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Acceptor binding energy in δ-doped GaAs/AlAs multiple-quantum wells

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A series of Be δ-doped GaAs/AlAs multiple-quantum wells with the doping at the well center were grown by molecular beam epitaxy. The photoluminescence spectra were measured at 4, 20, 40, 80, 120, and 200 K, respectively. The two-hole transitions of the acceptor-bound exciton from the ground state, $1S_{3/2}(\Gamma_6)$, to the first-excited state, $2S_{3/2}(\Gamma_6)$, have been clearly observed and the acceptor binding energy measured. A variational calculation is presented to obtain the acceptor binding energy as a function of well width. It is found that the experimental results are in good agreement with the theory. [DOI: 10.1063/1.1516872]

I. INTRODUCTION

Recently, studies of the internal transition of shallow impurities fresh impetus due to the potential applications in far-infrared detectors and a solid state terahertz laser.[1–5] Confinement of such impurities in quasi-two-dimensional GaAs/$Al_xGa_{1-x}As$ quantum wells (QWs) allows the tuning of these levels in a controlled way. For device applications the greater range the binding energy of the impurity can be tuned over the better. In particular it is desirable that the dipole-allowed *1s–2p* transition can be tuned to be outside of the main longitudinal optical phonon bands of the semiconductor.

In the GaAs beryllium is an acceptor species commonly used in devices, it is relatively stable with respect to diffusion and has a binding energy of 28 meV in the bulk.[6] It has been shown previously that when incorporated into GaAs/$Al_xGa_{1-x}As$ QWs this binding energy can be increased substantially. Furthermore, the use of δ doping avoids the extension of the impurity energy levels resulted from the distribution of the dopant atoms along the growth direct of the quantum wells. The electronic states and properties of the shallow acceptors in the GaAs/$Al_xGa_{1-x}As$ QWs are not understood as well as those of shallow donors as the valence band of bulk GaAs is fourfold degenerate and contains a cubic term that must be taken into account to describe the acceptor states. Theoretically, the calculation of the effect of confinement on the ideal acceptor state in GaAs/$Al_xGa_{1-x}As$ QWs was reported by Masselink *et al.*[7] Experimentally, Gammon *et al.* have observed the confinement-induced splittings of acceptor levels in the GaAs/$Al_xGa_{1-x}As$ QWs with the resonant Raman scattering experiments.[8] Subsequently, the transitions between splittings of the acceptor levels have also been investigated in detail with far-infrared transmission,[9] photoluminescence (PL), and photoluminescence excitation experiments.[10–14] However, for the Be δ-doped GaAs/AlAs multiple-quantum wells (MQWs) with doping at the well center, there have been few reports of either experimental or theoretical work. However, this system is of especial interest as it represents the maximum possible confinement for the acceptor states in the valence band and thus the maximum possible tuning range for the *1s–2p* transition of the acceptor. In this article we measure PL spectra at various temperatures for a series of GaAs/AlAs MQWs with Be δ-doped at the well center and the well width range from 30 to 200 Å. The acceptor binding energy has been measured experimentally. The acceptor binding energy is calculated using a variational calculation as a function of the quantum-well width and compared with the experimental results.

II. EXPERIMENT AND RESULTS

A series of Be δ-doped GaAs/AlAs MQWs were grown by molecular beam epitaxy with doping at the quantum-well center, on a semi-insulating (100) GaAs substrate in a VG V80 H reactor equipped with all solid sources. The growth of the layers was performed under exact stoichiometric condition using the technique of stoichiometric low-temperature growth,[5] which ensures high quality optical materials even at relatively low growth temperatures. Under these conditions, the quantum-well structures were grown at 550 or 540 °C and without interruptions at the quantum well interfaces, which ensured negligible diffusion of the Be δ layers. Prior to the growth of the MQWs a 3000 Å GaAs buffer layer was grown. Each of multiple-quantum well structures investigated contained a same 50 Å wide AlAs barrier, while every GaAs well layer was δ doped at the well center with Be acceptor atoms. The doping level and the main characteristics of each sample are summarized in Table I.

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TABLE I. Characteristics of the samples: the repeated period, the quantum-well width (L_w), the δ-doping Be concentration (P), and the growth temperature of the epitaxial layers (T)

Samples	Periods	L_w (Å)	P (cm^{-2})	T (°C)
RM1795	400	30	2×10^{10}	550
RM1794	200	100	5×10^{10}	550
R1303	50	150	2.5×10^{12}	540
R1392	40	200	2.5×10^{12}	540

Photoluminescence experiments were performed from liquid helium to room temperatures using Renishaw Raman imaging microscope. The samples were mounted on a cold finger of a continuous flow helium cryostat. The optical excitation for the PL experiments was provided by an argon-ion laser (5145 Å). The laser beam was focused onto a sample, and the light returning back from the sample was collected and passed into a spectrograph for analysis. The excitation was typically 5 mW.

The PL spectra with above-band gap excitation have been measured at the various temperatures for the samples in Table I. Figure 1 shows the PL spectra at 4, 20, 40, 80, 120, and 200 K for the sample RM1794. Three peaks are clearly resolved at 4 K, with positions located at 794, 795.88, and 806.26 nm, respectively. The first at 794 nm is the strongest peak and attributed to the transition of a heavy free exciton ($X_{\mathrm{CB1-HH1}}$). The second at 795.88 nm originates from the exciton recombination bound to the neutral beryllium acceptor (Be^0X). The energy separation between the $X_{\mathrm{CB1-HH1}}$ and the Be^0X is 3.7 meV, which is the energy required removing the exciton from the Be^0X complex. The intensity ratio of the Be^0X with the $X_{\mathrm{CB1-HH1}}$ is 0.76 at 4 K and decreases as the measuring temperature increases. As the temperature arrives at 80 K, the Be^0X peak is no longer detectable, and the light free exciton peak ($X_{\mathrm{CB1-LH1}}$) can be observed. In addition the third peak at 806.26 nm is weaker than the $X_{\mathrm{CB1-HH1}}$ and Be^0X, which is attributed to the free-to-bound recombination (eBe^0) between an electron of the $n=1$ quantized confinement level and a hole bound to a Be acceptor at the center of the GaAs well. As the measuring temperature rises to 40 K, the eBe^0 line becomes very weak.

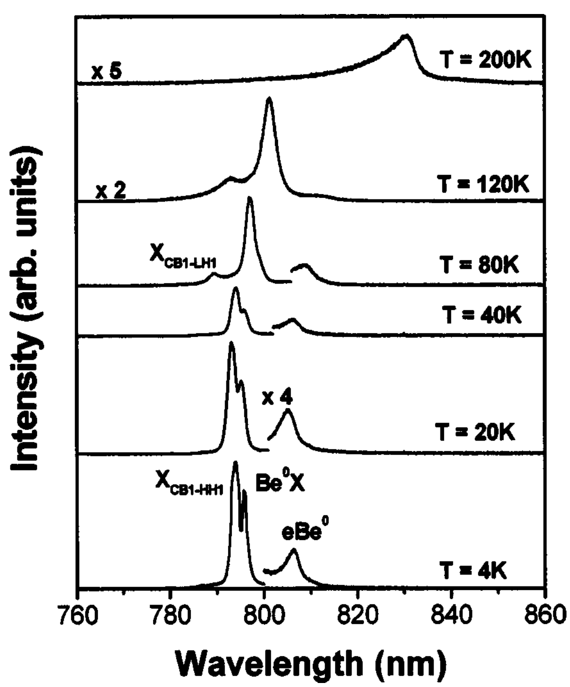

FIG. 1. Series of PL spectra for sample RM1794 ($L_w=100$ Å) at different measuring temperatures showing the $X_{\mathrm{CB1-LH1}}$, $X_{\mathrm{CB1-HH1}}$, Be^0X, and eBe^0 peaks.

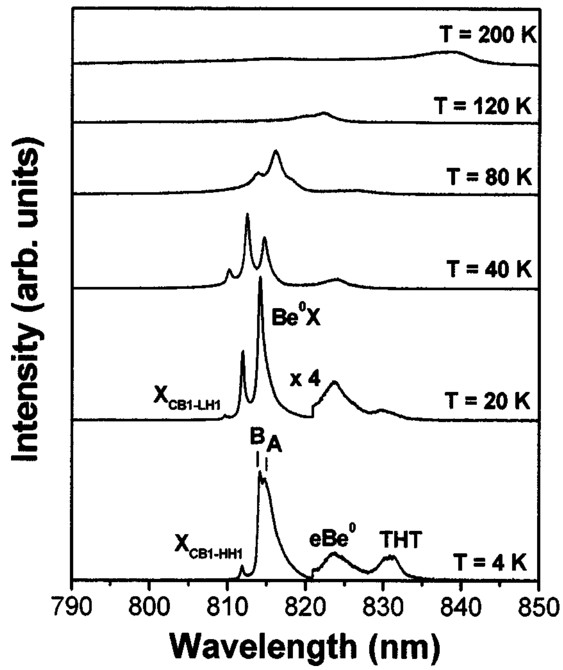

FIG. 2. Series of PL spectra for sample R1392 ($L_w=200$ Å) at different measuring temperatures showing the $X_{\mathrm{CB1-LH1}}$, $X_{\mathrm{CB1-HH1}}$, Be^0X, eBe^0, and THT peaks.

Figure 2 indicates the PL spectra of the sample R1392 at different measuring temperatures. In contrast to Fig. 1, the strongest peak at 4 K is attributed to the bound exciton, Be^0X, with a doublet structure labeled A and B. The energy splitting between A and B is 1.3 meV, which is 1.7 meV for a 100 Å wide $GaAs/Al_{0.3}Ga_{0.7}As$ QWs. [11] The B peak is due to an excited state of the Be acceptor bound exciton. For the sample R1303 with the same density of dopant atoms as the sample R1392 and instead of a 150 Å wide quantum well, the doublet structure is not observed clearly, but the bound exciton peak, Be^0X, has a weak shoulder. In Fig. 2 at the high energy side of the Be^0X is the heavy free exciton peak, $X_{\mathrm{CB1-HH1}}$, and the intensity ratio, $I_{\mathrm{Be^0X}}/I_{\mathrm{CB1-HH1}}$, is 6.8 at 4 K, greater than that of other three samples, respectively. This fact is mainly due to the reduction of the free exciton lifetime with decreasing quantum well width, since the probability for the electron-hole pairs to recombine as a free exciton instead of getting captured by impurities increases with decreasing quantum-well width.[15] In addition at the lower energy side of the eBe^0, a peak labeled THT is clearly observed and interpreted as the two-hole transitions (THT) of the acceptor bound exciton. As the exciton attached to the neutral acceptor atom recombines, there is a small probability that some of energy emitted is absorbed by the hole bound to the acceptor, which then undergoes a transition to an excited state leaving the acceptor, which binds the exciton, in an excited state instead of the ground state. The energy of this transition is equal to that of the separation between the Be^0X and THT, 29.5 meV. The THT is also observed in RM1795 and R1303 sample, but the THT signal intensity of the RM1795 is even weaker than that of R1303 and R1392 sample, respectively. This reason is attributed to the smaller concentration of dopant atoms and the smaller probability for the capture by an impurity, of an electron-hole pair in QWs instead of recombination as a free exciton. For the Be-doped GaAs/$Al_xGa_{1-x}As$ QWs structures, the degeneracy of the valence band is lifted at the Γ point due to the quantum well potential which reduces the point group symmetry from T_d to D_{2d}. Therefore, the ground state of the neutral acceptor Be at the center of a quantum well splits into two doublet levels, namely $1S_{3/2}(\Gamma_6)$ and $1S_{3/2}(\Gamma_7)$.[9] The THT peak in Fig. 2 is associated with the transition of $1S_{3/2}(\Gamma_6)\rightarrow 2S_{3/2}(\Gamma_6)$.

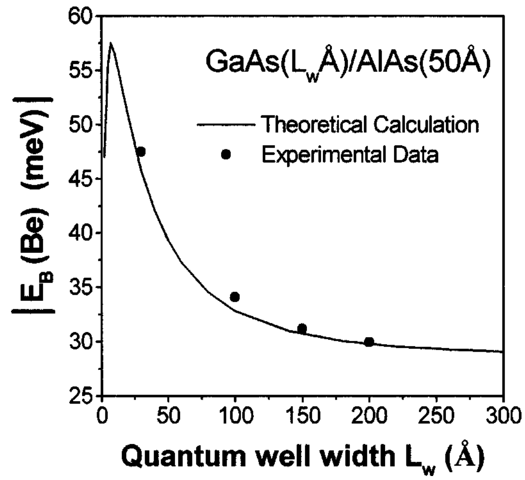

FIG. 3. Magnitude of the binding energy as a function of the quantum-well width (L_w) for neutral acceptors Be at the centers of the GaAs wells surrounded by AlAs barriers. The solid circle dots indicate the experimental data, and the curve represents the theoretical calculation.

III. CALCULATION AND DISCUSSION

A. Theory

Under the single-band effective mass and envelop function approximations, the Hamiltonian of a hole bound to the hydrogenic acceptor at position $(0,0,z_0)$ in the GaAs/AlAs MQWs can be written as

$$H=-\frac{\hbar^2}{2m^*}\nabla^2+V(z)-\frac{e^2}{4\pi\varepsilon_0\varepsilon_r r'}, \quad (1)$$

where m^* is the effective mass of a hole, ε_r is the relative dielectric permittivity, and $V(z)$ is the one-dimensional potential. The hole-acceptor separation r' is written as follows:

$$r'=\sqrt{x^2+y^2+(z-z_0)^2}. \quad (2)$$

The success of variational approaches centers around the general choice of the trial wave function. We choose the produce of two terms as the trial wave function of a hole, i.e.,

$$\Psi=\chi(z)e^{-r'/\lambda}, \quad (3)$$

where $\chi(z)$ is the wave function of the hole in the GaAs/AlAs MQWs without the acceptor impurity present and $e^{-r'/\lambda}$ is the simple hydrogenic wave function of the ground state, and where λ is known as the Bohr radius, but now it is employed as a variational parameter in order to minimize the total energy of the system. The Schrödinger equation corresponding to the Hamiltonian of Eq. (1) becomes

$$-\frac{\hbar^2}{2m^*}\{[\nabla_z^2\chi(z)]e^{-r'/\lambda}+2\nabla_z\chi(z)\nabla_z e^{-r'/\lambda}$$
$$+\chi(z)\nabla^2 e^{-r'/\lambda}\}-\frac{e^2}{4\pi\varepsilon_0\varepsilon_r r'}\chi(z)e^{-r'/\lambda}$$
$$+V(z)\chi(z)e^{-r'/\lambda}=E\chi(z)e^{-r'/\lambda}, \quad (4)$$

where E is the total energy of the system. The energy E can be solved for any choice of λ by expanding the derivatives in finite differences and forming an iterative shooting algorithm.[16] The variational parameter, λ, is varied systematically with the aim of minimizing the total energy. The acceptor binding energy, E_B, is equal to the difference between E and the ground-state energy, E_1, for the hole in the QWs without the acceptor impurity present, and hence,

$$E_B=E-E_1. \quad (5)$$

B. Results and discussion

In this subsection the numerical results of the binding energies of the Be acceptor at the center of the GaAs/AlAs MQWs as a function of the well width will be given. We use the formula $V(z)=0.33\Delta E_g(x)$,[16] where $\Delta E_g(x)$ is the difference in band gaps at $k=0$ between GaAs and $Al_xGa_{1-x}As$. $\Delta E_g(x)$ is taken to $1247x$ meV and x is the mole fraction of the $Al_xGa_{1-x}As$ barriers. Although the single-well potential $V(z)$ makes it more appropriate for completely decoupled quantum wells, it will also be appropriate for MQWs with thick barriers that no hole wave function can penetrate. The effective mass m^* is used as the heavy-hole mass of $0.62m_0$, the typical value in bulk GaAs, where m_0 is the mass of an electron in the free space. The relative dielectric permittivity, ε_r, is set to 17.2. In previous theoretical calculations, a great deal of attention has been paid to the roles of the effective mass and the dielectric constant mismatches at interfaces in semiconductor hererostructures, and their effects on the shallow impurity energy levels in the QWs.[17–19] With m^* and ε_r parameters given earlier, we deduce the acceptor binding energy of 28.0 meV for very large well widths, which is in good agreement with the experimental result in bulk GaAs.[6] Figure 3 illustrates the acceptor binding energy for the on-center Be δ-doped in GaAs/AlAs MQWs with a well width range from 5 to 300 Å. The solid circle dots show the acceptor binding energies gotten from the PL experimental measurements. According to the eBe^0 position of the PL spectra, the acceptor binding energy E_B(Be) can be determined using the relation[20]

$$E_B(\text{Be}) = E(X_{\text{CB1-HH1}}) + E_{BX} - E(\text{eBe}^0), \tag{6}$$

where $E(X_{\text{CB1-HH1}})$ and $E(\text{eBe}^0)$ are the energies of the $X_{\text{CB1-HH1}}$ and eBe^0 lines, respectively, and where E_{BX} is the binding energy of the heavy exciton and can be deduced from the accurate calculation of Andreani and Pasquarello.[21] In Fig. 3 it can be find that the theoretical calculation is in good agreement with the experimental results. The acceptor binding energy increases as the quantum-well width decreases, and has a peak with a maximum at $L_w = 7$ Å. This is due to the finite barrier and is similarly seen in quantized donor and exciton calculations.[22,23] For very narrow wells, the increasing confinement energy pushes the hole closer to the top of well, eventually forcing it to ''spill over the top,'' thus leading to decreases in the binding energy. As the well width increases, the acceptor binding energy tends towards its bulk value of 28 meV.[6] The acceptor binding energy in this article is consistent with that of the Γ_6 ground state in the literature.[7]

IV. CONCLUSIONS

We have investigated experimentally and theoretically the acceptor binding energy of a series of GaAs/AlAs MQWs δ-doped with Be at the center of wells. The PL spectra are measured at the various temperatures and the two-hole transitions of the acceptor-bound exciton from $1S_{3/2}(\Gamma_6) \rightarrow 2S_{3/2}(\Gamma_6)$ have been clearly observed. The binding energy of the shallow Be acceptor at the center of the MQWs has been measured experimentally. Using a variational calculation the acceptor binding energy has been calculated as a function of the quantum-well width. It is found that the experiment results are in good agreement with the theory.

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