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Quantum theory of electron tunneling into intersubband cavity polariton states

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Recent mid-infrared absorption and electroluminescence experiments on microcavities embedding quantum wells have shown strong coupling between a cavity photon mode and the transition between two conduction subbands, being the lowest one filled with a dense two-dimensional electron gas. Through a non-perturbative quantum theory, here we investigate how the electronic states are modified by the coupling to the microcavity vacuum field. We show that resonant electron tunneling from a narrow-band injector can selectively excite superradiant states and produce ultraefficient polariton electroluminescence.

(June 19, 2008)

Cavity quantum electrodynamics in the strong coupling regime is presently the subject of many fascinating investigations in several interesting systems, including ultracold atoms[1], Cooper pair quantum boxes [2] and semiconductor nanostructures[3]. In the strong coupling regime, the eigenstates of a cavity system are a coherent mixing of photonic and electronic excitations. This occurs when the light-matter interaction, quantified by the so-called vacuum Rabi frequency, is dominant with respect to loss mechanisms for the cavity photon field and for the electronic transition.

Recently, the strong coupling regime has been demonstrated also between a planar microcavity mode and an intersubband transition in a doped semiconductor quantum well. The normal modes of such a system are called intersubband cavity polaritons [4, 5, 6, 7, 8, 9, 10, 11]. The active electronic transition is between two conduction subbands, where a dense two-dimensional electron gas populates the lowest one. Large vacuum Rabi frequencies can be achieved thanks to the giant collective dipole associated to the dense electron gas and even an unusual ultra-strong coupling regime can be reached[8, 9, 12].

Electroluminescence experiments in microcavity-embedded quantum cascade devices[13] have recently demonstrated that it is possible to obtain intersubband cavity polariton emission after resonant electrical excitation even at room temperature. A fundamental question to address is how the strong interaction with the microcavity vacuum field modifies the quasi-electron states in the quantum well and how the electron tunneling is affected. In this Letter, we present a quantum theory to investigate such fundamental problem. We show that the electronic eigenstates originate from a Fano-like coupling between the bare injected electron and the continuum of cavity polariton modes. Our theory demonstrates that resonant electron tunnelling from a narrow-band injector contact can selectively excite polaritonic states leading to ultraefficient polariton electroluminescence.

In order to describe the system under study, we consider the following second quantization Hamiltonian (for

details on the approximations behind this Hamiltonian see [14, 15]):

$$H = \sum_{\mathbf{k}} \hbar\omega_{1,k} c_{1,\mathbf{k}}^\dagger c_{1,\mathbf{k}} + \sum_{\mathbf{k}} \hbar\omega_{2,k} c_{2,\mathbf{k}}^\dagger c_{2,\mathbf{k}} + \sum_{\mathbf{k}} \hbar\omega_{c,q} a_{\mathbf{q}}^\dagger a_{\mathbf{q}} + \sum_{\mathbf{k},\mathbf{q}} \hbar\chi_q a_{\mathbf{q}} c_{1,\mathbf{k}} c_{2,\mathbf{k}+\mathbf{q}}^\dagger + \sum_{\mathbf{k},\mathbf{q}} \hbar\chi_q^* a_{\mathbf{q}}^\dagger c_{2,\mathbf{k}+\mathbf{q}} c_{1,\mathbf{k}}^\dagger, \quad (1)$$

where $\hbar\omega_{1,k} = \frac{\hbar^2 \mathbf{k}^2}{2m^*}$ and $\hbar\omega_{2,k} = \hbar\omega_{12} + \frac{\hbar^2 \mathbf{k}^2}{2m^*}$ are the energy dispersions of the two quantum well conduction subbands as a function of the in-plane wavevector \mathbf{k} , m^* being the effective mass. The corresponding electron creation operators are $c_{1,\mathbf{k}}^\dagger$ and $c_{2,\mathbf{k}}^\dagger$. $\omega_{c,q}$ is the frequency dispersion of the cavity photonic mode and $a_{\mathbf{q}}^\dagger$ is the corresponding photon creation operator.

If we wish to study the tunneling injection of one electron at low temperature, we have to determine the electron spectral function[16], defined as:

$$A_j^+(\mathbf{k}, \omega) = \sum_{\zeta} |\langle \zeta | c_{j,\mathbf{k}}^\dagger | F_N \rangle|^2 \delta(\omega - \omega_{\zeta}), \quad (2)$$

where $|F_N\rangle$ is the N-electron Fermi sea ground state times the vacuum state for the cavity photon field and $j = 1, 2$ is the conduction subband index. The index ζ labels the excited (N+1)-electron eigenstates and $\hbar\omega_{\zeta}$ is the corresponding eigenenergy. As apparent from Eq. (2), the electron spectral function is the density of quasi-electron states, weighted by the overlap with the bare electron state $c_{j,\mathbf{k}}^\dagger | F_N \rangle$. This is the key quantity affecting the electron tunneling and can be non-trivially modified by interactions like in the case of superconductors. For a non-interacting electron gas, $c_{1,\mathbf{k}}^\dagger | F_N \rangle$ and $c_{2,\mathbf{k}}^\dagger | F_N \rangle$ are eigenstates of the Hamiltonian and thus all the other eigenstates are orthogonal to them. Therefore the non-interacting spectral functions are

$$A_1^+(\mathbf{k}, \omega) = \delta(\omega - \omega_1(k)) \theta(k - k_F), \quad (3)$$

$$A_2^+(\mathbf{k}, \omega) = \delta(\omega - \omega_2(k)), \quad (4)$$

where k_F is the Fermi wavevector and $\theta(x)$ is the Heaviside function. The Heaviside function is due to Pauli blocking: $c_{1,\mathbf{k}}^\dagger | F_N \rangle = 0$ for $k < k_F$.

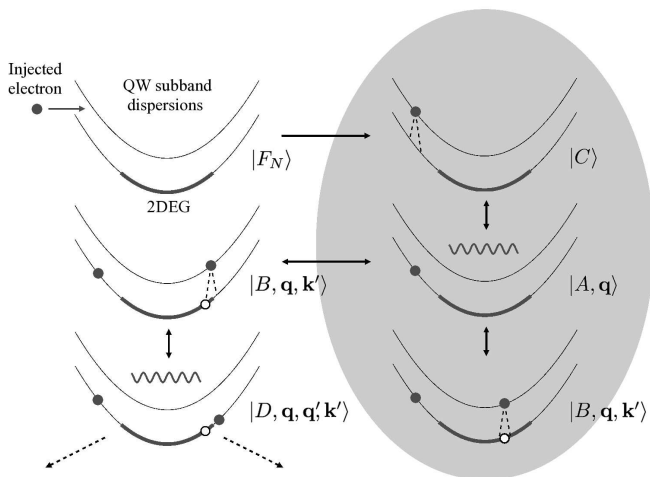


FIG. 1: Sketch of the dynamical coupling between quantum states in a microcavity-embedded quantum well (QW) containing a two-dimensional electron gas (2DEG). The electron gas partially fills the fundamental quantum well conduction subband, having a parabolic energy dispersion $\hbar\omega_{1,k}$ as a function of the electron in-plane wavevector k . The excited subband has a parallel energy dispersion $\hbar\omega_{2,k}$. The excited state obtained by injecting a bare electron in the second subband is not an eigenstate of the system. Due to spontaneous photon emission and reabsorption processes, such a state is coupled to a manifold of states sketched here. Being the relevant cavity photon wavevectors very small compared to the Fermi wavevector, spontaneous emission can occur only on narrow emission cone in momentum space. Due to the small probability of photon absorption by electrons on the border the Fermi sea, the relevant dynamics takes place only between the states in the shaded region.

In the interacting case, it is easy to verify that $c_{1\mathbf{k}}^\dagger|F_N\rangle$ is still an eigenvector of the Hamiltonian in Eq. (1) and thus the first subband spectral function is still given by Eq. (3). Instead for the electrons in the second subband we have to distinguish between two cases: k well inside or outside the Fermi sea. In the first case, an electron in

the second subband can not emit a photon because all the final states in the first subband are occupied (Pauli blocking), hence the spectral function will be given by the unperturbed one (Eq. (4)). Well outside the Fermi sea, it can emit and the spectral function will be modified by the interaction. A smooth transition between the two cases will take place for $|k - k_F|$ of the order of the resonant cavity photon wave-vector q_{res} , where $\omega_c(q_{res}) = \omega_{12}$. Being the ratio q_{res}/k_F typically very small, of the order of 10^{-2} [17], we can safely consider an abrupt transition at the Fermi edge.

For $k > k_F$ we need to find all the $(N+1)$ -electron eigenstates that have a nonzero overlap with $c_{2\mathbf{k}}^\dagger|F_N\rangle$. In order to do this we notice that the Hamiltonian in Eq. (1) commutes with the number of total fermions $\hat{N}_F = \sum_{j=1,2} \sum_{\mathbf{k}} c_{j,\mathbf{k}}^\dagger c_{j,\mathbf{k}}$, the total in plane wave-vector operator $\hat{\mathbf{K}} = \sum_{j=1,2} \sum_{\mathbf{k}} \mathbf{k} c_{j,\mathbf{k}}^\dagger c_{j,\mathbf{k}}$ and the excitation number operator $\hat{Q} = \sum_{\mathbf{k}} a_{\mathbf{k}}^\dagger a_{\mathbf{k}} + c_{2,\mathbf{k}}^\dagger c_{2,\mathbf{k}}$. Hence the eigenstates $|\zeta\rangle$ of H can be also labeled by the corresponding eigenvalues $N_\zeta, \mathbf{K}_\zeta$ and Q_ζ . We will thus identify an eigenstate of H in the subspace ($\hat{N}_F = N, \hat{\mathbf{K}} = \mathbf{K}, \hat{Q} = Q$) as $|N, \mathbf{K}, Q, \zeta\rangle$, where the index ζ now runs over all the eigenstates of the subspace. Having $|F_N\rangle$ quantum numbers $(N, \mathbf{0}, 0)$ it is easy to verify that the state $c_{2,\mathbf{k}}^\dagger|F\rangle$ is labeled by the quantum numbers $(N+1, \mathbf{k}, 1)$. We can thus limit ourselves to diagonalize H in this subspace, which is spanned by vectors of the form: (i) $c_{2,\mathbf{k}_0}^\dagger \prod_{j=1}^N c_{1,\mathbf{k}_j}^\dagger |0\rangle$, where $|0\rangle$ is the empty conduction band state and $\sum_{j=1}^N \mathbf{k}_j = \mathbf{k} - \mathbf{k}_0$; (ii) $a_{\mathbf{q}_0}^\dagger \prod_{j=1}^{N+1} c_{1,\mathbf{k}_j}^\dagger |0\rangle$ with $\sum_{j=1}^{N+1} \mathbf{k}_j = \mathbf{k} - \mathbf{q}_0$. For a large number of electrons, the exact diagonalization of the Hamiltonian in this subspace is an unmanageable task. Here, we show that by a judicious approximation, we can considerably simplify the diagonalization problem, keeping the relevant non-perturbative physics. Namely, we claim that the elements of $(N+1, \mathbf{k}, 1)$ subspace can be well approximated by vectors of the form:

$$|N+1, \mathbf{k}, 1, \zeta\rangle = \left\{ \mu_\zeta c_{2\mathbf{k}}^\dagger + \sum_{\mathbf{q}} \left[\alpha_\zeta(\mathbf{q}) a_{\mathbf{q}}^\dagger c_{1\mathbf{k}-\mathbf{q}}^\dagger + \sum_{|\mathbf{k}'| < k_F} \beta_\zeta(\mathbf{q}, \mathbf{k}') c_{2\mathbf{k}'+\mathbf{q}}^\dagger c_{1\mathbf{k}'} c_{1\mathbf{k}-\mathbf{q}}^\dagger \right] \right\} |F_N\rangle. \quad (5)$$

To understand the origin of our approximation, let us consider the time evolution picture sketched in Fig. 1. Suppose that initially the system is in its ground state $|F_N\rangle$. After injection of one bare electron, the state of the system is $|C\rangle = c_{2\mathbf{k}}^\dagger|F_N\rangle$. If \mathbf{k} is well inside the Fermi sphere, as we said before, it can not radiatively relax into the first subband. Instead, when $k > k_F$, the

injected electron can radiatively decay, emitting a photon and falling into the first subband. After the first emission the state will have the form $|A, \mathbf{q}\rangle = a_{\mathbf{q}}^\dagger c_{1\mathbf{k}-\mathbf{q}}^\dagger|F_N\rangle$. If the cavity system is closed and only the light-matter interaction is considered, the emitted photon will be reabsorbed. The system can evolve back to the state $|C\rangle$ or into one vector of the form $|B, \mathbf{q}, \mathbf{k}'\rangle = c_{2\mathbf{k}'+\mathbf{q}}^\dagger c_{1\mathbf{k}'} c_{1\mathbf{k}-\mathbf{q}}^\dagger|F_N\rangle$. If

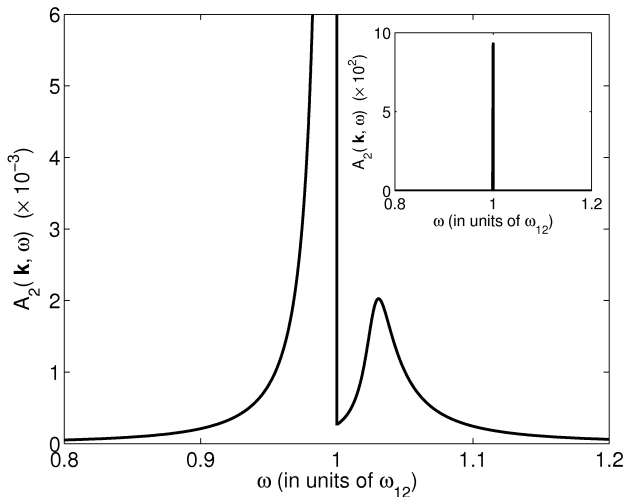


FIG. 2: Electron spectral function $A_2^+(\mathbf{k}, \omega)$ for the second subband, for all $k > k_F$. The spectral function, defined in Eq. (2), is the density of quasi-electron states, weighted by the overlap with the bare electron state. Inset: the same quantity on a larger vertical scale. Coupling parameter: $\Omega_0(q_{res}) = \chi(q_{res})\sqrt{N} = 0.1\omega_{12}$.

\mathbf{k}' is well inside the Fermi sea, when the second subband electron decay, it will go back to state $|A, \mathbf{q}\rangle$. If \mathbf{k}' is on the border of the Fermi sea, on the contrary, it can evolve into a state of the form $|D, \mathbf{q}, \mathbf{q}', \mathbf{k}'\rangle = a_{\mathbf{q}}^\dagger c_{1, \mathbf{k}'+\mathbf{q}-\mathbf{q}'}^\dagger c_{1, \mathbf{k}'}^\dagger c_{1, \mathbf{k}-\mathbf{q}}^\dagger |F_N\rangle$. The probability of ending in

any of the $|D, \mathbf{q}, \mathbf{q}', \mathbf{k}'\rangle$ states is proportional to the ratio $q_{res}/k_F \ll 1$ and is thus negligible. We can thus look for vectors of the form shown in Eq. (5). In the subspace spanned by $\{|C\rangle, |A, \mathbf{q}\rangle, |B, \mathbf{q}, \mathbf{k}'\rangle, |B, \mathbf{q}, \mathbf{k}''\rangle, \dots\}$, we have found[18] that the eigenstates of H with a finite overlap with the bare electron have the form

$$|N+1, \mathbf{k}, 1, \zeta\rangle = \mu_\zeta c_{2, \mathbf{k}}^\dagger |F_N\rangle + \sum_{q, \sigma=\pm} \lambda_{\zeta, \sigma, q} |\sigma, q\rangle, \quad (6)$$

where

$$|\pm, q\rangle = \sum_{|\mathbf{q}|=q} \frac{(\omega_{\pm, q} - \omega_{12})|A, \mathbf{q}\rangle + \chi_q \sum_{\mathbf{k}} |B, \mathbf{q}, \mathbf{k}\rangle}{\sqrt{Lq} \sqrt{(\omega_{\pm, q} - \omega_{12})^2 + |\chi_q|^2 N}} \quad (7)$$

and where $\omega_{\pm, q}$ are the frequencies of the lower and upper polariton branches with in-plane wavevector \mathbf{q} , namely

$$\omega_{\pm, q} = \frac{\omega_{c, q} + \omega_{12}}{2} \pm \sqrt{\left(\frac{\omega_{c, q} - \omega_{12}}{2}\right)^2 + N|\chi_q|^2}. \quad (8)$$

Hence, the spectral function of the second subband reads

$$A_2^+(\mathbf{k}, \omega) = \sum_{\zeta} |\mu_\zeta|^2 \delta(\omega - \omega_\zeta) \theta(k - k_F) + \delta(\omega - \omega_{2, k}) \theta(k_F - k). \quad (9)$$

The coefficients μ_ζ , $\lambda_{\zeta, \sigma, q}$ as well as the eigenfrequencies ω_ζ are obtained by the diagonalization of the matrix representation of H in the subspace $\{c_{2, \mathbf{k}}^\dagger |F_N\rangle, |+, q\rangle, |-, q\rangle, |+, q'\rangle, |-, q'\rangle, \dots\}$, namely:

$$\mathcal{H}_{N+1, \mathbf{k}, 1} = \hbar \begin{pmatrix} \omega_{1, k} + \omega_{12} & J_+^*(q)\sqrt{Lq} & J_-^*(q)\sqrt{Lq} & J_+^*(q')\sqrt{Lq'} & J_-^*(q')\sqrt{Lq'} & \dots \\ J_+(q)\sqrt{Lq} & \omega_{1, k} + \omega_{+, q} & 0 & 0 & 0 & \dots \\ J_-(q)\sqrt{Lq} & 0 & \omega_{1, k} + \omega_{-, q} & 0 & 0 & \dots \\ J_+(q')\sqrt{Lq'} & 0 & 0 & \omega_1(k) + \omega_{+, q'} & 0 & \dots \\ J_-(q')\sqrt{Lq'} & 0 & 0 & 0 & \hbar\omega_1(k) + \omega_{-, q'} & \dots \\ \vdots & \vdots & \vdots & \vdots & \ddots & \ddots \end{pmatrix}, \quad (10)$$

where L^2 is the sample area and

$$J_\pm^*(q) = \frac{\chi_q^*(\omega_{\pm, q} - \omega_{12})}{\sqrt{(\omega_{\pm, q} - \omega_{12})^2 + |\chi_q|^2 N}}. \quad (11)$$

The matrix in Eq. (10) shows that the dressed electronic states are given by a Fano-like coupling of the bare electron state in the second subband with the continuum of cavity polariton modes (associated to all the different photonic in-plane wavevectors \mathbf{q}). In Fig. 2, we show numerical results using a vacuum Rabi frequency $\Omega_{0, q_{res}} = |\chi_{q_{res}}| \sqrt{N} = 0.1\omega_{12}$. As it appears from Eq. (9), the broadening of the spectral function is intrinsic,

being associated to the continuum spectrum of frequencies ω_ζ corresponding to the quasi-electron states. The pronounced dip around $\omega = \omega_{12}$ in the spectral function is a quantum interference feature, typical of a Fano resonance[19].

The states $|N+1, \mathbf{k}, 1, \zeta\rangle$ have been obtained by diagonalizing the Hamiltonian (1), which takes into account only the coupling between the two-subband electronic system and the microcavity photon quantum field. If, as we have assumed, the light-matter interaction is the strongest one, all other residual couplings can be treated perturbatively. These residual interactions in-

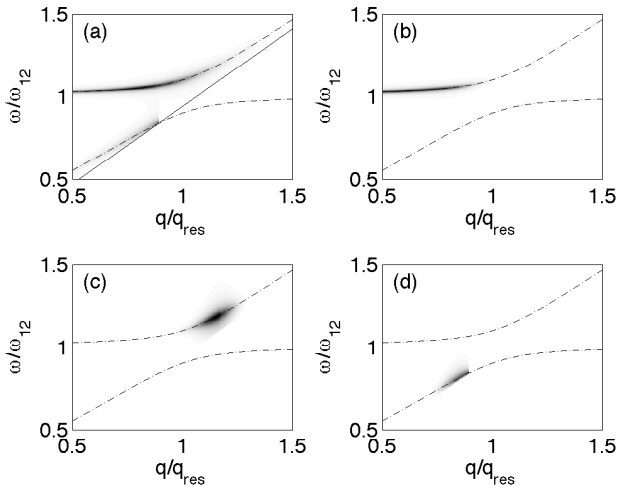


FIG. 3: Extracavity electroluminescence spectra $N_{ph}(\mathbf{q}, \omega)$. Panel (a): the case of a broadband electrical injector (bandwidth equal to ω_{12} , centered at $\omega = \omega_{12}$). The other panels show the results for a narrow-band injector (width $0.05\omega_{12}$) centered respectively at $\omega = \omega_{12}$ (b), $1.2\omega_{12}$ (c) and $0.8\omega_{12}$ (d). The non-radiative relaxation rate $1/\tau_{nr}$ has been taken equal to $0.005\omega_{12}$. In all panels, the dashed-dotted lines are the frequency dispersions $\omega_{\pm, q}$ of the two cavity polariton branches. In the first panel the solid line represents the edge of the light cone [17].

clude the coupling to the extracavity fields, the interaction with contacts, phonon and impurity scattering as well as Coulomb electron-electron interactions[20].

The states $|N + 1, \mathbf{k}, 1, \zeta\rangle$ can be excited by resonant electron tunneling. Treating the tunneling coupling per-

turbatively, we find that the injection rate reads

$$\Gamma_{inj}(\mathbf{k}, \zeta) = \frac{2\pi}{\hbar} |\mu_{\zeta}|^2 |V_{\mathbf{k}}^{tc}|^2 \rho_{inj}(\omega_{\zeta}) n_F(\omega_{\zeta}), \quad (12)$$

where $V_{\mathbf{k}}^{tc}$ is the tunneling coupling matrix element, $\rho_{inj}(\omega)$ is the density of electronic states inside the contact and $n_f(\omega)$ its Fermi distribution. $\rho_{inj}(\omega)n_f(\omega)$ determines the spectral shape of the injector. μ_{ζ} comes from Eq. (9).

The finite transmission of the cavity mirrors is responsible for a finite lifetime for the cavity photons and consequently for the dressed states $|N + 1, \mathbf{k}, 1, \zeta\rangle$. By using the Fermi golden rule and a quasi-mode coupling to the extracavity field, we find that the radiative lifetime $\tau_{r, \mathbf{k}, \zeta}$ reads:

$$\frac{1}{\tau_{r, \mathbf{k}, \zeta}} = \frac{2\pi}{\hbar} \sum_{\mathbf{q}, q_z} |\alpha_{\zeta}(\mathbf{q})|^2 |V_{\mathbf{q}, q_z}^{qm}|^2 \delta(\hbar\omega_{\zeta} - \hbar\omega_{\mathbf{q}, q_z}) \theta(k - k_F),$$

where $V_{\mathbf{q}, q_z}^{qm}$ is the quasi-mode coupling matrix element, $\omega_{\mathbf{q}, q_z}$ the extracavity photon frequency and $\alpha_{\zeta}(\mathbf{q}) = \langle A, \mathbf{q} | N + 1, \mathbf{k}, 1, \zeta \rangle$ as defined in Eq. (5). Having calculated the tunneling injection rate and the radiative lifetime for the different states, we are able to evaluate the electroluminescence spectra. It is convenient to introduce the normalized photon emission distribution corresponding to each eigenstate $|N + 1, \mathbf{k}, 1, \zeta\rangle$, namely

$$L(\mathbf{q}, \zeta) = \mathcal{N} \sum_{q_z} |\alpha_{\zeta}(\mathbf{q})|^2 |V_{\mathbf{q}, q_z}^{qm}|^2 \delta(\hbar\omega_{\zeta} - \hbar\omega_{\mathbf{q}, q_z}^{ph}), \quad (13)$$

where the normalization \mathcal{N} is fixed by imposing $\sum_{\mathbf{q}} L(\mathbf{q}, \zeta) = 1$. The number of photons with in-plane wave-vector \mathbf{q} and frequency ω emitted per unit time is

$$N_{ph}(\mathbf{q}, \omega) = \frac{1}{\pi} \sum_{\mathbf{k}, \zeta} \Gamma_{inj}(\mathbf{k}, \zeta) L(\mathbf{q}, \zeta) \frac{1/\tau_{r, \mathbf{k}, \zeta}}{(\omega - \omega_{\zeta})^2 + (1/\tau_{r, \mathbf{k}, \zeta} + 1/\tau_{nr, \mathbf{k}, \zeta})^2}, \quad (14)$$

where the last factor accounts for the Lorentzian broadening due to radiative and non-radiative processes. $\tau_{nr, \mathbf{k}, \zeta}$ is the non-radiative lifetime of the electronic excitations and $\Gamma_{inj}(\mathbf{k}, \zeta)$ is given by Eq. (12). Fig. 3 reports representative electroluminescence spectra in the case of a broadband (panel a) and narrowband (panel b,c,d) injector. In the broadband case, the emission is resonant at the intersubband cavity polariton frequencies (dashed lines) and it is significant in a wide range of in-plane wavevectors[14]. In contrast, in the case of narrowband electrical injector our theory shows that the photon in-plane momentum and the energy of the cavity polariton emission can be selectively excited by the resonant

electron tunneling process, in agreement with what suggested by recent experiments [13].

In free-space, the quantum efficiency of electroluminescent devices based on intersubband transitions is poor ($\approx 10^{-5}$ in the mid-infrared) due to the slow radiative recombination of long wavelength transitions. In the microcavity case, the efficiency of the emission from an excited state $|N + 1, \mathbf{k}, 1, \zeta\rangle$ is given by $(1 + \tau_{r, \mathbf{k}, \zeta}/\tau_{nr, \mathbf{k}, \zeta})^{-1}$. Being $1/\tau_{nr, \mathbf{k}, \zeta}$ essentially proportional to the matter component of the excitation and $1/\tau_{r, \mathbf{k}, \zeta}$ to its photonic fraction, we have found that it is possible to obtain a quantum efficiency approaching unity by selectively injecting electrons into dressed states with a high photonic frac-

tion. In particular, this is achievable by avoiding injection resonant with the central peak of the electron spectral function in Fig.2, which corresponds to states with strong overlap with the bare electron state.

In conclusion, we have determined in a non-perturbative way the quasi-electron states in a microcavity-embedded two-dimensional electron gas. Such states originate from a Fano-like coupling between the bare electron state and the continuum of cavity polariton excitations. We have proven that these states can be selectively excited by resonant electron tunneling and that the use of narrow-band injector may give rise to ultrahigh efficient polariton electroluminescence.

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