

Formation of a surface quantum dot near laterally and vertically neighboring dots

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The elastic-energy-release rate (EERR) of a surface quantum dot (QD) near laterally and vertically neighboring QDs in a linear anisotropic elastic substrate is calculated. The EERR is used to describe quantitatively the driving force for growth of a newly formed (i.e., small) QD in the presence of a grown (i.e., large) QD and hence quantitatively the driving force for their ordering by assuming that the variation of the total change of free energy (including elastic energy, surface energy and edge energy) with their relative location depends only upon the elastic-energy part. An efficient boundary-element method is employed to solve the three-dimensional boundary-value problem of anisotropic elasticity, which requires discretization only along the surface of the QDs and their interface with the substrate. Numerical results for InAs QDs of cuboidal shape on a GaAs substrate with a free surface along the (001) plane are reported. It is found that the presence of a large surface QD inhibits the growth of a small surface QD. The small QD “prefers” to align with the large one in the (100) and (010) directions. However, this effect is relatively small, of about a 1.5% change of the EERR. In contrast, the effect of a buried large QD on the growth of a small surface QD can be significant, of up to a 25% change of the EERR. The favorable location of a small surface QD may be either vertically above or at an angle to the buried large QD, depending upon the depth of the latter. The driving force for the growth of a small surface QD at the favorable locations is enhanced by the presence of the buried large QD. In addition, the theory predicts an optimum depth of the buried large QD for the driving force for the growth of a small QD at the surface.

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I. INTRODUCTION

Currently there is a strong interest in semiconductor quantum dots (QDs) because of their potential applications in interesting devices, such as ultralow threshold laser, resonant tunneling devices, and huge-capacity memory media.¹ An efficient method of fabricating semiconductor QDs is based on the spontaneous formation of small dots on the surface of a heteroepitaxial thin film. In this process, a QD may nucleate and grow in the elastic field of neighboring QDs. The long-range elastic effect may lead to lateral and vertical ordering of QD arrays that is essential to many QD applications, such as those mentioned above. Recently, Yang² introduced the elastic-energy-release rate (EERR) to quantify the driving force in the formation of quantum islands (QIs) including QDs and quantum wires (QWs). The EERR is defined as the reduction of elastic strain energy per unit volume of QI growth. It was used to examine the vertical ordering of QWs in an isotropic elastic system under the assumption that the variation of the total change of free energy (including elastic energy, surface energy and edge energy) with their relative location depends only upon the elastic energy part. It is the objective of the present work to apply the EERR under the same assumption to explore quantitatively the effects of laterally and vertically neighboring grown (i.e., large) QDs on the driving force for a newly formed (i.e., small) surface QD in a three-dimensional (3D) anisotropic elastic system.

Experimentally, a new QD is often found to grow on a free surface vertically above an existing QD.³⁻⁷ In other cases, a vertically oblique array of QDs is found.^{5,8,9} In their discussion of the formation of arrays of QWs, Shchukin *et al.*¹⁰ have calculated the interaction energy between two sheets of identical QWs in a cubic anisotropic elastic solid. They assumed that the QWs have the same properties as the

substrate. They found that there exists a bifurcation of correlation and anticorrelation of QW sheets, depending upon the distance between the two sheets, in order to minimize the energy of the system. This bifurcation has, however, not been found in the isotropic elastic system with QWs of a different shape based on the same concept of energy minimization.² Ponchet *et al.*¹¹ developed a similar model to investigate the lateral interaction of identical QDs. However, since these studies^{10,11} did not set up a reference level of energy change, it is not clear how strongly these QIs interact, for instance, compared to the environmental noises in the practical sense. In parallel, some research groups have used the spatial variation of strain (or strain energy density) of a single seed QI (or a group of seed QIs) to predict the favorable location of a new QI.^{3,12-14} Zhang *et al.*¹⁵ further developed the local-field approach

of QI formation by coupling to it a kinetic law. Such approaches in terms of strain or strain energy density, which are conceptually different from the aforementioned approaches^{2,10,11} based on the change of system energy in the course of QD formation, may be applicable to a kinetics-controlled process. In the self-assembly of equilibrated QI arrays, an approach based on the minimization of system energy is applicable.^{1,2,10,11}

In Sec. II, the three-dimensional boundary-value problem of QDs in a substrate is formulated. The QDs and substrate have generally different materials properties. An efficient boundary-element (BE) method^{16,17} is used to solve the anisotropic elastic inhomogeneity problem. Because it uses the half-space Green's function for the substrate, coupled with the infinite-space Green's function for the QDs, the BE method requires only numerical discretization along the surface of the QDs and their interface with the substrate. This numerical procedure is more efficient than the conventional

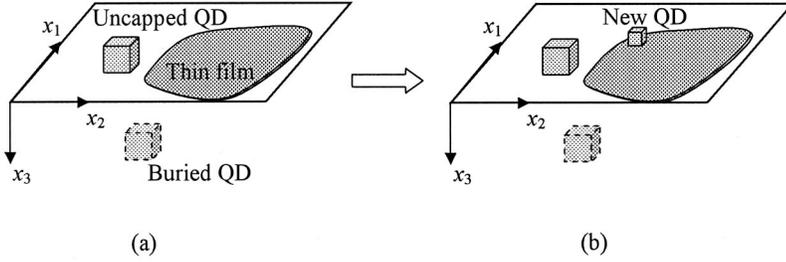


FIG. 1. (a) A half-space heterostructure with embedded and uncapped QDs and a thin wetting film. (b) Formation of a new QD by mass transport from the wetting film.

BE method using only the infinite-space Green's function and than both the domain-based numerical techniques of finite-element method¹⁸ and finite-difference method.¹⁹ In Sec. III, the EERR of a small InAs QD of cuboidal shape near a large InAs QD on a GaAs substrate is calculated. The variation of the EERR with their relative location is used to explore the driving force and favorable location of a newly formed QD in the presence of a laterally or a vertically neighboring grown QD. The present formulation describes quantitatively the driving force for formation of a QD and hence quantitatively the driving force for interaction and ordering of QDs. This is of practical significance in the strain engineering of QD nanostructures.²⁰ Conclusions are drawn in Sec. IV.

II. FORMULATION

The elastic-energy-release rate is defined as the reduction of elastic strain energy per unit volume of mass transport from a wetting layer to a QD of certain shape.² We analyze the critical EERR for nucleation of a self-similar QD of cuboidal shape near an existing grown QD. This physical process is schematically shown in Fig. 1. Figure 1(a) shows the initial state: a grown (seed) QD that is uncapped or buried, and a finite wetting layer on a substrate. The wetting layer is assumed to be very thin and to cover a very large area compared to the dimensions of the QD. The product of the thickness and the area is finite, yielding a finite volume. Thus, the elastic strain energy of the system can be obtained as a sum of those of the wetting layer and of the seed QD calculated separately, i.e., the interaction energy of the wetting layer and the seed QD is negligible. Figure 1(b) shows a target state of a small QD being nucleated by mass transport from the thin wetting layer. Similarly, because the wetting layer is very thin and covers a very large area compared to the dimensions of the QDs, the elastic strain energy of the system can be obtained as a sum of that of the wetting layer and of the seed QD plus the new QD calculated separately. From states (a) to (b), the mass is conserved. Therefore, the elastic strain energy of the two systems, W_{initial} and W_{target} , can be written, respectively, as

$$W_{\text{initial}} = W_{\text{seed}} + w_{\text{wet}} V_{\text{wet}} \quad \text{and} \quad W_{\text{target}} = W_{\text{seed+newdot}} + w_{\text{wet}}(V_{\text{wet}} - V_{\text{newdot}}), \quad (1)$$

where W_{seed} is the elastic strain energy of the system of only the seed QD and substrate, and w_{wet} and V_{wet} are, respectively, the elastic strain energy density and volume of the

wetting layer, $W_{\text{seed+newdot}}$ is the elastic strain energy of the system of the seed QD, new QD, and substrate, and V_{newdot} is the volume of the new QD.

By fixing the shape and volume of the seed QD, the EERR for formation of the new QD is derived by differentiating the second equation in Eq. (1):

$$\Gamma_{\text{newdot}} = - \frac{dW_{\text{target}}}{dV_{\text{newdot}}} = - \frac{dW_{\text{seed+newdot}}}{dV_{\text{newdot}}} + w_{\text{wet}}. \quad (2)$$

In later calculations, this quantity is evaluated approximately in a finite-difference scheme as

$$\Gamma_{\text{newdot}} = - \frac{W_{\text{seed+newdot}} - W_{\text{seed}}}{V_{\text{newdot}}} + w_{\text{wet}}. \quad (3)$$

The finite-difference formula is accurate for a new QD of small volume compared to the seed QD. It is valid also because W_{seed} is the limiting value of $W_{\text{seed+newdot}}$ when V_{newdot} approaches zero. Therefore, the evaluation of the EERR, Γ_{newdot} , is reduced to calculating the terms: W_{seed} , $W_{\text{seed+newdot}}$, and w_{wet} . Assuming that the higher the EERR, the more favorable is the condition for the growth of a new QD, the relation of a new QD to a seed QD can be determined.

To evaluate the term $W_{\text{seed+newdot}}$, we consider a heterostructure consisting of a semi-infinite substrate, a seed QD (uncapped or buried), and a small new surface QD, as shown in Fig. 1(b) (excluding the wetting layer). The materials properties of the QDs are the same, and are different from those of the substrate. The QDs and substrate are coherently bonded, and their lattice constants are different. This introduces a uniform eigenstrain field in each QD. The eigenstrain field induces a residual elastic field upon relaxation of the system, and hence causes the QDs to interact. A Cartesian frame of reference system (x_1, x_2, x_3) is established with the x_3 axis normal to the free surface and with the origin at the free surface. A numerical BE scheme is applied to solve the elastic problem with multiple bodies and eigenstrain field, leading to an efficient evaluation of the elastic strain energy of the system.

The boundary integral equation of displacement along boundaries of the substrate in equilibrium is given by

$$\frac{1}{2} \mathbf{u}_i^{(M)}(\mathbf{X}) = \int_{S^{(M)}} [u_{ij}^{*(M)}(\mathbf{X}, \mathbf{x}) p_j^{(M)}(\mathbf{x}) - p_{ij}^{*(M)}(\mathbf{X}, \mathbf{x}) u_j^{(M)}(\mathbf{x})] dS(\mathbf{x}), \quad (4)$$

where the superscript (M) indicates the association of a quantity to the substrate, S stands for surface, u_i and p_i ($\equiv \sigma_{ij}n_j$) are respectively the displacement and traction components, and $u_{ij}^{*(M)}$ and $p_{ij}^{*(M)}$ are respectively the half-space fundamental solutions of displacement and traction in the j th direction at a field point \mathbf{x} due to a unit point force acting in the i th direction at a source point \mathbf{X} . Repeated subscript implies summation over its range from 1 to 3. These fundamental solutions satisfy the traction-free boundary condition along the surface of the half space.²¹ In the above definition of traction p_i , σ_{ij} is the stress component, and n_k is the k th component of the unit outward normal at a smooth boundary point.

Similarly, the boundary integral equation of displacement along boundary of the n^{th} QD, D_n ($n=1,2$) with a uniform eigenstrain field in equilibrium is given by

$$\frac{1}{2}u_i^{(D_n)}(\mathbf{X}) = \int_{S^{(D_n)}} \{u_{ij}^{*(D_n)}(\mathbf{X}, \mathbf{x}) [p_j^{(D_n)}(\mathbf{x}) - F_j^{(D_n)}(\mathbf{x})] - p_{ij}^{*(D_n)}(\mathbf{X}, \mathbf{x}) u_j^{(D_n)}(\mathbf{x})\} dS(\mathbf{x}), \quad (5)$$

where the intrinsic traction $F_j^{(D_n)} \equiv C_{ijkl}^{(D_n)} \varepsilon_{lm}^{0(D_n)} n_k$,²² where C_{ijklm} is the elastic stiffness component, ε_{lm}^0 is the eigenstrain component, and $u_{ij}^{*(D_n)}$ and $p_{ij}^{*(D_n)}$ are, respectively, the infinite-space fundamental solutions of displacement and traction due to a point force,²¹ which are different from the above half-space ones, $u_{ij}^{*(M)}$ and $p_{ij}^{*(M)}$ for the substrate.

Along all the external boundaries, including the open surface of an uncapped QD and the top surface of substrate excluding the part covered by an uncapped QD, the traction-free condition, i.e., $p_i=0$, is imposed. Across all the interfaces between the substrate and QDs, the continuity conditions of displacement and traction, i.e., $u_i^{(M)}=u_i^{(D_n)}$ and $p_i^{(M)}=-p_i^{(D_n)}$, are imposed. Under these conditions, a unique solution to the elastic field in the system exists. This solution can be obtained by using a BE method based on Eqs. (4) and (5).^{16,17} Because Eq. (4) uses the half-space fundamental solution that satisfies the traction-free condition along the top surface, only the part of the top surface, where the traction-free condition is not satisfied due to the attachment of a surface QD, requires discretization. The iterative scheme of successive over-relaxation is used to solve the problem of multiple bodies.¹⁶

Once the solution on the boundaries is obtained, the elastic strain energy of the system with uniform eigenstrain field in each QD can be evaluated by²³

$$W_{\text{seed+newdot}} = \frac{1}{2} \int_{S^{(D)}} p_i u_i dS - \frac{1}{2} \int_{S^{(D)}} F_i u_i dS + \frac{1}{2} \int_{V^{(D)}} C_{ijkl} \varepsilon_{ij}^0 \varepsilon_{kl}^0 dV, \quad (6)$$

where $S^{(D)}$ and $V^{(D)}$ are, respectively, the surface and volume of the seed and new QDs. The first term represents the work done by the external traction, which is zero in the present case of zero traction along external boundaries and

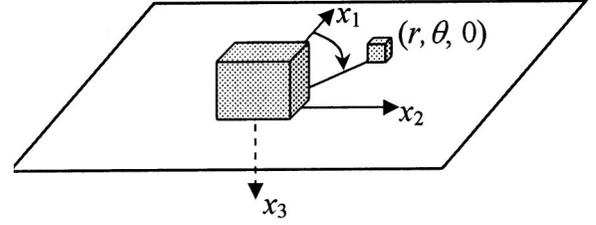


FIG. 2. Formation of a new QD in the presence of a laterally neighboring grown QD.

self-balanced traction along interfaces. The second term represents the work done by the intrinsic traction due to the uniform eigenstrain field in the QDs during the deformation of the body. The third term, which is a constant, corresponds to the ground state before relaxation of the eigenstrain field in the QDs.

The term W_{seed} can be evaluated as a special case in the same way as above. The wetting layer, which is assumed to consist of the same material as the QDs, has an eigenstrain field due to its lattice mismatch with the substrate. Its strain energy density w_{wet} corresponds to the state with its stress relaxed in the x_3 direction. This term, which is defined to be $1/2 \sigma_{ij}(\varepsilon_{ij} - \varepsilon_{ij}^0)$, can be obtained provided that the stress σ_{ij} and strain ε_{ij} are available. The stress and strain can be obtained by solving the twelve (independent) equations of $\sigma_{ij} = C_{ijkl}(\varepsilon_{kl} - \varepsilon_{kl}^0)$ (i.e., constitutive law) under six conditions of $\sigma_{i3}=0$ and $\varepsilon_{11}=\varepsilon_{22}=\varepsilon_{12}=0$.

III. FORMATION OF A QD NEAR NEIGHBORING DOTS

In this section, the previous formulation of the EERR for formation of a small QD in the presence of a seed QD is applied to examine InAs QDs on a GaAs substrate. Both materials are modeled as cubic anisotropic and linearly elastic. Their crystallographic directions are, respectively, parallel to each of the Cartesian axes. The free surface of the substrate is taken to be the (001) plane. The elastic constants for GaAs are $C_{11}=118$, $C_{12}=54$, $C_{44}=59$ (GPa), and, for InAs, $C_{11}=83$, $C_{12}=45$, and $C_{44}=40$ (GPa). The eigenstrain in the QDs is hydrostatic, i.e., $\varepsilon_{ij}^0 = \varepsilon^0 \delta_{ij}$. The shapes of the QDs are taken to be cuboidal with side dimensions: $a \times a \times 0.5a$ for seed dot (either uncapped or buried), and $0.2a \times 0.2a \times 0.1a$ for new surface dot. Since the elastic fields and total strain energy are, respectively, linearly and quadratically dependent upon ε^0 , these quantities are scaled correspondingly by the latter. All lengths are scaled by a .

A. Effect of a laterally neighboring dot

The case when the seed QD as well as the new QD is on the free surface is first considered, as shown in Fig. 2. The location of the QDs is defined by the coordinates of the center of their bottoms. Let us locate the seed QD at $(0, 0, 0)$ and the new QD at $(x_1, x_2, 0)$. Using the previous formulation, the EERR for self-similar formation of the new QD is calculated for various values of r and θ , where $x_1 = r \cos \theta$ and $x_2 = r \sin \theta$. The results along three radial lines, $\theta=0^\circ, 30^\circ$

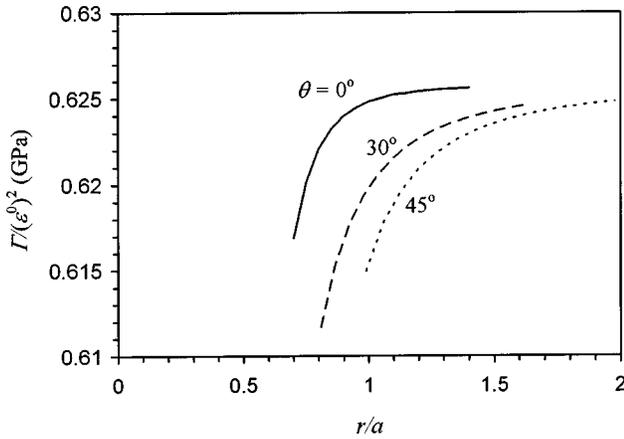


FIG. 3. Radial variation of the EERR for formation of a new QD at three angles $\theta=0^\circ$, 30° and 45° from the center of the large surface QD.

and 45° , and along a circular arc from $\theta=0^\circ$ to 90° at $r = a$, are plotted in Figs. 3 and 4. The angular variation of the EERR is symmetric relative to the x_1 and x_2 axes.

Figure 3 shows that the EERR for formation of the new QD decreases when the new QD gets closer to the seed QD. This shows that the presence of the surface seed QD inhibits the growth of a new QD nearby. This figure also shows that the EERR is the highest when the new QD is aligned in the (100) direction. This behavior is confirmed by Fig. 4, which shows that the angular variation of the EERR at $r=a$ has a minimum at $\theta=45^\circ$ and maxima at $\theta=0^\circ$ and 90° . Thus, an alignment of the new QD along the (100) or the (010) direction is favorable. However, the difference between the maximum and minimum values of the EERR is only approximately 1.5% of the average magnitude. Figure 3 also shows that the EERR decreases rapidly with r when the new QD gets very close to the seed QD. In this situation, the continuum model as such may not be valid, and a lattice-level model should be used.

B. Effect of a vertically neighboring dot

Now the case of a buried seed QD and a new surface QD is considered, as shown in Fig. 5. The location of the seed

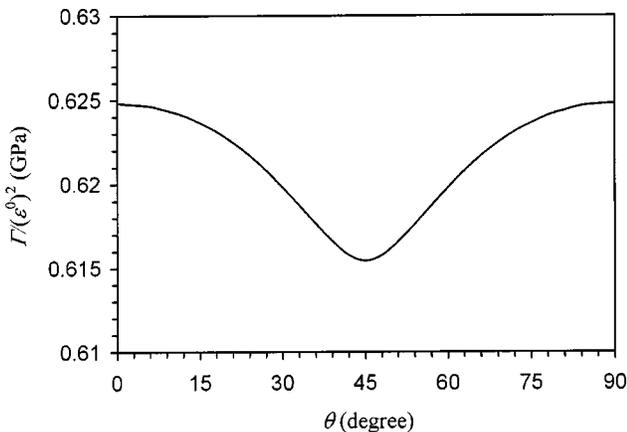


FIG. 4. Angular variation of the EERR for formation of a new QD between $\theta=0^\circ$ and 90° and at $r=a$ from the center of the large surface QD.

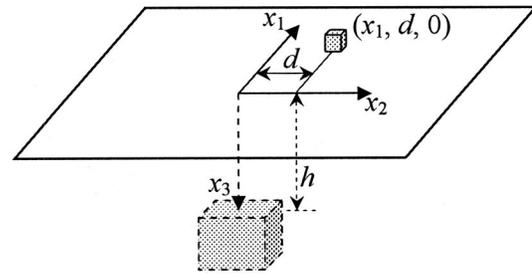


FIG. 5. Formation of a new QD in the presence of a vertically neighboring grown QD.

QD is defined by the coordinates at the center of its top surface, set at $(0, 0, h)$. The location of the new surface QD is defined in the same way as above by the coordinates of the center of its bottom surface, set at $(x_1, d, 0)$. The EERR for formation of the small surface QD is calculated as a function of the new-dot coordinate x_1 for $d=0, 0.3a$ and $0.6a$, and $h=0.1a, 0.3a$, and $0.6a$. The results are plotted in Figs. 6(a), 6(b), and 6(c).

The plots in Fig. 6 (except one curve with $h=0.1a$ and $d=0$ in Fig. 6(a), which will be discussed next) show that the EERR for the formation of the small surface QD is the highest when the new QD is vertically above the buried seed QD. When the new QD is moved aside and farther from the center of the buried seed QD, the EERR decreases, and then increases. It eventually approaches a constant value at a large distance. When this happens, the buried seed QD has no significant effect on the formation of the new QD.

When the depth of the buried QD is small, as in the case of $h=0.1a$ shown in Fig. 6(a), the behavior of the EERR as described above is different. In this case, the maximum value of the EERR does not occur when the new QD is right above the buried seed QD. Instead, the maximum value appears when the new QD is between the center and the edge of the buried seed QD. At the peak point, the EERR decreases rapidly with increasing distance between the two QDs. This implies that for small values of h , the favorable location for formation of the new QD is not vertically above the seed QD. Therefore, the calculation suggests that an oblique stacking of QD arrays can be energetically favorable if the separation depth, i.e., spacer thickness, is small. For large separation depths, the vertical stacking of QD arrays is favorable.

In addition, the variation of the EERR as a function of depth h for $x_1=d=0$ is examined. In this case, the new QD is vertically above the seed QD and the depth of the seed QD is varied. The result is shown in Fig. 7. It can be seen that the EERR has the maximum value at about $h=0.3a$. This implies that there is an optimum depth of the buried seed QD for the formation of a new QD at the free surface. Also, the magnitude of the EERR is always larger than that when the seed QD is remote to the new QD (equivalently, that in the absence of the seed QD). Thus, together with the results shown in Fig. 6, it is concluded that the presence of the buried seed QD enhances the growth of a new QD at the favorable locations on the free surface. The enhancement of the EERR can be up to 25% above the mean value.

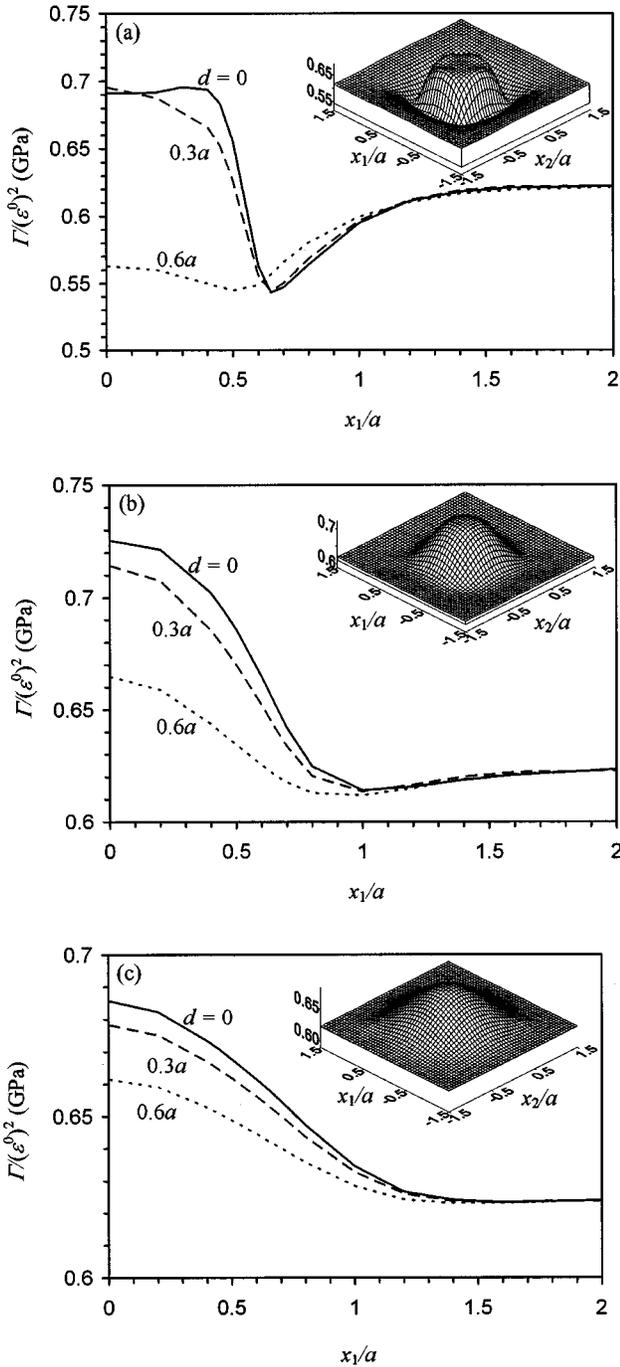


FIG. 6. Variation of the EERR for formation of a new QD along lines $(x_1, d, 0)$ for a fixed depth of the buried seed QD: (a) $h = 0.1a$, (b) $h = 0.3a$, and (c) $h = 0.6a$. The insets are the surface plots of the EERR over the area above the buried seed QD.

Finally, we remark that the presence of edges of the QDs contributes significantly to the spatial variation of the EERR for formation of a new QD. Therefore, the results presented above depend upon the shapes of the QDs which are assumed to be cuboidal. The characteristics of the EERR variation may be different for QDs of other shapes.

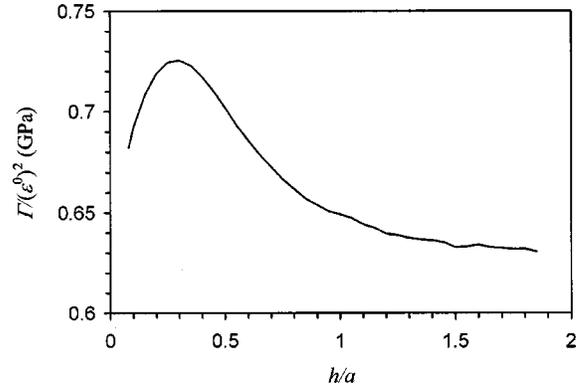


FIG. 7. Variation of the EERR for formation of a new QD right above the center of the buried seed QD with the depth of the buried seed QD.

IV. CONCLUSIONS

The EERR for the formation of a surface QD in the presence of laterally and vertically neighboring QDs in semiconductors has been examined. This problem of multiple bodies in anisotropic elasticity has been solved by applying an efficient BE method. Because it uses the half-space fundamental solution for the substrate, coupled with the infinite-space fundamental solution for the QDs, the BE method requires only numerical discretization along the surface of the QDs and their interface with the substrate. The formula for calculating the elastic strain energy of the QDs with uniform eigenstrain field requires only the displacement and traction along these boundary and interface, which are the direct outcome of the BE solution. Thus, the present BE method is computationally more efficient than the conventional BE method using only the infinite-space fundamental solution as well as the domain-based numerical techniques such as the finite-difference and the finite-element methods. Numerical results for the EERR of an InAs QD of cuboidal shape on a GaAs(001) substrate are reported.

It has been shown that the presence of a grown surface QD reduces the EERR for the growth of a small surface QD nearby. The small surface QD prefers to align along the (100) and (010) axes with the large one. However, this lateral effect is relatively small, of only about 1.5% change of the EERR. In contrast, the effect of a buried seed QD on the formation of a surface QD can be significant, of up to 25% change of the EERR. Depending on the depth of the buried seed QD the EERR or the formation of a small surface QD reaches the highest magnitude when the small QD is either vertically above or at an angle to the large grown QD. It implies that in the case of thick spacer, which covers the seed QD, a vertical array of QDs would exhibit an ordering of correlation. In the other case of thin spacer, an oblique stacking of QDs is energetically favorable. When the new QD is vertically above the seed QD, the EERR, plotted as a function of the depth of the seed QD, has the maximum value at about $h = 0.3a$. Thus, there is an optimum depth of the buried seed QD for the driving force of a new QD at the surface.

The above results have been derived for QDs of cuboidal shape. The EERR is sensitive to the shape of the QDs be-

cause of the effect of the edges. Therefore, the characteristics of the EERR as discussed above may not apply to QDs of other shapes. The EERR also depends on the volume of the QDs. The EERR calculated above is for the early stage of QD growth. Favorable locations for growth of a QD may change as the QD grows.

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