PERGAMON

Solid State Communications 117 (2001) 713–717

solid state communications

www.elsevier.com/locate/ssc

Acoustic phonon bottleneck in quantum dots: role of deformation variation of electron effective mass

V.I. Pipa[a,b], V.V. Mitin[a,*], M. Stroscio[c]

[a]*Department of Electrical and Computer Engineering, Wayne State University, Detroit, MI 48202, USA*
[b]*Institute of Semiconductor Physics, Kiev 252650, Ukraine*
[c]*US Army Research Office, P.O. Box 12211, Research Triangle Park, NJ 27709, USA*

Received 7 December 2000; accepted 3 January 2001 by D.J. Lockwood

Abstract

We demonstrate that phonon-induced variation of electron effective mass leads to a substantial increase of the electron–acoustic phonon interaction in semiconductor quantum dots. The contribution of this mechanism to electron scattering depends on a shape of the nanostructure. For dots of small sizes it exceeds the contribution of the conventional deformation potential and it prevents the reduction of the electron relaxation rate with decreasing dot size as predicted by the conventional theory. © 2001 Elsevier Science Ltd. All rights reserved.

Keywords: A. Semiconductors; D. Electron–phonon interactions; D. Phonons

PACS: 72.10.Di; 73.50.Bk

Research on electron energy relaxation in semiconductor quantum dots (QDs) has attracted much interest due to the critical importance of carrier-relaxation processes in the performance of novel semiconductor devices based on QDs (lasers, infrared photodetectors and others). The relaxation via phonon emission has been of a particular interest. This intrinsic mechanism influences fundamental properties of semiconductor nanostructures and may be the only nonradiative process controlling electron energy losses. Among various aspects of phonon-assisted relaxation, the effect of phonon bottleneck predicted in Ref. [1] has been receiving widespread and increasing attention in recent years; see, e.g. Ref. [2] (and references therein). This effect is the basis of large predicted reductions of electron relaxation rates in QDs when compared to 2D or 1D heterostructures. Due to the discrete electron energy spectrum, the interaction with phonons occurs only when an interlevel interval matches the energy of longitudinal optical (LO) phonons or it is smaller than the bandwidth of longitudinal acoustic (LA) phonons (a few meV). For dot sizes less than some critical value, the interaction or electrons with acoustic phonons weakens drastically due to the decreasing form factor. The existence of a phonon bottleneck effect is still discussed controversially [2]: some experimental results [3,4] support the theoretical prediction [1] while other studies [5,6] show that the bottleneck does not exist. Assuming that the intrinsic process of acoustic phonon emission cannot provide rapid relaxation, different extrinsic mechanisms have been investigated (Auger-like [7], multiphonon scattering [8] and others). While restricted for intrinsic phonon scattering, a large enhancement of the relaxation rate around the energy of LO phonons has been calculated by including the LO ± LA two-phonon mechanism [9] and the decay of LO phonons into acoustic phonons [2]. These intrinsic mechanisms do not restrict a large decrease in the relaxation in low-energy range.

In this paper, we investigate electron energy relaxation due to acoustic phonon emission by reexamining the electron–acoustic phonon coupling in QDs. It is worth emphasizing that the acoustic phonons bottleneck has been predicted using the conventional interaction energy, D div $\mathbf{u}$, where D is the deformation potential (DP) constant, and $\mathbf{u}$ is the lattice displacement. This energy takes into account only the shift of the conduction band edge given by deformation potential. In nanostructures this bulk form of interaction has to be modified by taking into account a shift

* Corresponding author. Tel.: +1-313-577-8944; fax: +1-313-577-1101.

E-mail address: mitin@ece6.eng.wayne.edu (V.V. Mitin).

PII: S0038-1098(01)00016-3

of energy levels of confined electrons relative to their band edge. The expression for a quantized energy, $E \sim \hbar^2/(m^* L^2)$ indicates that an additional coupling originates from phonon induced changes of the dot size, L; and the electron effective mass, m^*. For weak deformation, these changes give rise to additional mechanisms acting independently. The first of them which is caused, in the general case, by a change of a confinement potential has been investigated in [10–12]. We refer to it as the macroscopic deformation potential (MDP) [10]. The corresponding perturbation is given by

$$\hat{H}_1 = -\mathbf{u}(\mathbf{r})\nabla U(\mathbf{r}), \tag{1}$$

where $U(\mathbf{r})$ is a confinement potential. For electrons confined in quantum wells, only long wavelength phonons participate in scattering, and MDP interaction is weak compared with the DP interaction as $E/D \ll 1$. For electrons in QDs, due to the large wave vectors of phonons participating in scattering, the contribution of the MDP interaction can be comparable with that from the DP coupling [11]. The tight-binding approach for Bloch states of 2D electrons in a deformed crystal [12] and calculation of the electron mobility in quantum wells [13] show that the main additional interaction originates from deformation-related variation of the electron effective mass. To derive this interaction for electrons in QDs, we expand the tensor of the inverse effective mass, m_{ij}^{-1}, in terms of the strain tensor, u_{ij}. In a cubic crystal, $m_{ij}^{-1} = (1/m^*)(1 + \chi u_{ij})\delta_{ij}$, where m^* is the isotropic effective mass in the absence of deformation and χ is a phenomenological parameter. Using the kinetic energy operation of the Ben Daniel–Duke form $(-\hbar^2/2)\nabla(m^{-1}(\mathbf{r})\nabla)$, yields the interaction Hamiltonian

$$\hat{H}_2 = -\frac{\hbar}{2}\sum_i \nabla_i\left(\frac{\chi}{m^*}u_{ii}\nabla_i\right). \tag{2}$$

This operator takes into account a difference in the effective masses of the semiconductors forming a nanostructure. In narrow-gap semiconductors, the parameter χ can be expressed through the slope dE_g/dP of the dependence of the bandgap energy, E_g on pressure P. Using the relationship $1/m^* = 2P_\Gamma^2/(m_0^2 E_g)$, where P_Γ is the interband $\mathbf{p}$-matrix element from $\mathbf{k}\cdot\mathbf{p}$ perturbation theory we obtain

$$\chi \simeq \frac{(C_{11} + 2C_{12})}{E_g}\frac{dE_g}{dP} \simeq -\frac{3D}{E_g}, \tag{3}$$

where C_{11} and C_{12} are elastic moduli of a cubic crystal in the Voigt notation. The last term in the righthand part holds when a shift of conduction band edge provides the main contribution to the pressure dependence of E_g (this is true for practically all semiconductors [14]). Using the data for the elastic moduli and the pressure coefficients given in Ref. [15], we find $\chi \simeq 17$ for GaAs, $\chi \simeq 28$ for $In_{0.53}Ga_{0.47}As$ ternary, and $\chi \simeq 42$ for InAs. The estimation χ by help of the last term of Eq. (3) is in acceptable agreement with above obtained values when the $D \simeq -8.1, -7$, and -5.7 for the above listed semiconductors, correspondingly. The condition $\chi \gg 1$ implies that the interaction of Eq. (2) dominates over the MDP interaction.

In the present communication we calculate the scattering rate of an electron in a QD due to the interaction with acoustic phonons through the deformation potential and the additional interface mechanisms of Eqs. (1) and (2). We will ignore the effect of acoustic mismatches at the interfaces and assume bulk acoustic phonons. The rate of electron relaxation associated with acoustic phonon emission is calculated in the first-order of perturbation using the Fermi golden rule. For a single electron,

$$\frac{1}{\tau_{if}} = \frac{2\pi}{\hbar}\sum_{j\mathbf{q}}|M_{if}^j|^2(N(\hbar\omega_j/k_B T) + 1)\delta(E_i - E_f - \hbar\omega_j). \tag{4}$$

Here M_{if}^j is the matrix element of a transition between the initial, i, and the final, f, electron states, $\omega = s_j q$ and $\mathbf{q}$ are the frequency and wave vector of phonons, respectively, $j = l$ and $j = t_{1,2}$ label longitudinal and two transverse phonon modes, respectively, s_j is sound velocity, and N is the Planck distribution function. In contrast to the case of the bulk mechanism, which allows only the interaction with longitudinal phonons in isotropic media, transverse acoustic phonons contribute to the interactions of Eqs. (1) and (2) as well.

Consider a QD of parallelepiped shape with transverse dimensions $L_x = L_y = L$ and thickness L_z. We restrict our consideration to cubic QDs ($L_z = L$) and flattened QDs ($L_z \ll L$). In order to obtain analytical expressions for the relaxation rates, the conduction-band offset in the heterojunction system is assumed to be infinite. This assumption provided we separate the electron motion in all three spatial directions. The numerical calculations of the relaxation rates are carried out for flattened $In_{0.53}Ga_{0.47}As$ QDs embedded in InP serves as the 2D basis. For this semiconductor system which has been considered in Refs. [1,11], we take into account a finite height of potential barriers outside the quantum well region.

In a QD with potential barriers of infinite height outside the dot, the effective-mass electronic wave function is given by a product of harmonic functions $\sin(\pi n_\alpha x_\alpha/L_\alpha)$ where n_α are positive integers that label the states, $\alpha = (x, y, z)$. For the ground state the set n_α equals (111). For the threefold-degenerate first-excited state in a cubic dot, it equals (211), (121) or (112) (the x-, y- or z- states correspondingly) and (211) or (121) for the twofold-degenerate first-excited in a QD with the shape of parallelepiped ($L_z \ll L$). The energy of transition from the excited state to the ground state is $E_1 = 3\pi^2\hbar^2/(2m^* L^2)$.

The continuum approximation for lattice vibrations as well as the previously-given expressions for interaction Hamiltonian hold when the length of the phonon wave vector involved in scattering $q_0 = E_1/\hbar s_l$ is small compared to inverse lattice constant b^{-1}. For $In_{0.53}Ga_{0.47}As$ QDs, the criterion $q_0 b < 1$ is satisfied when $L > 70$ nm. For QDs with sizes satisfying this condition, the wave vectors q_0 of

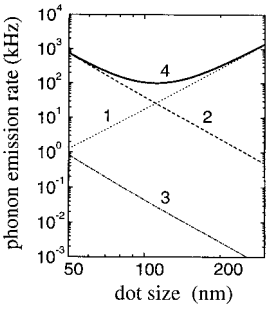

Fig. 1. Dependence of the acoustic phonon emission rates associated with electron scattering via the different mechanisms of interaction with acoustic phonons versus size L for $In_{0.53}Ga_{0.47}As$ cubic-shaped quantum dot at temperature $T = 300$ K : bulk deformation potential (1), deformation dependence of electron effective mass (2), macroscopical deformation potential (3), and total interaction (4).

emitted phonons are found to be substantially larger than the inverse dot size L^{-1}; i.e. $Q_0 \equiv q_0 L/2 \gg 1$. Analysis shows that the phonons which are emitted due to transition from the z-state have the wave vector oriented predominantly along the z axis (similar to that for DP and MDP scattering [1,11]). In cubic QDs, the matrix element of transitions from z-state to the ground state due to emission of LA phonons is expressed as

$$M_z = \left(1 + \frac{m^* s_l^2}{D} I_z(\mathbf{Q})\right) M_z^{(\mathrm{DP})}, \tag{5}$$

where $M_z^{(\mathrm{DP})}$ is the matrix element for DP interaction, $\mathbf{Q} = \mathbf{q}L/2$. The factor I_z ($\mathbf{Q}$) comprises the contributions of additional interactions given by Eqs. (1) and (2). For $Q_0 = q_0 L/2 \gg 1$, $I_z = -(4/(3\pi^2))^2(\chi - 1)Q_0^4$. We see that the partial contributions of all three types of interactions are in phase. As a result, $|M_z|^2$ is not represented by a sum of the squared matrix elements of the particular mechanisms and it depends on the sign of D. Since for small pressure $dE_g/dP > 0$ [15], we will assume, based on Eq. (3), that $D < 0$. Electron transitions from the x- and y-states give the same contribution. The contribution of TA phonons to the scattering rate was found to be smaller than the contribution of LA phonons due to interaction defined by Eq. (2) by a factor of the order of $Q_0^{-2} \ll 1$. Neglecting this contribution, one can express the total rate as a renormalized DP scattering rate:

$$\tau^{-1} = \frac{2^{11} D_{\mathrm{cub}}^2 L^2 m^{*5} s_l^3 \cos^2 Q_0}{9\pi^6 \hbar^6 \rho}(N + 1), \tag{6}$$

where ρ is the crystal density and the effective DP constant for cubic dots, D_{cub}, is given by

$$D_{\mathrm{cub}} = D - (\chi - 1)\frac{\hbar^2 q_0^2}{4m^*} \simeq D\left(1 + \frac{3E_l^2}{4m^* s_l^2 E_g}\right). \tag{7}$$

In obtaining the approximate expression of Eq. (7), we have neglected the small contribution of the MDP interaction and used the relationship of Eq. (3). It is important to stress here that the rates reported in Ref. [11] can be obtained from Eqs. (6) and (7) if one assumes that $\chi \equiv 0$ (as we already indicated, $\chi \gg 1$) and in addition the interference between the DP and MDP mechanisms is neglected. The interference term is of principal importance since under the assumption $\chi \equiv 0$, the absolute value of D_{cub} should decrease as the dot size decreases ($D_{\mathrm{cub}} < 0$ for $|D| \gg \frac{\hbar^2 q_0^2}{4m^*}$), turn into zero, and become positive for the limiting case of small dots, $\frac{\hbar^2 q_0^2}{4m^*} \gg |D|$ (these small dots were discussed in Ref. [11]). The correct result with $\chi \gg 1$ in Eq. (7) demonstrates that D_{cub} and the bulk deformation potential, D, have the same sign, and that the value of D_{cub} always exceeds the value of D. The effective deformation potential in cubic QDs depends on the dot size, electron effective mass and other parameters of a semiconductor material.

Dependence of the phonon emission rate (6) on the dot size is depicted in Fig. 1[1]. In order to illustrate the distinction between different mechanisms more clearly, we replace the rapidly oscillating function $\cos^2 Q_0$ with 1/2. It is seen that the conventional DP scattering (curve 1) dominates in QDs of large size. When the dot size decreases, the corresponding rate (Eq. (6) with $D_{\mathrm{cub}} = D$) decreases as L^2 due to decrease of the electron form factor as L^6. But due to the rapid increase (as L^{-6}) of the contribution of substantially nonlocal mechanism (2) (curve 2), the total rate (curve 4), increases. It is seen also that the contribution of MDP coupling (curve 3) is small. The scattering rates associated with the bulk DP coupling and interface mechanism of Eq. (2) are equal when the dot size is $L_c = 3^{1/2}\pi\hbar(\chi/(16m^{*3}s_l^2|D|))^{1/4}$. Estimation for $In_{0.53}Ga_{0.47}As$ QDs, $L_c \simeq 125$ nm, shows that the dimensions of a dot for which the emission rate due to mechanism of Eq. (2) prevails over that of DP coupling still satisfy the long-wavelength approximation.

Now consider parallelepiped shape QDs with transverse dimensions $L_x = L_y = L$ considerably exceeding their thickness L_z. Electron transitions from x- and y-states yield LA phonon emission with the rate

$$\tau_l^{-1} = \frac{2^{13} D_{\mathrm{par}}^2 L^2 m^{*5} s_l^3 \sin^2 Q_0}{81\pi^6 \hbar^6 \rho}(N + 1), \tag{8}$$

where the effective deformation potential is given by

$$D_{\mathrm{par}} = D - (\chi - 1)\frac{\hbar^2 q_0^2}{4m^*} + (\chi - 2)\frac{\pi^2 \hbar^2}{2m^* L_z^2}. \tag{9}$$

To obtain the rate (8) in the framework of the conventional bulk interaction, one has to use a tensor deformation potential, $D_{ij}u_{ij}$, with $D_{xx} = D_{yy} = D_{\mathrm{cub}}$ and $D_{zz} = (\chi - 2)\pi^2\hbar^2/(2m^* L_z^2)$. The necessity of the two different DP constants, in contrast to a single constant in cubic

[1] We use the following $In_{0.53}Ga_{0.47}As$ parameters: $D = -7.2$ eV, $m^* = 0.043\, m_0$, $s_l = 4.3 \times 10^5$, $s_t = 2.3 \times 10^5$ cm/s, $\rho = 5.4$ g/cm^3.

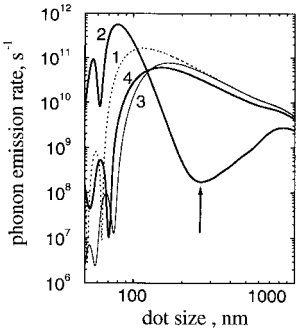

Fig. 2. Dependence of the acoustic phonon emission rates associated with scattering via bulk deformation potential (dotted curve 1 and thin solid curve 3) and total interaction (solid curves, 2, 4) versus size L for $In_{0.53}Ga_{0.47}As$ parallelepiped-shaped quantum dot with the in-plane dimension $L \times L$ and the thickness $L_z = 5$ nm at temperature $T = 300$ K. Curves 1, 2 correspond to the infinite potential barriers and 3, 4 to 0.22 eV.

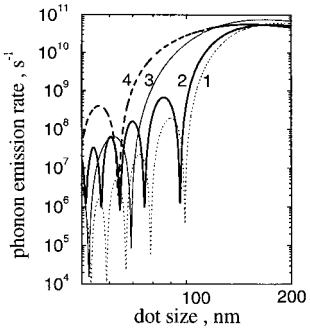

Fig. 3. Dependence of the acoustic phonon emission rates associated with scattering via bulk deformation potential (dotted curve 1 and thin solid curve 3) and total interaction (solid curve 2 and dashed curve 4) versus size L for $In_{0.53}Ga_{0.47}As$ parallelepiped-shaped quantum dot with the in-plane dimension $L \times L$ and the thickness $L_z =$ 10 nm (curves 1, 2) and $L_z = 4$ nm (curves 3, 4) at temperature $T =$ 300 K. Potential barrier is equal 0.22 eV.

dots, reflects the tetragonal symmetry of a parallelepiped shaped QD. The size-dependent component, D_{zz}, coincides with the effective DP constant [12] which leads to an intrasubband scattering rate of electrons in a quantum well with mechanisms (1) and (2) taken into account. Due to the difference in the parity of the electron states participating in intrasubband and interlevel transitions, the component D_{zz} differs by a sign from D_{cub} which describes interlevel scattering. This results in substantial suppression of electron relaxation in a QD having particular sizes, $L/L_z \simeq Q_0$.

The rate of emission of TA phonons, τ_t^{-1}, due to perturbation (2) is estimated as

$$\frac{\tau_l}{\tau_t} \sim \left(\frac{\chi E_1}{D}\right)^2 \left(\frac{s_t}{s_l}\right)^3 \left(\frac{L}{L_z}\right)^6. \tag{10}$$

It is seen that the emission of TA phonons can dominate only in very flattened QDs with a large transverse size L. TA phonons as well as LA phonons are emitted with wave vectors oriented predominantly along the z-axis. The overall phonon emission rate due to electron transitions from the first excited states to the ground state in parallelepiped — like shaped QDs as a function of the dot size L is shown in Fig. 2. In a QD with infinite potential barriers, the bottleneck effect [1] is demonstrated by curve 1 which exhibits a drastic reduction of the phonon emission rate in dots of small sizes. Curve 2 shows that this effect vanishes when the phonon induced variation of the electron effective mass is taken into account. The arrow marks a feature where the total rate appears to be smaller than the rate which corresponds to DP interaction only. This unusual behavior of electron scattering is caused by interference between the bulk DP mechanism and the interface mechanisms. The dependencies calculated for the QD with $In_{0.53}Ga_{0.47}$ As parameters are presented by curves 3 and 4. It is important that the total emission rate in this system, shown by curve 4, for dots with small sizes exceeds the conventional theoretical result [1] (curve 3) by an order of magnitude, providing for rapid electron energy relaxation. The dominant role of the additional mechanism (2) appears for dot sizes that satisfy the continuum approximation for phonons and the electron effective mass approximation. It is seen that the lateral size for which the contributions of the mechanism (2) and the DP coupling are equal remains practically unchanged when a barrier height decreases and this transition size is close to the corresponding size of cubic QDs.

The phonon emission rates in the parallelepiped-shaped QDs with different thicknesses, L_z, are plotted in Fig. 3. Curve 1 exhibits the bottleneck effect calculated in Ref. [1]; curve 2 demonstrates its modification caused by the deformation dependence of the electron effective mass. On comparing the pairs of curves -1, 2 and 3, 4 — one can see that the phonon emission rate increases substantially when the thickness L_z of QDs decreases. In summary, the dominant and noteworthy phenomenon reported in this paper is that mechanism (2) is more pronounced in narrow QDs.

Acknowledgements

This work was supported by US Army Research Office.

References

[1] U. Bockelmann, G. Bastard, Phys. Rev. B 42 (1990) 8947.
[2] X.Q. Li, H. Nakayama, Y. Arakawa, Phys. Rev. B 59 (1999) 5069.
[3] K. Brunner, U. Bockelmann, G. Abstreiter, M. Walther, G. Böhm, G. Tränkle, G. Weimann, Phys. Rev. Lett. 69 (1992) 3216.

[4] K. Mukai, N. Ohtsuka, H. Shoji, M. Sugawara, Phys. Rev. B 54 (1996) R5243.
[5] U. Bockelmann, W. Heller, A. Filoramo, Ph. Roussignol, Phys. Rev. B 55 (1997) 4456.
[6] R. Heitz, M. Veit, N.N. Ledentsov, A. Hoffman, D. Bimberg, V.M. Ustinov, P.S. Kop'ev, Zh.I. Alferov, Phys. Rev. B 56 (1997) 10435.
[7] R. Ferreira, G. Bastard, Appl. Phys. Lett. 74 (1999) 2818.
[8] X.Q. Li, Y. Arakawa, Phys. Rev. B 56 (1997) 10423.
[9] T. Inoshita, H. Sakaki, Phys. Rev. B 46 (1992) 7260.
[10] F.T. Vasko, V.V. Mitin, Phys. Rev. B 52 (1995) 1500.
[11] P.A. Knipp, T.L. Reinecke, Phys. Rev. B 52 (1994) 5923.
[12] V.I. Pipa, V.V. Mitin, M.A. Stroscio, Appl. Phys. Lett. 74 (1999) 1585.
[13] V.V. Mitin, V.I. Pipa, M.A. Stroscio, Microelectron. Engng. 47 (1999) 373.
[14] P.Y. Yu, M. Cardona, Fundamentals of Semiconductors, Springer, Berlin, 1999.
[15] S. Adachi, Physical Properties of III–V Semiconductor Compounds: InP, InAs, GaAs, GaP, and InGaAs, Wiley, New York, 1992.