

Stick-slip friction at the atomic scale: A model for van der Waals epitaxial strain

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It has been demonstrated experimentally that friction forces between a two-dimensional (2D) material and an amorphous substrate are sufficient to stabilize tensile and compressive strains on the order of 1% within the 2D material. Here, a simple atomic-scale model is used to explore the friction between a 2D material and an idealized substrate. The atomic-scale model shows that the observed strains can be sustained by bonds with strengths on the order of van der Waals bonds. The model also reveals interesting atomic-scale structures at the interface that include vortices, antivortices, and other more complicated structures.

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I. INTRODUCTION

Transition-metal dichalcogenide (TMDC) monolayers (e.g., MoS₂ and WSe₂) are promising materials for electronic [1,2] and optoelectronic [3,4] applications. Their band structures are known to be strain dependent [3,5–8] and to display direct-indirect band-gap transitions.

Experimentally, the friction forces between an amorphous substrate and a monolayer of WSe₂ bonded to that amorphous substrate can sustain strains on the order of 1% [9] (at room temperature). Moreover, this strain can be released by the evaporation of a liquid droplet in contact with the two-dimensional (2D) material [10]. An analysis rooted in continuum elasticity theory [11] suggests that forces on the order of those expected from van der Waals bonding (~ 0.01 eV/Å) are capable of sustaining the observed strains. Although there have been studies on the effects of conformal mapping on the structure of a growing film [12], no atomic-scale model has been proposed for the structure of the monolayer-substrate interface arising during the strain relaxation process.

The strains observed in Refs. [9,10] arise due to a mismatch in the thermal expansion coefficients. In the case of WSe₂ grown on amorphous silica (a-SiO₂), the thermal expansion coefficient of WSe₂ exceeds that of a-SiO₂. Thus, a film of WSe₂ grown on a-SiO₂ at high temperature will be strained in tension upon cooling provided the friction forces are sufficiently strong.

Lattice registry during epitaxy often results in covalent bonds across the interface between the film and the substrate.

These bonds offer substantial strength, and it is observed experimentally that they can sustain the strain within thin films. However, the amorphous substrate-film interface might necessarily have fewer, if any, covalent bonds. Thus, the friction force might stem entirely from van der Waals bonds.

The atomic-scale properties of the interface between the TMDC monolayer and the fused, amorphous silica surface are of interest. In this paper, an idealized model of this interface is examined, and the atomic-scale structure of the TMDC monolayer at the interface is studied.

II. MODEL

The model developed here is motivated by the properties of a van der Waals bond, as represented by a Lennard-Jones potential (see Eq. (1) of the Supplemental Material [13]). Consider two atoms bound by this potential. Fix one atom at the origin and apply a force on the other atom that tends to stretch the bond. For forces less than $\approx 2.69 \frac{\epsilon}{r_o}$, with ϵ the bond energy and r_o the zero-temperature equilibrium bond length, a stable stretched-bonded configuration can be found. However, for larger forces, the atoms will separate to infinity. Thus, there is a threshold force beyond which the bonds must break. A simple analysis based on the work of Grimme [14] suggests that these bonds have strengths between 0.01 and 0.02 eV/Å (see Table I of the Supplemental Material [13]).

An idealized stick-slip friction model for atoms at the 2D material-substrate interface [15], similar to the Burridge-Knopoff models developed in the study of earthquakes [16], is introduced and analyzed. The model shows that the observed residual tensile strains can be sustained by relatively weak bonds. The atomic-scale structures that emerge highlight the atomic-scale origins of the strain stabilization. Moreover, the atomic-scale structures that emerge display an interesting pattern of forces and displacements that reveal vortices, antivortices, and other more complicated defects.

In the idealized model, the interactions between atoms in the MoS₂ are simulated with the reactive empirical bond

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order (REBO) potential, including a Lennard-Jones potential developed by Liang *et al.* [17]. The calculations are carried out within LAMMPS [18].

The 2D material-substrate interface is then modeled as follows. MoS₂ has a three-layer structure consisting of a bottom layer of S atoms, a middle layer composed of Mo, and a top layer of S atoms. It is assumed that the S atoms in the bottom layer of the MoS₂ are bonded to a smooth, perfectly flat substrate. The net force on each of the bottom S atoms arises from two interactions: (1) the interaction of the S atoms with the other atoms in the monolayer and (2) the interaction with the substrate. The bonding to the substrate leads to a critical friction force on the bottom S atoms that must be overcome to allow the bottom S atoms to move. If the force on a bottom S atom arising from the other atoms in the film is less than the critical force, the atom in question cannot move. However, if the force from the other atoms exceeds the critical force of the atom in question, then that atom is free to move for a short time before it comes to rest at a different position.

A typical simulation begins with a circular film. The atoms at the very edge of the film are necessarily in defective environments. (These are allowed to relax as dictated by the interatomic potential, and the resulting relaxations play little role in the strain relaxation of the film.) The film is then heated up to 1100–1500 K using molecular dynamics (the sample is constrained to be flat by the bonding to the substrate—see the Supplemental Material [13]) and thus undergone thermal expansion similar to that for the WSe₂ films grown in the experiments [9,10]. The film is then relaxed using a simple steepest descent algorithm with a time step of 0.005 ps with the velocities of all atoms zeroed after each step. Before relaxation begins, each bottom layer S atom is assigned a threshold friction force, drawn from a Gaussian distribution with a typical mean of 0.01 eV/Å and a standard deviation of 1/10 of the mean. The forces on the atoms arising from the other atoms in the monolayer are evaluated at each time step using the atomic-scale model. If the force on a bottom layer S atom exceeds the atom's assigned threshold, the friction force is set to zero, and that atom is allowed to move in all three directions in response to the remaining forces upon it. All other atoms are assumed to be able to respond to the forces upon them. At the end of the time step, the bottom S atoms that had moved are assigned a threshold force drawn from the same Gaussian distribution. This process is repeated until a steady-state structure is determined.

III. RESULTS

Figure 1 shows the final strain and the evolution of the reaction force vectors as a function of time during stress relaxation. The local strain is calculated using the algorithm developed in Ref. [19] as implemented within Ovito [20]. Note that the extreme edges of the films appear to be highly strained by this method, but this is an artifact resulting from the disorder at the terminating edges. The neighbor cutoff for the strain calculation was chosen to be 3.6 Å to include only nearest-neighbor S atoms at the interface. The reaction force vectors are the force that the substrate is exerting on the film. At very short times (not shown), the reaction force vectors are all large and appear to point in random directions. As the

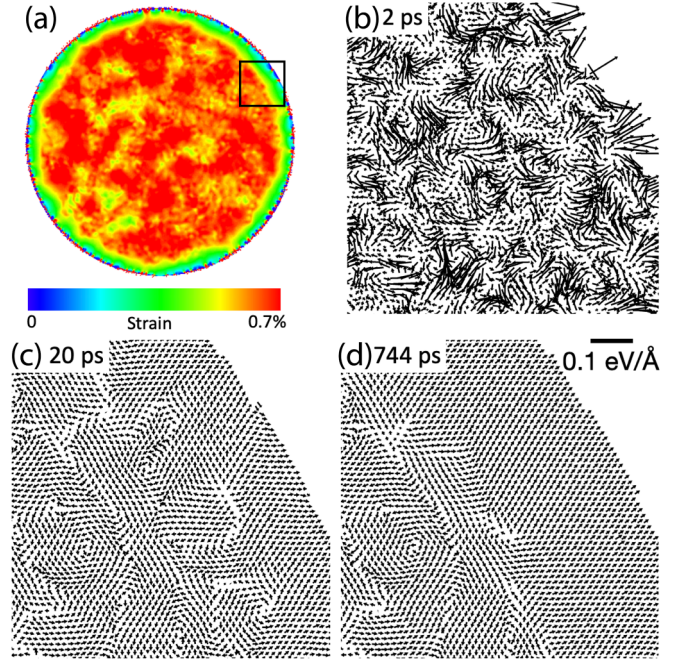


FIG. 1. (a) The strain distribution of a stabilized $r = 50$ nm structure with the force threshold sampling from a normal distribution with a mean 0.01 eV/Å and a standard deviation 0.001 eV/Å. (b)–(d) The evolution of the reaction force vectors in the black box of the strain map with time after the start of the relaxation from 2 ps to the steady state where all forces are lower than their assigned force thresholds. The scale bar for the force magnitudes is shown in panel (d).

system relaxes, the force vectors decrease in length and begin to show local order. Specifically, it is notable that an outer annulus of reaction force vectors with nearly uniform length pointing in the radial direction emerges. The length of these vectors is approximately equal to the mean threshold friction force $f_{\text{threshold}}$.

This outer annulus of oriented forces is similar to that assumed in the theory presented in Ref. [11]. One can derive an expression for the width of the relaxed region within the outer annulus by evaluating Eq. (7) of Ref. [11] with the value of Δ set equal to the unstrained radius of the circular film, R . (In Ref. [11], Δ is radius at which the film remains strained by the substrate.) Then, the width of the annular region is defined to be δ and the strain in the interior of the film defined as ϵ_o . ϵ_o is computed by averaging the strain over the region of the film defined by $R - \delta$. The theory predicts that the annular ring of width δ can stabilize the strain in the film if the average reaction force on the atoms in that strip, given by $\bar{f}$, is

$$\bar{f} = \epsilon_o \frac{3A_c(\lambda + \mu)(2R - \delta)}{\delta(3R - 2\delta)}, \quad (1)$$

where $\lambda = 2.76 \text{ eV}/\text{\AA}^2$ is the (2D) Lamé constant of MoS₂, $A_c = 8.694 \text{ \AA}^2$ is the area of the atomic unit cell, and $\mu = 3.27 \text{ eV}/\text{\AA}^2$ is the (2D) shear modulus of MoS₂. These parameters are calculated using the aforementioned REBO potential [17].

Figure 2(a) shows the predicted thicknesses of the outer annuli as a function of $\bar{f}$ for two values of the initial strain

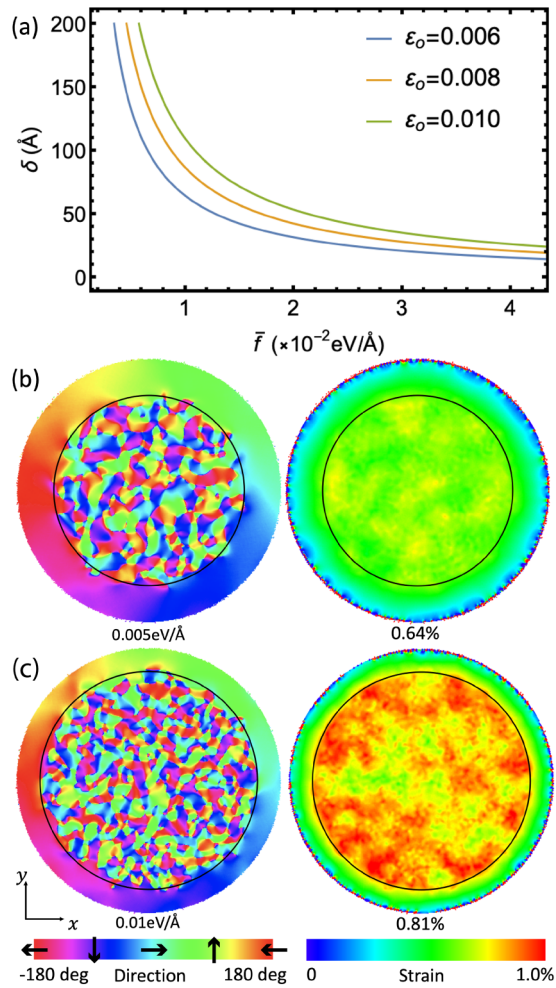


FIG. 2. (a) The width of the relatively relaxed rim plotted as a function of the average forces on atoms in the rim. Three different initial strains are shown. (b) The directional distribution of the reaction forces (the angles are relative to x direction) and the strain distribution map for an initial strain of $\sim 0.64\%$, force threshold mean value of $0.005 \text{ eV/\AA}$. (c) Steady-state structure for initial strain of $\sim 0.81\%$ and force threshold mean value of $0.01 \text{ eV/\AA}$. All the standard deviations of the force thresholds are $1/10$ of the mean value. The solid circle is the boundary of the axially oriented forces predicted by Eq. (1).

for a MoS_2 circular monolayer with an unstrained radius of $500 \text{ \AA}$. As noted in Ref. [11], the continuum model finds that relatively weak forces can sustain interior strains on the order of 1% , strains that are consistent with the strains observed experimentally. Moreover, the stabilized strain is not limited by the magnitude of $\bar{f}$ alone. Smaller values of $\bar{f}$ simply require larger annuli to stabilize the same strain. We note that in practice, however, the residual strain is determined by the initial strain arising from thermal expansion mismatch and the strain relaxation dynamics.

To compare the predictions of Eq. (1) with the atomic-scale simulations, one can set $\bar{f} = f_{\text{threshold}}$, and use the parameters that describe MoS_2 to determine δ . Figures 2(b) and 2(c) display the results of this comparison. Specifically, the color maps show the local orientation of the steady-state reaction

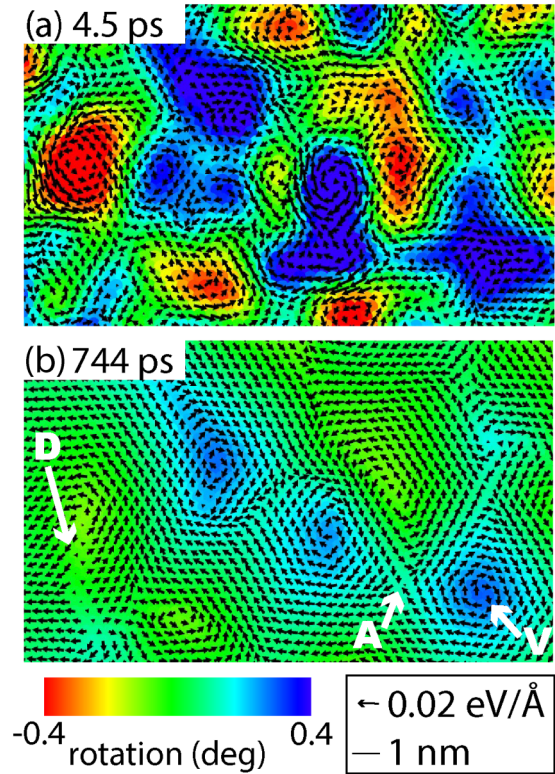


FIG. 3. The evolution of the force vectors on the left edge of the sample in the model with stick-slip friction (a) 4.5 ps and (b) 744 ps after relaxation. The final strain is $\varepsilon_o = 0.0065$, and the radius of the film is $500 \text{ \AA}$. The average $f_{\text{threshold}}$ is $\sim 0.01 \text{ eV/\AA}$ with a standard deviation of $1/10$ of the mean. The local rotation of the crystal is measured relative to its original orientation. A typical vortex (V), antivortex (A), and domain boundary (D) have been identified in panel (b).

force vectors. In addition, superimposed on each map is a circle of radius $R - \delta$ for the given parameters as predicted by Eq. (1). The portion of the color map exterior to this circle shows an orientation that smoothly varies with position, indicating that the reaction force vectors are pointing (on average) in the axial direction. The strain measured in this region is generally lower than the average, representing the effects of the strain relaxation, as expected from the continuum theory [11]. The interior portions of the maps show a more complicated structure, with small domains of aligned forces oriented in seemingly random directions. In all cases, the plotted circle serves to separate the highly ordered exterior region from the interior region. Thus, the predictions of Eq. (1) describe the atomic-scale patterns of residual forces very well. The strain relaxation pattern can be used, in principle, to measure the magnitude of $f_{\text{threshold}}$ experimentally. Specifically, if one can determine the interior strain and the width of the relaxed exterior region using, for example, four-dimensional scanning transmission electron microscopy [21–24], one can compute $\bar{f} \approx f_{\text{threshold}}$.

The patterns of rotations and deformations in the interior regions of the films (Fig. 2) are complex. They reveal that the thermal excitation of the film includes both lateral expansion and domain rotations. Figure 3 shows an example of the

patterns of the reaction force vectors between the substrate and the 2D film. There are several notable features of these figures. First, the reaction force vectors have a typical length approximately equal to $f_{\text{threshold}}$. Second, the arrangement of the reaction force vectors has interesting topologies. The patterns clearly reveal vortices and antivortices [Fig. 3(b)]. One can also see extended domain-wall-like structures that often seem to terminate in vortices. There are also regions in which the magnitudes of the reaction forces are clearly below $f_{\text{threshold}}$. Moreover, the regions with vortices correlate with the local rotations of the 2D crystal. Empirically, it appears that these rotational distortions are the longest-lived features of the thermal disorder during the relaxation process.

To better understand the topological patterns as observed in Fig. 3, the center of the vortices and antivortices was identified by calculating the winding number (WN), $n(x, y)$ [25], of the reaction force vectors:

$$n(x, y) = \frac{1}{2\pi} \oint_L \nabla \theta dr. \quad (2)$$

Here, θ is the angle between the reaction force vector and the reference direction, and L is a closed loop around the point of interest. WN is calculated at every atomic position, and the closed loop is formed by the second-nearest neighbors to reduce noise. Here, WN values of +1 and -1 represent the center of vortices and antivortices, respectively.

Figure 1 of the Supplemental Material [13] shows the final vortex-antivortex configuration for a sample with an initial strain of approximately 0.006. In the early stages of relaxation, the number of vortex-antivortex pairs within the circular sample with a diameter of 500 Å is approximately 1550. Initially, the number of pairs rapidly decreases. By 200 ps, the number of vortex-antivortex pairs has dropped to approximately 400.

Examples of the positions of the centers of vortices and antivortices are shown in Fig. 4 for times between 10.0 and 744 ps. These reductions in number are mediated, in part, by the annihilation of vortex-antivortex pairs (see discussion below). As can be seen in Fig. 4(c), the vortices and antivortices are mobile but do not wander far during the process. The maximum distance traveled during relaxation appears to be on the order of 10 Å.

The topological patterns that appear in the reaction force field lead to similar structures within the atomic-scale displacements. However, these displacements are difficult to discern from a global displacement map because the overall thermal expansion overwhelms the additional local displacements due to the reaction forces. However, the displacements can be discerned if one uses a lattice-template matching method. Figure 2 of the Supplemental Material [13] shows typical displacements associated with various topological features in the reaction force field. From this figure, it can be concluded that the atomic displacements associated with the topological features of the reaction force field can reach approximately 0.02 Å.

IV. DISCUSSION AND CONCLUSIONS

The overall pattern of relaxation, then, can be characterized as follows. As expected, thermal excitation causes the atoms to move from their zero-temperature equilibrium

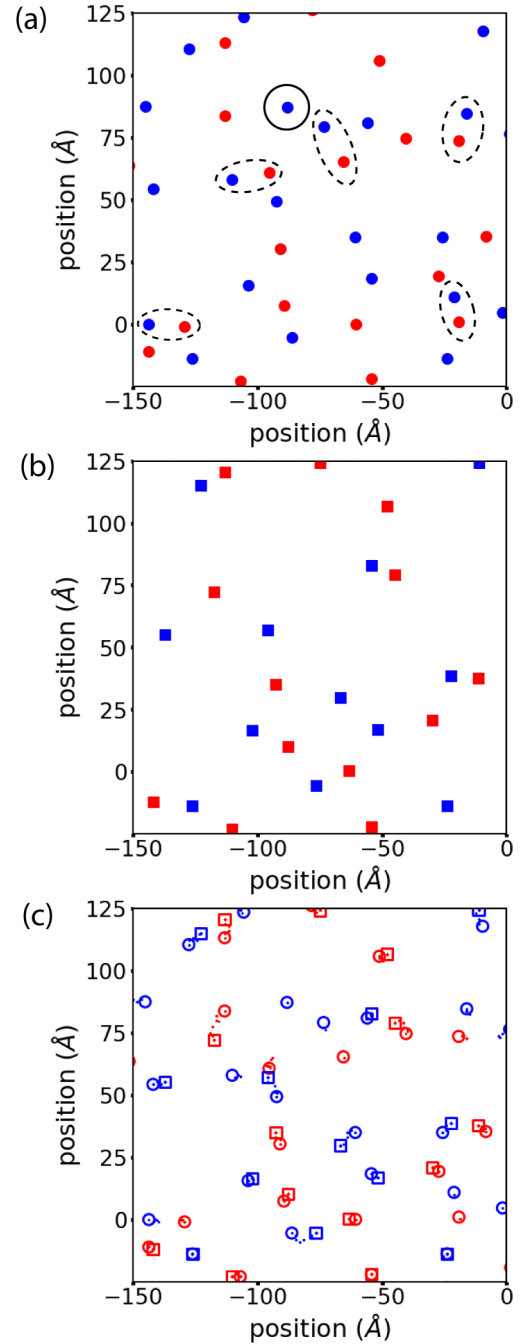


FIG. 4. (a) Positions of the centers of vortices (red) and antivortices (blue) taken from the interior region of a disk with a radius of 500 Å and an initial strain of approximately 0.006 recorded after 10 ps of relaxation time. The dashed ellipses highlight vortex-antivortex pairs that annihilate during the relaxation of the film. The solid circle indicates an antivortex that annihilates on its own. (b) Positions of vortices (red) and antivortices (blue) at the end of the relaxation, 744 ps. Note that the annihilated pairs are no longer present, nor is the isolated circled vortex. (c) A figure showing the trajectories of the vortices during the relaxation. The initial and final positions of the vortices (antivortices) are shown with open circles (squares). Note that many of the vortices and antivortices have only initial positions. This is because they have annihilated during the strain relaxation. The intermediate positions of the vortices (antivortices) are shown as solid red (blue) dots. The snapshots are not taken at equal time intervals; times are chosen to reveal the dynamics.

positions. These displacements include random displacements associated with the high-frequency phonon modes and local rotations. During the relaxation process, the small random displacements are the first to relax as the short-wavelength excitations have the highest relative energy. The local rotations relax more slowly over time. In the steady state, the reaction force vectors are nearly uniform in length, except in proximity to the remaining vortices, antivortices, and domain walls.

Topological defects within 2D fixed magnitude spin systems have been well studied. Isolated defects with winding numbers ± 1 are topologically stabilized; they cannot be removed without altering the configurations of the spins far from the defect [26].

It is likely that topological stability plays a role in stabilizing the vortices and antivortices seen in the reaction force field. However, there is at least one important difference between the reaction force field vectors and fixed magnitude spins. Although the vectors in the reaction force field have typical magnitudes approximately equal to the imposed threshold bond strength (Fig. 1), they are not strictly quantized. The implication is that, if conditions are right, it is possible for one of these topological defects to simply disappear by the shrinking of its force vectors, without the need to annihilate with its antidefect. An example of this is shown in Fig. 4(a) as the circled antivortex.

However, during the relaxation process, vortices more often annihilate antivortices, as also shown in Fig. 4(a). The vortex-antivortex pairs circled by dashed ellipses all annihilate during the strain relaxation. So, topological stability probably plays some role in the stabilization of these reaction force field patterns, and by association, the residual strain, but the structures are not rigorously topologically stable.

The fact that vortex-antivortex pairs arise during the strain relaxation of a thermally expanded film within a Burridge-Knopoff model is interesting in its own right, and hints

at a connection between topological structures and friction. However, we note that the conditions present during strain relaxation differ from those governing macroscopic sliding.

The detailed properties of the vortices and antivortices have not been explored yet. However, their “handedness” makes it likely that they will interact with circularly polarized light. There is some evidence for vortexlike structures emerging from angle-resolved photoemission spectroscopy studies of the band structure of bilayer graphene, but more exploration is needed [27,28]. The energy contributed by the vortices is small (~ 0.1 meV/atom) in this system, which suggests that these patterns could be induced by external fields arising from, for example, polarization skyrmions [29].

In conclusion, the van der Waals epitaxial strain in MoS₂ on amorphous substrates is studied with an atomic-scale model that describes the interaction between 2D materials and the substrate as stick-slip friction. The strain-stabilized state shows that the edge of the sample plays a crucial role in maintaining this strain. This observation aligns remarkably with predictions made from a continuum model. The displacement patterns that emerge include vortices and antivortices are observed in the force and the displacement vector field.

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DATA AVAILABILITY

The data used in the construction of the figures in the main text and the Supplemental Materials are available upon reasonable request from the corresponding author.

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