Cooperative Lamb shift and superradiance in an optoelectronic device

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Abstract
When a single excitation is shared between a large number of two-level systems, a strong enhancement of the spontaneous emission appears. This phenomenon is known as superradiance. This enhanced rate can be accompanied by a shift of the emission frequency, the cooperative Lamb shift, issued from the exchange of virtual photons between the emitters. In this work we present a semiconductor optoelectronic device allowing the observation of these two phenomena at room temperature. We demonstrate experimentally and theoretically that plasma oscillations in spatially separated quantum wells interact through real and virtual photon exchange. This gives rise to a superradiant mode displaying a large cooperative Lamb shift.

1. Introduction

Superradiance is one of the many fascinating phenomena predicted by quantum electrodynamics that have first been experimentally demonstrated in atomic ensembles [1, 2] and more recently in condensed matter systems like semiconductor quantum dots [3], superconducting q-bits [4], cyclotron transitions [5] and plasma oscillations in quantum wells (QWs) [6]. It occurs when a dense collection of $N$ identical two-level emitters are phased via the exchange of photons [7], giving rise to enhanced light–matter interaction, hence to a faster emission rate [8, 9]. Superradiance can be obtained by preparing the emitters in different ways: a well known procedure is to promote all of them in the excited state and observe their coherent decay through successive emission of $N$ photons into free space [9]. Of great interest is also the opposite regime where the ensemble interacts with one photon only and therefore all of the atoms, but one, are in the ground state. In this case the quantum superposition of all possible single emitter excitations produces a symmetric state that decays radiatively with a rate $N$ times larger than that of the individual oscillators. This phenomenon, also called single photon superradiance [10], was first predicted by Dicke [8], whose model describes the phasing of the emitters by the exchange of real photons. Yet, single photon superradiance is also associated with another collective effect that arises from virtual photon exchanges triggered by the vacuum fluctuations of the electromagnetic field. This phenomenon, known as cooperative Lamb shift [11–13], renormalizes the emission frequency, and was only recently evidenced experimentally in atomic systems [14–16].

In this work, we show that cooperative Lamb shift and superradiance can be engineered in a semiconductor device by coupling spatially separated plasma resonances arising from the collective motion of confined electrons in QWs. These resonances are associated with a giant dipole perpendicular to the QW plane. They have no mutual Coulomb coupling and interact only through absorption and re-emission of real and virtual free space photons. They thus behave as a collection of macro-atoms located on different positions along the growth direction $z$. Our device is therefore very valuable to simulate the low excitation regime of quantum electrodynamics in a solid state system.
This work is organized as follows. Section 2 presents the devices and their electrical characterization. The angle resolved electroluminescence measurements used to demonstrate cooperative Lamb shift and superradiance are also described. In section 3 we present our experimental results, which are then compared to the outcomes of our quantum model, presented in section 4. From the comparison between experimental and theoretical results, we demonstrate that all the observed effects can be interpreted in a quantum picture as a result of real and virtual photon exchange between plasma resonances in spatially separated QWs.

2. Samples and experiments

The two samples used in this study are based on GaInAs/AlInAs highly doped QWs grown by metal organic chemical vapor deposition on an InP substrate. The first one (SQW) consists of a single 45 nm GaInAs layer, n-doped with a surface density $N_s = 7.5 \times 10^{13} \text{ cm}^{-2}$, sandwiched between two 15 nm AlInAs barriers. The second sample (MQW) has been designed such that six 45 nm QWs, identical to that of SQW sample, n-doped with surface density $N_s$, are distributed within one wavelength and separated from one another by a sufficiently thick AlInAs barrier (15 nm) to avoid tunneling.

Both samples are processed into field effect transistor-like structures (insets of figure 1), consisting of two ohmic contacts for source–drain current injection and a Ti/Au 50 × 50 μm² top mirror. As for the SQW sample, the source and drain contacts are made of Ni/Ge/Au/Ni/Au metals annealed at 400 °C. For the MQW sample, we explicitly connected electrically only the top QW by depositing non-alloyed Ti/Au ohmic contacts directly on the first GaInAs layer. As a consequence in the MQW device electrons located in different wells only interact via the exchange of free space photons. Figure 1 shows that SQW (red lines) and MQW (blue lines) devices display the same source–drain voltage–current characteristics.

The samples have been prepared for angle-resolved thermal emission measurements, by mechanically polishing a facet. In order to span a wide angular range, devices with different polishing angles $\theta_p$ (20°, 45°, and, only for the SQW, 60° and 80°) are prepared. Thermal emission experiments are performed at room temperature by applying an in-plane current modulated at a frequency of 10 kHz with a 50% duty cycle and a fixed electrical power of 400 mW. The radiation emitted through the polished facet is focused onto an $f/1.5$ anti-reflection coated aspheric germanium lens through a polarizer, sent to a Fourier transform interferometer (FTIR) and then detected using a lock-in technique by a mercury cadmium telluride (MCT) detector cooled at 77 K. The signal is focused on the detector through another germanium lens ($f/0.5$). The measured spectra are then treated, following the procedure described in appendix A, in order to isolate the contribution of the plasma resonances to the incandescence spectra.

3. Experimental results

In previous work [6] we demonstrated that the plasma resonance of a highly doped QW, called multisubband plasmon [17], can be thermally excited by applying a current through a source–drain contact. The multisubband plasmons, issued from Coulomb interaction among electronic transitions within the conduction band of the well, superradiantly decay into free space, with a rate proportional to the electronic density in the QW, $N_e$. The emission spectrum measured at a given internal angle $\theta$ presents a unique peak, whose linewidth contains a non-
radiative contribution $\gamma$ and a radiative one $\Gamma(\theta)$ given by:

$$\Gamma(\theta) = \Gamma_0 \sin^2 \theta / \cos \theta \propto N_q \sin^2 \theta / \cos \theta,$$

(1)

where $\Gamma_0$ depends on $N_q$ and on the confining potential [18] (see also appendix B). The dependence on $\sin^2 \theta$ accounts for the fact that the plasmon collective dipole is oriented along the growth direction $z$ of the QW. The $1 / \cos \theta$ factor is due to the lack of wavevector conservation along $z$, resulting in the interaction of the plasmon with a one-dimensional density of photon states.

The radiative broadening $\Gamma(\theta)$ characterizes the strength of the light–matter interaction. By varying the angle we can therefore explore very different regimes of interaction as $\Gamma(\theta)$ varies from zero to a divergence [18].

At low values of $\theta$, when the coupling is weak, the linewidth is dominated by non-radiative effects. We have experimentally studied this regime between $6^\circ$ and $16^\circ$ by performing reflectivity measurements through the substrate on unprocessed samples, with only a gold mirror on the top surface. Figure 2(a) presents two spectra measured at $6^\circ$ internal angle on SQW (red line) and MQW (blue line) sample. The two normalized spectra are identical (see inset of figure 2(a)) with a plasmon resonance at $1650\text{meV}$ and very similar linewidths ($7.6 \text{meV}$ and $7.1 \text{meV}$ respectively for MQW and SQW).

When the angle is increased, the radiative broadening $\Gamma(\theta)$ increases and becomes dominant over $\gamma$. Figure 2(b) compares two emission spectra measured at $\theta = 35^\circ$ for the SQW (red line) and the MQW (blue line) samples. Although the two samples are made of identical QWs (one for SQW and six for MQW) their emission spectra have a completely different shape. The main multisubband plasmon peak is much broader for MQW sample than for SQW. Furthermore, a second resonance at $\approx 185 \text{meV}$, associated with an excited multisubband plasmon mode [20], is much more apparent in the MQW than in the SQW sample. Finally, the total incandescence signal of MQW device (directly related to absorption by Kirchhoff’s law [21]) is only twice

![Figure 2](image-url)
the SQW one. This is a strong evidence that, contrary to the low angle case, light–matter interaction is not perturbative and has to be described by an exact model \cite{18}.

Red (blue) bullets in figure 2(c) present the full width at half the maximum, $\gamma + \Gamma(\theta)$, of the main multisubband plasmon peak, extracted from emission and absorption measurements on the SQW (MQW) device, as a function of the internal angle $\theta$. For the SQW device, the data follow very well equation (1) (black line), with a rate $\hbar \Gamma_0 = 13$ meV corresponding to the nominal density of electrons in the QW. The larger broadening of the main emission peak in the MQW device indicates a much faster radiative decay for this sample than in SQW. This arises because multiple photon absorption and re-emission mediate an effective interaction between plasmons located in different wells. This light-mediated interaction between spatially separated plasmons gives rise to a superradiant mode extending over all the QWs in the structure, which gathers the oscillator strength of all plasmons. Similar effects have also been observed with excitons in periodic QW structures \cite{22, 23}.

Figure 2(d) presents the measured shift of the main peak position (with respect to the peak energy at $\theta = 6^\circ$) extracted from absorption and emission measurements. While the shift is negligible for the SQW sample (2 meV between 0° and 55°), it becomes substantial for MQW. In the following sections, we demonstrate that the observed blueshift corresponds to a cooperative Lamb shift arising from virtual photon emission and reabsorption processes \cite{11}.

4. Quantum model

In order to prove the physical interpretation of our experimental observations, we have extended the non-perturbative model developed in previous work \cite{18, 21}, which was based on a single infinitely thin charged plane, to a distribution of QWs in the z-direction. This model relies on quantum Langevin equations, describing the dynamics of plasmons, coupled with an electronic and a photonic bath, as schematized in the top panel of figure 3. The model is valid in the weakly excited limit, i.e. when the density of electronic excitations is much smaller than the doping density $N_e$. This description has been shown to be accurate for the experimental conditions considered in this study (thermal excitation slightly above room temperature or optical excitation from a low-power source \cite{17, 21}). In these cases, the plasmons can be treated as bosons despite the fermionic nature of their elementary constituents (electrons) \cite{24}, and the optical properties of the QWs can be derived using a semiclassical description based on non-local susceptibility formalism \cite{25}. Our model, however, is fully quantum and it provides without any additional assumption, the incandescent emission of the system, thus demonstrating Kirchhoff’s law of thermal emission for a gray body \cite{21}. This is calculated by considering an electronic input, corresponding to a thermal excitation of the electronic bath.

Thermal pumping guarantees to operate in the weak excitation regime. In this situation our system is the solid state equivalent of an atomic ensemble displaying single-photon superradiance \cite{13}. As a consequence, our model permits a clear cut quantum interpretation of the experimental results in terms of exchange of real and
virtual photons among plasmons located in different regions of space, giving rise respectively, to superradiance and Lamb shift.

The annihilation operator of the main plasmon mode located in the QW of index \( n \) (at position \( z_n \), see lower panels in figure 5) and characterized by an in-plane wavevector \( k \) is denoted \( P_{n,k} \). For simplicity, only the main plasmon mode at energy \( \hbar \omega_0 \) is included in this theoretical discussion (see appendix B for the full theoretical method, employed to simulate our experiments). The variations of \( P_{n,k} \) are given by quantum Langevin equations:

\[
\frac{dP_{n,k}}{dt} = \left[ -i\omega_0 - \frac{\gamma}{2} \right] P_{n,k}(t) - \sum_{n'} \frac{\Gamma_{n'}(\theta)}{2} P_{n',k}(t) + F_{n,k}(t). 
\]

In the above, \( \omega_0 \) and \( \gamma \) are considered independent on the QW index \( n \), as all the wells are identical. The operator \( F_{n,k} \) is the Langevin force, that arises from the interaction of plasmons with their fluctuating environment. This force is responsible for the thermal excitation of plasmons. The rates \( \Gamma_{n}(\theta) \) characterize the exchanges between plasmons through the bath of free space photons. Considering the spatial distribution of the electromagnetic field in the presence of the top mirror, the radiative rates \( \Gamma_{n}^{\text{r}} \) can be written as (see appendix B for their derivation):

\[
\text{Re}[\Gamma_{n}^{\text{r}}(\theta)] = \cos(qz_n)\cos(qz_{n'}) \Gamma(\theta), \\
\text{Im}[\Gamma_{n}^{\text{r}}(\theta)] = \frac{\sin(q|z_n - z_{n'}|) + \sin(q|z_n + z_{n'}|)}{2} \Gamma(\theta).
\]

The real part of the radiative rates is related to the exchange of real photons, and it determines the radiative broadening, while the imaginary part, associated with the exchange of virtual photons, gives rise to the Lamb shift of the emission energy. In the SQW case (i.e. \( n = n' = 1 \) and \( qz \ll 1 \) the real part of the radiative rate is given by (1). While its imaginary part, the SQW Lamb shift, is negligible (see the black line on figure 2(d)). In the MQW case, the rates \( \Gamma_{n}^{\text{r}}(\theta) \), that describe the radiative decay of plasmons in each well, depend on the QW index \( n \) and their imaginary part cannot be neglected. Furthermore, non-diagonal rates, corresponding to photon-mediated exchanges between separated wells, give rise to a single superradiant mode in which plasmons of all the different QWs oscillate in phase.

In order to gain further insight, we have derived the Langevin equation for this superradiant mode issued from the spatially separated plasmons. Due to the variations of the field over the structure thickness, the superradiant mode does not correspond exactly to the symmetric combination of all plasmons. Its annihilation operator is instead given by: \( P_{\text{S},k} = n_{\text{QW}}(\theta)^{-\frac{1}{2}} \sum_{n} \cos(qz_n)P_{n,k} \). Its dynamics is described by the single operator equation:

\[
\frac{dP_{\text{S},k}}{dt} = \left[ -i[\omega_0 + L_{\text{S}}(\theta)] - \frac{\gamma}{2} - n_{\text{QW}}(\theta) \frac{\Gamma(\theta)}{2} \right] P_{\text{S},k}(t) + F_{\text{S},k}(t).
\]

In this equation we can clearly identify the superradiant emission rate \( n_{\text{QW}}(\theta)\Gamma(\theta) \) and the frequency shift of the superradiant mode frequency \( L_{\text{S}}(\theta) \). The effect of the superradiant emission rate can be observed in figure 2(c). Indeed, the experimental data for MQW device in figure 2(c) are well reproduced by equation (1) replacing \( \Gamma_0 \) with \( n_{\text{QW}}(\theta) \Gamma_{n}^{\text{r}} \) (black line in figure 2(c)), with \( n_{\text{QW}}(\theta) \approx 4 \) at low angle and \( n_{\text{QW}}(\theta \to 90^\circ) \approx 6 \) (the actual number of QWs). The cooperative Lamb shift \( L_{\text{S}}(\theta) \) arises from virtual photon exchanges, that are described by the imaginary part of the rates \( \Gamma_{n}^{\text{r}} \). In appendix B we derive an analytical expression for \( L_{\text{S}}(\theta) \), which tends to increase with the number of QWs, but also depends in an intricate way on the well positions \( z_n \). The calculated Lamb shift is compared with the observed blueshift of the resonance in figure 2(d) (full black line), showing a remarkable agreement for both samples that confirms our physical interpretation.

Note that in the long wavelength approximation (i.e. if \( z_n \ll q^{-1} \) for all \( n \)), the overlap factor can be neglected, \( \text{Re}[\Gamma_{n,k}(\omega)] \propto \frac{k}{q} \) and it follows that \( \text{Im}[\Gamma_{n,k}(\omega)] = 0 \), so that the Lamb shift is exactly zero in thin structures. This approximation is valid for SQW sample, in which the center of the well is located only 90 nm away from the gold layer, but not in MQW sample. As a consequence, if all the electrons in the MQW sample were concentrated into the top QW, the measured thermal emission spectra would have been completely different, displaying a negligible Lamb shift.

### 5. Results and discussion

In order to simulate the full emission spectra, we consider an incoherent thermal input at temperature \( T \) in the electronic bath, corresponding to a Langevin force:
\[ T = \text{the temperature of the electron gas, which is higher than the substrate temperature due to Joule heating induced by the source–drain current. For the SQW sample, we have considered that the current flowing in the doped InGaAs well induces a temperature increase } \Delta T = 80 \text{ K for the electronic bath, with respect to the lattice temperature } T_0 = 300 \text{ K } (T = T_0 + \Delta T). \text{ The value of } \Delta T \text{ has been estimated by studying the increase of the emitted optical power as a function of the electrical power injected in the device, following } [26]. \text{ In the MQW device we assume that, although only one QW is electrically contacted, the electronic temperature increases equally in all the wells during the electrical pulses, due to the small thickness of the sample. For simplicity we also assume that the increase of the electronic temperature } \Delta T \text{ is identical in the MQW and SQW cases.} \]

The outcomes of our model (including all the plasmon modes) are summarized in the top panels of figure 4, which present the complete angular and energy behavior of the two devices, compared to the experimental results (middle panels). For the SQW device, the Lamb shift is negligible and the emission peak is maximum at \( \theta \approx 40^\circ \), where the critical coupling condition \([21]\) \( \gamma = \Gamma (\theta) \) is met. For MQW device, due to the stronger interaction with free space radiation, the blueshift of the main plasmon peak increases with \( \theta \) and critical coupling condition \( \gamma = n_{QW} (\theta) \Gamma (\theta) \) is fulfilled at lower angle \( \theta \approx 20^\circ \). Beyond \( \theta \approx 40^\circ \), the second plasmon peak at 185 meV becomes dominant.

The comparison between experimental and theoretical results shows that our model provides a very good understanding of the variations of the linewidth, position and amplitude of the plasmon modes in both samples and supports their interpretation in terms of superradiance and cooperative Lamb shift.

\[ \langle F_{n,k}^{(1)} (\omega') F_{n,k'}^{(1)} (\omega) \rangle = \frac{2 \pi \gamma}{\delta_k^2} \frac{\delta (\omega - \omega')}{\cos \theta - 1} \]
In the simulated spectra of MQW sample it is possible to observe, at high angles, an emission peak at energy close to $\hbar \omega_0$. This corresponds to a dark (or subradiant) mode which is coupled to the bright plasmon thanks to virtual photon processes. Indeed, as discussed in appendix B, the imaginary part of $\Gamma^{\alpha}(\theta)$ induces an effective coupling between bright and dark modes. This effect was also predicted by Svidzinsky and Scully in [27]: in large atomic ensembles (with dimensions much larger than the wavelength of light), the symmetric excitation is dark, but it acquires a finite radiative lifetime thanks to virtual photon processes. The fact that this mode is not seen in the experimental spectra is still under investigation. It might arise from spatial disorder and inhomogeneity between the different wells or from the finite coherence time of photons in the structure that is not accounted for in our model.

The bottom panels of figure 4 show the variations of the emitted power per solid angle with $\theta$ for the two devices. At very low angles, the emitted power increases proportionally to $\Gamma(\theta)$ for SQW (left) and to $n_{QW}(\theta)\Gamma(\theta)$ for MQW (right), as it would be expected from a perturbative treatment of the light–matter interaction. However, at higher angles light–matter interaction is non-perturbative and the two devices display different angular behaviors, with a maximum emission at $35^\circ$ ($55^\circ$) for the MQW (SQW) sample. The predictions of our model (full lines) are well corroborated by the experimental results for the total power (colored dots). This further confirms that the observed behavior is a consequence of the superradiant enhancement of the radiative rate in MQW device. Note that although the two devices have identical electrical characteristics, the maximum emitted power is significantly increased in MQW with respect to SQW, showing that multiple QW superradiance is a possible approach to improve the performance of thermal emitters in the mid-infrared.

In summary we demonstrated a room temperature semiconductor platform that allows probing some of the most fundamental properties of quantum electrodynamics, like superradiance and Lamb shift, which are usually the realm of atomic physics. Furthermore, the observed effects open new perspectives in the development of efficient mid-infrared sources.

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Appendix A. Extraction of the plasmonic contribution to the incandescence spectrum

In this section we present the data treatment realized on the measured spectra in order to extract the plasmonic contribution to the thermal emission. In figure A1 we present an example of this treatment on the data measured at $20^\circ$ from the SQW (a) and MQW (b) device.

The measured spectra are first corrected by the response curve of the MCT detector and by the measured transmission of the lenses, the FTIR and the polarizer. In order to compare data extracted from samples with different polishing angles, we also normalize the spectra by accounting for the Fresnel coefficients. The resulting spectra in transverse magnetic (TM) and transverse electric (TE) polarization are shown as blue lines and red lines respectively in the two panels of figure A1. It is important to remark that in TM configuration the signal is the sum of a contribution polarized along the growth axis and due to the multisubband plasmon emission, and of a contribution with polarization parallel to the plane of the QWs. On the contrary, the TE contribution corresponds to purely in-plane polarization, thus perpendicular to the plasmon dipole. It is thus related to the blackbody emission of the sample, without including the plasmon contribution. It can thus be considered as a background spectrum, that allows extracting the plasmon contribution from the spectrum measured in TM configuration.

In order to extract the multisubband plasmon emission, we fitted the TM emission spectra $P_{TM}(\omega)$ by the sum of two Lorentzian functions $L_0$ and $L_1$ (accounting respectively for the main multisubband plasmon, at $\hbar \omega_0 \sim 162$ meV, and for the second plasmon at $\hbar \omega_1 \sim 185$ meV), and of the blackbody background contribution proportional to the TE signal (the proportionality factor $h$ is taken as a fitting parameter):

$$P_{TM}(\omega) = L_0(\omega) + L_1(\omega) + h \times P_{TE}(\omega).$$

(A.1)

The result of this fitting procedure is displayed as dashed black lines. It is worth noting that for the SQW sample, a small additional peak appears in the TE signal around 151 meV which is absent from the TM spectra. To correct for this minor contribution (whose origin is still under investigation) we fitted the emission spectra of SQW sample with the following equation:
where $L_2$ is a Lorentzian peak centered at 151 meV. In the fitting procedure, the energy and the width of $L_1$ and $L_2$ were fixed to reduce the number of parameters (the energy of the main peak $L_0$ strongly varies with the angle due to the Lamb shift, as explained in the main text). The factor $h$ is approximately unity for the whole interval of emission angles exploited.

The final spectrum, only including the plasmon contributions, is represented by black lines in figure A1. This procedure has been used to extract all the spectra presented in the figures of the main article.

Appendix B. Full theoretical model for incandescent emission of several plasmons in spatially separated QWs

In this section we present the full theoretical model used in order to simulate the thermal emission spectra of a highly doped multi-QW structure. This model is based on the resolution of the quantum Langevin equations for thermal emission of plasmons and it is an extension to multiple QW devices of the theory presented in the references [18, 21].

We consider a system constituted of $N$ identical QWs labeled by the quantum number $n$ and located at distance $z_n$ from the reflecting gold layer. We denote $m = 1, \ldots, M$ the index of the multisubband plasmon issued from the diagonalization of the dipole–dipole interaction in each well, as presented in [20]. These plasmons are collective oscillation modes of the two-dimensional electron gas confined in the QW, their frequency is denoted $\omega_{m}$ and they are described by the creation and annihilation operators $P_{\alpha, m, k}$ and $P_{\alpha, m, k}^{\dagger}$ which follow bosonic commutation rules. Each plasmon is coupled to an electronic reservoir, which is responsible for both the non-radiative decay of the plasmon and its thermal excitation with a constant rate $\gamma$ that can be inferred from the low-angle measurements.

All plasmon modes are also coupled to a common photonic reservoir consisting of the free space photon modes of the structure. Due to the presence of a metallic mirror on the top of the samples, we quantize the

\[ P_{\text{TM}}(\omega) = L_0(\omega) + L_1(\omega) + h \times P_{\text{TE}}(\omega) - L_2(\omega), \]  

(A.2)
electromagnetic field by considering the TM modes of a double metal cavity of infinite thickness. The creation and annihilation operators associated with the photon with in-plane wavevector $k$ and frequency $\omega$ are denoted $a^\dagger_{k,\omega}$ and $a_{k,\omega}$. The associated out-of-plane component of the wavevector, $q$, is such that $\omega = \frac{c}{\sqrt{\varepsilon_r}} \sqrt{k^2 + q^2}$, with $c$ the velocity of light in the semiconductor. Therefore, for a given $k$, the photon reservoir consists of a continuum of bosonic modes with energy ranging from $\frac{\omega}{\sqrt{\varepsilon_r}}$ to infinity. The intensity of the light–matter coupling between plasmons and photons can be computed from a microscopic description of the collective electronic excitations [18]. It depends on all the quantum numbers $n$, $m$, $k$, and $\omega$. The full system Hamiltonian is written as:

$$H = \sum_{n,m,k} \hbar \omega_{nm} p^\dagger_{n,m,k} p_{n,m,k} + \sum_{n,m,k} \int d\omega \, \hbar \omega \, b^\dagger_{n,m,k,\omega} b_{n,m,k,\omega} + \sum_{k} \int d\omega \, \hbar \omega \, a^\dagger_{k,\omega} a_{k,\omega} + i\hbar \sum_{k,n,m} \int_0^{\infty} d\omega \, W_{n,m,k,\omega} [a^\dagger_{k,\omega} - a_{k,\omega}] [p^\dagger_{n,m,k} + p_{n,m,-k}]$$

$$+ i\hbar \sum_{k,n,m} \int_0^{\infty} d\omega \, \frac{\omega}{2\pi} [b^\dagger_{n,m,k,\omega} - b_{n,m,-k,\omega}] \times [p_{n,m,k} + p^\dagger_{n,m,-k}]. \quad (B.1)$$

The operators $b_{n,m,k,\omega}$ describe the annihilation of an excitation in the electronic bath, each plasmon being considered as coupled to an independent bath. The first three terms on the right-hand side of this equation correspond to the energy of each isolated subsystem (the plasmon energy, the electronic and photonic reservoirs energy), while the last two terms describe respectively light–matter interaction and non-radiative scattering. The expression for the last term is the simplest possible, where all non-radiative phenomena are described by a single phenomenological rate $\gamma$. The light–matter coupling parameters $W_{n,m,k,\omega}$ are given by:

$$W_{n,m,k,\omega} = \sqrt{\frac{d_{nm}}{\sqrt{2\pi} \omega_{nm} \sqrt{\varepsilon_r}}} \cos(qz_n). \quad (B.2)$$

The constant $d_{nm}$ computed from a numerical diagonalization procedure, characterizes the oscillator strength of the plasmon mode of index $m$ [20], while the denominator $\sqrt{\varepsilon_r}$ causes the spontaneous emission rate to diverge at 90° as it was analyzed extensively in [6, 18, 21]. The factor $\cos(qz_n)$ comes from the fact that the distance $z_n$ between the QWs and the gold surface can be a significant fraction of the optical wavelength, therefore, the variations of the electromagnetic field over the thickness of the structure cannot be neglected (see figure 3 of the main text). The QW used in this study has a very high electronic density with a Fermi energy more than 300 meV above the band edge of InGaAs. For this reason, band non-parabolicity effects become important and the numerical diagonalization procedure described in [20] is slightly modified in order to compute the parameters $\omega_{nm}$ and $f_{nm}$ with more accuracy. Indeed these parameters are obtained by calculating the dipole–dipole coupling between all the optically active intersubband excitations, characterized by an energy which depends on the electron in–plane momentum. In our simulations we consider this dependence, due to non-parabolicity, in an effective way. We consider that for each intersubband transition, due to Pauli blocking the only electrons involved in the interaction with light are those close to the Fermi level. As a consequence, we fix the energy of a given intersubband transition to its value close to the Fermi level. The rates $f_{nm}$ are multiplied by a global factor 0.75 to account for the increase of the effective mass with energy. We have verified that the absorption spectrum calculated at low angle by using this procedure is equivalent to the one simulated by using a Drude–Lorentz model, including in a more rigorous way non-parabolicity effects through a summation over the in–plane electron wavevector for each transition [17].

From the hamiltonian (B.1), we derive the quantum Langevin equations, by following the same procedure as in [18, 21]:

$$\frac{dp^\dagger_{n,m,k}(t)}{dt} = -i\omega p^\dagger_{n,m,k}(t) - \frac{\gamma}{2} [p^\dagger_{n,m,k}(t) + p_{n,m,-k}(t)]$$

$$- \sum_{n',m'} \int d\tau \left[ \frac{\Gamma^+_{n',m',k}(\tau)}{2} - \frac{\Gamma^-_{n',m',k}(\tau)}{2} \right] [p^\dagger_{n',m',k}(t - \tau) + p_{n',m',-k}(t - \tau)]$$

$$+ f_{n,m,k}(t) - p^\dagger_{n,m,-k}(t), \quad (B.3)$$

$$\frac{dp_{n,m,k}(t)}{dt} = i\omega p_{n,m,k}(t) + \frac{\gamma}{2} [p^\dagger_{n,m,k}(t) + p_{n,m,-k}(t)]$$

$$+ \sum_{n',m'} \int d\tau \left[ \frac{\Gamma^+_{n',m',k}(\tau)}{2} - \frac{\Gamma^-_{n',m',k}(\tau)}{2} \right] [p_{n',m',k}(t - \tau) + p^\dagger_{n',m',-k}(t - \tau)]$$

$$+ f_{n,m,k}(t) - p_{n,m,-k}(t), \quad (B.4)$$

The constants $f_{nm}$ are obtained by minimizing the classical mechanical energy of the system with respect to the Fermi energy $E_F$, which is given by the condition

$$\frac{d}{dE_F} \left( \sum_{n,m} \int d\omega \, W_{n,m,\omega} \right) = 0.$$
where we have defined the radiative damping function:

$$\Gamma_{n,m,k}'(\tau) = \Theta(\tau) \int d\omega \frac{2W_{n,m,k\omega}}{2} W_{n,m',k\omega} e^{-i\omega \tau}$$

with $\Theta(\tau)$ the Heaviside step function. This equation can be reduced to equation (2) of the main text by making three approximations: the rotating-wave approximation (RWA) that consists in discarding the rapidly oscillating terms $P_{n,m-k}$ and $F_{n,m-k}$; the single bright plasmon approximation (SBPA) that amounts to eliminate all plasmon modes $m$ except for the one with the highest coupling rate $f_{nm}$; and Markov approximation (MA). The last approximation relies on the fact that $\Gamma_{n,k}'(\tau)$ is a rapidly decaying function compared to the typical evolution time of the operator $P_{n,k}$. It can therefore be replaced by $\hat{\Gamma}_{n,k}^{\prime} (\omega_0) \delta(\tau)$, where the tilde denotes the Fourier transform. The equation obtained with these simplifications is:

$$\frac{dP_{n,k}}{dt} = \left[ -i\omega_0 - \frac{\gamma}{2} \right] P_{n,k}(t) - \sum_{m} \left[ \frac{\hat{\Gamma}_{n,k}^{\prime}(\omega_0)}{2} - \frac{\hat{\Gamma}_{n,k}^{\prime}(-\omega_0)^{\ast}}{2} \right] P_{n,m,k}(t) + F_{n,k}(t).$$

This is exactly equation (2) of the main text with the definition:

$$\hat{\Gamma}_{n,k}^{\prime}(\omega) = [\hat{\Gamma}_{n,k}^{\prime}(\omega_0) - \hat{\Gamma}_{n,k}^{\prime}(-\omega_0)^{\ast}],$$

where $\theta$ is the emission angle measured with respect to the normal to the QW plane. Using the general properties of the Heaviside function, we derive the following relations for the real and imaginary parts of $\hat{\Gamma}_{n,k}^{\prime}(\omega)$ describing real and virtual photon emission respectively:

$$\text{Re} \left[ \hat{\Gamma}_{n,k}^{\prime}(\omega) - \hat{\Gamma}_{n,k}^{\prime}(-\omega)^{\ast} \right] = 2\pi W_{n,m,k\omega} W_{n,m',k\omega}$$

$$\text{Im} \left[ \hat{\Gamma}_{n,k}^{\prime}(\omega) - \hat{\Gamma}_{n,k}^{\prime}(-\omega)^{\ast} \right] = \int_0^\infty d\omega' \frac{4\omega'}{\omega_{nm\omega'}^2} W_{n,m,k,\omega'} W_{n,m',k,\omega'}$$

$$\frac{ck^2}{\sqrt{2\pi}} \int_0^\infty d\omega' \frac{4\omega'}{\omega_{nm\omega'}^2} \int_0^{\infty} dq \frac{2\cos(q\sin(\theta))\cos(q\sin(\theta_0))}{q^2 - k^2 - q^2}$$

$$\frac{ck^2}{\sqrt{2\pi}} \int_0^\infty d\omega' \frac{4\omega'}{\omega_{nm\omega'}^2} \int_0^{\infty} dq \frac{2\cos(q\sin(\theta))\cos(q\sin(\theta_0))}{q^2 - k^2 - q^2}$$

From this we deduce equations (3) and (4) of the main text, within the three approximations mentioned above. The validity of RWA and MA in this context was explored in a previous article [18]. It was shown that they are accurate for low emission angles but fail at high angles when the system enters the ultra-strong coupling regime, i.e. when the radiative broadening becomes comparable with the bare plasmon frequency $\omega_0$. Furthermore, SBPA only allows describing the main emission peak (around 165 meV for our samples), and not the second peak that becomes particularly significant at high angles. Therefore equation (2) provides a very good estimate for the linewidth and for the cooperative Lamb shift of the main superradiant mode at moderate angles. However, to simulate the emission spectra over the full spectral and angular range experimentally accessible, we rely on a numerical resolution of (B.3).

The calculation of the emission spectrum is based on the input–output formalism. We first introduce initial time $t_0$ and final time $t$ and define the input and output reservoir operators:

$$a_{in}^{\dagger} = a_{in}(t_0) e^{i\omega t}$$
$$a_{in}^{\dagger} = a_{in}(t_0) e^{i\omega t}$$
$$b_{out}^{\dagger} = b_{out}(t) e^{i\omega t}$$
$$b_{out}^{\dagger} = b_{out}(t) e^{i\omega t}$$

The emission spectra are obtained by computing the output photon number $\langle a_{out}^{\dagger} a_{out} \rangle$. To calculate this quantity we first derive the input–output relation for the photon operators, which follows from the dynamical equation for $a_{in}$ considered in the limit $t \rightarrow \infty$ and $t_0 \rightarrow -\infty$:

$$a_{out} = a_{in} + \sum_{n,m} W_{n,m,k} [\hat{P}_{n,m,k}(\omega) + \hat{\Gamma}_{n,m-k}(\omega)].$$

We then determine the plasmon operators $\hat{P}_{n,m,k}(\omega)$ by performing a Fourier transform of (B.3), from which we obtain a linear relation between the Langevin forces and the plasmon operators:

$$\hat{P}_{n,m,k}(\omega) = \sum_{n',m'} \{ M_{k}(\omega) \} n',m' \hat{P}_{n',m',k}(\omega).$$
Hence, inverting numerically the matrix $M_k (\omega)$ allows us to compute the plasmon operators from the value of the Langevin forces. The latter can then be defined from the input operators of the reservoirs. In Fourier space they are given by:

$$\tilde{f}_{n,m,k}(\omega) = \sqrt{2\pi\gamma} \, b^\dagger_{n,m,k,\omega} + 2\pi \, W_{n,m,k}\omega \, a^\dagger_{k,\omega}. \quad (B.10)$$

In order to compute the thermal emission of the structure, we consider a purely non-radiative, thermal input in the electronic reservoirs, characterized by:

$$\langle \tilde{F}^+_{n,m,k}(\omega') \tilde{F}_{n,m,k'}(\omega) \rangle = 2\pi\gamma \, \phi^{kk'} \, \delta(\omega - \omega'). \quad (B.11)$$

From this method, it is possible to simulate the thermal emission spectra of SQW and MQW devices without using any of the three approximations mentioned above. However, the aforementioned approximations permit to obtain an analytical expression of the power emitted from a single QW [18, 21].

Let us now focus on the relevant approximations to describe thermal emission in multiple QW structures. We first define the MQW superradiant mode:

$$P_{S,k} = n_{QW} (\theta)^{-\frac{1}{2}} \sum_n \cos(qz_n) P_{n,k}, \quad (B.12)$$

with $n_{QW} (\theta) = \sum_n |\cos(qz_n)|^2$. \quad (B.13)

Note that the superradiant mode is not a perfectly symmetric excitation because individual plasmons in each well have different spontaneous emission rates:

$$\text{Re}[\Gamma^u_{n,k} (\omega_0)] \propto \cos^2(qz_n). \quad (B.14)$$

The approximated expressions (B.5) are combined to obtain:

$$\frac{dP_{S,k}}{dt} \simeq \left[-i\omega_0 - \frac{\gamma}{2} - n_{QW} (\theta) \frac{\Gamma(\theta)}{2} \right] P_{S,k} (t) - n_{QW} (\theta) \frac{1}{2} \sum_{n,n'} \cos(qz_{n'}) P_{n,k} (t)$$

$$\times \left[ \frac{1}{2} \text{Im}[\Gamma^u_{n,k} (\theta)] P_{n,k} (t) + F_{S,k} (t) \right]$$

$$\quad \text{with } \Gamma(\theta) = \frac{\Gamma_0}{\omega_0} \frac{ck^2}{\sqrt{m'q}} = \Gamma \frac{\sin^2 \theta}{\cos^2 \theta}. \quad (B.15)$$

Here $\Gamma_0$ is the effective oscillator strength of the bright multisubband plasmon. It can be shown [18] that $\Gamma_0 = f_0 \simeq \frac{eN}{2m'\sqrt{\epsilon'}}$, with $N_e$ the surface electronic density in the well and $m'$ the effective mass.

Due to the imaginary part of $\Gamma^u_{n,k} (\omega_0)$, i.e. to virtual photon processes, the equation above does not only involve $P_{S,k}$ but also other combinations of the $P_{n,k}$ operators, the dark modes. Therefore (B.15) induces an effective coupling between bright and dark modes: if only real photon processes were included, bright and dark modes would obey entirely independent Langevin equations and the dark ones would only decay non-radiatively, whereas due to virtual photon processes, they acquire a finite radiative lifetime. This effect was also predicted by Svidzinsky and Scully in [27]: in large atomic ensembles (with dimensions much larger than the wavelength of light), the symmetric excitation is dark, but it acquires a finite radiative lifetime thanks to virtual photon processes. In the simulated spectra provided in figure 4 of the main text, the effective coupling between bright and dark modes is responsible for the persistence of an emission peak close to the plasmon frequency at high angles. This corresponds to a dark (or subradiant) plasmon mode.

Finally to obtain the single operator Langevin equation (equation (5) of the main text) and derive an analytical expression of the cooperative Lamb shift, we project the imaginary term in (B.5) onto the bright operator and neglect the residual bright-dark coupling to obtain:

$$\frac{dP_{S,k}}{dt} = \left\{-i[\omega_0 + L_5 (\theta)] - \frac{\gamma}{2} - n_{QW} (\theta) \frac{\Gamma(\theta)}{2} \right\} P_{S,k} (t) + F_{S,k} (t), \quad (B.16)$$

where the superradiant mode frequency is shifted by the cooperative Lamb shift $L_5 (\theta)$ given by:

$$L_5 (\theta) = \frac{\Gamma(\theta)}{4n_{QW} (\theta)} \sum_{n,n'} \cos(qz_n) \cos(qz_{n'}) \sin (q|z_n - z_{n'}|) + \sin (q|z_n + z_{n'}|)). \quad (B.17)$$

This procedure provides a very accurate analytical expression for the MQW Lamb shift, as illustrated in the main text, although it does not allow for the description of the residual dark mode peak at high angle in the emission spectra, for which the full numerical resolution should be used.
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