PHYSICAL REVIEW B **68**, 161307(R) (2003)

Radiative lifetime of quantum confined excitons near interfaces

Kwang Jun Ahn and Andreas Knorr
Institut für Theoretische Physik, Nichtlineare Optik und Quantenelektronik, Technische Universität Berlin, Hardenbergstraße 36, PN 7-1, 10623 Berlin, Germany

(Received 7 February 2003; revised manuscript received 21 May 2003; published 20 October 2003)

Using a nonlocal susceptibility and Green's-function approach, lifetime calculations for quantum confined excitons are carried out beyond the dipole approximation. The radiative lifetimes close to a metal surface are shown to strongly depend on the excitonic confinement length.

DOI: 10.1103/PhysRevB.68.161307 PACS number(s): 68.65.Hb, 78.67.−n, 78.68.+m

It is a well established fact that the radiative lifetime of an atomic emitter depends on the dielectric properties and the geometry of its macroscopic surrounding.[1,2] A typical example is the lifetime variation as a function of the distance of the emitter from the surface of its host material.[3,4] This effect is caused by the interface induced modification of the electromagnetic field at the position of the emitter due to its mirror image. Usually, for a atomic emitter, the optical wavelength is much larger than its extension, and therefore, the dipole approximation is applied. Theoretical calculations in this regime including surface effects agree very well with experimental data.[3–5] In spite of its success for the description of atomic systems sandwiched within different refractive index,[6] the dipole approximation must be questioned for nanoscopic systems. Here, the spatial extension of the structures may not always be neglected in comparison to the wavelength of light. Typical examples are excitons (Coulomb correlated electron-hole pair in semiconductor), which are quantum confined on a length scale of several tens to hundreds of nanometers.[7–9] Already calculations focused on excitonic excitations in a homogeneous host medium, i.e., neglecting surface effects, have shown that the radiative lifetime can vary over several orders of magnitude by changing the localization length of the excitonic wave functions compared to the emission wavelength.[7,10–12] However, the approximation of a homogeneous hostmedium having no surface is often not the situation found in experiments aiming at the investigation of single quantum confined excitons. As discussed in many recent papers, aiming at application in quantum computing and entanglement,[8,13,14] typical surface sensitive methods, such as near-field microscopes and apertures in metalized surfaces, have to be used to address the single excitons.[8,15] In these situations, the investigated excitons are typically situated within a wavelength near the surface. *In this paper, we demonstrate that for such a near-field situation the dipole approximation may yield—depending on the interface materials—even qualitatively different results in comparison to the full solution of the wave equation for quantum confined excitons at interfaces.*

The paper is organized as follows: First, the self-consistent formulation of the problem in terms of the excitonic dipole density and the optical fields is given. Second, the excitonic susceptibility and the relevant Green's-function tensor for the electromagnetic fields are used to derive the radiative lifetime. Third, typical examples, such as the influence of different interface materials and the influence of the emitter-interface distance are discussed. In Fig. 1, the configuration of the investigated system is sketched. An interface at $z=0$ spreads infinitely in x and y directions and divides the region 1 of the dielectric constant ε_1 from the region 2 of ε_2. The quantum confined GaAs exciton within a potential of radius R is located at the position $z=z_0$ in the region 1, parallel to the interface and centered at the origin of the x and y coordinates.

The induced electric field $\mathbf{E}(\mathbf{r},\omega)$ satisfies the wave equation in the region 1:

$$-\nabla^2\mathbf{E}(\mathbf{r},\omega)+\boldsymbol{\nabla}[\boldsymbol{\nabla}\cdot\mathbf{E}(\mathbf{r},\omega)]-\epsilon_1\frac{\omega^2}{c_0^2}\mathbf{E}(\mathbf{r},\omega)=\mu_0\omega^2\mathbf{P}(\mathbf{r},\omega), \tag{1}$$

where $\mathbf{P}(\mathbf{r},\omega)$ is the excitonic polarization and c_0 is the light velocity in vacuum. As a solution of Eq. (1), the electric field produced by the exciton in this region is the sum of two electric fields, one of which is electric field emitted directly from the exciton and the other the reflected field from the interface:

$$\mathbf{E}(\mathbf{r},\omega)=-\omega^2\mu_0\int\{\underline{\underline{\mathbf{G}}}_0(\mathbf{r},\mathbf{r}')+\underline{\underline{\mathbf{G}}}_1(\mathbf{r},\mathbf{r}')\}\cdot\mathbf{P}(\mathbf{r}',\omega)d^3r', \tag{2}$$

where $\underline{\underline{\mathbf{G}}}_{0,1}(\mathbf{r},\mathbf{r}')$ are the 3×3 Green's function tensors (GFT) for the direct field (0) and the reflected field (1). To evaluate Eq. (2), the polarization must be known. In the linear optics, the polarization in material can be written as

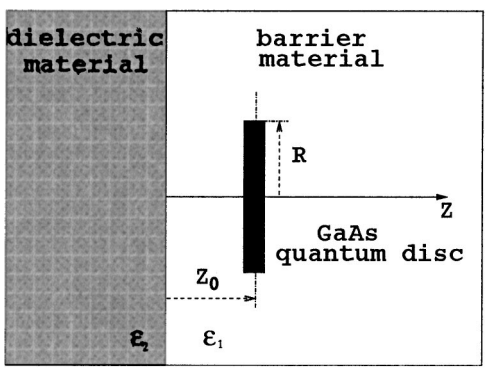

FIG. 1. Configuration of the system.

$$\mathbf{P}(\omega,\mathbf{r})=\epsilon_0\int \chi(\mathbf{r},\mathbf{r}',\omega)\mathbf{E}_{ex}(\mathbf{r}',\omega)d\mathbf{r}', \tag{3}$$

where $\chi(\mathbf{r},\mathbf{r}',\omega)$ is the nonlocal susceptibility of the material and $\mathbf{E}_{ex}(\mathbf{r}',\omega)$ an arbitrary external light source. To solve the self-consistently formulated problem of the radiation-matter coupling, Eqs. (1)–(3), the susceptibility and the GFT's must be derived.

To determine the nonlocal susceptibility, the equation of motion of the dipole density in a two-band model (conduction, valence band) is used.[11,12] A GaAs quantum well (QW) with ideal confinement in z direction and providing a potential in the xy plane ($\mathbf{r}_\parallel$) via interface fluctuations is considered. For a cylindrical interface fluctuation, this yields the formation of a quantum disc (QDc). For a QDc larger than exciton Bohr radius, the exciton motion is quantized via its center-of-mass coordinate. The corresponding wave function can be expanded in the product of the eigenfunction $\psi_n(\mathbf{r}_\parallel)$ in the in-plane confinement potential $V(\mathbf{r}_\parallel)$, here assumed to be a two-dimensional parabolic potential,[16] and its time-dependent amplitude function $\tilde{a}_n(t)$:

$$\mathbf{P}(\mathbf{r},\omega)=\sum_n \{\varphi_{1s}^{2d}(0)\mathbf{d}_{cv}\tilde{a}_n(t)\psi_n(\mathbf{r}_\parallel)+\text{c.c.}\}\delta(z-z_0). \tag{4}$$

The equation of motion for the amplitude function $a_n(\omega)$ in frequency domain is given as

$$\omega a_n(\omega)=\omega_n a_n(\omega)-\frac{\varphi_{1s}^{2d*}(0)\mathbf{d}_{cv}}{\hbar}\cdot\int \psi_n^*(\mathbf{r}_\parallel) \times\{\mathbf{E}_{ex}(\mathbf{r}_\parallel,\omega)+\mathbf{E}(\mathbf{r}_\parallel,\omega)\}d\mathbf{r}_\parallel, \tag{5}$$

where $\varphi_{1s}^{2d}(\mathbf{r}_\parallel)$ is the $1s$ eigenfuction of the exciton for the relative motion, ω_n is the eigenfrequency of the exciton in the state n, and $\mathbf{d}_{cv}=\langle c|e_0\mathbf{r}|v\rangle$ is the dipole matrix element over the elementary cell. The usual dipole approximation, valid for a QDc considerably smaller than the spatial variation of the light field (here, wavelength of light) is given by

$$\int \psi_n^*(\mathbf{r}_\parallel)[\mathbf{E}(\mathbf{r}_\parallel)+\mathbf{E}_{ex}(\mathbf{r}_\parallel)]d\mathbf{r}_\parallel \cong[\mathbf{E}(\mathbf{r}_\parallel=0)+\mathbf{E}_{ex}(\mathbf{r}_\parallel=0)]\int \psi_n^*(\mathbf{r}_\parallel)d\mathbf{r}_\parallel. \tag{6}$$

For a larger extension of the QDc, the spatial integral in Eq. (5) has to be fully calculated. Using Eqs. (3)–(6), the lateral part of the nonlocal susceptibility can be written as

$$\chi(\mathbf{r}_\parallel,\mathbf{r}_\parallel',\omega)=-\sum_n \frac{|\varphi_{1s}^{2d}(0)|^2|\mathbf{d}_{cv}|^2}{\epsilon_0\hbar(\omega-\omega_n+\Sigma_n)}\psi_n^*(\mathbf{r}_\parallel)\psi_n(\mathbf{r}_\parallel'), \tag{7}$$

where $\Sigma_n=\Sigma_n^{\text{se}}+\Sigma_n^{\text{intf}}$ and Σ_n^{se} are the radiation self-energies, see Eq. (8) for the homogeneous medium, and Σ_n^{intf} for the radiation self-energies induced by interface, see Eq. (9). In the following, we use GFTSs in Eq. (2) for an array of dielectric layer with arbitrary optical constants that have been already derived.[17] These GFT's are sum of two tensors for the s- (perpendicular to the incident plane) and the p-polarized (parallel to the incident plane) electric fields multiplied by the corresponding Fresnel reflection or transmission coefficients. In the numerical simulations, it is assumed that the QDc is polarized parallel to the x axis and only the heavy-hole excitons will be discussed. In addition in Eq. (7), only the diagonal parts of the self-energies $\Sigma_n=\delta_{nm}\Sigma_{nm}$ are taken into consideration because the nondiagonal parts were negligibly small in our calculations. The parameters used in the numerical calculations are listed in Ref. 19.

Equations (2)–(6) form a closed set and can be solved for several situations of interest. First, to understand the results where an interface is incorporated, we first generalize the solution for the radiation lifetime of QDc exciton ground state[7] in homogeneous medium to a multilevel system (i). Second, we investigate the influence of an interface on the radiation lifetime of the QDc ground state (ii).

(i) The radiative lifetime of a excitonic multilevel system in an homogeneous semiconductor host medium is considered. The radiative self-energy of the QDc in the state n in homogeneous medium is defined as

$$\Sigma_n^{\text{se}}=\mathcal{C}\int\int d\mathbf{r}_\parallel' d\mathbf{r}_\parallel \psi_n^*(\mathbf{r}_\parallel)\cdot\underline{\underline{\mathbf{G}}}_0(\mathbf{r}_\parallel,z_0,\mathbf{r}_\parallel' z_0)\cdot\psi_n(\mathbf{r}_\parallel'), \tag{8}$$

where $\mathcal{C}=\omega_n^2\mu_0|\mathbf{d}_{cv}|^2|\varphi_{1s}^{2d}(0)|^2/\hbar$ and $\underline{\underline{\mathbf{G}}}_0(\mathbf{r},\mathbf{r}')$ is the GFT of the homogeneous GaAs background with $\epsilon_1=\epsilon_2=12.5$.[20] Recent results[7,21] are verified by calculating the radiative lifetime of the excitonic ground state. In Fig. 2(a), the lifetimes $\tau_{homo}=1/\text{Im}[\Sigma_n^{\text{se}}]$ of the ground and the first excited excitonic state are plotted as a function of the QDc radius. The results show radiative lifetimes from the picosecond to nanosecond range.[7] As the size of the QDc increases, the lifetime of the ground state decreases down to a fixed value, approximately the lifetime of the bare QW in GaAs.[21] The qualitative behavior of the radiative lifetime can be explained as follows: The wave vector of the emitted light $\mathbf{k}$ can be decomposed into its lateral and z component, so that $\mathbf{k}=(\mathbf{k}_\parallel,k_z=\sqrt{\mathbf{k}^2-\mathbf{k}_\parallel})$. For real k_z, we have a propagating field and for imaginary k_z an evanescent field. Evanescent fields occur for $k_\parallel>k=\sqrt{\epsilon}\,\omega/c_0$, i.e., for QDc with a lateral dimension R with $k_\parallel\cong R^{-1}>\sqrt{\epsilon}\,\omega/c_0$. Similarly, the radiative self-energy Σ_n^{se}, Eq. (8), can be decomposed into two parts. The imaginary part of Σ_n^{se} is the contribution of the propagating modes of the field ($k_z<(\omega/c_0)\sqrt{\epsilon_{\text{GaAs}}}$) which is the inverse radiative lifetime, while the real part is that of the evanescent modes of the field ($k_z>(\omega/c_0)\sqrt{\epsilon_{\text{GaAs}}}$) which has no contribution to the emission. In the case of the QDc buried in GaAs, so-called *cutoff wave number* k_c between both contributions is given by $k_c=(\omega/c_0)/\sqrt{\epsilon_{\text{GaAs}}}(\epsilon_{\text{GaAs}}=12.5)$. In Fig. 2(b), the wave number resolved radiative contributions $f(k_\parallel)[\text{Im}(\Sigma_n^{\text{se}})=\int \text{dk}_\parallel f(k_\parallel)]$ of the ground state (00) is plotted for increasing radii ($R=20$, 130, 500 nm) of QDc as a function of the normalized wave number $x=k_\parallel/k_0(k_0=\omega/c_0)$. As the radius of the QDc increases, the spatial uncertainty $\Delta r_\parallel=\Delta x\Delta y$ for excitons increases, while the momentum uncertainty $\Delta k_\parallel=\Delta k_x\Delta k_y$ for excitons decreases. In

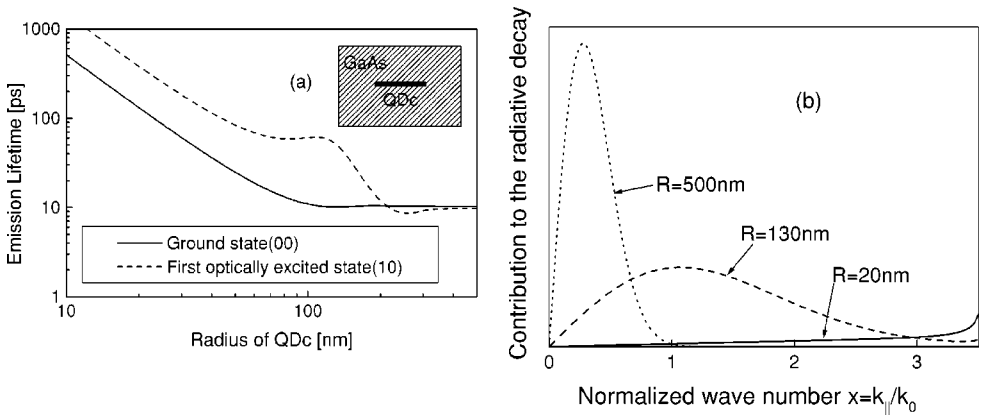

FIG. 2. (a) The radiative lifetime of the first two optically active states as the function of the QDc radius and (b) The radiative contributions to the lifetime of the ground state (00) as a function of the normalized wave number $x=k_{\parallel}/k_0$ for diverse radii of the QDc.

this process, the radiative contributions are enhanced by the excitonic momentum distribution $|\psi(k_{\parallel})|^2$ which becomes more dominant for wave numbers below k_c, corresponding to increased emission. That means, for increasing R, a conversion of evanescent to radiative modes takes place. From $R=130$ nm, the lifetime is determined basically by the waves with the wave numbers $k_{\parallel}$ smaller than k_c, saturating to the ideal QW limit $(k_{\parallel}=0)$. The small oscillation of the radiative lifetime of the first excitonic state results from the characteristics of the wave function which oscillates around the origin.

(ii) Next, the radiative lifetime of a QDc below a surface is considered, compare Fig. 1. Here, the full electric field in the QDc is the sum of the self-interaction and the reflected fields from the interface. The radiative self-energy Σ_n^{intf} induced by the interface is defined as

$$\Sigma_n^{\mathrm{intf}}=\mathcal{C}\int\int d\mathbf{r}_{\parallel}' d\mathbf{r}_{\parallel}\psi_n^*(\mathbf{r}_{\parallel})\cdot\underline{\underline{\mathbf{G}}}_1(\mathbf{r}_{\parallel},z_0,\mathbf{r}_{\parallel}';z_0)\cdot\psi_n(\mathbf{r}_{\parallel}'). \tag{9}$$

$\underline{\underline{\mathbf{G}}}_1(\mathbf{r},\mathbf{r}')$ is the GFT in the presence of the interface.[17] Equation (9) is applied to a semiconductor-vacuum interface[18] and a semiconductor-metal interface. All calculated lifetimes are normalized with respect to the lifetimes in homogeneous environment without interface. Figure 3(a) shows the lifetimes as a function of the distance from the vacuum-semiconductor interface for different QDc size. To highlight the results, the calculated lifetimes are compared with those of the dipolelike emitter, i.e., dipole approximation in Eq. (6) and the ideal QW case. The calculated lifetimes oscillate depending on the distance to the interface. These well known oscillations result from the interference of emitted and back reflected waves which accelerate or decelerate the emission.[3–6] The response of the small QDc can be approximated by an ideal dipole emitter. As the QDc becomes larger, the lifetime oscillations increase, finally, the lifetime of the QDc converges to the QW lifetime. For the vacuum-semiconductor interface, there is no qualitative, only quantitative difference in the behavior as a function of the radius. Next, the lifetimes of the QDc in the aluminum-semiconductor interface system are depicted in Fig. 3(b). Such metal coatings can be used in near-field optical situation.[15] The dielectric constant of aluminum $(\epsilon_{\mathrm{Al}}=-63.6+i47.3)$ is taken at the band gap of GaAs. For the Al-semiconductor interface, the lifetime of the dipolelike emitter shows a qualitatively different behavior, compared to the larger QDc's in the near-field (distance <50 nm) of the interface. Whereas the radiative decay of the ideal dipole is enhanced close to the metal surface, the QDc shows the opposite behavior if the radius is increased. The enhancement of the radiative decay of a pointlike dipole close to the metal interface had been already reported.[3,4,22,23] If the dipolelike emitter is close to (below 1/4 of the emission wavelength) the surface, the emission is enhanced by the resonance between the emitter and the surface-plasmon polariton (SPP) propagating along the surface.[3,22–25] This effect is reproduced for the dipolelike emitter and—to some extent—for small QDc's close to the surface. However, with

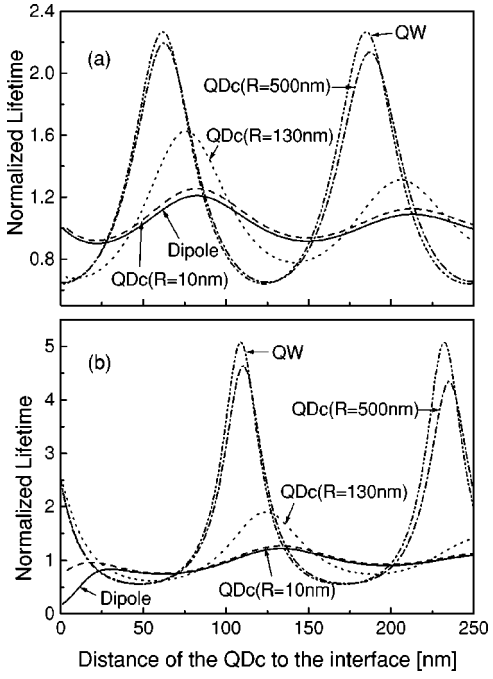

FIG. 3. (a) Normalized lifetime of QDc as a function of the distance to the vacuum-semiconductor interface and (b) to the Al-semiconductor interface.

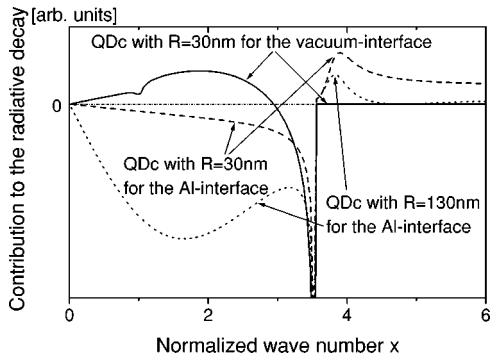

FIG. 4. Contribution to the decay rate of QDc ($R=30$, 130 nm) at $Z_0=1$ nm as a function of the normalized wave number $x=k_\parallel/k_0$ for Al semiconduction interface.

increasing size the QDc exciton exhibits an opposite trend, the radiative decay is suppressed up to a factor of 10 in comparison with the ideal dipole case.

To understand this inverted behavior close to the surface, the momentum distribution $f(k_\parallel)[\mathrm{Im}(\Sigma_n^{\mathrm{intf}})=\int dk_\parallel f(k_\parallel)]$ for two QDcs ($R=30$, 130 nm) are plotted as a function of the normalized wave number $x=k_\parallel/k_0$ in Fig. 4. The investigated QDc is separated by 1 nm from the interface, to pronounce the observed effect. No contribution to the radiative decay above k_c can be found in the case of the QDc with $R=30$ nm for the vacuum-semiconductor interface. Here, the radiative decay results from positive contributions of $f(k_\parallel)$ below $k_\parallel\cong 3.5k_0$ similar to Fig. 2(b). The negative contributions result from surface reflected fields, which reduce the radiation emission. For the case of the metal-semiconductor interface, an additional resonance at $k_\parallel=3.9k_0$ can be found, this absorbing contribution results from the metal-semiconductor surface plasmon.[24] Furthermore, due to the strong metallic response the surface-induced reflection (negative contribution) increases. These contributions become larger as the QDc grows and the contribution from the resonance with the surface plasmon becomes smaller. The enhancement of the radiative decay via the resonance with the SPP is smaller than its suppression by the back-propagating reflected field that is out of phase with the radiative wave emitted directly from the QDc.

In conclusion, the radiative lifetimes of quantum confined excitons in the near-field distance from an interface depend strongly on the dielectric properties of the interface. For dissipative interfaces, such as metal-semiconductor interface and QDc of increasing size, the contribution from the resonance with the surface-plasmon polariton to the radiative decay is negligibly small, compared with the contribution of the reflected field from the interface. These effects cannot be predicted within the dipole approximation for nanoscopic structures.

Future work should include the difference between the background refractive indices of the quantum disc and its host medium which is a further correction to the resonant interaction discussed here (Ref. 2).

This project was supported by the Deutsche Forschungsgemeinschaft (DFG) through Sonderfoschungsbereich 296.

[1] W. Vogel, D. G. Welsch, and S. Wallentowitz, *Quantum Optics: An Introduction*, 2nd ed. (Wiley-VCH, Berlin, 2001).
[2] A. Thränhardt *et al.*, Phys. Rev. B **65**, 035327 (2002).
[3] R.M. Amos and W.L. Barnes, Phys. Rev. B **55**, 7249 (1997).
[4] J.M. Wylie and J.E. Sipe, Phys. Rev. A **30**, 1185 (1984).
[5] J.E. Sipe, Surf. Sci. **105**, 489 (1981).
[6] A. Rahmani *et al.*, Phys. Rev. A **56**, 3245 (1997).
[7] M. Sugawara, Phys. Rev. B **51**, 10 743 (1995).
[8] J.R. Guest *et al.*, Phys. Rev. B **65**, 241310 (2002).
[9] J.L. Spithoven *et al.*, J. Vac. Sci. Technol. B **17**, 1632 (1999).
[10] J. Bellessa *et al.*, Phys. Rev. B **58**, 9933 (1998).
[11] A. Siarkos, E. Runge, and R. Zimmermann, Phys. Rev. B **61**, 10 854 (2000).
[12] B. Hanewinkel, A. Knorr, P. Thomas, S. W. Koch, *Optics of Nanostructured Materials* Wiley Series in Lasers and Application (Wiley, New York 2001), Chap. 4.
[13] T. Guenther *et al.*, Phys. Rev. Lett. **89**, 057401 (2002).
[14] M.S. Shahriar *et al.*, Phys. Rev. A **66**, 032301 (2002).
[15] N.H. Bonadeo *et al.*, Phys. Rev. Lett. **81**, 2759 (1998).
[16] S. Flügge, *Practical Quantum Mechanics*, (Springer-Verlag, Berlin, 1974).
[17] R.L. Hartman, S.M. Cohen, and P.T. Leung, J. Chem. Phys. **110**, 2189 (1999).
[18] V.I. Pipa *et al.*, Phys. Rev. B **65**, 235308 (2002).
[19] $\omega_0\cong 2.163\times 10^15\ \mathrm{s}^{-1}$ for the resonance frequency of the exciton in GaAs, $m_e=0.067m_0$, $m_{hh}=0.045m_0$ for the mass of the electron and heavy hole, where m_0 is the electron rest mass, $\varphi_{1s}^{2d}(0)=\sqrt{8/(\pi a_B^2)}$, where the Bohr radius $a_B=10$ nm, $|\mathbf{d}_{cv}|=0.4e_0$ nm for the dipole moment strength, where e_0 is fundamental electronic charge.
[20] Semiconductor Information web site, www.semiconductors.co.uk
[21] D. Ammerlahn *et al.*, Phys. Rev. B **61**, 4801 (2000).
[22] R.M. Amos and W.L. Barnes, Phys. Rev. B **59**, 7708 (1999).
[23] M. Moskovits, Rev. Mod. Phys. **57**, 783 (1985).
[24] S. W. Koch, A. Mysyrowicz, and N. Peygambarian, *Introduction to Semiconductor Optics*, Series in Solid State Physical Electronics (Prentice-Hall, Engleword Cliffs, NJ, 1993).
[25] C. F. Klingshirn, *Semiconductor Optics* (Springer, Berlin, 1997).