

Schwoebel-Ehrlich barrier: from two to three dimensions

S. J. Liu, Hanchen Huang,^{a)} and C. H. Woo

Department of Mechanical Engineering, The Hong Kong Polytechnic University, Hong Kong

(Received 22 January 2002; accepted for publication 4 March 2002)

The Schwoebel-Ehrlich barrier—the additional barrier for an adatom to diffuse down a surface step—dictates the growth modes of thin films. The conventional concept of this barrier is two dimensional (2D), with the surface step being one monolayer. We propose the concept of a three-dimensional (3D) Schwoebel-Ehrlich barrier, and identify the 2D to 3D transition, taking aluminum as a prototype and using the molecular statics method. Our results show that: (1) substantial differences exist between the 2D and 3D barriers; (2) the transition completes in four monolayers; and (3) there is a major disparity in the 3D barriers between two facets; further, alteration of this disparity using surfactants can lead to the dominance of surface facet against thermodynamics. © 2002 American Institute of Physics. [DOI: 10.1063/1.1475774]

Texture has been a recognized factor that controls the performance of thin films. For example, the $\langle 111 \rangle$ texture of aluminum interconnects in integrated circuits dictates their resistance to electromigration,¹ and $\langle 100 \rangle$ texture of TiN is preferred in mechanical coating.^{2,3} The Schwoebel-Ehrlich barrier is a key factor in surface processing.^{4–22} In our previous studies, it has been demonstrated that the dominance of $\langle 111 \rangle$ texture is a result of two-dimensional growth at initial stage, that is the formation of large $\{111\}$ facets.^{23,24} The large facet is the direct consequence of small adatom migration barrier and nearly zero conventional—hereafter referred to as the two-dimensional (2D)—Schwoebel-Ehrlich barrier in aluminum. Our ensuing studies show that the 2D Schwoebel-Ehrlich barrier is also very small for dimers and trimers diffusing down a $\{111\}$ facet in aluminum.²⁵ On the other hand, both experiments²⁶ and Wulff construction show that two large facets meet each other and form a ridge, as shown in Fig. 1. The intersection of two large facets is also common under normal deposition conditions; when deposition rate is not too high or substrate temperature is not too low.²³ For exchange of atoms between two such facets, an adatom has to cross over the ridge, effectively diffusing down a surface step of multiple layers; the Schwoebel-Ehrlich barrier is therefore 3D. During a growth process, a facet may not be so flat, and surface steps can be of one, two, or many layers. As a result, the Schwoebel-Ehrlich barrier experiences a gradual transition from 2D to 3D. For clarity, we define the Schwoebel-Ehrlich barrier as the *total* energy barrier over a step or a ridge; in contrast to the extra energy barrier when only a 2D case is considered.

The molecular statics method is described in detail in reference,²⁵ and will be briefly summarized here. A simulation cell with a flat surface, say $\{111\}$, $\{110\}$, or $\{100\}$, is first chosen. An island of multiple layers is introduced on top of the flat surface. The island is constructed so that the top surface is parallel to the substrate, and the side surfaces consist of $\{100\}$, $\{110\}$, and $\{111\}$ facets. A typical simulation cell

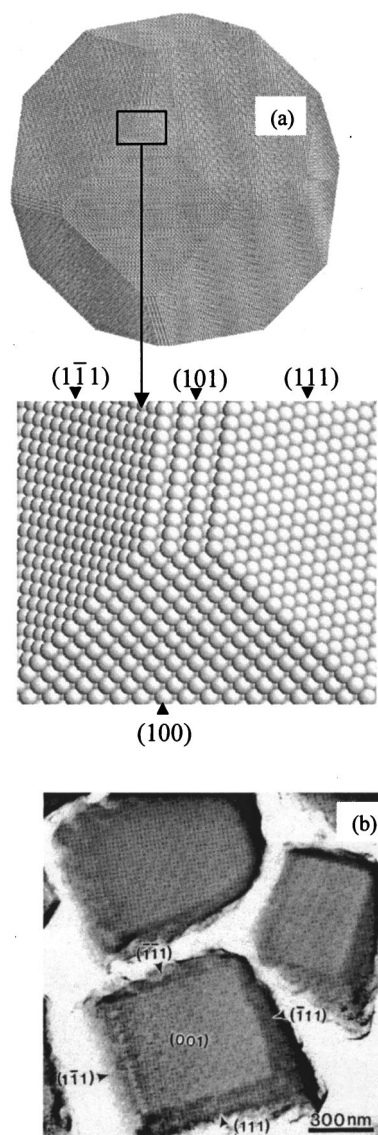


FIG. 1. (a) A Wulff construction of aluminum, and (b) an electron microscopy of aluminum thin-film surface after high-temperature annealing, with various surface orientation labeled.²⁶

^{a)}Corresponding author; electronic mail: Hanchen.Huang@polyu.edu.hk (and hanchen@rpi.edu after summer 2002)

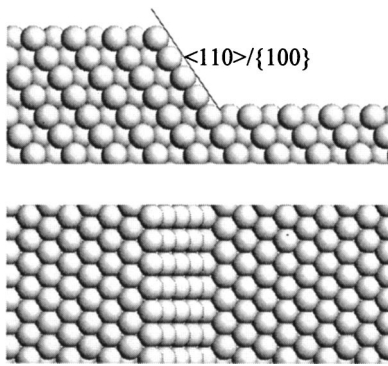


FIG. 2. Side (upper section) and top (lower section) view of a step along $\langle 110 \rangle$ of multiple layers forming a $\{100\}$ surface; the two horizontal surfaces bounding the step are of $\{111\}$.

of aluminum is shown in Fig. 2. Atoms in the bottom region of the simulation cell are fixed to their perfect lattice positions to mimic a semi-infinite large surface. The convergence of numerical results is tested against the size of the simulation cell, and the simulation cell is chosen so that the energy calculations are reliable up to ± 0.01 eV. It is worth mentioning that molecular statics, instead of the molecular dynamics method, is used because of the extremely small energy barrier of aluminum adatom on $\{111\}$ -0.04 eV.^{25,27} For such a small migration barrier, adatoms diffuse away from the desired configurations even at low temperatures.

The transition from the 2D to 3D Schwoebel-Ehrlich barrier is demonstrated in Fig. 3 for the case of an adatom diffusing from one $\{111\}$ to another $\{111\}$ facet, across a step along $\langle 110 \rangle$; thereafter referred to as diffusion from $\{111\}$ facet to $\langle 110 \rangle / \{111\}$ step/facet. Table I provides a complete set of the calculation results. Since direct hopping is consistently more difficult, only the barriers by exchange mechanism are listed. According to our definition, the 3D Schwoebel-Ehrlich barriers from facet A to facet B and from B to A are different. For example, Table I gives the 3D Schwoebel-Ehrlich barrier from $\{111\}$ to $\{100\}$ facet to be 0.30 eV, and that from $\{100\}$ to $\{111\}$ facet to be 0.68 eV—the difference being from adatom formation energies on the two facets. This difference will lead to the imbalance of adatom flux between the two facets, and thereby the dominance of $\{111\}$ over $\{100\}$. There is little ambiguity in the definition of the 3D Schwoebel-Ehrlich barrier. However, the definition of the Schwoebel-Ehrlich barrier for a step of multiple layers deserves a clarification. The flat surfaces bounding the step

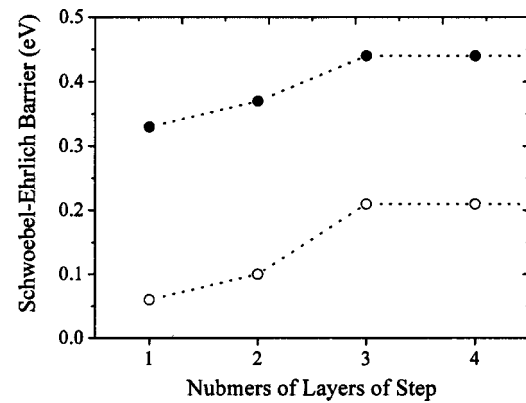


FIG. 3. Transition from the 2D to 3D Schwoebel-Ehrlich barrier of an adatom from one $\{111\}$ to another $\{111\}$ across a $\langle 110 \rangle$ step, by exchange (open circle) and direct-hopping (solid circle) mechanisms.

of multiple layers are parallel. For example, in the calculation of transition barriers from $\{111\}$ to $\{100\}$, the two bounding surfaces are of $\{111\}$ and the small facet in between is of $\{100\}$. The results show that: (1) the transition from 2D to 3D completes in four monolayers; (2) the 3D barrier can be substantially larger than its 2D counterpart; and (3) there is a major disparity of 3D barriers between two flat surfaces, such as $\{100\}$ and $\{111\}$. Accompanying this disparity, the transition from 2D to 3D can also be different for the two facets. For example, the barrier from $\{111\}$ to $\{100\}$ is insensitive to step thickness. However, the barrier from $\{100\}$ to $\{111\}$ increases substantially with the step thickness. This indicates that $\{111\}$ facets dominate more easily once they grow beyond a critical size, because the flow of adatoms from $\{100\}$ to $\{111\}$ is more difficult beyond this size.

It is interesting to discuss technological impacts of this concept of the 3D Schwoebel-Ehrlich barrier in materials processing. If one could modify this barrier, in particular, reverse the disparity, then thermodynamic faceting of thin films may be reversed. Indeed, our Monte Carlo simulations in a preliminary report have demonstrated²⁸ that this reversal is possible when surfactants are used. It is even more encouraging that our two recent and independent experiments, one using antimony as surfactant during silver film deposition, and another indium as surfactant, have shown the facet conversion.

The work described in this letter was substantially supported by grants from the Research Grants Council of the

TABLE I. Schwoebel-Ehrlich barrier (eV) as a function of number of layers of each step, for various step orientations and facets.

Initial Facet	Step Orientation/ Final Facet	Number of Layers				
		i	ii	iii	iv	Multi-
{111}	$\langle 110 \rangle / \{100\}$	0.30	0.31	0.30	0.30	0.30
	$\langle 110 \rangle / \{110\}$	0.06	0.02	0.04	0.04	0.04
	$\langle 110 \rangle / \{111\}$	0.06	0.10	0.21	0.21	0.21
{100}	$\langle 100 \rangle / \{100\}$	0.35	0.17	0.25	0.25	0.25
	$\langle 100 \rangle / \{110\}$	0.34	0.34	0.35	0.35	0.35
	$\langle 110 \rangle / \{111\}$	0.45	0.55	0.68	0.68	0.68
{110}	$\langle 100 \rangle / \{100\}$	0.63	0.68	0.70	0.72	0.72
	$\langle 111 \rangle / \{110\}$	0.33	0.38	0.44	0.47	0.47
	$\langle 110 \rangle / \{111\}$	0.70	0.78	0.81	0.83	0.83

Hong Kong Special Administrative Region (PolyU 1/99C, 5146/99E, 5152/00E, and 5161/01E), and partially by a central research grant from the Hong Kong PolyU (G-V943).

- ¹S. Vaidya and A. K. Sinha, *Thin Solid Films* **75**, 523 (1981).
- ²J. Greene, J. Sundgren, L. Hultman, I. Petrov, and D. Bergstrom, *Appl. Phys. Lett.* **67**, 2928 (1995).
- ³H. Onoda, M. Kageyama, and K. Hashimoto, *J. Appl. Phys.* **77**, 885 (1995).
- ⁴R. L. Schwoebel and E. J. Shipsey, *J. Appl. Phys.* **37**, 3682 (1966).
- ⁵G. Ehrlich and F. G. Hudda, *J. Chem. Phys.* **44**, 1039 (1966).
- ⁶G. Ehrlich, *Surf. Sci.* **299/300**, 628 (1994).
- ⁷A. Götzhäuser and G. Ehrlich, *Phys. Rev. Lett.* **77**, 1334 (1996).
- ⁸S. C. Wang and G. Ehrlich, *Phys. Rev. Lett.* **79**, 4234 (1997).
- ⁹K. Kyuno and G. Ehrlich, *Phys. Rev. Lett.* **84**, 2658 (2000).
- ¹⁰Z. Zhang and M. G. Lagally, *Phys. Rev. Lett.* **72**, 693 (1994).
- ¹¹Z. Zhang and M. G. Lagally, *Science* **276**, 377 (1997).
- ¹²D. Kandel, *Phys. Rev. Lett.* **78**, 499 (1997).
- ¹³D. Kandel and E. Kaxiras, *Phys. Rev. Lett.* **75**, 2742 (1995).
- ¹⁴S. Esch, M. Hohage, T. Michely, and G. Comsa, *Phys. Rev. Lett.* **72**, 518 (1994).
- ¹⁵H. A. van der Vegt, M. Breeman, S. Ferrer, V. H. Etgens, X. Torrelles, P. Fajardo, and E. Vlieg, *Phys. Rev. B* **51**, 14 806 (1995).
- ¹⁶J. Jacobsen, K. W. Jacobsen, P. Stoltze, and K. Nørskov, *Phys. Rev. Lett.* **74**, 2295 (1995).
- ¹⁷S. Kodiyalam, K. E. Khor, and S. D. Sarma, *Phys. Rev. B* **53**, 9913 (1996).
- ¹⁸P. J. Feibelman, *Phys. Rev. Lett.* **81**, 168 (1998).
- ¹⁹U. Kurpick and T. S. Rahman, *Phys. Rev. B* **57**, 2482 (1998).
- ²⁰M. V. Ramana Murty and B. H. Cooper, *Phys. Rev. Lett.* **83**, 352 (1999).
- ²¹J. Rottler and P. Maass, *Phys. Rev. Lett.* **83**, 3490 (1999).
- ²²S. Schinzer, S. Koehler, and G. Reents, *Eur. Phys. J. B* **15**, 161 (2000).
- ²³H. Huang, G. H. Gilmer, and T. Diaz de la Rubia, *J. Appl. Phys.* **84**, 3636 (1998).
- ²⁴G. H. Gilmer, H. Huang, T. Diaz de la Rubia, J. D. Torre, and F. Barumann, *Thin Solid Films* **365**, 189 (1999).
- ²⁵M. Bockstedte, S. J. Liu, O. Pankratov, C. H. Woo, and H. Huang, *Comput. Mater. Sci.* (to be published).
- ²⁶L. Hsiung (private communication).
- ²⁷R. Stumpf and M. Scheffler, *Phys. Rev. Lett.* **72**, 254 (1994); *Phys. Rev. B* **53**, 4958 (1996).
- ²⁸S. J. Liu, E. G. Wang, C. H. Woo, and H. Huang, *J. Computer-aided Mater. Design* **7**, 195 (2001).