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Step-step interactions on the vicinal Si(111)$\sqrt{3}\times\sqrt{3}$-Ga surface

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Steps on a Si(111)$\sqrt{3}\times\sqrt{3}$-Ga surface that was inclined toward the $[\bar{1}\bar{1}2]$ direction were observed at 550 °C with a scanning tunneling microscope. The step stiffness and step-step repulsion coefficient of single-bilayer steps were estimated to be 3×10^{-11} J m^{-1} ($\sim$20 meV Å^{-1}) and 2×10^{-29} J m ($\sim$1.3 eV Å). It was clarified that the elastic repulsion is dominant in step-step interactions on the surface. The formation energy of double-bilayer steps at 550 °C was estimated to be of the order of 1×10^{-13} J m^{-1} ($\sim$0.1 meV Å^{-1}). [S0163-1829(98)04927-3]

The vicinal Si(111)$\sqrt{3}\times\sqrt{3}$-Ga surface that is inclined toward the $[\bar{1}\bar{1}2]$ direction is of interest for constructing nanometer-scale structures, because alternate stripe structures consisting of Si(111)7×7 and Si(111)$\sqrt{3}\times\sqrt{3}$-Ga regions are formed on the surface by thermal desorption of Ga atoms.[1] Since the $\sqrt{3}\times\sqrt{3}$-Ga region is chemically less reactive than the 7×7 region, molecules such as oxygen,[2] disilane,[3] and antimony (Sb$_4$) (Ref. 4) are selectively adsorbed on the 7×7 region. Owing to this selectivity, nanoscale-stripe structures where Ga and Sb are alternately adsorbed can be constructed on the Si(111) surface without using conventional lithography. For the construction of the stripe structure, atomically straight boundaries must be formed between the stripe regions. When the vicinal Si(111)$\sqrt{3}\times\sqrt{3}$-Ga surface is annealed above 500 °C, step edges are bunched. The bunched step edges are so stable that they provide atomically straight boundaries between the stripe regions.

Step bunching is essential in constructing nanometer-scale structures on the vicinal Si(111)$\sqrt{3}\times\sqrt{3}$-Ga surface. Nonetheless, detailed understanding of bunching on the surface is still scant. The vicinal Si(111)$\sqrt{3}\times\sqrt{3}$-Ga surface inclined toward the $[\bar{1}\bar{1}2]$ direction shows mixtures of single- and double-bilayer steps within miscut angles from 0.6° to 0.9°. The presence of mixtures means that the step bunching on the surface takes place when the interactive energy due to step-step interactions overcomes the double-bilayer formation energy.[5,6] Step-step interactions result from elastic and entropic repulsions between steps.[7] The former is caused by elastic strain and/or dipole moment at step edges, and the latter arises from the no-overlap condition of steps. The dominant origin of step-step interactions on the Si(111)$\sqrt{3}\times\sqrt{3}$-Ga surface, however, has not been understood.

In this work, we determined the stiffness and step-step repulsion coefficient of single-bilayer steps on a Si(111)$\sqrt{3}\times\sqrt{3}$-Ga surface inclined toward the $[\bar{1}\bar{1}2]$ direction. For this purpose, we observed step wandering on the surface with a high-temperature scanning tunneling microscope (STM). Using the step stiffness and repulsion coefficient, we discussed the primary origin of step-step interactions on the surface and estimated the formation energy of double-bilayer steps.

The experiments were performed in a high-temperature STM chamber. The sample was cut from an edge of an on-axis Si(111) wafer whose miscut angle was less than 15 sec. The sample size was 7 mm along the [110] direction, and 1 mm along the $[\bar{1}\bar{1}2]$ direction. The sample was heated by passing a direct current through it. Temperatures corresponding to each current were calibrated by measuring sample resistance as a function of the temperature monitored with a pyrometer. The sample emissivity was calibrated by observing the phase transition between Si(111)7×7 and 1×1 by low-energy electron diffraction. Since the current was directed parallel to the steps, we ignored the effect of electromigration on step bunching. After cleaning of the surface by thermal flashing at 1200 °C, a one-third monolayer of Ga was deposited at room temperature.[8] The $\sqrt{3}\times\sqrt{3}$-Ga structure was created by annealing the sample at 500 °C. STM images were acquired in the constant-current mode at 550 °C, at which temperature Ga desorption is not frequent.[9] Before taking STM images, we waited more than 1 h to allow the instrument to stabilize thermally. Sample biases were in the range from 1.0 to 2.0 V and tunneling currents were from 0.1 to 0.3 nA. Miscut angles of the sample were varied from 0° to 1° by moving the sample along the $[\bar{1}\bar{1}2]$ direction mechanically. Since the miscut angles gradually varied within the sample width, the miscut angle in each STM image was regarded as constant.

Figure 1 shows an STM image of an isolated step that is wandering along the [110] direction on the Si(111)$\sqrt{3}\times\sqrt{3}$-Ga surface. Both edges of the step are not pinned. Similar isolated parts were selected from steps fluctuating freely on the surface. Figure 2 shows the mean-square fluctuation $\langle\Delta y^2\rangle$ of steps as a function of the step length L. The relationship between the mean-square fluctuation and step length is expressed as[10,11]

$$\langle\Delta y^2\rangle = kTL/12\tilde{\beta}, \tag{1}$$

where $\tilde{\beta}$ is the step stiffness, k the Boltzmann constant, and T the substrate temperature. The data are best fitted when $\tilde{\beta}$ is 2.7×10^{-11} J m^{-1} ($\sim$20 meV Å^{-1}), as indicated in the solid line in Fig. 2. While the data may include experimental error, the step stiffness is still estimated to be in the range of $(2-5)\times10^{-11}$ J m^{-1}. This value is about one-fourth of the step stiffness of the Si(111)1×1 surface, which was reported to be 1×10^{-10} J m^{-1} by Alfonso *et al.*, albeit at

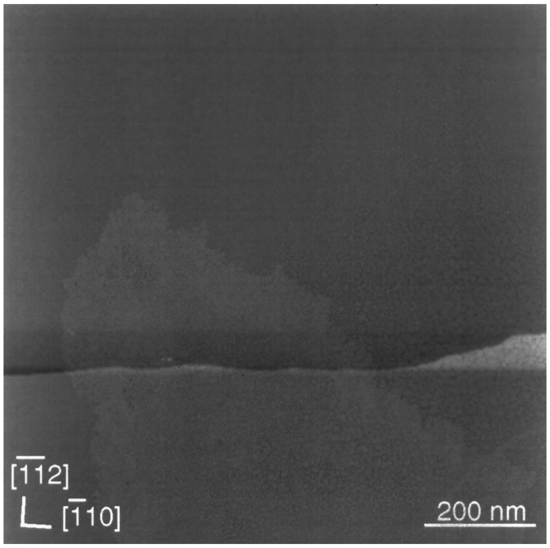

FIG. 1. STM image of an isolated step on a Si(111)$\sqrt{3}\times\sqrt{3}$-Ga surface. The step is almost parallel to the [110] direction. The substrate temperature is 550 °C. The area shown is $1.0\times 1.0\ \mu$m.

900 °C.[12] This difference is probably caused by the reduction in surface energy caused by Ga atoms passivating Si dangling bonds. The step stiffness is close to the step energy for a highly symmetrical surface such as Si(111). Since Ga passivation reduces the energy of the Si(111)$\sqrt{3}\times\sqrt{3}$-Ga surface and step edges, the step stiffness can be reduced on the surface.

Figure 3 shows a step train on the Si(111)$\sqrt{3}\times\sqrt{3}$-Ga surface at 550 °C. We acquired STM images by varying the miscut angle, and measured the mean step distances m and the standard deviation σ of the step distances by selecting 500–800 pairs from each STM image. Histograms of step distances were well fitted by a Gaussian distribution. The result is shown in Fig. 4. Step trains with mean step distances smaller than 30 nm were not analyzed because they included bunching steps. The standard deviation exhibits $\sigma=(0.26\pm 0.02)m$. Joós, Einstein, and Bartelt[13] reported that the ratio σ/m is 0.424 for purely entropic interactions and it is less than 0.424 when step-step interactions include both contributions of entropic and elastic repulsions. When the elastic repulsion outweighs the entropic one, distribution of step distances tends to a Gaussian distribution. Our result suggests that the elastic repulsion is dominant in the step-step interactions on the Si(111)$\sqrt{3}\times\sqrt{3}$-Ga surface. The relationship between the standard deviation and the mean step distance is[14]

$$\sigma=m(k^2T^2/48A\tilde{\beta})^{1/4}, \tag{2}$$

where A is the step-step repulsion coefficient, which describes the elastic-repulsion energy U between two steps separated by a spacing x as $U=A/x^2$. From $\sigma=0.26m$, the value of A is 2.2×10^{-29} J m ($\sim$1.4 eV Å) when $\tilde{\beta}=2.7$

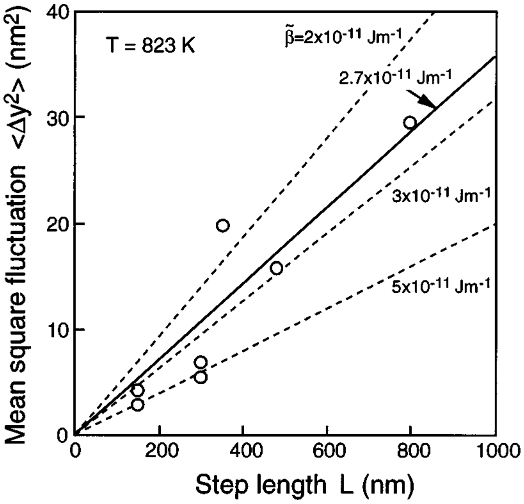

FIG. 2. Mean-square fluctuation of steps $\langle\Delta y^2\rangle$ as a function of the step length L. The solid line shows the best fit, resulting in the step stiffness $\tilde{\beta}=2.7\times 10^{-11}$ J m^{-1} from Eq. (1).

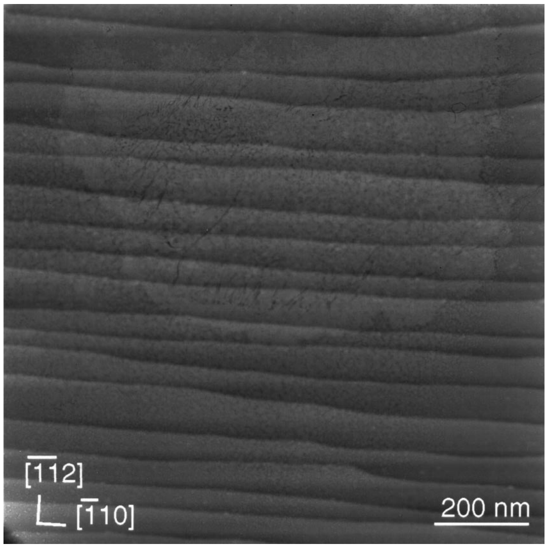

FIG. 3. STM image of step trains on a Si(111)$\sqrt{3}\times\sqrt{3}$-Ga surface inclined toward the [112] direction. The steps are almost parallel to the [110] direction. The substrate temperature is 550 °C. The area shown is $1.2\times 1.2\ \mu$m.

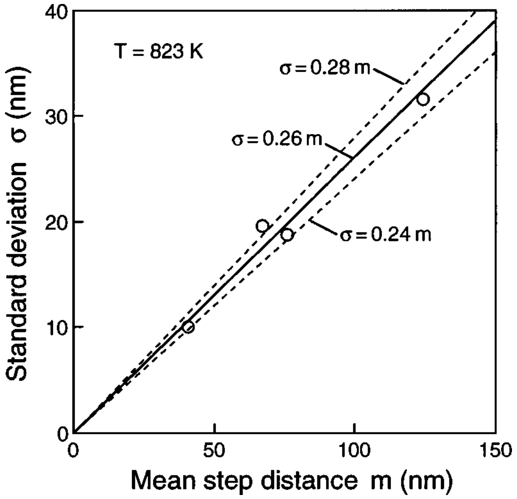

FIG. 4. Standard deviation of the step distance σ as a function of the mean-step distance m. The solid line shows $\sigma=0.26m$, resulting in the step-step repulsion coefficient $A=2.2\times 10^{-29}$ J m from Eq. (2).

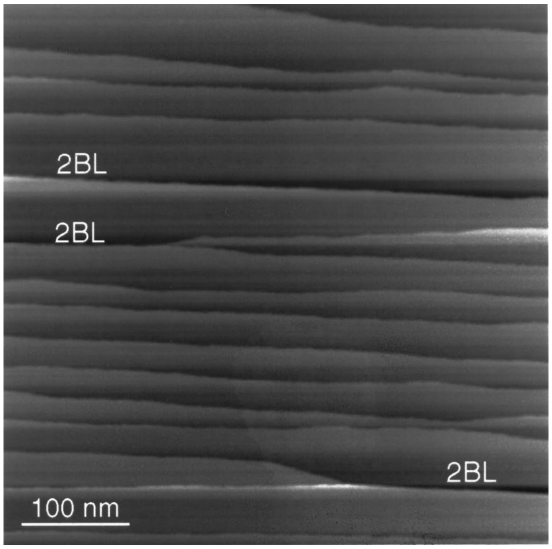

FIG. 5. STM image of double-bilayer (2 BL) steps present among single-bilayer steps on a vicinal Si(111)$\sqrt{3}\times\sqrt{3}$-Ga surface. The mean step distance is 29 nm when reducing each double-bilayer step to two single-bilayer steps. The substrate temperature is 550 °C. The area shown is 0.5×0.5 μm.

$\times 10^{-11}$ J m^{-1}. By taking into account the ambiguity of the value of $\tilde{\beta}$, the repulsion coefficient A is estimated to be in the range of $(2\pm 1)\times 10^{-29}$ J m (1.3 ± 0.6 eV Å). This value is larger than the repulsion coefficients of the Si(111)1×1 surface, 0.2 eV Å,[12] and the Si(111)7×7 surface, 0.4 eV Å.[15] Although the repulsion coefficient estimated in this work may be overestimated due to the ambiguity of $\tilde{\beta}$, the level of the repulsion coefficient suggests that the Si(111)$\sqrt{3}\times\sqrt{3}$-Ga surface exhibits stronger elastic step-step repulsions than clean Si(111) surfaces. When the miscut of the azimuthal angle is small, the contribution of entropy to step-step interactions is of the order of $\sim(kT)^2/\tilde{\beta}$.[15] In our case, it is estimated to be $\sim 4\times 10^{-30}$ J m, indicating that contribution of the elastic repulsion is larger than that of the entropic one on the Si(111)$\sqrt{3}\times\sqrt{3}$-Ga surface. This coincides with the fact that histograms of step distances were well fitted by a Gaussian distribution. Thus, we conclude from our results that the primary origin of step bunching on the Si(111)$\sqrt{3}\times\sqrt{3}$-Ga surface is the elastic step-step repulsion.

Next, we roughly estimate the energy for producing one double-bilayer step from two single-bilayer steps. When only single-bilayer steps are present with a density of ρ on a surface, the surface energy associated with steps and step-step interactions is expressed as $\beta\rho+\phi\rho^3$,[7] where β is the free-energy cost per unit length of creating an isolated single-bilayer step and ϕ is the free-energy cost per unit area due to single-bilayer step-step interactions. After step bunching, the single-bilayer step density decreases into $\rho-2\rho'$ by creating double-bilayer steps with a density of ρ'. The energy associated with steps per unit area is $\beta(\rho-2\rho')+\beta'\rho'$, where β' is the free energy per unit length of an isolated double-bilayer step. With respect to the step-step interaction energy, $\phi\rho^3$ represents the summation of interaction energies that are produced by each step interacting with an infinite number of steps. In our analysis, however, we use a first approximation where a pair of adjacent steps, separated by a spacing $1/\rho$, accumulates a step-step interaction energy of $\phi\rho^2$, because the interaction energy between adjacent steps is dominant in the total interaction energy. Within this approximation, when the interaction between double-bilayer and single-bilayer steps is α times larger than that between single-bilayer steps, the mean spacing between double and single steps is $\alpha^{1/3}$ times wider than that between neighboring single steps, l, and interactions energies of each single-single and double-single step pairs are ϕ/l^2 and $\alpha^{1/3}\phi/l^2$ $[=\alpha\phi/(\alpha^{1/3}l)^2]$. The mean spacing l is given by $(\rho-3\rho')l+2\rho'\alpha^{1/3}l=1$, which means that total terrace size is unity. When the density of bunched steps is sufficiently low, a unit area includes single-single step pairs with a density of $\rho-3\rho'$ and single-double step pairs with $2\rho'$. Then the energy associated with step-step interactions is approximated to $(\rho-3\rho')\phi/l^2+2\rho'\alpha^{1/3}\phi/l^2$. Therefore, the total surface energy per unit area, E, can be expressed as follows:

$$E\sim\beta(\rho-2\rho')+\beta'\rho'+\phi[\rho-(3-2\alpha^{1/3})\rho']^3. \qquad (3)$$

The general form of the interaction term ϕ is[15]

$$\phi=\pi^2k^2T^2a_n/24\tilde{\beta}a_p[1+(1+4Aa_p\tilde{\beta}/k^2T^2a_n)^{1/2}]^2, \qquad (4)$$

where a_n is the depth of a single kink and a_p is the minimum separation of kinks. In making our analysis, we assume a kink depth of 0.333 nm, and a kink separation of 0.384 nm, which are the same as those on the Si(111)1×1 surface, because the atomic configuration of Si atoms below Ga adatoms of the Si(111)$\sqrt{3}\times\sqrt{3}$-Ga structure is the 1×1 structure.[8,16] The step-step interaction term ϕ at 550 °C is estimated to be $\phi=5\times 10^{-29}$ J m.

The factor α is inferred to be around 2. When the elastic repulsion predominantly originates from surface stresses, the repulsion coefficient is proportional to step heights,[6,10] resulting in almost linear increase of the interaction term ϕ. To clarify step height dependence of the repulsion coefficient, we observed a 0.8°-miscut Si(111)$\sqrt{3}\times\sqrt{3}$-Ga surface, where single- and double-bilayer steps coexist, at 550 °C. The mean step-step distances of l_{ss}, l_{sd}, and l_{dd}, where ss, sd, and dd denote single-single, single-double, and double-double step pairs, were 28.0, 31.4, and 40.1 nm, respectively, from an analysis of total 680 step-step pairs. Ratios of step distances provide $(l_{sd}/l_{ss})^3=1.4$ and $(l_{dd}/l_{sd})^3=2.1$, indicating that the step-step interactions depend on step heights and α is around 2. More precise estimation of the factor α is difficult because the dipole moment at steps may also contribute to the elastic repulsion[7] and the step-height dependence of the step stiffness has a slight effect on the interaction term.

Since step bunching occurs when $\partial E/\partial\rho'\leqslant 0$ at $\rho'=0$, the formation energy of double-bilayer steps, $\beta'-2\beta$, is estimated to be $\beta'-2\beta\sim 3(3-2\alpha^{1/3})\phi\rho_c^2$, where ρ_c is the minimum value of the single-bilayer step density that causes double-bilayer step bunching. When $\alpha\sim 2$, the formation energy is of the order of $\sim 1.4\phi\rho_c^2$. The value of ρ_c can be measured directly by observing Si(111)$\sqrt{3}\times\sqrt{3}$-Ga surfaces for various miscut angles. In our STM observation, step bunching occurred when the mean distances of single-bilayer steps were narrower than 30 nm. A typical STM image is shown in Fig. 5, where 11 single-bilayer steps and 3 double-bilayer steps are seen. We determined the mean distance of single-bilayer steps to be 29 nm, by reducing each double-bilayer step to two single-bilayer steps. The step distance of 29 nm corresponds to $\rho_c = 3.4 \times 10^7\ \mathrm{m}^{-1}$ (0.6° miscut). The formation energy of double-bilayer steps is estimated to be of the order of $\beta' - 2\beta \sim 1 \times 10^{-13}\ \mathrm{J\,m}^{-1}$ ($\sim 0.1\ \mathrm{meV}\ \mathrm{Å}^{-1}$). This value is reasonable because it explains well the fact that bunched steps do not appear on Si(111)$\sqrt{3}\times\sqrt{3}$-Ga surfaces with wide terraces.[17] When $\beta' - 2\beta \sim 0.1\ \mathrm{meV}\ \mathrm{Å}^{-1}$, the formation energy of bunched steps with finite length, e.g., 1 μm in length, is of the order of 1 eV. Since this formation energy is larger than thermal energy, hardly any double-bilayer steps remain on the surface in the equilibrium state without the step-step interaction energy.

In summary, we measured the step stiffness and step-step repulsion coefficient of single-bilayer steps on the Si(111)$\sqrt{3}\times\sqrt{3}$-Ga surface inclined toward the [112] direction at 550 °C. The step stiffness was estimated to be $3 \times 10^{-11}\ \mathrm{J\,m}^{-1}$ ($\sim 20\ \mathrm{meV}\ \mathrm{Å}^{-1}$), and the repulsion coefficient $2 \times 10^{-29}\ \mathrm{J\,m}$ ($\sim 1.3\ \mathrm{eV}\ \mathrm{Å}$). The elastic step-step repulsion is the primarily origin of step-step interactions on the surface. The formation energy of double-bilayer steps is of the order of $1 \times 10^{-13}\ \mathrm{J\,m}^{-1}$ ($\sim 0.1\ \mathrm{meV}\ \mathrm{Å}^{-1}$).

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