Verification of very strong coupling in a semiconductor optical microcavity

Ming-Jay Yang1, Na Young Kim1, Yoshihisa Yamamoto2,3 and Neil Na1

1Institute of Photonics Technologies, National Tsing-Hua University, Hsinchu 30013, Taiwan
2Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305, USA
3National Institute of Informatics, Hitotsubashi, Chiyoda-ku, Tokyo 101-8430, Japan
E-mail: ycn@ee.nthu.edu.tw

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Abstract
We study the very strong coupling effect in a semiconductor optical microcavity in which the exciton radius is dramatically modified due to the presence of strong photon–exciton coupling by means of nonlinear numerical optimization. To experimentally verify the features of very strong coupling and distinguish them from those of strong coupling, we propose two schemes and show that our proposals can provide unequivocal experimental proof of the existence of very strong coupling.

Introduction

A semiconductor optical microcavity (MC) provides a unique platform for studying the coupling between MC photons and quantum well (QW) excitons. If the photon–exciton coupling constant is smaller than the linewidths of MC photons and QW excitons, a weak coupling occurs and manifests itself as an enhancement of exciton decay rate [1]. When the coupling constant is larger than the linewidths of MC photons and QW excitons, new eigenmodes of the strongly-coupled cavity quantum electrodynamics (QED) system are formed and often referred to as exciton-polaritons [2]. In this field, numerous experimental demonstrations of the coherent properties of exciton-polaritons have been made, such as Bose–Einstein condensation [3], superfluidity [4] and vortices [5]. If the coupling constant can be further enlarged to be of the same order of magnitude as the exciton binding energy, the system enters the so-called very strong coupling (VSC) regime [6]. In the VSC regime, it is theoretically predicted that the properties of exciton-polaritons are strongly modified, including exciton radius reduction/enlargement, unequal photon and exciton fractions at resonance, and strong asymmetric Rabi splitting between the upper polariton (UP) and lower polariton (LP) [6, 7]. In addition, because the effect of VSC increases the thermal dissociation temperature and nonlinear saturation density of LPs, it may be possible to construct a room temperature, ultralow threshold polariton laser using a GaAs-based optical MC if VSC can be achieved [7]. Compared to the cases of using wide bandgap semiconductors such as GaN, ZnO etc as MC material, this approach should feature significantly less inhomogeneous broadening and may be applicable to optical communication systems at near-infrared wavelengths. Nevertheless, despite the fact that a Rabi splitting larger than the exciton binding energy has been observed experimentally [8], the existence of VSC remains elusive for various reasons, which will be discussed later.

In this paper, we first review the analytical variational approach used in [6, 7], and then expand the theoretical concept by utilizing a nonlinear numerical optimization technique. Such a technique allows us to produce the exact shape of the exciton wavefunction in the relative coordinate for both UP and LP energy branches, even when an arbitrary potential seen by the exciton wavefunction in the relative coordinate is added. By means of this technique, we can further examine the problem when an external in-plane dc electrical field is applied to the exciton-polaritons, in which the excitonic dipole moment and the coupling constant are reduced so that the bare cavity energy is revealed. The value of the recovered bare cavity energy depends highly on whether photons and excitons are strongly coupled or very strongly coupled to each other, which serves as an experimental proof of the existence of VSC. In addition, we examine the problem when an external vertical dc
magnetic field is applied to the exciton–polaritons. Due to the dependence of exciton radius on the coupling constant in the VSC regime, the UP and LP manifest themselves in different diamagnetic energy blue-shifts that serve as another experimental proof of the existence of VSC. Detailed analyses on these two phenomena in a GaAs-based optical MC are presented with realistic experimental parameters.

**Formalism**

The exciton–polariton state can be written as \( |\Psi_{\text{pol}}\rangle = \alpha |\Phi_{\text{ex}}\rangle + \beta |\Phi_{\text{pol}}\rangle \) where the Hopfield coefficients \( \alpha \) and \( \beta \) are for QW exciton state \( |\Phi_{\text{ex}}\rangle \) and MC photon state \( |\Phi_{\text{pol}}\rangle \) respectively. \( \Phi_{\text{ex}} \) is the transverse wavevector and \( \Phi_{\text{pol}} \) is the longitudinal wavevector. The system Hamiltonian can be written as

\[
\hat{H} = -\frac{\hbar^2 \nabla_r^2}{2\mu} + \frac{-\hbar^2 \nabla_\Phi^2}{2\mu} + \frac{-e^2}{4\varepsilon_\varepsilon_0 r} + \hbar \omega \hat{a}_{\Phi_{\text{ex}}} \hat{a}_{\Phi_{\text{ex}}} + \frac{-e^2}{2\varepsilon_\varepsilon_0 \nu} \sqrt{\frac{\hbar \omega}{2\varepsilon_\varepsilon_0 \nu}} \left( \hat{a}_{\Phi_{\text{ex}}} + \hat{a}_{\Phi_{\text{pol}}} \right).
\] (1)

The first two terms are the kinetic energies including the relative motion and the center-of-mass motion of an exciton. The third term is the Coulomb interaction between an electron and a hole, and the fourth term is the MC photon energy \( \hbar \omega = \hbar \left( K_{\Phi_{\text{pol}}}^2 + K_{\Phi_{\text{pol}}}^2 \right)^{1/2} \) where \( K_{\Phi} \) is the discretized wavevector due to an optical confinement in the \( z \) direction. The last term is the coupling between the QW exciton and the MC photon and is proportional to the coupling constant \( g = \left( \hbar^2 \varepsilon_0^2 / 4 \varepsilon_\varepsilon_0 m_L \right)^{1/2} \) considering a two-dimensional (2D) exciton. The 2D exciton oscillator strength \( f_{2D} \) is linearly proportional to \( \nu \sqrt{\nu} \), i.e., the probability of locating an electron and a hole in the same position. \( L_z \) is the cavity effective length in the \( z \) direction. For most of the MCs in the literature, \( g \) is much smaller than the exciton binding energy \( \hbar \omega_{\text{exc}} \), so the photon–exciton coupling is treated as a perturbation to the total Hamiltonian. However, in the case of many QWs embedded in a high quality DBR MC, \( g \) and \( \hbar \omega_{\text{pol}} \) can be at the same order of magnitude which invalidates this assumption. The photon–exciton coupling and the electron–hole Coulomb interaction should be then treated on an equal footing, and the influence of this VSC effect was previously investigated by using a variational method with \( \alpha \) and \( \lambda \) as variational parameters, and \( \varphi (r) = (2/\pi)^{1/2} \left( \lambda / a_0 \right) \exp \left( -\lambda r / a_0 \right) \) as a trial exciton wavefunction [6, 7]. \( \lambda \) and \( a_0 \) are the exciton radius reduction factor and the 2D exciton Bohr radius respectively. It was predicted that the exciton wavefunction in the relative coordinate experiences a substantial radial change for both polariton energy branches in the regime of VSC, and the resulting coupling constant \( g \) becomes nonlinear and can be written as

\[
g = g_0 \frac{\varphi (0)}{\varphi_0 (0)}.
\] (2)

The subscript \( g \) means \( \lambda \) is taken as unity, i.e., when the coupling constant is much smaller than the exciton binding energy. Another way to understand the effect of VSC is to write down the 2 by 2 matrix

\[
H = \begin{pmatrix} -E_B & g_0 \\ g_0 & -E_B + \Delta - \frac{g_0^2}{E_B} \end{pmatrix}
\] (3)

which describes the effective system Hamiltonian [7] assuming the exciton ground state wavefunction is Gaussian. \( \Delta \) is the photon–exciton energy detuning. It is seen that the effect of VSC can be alternatively interpreted as an effective renormalization of the bare cavity energy by \(-g_0^2/E_B\). In addition, such an effect wears off when \( g_0 \ll E_B \) and (3) reduces to the usual strong coupling (SC) Hamiltonian. In figure 1, we plot the UP and LP energies \( E_{\text{pol}} \) considering various \( g_0 \) and \( \Delta \) using (3). It shows that for a specific detuning, the LP (UP) energy decreases (increases) with increasing coupling constant as expected. Nevertheless, compared to the conventional Rabi splitting where \( E_{\text{pol}} \) changes linearly with \( g_0 \), here the change is nonlinear and a significant asymmetrical splitting can be observed.

In order to handle a larger class of problems such as studying an exciton–polariton under an external electric or magnetic field, we extend the previous variational formalism to include the spatial profile of exciton wavefunction as variational parameters, so that the system energy functional can be written as
$$E = \left\{ \psi_{\text{pol}} \right\} H \left\{ \psi_{\text{pol}} \right\}$$

$$= \alpha^2 \int \phi(r) \left( -\frac{\hbar^2 \nabla^2}{2\mu} + \frac{-e^2}{4\pi\varepsilon_0 r} + U(r) \right)$$

$$\times \phi(r) d^2r + \beta^2 \left( -E_B + \Delta \right) - 2\alpha \beta g_0 \left| \frac{\phi(0)}{\phi(0)} \right|$$

$$= f(\phi(r), \alpha).$$

(4)

$U(r)$ is an external potential exerted on the exciton wavefunction. Note that $K_0 = 0$ has been assumed. The minimization of this energy functional will deliver the ground state exciton wavefunction, Hopfield coefficients, and polariton energy. Numerically, the QW exciton wavefunction is first discretized spatially as a 2D matrix

$$\phi(x, y) \Rightarrow \begin{bmatrix}
\varphi_{1,1} & \varphi_{1,2} & \cdots & \varphi_{1,n} \\
\varphi_{2,1} & \varphi_{2,2} & \cdots & \varphi_{2,n} \\
\vdots & \vdots & \ddots & \vdots \\
\varphi_{m,1} & \varphi_{m,2} & \cdots & \varphi_{m,n}
\end{bmatrix}$$

(5a)

with each matrix element representing the magnitude of exciton wavefunction at a specific spatial grid inside the simulation space. Its second-order derivative is then constructed by a finite difference approach

$$\nabla^2 \phi(x, y) \Rightarrow \frac{1}{h^2} \left( \varphi_{i-1,j} + \varphi_{i+1,j} - 4\varphi_{i,j} + \varphi_{i-1,j-1} + \varphi_{i-1,j+1} \right)$$

(5b)

where $(i,j)$ and $(m,n)$ stand for the label and the number of grid points in the $x$ and $y$ directions. $h = L/(n - 1)$ is the distance between the grid points assuming $m = n$, and $L$ is the size of simulation space. By using (5a) and (5b), we may rewrite (4) as

$$E = f(\varphi_{1,1}, \varphi_{1,2}, \cdots, \varphi_{i,j}, \cdots, \varphi_{h,m}, \alpha).$$

(6)

The energy functional now becomes a function with $(nm + 1)$ variables, and the problem is transformed into finding a vector that minimizes the objective function $f$ subject to the constraints of $\sum_{i,j}^m |\varphi_{i,j}|^2 = 1$ and $|\varphi|^2 + |\beta|^2 = 1$. The nonlinearity of both the objective function and the constraints render the minimization a nonlinear optimization problem (NLP) of the general form [9, 10]:

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**Figure 1.** Polariton energy plotted as a function of coupling constant and energy detuning for (a) LP and (b) UP. Note that the white dashed line in (b) indicates the boundary ($E_{\text{pol}} = 0$) between bound excitons and free electron–hole pairs.
\[ \begin{align*}
\min_{x \in \mathbb{R}^n} & \quad f(x) \\
\text{subjectto:} & \quad c_i(x) = 0 \quad i = 1, \ldots, p \\
& \quad g_i(x) \leq 0 \quad i = p + 1, \ldots, l
\end{align*} \]

Note that if \( f = 0 \) the problem is referred to as unconstrained optimization problem. In our numerical optimization \( s = nm + 1 \), and since the number of grid points involves a rather large scale, we apply the widely used sequential quadratic programming (SQP) \([11-13]\) algorithm to save the computational time. It begins with a given guess \( x^0 \) and then iterates through the quadratic programming to generate a next approximated solution \( x^{k+1} \). The successive iteration is performed to construct a sequence of solutions belonging to a specific set that converges to an optimum solution. We implement the iteration process by means of the Optimization Toolbox in the MATLAB software package \([14]\) to solve our NLP. By loading the system energy function together with its constraints into the SQP program where the 1s 2D exciton wavefunction is given as an initial guess, the solution corresponding to the exciton wavefunction \( \psi(r) \) and the exciton fraction \( \alpha \) of the LP is obtained. The UP solution can then be retrieved by implementing the parity condition of Hopfield coefficients (i.e. for UP \( \alpha \) and \( \beta \) are switched and have opposite signs) in the system energy functional and repeating the SQP program given the LP solution as an initial guess.

In figure 2(a), we plot the energy-detuning relations for both the UP and LP using the analytical solutions of Hamiltonian (3) and the numerical solutions of Hamiltonian (4) assuming no external potential presents. Note that the yellow region indicating the bound exciton no longer exists so any calculated UP state that penetrates this region should be ignored. It can be seen that the numerical results match the analytical results well, which attests to the applicability of our numerical technique. Note that the differences in analytical and numerical results are more apparent for the UP due to the fact the UP solution is iterated from the LP solution, and the accuracy can be further improved by for example increasing the number of grid points. In figure 2(b), \( E_{\text{pol}} \) at resonance for different \( g_0 \) is calculated to show the strong asymmetric Rabi splitting which nonlinearly depends on \( g_0 \). In figure 2(c), the LP exciton (photon) fraction at resonance is shown to be significantly smaller (larger) than 50% at a large \( g_0 \). Note that in both figures 2(b) and (c), again the analytical and numerical results agree well with each other.

In all above calculations, \( m = n = 89 \) and \( L = 10a_0 \) are used.

**Recovering the bare cavity energy**

From the experimental point of view, it is impossible to distinguish the effect of VSC from SC by simply measuring the UP and LP energy-detuning relations. This is due to the fact that although the UP and LP as well as the QW exciton energies can be determined by the energy-resolved photoluminescence measurements at a vertical angle (for the UP and LP) as well as at a large oblique angle (for the QW exciton), the MC photon energy is nevertheless treated as a fitting parameter. As a consequence, the ambiguity of whether one should use the black dashed line or the purple dashed line in figure 2(a) to represent the MC photon energy results in the uncertainty of whether the VSC model or the SC model should apply to the experimental system. We believe such a problem prevents a direct verification of the existence of VSC experimentally to date.

In viewing (3), if \( g_0 \) can be reduced in a controllable fashion so that the bare cavity energy \(-E_B + \Delta\) is recovered by the energy-resolved photoluminescence measurement at a vertical angle, the ambiguity of fitting the MC photo energy with the black dashed line (VSC model) or the purple dashed line (SC model) in figure 2(a) can be resolved. This can be realized by applying an in-plane electric field to the QWs so that the excitonic oscillator strength is bleached by separating the relative distance between an electron and a hole. The physical properties of such a system can be calculated by our numerical technique introduced previously with the external potential \( U(r) \) in (4) given as \( eF_0 r \). \( F_0 \) is the magnitude of an in-plane electric field. In our calculations, an electrical field is applied laterally in the \( y \) direction and the dimensionless field strength \( f = F_y/F_0 \) where \( F_y = E_y/ea_0 \) is introduced. The proposed experimental structure is shown in figure 3(a), in which the semiconductor optical MC is etched to dispose electrical contacts in close proximity to the QW layer for applying an in-plane electric field. To implement this structure, one thing that should be considered experimentally is how to precisely control the DBR etch depth so that the in-plane electrical field can be efficiently applied to the QW layers at the strongest cavity field antinodes. Moreover, the etched sidewalls may introduce undesired recombination centers that quench the LP signals so the area of etched mesa should be designed to be sufficiently larger than that of the LP or an efficient sidewall passivation should be applied.

We calculate the spatial profile of the exciton wavefunction in the relative coordinate under the influence of an in-plane electric field in figures 3(b) and (c), and the energy-detuning relations are calculated in figure 4. For the LP (UP) that is photon (exciton)-like, the dc Stark effect on the spatial profile is less obvious (obvious). Both UP and LP exciton wavefunctions are stretched by the in-plane electric fields as shown in figures 3(b) and (c), and the reduction of oscillator strength can be observed in figure 4. Note that the ‘stuck’ of the exciton
wavefunction at $y = -5a_0$ in figures 3(b) and (c) is due to the null-wavefunction boundary condition. In figure 4(a) where $f = 1$, the energy branches that are exciton-like ($\Delta \gg 0$ for LP; $\Delta \ll 0$ for UP) shift downward because of the dc Stark effect, similar to the energy red-shift of a hydrogen atom in a static electric field. In figure 4(b) where $f = 2$, the LP becomes a complete flat line corresponding to the red-shifted QW exciton energies and the UP unveils the bare cavity energies. The proposed experimental scheme shall provide a direct verification of the existence of VSC.

**Verification of the reduced (enlarged) radius of the LP (UP) in VSC**

The modification of exciton radius is the origin of the VSC phenomena discussed above. Here we propose another experimental scheme to verify the existence of the VSC by measuring the diamagnetic energy shifts in the presence of a vertical magnetic field. The diamagnetic energy shift is frequently used to determine the size of an exciton in a semiconductor and can be expressed as [15–17]
where the diamagnetic coefficient $\gamma_2$ is about $13 \mu$eV T$^{-2}$ for a 1s 2D exciton in a GaAs QW [17], i.e., a diamagnetic energy blue-shift of about 1.3 meV can be achieved for a 10 T vertical magnetic field. In the SC regime, a 'rigid' exciton model is assumed so that the exciton radius is independent of photon–exciton coupling. Accordingly, the net diamagnetic energy blue-shift, defined as the difference between UP and LP energy blue-shifts, should be zero at the resonance condition (equal photon–exciton fraction) and a small number at a detuned condition (unequal photon–exciton fraction). Nevertheless, in the VSC regime, a 'flexible' exciton model is assumed so that a considerable net diamagnetic energy blue-shift shall present due to the significant

\[ H_{\text{diam}} = \frac{e^2}{8\mu} \left| \vec{B} \times \vec{r} \right|^2 \gamma_2 \left( \frac{\vec{r}^2}{r^2} \right)_1 \hat{z} B^2, \quad (8) \]

Figure 3. (a) Schematic construction of our device where multiple QWs are inserted in the cavity region and the DBR region. The (b) LP and (c) UP exciton wavefunctions under the influence of an electrical field applied laterally in the $y$ direction with $f = 1$ given $\Delta = E_B$.

Figure 4. The energy-detuning relations of UP and LP under the influence of an electric field applied laterally in the $y$ direction with (a) $f = 1$ and (b) $f = 2$ given $g_0 = 1.2E_B$. 
difference in UP and LP exciton radii, which are enlarged and reduced respectively through the photon–exciton coupling. The apparent net diamagnetic energy shift serves as a direct verification of the existence of VSC, which can be experimentally measured through the photoluminescence signals from UPs and LPs in a vertical magnetic field.

We solve the NLP with diamagnetic interaction added and determine the exact exciton wavefunctions and the corresponding energy shifts. In figures 5(a)–(c), the exciton wavefunctions are presented respectively for the 1s 2D exciton (without applying a magnetic field) as well as LPs and UPs (applying a magnetic field). A diamagnetic energy blue-shift of about 2 meV and 0.2 meV can be observed for UPs and LPs respectively when $g_0 = 1.2E_B$ and $B = 10T$, which are accessible experimentally. An even larger energy blue-shift can be observed when $g_0 = 15T$. It is apparent that in the VSC regime, the significant energy blue-shift for the UP originates from the enlarged exciton radius, a phenomenon that is absent in the SC regime.

In figure 6(a), we calculate the net diamagnetic energy shifts $\Delta E_{\text{UP}} - \Delta E_{\text{LP}}$ for different coupling constants based on the VSC model solved by our numerical technique, and based on the SC model solved by an analytical calculation [18]. For the SC model (solid lines), the net diamagnetic energy shift at a given coupling constant vanishes at resonance, and remains as a small value in the small detuning region. For the VSC model (dashed lines), the net diamagnetic energy shift up to 2.5 meV can be observed at resonance and increases/decreases continuously with negative/positive detunings. Thus the nature of photon–exciton coupling can be clearly resolved on the level of the wavefunction by the proposed experiment, which provides another direct verification of the existence of VSC. Note that the above calculated diamagnetic energy shifts agree reasonably...
well with the results of using first-order perturbation theory [15, 16] assuming the cyclotron energy is small compared to the exciton binding energy.

**Conclusion**

To summarize, the VSC effect in semiconductor cavity QED systems where the QW exciton radius is dramatically modified has been verified by means of a nonlinear numerical optimization technique. Furthermore, we propose two experimental schemes, one by reducing the oscillator strength of QW exciton with an in-plane electric field to recover the bare cavity energy, and the other by inducing the diamagnetic energy shifts in both UP and LP energy branches with a vertical magnetic field to compare their energy difference, and they experimentally provide unequivocal proof of the existence of VSC. Our work offers further insight into the very strong light–matter interaction in semiconductor optical MCs, and the quest of developing a room-temperature GaAs polariton laser.

**References**

[1] Savona V, Andreani L C, Schendimann P and Quatropani A 1995 *Solid State Commun.* 93 733–9
[2] Skolnick M S, Fisher T A and Whitaker D M 1998 *Semicond. Sci. Technol.* 13 645
[3] Deng H et al 2002 *Science* 298 199
Kasprzak J et al 2006 *Nature* 443 409
Balili R, Hartwell V, Snoke D, Pfeiffer L and West K 2007 *Science* 316 1007
[4] Amo A et al 2009 *Nature* 457 291
Amo A et al 2009 *Nat. Phys.* 5 805
[5] Roumpos G et al 2010 *Nat. Phys.* 7 129
Lagoudakis K G et al 2008 *Nat. Phys.* 4 706
[6] Khurgin J B 2001 *Solid State Commun.* 117 307
[7] Zhang H, Kim N Y, Yamamoto Y and Na N 2013 *Phys. Rev. B* 87 115303
[8] Bloch J et al 1998 *Appl. Phys. Lett.* 73 1694
[9] Dimitri P B 1999 *Nonlinear Programming* (Cambridge: Athena Scientific)
[10] Andrzej R 2006 *Nonlinear Optimization* (Princeton, NJ: Princeton University Press)
[11] Bonnans et al 2006 *Numerical Optimization: Theoretical and Practical Aspects* (Berlin: Springer)
[12] Yang W Y et al 2005 *Applied Numerical Methods Using Matlab* (Hoboken, NJ: Wiley)
[13] Venkataraman P 2009 *Applied Optimization with Matlab Programming* (Hoboken, NJ: Wiley)
[14] www.mathworks.com
[15] Walck S N and Reinecke T L 1998 *Phys. Rev. B* 57 9088
[16] Grochol M, Grosse F and Zimmermann R 2005 *Phys. Rev. B* 71 125339
[17] Erdmann M, Ropers C, Wenderoth M and Ulbrich R G 2006 *Phys. Rev. B* 74 125412
[18] Deng H, Haug H and Yamamoto Y 2010 *Rev. Mod. Phys.* 82 1489