Equilibrium Analysis of Lattice-Mismatched Nanowire Heterostructures

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ABSTRACT

The quality of lattice-mismatched semiconductor heterojunctions is often limited by the presence of misfit dislocations. Nanowire geometries offer the promise of creating highly mismatched, yet dislocation free heterojunctions. A simple model, based upon the critical thickness model of Matthews and Blakeslee for misfit dislocation formation in planar heterostructures, illustrates that there exists a critical nanowire radius for which a coherent heterostructured nanowire system is unstable with respect to the formation of misfit dislocations. The model indicates that within the nanowire geometry, it should be possible to create perfect heterojunctions with large lattice-mismatch.

INTRODUCTION

The recent demonstration of coherent, lattice-mismatched nanowire semiconductor heterostructures [1,2] by the Vapor-Liquid-Solid (VLS) synthesis mechanism [3,4] represent the first foray into a new realm of heterojunction functionality and performance. These longitudinally heterostructured nanowires enable band gap engineering in one-dimensional structures, with potential applications towards unique quantum dot geometries, p-n and bipolar junctions, and nanowire superlattices (for example, see [5-8]). The design of such devices will be accelerated substantially if design engineers can use a simple model to predict circumstances under which misfit dislocations will form at the interface.

One-dimensional heterostructures differ from their planar counterparts via their elastic boundary conditions: while a thin film is constrained laterally during growth, a nanowire can relieve strain energy via elastic relaxation. Matthews and Blakeslee [9,10] investigated the lattice-mismatch phenomenon in heteroepitaxial systems, and developed an equilibrium critical thickness model for planar systems based on strain energy and misfit dislocation energy considerations. The goal of this study is to develop a simple model by extending the Matthews-Blakeslee model to one-dimensional systems.

A schematic illustration of the heterostructured nanowire system is shown in Fig. 1. The system is oriented along the z-axis and comprises an infinitely thick nanowire overlayer and nanowire substrate.

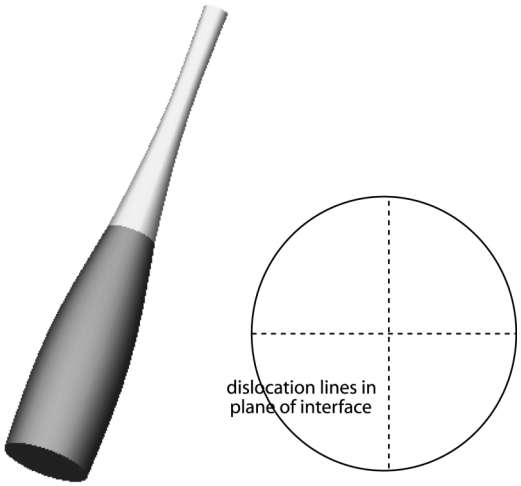

Figure 1. Schematic illustration of lattice-mismatched heterostructured nanowire system comprising an infinitely thick nanowire substrate and an infinitely thick nanowire overlayer. The misfit dislocation configuration in the plane of intersection is denoted as well by the dislocation lines in at the heterostructure interface.

COHERENCY LIMITS IN LATTICE-MISMATCHED NANOWIRES: MODEL

The equilibrium (i.e. unstrained) radius of the overlayer and substrate are denoted by R_o and R_s, respectively; it is assumed that $R_s > R_o$. The lattice mismatch between the overlayer and the substrate is denoted by f, such that $R_s = (1+f)R_o$. The relative stability of two systems is considered: a coherent, undislocated system and a system with one perpendicular pair of misfit dislocations (pure edge) that intersect at the geometric center of the interfacial plane. Mirroring the Matthews-Blakeslee approach, it is assumed that the lattice-mismatch f is partially accommodated by strain relaxation due to misfit dislocation formation, resulting in an effective lattice mismatch given by

$$\left|f_{eff}\right| = \left|f\right| - \frac{nb}{2R_o} \quad , \quad n = 0,1 \quad ; \tag{1}$$

with $n = 0$ corresponding to the coherent structure, and $n = 1$ corresponding to the structure with misfit dislocations. Unlike the Matthews-Blakeslee model, the relaxation of both the substrate and the overlayer is allowed, such that the effective lateral strain f_{eff} can be accommodated by a residual strain ε_{ro} in the overlayer and a residual strain ε_{rs} in the substrate:

$$\left|f_{eff}\right| = \left|\varepsilon_{ro}\right| + \left|\varepsilon_{rs}\right| \quad . \tag{2}$$

The materials of interest are assumed to be elastically isotropic. Implicit in this formulation is the assumption that, as with the planar thin film model, the one-dimensional heterostructure can be modeled as a system under biaxial stress. Shear stress components are assumed to be negligible in the formulation that follows. Further, it is assumed that the radial strain components ε_{rr} are everywhere uniform in the plane of the interface (both in the dislocated and undislocated structure), and that planes in the initial equilibrium formulation remain planar once

the interfacial bonding is affected. The implications of these assumptions are discussed below. With these assumptions, the expression for the strain energy density is:

$$e = \left(\frac{Y}{1-\nu} \right) \varepsilon_{rr}^2 \quad , \tag{3}$$

where Y denotes the Young's modulus and ν denotes the Poisson ratio for the medium.

Now, it is assumed that the strain energy density decays exponentially with distance from the interface, with a decay length given by $\lambda_o = \alpha_o R_o / 2$ in the overlayer and $\lambda_s = \alpha_s R_s / 2$ in the substrate (α_o and α_s are numerical factors of order unity). For an infinitesimally thin slab of the overlayer at $z = z_o$, the strain energy density is then given by

$$dE_o(z_o) = \pi R_o^2 \left(\frac{Y_o}{1-\nu_o} \right) \varepsilon_{ro}^2 \exp\left[-\frac{2z_o}{\alpha_o R_o} \right] dz \quad , \tag{4}$$

where the subscript 'o' indicates the overlayer. The total strain energy stored within the overlayer then is obtained by integrating Eq. (4) from $z = 0$ to $z = \infty$:

$$E_o = \left(\frac{\alpha_o \pi R_o^3}{2} \right) \left(\frac{Y_o}{1-\nu_o} \right) \varepsilon_{ro}^2 \quad . \tag{5}$$

One finds an analogous expression for the substrate. Incorporating Eq. (2), and minimizing with respect to ε_{ro} yields the expression for the total elastic strain energy in the system:

$$E_{elastic}^* = \frac{(1+f)^3 \pi R_o (bn - 2fR_o)^2 Y_o Y_s \alpha_o \alpha_s}{8\left((1-\nu_s) Y_o \alpha_o + (1+f)^3 (1-\nu_o) Y_s \alpha_s \right)} \quad . \tag{6}$$

where Y_s and ν_s are Young's modulus and Poisson's ratio for the substrate.

It is difficult to determine, *a priori*, the energetically favored misfit dislocation configuration in heterostructured nanowire systems; for example, consider the complex dislocated island morphologies in strained epitaxial systems [11,12]. For simplicity, the energetics of the simple dislocation geometry illustrated in Fig. 1(a) is considered. This simple geometry has the advantage that it symmetrically affects the strain distribution in the nanowire system. However, it is quite likely that there are other dislocation configurations that are energetically more stable than the simple one considered here. The consequence is that the present model is likely to overestimate the critical radius for the nanowires.

Misfit dislocations introduce both a core and strain energy. Here the strain and core energy arising from the dislocation pair are modeled using Hirthe and Lothe's formulation [13]:

$$E_{disl}^{n_i} = 2n(2R_o)\left(\frac{Y_o}{1-\nu_o}\right)\left(\frac{b^2}{8\pi(1+\nu_o)}\right)\mathrm{Log}\left[\frac{\beta R_o}{b}\right] \quad . \qquad (7)$$

In Eq. (7), b denotes the Burger's vector for each dislocation, β contains the lower limit for the integration of the strain field as well as the dislocation core energy. The nanowire radius R_o is chosen as the upper cutoff for the dislocation strain energy.

The final expression for the total energy is given:

$$E_n = \frac{(1+f)^3\pi R_o(bn-fR_o)^2 Y_o Y_s \alpha_o \alpha_s}{(1-\nu_s)Y_o\alpha_o + (1+f)^3(1-\nu_o)Y_s\alpha_s} + 2n(2R_o)\left(\frac{Y_o}{1-\nu_o}\right)\left(\frac{b^2}{8\pi(1+\nu_o)}\right)\mathrm{Log}\left[\frac{\beta R_o}{b}\right] \quad . \qquad (8)$$

By considering the relative values of E_1 and E_o, it is possible to determine the relative stabilities of the two systems discussed.

STRAIN ENERGY DISTRIBUTIONS IN LATTICE-MISMATCHED NANOWIRES

In this section, the implications of some of the assumptions in the above model are explored. A finite element analysis is employed. The heterojunction is modeled by enforcing the condition that the interface between the materials be coherent. Elastic constants are chosen to be appropriate for a Si-Ge heterojunction. For comparison, we note that the average strain energy per atom of the same order of magnitude as that of atoms in lattice-strained islands [14].

Finite element simulations reveal that the radial displacement varies linearly with the r-coordinate. Thus, the approximation that strain ε_{rr} is constant in the interface is reasonable. However, it is clear from the finite element analysis that shear components of the strain are not zero, and further that planes in the initial configuration do not remain planar in the bonded system. Therefore, the validity of Eqs. (4), (5) and (6) must be explored. Specifically, the approximation that the strain energy decays exponentially as one moves away from the interface must be examined.

Fig. 2(a), depicts the computed strain energy density in [J/nm] vs. distance from interface for a system with $Y_s = 80$ GPa, $\nu_s = 0.25$, $Y_o = 110$ GPa, $\nu_o = 0.22$, $R_o = 100$ nm, and various lattice mismatches $f = 0.01, 0.02, \ldots, 0.12$. Fig. 2(b) plots the logarithm of the strain energy density for the particular case corresponding to the silicon/germanium system with $f = 0.04$ and show least-squares fit to a linear function. The decay lengths λ_o and λ_s are determined from the slope of the linear fit. Fig. 2 indicates that the assumed exponential decay is reasonable. The total strain energy of the system, computed numerically from the exact data, differs from the total strain energy as computed by numerical determination of λ_o and λ_s and by a factor of $\gamma = 1.16$. The mismatch dependence of the decay parameters, α_o and α_s, were also studied for the same set of parameters. These parameters remain nearly constant with a value near $\alpha_o = \alpha_s = 0.375$. Hence the assumptions in the simple model lead to a reasonable description of the strain energy in the nanowire. Further, the assumption that the overlayer and substrate are infinitely long can now be restated: the model presented is reasonable when the overlayer and substrate are both longer than a few decay lengths.

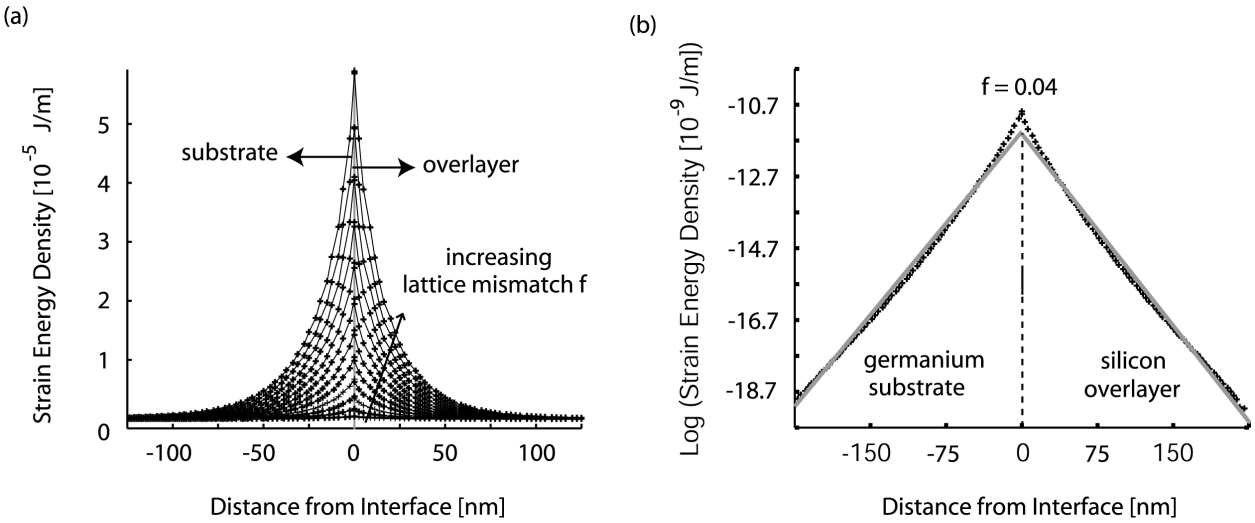

Figure 2. (a) Strain energy density as a function of distance from heterostructure interface. $Y_s = 80\,\text{GPa}$, $\nu_s = 0.25$, $Y_o = 110$ GPa, $\nu_o = 0.22$; lattice mismatches such that $R_o = 100$ nm, $R_s = (1+f)R_o$ with $f = 0.01, 0.02, \ldots, 0.12$. (b) Illustration of strain energy decay for silicon-germanium heterostructure system, fitted to an exponentially decaying function with decay constants $\lambda_o = \alpha_o R_o / 2$ in the overlayer and $\lambda_s = \alpha_s R_s / 2$.

RESULTS AND DISCUSSION

Eq. (8) can now be used to explore the relative energies of dislocated and undislocated nanowires. For each value of the the misfit f, one finds a critical overlayer radius, R_o^*, for which the dislocated and undislocated strain energies are equal. For $R_o > R_o^*$, the dislocated system has the lower energy while for $R_o < R_o^*$, the perfect interface is favored.

Fig. 3 plots the stability diagram for the material system considered above. The solid line gives the critical radius R_o^* as a function of lattice mismatch f using Eq. (8); the dashed line incorporates the numerical correction factor γ deduced from the finite element simulations. As the lattice-mismatch decreases, the undislocated structure remains stable for larger radii. There is not much difference between the dashed and solid lines, suggesting that Eq. (8) is adequate for design engineering.

A true test of the quality of the model is comparison with experiment. Unfortunately, experimental data for testing this model is not yet available. An alternative to experiment is comparison with a more complete model of the elastic energy. Development of such a model is left to future research.

CONCLUSIONS

A simple model to study the equilibrium and coherency limits in heterostructured nanowires is introduced. The model is straightforward to implement, and predicts conditions under which

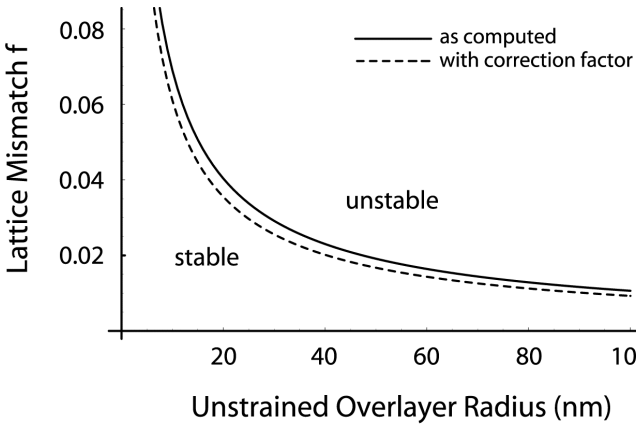

Figure 3. Stability diagram for heterostructured nanowire system with $Y_s = 80$ GPa, $\nu_s = 0.25$, $Y_o = 110$ GPa, $\nu_o = 0.22$, $b = 0.27$ nm, and $\beta = 4$. The region where the coherent structure remains stable with respect to formation of misfit dislocations and the regions where the coherent structure is unstable with respect to formation of misfit dislocations is delineated. The effect of correction factor $\gamma = 1.16$ for the exponential energy decay approximation is illustrated as well.

misfit dislocation formation can be expected. An experimental investigation of the quality of the model is still necessary.

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