We use linear response theory in order to compute the light absorption spectrum, in the terahertz band, of a polariton system composed by excitons in a quantum dot very strongly coupled to the lowest photon mode of a thin micropillar. In a thermalized (Bose condensed) system at low temperatures, the spectral function shows a peak associated to a $1s\rightarrow 2p$ exciton transition, enhanced by polariton effects. On the other hand, in a non-equilibrium system absorption is peaked at low energies. Thus, a measurement of terahertz absorption could give an indication of the degree of thermalization in the polariton system.

The strong coupling regime in the interaction between a confined photon mode and electron-hole pairs in semiconductor nanodevices has been demonstrated recently \[1\]. The quasiparticles, so called polaritons \[2, 3\], which are roughly half excitons and half photons, offer very interesting possibilities such as, for example, a new lasing mechanism (polariton lasing) based on their quasibosonic nature \[4\], with pumping threshold (related to ground-state occupation) two orders of magnitude lower than ordinary (photonic) lasing in the same devices \[5\], and operation at ambient temperatures \[6\].

In the present paper, we focus on the linear response of a model polariton system to terahertz radiation. The first motivation to carry on such a study is the intuitive idea that the interaction with the confined photon mode reinforces coherence of the excitonic subsystem and, thus, may reinforce the collective response of the excitons to the terahertz probe. This may result in a semiconductor version of the Giant Dipole Resonances (GDR), a phenomenon widely studied in nuclei \[7\] and electron clusters \[8\], with the possibility of controlling the position and intensity of the resonance by varying parameters such as the pumping rate or the photon-exciton detuning.

The second good reason to study terahertz absorption by excitonic polaritons is that it has proven to be very useful in order to observe exciton formation dynamics in quantum wells \[9\], and bulk systems \[10\]. We think, the available experimental techniques should be able to measure the degree of thermalization of the polariton system, not only under stationary conditions \[11\], but in the pumped regime as well \[12\]. Indicators following from interband emission alone are not enough because the main qualitative features (population of the lowest polariton state, behavior of the second order coherence function, etc) can be reproduced also from dynamical equations, without any thermalization mechanisms, both in the pumped \[13\] and in the stationary regimes \[14\].

Below, we compute terahertz absorption in two extreme situations. One is a Bose condensed state at low temperatures, described in a canonical ensemble. It is assumed that only the ground state has a significant occupation probability. The spectral function shows a GDR-like peak, whose position grows with the polariton number. The second case corresponds to a polariton system in a non-equilibrium stationary state (result of a balance between pumping and losses), with occupation probabilities that can not be described by a Gibbs distribution. The terahertz spectral function gets a completely different shape with a central peak at near zero energy. Intermediate situations would interpolate between the two extremes, and a measurement of the response in real systems would indicate their degree of thermalization. Let us stress, however, that calculations are carried on in a model with very strong light-matter coupling, in which Rabi splitting overpasses the exciton binding energy.

(i) Ground-state response of non-interacting polaritons

In order to get a preliminary estimate of the absorption spectrum, we first consider the ground-state response of non-interacting polaritons. We assume the system is in a Bose-condensed state, with $N_{pol}$ polaritons occupying a single state. Intraband absorption is described by the dipole operator acting only on the exciton functions. The absorption probability is then proportional to $|\alpha d_{10}|^2 N_{pol}$, where $\alpha$ is the Hopfield coefficient \[2\] (that is, amplitude of the exciton ground state function in the polariton function), and $d_{10}$ is the intraband dipole matrix element between ground-state exciton and an excited-state function. The latter is supposed to concentrate the oscillator strength for dipole transitions. Notice that the absorption probability increases with the number of polaritons in the ground state. The peak position, on the other hand, should be roughly constant, equal to the energy difference between the exciton ground- and excited states.

Finite, but low, temperatures, should lead to similar results. In a grand canonical description, on the other hand, which is more natural for the polariton system, sectors with polariton number near the mean value will contribute also to the spectral function with relatively high weights. The effects of polariton-polariton interactions is considered in the next paragraph.
FIG. 1: (Color online) (a) The ground-state spectral function, Eq. (1), for various \( N_{pol} \) numbers. (b) The non-equilibrium spectral function, Eq. (12), for pumping rates (in ps\(^{-1}\)) corresponding to mean polariton number in the steady state, \( \langle N_{pol} \rangle \), in the interval (1,10). The detuning parameter is \( \Delta = -3 \) meV.

(ii) Ground-state response of interacting polaritons

Polariton-polariton interactions come from residual Coulomb interactions between excitons. Instead of using a phenomenological approach, we start from a model in which Coulomb interactions are treated exactly, and the fermionic degrees of freedom are explicit. There is a finite number (10) of single-particle states for electron and holes, and a single photon mode. Saturation effects due to Fermi statistics are seen when the polariton number is around (or greater than) 10. A detailed description can be found elsewhere [14].

The very-low temperature (ground-state) response of the \( N_{pol} \)-polariton system is contained in the spectral function:

\[
S_0(\omega) = \sum_I |\langle I|d|J \rangle|^2 \frac{\Gamma_0}{\pi} \frac{1}{\Gamma^2_0 + (\omega_{IJ} - \omega)^2},
\]

where matrix elements, \( \langle I|d|J \rangle \), of the intraband dipole operator, \( d \sim \sum_i (\vec{r}_i^h - \vec{r}_i^e) \) (where \( \vec{r}_i^h \) and \( \vec{r}_i^e \) are, respectively, the hole and electron position vectors) shall be computed. \( |J \rangle \) is the ground state function of the \( N_{pol} \)-polariton system, and \( |I \rangle \) are excited states. \( \Gamma_0 = 0.1 \) meV/\( \hbar \) is a phenomenological damping parameter, and \( \omega_{IJ} = (E_I - E_J)/\hbar \) – the transition frequencies.

In our model, wave functions are constructed as linear combinations:

\[
|P \rangle = \sum_{S_e, S_h, n} C_{S_e, S_h, n} |S_e, S_h, n \rangle,
\]

where \( S_e \) and \( S_h \) are Slater determinants for electrons and holes, with electron and hole numbers \( N_e \) and \( N_h \), respectively, and \( n \) is the number of photons in the confined mode. Functions entering the combination preserve the polariton number:

\[
N_{pol} = N_e + n = N_h + n,
\]

and the total (envelope) angular momentum projection along the cavity axis (we assume a circular section):

\[
L = \sum_i (l_i^{(e)} + l_i^{(h)}).
\]

The ground-state function, \( |J \rangle \), has \( L = 0 \), whereas \( |I \rangle \) are \( L = 1 \) functions.

We show in Fig. 1 (a) the spectral function for different polariton numbers and detuning \( \Delta = -3 \) meV. In the model, the parameter \( \Delta \) measures the photon energy with respect to the nominal band gap, not the photon-exciton detuning. \( \Delta = -3 \) meV approximately corresponds to resonant conditions.

The GDRs can be identified as the dominant peaks in these curves. The peak position monotonously increases with increasing polariton number. This can be understood on intuitive grounds. The mass of the electron (or
hole) cloud is \( m \sim N_{\text{pairs}} \), and the Hooke coefficient for the force acting between clouds is \( k \sim N_{\text{pairs}}^2 \). Then, the excitation energy of the dipole mode is \( \hbar \omega \sim \sqrt{k/m} \sim \sqrt{N_{\text{pairs}}} \sim \sqrt{N_{\text{pol}}} \). The maximum intensity, on the other hand, has a non-trivial dependence on \( N_{\text{pol}} \), a kind of saturation effect is observed. The intensity first increases, as in the non-interacting case, but then, after reaching a maximum value, decays. These dependences are illustrated in Fig. 2 where the case \( \Delta = +3 \text{ meV} \), corresponding to an enhanced excitonic component of polaritons, is also shown. In this positive detuning situation, the absorption probability rises because the Hopfield parameter \( \alpha \) increases.

In spite of the fact that calculations are performed in a particular model, we expect that the statement about the existence of a peak in the absorption spectrum at relatively high excitation energies (of the order of the exciton \( 1s - 2p \) transition), whose intensity increases at least for polariton numbers well below saturation values, is general enough, and could be used as a criterium of a low-temperature, equilibrated (Bose-condensed) system.

(iii) Dynamical response of the non-equilibrium system (with non-resonant pumping and photon losses)

Below, we assume that relaxation mechanisms are not effective, and can not lead the polariton system to an equilibrium (thermal) state. The system is, thus, described by a density matrix, which is obtained from a master equation that takes care of photon losses through the cavity mirrors and incoherent (non-resonant) pumping. Details can be found in Ref. [14]. We solve the master equation in the stationary \((t \to \infty)\) limit in order to obtain the quasiequilibrium distribution, \( \rho^{(\infty)} \).

The response to the terahertz probe is computed in the linear approximation, where the probe does not modify the quasiequilibrium distribution. We adopt a computational scheme similar to the one used for the photoluminescence response [14]. The starting point is the first-order correlation function:

\[
\langle d^\dagger(t + \tau)d(t) \rangle = \sum_{I,J} \langle J|d^\dagger I \rangle g_{d,IJ}(t),
\]

written in terms of the auxiliary function:

\[
g_{d,IJ} = \langle \langle I \rangle(t + \tau) \rangle |d(t) \rangle,
\]

where \(|J\rangle\) are \( N_{\text{pol}} \)-polariton functions with total angular momentum \( L = 0 \), and the \(|I\rangle\) are \( N_{\text{pol}} \)-polariton functions with \( L = 1 \). Because of the Quantum Regression Theorem [15], \( g_{d,IJ} \) satisfies the same equation as the density matrix, that is [14]:

\[
\frac{d}{dt} g_{d,IJ} = (i\omega_{IJ} - \Gamma_{IJ}) g_{d,IJ} + \kappa \sum_{K,M} \langle J|a|I \rangle g_{d, MK} \langle K|a^\dagger I \rangle
\]

\[
- \frac{\kappa}{2} \sum_{K,M \neq I,J} \langle J|a|I \rangle \langle M|a|K \rangle g_{d, KJ} - \frac{\kappa}{2} \sum_{K,M \neq I,J} g_{d, KM} \langle M|a^\dagger |K \rangle \langle K|a|J \rangle, \tag{7}
\]

with boundary conditions at \( t \to \infty, \tau = 0 \):

\[
g_{d,IJ} = \sum_K \langle J|d|K \rangle \rho^{(\infty)}_{KJ}
\]

\[
\approx \langle J|d|J \rangle \rho^{(\infty)}_{JJ}, \tag{8}
\]

where, in the last step, we used the fact that \( \rho^{(\infty)}_{KJ} \) is approximately diagonal in the energy representation [13].

In Eq. (7), \( \kappa \) is the loss rate, \( 0.1 \text{ ps}^{-1} \) in our model. The widths, \( \Gamma_{IJ} \), are computed from:

\[
\Gamma_{IJ} = \frac{\kappa}{2} \sum_K \{ |\langle K|a|I \rangle|^2 + |\langle K|a|J \rangle|^2 \}
\]

\[
+ \frac{P}{2}(N_{\text{up}}(I) + N_{\text{up}}(J)), \tag{9}
\]

where \( P \) is the pumping rate, and \( N_{\text{up}}(I) \) is the number of states with polariton number \( N_{\text{pol}}(I) + 1 \) used to solve the equations.

The general solution of the linear system, Eq. (7), is written in terms of the eigenvalues, \( \lambda_n \), and eigenvectors, \( X^{(n)}_{IJ} \), of the matrix \( B_{IJ, MK} \) defined by the r.h.s. of Eq. (7), that is:

\[
g_{d,IJ}(t) = \sum_n C_n \exp(\lambda_n \tau) X^{(n)}_{IJ}, \tag{10}
\]

where the coefficients \( C_n \) are determined from the boundary conditions, Eq. (8).

The Fourier transform of Eq. (7) defines the response spectral function to the terahertz probe in the quasi-equilibrium system:

\[
S_{ne}(\omega) = -\frac{1}{2\pi} \sum_n \sum_{I,J} \frac{D^{(r)}_{IJ,n} \lambda^{(r)}_{n} + D^{(i)}_{IJ,n} \lambda^{(i)}_{n} - \omega}{(\lambda^{(r)}_{n})^2 + (\lambda^{(i)}_{n} - \omega)^2}, \tag{11}
\]

where \( D_{IJ,n} = \langle J|d^\dagger I \rangle C_n X^{(n)}_{IJ} \), and superscripts \((r), (i)\) refer to the real and imaginary parts of the magnitudes, respectively.

A simplified and more intuitive expression comes from the diagonal terms of Eq. (7), [14] Notice that, for excitation energies \( \hbar \omega > 1 \text{ meV} \), the diagonal is at least
Energy of \( N_{\text{pol}} \) states (meV)

State labels

1465
1470
1475
1480
1485
1490
1495

FIG. 3: (Color online) The lowest \( N_{\text{pol}} = 2 \) states with \( L = 0 \) and \( L = 1 \) in the model. \( N_{\text{pol}} = 1, L = 0 \) states are drawn as a reference, and also in order to show that \( L = 0 \) bands with different \( N_{\text{pol}} \) numbers are almost parallel. A big number of near zero-energy dipole transitions are possible in the \( N_{\text{pol}} = 2 \) sector.

10 times higher than the off-diagonal elements (because \( \kappa = 0.1 \text{ ps}^{-1} \)). Neglecting the off-diagonal terms, we get:

\[
S_{\text{nc}}(\omega) \approx \frac{1}{\pi} \sum_{I,J} |\langle I|d|J\rangle|^2 \rho^{(\infty)}_{IJ} \Gamma_{IJ}(\omega_{IJ} - \omega)^2.
\]  

As compared with \( S_0 \), the non-equilibrium spectral function includes also contributions from the excited states, \( |J\rangle \), which may have relatively high occupation probabilities, \( \rho^{(\infty)}_{IJ} \), as can be seen, for example, in Fig. 6 of Ref. [14]. On the other hand, the dipole matrix elements for transitions originated in excited states could be much stronger than ground-state dipole elements. This statement follows from the energy-weighted sum rule for dipole transitions [10]:

\[
\sum_I \Delta E_{IJ} |\langle I|d|J\rangle|^2 = C,
\]

where constant \( C \) does not depend on the indices \( J \).

The sum in Eq. (13) reduces to a single term when the oscillator strength from state \( |J\rangle \) is concentrated on a single state, \( |I\rangle \). Then, if there were excited states \( |J\rangle \) for which the dominant transitions have \( \Delta E_{IJ} \sim 0.1 \text{ meV} \), for example, their contribution to \( S_{\text{nc}} \) would be 100 times stronger than the ground state contribution. This is, indeed, what one sees in the spectral function, Fig. 1 (b). An extra factor of around 20 comes from the number of excited states. We have drawn in this picture the non-equilibrium spectral function for pumping rates, \( P \), corresponding approximately to the same situations depicted in Fig. 1 (a). That is, the mean polariton number \( \langle N_{\text{pol}} \rangle = \sum_J \rho^{(\infty)}_{JJ} N_{\text{pol}}(J) \) for \( P = 0.01 \text{ ps}^{-1} \), for example, is around 4, etc. In Fig. 3, we show that near zero-energy dipole transitions are very common in our model, and should be very common also in micropillars with enameled quantum wells because of the exciton near flat band.

In conclusion, we expect the absorption spectral function for a non-equilibrated polariton system to be peaked at near zero energies, in clear contrast with the Bose-condensed system, whose spectral function is peaked at the GDR. The dependence on \( N_{\text{pol}} \) is also very different. In the thermalized system absorption increases with increasing polariton number, whereas in the nonequilibrium system it decreases as the pumping rate increases. Thus, terahertz absorption could be a sharp criterion allowing to discriminate between the thermalized and the non-equilibrium scenarios.

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