TERAHERTZ ABSORPTION BY EXCITONIC POLARITONS

ABSORCIÓN EN LOS TERAHERTZ POR POLARITONES EXITÓNICOS

Carlos Andrés Vera-Ciro[a], Alain Delgado[b] and Augusto González[c †]

a) Kapteyn Astronomical Institute, University of Groningen, P.O. Box 800, 9700 AV Groningen, The Netherlands
b) Centro de Aplicaciones Tecnológicas y Desarrollo Nuclear, Calle 30 No 502, Miramar, CP 11300, La Habana, Cuba
c) Instituto de Cibernética, Matemática y Física, Calle E 309, Vedado, CP 10400, La Habana, Cuba, agonzale@icimaf.cu†
† corresponding author

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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-2p* like 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.

La teoría de respuesta lineal es utilizada para calcular el espectro de absorción, en la banda de terahertz, de un sistema polaritónico compuesto por excitones en un punto cuántico fuertemente acoplados al modo fotónico fundamental confinado en un micropilar. En un sistema termalizado (condensado de Bose) la función espectral muestra un pico, asociado a la transición excitónica *1s-2p*, reforzada por efectos polaritónicos. Por el contrario, en un sistema no equilibrado el pico de absorción se localiza a bajas energías. Luego, la medición de la absorción de terahertz podría indicar el grado de termalización de los polaritones.

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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 (photon) 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 absortion 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]. In the polariton system, a few years ago the common belief was that a thermalized Bose-condensed state is reached [11,12]. Very recently, however, this conclusion along with the interpretation of most experiments is being questioned [13]. 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 [14] and in the stationary regimes [15].

Below, we compute terahertz absorption in two extreme situations. One is a Bose condensed state at very low temperatures, in such a way that only the many-particle ground state has a significant occupation probability. In our model, with not very realistic parameters, the 1*s*-2*p* excitonic transition is located at around 10 *meV*, that is the temperature should be lower than 100 *K*, a common experimental situation. The spectral function shows a GDR-like peak, whose position grows with the polariton number, Fig. 1 (a).

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 which practically does not depend on the pumping rate, Fig. 1 (b).

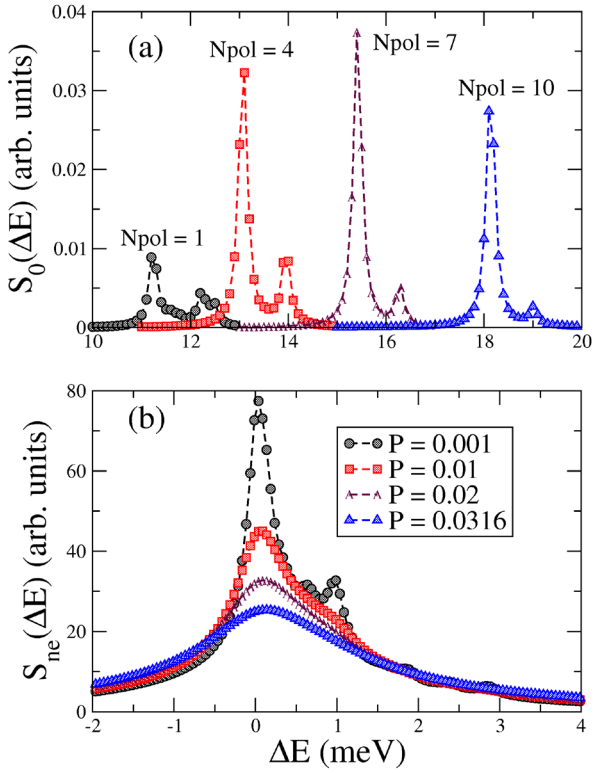

Figure 1. Comparison between the equilibrium and non-equilibrium terahertz absorption. (a) Ground-state spectral function, Eq. (1), for various N_{pol} numbers. At a given N_{pol}, the GDR is the highest peak. (b) Non-equilibrium spectral function, Eq. (12), for pumping rates (in *ps*) corresponding to mean polariton number in the interval (1,10). The detuning parameter is Δ = -3 *meV*.

Intermediate, real experimental, situations would interpolate between the two extremes, and a measurement of the response in real systems would indicate their degree of thermalization.

Calculations are carried on in a model for the quantum dot-microcavity system, detailed described in Ref. [15], with very strong light-matter coupling constant (3 *meV*), which leads to a significant blueshift of the GDR resonance with respect to the 1*s*-2*p* like exciton transition. The main qualitative conclusions of the paper are expected to be valid also for any relatively large quantum dot or thin quantum well micropillar working under the strong coupling regime.

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 α is the Hopfield coefficient [2] (that is, the weight of the exciton in the polariton function), and d_{10} is the intra-band 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 almost constant, roughly 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.

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 [15].

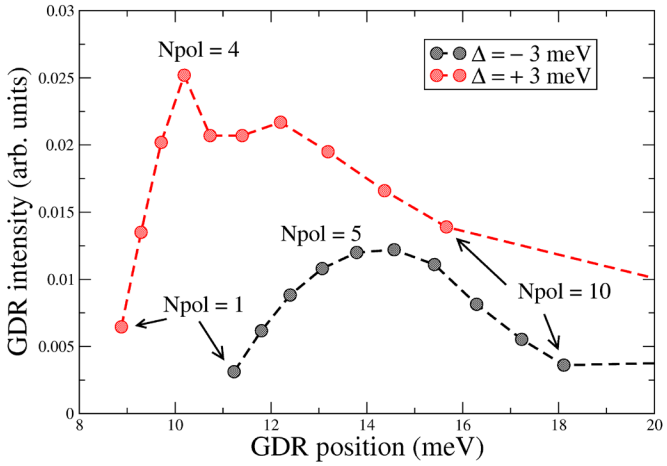

Figure 2. Intensity (that is, dipole matrix elements squared) and position of the GDR peak for two different values of the detuning, Δ. Each dot corresponds to a given N_{pol}.

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

$$S_0(\omega) = \sum_I \frac{|\langle I|d|J\rangle|^2\ \Gamma_0/\pi}{\Gamma_0^2 + (\omega_{IJ} - \omega)^2}, \qquad (1)$$

where matrix elements, $\langle I|d|J\rangle$, of the intraband dipole operator, $d \sim \Sigma_i(\vec{r}_i^{(h)} - \vec{r}_i^{(e)})$ (where $\vec{r}^{(h)}$ and $\vec{r}^{(e)}$ 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

$$|P\rangle = \sum_{S_e,S_h,n} C_{S_e,S_h,n}\,|S_e,S_h,n\rangle, \qquad (2)$$

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

$$N_{pol} = N_e + n = N_h + n \tag{3}$$

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

$$L = \sum_i (l_i^{(e)} + l_i^{(h)}). \tag{4}$$

In Eq. (4), the index i labels the particles. $l_i^{(e)}$, for example, corresponds to the angular momentum projection along the cavity axis of the i-th electron. 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 Δ = -3 *meV*. In the model, the parameter Δ measures the photon energy with respect to the nominal band gap, not the photon-exciton detuning. Δ = -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 $m \sim N_{pairs}$, and the Hooke coefficient for the force acting between clouds is $k \sim N_{pairs}^2$. Then, the excitation energy of the dipole mode is $\hbar\omega \sim \sqrt{k/m} \sim \sqrt{N_{pairs}} \sim \sqrt{N_{pol}}$. The maximum intensity, on the other hand, has a non-trivial dependence on N_{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 Δ = +3 *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 α 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 1*s*-2*p* 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 system in equilibrium (Bose-condensed) stae.

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. [15]. 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 [15]. The starting point is the first-order correlation function

$$\langle d\ (t+\tau) d(t)\rangle = \sum_{I,J} \langle J|d\ |I\rangle g_{d,IJ}, \tag{5}$$

written in terms of the auxiliary function

$$g_{d,IJ}(t+\tau, t) = \langle (|J\rangle\langle I|)(t+\tau) d(t)\rangle, \tag{6}$$

where $|J\rangle$ are N_{pol}-polariton functions with total angular momentum $L = 0$, and the $|I\rangle$ are N_{pol}-polariton functions with $L = 1$. Because of the Quantum Regression Theorem [16], $g_{d,IJ}$ satisfies the same equation as the density matrix, that is [15]

$$\begin{aligned} \frac{\mathrm{d}}{\mathrm{d}\tau} g_{d,IJ} &= (i\omega_{IJ} - \Gamma_{IJ}) g_{d,IJ} \\ &+ \kappa \sum_{K,M} \langle I|a|M\rangle g_{d,MK} \langle K|a\ |J\rangle \\ &- \frac{\kappa}{2} \sum_{K \neq I, M} \langle I|a\ |M\rangle\langle M|a|K\rangle g_{d,KJ} \\ &- \frac{\kappa}{2} \sum_{K, M \neq J} g_{d,IM} \langle M|a\ |K\rangle\langle K|a|J\rangle \end{aligned}, \tag{7}$$

with boundary conditions at $t \to \infty$, $\tau = 0$

$$\begin{aligned} g_{d,IJ} &= \sum_K \langle I|d|K\rangle \rho_{KJ}^{(\infty)} \\ &\approx \langle I|d|J\rangle \rho_{JJ}^{(\infty)} \end{aligned}, \tag{8}$$

where, in the last step, we used the fact that $\rho^{(\infty)}_{KJ}$ is approximately diagonal in the energy representation [14].

In Eq. (7), κ is the loss rate, 0.1 ps^{-1} in our model. The widths, Γ_{IJ}, are computed from

$$\begin{aligned} \Gamma_{IJ} &= \frac{\kappa}{2} \sum_K \{|\langle K|a|I\rangle|^2 + |\langle K|a|J\rangle|^2\} \\ &+ \frac{P}{2} \{N_{up}(I) + N_{up}(J)\} \end{aligned}, \tag{9}$$

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

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

$$g_{d,IJ}(\tau) = \sum_n C_n \exp(\lambda_n \tau) X_{IJ}^{(n)}, \tag{10}$$

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

The Fourier transform of Eq. (5) defines the response spectral function to the terahertz probe in the quasi-equilibrium system

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

where $D_{IJ,n} = \langle J \mid d \mid I \rangle C_n X_{IJ}^{(n)}$, 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) [15]. Notice that, for excitation energies $\hbar\omega > 1\ meV$, the diagonal is at least 10 times higher than the off-diagonal elements (because $\kappa = 0.1\ ps^{-1}$). Neglecting the off-diagonal terms, we get:

$$S_{ne}(\omega) \approx \frac{1}{\pi} \sum_{I,J} \frac{|\langle I \mid d \mid J \rangle|^2 \ \rho_{JJ}^{(\infty)} \Gamma_{IJ}}{\Gamma_{IJ}^2 + (\omega_{IJ} - \omega)^2}. \qquad (12)$$

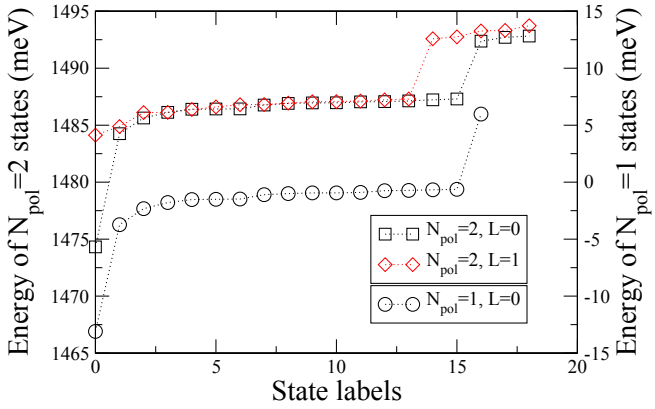

Figure 3. The lowest $N_{pol} = 2$ states with $L = 0$ and $L = 1$ in the model. A big number of near zero-energy dipole transitions are possible in the $N_{pol} = 2$ sector. We draw in the same figure, shifted by the nominal E_{gap}, the $N_{pol} = 1$, $L = 0$ states. Notice that $L = 0$ bands with different N_{pol} numbers are almost parallel.

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, $\rho^{(\infty)}_{JJ}$, as can be seen, for example, in Fig. 6 of Ref. [15]. 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 [17, 18]:

$$\sum_I \Delta E_{IJ} \, |\langle I \mid d \mid J \rangle|^2 = C, \qquad (13)$$

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 $\Delta E_{IJ} \sim 0.1\ meV$, for example, their contribution to S_{ne} 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 $(\langle N_{pol} \rangle = \Sigma_J \rho_{JJ}^{(\infty)} N_{pol}(J))$ for P = 0.01 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 embeebed quantum wells because of the exciton near flat band.

In conclusion, we expect the absorption spectral function for a non-equilibrium 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_{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 criterium allowing to discriminate between the thermalized and the non-equilibrium scenarios.

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