

MINIMIZATION OF PERIODIC-BOUNDARY-INDUCED STRAIN IN INTERFACE SIMULATIONS

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We present a discussion of the periodic-boundary-induced strain that is necessarily present in simulations of interfaces between materials with differing lattice structures. We propose a simple, systematic method of minimizing such strain, which effectively eliminates perturbations imposed by the periodic boundaries. A sequence is formulated that characterizes the degree of strain present. This sequence is used in several examples to determine the optimal choice of periodic cell dimensions that minimize the periodic-boundary-induced strain. We show that the method is easily generalized to any interface structure.

KEY WORDS: Periodic boundaries, surface strain, minimization method.

1. INTRODUCTION

In conventional Monte Carlo and molecular dynamics simulations, periodic boundary conditions (PBCs) are used to model an infinite or semi-infinite system. These PBCs eliminate perturbations in the dynamics of small systems that occur if wall boundaries or abrupt edges are used. The only constraint imposed by PBCs is that the system is now periodic with a unit cell the size of the periodic cell. In practice, this means that an upper limit exists on the wavelength for observing position dependent correlations, or equivalently, a lower limit exists in reciprocal (k) space.

For single phase systems, the choice of the appropriate PBCs is simply a matter of knowing the desired density and phase symmetry. For systems of cubic or rhombohedral symmetry, a periodic box is used. For disordered media, several choices of PBCs are possible, but the most prevalent is cubic [1, 2]. Such PBCs are well defined and have been tested extensively.

For the study of multi-phase systems, the appropriate PBCs are not uniquely defined and many ambiguities need to be resolved prior to performing simulations. This paper focuses on the use of PBCs to study interface structures between ordered materials of different symmetry and/or lattice constant. For interfaces, the problem is that only one set of PBCs must be chosen to satisfy two types of unit cells. As an example, consider an interface between two simple cubic (sc) materials, where the (100) faces of the two materials form the interface ($sc_a(100)/sc_b(100)$). The relative misfit of the two materials can be written in terms of the fractional lattice mismatch $M = (b - a)/b$, where a and b are the lattice constants of the two crystals (more exactly, they should be the lattice

constants parallel to the interface). The reciprocal of M is the vernier misfit [3], $P = b/(b - a)$. The vernier represents the wavelength (in terms of the number of unit cells of material a) of the misfit between the two lattices.

When choosing the PBCs, one must decide whether to select the periodic cell dimensions to be an integer multiple of the lattice constant of material a , material b , or some average of the two. The choice of periodic cell dimensions will affect the dynamics of the simulation because it may induce a strain (isotropic or anisotropic) in materials that do not fit perfectly into the cell. One might consider performing constant pressure simulations, in which the cell size and/or shape can change dynamically in response to the system [4, 5]. This is adequate for bulk phase changes, but for an interface, the problem remains that only one of the two phases forming the interface may be accommodated precisely in a given periodic cell. In addition, both bulk materials far from the interface have precisely preferred structures and therefore well-defined periodic box lengths. Thus, choosing a preferred box length for one of the materials will impose an unphysical strain on the other material. Furthermore, any chosen bulk periodic box dimensions will impose unphysical strain on the interface, which can cause structures to form that may not be lowest in energy for the layers immediately adjacent to and on either side of the interface.

One solution to this problem of periodic boundary choices would be to enumerate all possible choices of periodic cells for each phase under consideration. Then, examine the two lists of periodic cell dimensions to find a pair of periodic cells (one from each list) which are closest to one another, and choose this (or some compromise of the two dimensions) to be the chosen simulation cell. This enumeration is clearly feasible but open to pitfalls. For example, in the two phase case, an infinite number of continuous rotations could be considered for which different periodic cells would be applicable. Fortunately, in most cases, there are a few preferred orientational relationships between phases that are known to be most probable. An enumeration list can be created for each of the most probable interfaces in order to find a single periodic cell that could accommodate all of the most probable structures. The question becomes how to assess which periodic cell dimensions minimize the induced strain on both sides of the interface for this finite list of preferred orientations. Clearly, enumeration quickly becomes unwieldy as the number of possible orientations increases. This paper reports our development of an alternative, systematic methodology for the evaluation of PBC-induced strain at complex interfaces and hence a prescription for the proper choice of boundary conditions that minimizes PBC-induced strain in simulations of multiple ordered phases.

2. DEFINING A STRAIN SEQUENCE

Consider a two-dimensional interface between a bulk substrate and a thin film grown on the substrate. The fraction of PBC-induced strain for a given film/substrate system can be expressed in terms of the substrate-dependent PBCs. (The use of the terminology film/substrate does not limit the discussion to such systems. It applies equally well to bulk/bulk or film/film (superlattice) interfaces. The substrate here is merely the material for which the PBC will be restricted to be a simple integral multiple of that material's unit cell.) Let n be the number of substrate unit cells and a_n be the surface unit cell lattice constant of the

substrate. For a given choice of n , the length of the periodic cell for the simulation will be $L = na_n$. The optimal placement of the adsorbed film will depend upon m , the number of film unit cells, and a_m , the lattice constant for the film. The percent strain in the film, $\%S$, induced by the PBCs, is given by

$$\%S = S(m, a_n, n, a_m) \times 100 = \frac{na_n - ma_m}{na_n} \times 100, \quad (1)$$

where S is the fractional strain. For a given choice of n , a_n , and a_m , S is a discrete sequence, S_m . The S_m sequence is linear with a slope of a_m/na_n and an intercept, S_0 , of one. This sequence changes from positive to negative and the member of the sequence, S_{m^*} , with the value closest to zero will minimize the strain. We find it more convenient to replace S_m with a sequence, $f_m(a_m, n, a_n)$, that is not monotonic but will be nonnegative.

$$f_m = \left| \frac{na_n - ma_m}{na_n} \right| \quad (2)$$

This is essentially the PBC analog of the misfit, M , defined in the introduction. It is clear that f_m depends only parametrically upon n , a_n , and a_m . A consequence of the positive-definite form of the sequence is that at least one value of m will exist that produces a minimum in the sequence f_m .

Now consider the case in which only a_n and a_m are fixed. We wish to examine the properties of another sequence, f_n . Each member of the sequence, f_n , is selected by minimizing f_m with respect to m for each value of n . In other words, the optimal value of m for each n is used to construct the sequence f_n . This sequence can be shown to be convergent.

$$\lim_{n \rightarrow \infty} f_n = 0. \quad (3)$$

If $B > 0$ is some numerical bound, then there will be values of n for which $f_n < B$. Considering the computational limits placed upon simulations, the goal of this work is to find appropriate values of B that ensure minimal PBC-induced strain at the interface, thus causing only small perturbations to the simulation ensemble. Finally, we then select the smallest value of n for which $f_n < B$. The smaller the chosen value of n , the fewer atoms needed for the simulation, and the smaller the value of f_n , the less boundary-induced strain present in the film.

We define the strain sequence by

$$f_n = \left| \frac{(na_n - m^*a_m)}{na_n} \right|, \quad (4)$$

where m^* is the optimal value of m , i.e., that m which minimizes f_m for a given n . Note that f_n is defined again as an absolute value in order to ensure the sequence is always positive. The selection of m^* for a given n is now trivial, since we desire $ma_m \approx na_n$. It is simply given by

$$m^* = NINT(na_n/a_m), \quad (5)$$

where $NINT$ is the nearest integer to the value of the argument. The strain sequence f_n is now given by

$$f_n = \frac{(na_n - a_m NINT(na_n/a_m))}{na_n}. \quad (6)$$

Thus, the $NINT$ function simply serves to minimize f_n with respect to m for fixed n , a_n , and a_m . This strain sequence, f_n , represents the strain in material m as a function of the periodic box size (determined by n).

In the following sections, we will present examples and applications of this strain sequence that aid in choosing optimal periodic boundary conditions for simulations of lattice-mismatched interfaces.

3. TYPES OF INTERFACES

A. Commensurate Overlayers

First consider the case of a substrate with fixed lattice constant, a_n . If the adsorbed material is known to form commensurate submonolayer phases on the substrate, the choice of PBCs is governed by the desire to allow formation of various possible commensurate phases without formation of defects at the periodic boundaries. As an example, consider a system for which 2×1 , 2×4 and 4×4 commensurate phases exist on an fcc(100) square substrate. The 2×1 and 2×4 can be considered as subsets of the 4×4 . The PBC then should be chosen as an integer multiple of the 4×4 substrate unit cell

$$L_x = 4ka_n \quad (7)$$

$$L_y = 4la_n \quad (8)$$

where k and l are integers. It is important to choose correct PBC's in order to avoid the formation of PBC-induced defects, dislocations, etc. Note that by selecting PBCs with the above selection criteria one can study each of the submonolayer commensurate phases without introducing periodic-cell-derived anomalous behavior.

B. Incommensurate Interfaces

A second example for which PBCs play an important part in a simulation are incommensurate interfaces. Let us first consider a one-dimensional (line) interface between two semi-infinite, two-dimensional materials with different lattice constants, a_n and a_m . The condition that the PBCs should ideally obey is

$$L_x = na_n = ma_m \quad (9)$$

where n and m are integers and x is the direction parallel to the interface. Satisfaction of this condition is sufficient to guarantee no strain in either

material. Unfortunately, the values of n and m that will satisfy the above conditions within some threshold may be very large and the practical drawback exists that computations are only feasible for small n and m . A more practical condition may be obtained by algebraic manipulation of equation 9

$$\left| \frac{na_n - ma_m}{na_n} \right| < B \quad (10)$$

where zero has been replaced by B , a bounding value. The practical choice of n and m becomes the smallest values which satisfy the bounding condition. Using the previously mentioned substitution which minimizes the left hand side of equation 10 with respect to m , we obtain a sequence f_n which is that defined in an ad hoc manner in equation 6.

$$f_n = \left| \frac{na_n - NINT(na_n/a_m)a_m}{na_n} \right| < B \quad (11)$$

Let us consider such a one-dimensional interface system in which $a_n = 3.61$ and $a_m = 3.52$. By rearranging equation 9, we obtain the conditions which will identically solve our relation for L_x

$$\frac{3.52}{3.61} = \frac{352}{361} = \frac{n}{m} \quad (12)$$

Thus a strain-free simulation could be performed if $m = 361$ and $n = 352$ were chosen, giving a period box length of $L_x = (3.61)(352) = 1270.72$. If we instead use equation 11, where a small finite PBC-induced strain is deemed acceptable in the simulation, then we can use a much smaller periodic box. From equation 11 above, we can plot f_n vs. n (Figure 1). Note first that for small values of n , the strain remains constant because the least strain is achieved by having $m = n$. The length of this plateau in strain depends on how similar the two lattice constants are to each other. The closer they are, the longer the plateau (see Figure 1) because strain will be minimized for $m = n$ until larger n . The rise and fall of the f_n sequence demonstrates that various choices for n may be made for which f_n is small. The smallest value of n , for $a_n = 3.61$ and $a_m = 3.52$, that virtually eliminates strain occurs for $n = 39$. However, if some strain is acceptable, we can reduce the size of the periodic cell further. For example, if B is chosen to be 0.010 (i.e., 1% PBC-induced strain), then the smallest value of n for which $f_n < B$ is $n = 28$. This would correspond to a periodic cell with length of $(3.61)(28) = 101.08$, in which $m = 29$ adlayer unit cells would fit with less than 1% PBC-induced strain. While this value of n may seem large, it is much more feasible than $n = 361$, which yields a simulation cell an order of magnitude larger. The other case shown in Figure 1 is f_n for a system in which $a_n = 3.80$ and $a_m = 3.52$. For this sequence $f_n < 0.01$ when $n = 12$ and is nearly zero at $n = 25$. Note that the frequency of the oscillations in the strain sequence is seen to be much greater for the system which has the greater initial misfit, while the convergence toward zero strain ($n, m \rightarrow \infty$) is approximately independent of initial misfit.

Figure 1 illustrates that there are particularly bad choices of n for which the PBC-induced strain would be quite large. Selection of PBCs for which $f_n > B$

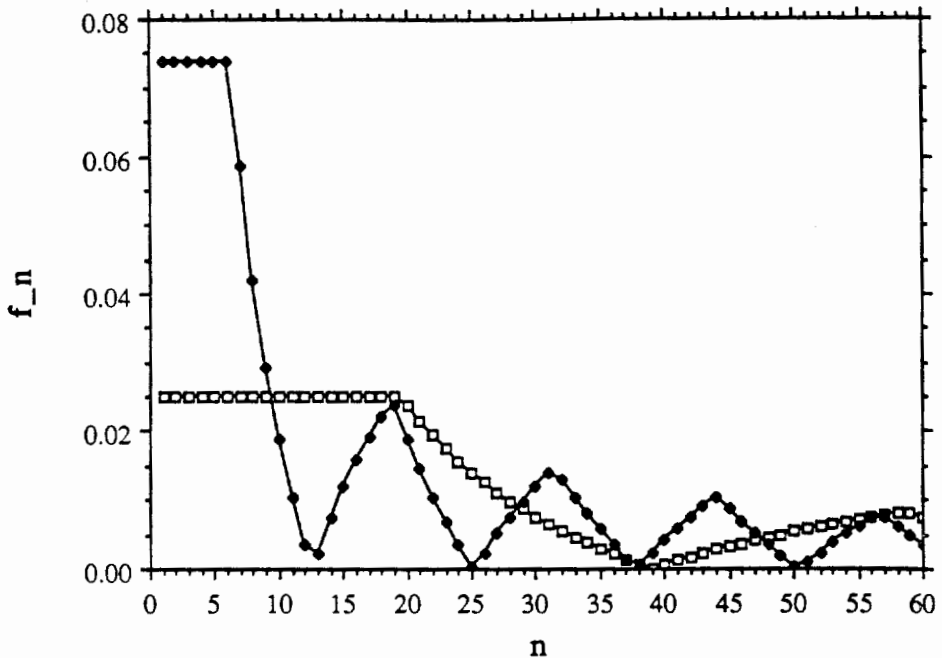


Figure 1 The strain sequence, f_n is shown for a system where $a_n = 3.61$, $a_m = 3.52$ (\square) and for $a_n = 3.80$, $a_m = 3.52$ (\blacklozenge).

could easily result in dynamics that would be drastically perturbed by the PBC-induced strain in the adlayer. Separate investigations should be implemented to determine the limiting values of B at which PBC-induced strain affects the results of the simulation. We reiterate that in our model, the PBC-induced strain is present only in the material with lattice constant a_m . No strain is present in the material a_n because L_x (the length of the periodic box) is constrained to be an integer multiple, n , of this lattice constant, as stated at the beginning Section II.

This simple example clearly demonstrates the dependence of PBC-induced strain on the number of substrate unit cells used to define the periodic boundaries. Let us now consider two-dimensional interfaces between two semi-infinite three-dimensional substances, A and B .

1. Square/Square fcc(100)/fcc(100)

A simple first case involves the interface between two square lattices with differing lattice constants. The two lattices are assumed to align such that the square unit cells for the two materials have the same orientation. This gives two possible interface structures, one in which $A[010] \parallel B[010]$ and the other rotated 45° , where $A[010] \parallel B[011]$ (see Figure 2). We are neglecting the infinite number of rotations in between, which could occur at the interface, since considering these rotations would render conventional periodic boundary methods intractable anyway. In such cases, one needs to consider islands as models of the semi-infinite materials [6].

Given the relative orientations shown in Figure 2, the effect of PBC-induced strain can be handled in exactly the same way as for the one-dimensional interface, with the only difference being that there will be two strain sequences, f_{n1} and f_{n2} , one for each of the two orientations depicted in Figure 2. The strain sequences along the x and y directions are the same for each of the two interface orientations, and so we need only include one sequence for each interface. A total strain sequence that we wish to minimize may then be defined as a simple sum of the strain sequences for the two interface orientations

$$f_n = f_{n1} + f_{n2}, \quad (13)$$

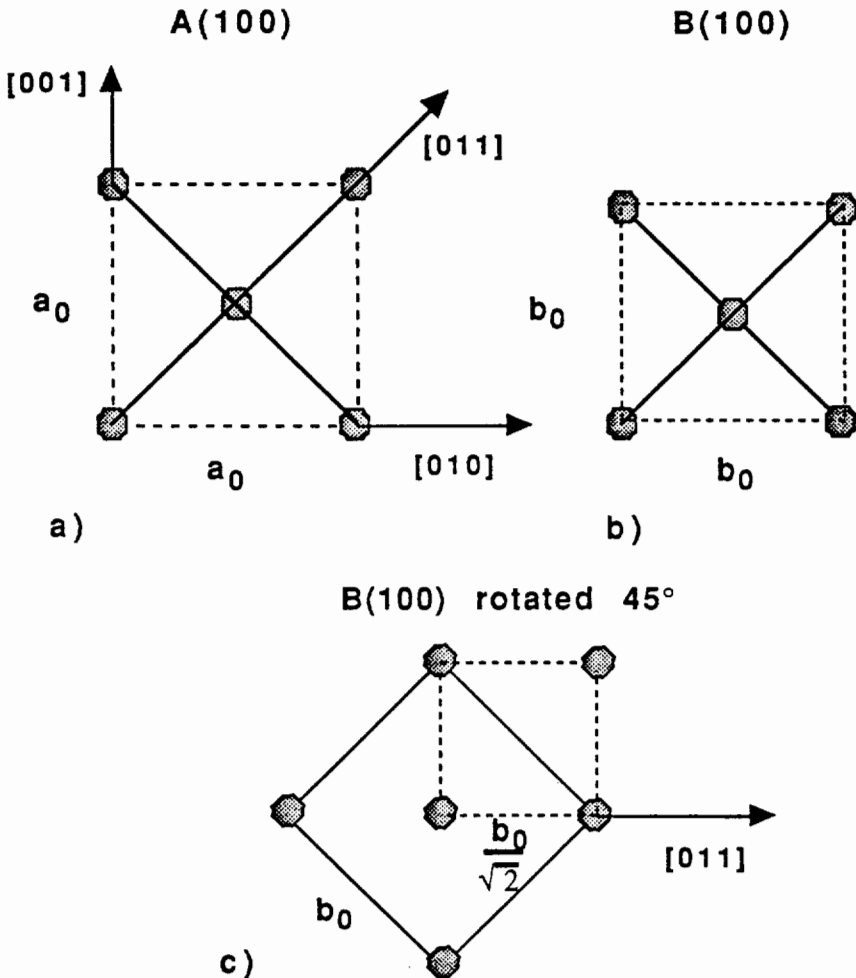


Figure 2 The geometry at the interface is shown for (a) the A(100) surface, for which the periodic box will be chosen to be an integer multiple of its unit cell, (b) the B(100) surface in the orientation where $A[010] \parallel B[010]$ and (c) the B(100) surface rotated 45° where $A[010] \parallel B[011]$. The dashed lines represent the surface unit cells used in calculating the f_{n1} and f_{n2} strain sequences.

where

$$f_{n1} = \left| \frac{n_1 a_0 - NINT(n_1 a_0 / b_0) b_0}{n_1 a_0} \right| < B_1 \quad (14)$$

and

$$f_{n2} = \left| \frac{n_2 a_0 - NINT(\sqrt{2} n_2 a_0 / b_0) b_0 / \sqrt{2}}{n_2 a_0} \right| < B_2. \quad (15)$$

As an example we consider the Pd(100)/Ag(100) interface, where $a_{Pd} = 3.89$ and $a_{Ag} = 4.09$. Figure 3 shows the strain sequences for each of the two possible interfaces, as well as the sum. The orientation with $A[010] \parallel B[010]$ has a periodicity of ~ 20 unit cells, while the orientation with $A[010] \parallel B[011]$ has a strain sequence that oscillates rapidly due to the large initial strain between the unit cells. Note that for $n < 16$, odd values of n have large PBC-induced strains compared to even values. It is clear that a choice of $n = 19$ for the periodic cell dimensions (in units of a_0) would ensure that the total strain would be approximately 1% and that for $n = 39$, the total PBC strain would be almost negligible. Both of these choices should be considered in picking an optimal periodic simulation cell. More importantly, this data tells us that the PBC-induced strain, in material B , increases rapidly for a periodic cell with dimensions of $n < 16$ and is also relatively large for $n = 26, 28, 30$. Thus arbitrarily increasing the size of the periodic cell does not guarantee that the strain decreases. The larger the PBC-induced strain, the more probable that the dynamics will be affected. It is important to note as well, that for this particular system, a change in the periodic cell dimensions of only $n \pm 1$ can cause large jumps in the PBC-induced strain (caused by the f_{n2} term).

2. Rectangular/Square fcc(111)/fcc(100)

An additional complexity may be introduced if an interface is formed between a material with a rectangular (or rhombohedral) surface unit cell (e.g., fcc(111)) and a material with a square surface unit cell (e.g., fcc(100)). (In order to simplify our construction of the periodic cell, we consider the fcc(111) surface in terms of periodically-replicated rectangles rather than the usual primitive unit cell, which is a rhombus.) In this case, the strain sequences along each of the perpendicular directions will not be equivalent. For notational simplicity, we have defined the lattice constants of the substrate to be a_x and a_y (for fcc(100), $a_x = a_y = a_0$) and the lattice constants of the film to be b_x and b_y (for the fcc(111) film $b_x = (b_0/\sqrt{2})$ and $b_y = (b_0\sqrt{3}/\sqrt{2})$). Since the substrate is isotropic along the x and y directions, then it makes no difference whether we align b_x parallel to a_x or a_y . The applicable strain sequences may then be written as

$$f_{nx} = \left| \frac{n_x a_x - NINT(n_x a_x / b_x) b_x}{n_x a_x} \right| < B \quad (16)$$

$$f_{ny} = \left| \frac{n_y a_y - NINT(n_y a_y / b_y) b_y}{n_y a_y} \right| < B. \quad (17)$$

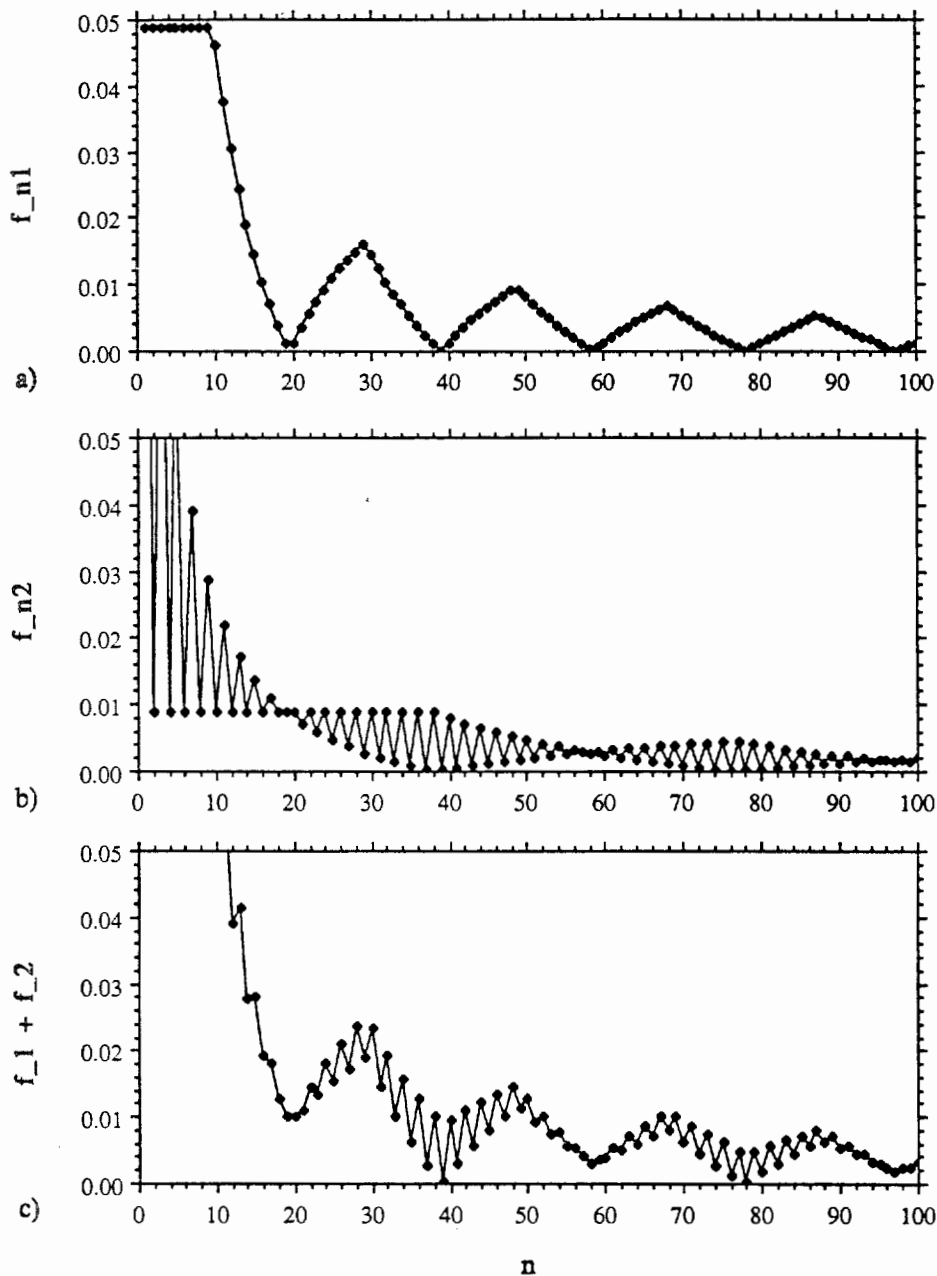


Figure 3 The strain sequences are shown for the two orientations of the Pd(100)/Ag(100) interface shown in (a) Figure 2a and Figure 2b and (b) Figure 2a and Figure 2c. (c) The sum of the two strain sequences.

The values of n which separately satisfy these two equations will not necessarily be the same (i.e., n_x does not have to equal n_y) and will not have identical PBC strains. To assess the overall picture, let us again define a sum of the individual strain sequences.

$$f_n = f_{n_x} + f_{n_y} < 2B \quad (18)$$

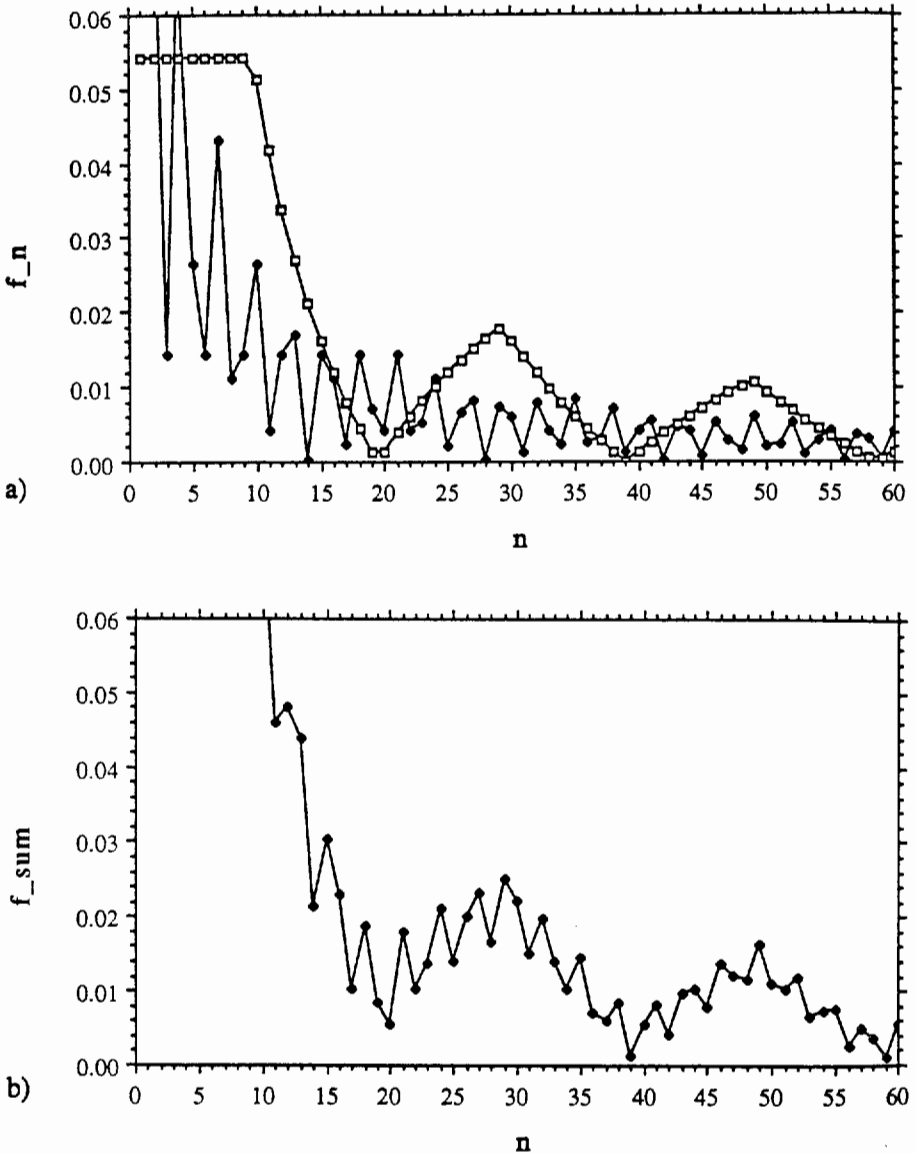


Figure 4 (a) The strain sequences for f_{n_x} (\blacklozenge) and f_{n_y} (\square) are shown for Ni(111)/Ag(100). (b) The sum of the strain sequences, $f_n = f_{n_x} + f_{n_y}$.

The bounding condition of $f_n < 2B$ can be used to determine optimal values of n . As an example of this situation, consider the Ni(111)/Ag(100) system in which a Ni(111) film forms on a Ag(100) substrate [7] Figure 4 shows the separate f_{nx} and f_{ny} sequences and the sum of the two. Note that f_{nx} oscillates more rapidly than f_{ny} because of the larger difference in lattice constants. We see that $n = 19$ or 20 are optimal periodic cell sizes for this system ($a_0 = 4.09$, $b_0 = 3.52$). If we are careful to keep the described alignment of the two surfaces, we could also separately minimize f_{nx} and f_{ny} rather than the sum. Clearly, eliminating the restriction that n_x and n_y be identical can minimize the strain still further. This suggests that a rectangular periodic cell could be used with, e.g., dimensions of 11×17 or 14×19 for this example of Ni(111) on Ag(100). The exact dimensions will depend upon the degree of PBC-induced strain which is deemed allowable for the simulation. This methodology can easily be extended to systems in which several different interfaces are possible, involving different surface unit cells.

3. Rectangular/Rectangular fcc(111)/fcc(111)

As another example of PBC-induced strain effects at interfaces, we consider the case in which the surface unit cells of both materials at the interface are rectangular (or rhombohedral). This introduces the additional complication that two different orientations of the interface are feasible, and that the PBC-induced strain along the perpendicular directions is different in both cases. The optimal periodic cell will most likely be rectangular, rather than square, in order to allow for a minimum of strain. Figure 5 shows the two orientations and their respective crystallographic directions. The relevant strain sequences are

$$f_{nx} = \left| \frac{n_x a_x - NINT(n_x a_x / b_x) b_x}{n_x a_x} \right| + \left| \frac{n_x a_x - NINT(n_x a_x / b_y) b_y}{n_x a_x} \right| < B \quad (19)$$

and

$$f_{ny} = \left| \frac{n_y a_y - NINT(n_y a_y / b_y) b_y}{n_y a_y} \right| + \left| \frac{n_y a_y - NINT(n_y a_y / b_x) b_x}{n_y a_y} \right| < B \quad (20)$$

where $a_x = (a_0/\sqrt{2})$, $a_y = (a_0\sqrt{3}/\sqrt{2})$, $b_x = (b_0/\sqrt{2})$ and $b_y = (b_0\sqrt{3}/\sqrt{2})$. Note that we have two terms in each strain sequence to account for the two different orientations that the B(111) surface may have with respect to the A(111) surface (see Figure 5 for one of them; the other is merely a 90° rotation of B(111) in Figure 5). Since either b_x or b_y may be aligned along a_x , we include both possibilities in f_{nx} and f_{ny} . The resulting strain sequences along the substrate x and y directions are plotted in Figure 6 for the example of Cu(111)/Au(111). We see from this that several choices for the periodic cell dimensions ($n_x \times n_y$) may be chosen. An 8×9 periodic cell seems to be the smallest reasonable choice, in which there will be an average of 0.0263 strain in the substrate x direction and 0.0386 strain in the substrate y direction. Alternative (better) choices would include 15×17 and 23×23 . Again we see that changing the periodic cell dimension by one surface lattice constant can change f_n by as much as 0.06 (6% change in strain).

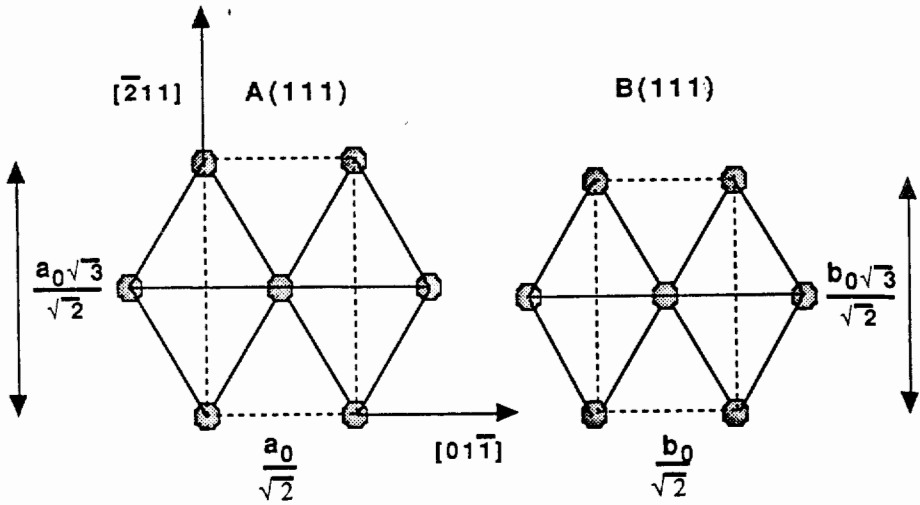


Figure 5 The relative geometries for an fcc(111)/fcc(111) substrate/adlayer with lattice constants a_0 and b_0 respectively. The second possible orientation at the interface can be seen by rotating the B(111) surface by 90° .

4. An example applicable to thin film growth on a square substrate

As a final example of PBC-induced strain, we consider the choice of an optimal periodic cell to be used for the simulation of thin film growth of an fcc metal on a square fcc(100) substrate. As our test example, we consider the deposition of Pd on Ag(100). Several possible interfaces may form in such a system including Pd(111)/Ag(100), Pd(110)/Ag(100), and Pd(100)/Ag(100). We wish to choose a single periodic cell which will allow the formation of all three interfaces with as little PBC-induced strain as possible. The dimensions of the surface unit cells for the three Pd surfaces are $b_0/\sqrt{2} \times b_0\sqrt{3}/\sqrt{2}$, $b_0 \times \sqrt{2}b_0$ and $b_0 \times b_0$, respectively. In addition, we will consider the formation of the 45° rotated Pd(100) unit cell mentioned in the previous example, which has dimensions of $b_0\sqrt{2} \times b_0/\sqrt{2}$ (see Figure 7). We thus consider four possible interface geometries and four unique unit cell lengths for the Pd film (b_0 , $\sqrt{2}b_0$, $b_0/\sqrt{2}$, $b_0\sqrt{3}/\sqrt{2}$). The resulting overall strain function (which is the same in both directions on a square lattice) is given by

$$f_n = f_{n1} + f_{n2} + f_{n3} + f_{n4} \quad (21)$$

where f_{ni} corresponds to the usual strain sequence with unit cell length $i = b_0$, $\sqrt{2}b_0$, $b_0/\sqrt{2}$, or $b_0\sqrt{3}/\sqrt{2}$. Figure 8 depicts this sum and shows that the optimal choices of periodic cell dimensions would include $n = 8, 12, 15, 16, 20$ substrate unit cells in both the x and y directions. A slightly more elegant approach might include weighting functions for each sequence, where the

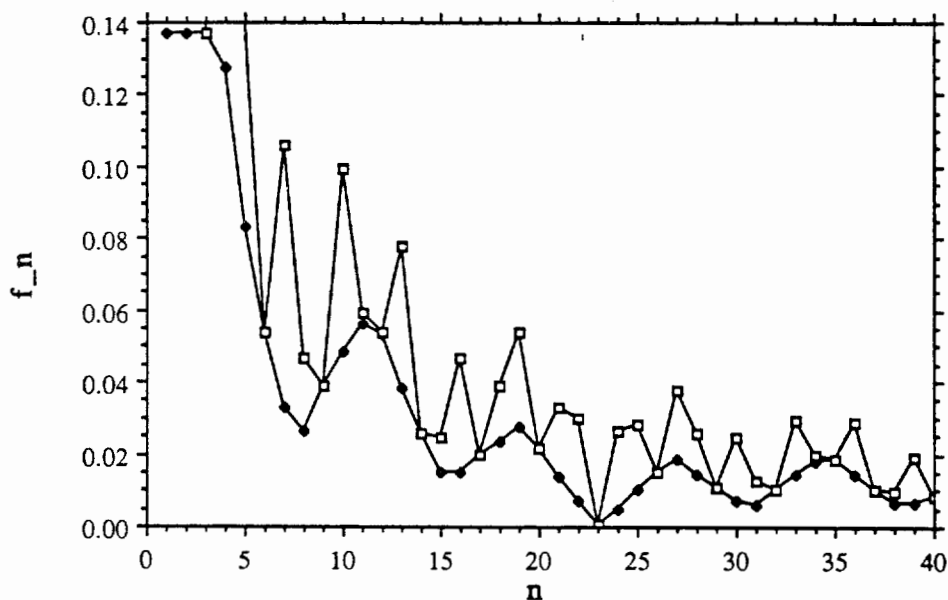


Figure 6 The strain sequences, f_{nx} (\blacklozenge) and f_{ny} (\square) shown for the Cu(111)/Au(111) system.

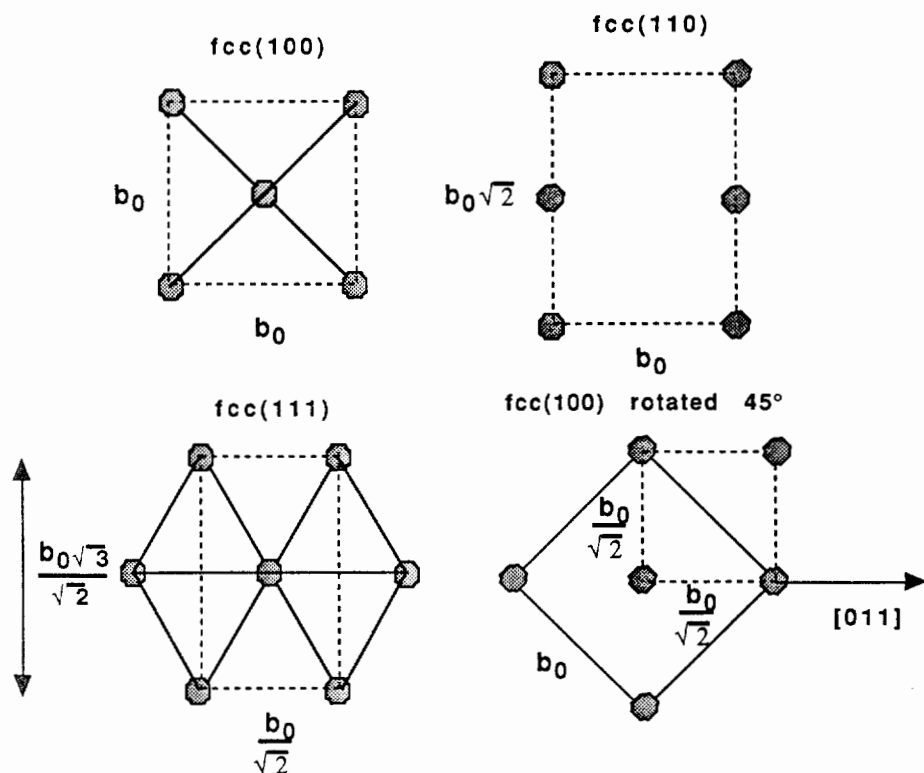


Figure 7 The four orientations of a thin film considered in example 4 in the text.

weighting could be derived from the surface free energies or elastic constants of the various adlayers.

$$f_n = w_1 f_{n1} + w_2 f_{n2} + w_3 f_{n3} + w_4 f_{n4}. \quad (22)$$

This would make the overall strain sequence more sensitive to those interfaces which are most probable based on thermodynamic or physical arguments.

IV. Summary

We have demonstrated the importance of taking into account the PBC-induced strain for simulations of interfaces. It is clear from the numerous examples given that there are optimal and non-optimal choices of the periodic boundaries that respectively minimize or enhance spurious effects in the dynamics and energetics of the simulated systems. In order to quantify the degree of PBC-induced strain, we defined an optimal strain sequence, which is a function only of the number of unit cells of one of the materials forming the interface. If the periodic cell dimensions are chosen to fit one of the materials perfectly, they will induce an expansive or compressive strain in the other material. This strain is characterized, in our description, completely by the surface lattice constants (parallel to the interface) of the two materials. For square lattice interfaces, a single strain sequence must be minimized, whereas for interfaces involving at least one nonsquare lattice, two strain sequences in orthogonal directions parallel to the interface should be minimized independently. Each strain sequence may consist

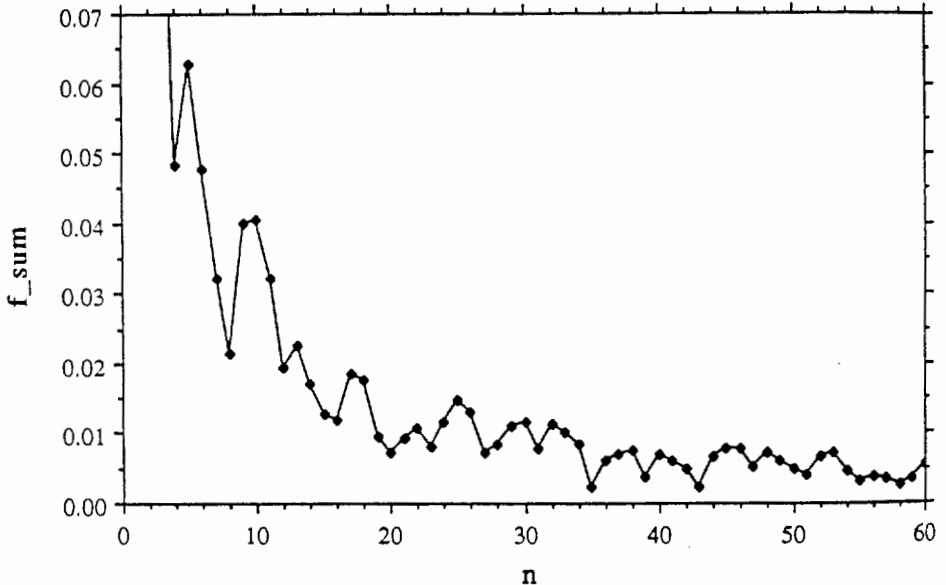


Figure 8 The sum of strain sequences for the four possible thin film structures shown in Figure 7 grown on an fcc(100) substrate. The lattice constants used correspond to that for growing Pd on Ag(100).

of a sum of strain sequences corresponding to different lattice orientations at the interface. The examples given here indicate that the PBC-induced strain can change drastically by changing the periodic cell dimensions only slightly. While our method does not provide a single choice for the periodic cell dimensions, it does narrow options by eliminating those dimensions that will induce the largest strain.

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References

- [1] S.S. Wang and J.A. Krunhansl, "Superposition Assumption. II. High Density Fluid Argon," *J. Chem. Phys.* **56**, 4287 (1972).
- [2] D.J. Adams, "Computer Simulation of Ionic Systems: The Distorting Effects of the Boundary Conditions," *Chem. Phys. Lett.* **62**, 329 (1979).
- [3] J.H. van der Merwe, "Crystal Interfaces. Part I. Semi-infinite Crystals," *J. Appl. Phys.* **34**, 117 (1963).
- [4] H.C. Andersen, "Molecular Dynamics Simulations at Constant Pressure and/or Temperature," *J. Chem. Phys.* **72**, 2384 (1980).
- [5] M. Parinello and A. Rahman, "Crystal Structure and Pair Potentials: A Molecular Dynamics Study," *Phys. Rev. Lett.*, **45**, 1196 (1980).
- [6] A. Kobayashi and S. Das Sarma, "Direct Calculation of Interfacial Energetics: Roles of Axial Commensuration and Strain in Epitaxial Growth," *Phys. Rev. B*, **35**, 8042 (1987).
- [7] B.C. Bolding and E.A. Carter, "Effect of Strain on Thin Film Growth: Deposition of Ni on Ag(100)," *Surface Sci.*, **268**, 142 (1992).