Near-equilibrium growth from borophene edges on silver

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Two-dimensional boron, borophene, was realized in recent experiments but still lacks an adequate growth theory for guiding its controlled synthesis. Combining ab initio calculations and experimental characterization, we study edges and growth kinetics of borophene on Ag(111). In equilibrium, the borophene edges are distinctly reconstructed with exceptionally low energies, in contrast to those of other two-dimensional materials. Away from equilibrium, sequential docking of boron feeding species to the reconstructed edges tends to extend the given lattice out of numerous polymorphic structures. Furthermore, each edge can grow via multiple energy pathways of atomic row assembly due to variable boron-boron coordination. These pathways reveal different degrees of anisotropic growth kinetics, shaping borophene into diverse elongated hexagonal islands in agreement with experimental observations in terms of morphology as well as edge orientation and periodicity. These results further suggest that ultrathin borophene ribbons can be grown at low temperature and low boron chemical potential.

INTRODUCTION

Borophene, a two-dimensional (2D) boron analog to graphene, is presently attracting interest due to its distinct structural, electronic, and optical properties for many potential applications (1–4). Early theories suggested that borophene is a network of hollow hexagons (HHs) in an otherwise triangular boron lattice (5–8), a characteristic that is first confirmed experimentally in large-sized planar boron clusters (9, 10). A mixture of HHs in a triangular lattice effectively compensates for the electronic deficiency of boron (11) and results in stabilization of borophene. However, borophene is polymorphic because of numerous possible arrangements of HHs in the triangular lattice (12, 13). The HH arrangements can be mediated by metal substrate (14, 15) and carrier density (16), such that borophene is a 2D material that does not have its own fixed lattice structure but depends on external physical conditions. Some borophene phases have been predicted to exhibit phonon-mediated superconductivity (17, 18) as well as Dirac fermions (19, 20) and visible range plasmonics (21). Recently, theoretical predictions of borophene (14, 15) were confirmed (22, 23) by its experimental synthesis on Ag(111). More borophene phases (24–27) and nanostructures (28, 29) were synthesized subsequently on Ag(110), Al(111), and Cu(111) substrates, but to date, these efforts have required molecular beam epitaxy, which is cumbersome and challenging to scale. Consequently, further fundamental and applied studies of borophene growth are needed to develop improved synthesis methods that can achieve large-scale, high-quality samples.

Extensive theoretical studies have been devoted to the growth mechanisms of nearly isotropic 2D materials including graphene (30, 31), hexagonal boron nitride (h-BN) (32), and transition metal dichalcogenides (33). A general picture is that feeding species land on the substrate and diffuse toward edges to nucleate a new atomic row, forming kinks that move via sequential addition of atoms. Such a nucleation–kink flow scenario has explained different experimental shapes for these 2D materials on growth substrates. In contrast, borophene is expressly anisotropic with a richer variety of edges, along which the growth kinetics are likely to be more complicated because of the multicenter characteristics of boron-boron bonding (34). In particular, feeding units of boron have many more possibilities of being organized during the edge growth in view of the polymorphism of borophene. This feature elicits a compelling question as to whether boron growth can proceed or be steered to form a given phase. These issues and new features underscore the importance of advancing the growth theory for anisotropic 2D materials of which borophene is the prototypical example.

Here, we report an atomistic growth mechanism for borophene by combining crystal growth theory with detailed first-principles calculations. First, the borophene edges are identified to be notably reconstructed on Ag(111), with the periodicities of zigzag edges doubled from the intrinsic lattice and edge energies much lower than those of graphene. The growth kinetics are then explored by extending our “nanoreactor” (30) model to the reconstructed edges, at which sequential docking of boron atoms not only drives the edge growth but also seamlessly extends the lattice with no change in its HH network. In contrast to graphene and h-BN, the edge growth can proceed via multiple energy pathways (EPs) with close barriers due to variable boron coordination. All EPs reveal a strong anisotropy of growth kinetics, which yields boron islands as greatly elongated hexagons with varied aspect ratios that agree well with experimental observations in terms of morphology and edge structures. The results also indicate that control of growth temperature and deposition rate could enable growth of ultrathin borophene ribbons. In addition to elucidating pathways toward improved borophene growth, these findings are likely applicable to the growth of other anisotropic 2D materials.

RESULTS AND DISCUSSION

The growth of 2D structures is energetically determined by the competition between bulk and edge energies. Thus, to elucidate the mechanisms of borophene growth, the structures and energy of
borophene edges must be determined for the Ag(111) growth substrate. We mainly focus on the $v_{1/6}$ and triangular sheets since they both have been proposed as the atomic models of recently synthesized borophene (22, 23). Different from graphene and h-BN, the two models are strongly anisotropic with four principal edges, two armchair ($A_1$ and $A_2$) and two zigzag ($Z_1$ and $Z_2$), as illustrated in Fig. 1A and fig. S1.

The most stable structure of each edge is determined by comparing the energy of several enumerated edge structures (figs. S1 and S2). Figure 1B presents the energetically favorable edge structures, which display substantial reconstruction. The lattices at the $A_1$ and $A_2$ edges show decreased HH concentration compared to that in the bulk-like region. The two armchair edges are purely flat and weakly interacting with the substrate, indicative of their high chemical stability. Similarly, flat edges are intrinsic to planar boron molecules (9, 34, 35) and boron sheets with large holes (36). Different behavior occurs at the $Z_1$ and $Z_2$ edges, which follows a zigzag shape but with a larger periodicity than that defined by the original borophene lattice. The boron atoms at sawtooth tips interact substantially with Ag, while all other edge atoms remain rather inert. The edge reconstruction results from a large variability of boron coordination that enables self-passivation of edge dangling bonds.

We calculate the edge energy using $\gamma = \frac{(E_{syst} - E_{sub} - n\mu_B)}{2L}$, where $E_{syst}$ is the energy of the whole system; $E_{sub}$ and $\mu_B$ are the energies of the Ag substrate and chemical potential of boron, respectively; $n$ is the number of boron atoms in the system; and $L$ is the edge length. We adopt $\mu_B$ as the energy per atom of a perfect borophene on Ag(111). Because of the reconstructions, the edges have exceptionally small energies on Ag(111), ranging from 0.06 to 0.18 eV/Å, about one-fifth of those for graphene on the same substrate. Note that one cannot exclude more stable structures for both the $Z_1$ and $Z_2$ edges, if larger edge protrusion and longer periodicity are considered (fig. S2); these larger periods are computationally unaffordable but can be the subject of future work. Similar edge reconstructions are also found for the triangular sheet, but the edge energies are notably higher because of the puckered geometry of the sheet (fig. S3).

On the basis of the energy of principal edges, we obtain the edge energy $\gamma$ for an arbitrary direction $\chi$ with respect to the $Z_1$ direction, through the same analysis developed for graphene. The edge energy $\gamma$ is expressed as (37) $\gamma(\chi) = |\gamma| \cos(\chi + C)$, where $|\gamma| = 2(\gamma_{A_2} + \gamma_{Z_2} - \sqrt{3}\gamma_{A_1}/\gamma_{Z_2})^{1/2}$ and $C = \arctan(\sqrt{3} - 2\gamma_{Z_2}/\gamma_{A_2})$, where $\gamma_{x} (U = A$ and $Z; x = 1$ and 2) represents the edge energy of the principal edges. The subscripted $\gamma_{x}$ can be $Z_1$ and $A_2$ for $0^\circ < \chi \leq 30^\circ$, $Z_2$ and $A_1$ for $30^\circ < \chi \leq 60^\circ$, and $Z_1$ and $A_1$ for $60^\circ < \chi < 90^\circ$. Then, thermodynamic Wulff construction can determine the equilibrium shape of borophene, the one with the minimal interface energy at a fixed area, by plotting $\gamma$ versus the edge direction $\chi$, as shown in Fig. 1C for the $v_{1/6}$ and triangular sheets. The equilibrium shape of the $v_{1/6}$ sheet is a rectangle (chopped by ~5% at its corners by the $Z_2$ edge), while that of the triangular sheet is a rhombus with two sharp corners truncated. Apparently, both shapes do not agree with experimental observations (22). However, the equilibrium shape of the $v_{1/6}$ sheet is closer to experiments than that of the triangular sheet. Moreover, a very recent experiment (27) has essentially excluded the triangular sheet as the model of synthesized borophenes (14). We thus turn to explore the growth kinetics of the $v_{1/6}$ sheet, and further analysis on the triangular sheet is not warranted.

The growth-advancement rate of an entire individual edge depends on the local growth kinetics along the edge. It is thus necessary to calculate the detailed sequence of atom accretion to each edge and then determine the kinetic barrier of growth. To this end, we invoke the idea of step-flow growth of crystals (38), which dictates that each edge moves forward by incorporating feeding species to its active site. The feeding species need to find a preferred state and then diffuse on the substrate. We thus compare the energy and dynamics of three candidate species: monomers, dimers, and trimers. The most stable adsorption site is in the subsurface for the three species: the octahedral site for monomers and the inlay interstitial site within the topmost Ag layer for dimers and trimers (fig. S4). The chemical potentials of boron for the three species at their preferred site are $-4.18$, $-4.89$, and $-5.19$ eV, respectively. The diffusion barriers for monomers and dimers within the subsurface region are 0.26 and 0.59 eV, respectively, indicating their high mobility at experimental growth temperatures of $300^\circ$ to $700^\circ$C. While trimers are more stable than monomers and dimers, trimers also have a significantly higher diffusion barrier. In reality, the entropy contribution at growth conditions further stabilizes monomers and dimers with respect to trimers. The trade-off between stability and mobility renders dimers as the most likely species for feeding growth. We thus take B dimers as the feeding species to compute the growth kinetics of borophene and will consider the effect of monomers later.

First, we add B dimers step by step to the $A_1$ edge. At each growth step, we scan a number of adsorption sites and determine the optimal configuration by comparing the system energy. However, since the configuration of the $n$th step has to be built on that of the $n-1$ step, the low-energy configuration at a given step may not lead to the optimal EP for the edge growth. The optimal EP must be the one with the overall lowest barrier (mathematically, lowest maximum point), not lowest at every step. This is particularly true for borophene in light of the highly variable coordination of boron that results from the multicenter characteristics of its chemical bonds. Therefore, we

![Fig. 1. Borophene edges on silver.](http://advances.sciencemag.org/) Fig. 1. Borophene edges on silver. (A) The $v_{1/6}$ sheet has two armchair edges, denoted as $A_1$ and $A_2$, and two zigzag edges, denoted as $Z_1$ and $Z_2$. (B) Preferred edges of the $v_{1/6}$ sheet on Ag(111) along with calculated edge energies. (C) Equilibrium shapes of the $v_{1/6}$ and triangular sheets. Black and blue lines stand for the $A_1$ and $Z_1$ edges, while black and blue dotted lines stand for the $A_2$ and $Z_2$ edges, respectively.
also examine possible branching of the EPs at each step by adding atoms to the second lowest and even third lowest previous configurations. Figure 2 (A and B) depicts the evolution of atomic configurations along the optimal EPs of sequential addition of B dimers to the A₁ and Z₁ edges. At the A₁ edge, the first added dimer is found to pull out a B atom that originally filled an HH at the edge, forming a deformed HH at the kink site. Then, the addition of the second dimer leads to the nucleation of a new row at the edges, which serves as a base for subsequent attachment of B dimers. For the Z₁ edge, the formation of HHs is also energetically preferred during the growth, yet more atoms are needed to nucleate a new row. The growth kinetics at other principal and screwed edges shows similar behavior (figs. S5 to S9). Along the minimum EPs, complete growth of a new row results in a seamless extension of the v₁/₆ lattice while shifting the reconstructed edges along the growth direction by one lattice constant. This structure “replication” is noteworthy in view of the polymorphic nature of borophene that generally allows a diverse arrangement of HHs.

The configurational spectra at each step of growth at the A₁ and Z₁ edges are shown in Fig. 2C. These results further confirm that the configurations that seamlessly extend the borophene lattice are in low-energy regions, while the topological defects and protrusions are in high-energy regions. Consequently, branching into dendrites is unlikely to occur during the borophene growth, consistent with experimental observations. The evolution of free energy during the edge growth also reveals strong kinetic anisotropy. For example, at a boron chemical potential bias of Δμ = 0.01 eV near the equilibrium, the kinetic barriers in the optimal EPs are 2.07 and 1.78 eV for the Z₁ and Z₂ edges and decrease to 1.1 and 0.6 eV for the A₁ and A₂ edges, respectively (Fig. 3A). The edge growth proceeds mainly via two steps, row nucleation and kink movement. Both the steps consume more than 10 atoms because of the edge reconstruction and structural anisotropy, in stark contrast to the cases of graphene (30) and h-BN (32) growths that need only 2 to 3 atoms. Because of the limitation imposed by the used supercell size, the energy profiles do not display the expected periodic up-down levels corresponding to kink flow. However, the energy barriers should be reliable for differentiating growth kinetics across different edges, as the kinetics are determined at the stage of row nucleation that is well accommodated by our supercells.

Different from the linear dependence of equilibrium shapes on the edge energy, the growth rates at the edges depend exponentially on the kinetic barriers, strongly amplifying the anisotropy (31). Therefore, all the skewed edges (figs. S7 to S9) and the two armchair edges advance quickly via kink flow. In particular, these edges disappear during growth, as confirmed by the slowest growth of zigzag edges. As a result, a borophene island is terminated only with the Z₁ and Z₂ edges. We first consider the case near thermodynamic equilibrium, where we set the boron chemical potential bias to Δμ = 0.01 eV and the growth temperature to kₜ = 0.08 eV (~900 K). The kinetic Wulff construction (30, 39) determines the shape of a crystal based on edge growth rates. By modifying the expression of growth velocity versus lattice orientation γ for graphene edges (30) to account for lattice anisotropy, we determine the borophene shape to be an

\[ G(\text{eV}) = \begin{cases} 
2 & \text{for } 0 < N < 12 \\
3 & \text{for } 12 < N < 32 
\end{cases} \]

\[ k_B T = 0.08 \text{ eV} \]

**Fig. 2. Growth kinetics of borophene on silver.** Energetically optimal atomic configurations during the step-by-step addition of boron dimers to (A) A₁ and (B) Z₁ edges. (C) EPs for the kinetic growth along the (left) A₁ and (right) Z₁ edges, with Δμ₉ = 0 eV. At each step, we scanned a number of candidate structures whose energies are shown by gray bars. The red solid lines present the optimal EP, while the black solid lines show another EP with a slightly higher maximum. The blue dotted lines show the case of a mixture of monomers and dimers as feeding species.
elongated hexagon, enclosed by two opposite $Z_1$ edges and four $Z_2$ edges in between, as shown by the leftmost image of Fig. 3B, in agreement with the experimental borophene islands shown in Fig. 3C. The elongated hexagon for borophene is unique among 2D materials grown on an isotropic substrate. Increasing the chemical potential of boron and the growth temperature both reduce the kinetic anisotropy of edge growth and shift the shape toward a standard hexagon, as exemplified by the cases of $\Delta \mu = 0.05 \text{ eV}$ at $k_5T = 0.08 \text{ eV}$ and $\Delta \mu = 0.01 \text{ eV}$ at $k_5T = 0.1 \text{ eV}$ (Fig. S10).

In addition to the optimal EPs, we also identify alternative EPs for edge growth, which essentially follow the minimum EPs, with deviations arisen mostly in the region around the energy barriers (Figs. 2C and 3A). These EPs give slightly higher growth barriers of 2.21 and 1.93 eV for the $Z_1$ and $Z_2$ edges and 1.54 and 0.68 eV for the $A_1$ and $A_2$ edges, respectively, at $\Delta \mu = 0.01 \text{ eV}$. We anticipate additional EPs with different barriers, but these EPs cannot be fully calculated because of prohibitively high computational cost imposed by the large-scale atomic models. Regardless, the additional computationally accessible EPs confirm the anisotropy of edge growth, but they mediate the competition between growth of $Z_1$ and $Z_2$ edges, ultimately leading to diversity of shapes. As seen in Fig. 3B, the shapes resulting from combinatorics of different barriers at $Z_1$ and $Z_2$ edges remain elongated hexagons yet show different aspect ratios. It is encouraging that these hexagons reproduce the experimentally observed shapes of borophene well, as marked by numbers 1 to 6 in Fig. 3C. Of particular interest are the needle-like ribbon structures that result from the combination of $E_{Z1} = 2.21 \text{ eV}$ and $E_{Z2} = 1.78 \text{ eV}$, which can potentially function as a conductive wire of atomic width for nanoelectronic devices. According to our theory, lowering the growth temperature and chemical potential of boron can enhance the kinetic anisotropy in a manner that will promote the growth of ultranarrow boron nanoribbons. In Fig. 3C, we find that the needle-like islands tend to appear separately from other islands (labeled by number 1), which, to some extent, supports our theory in a sense that the supply of boron for growing the narrow ribbons should be locally limited.

Our experimental islands show additional interesting points. First, as a result of the sixfold symmetry of the Ag(111) surface, the islands are oriented with an expected three-peak distribution (i.e., at $-60^\circ$, $0^\circ$, and $60^\circ$; see Fig. 3D), indicating that these islands share the same registry with the substrate, as adopted in our models. Second, a statistical analysis of the island aspect ratios, shown in Fig. 3E, suggests that the shapes are skewed toward elongation. The islands of the highest frequency have an aspect ratio of $\sim 3.5$, which agrees well with the value ($\sim 4$) determined by the optimal EPs at $\Delta \mu = 0.01 \text{ eV}$ and $k_5T = 0.08 \text{ eV}$. Despite the scattered distribution of aspect ratios, the island areas are relatively centered at $\sim 450 \text{ nm}^2$, which indicates a possible anticorrelation of the growth at the $Z_1$ and $Z_2$ edges; namely, rapid growth of one edge retards growth of the other while keeping the overall growth rate of an island unchanged. The aspect ratio of a specific island thus depends on the relative orientation between the edges and flow of deposited feeding species as well as the local nucleation density.

If boron were provided in the form of mixed monomers and dimers, then the accretion sequence possibilities would be more broadened. One benefit is that some high-energy states during the edge growth can be skipped, resulting in an EP with a lower kinetic barrier for each edge (Fig. 2C). As a result, the growth can be faster and perhaps have higher quality than that with dimers alone. The corresponding barriers for the $Z_1$ and $Z_2$ edges are reduced to 1.84 and 1.2 eV, respectively, shaping the borophene island into an elongated hexagon with an increased aspect ratio (more ribbonlike; Fig. S11). In any case, it is clear that the anisotropic growth kinetics of borophene are rather robust against the variation of feeding species.

While the borophene shapes have been well described by the growth kinetics, it is important to see whether the lattice structure and edge orientation of borophene islands also match the models. To this end, we examine the scanning tunneling microscopy (STM) image of a typical island with an elongated hexagonal shape (Fig. 4A). Atomically resolved images reveal an island in the striped phase reported in a previous work (22), with a rectangular unit cell (Fig. 4A, inset). A comparison of experimental and simulated STM topography images (Fig. 4, A and B) confirms that the $v_{1/6}$ sheet matches key features of the island, especially the lattice constant and the striped pattern. This comparison enables us to deduce that the long edge of the island is oriented along the HH rows, i.e., the direction of the $Z_1$ edge in our model (Fig. 4B), and the four short edges are oriented along the $Z_2$ edge. Moreover, the measured lattice periodicity based on the derivative of the STM topography is
0.6 Å for the Z₁ edge (Fig. 4C), again in agreement with the value of 0.58 Å predicted by the theoretical model (Fig. 4D). Similar agreement between theory and experiment is also obtained for the Z₂ edge.

CONCLUSIONS
On the basis of ab initio calculations and experimental characterization, we have identified the edges of borophene on silver and elucidated the growth mechanism including the island shape and morphology of the edges. Borophene is revealed to have significant edge reconstructions that result in low edge energy and doubled periodicities of its intrinsic lattice. The thermodynamic equilibrium shape is predicted to be approximately rectangular yet is absent in experimental observation. However, away from equilibrium, we determine the sequence of atomic row assembly at the edges, which not only moves the edges along the growth direction but also seamlessly extends the original borophene lattice. The growth kinetics follow multiple EPs with close barriers along each edge but with strong anisotropy across different edges. Consequently, the kinetic equilibrium shapes of borophene turn out to be a series of elongated hexagons that reproduce the shapes observed experimentally. We further show that the condition of low growth temperature and low boron chemical potential is likely to enable selective growth of ultrathin boron nanoribbons. Overall, these results provide the first atomistic understanding for the growth of borophene, which is likely to inform the high-quality synthesis of other highly anisotropic 2D materials.

MATERIALS AND METHODS
Theory and models
Our model systems consist of borophene nanostructures overlaid on Ag(111) of three atomic layers. The calculations were performed within the framework of density functional theory using a projector augmented wave method for the potential at the core region and generalized gradient approximation of the Perdew-Burke-Ernzerhof functional as implemented in the Vienna Ab initio Simulation Package code (40). We adopt a kinetic energy cutoff of 400 eV for the plane-wave expansion and set the vacuum region to 15 Å to isolate neighboring periodic images. The Brillouin zone was sampled at the Γ point only. Borophene nanoribbons were overlaid on Ag(111) to calculate energies of edges and of atom accretion therein. For the latter, the atoms at the nongrowth edge of nanoribbons were fixed to the relaxed positions before growth. The positions of other boron atoms plus metal atoms of the top two layers were relaxed using the conjugate gradient method until the force on each atom was less than 0.01 eV/Å.

Growth and STM of borophene
Borophene samples were grown on Ag(111) single-crystal substrates (99.999%, <0.01° miscut; MaTecK) by electron beam evaporation of a boron rod (99.999%; ESPI Metals) under ultrahigh vacuum. The substrate was prepared by cycles of Ar ion sputtering and annealing to 550°C. Samples were grown at 550°C, at a boron flux of ~0.05 monolayers/min. After growth, samples were cooled quickly to room temperature by immediate heater shutoff and placement of the sample on a room temperature heat sink. STM images were acquired in Omicron STM/atomic force microscopy systems, at temperatures of 55 and 4 K, respectively, using electrochemically etched W tips.

SUPPLEMENTARY MATERIALS
Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/5/9/eaax0246/DC1

Fig. 4. Edges and lattices of borophene islands. (A) STM topography image of a borophene island (\( V_{\text{sample}} = 0.1 \text{ V} \) and \( I_{\text{t}} = 1.0 \text{ nA} \)). Inset: Zoomed-in image of the area marked by the black rectangle. (B) Simulated STM image of the v\( \frac{1}{6} \) sheet on silver, overlaid with an atomic model (\( V_{\text{sample}} = 1.0 \text{ V} \)). The blue arrow indicates the Z₁ direction. (C) Top: STM topography image of the Z₁ and Z₂ edges of a typical borophene island (\( V_{\text{sample}} = 2.0 \text{ V} \) and \( I_{\text{t}} = 100 \text{ pA} \)). The lattice periodicities of the two edges are measured by x (middle) and y (bottom) derivatives of the STM topography. (D) Schematic model of a v\( \frac{1}{6} \) sheet island on Ag(111) along with calculated edge periodicities.


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