Vibrational frequency of a strong-coupling polaron in a quantum rod at finite temperatures*

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The Hamiltonian of a quantum rod with a boundary is presented after a coordinate transformation that changes the original ellipsoidal boundary into a spherical one. We then study the effect of temperature on the vibrational frequency and the ground state binding energy of the strong-coupling polaron in the rod. The two quantities are expressed as functions of the aspect ratio of the ellipsoid, the transverse and the longitudinal effective confinement lengths, the temperature and the electron–phonon coupling strength by linear combination operator and unitary transformation methods. It is found that the vibrational frequency and the ground state binding energy will increase rapidly with decreasing transverse and longitudinal effective confinement lengths. They are increasing functions of the electron–phonon coupling strength but become decreasing ones of the temperature and the aspect ratio.

Keywords: quantum rod, polaron, temperature effect, linear combination operator

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1. Introduction

Ever since the shape-controlled colloidal quantum rod was realized by modifying its synthesis,[1,2] it has become a major focus of attention because of the freedom it offers in material tailoring and wide functional applications, such as biological labeling [3,4]and optoelectronic devices.[5] Its novel electronic structures, optical properties and linearly polarized emission have become a hot field of investigation in solid-state condensed matter physics. Many investigators studied the properties of the quantum rod in many aspects by a variety of experimental[6−8] and theoretical methods[9−11]. Li and Xia[12] investigated the electronic structure and the optical properties of a wurtzite quantum rod in the framework of effective-mass envelope-function theory. Using a theory based on the dielectric continuum approach, Comas *et al.*[13] analysed surface polar-optical phonons in a semiconductor quantum rod. The interactions between electrons and the electron–hole pairs in semiconductor nanorods embedded in dielectric media were investigated by Climente *et al.*[14] in terms of the configuration-interaction method. Experimental data were compared by Talaat *et al.*[15] with those calculated from two different theoretical models, i.e., the effective mass approximation and the semi-empirical pseudopotential method. Theoretical values for the energy bandgap at varying radii were found to be in agreement with experimental results within 0.08 eV. To our knowledge, the polaron properties in a quantum rod, however, have not been studied so far by employing the linear-combination operator method; furthermore, the effect of temperature on the properties of the vibrational frequency of the polaron in a quantum rod have never been investigated.

In the present paper, we investigate the effect of temperature on the vibrational frequency and the ground state binding energy of a strongly coupled polaron in a quantum rod, by using the linear-combination operator combined with the unitary transformation methods, taking the aspect ratio of the rod, the transverse and longitudinal effective confinement lengths and the electron–phonon coupling strength into account. It should be noted that as long as the boundary potential of the quantum rod is transformed from the ellipsoidal form into a spherical one, we can conveniently use the linear-combination operator in the framework of the effective-mass approximation to calculate the relevant physical quantities, and do not have to use the real electron boundary

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condition.

2. Theoretical model

The electron under consideration is moving in a polar crystal quantum rod with three-dimensional anisotropic harmonic potential and is interacting with bulk longitudinal optical (LO) phonons. The Hamiltonian of the electron–phonon interaction system in the rod can be written as

$$H = \frac{P_{\parallel}^2}{2m} + \frac{P_Z^2}{2m} + \sum_{\boldsymbol{q}} \hbar\omega_{\rm LO} a_{\boldsymbol{q}}^{+} a_{\boldsymbol{q}} + \frac{1}{2} m\omega_{\parallel}^2 \rho^2 + \frac{1}{2} m\omega_z^2 z^2 + \sum_{\boldsymbol{q}} \left[V_q a_{\boldsymbol{q}} \exp\left(\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}\right) + \mathrm{h.c} \right], \quad (1)$$

where m is the band mass, $\omega_{\parallel}$ and ω_z are the measures of the transverse and longitudinal confinement strengths of the potential in the xy plane and the z direction, respectively. The $a_{\boldsymbol{q}}^{+}$ ($a_{\boldsymbol{q}}$) denotes the creation (annihilation) operator of the bulk LO phonons with wave vector $\boldsymbol{q}(q_z, \boldsymbol{q}_{\parallel})$; $\boldsymbol{P} = (\boldsymbol{P}_{\parallel}, P_z)$ and $\boldsymbol{r} = (\boldsymbol{\rho}, z)$ are, respectively the momentum and the position vectors of an electron, with

$$V_q = \mathrm{i}\left(\frac{\hbar\omega_{\rm LO}}{q}\right)\left(\frac{\hbar}{2m\omega_{\rm LO}}\right)^{\frac{1}{4}}\left(\frac{4\pi\alpha}{V}\right)^{\frac{1}{2}},$$
$$\alpha = \left(\frac{e^2}{2\hbar\omega_{\rm LO}}\right)\left(\frac{2m\omega_{\rm LO}}{\hbar}\right)^{\frac{1}{2}}\left(\frac{1}{\varepsilon_\infty} - \frac{1}{\varepsilon_0}\right). \quad (2)$$

For a quantum rod, the boundary condition is different from that of a spherical quantum dot. In order to simplify our boundary condition, we introduce a coordinate transformation to change the original boundary into a spherical one, which has a better symmetrical characteristic.[12] The transformation is $x' = x$, $y' = y$, $z' = z/e'$, where e' is the aspect ratio of the ellipsoid, (x, y, z) is the actual coordinate and (x', y', z') is the transformed one. The electron–phonon system Hamiltonian in the new coordinate is changed to H'. We subsequently introduce the following linear combinations of the momenta and positions of the electron as operators b^{+} and b:

$$p_j = \left[\frac{m\hbar\lambda}{2}\right]^{\frac{1}{2}}\left(b_j + b_j^{+}\right),$$
$$r_j = \mathrm{i}\left[\frac{\hbar}{2m\lambda}\right]^{\frac{1}{2}}\left(b_j - b_j^{+}\right), \quad (3)$$

where $j = x, y, z$, λ are the variational parameters, and b^{+} and b are Boson operators satisfying the Boson commutative relation. Equation (3) is then inserted into H' and a unitary transformation is performed to H' with the following unit operator:

$$U = \exp\left[\sum_{\boldsymbol{q}}\left(a_{\boldsymbol{q}}^{+} f_q - a_{\boldsymbol{q}} f_q^{*}\right)\right], \quad (4)$$

where $f_q(f_{\rm q}^{*})$ is the variational function. We choose a trial wavefunction for $|\psi\rangle$ at a finite temperature with

$$|\psi\rangle = |\{n_{\boldsymbol{q}}\}\rangle_{\rm a} |\{n_j\}\rangle_{\rm b}, \quad (5)$$

where $|\{n_{\boldsymbol{q}}\}\rangle_{\rm a}$ and $|\{n_j\}\rangle_{\rm b}$ are respectively the wavefunctions for the phonon and the polaron states with $\{n_{\boldsymbol{q}}\}$ and $\{n_j\}$ being the numbers of bulk LO phonons and the polarons. We now obtain

$$F(\lambda, f_q) = \langle\psi| U^{-1} H' U |\psi\rangle, \quad (6)$$

where

$$F(\lambda, f_q) = \frac{\hbar\lambda}{2}(2n+1) + \frac{\hbar\lambda e'^2}{4}(2n+1) + \frac{\hbar\omega_{\parallel'}^2}{2\lambda}(2n+1) + \frac{\hbar\omega_{z'}^2}{4\lambda e'^2}(2n+1) + \sum_q \hbar\omega_{\rm LO}\left(n_q + |f_q|^2\right) + \sum_q \left(V_q f_q \exp\left(-\frac{\hbar\left(q_{\parallel'}^2 + e'^2 q_{z'}^2\right)}{4m\lambda}\right) B(n) + \mathrm{h.c}\right), \quad (7)$$

where

$$B(n) = \langle\{n_j\}| \exp\left[-\left(\frac{\hbar}{2m\lambda}\right)^{1/2}\sum_j b_j^{+} q_j\right] \exp\left[\left(\frac{\hbar}{2m\lambda}\right)^{1/2}\sum_j b_j q_j\right] |\{n_j\}\rangle$$
$$= 1 - n\frac{\hbar\left(q_{\parallel'}^2 + e'^2 q_{z'}^2\right)}{2m\lambda} + 0\left(\frac{\hbar\left(q_{\parallel'}^2 + e'^2 q_{z'}^2\right)}{2m\lambda}\right)^2. \quad (8)$$

In the above calculating procedures, we have neglected the higher-order small quantity terms of the wave vector. We can obtain f_q by applying the variational method to f_q^*. Inserting f_q into F and replacing the summation by the integral, we have

$$F(\lambda) = \left(\frac{\hbar\lambda}{2} + \frac{\hbar\lambda e'^2}{4} + \frac{\hbar\omega_{\parallel'}^2}{2\lambda} + \frac{\hbar\omega_{z'}^2}{4\lambda}\right)(2n+1) + \bar{n}_{\boldsymbol{q}}\hbar\omega_{\rm LO} - \frac{2\alpha\sqrt{\omega_{\rm LO}\lambda}}{\sqrt{\pi}}\left(1-\frac{n}{2}\right)A(e'), \quad (9)$$

where $\bar{n}_{\boldsymbol{q}}$ and n are the average number of the bulk LO phonons and the number of the polaron. The $A(e')$ is written as

$$A(e') = \begin{cases} \dfrac{\arcsin\sqrt{1-e'^2}}{\sqrt{1-e'^2}} & (e'<1) \\ 1 & (e'=1) \\ \dfrac{1}{2\sqrt{e'^2-1}}\ln\dfrac{e'+\sqrt{e'^2-1}}{e'-\sqrt{e'^2-1}} & (e'>1). \end{cases} \quad (10)$$

Performing the variation of $F(\lambda)$ with respect to λ and choosing the usual polaron unit ($\hbar = 2m = \omega_{\rm LO} = 1$), we have

$$\lambda^2 - \frac{2\alpha\left(1-\frac{n}{2}\right)A(e')}{\sqrt{\pi}(2n+1)\left(1+\frac{e'^2}{2}\right)}\lambda^{\frac{3}{2}} - \frac{\left(\frac{4}{l_{\parallel}^4}+\frac{2}{l_z^4 e'^2}\right)}{\left(1+\frac{e'^2}{2}\right)} = 0, \quad (11)$$

where λ is the vibrational frequency of the polaron, and $l_{\parallel} = \sqrt{\hbar/m\omega_{\parallel'}}$, $l_z = \sqrt{\hbar/m\omega_{z'}}$ are the transverse and longitudinal effective confinement lengths, respectively. Solving Eq. (11), we obtain

$$\lambda = \lambda_0. \quad (12)$$

By minimizing the expectation value of the Hamiltonian, that is, substituting λ_0 into Eq. (9), we obtain the polaron ground state energy E_0. If $E_{\rm e}$ and $E_{\rm p}$ denote the energies of the uncoupled electron and the phonon, respectively, then the ground state binding energy of the strong-coupling polaron is given by

$$E_{\rm b} = E_{\rm e} + E_{\rm p} - E_0 = \frac{3\alpha\sqrt{\lambda_0}}{\sqrt{\pi}}\left(1-\frac{n}{2}\right)A(e') - \bar{n}_{\boldsymbol{q}} - \left(\frac{2}{\lambda_0 l_{\parallel}^4} + \frac{1}{\lambda_0 l_z^4 e'^2}\right)(2n+1). \quad (13)$$

3. Temperature effects

At finite temperature, the electron–phonon system is no longer entirely in the ground state. The lattice vibrations excite not only the real phonon but also the electron in a parabolic potential well. The properties of the polaron are statistical averages over various states. According to the quantum statistics theory, the statistical average number of the bulk LO phonons and the polaron are given by

$$\bar{n}_{\boldsymbol{q}} = \left[\exp(\delta) - 1\right]^{-1}, \qquad \bar{n} = \left[\exp(\delta\lambda) - 1\right]^{-1}, \quad (14)$$

where $\delta = \hbar/k_{\rm B}T$ is the temperature parameter and $k_{\rm B}$ is the Boltzmann constant. With the considerations mentioned above, the value of λ_0 determined by Eq. (11) relates not only to the value of n but also to $\bar{n}$, which should be self-consistently calculated with Eq. (14) to obtain the relations between λ, $E_{\rm b}$ and n, T.

4. Numerical results and discussion

Now we perform numerical calculations to show the effects of the aspect ratio e' of the ellipsoid, the transverse and longitudinal effective confinement lengths $l_{\parallel}$ and l_z, the electron–phonon coupling strength α and the temperature parameter δ on the strong-coupling polaron vibrational frequency λ and the ground state binding energy $E_{\rm b}$.

Figures 1 and 2 show the relationship between the vibrational frequency λ and the ground state binding energy $E_{\rm b}$ of the polaron varying with the aspect ratio of the ellipsoid e' for fixed $\delta = 0.8$, $l_z = 0.8$ and $l_{\parallel} = 0.7$, respectively. From the two figures we can see that the vibrational frequency λ and the ground state binding energy $E_{\rm b}$ increase with decreasing e'. This result is in agreement with those obtained in Refs. [12], [16] and [17]. For ellipsoidal quantum rods, we can define the aspect ratio of the ellipsoid as $e' = L/2R$, where L and R are the longitudinal length and the transverse radius of the rod, respectively. With the increase of e', that is, by increasing the longitudinal length L of the rod, the electron thermal motion energy and the interaction between electrons and the phonons, which take the phonons as medium, are reduced because of the larger motion space of the particles. As a result, the polaron vibrational frequency and the ground state binding energy all decrease with increasing e'. This result is similar to

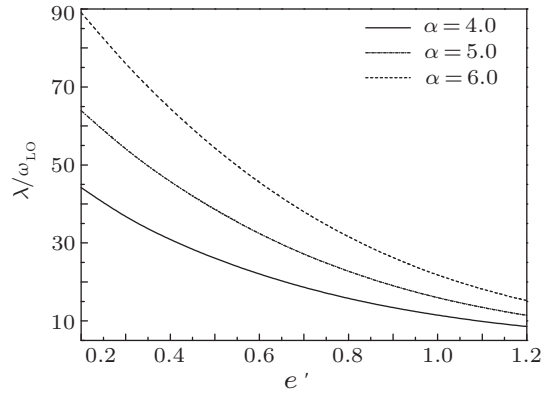


Fig. 1. Variations of the vibrational frequency λ with the aspect ratio e' for fixed $\delta = 0.8$, $l_z = 0.8$ and $l_{\|} = 0.7$.

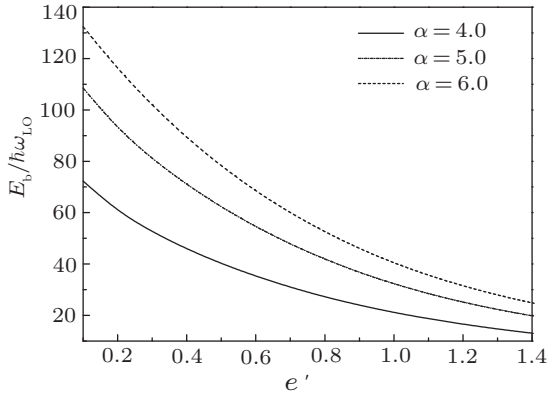


Fig. 2. Variations of the ground state binding energy $E_{\rm b}$ with the aspect ratio e' for fixed $\delta = 0.8$, $l_z = 0.8$ and $l_{\|} = 0.7$.

that of the quantum wire, which was early obtained in Ref. [18]. Conversely, with the decrease of the aspect ratio, the electron thermal motion energy and the electron–phonon interaction are enhanced because of the smaller particle motion space. Correspondingly, the vibrational frequency and the ground state binding energy all increase due to the interesting quantum size effects. This result is also similar to that of the quantum well case in Ref. [19]. This indicates that when the aspect ratio is quite small, the shape of the rod approaches a two-dimensional quantum well. At the extreme case of the aspect ratio being 1, the rod becomes a zero-dimensional dot; if the aspect ratio is large enough, the rod resembles a one-dimensional quantum wire. In other words, quantum rods constitute a bridge between two-dimensional quantum wells, zero-dimensional quantum dots and one-dimensional quantum wires. Studies of the transition regime from quantum wells to quantum dots and to quantum wires are of particular interest in the case of colloidal semiconductor quantum rods because size and shape control enable the synthesis of quantum rods with precise length and diameter. Therefore, by tuning the aspect ratio of the ellipsoid, one can follow the transition from two- to zero- to one-dimensional systems. This property offers the opportunity for designing new quantum devices. From the two figures one can also see that the vibrational frequency and the ground state binding energy are increasing functions of the electron–phonon coupling strength. This is because the larger the electron–phonon coupling strength is, the stronger the electron–phonon interaction is. Therefore, it leads to the increase of the electron energy and makes the electrons interact with more phonons, resulting in increased vibrational frequency and ground state binding energy. It is known that the electron–phonon interaction strength α is different in different crystal materials. Thus, the vibrational frequency and the ground state binding energy can be tuned by changing α. In quantum bit, the electron–phonon interaction strength will increase when the system is confined, resulting in larger transition frequency and the destroying of the super state (decoherence).[20−22] The present result of controlling the super state by tuning ground state may have practical applications in quantum information processes.

Figures 3 and 4 depict the vibrational frequency λ and the ground state binding energy $E_{\rm b}$ as functions of the temperature parameter δ for fixed $e' = 1.4$, $l_z = 0.8$ and $\alpha = 4.0$. From the two figures we can see that λ and $E_{\rm b}$ increase with decreasing temperature since the confusion extent of the lattice thermal motion is enhanced by increasing temperature and weakening electron-lattice polarisation field. In other words, the height of the parabolic potential for the strong-coupling polaron is lowered. With the increase of the temperature, the thermal motion of the phonons around the electron is enhanced, which weakens the interaction strength between the electron and the phonons and consequently reduces the polaron vibrational frequency and the ground state binding energy. This indicates that we provide a new way of controlling the rod's vibrational frequency and the ground state binding energy via the system temperature. In fact, the temperature effect plays an important role in low-dimensional materials, for example, both the decoherence and the electron density are affected by the temperature. Finding a suitable two-state system by adjusting the system temperature is crucial in constructing a quantum bit.[23] From the two figures one can also see that the vibrational frequency and the

ground state binding energy are decreasing functions of the transverse effective confinement length.

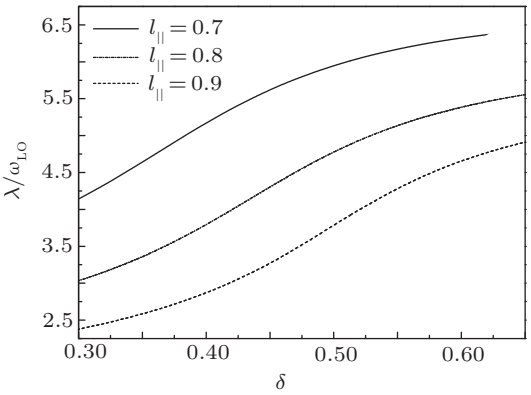


Fig. 3. Variations of the vibrational frequency λ with the temperature parameter δ for fixed $e' = 1.4$, $l_z = 0.8$ and $\alpha = 4.0$.

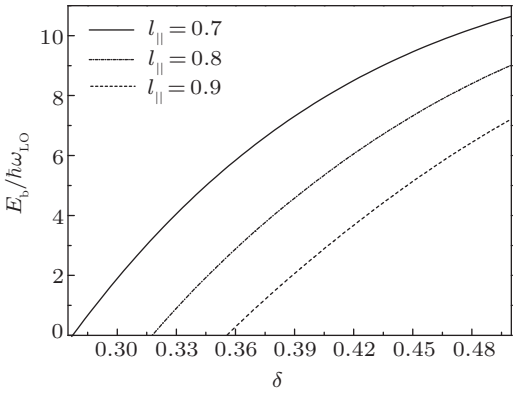


Fig. 4. Variations of the ground state binding energy with the temperature parameter δ for fixed $e' = 1.4$, $l_z = 0.8$ and $\alpha = 4.0$.

Figures 5 and 6 present the vibrational frequency λ and the ground state binding energy $E_{\rm b}$ as functions of the transverse and longitudinal effective confinement length $l_{\|}$ and l_z for fixed $\delta = 0.8$, $e = 1.4$ and $\alpha = 4.0$. From the two figures one can see that the vibrational frequency and the ground state binding energy increase rapidly with decreasing transverse and longitudinal effective confinement lengths. From the expressions of $l_{\|} = \sqrt{\hbar/m\omega_{\|'}}$ and $l_z = \sqrt{\hbar/m\omega_{z'}}$, we can see that the effective confinement length $l_{\|}$ and l_z are reciprocal with the square root of the confinement strength $\omega_{\|'}$ and $\omega_{z'}$, and then the vibrational frequency and the ground state binding energy will increase with increasing confinement strengths since the motion of the electrons is confined by the confining potential. With the increase of the confining potential ($\omega_{\|'}$, $\omega_{z'}$), that is, with decreasing ρ', z', the thermal motion energy of the electrons and the interaction between an electron and the phonons are enhanced because of the smaller particle motion space. As a result, the vibrational frequency and the ground state binding energy of the polaron all increase, which can also be attributed to the quantum size effects. From another point of view, since the presence of the parabolic potential is equivalent to introducing another confinement on the electrons, which leads to greater electron wavefunction overlapping with each other, the electron–phonon interactions will be enhanced, and the vibrational frequency and the ground state binding energy appears more obviously as shown in Figs. 5 and 6 in which the curves increase with increasing confinement strength.

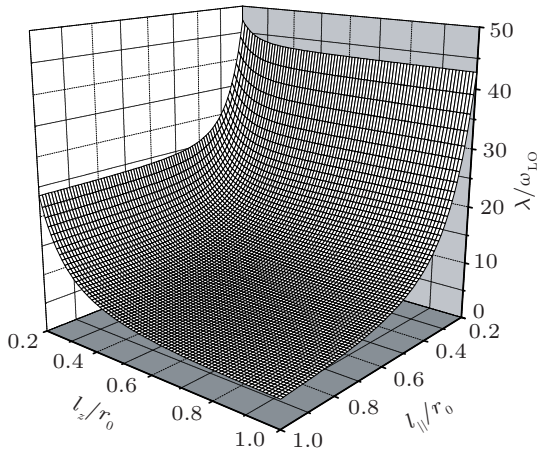


Fig. 5. Variations of the vibrational frequency λ with the transverse and longitudinal effective confinement length $l_{\|}$ and l_z for fixed $\delta = 0.8$, $e = 1.4$ and $\alpha = 4.0$.

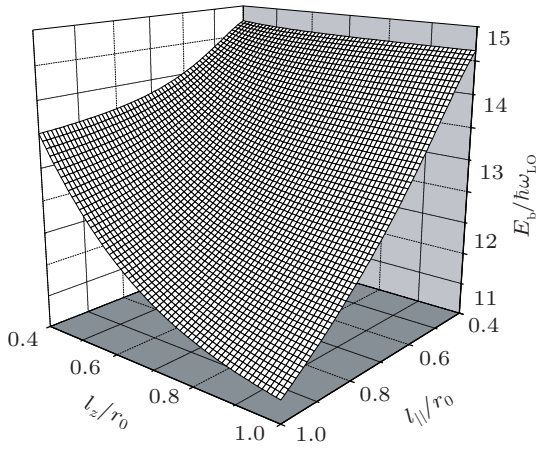


Fig. 6. Variations of the ground state binding energy $E_{\rm b}$ with the transverse and the longitudinal effective confinement length $l_{\|}$ and l_z for fixed $\delta = 0.8$, $e = 1.4$ and $\alpha = 4.0$.

5. Conclusion

Based on the linear combination operator method, we have investigated the effect of temperature on the vibrational frequency and the ground state binding energy of a strong-coupling polaron in a quantum rod. It is found that the vibrational frequency and the ground state binding energy will increase rapidly with decreasing transverse and longitudinal effective confinement lengths. They are increasing functions of the electron–phonon coupling strength while become decreasing ones of the temperature and the ellipsoid aspect ratio.

References

[1] Hu J T, Li L S, Yang W D , Manna L, Wang L W and Alvisatos A P 2001 *Science* **292** 2060

[2] Kan S H, Mokari T, Rothenberg E, Rothenberg E and Banin U 2003 *Nature* **2** 155

[3] Bruchez M, Moronne M, Gin P, Weiss S and Alivisatos S P 1998 *Science* **281** 2013

[4] Chi W, Chan W and Nie S 1998 *Science* **281** 2016

[5] Klimov V I, Mikhailovsky A A, Xu S, Malko A, Hollingsworth J A, Leatherdale C A, Eisler H J and Bawendi M G 2000 *Science* **290** 314

[6] Sek G, Podemski P, Misiewicz J, Li L H, Fiore A and Patriarche G 2008 *Appl. Phys. Lett.* **92** 021901

[7] Persano A, Leo G, Manna L and Cola A 2008 *J. Appl. Phys.* **104** 074306

[8] Bruhn B, Valenta J and Linnros J 2009 *Nanotechnology* **20** 505301

[9] Creti A, Rossi M Z, Lanzani G, Anni M, Manna L and Lomascolo M 2006 *Phys. Rev.* B **73** 165410

[10] Zhang X W and Xia J B 2005 *Phys. Rev.* B **72** 205314

[11] Sun Z X, Swart I, Delerue C, Vanmaekelbergh D and Liljeroth P 2009 *Phys. Rev. Lett.* **102** 196401

[12] Li X Z and Xia J B 2002 *Phys. Rev.* B **66** 115316

[13] Comas F, Studart N and Marques G E 2004 *Solid State Commun.* **130** 77

[14] Climente J I, Royo M, Movilla J L and Planelles J 2009 *Phys. Rev.* B **79** 161301(R)

[15] Talaat H, Abdallah T, Mohamed M B, Negm S, Mostafa A and El-Sayed M A 2009 *Chem. Phys. Lett.* **473** 288

[16] Li J B and Wang L W 2003 *Nano. Lett.* **3** 101

[17] Hu J T, Wang L W, Li L S, Yang W D and Alivisatos A P 2002 *J. Phys. Chem.* B **106** 2247

[18] Planelles J, Royo M, Ballester A and Pi M 2009 *Phys. Rev.* B **80** 045324

[19] Li S S and Xia J B 2008 *Appl. Phys. Lett.* **92** 022102

[20] Wang Z W and Xiao J L 2007 *Acta Phys. Sin.* **56** 678 (in Chinese)

[21] Yin J W, Xiao J L, Yu Y F and Wang Z W 2009 *Chin. Phys.* B **18** 446

[22] Li H J, Sun J K and Xiao J L 2010 *Chin. Phys.* B **19** 010314

[23] Chen Y J and Xiao J L 2008 *Acta Phys. Sin.* **57** 6758 (in Chinese)