Quantum theory of intersubband polarons

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We present a microscopic quantum theory of intersubband polarons, quasiparticles originated from the coupling between intersubband transitions and longitudinal optical phonons. To this aim, we develop a second quantized theory taking into account both the Fröhlich interaction between phonons and intersubband transitions and the Coulomb interaction between the intersubband transitions themselves. Our results show that the coupling between the phonons and the intersubband transitions is extremely intense, thanks both to the collective nature of the intersubband excitations and to the natural tight confinement of optical phonons. Not only is the coupling strong enough to spectroscopically resolve the resonant splitting between the modes (strong-coupling regime), but it can become comparable to the bare frequency of the excitations (ultrastrong-coupling regime). We thus predict the possibility to exploit intersubband polarons both for applied optoelectronic research, where a precise control of the phonon resonances is needed, and also to observe fundamental quantum vacuum physics, typical of the ultrastrong-coupling regime.

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I. INTRODUCTION

The theory of polarons, the quasiparticles describing electrons in a polarizable medium, dates back to the early days of quantum theory¹ and it has been an active field of research ever since.² In this paper, we will develop a microscopic theory of intersubband polarons, that is, a theory of intersubband transitions coupled to longitudinal optical (LO) phonons in semiconductor quantum wells.

The coupling between intersubband transitions and LO phonons is relevant for a number of optoelectronic applications, as it determines the lifetime of carriers in excited subbands.³ In particular, a precise knowledge of LO phonon intersubband scattering rates is important in the engineering of heterostructures for quantum cascade lasers.⁴ Normally, optoelectronic devices are designed to avoid being in resonance with optical phonon transitions, due to the high absorption between transverse and longitudinal optical (TO and LO) phonon frequencies (Restrahlen band). A notable exception is provided by quantum cascade lasers operating near such optical resonances^{5,6} in which instead the transitions between different subbands are almost resonant with LO phonon modes.

Even if the coupling between intersubband transitions and LO phonons in semiconductor quantum wells has indeed received some attention, and intersubband polaron resonances have been clearly and unambiguously observed, to the best of our knowledge, there is no microscopic theory of such excitations as the spectra of intersubband polarons are normally calculated with indirect methods. While such methods allow us to calculate, at least qualitatively, the polaron dispersions, missing a microscopic description makes it difficult to study more complex phenomena as nonequilibrium physics, quantum vacuum effects, or quantum phase transitions.

Using a second quantization formalism, we reduce the full electron-phonon Hamiltonian to a quadratic, bosonic form, from which we then calculate the polaron dispersions. In order to accomplish this task, we show that Coulomb interaction between electrons in conduction subbands naturally

separates into a dominant and a perturbative part, accordingly to the number of electrons that can participate to each transition.

Moreover, we show how, thanks to the tight confinement of LO phonons, the coupling between intersubband transitions and phonons can easily be in the ultrastrong-coupling regime, a regime characterized by a coupling strength comparable to the bare frequency of the excitations. ¹¹ Such a fact can have interesting observable consequences, as a whole new range of physics is *a priori* observable in this regime: spectral deformations, ^{12–14} quantum vacuum emission phenomena, ^{15–17} electroluminescence enhancement, ^{18,19} and even quantum phase transitions. ^{20–22}

This paper is organized as follows: In Sec. II, we develop the general theory of the coupling between intersubband transitions and LO phonons, which we then apply in Sec. III to the case of GaAs quantum wells, showing how the ultrastrong-coupling regime can be reached even with such relatively weakly polar material. In Sec. IV, we compare the calculated dispersions with those obtained using a homogeneous dielectric function approach. Finally, a few considerations on the impact of our results and on possible future developments are drawn in Sec. V.

II. THEORETICAL FRAMEWORK

A. Superradiant excitations

In 1954, Dicke²³ noticed that a set of coherently excited identical dipoles relaxes radiatively much faster than a single, isolated one. This is due to the phenomenon of superradiance: N identical dipoles behave as a single collective dipole \sqrt{N} times bigger. The concept of superradiance has been thoroughly applied to the study of intersubband polaritons^{24–27} in microcavity-embedded quantum wells. In such systems, the light couples to a collective electronic excitation and, as expected, ¹¹ the strength of the coupling between light and matter is proportional to the square root of the number of electrons involved.

We study the coupling of intersubband transitions with longitudinal optical phonons, considering also the role of Coulomb electron-electron interaction. Such couplings are extremely rich and, in order to limit the complexity of our investigation, we will need to determine which scattering channels are dominant and which are negligible. In general, if N electrons undergo a certain transition in a coherent way, the strength of the coupling is enhanced by a factor \sqrt{N} . Transitions involving a macroscopic number of electrons will thus be strongly enhanced and, for this reason, they will be treated exactly within a Hamiltonian formalism, while the others (involving only few electrons) will be treated perturbatively (or ignored altogether). The degree of collective enhancement of a scattering process will be evaluated looking at the number of electrons that can undergo the transition given fixed amounts of transferred impulsion and energy. In Fig. 1, we show a few illustrative examples of collective and noncollective transitions.

In the case of the Coulomb interaction, as we will see, only the intersubband terms, responsible for the depolarization

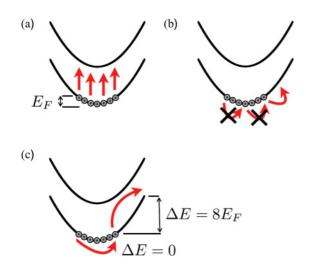


FIG. 1. (Color online) (a) An example of collective transition: an intersubband transition with small transferred momentum. All the electrons can undergo a transition resonant approximately at the same energy. (b) An example of noncollective transition: an intrasubband transition with transferred momentum much smaller than the Fermi wave vector $\hbar k_F$. The majority of electrons, being Pauli blocked, do not participate to the process. (c) Another example of noncollective transition: an intrasubband transition with transferred momentum of the order of $2\hbar k_F$, which is the minimum to allow all the electrons to undergo the transition avoiding Pauli blocking. Anyway, there is a large energy spread between the different single electron transitions. For an infinite potential well, electrons on the two opposite borders of the Fermi sea, with initial momenta $\pm \hbar k_F$ parallel to the transferred momentum, have initially the same energy $\frac{\hbar^2 k_F^2}{2m^*} = E_F$, where m^* is the electron effective mass and E_F the Fermi energy. After the transition, they will end up with final momenta $\hbar k_F$ and $3\hbar k_F$, corresponding to final energies $\frac{\hbar^2 k_F^2}{2m^*} = E_F$ and $\frac{9\hbar^2 k_F^2}{2m^*} = 9E_F$, respectively. This implies that, even if the transition is not blocked, only a small fraction of the single electron transitions can be resonant at the same time. In both cases, given that only few electrons can participate to the collective transition, the superradiant enhancement factor will be small.

shift, are collective. We will thus treat them exactly in the Hamiltonian, while considering all the other terms in a random phase approximation (RPA) linear response approach.

B. Free fields

We consider a symmetric quantum well of length $L_{\rm QW}$ in a bulk of height $L_{\rm BK}$, and S will be the surface of the sample. For the moment, we limit ourselves to the case of a single quantum well; the general case of multiple wells will be addressed later in this section. The quantum well is supposed to be doped in such a way that its Fermi level is between the first and the second conduction subbands, separated between them by the intersubband gap energy $\hbar\omega_{12}$.

We develop our theory using a zero-temperature formalism (T=0); anyway, our results remain quantitatively accurate while the thermal population of the second subband remains negligible. Depending on material parameters and doping level, this could imply the necessity to perform experiments in different kinds of cryogenic environments.

Electron states will be indexed by the subband index j and by the value of the in-plane wave vector \mathbf{k} . Their wave functions will be given by

$$\psi_{j,\mathbf{k}}(\rho,z) = \chi_j(z) \frac{e^{i\mathbf{k}\rho}}{\sqrt{S}}, \quad j = 1,2$$
 (1)

where, for simplicity, we choose $\chi_j(z)$ to be real and, due to the symmetry of the quantum well, the $\chi_j(z)$ have well-defined and opposite symmetry. Wave functions in Eq. (1) are chosen as the basis for second quantization, and the creation operator for an electron in the state described by Eq. (1) will be denoted as $c_{j,k}^{\dagger}$. The free Hamiltonian of the electron gas in the two considered subbands thus reads as

$$H_{el} = \sum_{j=\{1,2\},\mathbf{k}} \hbar \omega_j(\mathbf{k}) c_{j,\mathbf{k}}^{\dagger} c_{j,\mathbf{k}}, \tag{2}$$

where $\omega_1(\mathbf{k}) = \frac{\hbar^2 k^2}{2m^*}$ and $\omega_2(\mathbf{k}) = \omega_1(\mathbf{k}) + \omega_{12}$. In Eq. (2), as well as in the rest of this paper, we omit the electron spin index. This is justified by the fact that all interactions we consider are spin conserving. Given that we consider only in-plane wavevector exchanges \mathbf{q} much smaller than the typical electron wave vector \mathbf{k} , we can make the approximation $\omega_j(\mathbf{k} + \mathbf{q}) \simeq \omega_j(\mathbf{k})$, and introduce the operators describing intersubband transitions with a well-defined and dispersionless energy $\hbar\omega_{12}$ (Ref. 11):

$$b_{\mathbf{q}}^{\dagger} = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} c_{2,\mathbf{k}+\mathbf{q}}^{\dagger} c_{1,\mathbf{k}}, \quad b_{\mathbf{q}} = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} c_{1,\mathbf{k}}^{\dagger} c_{2,\mathbf{k}+\mathbf{q}}, \quad (3)$$

where N is the number of electrons in the quantum well. In the dilute regime, that is, if the number of excitations is much smaller than N, the $b_{\mathbf{q}}^{\dagger}$ operators are bosonic:

$$[b_{\mathbf{q}}, b_{\mathbf{q}'}^{\dagger}] \simeq \delta_{\mathbf{q}, \mathbf{q}'}.$$
 (4)

At higher excitation densities, which are out of the scope of this work, saturation effects start to appear and corrections to Eq. (4) have to be taken into account. For a detailed analysis of nonbosonicity effects in intersubband transitions, we invite the interested readers to refer to Ref. 28.

Using Eq. (3), we can rewrite the Hamiltonian of the free electron gas in Eq. (2) in terms of bosonic intersubband excitations

$$H_{el} = \sum_{\mathbf{q}} \hbar \omega_{12} b_{\mathbf{q}}^{\dagger} b_{\mathbf{q}}. \tag{5}$$

In this work, we are interested in the resonant case in which ω_{12} is equal, or close, to the LO phonon frequency ω_{LO} . We can thus neglect confinement effects on the phonons and consider bulk values for their frequencies. ^{29,30} We will thus describe LO phonons by means of the three-dimensional boson operators $d_{\mathbf{q},q_z}$:

$$[d_{\mathbf{q},q_z},d_{\mathbf{q}',q_z'}^{\dagger}] = \delta_{\mathbf{q},\mathbf{q}'}\delta_{q_z,q_z'},\tag{6}$$

indexed by their in-plane and out-of-plane wave vectors. While we know that LO phonon modes are confined inside the quantum well, we do not need to impose this constraint in the mode definition because, as we will see, intersubband transitions end up coupling with linear superpositions of phonon modes that are anyway confined inside the quantum well. We consider only the case of one single longitudinal optical branch, with the expansion to the case of multiple branches not presenting any fundamental difficulty.

Moreover, we are interested only in phonons with small in-plane wave vectors (in order to couple with coherent intersubband excitations), we can thus ignore phonon dispersion and write the free phonon Hamiltonian as

$$H_{ph} = \sum_{\mathbf{q},q_z} \hbar \omega_{\text{LO}} d_{\mathbf{q},q_z}^{\dagger} d_{\mathbf{q},q_z}. \tag{7}$$

C. Electron-phonon interaction

Interaction between electrons and LO phonons can be described using the Fröhlich Hamiltonian³¹

$$H_{\rm Fr} = \sqrt{\frac{\hbar \omega_{\rm LO} e^2}{2\epsilon_0 \epsilon_\rho S L_{\rm BK}}} \sum_{\mathbf{q}, q_z} \frac{e^{-i(\mathbf{q}\rho + q_z z)}}{\sqrt{q^2 + q_z^2}} d_{\mathbf{q}, q_z}^{\dagger} + \text{H.c.}, \quad (8)$$

where

$$\frac{1}{\epsilon_0} = \frac{1}{\epsilon_\infty} - \frac{1}{\epsilon_s},\tag{9}$$

where ϵ_s and ϵ_∞ are, respectively, the static and high-frequency dielectric constants.³⁰

The Hamiltonian in Eq. (8) can be written in second quantization (neglecting incoherent intrasubband scattering³) as

$$H_{\mathrm{Fr}} = \sqrt{\frac{\hbar \omega_{\mathrm{LO}} e^2}{2\epsilon_0 \epsilon_\rho S L_{\mathrm{BK}}}} \sum_{\mathbf{q}, q_z} \frac{F(q_z)}{\sqrt{q^2 + q_z^2}} \times (d_{\mathbf{q}, q_z}^{\dagger} + d_{-\mathbf{q}, -q_z}) (c_{1,\mathbf{k}}^{\dagger} c_{2,\mathbf{k}+\mathbf{q}} + c_{2,\mathbf{k}-\mathbf{q}}^{\dagger} c_{1,\mathbf{k}}), \quad (10)$$

where we have defined

$$F(q) = \int dz \,\chi_1(z)\chi_2(z)e^{-iqz}. \tag{11}$$

From Eq. (10), we see that, due to the three-dimensional character of the LO phonons,³⁰ each electronic transition couples to multiple phonon modes, indexed by different

values of the wave vector along the growth direction. It is thus convenient to introduce second quantized operators corresponding to the particular linear superpositions of phonon modes that are coupled to electronic transitions

$$r_{\mathbf{q}}^{\dagger} = \frac{1}{\sqrt{A}} \sum_{q_z} \frac{F(q_z) d_{\mathbf{q}, q_z}^{\dagger}}{\sqrt{q^2 + q_z^2}}, \quad r_{\mathbf{q}} = \frac{1}{\sqrt{A}} \sum_{q_z} \frac{\bar{F}(q_z) d_{\mathbf{q}, q_z}}{\sqrt{q^2 + q_z^2}}, \quad (12)$$

the spatial wave functions along the z axis of which are

$$\varphi_q(z) = \frac{1}{\sqrt{AL_{\text{BK}}}} \sum_{q_z} \frac{F(q_z)e^{iq_z z}}{\sqrt{q^2 + q_z^2}}.$$
(13)

From Eqs. (13) and (11), we see that the intersubband transitions naturally couple to phonon modes localized inside the quantum well [it is easy to verify that $\varphi_q(z)$ vanishes to the first order in q if z is outside the common support of χ_1 and χ_2].

The normalization factor A can be fixed by imposing bosonic commutation relations for the $r_{\mathbf{q}}^{\dagger}$ operators

$$[r_{\mathbf{q}}, r_{\mathbf{q}'}^{\dagger}] = \frac{1}{A} \sum_{q_z, q_z'} \frac{F(q_z)F(-q_z')}{\sqrt{(q^2 + q_z^2)(q'^2 + {q_z'}^2)}} [d_{\mathbf{q}, q_z}, d_{\mathbf{q}', q_z'}^{\dagger}]$$

$$= \frac{\delta_{\mathbf{q}, \mathbf{q}'} L_{\text{BK}} I(q)}{2Aq}, \tag{14}$$

and thus

$$A = \frac{L_{\rm BK}}{2} \frac{I(q)}{q},\tag{15}$$

where we have defined

$$I(q) = \int dz \, dz' \chi_1(z) \chi_2(z) \chi_2(z') \chi_1(z') e^{-q|z-z'|}.$$
 (16)

We can thus write Hamiltonians in Eqs. (7) and (10) in terms of coherent $r_{\bf q}^{\dagger}$ and $b_{\bf q}^{\dagger}$ operators as

$$H_{ph} = \sum_{\mathbf{q}} \hbar \omega_{\text{LO}} r_{\mathbf{q}}^{\dagger} r_{\mathbf{q}} \tag{17}$$

and

$$H_{\mathrm{Fr}} = \sum_{\mathbf{q}} \sqrt{N_{\mathrm{2DEG}} \hbar \omega_{\mathrm{LO}} \frac{e^2}{4\epsilon_0 \epsilon_{
ho}} \frac{I(q)}{q} (b_{\mathbf{q}}^{\dagger} + b_{-\mathbf{q}}) (r_{\mathbf{q}} + r_{-\mathbf{q}}^{\dagger})},$$

where

$$N_{\rm 2DEG} = \frac{N}{S} \tag{18}$$

is the density of the two-dimensional electron gas (2DEG). In order to pass from Eq. (7) to (17), we are neglecting higher-order phonon modes confined inside the quantum well. This is justified by the fact that we limit ourselves to long-wavelength modes.

D. Superradiant electron-electron interaction

In order to treat the Coulomb electron-electron interaction, we start by the second quantized form of the Hamiltonian

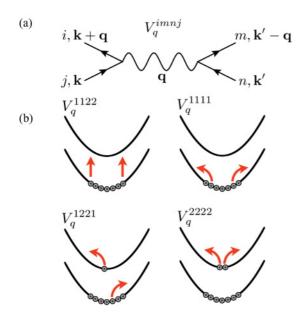


FIG. 2. (Color online) (a) Index convention of the matrix element V_q^{imnj} . Two electrons in subbands j and n, with momenta \mathbf{k} and \mathbf{k}' , are scattered into subbands i and m, with momenta $\mathbf{k} + \mathbf{q}$ and $\mathbf{k}' - \mathbf{q}$, respectively. (b) Graphical representation of the four qualitatively different kinds of scattering processes from Eq. (22).

describing the Coulomb interaction³² [see Fig. 2(a) for a graphical representation of the interaction coefficients]

$$H_c = \frac{1}{2} \sum_{i,j,m,n=1,2} \sum_{\mathbf{q},\mathbf{k},\mathbf{k}'} V_q^{imnj} c_{i,\mathbf{k}+\mathbf{q}}^{\dagger} c_{m,\mathbf{k}'-\mathbf{q}}^{\dagger} c_{n,\mathbf{k}'} c_{j,\mathbf{k}}, \qquad (19)$$

where

$$V_q^{imnj} = \frac{e^2}{2\epsilon_0 \epsilon_\infty q} \int dz \, dz' \chi_i(z) \chi_j(z) \chi_m(z') \chi_n(z') e^{-q|z-z'|},$$
(20)

is the two-dimensional Coulomb matrix element. It is important to notice that in Eq. (20) we used the high-frequency dielectric constant ϵ_{∞} instead of the static one. This is due to the fact that ϵ_s includes the effect of the coupling to LO phonons, which are already treated exactly in the Hamiltonian.

Due to the symmetry of the wave functions, a certain number of matrix elements in Eq. (20) can be seen to be zero, in particular, all the matrix elements with an odd number of 1 and 2 indices:

$$\begin{aligned} V_q^{1112} &= V_q^{1121} = V_q^{1211} = V_q^{2111} = 0, \\ V_q^{2111} &= V_q^{2212} = V_q^{2122} = V_q^{1222} = 0. \end{aligned} \tag{21}$$

The other elements can be evaluated as

$$V_{q}^{1122} = V_{q}^{1212} = V_{q}^{2121} = V_{q}^{2211} = \frac{e^{2}I(q)}{2\epsilon_{0}\epsilon_{\infty}q},$$

$$V_{q}^{1221} = V_{q}^{2112} = \frac{e^{2}}{2\epsilon_{0}\epsilon_{\infty}q} \int dz \, dz' \chi_{1}^{2}(z) \chi_{2}^{2}(z') e^{-q|z-z'|},$$

$$V_{q}^{1111} = \frac{e^{2}}{2\epsilon_{0}\epsilon_{\infty}q} \int dz \, dz' \chi_{1}^{2}(z) \chi_{1}^{2}(z') e^{-q|z-z'|},$$

$$V_{q}^{2222} = \frac{e^{2}}{2\epsilon_{0}\epsilon_{\infty}q} \int dz \, dz' \chi_{2}^{2}(z) \chi_{2}^{2}(z') e^{-q|z-z'|},$$

$$(22)$$

where I(q), defined in Eq. (16), is the same integral we encountered studying the electron-phonon Fröhlich interaction.

The four distinct nonzero possible values of the matrix elements correspond to different kinds of scattering processes. In Fig. 2(b), a graphical representation for each of these processes is shown. It is important at this point to notice a major difference between the elements in the first line of Eq. (22) and the others. The elements in the first line [upper left subpanel in Fig. 2(b)] represent intersubband excitations: each electron is scattered from one subband to the other. Such processes, responsible for the depolarization shift,³² describe a superradiant process in the sense defined above, that is, at least for small values of q, a great number of electrons can coherently undergo the same transition, approximately at the same energy. This is not the case for the interactions described in the other lines of Eq. (22), which instead describe intrasubband excitations that, either due to Pauli blocking or to the nonflat energy dispersions, involve only few electrons (see Fig. 1 for a graphical visualization of this crucial point).

Our previous discussion on superradiant processes thus implies that the terms in the first line of Eq. (22) strongly dominate over the others due to their superradiant enhancement. For this reason, we have to treat them exactly in a Hamiltonian formalism, while we can limit ourselves to treat the others within a perturbative approach.

Here, we will thus construct an exact, Hamiltonian approach to treat the effect of the depolarization shift terms, neglecting the others. We will analyze later the effect of the intrasubband terms. Let us start to rewrite the depolarization shift part of Eq. (19) in a more useful form

$$H_{c} = \sum_{\mathbf{q},\mathbf{k},\mathbf{k}'} \frac{e^{2}I(q)}{4\epsilon_{0}\epsilon_{\infty}q} (c_{1,\mathbf{k}+\mathbf{q}}^{\dagger}c_{1,\mathbf{k}'-\mathbf{q}}^{\dagger}c_{2,\mathbf{k}'}c_{2,\mathbf{k}} + c_{1,\mathbf{k}+\mathbf{q}}^{\dagger}c_{2,\mathbf{k}'-\mathbf{q}}^{\dagger}c_{1,\mathbf{k}'}c_{2,\mathbf{k}} + c_{2,\mathbf{k}+\mathbf{q}}^{\dagger}c_{1,\mathbf{k}'-\mathbf{q}}^{\dagger}c_{2,\mathbf{k}'}c_{1,\mathbf{k}} + c_{2,\mathbf{k}+\mathbf{q}}^{\dagger}c_{2,\mathbf{k}'}c_{1,\mathbf{k}})$$

$$= \sum_{\mathbf{q},\mathbf{k},\mathbf{k}'} \frac{e^{2}I(q)}{4\epsilon_{0}\epsilon_{\infty}q} (c_{1,\mathbf{k}+\mathbf{q}}^{\dagger}c_{2,\mathbf{k}}c_{1,\mathbf{k}'-\mathbf{q}}^{\dagger}c_{2,\mathbf{k}'} + c_{1,\mathbf{k}+\mathbf{q}}^{\dagger}c_{2,\mathbf{k}}c_{2,\mathbf{k}'-\mathbf{q}}^{\dagger}c_{1,\mathbf{k}'} + c_{2,\mathbf{k}+\mathbf{q}}^{\dagger}c_{1,\mathbf{k}}c_{1,\mathbf{k}'-\mathbf{q}}^{\dagger}c_{2,\mathbf{k}'} + c_{2,\mathbf{k}+\mathbf{q}}^{\dagger}c_{1,\mathbf{k}}c_{2,\mathbf{k}'-\mathbf{q}}^{\dagger}c_{1,\mathbf{k}'})$$

$$+ \sum_{\mathbf{q}} \frac{Ne^{2}I(q)}{4\epsilon_{0}\epsilon_{\infty}q}. \tag{23}$$

We see from Eq. (23) that, thanks to its collective, superradiant nature, the depolarization shift can be naturally written in terms of the bosonic intersubband excitations defined in Eq. (3) as

$$H_c = \sum_{\mathbf{q}} N_{2\text{DEG}} \frac{e^2}{4\epsilon_0 \epsilon_\infty} \frac{I(q)}{q} (b_{\mathbf{q}}^{\dagger} + b_{-\mathbf{q}}) (b_{-\mathbf{q}}^{\dagger} + b_{\mathbf{q}}), \quad (24)$$

where we have neglected the last constant term, which simply shifts the ground-state energy.

E. Residual electron-electron interaction

We treated exactly the depolarization shift terms of the Coulomb interaction in a bosonic excitation formalism. Moreover, we showed how the terms other than those responsible for the depolarization shift are strongly suppressed due to their lack of collective enhancement and can thus be treated perturbatively.

Here, we study the perturbative effect of such residual Coulomb contributions due to the intrasubband terms in the last three lines of Eq. (22) [schematized in the last three panels of Fig. 2(b)]. An important result due to Lee and Galbraith^{33,34} is that such intrasubband terms do not contribute to the screening of the intersubband ones at the level of the random phase approximation. This can be seen writing the Dyson equation for the dynamically screened Coulomb potential³⁵ $\mathcal{V}_q(\omega)$ as

$$V_q^{imnj}(\omega) = V_q^{imnj} + \sum_{rs} V_q^{irsj} \Pi_q^{sr}(\omega) V_q^{smnr}(\omega), \quad (25)$$

where $\Pi_q^{sr}(\omega)$ is the RPA polarization function. In the case of an intersubband contribution (e.g., V_q^{1122}), Eqs. (21) and (22) imply that

$$\begin{aligned} \mathcal{V}_{q}^{1122}(\omega) &= V_{q}^{1122} + \sum_{rs} V_{q}^{1rs2} \Pi_{q}^{sr}(\omega) \mathcal{V}_{q}^{s12r}(\omega) \\ &= V_{q}^{1122} + \sum_{r \neq s} V_{q}^{1rs2} \Pi_{q}^{sr}(\omega) \mathcal{V}_{q}^{s12r}(\omega) \\ &= V_{q}^{1122} + V_{q}^{1122} \left[\Pi_{q}^{12}(\omega) + \Pi_{q}^{21}(\omega) \right] \mathcal{V}_{q}^{1122}(\omega). \end{aligned} (26)$$

We have thus

$$\mathcal{V}_q^{1122}(\omega) = \frac{V_q^{1122}}{1 - V_q^{1122} \left[\Pi_q^{12}(\omega) + \Pi_q^{21}(\omega) \right]},\tag{27}$$

from which we see that the intrasubband Coulomb terms $(V_q^{1111},\ V_q^{2222},\ V_q^{1221},\ \text{and}\ V_q^{2112})$ do not intervene in the renormalization of the intersubband terms.

An analogous reasoning can be done for the phononelectron interaction. Calling M_{q,q_z} and $\mathcal{M}_{q,q_z}(\omega)$ the bare and screened versions of the potential defined in Eq. (10), we have the Dyson equation

$$\mathcal{M}_{q,q_{z}}(\omega) = M_{q,q_{z}} + \sum_{rs} V_{q}^{1mn2} \Pi_{q}^{sr}(\omega) \mathcal{M}_{q,q_{z}}(\omega)$$

$$= M_{q,q_{z}} + V_{q}^{1122} \left[\Pi_{q}^{12}(\omega) + \Pi_{q}^{21}(\omega) \right] \mathcal{M}_{q,q_{z}}(\omega)$$
(28)

and thus the formula for the screened potential is

$$\mathcal{M}_{q,q_z}(\omega) = \frac{M_{q,q_z}}{1 - V_q^{1122} \left[\Pi_q^{12}(\omega) + \Pi_q^{21}(\omega) \right]}.$$
 (29)

Being that the RPA screening is only due to terms that are exactly treated in the Hamiltonian, we can thus neglect the screening due to the two-dimensional electron gas.

F. Hopfield-Bogoliubov Hamiltonian

Putting together Eqs. (5), (18), and (24), we arrive to the full Hamiltonian for the intersubband transitions–LO phonons system

$$H = \sum_{\mathbf{q}} \hbar \omega_{12} b_{\mathbf{q}}^{\dagger} b_{\mathbf{q}} + \hbar \omega_{\text{LO}} r_{\mathbf{q}}^{\dagger} r_{\mathbf{q}}$$

$$+ \sqrt{N_{\text{2DEG}} \hbar \omega_{\text{LO}} \frac{e^{2}}{4\epsilon_{0}\epsilon_{\rho}} \frac{I(q)}{q}} (b_{\mathbf{q}}^{\dagger} + b_{-\mathbf{q}}) (r_{-\mathbf{q}}^{\dagger} + r_{\mathbf{q}})$$

$$+ N_{\text{2DEG}} \frac{e^{2}}{4\epsilon_{0}\epsilon_{\infty}} \frac{I(q)}{q} (b_{\mathbf{q}}^{\dagger} + b_{-\mathbf{q}}) (b_{-\mathbf{q}}^{\dagger} + b_{\mathbf{q}}). \tag{30}$$

The Hamiltonian in Eq. (30) can be rewritten in a more compact form by introducing the intersubband transitions—LO phonons coupling coefficient Ω and the Coulomb coefficient D:

$$\Omega = \sqrt{N_{2\text{DEG}}\omega_{\text{LO}}\frac{e^2}{4\epsilon_0\epsilon_\rho\hbar}\frac{I(q)}{q}}, \quad D = N_{2\text{DEG}}\frac{e^2}{4\epsilon_0\epsilon_\infty}\frac{I(q)}{q},$$
(31)

where we have dropped the dependences over the wave vector as we are interested in the long-wavelength limit [from Eq. (16), we can verify that $\lim_{q\to 0} \frac{I(q)}{q}$ tends to a constant valuel.

By using Eq. (31), Eq. (30) can be written as

$$H = \hbar \sum_{\mathbf{q}} \omega_{12} b_{\mathbf{q}}^{\dagger} b_{\mathbf{q}} + \omega_{\text{LO}} r_{\mathbf{q}}^{\dagger} r_{\mathbf{q}} + \Omega (b_{\mathbf{q}}^{\dagger} + b_{-\mathbf{q}}) (r_{-\mathbf{q}}^{\dagger} + r_{\mathbf{q}})$$
$$+ D(b_{\mathbf{q}}^{\dagger} + b_{-\mathbf{q}}) (b_{-\mathbf{q}}^{\dagger} + b_{\mathbf{q}}), \tag{32}$$

which can be cast in matrix form as

$$H = \frac{\hbar}{2} \sum_{\mathbf{q}} \hat{v}_{\mathbf{q}}^{\dagger} \, \eta \, \mathcal{H}_q \, \hat{v}_{\mathbf{q}}, \tag{33}$$

where the column vector of operators $\hat{v}_{\mathbf{q}}$ is defined as

$$\hat{v}_{\mathbf{q}} = [b_{\mathbf{q}}, r_{\mathbf{q}}, b_{-\mathbf{q}}^{\dagger}, r_{-\mathbf{q}}^{\dagger}]^{T}, \tag{34}$$

 η is the diagonal metric

$$\eta = \text{diag}[1, 1, -1, -1], \tag{35}$$

and the Hopfield-Bogoliubov³⁶ matrix \mathcal{H}_q is defined as

$$\mathcal{H}_{q} = \begin{pmatrix} \omega_{12} + 2D & \Omega & 2D & \Omega \\ \Omega & \omega_{LO} & \Omega & 0 \\ -2D & -\Omega & -\omega_{12} - 2D & -\Omega \\ -\Omega & 0 & -\Omega & -\omega_{LO} \end{pmatrix}.$$
(36)

Diagonalizing the matrix in Eq. (36) will yield the frequencies of the normal modes of the system ω_{\pm} , which are usually called polarons. ^{37,38} In our case, we name them more properly intersubband polarons because the electronic part of the mixed

excitations is an intersubband transition. The Hamiltonian in Eq. (32) can thus be put in the diagonal form

$$H = \sum_{j=\pm,\mathbf{q}} \hbar \omega_j \, p_{j,\mathbf{q}}^{\dagger} p_{j,\mathbf{q}} + E_{\Delta}, \tag{37}$$

where the $p_{j,\mathbf{q}}$ are the annihilation operators for the two polaronic branches, given by a linear superposition of $b_{\mathbf{q}}$, $r_{\mathbf{q}}$, $b_{-\mathbf{q}}^{\dagger}$, and $r_{-\mathbf{q}}^{\dagger}$ operators, and E_{Δ} is the energy of the new ground state relative to that of the uncoupled system.

G. Coupled ground state

The coupling between the intersubband transitions and the LO phonons does not modify only the system's resonances but it also qualitatively modifies the nature of its ground state. It is easy to verify that, if $|0\rangle$ is the ground state for the uncoupled phonons and intersubband excitations, defined in the usual way as

$$b_{\mathbf{q}}|0\rangle = r_{\mathbf{q}}|0\rangle = 0,$$

then

$$p_{i,\mathbf{q}}|0\rangle \neq 0$$
,

that is, $|0\rangle$ is not the ground state for the coupled system. The real ground state of the Hopfield matrix in Eq. (36), which has been thoroughly studied in Ref. 11, has the form of a two-mode squeezed vacuum.

Still, thanks to the bosonicity of the system, such a ground state does not influence the response of the system, which can be described as a gas of free bosonic excitations [from Eq. (37)]. A notable exception is the case in which the parameters of the system are nonadiabatically modulated in time. In this case, the sudden change in the ground state³⁹ can have observable effects, such as the emission of quantum vacuum radiation.^{15,16}

It is also interesting to notice that, from Eq. (31), we can write the Coulomb coefficient D as

$$D = \frac{\Omega^2}{\omega_{\text{LO}}} \frac{\epsilon_{\rho}}{\epsilon_{\infty}} \geqslant \frac{\Omega^2}{\omega_{\text{LO}}}.$$
 (38)

As it has recently been shown in Ref. 40, Eq. (38) implies that the ground state of the system will not undergo a Dicke phase transition, regardless of the strength of the coupling.

H. Multiple quantum wells

Until now, we considered the case of a single quantum well. This choice has been motivated by the fact that, as we will show, the presence of multiple wells does not modify our results. Given that we are considering rather large quantum wells (in order for the transition to be resonant with the LO phonon mode), the optical phonon spectrum is not modified and the optical phonon modes we consider are confined in each quantum well.

This is a rather important difference between the intersubband polaron case we consider in this paper and the physics of intersubband polaritons. For intersubband polaritons, the electromagnetic mode coupled to the intersubband transitions extends over all the structure. It thus couples to all the electrons, regardless of the quantum well they are in. This means that the only meaningful parameter for intersubband polaritons is the total density of electrons, and the light-matter coupling thus scales as $\sqrt{n_{\rm QW}N_{\rm 2DEG}}$, where $n_{\rm QW}$ is the number of quantum wells inside the microcavity.

In the present case instead, being the phonon modes confined inside each quantum well, electrons in different wells are completely decoupled. This can also be inferred from the coupling integral in Eq. (20). This integral does vanish, at least in the long-wavelength limit (first order in q), if the wave functions for the two integration variables z and z' do not have a common support, i.e., if the two interacting electrons are in different quantum wells. This means that, contrary to the intersubband polariton case, the intersubband polaron interaction scales only as $\sqrt{N_{\rm 2DEG}}$, and growing multiple quantum wells in the same sample will not increase the coupling.

III. RESULTS

In order to obtain some numerical predictions from the Hamiltonian in Eq. (32), we need to fix a few parameters concerning the material and the quantum well. For the sake of simplicity, we consider the quantum well to be correctly approximated by a rectangular, infinite potential well of length $L_{\rm QW}$. We thus have

$$\hbar\omega_{12} = \frac{3\hbar^2\pi^2}{2m^*L_{\text{OW}}^2},\tag{39}$$

and the electronic and phononic modes profiles are given by

$$\chi_1(z) = \sqrt{\frac{2}{L_{\text{QW}}}} \sin\left(\frac{\pi z}{L_{\text{QW}}}\right),$$

$$\chi_2(z) = \sqrt{\frac{2}{L_{\text{QW}}}} \sin\left(\frac{2\pi z}{L_{\text{QW}}}\right),$$

$$\varphi_0(z) = \sqrt{\frac{16}{5L_{\text{QW}}}} \sin^3\left(\frac{\pi z}{L_{\text{QW}}}\right)$$
(40)

inside the well and zero outside. As explained in Sec. II, we see here explicitly that the intersubband transitions couple to a linear superposition of phonon modes that is localized inside the quantum well [the cubic sinus in the third line of Eq. (40) comes from the integral of the first two, as can be verified performing the integral in Eq. (13)]. By inserting Eq. (40) into Eq. (16) and performing the integral, we have

$$\lim_{q \to 0} I(q) \to \frac{10}{9\pi^2} q L_{\text{QW}}.$$
 (41)

In Fig. 3, we plot the normalized coupling $\frac{\Omega}{\omega_{\text{LO}}}$ as a function of the density of the two-dimensional electron gas for a GaAs quantum well. In the inset of Fig. 3, we instead present a comparison of the values of $\frac{\Omega}{\omega_{\text{LO}}}$, at room temperature, for different semiconductors of the III-V and II-VI groups, ⁴¹ as a function of the respective LO phonon energies, for a reference doping $N_{\text{2DEG}} = 10^{12} \text{ cm}^{-2}$.

In Fig. 4, there is a plot of the intersubband polaron frequencies ω_{\pm} as a function of the intersubband frequency ω_{12} , in GaAs, for $N_{\rm 2DEG} = 10^{12} \, {\rm cm}^{-2}$. Notice that, due to the effect of Coulomb interaction, the resonant anticrossing is not

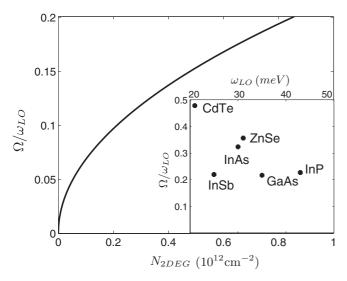


FIG. 3. Normalized coupling $\frac{\Omega}{\omega_{\rm LO}}$ in GaAs as a function of the doping density $N_{\rm 2DEG}$. Inset: the same quantity as a function of the LO phonon frequency $\omega_{\rm LO}$ for different materials, for $N_{\rm 2DEG} = 10^{12}~{\rm cm}^{-2}$.

at $\omega_{12}=\omega_{LO}$, but at a lower frequency. In the inset of the same figure we plot the same quantity as a function of the electron density. The length $L_{\rm QW}$ has been chosen in this case to have the two uncoupled modes at resonance ($\omega_{12}=\omega_{\rm LO}$, that is $L_{\rm OW}\simeq 23$ nm).

It is clear from the figures that intersubband polarons are not only strongly coupled, having coupling constants much larger than their linewidth (usual linewidths being not bigger than a few meV), but they are indeed in the ultrastrong-coupling regime, with values of the normalized coupling $\frac{\Omega}{\omega_{\text{LO}}}$ comparable or larger than the best ones reported in the literature. For physically realizable levels of doping, coupling values of a few tenths of the bare frequency of the excitation ω_{LO} are predicted in GaAs, and it seems that values much larger can be obtained using more polar materials. The reason

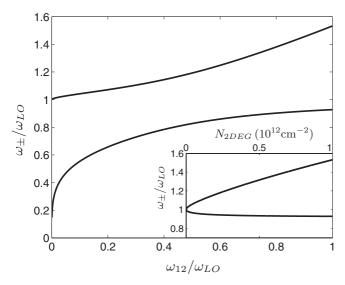


FIG. 4. Intersubband polaron frequencies ω_{\pm} as a function of the intersubband frequency ω_{12} in GaAs for $N_{\rm 2DEG}=10^{12}~{\rm cm}^{-2}$. Inset: the same quantity as a function of the doping for $\omega_{12}=\omega_{\rm LO}$.

of such large coupling can be found in the superradiant nature of intersubband excitations and in the natural confinement of the phonons inside the quantum well, which gives an extremely small mode volume when compared with what can be obtained with photonic microcavities.

The consequences of our results can be multiple, both for fundamental and applied research. On the fundamental side, intersubband polarons could become a new laboratory to test quantum vacuum physics, typical of the ultrastrong-coupling regime. On the applied side, our theory can be naturally exploited in the study of quantum cascade lasers working in or near the Restrahlen band. It can, for example, help in explaining the anticrossing observed in Ref. 5, near the LO phonon frequency. Moreover, the capability to strongly modify the LO phonon spectrum could have an impact on the performances of optoelectronic devices, as the electron–LO phonon scattering rate determines the lifetime of carriers in excited subbands.

IV. COMPARISON WITH DIELECTRIC FUNCTION THEORY

In the previous sections, we have developed a detailed microscopic theory for the intersubband transitions coupled to LO phonons. Here, we will compare the dispersions obtained from the microscopic theory with those obtained with an homogeneous dielectric function theory, as the one used in Ref. 10.

The propagation of an electromagnetic wave in a dispersive, homogeneous medium obeys the equation

$$\operatorname{div}[D(\omega)] = \operatorname{div}[\epsilon(\omega)E(\omega)] = 0. \tag{42}$$

This implies that it is possible to have propagating longitudinal waves, such as polarons, only at frequencies for which

$$Re[\epsilon(\omega)] = 0, \tag{43}$$

where Re indicates the real part.

The z component of the dielectric function of a homogeneous medium filled with quantum wells is given by 10,13

$$\epsilon(\omega) = \epsilon_{\infty} \frac{\omega^2 - \omega_{\text{LO}}^2}{\omega^2 - \omega_{\text{TO}}^2 + i\omega 0^+} - \epsilon_{\infty} \frac{\omega_P^2}{\omega^2 - \omega_{12}^2 + i\omega 0^+}, \quad (44)$$

where

$$\omega_P^2 = \frac{2\omega_{12}d_{12}^2 N_{\text{2DEG}}}{\hbar\epsilon_0 \epsilon_\infty L_{\text{QW}}} \tag{45}$$

is the plasma frequency of the two-dimensional electron gas and d_{12} is the intersubband dipole

$$d_{12} = e \int dz \,\chi_1(z) z \chi_2(z). \tag{46}$$

The equation

$$Re[\epsilon(\omega)] = 0 \tag{47}$$

thus reads as

$$\omega^4 - \omega^2 \left(\omega_{\rm LO}^2 + \omega_{12}^2 + \omega_P^2 \right) + \omega_{\rm LO}^2 \omega_{12}^2 + \omega_{\rm TO}^2 \omega_P^2 = 0. \eqno(48)$$

As Eq. (44) neglects both the dielectric response in the x-y plane and the nonhomogeneity in the z direction, in order to

recover the same result from our microscopic approach, we have to consider only phonon modes with $\mathbf{q} = 0$ and $q_z \to 0$. From Eqs. (11) and (46), we thus have

$$\frac{F(q_z)}{\sqrt{q^2 + q_z^2}} \to -i\frac{d_{12}}{e}.$$
 (49)

Following exactly the same procedure of Sec. II, but with the F(q) defined in Eq. (49) and considering only the $q_z \to 0$ mode, we get

$$\Omega = \sqrt{\frac{N_{\text{2DEG}}\omega_{\text{LO}}d_{12}^2}{2\epsilon_0\epsilon_\rho L_{\text{OW}}\hbar}},$$
(50)

and thus, from Eq. (38),

$$D = \frac{N_{2\text{DEG}}d_{12}^2}{2\epsilon_0\epsilon_\infty L_{\text{OW}}\hbar}.$$
 (51)

In order to obtain the polaronic eigenfrequencies, we have to diagonalize the matrix in Eq. (36) using the coupling coefficients for the homogeneous limit defined in Eqs. (50) and (51). We thus obtain the secular equation

$$\omega^{4} - \omega^{2} (\omega_{LO}^{2} + \omega_{12}^{2} + 4D\omega_{12}) + \omega_{LO}^{2} \omega_{12}^{2} + 4D\omega_{12} \omega_{LO}^{2} - 4\Omega^{2} \omega_{12} \omega_{LO} = 0, \quad (52)$$

which, by using Eqs. (51) and (45), can be put into the form

$$\omega^4 - \omega^2 (\omega_{LO}^2 + \omega_{12}^2 + \omega_P^2) + \omega_{LO}^2 \omega_{12}^2 + \omega_P^2 \omega_{LO}^2 \frac{\epsilon_{\infty}}{\epsilon_s} = 0.$$
(53)

Equating the coefficients of Eqs. (48) and (53), we obtain

$$\omega_{\text{TO}}^2 = \omega_{\text{LO}}^2 \frac{\epsilon_{\infty}}{\epsilon_{\text{s}}},\tag{54}$$

which is the well-known Lyddane-Sachs-Teller relation.³⁰ We have thus proved that the homogeneous version of our theory gives the same results as the homogeneous dielectric function approach. It is anyway important to notice that the homogeneous limit is not exact, as a quantum well is, by definition, spatially inhomogeneous. Ignoring the higher q_z modes leads to underestimate the intersubband dipole of a factor roughly equal to $\sqrt{2}$.

V. CONCLUSIONS

In this paper, we have developed a microscopic theory of intersubband polarons, mixed excitations resulting from the coupling between intersubband transitions in doped quantum wells and LO phonons. We took into account the electron-electron Coulomb interaction and we were able to treat exactly the resulting depolarization shift. We proved that intersubband polarons can be in the ultrastrong-coupling regime, reaching extremely high values of the coupling constant. We critically discussed the relevance of our results both for fundamental and applied research.

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