

## Calculation of the radiative lifetime of Wannier-Mott excitons in nanoclusters

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*(Received April 18, 2013, Revised September 27, 2013, Accepted October 1, 2013)*

**Abstract.** This study is aimed to calculate the radiative lifetime of Wannier-Mott excitons in nanoclusters of a narrow-bandgap semiconductor embedded in a wide-bandgap one. The nanocluster linear dimensions are assumed to be much larger than the radius of the exciton so that the latter is not destructed by the confinement potential as it takes place in small quantum dots. The calculations were carried out for an example of InAs nanoclusters put into the GaAs matrix. It is shown that the radiative lifetime of Wannier-Mott excitons in such clusters increases with the decrease of the cluster dimensions, this tendency being more pronounced at low temperatures. So, the creation of excitons in nanoclusters of a narrow-bandgap material embedded in a wide-bandgap one can be used to significantly prolong their radiative lifetime in comparison with that of excitons in a bulk semiconductor.

**Keywords:** exciton radiative lifetime; nanoclusters; Wannier-Mott excitons

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### 1. Introduction

Excitons play an important role in the study of optical properties of solids. There exist two main types of excitons: Frenkel excitons and Wannier-Mott excitons (Knox 1963). A Frenkel exciton (Frenkel 1931) is also called the exciton of a small radius because it stands for a bound electron-hole pair, localized at one and the same crystal lattice site and migrating in a crystal from one site to another. In contrast to it, a Wannier-Mott exciton (Wannier 1937) is the exciton of a large radius in which the bound electron and hole may occupy different crystal lattice sites or the space between them. In semiconductors and based on them nanoheterostructures it is Wannier-Mott excitons that are more frequently observed. So, for the theoretical description of nanodevices, this exciton type is more important and therefore is considered here.

One of the main characteristics of an exciton is its lifetime. This is because all the processes in which it is involved obviously have to proceed on timescales shorter than this value. So, the increase of the exciton lifetime allows to broaden the scope of exciton-involved phenomena which can be observed and which can be employed in practice.

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The exciton lifetime is obviously determined by the time of its radiative recombination with the emission of a photon and its non-radiative recombination consisting in the exciton capture by traps and its subsequent recombination with the emission of a photon whose energy, however, is significantly smaller than the energy of the photon, emitted under process of radiative recombination. The rate of the exciton capture by traps depends on the density and type of the latter and therefore is determined by the crystal type, its quality and purity and therefore is strongly related to a concrete technology of the crystal fabrication. In contrast to that, the rate of the exciton radiative recombination is independent on technology features and determined only by the fundamental crystal parameters. So, it remains the same for crystals of a certain type grown by means of different technology processes and thereby puts the lower limit to the exciton recombination rate in this crystal type.

The aim of the present article is to calculate the Wannier-Mott exciton lifetime due to its radiative recombination and propose a way to increase this value. For this purpose in the next section the structure of the Wannier-Mott exciton is outlined and its interaction with quantized electromagnetic field is depicted. In section 3 for the example of the Wannier-Mott exciton in InAs its lifetime due to its radiative recombination is calculated. Then it is shown that this time can be significantly prolonged if InAs forms nanoclusters in a wide-bandgap material, e.g., GaAs. In the Conclusion the main results of the article are recapitulated.

## 2. Wannier-Mott exciton in quantized electromagnetic field

Let us represent the electron state in a crystal by the Bloch wavefunction  $\phi_{j\mathbf{k}} = u_j(\mathbf{r})\exp(i\mathbf{k}\mathbf{r})/\sqrt{V}$ , where index  $j$  stands for the zone number,  $j = v, c$  for the valence and conduction bands respectively,  $\mathbf{r}$  is the electron radius-vector,  $\mathbf{k}$  – its quasiwavevector,  $V$  – the crystal volume, and the function  $u_j(\mathbf{r})$  – a periodic spinor function,  $u_j(\mathbf{r}) = u_j(\mathbf{r}+\mathbf{a})$ , where  $\mathbf{a}$  – any translational vector of the crystal Bravais lattice.

The crystal ground state is depicted by a wavefunction  $\Psi_0 = (1/N!) \sum_{\mathbf{k}} \prod \phi_{v\mathbf{k}}$ , where the sum sign stands for the antisymmetrization over electrons radius-vectors and spins,  $\mathbf{k}$  runs through all the first Brillouin zone, its discrete values being determined by the periodic boundary conditions, and  $N$  is the number of allowed states in the valence band. Let us call an exciton (Elliott 1957) the crystal excited state described by a wavefunction  $\Psi_{\mathbf{k}_e n} = (1/N!) \sum_{\mathbf{k}} \sum_{\mathbf{c}\mathbf{k}} c_{\mathbf{k}n} \phi_{\mathbf{c}\mathbf{k}} \prod_{\mathbf{k}' \neq \mathbf{k} - \mathbf{k}_e} \phi_{v\mathbf{k}'}$ , where

$\mathbf{k}_e$  is the exciton center-of-mass quasiwavenumber,  $n$  stands for the exciton quantum state number, and  $c_{\mathbf{k}n}$  – some coefficients determined in the following way. Let us consider a formal Fourier transform

$$\begin{aligned} \psi(\mathbf{r}_{el}, \mathbf{r}_h) &\equiv \sum_{\mathbf{k}} c_{\mathbf{k}n} \exp[i\mathbf{k}\mathbf{r}_{el} - i(\mathbf{k} - \mathbf{k}_e)\mathbf{r}_h] / V \equiv \\ \exp[i\mathbf{k}_e \mathbf{r}_e] / V^{1/2} &\sum_{\mathbf{k}} c_{\mathbf{k}n} \exp\{i[\mathbf{k} - \mathbf{k}_e m_{el} / (m_{el} + m_h)](\mathbf{r}_{el} - \mathbf{r}_h)\} / V^{1/2} \equiv \\ \exp[i\mathbf{k}_e \mathbf{r}_e] / V^{1/2} &\psi_n(\mathbf{r}_{el} - \mathbf{r}_h) \end{aligned} \quad (1)$$

with some formal radius-vectors  $\mathbf{r}_{el}$  and  $\mathbf{r}_h$ . Here  $\mathbf{r}_e = (m_{el}\mathbf{r}_{el} + m_h\mathbf{r}_h) / (m_{el} + m_h)$ ,  $m_{el}$  and  $m_h$  are

the effective electron and hole masses respectively. Then, let us chose  $c_{k_n}$  in such a way that  $\psi_n(\mathbf{r}_{e1}-\mathbf{r}_h)$  be the wavefunction of the hydrogen atom in a quantum state  $n$ , in which, though, the electron mass is replaced with the reduced effective mass of the electron and hole,  $m_{e1}m_h/(m_{e1}+m_h)$ , and the Coulomb potential is diminished by  $\varepsilon_{st}$  times, where  $\varepsilon_{st}$  stands for the crystal static dielectric constant.

So, the wavefunction in Eq. (1) formally represents the motion of a hydrogen-like atom consisting of two particles with charges  $\pm e$ , where  $e$  is the absolute value of the elementary charge, radius-vectors  $\mathbf{r}_{e1}$  and  $\mathbf{r}_h$ , and masses  $m_{e1}$  and  $m_h$ . The function  $\exp[i\mathbf{k}_e\mathbf{r}_e]/V^{1/2}$  in Eq. (1) depicts the free motion of such an atom center-of-mass with the radius-vector  $\mathbf{r}_e$ , and  $\psi_n(\mathbf{r}_{e1}-\mathbf{r}_h)$  describes the relative motion of two particles comprising this atom. The energy of such a state is therefore  $\hbar^2 k_e^2/[2(m_{e1}+m_h)] - e^4 m_{e1}m_h/[2\hbar^2 \varepsilon_{st}^2 (m_{e1}+m_h)n^2]$  and the characteristic radius of the ground quantum state (corresponding to  $n = 1$ ) of such a hydrogen-like atom is  $r_{ex} = \hbar^2 \varepsilon_{st} (m_{e1}+m_h)/(e^2 m_{e1}m_h)$ . Let us call this energy plus the crystal bandgap energy  $E_g$  the exciton energy  $E_{k_e n}$  and this radius the exciton radius.

Let us note here that, for such a treatment to be valid, the linear dimensions of a cluster (in which an exciton is confined), that are of order  $V^{1/3}$ , have to significantly exceed  $r_{ex}$ . Only in this case the above described structure of a Wannier-Mott exciton is not destructed by the cluster confining potential. In this connection it is important to stress here that, as a rule, under the consideration of the exciton radiative lifetime (please see, e.g., Bauer *et al.* (2013), Boggess *et al.* (2001), Harbord *et al.* (2009), Musa *et al.* (2011), Schmidt *et al.* (2012), Wu and Lin (2012)) its dependence on the cluster dimensions is investigated in the opposite case, in which the exciton radius is larger than a characteristic cluster radius. Obviously, in such a case the Wannier-Mott exciton is disrupted by the cluster confining potential so that the obtained radiative exciton lifetime dependence on the cluster dimensions refers to an exciton of another (not of a Wannier-Mott) type.

The interaction of such an exciton with quantized electromagnetic field is described by the matrix element  $\langle \Psi_0 | -i\hbar e/(mc) \sum_{\mathbf{r}} \hat{\mathbf{A}}(\mathbf{r}, t) \nabla_{\mathbf{r}} | \Psi_{k_e n} \rangle$  of the resonant part of the electron-electromagnetic field non-relativistic interaction Hamiltonian (Landau and Lifshitz 1991), where  $m$  is the free electron mass,  $c$  – the light velocity in vacuum,  $\hbar$  – the reduced Plank constant, the sum is performed over radius-vectors  $\mathbf{r}$  of all the electrons,  $\nabla_{\mathbf{r}}$  – the nabla-operator, acting on the coordinates of the radius-vector  $\mathbf{r}$ ,  $\hat{\mathbf{A}}(\mathbf{r}, t) = \sum_{\mathbf{k}_{ph}\alpha} [\hat{a}_{\mathbf{k}_{ph}\alpha} \mathbf{A}_{\mathbf{k}_{ph}\alpha} \exp(-i\omega_{ph}t + i\sqrt{\varepsilon} \mathbf{k}_{ph} \mathbf{r}) + \hat{a}_{\mathbf{k}_{ph}\alpha}^+ \mathbf{A}_{\mathbf{k}_{ph}\alpha}^* \exp(i\omega_{ph}t - i\sqrt{\varepsilon} \mathbf{k}_{ph} \mathbf{r})]$  – the operator of the electromagnetic field vector-potential (Berestetskii *et al.* 1982),  $\omega_{ph}$  and  $\mathbf{k}_{ph}$  – the photon frequency and vacuum wave-vector, connected by the dispersion relation  $\mathbf{k}_{ph} = \omega_{ph}/c$ ,  $\varepsilon$  – the crystal dielectric function at  $\omega_{ph}$ , index  $\alpha$  stands for two orthogonal to  $\mathbf{k}_{ph}$  photon polarizations,  $\hat{a}_{\mathbf{k}_{ph}\alpha}$ ,  $\hat{a}_{\mathbf{k}_{ph}\alpha}^+$  – the operators of the annihilation and creation of a photon with the wavevector  $\mathbf{k}_{ph}$  and polarization  $\alpha$ ,  $\mathbf{A}_{\mathbf{k}_{ph}\alpha} = c\sqrt{2\pi\hbar}/(\omega_{ph}V') \mathbf{e}_{\mathbf{k}_{ph}\alpha}$ , where  $V'$  – normalization volume, much larger than  $V$ ,  $\mathbf{e}_{\mathbf{k}_{ph}\alpha}$  – the unit photon polarization vectors,  $\mathbf{k}_{ph} \mathbf{e}_{\mathbf{k}_{ph}\alpha} = 0$ ,  $\mathbf{e}_{\mathbf{k}_{ph}\alpha}^* \mathbf{e}_{\mathbf{k}_{ph}\alpha'} = \delta_{\alpha\alpha'}$ ,  $\delta_{\alpha\alpha'}$  – the Kronecker symbol.

Then, it can be shown that, retaining only the resonant term

$$\begin{aligned} & \langle \Psi_0 | \frac{-i\hbar e \sum_{\mathbf{r}} \hat{\mathbf{A}}(\mathbf{r}, t) \nabla_{\mathbf{r}}}{mc} | \Psi_{\mathbf{k}_e n} \rangle = \\ & - \sum_{\mathbf{k}_{ph\alpha}} \mathbf{E}_{\mathbf{k}_{ph\alpha}}^* \hat{a}_{\mathbf{k}_{ph\alpha}}^+ \sum_{\mathbf{k}} \mathbf{d}_{cv} c_{\mathbf{k}n} \sum_{\mathbf{r}_c} \exp[i\omega_{ph}t + i(\mathbf{k}_e - \sqrt{\varepsilon} \mathbf{k}_{ph}) \cdot \mathbf{r}_c] \frac{V_c}{V}, \end{aligned} \quad (2)$$

where  $\mathbf{E}_{\mathbf{k}_{ph\alpha}} = i\sqrt{2\pi\hbar\omega_{ph}/V} \mathbf{e}_{\mathbf{k}_{ph\alpha}}$ ,

$\mathbf{d}_{cv} = -\frac{e\hbar^2}{mE_{g\text{ cell}}} \int u_v^*(\mathbf{r}) [\nabla u_c(\mathbf{r}) + \mathbf{k}u_c(\mathbf{r})] \exp[i(\mathbf{k}_e - \sqrt{\varepsilon} \mathbf{k}_{ph}) \cdot \mathbf{r}] d^3r / V_c$  – the interband matrix element

having the dimension of a dipole moment, where the summation over spinor indices is assumed, the integration is carried out over a crystal primitive cell with the volume  $V_c$ , and  $\mathbf{r}_c$  – the radius-vectors of the crystal Bravais lattice sites each of which is associated with a crystal primitive cell.

In what follows I shall consider a case when  $|\mathbf{k}_e - \sqrt{\varepsilon} \mathbf{k}_{ph}|$  is much smaller than the primitive cell

inverse dimensions so that in the formula for  $\mathbf{d}_{cv}$  it can be put that  $\exp[i(\mathbf{k}_e - \sqrt{\varepsilon} \mathbf{k}_{ph}) \cdot \mathbf{r}] = 1$ . Due to

the orthogonality of  $u_v(\mathbf{r})$  and  $u_c(\mathbf{r})$ , it means that  $\mathbf{d}_{cv}$  does not depend on  $\mathbf{k}$  and therefore can be placed outside the sum over  $\mathbf{k}$  in Eq. (2). Then, according to the above definition of  $c_{\mathbf{k}n}$  and considering the case when the exciton is in its ground state, one has

$$\sum_{\mathbf{k}} c_{\mathbf{k}1} = \sqrt{V} \psi_1(0) = \sqrt{V / (\pi r_{ex}^3)}.$$

The sum over  $\mathbf{r}_c$  in Eq. (2), due to the considered situation in which  $|\mathbf{k}_e - \sqrt{\varepsilon} \mathbf{k}_{ph}|$  is much smaller than the primitive cell inverse dimensions, can be calculated by its replacement with an integral. This, under the assumption that the cluster with volume  $V$  has a spherical form with a radius  $R$  (so that  $V = 4\pi R^3/3$ ), gives that  $\sum_{\mathbf{r}_c} \exp[i(\mathbf{k}_e - \sqrt{\varepsilon} \mathbf{k}_{ph}) \cdot \mathbf{r}_c] / V = 3(\sin \kappa - \kappa \cos \kappa) / \kappa^3$ ,

where  $\kappa = R\sqrt{k_e^2 + \varepsilon k_{ph}^2 - 2k_e \sqrt{\varepsilon} k_{ph} \cos \theta}$ ,  $\theta$  being the angle between  $\mathbf{k}_e$  and  $\mathbf{k}_{ph}$ .

Then, let us represent the wavefunction of the system crystal+photons as  $\sum_{\mathbf{k}_{ph\alpha}} c_{\mathbf{k}_{ph\alpha}} |\Psi_0\rangle |\mathbf{k}_{ph\alpha}\rangle \exp(-it\omega_{ph}) + c_{\mathbf{k}_e} |\Psi_{\mathbf{k}_e 1}\rangle |0\rangle \exp(-itE_{\mathbf{k}_e 1}/\hbar)$ , where  $|0\rangle$  stands for a state without

photons and  $|\mathbf{k}_{ph\alpha}\rangle$  – for a state with one photon with a wavevector  $\mathbf{k}_{ph}$  and polarization  $\mathbf{e}_{\mathbf{k}_{ph\alpha}}$ ,

and it is assumed that the exciton resides only in its ground state. The system of equations for coefficients  $c_{\mathbf{k}_e}$ ,  $c_{\mathbf{k}_{ph\alpha}}$ , following from the Schrodinger equation, is

$$i\hbar \dot{c}_{\mathbf{k}_e} = - \sum_{\mathbf{k}_{ph\alpha}} (\mathbf{d} \mathbf{E}_{\mathbf{k}_{ph\alpha}}) \exp[-it(\hbar\omega_{ph} - E_{\mathbf{k}_e 1})/\hbar] c_{\mathbf{k}_{ph\alpha}} 3 \frac{\sin \kappa - \kappa \cos \kappa}{\kappa^3}, \quad (3)$$

$$i\hbar \dot{c}_{\mathbf{k}_{ph\alpha}} = - (\mathbf{d}^* \mathbf{E}_{\mathbf{k}_{ph\alpha}}^*) \exp[it(\hbar\omega_{ph} - E_{\mathbf{k}_e 1})/\hbar] c_{\mathbf{k}_e} 3 \frac{\sin \kappa - \kappa \cos \kappa}{\kappa^3}, \quad (4)$$

where  $\mathbf{d} = \mathbf{d}_{cv} \sqrt{V/(\pi r_{ex}^3)}$ .

Finding  $c_{\mathbf{k}_{ph}\alpha}$  from Eq. (4) with the initial conditions  $c_{\mathbf{k}_{ph}\alpha}(t=0) = 0$  and substituting them into Eq. (3), one obtains an integro-differential equation for  $c_{\mathbf{k}_e}$

$$i\hbar\dot{c}_{\mathbf{k}_e} = -\frac{i}{\hbar} \int_0^t \sum_{\mathbf{k}_{ph}\alpha} |\mathbf{d}\mathbf{E}_{\mathbf{k}_{ph}\alpha}|^2 \exp[-i(t-t')(\hbar\omega_{ph} - E_{\mathbf{k}_e})/\hbar] c_{\mathbf{k}_e}(t') \left(3 \frac{\sin\kappa - \kappa \cos\kappa}{\kappa^3}\right)^2 dt' \quad (5)$$

Replacing summation over  $\mathbf{k}_{ph}$  with integration according to a formula  $\sum_{\mathbf{k}_{ph}\alpha} \rightarrow V'/(2\pi)^3 \sum_{\alpha} \int d^3k_{ph}$ , taking the Laplace transform of Eq. (5), solving the issuing algebraic equation for the Laplace transform of  $c_{\mathbf{k}_e}$ ,  $c_{\mathbf{k}_e,p}$ , and taking the inverse Laplace transform of  $c_{\mathbf{k}_e,p}$ , it can be shown that under the initial condition  $c_{\mathbf{k}_e}(t=0) = 1$  one has  $|c_{\mathbf{k}_e}(t)|^2 \approx \exp[-\gamma(\mathbf{k}_e)t]$ . Here  $\gamma(\mathbf{k}_e)$ , after its averaging over the random orientations of  $\mathbf{k}_e$  with respect to  $\mathbf{d}$ , is given by a formula

$$\bar{\gamma}(k_e) = \frac{2\omega_{ph0}^2 R}{\hbar c^2 \sqrt{\varepsilon} k_e r_{ex}^3} |\mathbf{d}_{cv}|^2 \left[ \frac{\sin(2\kappa_+)}{\kappa_+^3} - \frac{1}{\kappa_+^2} - \frac{\sin^2 \kappa_+}{\kappa_+^4} - \frac{\sin(2\kappa_-)}{\kappa_-^3} + \frac{1}{\kappa_-^2} + \frac{\sin^2 \kappa_-}{\kappa_-^4} \right] \quad (6)$$

where  $\omega_{ph0} = E_{01}/\hbar$  is the resonant recombination photon frequency (the small exciton quasikinet energy  $\hbar^2 k_e^2 / [2(m_{el} + m_h)]$  is neglected here in comparison with  $E_g - e^4 m_{el} m_h / [2\hbar^2 \varepsilon_{st}^2 (m_{el} + m_h)]$ ),  $\kappa_+ = R \sqrt{(k_e + \sqrt{\varepsilon} k_{ph0})^2}$ ,  $\kappa_- = R \sqrt{(k_e - \sqrt{\varepsilon} k_{ph0})^2}$ ,  $k_{ph0} = \omega_{ph0}/c$ .

Then, it is necessary to average  $\bar{\gamma}(k_e)$  over the Maxwell distribution of exciton quasiwavevector values, so that the final formula for the exciton radiative recombination rate is

$$\langle \bar{\gamma} \rangle = \int_0^{+\infty} \bar{\gamma}(k_e) 4\pi \hbar^3 k_e^2 \exp[-(\hbar k_e)^2 / (2m_e k_B T)] / (2\pi m_e k_B T)^{3/2} dk_e, \quad (7)$$

where  $m_e = m_{el} + m_h$  is the exciton mass,  $T$  – temperature,  $k_B$  – the Boltzmann constant.

### 3. Wannier-Mott exciton lifetime in InAs nanoclusters embeded in GaAs

Now, let us apply the general Eq. (7) to the calculation of the exciton radiative lifetime in InAs nanoclusters embeded in GaAs. As the InAs bandgap energy is significantly lower than that of GaAs, such clusters form three-dimensional potential wells for excitons which are practically fully confined in them. For example, let us consider a Wannier-Mott exciton formed by an electron and a heavy hole. The InAs parameters taken from Madelung (2004) and Vurgaftman *et al.* (2001) are the following: bandgap energy is  $E_g = 0.417\text{eV}$ ,  $|\mathbf{d}_{cv}| \approx 100 \text{ D}$  ( $1\text{D} \approx 3.34 \cdot 10^{-30} \text{C}\cdot\text{m}$ ), the dielectric function at the exciton resonant recombination frequency is  $\varepsilon = 12.4$ , the static dielectric constant  $\varepsilon_{st} \approx 15.15$ , the effective electron mass is  $m_{el} \approx 0.0265m$ , the averaged heavy hole mass is  $m_h \approx 0.57m$ , and, according to the above indicated formulas,  $r_{ex} \approx 30 \text{ nm}$ , and the exciton ground state binding

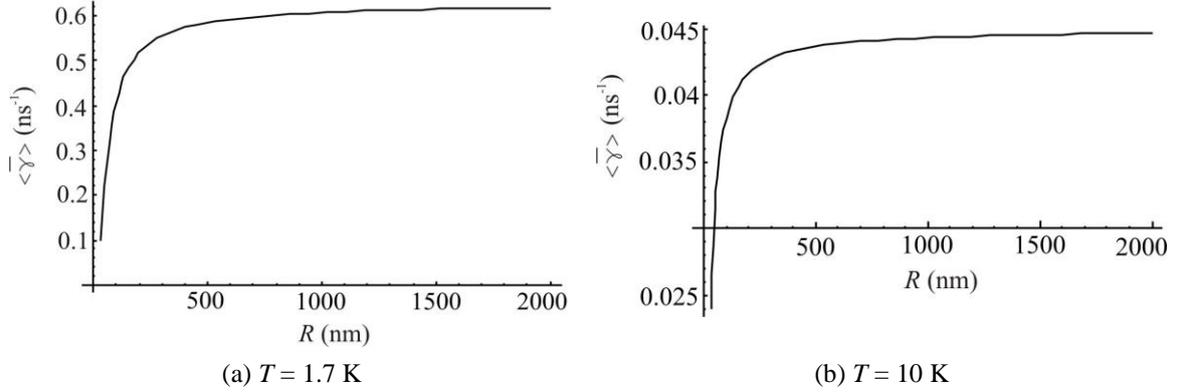


Fig. 1 The exciton radiative recombination rate  $\langle \bar{\gamma} \rangle$ , calculated according to Eq. (7) for an InAs nanocluster, as a function of its radius  $R$  at different temperatures

energy  $e^4 m_{\text{el}} m_{\text{h}} / [2 \hbar^2 \varepsilon_{\text{st}}^2 (m_{\text{el}} + m_{\text{h}})] \approx 1.6$  meV.

Let us calculate the exciton radiative recombination rate according Eq. (7) for temperatures, corresponding to the thermal energy  $k_B T$  smaller than the exciton ground state binding energy lest it be destructed by thermal fluctuations. Let us chose the nanocluster radius  $R$  grater than  $r_{\text{ex}}$  to avoid the exciton disruption by the confinement potential. The results are shown in Fig. 1.

From it one can see that at  $T = 1.7$  K the exciton radiative lifetime (i.e.,  $1/\langle \bar{\gamma} \rangle$ ) increases by almost 6 times as the InAs nanocluster radius decreases from infinity to the exciton radius. At larger temperature  $T = 10$  K (Fig. 1(b)) this effect still exists, but becomes less pronounced. So, these results allow to conclude that using nanoclusters of a narrow bandgap material with dimensions of order the Wannier-Mott exciton radius makes it possible to significantly increase the exciton radiative lifetime.

#### 4. Conclusions

Upon the whole, in the present work by means of analytical and numerical investigation it is shown that the radiative recombination lifetime of Wannier-Mott excitons can be increased by several times by exciting them in nanoclusters of a narrow-bandgap material inside a wide-bandgap one if the linear dimensions of such nanoclusters are of order the exciton radius.

#### Acknowledgments

The research, described in this article, was financially supported by the Russian Foundation for Basic Research (grants nos. 11-02-00364-a, 12-02-00855-a, and 13-02-97130-r\_povolzh'e\_a), by the Council for Grants of the President of Russian Federation for the State Support of Leading Scientific Schools (grant no. SS-4185.2012.2), and by the Skolkovo Institute of Science and Technology, Research Center "Scalable Hardware for Big Data".

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