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Invited Paper

Exciton polaritons in single and coupled microcavities

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Abstract

Recent work on strong coupling exciton–polariton phenomena in single and coupled microcavities is presented. We describe experiments for single cavities where the strong coupling nature of the excitations manifests itself. It is also shown that coupled cavities enable optically induced coupling between macroscopically separated exciton states to be achieved, and polaritons with strongly anisotropic properties to be realised. Results for both inorganic and organic microcavities are presented.

Keywords: Polaritons; Microcavities; Photonic properties

1. Introduction

There has been a great deal of interest in the study of exciton–polariton phenomena in semiconductor microcavities in the last few years [1]. This article describes some of the principal topics of interest, with emphasis on investigations where new physics is revealed. We first explain why microcavities are favourable for the study of polariton phenomena. We then describe the results of reflectivity, Raman scattering and ultrafast experiments on single cavities, and reflectivity and photoluminescence studies of coupled cavities, which exemplify some of the recent advances. Finally, we summarise the main points.

Semiconductor microcavities (for reviews see Refs. [2,3] are planar Fabry–Perot cavities (one-dimensional photonic structures) in which the mirrors are formed from distributed Bragg reflectors, alternating quarter wavelength layers of materials of low and high and dielectric constant. The confined photon modes interact with the excitonic states of QWs embedded in the cavity. A typical GaAs-based microcavity is shown schematically in Fig. 1.

2. Light–matter interaction in semiconductors, the significance of microcavities

Ever since the late 1950s the importance of polariton phenomena in determining the low-temperature optical properties of high-purity semiconductors has been recognised [4,5]. Polaritons are the stable coupled mode eigenstates of excitons and photons in a crystal. As a result of translational invariance, for each value of wave vector the exciton and photon states are (strongly) coupled together. However, since in the bulk of the material the photon lifetime is infinite, and in a high purity material exciton scattering is improbable, polariton decay within the bulk cannot occur. When an exciton is created by an external photon, in the absence of scattering, the excitation oscillates (vacuum Rabi oscillations) between the exciton and photon states without irreversible decay. Transformation into external photons does occur at the surface, but due to spatial dispersion [4,5]

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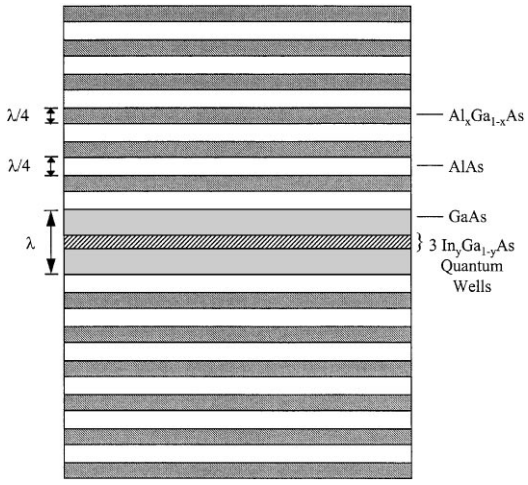

Fig. 1. Schematic diagram of semiconductor microcavity.

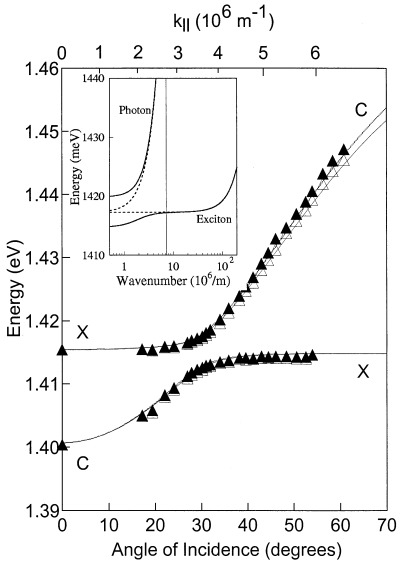

Fig. 2. Reflectivity dip positions as a function of angle and in-plane wave vector for single microcavity. TE polarisation open symbols, TM filled symbols. Inset – calculated polariton dispersion curves.

and the presence of an exciton free surface layer, reflectivity spectra do not provide reliable information on polariton energies and dispersion. Furthermore, as a result of scattering, photoluminescence (PL) does not provide direct information on the energy distribution of photocreated polaritons in bulk materials. The situation is very different in QWs. Excitonic wave vector is no longer conserved along the growth direction (z), and irreversible decay can occur [6] since there is no longer the one-to-one correspondence between exciton and photon states along all three directions. The exciton and photon states in QWs are thus weakly coupled; this is why excitonic recombination dominates the PL spectra of QWs, in contrast to bulk materials.

In a microcavity, the photon states are also quantised along z, and the correspondence in k-space between (QW) exciton and photon states, and the strong coupling limit, is restored. However, along z the photon has a finite lifetime due to leakage through the mirrors. The polaritons are thus directly coupled to external photons, and as a result polariton-related phenomena can be studied directly. This situation contrasts strongly with that in either bulk materials or QWs, and is the principal reason why new polariton physics can be studied in microcavities. The polariton dispersion curves (Fig. 2 inset) in a microcavity are also of strong significance. For accessible values of in-plane wave vector the photon dispersion is approximately parabolic, with very small "effective mass" of $\sim 3\times10^{-5}m_e$ [2,3]. In the region of strong coupling, the polaritons also have a small mass and thus small density of states (DOS). At large k the lower polariton branch is exciton-like, with effective mass $\sim 0.25m_e$, and thus large DOS.

To observe strong coupling the exciton–photon coupling strength must be greater than the widths of both the exciton and cavity modes. If this condition is not fulfilled then either the photon leaks from the cavity, or the exciton undergoes dephasing before one period of oscillation between the exciton and photon modes can occur. In this case, the system is in the weak coupling limit and the exciton decays by spontaneous emission.

3. Observation of strong coupling and the polariton dispersion curves

Strong exciton–photon coupling in microcavities was first reported in 1992 [1] as an anti-crossing and exchange of intensity between exciton and photon modes in reflectivity. Results as a function of angle of incidence (θ) are shown in Fig. 2 [7]. Since θ is related to $k_{||}(=(E_{photon}/\hbar c)\sin\theta)$, the angular dependence corresponds to a direct mapping of the polariton dispersion. The results demonstrate that the polariton dispersion can be observed directly in microcavities [8] as discussed in the previous section. Fig. 2 also shows a splitting between TE and TM polarisations, due to the slightly different phase delays of the two polarisations [7,9].

The size of the vacuum Rabi splitting (Δ_{VRS}) between the polariton modes on resonance is controlled principally by the exciton oscillator strength $f(\Delta\propto f^{1/2})$. In GaAs-based microcavities this limits the maximum values of mode splitting to $\sim$10 meV, although in II–VI microcavities values up to 20 meV have been reported

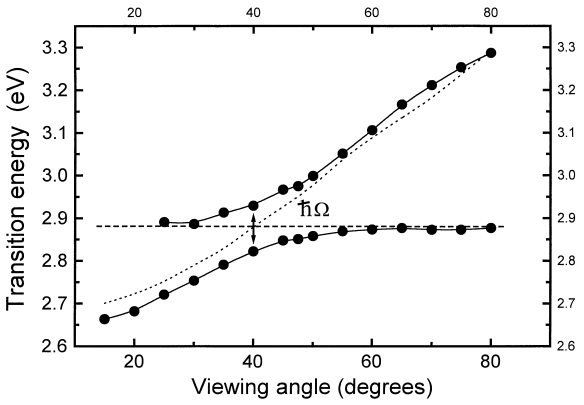

Fig. 3. Strong coupling in microcavity containing zinc porphyrin derivative. The vacuum Rabi splitting on resonance is 160 meV.

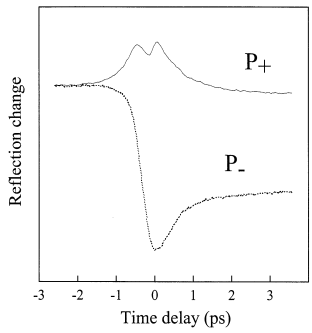

Fig. 4. Probe reflectivity spectrally integrated over pump pulse spectral width as a function of time delay for pumping of the upper (P_-) and lower (P_+) branches. The change in reflectivity for the lower branch is $\sim 10\%$.

[2,3]. Very much larger values of Δ_{VRS} have been reported in the last year in organic microcavities, as a result of the very large values of f for the Frenkel excitons of organic materials. By special choice of organic material with relatively narrow exciton lines (zinc porphyrin derivative or *j*-aggregate cyanine dye compound) the strong coupling limit was achieved at 300 K, with giant values of Δ_{VRS} up to 160 meV observed [10,11], as shown in Fig. 3. This result is significant since for large values of Δ_{VRS}, and for the large values of exciton binding energy of Frenkel excitons, the strong coupling limit is likely to persist into the regime of high carrier densities relevant to e.g. lasers. Advantage can then be taken of the very short polariton radiative lifetime relative to that of bare excitons which may lead to very low threshold lasing. Such large values of Δ_{VRS} are also likely to be very favourable for the study of stimulated polariton scattering [12].

4. Asymmetry between the polariton branches and the effect of disorder

The results of e.g. the experiments of Figs. 2 and 3 can be fully explained by a simple two-oscillator model. However, two pieces of experimental evidence cannot be explained by this approach: the line width of the upper polariton branch on-resonance is greater than that for the lower branch, and secondly the average line width on-resonance is less than the average of the off-resonance widths [13].

Despite much discussion [13–15], it was only in 1998 that a comprehensive understanding of these phenomena was achieved [16–18]. It is now accepted that both observations arise from the inhomogeneous nature of the exciton broadening. In Ref. [16] it is shown that as a result of the small polariton density of states, multiple k non-conserving disorder scattering makes a negligible contribution to the line widths (equivalent to "motional narrowing" of the disorder broadening in a real-space description) [16]. Instead, scattering of optically created polaritons into disorder-induced or high k exciton states dominates. The greater width of the upper branch is explained naturally as a result of the asymmetric exciton line shape (width Δ_{inh}) [16] which leads to stronger scattering into exciton states for the upper branch. Furthermore, for Δ_{VRS} greater than Δ_{inh}, scattering into exciton states will be significantly weaker than for smaller Δ_{VRS}, leading to the small on-resonance widths. Similar conclusions have been reached [17,18] from inclusion of an asymmetrically broadened exciton line in transfer matrix simulations. The assumption of translational invariance in these treatments is justified as a result of the very light polariton mass, which leads to averaging over the disorder potential.

Further evidence for the strongly differing properties of the two branches is provided by Raman spectroscopy [19] and ultrafast reflectivity [20]. In Ref. [19] it was found that the Raman cross-section of the upper polariton mode was much weaker than that for the lower mode. This behaviour was explained as resulting from rapid dephasing of upper-branch polaritons into disorder-related high-k exciton states. Confirmation of this result is provided by ultrafast reflectivity studies [20]. Fig. 4 shows that long-lived changes arise when the upper branch is pumped, whereas when the lower branch is pumped, the reflectivity returns close to its unpumped value after $\sim$1ps, providing direct evidence for efficient scattering of upper-branch polaritons into non-radiative states. The return to radiative $k \sim 0$ modes is slow due to the small polariton DOS, thus accounting for the long-lived change when the upper branch is pumped. By contrast when the lower branch is pumped, the probability for scattering out of the $k = 0$ states is much lower, due to the small overlap in energy with high k states

(Fig. 2 inset). When the upper branch is pumped a marked decrease ($\sim 10\%$) in reflectivity occurs, arising probably from screening effects from the high density of high k excitons; by contrast when the lower branch is pumped the change is very short lived and arises from an optical Stark effect which leads to an increase in reflectivity.

5. Coupled microcavities

Coupled microcavities provide an extra degree of freedom in the engineering of photonic modes and of their interaction with excitons. A typical coupled cavity is shown in Fig. 5a. It consists of two cavities, with coupling between them controlled by the central Bragg mirror. The photonic eigenstates have symmetric (S) and antisymmetric (AS) symmetry (Fig. 5b). Reflectivity spectra as a function of θ are shown in Fig. 6 [21,7]. At low θ, S and AS cavity modes are seen, together with the exciton feature X. With increasing θ, the exciton mode gains energy, and on resonance at $\sim 29^\circ$, a four-mode spectrum is observed. Increasing θ further results in the restoration of the three-mode spectrum. The four-mode spectrum on-resonance results from lifting of the degeneracy of the excitonic states from the upper and lower cavities, even though they are separated by over 2 μm. The S combination of exciton states couples to the S photon mode and likewise the AS combinations couple, leading to the four-mode spectrum. The long-range interaction between the exciton states results from coupling to the long-wavelength optical fields. For example, for two QWs embedded in a single cavity only the S exciton state is strongly coupled to light; the other AS combination is dark. The symmetries of the optical fields in a coupled cavity enable both S and AS combinations to be observed and permit coupling between exciton states separated by macroscopic distances.

Qualitatively, new phenomena are observed for coupled cavities when QWs are embedded in only one of the cavities [22]. The structure then consists of a three-oscillator system composed of the upper (C_U) and lower (C_L) cavity modes and the excitonic (X) states from the QWs. C_U is coupled to both X and C_L, whereas C_L is only coupled to C_U. Away from resonance reflectivity spectra were found to consist of three features [22]. However, for the central polariton mode, the interactions of C_U with C_L and X are in antiphase and cancel exactly on resonance, with the result that the optical field in C_U is zero. This cancellation occurs even though the exciton–cavity and cavity–cavity interaction strengths are not the same (~ 5 and 9 meV, respectively), since the mode admixtures depend on both the coupling strengths and on the relative uncoupled energies. On resonance the mode energies are equal and exact cancellation results.

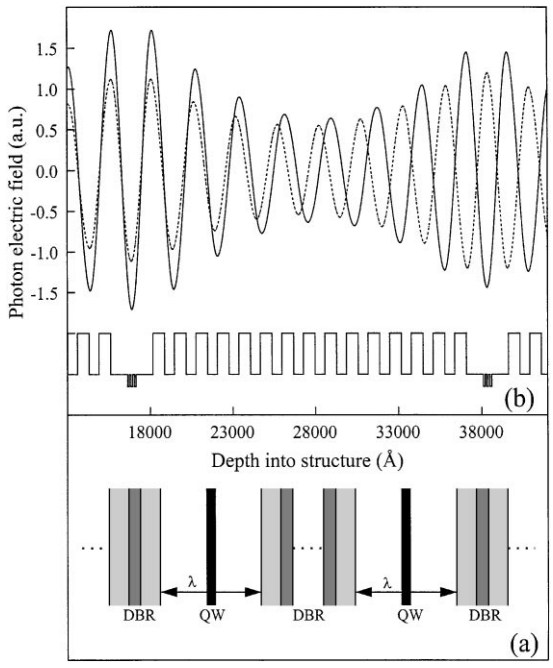

Fig. 5. (a) Schematic diagram of coupled cavity containing QWs in upper and lower cavities. (b) Calculated optical fields showing symmetric and antisymmetric photon fields.

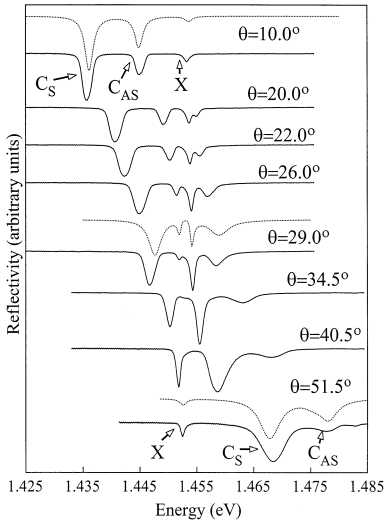

Fig. 6. Reflectivity spectra for sample of Fig. 5. Four modes are observed on resonance due to the lifting of the degeneracy of the exciton states from the upper and lower cavities. The dotted curves are theoretical fits using a transfer matrix model.

In a reflectivity experiment from the front surface the optical field in the upper cavity is probed. Since the optical field in C_U is zero on resonance, the central mode is dark. However, when the rear surface is probed,

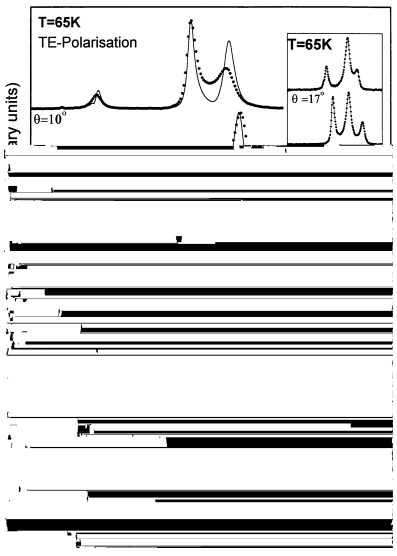

Fig. 7. PL spectra for coupled cavity containing QWs only in the upper cavity. Main figure – PL spectra from the top surface, inset – PL from the substrate side. Dotted curves – experimental, full lines – theory using transfer matrix model.

the central mode is found to be strong. Thus, for coupled cavities in which inversion symmetry is broken by embedding QWs in only one cavity, unidirectional polaritons are created.

PL spectra observed from the front surface (cavity containing the QWs) are presented in the main part of Fig. 7. The central mode is absent on resonance at 22.5°, but is strong when observed from the lower cavity, as seen in the inset to Fig. 7. This result is particularly striking when it is remembered that the optical field in the upper cavity is zero on resonance. The excitons thus emit light by non-local interaction with an optical field in a cavity which is spatially separated from the QWs by over 2 μm.

6. Summary

We have described recent experiments which reveal the polariton nature of the optical excitations in semiconductor microcavities. In single cavities new phenomena arising from the form of the polariton dispersion curves and from the effects of disorder have been described. The ability to engineer photonic states in coupled cavities, and hence to study novel physics, has also been demonstrated.

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References

[1] C. Weisbuch, M. Nishioka, A. Ishikawa, Y. Arakawa, Phys. Rev. Lett. 69 (1992) 3314.
[2] M.S. Skolnick, T.A. Fisher, D.M. Whittaker, Semicond. Sci. Technol. 13 (1998) 645.
[3] M.S. Skolnick et al., International Conference on Physics of Semiconductors, Jerusalem, World Scientific, Singapore, 1999.
[4] J.J. Hopfield, Phys. Rev. 112 (1958) 1555.
[5] S.I. Pekar, Sov. Phys. JETP 6 (1958) 785.
[6] L.C. Andreani, F. Tassone, F. Bassani, Solid State Commun. 74 (1991) 641.
[7] D. Baxter, M.S. Skolnick, A. Armitage, V.N. Astratov, D.M. Whittaker, T.A. Fisher, J.S. Roberts, D.J. Mowbray, M.A. Kaliteevski, Phys. Rev. B 56 (1997) R10032.
[8] R. Houdré, C. Weisbuch, R.P. Stanley, U. Oesterle et al., Phys. Rev. Lett. 73 (1994) 2043.
[9] G. Panzarini, L.C. Andreani, A. Armitage, M.S. Skolnick et al., Phys. Rev. B 59 (1999) 5082.
[10] D.G. Lidzey, D.D.C. Bradley, T. Virgili, M.S. Skolnick, D.M. Whittaker, S. Walker, Nature 395 (1998) 53.
[11] D.G. Lidzey, D.D.C. Bradley, T. Virgili, M.S. Skolnick, D.M. Whittaker, S. Walker, Phys. Rev. Lett. 82 (1999) 3316.
[12] P. Senellart, J. Bloch, Phys. Rev. Lett. 82 (1999) 1233.
[13] D.M. Whittaker, P. Kinsler, T.A. Fisher, M.S. Skolnick, A. Armitage, A.M. Afshar, J.S. Roberts, Phys. Rev. Lett. 77 (1996) 4792.
[14] R. Houdré, R.P. Stanley, M. Ilegems, Phys. Rev. A 53 (1996) 2711.
[15] V. Savona, C. Weisbuch, Phys. Rev. B 54 (1996) 10835.
[16] D.M. Whittaker, Phys. Rev. Lett. 80 (1998) 4791.
[17] A.V. Kavokin, Phys. Rev. B 57 (1998) 3757.
[18] C. Ell et al., Phys. Rev. Lett. 80 (1998) 4795.
[19] W.R. Tribe, D. Baxter, M.S. Skolnick, D.J. Mowbray et al., Phys. Rev. B 56 (1997) 12429.
[20] J.M. Baumberg, A. Armitage, M.S. Skolnick, J.S. Roberts, Phys. Rev. Lett. 81 (1998) 661.
[21] A. Armitage, M.S. Skolnick, V.N. Astratov, D.M. Whittaker, G. Panzarini, L.C. Andreani, T.A. Fisher, J.S. Roberts, A.V. Kavokin et al., Phys. Rev. B 57 (1998) 14887.
[22] A. Armitage, M.S. Skolnick, A.V. Kavokin, D.M. Whittaker, V.N. Astratov, G.A. Gehring, J.S. Roberts, Phys. Rev. B 58 (1998) 15367.