

Inversion of the excitonic spectrum in semiconductor microcavities

A.V. Kavokin

Physics and Astronomy School, University of Southampton, SO17 1BJ Southampton, United Kingdom.

G. Malpuech

LASMEA, UMR 6602 CNRS, Université Blaise-Pascal, 24, av des Landais, 63177, Aubière, France.

We show that coupling of odd photonic modes in microcavities with odd excited exciton states in wide quantum wells (WQWs) situated at the nodes of the electric field of the cavity mode may lead to the inversion of the excitonic spectrum: an odd exciton-polariton state (X2) may lie at lower energy than even (X1) exciton state. The kinetics of exciton-polariton relaxation is expected to be very peculiar in this case: the excitons are accumulated in the optically inactive even state at the beginning, and then scatter to the upper and lower odd polariton modes. This regime is favourable for Bose-Einstein condensation of X1 excitons whose life-time becomes much longer because of their decoupling from the cavity mode. Experimentally, the evidence for condensation of “dark” X1 excitons would come from the coherent emission of light from the ground odd exciton-polariton state. If the WQW is shifted from the node of the field, X1 excitons get coupled to light, and the anticrossing between even and odd polaritons can be distinguished in reflection spectra.

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1. INTRODUCTION

Semiconductor microcavities [1] represent a unique laboratory for studying of light-matter coupling effects in solids. Due to photonic confinement, the strength of interaction between photons and excitons can be enhanced in such extent that the splitting of exciton-polariton modes (Rabi splitting) exceeds the broadening of these modes. This happens in the strong-coupling regime, first evidenced by Weisbuch and co-workers [2] and then extensively studied in various semiconductor systems [3-6]. A typical microcavity structure consists of a semiconductor layer with embedded quantum wells (QWs) sandwiched between two Bragg mirrors. Usually, the QWs are placed at the anti-nodes of the electric field of the standing light mode of the cavity in order to enhance coupling between light and e1hh1 exciton resonances [7]. In this configuration, two exciton-polariton modes are formed having an even profile with respect to the cavity centre. In the strong coupling regime they demonstrate an anticrossing behaviour. Relaxation of exciton-polaritons along the lower dispersion branch may lead to their Bose-Einstein condensation and polariton lasing largely discussed in literature in the recent years [8-10]. In this picture excited exciton states do not play any role remaining “dark” (i.e. uncoupled to the cavity mode).

In this paper, we propose to study a different geometry of the microcavities allowing for the

coupling of odd cavity modes with odd excited exciton states. By odd exciton states we mean those excited states that have odd center of mass wave-functions $\Phi(z)$ in a wide quantum well (WQW) where an exciton is confined as a whole particle. The QW width must exceed the exciton Bohr radius in this case. We shall consider, in particular, the second exciton state (X2). This state is originated from e1hh2, e2hh1, and e2hh2 quantum states of a narrow QW mixed by the electron-hole Coulomb interaction. It is optically active indeed. We show that coupling of such a state with an odd cavity mode, though it is weaker than coupling of even cavity modes with the even X1 exciton state (originated from the ground e1hh1 exciton of a narrow well) in the traditional geometry, may lead to the Rabi splitting and anticrossing of exciton-polariton modes. A very peculiar situation may take place if this splitting exceeds the splitting between X1 and X2 states: X1 state remaining uncoupled to the cavity mode lies between two exciton-polariton modes formed by odd excitons. The inversion of the excitonic spectrum thus takes place: the state with higher electron and hole quantum numbers has the lower energy.

This inversion has an important impact on the polariton relaxation. Under non-resonant excitation, optically created electron-hole pairs are first bound mostly to X1 excitons. These excitons have a huge radiative live-time as they are decoupled from the cavity mode (kind of Purcell effect). That is why the relaxation time

of these excitons may be shorter than their life-time that simplifies their Bose-condensation at the bottom of their dispersion curve. The condensate of X1 excitons can be depleted due to the polariton-polariton scattering toward lower and upper odd exciton-polariton branches. Such a scattering requires energy and momentum conservation and therefore goes only to a limited number of quantum states within lower and upper polariton branches. Under a proper choice of parameters, at negative detuning between the photon mode and odd exciton resonance, the scattering from the condensate of dark X1 excitons directly populates the ground odd polariton state. Radiation from this state may have all characteristics of the laser light.

Thus, microcavities with inverted excitonic spectrum are good candidates for observation of the Bose-Einstein condensation of excitons. They have two advantages with respect to the conventional polariton laser structures: (i) the realisation of X1 excitons is possible because of their decoupling from light, (ii) the condensate of exciton-polaritons is formed by a single-step scattering process from the excitonic condensate, which allows to avoid problems of kinetic blocking or bottleneck effect [11] preventing the polaritons from relaxation to their ground state. The main difficulty in realisation of microcavities with the inverted spectrum comes from the weaker exciton-light coupling strength in these structures. We show numerically that placing 5 WQWs at the nodes of the electric field of the cavity mode in a $5\lambda/2$ CdTe-based cavity one can achieve the Rabi-splitting of about 3-4 meV, which can be experimentally observed. No spectral features associated to X1 excitons would be seen in this case. Further enhancement of the Rabi-splitting will require the use of new materials having enhanced exciton oscillator strength (GaN, ZnSe or ZnO, for example). In any case, Bose-Einstein condensation of X1 excitons is expected to take place at the liquid Helium temperature.

In Section II of this paper we derive the reflection and transmission coefficients of a WQW in the vicinity of the X2 exciton-resonance. In Section III we use these coefficients to obtain the eigen-energies of the exciton-polariton modes of microcavities with WQWs at the nodes of the confined light mode. In Section IV we present the results of

numerical calculations of the reflectivity of such microcavities and conclude.

2. REFLECTION AND TRANSMISSION OF LIGHT BY WIDE QUANTUM WELLS IN THE VICINITY OF AN ODD EXCITON RESONANCE

Let us consider a light wave propagating in a dielectric medium homogeneous in the plane of the wave (xy -plane), but possibly inhomogeneous in the propagation direction (z -direction). The electric \vec{E} and magnetic \vec{B} fields of the wave are given by Maxwell's equations:

$$\begin{aligned} \text{curl } \vec{E} &= -\frac{1}{c} \frac{\partial \vec{B}}{\partial t}; \\ \text{div } \vec{B} &= 0; \\ \text{curl } \vec{B} &= \frac{1}{c} \frac{\partial \vec{D}}{\partial t}; \\ \text{div } \vec{D} &= 0; \end{aligned} \quad (1)$$

where $\vec{D} = \vec{E} + 4\pi\vec{P}$, and \vec{P} is the dielectric polarisation vector. Consider a QW parallel to the plane xy and characterised by an exciton resonance frequency ω_0 . The displacement field near this frequency can be written as:

$$\vec{D} = \varepsilon_B \vec{E} + 4\pi\vec{P}_{exc}, \quad (2)$$

where \vec{P}_{exc} is the excitonic contribution to the dielectric polarisation and ε_B is the background dielectric constant, which is assumed to be the same in the QW and surrounding barriers for simplicity. Eqs. (1) and (2) yield:

$$-\frac{\partial^2 \vec{E}}{\partial z^2} = k_0^2 (\varepsilon_B \vec{E} + 4\pi\vec{P}_{exc}(z)), \quad (3)$$

where $k_0 = \omega_0/c$ is the wave-vector of light in a vacuum.

The exciton-induced dielectric polarisation can be written in the form [1,12]:

$$4\pi\vec{P}_{exc}(z) = \int_{-\infty}^{\infty} \chi(z, z') \vec{E}(z') dz', \quad (4)$$

where

$$\chi(z, z') = \tilde{\chi}(\omega) \Phi(z) \Phi(z'), \quad (5)$$

with

$$\tilde{\chi}(\omega) = \frac{Q}{\omega_0 - \omega - i\gamma}, \quad Q = \varepsilon_B \omega_{LT} \pi a_B^3.$$

Here, $\Phi(z)$ is the exciton center of mass wave-function, ω is the frequency of the incident light, γ is the *homogeneous* broadening of the exciton resonance caused by acoustic phonons, and ω_{LT} and a_B are exciton *longitudinal-transverse* splitting and *Bohr radius*.

Once the polarisation (4) is introduced, Eq. (3) becomes an integro-differential equation and can be solved exactly by using of the Green's function method. Within this method the solution of Eq. (3) is represented in the form

$$E(z) = E_0 \exp(ikz) + k_0^2 \int dz' 4\pi P_{exc}(z') G(z - z') \quad (6)$$

where E_0 is the amplitude of the incident light, and the Green's function G satisfies the equation

$$\left(\frac{\partial^2}{\partial z^2} + k^2 \right) G(z) = -\delta(z), \quad k = \sqrt{\epsilon_B} k_0, \quad (7)$$

Bearing in mind that

$$\int_{-\infty}^{\infty} dz f(z') \delta(z - z') = f(z), \quad \text{one can easily}$$

check that G is given by

$$G(z) = \frac{i \exp(ik|z|)}{2k}. \quad (8)$$

Eq. (6) can be solved with respect to $E(z)$ as it is contained both in the left-hand side and right-hand side (in the polarisation P). In order to do it, let us multiply both parts by $\Phi(z)$ and integrate over z . This procedure yields:

$$\int dz E \Phi(z) = E_0 \int dz \phi(z) \exp(ikz) + k_0^2 \tilde{\chi} \times \int \int dz dz' \phi(z) \phi(z') G(z - z') \int dz' E \phi(z''), \quad (9)$$

which means that

$$\int dz E \Phi(z) = \frac{E_0 \int dz \Phi(z) \exp(ikz)}{1 - k_0^2 \tilde{\chi} \int \int dz dz' \Phi(z') \Phi(z) G(z - z')} \quad (10)$$

We now return to Eq. (6) and substitute Eq. (10) into its right-hand side:

$$\int dz \int dz' e^{ik|z-z'|} \Phi(z) \Phi(z') = \left[\int dz \Phi(z) \sin(kz) \right]^2 + i \int \int dz dz' \Phi(z) \Phi(z') \sin k|z - z'| \quad (10)$$

$$E = E_0 \exp(ikz) + k_0^2 \tilde{\chi} \int dz' \Phi(z') G(z - z') \int dz'' E(z'') \Phi(z'') \\ = E_0 \left[e^{ikz} + \frac{k_0^2 \tilde{\chi} \int dz' \Phi(z') G(z - z') \int e^{ikz''} \Phi(z'') dz''}{1 - k_0^2 \tilde{\chi} \int \int dz dz' G(z - z') \Phi(z) \Phi(z')} \right] \quad (11)$$

Using Eq. (8) we finally obtain

$$E(z) = E_0 e^{ikz} + \frac{\frac{ik_0}{2\sqrt{\epsilon_B}} Q E_0 \int \Phi(z'') e^{ikz''} dz'' \int \Phi(z') e^{ik|z-z'|} dz'}{\omega_0 - \omega - i\gamma - Q \frac{ik_0}{2\sqrt{\epsilon_B}} \int \int dz' dz'' e^{ik|z'-z''|} \Phi(z') \Phi(z'')} \quad (12)$$

The amplitude reflection (r) and transmission (t) coefficients of the QW can then be obtained as:

$$r \equiv \frac{E(z) - E_0(z) e^{ikz}}{E_0(z) e^{-ikz}} \Big|_{z \rightarrow -\infty}, \\ t \equiv \frac{E(z)}{E_0 e^{ikz}} \Big|_{z \rightarrow \infty}. \quad (13)$$

In the case of X2 exciton, $\Phi(z)$ is an odd function, $\Phi(-z) = -\Phi(z)$, thus the integrals on the right-hand side of Eq. (12) can be easily simplified. If $z \rightarrow +\infty$:

$$\int dz' \Phi(z') e^{ik|z-z'|} = -ie^{ikz} \int dz' \sin(kz') \Phi(z')$$

;

and in the case $z \rightarrow -\infty$:

$$\int dz' \Phi(z') e^{ik|z-z'|} = ie^{-ikz} \int dz' \sin(kz') \Phi(z')$$

This allows us to obtain the reflection and transmission coefficients of the WQW in a simple form:

$$r(\omega) = -\frac{i\Gamma_0}{\tilde{\omega}_0 - \omega - i(\Gamma_0 + \gamma)}, \quad (14)$$

$$t(\omega) = 1 - r(\omega), \quad (15)$$

$$\Gamma_0 = \frac{Qk_0}{2\sqrt{\epsilon_B}} \left[\int \Phi(z) \sin kz dz \right]^2 \quad (16)$$

is the same as in the case of X1 exciton [12], except that there is sine under integral instead of the cosine, and

$$\tilde{\omega}_0 = \omega_0 + \frac{Qk_0}{2\sqrt{\epsilon_B}} \int \int dz dz' \Phi(z) \Phi(z') \sin k|z - z'| \quad (17)$$

is the renormalisation of the exciton resonance frequency due to the polariton effect, which is formally the same as in the X1 case. Let us note at this point that for the X1 exciton state the reflection coefficient is given by the expression in the right part of Eq. (14) taken with the positive sign, while $t(\omega) = 1 + r(\omega)$ [1,12].

3. DISPERSION OF EXCITON-POLARITONS FORMED BY X2 EXCITONS IN MICROCAVITIES

A Bragg mirror is a periodic structure composed of pairs of layers of dielectric or semiconductor materials characterised by different refractive indices (say n_a and n_b). The thicknesses of the layers (a and b , respectively) are chosen so that

$$n_a a = n_b b \equiv \bar{\lambda}/4. \quad (18)$$

Condition (18) is usually called the *Bragg interference condition*. The wavelength of light $\bar{\lambda}$ marks the centre of the *stop-band* of the mirror, i.e. the band of the wavelengths for which the reflectivity of the mirror is close to unity. In the following we assume $n_a < n_b$.

We shall describe the optical properties of the mirror within its stop-band using the transfer matrix approach.

At the central frequency of the stop-band, given by

$$\bar{\omega} = \frac{2\pi c}{\bar{\lambda}}, \quad (19)$$

one can derive a simple and useful expression for the reflection coefficient of a semi-infinite Bragg-

$$\text{mirror: } r_B = \frac{n_0 \left(\frac{n_A}{n_B} - \frac{n_B}{n_A} \right) - i(n_A + n_B)x}{n_0 \left(\frac{n_A}{n_B} - \frac{n_B}{n_A} \right) + i(n_A + n_B)x} = \exp \left(i \frac{n_A n_B \bar{\lambda}}{2n_0(n_B - n_A)C} ((\omega - \bar{\omega})) \right), \quad (20)$$

where n_0 is the refractive index of the media from where light is incident. The coefficient

$$L_{DBR} \equiv \frac{n_A n_B \bar{\lambda}}{2(n_B - n_A)} \quad (21)$$

is frequently called the *effective length* of a Bragg mirror.

Consider a symmetric $\lambda/2$ microcavity with a single QW embedded in the centre. In the basis of amplitudes of light waves propagating in positive and negative directions along the z -axis, the transfer matrix across the QW has the

$$\hat{T}_{QW} = \frac{1}{t} \begin{bmatrix} t^2 - r^2 & r \\ -r & 1 \end{bmatrix}, \quad (22)$$

where r and t are the angle- and polarisation-dependent amplitude reflection and transmission coefficients of the QW derived above.

The transfer matrix across the cavity from one Bragg mirror to the other is the product:

$$\hat{T}_c = \begin{bmatrix} e^{ikL_c/2} & 0 \\ 0 & e^{-ikL_c/2} \end{bmatrix} \frac{1}{t} \begin{bmatrix} t^2 - r^2 & r \\ -r & 1 \end{bmatrix} \begin{bmatrix} e^{ikL_c/2} & 0 \\ 0 & e^{-ikL_c/2} \end{bmatrix}, \quad (23)$$

where L_c is the cavity width. The elements of

$$T_{11}^c = \frac{t^2 - r^2}{t} e^{ikL_c}, \quad T_{12}^c = \frac{r}{t}, \quad T_{21}^c = -\frac{r}{t}, \quad T_{22}^c = \frac{1}{t} e^{-ikL_c}. \quad (24)$$

To find the eigenfrequencies of the exciton-polariton modes of the microcavity, one should search for non-trivial solutions of Maxwell's equations under the requirement of no light incident on the cavity from outside.

$$\hat{T}_c \begin{bmatrix} r_B \\ 1 \end{bmatrix} = A \begin{bmatrix} 1 \\ r_B \end{bmatrix}, \quad (25)$$

where r_B is the reflection coefficient of the Bragg mirrors for light incident from inside the cavity. It is angle-dependent, in general. At normal incidence, it can be conveniently approximated by Eq. (20). Excluding the coefficient A from Eq. (25), we obtain the

$$\frac{T_{21}^c r_B + T_{22}^c}{T_{12}^c + T_{11}^c r_B} = r_B. \quad (26)$$

This is already a dispersion equation because the coefficients of the transfer matrix and r_B are dependent on the in-plane wave-vector of light. Substituting into Eq. (26) the coefficients

(24), one can represent the equation for the \bar{r}

$$\left[(t+r) r_B e^{ikL_c} - 1 \right] \left[(t-r) r_B e^{ikL_c} + 1 \right] = 0 \quad (27)$$

The zeros of the first bracket describe the eigenstates of exciton-polaritons resulting from coupling of *even* optical modes with the exciton ground state. In our case $t+r=1$, consequently, the even cavity modes are not coupled with the exciton state we consider (which is quite natural). Note also that if an odd cavity mode is in the resonance with the X2 exciton, the nearest even cavity mode is very far from the X1 exciton resonance, which remains decoupled from light (“dark”) therefore. Solutions of Eq. (27), coming from zeros of the second bracket on the left-hand side represent *odd* eigen-modes of the microcavity. They result from coupling of the odd photon modes with the antisymmetric exciton state. Further we consider only them.

At normal incidence we take

$$r_B \approx \bar{r} \left(1 + iL_{DBR} \frac{n_c}{c} (\omega - \omega_c) \right), \quad (28)$$

where \bar{r} is close to 1, and assume $e^{ikL_c} \approx -1 - i \frac{\omega - \omega_c}{c} n_c L_c$. We obtain, using the explicit form for the reflection

$$\begin{aligned} & \bar{r} \left(1 + iL_{DBR} \frac{n_c}{c} (\omega_0 - \omega) \right) (\omega_0 - \omega - i(\gamma - \Gamma_0)) \\ & \times \left(1 + i \frac{\omega - \omega_c}{c} n_c L_c \right) \\ & = \omega_0 - \omega - i(\gamma - \Gamma_0) \end{aligned}$$

which finally yields, after algebraic

$$(\tilde{\omega}_0 - \omega - i\gamma)(\omega_c - \omega - i\gamma_c) \approx V^2, \quad (29)$$

Where

$$\begin{aligned} \gamma_c &= \frac{1 - \bar{r}}{\bar{r} \frac{n_c}{c} (L_{DBR} + L_c)} \\ V^2 &= \frac{1 + \bar{r}}{\bar{r}} \frac{\Gamma_0 c}{n_c (L_{DBR} + L_c)}. \end{aligned} \quad (30)$$

Here, quadratic terms in $(\omega_c - \omega)$ are omitted. In all further calculations we shall assume $\tilde{\omega}_0 = \omega_0$ for simplicity. Eq. (29) is an equation for eigen states of a system of two coupled harmonic oscillators, namely, the X2 exciton

resonance and the cavity mode. Note, that in this form Eq. (29) is formally identical to the well-known equation for the eigen-modes of a λ - microcavity having an X1 exciton coupled to light. That solutions come from zeros of the first bracket of Eq. (27). The Rabi-splitting of the odd modes in general is less than the splitting of the even modes because the radiative decay rate Γ_0 is an order of magnitude less for X2 than for X1 states.

Generalization for the case of a multiple quantum well system inside the microcavity is straightforward: one simply has to replace in Eq. (27) the reflection and transmission coefficients of a single QW (r and t) by corresponding coefficients of the multiple quantum well structure. Clearly, Rabi-splitting can be enhanced by putting multiple WQWs at the nodes of the cavity light mode.

4. NUMERICAL MODELLING OF THE MICROCAVITIES WITH AN INVERSED EXCITONIC SPECTRUM AND CONCLUSIONS.

In modelling we shall consider a $5\lambda/2$ -cavity with 5 WQWs placed at the nodes of the cavity field. We have taken $\hbar\Gamma_0 = 0.004$ meV for X2 excitons and $\hbar\Gamma_0 = 0.026$ meV for X1 excitons, which corresponds to CdTe/CdMnTe QWs a bit wider than those of Ref. [3]. We have taken the splitting of 2 meV between X1 and X2 states, which corresponds to quite large wells (of about 20 nm width). This parameter was taken so small in order to achieve X1 resonance lying between lower and upper X2 polariton modes. It has no impact on the reflectivity spectra as X1 state is fully decoupled from the cavity mode both at normal and oblique incidence. We supposed that at normal incidence the bare photon mode of the cavity lies 2 meV lower than the bare X2 exciton resonance. Eigen polariton modes of this more complex system are still given by Eq. (29) where the parameter Γ_0 (for X2) should be multiplied by 5 reflecting the number of QWs. Figure 1 shows the reflectivity spectra of the structure. One can clearly see the anticrossing between X2 exciton and the cavity mode. No spectral feature is associated with the “dark” X1 exciton, while we have taken it into account in the numerical computation. The dispersion of these dark excitons remain parabolic with the excitonic effective mass. At normal incidence, the ground exciton state is no more the ground state of the

system, it is situated above the lower X2 polariton mode. The Rabi splitting of X2 polariton branches is relatively weak as the important parameter Γ_0 is approximately 7 times smaller for X2 exciton than for X1 exciton even in wide QWs.

In conclusion, embedding WQWs at the nodes of the electric field of the cavity mode in microcavities, one can still see the polariton anticrossing and vacuum field Rabi splitting while it is expected to be about $\sqrt{7}$ times smaller than if the QWs are placed at the antinodes of the field. The interest to put the wells at the nodes is to decouple the ground X1 exciton state from light. This may lead to the Purcell effect i.e. significant increase of the exciton radiative life-time which is favorable for the Bose-Einstein condensation of ehh1 excitons. We expect that the condensates of X1 excitons will be slightly coupled to light anyway because the polariton-polariton scattering which would mix them with the polariton modes. This scattering cannot be very efficient because it is parity-forbidden in the first order. All this makes us to expect very peculiar non-linear emission spectra from microcavities having the QWs at the nodes of the field. Inversion of the excitonic spectrum would make the dynamics of energy relaxation of exciton polaritons in the cavities we consider

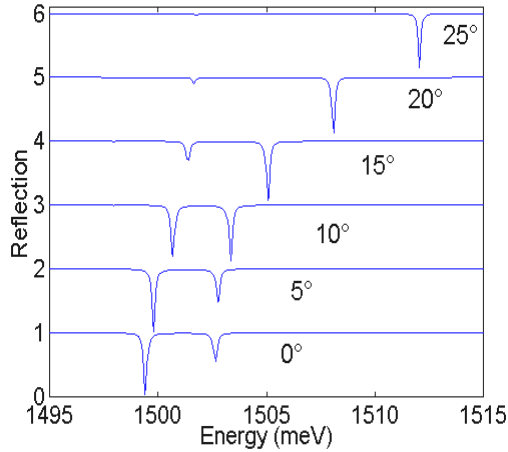


FIG. 1: Calculated reflectivity spectra of the model $5\lambda/2$ microcavity containing 5 WQWs embedded at the *nodes* of the effective. The detuning between X2 exciton and cavity modes is -2 meV at zero angle. The X1 exciton is situated at 1500 meV (invisible in the spectra)

strongly different from their dynamics in conventional microcavities. We hope that unusual properties of the unusual microcavities theoretically described here will attract attention of experimentalists who will check our predictions.

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