Strong and weak light-matter coupling in microcavity-embedded double quantum wells

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We demonstrate an efficient switching between strong and weak exciton-photon coupling regimes in microcavity-embedded asymmetric double quantum wells, controlled by an applied electric field. We show that a fine tuning of the electric field leads to drastic changes in the polariton properties, with the polariton ground state being red-shifted by a few meV and having acquired prominent features of a spatially indirect dipolar exciton. We study the properties of dipolar exciton polaritons, called dipolaritons, on a microscopic level and show that, unlike recent findings, they cannot be dark polaritons but are rather mixed states with comparable contribution of the cavity photon, bright direct, and dark indirect exciton modes.

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Semiconductor microcavities are well known due to their importance in fundamental physics and have a wide range of various applications [1]. Of particular interest are exciton polaritons created in microcavities in the regime of strong light-matter coupling [2]. The demonstrated possibilities of polariton condensation [3, 4] and room-temperature lasing [5, 6] have made microcavity polaritons the subject of intensive studies. If a semiconductor quantum well (QW) is placed in the antinode position of the resonant electro-magnetic field inside the microcavity, Coulomb bound electrons and holes localized in the QW layer can strongly interact with a high-quality cavity mode (CM) and form mixed exciton-photon states called polaritons. Double QWs have attracted much attention in recent years due to formation of long-living spatially indirect excitons (IXs) in such structures in the presence of an applied electric field (EF) [8]. Intensive studies of IXs in double QWs have resulted in demonstration of their electrostatic [9, 10] and optical control [11] as well as in engineering of the dipolar exciton-exciton interaction [12, 13] necessary for exploring different many-body effects, including an intriguing possibility of Bose-Einstein condensation of excitons [8, 9, 15, 17].

Very recently, asymmetric double quantum wells (ADQWs) have been embedded in a planar Bragg-mirror microcavity [18] in order to create a special type of polariton with a static dipole moment enhanced at the resonant tunneling condition [19]. Indeed, at the electron tunneling resonance, the asymmetry in the conduction band potential of the ADQW is compensated by an applied EF, resulting in a formation of resonant symmetric and antisymmetric electron states while keeping the hole state asymmetric. The electron and the hole are then bound together to form either a direct or an indirect exciton. When this structure is embedded into the cavity, the direct exciton (DX) strongly couples to the CM and creates a polariton. The IX itself does not form a polariton, as it has a much smaller oscillator strength, but it is electronically coupled to the DX via the tunneling across the barrier. A resulting mixed state of such a three-level system is a polariton that acquires a large electric dipole moment, typical for IX. This allows one to reinforce and control, both electrically and optically, the polariton-polariton interaction. These new hybrid quasi-particles called dipolaritons were introduced in Refs. 10 and 20 and have already been suggested for observation of superradiant terahertz (THz) emission [21], continuous THz lasing [22] and polariton bistability [23].

It has also been claimed [20, 24] that for zero detuning between IX and CM, the middle dipolariton state which in the case of zero detuning yields a strictly dark polariton. While this result sounds rather counterintuitive (indeed, the coupling is proportional to the exciton “brightness”), the argumentation which could support this conclusion is similar to that used in atomic physics in order to describe a dark polariton in a three-level Λ-system [25, 26] and to demonstrate the electromagnetically induced transparency and slowing of light propagation. Assuming an impenetrable barrier separating the QWs, one can calculate “pure” DX and IX states, with the IX and CM completely uncoupled due to the vanishing electron-hole overlap integral. By lowering the potential barrier between the wells both exciton states are modified, owing to the tunneling through the barrier. By treating tunnel coupling as a perturbation and keeping only two exciton states (the bare DX and IX) in the basis, a three-level Hamiltonian can then be obtained which in the case of zero detuning yields a strictly dark polariton state, as shown in Ref. 20.

We believe, however, that this last result is incorrect and the concept of dark polariton is misleading in the context of dipolaritons. On a qualitative level, exciton states in an ADQW structure are the eigenstates of this electronic system and thus do not couple to each other. Modifying the system by changing the potential barrier or applied EF cannot alter this fundamental property of the eigenstates. On the other hand, it is well known that
the coupling of an exciton to a photonic mode is proportional to its oscillator strength, so obviously a dark exciton produces no polariton. From the formal mathematical point of view, the dark polariton obtained in Ref. [20] is just an artifact of the basis truncation and reduction of the full system to the three-level model. In fact, the dark polariton state is obtained when the DX-IX coupling is large, i.e. comparable to the DX-CM coupling, otherwise a negligible contribution of the CM leaves the IX mode decoupled [20]. On the other hand, large DX-IX tunnel coupling means strong intrinsic perturbation of the electronic system which significantly affects the whole spectrum of exciton states. Therefore, the basis of only two bare DX and IX states is insufficient for the correct treatment of the system and makes the three-level model inadequate. The reason why the concept of dark polaritons is valid in atomic Λ-systems is that the dark and bright levels are coupled in that case by an extrinsic perturbation. This can be a resonant laser source or interactions which flip the electron spin. Such perturbations can resonantly couple the bright and dark states leaving all other states of the spectrum untouched.

The purpose of this Letter is twofold. One is to demonstrate an efficient mechanism of switching on/off the exciton-photon strong coupling regime in cavity-embedded ADQWs and EF-control of their polariton properties. We calculate the optical reflection and absorption spectra and analyze them in terms of the exciton-CM coupling and DX-IX crossover [27] and show that the results of our calculation are in quantitative agreement with recent experimental observations [18–20].

The other goal is to study dipolaritons on a microscopic level by calculating the full spectrum of exciton states in an ADQW and their coupling to the cavity photon. In this calculation, the polariton brightness and static dipole moment are deduced from the microscopic optical polarization. The relative fractions of the DX, IX, and CM are calculated and analyzed, showing in particular that no dark polariton can be observed in such systems.

We concentrate on a microcavity structure used in experiments [18,20], which consists of four InGaAs ADQWs placed at the antinodes of the electromagnetic field inside a 5λ/2 cavity sandwiched between 17 and 21 pairs of GaAs/AlAs distributed Bragg reflectors (DBRs), see Fig. 1. Each ADQW contains two 10-nm InGaAs-GaAs QW layers with the In content of 8% and 10% separated by a 4-nm GaAs barrier. To study the light-matter strong coupling in this system, we solve coupled Maxwell’s and material equations for the microscopic excitonic polarization $P(z_e, z_h, \rho)$ and the local optical electric field $\mathcal{E}(\omega, k; z)$, using the Green’s function approach [28–31]. Technically, we expand the polarization into the complete set of exciton eigenfunctions:

$$P(z_e, z_h, \rho) = |d_{cv}|^2 \sum_\nu \Psi_\nu(z_e, z_h, \rho) X_\nu(\omega, k)$$

The expansion coefficients are then given by

$$X_\nu(\omega, k) = \int \mathcal{E}(\omega, k; z) \Psi_\nu(z, z, 0) dz$$

and are found using the local electric field $\mathcal{E}$. The latter in turn satisfies Maxwell’s integro-differential equation which includes the macroscopic excitonic polarization $P(z, 0)$ bringing into the system a nonlocal optical susceptibility [31]. Here $z_e$ are $z_h$ are the electron and hole coordinates in the growth direction of the ADQW, $\rho$ is the coordinate of the electron-hole in-plane relative motion, $d_{cv}$ is the matrix element of the microscopic dipole moment between the valence and conduction bands, $M$ and $k$ are the in-plane exciton effective mass and wavevector, and $\omega$ is the frequency of the s-polarized electro-magnetic field. To take into account the homogeneous broadening of the excitonic system, a phenomenological damping constant $\gamma$ is added by hand [32].

In the calculation of the optical polarization Eq. (1) we keep up to 200 exciton states and for each state $\nu$ we calculate its energy $E_\nu$ and the energy wave function $\Psi_\nu(z_e, z_h, \rho)$, expanding the latter into electron-hole pair states, localized in the ADQW, and solving a radial matrix Schrödinger equation in real space [27]. The in-plane excitonic continuum is discretized by introducing a rigid wall at $|\rho| = R$ with $R = 800$ nm used everywhere except the last two figures of this Letter. The Maxwell equation is then solved in a strict manner, by reducing it to an effective matrix Fredholm problem with a factorizable kernel incorporated into the scattering matrix method [33, 34].

Figure 2 shows the reflectivity spectra for different values of the static EF $F$ applied in the growth direction and for different detuning between the CM (dashed vertical lines) and bare exciton modes (red circles). For small detuning, the three brightest exciton modes shown in Fig. 2(a), namely the exciton ground state (GS) and two DX excited states ($1s$ and $2s$ modes) [35] are all strongly coupled to the cavity. This is obvious from multiple anticrossings of polariton states seen as dips in the reflection. As a result of these anticrossings, the spectral positions of the polaritons strongly depend on the
EF. In particular, the lowest mode demonstrates a red shift by a few meV, following the exciton GS which in turn experiences a direct-to-indirect crossover \( \theta = 19 \text { kV/cm} \). In the case of a larger detuning shown in Fig. 2(b), which can be achieved e.g. by changing the cavity widths or the angle of light incidence \( \theta \), bright exciton states, i.e. those having a considerable contribution of the DX, are still strongly coupled to the CM. However, the polariton properties change abruptly with the EF – see the on/off switching of the strong coupling at around \( F = 19 \text { kV/cm} \) in Fig. 2(b).

The spectra shown in Fig. 2 are in good agreement with the measured reflection \( \theta = 35^\circ \). To reach a quantitative agreement with the data in Ref. 20, we have calculated the absorption spectra, making a 2 meV full width at half maximum Gaussian convolution of the excitonic-induced susceptibility, in order to include the actual effect of inhomogeneous broadening of the exciton lines \( \Delta \). The result is shown in Fig. 3 for two different angles of incidence. At normal incidence, the CM is weakly coupled to ADQWs as demonstrated by a narrow peak in the absorption. At \( \theta = 35^\circ \), one can clearly see three wide polariton bands corresponding to the three polariton modes analyzed in Ref. 21 with the help of the above mentioned three-level model. Apart from the missing temperature-dependent state occupation factor and a rigid shift of all spectra by 30 meV, a quantitative agreement with the measured photoluminescence (PL) \( \theta = 35^\circ \) is achieved that demonstrates the quality of our microscopic approach and justifies our microscopic analysis of dipolariton states given below.

To study the dipolaritons we switch off the inhomogeneous broadening in our calculation and reduce the in-plane confinement radius to \( R = 200 \text { nm} \), which leads to a less fine discretization of the continuum. The result for \( \theta = 35^\circ \) is seen in Fig. 3 as a series of peaks in the absorption spectrum [similar to Fig. 3(b)] – polariton states, indicated by black lines. Concentrating on a few lowest polariton states, we estimate the fraction of DX, IX, and CM in each of them by evaluating the polariton bands corresponding to the three polariton modes \( \theta = 35^\circ \), with the help of the above mentioned three-level model. Apart from the missing temperature-dependent state occupation factor and a rigid shift of all spectra by 30 meV, a quantitative agreement with the measured photoluminescence (PL) \( \theta = 35^\circ \) is achieved that demonstrates the quality of our microscopic approach and justifies our microscopic analysis of dipolariton states given below.

\[
F = N^{-1} \left| \int P(z, z, 0) dz \right|^2, \tag{3}
\]

\[
D = N^{-1} \int \int |P(z_e, z_h, \rho)|^2 (z_e - z_h) d\rho dz_e dz_h, \tag{4}
\]

where \( N \) is a normalization constant. The relationship between the DX, IX, and CM components \( (C_{DX}, C_{IX}, \text{and } C_{CM}) \) respectively) is given by

\[
(C_{IX}/C_X)^2 \approx \alpha D \quad (C_{CM}/C_X)^2 \approx \beta F, \tag{5}
\]
where $C_X^2 = C_{DX}^2 + C_{IX}^2$. The coefficients of proportionality are estimated to be $\alpha = 1/d$, where $d$ is the center-to-center distance between QWs in the ADQW, and $\beta = e^2|d_{cv}|^2\epsilon_b(\tilde{E}/\tilde{P})^2/(2\pi E_{CM}d)$, following from the standard expression for the polariton Rabi splitting, where $E_{CM}$ is CM energy, $\epsilon_b$ is the background dielectric constant, and $\tilde{E}$ and $\tilde{P}$ are the mean values of the local electric field and polarization inside the ADQW: $\tilde{P}^2d \approx \int |P(z, z, 0)|^2dz$. Normalized fractions $C_{DX}^2$, $C_{IX}^2$, and $C_{CM}^2$ are then calculated by taking the values of $F$, $D$, and $N$ at the polariton frequencies.

The relative contribution of DX, IX, and CM to the six lowest polariton states is shown in Fig. 5. The ground state ($n = 1$) has a small CM component – the corresponding exciton state is detuned from the CM – and demonstrates a DX-IX crossover typical for the exciton GS. Higher exciton states producing polariton modes $n = 2$ to 6 are resonant to the CM and thus for some values of the EF demonstrate a considerable photon contribution. At the same time, they have hybrid DX-IX properties as is also clear from Fig. 5. The maximum dipolariton effect is achieved for $n = 3$ and $n = 5$ states at around $F = 12$ kV/cm when the contribution of all three components is nearly the same ($\sim 1/3$). However, increasing the IX component and consequently the polariton static dipole moment always leads to decreasing photon contribution, due to the decreased coupling to the CM. In other words, an appreciable static dipole moment of the polariton is achievable only at the cost of a good balance between bright DX and dark IX.

In conclusion, we have demonstrated theoretically, on the microscopic level, the existence of hybrid exciton polariton states with a large static dipole moment, called dipolaritons. These have comparable contribution of spatially indirect exciton, optically bright direct exciton and photon cavity mode. We have also shown that no dark polaritons are created in microcavity-embedded asymmetric coupled quantum wells, but such states are just an artifact of an oversimplified three-level model. The reflectivity and absorption spectra calculated at different angles of incoming light are in quantitative agreement with experimental observations and demonstrate an efficient on/off switching of the strong coupling regime by means of adjusting the electric field.

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