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The Emission Spectra due to Exciton-Exciton Collisions in GaAs/AlGaAs Quantum Well System

Surendra K Pandey

***Abstract*—** Optical emission based on excitonic scattering processes becomes important in dense exciton systems in which the average distance between excitons is of the order of a few Bohr radii but still below the exciton screening threshold. The phenomena due to interactions among excited states play significant role in the emission near band edge of the material. The theory of two-exciton collisions for GaAs/AlGaAs quantum well systems is a mild attempt to understand the physics associated with the optical spectra due to excitonic scattering processes in these novel systems. The four typical processes considered give different spectral shape, peak position and temperature dependence of the emission spectra. We have used the theory of scattering together with the second order perturbation theory to derive the radiative power spontaneously emitted at an energy $\hbar\omega$ by these processes. The results arrived at are purely qualitative in nature. The intensity of emitted light in quantum well systems varies inversely to the square of temperature, whereas in case of bulk materials it simply decreases with the temperature.

***Keywords*—** Exciton-Exciton Collisions, Excitonic Scattering Processes, Interacting Excitonic States, Quantum Wells.

I. Introduction

STRONG enhancement of excitonic effects in the optical spectra of quantum well (QW) systems is observed due to carrier confinement [1]. Radiative recombination processes based on interacting excitonic states, become important in dense exciton systems in which the average distance between excitons is of the order of a few Bohr radii but still below the exciton screening threshold. Under these conditions van der Waals like interactions between excitons play significant role in the spontaneous emission near the band edge of the material. The spontaneous radiative decay of excitons and excitonic molecules (biexcitons) induced by inelastic scattering between quasi-particles, are the phenomena which contribute to the characteristic optical spectra for medium density of excitons in quantum wells [2].

The main feature of the collective processes in semiconductors is that they result in a modified ground level for the crystal, and the resulting emission always occurs at lower energy than the intrinsic emission of the unperturbed crystal. This qualitatively reflects the amount of energy spent by the interaction responsible for the formation of some new quasi-particles in the crystal. In terms of mean inter-particles distance r_s and the exciton Bohr radius a_0, the medium or intermediate density regime is characterized as $r_s \geq a_0$. Two-exciton collision processes have been reported in various semi-conducting compounds and also suggested to be important in the optical amplification or the laser action [3]-[4]. A below band gap emission band (P band) observed in CdS, CdSe and ZnO crystals have been interpreted in terms of inelastic collision process of two excitons leaving a free electron-hole pair and a luminescent photon. Moriya *et al.* have developed a detailed theory for the emission spectra due to exciton-exciton collisions in bulk GaAs at non-zero temperatures [5]. An extension of the theory for GaAs/AlGaAs quantum well systems provides some striking qualitative results.

We consider four typical exciton-exciton scattering processes to produce spontaneous emission, which occur in bulk semiconductors also [2]. These are symbolically expressed as:

(i) $(E_K^{1S}, E_{K'}^{1S}) \rightarrow (\hbar\omega, \text{e-h})$
(ii) $(E_K^{1S}, E_{K'}^{2S,2P}) \rightarrow (\hbar\omega, \text{e-h})$
(iii) $(E_K^{1S}, E_{K'}^{1S}) \rightarrow (\hbar\omega, E_{K''}^{1S})$
(iv) $(E_K^{1S}, E_{K'}^{1S}) \rightarrow (\hbar\omega, E_{K''}^{2S,2P})$

where $\hbar\omega$ is the photon energy, $E_K^{1S,\,2S,\,2P}$ correspond to the energies of 1S, 2S, 2P excitons with momentum $\hbar K$, and (e-h) is the energy of an electron-hole pair in the continuum state.

The use of scattering theory together with the second order perturbation theory under desired approximations gives the expression for radiative power spontaneously emitted at an energy $\hbar\omega$ by these processes. Our results and the subsequent explanations are purely qualitative in nature.

II. Excitonic Scattering Processes

In highly excited semiconductors the phenomena like exciton-exciton or exciton-carrier scattering, formation of excitonic molecules, Bose condensation of excitons or excitonic molecules and transition to metallic electron-hole systems, play significant roles in the optical emission near the band edge of the material. In the four typical exciton-exciton scattering processes considered as (i)-(iv), we have denoted the photon energy as $\hbar\omega$, the binding energy of exciton as E_b, the energies of 1S, 2S, 2P excitons with momentum $\hbar K$ as

Surendra K Pandey is with the Department of Physics, Government Model Science College, Jabalpur (Madhya Pradesh) 482 001 India (Phone: +91 761 2678893, 2678737; Fax: +91 761 2621272; E-mail: skpandey.7008@gmail.com).

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$E_K^{1S,\ 2S,\ 2P}$ and the energy of an electron-hole pair in the continuum state as (e-h). We consider the exciton as a composite particle of an electron and a hole with two kinds of freedom of motion, the internal and the translational motions. The coordinate for the internal motion of excitons, r and the center-of-mass coordinate, R are related as

$$r = r_e - r_h; R = (m_e r_e + m_h r_h)/M \quad (1)$$

The bound states of internal motion are labeled by 1S, 2S, 2P and K, K′ are the wave vectors related to the translational motion of excitons. In a similar way, we have used k and k′ respectively for the internal and translational motions of the free e-h pair. They have the following relations with the electron and hole wave vectors

$$k_e = k + \frac{m_e}{M}k'; k_h = -k + \frac{m_h}{M}k' \quad (2)$$

where, M (m_e+m_h) is the translational motion mass of the exciton.

With these noitations, the scattering process (i) is described as a radiative scattering via the exciton-exciton and exciton-photon interactions with the initial, intermediate and final states as

Exciton |1S, K> + Exciton |1S, K′>
→ Exciton |1S, 0> + electron-hole pair |k, k′>
→ Photon ħω + electron-hole pair |k, k′>

The other three processes can also be described in exactly the same way. The interaction potential of two excitons is very complicated, because the exciton has internal structure and the direct Coulomb and exchange interactions among the electrons and holes must be taken into account. Because of the electrical charge neutrality of the exciton, however, this potential is considered to be of short-range nature like inter-atomic or inter-molecular force which spreads over only a small region around each particle. Therefore, the effective interaction potential is assumed of the form

$$U_{12}(R_{12}) = \frac{4\pi\hbar^2}{M}\bar{a}\,\delta(R_1 - R_2) \quad (3)$$

In (3), R_1 and R_2 are the center-of-mass co-ordinates of the two excitons with $R_{12} = R_1$-R_2, and ā is the direction independent effective scattering length of the excitons.

If N(ħω), n_K^{1S}, n_e and n_h are the mean occupation numbers of the photon, 1S exciton, electron and hole in their respective states, then under the assumption that they are much smaller than unity (which is the case except for very high excitation rate), Boltzmann distribution is a good approximation for both excitons and free carriers. For a QW structure and 2D density of states, we can safely assume the relations

$$n_K^{1S} = N_{1S}(2\pi\hbar^2/Mk_BT)\exp(-\hbar^2K^2/2Mk_BT) \quad (4)$$

and

$$n_{e,h} = N_{e,h}(2\pi\hbar^2/m_{e,h}k_BT)\exp(-\hbar^2k_{e,h}^2/2m_{e,h}k_BT) \quad (5)$$

where T is the effective temperature of each particle system and N_{1S}, N_e and N_h are the densities of 1S excitons, electrons and holes, respectively.

The four scattering processes outlined above can be described as:

(i) (E_K^{1S}, $E_{K'}^{1S}$) → (ħω, e-h): By the use of the second order perturbation theory, the radiation power spontaneously emitted at an energy ħω by this process is finally obtained as

$$I(\hbar\omega) = A\frac{N_{1S}^2}{(k_BT)^2}\exp\left[-\frac{\hbar^2k'^2}{2Mk_BT} + \frac{1}{k_BT}\left(E_0^{1S} - E_b - \hbar\omega - \frac{\hbar^2k^2}{2\mu}\right)\right] \quad (6)$$

where, N_{1S} is the density of 1S excitons and μ is the reduced mass of electron-hole system given by $\mu^{-1} = m_e^{-1} + m_h^{-1}$.

The emission peak will lie at an energy slightly lower than (E_0^{1S}-E_b) at low temperatures and the spectrum is symmetric. The shift in the peak (E_0^{1S}) due to this process is by E_b. In quantum well structures, E_b being larger, the shift will be by greater amount as compared to that in bulk.

(ii) (E_K^{1S}, $E_{K'}^{2S,\ 2P}$) → (ħω, e-h): At higher temperature, the population of excitons in the higher states increases, so that the collisions between 1S and 2S, 2P excitons are considered to become important [6]. The emission spectrum due to this process is peaked at [E_0^{1S} - (1/9) E_b] and the spectral shape is more symmetric. The spontaneous emission spectrum is given by

$$I(\hbar\omega) = A\frac{N_{1S}N_{2S,2P}}{(k_BT)^2}\exp\left[-\frac{\hbar^2k'^2}{2Mk_BT} + \frac{1}{k_BT}\left(E_0^{1S} - \frac{1}{9}E_b - \hbar\omega - \frac{\hbar^2k^2}{2\mu}\right)\right] \quad (7)$$

As T increases, the emission channel shifts towards higher energies and is reduced in peak amplitude.

(iii) (E_K^{1S}, $E_{K'}^{1S}$) → (ħω, $E_{K''}^{1S}$): The spontaneous emission spectrum for this process is calculated as

$$I(\hbar\omega) = B\frac{N_{1S}^2}{(k_BT)^2}\exp\left[-\frac{\hbar^2K''^2}{2Mk_BT} + \frac{1}{k_BT}\left(E_0^{1S} - \hbar\omega\right)\right] \quad (8)$$

The spectrum is completely symmetric with its peak at E_0^{1S}. With the increase in temperature, the band broadens symmetrically without change in its peak position.

(iv) (E_K^{1S}, $E_{K'}^{1S}$) → (ħω, $E_{K''}^{2S,\ 2P}$): The calculated spontaneous emission spectrum is

$$I(\hbar\omega) = B\frac{N_{1S}^2}{(k_BT)^2}\exp\left[-\frac{\hbar^2K''^2}{2Mk_BT} + \frac{1}{k_BT}\left(E_0^{1S} - \hbar\omega - \frac{8}{9}E_b\right)\right] \quad (9)$$

The peak position of the emission spectrum is [E_0^{1S} - (8/9) E_b], which is nearly close to that obtained by process (i). The band is not symmetric in shape in this process.

III. Results and Discussion

To summarize the results, we have found that the four typical processes considered give different spectral shape, peak position and temperature dependence of the emission spectra. In QW structures, the intensity of emitted light due to excitonic scattering processes decreases inversely to the square of temperature, whereas in case of bulk it decreases inversely with the temperature. The emission channels (ii) and (iv) are rather symmetric and located between E_0^{1S} and (E_0^{1S}-E_b), while the spectrum (iii), which is peaked at E_0^{1S}, is perfectly symmetric in shape. The channel (i) is asymmetric with a width broader than the others and is situated on the low energy side of (E_0^{1S}-E_b).

In the energy region between E_0^{1S} and E_0^{1S}-E_b, usually there exists emission due to many types of bound excitons as well as that due to free exciton or exciton-polariton. Further, emission bands arising from free carrier-exciton collision and biexciton recombination processes are expected to lie around this energy region. Therefore, distinct observation of emissions due to the exciton-exciton scattering processes (ii)-(iv) may be obstructed by these overlapped emissions. In the low energy side of (E_0^{1S}-E_b), there usually exist almost no bound-exciton emissions. Sometimes, emissions due to impurities and phonon-assisted exciton recombination appear in this spectral region.

In process (i), the emission peak will lie at an energy slightly lower than (E_0^{1S}-E_b) at low temperatures and the spectrum is symmetric. The shift in the peak (E_0^{1S}) due to this process is by E_b. In quantum well structures, E_b being larger, the shift will be by greater amount as compared to that in bulk. Process (ii) usually takes place at higher temperatures, where the population of excitons in the higher states increases, so that the collisions between 1S and 2S, 2P excitons become important. The emission spectrum due to this process is peaked at [E_0^{1S} - (1/9) E_b] and the spectral shape is more symmetric. The spectrum in process (iii) is completely symmetric with its peak at E_0^{1S}. With the increase in temperature, the band broadens symmetrically without change in its peak position. And, finally the peak position of the emission spectrum in process (iv) is at [E_0^{1S} - (8/9) E_b], which is nearly close to that obtained by process (i). The band is not symmetric in shape in this process.

Our results and observations are purely qualitative in nature. The experimental observations of Moriya *et al.* [5], Göbel *et al.* [7], and Cingolani *et al.* [8] are qualitatively in agreement with our findings.

IV. Conclusion

In quantum well systems of III-V semiconductors, the theory of exciton-exciton scattering processes suggests four typical phenomena of scattering between different excitons.

1. The emission channels (ii) and (iv) are rather symmetric and located between E_0^{1S} and (E_0^{1S}-E_b), while the spectrum (iii) which is peaked at E_0^{1S} is perfectly symmetric in shape. The channel (i) is asymmetric with a width broader than the others and is situated on the low energy side of (E_0^{1S}-E_b).
2. In the low energy side of (E_0^{1S}-E_b), there usually exist almost no bound-exciton emissions. Sometimes, emissions due to impurities and phonon-assisted exciton recombination appear in this spectral region. However, because of the broad characteristic low energy band-tail, the identification of process (i) is considered to be much easier.
3. In the energy region between E_0^{1S} and E_0^{1S}-E_b, usually there exists emission due to many types of bound excitons as well as that due to free exciton or exciton-polariton. Further, emission channels arising from free carrier-exciton collision and biexciton recombination processes are expected to lie around this energy region. Therefore, distinct observation of emissions due to the exciton-exciton scattering processes (ii)-(iv) may be obstructed by these overlapped emissions.
4. The peak position, symmetry of the spectrum and the temperature dependence of the spectrum in these four typical scattering processes are different and they cohesively depend on the exciton binding energy, which in turn, is a function of well width of QW systems.

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Surendra K Pandey obtained his B.Sc. degree from Science College, Patna in 1988, M.Sc. (Physics) from IIT Delhi in 1990, Ph.D. & LL.B. from R. D. University, Jabalpur in 2002 & 2006. He has qualified the Joint CSIR-UGC National Eligibility Test (NET) for Lectureship and the Graduate Aptitude Test in Engineering (GATE).

He joined the Madhya Pradesh Educational (Gazetted) Service under the Dept. of Higher Education, Govt. of MP as an Asst. Professor of Physics in 1993 and was directly recruited as Professor of Physics in 2012. He has 37 Scientific Publications to his credit. His current research interests include the Physics of Low Dimensional Systems.

Dr. Pandey is a life member of the Society of Physical Sciences, Veer Kunwar Singh University, Ara.