Superradiant Electron Energy Loss Spectroscopy

Supporting Information

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Section I – Interaction between a free electron and non-interacting emitters

In this section, we will derive Eqs. 1-3 of the main text, starting from the basic one-dimensional (1D) Hamiltonian. This 1D Hamiltonian is based on the paraxial approximation for the electron, which is justified in the limit where the initial energy of the electron is considerably greater than the energy that electron loses during the interaction with multiple emitters. This approximation is well justified under typical conditions inside an electron microscope where the probing electron energy lay in the 100keV range and the energy range of the emitters' excitations are in the range of 1eV.

Let us start from the Hamiltonian of free relativistic (spin-less) electron, which Hamiltonian is derived from Klein-Gordon equation [1]:

$$H_{\mathrm{e}} = \sqrt{m^2c^4 + c^2\hat{\boldsymbol{p}}^2}. \#(\mathrm{SI1})$$

We now simplify the Klein–Gordon Hamiltonian under the paraxial approximation, which results from linearizing the dispersion relation of the electron around its central momentum. We consider that initially the electrons' momentum distribution is narrowly distributed around $p_0 \cdot \boldsymbol{e}_z$ and the change of momentum during the interaction is much smaller than p_0. In this case we can write Taylor expansion of Hamiltonian Eq. (SI1) and get:

$$H_e \approx E_0 + v_0 \cdot (\hat{p}_z - p_0), \#(\mathrm{SI2})$$

where $E_0 = \sqrt{m^2c^4 + c^2p_0}$, $v_0 = c^2p_0/E_0$ and $\hat{p}_z = -i\hbar\partial_z$ in the coordinate representation. Since the Hamiltonian is defined up to the constant, we can write the Hamiltonian without E_0 and p_0v_0, so that:

$$H_e = -i\hbar v_0 \partial_z. \#(\mathrm{SI3})$$

Physically, this approximation states that, regardless of the electron's momentum, its velocity is v, which is a type of "no-recoil approximation".

Now we consider the Hamiltonian of multiple non-interacting emitters:

$$H_a = \sum_i \sum_p \hbar\omega_i^p |p_i\rangle\langle p_i|, \#(\mathrm{SI4})$$

where the n'th energy eigenstate of the i'th emitter is labeled $|p_i\rangle$ and the corresponding energy eigenvalue is $\hbar\omega_i^p$. In Eq. (SI4) we have two sums – over all the states $|p_i\rangle$ of i^{th} emitter and over all the emitters. Eq. (SI4) gives general Hamiltonian with the only assumption that the emitters are far enough from each other that we can neglect interactions between them

We describe the interaction between multiple emitters and a free electron in assumption that the emitters are at such distance from the free electron that we can neglect the spatial size of the emitters. In this case, we apply dipole approximation and the Hamiltonian of the interaction between emitters and the free electron is:

$$H_{\text{int}} = -\sum_i \hat{\boldsymbol{d}}^i \cdot \boldsymbol{E}\left(\boldsymbol{r}^i{}_\perp, z - z_i\right), \#(\text{SI5})$$

where $\hat{\boldsymbol{d}}^i$ is the operator of dipole moment of i^{th} emitter and $\boldsymbol{E}\left(\boldsymbol{r}^i{}_\perp, z - z_i\right)$ is the electric field generated by relativistic electron with position $(\boldsymbol{0}, z)$ at the position of i^{th} emitter $(-\boldsymbol{r}^i{}_\perp, z_i)$. This classical electric field according to [2] equals to:

$$\boldsymbol{E}\left(\boldsymbol{r}^i{}_\perp, z - z_i\right) = -\frac{e\gamma}{4\pi\varepsilon_0} \frac{(z - z_i)\boldsymbol{e}_z + r^i{}_\perp \boldsymbol{e}_\perp}{\left(\gamma^2 (z - z_i)^2 + r^i{}_\perp{}^2\right)^{\frac{3}{2}}}. \#(\text{SI6})$$

The dipole moment operator can be written in the basis of eigen state of the emitter:

$$\hat{\boldsymbol{d}}^i = \sum_{p,q} |p_i\rangle\langle q_i| \left(d^i_{z,pq} \boldsymbol{e}_z + d^i{}_{\perp,pq} \boldsymbol{e}_\perp\right),$$

where $d^i_{z,pq} = \left\langle p_i \middle| \hat{\boldsymbol{d}}^i \middle| q_i \right\rangle \cdot \boldsymbol{e_z}$ and $d^i{}_{\perp,pq} = \left\langle p_i \middle| \hat{\boldsymbol{d}}^i \middle| q_i \right\rangle \cdot \boldsymbol{e}_\perp$.

Thus, within this model, the final Hamiltonian of free electron and multiple emitters has the following form:

$$H = H_e + H_a + H_{\text{int}}, \#(\text{SI7})$$

where all the parts of Hamiltonian are defined in Eqs. (SI 3-5). The Schrodinger equation is given by:

$$i\hbar \frac{\partial}{\partial t} |\Psi\rangle = H|\Psi\rangle, \#(\text{SI8})$$

where $|\Psi\rangle$ is the joint wavefunction of emitters and the free electron. We move to the interaction picture and get the following equation:

$$i\hbar \frac{\partial |\Psi_I\rangle}{\partial t} = \mathcal{H}_I |\Psi_I\rangle, \#(\text{SI9})$$

where $|\Psi_I\rangle = U^\dagger(t)|\Psi\rangle$, $\mathcal{H}_I = U^\dagger H_{\text{int}} U$ and $U = \exp\left(-i(H_a + H_e)t/\hbar\right)$.

$$\mathcal{H}_I = U^\dagger H_{\text{int}} U = \frac{e\gamma}{4\pi\varepsilon_0} \sum_i \sum_{pq} e^{-i\omega^i_{pq} t} \hat{t}^i_{pq} \frac{(z + v_0 t - z_i) d^i_{z,pq} + r^i{}_\perp d^i{}_{\perp,pq}}{\left(\gamma^2 (z + v_0 t - z_i)^2 + r^i{}_\perp{}^2\right)^{\frac{3}{2}}}, \#(\text{SI10})$$

where $\omega^{pq}_i = \omega^p_i - \omega^q_i$ and $\hat{t}^i_{pq} = |p_i\rangle\langle q_i|$. The scattering matrix for Eq. (SI9) is given by:

$$\mathbb{S} = \hat{\text{T}} \exp\left(-\frac{i}{\hbar} \int_{-\infty}^{+\infty} \mathcal{H}_I(t) dt\right), \#(\text{SI11})$$

where $\hat{\text{T}}$ is time-ordering exponent and scattering matrix allows to find the state of the system after the interaction ($\mathbb{S}|\Psi(-\infty)\rangle = |\Psi(+\infty)\rangle$). We consider weak coupling between free electron and the emitters, and thus consider only 1st order in Magnus expansion [3] of Eq. (11):

$$\mathbb{S} \approx \exp\left(-\frac{i}{\hbar} \int_{-\infty}^{+\infty} \mathcal{H}_I(t) dt\right). \#(\text{SI12})$$

The integral inside the exponent can be calculated analytically [4] and we get:

$$\mathbb{S} = \exp\left(i \sum_i \sum_{pq} g^i_{pq} \hat{t}^i_{pq} \hat{b}^i_{pq}\right), \#(\text{SI13})$$

where

$$g_{pq}^{i}=\left(\frac{ed^{i}{}_{\perp,pq}\omega_{i}^{pq}K_{1}\left(\frac{\omega_{i}^{pq}r^{i}{}_{\perp}}{\gamma v_{0}}\right)}{2\pi\varepsilon_{0}\gamma\hbar v_{0}^{2}}+i\frac{ed_{z,pq}^{i}\omega_{i}^{pq}K_{0}\left(\frac{\omega_{i}^{pq}r^{i}{}_{\perp}}{\gamma v_{0}}\right)}{2\pi\varepsilon_{0}\gamma^{2}\hbar v_{0}^{2}}\right)e^{-i\frac{\omega_{i}^{pq}z_{i}}{v_{0}}},$$

$$\hat{t}_{pq}^{i}=|p_{i}\rangle\langle q_{i}|,\ \hat{b}_{pq}^{i}=e^{-i\frac{\omega_{i}^{pq}}{v}z}.$$

t_i^{pq} is the transition operator $|p_i\rangle\langle q_i|$, K_m represents modified Bessel functions of the second kind, ε_0 is the vacuum permittivity, and $\gamma=\frac{1}{\sqrt{1-v^2/c^2}}$. Notice that b_i^{pq} is an energy translation operator for a paraxial electron, corresponding to an energy translation of $\hbar\omega_i^{pq}=\hbar\omega_i^{p}-\hbar\omega_i^{q}$.

To sum up, Eq. (SI13) describes the interaction of the relativistic free electron with multiple emitters. In the derivation of Eq. (SI13) a few approximations have been done: 1) We assume that the electron is relativistic and the energy change of the electron due to the interaction is much smaller than initial energy. This assumption give rise to the "no-recoil" approximation; 2) We assume that all emitters do not interact with each other and can be treated as point particles, which leads to the dipole approximation for the emitters; 3) We assume that interaction between the emitters and free electron is weak enough that we can use only the first term in Magnus expansion.

Let us elaborate more on the assumption of non-interacting emitters. This assumption is correct when the energy of the interaction between neighboring emitters is much smaller than the energy of the interaction between the free electron and the emitters. According to Eq. (SI6), the maximum energy of the interaction between the free electron and the emitter is $H_{\mathrm{int}}^{\max}=\frac{e\gamma\, d_{\perp}}{4\pi\varepsilon_0 r_{\perp}^2}$, while the energy of dipole-dipole interaction between neighboring emitters can be estimated as $H_{\mathrm{d-d}}\sim\frac{|\boldsymbol{d}|^2}{4\pi\varepsilon_0 a^3}$, where a is the distance between neighboring emitters. Thus, the assumption of non-interacting emitters holds true, when $H_{\mathrm{int}}^{\max}\gg H_{\mathrm{d-d}}$ which leads to:

$$a\gg\sqrt{dr_{\perp}^2/e\gamma}\sim 5\ \mathrm{nm}.$$

The estimation was done for $d=e\cdot 1$ nm and $r_{\perp}=10$ nm.

However, even with applied approximation the scattering matrix $\mathbb{S}$ lies in an enormous Hilbert space. To proceed with a tractable problem that still exhibits the ideas of our work, we consider the special case when all the emitters are two-level systems with the same energy separation $\hbar\omega_0$, the same transition dipole, and are located at an approximately equal transverse distance from the electron trajectory compared to the wavelength of light (e.g., satisfy $|\boldsymbol{r}^{i}{}_{\perp}-\boldsymbol{r}^{j}{}_{\perp}|\ll\frac{c}{2\pi\omega}$). In this case, $b_i\equiv b\equiv e^{-\frac{i\omega_0}{v_0}z}$ and $\boldsymbol{r}^{\boldsymbol{i}}{}_{\perp}\equiv\boldsymbol{r}_{\perp}$, $\boldsymbol{d}^{i}\equiv\boldsymbol{d}$ and the scattering matrix from Eq. (SI13) can be simplified to:

$$\mathbb{S}=\exp\left(i\left(gS_{+}b+g^{*}S_{-}b^{\dagger}\right)\right),\#(\mathrm{SI14})$$

with $S_{\pm}=\sum_i\sigma_{\pm}^{i}e^{\mp\frac{i\omega_0 z_i}{v}}$ and $g=\frac{e\omega_0}{2\pi\gamma\varepsilon_0\hbar v_0^2}\left(d_{\perp}K_1\left(\frac{\omega_0 r_{\perp}}{\gamma v_0}\right)+\frac{i}{\gamma}d_z K_0\left(\frac{\omega_0 r_{\perp}}{\gamma v_0}\right)\right)$. The validity of the two-level model is further discussed in the main text. This scattering matrix still resides in an enormous Hilbert space; however, we can split the space to sub-Hilbert spaces that are invariant under the operators $S_{\pm}$ and under the evolution by the scattering matrix. This invariance makes the electron interaction with ensembles of emitters a promising probe for investigating superradiating systems, because the superradiance dynamics is itself also restricted to such sub-Hilbert states. Notice that if all the emitters are confined in a small volume, s.t $\Delta\boldsymbol{r}_i\ll\frac{c}{\lambda}$, this sub-Hilbert space just reduces to the Dicke states [5].

Section II – Estimations of the scattering matrix in the superradiant sub-Hilbert space

In this section, we consider the scattering matrix from Eq. (SI14) and investigate the limiting case in which it reduced to Eq. 6 of the main text. We define the superradiant sub-Hilbert space as the space spanned by the states $|n\rangle = \frac{1}{\sqrt{\binom{N}{m}}} S_+^n |gg...g\rangle$. These states define the superradiant ladder, which is a $N+1$ dimensional sub-Hilbert space where N is the number of emitters. The scattering matrix is closed in this sub-Hilbert space, allowing us to write a matrix element:

$$s_{nm} = \langle n|\mathbb{S}|m\rangle = b^{n-m} e^{i(n-m)\arg(g)} (\cos|g|)^N (i\tan|g|)^{n-m} \times$$

$$\sqrt{m!n!(N-n)!(N-m)!} \sum_{k=0}^{m} \frac{(-1)^k (\tan|g|)^{2k}}{k!(m-k)!(n-m+k)!(N-n-k)!}. \#(\text{SI15})$$

Expanding to lowest non-zero order in $|g|$, we get:

$$s_{nm} \approx b^{n-m} e^{i(n-m)\arg(g)} i^{n-m} |g|^{n-m} \times$$

$$\sqrt{m!n!(N-n)!(N-m)!} \sum_{k=0}^{m} \frac{(-1)^k |g|^{2k}}{k!(m-k)!(n-m+k)!(N-n-k)!}. \#$$

If we assume that $N-m, N-n \gg 1$ and additionaly that $n,m \gg 1$, meaning that the initial and final state are both far from the edges of the superradiant ladder, we can take the upper limit of the sum to infinity and use the Stirling's approximation to evaluate the factorials, $n! \sim \sqrt{2\pi n}\left(\frac{n}{e}\right)^n$. Then, the matrix element s_{nm} can be simplified further:

$$s_{nm} \approx b^{n-m} e^{i(n-m)\arg(g)} i^{n-m} |g|^{n-m} \times$$

$$\sqrt{\sqrt{n \cdot m \cdot (N-m) \cdot (N-n)} \cdot \left(\frac{n^n m^m (N-m)^{(N-m)} (N-n)^{N-n}}{e^{2N}}\right)} \times$$

$$\sum_{k=0}^{\infty} \frac{(-1)^k |g|^{2k}}{k!(n-m+k)! \cdot \sqrt{(m-k) \cdot (N-n-k)} \cdot \left(\frac{(m-k)^{(m-k)} (N-n-k)^{(N-n-k)}}{e^{N+m-n-2k}}\right)}.$$

$$s_{nm} \approx b^{n-m} e^{i(n-m)\arg(g)} i^{n-m} |g|^{n-m} e^{n-m} \times$$

$$\sqrt{n^n m^m (N-m)^{(N-m)} (N-n)^{N-n}} \times$$

$$\sum_{k=0}^{\infty} \frac{(-1)^k |g|^{2k}}{k!(n-m+k)! \cdot (m-k)^{(m-k)} (N-n-k)^{(N-n-k)} e^{-2k}}.$$

$$s_{nm} \approx b^{n-m} e^{i(n-m)\arg(g)} i^{n-m} |g|^{n-m} e^{n-m} \times$$

$$\sqrt{\frac{n^n (N-m)^{(N-m)}}{m^m (N-n)^{N-n}}} \times \sum_{k=0}^{\infty} \frac{(-1)^k |g|^{2k} m^k (N-n)^k e^{2k}}{k!(n-m+k)!}.$$

Additionally, assuming that n and m are close enough compared to the size of the ladder (i.e., $|n-m| \ll N$), and defining $\beta \equiv \sqrt{N-m}|g| \approx \sqrt{N-n}|g|$, $l \equiv n-m$, we get:

$$s_{nm} \approx b^{n-m} e^{i(n-m)\arg(g)} i^{n-m} (\sqrt{m}\beta e)^l \sum_{k=0}^{\infty} \frac{(-1)^k (\beta e)^{2k} m^k}{k!(l+k)!}.$$

This sum can be expressed analytically as a Bessel function of the first kind:

$$s_{nm} \approx b^{n-m} e^{i(n-m)\arg(g)} i^{n-m} J_l\left(2\sqrt{Nm-m^2}|g|\right), \#(\text{SI16})$$

which is exactly the scattering matrix that describes free electron Rabbi oscillations [6] with $|g_{\text{eff}}| = \sqrt{Nm-m^2}|g|$. This result can be understood intuitively from the fact that a $N+1$ level ladder looks like a harmonic oscillator when far from the edges. If we assume that the emitters occupy only levels surrounding the level m on the superradiant ladder, such that $m \gg 1$, $N-m \gg 1$, $|n-m| \ll N$, then all the non-zero elements of the operator S_+ will be roughly proportional to $\sqrt{Nm-m^2}$, and the scattering matrix for the electron will look like:

$$\mathbb{S} = e^{i\sqrt{Nm-m^2}(gb+g^* b^\dagger)}. \#(\text{SI17})$$

This is exactly the free electron Rabbi oscillations scattering matrix. If we excite the emitters using coherent-control pulses, and the duration of our laser corresponds to ϕ radians of the Rabbi cycle, the emitters will be distributed around $m = \sin^2\left(\frac{\phi}{2}\right) \cdot N$, and so we can approximate the effective coupling in this case as:

$$g_{\text{eff}} = \frac{\sin(\phi)}{2} Ng. \#(\text{SI18})$$

While this system behaves like free electron Rabbi oscillations, it is interesting to note that when we do not assume $|g| \ll 1$, the systems behave fundamentally different regardless of the size of the ladder. To see that, we can write the original scattering matrix from Eq. (SI4) in a different equivalent form:

$$S = \prod_i \left[\cos(|g|) - i\sin(|g|)\left(e^{i\cdot\arg(g) - \frac{i\omega_0 z_i}{v}} \sigma^i_+ b + e^{-i\cdot\arg(g) + \frac{i\omega_0 z_i}{v}} \sigma^i_- b^+\right)\right]. \#(\text{SI19})$$

In this case, we can see that the scattering matrix behaves fundamentally different for high values of $|g|$. Two important examples are $|g| = \frac{\pi}{2}, \pi$:

$$S_{|g|=\frac{\pi}{2}} = \prod_i \left[-i\left(e^{i\cdot\arg(g) - \frac{i\omega_0 z_i}{v}} \sigma^i_+ b + e^{-i\cdot\arg(g) + \frac{i\omega_0 z_i}{v}} \sigma^i_- b^+\right)\right],$$

$$S_{|g|=\pi} = I. \#(\text{SI20})$$

For the case of $|g| = \frac{\pi}{2}$, the matrix representation in the superradiant sub-Hilbert space contains only elements on the secondary diagonals. Meaning that the state $|m\rangle$ can only transition to the state $|N-m\rangle$ (similar to the application of a π pulse in coherent control). This corresponds to a free electron changing the state of every two-level system. Similarly, the case of $|g| = \pi$ corresponds to a free electron that does not change any two-level system at all, or more precisely, the electron induces exactly one Rabbi cycle on each two-level system. These kinds of effects have no correspondence in the analogy of harmonic oscillator and represent a fundamental difference between the systems even in the limit of $N \gg 1$ emitters.

Section III – Interaction dependence on disorder and interaction length.

This section discusses the results of Section 3 of the main text and presents further analysis of their resilience to disorder in position, dipole strength, and energy of the emitters. In Eq. 10 of the main text, we presented the average dipole moment as seen by moving electron, when all the dipole moments of the emitters are taken to be of the same size. If we allow the different sizes of dipole moments for different emitters, the equation can be re-written as:

$$|\langle d(\theta)\rangle|^2 = \left|\sum_i \boldsymbol{d}_i e^{i\omega_0 z_i\left(\frac{n\cos\theta}{c}-\frac{1}{v}\right)}\right|^2 .\#(\text{SI21})$$

From this equation, it is apparent that when the Cherenkov condition is satisfied $\left(\frac{n\cos\theta}{c}-\frac{1}{v}=0\right)$, the equation reduces to $|\sum \boldsymbol{d}_i|^2$ independently on their position. Thus, the Cherenkov resonance is robust to disorder in the emitters' positions.

If the emitters are of varying dipole strength or direction, the equation can be reduced to $N^2|d_{\text{avg}}|^2$, and so the superradiant (N^2) enhancement is still apparent with the replacement of the dipole of the individual emitter with the average dipole. Only in the case when the average dipole moment is zero (i.e., $|\sum \boldsymbol{d}_i|^2 = 0$) the Cherenkov resonance disappears. However, in typical systems of emitters such as quantum dots grown on a substrate, or emitter ensembles forming superlattices, there is often a shared orientation of the dipole moment (or an approximated one) for all the emitters.

If additionally, we assume that the emitters are in periodic positions with distance Δz, the superradiant enhancement can be achieved for additional angles. By plugging this case into SI21, we can obtain the generalized hybrid Cherenkov-Smith-Purcell condition presented in Eq. 11 of the main text. Notice that the angular dependence in SI21 is multiplied in this case by $\frac{\Delta z\omega_0 n}{c}=\frac{2\pi\Delta z n}{\lambda}$, where λ is the emitters' radiation wavelength in free space. We can conclude that positional variations on the order of $\Delta z \ll \lambda$ does not change the superradiant enhancement significantly. For NIR emitters, this is a fairly simple requirement for modern fabrication abilities.

In Fig. 1, we present the EELS and observed dipole of the scattering of the free electron by 10 emitters in periodic positions in 4 exemplary cases (the parameters of the emitters are taken to be the same as these in Fig. 2 of the main text): (1, Fig.1a) Emitters are spaced by 1 μm, more than their radiation wavelength (800 nm) s.t. Smith-Purcell peaks can be observed. (2, Fig.1b) Emitters are spaced by 1μm, more than their radiation wavelength s.t. the Smith-Purcell peak can be observed. We further introduce Gaussian noise of 150 nm (smaller then but not negligible compared to the wavelength) on their position, showing that while some of the features of the Smith-Purcell peak can be observed, it is generally smeared out. (3, Fig.1c) Emitters are spaced by 300 nm, smaller but comparable to their radiation wavelength. In this case, the Smith-Purcell peak cannot be observed, and the Cherenkov resonance is broadened. (3, Fig.1d) Emitters are spaced by 20 nm, much smaller than their radiation wavelength. In this case, the Smith-Purcell peak cannot be observed and the Cherenkov resonance is completely broadened over the entire interaction as all the emitters are located within a single wavelength.

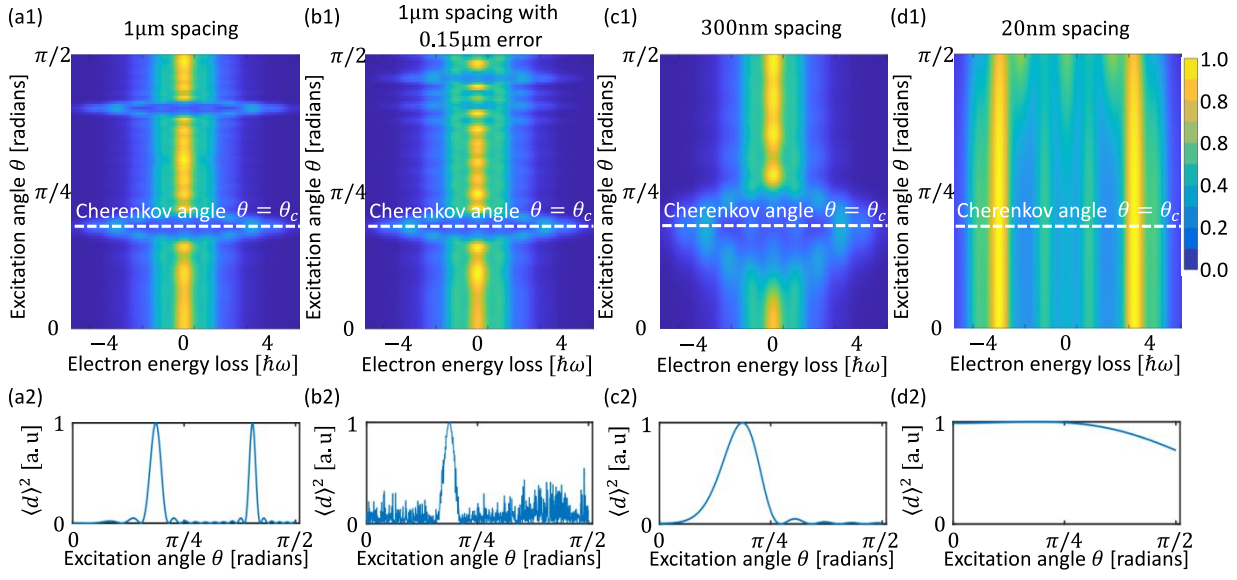

Fig. S1. Free-electron interaction with emitters of different spacings. (a) EELS as a function of excitation angle by a $\pi/2$ pulse. **(b)** The observed dipole from the perspective of the electron according to SI21.

Fig. S1 shows that the Cherenkov resonance is smeared over the entire scale of excitation angles when the spacing between emitters is much smaller than the emitters' radiation wavelength. However, the width of the Cherenkov resonance depends on the interaction length, which can be many times the radiation wavelength in state-of-the-art grazing angle experiments [7]. In such experiments, the electron can interact with thousands of emitters. We cannot perform a full quantum mechanical calculation for such a situation because the Hilbert-space is enormous, but we can evaluate the observed dipole by using SI21 to analyze the structure of the Cherenkov resonance. If we assume uniform spacing Δz between N emitters, the sum can be evaluated as:

$$|\langle d(\theta)\rangle|^2 = d_0^2\left|\sum_k e^{i\omega_0\Delta z\cdot k\cdot\left(\frac{n\cos\theta}{c}-\frac{1}{v}\right)}\right|^2 = d_0^2\left|\frac{e^{iN\omega_0\Delta z\left(\frac{n\cos\theta}{c}-\frac{1}{v}\right)}-1}{e^{i\omega_0\Delta z\left(\frac{n\cos\theta}{c}-\frac{1}{v}\right)}-1}\right|^2 .\#(\text{SI22})$$

We denote $\omega_0\Delta z\left(\frac{n\cos\theta}{c}-\frac{1}{v}\right)=X(\theta)$. When the Cherenkov condition is satisfied, we have $X(\theta)=0$ and $|\langle d(\theta)\rangle|^2 \to N^2 d_0^2$. We expand the expression from small values around that and get:

$$|\langle d(\theta)\rangle|^2 \approx d_0^2\left|\frac{\sin\left(\frac{NX(\theta)}{2}\right)}{\frac{X(\theta)}{2}}\right|^2 .\#(\text{SI23})$$

The sinc function implies that we should expect the width of the resonance (in terms of $X(\theta)$) to scale as $1/N$, and hence the resonance width should scale as $\frac{\lambda}{L}$, where L is the entire interaction length $N\Delta z$, and λ is the emitters' radiation wavelength $\propto \frac{c}{\omega_0}$.

In Fig. 2, we show the scaling law of the width of the resonance using a simple single-parameter fit, showing the calculation of full-width-half-max (FWHM) of the Cherenkov resonance for varying total interaction length.

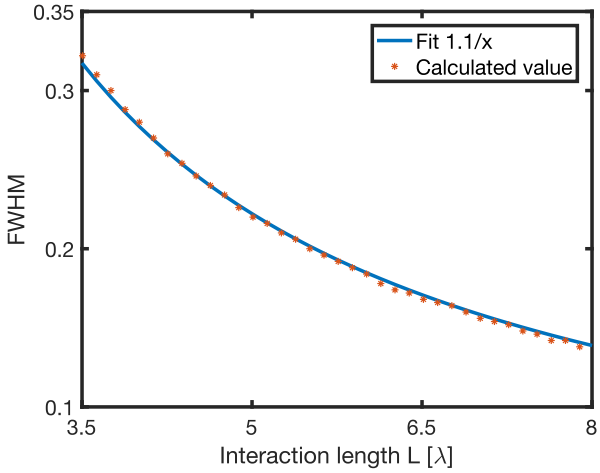

Fig. 1. The scaling law of the Cherenkov resonance FWHM, plotted for emitters with fixed spacings and varying total interaction length, as calculated from SI22. The fitted curve has the form $a\cdot\lambda/L$,

demonstrating the general scaling law. For the calculation, we chose $n = 1.5$, $\lambda = 800$ nm, $\Delta z = 10$ nm, and $N \equiv L/\Delta z$.

The last point to discuss in this section is how disorder in energy can influence the effect. Disorder in the emitters and inter-emitter interactions typically results in broadening of their energy distribution. This effect is captured inside the effective decoherence time of the emitters T_2^*. If the emitters are initially excited to be in the same phase, T_2^* will result in them losing their phase relation due to the dipoles rotating with different rates. Once the relative phase is lost, the analysis in the manuscript becomes invalid. To quantify this limit on our predictions, we consider how many emitters a single electron can interact with in time shorter than T_2^*. In such a short time, the energy disorder of the emitters can be neglected, and the analysis performed in this manuscript is valid. For a given T_2^*, the number of emitters effectively interacting coherently with the electron is approximately $\frac{vT_2^*}{\Delta z}$, where v is the velocity of the free electron and Δz is the spacing between emitters. In modern quantum dot systems, advanced fabrication techniques allow for the observation of T_2^* on the ps scale even in room temperature [8]. Then, for $T_2^* = 1$ ps, if we consider 200 keV electrons (velocity of $0.7c$), as is typical inside an ultrafast transmission electron microscope, we get that up to 1,000 emitters can interact coherently with each electron over 200 um, for emitter spacing of 200 nm.

Section IV – Reconstruction of the superradiant dynamics.

This section discusses Fig. 4 of the main text. We consider an initial state containing the emitters in a general state (inside the superradiant sub-Hilbert space) and an electron with initial energy E_0. The joint electron-emitters wavefunction is $|\Psi_\mathrm{i}\rangle = \sum_m c_m |m\rangle \otimes |E_0\rangle$. Then the final state is given by:

$$|\Psi_\mathrm{f}\rangle = S|\Psi_\mathrm{i}\rangle,$$

$$|\Psi_\mathrm{f}\rangle = \sum_m c_m \sum_k (\cos|g|)^N (i\tan|g|)^k \sqrt{m!(k+m)!(N-k-m)!(N-m)!} \times$$

$$\sum_{l=0}^{m} \frac{(-1)^l (\tan|g|)^{2l}}{l!(m-l)!(k+l)!(N-k-m-l)!} |k+m\rangle \otimes |E_0 - k\cdot\hbar\omega_0\rangle. \#(\mathrm{SI21})$$

The probability to measure the electron with energy $E_0 - k\cdot\hbar\omega_0$ is then given by:

$$P_{-k} = \sum_m d_{km} p_m,$$

$$d_{km} = (\cos^2|g|)^N (\tan^2|g|)^k m!(k+m)!(N-k-m)!(N-m)! \times \left| \sum_{l=0}^{m} \frac{(-1)^l (\tan|g|)^{2l}}{l!(m-l)!(k+l)!(N-k-m-l)!} \right|^2, \#(\mathrm{SI22})$$

where $p_m = |c_m|^2$.

To generate Fig. 4, we need to evaluate d_{km} numerically for some specific parameters and provide from some theory the statistics p_m as a function of time for the superradiating sample. In the case of Dicke superradiance [5], where all the emitters are located in a small volume, the statistics can be calculated numerically using closed-form differential equations provided by Bonifacio et al. [9]. However, for the long sample superradiance (Fig. 4b in the main text), no such equations are known. Thus, we use the Truncated-Wigner approximation (TWA) 10], where we evaluate the classical (or mean-field) equations of motion provided by Bonifacio et al. [11] for an ensemble of initial conditions. The TWA can provide an approximation for the moments of the probability distribution p_m, and from that we find the distribution using linear least square optimization.

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