Coherent exciton lasing in ZnSe/ZnCdSe quantum wells?

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A new mechanism for exciton lasing in ZnSe/ZnCdSe quantum wells is proposed. Lasing, occurring below the lowest exciton line, may be associated with a BCS-like condensed (coherent) exciton state. This state is most stable at low temperatures for densities in the transition region separating the exciton Bose gas and the coherent exciton state. Calculations show the gain region to lie below the exciton line and to be separated from the absorption regime by a transparency region of width, for example, about 80 meV for a 90 Å ZnSe/Zn0.75Cd0.25Se quantum well. Experimental observation of the transparency region using differential spectroscopy would confirm this picture. © 1995 American Institute of Physics.

Time-resolved pump-probe measurements of absorption in ZnSe/ZnCdSe quantum wells indicate that lasing in these systems may originate from exciton states. Excitons in this system are unusually robust; the binding energy (40 meV in a 90 Å quantum well) exceeds both $k_B T$ at room temperature and the LO-phonon energy (31 meV). This situation sharply contrasts with that in GaAs, where the quantum-well-enhanced binding energy can be 10 meV, and the LO-phonon energy is 36 meV. Carriers in the ZnSe/ZnCdSe systems relax quickly to quasiequilibrium distributions due to LO-phonon coupling.

Photon emission in this system exhibits two unusual characteristics: emission occurs at energies below the exciton line and at densities for which the excitons can no longer be regarded as independent. As a criterion for the importance of many-body exciton interactions, the characteristic exciton density $n_0$ is most simply defined as that for which the binding energy of an effective exciton in a Bose gas vanishes. In the presence of an increasing density of other excitons, the exciton wave functions begin to overlap. At low temperatures this overlap marks the transition to the coherent state we describe below, while at higher temperatures it marks the transition to an electron-hole plasma. Our value for $n_0$ agrees roughly with that expected on the basis of a Mott criterion. Both values are an order of magnitude smaller than a criterion used by Nurmikko1–3 based on densities when isolated excitons, characterized by density-independent Bohr orbits, first overlap. This letter will show how an excitonic picture survives, with modifications, at such high densities and how emission occurs below the exciton line.

The mechanism suggested by Nurmikko et al. in their seminal work1–3 requires an inhomogeneous exciton linewidth due to imperfections which substantially exceeds the homogeneous linewidth. The exciton states in their model are nonoverlapping and have a density below the electron-hole plasma transition. The occupied states lie at the bottom of the inhomogeneous exciton band. As a result lasing involves excitons lying below the center of the exciton line.

This mechanism could not be operate in the absence of crystal imperfections. We propose here a new mechanism for

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The Hamiltonian of this system is

\[ H = \sum_{\mathbf{k}} \epsilon_c(k) c_{\mathbf{c}, \mathbf{k}}^\dagger c_{\mathbf{c}, \mathbf{k}} + \sum_{\mathbf{k}} \epsilon_v(k) c_{\mathbf{v}, \mathbf{k}}^\dagger c_{\mathbf{v}, \mathbf{k}} \]  \tag{1}

where \( c_{\mathbf{c}, \mathbf{k}}^\dagger \) (\( c_{\mathbf{v}, \mathbf{k}} \)) creates a conduction electron (valence hole) with momentum \( \mathbf{k} \). Spin indices and sums are implied. Here

\[ \epsilon_c(k) = \frac{k^2}{2m_e} + E_G - \mu_e, \quad \epsilon_v(k) = -\frac{k^2}{2m_h} + \mu_h, \]

\[ \mu_{eh} = \mu_e + \mu_h, \]  \tag{2}

and \( E_G \) is the band gap; \( m_e \) and \( m_h \) are the effective masses. In the presence of the electron-hole interaction

\[ H_{e-h} = - \sum_{\mathbf{q}, \mathbf{k}, \mathbf{k}'} V(\mathbf{q}) c_{\mathbf{c}, \mathbf{k}+\mathbf{q}}^\dagger c_{\mathbf{v}, \mathbf{k}'} c_{\mathbf{v}, \mathbf{k}'}^\dagger c_{\mathbf{c}, \mathbf{k}}, \]

\[ \]  \tag{3}

a quasiparticle gap forms at the conduction-electron and valence-hole Fermi surfaces [shown in Fig. 1(b)]. Here \( V(\mathbf{q}) \) is the attractive effective electron-hole Coulomb interaction modified by screening due to the exciton polarizability. For our calculations we use a state-independent \( V(\mathbf{q}) \) whose magnitude and functional form is determined by the exciton model specified below.

The assumption of singlet pairing between electrons and holes having momentum \( \mathbf{k} \) and \( -\mathbf{k} \), respectively, leads to a gap equation:

\[ 2\Delta(k) = \sum_q V(q) \frac{2\Delta(k+q)}{E_{eh}(k+q)}. \]  \tag{4}

Here

\[ E_{eh}(k) = \sqrt{\left[ \epsilon_c(k) - \epsilon_v(k) \right]^2 + 4|\Delta(k)|^2}, \]

\[ \Delta(k) \] is analogous to the gap parameter in the BCS theory. The number of excitons in a quantum well having unit volume is

\[ n = \sum_k \frac{1}{2} \left[ 1 - \frac{\epsilon_c(k) - \epsilon_v(k)}{2E_{eh}(k)} \right]. \]  \tag{6}

The pairing assumption used here is analogous to the pairing assumption in superconductivity involving spin-up electrons of momentum \( \mathbf{k} \) and spin-down electrons of momentum \( -\mathbf{k} \). The gaps in the valence and conduction band labeled \( E_{eh}/2 \) in Fig. 1(b) each correspond to half the energy \( E_{eh} \) required to break up the exciton, one half being assigned to the electron \( e \) and hole \( h \), respectively.\(^5\)

The sketch in Fig. 1(b) satisfies the condition \( \mu_{eh} = E_G \) defining the characteristic density \( n_0 \). The valence-conduction band gap is seen to be smaller than the corresponding quantity \( E_G \) of Fig. 1(a) because of band-gap renormalization effects associated with the electron-hole Coulomb interaction. (The corresponding electron-electron and hole-hole interactions neglected here would further increase this gap shrinkage.) The gain and absorption regions are illustrated by the arrows marked “g” and “a,” respectively. The gaps \( E_{eh} \) produce a transparency regime, illustrated by the arrow “tr”, separating the two regions. (The transparency regime width 2\( \Delta \) defined by the exciton binding energy, which is of the order of the exciton binding energy, should be experimentally observable if this model is applicable to the present situation.) In addition, the enlarged density of states near the gap edge enhances the gain near the transparency edge. These features are qualitatively consistent with the observed lasing spectrum in the \( \text{ZnSe/ZnCdSe} \) quantum wells at energies well below the center of the exciton line.\(^1,3\)

Equation (4) has been solved for \( \Delta(k) \) for 30 and 90 Å quantum wells as well as the idealized two-dimensional case. The results are shown in Fig. 2. Since the heavy-hole, light-hole band splitting is substantial in these systems (79 meV in the 90 Å case), the independent subband model and the rod model, used by Young et al.\(^7\) are valid. These approximations have been used in quantitatively accurate calculations of exciton binding energies and optical absorption coefficients in III–V\(^8\) and II–VI\(^7\) superlattices and quantum wells. In the rod model, the interaction potential is approximated by considering the electron and hole to be rods having length equal to the well width. Screening effects of \( V(\mathbf{q}) \) due to the presence of other excitons are small within this approximation, particularly for the 90 Å quantum well, and will be neglected.

The fundamental absorption coefficient in the coherent state is given by\(^6\)

\[ \alpha(\omega) = \frac{2\pi^2 e^2 \hbar}{m e c \omega} \sum_k \left| \mathbf{e} \cdot \mathbf{p}_{c,v}(\mathbf{k}) \right|^2 \times \left[ \frac{1}{2} - \frac{\left[ \epsilon_c(k) - \epsilon_v(k) \right]^2}{2E_{eh}(k)} \right] \delta(\omega - \mu_{eh} + E_{eh}(k)) \]

\[ + \left[ \frac{1}{2} + \frac{\left[ \epsilon_c(k) - \epsilon_v(k) \right]^2}{2E_{eh}(k)} \right] \delta(\omega - \mu_{eh} - E_{eh}(k)) \].  \tag{7}

Here \( n \) is the index of refraction, \( \mathbf{e} \) is the photon’s polarization, and the momentum matrix element \( \mathbf{p}_{c,v}(\mathbf{k}) \) is obtained from band-structure calculations using a \( \mathbf{k} \cdot \mathbf{p} \) method.\(^7,8\)
sured at low temperatures, which is presumably associated
with inhomogeneous broadening. Each curve shows a
sharply peaked gain region and a corresponding absorption
region below and above $\mu_{eh}$, respectively. In a collisionless
ideal quantum well these regions would be separated by a
transparency region of width $2E_{eh}$ around $\mu_{eh}$. This feature
is somewhat blurred by the phenomenological linewidth. By
contrasting the gain at $3.7 \times 10^{11}$ cm$^{-2}$ with that at $4.1 \times 10^{11}$ cm$^{-2}$, just above $n_0$, it is clear that a pronounced
increase in gain occurs in the vicinity of the characteristic
density.

The peak is less than a meV wide and very large in the
absence of broadening effects. For $n > n_0$ and in the absence of
broadening, the gain has a square-root singularity at the
transparency edge. This divergence is associated with the
flat-band regions on either side of the quasiparticle gaps
shown in Fig. 1(b). Because of the sensitivity of the peak
to broadening, the calculated threshold density is also
sensitive to this effect. The threshold density corresponding
to $100$ cm$^{-1}$ gain, the value cited in Refs. 1–3, is calculated
to be $3.3 \times 10^{11}$ cm$^{-2}$, and lies just 20% below $n_0$. In these
experiments, the typical excitation density by optical pumping
was $5 \times 10^{11}$ cm$^{-2}$. The experimental threshold densities
were estimated to be less than $7.3 \times 10^{11}$ cm$^{-2}$, in agree-
ment with the present estimates.$^{1–3}$

The coherent exciton state may therefore provide an al-
ternative explanation to the proposed inhomogeneous-
linewidth theory of lasing in ZnSe/ZnCdSe quantum wells.
Since this system is the first lasing semiconductor having
excitons that are sufficiently robust at ordinary temperatures,
these alternative explanations are both important. Both view-
points lead to lasing below the exciton line. The coherent
exciton state becomes important at the characteristic density
$n_0$. Numerical calculations of the gap parameter characteriz-
ing this state yield results that bear qualitative similarity to
experiment. An experimental attempt to measure the quasi-
particle gap with IR radiation, akin to that first used to mea-
sure the superconducting gap,$^9$ may be used to decide the
issue. The experiment in this case, however, is far simpler
since the magnitude of the gap is about 40 meV, which
should be observable even in the presence of broadening
effects using differential optical spectroscopy.

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