Theoretical comparative study of negatively and positively charged excitons in GaAs/$Ga_{1-x}Al_xAs$ semiconductor quantum wells

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We compare the binding energies of the singlet ground states of negatively and positively charged excitons in a GaAs/$Ga_{1-x}Al_xAs$ semiconductor quantum well. We found that the binding energies are very close, contrary to what happens in three-dimensional and two-dimensional semiconductors where the positively charged exciton is always more stable than the negatively charged exciton. This result is in agreement with recent experimental observations.

Since the first observation[1] by Kheng *et al.*, much interest has been devoted to charged excitons in semiconductor quantum wells (QW). The existence of singlet (s) and triplet (t) negatively (X_s^- and X_t^-) and positively ($X_{2,s}^+$ and $X_{2,t}^+$) charged excitons has been widely reported in GaAs and CdTe heterostructures. The triplet states X_t^- and $X_{2,t}^+$ have only been observed using a magnetic field. However, generally speaking, the X^- and X_2^+ states cannot be observed in the same sample because the corresponding optical transitions require the presence of a free electron *or* a free hole gas. Thus it is difficult to compare the experimental binding energies. Recently it has been reported[2] how it is possible to control the density and type of the excess carriers in the QW by changing the illumination conditions and to study the X^- and X_2^+ spectra in the same sample. It was found[2] that the X_s^- and the $X_{2,s}^+$ binding energies are very close at zero magnetic field. At first sight, this result seems rather surprising. Indeed, it is well known that the hydrogen ion H^- is less stable than the hydrogen molecule ion H_2^+. Moreover, it has been shown[3] in bulk semiconductors that the $X_{2,s}^+$ state has always the highest binding energies. Nevertheless, this conclusion may be different in a QW, where the heavy holes are more localized than the electrons. To our knowledge, no theoretical comparison of the binding energies of the different kind of charged excitons in a QW has been reported up to now. Only the singlet state of the negatively charged exciton has been studied.[4]

In this paper we present a theoretical study of the $X_{2,s}^+$ states in a QW. In particular, we compare the binding energies of the singlet states of negatively and positively charged excitons in a GaAs/$Ga_{1-x}Al_xAs$ QW and show how the quantum confinement may explain the experimental observation.[2]

We extend here our previous theory[4] to the case of a positively charged exciton X_2^+. In a two-bands model, we assume that in the bulk materials, the constant energy surfaces in the reciprocal space are isotropic and spherical. Furthermore, we suppose that the electron and hole effective masses m_e^* and m_h^* are the same in the well and in the barrier materials, and we neglect the electron-hole exchange interaction. Within the envelope-function approximation, the effective-mass total Hamiltonian of the system consisting of two holes (1, 2) and an electron (e) confined in a well material with thickness L reads

$$H=-\frac{\hbar^2}{2m_h^*}(\Delta_1+\Delta_2)-\frac{\hbar^2}{2m_e^*}\Delta_e - \frac{e^2}{\kappa}\left(\frac{1}{r_{1e}}+\frac{1}{r_{2e}}-\frac{1}{r_{12}}\right)+V_w, \tag{1}$$

where r_{1e}, r_{2e}, and r_{12} are the distances between the three particles. The effective dielectric constant κ, assumed to be the same in the two materials, takes into account possible polarization effects. The well potential V_w arising from the band offsets is modeled by the sum of three square-well potentials. The total envelope energy reads

$$E^{tot}=E^{rel}+\frac{\hbar^2 K_{0,p}^2}{2M_0}, \tag{2}$$

where E^{rel} is the ''relative'' envelope energy, $\mathbf{K}_{o,p}$ is the in-plane wave vector of the center of mass, and $M_0=2m_h^*+m_e^*$ is the total effective mass of the system. We determine the relative energies in the frame of the variation method using a trial wave function similar to that we used previously[4] in the study of the negatively charged exciton. However, though this atomiclike wave function becomes less well adapted in the case of very heavy holes, i.e., $\sigma=m_e^*/m_h^*\ll 0.1$, it is expected to lead to reasonable results in the case of a GaAs/$Ga_{1-x}Al_xAs$ QW where $\sigma\simeq 0.2$. It reads

$$\psi=f_h(z_1)f_h(z_2)f_e(z_e)\exp[-ks/2-\alpha_h(z_1^2+z_2^2)-\alpha_e z_e^2] \times\sum_{lmnpqr} C_{lmnpqr}s^l u^m t^n (z_1^p z_2^q+z_1^q z_2^p)z_e^r, \tag{3}$$

where s, t, and u are the in-plane elliptic coordinates defined by

$$s=\rho_{1e}+\rho_{2e},\quad t=\rho_{1e}-\rho_{2e},\quad u=\rho_{12}, \tag{4}$$

$$0\leqslant s, -u\leqslant t\leqslant +u,\quad 0\leqslant u\leqslant s, \tag{5}$$

and ρ_{12}, ρ_{1e}, and ρ_{2e} are the in-plane distances between the particles. The functions $f_e(z)$ and $f_h(z)$ describe the ground states of the electron and the holes in a quantum well. The indices l, m, n, p, q, and r are positive integers or zero. In the case of singlet states, n must be even because of the symmetry of the envelope function against the permutation of the

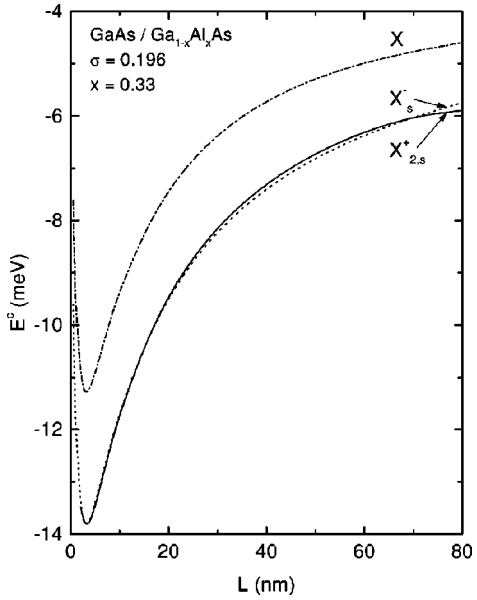

FIG. 1. Comparison between the X, X_s^-, and $X_{2,s}^+$ correlation energies against the well width L.

two holes. The variational parameters C_{lmnpqr}, α_e, α_h, and k are determined by minimizing the mean value of the energy. We have calculated the energies E of the singlet ground state of the charged excitons using a symmetrical 66-terms wave function defined by $l+m+n\leqslant 4$ and $\{pqr\}=(000,101,110)$. In order to estimate the influence of the quantum confinement on the Coulombic correlations, we have defined the ''correlations energies,'' $E_{X^-}^c$, $E_{X_2^+}^c$, and E_X^c of the charged excitons and the exciton

$$E_{X^-}^c = E_{X^-}^{rel} - 2E_e - E_h,$$
$$E_{X_2^+}^c = E_{X_2^+}^{rel} - 2E_h - E_e, \tag{6}$$
$$E_X^c = E_X^{rel} - E_e - E_h,$$

where E_e and E_h are the electron and hole ground-state energies in a quantum well. The conditions of stability against dissociation into an exciton and a free electron or hole read

$$W_{X^-} = E_{X^-}^c - E_X^c \leqslant 0,$$
$$W_{X_2^+} = E_{X_2^+}^c - E_X^c \leqslant 0, \tag{7}$$

where $-W_{X^-}$ and $-W_{X_2^+}$ are the X^- and X_2^+ binding energies. In order to get the exciton energies, we have performed an analogous variational calculation using the wave function

$$\psi_X = f_e(z_e) f_h(z_h) e^{-\beta\rho - \beta_e z_e^2 - \beta_h z_h^2} \sum_{pr} c_{pr} z_e^p z_h^r, \tag{8}$$

where β, β_e, β_h, and c_{pr} are variational parameters, p and r are positive integers chosen such that $p+r$ is even. It was found that the use of a nine-term wave function, defined by the condition $p+r\leqslant 4$, yields to an accuracy comparable with that we obtained for the trions.

We present here the results obtained in the case of a $GaAs/Ga_{1-x}Al_xAs$ QW with $x=0.33$. We use the following material data:[5] $m_e^*/m_0=0.0665$ for the electron mass and $m_{hh}^*/m_0=0.34$ for the heavy-hole mass. Thus, the value of the electron to hole effective-mass ratio amounts to $\sigma=0.196$. The band offsets are given by $V_e=Q_e\Delta\epsilon_g$ and $V_h=Q_h\Delta\epsilon_g$ where $Q_e=0.57=1-Q_h$. Further we assume that the band-gap difference $\Delta\epsilon_g$ and the aluminum concentration x are related by[6] $\Delta\epsilon_g=1.155x+0.37x^2$ eV and we use the value $\kappa=12.5$ for the dielectric constant.[7] In Fig. 1 we compare the correlation energies of the X_s^- and $X_{2,s}^+$ singlet states. It appears that they remain stable for all values of the well width L and that their correlation energies are very close. When L tends to infinity, we should get the ''exact'' excitonic three-dimentional (3D) value, $E_X=-4.84$ meV, and the 3D singlet charged excitons values we obtain using the method reported previously:[3] $E_{X_s^-}=-5.07$ meV, $E_{X_{2,s}^+}=-5.24$ meV. It appears that the values we obtain in this limit are a little bit higher than the 3D values. This is due to the fact that our wave functions are not well adapted to the 3D case. However, we expect to get better values for the binding energies, Eq. (7). From the 3D values of the binding energies: $-W_{X_s^-}=0.23$ meV and $-W_{X_{2,s}^+}=0.4$ meV, it results that in this limit $X_{2,s}^+$ is more stable than X_s^-. This behavior is well known in atomic physics, where the hydrogen ion H^- is less stable than the hydrogen molecule ion H_2^+. However, at first sight, it may seem rather surprising that for lower L values, the X_s^- and $X_{2,s}^+$ correlation energies become very close, but this result is in agreement with recent observations[2] of X_s^- and $X_{2,s}^+$ in the same sample. It may be explained by the fact that the quantum confinement localizes more the heavy holes in the X_2^+ states than the electrons in the X^- states. Thus the Coulomb repulsion is more important for the holes than for the electrons. This explains why the X_2^+ state becomes less stable in a QW than in a 3D material. It must be stressed that this result is largely insensitive to the mass ratio as it results from calculations carried out in the case of $0\leqslant\sigma\leqslant 1$, and thus does not much depend on the choice of the material parameters.

As a conclusion, we have shown that in a quantum well the negatively and positively charged excitons correlation energies are very close in agreement with the experimental observations.[2]

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