Spatial ordering of stacked quantum dots

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We investigate the growth conditions necessary to form an ordered quantum dot crystal by capping spatially ordered quantum dots and growing a new layer of dots on top of the capping layer. Performing Monte Carlo simulations and developing analytic arguments based on the stress energy function, we demonstrate the existence of an optimal capping layer thickness, external flux, and temperature for the formation of quantum dot crystals. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1347391]

Quantum dots (QDs) are attractive as potential candidates for many optoelectronic applications, including lasers or detectors.[1] While they have been fabricated using lithographic techniques,[2] small enough dots with high spatial density, necessary for device applications, have not yet been achieved with this technique. Consequently, much research has focused on an alternative method, based on self-assembly,[3] that generates dots through the unique combination of strain and growth kinetics, offering relatively uniform islands. To obtain high spatial density of QDs, the islands have been grown in layers, generating a three dimensional superlattice of capped islands[4] and leading to an order of magnitude increase in the QD density. In such multilayer structures the buried dots influence the nucleation of the islands in the subsequent layers, making the dots line up vertically. A number of experiments have demonstrated that the alignment quality depends strongly on the number of QD layers,[4–7] as well as the capping layer thickness,[8] with thinner layers yielding better vertical correlation.[9] Depending on the nature of the strain anisotropy in the system, the vertical alignment of the dots in subsequent layers can accompany lateral ordering as well,[4] followed by the formation of a face-centered cubic-like ABCABC vertical dot stacking sequence.[10]

Fueled by experimental interest, a number of theoretical approaches have been developed to address the alignment of the QDs. A conceptually simple, but rather successful approach assumes that the buried QDs are pointlike embedded islands, allowing the determination of the stress field on the surface of the capping layer. As Tersoff *et al.* have demonstrated, the superposition of the stress field generated by the buried islands leads to minima in the strain surface energy, that act as preferred nucleation sites for the next layer of islands.[4] With the deposition of multiple layers, the QDs gradually undergo self-organized spatial ordering, forming vertically aligned columns. While its influence in uncovering the mechanism of stacked QD formation is unquestionable, Ref. 4 has not addressed the optimal conditions under which the islands are best ordered. Furthermore, while leading to good qualitative predictions, it is always questionable if the stress field from a pointlike source reproduces correctly the stress field of finite islands. However, recent molecular dynamics simulations[11] have concluded that the numerical results on the surface stress in the Ge/Si system can be fitted by the analytic solution of a pointlike source if the parameters in the formula are chosen appropriately.[12] In this letter we use this result to determine the stress field on the surface, allowing us to investigate the growth conditions necessary to form a QD superlattice ordered in all three spatial directions, leading to a highly regular QD crystal. In order to obtain a highly regular QD superlattice, we assume that QDs on the first layer have been ordered using either stamping[13] or lithographic techniques. After growing a new layer of islands on the top of this structure, we investigate the quality of spatial ordering of the islands in this new layer. We demonstrate the existence of optimal conditions for stacked QD formation in terms of the capping layer thickness, external flux rate, and substrate temperature. Growing the islands using these optimal parameters, we find that the obtained QD crystal has potentially superior electronic properties due to decreased randomness in both island size and position.

Our investigation is based on the analytic formula of the stress energy tensor σ_{ij} generated by an embedded pointlike island in a semi-infinite system. In the Ge/Si system, it was found that the off-diagonal components of the stress energy tensor (σ_{ij}, $i \neq j$) and the vertical component σ_{zz} were negligible, thus the stress distribution is mainly due to $\sigma = \sigma_{xx} + \sigma_{yy}$.[11] When a single QD is located at the origin of the coordinate system, the stress energy at a position $\mathbf{r} = (x,y,z)$ is given by

$$\sigma(\mathbf{r}) = \frac{P}{R^3}\left\{1 - \frac{3}{R^2}\left[\frac{4-4v}{4-8v}(x^2+y^2) - \frac{8vz^2}{4-8v}\right]\right\}, \qquad (1)$$

where $P \approx 110.6$ eV is the strength of the dipole, $R = (x^2 + y^2 + z^2)^{1/2}$, and $v = 0.218$ is the Poisson ratio for the Si embedding matrix. The stress energy displays a deep potential well at the center ($x = y = 0$) and a circular barrier around

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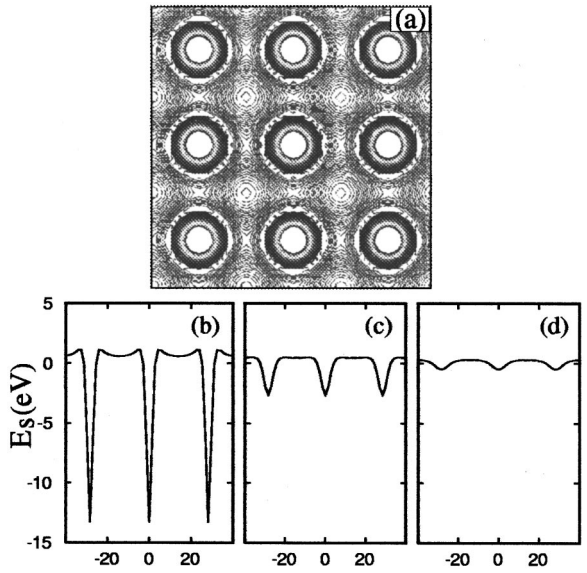

FIG. 1. (a) Contour plot of the stress energy E_s generated by nine embedded quantum dots underneath at $z=0$. (b)–(d) Profile of E_s as a function of the distance along the diagonal direction at (b) $z=3$, (c) $z=5$, and (d) $z=8$.

it, and decays as R^{-3}, a behavior similar to the electric field of a dipole. In order to investigate the formation of stacked QDs, we assume that a mesh of $N\times N$ pointlike QDs are embedded in the $z=0$ plane with a separation a between nearest neighboring QDs. Each QD generates the stress energy $\sigma(\mathbf{r})$, given by Eq. (1), thus an adatom on the top of the capping layer of thickness z is affected by the total stress energy E_s, the sum of all the contributions from each embedded QD. As shown in Fig. 1(a), the stress energy E_s at height z has a structure dominated by potential wells, located at $\{P_1\}=(ma,na)$ $(m,n=1,2\ldots N)$ on top of the embedded QDs, each potential well being surrounded by a circular barrier. To simplify our discussion, we will refer to the region with $E_s<0$ as core region [Fig. 1(b)], covering most of the area inside the barrier. In addition, there are shallow potential wells centered between four neighboring QDs, denoted by $\{P_2\}=(ma+1/2,na+1/2)$ $(m,n=0,1,2,\ldots N-1)$. These wells are generated by the superposition of the stress energy function contributed by each buried island. The depths of the potential wells at $\{P_1\}$ and $\{P_2\}$ and the height of the barrier decrease with increasing capping layer thickness z [Figs. 1(b)–1(d)]. Furthermore, with increasing z the potential well becomes broader by increasing the radius of the barrier.

The adatom dynamics on the top of the capping layer is determined by the diffusion constant D which is a function of the stress energy E_s and temperature T. Since E_s depends on the capping layer thickness z the diffusion constant also has a z dependence. On the other hand, since E_s depends on x and y as well, the coordinates along the surface, the diffusivity depends on the position of the adatom: when the stress energy is small (large), the chemical potential of the adatom is low (enhanced). In particular, when $E_s<0$ and its magnitude is large (e.g., in the core region) the chemical potential is extremely diminished, while the chemical potential of the adatom near the barrier, where $E_s>0$, is enhanced.

When the potential well at $\{P_1\}$ is deep enough, the nucleation could take place in the vicinity of $\{P_1\}$ inside the barrier, but not at $\{P_1\}$, because as the adatoms approach $\{P_1\}$, they drastically slow down. On the other hand, the atoms outside of the core region have higher chemical potential, thus they meet and form stable islands around $\{P_1\}$. This situation can be avoided by increasing the chemical potential in the core region, accomplished either by increasing the capping layer thickness or the substrate temperature. When the chemical potential is enhanced, the atoms will reach $\{P_1\}$, where the stress energy has its minimum. Thus, the first condition for spatial ordering is that the chemical potential of the adatoms needs to be high enough to reach the minimum $\{P_1\}$.

On the other hand, the adatoms diffusing on the substrate can meet and form islands at random positions. The mean separation l between these islands depends on the flux F and the diffusion constant D.[14] Since the diffusion driven nucleation is random, the resulting islands are not spatially ordered. For example, when an array of pointlike impurities are present, the islands are nucleated on the impurities only when $l>a$.[15] Accordingly, the second condition for spatial ordering in stacked QD formation is that $l>a$. This and the condition discussed earlier compete with each other such that with increasing z the chemical potential is enhanced but the mean free distance l decreases. This competition leads to an optimal thickness z, where the vertical correlation between the embedded and the self-organized islands has a maximum.

To investigate the appearance of this optimal thickness, we performed Monte Carlo simulations using the lookup table algorithms of the bond-counting method.[16] First, we choose a $N\times N=6\times 6$ array of QDs buried in the $z=0$ plane with lattice constant $a=20$, and the adatoms diffuse along the two dimensional plane at the top of the capping layer of thickness z. The length scales, a and z, are considered to be integer multiples of the Si layer thickness 3 Å. Atoms are deposited with flux F, and diffuse with the hopping rate $r=(2kT/\hbar)\exp(-E_{\mathrm{tot}}/kT)$, where kT is thermal energy and $\hbar$ is the Planck constant. The energy E_{tot} is given by $E_{\mathrm{tot}}=E_0+nE_N-E_s$, where E_0 and E_N denote the binding energies between Ge/Si and Ge/Ge atoms, respectively, n is the number of nearest neighboring atoms, and E_s is the stress energy. The effect of the stress on the activation energy has been considered to be linear, following Ref. 17. While the numerical values of E_0 and E_N depend on the orientation of the surface, we used $E_0=1.7$ eV and $E_N=0.9$ eV.[18] We limit the adatom diffusion to the capping layers, excluding the formation of three dimensional islands. We measured the probability $P(z)$ that an island is nucleated on the top of the embedded QDs in function of the layer thickness z, flux (F), and temperature (T). As shown in Figs. 2 and 3, for all flux and temperature values $P(z)$ has a maximum, indicating the existence of an optimal capping layer thickness. Furthermore, we find that $P(z)$ reaches 1 when the temperature is high enough (such as $T=700$ K, see Fig. 2), and the flux is low enough (such as $F=0.05$ ML/s, see Fig. 3). $P(z)=1$ corresponds to perfect nucleation, when all buried islands nucleate new islands on the top.

The emergence of the peak in $P(z)$ is the result of the two competing effects discussed above: for small z, the nucleation occurs around rather than at $\{P_1\}$ due to the diminished chemical potential in the core region. Also, if z is

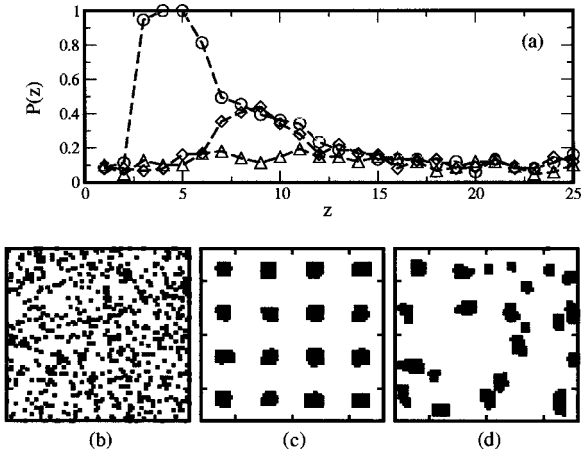

FIG. 2. (a) The probability $P(z)$ vs the height z for a fixed flux rate $F=0.05$ ML/s, but various temperatures, $T=700$ K (○), $T=680$ K (◇), and $T=660$ K (△). (b)–(d) Surface morphologies at the coverage 0.01 ML for the fixed flux rate $F=0.05$ ML/s and the temperature $T=700$ K, but various height at (b) $z=1$, (c) $z=5$, and (d) $z=13$.

very small, the strain energy on the surface is too high and islands do not form. For larger z, the mean free distance becomes shorter than the lattice constant a of the QD array due to the reduced diffusion constant. Thus, for small z, island nucleation is either prohibited or they form in the vicinity but not at $\{P_1\}$ [see Fig. 2(b)], while they are formed randomly for large z [see Fig. 2(d)]. Those effects reduce the probability of nucleation $P(z)$, but allow it to take up a maximum for intermediate z. The peak of $P(z)$ can reach 1 for $l>a$, when the chemical potential is moderated in the core region. For lower temperature such as $T=680$ and 660 K for the flux rate $F=0.05$ ML/s, however, the peak of the probability $P(z)$ is smaller than 1 [see Fig. 2(a)], because $l<a$ at the optimal layer thickness, even though the chemical potential in the core region is moderated. Similar behavior can be observed for high fluxes (e.g., $F=0.1$ and 0.2 ML/s), and fixed temperature ($T=700$ K) (Fig. 3), where the mean free distance l decreases below a, decreasing $P(z)$ below 1. Meanwhile, one may notice in Figs. 2(d) and 3(d) that the mean island size is larger for higher z, confirming the experimental observations.[9] So far, we have considered the case where the lattice constant of the QD array is fixed at $a=20$, but similar behavior can be obtained for other values of a. Since the peak can reach 1 when $l>a$ at the optimal height, the probability $P(z)$ could reach 1 for lower temperature T and smaller flux if the QD lattice constant a is smaller.

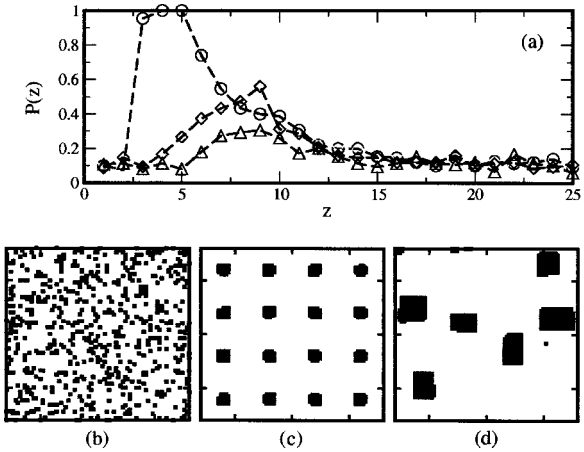

FIG. 3. (a) The probability $P(z)$ vs the height z for a fixed temperature $T=700$ K, but various flux rates, $F=0.05$ ML/s (○), $F=0.1$ ML/s (◇), and $F=0.2$ ML/s (△). (b)–(d) Surface morphologies at the coverage, 0.01 ML for the fixed temperature $T=700$ K and the flux rates $F=0.05$ ML/s, and various height at (b) $z=1$, (c) $z=5$, and (d) $z=13$.

In conclusion, we have investigated the optimal condition necessary to form spatially ordered stacked QDs in terms of the capping layer thickness, flux rate F, and temperature T. We find that there exists an optimal thickness of the capping layer, at which the probability $P(z)$ that islands nucleate on the top of embedded QDs reaches 1. This optimal thickness arises due to two competing effects: For a thin capping layer the chemical potential in the core region is diminished due to the strong stress energy, which is cured by increasing the capping layer thickness, while for a thicker capping layer the mean free distance of the adatoms diffusing without meeting other atoms decreases and the islands are nucleated randomly. Performing Monte Carlo simulations with varying the capping layer thickness, flux rate, and temperature, we confirmed numerically the existence of the optimal capping layer thickness, which can lead to the growth of uniform and spatially ordered QDs with potentially superior optical properties.

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[1] For a review, see for example, L. Jacak, P. Hawrylak, and A. Wojs, *Quantum Dots* (Springer, Berlin, 1998).
[2] M. A. Reed, R. T. Bate, K. Bradshaw, W. M. Duncan, W. M. Frensley, J. W. Lee, and H. D. Smith, J. Vac. Sci. Technol. B **4**, 358 (1986).
[3] For a review, see V. Shchukin and D. Bimberg, Rev. Mod. Phys. **71**, 1125 (1999).
[4] J. Tersoff, C. Teichert, and M. G. Lagally, Phys. Rev. Lett. **76**, 1675 (1996).
[5] D. Leonard, M. Krishnamurthy, C. M. Reaves, S. P. Denbaars, and P. M. Petroff, Appl. Phys. Lett. **63**, 3203 (1993).
[6] Q. Xie, A. Madhukar, P. Chen, and N. P. Kobayashi, Phys. Rev. Lett. **75**, 2542 (1995).
[7] G. S. Solomon, J. A. Trezza, A. F. Marshall, and J. S. Harris, Jr., Phys. Rev. Lett. **76**, 952 (1996).
[8] Other stacked quantum dot systems include Ge/Si, A. A. Darhuber, J. Stangl, G. Bauer, P. Schittenhelm, and G. Abstreiter, Thin Solid Films **294**, 296 (1997); GaN/AIN, F. Widmann, B. Daudin, G. Feuillet, Y. Samson, J. L. Rouvière, and N. Pelekanos, J. Appl. Phys. **83**, 7618 (1998); and InP/GaInP, M. K. Zundel, P. Specht, K. Eberl, N. Y. Jin-Phillipp, and F. Phillipp, Appl. Phys. Lett. **71**, 2972 (1997).
[9] Y. Nakata, Y. Sugiyama, T. Futatsugi, and N. Yokoyama, J. Cryst. Growth **175**, 713 (1997); G. S. Solomon, S. Komarov, J. S. Harris, and Y. Yamamoto, *ibid.* **175**, 707 (1997).
[10] G. Springholz, V. Holly, M. Pinczolits, and G. Bauer, Science **282**, 734 (1998); V. Holly, G. Springholz, M. Pinczolits, and G. Bauer, Phys. Rev. Lett. **83**, 356 (1999).
[11] I. Daruka, A.-L. Barabási, S. J. Zhou, T. C. Germann, P. S. Lomdahl, and A. R. Bishop, Phys. Rev. B **60**, R2150 (1999).
[12] S. M. Hu, J. Appl. Phys. **66**, 2741 (1989).
[13] T. I. Kamins and R. S. Williams, Appl. Phys. Lett. **71**, 120 (1997).
[14] D. E. Wolf, in *Dynamics of Fluctuating Interfaces and Related Phenomena*, edited by D. Kim, H. Park, and B. Kahng (World Scientific, Singapore, 1997).
[15] C. Lee and A.-L. Barabási, Appl. Phys. Lett. **73**, 2651 (1998).
[16] M. E. J. Newman and G. T. Barkema, *Monte Carlo Methods in Statistical Physics* (Clarendon, Oxford, 1999).
[17] M. Schroeder and D. E. Wolf, Surf. Sci. **375**, 129 (1997).
[18] A. Kobayashi and S. Das Sarma, Phys. Rev. B **37**, 1039 (1988); A. Kobayashi, S. M. Paik, and S. Das Sarma, J. Vac. Sci. Technol. B **6**, 1145 (1988); R. D. Meade and D. Vanderbilt, Phys. Rev. B **40**, 3905 (1989).