Microcavity quantum-dot systems for non-equilibrium Bose-Einstein condensation

I M Piper¹, P R Eastham², M Ediger¹, A M Wilson¹, Y Wu¹, M Hugues³, M Hopkinson³ and R T Phillips¹

¹ Cavendish Laboratory, University of Cambridge, J J Thomson Avenue, Cambridge CB3 0HE, UK
² School of Physics, Trinity College Dublin, Dublin 2, Ireland
³ Department of Electronic and Electrical Engineering, University of Sheffield, Mappin Street, Sheffield, S1 3JD UK

E-mail: imp24@cam.ac.uk

Abstract. We review the practical conditions required to achieve a non-equilibrium BEC driven by quantum dynamics in a system comprising a microcavity field mode and a distribution of localised two-level systems driven to a step-like population inversion profile. A candidate system based on eight 3.8nm layers of In₀.₂₃Ga₀.₇₇As in GaAs shows promising characteristics with regard to the total dipole strength which can be coupled to the field mode.

1. Introduction
Experimental observation of polariton Bose-Einstein condensates (BEC) has been reported in several different systems [1, 2, 3]. In all of these cases, the ground state is populated by relaxation from an excited state, rather than populating the ground state resonantly. A different approach to creating BEC in a polariton system proposed by Eastham and Phillips [4], does not involve dissipation or inelastic scattering. Instead the condensation occurs as a result of the coherent dynamics of a specific coupled photon-exciton system.

In general the proposed dynamical condensation could be implemented in any system of discrete atomic-like states which are dipole-coupled to a long-lived mode of the electromagnetic field. We consider the practical aspects of implementing this proposal in semiconductor systems.

2. Theoretical requirements
The proposed experiment involves two stages, which are separated in time and can be regarded as independent. The first stage is the creation of a tailored exciton population in the inhomogeneously-broadened exciton line using a controlled pump pulse. For certain population profiles there is then a second stage, in which the photon-mediated interactions between the localized excitons lead to condensation. More specifically, the normal modes of a population of dipole-coupled localized excitons obey an equation [(1)], which is essentially the Cooper equation of BCS theory. It predicts that steps in the energy-dependent exciton population result in new collective modes, leading to condensation. The theoretical requirements for the condensation
stage are described by the equation ($\hbar = 1$)

$$\lambda_k - \omega_k + \kappa^2 n \int \frac{\nu(E)[2n(E) - 1]}{\lambda_k - E} dE = 0. \quad (1)$$

This equation determines the complex normal-mode frequencies $\lambda_k$ of the electromagnetic field, in a planar microcavity containing an ensemble of localized dipole oscillators. These normal modes are approximately plane waves, with in-plane wavevector $k$. $\omega_k$ is the frequency of the bare cavity mode, whose imaginary part $-\gamma$ describes the cavity losses. The localized exciton states are assumed to be two-level systems, which can be either unoccupied or occupied. $\nu(E)$ is the normalized distribution of the energies of these transitions, and $n$ is their area density. Their average occupation at energy $E$ is $n(E)$. Adiabatic rapid passage allows the creation of tailored population profiles $n(E)$, with an energy resolution set by the duration of the pump pulse [4].

The coupling $\kappa$ is the matrix element for a transition from the unoccupied to the occupied state, assumed to be a constant for simplicity. In the dipole gauge its explicit form is

$$\kappa = \langle er \rangle \sqrt{\frac{\hbar \omega}{2e_0 L_{\text{eff}}}}, \quad (2)$$

where $\langle er \rangle$ is the dipole moment of the transition, $\hbar \omega$ its energy, and $L_{\text{eff}}$ the effective width of the cavity. A typical value for a semiconductor in the bandgap region is $\langle er \rangle \approx 20 \text{ D}$, corresponding to an electron moving approximately 0.5 nm [5]. For $\hbar \omega \approx 1 \text{ eV}$ and $L_{\text{eff}} \approx 20 \mu \text{m}$ this gives an estimate $\kappa \approx 8 \times 10^{-7} \text{ meV cm}^{-1}$.

A solution to (1) with $\lambda_k'' = 3(\lambda_k') > 0$ corresponds to an exponentially growing normal mode, which leads to a large population. Thus the presence of such solutions implies a dynamical phase transition from an exciton population to a non-equilibrium condensate. The character of this condensate depends on that of the unstable normal mode. If it is essentially a pure electromagnetic field mode we have a photon condensate, i.e. a laser. If it has a significant excitonic polarisation component then we have condensation of coupled light-matter states.

Close to a condensation threshold the imaginary part of (1) is a gain-loss equation for the growth rate of the mode: $\lambda_k'' = -\gamma + \pi \kappa^2 n \nu(\lambda_k)[2n(E) - 1]$. The energy range of the optically-active localized states is approximately the inhomogeneous linewidth $\gamma_{\text{inh}}$, so typically $\nu(E) \sim 1/\gamma_{\text{inh}}$. Thus a necessary condition for condensation is, up to numerical factors of order one, $\kappa^2 n > \gamma_{\text{inh}}$. A second requirement comes from the real part of (1), which determines the shifting of the normal mode energies due to the exciton population. Large shifts indicate a significant exciton component, and hence distinguish exciton-photon condensation from lasing. For realistic $n(E)$ and $\nu(E)$ equation (1) gives shifts of order $\kappa^2 n/\gamma_{\text{inh}}$. Thus if we require energy shifts of order $E_{\text{res}}$, as well as the gain-loss criterion to be satisfied, we have $\kappa^2 n > \gamma_{\text{inh}} \max(\gamma, E_{\text{res}})$.

An energy shift greater than the inverse of the exciton’s decoherence time implies an excitonic component to the condensate. We hope to achieve somewhat larger shifts, of order 1 meV. This is about one order of magnitude smaller than the vacuum Rabi splittings now routinely observed, and is a reasonable benchmark for a new exciton-photon coupling phenomenon. Assuming an inhomogeneous linewidth also of order 1 meV, and taking the estimate of the coupling strength above, gives an order-of-magnitude estimate of the total area density required near the field antinodes as $n > 10^{12} \text{ cm}^{-2}$. The previous demonstration of lasing in very similar vertical cavity surface-emitting structures [6] indicates that the required physical régime should be accessible.

3. Candidate semiconductor systems

Stranski-Krastanow (S-K) dots are limited in density by the requirements of the growth mode, and usually are strongly inhomogeneously broadened, which reduces the spectral density. We
Figure 1. (a) Confocal µPL from a region of area ≈ 1µm^2, showing both resolved and unresolved emission from the dot ensemble. Excitation was at 633nm, at a power density ≈ 10Wcm^{-2}. A model density of states of n = 10^{13} cm^{-2} (c) is too high to resolve individual transitions. Localised states can be resolved for an order of magnitude lower density (10^{12} cm^{-2}, (b)), indicating that the density of states in the sample lies between that of (b) and (c).

Figure 2. (a) The spectral dependence of the weak RRS enhancement shows a dip on the high energy side of the line associated with reabsorption in the eight-layer stack. The low degree of RRS enhancement indicates 0D rather than 2D states. (b) The sample is excited at an oblique angle and the RRS normal to the sample collected. The scattered intensity as the CW laser is tuned across the transition is recorded.

have therefore considered a system based on a "poor" quantum well, in which alloy and thickness fluctuations localise the nominally 2D states into quantum dots (QD). The key issues here relate to the dipole coupling strength, density of dots, and proof of their behaviour as isolated two-level systems. The sample consists of eight 3.8nm layers of In_{0.23}Ga_{0.77}As with GaAs barriers, grown by molecular beam epitaxy on an insulating GaAs substrate. Microphotoluminescence (µPL) collects PL from an area < 1µm^2 and resolves emission from individual localised states, as shown in Figure 1. By modelling the PL as a superposition of Lorentzians in a Gaussian distribution we estimate the total density of states (in eight layers) to be at least 10^{12} cm^{-2} (Figure 1(b,c)). This system therefore appears capable of satisfying the density requirement implied in Section 2. A further important feature of the PL evidence is that the total inhomogeneous linewidth of 10meV is very well suited to the proposed cavity BEC experiment, and is narrower than typically found for high density (n > 10^{10} cm^{-2}) S-K dot samples.

Resonant Rayleigh scattering (RRS) is sensitive to localisation, and for 2D states shows enhancement by a factor up to 100 on resonance [7, 8]. However, as localisation increases, the intensity of the RRS signal decreases and becomes broader, as the RRS efficiency depends on the ratio between the homogeneous and inhomogeneous broadening of the transitions [7]. The weak enhancement in the present samples (Figure 2(a)) is consistent with a high degree of localisation.

The electron-hole correlation length \( \rho \) is given by \( \langle \rho^2 \rangle = \frac{8\gamma_2 B}{\omega_0^2} \), where \( \gamma_2 B \) is the diamagnetic shift and \( \mu \) the reduced exciton mass [9]. The diamagnetic coefficient of one state measured using magneto-optical µPL [10] is found to be 20.1 \( \mu eV \) T^{-2} in the Faraday geometry and 7.6 \( \mu eV \) T^{-2} in Voigt, shown in Figure 3. We estimate the extent of the e-h correlation length to be approximately 7nm and 4nm respectively. As this is a simple model, these values are consistent with those of localised states in QDs. Electron microscopy of a QD system with
Figure 3. Diamagnetic shifts and spin fine structure of a single localised state in both the Faraday and Voigt geometries, illustrating the change in e-h correlation length as the field is moved from parallel to perpendicular to the growth axis. Markers show every tenth data point; lines show a least-squares minimised regression fit.

Figure 4. Map of µPL emission from a 15µm × 15µm region at six different energies.

similar composition shows the QDs are of length 40-100nm and width 20-30nm; these states have similar diamagnetic shifts to those measured in the present work [11].

µPL emission maps at six photon energies are shown in Figure 4 for a 15µm × 15µm region indicating elongation of the emitting regions in the [110] direction. The combination of these experiments indicates that the localised states in narrow In_{0.23}Ga_{0.77}As/GaAs quantum wells are promising candidates for the proposed dynamically-driven BEC, as they have a high density of discrete localised states distributed over a suitably narrow energy range.

4. Conclusions
We have outlined the theoretical origin of the requirements for a suitable system in which to realise the proposed condensation scheme. Narrow In_{0.23}Ga_{0.77}As/GaAs quantum wells show a suitably high density of localised states to offer a good system for attempted observation of the dynamically-driven BEC.

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