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# Electric forces on a confined *advacancy* island

F. Leroy,<sup>1,\*</sup> A. El Barraj,<sup>1</sup> F. Cheynis,<sup>1</sup> P. Muller,<sup>1</sup> and S. Curiotto<sup>1</sup>

<sup>1</sup>*Aix Marseille Univ, CNRS, CINAM, Marseille, France*

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The passage of an electric current in a material can cause a biased mass transport at its surface. This migration phenomenon is intimately related to the microscopic details of atomic processes of diffusion and attachment/detachment at step edges. Using low energy electron microscopy we have examined *in operando* under an electric current the migration of Si(111)- $1\times 1$  monoatomic *advacancy* islands confined on Si(111)- $7\times 7$  terraces. The islands move opposite to the current direction, with velocity increasing with the radius. We show that the kinetics of attachment/detachment of atoms at step edges and the migration of *adatoms* on terraces are in competition. The effective valence of Si *adatoms* is  $3.4\pm 0.6$  and the kinetic length of attachment-detachment is about 500 nm. The analysis of the islands shape reveals that the electric current biases significantly the kinetic rate of mass transfers at step edges modifying the overall island shape.

Atomic steps are the most abundant structures at surfaces and play a key role in mass transfers. They are involved in complex atomic mechanisms such as the attachment-detachment of atoms or the diffusion along the step edge [1 and 2]. These elementary mechanisms may occur simultaneously and their experimental study is all the more difficult that the steps are in competition as they share a common *adatom* bath. To study the mass transport mechanisms, different experimental strategies have been carried out based on the spatio-temporal fluctuations of the position of isolated/interacting steps [3–5] or on the step displacement velocity when a driving force intervenes [6–8]. In particular the electromigration force, even though it is extremely weak, is known to bias the diffusion of mobile *adatoms* [9–13]. It may also modify the atomic steps properties such as the local *adatom* equilibrium concentration and/or the kinetic coefficients of attachment-detachment [5, 14, and 15]. These effects arise since the force felt by atoms depends on their local environment that differs at step edges or kink sites. The electromigration force can cause substantial changes in the surface morphology such as step bunching for vicinal surfaces [16–19] or shape instabilities of 2D islands [20–23]. The experimental determination of the underlying elementary mechanisms of mass transport at surfaces and the impact of electromigration on these mechanisms appeal for dedicated studies.

In this letter we analyze quantitatively the atomic mechanisms of mass transport on Si(111) under an electric bias by addressing precisely the boundary conditions to disentangle all the contributions. In that purpose we have met two essential conditions: (1) A monoatomic height island where atomic displacements occur at the interior of a confined 2D space closed by a step edge; and (2) a driving force induced by an electric current to move the island. By adjusting the perimeter of the island and measuring its drift velocity induced by an electric current we show a transition from a kinetics of mass transfer limited by attachment-detachment of atoms at step edges to a kinetics limited by terrace diffusion. We deduce that the kinetic length for attachment/detachment is  $d \sim 500$  nm and the effective valence  $Z^*$  of the Si *adatoms* at the

surface is  $3.4\pm 0.6$ . Importantly our detailed analysis of the stationary shape of the electromigrating *advacancy* islands is consistent with a strong modification of the kinetic rate of attachment/detachment at step edges induced by the electric current.

The experiments were performed in an ultra high vacuum (UHV) setup equipped with a low energy electron microscope (LEEM III, Elmitec GmbH) [24]. Si(111) substrates (n or p-doped,  $\rho=1 \Omega\text{cm}$ ) were cut into pieces of  $15\times 3\times 0.5 \text{ mm}^3$ , cleaned with acetone and ethanol before introduction in UHV. An electric current is applied through the sample *via* two Mo electrodes clamped to its extremities. The samples were degassed in UHV for several hours at about 1100 K and then flashed above 1500 K for a few seconds by direct current heating. *Advacancy* islands are created by Si sublimation in the middle of large terraces [25]. The surface evolution under electromigration is studied by LEEM in bright field mode, with an electron beam energy of 3 eV. To change the *advacancy* islands size, Si was deposited *in situ* by a homemade direct current evaporator made of a piece of Si wafer clamped between Mo electrodes.

LEEM images in Fig. 1(a) show the time evolution of the Si(111) surface while crossing the  $1\times 1 \rightarrow 7\times 7$  phase transition temperature (1133 K). The low temperature  $7\times 7$  surface reconstruction nucleates at the step edges on the upper terraces [26] and appears as bright lines (Fig. 1(a)-i). Upon slow cooling, by decreasing the electric current, the  $7\times 7$  phase extends onto the terraces (Fig. 1(a)-ii). Since the crystallographic arrangement of the different  $7\times 7$  domains does not necessarily coincide,  $1\times 1$  out-of-phase boundaries persist at their intersections. Moreover the nucleation of the  $7\times 7$  phase is hindered at the lower step edges and on terraces [7], therefore the *advacancy* island in the middle of Fig. 1(a)-iii stays in a metastable supercooled  $1\times 1$  state [27]. This effect was originally described as a hysteresis of the  $1\times 1 \leftrightarrow 7\times 7$  phase transition temperature [28]. Interestingly this *advacancy* island migrates in the direction opposite to the electric current (Fig. 1(a)-iii-iv). During the displacement, the out-of-phase boundaries attached at the rear of the island merge from time to time and/or spon-

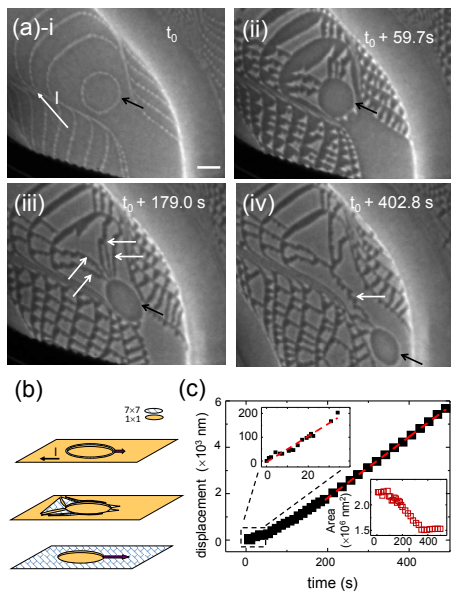


FIG. 1. (a) Sequence of LEEM images during the  $1\times 1 \rightarrow 7\times 7$  phase transition and under electric heating (see complete movie S1 in the supplementary materials). (i) Nucleation of  $7\times 7$  surface reconstruction at the step edges on the upper terraces. (ii) Spreading of the  $7\times 7$  onto the terraces except in the *advacancy* island (black arrow) where the  $7\times 7$  nucleation is hindered. (iii) Formation of  $1\times 1$  out-of-phase boundaries at the  $7\times 7$  domain intersections (white arrows). (iv) Migration of the *advacancy* island in the  $\langle 1\bar{1}2 \rangle$  direction, opposite to the electric current. Out-of-phase boundaries merging at the rear side of the island. Electron energy  $E=3$  eV. Scale bar  $1\mu\text{m}$ . (b) Scheme of the surface evolution under slow cooling. (c) Time evolution of the displacement of the *advacancy* island (black square). The steady velocity is  $13.1\pm 0.1$   $\text{nm}\cdot\text{s}^{-1}$  (velocity in (a)-i is  $5.4\pm 0.3$   $\text{nm}\cdot\text{s}^{-1}$ , see top inset) and the area is  $1.5\pm 0.1$   $10^6$   $\text{nm}^2$  (see bottom inset).

90 taneously detach. The velocity of the *advacancy* island  
 91 increases up to  $13.1\pm 0.1$   $\text{nm}\cdot\text{s}^{-1}$  and reaches a station-  
 92 ary value when the  $7\times 7$  phase significantly covers the  
 93 surrounding surface. Simultaneously, after an initial size  
 94 reduction due to mass transfers with the exterior, the is-  
 95 land size reaches also a steady state. Mass transfers have  
 96 two contributions: The Gibbs-Thompson effect favors the  
 97 capture of *adatoms* as the *advacancy* island curvature is  
 98 locally the largest one (in absolute). The phase transi-  
 99 tion expels the excess atoms of the  $1\times 1$  that diffuse to  
 100 the step edge [29]. The fact that the island area stabi-  
 101 lizes indicates that mass transfers from the exterior are  
 102 nearly entirely suppressed when the  $7\times 7$  covers most of  
 103 the surface. This diffusion barrier effect [30] is due to the  
 104 large surface diffusivity of Si *adatoms* on the  $1\times 1$  with  
 105 respect to the  $7\times 7$  (ratio  $\sim 20$ , [7]). During its displace-  
 106 ment the *advacancy* island can reach a step edge or a de-  
 107 fect that may induce the nucleation of the  $7\times 7$  inside the  
 108 island. To prevent this process from occurring the elec-  
 109 tric current direction is regularly reversed to change the  
 110 drift direction by electromigration while keeping a con-

111 stant temperature ( $\pm 1$  K). The islands move back and  
 112 forth over a distance larger than  $10\mu\text{m}$  on extended ter-  
 113 races without meeting any surface defect or step (Fig.  
 114 2(b)). Concomitantly this process allows for the disap-  
 115 pearance of all  $1\times 1$  out-of-phase boundaries attached to  
 116 the islands by merging and detachment from the rear  
 117 side and by removal at the front side. Let us note that  
 118 a few out-of-phase boundaries have barely no effect on  
 119 the measured velocity but their removal is important to  
 120 determine the stationary shape of the *advacancy* island  
 121 without ambiguity.

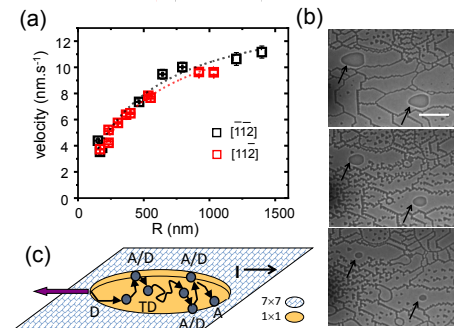


FIG. 2. (a) *Advacancy* island velocity versus radius. The islands electromigrate in the  $\langle 1\bar{1}2 \rangle$  (black square) and  $\langle 11\bar{2} \rangle$  (red square) directions. Fit of the velocity (dotted lines). (b) LEEM images of islands of different sizes electromigrating in the  $\langle 11\bar{2} \rangle$  direction (scale bar  $5\mu\text{m}$ , see complete movie S2 in the supplementary materials). (c) Scheme of mass transfer process: detachment of atoms (D), biased terrace diffusion (TD) and attachment (A).

122 To address the mass transport mechanisms that are  
 123 occurring inside the *advacancy* islands under electromi-  
 124 gration we have studied the size-dependence of the island  
 125 velocity in the stationary regime. Figure 2(a) shows that  
 126 the velocity increases with the island effective radius  $R$   
 127 ( $R = \sqrt{A}/\pi$  where  $A$  is the island area). Pierre-Louis *et*  
 128 *al.* [20] have analyzed the island velocity in the frame-  
 129 work of the linear response theory with weak electromi-  
 130 gration. Considering a kinetics of mass transport by at-  
 131 tachment (A), detachment (D) and terrace diffusion (TD)  
 132 inside the  $1\times 1$  *advacancy* island (see Fig. 2(c)), and ne-  
 133 glecting the *adatom* flux from the upper terrace ( $7\times 7$ ),  
 134 the island drift velocity resulting from these processes is  
 135 [20]:

$$V_{isl} = c_{eq}v_{1\times 1} \frac{R}{R+d} \quad (1)$$

138 where  $c_{eq}$  is the equilibrium surface concentration of mo-  
 139 bile *adatoms*,  $v_{1\times 1}$  is the *adatoms* velocity on the ( $1\times 1$ )  
 140 terrace and  $d = D_{1\times 1}/k$  is the kinetic length of attach-  
 141 ment/detachment and is defined as the ratio of the sur-  
 142 face diffusion coefficient  $D_{1\times 1}$  to the rate  $k$  of *adatom*  
 143 attachment to the step from the terrace. The fit of the  
 144 experimental plots give two key parameters  $c_{eq}v_{1\times 1}$  and  
 145  $d$ . The first term is deduced from the asymptotic veloc-  
 146 ity at large radius ( $15\pm 1$   $\text{nm}\cdot\text{s}^{-1}$ ) and is only related to

147 terrace diffusion of the electromigrating *adatoms*. Con-  
 148 sidering that  $c_{eq} \sim 0.2$  ML [29] and correcting the veloc-  
 149 ity with advection [31] (sweeping effect on the *adatoms*  
 150 due to the step motion) we get the *adatom* velocity  
 151  $v_{1 \times 1} = 135 \pm 9$  nm.s<sup>-1</sup> on the  $1 \times 1$  surface reconstruc-  
 152 tion at the phase transition temperature. This velocity  
 153 derives from the Einstein relation  $v_{1 \times 1} = \frac{D_{1 \times 1}}{k_B T} F$  where  
 154  $k_B$  is the Boltzmann constant,  $T$  the temperature and  
 155  $F = Z^* e E$  is the electromigration force. Therefore the  
 156 effective charge  $Z^*$  of Si *adatoms* can be obtained if the  
 157 diffusion coefficient  $D_{1 \times 1}$  is known. Hibino *et al.* have  
 158 found  $D_{1 \times 1} c_{eq} = 3.0 \cdot 10^7$  s<sup>-1</sup> [7 and 27] at the phase  
 159 transition temperature. Pang *et al.* have obtained by  
 160 different approaches  $D_{1 \times 1} c_{eq} = 2.0 \pm 0.2 \cdot 10^7$  s<sup>-1</sup> [8] in  
 161 a slightly higher temperature regime (1163 K). Consid-  
 162 ering an average value for  $D_{1 \times 1}$  we can deduce the only  
 163 free parameter, *i.e.* the effective charge of Si *adatoms*  
 164  $Z^* = 3.4 \pm 0.6$  ( $E = 490$  V.m<sup>-1</sup>, atomic area: 0.064  
 165 nm<sup>2</sup>). The deduced value of  $Z^*$  is larger by one order of  
 166 magnitude with earlier reports [32 and 33] except for [34]  
 167 ( $Z^* > 1.3$ ). The second term that is deduced from the  
 168 fit is the kinetic length of attachment/detachment  $d$ . We  
 169 obtain  $d_{\langle 11\bar{2} \rangle} = 450 \pm 100$  nm and  $d_{\langle \bar{1}\bar{1}2 \rangle} = 500 \pm 30$  nm  
 170 respectively for an island displacement in the  $\langle 11\bar{2} \rangle$  and  
 171 the  $\langle \bar{1}\bar{1}2 \rangle$  directions. These large values are comparable  
 172 with previous measurements of the kinetic length of at-  
 173 tachment/detachment on the  $7 \times 7$  [35]. To explain this  
 174 large kinetic length we can note that the step advance  
 175 needs to build  $7 \times 7$  unit cells. A complex ordering is  
 176 necessary with potentially concerted events and this pro-  
 177 cess could have a rather low frequency of occurrence [36  
 178 and 37]. From the evaluation of  $d$  we can estimate the  
 179 rate of attachment/detachment at a step edge per atomic  
 180 site  $kc_{eq}a = c_{eq}D_{1 \times 1}a/d \sim 1.9 \times 10^4$  s<sup>-1</sup> ( $a=0.384$  nm is  
 181 the lattice parameter). It is also instructive to estimate  
 182 the average macroscopic time for *adatoms* to detach from  
 183 the front side, cross the island and attach at the rear side.  
 184 The traveling time across the terrace by diffusion is about  
 185  $t_d \sim 2R/v_{1 \times 1}$  and for a typical island of 1  $\mu$ m radius  $t_d \sim$   
 186 15 s. As a comparative time scale, the delay time for  
 187 an atom to make all attachment/detachment processes  
 188 to cross the island is about  $t_{AD} \sim 2d/v_{1 \times 1} \sim 6$  to 8  
 189 s. This indicates that many events of (re)-attachment-  
 190 detachment occur during this traveling ( $kc_{eq}a \times t_{AD} \sim$   
 191  $10^5$ , see Fig.2(c)).

193 In addition to velocity, the *advacancy* island shape in  
 194 the stationary regime is measured and depends both on  
 195 the island size and electric current direction. In Fig. 3(a),  
 196 the *advacancy* islands have a faceted front and an overall  
 197 triangular shape when they migrate in the  $\langle 11\bar{2} \rangle$  direction  
 198 whereas they have a lozenge shape (elongated head and  
 199 lateral facets) in the opposite direction. If they move  
 200 in the  $\langle \bar{1}\bar{1}0 \rangle$  direction the shape is no more symmetric  
 201 (Fig. 3(d)). In all cases the shape is elongated in the  
 202 migration direction. This elongation increases with the  
 203 island size and tends to be circular for small sizes (the  
 204 typical crossover is about the attachment-detachment ki-  
 205 netic length  $d$ ). To describe the island shape, we use the

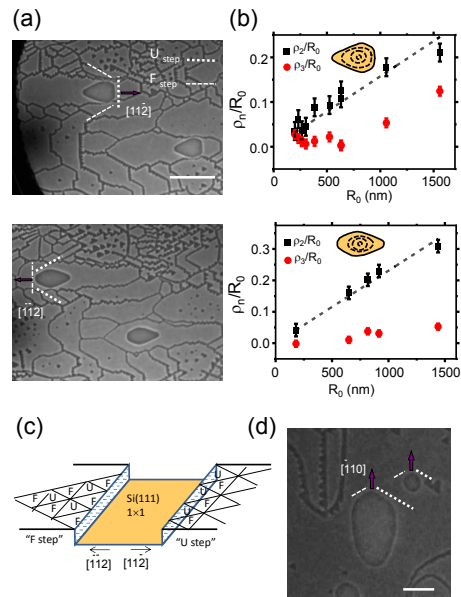


FIG. 3. (a) LEEM images of *advacancy* islands electromi-  
 grating in the  $\langle 11\bar{2} \rangle$  (top) and the  $\langle \bar{1}\bar{1}2 \rangle$  (bottom) directions  
 (scale bar 5  $\mu$ m). The shape is respectively a triangle and a  
 lozenge. Unfaulted steps ( $U_{step}$ ) are shown as dotted lines and  
 faulted steps ( $F_{step}$ ) as dashed lines [38]. (b) Fourier coeffi-  
 cients of the island shape as function of island radius for both  
 directions. (c) Scheme of the  $U$  and  $F$  step edge structure.  
 (d) LEEM image of two *advacancy* islands electromigrating  
 in the  $\langle \bar{1}\bar{1}0 \rangle$  direction. The shape is asymmetric (scale bar 1  
 $\mu$ m, see complete movie S3 in the supplementary materials).

206 polar coordinates  $R(\theta) = R_0 + \rho(\theta)$  ( $R_0$  is the mean ra-  
 207 dius), and we apply the Fourier series expansion of  $\rho(\theta)$ :

$$\rho(\theta) = \sum_{n \geq 2}^{\infty} \rho_n \cos(n\theta) + \nu_n \sin(n\theta) \quad (2)$$

208 where  $\rho_n$  and  $\nu_n$  are the Fourier coefficients ( $\nu_n = 0$  for  
 209 symmetric islands and  $\rho_1$  and  $\nu_1$  are not considered be-  
 210 cause they correspond to a simple shape translation). In  
 211 Fig. 3(b) are plotted the normalized Fourier coefficients  
 212  $\rho_n/R_0$  as function of the island radius  $R_0$  when the is-  
 213 lands are migrating along the  $\langle 11\bar{2} \rangle$  and  $\langle \bar{1}\bar{1}2 \rangle$  directions.  
 214 The main term of elongation is the  $n=2$  mode  $\rho_2/R_0$   
 215 and in both cases it increases approximately linearly with  
 216 the island radius.  $\rho_2/R_0$  is larger when the island has a  
 217 lozenge shape. The triangular shape of the islands mi-  
 218 grating in the  $\langle 11\bar{2} \rangle$  direction is given by a strong  $n=3$   
 219 mode  $\rho_3/R_0$  that is increasing non-linearly with the is-  
 220 land radius.

221 Our first insight into the island shape and symmetry  
 222 is based on crystallographic considerations. The step  
 223 edge properties of the  $7 \times 7$  have a threefold symmetry  
 224 [39 and 40]. However due to symmetry breaking by elec-  
 225 tromigration, maximum a mirror symmetry can be ex-  
 226 pected. As observed experimentally if the electric cur-  
 227 rent is along the symmetry axis  $\langle 11\bar{2} \rangle$  the shape has a

228 mirror line whereas it is not the case when the electric  
229 current is along the non-symmetric  $\langle 110 \rangle$  direction (Fig.  
230 3(d)). As the shape is far from equilibrium the kinetic  
231 of mass transfers such as the one involved in the attach-  
232 ment/detachment of atoms at step edges is expected to  
233 play a major role.

234 In the framework of a continuous step model, the shape  
235 of *advacancy* islands driven by an electromigration force  
236 on *adatoms* and considering mass transfers by terrace  
237 diffusion and attachment/detachment at step edges has  
238 been calculated [20, 31, and 41]. The elongation of the  
239 *advacancy* islands is perpendicular to the migration di-  
240 rection considering isotropic surface properties. This  
241 shape can be qualitatively interpreted as resulting from a  
242 mass flux towards the migration axis. Indeed in presence  
243 of a slow kinetics of attachment, the *adatoms* make sev-  
244 eral trials before attaching to the step and have a residual  
245 drift towards the migration axis. In a steady state the  
246 local curvature of the island is modified to compensate  
247 this mass flux by a capillary effect. Quantitatively the  
248 change of shape involves the  $n = 2$  mode as  $\rho_2/R_0$  ratio  
249 (elongation) and reads ([20] for  $d \ll R_0$ ):

$$\frac{\rho_2}{R_0} = -\frac{1}{12\Gamma} \frac{R_0^2}{\xi^2} d < 0 \quad (3)$$

250 where  $\Gamma = \frac{a^2 \tilde{\beta}}{k_B T}$  is the capillary length (Gibbs-Thomson  
251 effect),  $\tilde{\beta}$  is the step edge stiffness and  $\xi$  is a character-  
252 istic length associated with the electromigration force:  
253  $\xi = \frac{k_B T}{F} = 58 \mu\text{m}$ . This result is opposite to the ex-  
254 perimental shape since the elongation of the *advacancy*  
255 islands is along the displacement direction ( $\frac{\rho_2}{R_0} > 0$ )  
256 whatever this direction is. Therefore we conclude that  
257 the anisotropy of the surface properties cannot solely ex-  
258 plain it. We propose that the electric current modifies  
259 not only the *adatom* displacement but also the atomic  
260 step properties. As a minimum model, the electric cur-  
261 rent breaks the threefold symmetry of the kinetic rate of  
262 attachment-detachment at the step edges [42]. To study  
263 this effect we expand the kinetic length  $d$  as a Fourier  
264 series  $d = \bar{d} + \sum_n d_n \cos(n\theta)$  where  $\bar{d}$  is the mean kinetic  
265 length of attachment/detachment and  $d_n$  are the Fourier  
266 coefficients for a symmetric shape. The main Fourier  
267 term acting as an electrobias, *i.e.* changing the kinetics  
268 of attachment-detachment at the step edge, is expected  
269 to be  $d_1$  (without electrobias only  $d_{3n}$  exists by sym-  
270 metry). Expanding linearly the shape of the *advacancy*  
271 island with this electrobias effect we obtain:

$$\frac{\rho_2}{R_0} = -\frac{1}{12\Gamma} \left[ \frac{R_0^2}{\xi^2} \left( \bar{d} + \frac{d_2}{2} \right) - 2 \frac{R_0}{\xi} (d_1 + d_3) \right] \quad (4)$$

272 The shape elongation  $\frac{\rho_2}{R_0}$  shows a new contribution that

273 increases linearly with the island radius  $R_0$  as in the mea-  
274 surements, and is along the migration axis if  $d_1 + d_3 > 0$ .  
275 As  $\frac{R_0}{\xi} \sim 0.017 \ll 1$  we can neglect the second order  
276 contribution in eq. (4). To estimate only the electro-  
277 bias effect  $d_1$  we use the change of the current direc-  
278 tion in the experiment. Assuming that  $d_3$  is not signifi-  
279 cantly affected by the electrobias effect, the inversion of  
280 the current direction changes this value by  $-d_3$ , there-  
281 fore the kinetic length of electrobias  $d_1$  is obtained by  
282 averaging both shape elongation  $\frac{\rho_2}{R_0}$  in the  $\langle 11\bar{2} \rangle$  and  
283 the  $\langle \bar{1}12 \rangle$  directions. We estimate that  $d_1 \sim 68 \pm 10$   
284 nm ( $\Gamma = 1$  nm [8]) and considering that this contribution  
285 is thermally activated, we extract the activation energy  
286  $E_1 = k_B T \ln \left( 1 + \frac{d_1}{\bar{d}} \right) = 1.2 \cdot 10^{-2}$  eV. This electrobias ef-  
287 fect on the step edge is much larger than on *adatoms*  
288 ( $E = Fa/2 \sim 3.2 \cdot 10^{-7}$  eV). This result could be related  
289 to an intrinsic change of step properties induced by the  
290 current but it may also arise from a change of kink den-  
291 sity at step edges. Indeed it has been shown [43] that an  
292 electric current in the  $\langle 11\bar{2} \rangle$  direction along a step and as-  
293 cending the kinks favors the formation of an atomically  
294 straight step edge. Therefore considering that the kinet-  
295 ics of mass transfers at step edges is mediated by kinks  
296 then the rate of attachment/detachment could be indeed  
297 strongly modified by the electric current. Such an elec-  
298 trobias effect on steps or kinks has already been found  
299 on metal surfaces [5 and 14] but not on semiconductors  
300 even though it has been suspected to occur [44].

301 In conclusion we have shown on Si(111) surface that  
302 a monoatomic *advacancy* island in the  $1 \times 1$  high tem-  
303 perature phase and surrounded by the  $7 \times 7$  low temper-  
304 ature phase can be stabilized. This regime allows keep-  
305 ing the 2D island in a confined state in terms of atomic  
306 exchanges. Then under the influence of an electric cur-  
307 rent, the island is moving. The analysis of the velocity  
308 and shape of the island as function of its radius show  
309 that (i) Si *adatoms* migration on the terrace is biased  
310 and they have an effective valence  $Z^*$  of  $3.4 \pm 0.6$  (ii) the  
311 kinetic of attachment/detachment of atoms at the step  
312 edges is very slow and we evaluate the kinetic length as  
313  $\sim 500$  nm. (iii) An electrobias effect on the kinetics of  
314 attachment/detachment at step edges elongates the is-  
315 land shape in the direction of the electric current. We  
316 believe that a complete modeling including all the effects  
317 of anisotropy, non-linearities and high density of *adatoms*  
318 would be necessary to describe precisely the island shape.

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323 \* leroy@cinam.univ-mrs.fr

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