Nonlinear Emission of Semiconductor Microcavities in the Strong Coupling Regime

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We report on the nonlinear laserlike emission from semiconductor microcavities in the strong coupling regime. Under resonant continuous wave excitation we observe a highly emissive state. The energy, dispersion, and spatial extent of this state is measured and is found to be dispersionless and spatially localized. This state coexists with luminescence that follows the usual cavity-polariton dispersion. It is attributed to the amplification of luminescence by a parametric gain due to cavity-polariton scattering. Despite the resonant excitation at 1.6 K, we observe no sign of Bose-Einstein condensation nor Boser action.

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A quantum well in a semiconductor microcavity exhibits rich physics when the coupling between the cavity photon and the quantum well (QW) exciton is in the strong nonperturbative regime [1,2]. The resulting collective excitation, known as a cavity polariton (CP), is a composite boson and has the potential to produce collective behavior such as Bose-Einstein condensation (BEC) as observed in atomic systems. The experimental observation of the energy-momentum (E-k) dispersion of the CP has been essential in understanding the physics of CP's [3]. The form of the dispersion determines both the density of states and CP-CP scattering channels allowed by energy and momentum conservation. The CP dispersion is mainly photonlike and has a curvature 10^4 times greater than that of a QW exciton, while CP-CP interactions are excitonlike. The large elastic mean-free path implied by their dispersion has led many to search and claim quantum effects [4–10]. Unique to this system is the possibility to engineer the dispersion by changing the photon-exciton detuning. Although the linear properties [low intensity photoluminescence (PL), resonant Rayleigh scattering] of CP are more involved than that of a bare QW exciton, the physics is unambiguous: This is well explained by the properties of the CP's as the elementary excitations of the system. However, some nonlinear phenomena, in particular, the laserlike nonlinear emission observed under both nonresonant and resonant (this paper) excitation has been the subject of considerable debate. The main mechanisms used to explain such nonlinear emission in this system have been the Boser effect [4] and, more recently, boson assisted stimulation [10], as well as BEC and biexciton effects. The Boser effect is the stimulated scattering of bosons into their ground state when the ground state occupancy is greater than unity. For CP the scattering could be mediated by phonons (Boser) or by the CP themselves (Baser). Part of the problem is that some previous experiments dwelt on the energy spectrum of the emission, neglecting the dispersion which is fundamental to the nature of CP; other experiments neglect spatial effects. We will show that a complete view is necessary to distinguish between competing theories.

This paper presents novel results on the superlinear emission of semiconductor microcavities in the strong coupling regime. The novelty lies in the ability to simultaneously measure the energy, momentum, and spatial extent of the nonlinear emission. Among the wealth of phenomena we observed, the most significant is the coexistence of an intense spectrally narrow but *dispersionless* nonlinear emission line along with emission that follows the CP dispersion curve.

The experimental setup is shown in Fig. 1. It consists of a tunable Ti:sapphire laser beam impinging at the edge of a $f/2$ lens. The beam is focused to a 100 μm spot on the sample at an incidence angle of 10° and the same lens is used to collect the light emitted from the sample. To diminish the thermal load the exciting beam is modulated with a duty factor of 10^{-3}. The measurements are performed with an incident average intensity of 0.1 W cm^{-2} or below. The collected light either forms an image of the far field pattern on a visualization screen or is coupled into a 1 m monochromator. A movable diaphragm after the collection/excitation lens allows spectral measurements to be taken as a function of the emission angle. In all cases, the excitation is resonant with the lower cavity-polariton branch and the sample temperature is between

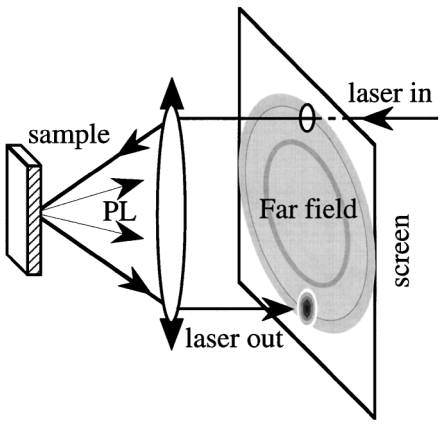

FIG. 1. Far field emission pattern setup.

1.5 and 4.2 K. The sample consists of a high quality factor GaAs/AlAs 2-λ cavity, with three low indium content $In_{0.04}Ga_{0.96}As$ QWs, one at each antinode of the light field inside the cavity. Such high quality samples exhibit extremely narrow polariton linewidths in the 100–200 μeV range at 4 K [11]. A wedge in the cavity thickness allows us to investigate cavity-exciton detunings from +8 down to −4 meV (negative for cavity mode energies less than the exciton energies).

A far field image at *low pump intensity* is shown in Fig. 2a. When the laser energy and incidence angle is exactly resonant with the lower branch CP energy and angle, absorption can be observed in the image of the reflected beam [denoted region (iv) in Fig. 2a] because the angular width of the cavity resonance is narrower than that of the incident laser beam. It appears as a weaker (dark) line inside the reflected beam spot. Simultaneously a bright ring appears at the same resonant angle outside the reflected beam, region (i), corresponding to resonant Rayleigh (elastic) scattering (RRS) of the incident photon (for further discussion on Rayleigh scattering, see [12]).

On increasing the excitation power, a sudden transition occurs and an intense emission occurs as either a disk, a ring, or a more complex pattern (depending on conditions), [region (vi) in Fig. 2b] inside the elastic Rayleigh ring and a crescent shaped resonant photoluminescence becomes clearly visible [region (v)]. Performing angular resolved photoluminescence measurements, we find three components in the emitted spectra, of varying amplitude for different emission angles (Fig. 3 inset): a Rayleigh component at the exciting energy and centered on the Rayleigh cone, a CP luminescence line the energy of which shifts with angle according to the CP dispersion curve, and a strong nonlinear emission line which is dispersionless (Fig. 3), at an energy slightly above the bottom of the CP dispersion curve [13]. This shift rules out an interpretation of the new emission line as a Boser-type effect as it would be unshifted compared to the bottom of the CP band.

Figures 2–5 show the principal results of this Letter; however, other key phenomena were observed under various experimental conditions and are included in the following summary: (i) At threshold, depending on the detuning, either an angular point or an actual discontinuity can be observed in the light in–light out curves (Fig. 4). (ii) Around −1 meV detuning, the threshold can reach values as low 17 mW peak incident power, corresponding to an estimated absorbed photon density of 7.10^{19} cm^{-2} s^{-1}. As will be discussed below, it is difficult to translate this number into a steady-state population as both lifetime and excited volume depend on excitation intensity via changes in relative populations of CP states with widely different lifetimes, resonance energy shift, and spatial pattern formation. (iii) The nonlinear emission energy, angular spread, even emission pattern depend on the excitation conditions. For a given cavity detuning, the nonlinear emission *follows* the excitation variation, both in energy (i.e., nonlinear emission energy vs pump energy) and angular extension (i.e., nonlinear emission angular spread vs incident pump angle), although in a nontrivial manner. (iv) For pump power densities accessible in our experiment, this nonlinear behavior is observed only while

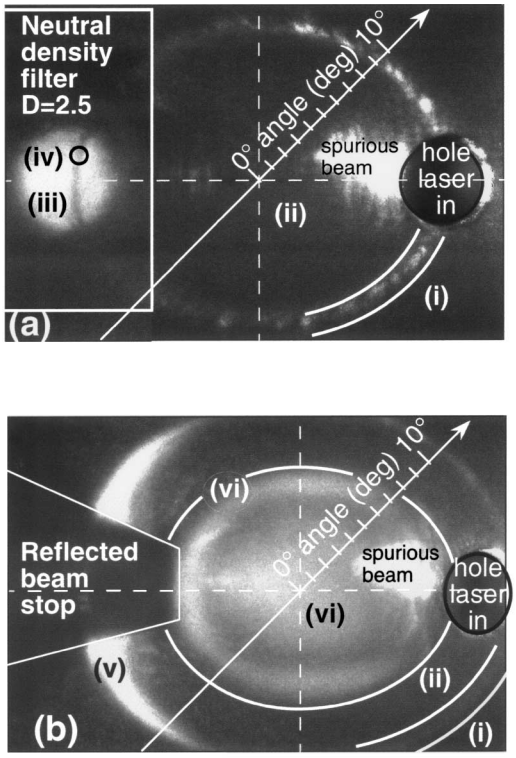

FIG. 2. (a) Far field image under resonant excitation of the lower cavity-polariton branch in the linear regime: (i) Elastic Rayleigh circle, (ii) photoluminescence from the lower cavity-polariton branch, (iii) reflected laser beam, (iv) polariton absorption. (b) Far field image in the nonlinear regime, above threshold: (i) Elastic Rayleigh circle, (ii) photoluminescence from the lower cavity-polariton branch, (v) resonant photoluminescence, (vi) nonlinear structure.

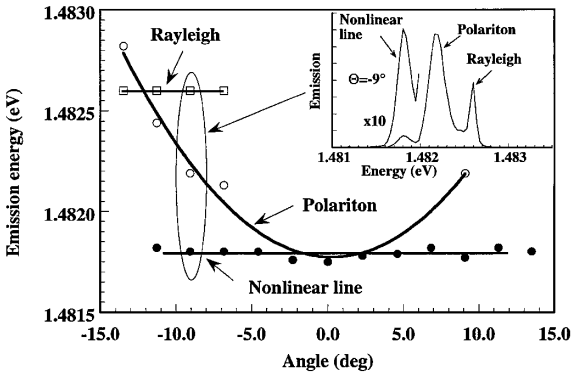

FIG. 3. Line energies vs emission angle from angular resolved PL in the nonlinear regime, $T = 4$ K and detuning $= -2.5$ meV. Inset: Photoluminescence spectrum at −9° emission angle.

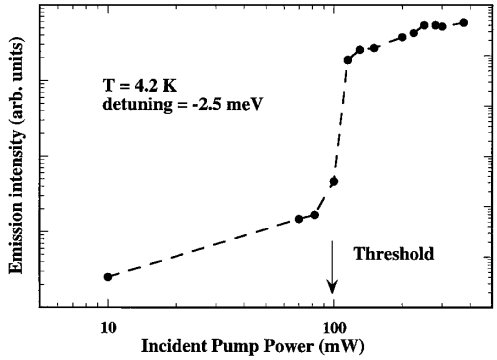

FIG. 4. Light in–light out plot at -2.5 meV detuning, $T = 4$ K.

exciting the lower polariton branch. (v) From a spatially filtered image, one observes that the strong nonlinear emission originates from a region which is no greater than half of the excitation spot (measurement limited by our spatial resolution at the large numerical aperture of the experiment). (vi) No clear differences are found between linear and circular polarization excitation. This latter result rules out *a priori* nonlinear mechanisms based on biexcitonic effects as it can be assumed that under resonant excitation little spin depolarization occurs, and therefore circular polarization should inhibit biexciton formation as the total biexciton angular momentum must be 0.

It is well known that by pumping a strongly coupled microcavity intensely the system evolves toward the weak coupling regime [14]. Therefore it was important to check whether the cavity was still under strong coupling conditions when reaching threshold. While the persistence of the Rayleigh scattering of the polariton state and observation of a cavity-polariton photoluminescence line would imply that strong coupling was not bleached, it can be seen in Fig. 5 that the experimental features are far more complex. Under high excitation the polariton absorption observed in

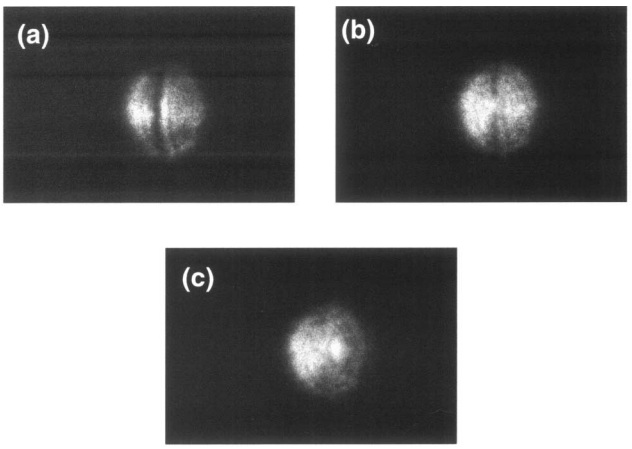

FIG. 5. Reflected beam showing transverse effects (a) below threshold, incident power <10 mW, (b) 75 mW incident power: screening of the cavity polariton in the center, and (c) 220 mW incident power: full screening of the cavity polariton.

the reflected beam is bleached at the center of the excitation spot, while absorption continues to occur in the outer part of the spot. The observation of this inhomogeneous situation greatly helps to solve the puzzle of the many apparently incompatible observed facts, the main one being the coexistence of "well-behaved" CP photoluminescence with a new emission line incompatible with a CP description: they are due to the coexistence of regions in strong coupling (periphery of excited spot) and regions (center of excited spot) where the resonance energy is spatially shifted, by either carrier renormalization or bleaching or by homogeneous broadening.

Any model however crude must address several major issues: (i) Below threshold, where does the pump energy go? (ii) What is the nonlinear emission mechanism? (iii) How can the nonlinear emission from the cavity have a linewidth similar to that of the cavity and yet be dispersionless?

Below threshold radiative CP states are directly excited through the resonant excitation. The energy is then redistributed in energy, direction, eventually space, through inelastic processes (relaxation) or elastic processes (Rayleigh scattering). Neither the thermalized PL nor the elastic ring changes in intensity at threshold. Therefore there must be another channel not visible in our experiment into which the pump energy is scattered below threshold. Such channels could be leaky and guided modes that remain trapped inside the GaAs material. We postulate that the energy is scattered into these channels by resonant Rayleigh scattering (RRS). This is justified from the Rayleigh scattering of bare QW excitons: the pioneering experiments of Hegarty *et al.* [15,16] show that RRS is efficient because the correlation length of the QW roughness is short enough to allow exciton scattering to all available k vectors. Similarly we expect that CP can be elastically scattered through disorder to other k vectors than the resonant cavity mode. A simple density of modes evaluation for our microcavities indicates that the scattering to guided modes (therefore not observed outside the sample in the experiment) should be up to 2 orders of magnitude greater than scattering into the observable elastic ring.

We have already ruled out biexcitonic or Boser-based gain mechanisms as the sources of the nonlinear emission. Instead CP-CP scattering, which conserves both energy and momentum, can be viewed as parametric amplification (or nondegenerate four wave mixing) where 2 CP scatter simultaneously from $k = k'$ to $k = 0$ and $k = 2k'$. Such phenomena has been recently observed by Savvidis *et al.* [10], in pump-probe experiments, where the CP's were excited resonantly at $k = k'$ and $k = 0$, with resulting emission at $k = 2k'$. Their results (however originally explained in terms of Boser) and ours triggered a novel theoretical treatment [17] based on a phase-matched four wave mixing or parametric amplification. In our experiments, the thermal PL acts as an idler (probe) which seeds the parametric process. After a certain pump intensity there

is sufficient power to provide gain at $k = 0$. Many of the features observed in our experiments such as the blueshift of the nonlinear emission with respect to the cavity mode by a quantity of the order of the CP homogeneous linewidth are also predicted in this model [17].

The remaining issue is therefore the dispersionless nature of the nonlinear emission. As only the center of the Gaussian excited region will reach threshold, the amplified emission will originate from a spatial region that has collapsed to a size smaller than the excitation Gaussian spot, and can thus have a flat dispersion as its angular width is not determined by population along the CP dispersion curve but by diffraction of the small emitting region. The CP mode has an angular width of about 0.01 rad (as taken from the width of the elastic ring or absorption line; Fig. 2a), while the nonlinear emission has a width of about 0.13 rad. The latter value gives a diffraction limited emission spot size of 3 μm. Furthermore, the emission must be spatially coherent on this scale.

While these arguments have yet to be put into a quantitative basis we believe that they explain the essential elements of our experiment. However, there remain several features to be explained, in particular, the transverse pattern formation. A detailed analysis of the energy coupled into the system is quite complex: Concurrently to the spatial collapse of the emitting region, absorption was shown to occur only on the periphery of the excitation spot (Fig. 5), some carrier redistribution has then to take place. This could give rise to spatial soliton [18] phenomena and may be the origin of spatial pattering.

In conclusion, we have shown in high-quality semiconductor microcavities that a transverse nonlinear optical pattern develops which displays a high efficiency. Boser and biexciton phenomena can be ruled out. While this new nonlinear emission mechanism has its major features well explained by the nonlinear parametric amplification scheme due to the same CP-CP scattering mechanism [17] which also explains four-wave amplication [10], it is here triggered by the spontaneous population of cavity-polariton states. To further investigate these phenomena one has to use advanced theoretical tools. They should rely on those used in the studies of optical pattern formation phenomena, this time including strong coupling, disorder, energy relaxation (both in real and k space), and accurate exciton interactions, a huge computational effort. The present experiments, while not reproducing conditions of the previous reports on nonlinear emission in strongly coupled microcavities (they were not under resonant excitation), do give some insight on novel ways to carry out experiments to sort out inhomogeneities, both in real and momentum spaces.

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