Acceptor-related photoluminescence spectra of GaAs quantum-wire microcrystals: A model calculation

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The acceptor-related photoluminescence spectrum of a GaAs quantum-wire microcrystal is investigated theoretically using a model calculation within the effective-mass approximation, with the acceptor envelope wave functions and binding energies calculated by a variational procedure. Typical theoretical photoluminescence spectra show two peaks associated with transitions from the $n=1$ conduction-subband electron gas to acceptors at the on-center and on-edge positions in the wire, in good agreement with the recent experimental results by Hiruma *et al.* [Appl. Phys. Lett. **59**, 431 (1991)].

Understanding the electronic and impurity properties in semiconductor heterostructures has attracted both experimental and theoretical attention in the last two decades due to the wide-ranging potential applications of these structures in electronic devices. In particular, in the past few years, a considerable effort has been made in the study of the properties of impurity states in low-dimensional semiconducting structures having quantum confinement in one, two, or three dimensions, such as quantum wells (QW's), quantum-well wires (QWW's), and quantum dots (QD's).

Fabrication of QW's has been very successful, whereas fabrication techniques of QWW's and QD's are not as advanced. In the case of QWW's, although there have been considerable improvements in experimental techniques for the construction of GaAs QWW's, these techniques have not provided high-quality wires due to a number of reasons, including surface damage and interface fluctuations. Petroff *et al.*[1] used etching and epitaxial regrowth in a pioneering work on GaAs QWW's, while several groups have studied quasi-one-dimensional transport on laterally confined inversion layers.[2] QWW's have been fabricated by employing molecular-beam epitaxy (MBE),[3] ion-beam technology,[4] organometallic vapor-phase epitaxy (OMVPE),[5] and electron-beam lithography combined with reverse mesa wet etching.[6]

Recently, Hiruma *et al.*[7] have performed the optical characterization of GaAs quantum-wire microcrystals grown by OMVPE. A typical microcrystal (1–5 μm in length) would have a hexagonal-like cross section which would decrease in a stepped fashion, and with a diameter of 200 nm at the base to 10 nm at the peak. In their work, spectral features dominated by free-carrier to acceptor-impurity recombination have clearly appeared in the photoluminescence measurements. Hiruma *et al.*[7] have assigned the impurity features to the presence of carbon acceptors in the wire microcrystals. As the carriers are confined in a quantum-wire-shaped microcrystal surrounded by air, a modeling of the system with an infinite confining potential should be realistic. Moreover, one may think of a cylindrical quantum wire as the simplest way to describe one of the wire microcrystals in the experiment of Hiruma *et al.*[7] In this work we present, therefore, a model calculation of the photoluminescence spectra associated with transitions between the $n=1$ conduction-subband electron gas and the acceptor-impurity band for an infinite-barrier cylindrical GaAs QWW, in order to attempt a theoretical understanding of the experimental results by Hiruma *et al.*[7]

We consider a single GaAs QWW with a $P_a(\rho_i)$ distribution of acceptor impurities, in which electrons have been optically injected into the conduction band and recombine with holes in the acceptor band. We also assume that the temperature is low enough (e.g., $T=4$ K, such as in the experiment of Hiruma *et al.*[7]) to guarantee that each acceptor state is filled with a hole. The acceptor-related photoluminescence spectrum can be written as[8]

$$L_R(\omega)=2\pi\int_0^R d\rho_i\rho_i W_R(\rho_i,\omega)P_a(\rho_i)f(E_k)\ , \tag{1}$$

where $f(E_k)=1/\{1+\exp[\beta(\Delta-E_F)]\}$ is the Fermi distribution function for the conduction-subband electron gas, with $\beta=1/k_BT$ and E_F the quasi-Fermi-energy[9] level of the electron gas, measured from the bottom of the subband. In the above expression,

$$\Delta=\hbar\omega-\varepsilon_g+E_b(\rho_i,R)\ , \tag{2}$$

$$E_g(\text{eV})=1.519-\frac{5.405\times10^{-4}T^2}{T+204}\ , \tag{3}$$

where $\hbar\omega$ is the photon energy, E_b the binding energy of the acceptor impurity, and $\varepsilon_g=E_g+E_{10}^c+E_{10}^v$, with E_g being the temperature-dependent[10] bulk GaAs band gap, and E_{10}^c (E_{10}^v) the bottom (top) of the first conduction (valence) subband.

For an infinite-barrier GaAs QWW of radius R and length L, the transition probability per unit time for conduction-to-acceptor transitions associated with a single impurity at ρ_i is given by[11]

$$W_R(\rho_i,\omega)=W_0\left[\frac{(m_c)^{1/2}\hbar L}{2^{1/2}m_0a_0^2}\right]\times\frac{S_{fi}^2\left[\rho_i,\lambda,\left[\frac{2m_c\Delta}{\hbar^2}\right]^{1/2}\right]}{\Delta^{1/2}}\Theta(\Delta)\ , \quad (4)$$

where a_0 is the Bohr radius, m_0 is the free-electron mass, m_c is the conduction-band effective mass, $\Theta(\Delta)$ is the step function, and λ is the acceptor wave-function variational parameter.[12] In this expression, we have

$$W_0=\frac{4m_0}{\hbar^3}a_0^2|C|^2|\mathbf{e}\cdot\mathbf{P}_{fi}|^2\ . \quad (5)$$

in which $\mathbf{e}\cdot\mathbf{P}_{fi}$ is a matrix element,[11] $\mathbf{e}$ is the polarization vector in the direction of the electric field of the radiation, and C is a prefactor which contains the photon vec-

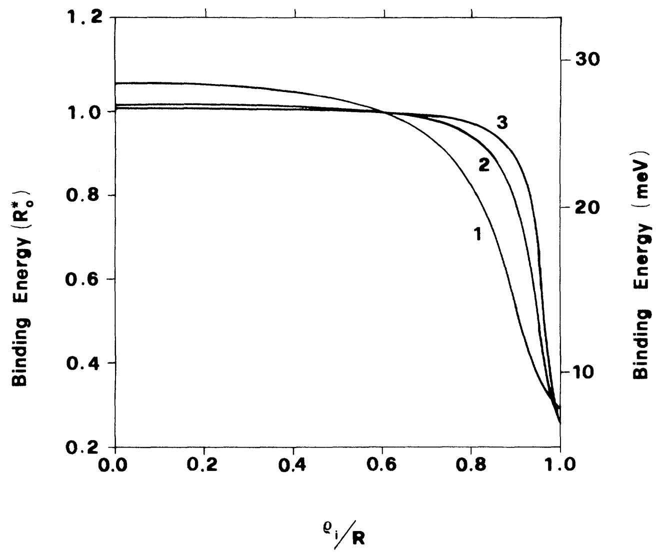

FIG. 1. Binding energies of shallow-acceptor impurities in infinite-barrier-potential cylindrical GaAs QWW's of radii R as functions of the ρ_i/R impurity positions. Curves 1, 2, and 3 correspond to QWW radii of 200, 400, and 600 Å.

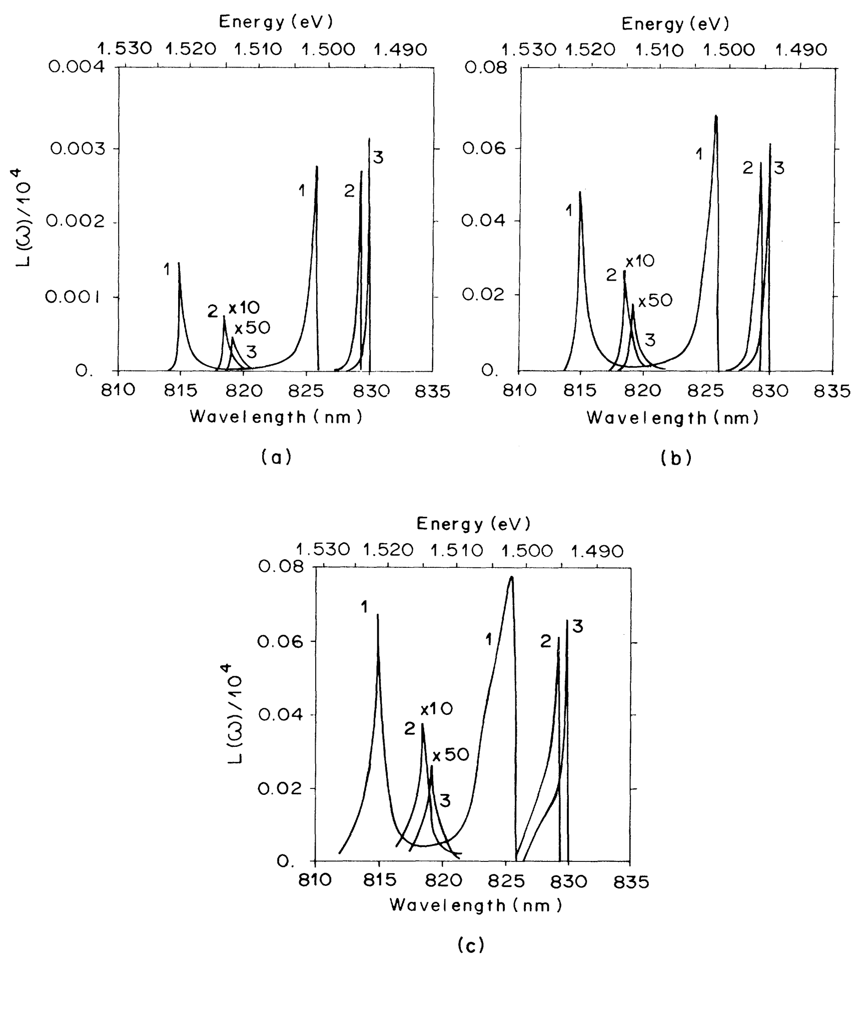

FIG. 2. Photoluminescence spectra L (in units of W_0; see text), at $T=4$ K, associated with electron-to-acceptor recombinations for infinite-barrier cylindrical GaAs QWW's and for different choices of the quasi-Fermi-energy levels: (a) $E_F=-1$ meV, (b) $E_F=1$ meV, and (c) $E_F=5$ meV. In each figure, curves 1, 2, and 3 correspond to QWW radii of 200, 400, and 600 Å, respectively.

tor potential. The expression for $S=S_{fi}$ in Eq. (4) is similar to the ones found in earlier work.[8,11]

In the calculation of acceptor states, we used an average spherical[13] effective mass $m_v \simeq 0.30 m_0$, which—together with a value of the dielectric constant $\varepsilon = 12.58$—would yield an effective Rydberg of 26 meV, which corresponds to the experimental binding energy of carbon acceptors in bulk GaAs, although a more realistic description should certainly consider the effects of the coupling of the top four valence bands.[14]

The doping profile used in our photoluminescence-line-shape calculations corresponds to a homogeneous distribution of acceptors along the well wire, i.e., $P_a(\rho_i) = 1/\pi R^2$, which we believe would be appropriate for comparison with the experimental situation in the work of Hiruma *et al.*[7], as there was no intentional doping.

The acceptor envelope wave functions and binding energies are obtained within an effective-mass variational procedure outlined in previous work;[12] results for the acceptor-position (ρ_i) dependence of the binding energies for different GaAs QWW radii are shown in Fig. 1. Note that values of the QWW radii of 200, 400, and 600 Å are compatible with the GaAs microcrystals' characteristics used in the study by Hiruma *et al.*[7]

The acceptor-related photoluminescence spectra for $R = 200$, 400, and 600 Å infinite-barrier cylindrical GaAs QWW's are shown in Fig. 2, for different values of the quasi-Fermi level[9] of the electron gas—associated to the $n=1$ conduction subband—in the quasiequilibrium steady state. As shown in previous work[9] for GaAs-(Ga, Al)As QW's, the quasi-Fermi energy E_F is related to experimental parameters such as the laser power, the temperature, and the profile and density of impurities, etc. For QWW's, there are no calculations of E_F and, therefore, we display our results for the impurity-related photoluminescence line shape in the cases of $E_F = -1$ (which would correspond to a Maxwell-Boltzmann distribution for the excited electron gas in the conduction subband), 1 (which is an estimate by Mahan and Oliveira[9] for QW's), and 5 meV.

In Fig. 2, one should note that results for the acceptor-related photoluminescence line shape present essentially two peaks, one at lower energies associated with impurities mostly at the QWW center, and another at higher energies corresponding to transitions involving acceptors at the QWW edge. Note also that the spectra in curves 2 and 3, which correspond to $R = 400$ and 600 Å, respectively, are multiplied by factors of 10 (for $R = 400$ Å) and 50 (for $R = 600$ Å) in the energy region corresponding to acceptors at the edge of the QWW. Of course, the spectra corresponding to $E_F = 5$ meV present peaks with larger halfwidths, as one would expect. It is clear from our results in Fig. 2 that the 830-nm peak in the experimental results of Hiruma *et al.*[7] is associated with free-electron-to-neutral-acceptor impurity recombination *with the acceptors close to the QWW on-center position,* as may be seen by examining the theoretical QWW on-center peaks at 829–830 nm, in the case where $R = 400$ and 600 Å.

In Fig. 3, we compare the experimental results of Hiruma *et al.*[7] with our model calculation for GaAs QWW's of radii 400, 600, and 1000 Å. It is clear that the experimental photoluminescence spectrum is consistent with a convolution of features characteristic of each QWW with a different radius. This is not surprising, as a typical GaAs microcrystal has a hexagonal-like cross section with decreasing "radii" going from 1000 to 50 Å. The 830-nm experimental peak agrees very well with the on-center acceptor theoretical peaks, corresponding to wells of $R = 400$, 600, and 1000 Å. The theoretical peaks associated with transitions from the conduction-subband electron gas to on-edge acceptors, for $R = 600$ and 1000 Å, are in good agreement with the experimental peak at 819.7 nm, although Hiruma *et al.*[7] have claimed this last structure to be due to neutral-acceptor–bound-exciton recombination. As there is another experimental structure at a slightly lower energy, and as our calculations do not, of course, include excitonic effects, more work would be necessary for a complete explanation of the experimental photoluminescence results of Hiruma *et al.*[7]

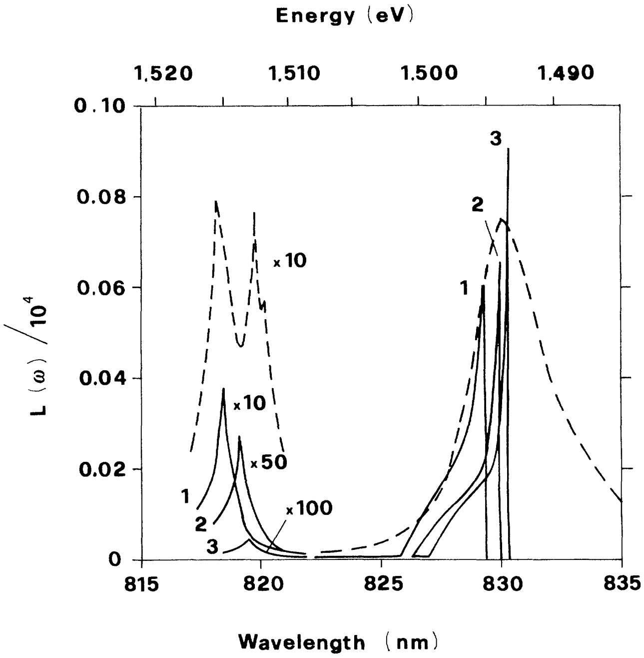

FIG. 3. Photoluminescence spectra L (in units of W_0; see text), at $T=4$ K, associated with electron-to-acceptor recombinations for infinite-barrier cylindrical GaAs QWW's, for a quasi-Fermi energy $E_F = 5$ meV, and for different choices of the QWW radii: curves 1, 2, and 3 correspond to QWW radii of 400, 600, and 1000 Å, respectively. The dashed curve displays the recent experimental results of Hiruma *et al.* (Ref. 7) (in arbitrary units).

In summary, in this work we have presented a model calculation of the acceptor-related photoluminescence spectrum of a GaAs quantum-wire microcrystal. Typical theoretical photoluminescence spectra show two peaks associated with transitions from the $n=1$ conduction-subband electron gas to acceptors at the on-center and on-edge positions in the wire, in good agreement with the experimental results of Hiruma *et al.*[7] Further theoretical work to explain the line shape of the excitonic peak (at 818.2 nm) and to properly identify the position of the neutral-acceptor–bound-exciton recombination is certainly necessary.

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