Resonant Interaction of Modulation-correlated Quantum Electron Wavepackets with Bound Electron States

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Free-Electron Bound-Electron Resonant Interaction (FEBERI) is the resonant inelastic interaction of periodically density-bunched free electrons with a quantum two level system. We present a comprehensive relativistic quantum mechanical theory for this interaction in a model in which the electrons are represented as quantum electron wavepackets (QEW). The analysis reveals the wave-particle duality nature of the QEW, delineating the point-particle-like and wave-like interaction regimes, and manifesting the physical reality of the wavefunction dimensions and its density modulation characteristics in interaction with matter. The analysis comprehends the case of laser-beam-modulated multiple QEWs that are modulation-phase correlated. Based on the Born interpretation of the electron wavefunction we predict quantum transitions enhancement proportional to the number of electrons squared, analogous to superradiance.

We present here a comprehensive quantum model for the recently proposed new concept of Free-Electron-Bound-Electron Resonant Interaction (FEBERI) [1]. In this process, a probability-density modulated Quantum Electron Wavepacket (QEW) interacts resonantly with a Two-Level System (TLS) when its optical frequency modulation matches the TLS quantum energy level transitions:

\[ n\hbar \omega_b = E_{2,1} \]  \hspace{1cm} (1)

where \( \omega_b \) is the periodic temporal modulation frequency of the QEW density distribution as seen by a stationary observer, equal to the laser beam frequency that modulates it. \( n\omega_b \) is an harmonic frequency of the QEW periodic bunching and \( E_{2,1} = E_2 - E_1 \) is the quantum energy gap of the TLS.
Optical frequency modulation of single quantum electron wavepackets is a direct outcome of the process of Photon-Induced Near-Field Electron Microscopy (PINEM) [2-12]. In this process, the energy spectrum of single Quantum Electron Wavepackets (QEW) is modulated at optical frequencies by interaction with a laser beam of frequency $\omega_b$, exhibiting discrete energy sidebands $\Delta E_n = n\hbar\omega_b$. The interaction is made possible by a multiphoton emission/absorption process in the near field of a nanostructure [8, 10], a foil [9,7] or a laser-beat (pondermotive potential) [11, 12]. It was also shown [8,9] that due to the nonlinear energy dispersion of electrons in free space drift, the discrete energy modulation of the QEW is converted into tight periodic density modulation (bunching) at atto-second short levels, corresponding to high spectral harmonics contents $\nu_n = n\omega_b$ in the expectation value of the QEW density:

$$n(r,t) = \left| \Psi(r,t) \right|^2$$ (2)

The interpretation of the quantum electron wavefunction (QEW) has been a matter of debate since the inception of quantum theory [13, 14]. The accepted Born interpretation of the quantum electron wavefunction is that the expectation value (2) represents the probability density of finding the electron at point $r$ at time $t$. The semiclassical interpretation of $-e\left| \Psi(r,t) \right|^2$ as charge density may have limited validity in situations where it is possible to take an ensemble average over multiple electrons [15]. The reality of the QEW and the measurability of its dimensions, and the transition from the quantum wavefunction presentation to the classical point-particle theory (the wave-particle duality) were considered recently in the context of radiative interaction of single electron QEWs with light near polarizable structures, such as in Smith-Purcell radiation [16-18]. Semiclassical analysis of such stimulated interaction (under external radiation field) reveals the transition from the quantum electron wavefunction interaction regime that is characterized by the multi-sidebands ($\Delta E_n = n\hbar\omega_b$) electron energy spectrum of PINEM [2-12] to the classical point-particle-like acceleration\deceleration regime. This transition takes place when

$$\Gamma = \omega\sigma_{\text{el}} = 2\pi\sigma_{\text{ex}}/\beta_0\lambda < 1$$ (3)

where $\beta_0 = v_0/c$ is the centroid velocity of the QEW [16, 45]. Namely, the transition takes place when the wavepacket duration $\sigma_{\text{el}}$ or its length $\sigma_{\text{ex}}$ are short relative to the optical radiation period $2\pi/\omega$ or wavelength $\lambda$ respectively. This result, indicating the reality and measurability of the QEW dimensions in interaction with light has been confirmed also by a QED theory analysis [19].

Furthermore, both semiclassical and QED model analyses [18,19] suggest that stimulated interaction of radiation with modulated QEWs are sensitive also to modulation features of the QEW. So, the density bunching of the QEW after a PINEM interaction and a subsequent free drift step, is measurable by interaction with a second synchronous laser beam. The physical reality of the periodic sculpting of the QEW in the time and space (propagation coordinate – $z$) dimensions, has been demonstrated recently experimentally by interaction
with a second laser beam, phase-locked to the bunching frequency $\omega_b$ or its harmonic [9,10,20,21].

The physical reality of the wavepacket dimensions and its modulation features in the case of spontaneous radiative interaction in different schemes of QEWR interactions with radiation is still being studied, and is subject of controversies [22-24]. QED analyses of spontaneous Smith-Purcell radiative emission by a single QEWR disaffirms dependence on the transverse dimensions of the QEWR [24] and on its longitudinal dimension and modulation features [19]. However, the case may be different when multiple electrons are considered. In the classical point-particle regime, a multiple particles beam exhibits an effect of spontaneous superradiance (in the sense of Dicke [25]) when bunched within less than an optical wavelength [26]. Namely, they emit coherent radiation proportional to the square of the number of particles - $N^2$. One would expect that a bunch of identical QEWRs that satisfy condition (3) would likewise exhibit similar $N^2$ scaling [18], and that it would turn into the N scaling dependence of “shot-noise radiation” when the condition is not satisfied. This establishes a dependence of the coherent superradiance emission on the QEWRs dimension in the case of multiple QEWRs. Furthermore, as reviewed in [27], periodically bunched point-particle electron beams emit superradiantly at the frequency and harmonics of their density modulation frequency with the same quadratic scaling. Based on the Born probability interpretation, one would expect that similar $N^2$ scaling of superradiant emission will take place also with a multi-QEWRs beam when the density expectation value of their individual wavefunctions is modulated at optical frequency, under the condition that their modulation phases are correlated. Such a correlated QEWRs beam can be generated in the PINEM interaction process if all QEWRs are exposed at the interaction point to the near field of the same coherent laser beam as in [8]. In this case of multiple particles, a semiclassical model is valid [18] and predicts the same quadratic scaling of superradiant emission by bunched QEWRs.

In the FEBERI concept of Ref. 1 the idea of the reality of the QEWR modulation features is implemented in the case of interaction with matter. Ref. 1 used a simple semiclassical model in order to bring to light the feasibility of the FEBERI effect. In this model the modulated QEWR probability density is taken to represent periodic space-charge distribution of the QEWR. Such a semiclassical model is questionable in the case of single electrons, but is expected to have partial validity in appropriate limits of multiple correlated QEWRs. Nevertheless, a more complete quantum mechanical theory analysis is called for, in order to affirm the feasibility of the effect and the limits of its validity. Furthermore, the interaction with multiple QEWRs involves their entanglement with the bound electron (TLS) states and their entanglement with each other. In a future publication we expect to refer to this additional interesting feature of the multi-particle FEBERI interaction. In the present publication we limit our scope to formulation of the FEBERI problem in terms of a complete quantum mechanical theory model, and to identifying the operating regimes where the semiclassical model [1] is valid.
1. **Quantum Formulation of FEBERI**

![Fig. 1 A Two-Level System (TLS) quantum interaction model of Quantum Electron Wavepacket interaction with a bound electron.](image)

Our comprehensive quantum mechanical theory model is based on the setup shown in Fig. 1, showing a thin free electron QEW propagating in proximity to a TLS that is modeled as a Hydrogen-like atom. For simplicity we assume that the interaction of the free and bound electrons is only through Coulomb interaction. We start with a Schrödinger equation for the combined wavefunction of the free and bound electrons:

\[
i \hbar \frac{\partial \Psi(r, r', t)}{\partial t} = (H_0 + H_I) \Psi(r, r', t) \tag{4}
\]

\[
H_0 = H_{0F} + H_{0B} \tag{5}
\]

where \(H_{0F}, H_{0B}\) are the Hamiltonians of the free and bound electrons respectively, and \(H_I\) is the interaction Hamiltonian. In order to apply the analysis also to relativistic electrons, we use the “relativistic” Schrödinger equation Hamiltonian for a free electron of energy \(E_0 = \gamma_0 mc^2\) and momentum \(p_0 = \gamma_0 \text{m}\vec{v}_0\):

\[
H_{0F}(r) = E_0 + \vec{v}_0 \cdot (-i\hbar \nabla - \vec{p}_0) + \frac{1}{2\gamma_0^3 m}(-i\hbar \nabla - \vec{p}_0)^2 \tag{6}
\]

This Hamiltonian was derived in Ref. [28] and the Supplementary of [16] by a second order iterative approximation of Klein-Gordon equation, and therefore does not include spin effects. It has been derived recently also directly from Dirac equation [29] without the quadratic term that is needed here to account for electron wavepacket chirp and size expansion in free drift. It corresponds to second order expansion of the relativistic energy dispersion of a free electron:

\[
E_p = E_0 + \vec{v}_0 \cdot (\vec{p} - \vec{p}_0) + \frac{1}{2\gamma_0^3 m}(\vec{p} - \vec{p}_0)^2 \tag{7}
\]

The eigenfunction solutions of the bound electron Hamiltonian \(H_{0B}\) are modelled as the solutions of a two-level system (TLS):

\[
H_{0B} \psi_j(r', t) = E_j \psi_j(r', t) \quad (j=1,2) \tag{8}
\]
The wavefunction solution of the free electron in zero order is taken to be a wavepacket:

$$\Psi_j (r', t) = \varphi_j (r') e^{-iE_j t / \hbar}$$  \hspace{1cm} (9)$$

$$\Psi_B (r', t) = \sum_{j=1}^{2} C_j \Psi_j (r', t)$$  \hspace{1cm} (10)$$

The wavefunction solution of the free electron in zero order is taken to be a wavepacket:

$$\Psi_F^{(0)} (z, t) = \frac{dp}{\sqrt{2\pi \hbar}} c_p^{(0)} e^{-iE_p t / \hbar} e^{i\varphi_p / \hbar}$$ \hspace{1cm} (11)$$

For a Gaussian wavepacket [16]:

$$c_p^{(0)} (t_0) = \frac{1}{(2\pi \sigma_{p0}^2)^{1/4}} e^{-\left( p - p_{0} \right)^2 / 4\sigma_{p0}^2} e^{i(p_{0}t_0 - E_{p0}) / \hbar}$$ \hspace{1cm} (12)$$

and in space coordinates:

$$\Psi_F^{(0)} (z, t) = \frac{\sqrt{\sigma_{z0}}}{(2\pi \sigma_{z0}^2 (t + t_D) )^{1/4}} \exp \left\{ -\frac{(z - v_0 (t - t_0))^2}{4\sigma_{z0}^2 (t + t_D)} \right\} e^{i(p_{0}(z + t_0) - E_{p0} (t + t_0))) / \hbar}$$ \hspace{1cm} (13)$$

These equations for a freely drifting QEW, were derived in [16] for the Hamiltonian (6) that includes wavepacket chirp and expansion effects due to the dispersive second order term in (6). For simplicity we assume here that the QEW arrives at the interaction point \(z=0\) at time \(t_0\) at its longitudinal waist, so that \(\tilde{\sigma}_z (t) = \sigma_{z0} = \sigma_{p0}, \) and \(\sigma_{z0} = \hbar / 2\sigma_{p0}.\)

For a modulated wavepacket, with the same assumption [18]:

$$c_p^{(0)} = (2\pi \sigma_{p0}^2)^{-1/4} \sum_{n=-\infty}^{\infty} J_n (2|g|) \exp \left\{ -\frac{(p - p_{0} - n\delta p)^2}{4\sigma_{p0}^2} \right\} e^{i\varphi_p} e^{-i(p_{0}t_0 - E_{p0}) / \hbar}$$ \hspace{1cm} (14)$$

$$\Psi_F^{(0)} (z, t) = \frac{e^{-i(t_0 - \delta pt_0 / 2) / \hbar}}{(2\pi \sigma_{z0}^2)^{1/4}} \sum_{n=-\infty}^{\infty} J_n (2|g|) \exp \left\{ -\frac{(z - v_0 (t - t_0) - n\delta pt / 2\gamma m)^2}{4\sigma_{z0}^2} \right\} e^{i(\mu_{0} - v_{0})z - n\delta pt / 2\gamma m}$$ \hspace{1cm} (15)$$

Where \(\delta p = \hbar \omega_b / v_0.\)

In a simplified model, the spin is neglected, and we assume that the free and bound electrons do not overlap spatially. Therefore, there are no exchange energy or spin–orbit interaction effects, and we can avoid the intricate second quantization of many body interaction theory [30]. We assume that the only interaction is Coulomb interaction, and in the near field, neglecting retardation [36] and with gauge \(A = 0\), the interaction Hamiltonian is

$$H_1 (r, r') = \frac{e^2}{4\pi \varepsilon_0} \frac{\gamma}{\left[ (\gamma Z - \gamma')^2 + (r_r - r'_r)^2 \right]^{3/2}}$$ \hspace{1cm} (16)$$
\[ H_i(r, r') = \frac{e^2}{4\pi\varepsilon_0} \left[ \frac{1}{\left( \gamma^2 z^2 + r_{10}^2 \right)^{1/2}} + \frac{r'(\hat{e}_z \gamma Z - \hat{e}_r r_{10})}{\left( \gamma^2 z^2 + r_{10}^2 \right)^{3/2}} \right] \]  \tag{17}

where \( r, r' \) are the coordinates of the free and bound electrons respectively. Here we used Feynman's expression for the Coulomb potential \([46]\) in order to keep the analysis valid in the relativistic regime. A more accurate form would be to use the Darwin potential for relativistic Coulomb interaction between moving charged particle \([33,34]\). We believe, however, that for the parameters of the cases delineated here, the corrections due to this model are negligible.

In the interaction process, the expansion coefficients of the QEW \( c_p^{(0)} \) (14) are entangled with the coefficients of the bound electron \( C_j \) (10) and the combined wavefunction is:

\[ \Psi(r, r', t) = \sum_{j=1}^{2} \int dp c_{j, p}(t) \phi_j(r') e^{-iE_j t/h} e^{-iE_p t/h} e^{ipz/h} \]  \tag{18}

and after substitution in (4):

\[ i\hbar \frac{\partial \Psi}{\partial t} = \sum_{j=1}^{2} \int dp \left[ \hat{c}_{j,p}(t) - \frac{E_j + E_p}{\hbar} c_{j,p}(t) \right] \phi_j(r') e^{-iE_j t/h} e^{-iE_p t/h} e^{ipz/h} = \]

\[ = (H_{0B} + H_{0E} + H_1) \sum_{j=1}^{2} \int dp c_{j, p}(t) \phi_j(r') e^{-iE_j t/h} e^{-iE_p t/h} e^{ipz/h} \]  \tag{19}

After cancelling out the no-interaction terms, we are left with:

\[ i\hbar \sum_{j=1}^{2} \int dp \hat{c}_{j, p}(t) \phi_j(r') e^{-iE_j t/h} e^{-iE_p t/h} e^{ipz/h} = H_1(r, r') \sum_{j=1}^{2} \int dp c_{j, p}(t) \phi_j(r') e^{-iE_j t/h} e^{-iE_p t/h} e^{ipz/h} \]  \tag{20}

We now multiply by \( \phi_i^*(r') \) and integrate over space. Using the ortho-normality relation

\[ \int \phi_i^*(r') \phi_j^*(r') \, d^3 r' = \delta_{i,j} \]

\[ i\hbar \int \int dp d\hat{c}_{i, p}(t) \phi_i^*(r') e^{-iE_i t/h} e^{-iE_p t/h} e^{ipz/h} = \]

\[ e^{-iE_i t/h} \int \int dp d\hat{c}_{i, p}(t) \left\{ \int \hat{H}_1(r, r') \, d^3 r' \right\} e^{-iE_j t/h} e^{ipz/h} + e^{-iE_j t/h} \int \int dp d\hat{c}_{j, p}(t) \left\{ \int \hat{H}_1(r, r') \, d^3 r' \right\} e^{-iE_p t/h} e^{ipz/h} \]

\[ = M_{i,j}(r') = \left\{ \int \phi_i^*(r') \hat{H}_1(r, r') \phi_j(r') \right\} \]  \tag{21}

where

\[ M_{i,j}(r') = \left\{ \int \phi_i^*(r') \hat{H}_1(r, r') \phi_j(r') \right\} = \int d^3 r' \phi_i^*(r') \hat{H}_1(r, r') \phi_j(r') \]  \tag{22}

For simplicity we redefine the self-interaction terms, so that \( \left\{ \int \hat{H}_1(r, r') \, d^3 r' \right\} = 0 \), then:
This is an integro-differential equation that needs to be solved as a function of time for the initial condition \( c_{j,p}(t^-) = C_j^{(0)}(t^-)c_p^{(0)} \)

If \( |\mathbf{r}^-| << |\mathbf{r} - \mathbf{r}'| \approx (r_{10}^- + \gamma^2 z^2)^{1/2} \) then the integration over \( \mathbf{r}' \) can be carried out independently of \( \mathbf{r} \):

\[
M_{i,j}(\mathbf{r}_{10}, \mathbf{r}) = \int \varphi_i^*(\mathbf{r}') H_i(\mathbf{r}, \mathbf{r}') \varphi_j(\mathbf{r}') d^3\mathbf{r}'
\]  

(24)

For the interaction (17):

\[
M_{i,j} = \frac{e^2}{4\pi\varepsilon_0} \frac{\mathbf{r}_{i,j} \cdot (\hat{\mathbf{r}}Z - \hat{\mathbf{z}} \mathbf{r}_{10})}{(\gamma^2 z^2 + r_{10}^2)^{3/2}}
\]  

(25)

where

\[
-\mathbf{e} \mathbf{r}_{2,1} \equiv -e \int \varphi_2^*(\mathbf{r}') \mathbf{r}' \varphi_1(\mathbf{r}') d^3\mathbf{r}'
\]  

(26)

is the dipole transition matrix element \( \mathbf{\mu}_{2,1} = -e \mathbf{r}_{2,1} \).

2. **Projection to Momentum space**

We project the integro-differential equation (23) onto momentum space by multiplying with \( e^{-ipz/h} \) and integrating over \( z \). With \( \int e^{i(p'-p)z/h} dz = 2\pi i\hbar \delta(p' - p) \), we get:

\[
2\pi i\hbar \int dp \dot{c}_{i,p}(t)e^{-iE_i t/h} = e^{-i(E_j - E_i) t/h} \int dp \dot{c}_{j,p}(t) \int d\mathbf{r} M_{i,j}(\mathbf{r}) e^{i(p-p')Z} e^{-iE_j t/h}
\]  

(27)

\[
\dot{c}_{i,p}(t) = \frac{1}{2\pi i\hbar} \int dp \tilde{M}_{i,j}(p' - p) c_{j,p}(t) e^{-i(E_j - E_i - E_{ij}) t/h}
\]  

(28)

where

\[
E_{i,j} = E_i - E_j
\]  

(29)

\[
\tilde{M}_{i,j}(p' - p) = \int_{-\infty}^{\infty} d\mathbf{r} M_{i,j}(\mathbf{r}) e^{i(p-p')Z/h}
\]  

(30)

In Appendix A we present the explicit expressions of \( \tilde{M}_{i,j}(p) \) for the dipole matrix elements (25) of the longitudinally and transversely aligned dipoles. This function is related to the momentum space matrix element of the interaction Hamiltonian (25) by \( \tilde{M}_{i,j}(p' - p) = 2\pi i \langle p'|M_{i,j}|p\rangle = 2\pi i \langle p'\{i|H_j|p,j\} \rangle \).
The differential equation (28) describes the dynamic evolution of the entangled free electron and bound electron in momentum space. Before the start interaction time $t_0$, the free and bound electrons are not entangled:

$$c_{jp}(t) = C_j(t) c_p^{(0)}$$  \hspace{1cm} (31)

Equation (28) can be solved by an iterative process, in which we assume to first order that the free and bound electrons are not evolving in time during an effective interaction time:

$$c_{jp}(t) = C_j(t_0) c_p^{(0)}.$$  \hspace{1cm} (32)

Substituting this time independent amplitude in the RHS of (28), it is possible to integrate over time, and get a factor:

$$\int_{t_0}^{t} dt e^{-i(E_p-E_{p'}-E_{ij})t/\hbar} (t_0 - t) \sin c \left[ (E_p - E_{p'} - E_{ij})(t_0 - t) / 2\hbar \right] =$$

$$e^{-i(E_p-E_{ij})t/\hbar} 2t \sin c \left[ (E_p - E_{p'} - E_{ij})t / \hbar \right] \rightarrow 2\pi \hbar \delta(E_p - E_{p'} - E_{ij})$$  \hspace{1cm} (33)

where the last limit is taken for an infinite interaction time. This dictates a conservation of energy transfer condition:

$$E_{p'} - E_p = -E_{ij}$$  \hspace{1cm} (34)

This condition, used in the dispersion relation (7) determines the recoil momentum of the QEW during the interaction:

$$p_{rec} = p' - p = -E_{ij} / \nu_0$$  \hspace{1cm} (35)

The momentum recoil $p_{rec}$ is defined here so that if the transition is from the lower level $j = 1$ to the upper level $i = 2$, the momentum recoil is negative, and vice versa. Here we used only the first order expansion of the dispersion equation (7) (the dispersive second order term would introduce a small interaction quantum recoil correction differentiating up and down transitions [16,28] that can be neglected in the present context). Thus integration of (28) using (31),(32) results in:

$$c_{i,p'}(t_0^+) = C_i^{(0)}(t_0^-) c_p^{(0)} + \Delta c_{i,p'}$$  \hspace{1cm} (36)

$$\Delta c_{i,p'} = \frac{1}{i\hbar \nu_0} \tilde{M}_{ij} (p_{rec}) C_i^{(0)}(t_0^-) c_p^{(0)}$$  \hspace{1cm} (37)

The transition probability is:

$$P_i \left( t_0^+ \right) = \int_{p'} c_{i,p'}(t_0^-) dp' = \int_{p'} \left| C_i(t_0^-) c_p^{(0)}(t_0^-) + \Delta c_{i,p'}(t_0^-) \right|^2 dp' =$$

$$P_i^{(0)} + \Delta P_i^{(1)} + \Delta P_i^{(2)}$$  \hspace{1cm} (38)

where
\[ P^{(0)}_i = \left| C_i (t_0) \right|^2 \int_{p} \left| c^{(0)}_{i,p} (t_0) \right|^2 dp' = \left| C_i (t_0) \right|^2 \] (39)

is the initial occupation probability of level \( i \), and the incremental probabilities are:

\[ \Delta P^{(1)}_i = 2 \text{Re} \left[ C^{(0)*}_i (t_0) \int_{-\infty}^{\infty} dp' c^{(0)*}_{i,p'} (t_0) \Delta c_{i,p'} \right] \] (40)

\[ \Delta P^{(2)}_i = \int_{-\infty}^{\infty} dp' \left| \Delta c_{i,p'} \right|^2 \] (41)

Substituting (37) in (41), and integrating over momentum, using \( \int dp \left| c^{(0)}_{p-p_{\text{rec}}} \right|^2 = 1 \), one obtains:

\[ \Delta P^{(2)}_i (t_0) = \int dp' \left| c^{(0)}_{i,p'} \right|^2 = \frac{1}{\hbar^2 v_0^2} \left| \tilde{M}_{i,j} (p_{\text{rec}}) \right|^2 \left| C^{(0)}_j (t_0) \right|^2 \] (42)

In the case of excitation of a TLS from ground potential: \( C^{(0)}_2 (t_0) = 0, \quad C^{(0)}_1 (t_0) = 1 \), the excitation probability of the TLS is given by:

\[ P_2 (t_0^*) = \Delta P^{(2)}_2 = \int dp' \left| c^{(0)}_{i,p'} \right|^2 = \frac{1}{\hbar^2 v_0^2} \left| \tilde{M}_{i,j} (p_{\text{rec}}) \right|^2 \] (43)

Evidently the excitation probability in this case is independent of the QEW shape or dimensions. However, considering the case where the TLS is in a superposition state at the interaction time, the first order incremental probability term (40) may be dominant. Substituting (37) in (40) and integrating over momentum results in:

\[ \Delta P^{(1)}_i = \frac{2}{\hbar v_0} (\tilde{M}_{i,j} (p_{\text{rec}}) / i) \text{Re} \left[ C^{(0)*}_i (t_0) C^{(0)}_j (t_0) \right] I(p_{\text{rec}}) \] (44)

\[ I(p_{\text{rec}}) = \int c^{(0)*}_{p} c^{(0)}_{p-p_{\text{rec}}} dp \] (45)

This integral is evaluated in Appendix B for a Gaussian distribution of the QEW:

\[ I(p_{\text{rec}}) = e^{-\frac{1}{2} \left( p_{\text{rec}}/2\sigma_{p_0} \right)^2} \] (46)

Substituting \( p_{\text{rec}} = \hbar \omega_{1,2} / v \), \( \sigma_{z_0} = \hbar / 2\sigma_{p_0} \), \( \sigma_{t_0} = \sigma_{z_0} / v \), one gets:

\[ \Delta P^{(1)}_i = \frac{2}{\hbar v_0} \text{Re} \left[ C^{(0)*}_i (t_0) C^{(0)}_j (t_0) (\tilde{M}_{i,j} (p_{\text{rec}}) / i) \right] e^{-\Gamma^2/2} \] (47)

where:

\[ \Gamma = \frac{p_{\text{rec}}}{2\sigma_{p_0}} = \frac{\hbar \omega_{2,1}}{2v\sigma_{p_0}} = \omega_{2,1} \sigma_{10} = 2\pi \frac{\sigma_{x_0}}{\beta_{2,1}} \] (48)
in complete analogy to the case of stimulated radiative interaction of a finite size QEW (3) [16]. This means that the incremental probability is dependent on the wavepacket dimensions if the phase of the superposition state of the TLS is pre-determined, and it vanishes for a long wavepacket.

We note that the formulation in this section has only partial validity because its derivation involved some crude approximations. In particular, the integration of (28) in time, using the approximate relation (33) would be valid only if one can neglect the time dependence of \( c_{j,p}(t) \) during the interaction (32). Numerical solution of the Schrödinger equation that we carried out elsewhere, suggest that this amplitude does change substantially during the interaction when the electron wavepacket is long \( (\Gamma = \omega_{2,1} \sigma_{e_0} > 1) \). The results (43), (47) turn out to match the numerical solution only in the short interaction time limit

\[
E_{2,1} t_{\text{int}} < \hbar / 2
\]

\[
t_{r,\sigma_{t}} < t_{\text{int}} < E_{2,1} \hbar / \omega_{2,1}
\]

where \( t_{\text{int}} \) is the longer of the transit time \( t_r = \omega_{1,0} / \gamma \nu_0 \) and the wavepacket duration \( \sigma_{e_0} \). Instructively, if \( \Gamma = \omega_{2,1} \sigma_{e_0} < 1 / 2 \), then \( \sigma_{e_p} > E_{2,1} \sqrt{\nu_0} \), namely, the momentum spread is larger than the recoil (35).

In the next section we present an alternative approximate solution of the Schrödinger equation that contrary to the momentum projection approach takes into consideration also the dynamics of the TLS transition, and may better describe the short interaction regime (50).

3. Probabilistic Model for FEBERI Interaction

We go back to the source equation (28) and solve it directly with a first order iteration approximation by substituting on the RHS of (28):

\[
c_{j,p}(t) = C_j^{(0)}(t) c_p^{(0)}
\]

which is the same as in the previous assumption (32), but here allowing development in time of the TLS. After multiplying (23) by the complex conjugate of the free electron wavefunction (11) and integrating over space: \( \int d^3 r \psi_F^{(0)*}(r,t) \{ \cdots \}, \) one obtains:

\[
\frac{i}{2\pi} \int dp \int \hat{c}_{i,p}(t) c_p^{(0)*} e^{i(E_p - E_i) \hbar / \hbar} \int dz e^{i(p' - p)z / \hbar} = C_j^{(0)}(t) e^{i(E_i - E_j) \hbar / \hbar} \int d^3 r M_{i,j}(r) |\psi_F^{(0)}(r,t)|^2
\]

With \( \int dz e^{i(p' - p)z / \hbar} = 2\pi \hbar \delta(p' - p), \)

\[
2\pi \hbar \int dp' \hat{c}_{i,p}(t) c_p^{(0)*} = C_j^{(0)}(t) e^{i(E_i - E_j) \hbar / \hbar} \int d^3 r M_{i,j}(r) |\psi_F^{(0)}(r,t)|^2
\]
This presentation is reminiscent of point-particle interaction with the Born quantum wavefunction probability $|\Psi_{r}^{(0)}(r, t)|^2$ determining the electron arrival time $t$ at the TLS location $z=0$.

It should be stressed that $|\Psi_{r}^{(0)}(r, t)|^2$ is not well determined for a single electron. We assume that it is possible to solve (53) with substitution of its expectation value $\left\langle |\Psi_{r}^{(0)}(r, t)|^2 \right\rangle$, and the solution will then represent the result of interaction with an ensemble of identical QEWs.

The probability distribution of a single electron QEW of narrow width is:

$$\left\langle |\Psi_{r}^{(0)}(r, t)|^2 \right\rangle = \delta(r_z)f_{ct}(z-v_0(t-t_0)) = \delta(r_z)f_{ct}(t-t_0-z/v_0)/v_0$$  \hspace{1cm} (54)

where $f_{ct}$ is normalized over time. Then:

$$i\hbar \int p \hat{c}_{i,p}^\dagger(t)c_{p}^{(0)*} = C_j^{(0)}(t)e^{i\omega_j t}f(t-t_0)$$  \hspace{1cm} (55)

$$f(t-t_0) = \frac{1}{v_0} \int dz M_{i,j}(z)f_{ct}(t-t_0-z/v)$$  \hspace{1cm} (56)

where $M_{i,j}(z)$ is given in (25).

With approximation (51), assuming negligible change in the QEW around the interaction time $t_0$, we can turn (55) into coupled differential equations for the TLS $(i, j = 1, 2)$:

$$\dot{C}_i(t) = \frac{1}{i\hbar}C_j(t)e^{-i\omega_j t}f(t-t_0)$$  \hspace{1cm} (57)

and after integration:

$$C_i(t_0) = C_i(t_0) + \frac{1}{i\hbar} \int_{t_0}^{t_0} dt C_j(t)e^{-i\omega_j t}f(t-t_0).$$  \hspace{1cm} (58)

The weighed interaction probability $f(t-t_0)(56)$ depends on both the $z$ dependence of the interaction Hamiltonian (25) and the quantum probability (2), which for a Gaussian or modulated Gaussian QEW, can be calculated from the wavepacket functions (13) or (15).

For a single Gaussian wavepacket (13) at its longitudinal waist ($\tilde{\sigma}_z(t) = \sigma_{z0} = v_0 \sigma_{t0}$): $f_{ct}(t-t_0-z/v) = \frac{1}{\sqrt{2\pi\sigma_{t0}^2}} e^{-\frac{(t-t_0-z/v)^2}{2\sigma_{t0}^2}}$  \hspace{1cm} (59)
Normalizing time to the transit time parameter $\bar{T} = t / t_r$, and defining $\bar{T}' = z / \nu_0 t_r$, the weighed interaction probability function (56) can be recast into a convolution relation with the ratio $\bar{\sigma}_{\text{in}} = \sigma_{\text{in}} / t_r$ between the wavepacket duration and the transit time as a parameter:

$$f_\parallel(t-t_0) = K_\parallel \int_{-\infty}^{\infty} d\bar{T}' \frac{1}{(\bar{T}'^2+1)^{3/2}} \frac{1}{\sqrt{2\pi \bar{\sigma}_{\text{in}}}} e^{-(\bar{T}'-\bar{T})^2/2\bar{\sigma}_{\text{in}}^2}$$  \hspace{1cm} (60)

$$f_\perp(t-t_0) = K_\perp \int_{-\infty}^{\infty} d\bar{T}' \frac{1}{(\bar{T}'^2+1)^{3/2}} \frac{1}{\sqrt{2\pi \bar{\sigma}_{\text{in,0}}}} e^{-(\bar{T}'-\bar{T})^2/2\bar{\sigma}_{\text{in,0}}^2}$$  \hspace{1cm} (61)

$$K_\parallel = \frac{e^2}{4\pi \varepsilon_0 (\nu_0/\nu t_r)^2} r_{ij} \cdot \hat{e}_z = \frac{e^2}{4\pi \varepsilon_0} \frac{r_{ij} \cdot \hat{e}_z}{r_{0z}^2}$$  \hspace{1cm} (62)

$$K_\perp = \frac{e^2}{4\pi \varepsilon_0 (\nu_0/\nu t_r)^2} r_{ij} \cdot \hat{e}_r = \frac{e^2}{4\pi \varepsilon_0} \frac{r_{ij} \cdot \hat{e}_r}{r_{0r}^2}$$  \hspace{1cm} (63)

![Fig. 2 The weighed interaction probability $f(t,t_0)$ for $M_{i,j\parallel}(z)$, $M_{i,j\perp}(z)$.](image)

Substituted in (58), this already indicates dependence of the transition probability amplitude on the QEW dimension.

Now we proceed in calculating the TLS transition amplitude (58). Assuming small change in the TLS probability amplitude during the interaction:

$$C_i(t) = C_i(t_0)$$  \hspace{1cm} (64)

$$C_i(t + t_r) = C_i^{(0)}(t_0) + \Delta C_i$$  \hspace{1cm} (65)

$$\Delta C_i = \frac{1}{i\hbar} C_i(t_0) \int_{t_0}^{t+ \nu_0 t_r} f(t-t_0) \, dt.$$  \hspace{1cm} (66)

Within the range of short interaction time (50), and with the definition of Fourier transform:
\[ F(\omega) = \mathcal{F}\{f(t-t_0)\} = \int_{-\infty}^{\infty} e^{i\omega t} f(t-t_0) dt \quad (67) \]

\[ \Delta C_i = \frac{1}{i\hbar} C_j(t_0) F(-\omega_{i,j}) \quad (68) \]

where \( F_i(\omega) = \mathcal{F}\{f_i(t)\} \) is the Fourier transform of the probability function. Rather than using the explicit expressions (60), (61), we evaluate (66) by substitution of (56) in (67) and changing the integrations order:

\[ F(-\omega_{h,i}) = \frac{1}{v} \int_{-\infty}^{\infty} dt e^{-i\omega_{h,i}t} \int_{-\infty}^{\infty} dz M_{i,j}(z) f_{et}(t-t_0-z/v) \]

\[ = \frac{1}{v} \int dz e^{i\omega_{h,i}(t_0+z/v)} M_{i,j}(z) F_{et}(-\omega_{h,i}) \]

\[ = \frac{1}{v} e^{i\omega_{h,i}v} \bar{M}_{i,j} \left( \frac{\omega_{h,i}}{v} \right) F_{et}(-\omega_{h,i}) \quad (69) \]

For a Gaussian QEW:

\[ f_{et} = \frac{1}{(2\pi\sigma_v^2)^{1/2}} e^{-t^2/2\sigma_v^2} \quad (70) \]

\[ F(\omega_{h,i}) = e^{-\omega_{h,i}^2\sigma_v^2/2} \quad (71) \]

\[ \Delta C_i = \frac{1}{i\hbar v} C_j(t_0) e^{i\omega_{h,i}v} \bar{M}_{i,j} \left( \frac{\hbar \omega_{h,i}}{v_0} \right) e^{-\omega_{h,i}^2\sigma_v^2/2} \quad (72) \]

Similarly to (38):

\[ P_i(t_0^+) = \left| C_i(t_0^+) + \Delta C_i \right|^2 = P_i^{(0)} + \Delta P_i^{(1)} + \Delta P_i^{(2)} \quad (73) \]

\[ P_i^{(0)} = \left| C_i^{(0)}(t_0^-) \right|^2 \quad (74) \]

\[ \Delta P_i^{(1)} = 2 \text{Re} \left[ C_i^{(0)*}(t_0^-) \Delta C_i \right] \quad (75) \]

\[ \Delta P_i^{(2)} = |\Delta C_i|^2 \quad (76) \]

For finite size QEW:

\[ \Delta P_i^{(2)}(t_0^+) = \left[ \frac{1}{\hbar v_0} C_j(t_0) \bar{M}_{i,j} \left( \frac{\hbar \omega_{h,i}}{v_0} \right) \right]^2 e^{-t^2} \quad (77) \]
Which for \( i = 2, j = 1 \), \( C_2(t_0) = 1 \), can be written, in contrast to (43), as

\[
P_2(t_0) = \left[ \frac{1}{\hbar v_0} \tilde{M}_{i,j} \left( \frac{\hbar \omega_{ij}}{v_0} \right) \right]^2 e^{-t^2}.
\]

\[
\Delta P^{(1)}(t_0) = \frac{2}{\hbar} \operatorname{Re} \left[ C_i^{(0)*} (t_0) C_j^{(0)} (t_0) e^{i \omega_{ij} t_0} \tilde{M}_{i,j} \left( \frac{\hbar \omega_{ij}}{v_0} \right) / i \right] e^{-t^2/2} \tag{78}
\]

These are consistent with Eqs. 43, 47 in the limit of short interaction time (50), and manifest the wavepacket dependence of the transition probabilities through the parameter \( \Gamma = \omega_{i,j} \sigma_i \) (48). In the long wavepacket regime \( \Gamma > 1 \), (77) suggests decay of \( \Delta P^{(2)} \), which is inconsistent with (43). On the other hand, both (77) and (47) predict the same wavepacket dependent decay of the incremental probability \( \Delta P^{(1)} \). The dependence of (78) on the resonant phase-match timing of the short interaction impulse due to the QEW arrival relative to the dipole oscillation phase, is indicative of a possible coherent interaction enhancement by multiple electrons with correlated arrival timing, as is hypnotized in the coming sections.

We note that numerical solution of the Schrödinger equation that has been carried out elsewhere, support the prediction of finite transition probability and inelastic scattering predicted by (43), (77) in the short QEW regime \( \Gamma = \omega_{i,j} \sigma_i < 1 \) and the decayed transition probability (elastic scattering) of (77) in the long QEW regime \( \Gamma = \omega_{i,j} \sigma_i > 1 \) (in contrast to (43)). Both approximate formulations of the present and the previous sections are not valid in the intermediate regime of resonant interaction \( \Gamma = \omega_{i,j} \sigma_i \approx 2 \pi \), where enhanced transition rate takes place.

4. FEBERI interaction with a modulated QEW

In this case we use in (57)

\[
\left\langle \left| H_{\text{mod}} \right| \right| \left( \mathbf{r}, t \right) \right\rangle = \delta(t - t_{0k} - z / v_0) f_{\text{mod}}(t - z / v - t_L) \tag{79}
\]

where the modulation function is periodic [8,18] (see Fig. 4):

\[f_{\text{mod}}(t) = f_{\text{mod}}(t + 2\pi/\omega_b) \tag{80}\]

and therefore:

\[
f_{\text{mod}}(t) = \sum_{m=-\infty}^{\infty} f_m e^{im\omega_b t} \tag{81}\]

The coefficients \( f_m \) were derived in [18] for the case of the wavefunction of a modulated Gaussian QEW (15), \( \omega_b t_L \) is a modulation phase, determined by the modulating laser beam.

The incremental excitation probabilities (73) are derived in Appendix C. Explicitly:
\[ 
\Delta P^{(1)}_i = 2 \text{Re} \left\{ \frac{1}{2\pi i\hbar v_0} \tilde{M}_{i,j} \left( \frac{\hbar \omega_{i,j}}{v_0} \right) C_i^*(t_{0K}) C_j(t_{0K}) f_n e^{-i\omega_{i,j}t} e^{-(\omega_{i,j} - \omega_n)^2 \sigma_n^2 / 2} \right\} 
\]

\[ 
\Delta P^{(2)}_i = \frac{1}{\hbar v_0} \tilde{M}_{i,j} \left( \frac{\hbar \omega_{i,j}}{v_0} \right) C_j(t_{0K}) \left| f_n \right|^2 e^{-(\omega_{i,j} - \omega_n)^2 \sigma_n^2} 
\]

where \( n \) is the bunching frequency harmonic that matches the TLS quantum energy levels (1):

\[ \omega_{i,j} = n \omega_b \]

Remarkably, both incremental probabilities display resonant excitation characteristics around condition (84), which would manifest the QEW modulation characteristics in a properly set experiment. Note that in a modulated QEW \( \omega_{i,j} \sigma_{\omega} > 1 \), and according to (43), (47), in this range the second order incremental modulation of an unmodulated QEW is not wavepacket dependent, and the first order increment decays. In comparison, Eqs. 82, 83 suggest resonant behavior at harmonics of the modulation frequency, but no enhancement.

5. FEBERI interaction with Multiple QEWs:

The FEBERI interaction with multiple QEWs is theoretically an intricate problem that involves entanglement of the free electron wavefunction with the TLS quantum states, and consequently – entanglement with all subsequent interacting electrons. We carry out such analysis elsewhere in a rigorous multi-particle density matrix formulation. At present, we resort to a simple model, in which we extend (54), (57), (58) to multiple particles by the substitution:

\[ \left\langle \left| \Psi^{(0)}_\kappa (\mathbf{r}, t) \right|^2 \right\rangle \to \sum_{K=1}^{N} \left\langle \left| \Psi^{(0)}_\kappa (\mathbf{r}, t) \right|^2 \right\rangle \]

\[ \left\langle \left| \Psi^{(0)}_\kappa (\mathbf{r}, t) \right|^2 \right\rangle = \delta(\mathbf{r}) f_{\kappa}(t - t_{0K} - z / v_0) \]

We then solve for the cumulative incremental FEBERI transition probability from ground state \( (j=1) \) to upper state \( (i=2) \) under the assumption that the relaxation time of the upper level is much longer than the duration of the N QEWs pulse [44]. This results in (Appendix D):

\[ P_2 = N^2 \left\{ \frac{1}{\hbar v_0} \tilde{M}_{2,1} \left( \frac{\hbar \omega_{2,1}}{v_0} \right) \right\} \left| f_{\kappa} \right|^2 e^{-\sigma_n^2 / 2} 
\]

This can be termed the case of the “Quantum Klystron”, that was analyzed in [35] in the point particle limit. It is the quadratic approximation of the \( \sin^2 (\Omega_{s} t / 2) \) scaling of Rabi oscillation, and it is analogous to the bunched particles beam superradiance effect [27].
Note that in the classical point particle limit and low (microwave) frequencies \[35\] there may be multiple electrons per period and \(N\) is just the number of electrons in the modulated electron beam pulse: \(N = I_{\text{mod}} I_{\text{pulse}} / e\).

6. FEBERI interaction with multiple modulation-correlated QEWs:

We now combine the cases of the last two sections, and consider the case of multiple QEWs, all modulated at the level of their quantum wavefunctions by the same coherent laser beam of frequency \(\omega_b\) and phase \(\omega_b t_L\). We use the multi-particles probability function \((85)\), with

\[
\left|\psi_k^{(0)}(r, t)\right|^2 = \delta(r_z) f_{\alpha}(t - t_{0K} - z / v) f_{\text{mod}}(t - z / v_0 - t_L) \tag{88}
\]

Where \(t_{0K}\) are the centroid arrival times of the envelopes of the modulated QEWs, and the modulation function, common to all QEWs is periodic as in time \((80)\):

\[
f_{\text{mod}}(t) = f_{\text{mod}}(t + 2\pi / \omega_b) \tag{89}
\]

With \((81)\), the multi-electron probability distribution function is:

\[
f(t - t_0) = \sum_{K=1}^{N} \frac{1}{\sqrt{\nu}} \int dz M_{i,j}(z) f_{\alpha}(t - t_{0K} - z / v_0) \sum_{m=-\infty}^{\infty} f_m e^{i\omega_b(t - t_{0K} - t_L)} \tag{90}
\]

Then, as in Appendix. C, substitution in \((58)\), changing the integration order of \(z\) and \(t\), results in:

\[
C_i(t_{0N}^+) = C_i(t_0^+) + \frac{1}{i\hbar v_0} \int dz M_{i,j}(z) \sum_{K=1}^{N} C_j(t_{0K}) \sum_{m} \int dt e^{-i\omega_b t} f_{\alpha}(t - t_{0K} - z / v_0) f_m e^{i\omega_b(t - t_{0K} - t_L)} \tag{91}
\]

\[
= C_i(t_0^+) + \frac{1}{i\hbar v_0} \int dz M_{i,j}(z) e^{i\omega_b t} \sum_{K=1}^{N} C_j(t_{0K}) \sum_{m} f_m \int dt e^{-i(\omega_b - \omega_b^*) t} f_{\alpha}^*(t - t_{0K}) e^{-i\omega_b^* t_L} \tag{92}
\]

where \(F_{el}(\omega_{i,j} - \omega_b) = \mathcal{F}\{f_{\alpha}(t)\}_{\omega = \omega_{i,j} - \omega_b}\) is the Fourier transform of the single QEW probability function. For a Gaussian distribution the envelope distribution \((70)\) is a wide function - \(\sigma_{\alpha} > 2\pi / \omega_b\), and the spectral function
\[ F(\omega_{i,j} - m\omega_b) = e^{-(\omega_{i,j} - m\omega_b)^2/2} \] (93)

is a narrow function around a harmonic \(m=n\) that is resonant at the transition frequency:

\[ \omega_{i,j} = n\omega_b \] (94)

Take \(i=2, j=1\) (upper and lower levels), then with the approximation of small change in the amplitude:

\[ C_1(t_{0K}) \approx C_1(t_0) = \text{const} \approx 1, \quad |C_2(t_0)| << 1 \] (95)

\[ C_2(t_{0N}^+) \approx \frac{1}{\hbar \nu_0} \tilde{M}_{2,1} \left( \frac{\hbar \omega_{2,1}}{\nu_0} \right) \sum_m f_m \sum_{k=1}^N e^{i(\omega_{i,j} - m\omega_b)t_0} e^{-i\omega_{i,j}t_0} e^{-(\omega_{i,j} - m\omega_b)^2/2} \] (96)

This averages to zero for random arrival times \(t_{0K}\) of the wavepacket centroids, except at the resonance condition (94), where independently on the arrival times \(t_{0K}\):

\[ C_2(t_{0N}^+) \big|_{t_{0K}=\pi m\omega_b} \approx N \frac{1}{\hbar \nu_0} \tilde{M}_{2,1} \left( \frac{\hbar \omega_{2,1}}{\nu_0} \right) f_n e^{-i\omega_{i,j}t_0} \] (97)

\[ P_2 = N^2 \left| \frac{1}{\hbar \nu_0} \tilde{M}_{2,1} \left( \frac{\hbar \omega_{2,1}}{\nu_0} \right) f_n \right|^2 \] (98)

This expression, explicitly manifests the \(N^2\) scaling buildup of the upper quantum level probability in the case of multiple modulation-correlated QEWs, similarly to the case of periodically modulated point particles (section 4) and in analogy to superradiance of bunched particles [27].

7. Simulation of FEBERI with multiple correlated QEWs

To affirm the validity of the FEBERI effect and its quadratic scaling \(N^2\) with the number of modulation-correlated QEWs, we simulate the TLS transitions and the quantum levels population dynamics in a model where the electron interaction times are determined by Born’s combined probability function (85,88,89), that accounts for both the classical random injection times of the centroids of the QEWs \(t_{0K}\) and the quantum probability timings of the electrons arrival corresponding to the probability function \(f_{\text{mod}}(t-Z/v_0-t_L)\) common to all modulation-correlated QEWs. It has the same modulation phase for all electrons - \(\omega_b t_L\) (the phase of the modulating laser).

We use for our simulations the parameters of [8] corresponding to a PINEM experiment with a 200keV electron beam. The modulation probability function is the squared
absolute value of the QEW wavefunction after PINEM laser interaction (15), evaluated at an optimal post-interaction drift time \( t_D = T_b / 2(\Delta p_{\text{mod}} / p_0) \), where \( \Delta p_{\text{mod}} \) is the momentum modulation amplitude at the PINEM interaction point [18]. This distribution is shown in Fig. 4 for the parameters of [8, 18]. The density modulation bunches in this case are of attosecond time duration, much shorter than the optical frequency period of the modulation.

Fig. 4. The density modulated QEW distribution displayed in terms of time \( t - t_{0K} \) relative to \( t_{0K} \) - the centroid arrival time of electron \( K \) (from [8, 18]).

Therefore, we assert that even in case that the wavepacket duration \( \sigma_t \) of an unmodulated QEW is long relative to the TLS transition resonant frequency \( \omega_{2,1} \), not satisfying the near-point-particle condition (50), such tightly bunched modulated QEWs may still operate in this regime.

Contrary to the analytical approach in section 6, in the simulations we need to calculate the accumulative dynamics of the TLS transitions due to multi-electron interactions by calculating separately the transition dynamics of single near-point-particle QEWs (56, 57). Guided by the probabilistic interpretation of (53), we use the probability distribution function (88) only as the algorithm for determining the interaction time \( t_K \) of a modulated QEW of centroid arrival time \( t_{0K} \) (see Fig. 4). For simplicity we take the approximation of very tight density bunching (see [8,9,12], where attoSecond short bunches were demonstrated, while the optical period \( T_b = 2\pi / \omega_\nu \) was of the order of femtoSeconds). Therefore, under the assumption that the QEW’s envelope duration \( \sigma_t \) is longer than the optical period, the arrival times of the electrons are determined by the modulation function peaks of the quantum probability distribution displayed in Fig. 4:

\[
t_K = t_{0K} + n_K T_b
\]

where \( n_K \) is an ascending series of randomly spaced integers \( K=1..N \). The sample electrons \( K \) that contribute to the transition amplitude dynamics through Eq. 57 are only the ones that their arrival times \( t_K \) fall within the range of the QEW envelope width (see Fig. 4) of any QEW.
that arrives at a centroid random time $t_{0K}$. This is almost the only role of $t_{0K}$, and the interaction time (99) is determined primarily by the peaks (Fig. 4) of the Born quantum probability distribution function (88). In this picture, Eq. 99 represents a train of near point-particle wavepackets spaced with integral multiple optical periods, such that only one electron is counted within the envelope of each modulated QEW. Therefore, as predicted in the analytical calculation of Section 6, it would be expected that simulation of such a train of phase-correlated particles would resonantly buildup the transition to the upper level when the modulation frequency $\omega_2$ is a sub-harmonic of the transition frequency:

$$\omega_{2,1} = n\omega_b$$

(100)

The transition probability of a single electron wavepacket was calculated for parameters choice $E_0 = 200keV$, $r_\perp = 4.8nm$, $\omega_{2,1} = 3x10^{15} \text{ rad} / S$ ($E_{2,1} = 1.98eV$), $\mu_{ij}^{\perp} = 5\text{Debye}$, by solving the coupled equations (57) (i,j=1,2) for a QEW in the near-point-particle limit. This is shown in Fig. 5.

Fig. 5: Numerical solution of the coupled equations (57) for the transition between the TLS quantum states. $P_1$ and $P_2$ represent the occupation probability of the ground state and upper state respectively, satisfying the relation $P_1 + P_2 = 1$. The parameters of the exciting QEW are in the near-point-particle regime.

Fig. 6 shows simulation of the buildup of the TLS upper level probability, solving (57) with $N_1=20$ particles arriving at times (99), where $n_k$ is a random number (blue curve). The growth is evidently quadratic, $P_2 \propto N^2$ as claimed. For comparison, we show in the upper (magenta) curve the case where instead of (99), $t_k$ is taken to be entirely random. The growth rate is linear, and the upper TLS level arrives at the same excitation level only with $N_1=N_1^2=400$. 

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Fig. 6: Simulation results of the upper level probability buildup by electrons arriving to the interaction point at random (magenta) and by electrons arriving at times commensurate with the modulation period $T_b = 2\pi / \omega_b$ (99) at the resonance condition (100) (blue). The red-dash and cyan-dash curves are the linear and quadratic curve-fittings, respectively.

Conclusion

We studied the interaction between free electrons and a bound electron (FEBERI) in a framework of a comprehensive quantum model, in which the bound electron is represented by a two level system (TLS) and the free electrons are represented as quantum electron wavepackets (QEW). The electric dipole interaction of the two body system was solved in terms of the Schrodinger equation, extended to the relativistic regime. Since we start from a wavepacket model for the free electron, the analysis can provide two limits of interaction: wavelike and point-particle-like interactions, manifesting the wave-particle duality nature of quantum mechanics and the transition from quantum to classical point particle presentations. This observation is similar to the analogous cases of QEWs interactions with light: stimulated radiative interaction and superradiance [16-19]. The results show that the electron wavepacket dimension and its density modulation characteristics [8] are physically observable parameters that can be measured in appropriately designated experimental setups of electron interaction with matter or light. However, since the QEW dimensions and modulation characteristics are defined only in terms of expectation values, this assertion applies only to measurement of interaction with multiple identical QEWs.

The analysis shows that optical-frequency-modulated QEWs interact resonantly with a TLS at the condition that the TLS transition frequency is a harmonic of the modulation frequency of the QEWs. This resonant excitation may be observable in measurement of cathodoluminescence of the TLS.

Extending the analysis of FEBERI interaction to multiple electrons, interacting with a TLS under the condition that the duration of the electron beam pulse is shorter than the relaxation time of the TLS, we have found that resonant transition probability buildup of the upper TLS
quantum level takes place in proportion to $N^2$. This would happen when the QEWs are short, namely in the near-point-particle limit of the QEW, and their wavefunction centroids are periodically density-bunched at a subharmonic of the TLS quantum transition frequency. This case corresponds to a “quantum klystron” [35], where the QEWs interaction can be described in terms of periodic bunching of classical single particles, analogous to classical superradiance of a bunched particle beam [26, 27].

The analysis was further extended to the wave-like limit of modulated QEWs, in which the duration of the QEWs is long relative to the modulation period, so that the expectation value of the density distribution of each QEW is modulated at the optical frequency of the modulating laser (Fig. 4). Our quantum formulation of the FEBERI interaction leads to an interpretation of point-particle interaction, where the interaction time is determined by the Born probability distribution of the modulated QEW. Under the hypothesis that this applies to multiple modulation-correlated QEWs (namely, all QEWs are modulated by the same coherent laser beam), we show that similar quadratic $N^2$ buildup of the upper TLS level is expected when the QEWs are tightly modulated, and a harmonic of their common modulation frequency is resonant with the TLS quantum transition frequency. This would be the case, because the arrival phase of the train of collapsed point-particles is commensurate with the TLS transition frequency even though the centroids of the QEWs arrive at random. Such a coherent buildup process of the quantum transitions of the TLS would be expressed by enhanced cathodoluminescence in ultrafast Transmission Electron Microscope settings, and may have potential applications in atomic scale probing of quantum excitation in matter and coherent control of Qbits and quantum emitters.

The analysis here, though comprehensively quantum, is still incomplete and particularly the multiple particle FEBERI process requires further elaboration, considering the entanglement that is established in the process between the QEW and the TLS quantum state, and further with the subsequent incoming electrons. This kind of analysis is under way elsewhere in terms of density matrix formulation. Moreover, experimental verification of the new FEBERI process is called for, requiring advancement of the technological state of the art of electron microscopy. Shaping the wavepacket length in the range of an optical period requires development of filtering and wavepacket compression techniques [48, 49]. One must be also concerned about mitigation of the electron beam deterioration due to Coulomb interaction between the electrons [38] (e.g. by use of a high repetition rate mode locked laser [39]). One must be aware also that the expected probabilities of transition per single TLS in the given examples are very small, and experimental verification would probably require use of multiple TLS schemes [50].

Finally, we stress that the present single QEW analysis of the FEBERI interaction, and the conclusions about the measurability of the QEW dimensions, are based on the expectation value of the density probability distribution, and there is an explicit assumption there that the measurements are done with an ensemble of properly prepared identical multiple electrons. We mention however, the prevalent research interest in non-projective direct measurability of single particle wavefunctions using weak [40, 41] or protective measurement [42, 43] schemes. This aspect, representing an alternative realistic interpretation of the QEW, is not covered in the present article.
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References:

1. A. Gover, A. Yariv, “Free-Electron-Bound-Electron Resonant Interaction”, PRL 124, 064801 (2020).
2. Barwick, B., Flannigan, D.J. and Zewail, A.H. "Photon-induced near-field electron microscopy". *Nature*, 462 (7275), p. 902 (2009).
3. García de Abajo, F.J., Asenjo-Garcia, A. and Kociak, M., "Multiphoton absorption and emission by interaction of swift electrons with evanescent light fields". *Nano letters*, 10 (5), pp.1859-1863, (2010).
4. Park, S.T., Lin, M. and Zewail, A.H., "Photon-induced near-field electron microscopy (PINEM): theoretical and experimental". *New Journal of Physics*, 12 (12), p.123028, (2010).
5. Park, S.T. and Zewail, A.H., "Relativistic effects in photon-induced near field electron microscopy". *The Journal of Physical Chemistry A*, 116 (46), pp.11128-11133. (2012).
6. Echternkamp, K.E., Feist, A., Schäfer, S. and Ropers, C., "Ramsey-type phase control of free-electron beams", *Nature Physics*, 12 (11), p.1000, (2016).
7. Vanacore, G.M., Madan, I., Berruto, G., Wang, K., Pumarico, E., Lamb, R.J., McGrouther, D., Kaminer, I., Barwick, B., de Abajo, F.J.G. and Carbone, F. "Attosecond coherent control of free-electron wave functions using semi-infinite light fields". *Nature communications*, 9 (1), p.2694, (2018).
8. Feist, A., Echternkamp, K. E., Schauss, J., Yalunin, S. V., Schfer, S., Ropers, C. "Quantum coherent optical phase modulation in an ultrafast transmission electron microscope". Nature, 521 (7551), p. 200-203 (2015).
9. Priebe, K.E., Rathje, C., Yalunin, S.V., Hohage, T., Feist, A., Schäfer, S. and Ropers, C., "Attosecond electron pulse trains and quantum state reconstruction in ultrafast transmission electron microscopy". *Nature Photonics*, 11 (12), p.793, (2017).
10. Piazza, L. U. C. A., Lummen, T. T. A., Quinonez, E., Murooka, Y., Reed, B. W., Barwick, B., & Carbone, F. (2015). Simultaneous observation of the quantization and the interference pattern of a plasmonic near-field. *Nature communications*, 6, 6407.
11. Kozák, M., N. Schönenerberger, and P. Hommelhoff. "Ponderomotive generation and detection of attosecond free-electron pulse trains." Physical review letters 120.10 (2018): 103203.
12. Kozák, M., Eckstein, T., Schönenberger, N., & Hommelhoff, P. (2018). Inelastic ponderomotive scattering of electrons at a high-intensity optical travelling wave in vacuum. *Nature Physics, 14*(2), 121.

13. Schrödinger, E. (1926). An undulatory theory of the mechanics of atoms and molecules. Physical Review, 28(6), 1049.

14. Born, M. Z. Physik (1926) 37: 863. https://doi.org/10.1007/BF01397477.

15. http://www.feynmanlectures.caltech.edu/III_21.html

16. Gover, A. and Pan, Y., "Dimension-dependent stimulated radiative interaction of a single electron quantum wavepacket", *Physics Letters A*, 382 (23), pp.1550-1555, (2018).

17. Y. Pan, B. Zhang, A. Gover, "Anomalous photon-induced near-field electron microscopy" Phys. Rev. Lett. 122, 183204 (2019)

18. Pan, Y. and Gover, A., "Spontaneous and stimulated radiative emission of modulated free-electron quantum wavepackets—semiclassical analysis", *Journal of Physics Communications*, 2 (11), p.115026, (2018).

19. Pan, Yiming, and Avraham Gover. "Spontaneous and stimulated emissions of a preformed quantum free-electron wave function." Physical Review A 99.5 (2019): 052107.

20. Black, D. S., Niedermayer, U., Miao, Y., Zhao, Z., Solgaard, O., Byer, R. L., & Leedle, K. J. Net acceleration and direct measurement of attosecond electron pulses in a silicon dielectric laser accelerator. Phys. Rev. Lett. 123, 264802 (2019)

21. Schönenberger, N., Mittelbach, A., Yousefi, P., Niedermayer, U., & Hommelhoff, P. (2019). Generation and Characterization of Attosecond Micro-Bunched Electron Pulse Trains via Dielectric Laser Acceleration. *Phys. Rev. Lett. 123, 264803 (2019)*.

22. Corson, J. P., & Peatross, J. (2011). Quantum-electrodynamic treatment of photoemission by a single-electron wave packet. Physical Review A, 84(5), 053832.

23. I. Kaminer, M. Mutzafi, A. Levy, G. Harari, H. Herzig Sheinfux, S. Skirlo, J. Nemirovsky, J. D. Joannopoulos, M. Segev, andM. Soljac’ic’, Quantum C^-erenkov Radiation: Spectral Cutoffs and the Role of Spin and Orbital Angular Momentum, Phys. Rev. X 6, 011006 (2016).

24. Remez, R., Karnieli, A., Trajtenberg-Mills, S., Shapira, N., Kaminer, I., Lereah, Y., & Arie, A. (2019). Observing the Quantum Wave Nature of Free Electrons through Spontaneous Emission. *Physical review letters*, 123(6), 060401.

25. Dicke, R. H. Coherence in spontaneous radiation processes. Physical Review, 93(1), 99 (1954).

26. Gover, A. Superradiant and stimulated-superradiant emission in prebunched electron-beam radiators. I. Formulation. Physical Review Special Topics-Accelerators and Beams, 8(3), 030701 (2005).
27. A. Gover, R. Ianconescu, A. Friedman, C. Emma, N. Sudar, P. Musumeci, C. Pellegrini, "Superradiant and stimulated superradiant emission of bunched electron beams", Reviews of Modern Physics 91, (3), 035003 (2019).

28. Friedman, A., A. Gover, G. Kurizki, S. Ruschin, and A. Yariv. "Spontaneous and stimulated emission from quasifree electrons." Reviews of modern physics 60, (1988): 471.

29. Di Giulio, Valerio, Mathieu Kociak, and F. Javier García de Abajo. "Probing quantum optical excitations with fast electrons." Optica 6.12 (2019): 1524-1534.

30. Bartlett R J. Many-body perturbation theory and coupled cluster theory for electron correlation in molecules[J]. Annual Review of Physical Chemistry, 1981, 32(1): 359-401.

31. Feynman lecture: https://www.feynmanlectures.caltech.edu/II_26.html

32. Kociak, M., and L. F. Zagonel. "Cathodoluminescence in the scanning transmission electron microscope." Ultramicroscopy176 (2017): 112-131.

33. Breit, G. “Dirac's Equation and the Spin-Spin Interactions of Two Electrons.” Physical Review 39, (1932); 616-624

34. Lhuillier, C., and Faroux, J.P. “Hamiltonian of a Many-Electron Atom in an External Magnetic Field and Classical Electrodynamics”, Le Journal De Physique 7 (1977); 747-755.

35. Rätzel, D., Hartley, D., Schwartz, O., & Haslinger, P. (2020). A Quantum Klystron--Controlling Quantum Systems with Modulated Electron Beams. arXiv preprint arXiv:2004.10168.

36. De Abajo, FJ García. "Optical excitations in electron microscopy." Reviews of modern physics 82.1 (2010): 209.

37. Bateman, Tables of Integral Transforms, Vol. 1, chapter 1 and 2

38. Feist, A., Bach, N., da Silva, N. R., Danz, T., Möller, M., Priebe, K. E., ... & Strauch, S. (2017). Ultrafast transmission electron microscopy using a laser-driven field emitter: Femtosecond resolution with a high coherence electron beam. Ultramicroscopy, 176, 63-73.

39. Schaeres, L., Paschotta, R., Occhi, L. and Guekos, G., 2004. 40-GHz mode-locked fiber-ring laser using a Mach-Zehnder interferometer with integrated SOAs. Journal of lightwave technology, 22(3), pp.859-873.

40. Lundeen, J. S., Sutherland, B., Patel, A., Stewart, C., & Bamber, C. (2011). Direct measurement of the quantum wavefunction. Nature, 474(7350), 188-191.

41. Thekkadath, G. S., et al. (2016). Direct measurement of the density matrix of a quantum system, Phys. Rev. Lett. 117, 120401.

42. Aharonov, Y. and Vaidman L. (1993). Measurement of the Schrödinger wave of a single particle, Phys. Lett. A 178, 38-42.

43. Piacentini, F. et al. (2017) Determining the quantum expectation value by measuring a single photon, Nat. Phys. 13, (2017): 1191-1194.
44. Collins, A. T., M. F. Thomaz, and Maria Isabel B. Jorge. "Luminescence decay time of the 1.945 eV centre in type Ib diamond." *Journal of Physics C: Solid State Physics* 16.11 (1983): 2177.

45. Fares, Hesham. "The quantum effects in slow–wave free–electron lasers operating in the low–gain regime." *Physics Letters A* (2020): 126883.

46. Feynman Richard P. "Feynman lectures on physics. Volume 2: Mainly electromagnetism and matter." *flp* (1964).

47. Bateman, Harry. "Tables of integral transforms." (1954).

48. Baum, Peter. "Quantum dynamics of attosecond electron pulse compression." *Journal of Applied Physics* 122.22 (2017): 223105.

49. Niedermayer, U., Egenolf, T., Boine-Frankenheim, O., & Hommelhoff, P. (2018). Alternating-phase focusing for dielectric-laser acceleration. *Physical review letters, 121*(21), 214801.

50. A. Halperin, A. Gover, and A. Yariv. "Electron-beam-induced super-radiant emission from a grating." *Physical Review A* 50.4 (1994): 3316.
Appendix A: Coulomb Interaction Matrix element

Here we evaluate the momentum space representations of the interaction matrices of the longitudinally and transversely aligned dipoles (25).

For the longitudinally aligned dipole:

\[
M_{i,j,\parallel}(z) = \frac{e^2 r_{i,j}}{4\pi\varepsilon_0} \frac{yz}{(y^2z^2 + r_\parallel^2)^{3/2}}
\]

(A1)

Then:

\[
\tilde{M}_{i,j,\parallel}(p) = \int dz \ M_{i,j,\parallel}(z)e^{-ipz/h} = \frac{e^2 r_{i,j}}{4\pi\varepsilon_0 y^2} \int dz \ \frac{Z}{(z^2 + r_\parallel^2 / y^2)^{3/2}} e^{-ipz/h}
\]

(A2)

Using a relation from an integrals table [47]:

\[
\tilde{M}_{i,j,\parallel}(p) = -i \frac{e^2 r_{i,j}}{2\pi\varepsilon_0 y^2} \frac{p}{h} K_0 \left( \frac{pr_\parallel}{h} \right)
\]

(A3)

For the transversely aligned dipole:

\[
M_{i,j,\perp}(z) = \frac{e^2 r_{i,j}}{4\pi\varepsilon_0} \frac{yr_{\perp}}{(y^2z^2 + r_\perp^2)^{3/2}}
\]

(A4)

So the Fourier transformation of the function is

\[
\tilde{M}_{i,j,\perp}(p) = \int dz \ M_{i,j,\perp}(z)e^{-ipz/h} = \frac{e^2 r_{i,j}r_{\perp}}{4\pi\varepsilon_0 y^2} \int dz \ \frac{1}{(z^2 + r_\perp^2 / y^2)^{3/2}} e^{-ipz/h} =
\]

(A5)

\[
\tilde{M}_{i,j,\perp}(p) = -i \frac{e^2 r_{i,j}}{2\pi\varepsilon_0} \frac{p}{h} K_0 \left( \frac{pr_{\perp}}{h} \right)
\]

(A6)
Append. B: Calculate $I = \int c_p^{(0)} c_p^{(0)} dp$

We calculate the coefficient $I(p)$ of Eq. 45 for a finite Gaussian QEW using Eq. 12, and assuming for simplicity that the QEW arrives at its longitudinal waist (no chirp) - $t_D = L_D = 0$, $\sigma_p = \sigma_{p_0}$:

$$I(p_{\text{rec}}) = \frac{1}{(2\pi \sigma_{p_0}^2)^{1/2}} \int dp \frac{(p-p_0)^2}{4\sigma_{p_0}^2} e^{-\frac{(p-p_0)^2}{4\sigma_{p_0}^2}}$$

(B1)

$$I(p_{\text{rec}}) = \frac{1}{(2\pi \sigma_{p_0}^2)^{1/2}} \int dp \frac{(p-p_0)^2}{2\sigma_{p_0}^2} e^{-\frac{(p-p_0)^2}{2\sigma_{p_0}^2}}$$

(B2)

Complete the polynomial to a square:

$$\frac{1}{(2\pi \sigma_{p_0}^2)^{1/2}} \int dp \left[ e^{-\frac{(p-p_0)^2}{2\sigma_{p_0}^2}} \right] = 1 \cdot e^{-\frac{1}{2}(p_{\text{rec}}/2\sigma_{p_0})^2}$$

(B3)

Note:

With $p_{\text{rec}} = \hbar \omega_{1,2}/v$, $\sigma_{p_0} = \hbar / 2\sigma_{p_0}$, $\sigma_{\omega} = \sigma_{\omega_0} / v_0$, we define (3), in analogy to the case of QEW interaction with light [16]:

$$\Gamma = \frac{p_{\text{rec}}}{2\sigma_{p_0}} = \frac{\hbar \omega_{1,2}}{2v_0\sigma_{p_0}} = \omega_{1,2}\sigma_{\omega_0} = 2\pi \frac{\sigma_{\omega_0}}{\beta \lambda_{1,2}}$$

(B4)

then

$$I(p_{\text{rec}}) = e^{-\Gamma^2/2}$$

(B5)

This factor is independent of the sign of $p_{\text{rec}}$. 
Appendix C. FEBERI Interaction with a modulated QEW

Starting from (79), (81)

\[
\left\langle |\Psi^{(0)}_k(r,t)|^2 \right\rangle = \delta(t) f_{ct}(t-t_{0K} - z/v) f_{mod}(t-z/v_0 - t_L)
\]

(C1)

\[
f_{mod}(t) = \sum_{m=-\infty}^{\infty} f_m e^{im\omega_b t}
\]

(C2)

The distribution function (56) becomes

\[
f(t-t_0) = \frac{1}{v_0} \int dz M_{i,j}(z) f_{ct}(t-t_{0K} - z/v) \sum_{m=-\infty}^{\infty} f_m e^{im\omega_b (t-z/v_0 - t_L)}
\]

(C3)

We substitute this probability distribution of a modulated QEW in (58) and change order of integrations:

\[
C_i(t_i^*) = C_i(t_0^*) + \frac{1}{2\pi i \hbar v_0} \int dz M_{i,j}(z) C_j(t_{0K}) \sum_m dt e^{-im\omega_b t} f_{ct}(t-t_{0K} - z/v_0) f_m e^{im\omega_b (t-z/v_0 - t_L)}
\]

(C4)

With change of variables \( t' = t - z/v_0 \),

\[
C_i(t_0^*) = C_i(t_0^*) + \frac{1}{2\pi i \hbar v_0} \int dz M_{i,j}(z) e^{\frac{i \omega_b z}{v_0}} C_j(t_{0K}) \sum_m dt' e^{-im\omega_b t'} f_{ct}(t'-t_{0K}) e^{-im\omega_b t_L}
\]

\[
= C_i(t_{0K}) + \frac{1}{2\pi i \hbar v_0} \tilde{M}_{i,j} \left( \frac{\hbar \omega_{i,j}}{v_0} \right) C_j(t_{0K}) \sum_m f_m e^{i(\omega_{i,j}-\omega_b) t_{0K}} F_{ct} \left( \omega_{i,j} - \omega_b \right) e^{-im\omega_b t_L}
\]

(C5)

We calculate the incremental transition probabilities (75), (76):

\[
\Delta P_i^{(1)} = 2 \text{Re} \left[ C_i^{(0)*} \left( t_0^* \right) \Delta C_i \right] = \\
2 \text{Re} \left[ \frac{1}{2\pi i \hbar v_0} \tilde{M}_{i,j} \left( \frac{\hbar \omega_{i,j}}{v} \right) C_i^{*} \left( t_{0K} \right) C_j(t_{0K}) \sum_m f_m e^{i(\omega_{i,j}-\omega_b) t_{0K}} F_{ct} \left( \omega_{i,j} - \omega_b \right) e^{-im\omega_b t_L} \right]
\]

(C6)

If the Gaussian distribution (70) is a wide function - \( \sigma_{w} > 2\pi / \omega_b \), then the spectral function

\[
F \left( \omega_{i,j} - \omega_b \right) = e^{-\left( \omega_{i,j} - \omega_b \right)^2 \sigma_w^2 / 2}
\]

is a narrow function around a harmonic \( m=n \) that is resonant with the transition frequency:

\[
\omega_{i,j} = \omega_b.
\]

(C8)

Then only one harmonic - \( n \) can excite resonantly the transition in (C6):

28
\[ \Delta P^{(i)} = 2 \text{Re} \left\{ \frac{1}{2\pi \hbar v_0} \tilde{M}_{i,j} \left( \frac{\hbar \omega_{i,j}}{v_0} \right) C_i^* (t_{0K}) C_j (t_{0K}) \Gamma_i e^{i(\omega_{i,j} - n_0 \omega_b) t_{0K}} e^{-i\omega_{i,j} \sigma_a^2/2} \right\} \]  

(C9)

under the condition that it is phase-matched to the phase of the initial dipole moment \( C_i^* (t_{0K}) C_j (t_{0K}) \).

Likewise:

\[ \Delta P^{(2)} = \left| \Delta C_i \right|^2 = \left| \frac{1}{2\pi \hbar} \tilde{M}_{i,j} \left( \frac{\hbar \omega_{i,j}}{v_0} \right) C_j (t_{0K}) \right|^2 \left| \sum_m f_m e^{i(\omega_{i,j} - m\omega_b) t_{0K}} F_m \left( \omega_{i,j} - m\omega_b \right) e^{-i m\omega_b t_{0K}} \right|^2 = \]

\[ = \left| \frac{1}{2\pi \hbar v_0} \tilde{M}_{i,j} \left( \frac{\hbar \omega_{i,j}}{v_0} \right) C_j (t_{0K}) \right|^2 \left| f_m \right|^2 \left| F_m \left( \omega_{i,j} - n\omega_b \right) \right| \]

(C10)

\[ \Delta P^{(2)} = \left| \frac{1}{2\pi \hbar v_0} \tilde{M}_{i,j} \left( \frac{\hbar \omega_{i,j}}{v_0} \right) C_j (t_{0K}) \right|^2 \left| f_m \right|^2 \left( e^{-(\omega_{i,j} - n\omega_b)^2 \sigma_a^2} \right) \]

(C11)

Both incremental probabilities are dependent on the QEW shape and modulation features.
Appendix D: FEBERY interaction with multiple QEWs

Starting from equations (85), (86):

\[
\langle \left| \Psi_r^{(0)}(r,t) \right| \rangle^2 \to \sum_{k=1}^{N} \left( \left| \Psi_k^{(0)}(r,t) \right| \right)^2 \tag{D1}
\]

\[
\langle \left| \Psi_k^{(0)}(r,t) \right| \rangle^2 = \delta(r_\perp)f_{\text{et}}^k(t-t_{0K}-z/v_0)
\]

we substitute the N particles probability function

\[
f(t-t_0) = \sum_{k=1}^{N} \frac{1}{v} \int dz M_{i,j}(z) f_{\text{et}}(t-t_{0K}-z/v_0) \tag{D3}
\]

in (58), and changing order of integrations in z and t:

\[
C_i(t_{0N}^+) = C_i(t_0^-) + \frac{1}{2\pi i h v_0} \int dz M_{i,j}(z) \sum_{k=1}^{N} C_j(t_{0K}) \int dt e^{-i\omega_{i,j} t} f_{\text{et}}^i(t-t_{0K}-z/v_0) \tag{D4}
\]

With change of variables \( t' = t - z/v_0 \),

\[
C_i(t_{0N}^+) = C_i(t_0^-) + \frac{1}{2\pi i h v_0} \int dz M_{i,j}(z) \sum_{k=1}^{N} C_j(t_{0K}) \int dt' e^{-i\omega_{i,j} t'} f_{\text{et}}^i(t'-t_{0K})
\]

\[
= C_i(t_0^-) + \frac{1}{2\pi i h v_0} \tilde{M}_{i,j}(\frac{\hbar \omega_{i,j}}{v_0}) C_j(t_{0K}) \sum_{k=1}^{N} e^{i\omega_{i,j} t_{0K}} F_{\text{et}}(\omega_{i,j}) \tag{D5}
\]

where \( F_{\text{et}}(\omega) = \mathcal{F}\{f_{\text{et}}(t)\} \) is the Fourier transform of the single QEW probability function.

The incremental probability amplitude in (D5) averages to zero for random \( t_{0K} \), except when:

\[
\omega_{i,j} = n\omega_b \tag{D6}
\]

where

\[
t_{0K} = K2\pi/\omega_b \tag{D7}
\]

namely, when the QEWs arrive to the interaction point at a rate that is a sub-harmonic of the transition frequency \( \omega_{i,j} \). Take \( i=2, j=1 \) (upper and lower levels respectively), then with the approximation of small change in the amplitude:

\[
C_i(t_{0K}) \approx C_i(t_0^-) = \text{const} = 1, \quad \left| C_2(t_0^-) \right| << 1 \tag{D8}
\]

\[
C_2(t_{0N}^+)_{\omega_{i,j}=n\omega_b} \approx N \frac{1}{2\pi i h v} \tilde{M}_{2,1}(\frac{\hbar \omega_{2,1}}{v}) F_{\text{et}}(\omega_{2,1}) \tag{D9}
\]

For a Gaussian QEW,
\[ f_{e\tau} = \frac{1}{(2\pi \sigma_{e\tau}^2)^{1/2}} e^{-t^2/2\sigma_{e\tau}^2} \quad \text{(D10)} \]

\[ P_2 = N^2 \left\{ \frac{1}{2\pi \hbar v_0} \left| \tilde{M}_{2,1} \left( \frac{\hbar \omega_{2,1}}{v_0} \right) \right| \right\}^2 e^{-\alpha_{e\tau}^I \sigma_{e\tau}^I} \quad \text{(D11)} \]