Superradiance and subradiance due to quantum interference of entangled free electrons: Supplementary Material

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S1. Electric field autocorrelations for spontaneous emission by free electrons

In this section we show that the field-field quantum optical correlations of light $\langle \mathbf{E}^\dagger(\mathbf{r}',\omega')\mathbf{E}(\mathbf{r},\omega)\rangle$ emitted by free electrons, are related to the current-current quantum correlations of the electrons, $\langle \mathbf{j}^\dagger(\mathbf{r}',\omega')\mathbf{j}(\mathbf{r},\omega)\rangle_{\mathrm{el}}$, through a simple relation involving the dyadic Green's function of the medium, $\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)$, as follows:

$$\langle \mathbf{E}^\dagger(\mathbf{r}',\omega')\mathbf{E}(\mathbf{r},\omega)\rangle = \mu_0^2\omega\omega' \int d^3\mathbf{R}'\, \mathbf{G}^\dagger(\mathbf{r}',\mathbf{R}',\omega') \int d^3\mathbf{R}\, \mathbf{G}(\mathbf{r},\mathbf{R},\omega) \langle \mathbf{j}^\dagger(\mathbf{R}',\omega')\mathbf{j}(\mathbf{R},\omega)\rangle_{\mathrm{el}}$$

which we employ for analyzing the results of our paper. We shall derive this relation using quantum electrodynamics and perturbation theory, which is more commonly used in the literature for describing these processes.

First, we consider a system of a Dirac electron and radiation field with an initial density operator

$$\boldsymbol{\rho}_i = \sum_{\mathbf{i},\mathbf{i}'} \rho_{\mathrm{el}}(\mathbf{i},\mathbf{i}')|\mathbf{i}0\rangle\langle \mathbf{i}'0|, \qquad \text{(S1.1)}$$

where $\mathbf{i} = (\mathbf{k}_i, s_i)$ are pure spinor states with wavefunctions

$$\boldsymbol{\psi}_\mathbf{i} = \langle \mathbf{r}|\mathbf{i}\rangle = \frac{1}{\sqrt{(2\pi)^3}} \mathbf{u}_{s_i}(\mathbf{k}_i) e^{i\mathbf{k}_i\cdot\mathbf{r} - i\frac{E_i}{\hbar}t}, \qquad \text{(S1.2)}$$

where $E_i = \sqrt{\hbar^2k^2c^2 + m^2c^4}$ is the initial electron energy and $\mathbf{u}_{s_i}(\mathbf{k}_i)$ denotes the Dirac particle spinor. The state $|0\rangle$ denotes the vacuum of the electromagnetic (EM) field, which is quantized in a weakly dispersive, homogeneous medium as

$$\mathbf{A}(\mathbf{r},t) = \sum_{\mathbf{q}\sigma} \sqrt{\frac{\hbar v_{g\mathbf{q}\sigma}}{2\omega_{\mathbf{q}\sigma}\epsilon_0 n_{\mathbf{q}\sigma} c}}\, \boldsymbol{u}_{\mathbf{q}\sigma}(\mathbf{r}) e^{-i\omega_{\mathbf{q}\sigma}t} a_{\mathbf{q}\sigma} + h.c., \qquad \text{(S1.3)}$$

with the $\boldsymbol{u}_{\mathbf{q}\sigma}(\mathbf{r})$ denoting the EM modes of the Maxwell equations. After the interaction, the total density operator to first order in QED (post-selected to include only emission events via the projection operator $P = 1 - \sum_\mathbf{f}|\mathbf{f}0\rangle\langle \mathbf{f}0|$) is

$$\boldsymbol{\rho}_f = \sum_{\mathbf{i},\mathbf{i}'} \rho_{\mathrm{el}}(\mathbf{i},\mathbf{i}') \sum_{\mathbf{f};\mathbf{q}\sigma} M_{\mathbf{fi};\mathbf{q}\sigma} \sum_{\mathbf{f}';\mathbf{q}'\sigma'} M^*_{\mathbf{f}'\mathbf{i}';\mathbf{q}'\sigma'} \left|\mathbf{f}1_{\mathbf{q}\sigma}\right\rangle\left\langle\mathbf{f}'1_{\mathbf{q}'\sigma'}\right|, \qquad (\mathrm{S}1.4)$$

where $M_{\mathbf{fi};\mathbf{q}\sigma} = \frac{i}{\hbar} ec \int d\tau \Theta(t-\tau) \left\langle \mathbf{f}; 1_{\mathbf{q}\sigma} \middle| \boldsymbol{\alpha}\cdot\mathbf{A} \middle| \mathbf{i}; 0\right\rangle$ is the transition matrix element, and $\Theta(t)$ denotes the Heaviside step function. Explicitly the transition matrix element is given by

$$M_{\mathbf{fi};\mathbf{q}\sigma} = \frac{i}{\hbar}\sqrt{\frac{\hbar v_{g\mathbf{q}\sigma}}{2\omega_{\mathbf{q}\sigma}\epsilon_0 n_{\mathbf{q}\sigma}c}} \int d^3\mathbf{r}\, \boldsymbol{u}^*_{\mathbf{q}\sigma}(\mathbf{r}) \cdot \int d\tau \Theta(t-\tau)\left[ec\boldsymbol{\psi}^\dagger_{\mathbf{f}}(\mathbf{r},\tau)\boldsymbol{\alpha}\boldsymbol{\psi}_{\mathbf{i}}(\mathbf{r},\tau)\right]e^{i\omega_{\mathbf{q}\sigma}\tau}, \qquad (\mathrm{S}1.5)$$

The expression in the square brackets can be identified as the matrix element of the 3-current ***operator*** in first quantization:

$$\mathbf{j}(\mathbf{r}) = e\mathbf{n}(\mathbf{r})\mathbf{v} = e\delta(\mathbf{r}-\hat{\mathbf{r}})c\boldsymbol{\alpha}, \qquad (\mathrm{S}1.6)$$

note that in relativistic quantum mechanics, S1.6 is a Hermitian operator since $[\hat{\mathbf{r}},\boldsymbol{\alpha}] = 0$ making $\mathbf{v} = c\boldsymbol{\alpha}$. With this definition we find that

$$\begin{aligned}\langle\mathbf{f}|\mathbf{j}|\mathbf{i}\rangle = \langle\mathbf{f}(t)|\mathbf{j}(\mathbf{r})|\mathbf{i}(t)\rangle &= ec\langle\mathbf{f}(t)|\delta(\mathbf{r}-\hat{\mathbf{r}})\boldsymbol{\alpha}|\mathbf{i}(t)\rangle \\ &= ec\int d^3\mathbf{r}'\langle\mathbf{f}(t)|\delta(\mathbf{r}-\hat{\mathbf{r}})|\mathbf{r}'\rangle\langle\mathbf{r}'|\boldsymbol{\alpha}|\mathbf{i}(t)\rangle \\ &= ec\int d^3\mathbf{r}'\delta(\mathbf{r}-\mathbf{r}')\langle\mathbf{f}(t)|\mathbf{r}'\rangle\langle\mathbf{r}'|\boldsymbol{\alpha}|\mathbf{i}(t)\rangle = ec\langle\mathbf{f}(t)|\mathbf{r}\rangle\langle\mathbf{r}|\boldsymbol{\alpha}|\mathbf{i}(t)\rangle \\ &= ec\boldsymbol{\psi}^\dagger_{\mathbf{f}}(\mathbf{r},t)\boldsymbol{\alpha}\boldsymbol{\psi}_{\mathbf{i}}(\mathbf{r},t), \qquad (\mathrm{S}1.7)\end{aligned}$$

In the non-relativistic case, we can formally split the current operator into positive and negative frequency parts

$$\mathbf{j}(\mathbf{r}) = \mathbf{j}^{(+)}(\mathbf{r}) + \mathbf{j}^{(-)}(\mathbf{r}), \qquad (\mathrm{S}1.8a)$$

where

$$\mathbf{j}^{(+)}(\mathbf{r}) = \frac{e}{2m}\delta(\mathbf{r}-\hat{\mathbf{r}})\mathbf{p} = \mathbf{j}^{(-)\dagger}(\mathbf{r}), \qquad (\mathrm{S}1.8b)$$

The same ideas apply for replacing the Hermitian $\mathbf{j}(\mathbf{r})$ with $2\mathbf{j}^{(+)}(\mathbf{r})$, since in that case

$$\left\langle\mathbf{f}\middle|2\mathbf{j}^{(+)}(\mathbf{r})\middle|\mathbf{i}\right\rangle = e\psi^*_f(\mathbf{r},t)\frac{\hbar\mathbf{k}_i}{m}\psi_i(\mathbf{r},t), \qquad (\mathrm{S}1.8c)$$

the reason being that we consider only positive frequencies in the emission process. Since in emission processes $E_f < E_i$ it follows that the products $\psi^*_f(\mathbf{r},t)\psi_i(\mathbf{r},t) \propto e^{-i(E_i-E_f)t/\hbar}$ contain only positive frequencies.

And thus, writing $\mathbf{j}_{\mathrm{fi}}(\mathbf{r},t) = \langle\mathbf{f}|\mathbf{j}|\mathbf{i}\rangle$

$$M_{\mathbf{fi};\mathbf{q}\sigma} = \frac{i}{\hbar}\sqrt{\frac{\hbar v_{g\mathbf{q}\sigma}}{2\omega_{\mathbf{q}\sigma}\epsilon_0 n_{\mathbf{q}\sigma}c}} \int d^3\mathbf{r}\, \boldsymbol{u}^*_{\mathbf{q}\sigma}(\mathbf{r}) \cdot \int d\tau \Theta(t-\tau)\mathbf{j}_{\mathrm{fi}}(\mathbf{r},\tau)e^{i\omega_{\mathbf{q}\sigma}\tau}, \qquad (\mathrm{S}1.9)$$

When we detect only the photon component of the state, the electron's degrees of freedom are traced out, and the photon is described by a reduced density operator

$$\boldsymbol{\rho}_{\mathrm{ph}} = \mathrm{Tr}_{\mathrm{el}}\{\boldsymbol{\rho}_f\} = \sum_{\mathbf{q}\sigma}\sum_{\mathbf{q}'\sigma'}\sum_{\mathbf{f}}\sum_{\mathbf{i},\mathbf{i}'} \rho_{\mathrm{el}}(\mathbf{i};\mathbf{i}')M_{\mathbf{fi};\mathbf{q}\sigma}M^*_{\mathbf{fi}';\mathbf{q}'\sigma'}\left|1_{\mathbf{q}\sigma}\right\rangle\left\langle1_{\mathbf{q}'\sigma'}\right|, \qquad (\mathrm{S}1.10)$$

The positive frequency part of the electromagnetic field operator is

$$\mathbf{E}^{(+)}(\mathbf{r},t) = i\sum_{\mathbf{q}\sigma}\sqrt{\frac{\hbar\omega_{\mathbf{q}\sigma}v_{g\mathbf{q}\sigma}}{2\epsilon_0 n_{\mathbf{q}\sigma}c}}\boldsymbol{u}_{\mathbf{q}\sigma}(\mathbf{r})e^{-i\omega_{\mathbf{q}\sigma}t}a_{\mathbf{q}\sigma}, \qquad (\mathrm{S}1.11)$$

The field-field correlations are then

$$\left\langle \mathbf{E}^{(-)}(\mathbf{r}',t')\mathbf{E}^{(+)}(\mathbf{r},t)\right\rangle = \mathrm{Tr}\{\boldsymbol{\rho}_{\mathrm{ph}}\mathbf{E}^{(-)}(\mathbf{r}',t')\mathbf{E}^{(+)}(\mathbf{r},t)\}$$
$$= \sum_{\mathbf{f}}\sum_{\mathbf{i},\mathbf{i}'}\rho_{\mathrm{el}}(\mathbf{i};\mathbf{i}')\sum_{\mathbf{q}\sigma}\sum_{\mathbf{q}'\sigma'} M_{\mathbf{fi};\mathbf{q}\sigma}(t)M^*_{\mathbf{fi}';\mathbf{q}'\sigma'}(t')\left\langle 1_{\mathbf{q}'\sigma'}\right|\mathbf{E}^{(-)}(\mathbf{r}',t')\mathbf{E}^{(+)}(\mathbf{r},t)\left|1_{\mathbf{q}\sigma}\right\rangle$$
$$= \sum_{\mathbf{f}}\sum_{\mathbf{i},\mathbf{i}'}\rho_{\mathrm{el}}(\mathbf{i};\mathbf{i}')\sum_{\mathbf{q}\sigma}\sum_{\mathbf{q}'\sigma'} M_{\mathbf{fi};\mathbf{q}\sigma}(t)M^*_{\mathbf{fi}';\mathbf{q}'\sigma'}(t')\sqrt{\frac{\hbar\omega_{\mathbf{q}\sigma}v_{g\mathbf{q}\sigma}}{2\epsilon_0 n_{\mathbf{q}\sigma}c}}\sqrt{\frac{\hbar\omega_{\mathbf{q}'\sigma'}v_{g\mathbf{q}'\sigma'}}{2\epsilon_0 n_{\mathbf{q}'\sigma'}c}}\boldsymbol{u}^*_{\mathbf{q}'\sigma'}(\mathbf{r}')e^{i\omega_{\mathbf{q}'\sigma'}t'}\boldsymbol{u}_{\mathbf{q}\sigma}(\mathbf{r})e^{-i\omega_{\mathbf{q}\sigma}t},$$
(S1.12)

Substituting the expressions for $M_{\mathbf{fi};\mathbf{q}\sigma}(t)$ and employing index notation:

$$\left\langle E^{(-)}_{\alpha}(\mathbf{r}',t')E^{(+)}_{\beta}(\mathbf{r},t)\right\rangle$$
$$= \sum_{\mathbf{f}}\sum_{\mathbf{i},\mathbf{i}'}\rho_{\mathrm{el}}(\mathbf{i};\mathbf{i}')$$
$$\times\int d^3\mathbf{R}'\int d\tau'\left[\Theta(t'-\tau')\sum_{\mathbf{q}'\sigma'}\frac{v_{g\mathbf{q}'\sigma'}}{2\epsilon_0 n_{\mathbf{q}'\sigma'}c}u^*_{\mathbf{q}'\sigma',\alpha}(\mathbf{r}')u_{\mathbf{q}'\sigma',\gamma}(\mathbf{R}')e^{i\omega_{\mathbf{q}'\sigma'}(t'-\tau')}\right]j^*_{\mathrm{fi}',\gamma}(\mathbf{R}',\tau')$$
$$\times\int d^3\mathbf{R}\int d\tau\left[\Theta(t-\tau)\sum_{\mathbf{q}\sigma}\frac{v_{g\mathbf{q}\sigma}}{2\epsilon_0 n_{\mathbf{q}\sigma}c}u_{\mathbf{q}\sigma,\beta}(\mathbf{r})u^*_{\mathbf{q}\sigma,\delta}(\mathbf{R})e^{-i\omega_{\mathbf{q}\sigma}(t-\tau)}\right]j_{\mathrm{fi},\delta}(\mathbf{R},\tau), \qquad (\mathrm{S1.13})$$

Note that the Fourier transform of the square brackets is

$$\int dte^{i\omega t}\,\Theta(t)\sum_{\mathbf{q}\sigma}\frac{v_{g\mathbf{q}\sigma}}{2\epsilon_0 n_{\mathbf{q}\sigma}c}u_{\mathbf{q}\sigma,\beta}(\mathbf{r})u^*_{\mathbf{q}\sigma,\delta}(\mathbf{R})e^{-i(\omega_{\mathbf{q}\sigma}-i0^+)t}$$
$$= i\sum_{\mathbf{q}\sigma}\frac{v_{g\mathbf{q}\sigma}}{2\epsilon_0 n_{\mathbf{q}\sigma}c}\frac{u_{\mathbf{q}\sigma,\beta}(\mathbf{r})u^*_{\mathbf{q}\sigma,\delta}(\mathbf{R})}{\omega-\omega_{\mathbf{q}\sigma}+i0^+}, \qquad (\mathrm{S1.14})$$

and that the dyadic Green tensor is

$$G_{\beta\delta}(\mathbf{r},\mathbf{R},\omega) = \sum_{\mathbf{q}\sigma}\frac{cv_{g\mathbf{q}\sigma}}{n_{\mathbf{q}\sigma}}\frac{u_{\mathbf{q}\sigma,\beta}(\mathbf{r})u^*_{\mathbf{q}\sigma,\delta}(\mathbf{R})}{\omega^2_{\mathbf{q}\sigma}-\omega^2}$$
$$= -\sum_{\mathbf{q}\sigma}\frac{cv_{g\mathbf{q}\sigma}}{2\omega_{\mathbf{q}\sigma}n_{\mathbf{q}\sigma}}u_{\mathbf{q}\sigma,\beta}(\mathbf{r})u^*_{\mathbf{q}\sigma,\delta}(\mathbf{R})\left[\frac{1}{\omega-\omega_{\mathbf{q}\sigma}+i0^+}-\frac{1}{\omega+\omega_{\mathbf{q}\sigma}+i0^+}\right]$$
$$= -\frac{1}{\mu_0\omega}\sum_{\mathbf{q}\sigma}\frac{v_{g\mathbf{q}\sigma}}{2\epsilon_0 n_{\mathbf{q}\sigma}c}\frac{u_{\mathbf{q}\sigma,\beta}(\mathbf{r})u^*_{\mathbf{q}\sigma,\delta}(\mathbf{R})}{\omega-\omega_{\mathbf{q}\sigma}+i0^+}$$
$$-\frac{1}{\mu_0\omega}\sum_{\mathbf{q}\sigma}\frac{v_{g\mathbf{q}\sigma}}{2\epsilon_0 n_{\mathbf{q}\sigma}c}\frac{u_{\mathbf{q}\sigma,\beta}(\mathbf{r})u^*_{\mathbf{q}\sigma,\delta}(\mathbf{R})}{\omega+\omega_{\mathbf{q}\sigma}+i0^+}, \qquad (\mathrm{S1.15})$$

Therefore Eq. (S1.14) can be simplified to

$$i\sum_{\mathbf{q}\sigma}\frac{v_{g\mathbf{q}\sigma}}{2\epsilon_0 n_{\mathbf{q}\sigma}c}\frac{u_{\mathbf{q}\sigma,\beta}(\mathbf{r})u^*_{\mathbf{q}\sigma,\delta}(\mathbf{R})}{\omega-\omega_{\mathbf{q}\sigma}+i0^+} = -i\Theta(\omega)\mu_0\omega G_{\beta\delta}(\mathbf{r},\mathbf{R},\omega), \qquad (\mathrm{S1.16})$$

i.e., the positive frequency part of the Green tensor. Now define the frequency field in the following manner:

$$\mathbf{E}^{(+)}(\mathbf{r},t) = \int_0^\infty d\omega e^{-i\omega t}\mathbf{E}(\mathbf{r},\omega), \qquad (\mathrm{S1.17}a)$$

$$\mathbf{E}^{(-)}(\mathbf{r},t) = \int_0^\infty d\omega e^{i\omega t}\mathbf{E}^\dagger(\mathbf{r},\omega)\,, \qquad (\text{S}1.17b)$$

Identifying Eq. (S1.13) as a convolution and moving to the frequency domain one has

$$\begin{aligned}&\left\langle E_\alpha^\dagger(\mathbf{r}',\omega')E_\beta(\mathbf{r},\omega)\right\rangle \\ &\qquad = \mu_0^2\omega\omega'\int d^3\mathbf{R}'\, G_{\alpha\gamma}^*(\mathbf{r}',\mathbf{R}',\omega')\int d^3\mathbf{R}\, G_{\beta\delta}(\mathbf{r},\mathbf{R},\omega) \\ &\qquad \times\sum_{\mathbf{f}}\sum_{\mathbf{i},\mathbf{i}'}\rho_{\text{el}}(\mathbf{i};\mathbf{i}')j_{\text{fi}',\gamma}^*(\mathbf{R}',\omega')j_{\text{fi},\delta}(\mathbf{R},\omega)\,, \qquad (\text{S}1.18)\end{aligned}$$

The double-sum expression in the above equation can be simplified further, because

$$\begin{aligned}\sum_{\mathbf{f}}\sum_{\mathbf{i},\mathbf{i}'}\rho_{\text{el}}(\mathbf{i};\mathbf{i}')\mathbf{j}_{\text{fi}'}^*(\mathbf{R}',\omega')\mathbf{j}_{\text{fi}}(\mathbf{R},\omega) &= \sum_{\mathbf{f}}\sum_{\mathbf{i},\mathbf{i}'}\rho_{\text{el}}(\mathbf{i};\mathbf{i}')\left\langle\mathbf{i}'\middle|\mathbf{j}^\dagger(\mathbf{R}',\omega')\middle|\mathbf{f}\right\rangle\langle\mathbf{f}|\mathbf{j}(\mathbf{R},\omega)|\mathbf{i}\rangle \\ &= \sum_{\mathbf{i},\mathbf{i}'}\rho_{\text{el}}(\mathbf{i};\mathbf{i}')\left\langle\mathbf{i}'\middle|\mathbf{j}^\dagger(\mathbf{R}',\omega')\mathbf{j}(\mathbf{R},\omega)\middle|\mathbf{i}\right\rangle = \text{Tr}\left\{\rho_{\text{el}}\mathbf{j}^\dagger(\mathbf{R}',\omega')\mathbf{j}(\mathbf{R},\omega)\right\} \\ &= \left\langle\mathbf{j}^\dagger(\mathbf{R}',\omega')\mathbf{j}(\mathbf{R},\omega)\right\rangle_{\text{el}}\end{aligned}$$

Hence we arrive at the key result

$$\boxed{\left\langle\mathbf{E}^\dagger(\mathbf{r}',\omega')\mathbf{E}(\mathbf{r},\omega)\right\rangle = \mu_0^2\omega\omega'\int d^3\mathbf{R}'\,\mathbf{G}^\dagger(\mathbf{r}',\mathbf{R}',\omega')\int d^3\mathbf{R}\,\mathbf{G}(\mathbf{r},\mathbf{R},\omega)\left\langle\mathbf{j}^\dagger(\mathbf{R}',\omega')\mathbf{j}(\mathbf{R},\omega)\right\rangle_{\text{el}}, \qquad (\text{S}1.19)}$$

So, a knowledge of the current correlations $\left\langle\mathbf{j}^\dagger(\mathbf{R}',\omega')\mathbf{j}(\mathbf{R},\omega)\right\rangle_{\text{el}}$ of the electrons provides the value for the electric field autocorrelation$\left\langle\mathbf{E}^\dagger(\mathbf{r}',\omega')\mathbf{E}(\mathbf{r},\omega)\right\rangle$.

S2. Current correlations of free quantum emitters

In this section we wish to show that the quantum current-current correlations of free charged particles appearing in Eq. (S1.19) can be re-written in terms of the first- and second-order correlation functions of the particle field, $G_{\text{e}}^{(1)}$ and $G_{\text{e}}^{(2)}$, respectively, as:

$$\langle\mathbf{j}(\mathbf{x}')\mathbf{j}(\mathbf{x})\rangle_{\text{el}} = e^2\mathbf{v}_0\mathbf{v}_0\left[G_{\text{e}}^{(2)}(\mathbf{x}',\mathbf{x}) + \delta(\mathbf{x}-\mathbf{x}')G_{\text{e}}^{(1)}(\mathbf{x},\mathbf{x})\right]$$

with $\mathbf{x} = \mathbf{x}(\mathbf{r},t) = \mathbf{r} - \mathbf{v}_0 t$ and where $\mathbf{v}_0$ is the carrier velocity shared by the particle wavepackets under the paraxial approximation. This will help us connect particle quantum correlations and optical quantum correlations, which is at the core of super- and subradiance.

By a "free quantum emitter", we refer to particles which emit waves as they propagate through a medium due to recoil of their center of mass motion, and not due to any internal degree of freedom like an atom. Below we detail how to derive the current-current correlations for an arbitrary number of particles, wherein electrons emitting Cherenkov radiation are considered as a case study.

First quantization

In first quantization, a generalization of the current operator to N particles is:

$$\mathbf{j}(\mathbf{r}) = ec\sum_{i=1}^N \delta\left(\mathbf{r}^{(i)} - \mathbf{r}\right)\boldsymbol{\alpha}^{(i)}\,, \qquad (\text{S}2.2)$$

in the relativistic case, and

$$\mathbf{j}^{(+)}(\mathbf{r}) = \frac{e}{2m}\sum_{i=1}^{N} \delta\big(\mathbf{r}^{(i)} - \mathbf{r}\big)\mathbf{p}^{(i)}, \qquad \text{(S2.1)}$$

in the non-relativistic case, with $\mathbf{j}(\mathbf{r}) \to 2\mathbf{j}^{(+)}(\mathbf{r})$ in the equations derived above (see boxed comment following Eq. ()).

In order to avoid the redundancy of first quantization when dealing with identical particles, from now on we'll employ second quantization to describe both the current operator and the electron state ρ_{el}.

Second quantization: nonrelativistic current

Now we promote the wavefunction $\psi(\mathbf{r}, t)$ to an **operator** $\hat{\psi}(\mathbf{r}, t)$.

In second quantization of either bosons, $\left[b_{\mathbf{k}}, b^{\dagger}_{\mathbf{k}'}\right] = \delta_{\mathbf{kk}'}$ or fermions $\left\{b_{\mathbf{k}}, b^{\dagger}_{\mathbf{k}'}\right\} = \delta_{\mathbf{kk}'}$ the current density *operator* of free, massive particles described by nonrelativistic quantum mechanics is

$$\mathbf{j}(\mathbf{r}, t) = \frac{e\hbar}{2im}\left[\hat{\psi}^{\dagger}(\mathbf{r}, t)\nabla\hat{\psi}(\mathbf{r}, t) - \left(\nabla\hat{\psi}^{\dagger}(\mathbf{r}, t)\right)\hat{\psi}(\mathbf{r}, t)\right] = \mathbf{j}^{(+)}(\mathbf{r}, t) + \mathbf{j}^{(-)}(\mathbf{r}, t), \qquad \text{(S2.3)}$$

where $\hat{\psi}(\mathbf{r}, t) = \sum_{\mathbf{k}} \frac{e^{i\left(\mathbf{k}\cdot\mathbf{r} - \frac{E_{\mathbf{k}}}{\hbar}t\right)}}{\sqrt{V}} b_{\mathbf{k}}$ is the position space annihilation operator in the Heisenberg picture for a free particle. Let us now find a simple expression for the current that holds for free particles in the approximation of zero-recoil, as well as in the paraxial approximation. Our results will qualitatively hold even upon relaxation of these approximations. Substituting the wavefunctions into the current operator we find

$$\mathbf{j}(\mathbf{r}, t) = \frac{e\hbar}{2mV}\left[\sum_{\mathbf{k}'}\sum_{\mathbf{k}} e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}} e^{-i\frac{E_{\mathbf{k}} - E_{\mathbf{k}'}}{\hbar}t} (\mathbf{k} + \mathbf{k}')\, b^{\dagger}_{\mathbf{k}'} b_{\mathbf{k}}\right]. \quad \text{(S2.4)}$$

Taking the Fourier transform, we find:

$$\mathbf{j}(\mathbf{q}, t) = \int d^3\mathbf{r} e^{-i\mathbf{q}\cdot\mathbf{r}} \mathbf{j}(\mathbf{r}, t) = \frac{e\hbar}{mV}\sum_{\mathbf{k}} e^{i\frac{E_{\mathbf{k}-\mathbf{q}} - E_{\mathbf{k}}}{\hbar}t}\left(\mathbf{k} - \frac{\mathbf{q}}{2}\right) b^{\dagger}_{\mathbf{k}-\mathbf{q}} b_{\mathbf{k}}. \quad \text{(S2.5)}$$

In the paraxial and zero-recoil limits, we replace $\mathbf{k}$ with $\mathbf{k}_0$ and demand $q \ll k_0$ such that $\frac{E_{\mathbf{k}-\mathbf{q}} - E_{\mathbf{k}}}{\hbar} = \mathbf{q}\cdot\mathbf{v}_0$. We then find for both bosons and fermions:

$$\mathbf{j}(\mathbf{q}, t) = e\mathbf{v}_0 e^{-i\mathbf{q}\cdot\mathbf{v}_0 t}\frac{1}{V}\sum_{\mathbf{k}} b^{\dagger}_{\mathbf{k}-\mathbf{q}} b_{\mathbf{k}}, \qquad \text{(S2.6)}$$

where $\mathbf{v}_0$ denotes the emitter's carrier velocity. Going back to the position-space representation, we have:

$$\mathbf{j}(\mathbf{r}, t) = e\mathbf{v}_0\hat{\psi}^{\dagger}(\mathbf{r} - \mathbf{v}_0 t)\hat{\psi}(\mathbf{r} - \mathbf{v}_0 t), \qquad \text{(S2.7)}$$

It turns out that this approximate result for the Hermitian current operator $\mathbf{j}(\mathbf{r}, t)$, also holds if we choose to start from the nonrelativistic positive-frequency current as in the first-quantization case, applying the same approximations:

$$2\mathbf{j}^{(+)}(\mathbf{r},t) = 2\frac{e\hbar}{2im}\hat{\psi}^\dagger(\mathbf{r},t)\nabla\hat{\psi}(\mathbf{r},t) = e\mathbf{v}_0\hat{\psi}^\dagger(\mathbf{r}-\mathbf{v}_0 t)\hat{\psi}(\mathbf{r}-\mathbf{v}_0 t)$$

making the non-Hermiticity of $\mathbf{j}^{(+)}(\mathbf{r},t)$ negligible up to corrections to the paraxial and zero-recoil approximations. Thus, from here onwards we shall refer to Eq. (S2.7) as the nonrelativistic current operator.

Second quantization: relativistic current

Similarly, now we promote the spinor $\boldsymbol{\psi}(\mathbf{r},t)$ to an **operator** $\hat{\boldsymbol{\psi}}(\mathbf{r},t)$.

Here, we start from the 4-current

$$j^\mu = \bar{\hat{\boldsymbol{\psi}}}\gamma^\mu\hat{\boldsymbol{\psi}} = \hat{\boldsymbol{\psi}}^\dagger\gamma^0\gamma^\mu\hat{\boldsymbol{\psi}}, \qquad \text{(S2.8)}$$

The Hermitian 3-current is, in turn,

$$j^i = ec\hat{\boldsymbol{\psi}}^\dagger\gamma^0\gamma^i\hat{\boldsymbol{\psi}} = ec\hat{\boldsymbol{\psi}}^\dagger\alpha^i\hat{\boldsymbol{\psi}}, \qquad \text{(S2.9)}$$

Let us write

$$\hat{\boldsymbol{\psi}}(\mathbf{r},t) = \sum_{\mathbf{p},\sigma} b_{\mathbf{p}\sigma}\mathbf{u}_{\mathbf{p}\sigma}\frac{e^{i(\mathbf{p}\cdot\mathbf{r}-E_{\mathbf{p}\sigma}t)/\hbar}}{\sqrt{V}}, \qquad \text{(S2.10)}$$

with $\sigma = \uparrow_+, \downarrow_+, \uparrow_-, \downarrow_-$ denoting spin and particle/antiparticle. Substituting, we have that

$$\mathbf{j}(\mathbf{r},t) = ec\hat{\boldsymbol{\psi}}^\dagger\boldsymbol{\alpha}\hat{\boldsymbol{\psi}} = \frac{ec}{V}\sum_{\mathbf{p},\sigma}\sum_{\mathbf{p}',\sigma'} b^\dagger_{\mathbf{p}'\sigma'}b_{\mathbf{p}\sigma}\mathbf{u}^\dagger_{\mathbf{p}'\sigma'}\boldsymbol{\alpha}\mathbf{u}_{\mathbf{p}\sigma}e^{i\left[(\mathbf{p}-\mathbf{p}')\cdot\mathbf{r}-\left(E_{\mathbf{p}\sigma}-E_{\mathbf{p}'\sigma'}\right)t\right]/\hbar}, \qquad \text{(S2.11)}$$

Although a thorough treatment of the role of spin and recoil corrections in CR was discussed in Ref. [50], for simplicity we choose here to neglect any recoil and spin-flip contributions to the current operator. Recoil corrections are typically orders of magnitude smaller than the electron momentum, making this approximation quite accurate. Also, we keep only contributions where a particle scatters to a particle (no particle-antiparticle transitions), and thus restrict ourselves to $\sigma = \uparrow, \downarrow$. Again, employing the zero-recoil and paraxial approximations, we find that:

$$\mathbf{u}^\dagger_{\mathbf{p}'\sigma'}\boldsymbol{\alpha}\mathbf{u}_{\mathbf{p}\sigma} \cong \frac{1}{c}\mathbf{v}_0\delta_{\sigma\sigma'}, \qquad \text{(S2.12)}$$

$$E_{\mathbf{p}} - E_{\mathbf{p}'} \cong (\mathbf{p}-\mathbf{p}')\cdot\mathbf{v}_0, \qquad \text{(S2.13)}$$

$$\mathbf{j}(\mathbf{r},t) \cong \frac{e\mathbf{v}_0}{V}\sum_\sigma\sum_{\mathbf{k},\mathbf{k}'} b^\dagger_{\mathbf{k}'\sigma}b_{\mathbf{k}\sigma}e^{i(\mathbf{k}-\mathbf{k}')\cdot(\mathbf{r}-\mathbf{v}_0 t)}, \qquad \text{(S2.14)}$$

where now the carrier velocity is $\mathbf{v}_0 = \frac{\mathbf{p}_0}{m\gamma}$. Now, since $\{b_{\mathbf{k}\sigma}, b^\dagger_{\mathbf{k}'\sigma'}\} = \delta_{\mathbf{k}\mathbf{k}'}\delta_{\sigma\sigma'}$, if we redefine for each spin σ a scalar operator

$$\hat{\psi}_\sigma(\mathbf{r}-\mathbf{v}_0 t) = \sum_{\mathbf{k}} b_{\mathbf{k}\sigma}\frac{e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{v}_0 t)}}{\sqrt{V}}, \qquad \text{(S2.15)}$$

Then the spatial anti-commutation relation (at equal times) of these new operators becomes

$$\{\hat{\psi}_\sigma(\mathbf{r}-\mathbf{v}_0t),\hat{\psi}^\dagger_{\sigma'}(\mathbf{r}'-\mathbf{v}_0t)\}=\sum_{\mathbf{k}}\frac{e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{v}_0t)}}{\sqrt{V}}\sum_{\mathbf{k}'}\{b_{\mathbf{k}\sigma},b^\dagger_{\mathbf{k}'\sigma'}\}\frac{e^{-i\mathbf{k}'\cdot(\mathbf{r}'-\mathbf{v}_0t)}}{\sqrt{V}}$$
$$=\delta_{\sigma\sigma'}\sum_{\mathbf{k}}\frac{e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}}{V}=\delta_{\sigma\sigma'}\delta(\mathbf{r}-\mathbf{r}'),\qquad \text{(S2.16)}$$

from which it follows that

$$\mathbf{j}(\mathbf{r},t)=e\mathbf{v}_0\sum_\sigma\hat{\psi}^\dagger_\sigma(\mathbf{r}-\mathbf{v}_0t)\hat{\psi}_\sigma(\mathbf{r}-\mathbf{v}_0t),\qquad \text{(S2.17)}$$

quite similarly to the non-relativistic case.

Current correlations

Since, for both nonrelativistic and relativistic particles, the current operator obtains a similar form, we may proceed by analysing the general behaviour. The current correlations appearing in Eq. (S1.20) can be rewritten

$$\langle j_\alpha(\mathbf{r}',t')j_\beta(\mathbf{r},t)\rangle_{\mathrm{el}}=e^2v_{0\alpha}v_{0\beta}\sum_{\sigma'}\sum_\sigma\langle\hat{\psi}^\dagger_{\sigma'}(\mathbf{r}'-\mathbf{v}_0t')\hat{\psi}_{\sigma'}(\mathbf{r}'-\mathbf{v}_0t')\hat{\psi}^\dagger_\sigma(\mathbf{r}-\mathbf{v}_0t)\hat{\psi}_\sigma(\mathbf{r}-\mathbf{v}_0t)\rangle,$$
$$\text{(S2.18)}$$

We bring the expression $\langle\hat{\psi}^\dagger_{\sigma'}\hat{\psi}_{\sigma'}\hat{\psi}^\dagger_\sigma\hat{\psi}_\sigma\rangle$ to normal order using the commutation relations for bosons, or anticommutation relations for fermions, to obtain the same results:

$$\langle j_\alpha(\mathbf{r}',t')j_\beta(\mathbf{r},t)\rangle_{\mathrm{el}}$$
$$=e^2v_{0\alpha}v_{0\beta}\sum_{\sigma'}\sum_\sigma\langle\hat{\psi}^\dagger_{\sigma'}(\mathbf{r}'-\mathbf{v}_0t')\hat{\psi}^\dagger_\sigma(\mathbf{r}-\mathbf{v}_0t)\hat{\psi}_\sigma(\mathbf{r}-\mathbf{v}_0t)\hat{\psi}_{\sigma'}(\mathbf{r}'-\mathbf{v}_0t')\rangle$$
$$+e^2v_{0\alpha}v_{0\beta}\delta[\mathbf{r}-\mathbf{r}'-\mathbf{v}_0(t-t')]\sum_\sigma\langle\hat{\psi}^\dagger_\sigma(\mathbf{r}-\mathbf{v}_0t)\hat{\psi}_\sigma(\mathbf{r}-\mathbf{v}_0t)\rangle,\qquad \text{(S2.19)}$$

Now, define the first and second order correlation functions of the emitters as

$$G^{(1)}_{\mathrm{e}}(\mathbf{x},\mathbf{x})=\sum_\sigma\langle\hat{\psi}^\dagger_\sigma(\mathbf{x})\hat{\psi}_\sigma(\mathbf{x})\rangle=\rho_{\mathrm{e}}(\mathbf{x},\mathbf{x}),\qquad \text{(S2.20)}$$

$$G^{(2)}_{\mathrm{e}}(\mathbf{x}',\mathbf{x})=\sum_{\sigma'}\sum_\sigma\langle\hat{\psi}^\dagger_{\sigma'}(\mathbf{x}')\hat{\psi}^\dagger_\sigma(\mathbf{x})\hat{\psi}_\sigma(\mathbf{x})\hat{\psi}_{\sigma'}(\mathbf{x}')\rangle,\qquad \text{(S2.21)}$$

with $\mathbf{x}=\mathbf{r}-\mathbf{v}_0t$ and $\rho_{\mathrm{e}}(\mathbf{x},\mathbf{x})$ is the diagonal of the emitter density matrix, so that

$$\langle j_\alpha(\mathbf{x}')j_\beta(\mathbf{x})\rangle_{\mathrm{el}}=e^2v_{0\alpha}v_{0\beta}G^{(2)}(\mathbf{x}',\mathbf{x})+e^2v_{0\alpha}v_{0\beta}\delta(\mathbf{x}-\mathbf{x}')G^{(1)}_{\mathrm{e}}(\mathbf{x},\mathbf{x}).\ \text{(S2.22)}$$

In Fourier space:

$$\langle j^\dagger_\alpha(\mathbf{q}',\omega')j_\beta(\mathbf{q},\omega)\rangle_{\mathrm{el}}$$
$$=\int dt'\int d^3\mathbf{r}'\,e^{-i(\mathbf{q}'\cdot\mathbf{r}'-\omega't')}\int dt\int d^3\mathbf{r}e^{i(\mathbf{q}\cdot\mathbf{r}-\omega t)}\,\langle j^\dagger_\alpha(\mathbf{r}',t')j_\beta(\mathbf{r},t)\rangle_{\mathrm{el}}$$
$$=(2\pi)^2e^2v_{0\alpha}v_{0\beta}\delta(\omega'-\mathbf{q}'\cdot\mathbf{v}_0)\delta(\omega-\mathbf{q}\cdot\mathbf{v}_0)$$
$$\times\left[\iint d^3\mathbf{x}\int d^3\mathbf{x}'\,e^{i\mathbf{q}\cdot\mathbf{x}-i\mathbf{q}'\cdot\mathbf{x}'}G^{(2)}_{\mathrm{e}}(\mathbf{x}',\mathbf{x})+\int d^3\mathbf{x}e^{i(\mathbf{q}-\mathbf{q}')\cdot\mathbf{x}}G^{(1)}_{\mathrm{e}}(\mathbf{x},\mathbf{x})\right],$$
$$\text{(S2.23)}$$

S3. Total radiated power of coherent Cathodoluminescence

In this section we combine the results of the previous sections to obtain the emitted optical power spectrum of any coherent cathodoluminescence process, and then focusing on Cherenkov radiation.

The radiated power detected in the far-field, at a distance r and direction $\hat{\mathbf{n}}$ is proportional to the time-average of the quantum expectation value

$$\frac{dP}{d\Omega} = 2r^2\epsilon_0 c\frac{1}{T}\int dt\,\langle|\mathbf{E}(r\hat{\mathbf{n}},t)|^2\rangle = 2r^2\epsilon_0 c\frac{1}{2\pi T}\int d\omega\,\langle|\mathbf{E}(r\hat{\mathbf{n}},\omega)|^2\rangle, \qquad \text{(S3.1)}$$

and, per unit frequency, reads

$$\frac{d^2P}{d\Omega d\omega} = 2r^2\epsilon_0 c\frac{1}{2\pi T}\langle|\mathbf{E}(r\hat{\mathbf{n}},\omega)|^2\rangle. \quad \text{(S3.2)}$$

Using Eq. (S1.19) let us write the expression for the field spectral intensity

$$\langle|\mathbf{E}(\mathbf{r},\omega)|^2\rangle = \mathrm{Tr}\langle\mathbf{E}^\dagger(\mathbf{r},\omega)\mathbf{E}(\mathbf{r},\omega)\rangle$$
$$= \mu_0^2\omega^2\int d^3\mathbf{R}'\int d^3\mathbf{R}\,\mathrm{Tr}\mathbf{G}^\dagger(\mathbf{r},\mathbf{R}',\omega)\mathbf{G}(\mathbf{r},\mathbf{R},\omega)\langle\mathbf{j}^\dagger(\mathbf{R}',\omega')\mathbf{j}(\mathbf{R},\omega)\rangle_{\mathrm{el}},$$
$$\text{(S3.3)}$$

so that, we find the general result:

$$\boxed{\frac{d^2P}{d\Omega d\omega} = 2r^2\epsilon c\mu_0^2\omega^2\frac{1}{2\pi T}\int d^3\mathbf{R}'\int d^3\mathbf{R}\,\mathrm{Tr}\mathbf{G}^\dagger(\mathbf{r},\mathbf{R}',\omega)\mathbf{G}(\mathbf{r},\mathbf{R},\omega)\langle\mathbf{j}^\dagger(\mathbf{R}',\omega)\mathbf{j}(\mathbf{R},\omega)\rangle_{\mathrm{el}}. \quad \text{(S3.4)}}$$

For example, for Cherenkov radiation, the far field Green function is

$$\mathbf{G}(\mathbf{r},\mathbf{r}',\omega) = \frac{e^{iqr}}{4\pi r}(\mathbf{I}-\hat{\mathbf{r}}\hat{\mathbf{r}})e^{-i\mathbf{q}\cdot\mathbf{r}'}, \qquad \text{(S3.5)}$$

where $\mathbf{q} = \hat{\mathbf{r}}n\omega/c$ is the wavevector of the emitted radiation in the observation direction. So, we find

$$\frac{d^2P}{d\Omega d\omega} = \frac{1}{(4\pi)^2}\frac{1}{2\pi T}2\epsilon_0 c\mu_0^2\omega^2\sin^2\theta\left\langle j_z^\dagger\left(\frac{n\omega}{c}\hat{\mathbf{n}},\omega\right)j_z\left(\frac{n\omega}{c}\hat{\mathbf{n}},\omega\right)\right\rangle_{\mathrm{el}}, \qquad \text{(S3.6)}$$

with

$$\left\langle j_z^\dagger\left(\frac{n\omega}{c}\hat{\mathbf{n}},\omega\right)j_z\left(\frac{n\omega}{c}\hat{\mathbf{n}},\omega\right)\right\rangle_{\mathrm{el}}$$
$$= \frac{(2\pi)^2e^2v_0^2}{\omega n\beta}\underbrace{\delta(0)}_{T/2\pi}\,\delta\left(\cos\theta-\frac{1}{n\beta}\right)$$
$$\times\left[\iint d^3\mathbf{x}G_{\mathrm{e}}^{(1)}(\mathbf{x},\mathbf{x}) + \int d^3\mathbf{x}\int d^3\mathbf{x}'\,e^{i\mathbf{q}\cdot(\mathbf{x}-\mathbf{x}')}G_{\mathrm{e}}^{(2)}(\mathbf{x}',\mathbf{x})\right], \qquad \text{(S3.7)}$$

Finally giving for Cherenkov radiation:

$$\boxed{\frac{d^2P}{d\Omega d\omega} = \frac{\hbar\omega\alpha\beta}{2\pi}\sin^2\theta\,\delta\left(\cos\theta-\frac{1}{n\beta}\right)\left[\iint d^3\mathbf{x}G_{\mathrm{e}}^{(1)}(\mathbf{x},\mathbf{x}) + \int d^3\mathbf{x}\int d^3\mathbf{x}'\,e^{i\mathbf{q}\cdot(\mathbf{x}-\mathbf{x}')}G_{\mathrm{e}}^{(2)}(\mathbf{x}',\mathbf{x})\right], \text{(S3.8)}}$$

S4. Entangled vs. classically correlated emitter pairs

In this section we calculate the emission rates from pairs of free emitters, showing that one obtains a new kind of super- and sub-radiance for entangled pairs, that cannot be achieved by a classically-correlated pair.

We now examine how the emission pattern of a general spontaneous emission process from free emitters is influenced by the quantum mechanical state of an emitter pair. Consider a two-electron state prepared in two spectrally distinguishable wavepacket states: $\varphi_1(\mathbf{r}), \varphi_2(\mathbf{r})$ and different spins ↑↓. The wavepackets share a common carrier wavevector $\mathbf{k}_0$, and differ by a small wavevector difference $\pm\boldsymbol{\kappa}$ $(\kappa \ll k_0)$ in the following manner.

The relation between the wavepackets $\varphi_1(\mathbf{r}), \varphi_2(\mathbf{r})$ (decoupled from their respective spinors) and their shifted momentum representation $\phi(\mathbf{k})$ is then

$$\varphi_{1,2}(\mathbf{r}) = \int d^3\mathbf{k}\,\phi(\mathbf{k} - \mathbf{k}_0 \mp \boldsymbol{\kappa})e^{i(\mathbf{k}\cdot\mathbf{r} - \omega_k t)}$$
$$\cong e^{\pm i\boldsymbol{\kappa}\cdot(\mathbf{r} - \mathbf{v}_0 t)}\varphi(\mathbf{r} - \mathbf{v}_0 t)e^{i(\mathbf{k}_0\cdot\mathbf{r} - \omega_{|\mathbf{k}_0|}t)}, \qquad (S4.1)$$

In what follows, it is useful for us to define (with $\mathbf{x} = \mathbf{r} - \mathbf{v}_0 t$)

$$\varphi_{1,2}(\mathbf{x}) = e^{\pm i\boldsymbol{\kappa}\cdot\mathbf{x}}\varphi(\mathbf{x}). \quad (S4.2)$$

In Eq. (S.4.1), we take walk-off effects (longitudinal and transverse) due to the small difference in the group velocities of $\varphi_1(\mathbf{r})$ and $\varphi_2(\mathbf{r})$ to be negligible as we shall assume a large wavefunction in the respective dimension. The overlap between the wavepackets is given as:

$$\langle\varphi_2|\varphi_1\rangle = \int d^3\mathbf{r}\varphi_2^*(\mathbf{r})\varphi_1(\mathbf{r}) = \int d^3\mathbf{r}|\varphi(\mathbf{r})|^2 e^{2i\boldsymbol{\kappa}\cdot\mathbf{r}}$$
$$= \begin{cases} 0, \text{when } |\varphi(\mathbf{r})|^2 \text{ is wider than } \dfrac{2\pi}{\kappa} \text{ in the } \hat{\boldsymbol{\kappa}} \text{ direction} \\ 1, \text{when } |\varphi(\mathbf{r})|^2 \text{ is narrower than } \dfrac{2\pi}{\kappa} \text{ in the } \hat{\boldsymbol{\kappa}} \text{ direction} \end{cases}. \quad (S4.3)$$

This same condition, together with the requirement of negligible temporal walkoff, can be recast in momentum space as

$$\Delta k \ll |\mathbf{k} - \mathbf{k}'| \ll \frac{m}{2\hbar t\Delta k}, \qquad (S4.4)$$

where Δk is the wavepacket coherent momentum uncertainty and t the interaction time. For $\Delta k \to 0$ this condition is naturally satisfied.

Below we consider the regime where $\langle\varphi_2|\varphi_1\rangle \to 0$, in agreement with the extended-wavefunction approximation that we employ.

Classically correlated state:

We first consider a classical (probabilistic) correlation between the two electrons, wherein if one is found in state φ_1 ↑, it is correlated with the other to be found in φ_2 ↓, and vice versa. The density operator defining this state is

$$\rho = \frac{1}{2}|\varphi_{1\uparrow}\varphi_{2\downarrow}\rangle\langle\varphi_{1\uparrow}\varphi_{2\downarrow}| + \frac{1}{2}|\varphi_{2\uparrow}\varphi_{1\downarrow}\rangle\langle\varphi_{2\uparrow}\varphi_{1\downarrow}|, \qquad (S4.5)$$

One easily shows that for this density matrix, the correlation functions are

$$G_{\mathrm{e}}^{(1)}(\mathbf{x},\mathbf{x}) = \sum_{\sigma}\left\langle\hat{\psi}_{\sigma}^{\dagger}(\mathbf{x})\hat{\psi}_{\sigma}(\mathbf{x})\right\rangle = 2|\varphi(\mathbf{x})|^2, \qquad \text{(S4.6)}$$

$$G_{\mathrm{e}}^{(2)}(\mathbf{x}',\mathbf{x}) = \sum_{\sigma'}\sum_{\sigma}\left\langle\hat{\psi}_{\sigma'}^{\dagger}(\mathbf{x}')\hat{\psi}_{\sigma}^{\dagger}(\mathbf{x})\hat{\psi}_{\sigma}(\mathbf{x})\hat{\psi}_{\sigma'}(\mathbf{x}')\right\rangle = 2|\varphi(\mathbf{x})|^2|\varphi(\mathbf{x}')|^2, \qquad \text{(S4.7)}$$

According to Eq. (2) in the main text,

$$\left\langle\mathbf{j}^{\dagger}(\mathbf{x}')\mathbf{j}(\mathbf{x})\right\rangle = e^2\mathbf{v}_0\mathbf{v}_0\left[G_{\mathrm{e}}^{(2)}(\mathbf{x}',\mathbf{x}) + \delta(\mathbf{x}-\mathbf{x}')G_{\mathrm{e}}^{(1)}(\mathbf{x},\mathbf{x})\right], \qquad \text{(S4.8)}$$

this means that the current fluctuations of the *classically* correlated state are

$$\left\langle\mathbf{j}^{\dagger}(\mathbf{x}')\mathbf{j}(\mathbf{x})\right\rangle_{\mathrm{c}} = e^2\mathbf{v}_0\mathbf{v}_0[2|\varphi(\mathbf{x})|^2|\varphi(\mathbf{x}')|^2 + 2\delta(\mathbf{x}-\mathbf{x}')|\varphi(\mathbf{x})|^2], \qquad \text{(S4.9)}$$

<u>Entangled state</u>:

We now consider the emitters being prepared in an entangled, pure state (Bell state):

$$|\Psi\rangle = \frac{|\varphi_{1\uparrow}\varphi_{2\downarrow}\rangle + e^{i\zeta}|\varphi_{1\downarrow}\varphi_{2\uparrow}\rangle}{\sqrt{2}}, \qquad \text{(S4.10)}$$

with ζ being a phase factor. We emphasize that this state is written in the Fock basis using wavepacket quantization, and that the two electrons considered here are distinguishable by spin. Computing the correlation functions using:

$$\hat{\psi}_{\uparrow}(\mathbf{r})|\Psi\rangle = \frac{\varphi_1^*(\mathbf{r})|\varphi_{2\downarrow}\rangle - e^{i\zeta}\varphi_2^*(\mathbf{r})|\varphi_{1\downarrow}\rangle}{\sqrt{2}}, \qquad \text{(S4.11)}$$

$$\hat{\psi}_{\downarrow}(\mathbf{r})|\Psi\rangle = \frac{-\varphi_2^*(\mathbf{r})|\varphi_{1\uparrow}\rangle + e^{i\zeta}\varphi_1^*(\mathbf{r})|\varphi_{2\uparrow}\rangle}{\sqrt{2}}, \qquad \text{(S4.12)}$$

$$\hat{\psi}_{\downarrow}(\mathbf{r}')\hat{\psi}_{\uparrow}(\mathbf{r})|\Psi\rangle = \frac{\varphi_1^*(\mathbf{r})\varphi_2^*(\mathbf{r}') - e^{i\zeta}\varphi_2^*(\mathbf{r})\varphi_1^*(\mathbf{r}')}{\sqrt{2}}|0\rangle, \qquad \text{(S4.13)}$$

$$\hat{\psi}_{\uparrow}(\mathbf{r}')\hat{\psi}_{\downarrow}(\mathbf{r})|\Psi\rangle = \frac{-\varphi_1^*(\mathbf{r}')\varphi_2^*(\mathbf{r}) + e^{i\zeta}\varphi_1^*(\mathbf{r})\varphi_2^*(\mathbf{r}')}{\sqrt{2}}|0\rangle, \qquad \text{(S4.14)}$$

gives:

$$G_{\mathrm{e}}^{(1)}(\mathbf{x},\mathbf{x}) = 2|\varphi(\mathbf{x})|^2, \qquad \text{(S4.15)}$$

and, via direct calculation

$$\begin{aligned}
&\langle\Psi|\hat{\psi}_{\uparrow}^{\dagger}(\mathbf{r})\hat{\psi}_{\downarrow}^{\dagger}(\mathbf{r}')\hat{\psi}_{\downarrow}(\mathbf{r}')\hat{\psi}_{\uparrow}(\mathbf{r})|\Psi\rangle \\
&\quad = \frac{\left[\varphi_1(\mathbf{r})\varphi_2(\mathbf{r}') - e^{-i\zeta}\varphi_2(\mathbf{r})\varphi_1(\mathbf{r}')\right]\left[\varphi_1^*(\mathbf{r})\varphi_2^*(\mathbf{r}') - e^{i\zeta}\varphi_2^*(\mathbf{r})\varphi_1^*(\mathbf{r}')\right]}{2} \\
&\quad = |\varphi(\mathbf{r})|^2|\varphi(\mathbf{r}')|^2 - \frac{1}{2}e^{i\zeta}e^{i2\boldsymbol{\kappa}\cdot(\mathbf{r}-\mathbf{r}')}|\varphi(\mathbf{r})|^2|\varphi(\mathbf{r}')|^2 \\
&\quad - \frac{1}{2}e^{-i\zeta}e^{-i2\boldsymbol{\kappa}\cdot(\mathbf{r}-\mathbf{r}')}|\varphi(\mathbf{r})|^2|\varphi(\mathbf{r}')|^2
\end{aligned}$$

$$\langle\Psi|\hat{\psi}_\downarrow^\dagger(\mathbf{r})\hat{\psi}_\uparrow^\dagger(\mathbf{r}')\hat{\psi}_\uparrow(\mathbf{r}')\hat{\psi}_\downarrow(\mathbf{r})|\Psi\rangle$$
$$= \frac{\left[-\varphi_1(\mathbf{r}')\varphi_2(\mathbf{r}) + e^{-i\zeta}\varphi_1(\mathbf{r})\varphi_2(\mathbf{r}')\right]\left[-\varphi_1^*(\mathbf{r}')\varphi_2^*(\mathbf{r}) + e^{i\zeta}\varphi_1^*(\mathbf{r})\varphi_2^*(\mathbf{r}')\right]}{2}$$
$$= |\varphi(\mathbf{r})|^2|\varphi(\mathbf{r}')|^2 - \frac{1}{2}e^{-i\zeta}e^{i2\boldsymbol{\kappa}\cdot(\mathbf{r}-\mathbf{r}')}|\varphi(\mathbf{r})|^2|\varphi(\mathbf{r}')|^2$$
$$-\frac{1}{2}e^{i\zeta}e^{-i2\boldsymbol{\kappa}\cdot(\mathbf{r}-\mathbf{r}')}|\varphi(\mathbf{r})|^2|\varphi(\mathbf{r}')|^2$$

we find

$$G_\mathrm{e}^{(2)}(\mathbf{x}',\mathbf{x}) = 2|\varphi(\mathbf{x})|^2|\varphi(\mathbf{x}')|^2 - \cos\zeta\, 2|\varphi(\mathbf{x})|^2|\varphi(\mathbf{x}')|^2\cos[\Delta\mathbf{k}\cdot(\mathbf{x}-\mathbf{x}')], \qquad \text{(S4.16)}$$

with $\Delta\mathbf{k} = 2\boldsymbol{\kappa}$.

<u>Comparison of the emission patterns</u>:

As a consequence of Eq. (1) of the main text, it is obvious that the emission patterns satisfy the relation

$$P_\mathrm{ent} = P_\mathrm{c} - \cos\zeta\, P_\mathrm{q}, \qquad \text{(S4.17)}$$

which shows that spontaneous emission is sensitive to the phase of the entangled emitter pair. This effect cannot be described using the classical correlation.

For Cherenkov radiation specifically, we find

$$\frac{d^2P}{d\Omega d\omega} = \frac{\hbar\omega\alpha\beta}{2\pi}\sin^2\theta\,\delta\left(\cos\theta - \frac{1}{n\beta}\right)\left\{\underbrace{2 + 2\left|\int d^3\mathbf{x}e^{-i\frac{n\omega}{c}\hat{\mathbf{r}}\cdot\mathbf{x}}|\varphi(\mathbf{x})|^2\right|^2}_{\text{classical}}\right.$$
$$\left. - \cos\zeta\underbrace{\left[\left|\int d^3\mathbf{x}e^{-i\left(\frac{n\omega}{c}\hat{\mathbf{r}}-\Delta\mathbf{k}\right)\cdot\mathbf{x}}|\varphi(\mathbf{x})|^2\right|^2 + \left|\int d^3\mathbf{x}e^{-i\left(\frac{n\omega}{c}\hat{\mathbf{r}}+\Delta\mathbf{k}\right)\cdot\mathbf{x}}|\varphi(\mathbf{x})|^2\right|^2\right]}_{\text{quantum}}\right\},$$
(S4.18)

Now, we model the wavepacket as Gaussian with transverse width Δr_{Te} and longitudinal length Δz_e:

$$|\varphi(\mathbf{x})|^2 = \frac{1}{(2\pi)^{\frac{3}{2}}\Delta z_e\Delta r_{T\mathrm{e}}^2}e^{-\frac{x^2+y^2}{2\Delta r_{T\mathrm{e}}^2}}e^{-\frac{z^2}{2\Delta z_e^2}}, \qquad \text{(S4.19)}$$

This gives

$$\int d^3\mathbf{x}e^{-i\frac{n\omega}{c}\hat{\mathbf{r}}\cdot\mathbf{x}}|\varphi(\mathbf{x})|^2 = e^{-\frac{\omega^2\cos^2\theta}{2(c/n\Delta z_\mathrm{e})^2}}e^{-\frac{\omega^2\sin^2\theta}{2(c/n\Delta r_{T\mathrm{e}})^2}} = e^{-\frac{\omega^2}{2(v/\Delta z_\mathrm{e})^2}}e^{-\frac{\omega^2\sin^2\theta}{2(c/n\Delta r_{T\mathrm{e}})^2}}, \qquad \text{(S4.20)}$$

and

$$\int d^3\mathbf{r}|\varphi(\mathbf{r})|^2e^{i\Delta\mathbf{k}\cdot\mathbf{r}} = e^{-\frac{1}{2}\Delta z_\mathrm{e}^2\Delta k_z^2}e^{-\frac{1}{2}\Delta r_{T\mathrm{e}}^2\Delta k_T^2}. \text{ (S4.21)}$$

<u>**First case**</u>: longitudinal beat $\Delta\mathbf{k} = \Delta k\hat{\mathbf{z}}$:

$$\int d^3\mathbf{x}e^{-i\left(\frac{n\omega}{c}\hat{\mathbf{r}}\mp\Delta\mathbf{k}\right)\cdot\mathbf{x}}|\varphi(\mathbf{x})|^2 = e^{-\frac{(\omega\mp v\Delta k)^2}{2(v/\Delta z_\mathrm{e})^2}}e^{-\frac{\omega^2\sin^2\theta}{2(c/n\Delta r_{T\mathrm{e}})^2}}, \qquad \text{(S4.22)}$$

by choosing the wave function dimensions to satisfy

$$\frac{v}{\Delta z_e} \ll \omega \ll \frac{c}{n\Delta r_{T\mathrm{e}}}, \qquad \text{(S4.23)}$$

(e.g., a longitudinally long and transversely narrow wavefunction with respect to the emitted wavelength) and ensuring $\Delta z_e \Delta k \gg 1$, we have

$$\frac{d^2P}{d\Omega d\omega} = \frac{\hbar\omega\alpha\beta}{2\pi}\sin^2\theta\,\delta\left(\cos\theta - \frac{1}{n\beta}\right)\left\{2 + 2e^{-\frac{\omega^2}{2(v/\Delta z_\mathrm{e})^2}} - \cos\zeta\, e^{-\frac{(\omega - v\Delta k)^2}{(v/\Delta z_\mathrm{e})^2}}\right\}, \qquad \text{(S4.24)}$$

Integrated over angles, the emission rate becomes:

$$\Gamma(\omega) = \Gamma_0\left[2 + 2e^{-\frac{\omega^2}{2(v/\Delta z_\mathrm{e})^2}} - \cos\zeta\, e^{-\frac{(\omega - v\Delta k)^2}{(v/\Delta z_\mathrm{e})^2}}\right], \qquad \text{(S4.25)}$$

with $\Gamma_0 = \alpha\beta\sin^2\theta_c$ denoting the classical emission rate for one particle. So, at the frequency $\omega = v\Delta k$ corresponding to the beat note we can obtain a peak or a dip in the emission spectrum, as a function of the phase angle ζ.

<u>Second case</u>: transverse beat $\Delta\mathbf{k} = \Delta k\hat{\mathbf{x}}$:

Choosing a transverse beat $\Delta k = \frac{n\omega}{c}\sin\theta$ corresponding to the transverse emission recoil, we have that

$$\int d^3\mathbf{x} e^{-i\left(\frac{n\omega}{c}\hat{\mathbf{r}} \mp \Delta\mathbf{k}\right)\cdot\mathbf{x}}|\varphi(\mathbf{x})|^2 = e^{-\frac{\omega^2}{2(v/\Delta z_\mathrm{e})^2}} e^{-\frac{1\mp\cos\varphi}{(1/\Delta k\Delta r_{T\mathrm{e}})^2}}, \qquad \text{(S4.26)}$$

and, by choosing the wave function dimensions to satisfy

$$\frac{c}{n\sin\theta\,\Delta r_{T\mathrm{e}}} \ll \omega \ll \frac{v}{\Delta z_\mathrm{e}}, \qquad \text{(S4.27)}$$

e.g., a transversely wide and longitudinally short wavefunction with respect to the emitted wavelength, we have that

$$\frac{d^2P}{d\Omega d\omega} = \frac{\hbar\omega\alpha\beta}{2\pi}\sin^2\theta\,\delta\left(\cos\theta - \frac{1}{n\beta}\right)\Big\{2 - \cos\zeta\left[e^{-2(\Delta k\Delta r_{T\mathrm{e}})^2(1-\cos\varphi)} + e^{-2(\Delta k\Delta r_{T\mathrm{e}})^2(1+\cos\varphi)}\right]\Big\}, \qquad \text{(S4.28)}$$

Integrated over θ, we obtain the emission pattern on the Cherenkov cone per unit frequency

$$\frac{1}{\Gamma_0}\frac{d\Gamma}{d\varphi} = 2 - 2\cos\zeta\, e^{-\eta}\cosh(\eta\cos\varphi), \qquad \text{(S4.29)}$$

with $\Gamma_0 = \frac{\alpha\beta}{2\pi}\sin^2\theta_\mathrm{c}$ is the classical emission pattern and $\eta = 2(\Delta k\Delta r_{T\mathrm{e}})^2 \gg 1$ for a transversely wide wavefunction. The expression $e^{-\eta}\cosh(\eta\cos\varphi)$ equals $1/2$ at $\varphi = 0, \pi$ and vanishes otherwise. Thus, the emission pattern demonstrates two new peaks (or dips) at $\varphi = 0, \pi$ on the Cherenkov cone, corresponding to the direction of the momentum beat. The new emission pattern depends on the phase angle ζ of the electron Bell state.

S5. Comparison with emission by electron product states and classical superradiance by delayed electrons

This section shows that classical super- and subradiance (due to, for example, to a temporal delay between free electrons or a spatio-temporal modulation of the electron charge density) is qualitatively and quantitatively different from the super- and subradiance effects that we find due to entanglement.

Now let us show that classical super- and subradiance due to (for example) a temporal delay between free electrons can be told apart from the quantum effect. Let us consider first two electrons of waepackets $\varphi_1(\mathbf{r})$ and $\varphi_2(\mathbf{r})$. Without loss of generality let these electrons be distinguishable (e.g. by spin), such that their quantum state is a product state

$$|\Psi\rangle = |\varphi_{1\uparrow}\varphi_{2\downarrow}\rangle, \qquad \text{(S5.1)}$$

Computing the correlation functions using:

$$\hat{\psi}_\uparrow(\mathbf{r})|\Psi\rangle = \varphi_1^*(\mathbf{r})|\varphi_{2\downarrow}\rangle, \qquad \text{(S5.2)}$$

$$\hat{\psi}_\downarrow(\mathbf{r})|\Psi\rangle = -\varphi_2^*(\mathbf{r})|\varphi_{1\uparrow}\rangle, \qquad \text{(S5.3)}$$

$$\hat{\psi}_\downarrow(\mathbf{r}')\hat{\psi}_\uparrow(\mathbf{r})|\Psi\rangle = \varphi_2^*(\mathbf{r}')\varphi_1^*(\mathbf{r})|0\rangle, \qquad \text{(S5.4)}$$

$$\hat{\psi}_\uparrow(\mathbf{r}')\hat{\psi}_\downarrow(\mathbf{r})|\Psi\rangle = -\varphi_1^*(\mathbf{r}')\varphi_2^*(\mathbf{r})|0\rangle, \qquad \text{(S5.6)}$$

We find

$$G_\mathrm{e}^{(1)}(\mathbf{r},\mathbf{r}) = |\varphi_1(\mathbf{r})|^2 + |\varphi_2(\mathbf{r})|^2, \qquad \text{(S5.7)}$$

$$G_\mathrm{e}^{(2)}(\mathbf{r},\mathbf{r}') = |\varphi_1(\mathbf{r})|^2|\varphi_2(\mathbf{r}')|^2 + |\varphi_1(\mathbf{r}')|^2|\varphi_2(\mathbf{r})|^2, \qquad \text{(S5.8)}$$

Giving the classical result

$$\frac{d^2P}{d\Omega d\omega} = \frac{\hbar\omega\alpha\beta}{2\pi}\sin^2\theta\,\delta\left(\cos\theta - \frac{1}{n\beta}\right)\left\{2 + 2\mathrm{Re}\int d^3\mathbf{x}\int d^3\mathbf{x}'\,e^{-i\frac{n\omega}{c}\hat{\mathbf{n}}\cdot(\mathbf{x}-\mathbf{x}')}|\varphi_1(\mathbf{x})|^2|\varphi_2(\mathbf{x}')|^2\right\}, \qquad \text{(S5.9)}$$

in comparison, for the entangled state

$$|\Psi\rangle = \frac{|\varphi_{1\uparrow}\varphi_{2\downarrow}\rangle + e^{i\zeta}|\varphi_{1\downarrow}\varphi_{2\uparrow}\rangle}{\sqrt{2}}, \qquad \text{(S5.10)}$$

we obtain the quantum result

$$\frac{d^2P}{d\Omega d\omega} = \frac{\hbar\omega\alpha\beta}{2\pi}\sin^2\theta\,\delta\left(\cos\theta - \frac{1}{n\beta}\right)\left\{\underbrace{2 + 2\mathrm{Re}\int d^3\mathbf{x}\int d^3\mathbf{x}'\,e^{-i\frac{n\omega}{c}\hat{\mathbf{n}}\cdot(\mathbf{x}-\mathbf{x}')}|\varphi_1(\mathbf{x})|^2|\varphi_2(\mathbf{x}')|^2}_{\text{classical}}\right.$$
$$\left. - \underbrace{\cos\zeta\int d^3\mathbf{x}\int d^3\mathbf{x}'\,e^{-i\frac{n\omega}{c}\hat{\mathbf{n}}\cdot(\mathbf{x}-\mathbf{x}')}2\mathrm{Re}\{\varphi_1(\mathbf{x})\varphi_2^*(\mathbf{x})\varphi_2(\mathbf{x}')\varphi_1^*(\mathbf{x}')\}}_{\text{quantum}}\right\}, \qquad \text{(S5.11)}$$

Clearly, there is a distinct difference between the entangled and product states, as long as the quantum interference term does not vanish identically. This property does not depend on the choice of wavepacket modes φ_1 and φ_2.

S6. Electron-electron decoherence and entanglement generation by post-selection

In this section we derive the final state of a system of free electrons (a single particle or a pair of opposite spins) following spontaneous emission of a photon in an arbitrary optical medium. The results are summarized in the table below.

Property	one electron	two electrons of opposite spins
Initial pure state	$\lvert\psi_{1\mathrm{el}}^{(i)}\rangle = \sum_{\mathbf{k}} \psi_{1\mathrm{el}}^{(i)}(\mathbf{k})\lvert\mathbf{k}\rangle$	$\lvert\psi_{2\mathrm{el}}^{(i)}\rangle = \sum_{\mathbf{k}_1\mathbf{k}_2} \psi_{2\mathrm{el}}^{(i)}(\mathbf{k}_1,\mathbf{k}_2)\lvert\mathbf{k}_1\mathbf{k}_2\rangle$ (may or may not be entangled)
Interaction	$H_{\mathrm{int}} = \frac{e}{m}\mathbf{A}(\hat{\mathbf{r}})\cdot\hat{\mathbf{p}}$	$H_{\mathrm{int}} = \frac{e}{m}\sum_{j=1}^{2}\mathbf{A}(\hat{\mathbf{r}}_j)\cdot\hat{\mathbf{p}}_j$
Optical medium response	$\mathrm{Im}\mathbf{G}(\mathbf{q},\omega)$ (uniform, dispersive and lossy medium)	
Recoiled state, for a given recoil $\mathbf{q}$	$\lvert\psi_{1\mathrm{rec}}^{(i)}(\mathbf{q})\rangle = \sum_{\mathbf{k}} \psi_{1\mathrm{el}}^{(i)}(\mathbf{k})\lvert\mathbf{k}-\mathbf{q}\rangle$	$\lvert\psi_{2\mathrm{rec}}^{(i)}(\mathbf{q})\rangle = \frac{1}{\sqrt{N_{\mathbf{q}}}}\sum_{\mathbf{k}_1\mathbf{k}_2} \psi_{2\mathrm{el}}^{(i)}(\mathbf{k}_1,\mathbf{k}_2)(\lvert\mathbf{k}_1-\mathbf{q};\mathbf{k}_2\rangle + \lvert\mathbf{k}_1;\mathbf{k}_2-\mathbf{q}\rangle)$ $N_{\mathbf{q}} = 2 + 2\mathrm{Re}\sum_{\mathbf{k}_1\mathbf{k}_2} \psi_{2\mathrm{el}}^{(i)*}(\mathbf{k}_1+\mathbf{q},\mathbf{k}_2-\mathbf{q})\psi_{2\mathrm{el}}^{(i)}(\mathbf{k}_1,\mathbf{k}_2)$
Final mixed state	$\boldsymbol{\rho}_{1\mathrm{el}}^{(f)} = \int d^3\mathbf{q}\, p_{1\mathbf{q}}\, \lvert\psi_{1\mathrm{rec}}^{(i)}(\mathbf{q})\rangle\langle\psi_{1\mathrm{rec}}^{(i)}(\mathbf{q})\rvert$	$\boldsymbol{\rho}_{2\mathrm{el}}^{(f)} = \int d^3\mathbf{q}\, p_{2\mathbf{q}}\, \lvert\psi_{2\mathrm{rec}}^{(i)}(\mathbf{q})\rangle\langle\psi_{2\mathrm{rec}}^{(i)}(\mathbf{q})\rvert$
Recoil probabilities	$p_{1\mathbf{q}} = \frac{\mathrm{Im}G_{zz}(\mathbf{q},\mathbf{q}\cdot\mathbf{v})}{\int d^3\mathbf{q}\,\mathrm{Im}\,G_{zz}(\mathbf{q},\mathbf{q}\cdot\mathbf{v})}$	$p_{2\mathbf{q}} = \frac{N_{\mathbf{q}}\mathrm{Im}G_{zz}(\mathbf{q},\mathbf{q}\cdot\mathbf{v})}{\int d^3\mathbf{q}\,N_{\mathbf{q}}\mathrm{Im}\,G_{zz}(\mathbf{q},\mathbf{q}\cdot\mathbf{v})}$
Post-selection chain	$\lvert\mathbf{k}\rangle \to \lvert\mathbf{k}-\mathbf{q}\rangle \to \lvert\mathbf{k}-2\mathbf{q}\rangle \ldots$	$\lvert\mathbf{k};\mathbf{k}\rangle \to \frac{\lvert\mathbf{k}-\mathbf{q};\mathbf{k}\rangle + \lvert\mathbf{k};\mathbf{k}-\mathbf{q}\rangle}{\sqrt{2}}$ $\to \frac{\lvert\mathbf{k};\mathbf{k}-2\mathbf{q}\rangle + \lvert\mathbf{k}-2\mathbf{q};\mathbf{k}\rangle + 2\lvert\mathbf{k}-\mathbf{q};\mathbf{k}-\mathbf{q}\rangle}{\sqrt{6}} \to \cdots$ (two-electron entanglement creation)

We investigate what happens to a free-electron system (single or two distinguishable electrons) after interaction with an optical medium. We consider a pure initial state:

$$\text{one electron:}\quad \lvert\psi_{1\mathrm{el}}^{(i)}\rangle = \sum_{\mathbf{k}} \psi_{1\mathrm{el}}^{(i)}(\mathbf{k})\lvert\mathbf{k}\rangle\,, \qquad (\mathrm{S}6.1a)$$

$$\text{two electrons:}\quad \lvert\psi_{2\mathrm{el}}^{(i)}\rangle = \sum_{\mathbf{k}_1\mathbf{k}_2} \psi_{2\mathrm{el}}^{(i)}(\mathbf{k}_1,\mathbf{k}_2)\lvert\mathbf{k}_1\mathbf{k}_2\rangle\,, \qquad (\mathrm{S}6.1b)$$

The electronic quantum state decoheres, but at the same time, retains a partial coherence.[1] The reason is that the final state can be written as a statistical mixture of ***recoiled copies*** of the original initial state

$$\text{one recoiled electron:}\quad \lvert\psi_{1\mathrm{rec}}^{(i)}(\mathbf{q})\rangle = \sum_{\mathbf{k}} \psi_{1\mathrm{el}}^{(i)}(\mathbf{k})\lvert\mathbf{k}-\mathbf{q}\rangle\,, \qquad (\mathrm{S}6.2a)$$

$$\text{two recoiled electrons:}\quad \lvert\psi_{2\mathrm{rec}}^{(i)}(\mathbf{q})\rangle = \frac{1}{\sqrt{N_{\mathbf{q}}}}\sum_{\mathbf{k}_1\mathbf{k}_2} \psi_{2\mathrm{el}}^{(i)}(\mathbf{k}_1,\mathbf{k}_2)(\lvert\mathbf{k}_1-\mathbf{q};\mathbf{k}_2\rangle + \lvert\mathbf{k}_1;\mathbf{k}_2-\mathbf{q}\rangle)\,,$$
$$(\mathrm{S}6.2b)$$

for the latter state the normalization is state-dependent:

[1] For a single electron we have seen (in the first Cherenkov paper) that this partial coherence is sufficient to conserve the off-diagonal momentum coherence $\int d^3\mathbf{k}\langle\mathbf{k}+\mathbf{q}\rvert\boldsymbol{\rho}\lvert\mathbf{k}-\mathbf{q}\rangle$ that determines the optical coherence of the emitted light.

$$N_{\mathbf{q}} = 2 + 2\mathrm{Re}\sum_{\mathbf{k}_1\mathbf{k}_2} \psi_{2\mathrm{el}}^{(i)*}(\mathbf{k}_1+\mathbf{q},\mathbf{k}_2-\mathbf{q})\psi_{2\mathrm{el}}^{(i)}(\mathbf{k}_1,\mathbf{k}_2), \qquad \text{(S6.3)}$$

its value changes according to the interference between recoil paths (the same mechanism that results in super- and subradiance of light by the two-electron system).

The final states then read:

$$\boldsymbol{\rho}_{1\mathrm{el}}^{(f)} = \int d^3\mathbf{q}p_{1\mathbf{q}}\,|\psi_{1\mathrm{rec}}^{(i)}(\mathbf{q})\rangle\langle\psi_{1\mathrm{rec}}^{(i)}(\mathbf{q})|, \qquad \text{with} \quad p_{1\mathbf{q}} = \frac{\mathrm{Im}G_{zz}(\mathbf{q},\mathbf{q}\cdot\mathbf{v})}{\int d^3\mathbf{q}\mathrm{Im}\,G_{zz}(\mathbf{q},\mathbf{q}\cdot\mathbf{v})}, \text{(S6.4}a\text{)}$$

$$\boldsymbol{\rho}_{2\mathrm{el}}^{(f)} = \int d^3\mathbf{q}p_{2\mathbf{q}}\,|\psi_{2\mathrm{rec}}^{(i)}(\mathbf{q})\rangle\langle\psi_{2\mathrm{rec}}^{(i)}(\mathbf{q})|, \qquad \text{with} \quad p_{2\mathbf{q}} = \frac{N_{\mathbf{q}}\mathrm{Im}G_{zz}(\mathbf{q},\mathbf{q}\cdot\mathbf{v})}{\int d^3\mathbf{q}N_{\mathbf{q}}\mathrm{Im}\,G_{zz}(\mathbf{q},\mathbf{q}\cdot\mathbf{v})}, \text{(S6.4}b\text{)}$$

by interpreting $p_{1\mathbf{q}}$ and $p_{2\mathbf{q}}$ as emission probabilities, we see that for a single electron $p_{1\mathbf{q}}$ is ***wavefunction-independent***, and that $p_{2\mathbf{q}}$ is ***wavefunction-dependent*** because of the $N_{\mathbf{q}}$ term. As noted earlier, this exactly corresponds to the super- and subradiant emission we found in our paper. Here, the optical medium response $\mathrm{Im}G_{zz}(\mathbf{q},\omega)$ serves as a weight for different emission processes according to their relative strength.

Final state purity

The purities can be found via

$$\text{purity} = \mathrm{Tr}\left[\boldsymbol{\rho}_{n\mathrm{el}}^{(f)}\right]^2 = \int d^3\mathbf{q}\int d^3\mathbf{q}' p_{n\mathbf{q}}\,p_{n\mathbf{q}'}\left|\langle\psi_{n\mathrm{rec}}^{(i)}(\mathbf{q}')|\psi_{n\mathrm{rec}}^{(i)}(\mathbf{q})\rangle\right|^2, \qquad \text{(S6.5)}$$

with $n = 1{,}2$. The purity is maximal (purity → 1), if all recoiled states significantly overlap in momentum space (e.g., due to a large initial momentum uncertainty), giving

$$\text{purity} = \int d^3\mathbf{q}\int d^3\mathbf{q}' p_{n\mathbf{q}}\,p_{n\mathbf{q}'}\underbrace{\left|\langle\psi_{n\mathrm{rec}}^{(i)}(\mathbf{q}')|\psi_{n\mathrm{rec}}^{(i)}(\mathbf{q})\rangle\right|^2}_{\to 1} = \int d^3\mathbf{q}p_{n\mathbf{q}}\int d^3\mathbf{q}'\,p_{n\mathbf{q}'} = 1, \qquad \text{(S6.6)}$$

this justifies the classical limit of point particles $|\psi|^2 \to \delta(\mathbf{r}-\mathbf{v}t)$ with infinite momentum uncertainty: in this limit the state does not change upon recoil and stays pure.

The other limit is that of minimal purity, obtained when the recoiled states are all orthogonal to each other (e.g., when starting with plane-wave electrons), giving

$$\text{purity} = \int d^3\mathbf{q}\int d^3\mathbf{q}' p_{n\mathbf{q}}\,p_{n\mathbf{q}'}\underbrace{\left|\langle\psi_{n\mathrm{rec}}^{(i)}(\mathbf{q}')|\psi_{n\mathrm{rec}}^{(i)}(\mathbf{q})\rangle\right|^2}_{\to\delta(\mathbf{q}-\mathbf{q}')} = \int d^3\mathbf{q}p_{n\mathbf{q}}^2, \qquad \text{(S6.7)}$$

For both a single electron and an electron pair, the purity then satisfies

$$\frac{\int d^3\mathbf{q}|\mathrm{Im}\,G_{zz}(\mathbf{q},\mathbf{q}\cdot\mathbf{v})|^2}{|\int d^3\mathbf{q}\mathrm{Im}\,G_{zz}(\mathbf{q},\mathbf{q}\cdot\mathbf{v})|^2} < \text{purity} < 1, \qquad \text{(S6.8)}$$

Post selection

This decoherence however, does not imply that all quantum coherence is lost: if one post-selects a recoil $\mathbf{q}$ by detecting a photon $\mathbf{q}$ with $\omega = \mathbf{q}\cdot\mathbf{v}$, the resulting single- or two-electron state "collapses" to the recoiled subspace, and becomes the corresponding recoiled copy: $|\psi_{n\mathrm{rec}}^{(i)}(\mathbf{q})\rangle$, $n = 1{,}2$ which by itself is a ***pure state***. Within this recoiled state all the original quantum coherences of the original initial quantum state are preserved.

Interestingly, one may repeat this process, to ***create*** entanglement between two initially separable electrons:

$$|\mathbf{k};\mathbf{k}\rangle \to \frac{|\mathbf{k}-\mathbf{q};\mathbf{k}\rangle + |\mathbf{k};\mathbf{k}-\mathbf{q}\rangle}{\sqrt{2}} \to \frac{|\mathbf{k};\mathbf{k}-2\mathbf{q}\rangle + |\mathbf{k}-2\mathbf{q};\mathbf{k}\rangle + 2|\mathbf{k}-\mathbf{q};\mathbf{k}-\mathbf{q}\rangle}{\sqrt{6}} \to \cdots , \quad \text{(S6.9)}$$

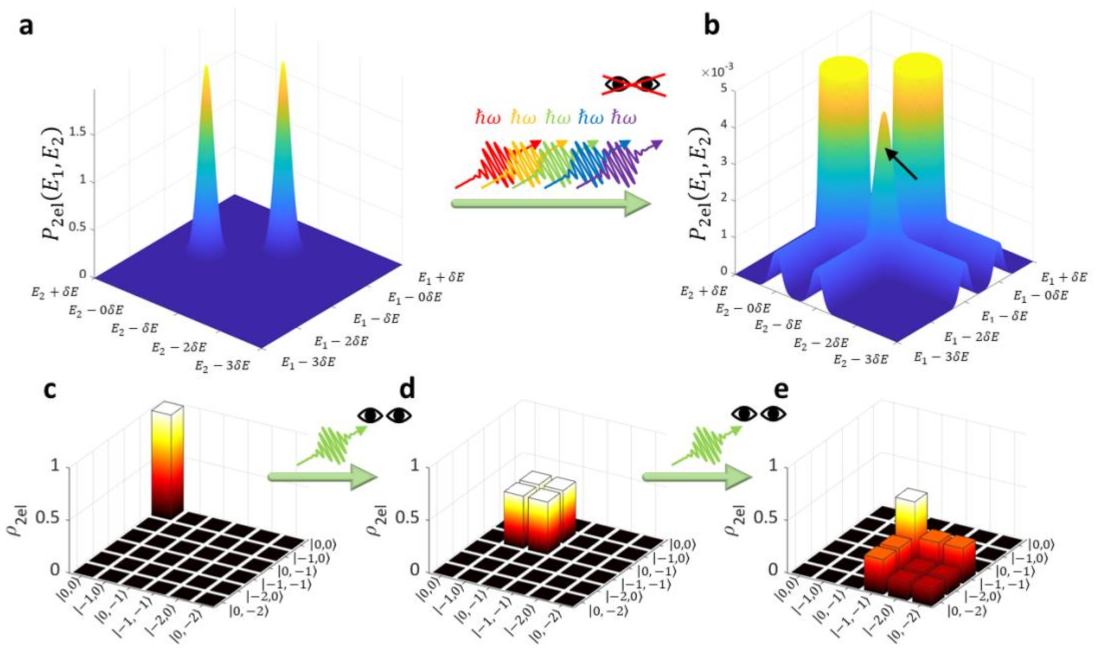

Fig. S1: Electron-electron decoherence and entanglement generation by post-selection. (a) joint two-electron probability $P_{2\mathrm{el}}$ for the initial Bell state $(|E_0-\delta E;E\rangle + |E_0;E-\delta E\rangle)/\sqrt{2}$. The electrons interact with an environment hosting a range of possible optical excitations. The emitted photon is not observed. (b) joint probability for the final mixed state of Eq. (S6.4b) (z-axis truncated to make the distribution visible). Quantum interference corresponding to emission of $\hbar\omega = \delta E$ is still present (black arrow). (c-e) Electron-electron entanglement generated by post-selection, done by observing a photon at $\hbar\omega = \delta E$. Density matrices of (c) an initial product state $|E_0;E_0\rangle$; (d) 2-level Bell state $(|E_0-\delta E;E\rangle + |E_0;E-\delta E\rangle)/\sqrt{2}$ and (e) 3-level entangled state $(|E_0-2\delta E;E\rangle + |E_0;E-2\delta E\rangle + 2|E_0-\delta E;E-\delta E\rangle)/\sqrt{6}$. Axis labels $|-j,-k\rangle$ correspond to the states $|E_0-j\delta E;E_0-k\delta E\rangle$.

<u>Derivation of the results</u>:

Consider a two-electron initial state:

$$\boldsymbol{\rho}_{\text{2el}}^{(i)} = \sum_{\substack{\mathbf{i}_1,\mathbf{i}_2 \\ \mathbf{i}_1',\mathbf{i}_2'}} \rho_{\text{2el}}^{(i)}(\mathbf{i}_1, \mathbf{i}_2; \mathbf{i}_1', \mathbf{i}_2')|\mathbf{i}_1, \mathbf{i}_2\rangle\langle\mathbf{i}_1', \mathbf{i}_2'|, \qquad (1)$$

The total state: two-electron + electromagnetic vacuum

$$\boldsymbol{\rho}_i = \boldsymbol{\rho}_{\text{2el}}^{(i)} \otimes |0\rangle\langle 0| = \sum_{\substack{\mathbf{i}_1,\mathbf{i}_2 \\ \mathbf{i}_1',\mathbf{i}_2'}} \rho_{\text{2el}}^{(i)}(\mathbf{i}_1, \mathbf{i}_2; \mathbf{i}_1', \mathbf{i}_2')|\mathbf{i}_1, \mathbf{i}_2\rangle\langle\mathbf{i}_1', \mathbf{i}_2'| \otimes |0\rangle\langle 0|, \qquad (2)$$

Following light-matter interaction, the total (electron pair + light) final state is:

$$\boldsymbol{\rho}_f = \sum_{\substack{\mathbf{i}_1,\mathbf{i}_2 \\ \mathbf{i}_1',\mathbf{i}_2'}} \rho_{\text{2el}}^{(i)}(\mathbf{i}_1, \mathbf{i}_2; \mathbf{i}_1', \mathbf{i}_2') \sum_{\substack{\mathbf{f}_1\mathbf{f}_2 \\ \mathbf{r}\omega\sigma}} M_{\mathbf{i}_1\mathbf{i}_2\to\mathbf{f}_1\mathbf{f}_2;\mathbf{r}\omega\sigma} \sum_{\substack{\mathbf{f}_1'\mathbf{f}_2' \\ \mathbf{r}'\omega'\sigma'}} M^*_{\mathbf{i}_1'\mathbf{i}_2'\to\mathbf{f}_1'\mathbf{f}_2';\mathbf{r}'\omega'\sigma'} |\mathbf{f}_1\mathbf{f}_2; 1_{\mathbf{r}\omega\sigma}\rangle\langle\mathbf{f}_1'\mathbf{f}_2'; 1_{\mathbf{r}'\omega'\sigma'}|, \qquad (3)$$

Note that $|1_{\mathbf{r}\omega\sigma}\rangle$ stands for an optical excitation at position $\mathbf{r}$, frequency ω and polarization σ. The transition amplitudes are:

$$M_{\mathbf{i}_1\mathbf{i}_2\to\mathbf{f}_1\mathbf{f}_2;\mathbf{r}\omega\sigma} = \frac{i}{\hbar}\int dt\,\langle\mathbf{f}_1\mathbf{f}_2; 1_{\mathbf{r}\omega\sigma}|H_{\text{int}}|\mathbf{i}_1\mathbf{i}_2; 0\rangle, \qquad (4)$$

where

$$H_{\text{int}} = \frac{e}{m}\sum_j \mathbf{A}(\hat{\mathbf{r}}_j)\cdot\hat{\mathbf{p}}_j, \qquad (5)$$

is the interaction Hamiltonian, summing over the two electrons. The electromagnetic vector potential in macroscopic QED is given by

$$\mathbf{A}(\mathbf{r}) = \sqrt{\frac{\hbar}{\pi\epsilon_0}}\frac{1}{c^2}\int \omega\, d\omega \int d^3\mathbf{r}'\sqrt{\text{Im}\,\epsilon(\mathbf{r}',\omega)}\,\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)\mathbf{f}(\mathbf{r}',\omega) + h.c., \qquad (6)$$

where $\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)$ is the dyadic Green's function of the medium, $\epsilon(\mathbf{r}',\omega)$ the relative permittivity, and the operators annihilation operators $\mathbf{f}(\mathbf{r},\omega)$ satisfying

$$\left[f_\alpha(\mathbf{r},\omega), f_\beta^\dagger(\mathbf{r}',\omega')\right] = \delta_{\alpha\beta}\delta(\mathbf{r}-\mathbf{r}')\delta(\omega-\omega'), \qquad (7)$$

The transition amplitude is found to be:

$$M_{\mathbf{i}_1\mathbf{i}_2\to\mathbf{f}_1\mathbf{f}_2;\mathbf{r}\omega\sigma} = \frac{i}{\hbar}e\sqrt{\frac{\hbar}{\pi\epsilon_0}}\frac{v_0}{c^2}\omega 2\pi\delta\left(\omega-\omega_{\mathbf{i2;f2}}^{\mathbf{i1;f1}}\right)\sqrt{\text{Im}\,\epsilon(\mathbf{r},\omega)}$$
$$\times\sum_j\int d^3\mathbf{r}_1\int d^3\mathbf{r}_2\, G^*_{\sigma z}\left(\mathbf{r}_j,\mathbf{r},\omega\right)e^{i(\mathbf{k}_{i1}-\mathbf{k}_{f1})\cdot\mathbf{r}_1}e^{i(\mathbf{k}_{i2}-\mathbf{k}_{f2})\cdot\mathbf{r}_2}, \qquad (8)$$

where

$$\omega_{\mathbf{i2;f2}}^{\mathbf{i1;f1}} = \frac{E_{\mathbf{i1}}-E_{\mathbf{f1}}+E_{\mathbf{i2}}-E_{\mathbf{f2}}}{\hbar}, \qquad (9)$$

<u>Derivation of Eq. (8)</u>

Consider

$$M_{\mathbf{i}_1\mathbf{i}_2\to\mathbf{f}_1\mathbf{f}_2;\mathbf{r}\omega\sigma} = \frac{i}{\hbar}\frac{e}{m}\sum_j \int dt\, \left\langle \mathbf{f}_1\mathbf{f}_2; 1_{\mathbf{r}\omega\sigma} \middle| \mathbf{A}(\hat{\mathbf{r}}_j)\cdot\hat{\mathbf{p}}_j \middle| \mathbf{i}_1\mathbf{i}_2; 0 \right\rangle$$

The matrix element of the jth electron is

$$\left\langle \mathbf{f}_1\mathbf{f}_2; 1_{\mathbf{r}\omega\sigma} \middle| \mathbf{A}(\hat{\mathbf{r}}_j)\cdot\hat{\mathbf{p}}_j \middle| \mathbf{i}_1\mathbf{i}_2; 0 \right\rangle = \int d^3\mathbf{r}_1 \int d^3\mathbf{r}_2 \left\langle \mathbf{f}_1\mathbf{f}_2; 1_{\mathbf{r}\omega\sigma} \middle| \mathbf{A}(\hat{\mathbf{r}}_j) \middle| \mathbf{r}_1\mathbf{r}_2; 0 \right\rangle \cdot \left\langle \mathbf{r}_1\mathbf{r}_2; 0 \middle| \hat{\mathbf{p}}_j \middle| \mathbf{i}_1\mathbf{i}_2; 0 \right\rangle$$

Note that

$$\left\langle \mathbf{r}_1\mathbf{r}_2; 0 \middle| \hat{\mathbf{p}}_j \middle| \mathbf{i}_1\mathbf{i}_2; 0 \right\rangle = \hbar\mathbf{k}_{ij} e^{i\mathbf{k}_{i1}\cdot\mathbf{r}_1} e^{i\mathbf{k}_{i2}\cdot\mathbf{r}_2} e^{-i\frac{E_{i1}+E_{i2}}{\hbar}t}$$

And that

$$\left\langle \mathbf{f}_1\mathbf{f}_2; 1_{\mathbf{r}\omega\sigma} \middle| \mathbf{A}(\hat{\mathbf{r}}_j) \middle| \mathbf{r}_1\mathbf{r}_2; 0 \right\rangle$$

$$= \sqrt{\frac{\hbar}{\pi\epsilon_0}\frac{1}{c^2}} \int \omega'\, d\omega' \int d^3\mathbf{r}' \sqrt{\mathrm{Im}\,\epsilon(\mathbf{r}',\omega')} \sum_\beta \left\langle \mathbf{f}_1\mathbf{f}_2; 1_{\mathbf{r}\omega\sigma} \middle| G^*_{\beta\alpha}(\hat{\mathbf{r}}_j, \mathbf{r}', \omega') f^\dagger_\beta(\mathbf{r}',\omega') \middle| \mathbf{r}_1\mathbf{r}_2; 0 \right\rangle$$

$$= \sqrt{\frac{\hbar}{\pi\epsilon_0}\frac{1}{c^2}} \int \omega'\, d\omega' e^{i\omega' t} e^{i\frac{E_{f1}+E_{f2}}{\hbar}t} \int d^3\mathbf{r}' \sqrt{\mathrm{Im}\,\epsilon(\mathbf{r}',\omega')}\, \delta(\mathbf{r}-\mathbf{r}')\delta(\omega-\omega')$$

$$\times \left\langle \mathbf{f}_1\mathbf{f}_2 \middle| G^*_{\sigma\alpha}(\hat{\mathbf{r}}_j, \mathbf{r}', \omega') \middle| \mathbf{r}_1\mathbf{r}_2 \right\rangle$$

$$= \sqrt{\frac{\hbar}{\pi\epsilon_0}\frac{1}{c^2}}\, \omega e^{i\omega t} e^{i\frac{E_{f1}+E_{f2}}{\hbar}t} \sqrt{\mathrm{Im}\,\epsilon(\mathbf{r},\omega)} \left\langle \mathbf{f}_1\mathbf{f}_2 \middle| G^*_{\sigma\alpha}(\hat{\mathbf{r}}_j, \mathbf{r}, \omega) \middle| \mathbf{r}_1\mathbf{r}_2 \right\rangle$$

$$= \sqrt{\frac{\hbar}{\pi\epsilon_0}\frac{1}{c^2}}\, \omega e^{i\omega t} e^{i\frac{E_{f1}+E_{f2}}{\hbar}t} \sqrt{\mathrm{Im}\,\epsilon(\mathbf{r},\omega)}\, G^*_{\sigma\alpha}(\mathbf{r}_j, \mathbf{r}, \omega) e^{-i\mathbf{k}_{f1}\cdot\mathbf{r}_1} e^{-i\mathbf{k}_{f2}\cdot\mathbf{r}_2}$$

Finally (approximating $\mathbf{k}_{ij} \cong \hat{\mathbf{z}}k_0$):

$$\left\langle \mathbf{f}_1\mathbf{f}_2; 1_{\mathbf{r}\omega\sigma} \middle| \mathbf{A}\cdot\mathbf{p}_j \middle| \mathbf{i}_1\mathbf{i}_2; 0 \right\rangle$$

$$= \sqrt{\frac{\hbar}{\pi\epsilon_0}}\frac{\hbar k_0}{c^2}\, \omega e^{i\left(\omega - \frac{E_{i1}+E_{i2}-E_{f1}-E_{f2}}{\hbar}\right)t} \sqrt{\mathrm{Im}\,\epsilon(\mathbf{r},\omega)}$$

$$\times \int d^3\mathbf{r}_1 \int d^3\mathbf{r}_2\, G^*_{\sigma z}(\mathbf{r}_j, \mathbf{r}, \omega) e^{i(\mathbf{k}_{i1}-\mathbf{k}_{f1})\cdot\mathbf{r}_1} e^{i(\mathbf{k}_{i2}-\mathbf{k}_{f2})\cdot\mathbf{r}_2}$$

Giving

$$M_{\mathbf{i}_1\mathbf{i}_2\to\mathbf{f}_1\mathbf{f}_2;\mathbf{r}\omega\sigma} = \frac{i}{\hbar} e \sqrt{\frac{\hbar}{\pi\epsilon_0}}\frac{v_0}{c^2}\, \omega 2\pi\delta\left(\omega - \frac{E_{\mathbf{i1}}-E_{\mathbf{f1}}+E_{\mathbf{i2}}-E_{\mathbf{f2}}}{\hbar}\right) \sqrt{\mathrm{Im}\,\epsilon(\mathbf{r},\omega)}$$

$$\times \sum_j \int d^3\mathbf{r}_1 \int d^3\mathbf{r}_2\, G^*_{\sigma z}(\mathbf{r}_j, \mathbf{r}, \omega) e^{i(\mathbf{k}_{i1}-\mathbf{k}_{f1})\cdot\mathbf{r}_1} e^{i(\mathbf{k}_{i2}-\mathbf{k}_{f2})\cdot\mathbf{r}_2}$$

Now, we want to find the final state of the electron pair: this is the reduced density matrix

$$\boldsymbol{\rho}^{(\mathrm{f})}_{2\mathrm{el}} = \mathrm{Tr}_{\mathrm{ph}}\boldsymbol{\rho}_f = \sum_{\substack{\mathbf{f}_1\mathbf{f}_2\\ \mathbf{f}'_1\mathbf{f}'_2}} \sum_{\substack{\mathbf{i}_1,\mathbf{i}_2\\ \mathbf{i}'_1,\mathbf{i}'_2}} \rho^{(\mathrm{i})}_{2\mathrm{el}}(\mathbf{i}_1,\mathbf{i}_2;\mathbf{i}'_1,\mathbf{i}'_2) \sum_{\mathbf{r}\omega\sigma} M_{\mathbf{i}_1\mathbf{i}_2\to\mathbf{f}_1\mathbf{f}_2;\mathbf{r}\omega\sigma} M^*_{\mathbf{i}'_1\mathbf{i}'_2\to\mathbf{f}'_1\mathbf{f}'_2;\mathbf{r}\omega\sigma} |\mathbf{f}_1\mathbf{f}_2\rangle\langle\mathbf{f}'_1\mathbf{f}'_2|, \qquad (10)$$

it is now our task to evaluate the sum $\sum_{\mathbf{r}\omega\sigma} M_{\mathbf{i}_1\mathbf{i}_2\to\mathbf{f}_1\mathbf{f}_2;\mathbf{r}\omega\sigma} M^*_{\mathbf{i}'_1\mathbf{i}'_2\to\mathbf{f}'_1\mathbf{f}'_2;\mathbf{r}\omega\sigma}$. It takes the form

$$\sum_{\mathbf{r}\omega\sigma} M_{\mathbf{i}_1\mathbf{i}_2\to\mathbf{f}_1\mathbf{f}_2;\mathbf{r}\omega\sigma} M^*_{\mathbf{i}'_1\mathbf{i}'_2\to\mathbf{f}'_1\mathbf{f}'_2;\mathbf{r}\omega\sigma} = \frac{4\alpha}{c} v_0^2 4\pi^2 \delta\left(\omega^{\mathbf{i1;f1}}_{\mathbf{i2;f2}} - \omega^{\mathbf{i'1;f'1}}_{\mathbf{i'2;f'2}}\right) \begin{bmatrix} \delta(\mathbf{k}_{i2}-\mathbf{k}_{f2})\delta(\mathbf{k}_{i'2}-\mathbf{k}_{f'2})\mathrm{Im}\,G_{zz}(\mathbf{k}_{i1}-\mathbf{k}_{f1},\mathbf{k}_{i'1}-\mathbf{k}_{f'1},\omega) \\ +\delta(\mathbf{k}_{i'1}-\mathbf{k}_{f'1})\delta(\mathbf{k}_{i2}-\mathbf{k}_{f2})\mathrm{Im}\,G_{zz}(\mathbf{k}_{i1}-\mathbf{k}_{f1},\mathbf{k}_{i'2}-\mathbf{k}_{f'2},\omega) \\ +\delta(\mathbf{k}_{i1}-\mathbf{k}_{f1})\delta(\mathbf{k}_{i'2}-\mathbf{k}_{f'2})\mathrm{Im}\,G_{zz}(\mathbf{k}_{i2}-\mathbf{k}_{f2},\mathbf{k}_{i'1}-\mathbf{k}_{f'1},\omega) \\ +\delta(\mathbf{k}_{i1}-\mathbf{k}_{f1})\delta(\mathbf{k}_{i'1}-\mathbf{k}_{f'1})\mathrm{Im}\,G_{zz}(\mathbf{k}_{i2}-\mathbf{k}_{f2},\mathbf{k}_{i'2}-\mathbf{k}_{f'2},\omega) \end{bmatrix}, \qquad (11)$$

Derivation of Eq. 11

$$\sum_{\mathbf{r}\omega\sigma} M_{\mathbf{i}_1\mathbf{i}_2\to\mathbf{f}_1\mathbf{f}_2;\mathbf{r}\omega\sigma} M^*_{\mathbf{i}'_1\mathbf{i}'_2\to\mathbf{f}'_1\mathbf{f}'_2;\mathbf{r}\omega\sigma}$$

$$= \frac{e^2}{\hbar^2}\frac{\hbar}{\pi\epsilon_0}\frac{v_0^2}{c^2}\,4\pi^2\sum_\sigma\int d\omega\,\delta\left(\omega-\omega^{\mathbf{i1;f1}}_{\mathbf{i2;f2}}\right)\delta\left(\omega-\omega^{\mathbf{i'1;f'1}}_{\mathbf{i'2;f'2}}\right)$$

$$\times\sum_j\int d^3\mathbf{r}_1\int d^3\mathbf{r}_2\,e^{i(\mathbf{k}_{i1}-\mathbf{k}_{f1})\cdot\mathbf{r}_1}e^{i(\mathbf{k}_{i2}-\mathbf{k}_{f2})\cdot\mathbf{r}_2}\sum_{j'}\int d^3\mathbf{r}'_1\int d^3\mathbf{r}'_2\,e^{-i\left(\mathbf{k}_{i'1}-\mathbf{k}_{f'1}\right)\cdot\mathbf{r}'_1}e^{-i\left(\mathbf{k}_{i'2}-\mathbf{k}_{f'2}\right)\cdot\mathbf{r}'_2}$$

$$\times\underbrace{\int d^3\mathbf{r}\frac{\omega^2}{c^2}\,\mathrm{Im}\,\epsilon(\mathbf{r},\omega)G^*_{\sigma z}(\mathbf{r}_j,\mathbf{r},\omega)G_{z\sigma}(\mathbf{r},\mathbf{r}'_{j'},\omega)}_{\mathrm{Im}\,G_{zz}\left(\mathbf{r}_j,\mathbf{r}'_{j'},\omega\right)}$$

Giving

$$\sum_{\mathbf{r}\omega\sigma} M_{\mathbf{i}_1\mathbf{i}_2\to\mathbf{f}_1\mathbf{f}_2;\mathbf{r}\omega\sigma} M^*_{\mathbf{i}'_1\mathbf{i}'_2\to\mathbf{f}'_1\mathbf{f}'_2;\mathbf{r}\omega\sigma}$$

$$=\frac{4\alpha}{c}v_0^2 4\pi^2\delta\left(\omega^{\mathbf{i1;f1}}_{\mathbf{i2;f2}}-\omega^{\mathbf{i'1;f'1}}_{\mathbf{i'2;f'2}}\right)\sum_j\sum_{j'}\int d^3\mathbf{r}_1\int d^3\mathbf{r}_2\int d^3\mathbf{r}'_1\int d^3\mathbf{r}'_2$$

$$\times\,e^{i(\mathbf{k}_{i1}-\mathbf{k}_{f1})\cdot\mathbf{r}_1}e^{i(\mathbf{k}_{i2}-\mathbf{k}_{f2})\cdot\mathbf{r}_2}\,\mathrm{Im}\,G_{zz}\left(\mathbf{r}_j,\mathbf{r}'_{j'},\omega^{\mathbf{i1;f1}}_{\mathbf{i2;f2}}\right)e^{-i\left(\mathbf{k}_{i'1}-\mathbf{k}_{f'1}\right)\cdot\mathbf{r}'_1}e^{-i\left(\mathbf{k}_{i'2}-\mathbf{k}_{f'2}\right)\cdot\mathbf{r}'_2}$$

The sums of integrals in the last term can be simplified

$$\sum_j\sum_{j'}\int d^3\mathbf{r}_1\int d^3\mathbf{r}_2\int d^3\mathbf{r}'_1\int d^3\mathbf{r}'_2\,e^{i(\mathbf{k}_{i1}-\mathbf{k}_{f1})\cdot\mathbf{r}_1}e^{i(\mathbf{k}_{i2}-\mathbf{k}_{f2})\cdot\mathbf{r}_2}\,\mathrm{Im}\,G_{zz}\left(\mathbf{r}_j,\mathbf{r}'_{j'},\omega\right)e^{-i\left(\mathbf{k}_{i'1}-\mathbf{k}_{f'1}\right)\cdot\mathbf{r}'_1}e^{-i\left(\mathbf{k}_{i'2}-\mathbf{k}_{f'2}\right)\cdot\mathbf{r}'_2}$$

$$=\int d^3\mathbf{r}_1\int d^3\mathbf{r}_2\int d^3\mathbf{r}'_1\int d^3\mathbf{r}'_2\,e^{i(\mathbf{k}_{i1}-\mathbf{k}_{f1})\cdot\mathbf{r}_1}e^{i(\mathbf{k}_{i2}-\mathbf{k}_{f2})\cdot\mathbf{r}_2}\,\mathrm{Im}\,G_{zz}(\mathbf{r}_1,\mathbf{r}'_1,\omega)e^{-i\left(\mathbf{k}_{i'1}-\mathbf{k}_{f'1}\right)\cdot\mathbf{r}'_1}e^{-i\left(\mathbf{k}_{i'2}-\mathbf{k}_{f'2}\right)\cdot\mathbf{r}'_2}$$

$$+\int d^3\mathbf{r}_1\int d^3\mathbf{r}_2\int d^3\mathbf{r}'_1\int d^3\mathbf{r}'_2\,e^{i(\mathbf{k}_{i1}-\mathbf{k}_{f1})\cdot\mathbf{r}_1}e^{i(\mathbf{k}_{i2}-\mathbf{k}_{f2})\cdot\mathbf{r}_2}\,\mathrm{Im}\,G_{zz}(\mathbf{r}_1,\mathbf{r}'_2,\omega)e^{-i\left(\mathbf{k}_{i'1}-\mathbf{k}_{f'1}\right)\cdot\mathbf{r}'_1}e^{-i\left(\mathbf{k}_{i'2}-\mathbf{k}_{f'2}\right)\cdot\mathbf{r}'_2}$$

$$+\int d^3\mathbf{r}_1\int d^3\mathbf{r}_2\int d^3\mathbf{r}'_1\int d^3\mathbf{r}'_2\,e^{i(\mathbf{k}_{i1}-\mathbf{k}_{f1})\cdot\mathbf{r}_1}e^{i(\mathbf{k}_{i2}-\mathbf{k}_{f2})\cdot\mathbf{r}_2}\,\mathrm{Im}\,G_{zz}(\mathbf{r}_2,\mathbf{r}'_1,\omega)e^{-i\left(\mathbf{k}_{i'1}-\mathbf{k}_{f'1}\right)\cdot\mathbf{r}'_1}e^{-i\left(\mathbf{k}_{i'2}-\mathbf{k}_{f'2}\right)\cdot\mathbf{r}'_2}$$

$$+\int d^3\mathbf{r}_1\int d^3\mathbf{r}_2\int d^3\mathbf{r}'_1\int d^3\mathbf{r}'_2\,e^{i(\mathbf{k}_{i1}-\mathbf{k}_{f1})\cdot\mathbf{r}_1}e^{i(\mathbf{k}_{i2}-\mathbf{k}_{f2})\cdot\mathbf{r}_2}\,\mathrm{Im}\,G_{zz}(\mathbf{r}_2,\mathbf{r}'_2,\omega)e^{-i\left(\mathbf{k}_{i'1}-\mathbf{k}_{f'1}\right)\cdot\mathbf{r}'_1}e^{-i\left(\mathbf{k}_{i'2}-\mathbf{k}_{f'2}\right)\cdot\mathbf{r}'_2}$$

$$=\delta(\mathbf{k}_{i2}-\mathbf{k}_{f2})\delta(\mathbf{k}_{i'2}-\mathbf{k}_{f'2})\int d^3\mathbf{r}_1\int d^3\mathbf{r}'_1\,e^{i(\mathbf{k}_{i1}-\mathbf{k}_{f1})\cdot\mathbf{r}_1}\,\mathrm{Im}\,G_{zz}(\mathbf{r}_1,\mathbf{r}'_1,\omega)e^{-i\left(\mathbf{k}_{i'1}-\mathbf{k}_{f'1}\right)\cdot\mathbf{r}'_1}$$

$$+\delta(\mathbf{k}_{i'1}-\mathbf{k}_{f'1})\delta(\mathbf{k}_{i2}-\mathbf{k}_{f2})\int d^3\mathbf{r}_1\int d^3\mathbf{r}'_2\,e^{i(\mathbf{k}_{i1}-\mathbf{k}_{f1})\cdot\mathbf{r}_1}\,\mathrm{Im}\,G_{zz}(\mathbf{r}_1,\mathbf{r}'_2,\omega)e^{-i\left(\mathbf{k}_{i'2}-\mathbf{k}_{f'2}\right)\cdot\mathbf{r}'_2}$$

$$+\delta(\mathbf{k}_{i1}-\mathbf{k}_{f1})\delta(\mathbf{k}_{i'2}-\mathbf{k}_{f'2})\int d^3\mathbf{r}_2\int d^3\mathbf{r}'_1\,e^{i(\mathbf{k}_{i2}-\mathbf{k}_{f2})\cdot\mathbf{r}_2}\,\mathrm{Im}\,G_{zz}(\mathbf{r}_2,\mathbf{r}'_1,\omega)e^{-i\left(\mathbf{k}_{i'1}-\mathbf{k}_{f'1}\right)\cdot\mathbf{r}'_1}$$

$$+\delta(\mathbf{k}_{i1}-\mathbf{k}_{f1})\delta(\mathbf{k}_{i'1}-\mathbf{k}_{f'1})\int d^3\mathbf{r}_2\int d^3\mathbf{r}'_2\,e^{i(\mathbf{k}_{i2}-\mathbf{k}_{f2})\cdot\mathbf{r}_2}\,\mathrm{Im}\,G_{zz}(\mathbf{r}_2,\mathbf{r}'_2,\omega)e^{-i\left(\mathbf{k}_{i'2}-\mathbf{k}_{f'2}\right)\cdot\mathbf{r}'_2}$$

$$=\delta(\mathbf{k}_{i2}-\mathbf{k}_{f2})\delta(\mathbf{k}_{i'2}-\mathbf{k}_{f'2})\mathrm{Im}\,G_{zz}(\mathbf{k}_{i1}-\mathbf{k}_{f1},\mathbf{k}_{i'1}-\mathbf{k}_{f'1},\omega)$$
$$+\delta(\mathbf{k}_{i'1}-\mathbf{k}_{f'1})\delta(\mathbf{k}_{i2}-\mathbf{k}_{f2})\mathrm{Im}\,G_{zz}(\mathbf{k}_{i1}-\mathbf{k}_{f1},\mathbf{k}_{i'2}-\mathbf{k}_{f'2},\omega)$$
$$+\delta(\mathbf{k}_{i1}-\mathbf{k}_{f1})\delta(\mathbf{k}_{i'2}-\mathbf{k}_{f'2})\mathrm{Im}\,G_{zz}(\mathbf{k}_{i2}-\mathbf{k}_{f2},\mathbf{k}_{i'1}-\mathbf{k}_{f'1},\omega)$$
$$+\delta(\mathbf{k}_{i1}-\mathbf{k}_{f1})\delta(\mathbf{k}_{i'1}-\mathbf{k}_{f'1})\mathrm{Im}\,G_{zz}(\mathbf{k}_{i2}-\mathbf{k}_{f2},\mathbf{k}_{i'2}-\mathbf{k}_{f'2},\omega)$$

Substituting Eq. 11 into Eq. 10, changing sums to integrals and changing the integration variable from the initial wavevectors $\mathbf{k}_i$ to the recoils $\mathbf{q}$, gives the general result

$$\rho^{(\mathrm{f})}_{2\mathrm{el}}(\mathbf{k}_{f1}\mathbf{k}_{f2};\mathbf{k}_{f'1}\mathbf{k}_{f'2})$$
$$=\int d^3\mathbf{q}_1\int d^3\mathbf{q}_2\int d^3\mathbf{q}'_1\int d^3\mathbf{q}'_2\,\rho^{(\mathrm{i})}_{2\mathrm{el}}(\mathbf{k}_{f1}+\mathbf{q}_1,\mathbf{k}_{f2}+\mathbf{q}_2;\mathbf{k}'_{f1}+\mathbf{q}'_1,\mathbf{k}'_{f2}+\mathbf{q}'_2)$$
$$\times\,\delta\left(\omega^{\mathbf{k}'_{f1};\mathbf{q}'_1}_{\mathbf{k}'_{f2};\mathbf{q}'_2}-\omega^{\mathbf{k}_{f1};\mathbf{q}_1}_{\mathbf{k}_{f2};\mathbf{q}_2}\right)\begin{bmatrix}\delta(\mathbf{q}_2)\delta(\mathbf{q}'_2)\mathrm{Im}\,G_{zz}\left(\mathbf{q}_1,\mathbf{q}'_1,\omega^{\mathbf{k}_{f1};\mathbf{q}_1}_{\mathbf{k}_{f2};\mathbf{q}_2}\right)\\+\delta(\mathbf{q}_2)\delta(\mathbf{q}'_1)\mathrm{Im}\,G_{zz}\left(\mathbf{q}_1,\mathbf{q}'_2,\omega^{\mathbf{k}_{f1};\mathbf{q}_1}_{\mathbf{k}_{f2};\mathbf{q}_2}\right)\\+\delta(\mathbf{q}_1)\delta(\mathbf{q}'_2)\mathrm{Im}\,G_{zz}\left(\mathbf{q}_2,\mathbf{q}'_1,\omega^{\mathbf{k}_{f1};\mathbf{q}_1}_{\mathbf{k}_{f2};\mathbf{q}_2}\right)\\+\delta(\mathbf{q}_1)\delta(\mathbf{q}'_1)\mathrm{Im}\,G_{zz}\left(\mathbf{q}_2,\mathbf{q}'_2,\omega^{\mathbf{k}_{f1};\mathbf{q}_1}_{\mathbf{k}_{f2};\mathbf{q}_2}\right)\end{bmatrix},\qquad(12)$$

where here after the change of variables

$$\omega^{\mathbf{k}_{f1};\mathbf{q}_1}_{\mathbf{k}_{f2};\mathbf{q}_2}=\frac{E_{\mathbf{k}_{f1}+\mathbf{q}_1}-E_{\mathbf{k}_{f1}}+E_{\mathbf{k}_{f2}+\mathbf{q}_2}-E_{\mathbf{k}_{f2}}}{\hbar},\qquad(13)$$

Eq. (12) dramatically simplifies when considering an Isotropic medium, for which:

$$\mathrm{Im}\,G_{zz}(\mathbf{q},\mathbf{q}',\omega)=(2\pi)^3\delta(\mathbf{q}-\mathbf{q}')\mathrm{Im}G_{zz}(\mathbf{q},\omega),\qquad(14)$$

and, by considering the small-recoil and paraxial approximations. The simplified expression for the final density matrix reads

$$\rho_{2\mathrm{el}}^{(\mathrm{f})}(\mathbf{k}_1,\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4)=\int d^3\mathbf{q}\,\mathrm{Im}\,G_{zz}(\mathbf{q},\mathbf{q}\cdot\mathbf{v})\begin{bmatrix}\rho_{\mathrm{el}}^{(\mathrm{i})}(\mathbf{k}_1+\mathbf{q},\mathbf{k}_2;\mathbf{k}_3+\mathbf{q},\mathbf{k}_4)\\+\rho_{\mathrm{el}}^{(\mathrm{i})}(\mathbf{k}_1+\mathbf{q},\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4+\mathbf{q})\\+\rho_{\mathrm{el}}^{(\mathrm{i})}(\mathbf{k}_1,\mathbf{k}_2+\mathbf{q};\mathbf{k}_3+\mathbf{q},\mathbf{k}_4)\\+\rho_{\mathrm{el}}^{(\mathrm{i})}(\mathbf{k}_1,\mathbf{k}_2+\mathbf{q};\mathbf{k}_3,\mathbf{k}_4+\mathbf{q})\end{bmatrix},\qquad(15)$$

<u>Derivation of Eq. (15)</u>

$$\rho_{2\mathrm{el}}^{(\mathrm{f})}(\mathbf{k}_{f1}\mathbf{k}_{f2};\mathbf{k}_{f'1}\mathbf{k}_{f'2})$$
$$=\int d^3\mathbf{q}_1\int d^3\mathbf{q}_2\int d^3\mathbf{q}_1'\int d^3\mathbf{q}_2'\,\rho_{2\mathrm{el}}^{(\mathrm{i})}(\mathbf{k}_{f1}+\mathbf{q}_1,\mathbf{k}_{f2}+\mathbf{q}_2;\mathbf{k}_{f1}'+\mathbf{q}_1',\mathbf{k}_{f2}'+\mathbf{q}_2')$$
$$\times\,\delta\left(\omega_{\mathbf{k}_{f2}';\mathbf{q}_2'}^{\mathbf{k}_{f1}';\mathbf{q}_1'}-\omega_{\mathbf{k}_{f2};\mathbf{q}_2}^{\mathbf{k}_{f1};\mathbf{q}_1}\right)\begin{bmatrix}\delta(\mathbf{q}_2)\delta(\mathbf{q}_2')\delta(\mathbf{q}_1-\mathbf{q}_1')\mathrm{Im}\,G_{zz}(\mathbf{q}_1,\omega)\\+\delta(\mathbf{q}_2)\delta(\mathbf{q}_1')\delta(\mathbf{q}_1-\mathbf{q}_2')\mathrm{Im}\,G_{zz}(\mathbf{q}_1,\omega)\\+\delta(\mathbf{q}_1)\delta(\mathbf{q}_2')\delta(\mathbf{q}_2-\mathbf{q}_1')\mathrm{Im}\,G_{zz}(\mathbf{q}_2,\omega)\\+\delta(\mathbf{q}_1)\delta(\mathbf{q}_1')\delta(\mathbf{q}_2-\mathbf{q}_2')\mathrm{Im}\,G_{zz}(\mathbf{q}_2,\omega)\end{bmatrix}$$

Finally, we have

$$\rho_{2\mathrm{el}}^{(\mathrm{f})}(\mathbf{k}_1,\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4)=\begin{array}{l}\int d^3\mathbf{q}\,\mathrm{Im}\,G_{zz}\left(\mathbf{q},\omega_{\mathbf{k}_2,\mathbf{0}}^{\mathbf{k}_1,\mathbf{q}}\right)\delta\left(\omega_{\mathbf{k}_2,\mathbf{0}}^{\mathbf{k}_1,\mathbf{q}}-\omega_{\mathbf{k}_4,\mathbf{0}}^{\mathbf{k}_3,\mathbf{q}}\right)\rho_{2\mathrm{el}}^{(\mathrm{i})}(\mathbf{k}_1+\mathbf{q},\mathbf{k}_2;\mathbf{k}_3+\mathbf{q},\mathbf{k}_4)\\+\int d^3\mathbf{q}\,\mathrm{Im}\,G_{zz}\left(\mathbf{q},\omega_{\mathbf{k}_2,\mathbf{0}}^{\mathbf{k}_1,\mathbf{q}}\right)\delta\left(\omega_{\mathbf{k}_2,\mathbf{0}}^{\mathbf{k}_1,\mathbf{q}}-\omega_{\mathbf{k}_4,\mathbf{q}}^{\mathbf{k}_3,\mathbf{0}}\right)\rho_{2\mathrm{el}}^{(\mathrm{i})}(\mathbf{k}_1+\mathbf{q},\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4+\mathbf{q})\\+\int d^3\mathbf{q}\,\mathrm{Im}\,G_{zz}\left(\mathbf{q},\omega_{\mathbf{k}_2,\mathbf{q}}^{\mathbf{k}_1,\mathbf{0}}\right)\delta\left(\omega_{\mathbf{k}_2,\mathbf{q}}^{\mathbf{k}_1,\mathbf{0}}-\omega_{\mathbf{k}_4,\mathbf{0}}^{\mathbf{k}_3,\mathbf{q}}\right)\rho_{2\mathrm{el}}^{(\mathrm{i})}(\mathbf{k}_1,\mathbf{k}_2+\mathbf{q};\mathbf{k}_3+\mathbf{q},\mathbf{k}_4)\\+\int d^3\mathbf{q}\,\mathrm{Im}\,G_{zz}\left(\mathbf{q},\omega_{\mathbf{k}_2,\mathbf{q}}^{\mathbf{k}_1,\mathbf{0}}\right)\delta\left(\omega_{\mathbf{k}_2,\mathbf{q}}^{\mathbf{k}_1,\mathbf{0}}-\omega_{\mathbf{k}_4,\mathbf{q}}^{\mathbf{k}_3,\mathbf{0}}\right)\rho_{2\mathrm{el}}^{(\mathrm{i})}(\mathbf{k}_1,\mathbf{k}_2+\mathbf{q};\mathbf{k}_3,\mathbf{k}_4+\mathbf{q})\end{array}$$

Note that, under the small recoil approximation

$$\omega_{\mathbf{k}_2,\mathbf{0}}^{\mathbf{k}_1,\mathbf{q}}=\mathbf{q}\cdot\frac{\hbar\mathbf{k}_1}{m}=\mathbf{q}\cdot\mathbf{v}_1$$
$$\omega_{\mathbf{k}_2,\mathbf{0}}^{\mathbf{k}_1,\mathbf{q}}-\omega_{\mathbf{k}_4,\mathbf{0}}^{\mathbf{k}_3,\mathbf{q}}=\mathbf{q}\cdot(\mathbf{v}_1-\mathbf{v}_3)$$

etc. Replacing all $\mathbf{v}_i$'s with the carrier velocity $\mathbf{v}$ we can factor out $\int d^3\mathbf{q}\,\mathrm{Im}\,G_{zz}(\mathbf{q},\mathbf{q}\cdot\mathbf{v})$ and a constant $\delta(0)$ term which falls in normalization, giving

$$\rho_{2\mathrm{el}}^{(\mathrm{f})}(\mathbf{k}_1,\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4)=\int d^3\mathbf{q}\,\mathrm{Im}\,G_{zz}(\mathbf{q},\mathbf{q}\cdot\mathbf{v})\begin{bmatrix}\rho_{2\mathrm{el}}^{(\mathrm{i})}(\mathbf{k}_1+\mathbf{q},\mathbf{k}_2;\mathbf{k}_3+\mathbf{q},\mathbf{k}_4)\\+\rho_{2\mathrm{el}}^{(\mathrm{i})}(\mathbf{k}_1+\mathbf{q},\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4+\mathbf{q})\\+\rho_{2\mathrm{el}}^{(\mathrm{i})}(\mathbf{k}_1,\mathbf{k}_2+\mathbf{q};\mathbf{k}_3+\mathbf{q},\mathbf{k}_4)\\+\rho_{2\mathrm{el}}^{(\mathrm{i})}(\mathbf{k}_1,\mathbf{k}_2+\mathbf{q};\mathbf{k}_3,\mathbf{k}_4+\mathbf{q})\end{bmatrix}$$

If the initial two-electron state is a pure state:

$$\boldsymbol{\rho}_{2\mathrm{el}}^{(i)}=|\psi_{2\mathrm{el}}^{(i)}\rangle\langle\psi_{2\mathrm{el}}^{(i)}|,\qquad(16a)$$

$$|\psi_{2\mathrm{el}}^{(i)}\rangle=\sum_{\mathbf{k}_1\mathbf{k}_2}\psi_{2\mathrm{el}}^{(i)}(\mathbf{k}_1,\mathbf{k}_2)|\mathbf{k}_1\mathbf{k}_2\rangle,\qquad(16b)$$

then the final two-electron density matrix can be written as the mixture

$$\boldsymbol{\rho}_{2\mathrm{el}}^{(f)}=\int d^3\mathbf{q}\,p_{\mathbf{q}}\,|\psi_{\mathrm{rec}}^{(i)}(\mathbf{q})\rangle\langle\psi_{\mathrm{rec}}^{(i)}(\mathbf{q})|,\qquad(17)$$

where the "recoiled" states are given by

$$|\psi_{\rm rec}^{(i)}(\mathbf{q})\rangle = \frac{1}{\sqrt{N_\mathbf{q}}}\sum_{\mathbf{k}_1\mathbf{k}_2}\psi_{\rm 2el}^{(i)}(\mathbf{k}_1,\mathbf{k}_2)(|\mathbf{k}_1-\mathbf{q};\mathbf{k}_2\rangle + |\mathbf{k}_1;\mathbf{k}_2-\mathbf{q}\rangle)\,, \qquad (18a)$$

$$N_\mathbf{q} = 2 + 2\mathrm{Re}\sum_{\mathbf{k}_1\mathbf{k}_2}\psi_{\rm 2el}^{(i)*}(\mathbf{k}_1+\mathbf{q},\mathbf{k}_2-\mathbf{q})\psi_{\rm 2el}^{(i)}(\mathbf{k}_1,\mathbf{k}_2)\,, \qquad (18b)$$

Each recoiled state has a probability

$$p_\mathbf{q} = \frac{N_\mathbf{q}\mathrm{Im}G_{zz}(\mathbf{q},\mathbf{q}\cdot\mathbf{v})}{\int d^3\mathbf{q}N_\mathbf{q}\mathrm{Im}\,G_{zz}(\mathbf{q},\mathbf{q}\cdot\mathbf{v})}, \qquad (19)$$

<u>Deriving Eq. (16-19)</u>

First, we find that, for a given recoil $\mathbf{q}$:

$$\begin{aligned}&\rho_{\rm 2el}^{(\rm i)}(\mathbf{k}_1+\mathbf{q},\mathbf{k}_2;\mathbf{k}_3+\mathbf{q},\mathbf{k}_4)\\ &+\rho_{\rm 2el}^{(\rm i)}(\mathbf{k}_1+\mathbf{q},\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4+\mathbf{q})\\ &+\rho_{\rm 2el}^{(\rm i)}(\mathbf{k}_1,\mathbf{k}_2+\mathbf{q};\mathbf{k}_3+\mathbf{q},\mathbf{k}_4)\\ &+\rho_{\rm 2el}^{(\rm i)}(\mathbf{k}_1,\mathbf{k}_2+\mathbf{q};\mathbf{k}_3,\mathbf{k}_4+\mathbf{q})\\ &= \left[\psi_{\rm 2el}^{(i)}(\mathbf{k}_1+\mathbf{q},\mathbf{k}_2)+\psi_{\rm 2el}^{(i)}(\mathbf{k}_1,\mathbf{k}_2+\mathbf{q})\right]\left[\psi_{\rm 2el}^{(i)*}(\mathbf{k}_3+\mathbf{q},\mathbf{k}_4)+\psi_{\rm 2el}^{(i)*}(\mathbf{k}_3,\mathbf{k}_4+\mathbf{q})\right]\end{aligned}$$

Which looks like a pure state. We define this new "recoiled state" as

$$\begin{aligned}|\psi_{\rm rec}^{(i)}(\mathbf{q})\rangle &= \frac{1}{\sqrt{N_\mathbf{q}}}\sum_{\mathbf{k}_1\mathbf{k}_2}\left[\psi_{\rm 2el}^{(i)}(\mathbf{k}_1+\mathbf{q},\mathbf{k}_2)+\psi_{\rm 2el}^{(i)}(\mathbf{k}_1,\mathbf{k}_2+\mathbf{q})\right]|\mathbf{k}_1;\mathbf{k}_2\rangle\\ &= \frac{1}{\sqrt{N_\mathbf{q}}}\sum_{\mathbf{k}_1\mathbf{k}_2}\psi_{\rm 2el}^{(i)}(\mathbf{k}_1,\mathbf{k}_2)(|\mathbf{k}_1-\mathbf{q};\mathbf{k}_2\rangle+|\mathbf{k}_1;\mathbf{k}_2-\mathbf{q}\rangle)\end{aligned}$$

For this state to be normalized we demand that:

$$\begin{aligned}1 = \langle\psi_{\rm rec}^{(i)}(\mathbf{q})|\psi_{\rm rec}^{(i)}(\mathbf{q})\rangle &=\\ &= \frac{2}{N_\mathbf{q}}\sum_{\mathbf{k}_1\mathbf{k}_2}\psi_{\rm 2el}^{(i)*}(\mathbf{k}_1,\mathbf{k}_2)\psi_{\rm 2el}^{(i)}(\mathbf{k}_1,\mathbf{k}_2)\\ &+\frac{2}{N_\mathbf{q}}\mathrm{Re}\sum_{\mathbf{k}_1\mathbf{k}_2}\psi_{\rm 2el}^{(i)*}(\mathbf{k}_1+\mathbf{q},\mathbf{k}_2-\mathbf{q})\psi_{\rm 2el}^{(i)}(\mathbf{k}_1,\mathbf{k}_2)\end{aligned}$$

giving

$$N_\mathbf{q} = 2 + 2\mathrm{Re}\sum_{\mathbf{k}_1\mathbf{k}_2}\psi_{\rm 2el}^{(i)*}(\mathbf{k}_1+\mathbf{q},\mathbf{k}_2-\mathbf{q})\psi_{\rm 2el}^{(i)}(\mathbf{k}_1,\mathbf{k}_2)$$

The recoiled state density matrix, for a given $\mathbf{q}$ is

$$\boldsymbol{\rho}_{\rm rec}^{(i)}(\mathbf{q}) = |\psi_{\rm rec}^{(i)}(\mathbf{q})\rangle\langle\psi_{\rm rec}^{(i)}(\mathbf{q})|$$

Substituting into Eq. (15) we get

$$\boldsymbol{\rho}_{\rm 2el}^{(\rm f)} = \int d^3\mathbf{q}N_\mathbf{q}\mathrm{Im}\,G_{zz}(\mathbf{q},\mathbf{q}\cdot\mathbf{v})\,|\psi_{\rm rec}^{(i)}(\mathbf{q})\rangle\langle\psi_{\rm rec}^{(i)}(\mathbf{q})|$$

Upon normalizing $\mathrm{Tr}\boldsymbol{\rho}_{\rm 2el}^{(\rm f)} = 1$, we can write the final result

$$\boldsymbol{\rho}_{\rm 2el}^{(\rm f)} = \int d^3\mathbf{q}p_\mathbf{q}|\psi_{\rm rec}^{(i)}(\mathbf{q})\rangle\langle\psi_{\rm rec}^{(i)}(\mathbf{q})|$$

where the probabilities are

$$p_\mathbf{q} = \frac{N_\mathbf{q}\mathrm{Im}G_{zz}(\mathbf{q},\mathbf{q}\cdot\mathbf{v})}{\int d^3\mathbf{q}N_\mathbf{q}\mathrm{Im}\,G_{zz}(\mathbf{q},\mathbf{q}\cdot\mathbf{v})}$$