Exciton Green's Function Approach to Absorption in II-VI Semiconductor Quantum Wells

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The optical absorption spectra of undoped II-VI semiconductor quantum wells have been calculated by using the exciton Green's function formalism. Unlike traditional variational methods employed in most of the exciton binding energy studies, the method we use allows to take into account several exciton's states, included those of the continuum, and at the same time to consider both Coulomb electron-hole interaction and confinement potentials on the same feet. In particular, the system ZnSe/ZnS, whi has been proposed as a vanishing conduction-band offset quantum well, is addressed in this work. The optical absorption spectra are analysed as a function of the screening of the Coulomb interaction for two different effective depth of the electron well.

I. Introduction

Quantum well optoelectronics has become a very interesting and dynamic field of research because of its potential applications in devices such as improved lasers. Strong quantum-confinement effects widen the range of possibilities for electronic, and all optical devices. Indeed the confinement-induced strenghtening of the binding energy allows the presence of Wannier-Mott excitons up to room temperature[1]. This completely alters the shape and intensity of the absorption spectrum, comparing to that produced by interband transitions which involve uncorrelated carriers.

Recently, there has been considerable research interest in the optical properties of wide- gap II-VI semiconductors heterostructures. Among other things, experimental evidence from optical spectroscopy on this type of heterostructure suggested that the valence band offset in a $CdSe/Cd_{1-x}Zn_xSe$ or CdTe/ZnTe superlattices[2-4] is rather small. An exciton created inside such a quantum well by optical excitations consists of a quasi-two-dimensional electron in the conduction band interacting with a quasi-three-dimensional hole in the valence band. The system ZnSe/ZnS[5,6], which has been proposed as a vanishing conduction band offset quantum well[7], in this case the created exciton consists of a quasi-two-dimensional hole in the valence band and a three dimensional electron in the conduction band. Accurate theoretical description on such excitons is important both from a fundamental point of view as for potential device applications. Sec. II deals with a sketcht of the relation between the absorption coefficient and the exciton Greens function for a quantum well with a general profile. The results for II-VI ZnSe-ZnS heterostructure are presented in Sec. III. The conclusions are given in Sec. IV. Only linear susceptibility is considered in this paper.

II. The theoretical approach

We present in this section only an outline of the method, which is reported in ref.[8]. We first solve the free carrier problem for electron and hole separately. The free electron and hole carrier states interact through the screened Coulomb force to form excitons. The exciton wave function is made up of linear combination of direct products of quantum well electron and hole eigenstates.

$$|X>=\Sigma_{nmk}\Phi^X_{nm}(\mathbf{r})|nm\mathbf{k}> \qquad (1)$$

where n and m are the electron and hole subband indices and $\mathbf{k}$ is the wave vector in the x-y plane. The linear optical susceptibility can be written as

$$\epsilon_0 X_{ij}(\Omega) = \Sigma_x \left(\frac{M^i_{gX} M^j_{Xg}}{E_x - \hbar\Omega - i\Gamma} \right) \tag{2}$$

The matrix element can be reduced from an N-particle operator form to a single particle operator form:

$$< nmk|eR|g > = < \Psi^c_{nk}|er|\Psi^h_{mk} > = \mu_{nm}(\mathbf{k}) \tag{3}$$

Ψ^h_{mk} and Ψ^c_{nk} are the eigenfunctions of the carrier in the finite quantum well. The optical matrix elements becomes

$$< X|eR|g > = \sum_{nmk} \Phi^X_{*nm}(\mathbf{k})_{nm}(\mathbf{k}) \tag{4}$$

The amplitude $\Phi^x_{nm}(\mathbf{k})$ satisfies the effective-mass equation for exciton in a quantum well

$$E_{nm}(\mathbf{k})\Phi^x_{nm}(\mathbf{k}) \tag{5}$$

$-\sum_{n'm'k'} < nm\mathbf{k}|V_{k-k'}|n'm' > \Phi^x_{n'm'}(\mathbf{k}') = E_X(\mathbf{k})\Phi^x_{nm}(\mathbf{k})$ where

$$E_{nm}(\mathbf{k}) = E_G + E^e_n(\mathbf{k}) + E^h_m(\mathbf{k}) \tag{6}$$

and

$$E^e_n(\mathbf{k}) = E^e_n + \frac{\hbar^2 k^2}{2m^*_e} \tag{7}$$

$$E^e_h(\mathbf{k}) = E^h_n + \frac{\hbar^2 k^2}{2m^*_h} \tag{8}$$

The exciton Greens function satisfies the following integral equation in momentum space

$$[E^e_n(\mathbf{k}) - \Omega]G_{nm}((\mathbf{k},\Omega) - \Sigma_{n'm'k'} < nm|V_{k-k'}|n'm' > G_{nm}((\mathbf{k},\Omega) = \mu_{nm}(\mathbf{k}) \tag{}$$

The singularity of the Coulomb potential must be taken into account properly. Here we assume that G depends on the magnitude of $\mathbf{k}$ only, since only the s states of the exciton wave functions contribute to the linear absorptio We then can obtain the imaginary part of dielectric function as

$$\epsilon_2 = Im \left[\frac{2}{V} \sum_{nmk} \mu_{*nm}(\mathbf{k}) G_{nm}((\mathbf{k}), \hbar\Omega + \Gamma) \right) \tag{10}$$

and then the absorption coefficient as

$$\alpha = \frac{\Omega\epsilon_2}{n_R c_0} \tag{11}$$

III. Results

Wide gap semiconductors such as ZnSe and ZnS_xSe_{1-x} are direct semiconductors. Their gaps correspond to an operation in the blue region of the visible light. It has been assumed that for electrons a potential well does not exist in the conduction band. Therefore the whole band offset equals the valence band discontinuity. Thus only holes are affected by the quantum size effect. The strained II-VI system ZnSe-ZnS has been proposed as a good candidate material to observe new optoelectronic effects due to the fact that some optical transitions are allowed in it while they are forbidden in other II-VI heterostructures. Excitons in this system have been first proposed[9,10] and experimentally found[11].

We first apply the Green's function formalism to optical absorption coefficient for heavy excitons, including the Coulomb interaction, for the case of an electronic well 59.2 meV and 30 meV, and fixed values for effective masses: $m^*_e(\text{ZnSe}) = 0.19$, $m_0, m^*_h(\text{ZnSe}) = 0.528$, $m_0, m^*_e(\text{ZnS}) = 0.367$, $m_0, m^*_h(\text{ZnS}) = 0.569$, m_0, refraction index = 8.7, and effective Rydberg $E_0 = 25.088$ meV. We assume a electronic well different from zero because the behaviour of electrons in the conduction band differs from the complete free particle one due to the variation of the electronic effective mass between these two materials without scattering events produced

by any potential discontinuity. The confinement properties of the lowest conduction states in this case are very different from the particle in a box model. For comparison purposes only one conduction subband and one heavy-hole subband are considered since the other methods usually asume the same asumptions, which is a good approximation when the subband energy level difference is larger than the exciton binding energy. This is true in our case. In Fig.1 the horizontal axes represents energies of the incident photons respect to the effective gap of the quantum well: E_G is the gap of the well material, E_{0e} and E_{0h} are the groung state energies of electron and hole in their corresponding wells, and E_0 is the effective Rydberg. We illustrate the dependence of absorption from Coulomb interaction screening. The lower continuos curve shows the absorption neglecting completely the Coulomb interaction. It corresponds exactly to the joint density for the transition. On the contrary the upper continuos curve is the optical absorption with Coulomb interaction without any screening (un-doped weakly optical excited systems). Between these two extremes we can observe the variation of the absorption as function of the intensity of the optical excitation, which corresponds to different screening levels for the Coulomb interaction depending on the density of electron-hole pairs creating by light. We observe that with increasing interaction the exciton formed has larger binding energy, because the excitonic absorption peak shifts to lower energies with respect to the gap and the intensity of absorption increases as well (oscillator strength). With lower depth of the well we observe that (Fig.1b) both the binding energy of the exciton and the oscillator strength decrease. This result can be explained because electron confinement diminishes and so the exciton is the three dimensional limit.

The Greens function method used in this work has been previously tested in the III-V quantum wells[12] with very stimulating results such as oscillator strengh larger in 20 per cent than the calculated by using variational methods and thus in better agreement to the experiment. We choose this method because II-VI quantum wells have shown an increase in the exciton oscillator strength compared to the II-VI's therefore the calculation method should be sufficienty accurate to fit with experiment. In addition to the fact that exciton binding energy exceed the LO-phonon energy and their exciton absorption features exist up to room temperature.

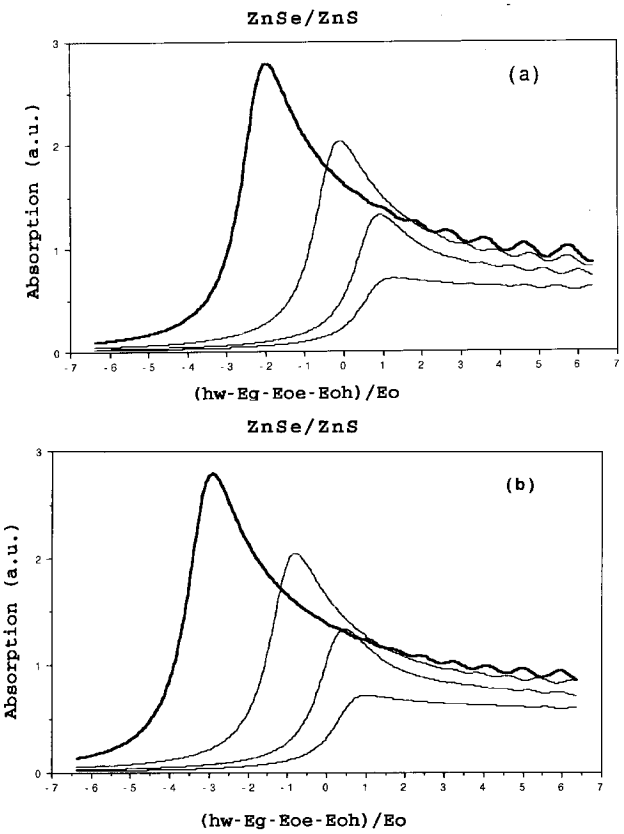

Figure 1. Absorption spectra of the ZnSe/ZnS quantum well as function of Coulomb interaction screening for an electronic quantum well of masses: m_e^*(ZnSe) = 0.19, m_0, m_h^*(ZnSe) = 0.528, m_0, m_e^*(ZnS) = 0.367, m_0, m_h^*(ZnS) = 0.569, m_0, refraction index = 8.7, and effective Rydberg E_0= 25.088 meV.: (a) 59.3 meV and (b) 30 meV.

IV. Conclusions

Absorption calculations are appreciated as a tool to characterize potential wells. We present the absorption spectra for the wide gap heterostructure system ZnSe/ZnS as function of the screening of the Coulomb interaction for two electronic-well depths. Our results allow a comparison with measurements in order to calibrate the role played by two fundamental effects: confinement and electrostatic interaction. Although at present there are no systematic experimental data available for direct comparison with our results, we consider that this is new step toward the understanding of opto-electronic structure of this important II-VI heterostructure.

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