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Comments on Lasing and Many-exciton Physics in Strongly Confined Systems

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Abstract

The stimulated emission of strongly confined heterostructures, namely highly polar II–VI quantum wells, is discussed in terms of a phase transition between the exciton gas and the free-carrier gas. The extent to which the transition has proceeded under the operation condition of the device (high temperature and high injection density) depends on the many-body renormalization of the exciton binding energy. The general applications of the model are finally discussed.

Lasing in semiconductors usually involves free carrier recombination, but in more polar materials, such as the II–VI semiconductors for blue-green lasers, the stronger Coulomb correlation may lead to stable excitons even in the presence of a dense free-carrier background. In such materials lasing may occur in principle through stimulated recombination of excitons, provided that an intermediate level – a localized exciton state or the final state of a scattering process – exists to form the required three level system [1].

In the last few years, indeed, the impressive development of material technology has allowed fabrication of thin ZnCdSe/ZnSe and ZnSe/ZnS quantum wells whose large band discontinuities and polarity result in exciton binding energies exceeding the LO-phonon energy (up to 40 meV or more). Similarly, advances in nanotechnologies have recently permitted the achievement of exciton binding energy larger than the room temperature kT in narrow quantum wires [2]. Moreover, even biexcitons in II–VI quantum wells have been demonstrated to contribute to lasing under particular circumstances (high crystalline quality, low temperature and strong confinement) [3].

The unexpectedly important role of excitons in the lasing processes of the newest quantum structures has been pointed out in the pioneering work of the Brown-Purdue groups in 1992 [4], (see the paper of A. Nurmikko in this book), and since that time it has been the subject of intense discussion about the excitonic lasing and its relationships to the more conventional electron-hole plasma stimulated recombination [4–12]. In recent times, even excitonic lasing of quantum wires has been claimed in samples exhibiting substantial enhancement of the exciton binding energy with respect to the temperature of the experiment [2, 13].

In this contribution we would like to evidence that excitonic lasing or free-carrier lasing are inherent and competing aspects of strongly confined systems. The prevalence of one of the two mechanisms can be forecast according to a thermodynamical treatment of the exciton and free-carrier phase diagram in thermal equilibrium. The key-parameter is

found to be the exciton binding energy renormalized by the many-body interactions and by the finite temperature of the distribution functions. This controls the dissociation of the exciton phase into the electron-hole plasma phase and, in turn, determines the main stimulated emission mechanism.

Such an approach is quite general, and can be applied to any material system having large electrostatic and thermal stability of the exciton. This is based on the evaluation of the many body-corrections to the exciton binding energy under the typical operation conditions of the device, i.e. the temperature and density of injected elementary excitations (no matter whether these are electrically or optically injected). For weakly confined systems excitons are screened quite promptly with increasing density or temperature, thus leading to the usual free-carrier recombination [14, 15]. As soon as the confining effects become large excitons become more stable. Under this condition they coexist with free-carriers and eventually prevail under the operating condition of the device.

The equilibrium between the two phases is established by a mass-action law, and depends on the equilibrium temperature of the whole system and on the density dependent exciton binding energy $E'_b(n_{eh}, n_{exc})$. This results in a set of coupled equations for the total density of photogenerated elementary excitations and for the balance of free-carriers and excitons [10]:

$$N = n_{eh}(T) + n_{exc}(T) \quad (1)$$

$$\frac{n_{eh}^2}{n_{exc}} = \frac{m_e m_h k_b T}{2\pi\hbar^2(m_e + m_h)} \exp\left(\frac{-E'_b[n_{exc}(T), n_{eh}(T)]}{k_b T}\right) \quad (2)$$

N is the total density of elementary excitations, given by the sum of free-carrier (n_{eh}) and exciton density (n_{exc}) and E'_b is the renormalized exciton binding energy.

Unlike the common case of III–V heterostructures, both the excitonic and free-carrier energy renormalization have to be included in E'_b because the dense exciton gas and free-carrier gas coexist in II–VI quantum wells (see Ref. [10] for details). Though the exact evaluation of these many-body corrections is a rather difficult computational task, first-order (perturbative) approximations can reasonably be used in order to estimate the density dependence of E'_b , which reads

$$E'_b(n_{exc}, n_{eh}) = E_b - \Delta E_g(n_{eh}, n_{exc}) - \pi a_0^2 [3.84 n_{exc} + 13.1 n_{eh}] * E_b \quad (3)$$

where a_0 is the unperturbed quasi-two-dimensional Bohr radius of the exciton. $\Delta E_g(n_{eh}, n_{exc})$ is the band gap renormalization, which includes the contribution of free-carriers (obtained by evaluating the exchange and coulomb-hole

self-energies given in Ref. [16]) as well as exciton populations. This is obtained by solving analytically [10]

$$\Delta E_g(n_{\text{exc}}) \simeq 256\pi \frac{e^2}{\varepsilon} n_{\text{exc}} \int_0^\infty \frac{k}{k + \chi} \frac{a_0^2}{[4 + (a_0 k)^2]^3} dk \quad (4)$$

The inverse screening length χ was computed following Ref. [16]. The third term of eq. (3) includes the hard-core exchange repulsion and the effects of phase space filling on the exciton energy due to the presence of the other excitons and free-carriers.

The above model can be used to evaluate the phase diagram self-consistently, and for the quantitative analysis of pump and probe transmission experiments. Typical results of these experiments are shown in Fig. 1 together with the calculated curves. The sample investigated in this figure has a rather large exciton binding energy, of the order of 37 meV. Stimulated emission occurs at power densities of the order of 5 kW cm^{-2} , when the exciton resonance is still well resolved in the pump and probe spectra.

A systematic comparison of the pump and probe and stimulated emission spectra in samples of different exciton binding energy reveals that the stimulated emission threshold can be either larger or smaller than the saturation intensity, depending on the exciton stability of the investigated sample (i.e. on the size and composition of the quantum well). Two limiting cases can be clearly identified:

(a) The deep and narrow quantum wells ($E_b \geq \hbar\omega_{\text{LO}}$, where $\hbar\omega_{\text{LO}} = 31.8 \text{ meV}$ is the LO phonon energy) exhibit stimulated emission at power densities such that distinguish-

able excitonic features can still be observed in the pump and probe spectra, i.e. $I_{\text{lasing}} < I_{\text{sat}}$. For example, the estimated density of excitons in Fig. 1 around lasing threshold is about 80% of the total particle density.

(b) In relatively shallow and wide quantum wells ($E_b \leq 25 \text{ meV}$) lasing occurs at $I_{\text{lasing}} > I_{\text{sat}}$, when the exciton resonance is saturated. In the wide range of intermediate sizes and compositions ($E_b \simeq \omega_{\text{LO}}$) I_{sat} and I_{lasing} are comparable, preventing a straightforward correlation between the two processes. Qualitatively, this indicates that above the lasing threshold excitons can play a role only in the strongly confined quantum wells, whereas they are already ionized in shallow potential wells.

The spectral shape of the non-linear absorption is calculated assuming a simple $1/(1 + N/N_{\text{sat}})$ saturation law for the oscillator strength (N_{sat} is the saturation density), a Gaussian resonance for the excitonic resonance and a step-like interband continuum modified by the Sommerfeld factor (curves in Fig. 1). The spectra are calculated self-consistently with eqs (1)–(4), sharing the total density N between excitons and free-carriers at any injection level. The hot-carrier temperatures were directly obtained from the slope of the high-energy tails of the luminescence spectra recorded under the same experimental conditions of the pump and probe measurements (typical values ranged between 2 meV and 5 meV in the investigated excitation intensity range, and have been found to vary roughly sub-linearly with the total density N).

In Fig. 2 we display the phase diagrams obtained from the analysis of the pump and probe spectra of the deep quantum well sample studied in Fig. 1, together with that obtained from a shallow sample with bulk-like exciton binding energy [Fig. 2(b), with $E_b \simeq 25 \text{ meV}$]. A careful inspection of these diagrams elucidates several important aspects of the exciton/free-carrier coexistence:

(i) A smooth and continuous phase transition occurs with increasing injection rate, reflecting the progressive transformation of the Coulomb correlated electron-hole pair into an unbound couple of carriers. This is ultimately controlled by the total density N and by the carrier temperature (i.e. by the resonant or off-resonant optical excitation). The extent to which the phase transition has proceeded at the lasing threshold density determines the dominant recombination process in II–VI quantum well lasers. The boundary between the exciton phase and the plasma phase changes dramatically with the hot-carrier temperature. The smaller the temperature the higher the critical density of the transition.

(ii) The carrier density (n_{lasing}) corresponding to the stimulated emission threshold (I_{lasing}) ranges between $5 \times 10^{10} \text{ cm}^{-2}$ and $9 \times 10^{10} \text{ cm}^{-2}$ in most samples. These values clearly fall in the exciton-rich phase for the deep quantum wells and in the free-carrier-rich phase for the shallow samples. For the intermediate sizes and compositions, the actual lasing mechanism can be tuned by varying the initial excess energy of the photoinjected carriers (carrier temperature) or the excitation intensity, leading to a subtle competition between the two recombination mechanisms.

(iii) The density at which the exciton resonance is totally bleached corresponds to the condition $n_{\text{exc}}/N_{\text{eh}} \simeq 1$ (white arrows in Fig. 2), i.e. to the transition from exciton-rich to free-carrier-rich phase. The exciton oscillator strength van-

Fig. 1. Experimental (symbols) and calculated (curves) pump and probe transmission spectra of a ZnCdSe/ZnSe multiple quantum well sample (well width 3 nm, Cd content 26%), measured at 10 K and under different pump power densities. The table quotes the total density N and the ratio between free-carrier and exciton populations ($n_{\text{eh}}:n_{\text{exc}}$) obtained from the line shape analysis of the pump and probe spectra at different pump intensities.

Fig. 2. (a) Phase-diagram of the exciton-free carrier gas for the sample investigated in Fig. 1. The dashed vertical line indicates N_{sat} expected for free-carriers screening only. n_{exc} and n_{eh} are the exciton and electron-hole pair densities. n_{lasing} (full arrows) indicates the carrier density corresponding to the threshold intensity for stimulated emission. The empty arrow indicates the density for total exciton bleaching obtained from the line shape analysis of the pump and probe spectra; (b) Same as in Fig. 2(a) but for the shallow quantum well sample of binding energy $E_b \approx 25$ meV (well width 11 nm, Cd content 11%).

ishes around $1.3 \times 10^{11} \text{ cm}^{-2}$ and $6.5 \times 10^{10} \text{ cm}^{-2}$ in the two samples investigated in Fig. 2(a) and (b) (empty arrows), respectively, consistent with the different exciton binding energies. These values are well below the N_{sat} value calculated for the usual electron-hole plasma condition [17] [dashed vertical lines in Fig. 2(a) and (b)], provided the actual carrier temperature and the excitonic interactions are taken into account.

An unambiguous confirmation of these results is obtained by measuring the diamagnetic shift of the stimulated emis-

sion in high magnetic field [9, 18]. A weak excitonic (parabolic) shift, of the order of $2 \mu\text{eV} \cdot \text{T}^{-2}$, is indeed observed in deep quantum well, whereas a linear Landau shift (of the order of 0.5 meV/T) typical of free-carrier recombination is observed in shallow samples with relatively weak exciton binding energy.

Finally, we would like to shortly comment about the general validity of the present model. The occurrence of excitonic lasing clearly needs an additional localization level in order to form the three level system. The localized exciton is supposed to be less sensitive to Coulomb screening thus having an enhanced screening threshold. Though this effect is not included in our phase diagram model, it can be phenomenologically accounted for by adding the average localization energy to the exciton binding energy. This will result in a further increase of N_{sat} . On the same way, we can evaluate the stability of the biexciton in II-VI quantum wells. Since the biexciton binding energy is of the order of 5 meV [19], simple thermodynamical arguments suggest that biexcitons cannot be responsible for lasing at temperatures above 60 K. This is indeed verified in several samples [20].

The above arguments are summarized in Fig. 3, where we plot a schematic phase diagram picture for II-VI quantum wells. The main reservoir of injected electrons and holes, reaches a steady state equilibrium condition with the exciton gas, when the exciton formation and dissociation rates are equal. This condition depends on the total density of particles N (through many-body renormalization), on temperature and on the unperturbed exciton binding energy. Localization of excitons occurs due to the presence of compositional disorder. The balance between free- and localized excitons can also be described by a phase diagram (left hand side of Fig. 3), provided two additional parameters are taken into account: the additional localization energy E_{loc} and the density of localization centers n_{loc} , which in turn depends on the degree of disorder existing in the structure. This latter quantity has been recently measured by time resolved spectroscopy and is found to be an increasing function of the Cd content in the well [21]. Finally the biexciton phase diagram (right hand side in Fig. 3) is described by the mass action law without inclusion of many-body renormalization, and adopting the biexciton binding energy E_b^{xx} as main stability parameter.

Fig. 3. Schematic phase diagram including localization of excitons and biexciton formation. See text for explanations.

In conclusion, we have shown that the competition between free-carrier and exciton stimulated emission in strongly confined heterostructures is inherent to the smooth phase-transition from excitonic to free-carrier gas occurring at high temperatures and/or in the presence of a dense carrier background. The phase transition is described by means of a self-consistent mass-action law including hot-carrier temperature and many-body renormalization of the exciton binding energy. Simple corrections can be applied in order to account for localized excitons or biexcitons.

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