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Polaritons in Confined Systems

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Polaritons in confined systems are introduced and their dispersion is calculated. The amount of squeezing of confined excitons–polaritons in GaAs quantum wells is evaluated.

KEY WORDS: Confined quantum systems; excitons–polaritons; squeezing.

1. INTRODUCTION

The quantum mechanical description of radiation propagation in a polarizable medium through polaritons is well known since the work by Hopfield.[1] The dispersion curves of these excitations as well as their role in the description of radiation–matter interaction in solids have been widely discussed both theoretically and experimentally (e.g., refs. 2). Recently the statistical properties of polariton states in a bulk crystal have been studied[3] and it has been shown that polariton states exhibit intrinsic squeezing properties. As we shall see, this means that the variance of a linear combination of polariton amplitudes is smaller than $\hbar/2$. This effect is well known from quantum optics, where it has been observed for the radiation field. One interesting feature of polariton squeezing is that it is intrinsic to the polariton, being a consequence of the transformation from free photons and excitons (or phonons) into polaritons.[3] For bulk polaritons this effect may be relevant either for phonon–polaritons or for exciton–polaritons in materials like alkali halides, where the coupling may

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be large. It is of some relevance also in confined quantum systems because of the stronger localization of the exciton. Furthermore, the polariton effect in a confined system strongly differs from that in the bulk; in particular polaritons show a characteristic radiative lifetime depending on the energy of the photons which interact on the quantum system.

Polaritons in confined systems like quantum wells or quantum wires have already been considered in the literature both from a classical[4] and from a quantum mechanical viewpoint.[5] In this paper we present a unified description from which the classical and the quantum mechanical results follow and which allows predictions about the statistical properties of the polaritons in quantum wells.

2. DISPERSION OF THE POLARITONS

Here we consider the interaction between photons and excitons in the confined system with the Hamiltonian discussed in ref. 5, which describes the interaction of a quantized electromagnetic field with a quasi-two-dimensional polarizable medium. The medium is strongly confined in the growth direction (z direction) and behaves like a two-dimensional crystal in the transverse direction. The Hamiltonian reads

$$\begin{aligned} H = &\sum_{\mathbf{q}} \hbar\omega_q A_{\mathbf{q}}^{2+} A_{\mathbf{q}}^{2} + \sum_{Q,\lambda} \hbar v\, |\mathbf{Q}|\, A_{\mathbf{Q},\lambda}^{1+} A_{\mathbf{Q},\lambda}^{1} \\ &+ i \sum_{\mathbf{Q},\lambda} C_{\mathbf{Q}}^{\lambda} (A_{-\mathbf{q}}^{2} - A_{\mathbf{q}}^{2+})(A_{\mathbf{Q},\lambda}^{1} + A_{-\mathbf{Q},\lambda}^{1+}) \\ &+ \left(\frac{1}{\hbar\omega_q}\right) \sum_{\mathbf{Q}',\mathbf{Q},\lambda,\lambda'} C_{\mathbf{Q}}^{\lambda} C_{\mathbf{Q}'}^{\lambda'*} (A_{\mathbf{Q}',\lambda'}^{1} + A_{-\mathbf{Q}',\lambda'}^{1+})(A_{\mathbf{Q},\lambda}^{1} + A_{-\mathbf{Q},\lambda}^{1+}) \end{aligned} \tag{1}$$

where $A_{\mathbf{q}}^{2+}$ and $A_{\mathbf{q}}^{2}$ are the creation and annihilation operators of the two-dimensional excitons, $\mathbf{q}$ is the component of the wave vector $\mathbf{Q}$ perpendicular to the z direction, and k_z is the component of $\mathbf{Q}$ in the z direction. $A_{\mathbf{q}}^{1+}$ and $A_{\mathbf{q}}^{1}$ are the field operators. The coupling constants are defined as

$$C_Q^{\lambda} = C_{\mathbf{q},k_z}^{\lambda} = C_{-\mathbf{q},-k_z}^{\lambda *} = \left(\frac{2\pi\hbar v}{L'\,|\mathbf{Q}|}\right)^{1/2} \omega_q \boldsymbol{\mu}_{cv} \cdot \hat{\mathbf{e}}_{\mathbf{q},k_z,\lambda} \frac{1}{c} F(0) \frac{1}{L} \int_{-L/2}^{L/2} dz\, \rho(z)\, e^{ik_z z} \tag{2}$$

where $v = c/\sqrt{\varepsilon_\infty}$ is the velocity of light in the medium, $\omega_q = \omega_0 + \gamma q^2$ is the exciton frequency, $A_{\mathbf{Q},\lambda}^{1} = A_{\mathbf{q},k_z,\lambda}^{1}$, and $A_{-\mathbf{Q},\lambda}^{1+} = A_{-\mathbf{q},-k_z,\lambda}^{1+}$.

The electromagnetic field has been quantized inside a box of dimension L' with periodic boundary conditions. The coupling constant is derived from a phenomenological model for the exciton in a quantum well and we have included the A^2 term which was neglected in ref. 5. It will turn out to be important in order to give the correct behavior of the polariton dispersion for $q \to 0$. For simplicity we consider only one polarization for the field and diagonalize the Hamiltonian using the transformation

$$B^l_{\mathbf{q}} = \sum_{k_z} W_l(k_z, \mathbf{q})\, A^1_{\mathbf{q},k_z} + X_l(\mathbf{q})\, A^2_{\mathbf{q}} + \sum_{k_z} Y_l(k_z, \mathbf{q})\, A^{1^+}_{-\mathbf{q},-k_z} + Z_l(\mathbf{q})\, A^{2^+}_{-\mathbf{q}} \tag{3}$$

where l indicates the polariton mode.

We consider now the limit $L' \to \infty$; in this case l becomes a continuous index which will be omitted in the following. Furthermore we define the L'-independent coupling constant $|\tilde{C}_{\mathbf{q},k_z}|^2 = |C^\lambda_{\mathbf{q},k_z}|^2\, L'/2\pi$. The polariton operators $B^l_{\mathbf{q}}$ will be denoted by $B_{\mathbf{q}}(\Omega)$ and the coefficients of the transformation (3) are in this limit

$$Z(\mathbf{q}) = \frac{\omega_q - \Omega}{\omega_q + \Omega}\, X(\mathbf{q}) \tag{4}$$

$$W(k_z, \mathbf{q}) = \frac{-i}{\hbar}\, \frac{2\omega_q}{\omega_q + \Omega} \left[\frac{1}{v\,|\mathbf{Q}| - \Omega} + M\delta(v^2\mathbf{Q}^2 - \Omega^2) \right] \frac{\Omega^2}{\omega_q^2}\, C^\lambda_{\mathbf{q},k_z} X(\mathbf{q}) \tag{5}$$

$$Y(k_z, \mathbf{q}) = \frac{v\,|\mathbf{Q}| - \Omega}{v\,|\mathbf{Q}| + \Omega}\, W(k_z, \mathbf{q}) \tag{6}$$

where $X(\mathbf{q})$ is determined from $[B_q(\Omega), B_q(\Omega')^+] = \delta(\Omega - \Omega')$ and M will be determined later from the dispersion. Here Ω is the polariton frequency. We distinguish between two different frequency regions, i.e., the lower and the upper polariton.

For $\Omega(q) < v(k_z^2 + q^2)^{1/2}$ the delta function in (5) vanishes. In this case the consistency condition for the transformation (3) leads to the eigenvalue equation

$$\Omega^2 - \omega_q^2 + \frac{4\omega_q}{\hbar^2} \frac{\Omega^2}{\omega_q^2} \int \frac{v\,|\mathbf{Q}|}{v^2\,|\mathbf{Q}|^2 - \Omega^2}\, |\tilde{C}_{\mathbf{q},k_z}|^2\, dk_z = 0 \tag{7}$$

which defines the lower polariton. Its dispersion curve is presented in Fig. 1 and does not differ much from the one for the bulk polariton. Notice that the dispersion curve for the lower polariton starts at $q = 0$ and coincides with the one obtained from the classical approach.[4] This behavior is a consequence of having included the A^2 in the Hamiltonian and was not considered in the previous quantum approaches,[5] where the dispersion curves started at $q \neq 0$.

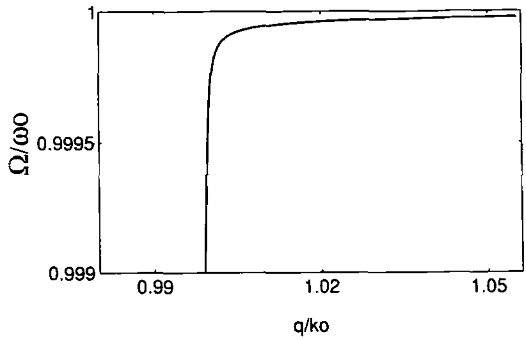

Fig. 1. Dispersion curve of the lower polariton in a GaAs quantum well of 60 Å width. The polariton frequency Ω is normalized with respect to the exciton frequency ω_0, the wave vector is normalized with respect to $k_0 = \omega_0/v$. The material parameters are defined in the text.

For $\Omega(q) \geqslant v(k_z^2 + q^2)^{1/2}$ the term proportional to the delta function in (5) may be different from zero and the quantity M is determined from the consistency condition for the transformation. It reads

$$M = -\left(\Omega^2 - \omega_q^2 + \frac{4\omega_q}{\hbar^2}\frac{\Omega^2}{\omega_q^2}\int \frac{v\,|\mathbf{Q}|}{v^2\,|\mathbf{Q}|^2 - \Omega^2}\,|\tilde{C}_{\mathbf{q},k_z}|^2\,dk_z\right) \times \left(\frac{2\omega_q}{\hbar^2}\frac{\Omega^2}{\omega_q^2}\int |\tilde{C}_{\mathbf{q},k_z}|^2\,\delta(v^2\mathbf{Q}^2 - \Omega^2)\,dk_z\right)^{-1} \tag{8}$$

We call the excitations which are present in this energy region upper polaritons although they differ from the bulk upper polariton as we will see later. The coefficient $X(\mathbf{q})$ in the transformation (3) is determined from the commutator $[B_\mathbf{q}(\Omega), B_\mathbf{q}(\Omega')^+] = \delta(\Omega - \Omega')$, and we obtain

$$|X_{(\mathbf{q})} + Z_{(\mathbf{q})}|^2 = \frac{1}{\pi}\frac{\omega_q^2}{\Omega^2}\frac{\Gamma}{R^2 + \Gamma^2} \tag{9a}$$

where

$$R = \frac{1}{2\omega_q}\left(\Omega^2 - \omega_q^2 + \frac{4\omega_q}{\hbar^2}\frac{\Omega^2}{\omega_q^2}\int \frac{v\,|\mathbf{Q}|}{v^2\,|\mathbf{Q}|^2 - \Omega^2}\,|\tilde{C}_{\mathbf{q},k_z}|^2\,dk_z\right) \tag{9b}$$

The right-hand side of this expression shows the typical Lorentzian behavior with a linewidth defined as

$$\Gamma = \frac{2\pi\Omega^3}{\hbar^2\omega_q^2}\int |\tilde{C}_{\mathbf{q},k_z}|^2\,\delta(v^2\mathbf{Q}^2 - \Omega^2)\,dk_z \tag{10}$$

This expression coincides with the classical and quantum linewidth as discussed in refs. 4 and 5, respectively. In the quantum well, states with zero linewidth correspond to two-dimensional polaritonlike excitations in the q plane, i.e., the lower polariton.

Frequency Ω and linewidth Γ defining the upper polaritons may be interpreted as the solution of a Fano-like problem in which the exciton state, for a fixed value of q, corresponds to the discrete state. The continuum is related to the k_z component of the wave vector of the electromagnetic field for fixed q. In contrast to the bulk case, the upper polaritons have a continuous energy spectrum. However, they are characterized by a resonance near the exciton energy which is obtained from the equation $M=0$ and which coincides with the classical resonance of ref. 4. We can interpret this resonance in our scheme as follows: consider the mean value of the polariton number operator in a one-exciton state as a function of the polariton energy. Its expression is given by

$$\begin{aligned}\langle B_{\mathbf{q}}^{+}(\Omega)\, B_{\mathbf{q}}(\Omega)\rangle &= |X_{(\mathbf{q})}|^2 + |Z_{(\mathbf{q})}|^2 + (L'/2\pi)\int |Y_{(k_z,\mathbf{q})}|^2\, dk_z \\ &= \left[2\hbar^2\Omega\left(\frac{\Omega^2+\omega_q^2}{2\omega_q^2} + \frac{\Omega^4}{\omega_q^4}\int \frac{|\tilde{C}_{\mathbf{q},k_z}|^2}{\hbar^2(vQ+\Omega)^2}\, dk_z\right)\right] \\ &\quad \times \left\{\frac{\Omega^4}{\omega_q^4}\int dk_z\, |\tilde{C}_{\mathbf{q},k_z}|^2\, \delta(v^2\mathbf{Q}^2-\Omega^2)[M^2+4\pi^2\Omega^2]\right\}^{-1}\end{aligned} \tag{11}$$

and its maximum is found when $M=0$. Therefore this condition gives the maximum number of upper polaritons which are compatible with a given exciton state.

The lower and the upper polaritons can also be introduced through the retarded Green's function for the exciton, as can be shown defining the inverse of the transformation (3) and expressing the time evolution of the exciton and photon operators through that of the polariton operators whose eigenfrequencies are known. A particular upper polariton is defined by the resonance of the Green's function for $M=0$. This particular polariton mode is the counterpart of the upper polariton in the bulk. The analogous treatment for the Fano model is discussed in ref. 6.

3. STATISTICAL PROPERTIES

The transformation (3) allows us to construct the polariton states out of the product of free particle states and we give some indications on the quantum statistical properties of polaritons in a confined system.

Here we discuss one of these properties: polariton squeezing. This property is introduced through the analogy with squeezing for a harmonic oscillator described by the operators A_1 and A_1^+. Consider its so-called "in-quadrature" component $d_1 = (A_1 + A_1^+)/2$ and its "in-phase" component $d_2 = (A_1 - A_1^+)/2i$. In a coherent state the product of the mean values of the variances of this quantity is $\Delta d_1 \, \Delta d_2 = 1/4$. A squeezed state is characterized by $\Delta d_1 \, \Delta d_2 \geqslant 1/4$ but $\Delta d_1 < 1/2$ or $\Delta d_2 < 1/2$. This implies that for one component of the amplitude of the electromagnetic field the noise is reduced below the quantum limit. Here we show that this effect is also present for polaritons in confined systems as an intrinsic property. We introduce the quantities

$$d_1 = \frac{1}{2^{3/2}} [B_{\mathbf{q}} + B_{-\mathbf{q}} + B_{-\mathbf{q}}^+ + B_{\mathbf{q}}^+] \tag{12a}$$

$$d_2 = \frac{-i}{2^{3/2}} [B_{\mathbf{q}} + B_{-\mathbf{q}} - B_{-\mathbf{q}}^+ - B_{\mathbf{q}}^+] \tag{12b}$$

and as an example we evaluate the variances in the exciton–photon vacuum $|0\rangle$. This leads to the result

$$\begin{aligned} \langle (\Delta d_2)^2 \rangle \equiv \langle 0 | \, (\Delta d_2)^2 \, | 0 \rangle \\ = \frac{1}{4} \left[|Z(\mathbf{q})|^2 + |X(\mathbf{q})|^2 + \frac{L'}{2\pi} \int dk_z \, \{ |W(k_z, \mathbf{q})|^2 + |Y(k_z, \mathbf{q})|^2 \} \right] \\ - \frac{1}{2} \operatorname{Re} X(\mathbf{q}) \, Z(\mathbf{q}) - \frac{1}{2} \operatorname{Re} \frac{L'}{2\pi} \int dk_z \, W(k_z, \mathbf{q}) \, Y(-k_z, -\mathbf{q}) \end{aligned} \tag{13}$$

We have calculated (13) for the lower polariton in a GaAs quantum well with a width $L = 60$ Å. Cavity and quantum well have the same dielectric constant $\varepsilon = 12$ and an exciton energy of 1.6 eV. We also use the relation $|\boldsymbol{\mu}_{cv}|^2 \, |F(0)|^2/\hbar = fe^2/2m\omega_0$ between the oscillator strength f and the quantity used here; the value of $f = 36 \times 10^{-5}$ Å^{-2} is taken from ref. 4. The result is presented in Fig. 2 in terms of the quantity

$$\Delta_{\text{sq}} \equiv \left[1 - \left(\frac{\langle (\Delta d_2)^2 \rangle}{1/4} \right)^{1/2} \right] \times 100 \tag{14}$$

One obtains an amount of squeezing of about 0.06 %, which is small, but larger than in the bulk case[3] as a consequence of a larger oscillator strength of the confined exciton. For the upper polariton squeezing is

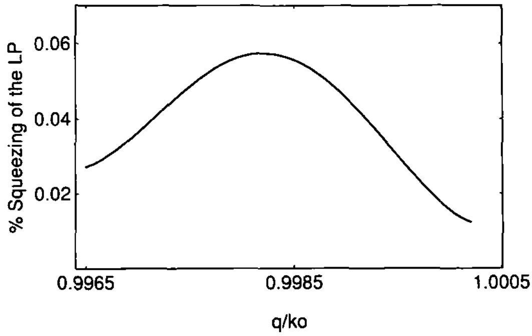

Fig. 2. Squeezing percentage Δ_{sq} of the d_2 operator for the lower polariton in a GaAs quantum well of 60 Å as a function of the normalized wave vector q/k_0 with $k_0 = \omega_0/v$. The material parameters are defined in the text.

defined in the same way. Here we have evaluated the squeezing for a quantum well in an infinite cavity; the effect should be measurable in realistic cavities of finite length and reflectivity close to one.

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