes [149-152]. An approximate analytical expression for $L_{\text{opt}}$ is presented in SM Section III. As we see in Figs. 3(a),(b), the modulation factor varies in an oscillatory pattern with $L$, with a central spatial period of $\lambda_{\text{SE}} = 4\pi\nu_0/\delta_3\omega_a$. The white region in Fig. 3(a) denotes combinations of QEW $E_K$ and $\omega_a$ that lie within the strong coupling regime, which is outside the scope of this work. Although we have focused on SnV as an example, our conclusions remain qualitatively unchanged even when we consider other systems (SM Section IV), as long as the transition dipole moment falls within the range $\sim 10^{-31}$ Cm to $\sim 10^{-29}$ Cm.

The modulation factor $\gamma$ can also be controlled via the initial excited state population and coherence of the atomic system. As Fig. 3 shows, a larger $L$ generally favors a smaller initial excited state population for maximal $|\gamma|$. This is more clearly depicted when we plot the SE modulation strength $|\gamma|$ using the Bloch sphere representation (polar colormaps, inset). Here, $\theta_a$ (radial coordinate) and $\phi_a$ (angular coordinate) are related to the excited state population $\rho_{ee}$ and coherence $\rho_{eg}$ through the relations $\rho_{ee} = \cos^2(\theta_a/2)$ and $\rho_{eg} = (1/2)e^{i\phi_a} \sin \theta_a$ respectively. For longer $L$, the peaks corresponding to $\max(|\gamma|)$ shift towards $\theta_a = \pi$, which corresponds to the ground state. As can also be seen from Eq. (2), the bunching phase $\Psi_b$ can be used to azimuthally rotate the profile of $\gamma$ on the atomic Bloch sphere.

**Discussion** Our work shows that atom-photon and electron-photon interactions cannot always be treated as separate processes and summed incoherently, even when we can ignore higher-order processes involving all three bodies (atom, electron and photon). Instead, our results reveal that first-order atom-photon and electron-photon interactions can interfere with each other, resulting in a total SE rate substantially different from sum of SE rates due to each interaction alone. It is noteworthy that the interference can be significant even when the atom and QEW are arbitrarily far apart, so long as they are each interacting with the same photon mode. This is in sharp contrast to the Coulomb interaction between the atomic system and the QEW, which relies on the proximity between the atom and the QEW, and which has been leveraged in recent works [155,157] to encode information on atomic coherence and dephasing in electron spectra. In this regard, our work could provide a route towards free-electron quantum metrology without the requirement of the atomic system and QEW being physically near each other.

Our results also indicate that strong SE enhancement and suppression can be attained for atomic emission frequencies from the optical to the terahertz regime. This can potentially be used to tailor and probe emitters...
FIG. 4. Dependence of quantum interference contribution on initial excited atomic state population. The optimal excited state population decreases with increasing interaction length $L$ (solid red curve), revealing that smaller initial excited state populations favor larger $|\gamma|$ at longer interaction lengths $L$. The polar colormaps show the value of $|\gamma|$ on the Bloch sphere representing the initial atomic state, at various values of $L$. As illustrated for the case of $L = 68.66 \mu m$, the location of the maximal $|\gamma|$ on the Bloch sphere can be manipulated by varying the bunching factor phase $\Psi_b$. For the polar plots, the azimuthal angle $\phi_a$ is the phase of the coherent state and the polar angle $\theta_a$ (radial coordinate) is such that cos $^2 (\theta_a/2)$ is the excited state population. Here, we use the same parameters for the atomic state as Figs. 1 and 2 and consider a 30 keV modulated QEW with $\langle b \rangle = 0.58$.

in a wide variety of systems, such as superconducting qubits [162], quantum dots [46–54], and crystal and vacancy centers and defects in crystals [52, 53, 60–64] – capabilities that are increasingly sought-after in the development of quantum information technologies [18–45]. Additionally, the electron energy can be used as a degree of freedom to simultaneously tune $L_{opt}$ and $\lambda_{SE}$ for a given emission frequency, granting a degree of flexibility in cavity/waveguide designs.

Our findings are highly complementary to recent works exploring the use of shaped QEWs to tailor quantum mechanical and radiative processes. These include the use of shaped QEWs to shape spontaneous and stimulated emission from free electrons [138, 140–142, 147] and to realize free-electron-bound-electron-resonant interaction (FEBERI) [158, 159]. The combination of modulated electron wavepackets and synchronized external light was recently shown to suppress the resulting cathodoluminescence in a way that can be useful for ultrafast electron microscopy [111]. The physics we study here is completely different from all the above, as this work concerns interference between atomic and electron SE processes, where the total emission can be either enhanced or suppressed compared to the incoherent sum of the individual SE processes. Our theory can be readily extended to alternative configurations such as one where the QEW travels at an angle (instead of parallel, as in this work) to the photon mode of the EM environment. Additionally, our framework supports the study of stimulated emission and the possibility of entanglement among the input states, opening up a rich field of exploration. Our findings also suggest exciting prospects for applying QEWs that go beyond controlling photon emission, such as interference between electron-photon and electron-atom interactions for manipulation of electron wavepackets, and interference between electron-atom and atom-photon interactions for manipulation of atomic population and coherence.

Conclusion In summary, we present a regime of light-matter interaction where interference between electron-photon and atom-photon interactions can lead to strong enhancement or suppression of the total SE rate. This interference is facilitated by modulated QEWs, which provide a means of tailoring the total SE through their shape – specifically, the magnitude and phase of bunching factor $\langle b \rangle$. The sensitivity of the total SE to the coherence and population of the initial atom also makes this a promising way of measuring the atomic state using first-order processes. Furthermore, unlike electron-atom interactions, the interference can be strong even if the electron and atom are not physically near each other. Thanks to the interference, the magnitude of the total SE varies significantly with a relatively small change in the interaction length. With rapid advances in electron waveshaping techniques, our findings open the doors to unprecedented control over photon emission processes and related diagnostics.

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