Exciton linewidth due to scattering by polar optical phonons in semiconducting cylindrical quantum wire structures

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The exciton linewidth has been calculated in semiconducting cylindrical quantum wire structures as a function of the radius of the wire when the electron-hole pair which are bound into the exciton are scattered by polar optical phonons. Because in quantum wells, we found that the dominant contributions to the linewidth come from processes where the exciton is scattered in its ground state with a change in its center of mass motion and where the exciton is ionized into a free electron-hole pair by the polar optical phonons, we only consider such processes here. The linewidth for this scattering mechanism is found to increase as the radius of the wire decreases. Therefore, confinement of the exciton in a quantum wire structure will increase the linewidth of the exciton peaks when the exciton is scattered by polar optical phonons. As in the case of quantum wells for this scattering mechanism, the contribution to the linewidth from the inelastic scattering is larger than the contribution from the elastic scattering.

I. INTRODUCTION

In the past few decades, it has been possible to grow semiconducting structures in which the free carriers (electrons and holes) are confined in their motion and therefore behave as two-,[1] one-[2] and zero-dimensional[3] gases of particles. The confinement of electrons and holes in quantum wells, quantum wires, and quantum dots has lead to these semiconducting quantum structures having interesting optical and electronic properties.[4,5] Excitonic effects in these semiconducting nanostructures are greatly enhanced over those in bulk semiconductors leading to a dramatic alternation in their optical properties. Hassan and Spector[6] and Bockelman and Bastard[7] have calculated the interband optical absorption in quantum wires. Calculations of the binding energy and oscillator strength of excitons in quantum wires have been performed by many authors both for cylindrical[8] and rectangular quantum wires[9,10] using both the infinite and finite well models. Glutsch and co-workers[11–13] have theoretically investigated the optical absorption in quantum wires taking excitonic effects into account while Kohl *et al.*[14–17] and Tsuchiya *et al.*[18] have experimentally investigated the optical properties of such quantum wires. Branis *et al.*[19] have investigated theoretically the effect of magnetic fields applied along the axis of a cylindrical quantum wire on the energies of the excitons. Wegscheider *et al.*[20] have studied lasing from excitons in quantum wires while Nagamune *et al.*[21] have experimentally investigated the photoluminescence in GaAs quantum wires in high magnetic fields. Someya *et al.*[22] have also investigated the photoluminescence spectra in T-shaped quantum wires.

Because of the presence of the excitons in a semiconductor, an excitonic peak in the optical absorption occurs below the interband absorption edge. Because of the increase in the exciton binding energy with increasing confinement, the exciton peak shifts to lower energies below the absorption edge. The exciton peak has a finite width due to the scattering of excitons by phonons and other imperfections in the semiconductor.[23–26] Of particular importance in III-V semiconducting compounds such as GaAs, from which many semiconducting nanostructures have been fabricated, is scattering from polar optical phonons.[16,18] Theoretical investigations of the effect of carrier confinement on the contribution to the exciton linewidth due to scattering from polar optical phonons in quantum wells have shown that the linewidth increases with decreasing width of the well.[24] In this paper, we use similar methods to calculate the behavior of the exciton linewidth due to scattering by polar optical phonons in quantum wires.

II. CALCULATION OF THE EXCITON LINEWIDTH

In this paper we will follow the same approach as Spector *et al.*[24] in calculating the exciton linewidth. Knox[27] has presented a theoretical calculation of the exciton linewidth using first order perturbation theory. The linewidth of the exciton can be directly related to the exciton lifetime due to the interaction of the bound electron-hole pair with various imperfections in the semiconductor. This in turn can be calculated using time dependent perturbation theory to find the transition probability between some initial state of the exciton and all final states for which the transition is allowed by the selection rules and energy conservation. The exciton linewidth in energy units is thus given by

$$\Gamma = 2\pi \Sigma_f |\langle f|V|i\rangle|^2 \delta(E_f - E_i), \tag{1}$$

where V is the interaction potential between the exciton and the scatterers, E_f and E_i are the final and initial energies of the system and the matrix element $\langle f|V|i\rangle$ is evaluated using the exciton wave functions in the initial and final states. In our calculations, we will use the exciton wave functions obtained via variational methods for a cylindrical quantum wire of radius d by Brown and Spector.[6] Using the infinite poten-

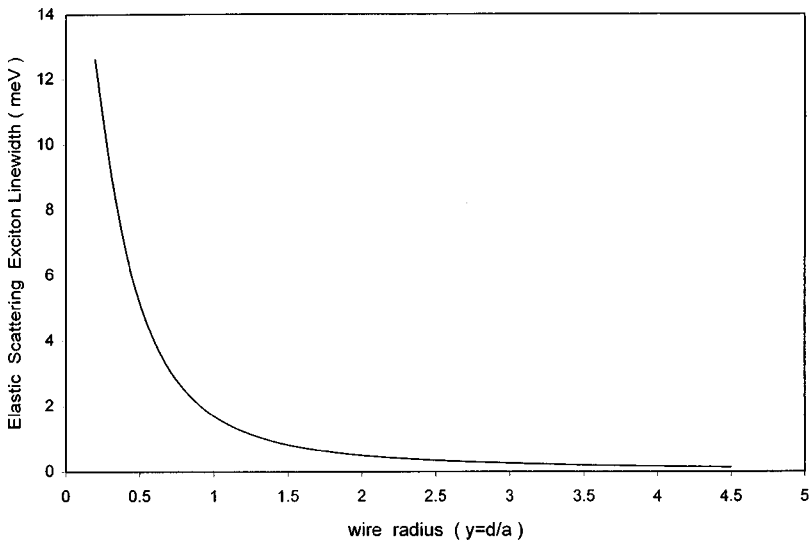

FIG. 1. The contribution to the exciton linewidth due to the elastic scattering of the exciton is shown as a function of the wire radius. Here the wire radius is given in units of Bohr radii, $y = d/a$, and the linewidth in meV. The parameters are those for the heavy hole exciton in GaAs.

tial well model, the ground state exciton wave function in a quantum wire of radius d was found to be[6]

$$\Psi(r_1,r_2)=\begin{cases} NJ_0(k\rho_1)J_0(k\rho_2)\exp\{-\lambda[(\vec{\boldsymbol{\rho}}_1-\vec{\boldsymbol{\rho}}_2)^2+z^2]^{1/2}\}\exp(iKZ_{\text{c.m.}}), & \rho_1 \text{ and } \rho_2 \leqslant d, \\ 0, \quad \rho_1 \text{ or } \rho_2 \geqslant d & \end{cases} \tag{2}$$

where the normalization coefficient N is given by

$$N^{-2}=-8\pi^2 d^5 L H_1 \tag{3}$$

and

$$H_1=\int_0^1 d\xi_1 \xi_1 J_0^2(kd\xi_1)\int_0^1 d\xi_2\xi_2 J_0^2(kd\xi_2) \times[\xi_< I_1(2\lambda d\xi_<)K_0(2\lambda d\xi_>) - \xi_> I_0(2\lambda d\xi_<)K_1(2\lambda d\xi_>)]. \tag{4}$$

Here, λ is a variational parameter which was found in the previous calculations as a function of the wire radius by minimizing the exciton energy in the wire. In Eq. (2), ρ_1 and ρ_2 are the coordinates of the electron and hole perpendicular to the wire axis, z is the relative coordinate of the electron-hole pair along the wire axis, and $Z_{\text{c.m.}}$ is the center of mass coordinate of the exciton along the wire axis. K is the center of mass wave vector of the exciton along the axis of the wire and is zero in the initial state when the exciton is formed by the optical excitation of a bound electron-hole pair while $kd=2.405$, which is the first zero of the Bessel function $J_0(x)$. In the integral H_1, we have made the substitution $\xi_i = \rho_i/d$. Also, $l_n(x)$ and $K_n(x)$ are modified Bessel functions of the first and second kind, respectively.

The interaction potential between the exciton and the polar optical phonons is given by

$$V(\vec{\mathbf{r}}_e,\vec{\mathbf{r}}_h)=ie[2\pi(\varepsilon_\infty^{-1}-\varepsilon_0^{-1})\hbar\omega_L]^{1/2} \times\int d^3q q^{-1}\{[a_q^+\exp(i\vec{\mathbf{q}}\cdot\vec{\mathbf{r}}_h)-a_q\exp(-i\vec{\mathbf{q}}\cdot\vec{\mathbf{r}}_h)] -[a_q^+\exp(i\vec{\mathbf{q}}\cdot\vec{\mathbf{r}}_e)-a_q\exp(-i\vec{\mathbf{q}}\cdot\vec{\mathbf{r}}_e)]\}, \tag{5}$$

where we have assumed that the interaction is with the bulk optical phonons. Here ω_L is the optical phonon frequency, ε_∞ and ε_0 are the high and low frequency dielectric constants, and a_q^+ and a_q are the phonon creation and annihilation operators.

Previous calculations for the exciton linewidth in quantum well structures have shown that the main contribution to the exciton linewidth comes from the elastic scattering of excitons with a change in the motion of their center of mass and from the ionization scattering of excitons in which the exciton is disassociated into a free electron hole pair.[16]

Therefore here we shall consider only these two contributions to the exciton linewidth in quantum wire structures assuming that as in the quantum well case, the contributions due to the scattering of the exciton into one of its excited bound states make a negligible contribution to the exciton linewidth. One of the problems with considering processes in which the exciton is scattered to an excited bound state is the lack of good excited state wave functions for excitons in a quantum wire in contrast to the case in quantum wells.

A. Elastic scattering

Here we consider the scattering of the exciton from its ground state with no center of mass motion $(K=0)$ to its ground with center of mass motion of the exciton. Using the variational exciton wave functions given in Eq. (2) to calculate the matrix elements of the scattering potential, we obtain the following result for the linewidth:

$$\Gamma=e^2[(\varepsilon_\infty^{-1}-\varepsilon_0^{-1})]N(\omega_L)(2m\omega_L/\hbar)^{1/2}(\lambda a H_2/H_1)^2, \tag{6}$$

where

$$H_2=\int_0^1 d\xi_1 J_0^2(kd\xi_1)\int_0^1 d\xi_2 J_0^2(kd\xi_2)\{(\xi_1/\delta)\exp[-(m/\mu)^{1/2}Qd\xi_2/a][\xi_> I_0(\delta d\xi_</a)K_1(\delta d\xi_>/a) - \xi_< I_1(\delta d\xi_</a)K_0(\delta d\xi_>/a)] - (\xi_2/\gamma)\exp[-(m/\mu)^{1/2}Qd\xi_1/a][\xi_> I_0(\gamma d\xi_</a)K_1(\gamma d\xi_>/a) - \xi_< I_1(\gamma d\xi_</a)K_0(\gamma d\xi_>/a)]\} \tag{7}$$

and

$$\gamma=[(2\lambda a)^2+(m_eQ/m)^2]^{1/2},$$

$$\delta=[(2\lambda a)^2+(m_hQ/m)^2]^{1/2}, \quad Q=(\hbar\omega_L/Ry)^{1/2}.$$

Here $a=(\varepsilon\hbar^2/\mu e^2)$ is the exciton Bohr radius, $Ry=(e^2/2\varepsilon a)$ is the exciton Rydberg unit, ε is the dielectric constant, μ is the exciton reduced mass, $N(\omega_L)$ is the thermal equilibrium distribution for the optical phonons, m_e and m_h are the electron and hole effective masses, respectively, and $m=m_e+m_h$ is the mass of the exciton. The contribution to the linewidth due to the elastic scattering of the exciton by polar optical phonons is shown as a function of the wire radius d in Fig. 1 for a temperature of 300 K. The parameters used in obtaining this figure are those for the heavy hole exciton in GaAs. Using these parameters, the exciton Rydberg unit is 4.61 meV and the exciton Bohr radius is 118 Å. The linewidth increases dramatically as the radius of the quantum wire structure decreases. The increase in the linewidth with decreasing wire radius is due to the increasing confinement of the electron-hole pair in the exciton. The only temperature dependence of the linewidth for the scattering of the exciton by optical phonons comes in through the temperature dependence of the optical phonon population. However, it should be noted that at low temperatures such that $\hbar\omega_L \gg k_BT$, the optical phonon modes will be frozen out and the contribution to the linewidth due to scattering of the exciton from polar optical phonons will decrease rapidly with temperature as $\exp(-\hbar\omega_L/k_BT)$. Therefore, while scattering from polar optical phonons will play an important role in determining the exciton linewidth at temperatures where $\hbar\omega_L \leqslant k_BT$, it will play a negligible role at low temperatures.

B. Ionization scattering

Here we consider the scattering of the exciton from its ground state with no center of mass motion $(K=0)$ to a state in which the electron-hole pair are free and the exciton is ionization by its interaction with the polar optical phonons. The final state exciton wave function in this case is given by

$$\Psi(r_1,r_2)=\begin{cases} N'J_0(k\rho_1)J_0(k\rho_2)\exp(ik_ez_e)\exp(ik_hz_h), & \rho_1 \text{ and } \rho_2\leqslant d \\ 0 & \rho_1 \text{ or } \rho_2 \geqslant d \end{cases}, \tag{8}$$

where the normalization coefficient N' is given by

$$N'=[\pi d^2 L J_1^2(kd)]^{-1}. \tag{9}$$

Here, we have neglected the correlation of the free electron-hole pair as a result of their Coulomb interaction by using plane wave functions for them instead of free state Coulomb functions. Using the variational exciton wave functions given in Eqs. (2) and (8) to calculate the matrix elements of the scattering potential, we obtain the following result for the linewidth:

$$\Gamma=(e^2/\pi)[(\varepsilon_\infty^{-1}-\varepsilon_0^{-1})](m_hm_e)^{1/2}N(\omega_L) \times(\omega_L/\hbar)d(\lambda a)^2(H_3/H_1)[J_1(kd)]^{-4}, \tag{10}$$

where

$$H_3=\int_{-\pi/2}^{+\pi/2} d\alpha\{|H_4|^2+|H_5|^2\}, \tag{11}$$

with

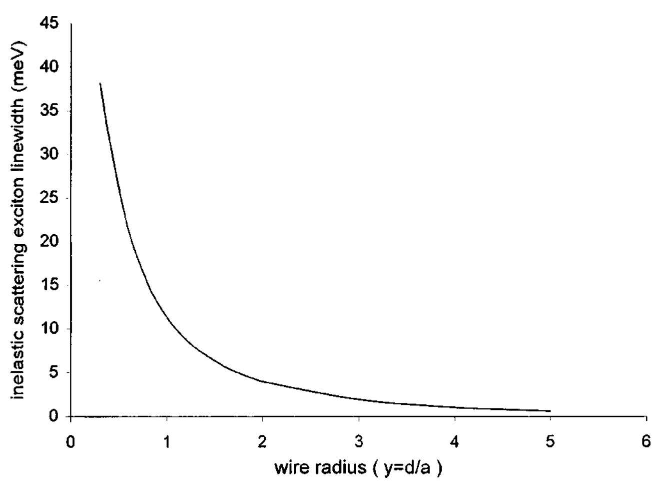

FIG. 2. The contribution to the exciton linewidth due to the ionization scattering of the exciton is shown as a function of the wire radius. Here the wire radius is given in units of Bohr radii, $y=d/a$, and the linewidth in meV. The parameters are those for the heavy hole exciton in GaAs.

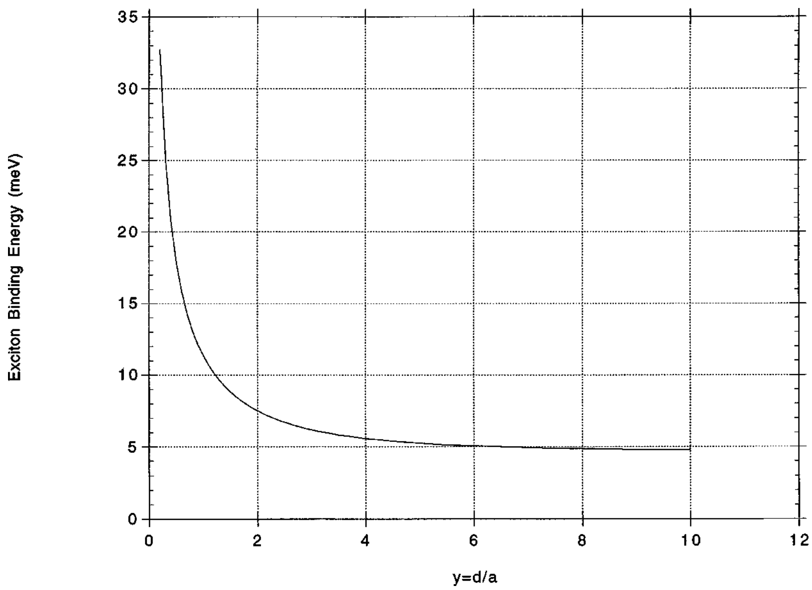

FIG. 3. The exciton binding energy is shown as a function of wire radius. Here the wire radius is given in units of Bohr radii, $y=d/a$, and the binding energy in meV. The parameters are those for the heavy hole exciton in GaAs.

$$H_4=\int_0^1 d\xi_1 J_0^2(kd\xi_1)\int_0^1 d\xi_2 J_0^2(kd\xi_2)\{(\xi_2/P)\exp(-|c|d\xi_1/a)[\xi_> I_0(Pd\xi_>/a)K_1(Pd\xi_>/a) - \xi_< I_1(Pd\xi_</a)K_0(Pd\xi_>/a)]-(\xi_1/W)\exp(-|c|d\xi_2/a)[\xi_> I_0(Wd\xi_</a)K_1(Wd\xi_>/a) - \xi_< I_1(Wd\xi_</a)K_0(Wd\xi_>/a)]\} \tag{12a}$$

and

$$H_5=\int_0^1 d\xi_1 J_0^2(kd\xi_1)\int_0^1 d\xi_2 J_0^2(kd\xi_2)\{(\xi_2/P)\exp(-|c'|d\xi_1/a)[\xi_> I_0(Pd\xi_</a)K_1(Pd\xi_>/a) - \xi_< I_1(Pd\xi_</a)K_0(Pd\xi_>/a)]-(\xi_1/W)\exp(-|c'|d\xi_2/a)[\xi_> I_0(Wd\xi_</a)K_1(Wd\xi_>/a) - \xi_< I_1(Wd\xi_</a)K_0(Wd\xi_>/a)]\}. \tag{12b}$$

Here

$$P=[(\lambda a)^2+v^2\sin^2\alpha]^{1/2},$$

$$W=[(\lambda a)^2+(m_h/m_e)\cos^2\alpha]^{1/2},$$

$$c=v[\sin\alpha+(m_h/m_e)^{1/2}\cos\alpha],$$

$$c'=v[\sin\alpha-(m_h/m_e)^{1/2}\cos\alpha],$$

$$v=[(m_e/\mu)(Q^2-E_b/R_y)]^{1/2},$$

where E_b is the exciton binding energy in the wire.

The contribution to the exciton linewidth due to ionization of the exciton by the polar optical phonons is shown as a function of the wire radius for a temperature of 300 K in Fig. 2. As in the case for elastic scattering of the exciton by the optical phonons, the linewidth here increases with a decrease of the radius of the quantum wire. The order of magnitude of this contribution to the exciton linewidth is greater than that for the elastic scattering case. This is similar to the results obtained for the same scattering mechanism in quantum wells,[24] where the contribution to the linewidth due to the ionization of the exciton is much greater than that due to elastic scattering of the exciton by polar optical phonons. At smaller wire radii, we would expect that there would be no inelastic contribution to the exciton linewidth as the exciton binding energy also increases with decreasing wire radii. Therefore, at smaller radii than we have considered in our calculations, the binding energy would become larger than the optical phonon energy and the exciton could no longer be ionized by absorbing an optical phonon. This possibility would not occur in quantum wells since the maximum exciton binding energy there using the infinite model is 4 Ry which is less than the optical phonon energy in GaAs. In Fig. 3, we show the exciton binding energy in meV as a function of the wire radius.[6] Again, the parameters used are those for the heavy hole exciton in GaAs. For small wire radii, the linewidth due to the inelastic scattering of the excitons by polar optical phonons can be greater than the exciton binding energy. Since the temperature dependence of the linewidth due to scattering from polar optical phonons comes in only through the phonon population factor, $N(\omega_L)$, as in the case for elastic scattering, the linewidth will decrease as exp

($-\hbar\omega_L/k_BT$) at low temperatures. In their measurements of the photoluminescence spectra in T-shaped quantum wires, Someya *et al.*[22] found their photoluminescence lines very sharp with linewidths of 4–7 meV. However, they performed their measurements at liquid helium temperatures where the contribution to the linewidth due to scattering from polar optical phonons should be negligible because of the diminished population of such phonons. Also, in T-shaped quantum wires, the infinite potential well model which we used in our calculations should not be too good as the carriers would not be as completely confined as in cylindrical wires.

It should also be noted that both the elastic and inelastic contributions to the exciton linewidth due to scattering by polar optical phonons are larger in quantum wires than in quantum wells.[24] This is due to the greater confinement of the exciton in quantum wires.

III. SUMMARY

Our calculations show that at temperatures where there is a significant population of polar optical phonons present, the linewidth should increase as the radius of the cylindrical quantum wire decreases using the infinite confining potential well model. As the temperature decreases, the linewidth should also decrease exponentially due to freezeout of the population of polar optical phonons. Therefore, in good quality quantum wires where the alloy scattering contribution to the linewidth has been reduced, the linewidth should increase dramatically with decreasing wire radius at temperatures of order $T \geqslant \hbar\omega_L/k_B$.

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