

# Atomistic approach to the strain field in finite-sized heterostructures

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## Supplementary Material

### A. Elastic energy and its gradient

For the valence-force-field (VFF) atomistic approach we consider the Keating interaction [1-3]. The Keating potential expression for zinc-blende structure [1] written in programmable way is

$$\begin{aligned}
 U &= \sum_{\substack{m\_n \\ m \neq n}} \sum_{i(m)=1}^{N(m)} \sum_{j(n)=1}^4 \gamma_{i(m)-j(n)} \left( \mathbf{r}_{i(m)-j(n)} \cdot \mathbf{r}_{i(m)-j(n)} - d_{i(m)-j(n)}^2 \right) \\
 &+ \sum_{\substack{m\_n\_p \\ m \neq n, m \neq p}}^{PA} \sum_{i(m)=1}^{N(m)} \sum_{\substack{j(n), k(p)=1 \\ j(n) \neq k(p)}}^4 \mu_{i(m)-j(n)-k(p)} \left( \mathbf{r}_{i(m)-j(n)} \cdot \mathbf{r}_{i(m)-k(p)} - d_{i(m)-j(n)} d_{i(m)-k(p)} \cos \theta_{i(m)-j(n)-k(p)} \right) \\
 &= U_1 + U_2
 \end{aligned} \tag{S1}$$

with

$$\begin{aligned}
 \gamma_{i(m)-j(n)} &= \frac{3\alpha_{i(m)-j(n)}}{8d_{i(m)-j(n)}^2} \left( \mathbf{r}_{i(m)-j(n)} \cdot \mathbf{r}_{i(m)-j(n)} - d_{i(m)-j(n)}^2 \right), \\
 \mu_{i(m)-j(n)-k(p)} &= \frac{3\beta_{i(m)-j(n)-k(p)}}{8d_{i(m)-j(n)} d_{i(m)-k(p)}} \left( \mathbf{r}_{i(m)-j(n)} \cdot \mathbf{r}_{i(m)-k(p)} - d_{i(m)-j(n)} d_{i(m)-k(p)} \cos \theta_{i(m)-j(n)-k(p)} \right).
 \end{aligned}$$

In eq. (S1)  $m, n, p$  denote types of atom,  $i(m)$  is the label for the  $i$ th atom of type  $m$ ,  $N(m)$  is the total number of atoms of  $m$  type,  $m\_n$  denotes the pair of atoms of type  $m$  and  $n$  ( $m \neq n$ ),  $m\_n\_p$  denotes the group of three tetrahedral atoms (belonging to a primitive cell) of type  $m, n$ , and  $p$ , with  $m$  the tetrahedron central atom (we name such a central atom as a principal atom (PA)), and  $n$  and  $p$  ( $n \neq m, p \neq m$ ) as neighbor atoms (NAs),  $\alpha_{i(m)-j(n)}$  and  $\beta_{i(m)-j(n)-k(p)}$  are the bond-stretching and bond-bending parameters, respectively,  $\mathbf{r}_{i(m)}$  is the position vector of the  $i(m)$  atom,  $\mathbf{r}_{i(m)-j(n)} = \mathbf{r}_{j(n)} - \mathbf{r}_{i(m)}$  is the nearest-neighbor position vectors of the atoms  $j(n)$  relative to the atom  $i(m)$ ,  $d_{i(m)-j(n)}$ ,  $d_{i(m)-k(p)}$  are the ideal bond lengths, and  $\theta_{i(m)-j(n)-k(p)}$  is the tetrahedral primitive cell bond angle ( $109.5^\circ$ );  $U_1$  denotes the first and  $U_2$  the second sum term in eq. (S1). For a given atom of type  $m$ , the sum in eq. (S1) run over the 4 nearest-NAs.

The L-BFGS minimization procedure involves the calculus of the elastic energy gradient. For this we consider the derivatives

$$\frac{\partial \left( \mathbf{r}_{i(m)-j(n)} \cdot \mathbf{r}_{i(m)-j(n)} \right)}{\partial r_{l(q)}^c} = 2 \left( r_{j(n)}^c - r_{i(m)}^c \right) \left( \delta_{jl} \delta_{nq} - \delta_{il} \delta_{mq} \right), \tag{S2a}$$

$$\frac{\partial \left( \mathbf{r}_{i(m)-j(n)} \cdot \mathbf{r}_{i(m)-k(p)} \right)}{\partial r_{l(q)}^c} = 2 \left( r_{j(n)}^c - r_{i(m)}^c \right) \left( \delta_{j(n)l(q)} - \delta_{i(m)l(q)} \right), \tag{S2b}$$

$$\frac{\partial \left( \mathbf{r}_{ij} \cdot \mathbf{r}_{ij} \right)}{\partial r_l^c} = 2 \left( r_j^c - r_i^c \right) \left( \delta_{jl} - \delta_{il} \right), \tag{S2c}$$

$$\frac{\partial \left( \mathbf{r}_{i(m)-j(n)} \cdot \mathbf{r}_{i(m)-k(p)} \right)}{\partial r_{l(q)}^c} = \left( r_{j(n)}^c - r_{i(m)}^c \right) \left( \delta_{kl} \delta_{pq} - \delta_{il} \delta_{mq} \right) + \left( r_{k(p)}^c - r_{i(m)}^c \right) \left( \delta_{jl} \delta_{nq} - \delta_{il} \delta_{mq} \right), \tag{S2d}$$

$$\frac{\partial(\mathbf{r}_{i(m)-j(n)} \cdot \mathbf{r}_{i(m)-k(p)})}{\partial r_{l(q)}^c} = (r_{j(n)}^c - r_{i(m)}^c)(\delta_{k(p)l(q)} - \delta_{i(m)l(q)}) + (r_{k(p)}^c - r_{i(m)}^c)(\delta_{j(n)l(q)} - \delta_{i(m)l(q)}), \quad (\text{S2e})$$

$$\frac{\partial(\mathbf{r}_{ij} \cdot \mathbf{r}_{ik})}{\partial r_l^c} = (r_j^c - r_i^c)(\delta_{kl} - \delta_{il}) + (r_k^c - r_i^c)(\delta_{jl} - \delta_{il}), \quad (\text{S2d})$$

where the superscript  $c$  denotes one of the three Cartesian components of position vector  $\mathbf{r}$ ,  $l(q)$  denotes the  $l$ th atom of type  $q$ , and  $\delta$  denotes the Kronecker delta symbol. For the gradient, the derivatives of  $U_1$  and  $U_2$  are as follows

$$\frac{\partial U_1}{\partial r_{l(q)}^c} = 4 \sum_{\substack{m=1 \\ n=2,3}}^{N(1)} \sum_{i(1)=1}^{N(1)} \gamma_{i(1)-l(q)} (r_{l(q)}^c - r_{i(1)}^c) - 4 \sum_{\substack{j(n)=1 \\ n \neq q}}^4 \gamma_{l(q)-j(n)} (r_{j(n)}^c - r_{l(q)}^c), \quad (\text{S3a})$$

$$\frac{\partial U_2}{\partial r_{l(q)}^c} = 4 \sum_{\substack{m=n \\ m \neq n}}^{N(m)} \sum_{i(m)=1}^{N(m)} \sum_{\substack{j(n)=1 \\ j(n) \neq l(q) \\ m \neq n, m \neq q}}^4 \mu_{i(m)-j(n)-l(q)} (r_{j(n)}^c - r_{i(m)}^c) - 4 \sum_{\substack{j(n), k(p)=1 \\ j(n) \neq k(p) \\ q \neq n, q \neq p}}^4 \mu_{l(q)-j(n)-k(p)} (r_{j(n)}^c - r_{l(q)}^c), \quad (\text{S3b})$$

and the derivative of  $U$  relative to  $r_{l(q)}^c$  is

$$\frac{\partial U}{\partial r_{l(q)}^c} = \frac{\partial U_1}{\partial r_{l(q)}^c} + \frac{\partial U_2}{\partial r_{l(q)}^c}. \quad (\text{S3c})$$

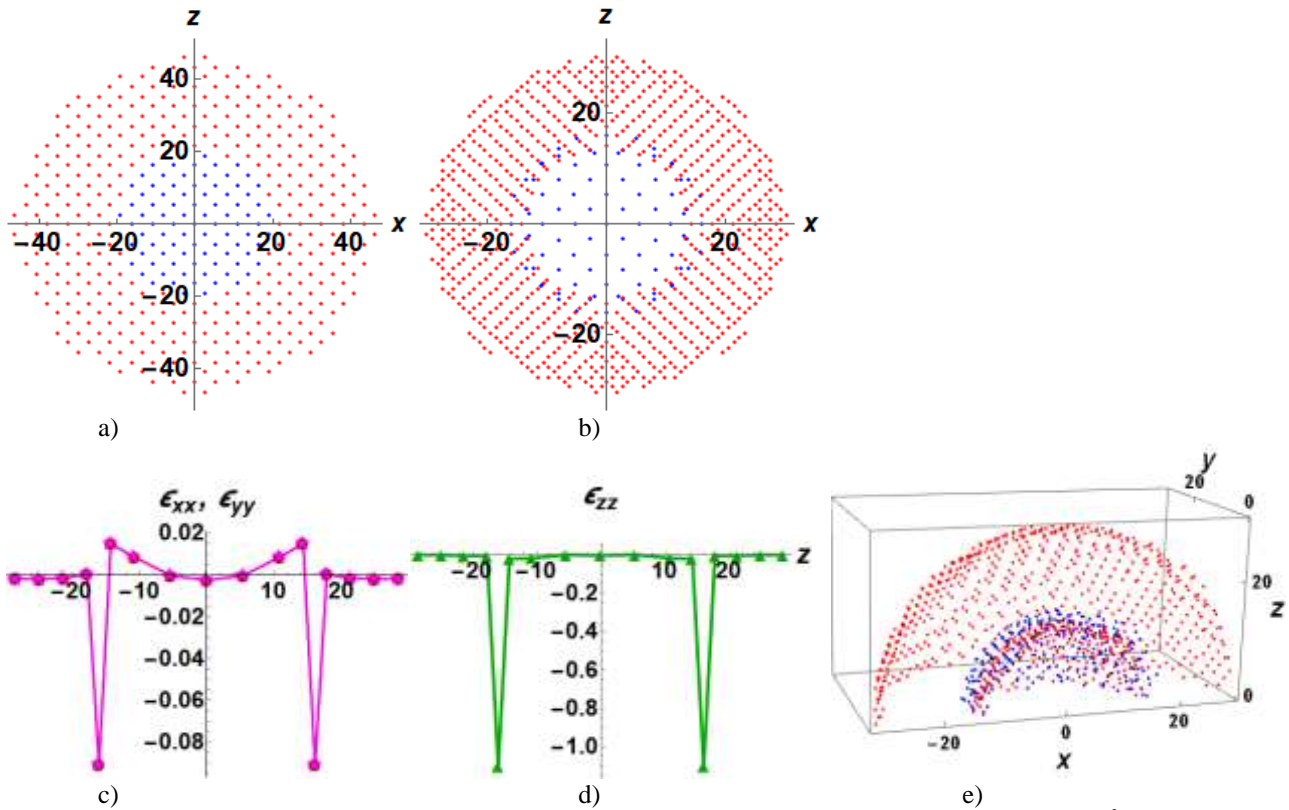
The PAs can have from zero to 4 bonds. In eq. (S3a) in the first (second) sum the atom  $i(m)$  ( $l(q)$ ) is a PA and the atom  $l(q)$  ( $j(n)$ ) is a NA. In eq. (S3b) in the first (second) sum the atom  $i(m)$  ( $l(q)$ ) is a PA and the atoms  $j(n)$ ,  $l(q)$  ( $j(n)$ ,  $k(p)$ ,) are NAs. The sums over the NAs in eqs. (S3a, b) also run (as in eq. (S1)) over the 4 nearest-NAs.

The code can successively re-start by setting more than one value for the radiuscut; in this case the final configuration computed for a value of radiuscut becomes the initial configuration of the next minimization process initiated for another value of radiuscut (see the flowchart in the User Manual). The default dimension of vector radiuscut is set as strtmax=5 in the code; the setting strtmax=1 initiates a one step minimization (relaxed configuration corresponding to a certain value of radiuscut is obtained).

## B. Simulation for heterostructures with large lattice mismatch

The AS\_LBFG\_v2 code can confidently simulate the strain field in heterostructures where the constituents have a close value of the lattice constant. In this case, the use of Keating interaction potential, which assumes elastic atomic interaction, is justified. In the case of nonlinear elastic effects due to large lattice misfit between the heterostructure constituents (see, e.g. ref. [4]), the simulations should cautiously be interpreted. For example, in Fig. S1, the strain field and misfit dislocations are simulated for core-shell Si/C, which is a heterostructure with large lattice mismatch

of the two materials,  $a_{\text{Si}}=5.431020511 \text{ \AA}$ ,  $a_{\text{C}}=3.567 \text{ \AA}$  [5]. In Fig. S1d) the strain tensor element  $\epsilon_{zz}$  in  $z$  direction, for a (001) orientation of crystal layers, obtained by the current code (which uses the Keating interaction potential) has a negative peak of absolute magnitude greater than unity at the interface. Comparison of the simulated strain field distribution with that obtained using more advanced interaction potentials, for example the Stillinger-Weber interaction for Si-C (see e.g. ref. [6]-[8]), could answer whether the Keating interaction is a sufficiently reliable approach for this type of heterostructure. Such advanced potentials can easily be implemented in the code as the gradient at each atom location is already coded in the current source code.



**Figure S1.** Simulation results of the strain field for Si/C core-shell quantum dot of Si core radius 20 Å and the setting  $n_x=n_y=n_zmin=n_zmax=13$ ,  $misfit=0.032$ . a) thin layer of thickness  $a_{Si}/2$  of the initial configuration, centered on the middle of structure and parallel to the  $xz$  plane, projected on the  $xz$  plane; b) relaxed configuration corresponding to a); c) and d)  $\epsilon_{xx}$ ,  $\epsilon_{yy}$ , and  $\epsilon_{zz}$ , respectively, strain field along  $z$  symmetry axis; e) misfit dislocations. Color in a), b), e): blue for Si atoms; red for C atoms. The unit length in the figures is 1 Å.

## References

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