Non-contact atomic force microscopy investigation of the (1×1) and $(\sqrt{3} \times \sqrt{3})$ phases on the Pb/Si(111) surface*

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The (1×1) and $(\sqrt{3} \times \sqrt{3})$ phases of the Pb/Si(111) surface are investigated at room temperature using non-contact atomic force microscopy. The topographic height difference between these phases is determined to be 1.6 ± 0.1 Å, which is larger than the theoretical value (1.2 Å) previously predicted. Kelvin probe force microscopy measurements show that the work function on the (1×1) region is 201 ± 16 meV higher than that on the $(\sqrt{3} \times \sqrt{3})$ region. The effective dipole moments by electron transfer from the first layer Si atom to the Pb adatom on the (1×1) phase is discussed. We also succeed in obtaining atomically resolved images of the two phases, and demonstrate that the (1×1) phase has small atomic corrugation compared with the $(\sqrt{3} \times \sqrt{3})$ phase when imaging at same tip condition and the same acquisition parameters. [DOI: 10.1380/ejssnt.2007.67]

Keywords: Atomic force microscopy; Surface structure, morphology, roughness, and topography; Si; Pb

I. INTRODUCTION

Non-contact atomic force microscopy (NC-AFM) in ultra high vacuum (UHV) has become a powerful tool for imaging various surfaces, including insulator surfaces, with atomic resolution [1, 2], measuring the interaction forces between a tip apex atom and an individual atom on surfaces [3–5], identifying chemical atom species [6], and manipulating single atoms [7–10]. It should be noted that the spatial resolution of NC-AFM on some semiconductor surfaces is higher than that of scanning tunneling microscopy (STM) [11–13]. On the other hand, there are only a few reports on observation of metallic surfaces using NC-AFM [14–16], while many metallic surfaces have been investigated by STM. It is challenging task to investigate metallic surfaces by the interaction force using NC-AFM because of the short interatomic distances and low charge corrugation.

The Pb/Si(111) surface, which is a model system for the growth of metals on semiconductors, is an interesting system because the interface between the Pb adatoms layer and the Si substrate is well defined due to the negligible solubility between them. This surface at room temperature has various phases dependent on sample preparation conditions of coverage of Pb atoms, annealing temperature after Pb deposition, and even annealing history [17–19]. In these phases, the Pb/Si(111)- $(\sqrt{3} \times \sqrt{3})$ pure phase

and the Pb/Si(111)- (1×1) phase are intriguing systems and have been well investigated since below room temperature, the phase transitions to the (3×3) and $(\sqrt{3} \times \sqrt{7})$ occur, respectively [19–22]. Both the two phases show the metallic behavior and especially the (1×1) phase has higher conductivity [23]. The structure model for the $(\sqrt{3} \times \sqrt{3})$ pure phase has been well established: Pb atoms adsorb on the T_4 sites on the Si(111) surface and saturate all dangling bonds of the first layer Si atoms [Fig. 1(a)]. In this phase, some Pb adatoms are substituted by Si atoms due to the entropy effect. On the other hand, the structure model for the (1×1) phase has not been determined although the (1×1) patterns were experimentally observed by using STM [23], low energy electron diffraction [23], and high energy electron diffraction [24]. A model proposed as in Fig. 1(b) is that each Pb atom locates a T_1 site on the Si(111) surface [25]. In another model, Pb atoms on the (1×1) phase move around at room temperature and preferential residence sites show the (1×1) pattern [26]. It is important to obtain further information regarding the atomic structure of the (1×1) phase from the aspect of understanding the novel physical phenomena observed in this surface [27].

In this study, we investigated the differences of the topographic height, the work function, and the atomic corrugation between the $(\sqrt{3} \times \sqrt{3})$ pure phase and the (1×1) phase on the Pb/Si(111) surface using NC-AFM. By using NC-AFM, useful information can be obtained. Especially, as we have previously demonstrated, NC-AFM topography obtained at the onset of the short-range interaction force can access the real topography i.e. atomic positions [5]. Furthermore, by Kelvin probe force microscopy (KPFM) measurements, the work function on the surface can be measured. To compare the two phases, the experi-

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FIG. 1: The schematic view of the structure model of (a, c) the Pb/Si(111)- $(\sqrt{3} \times \sqrt{3})R30^\circ$ pure phase and (b, d) the Pb/Si(111)- (1×1) surface. The unit cells are indicated by the red rhombuses. The $(\sqrt{3} \times \sqrt{3})$ phase has some substitutional Si adatoms. The vertical height differences between the adatom layer and the first layer and that between the first layer and the second layer are shown [29].

ments were performed under the same tip conditions and the same acquisition parameters since NC-AFM observation strongly depends on the atomic structure and chemical composition of the tip apex [6, 28]. The NC-AFM topographic height differences between the two phases, which were carefully measured at relatively far tip-surface distances with compensating the contact potential differences (CPD) using KPFM technique, are not consistent with the theoretically predicted value [29]. The work function difference between the two phases could be explained by effective electric dipole moments on the (1×1) phase previously suggested [30]. The atomic corrugation on the (1×1) phase is lower than that on the $(\sqrt{3} \times \sqrt{3})$ phase under the same tip conditions and the same acquisition parameters. These experimental results can shed some lights on not only the structure model of the (1×1) phase but also the NC-AFM imaging mechanism of metallic surfaces that has not been very well understood.

II. EXPERIMENT

We used a home made UHV-STM/NC-AFM operated at room temperature, equipped with an optical interferometer for the detection of the cantilever deflection. Using NC-AFM, the frequency shift of the cantilever resonance

is detected as the interaction force between tip apex atoms and surface atoms [31]; in this study, the contamination and the oxide layer of the Si tip were carefully removed by Ar ion sputtering in UHV. When large areas including both the (1×1) and the $(\sqrt{3} \times \sqrt{3})$ phases were scanned (Fig. 2), we applied the KPFM technique to compensate the CPD on the two phases during scanning. Our KPFM setup is the same as the method in [32]. The frequency and the oscillation amplitude of the AC voltage applied to the sample with respect to the grounded cantilever were 700 Hz and $0.2 V_{rms}$, respectively. The Pb/Si(111)- (1×1) and $(\sqrt{3} \times \sqrt{3})$ mixed surfaces were prepared as follows. After the Si(111)- (7×7) surface was prepared [7], Pb was evaporated on the surface followed by sample annealing at 420°C .

III. RESULT AND DISCUSSION

To investigate the topographic height difference between the $(\sqrt{3} \times \sqrt{3})$ and the (1×1) phases of the Pb/Si(111) surface, the large area of $853 \text{ \AA} \times 853 \text{ \AA}$ including both the phases, was imaged by NC-AFM [Fig. 2(a)]. Since the work function on the $(\sqrt{3} \times \sqrt{3})$ region is different from that on the (1×1) region, the CPD was compensated during scanning by KPFM technique. It

FIG. 2: (a) A NC-AFM image showing a Si(111) single step and a phase boundary between the $(\sqrt{3} \times \sqrt{3})R30^\circ$ and the (1×1) phases. The image size is $853 \text{ \AA} \times 853 \text{ \AA}$. Acquisition parameters are $f_0 = 166.008 \text{ kHz}$, $k = 31.6 \text{ N/m}$, $A = 15 \text{ nm}$ and $\Delta f = -2.2 \text{ Hz}$. Here, f_0 , k , A , and Δf are the first resonance frequency of the oscillated cantilever, the spring constant, the oscillation amplitude, and the frequency shift, respectively. The CPD was compensated by the KPFM technique. (b, d) Zoom in images of the center part of Fig. 2(a). The image size is $384 \text{ \AA} \times 384 \text{ \AA}$. $\Delta f = -3.0 \text{ Hz}$ for (b) and $\Delta f = -2.7 \text{ Hz}$ for (d). (c) The line profile of Fig. 2(b).

was reported that the additional electrostatic force by the component of the AC sample voltage affects the NC-AFM topography under the KPFM measurements [33]. However, our AC voltage and the difference of the CPD between the two phases were small enough to neglect this effect, and actually the topographic height difference using KPFM was same as that without KPFM within measurements error. The NC-AFM image in Fig. 2(a) includes two steps: the one is a step between upper (1×1) phase and the $(\sqrt{3} \times \sqrt{3})$ phase, and the other is a step between lower (1×1) phase and the $(\sqrt{3} \times \sqrt{3})$ phase. The former is a phase boundary and the latter is attributed to a Si(111) single step. Figs. 2(b) and (c) show a zoom in image of the center part of Fig. 2(a) and the line profile. The height difference between the upper and the lower (1×1) phases corresponds to the characteristic height of a Si(111) single step ($3.15 \text{ \AA}$). The height of the (1×1) phase is higher than that of the $(\sqrt{3} \times \sqrt{3})$ phase on the same terrace of the Si(111) substrate by a typical height of $1.6 \pm 0.1 \text{ \AA}$ (this measurement comes from the averaging of several images measured with different tips). To

obtain the real topographic information, we scanned the surface at the onset of the short-range chemical interaction force, where the tip-surface distance is relatively far [5]. In this situation, the differences of the tip-induced relaxation effect and the short-range interaction force between the two phases become negligible. Furthermore, we confirmed that the long-range force on the (1×1) region is the same with that on the $(\sqrt{3} \times \sqrt{3})$ region by frequency shift vs. tip-surface distance curves. Therefore, the measured height difference between the (1×1) and $(\sqrt{3} \times \sqrt{3})$ phases should be attributed to the height difference of the Pb adatom positions between the two phases. Recently, atomic positions of some phases of the Pb/Si(111) surface were investigated by first-principles calculations [29]. The side views of the schematic models for the $(\sqrt{3} \times \sqrt{3})$ and the (1×1) phases are shown in Figs. 1(c) and (d), respectively. In this calculation, the height difference between the adatom layer and the first layer is $1.71 \text{ \AA}$ for the $(\sqrt{3} \times \sqrt{3})$ phase and $2.68 \text{ \AA}$ for the (1×1) phase. Furthermore, the first layer atoms are relaxed from the unrelaxed Si(111) surface due to the bonding of the first

FIG. 3: A KPFM measurement of the area including both the Pb/Si(111)-($\sqrt{3} \times \sqrt{3}$) pure phase and (1×1) phase. (a) NC-AFM topographic image. (b) The work function image simultaneously obtained. (c) and (d) are the line profiles of (a) and (b), respectively. The image size is $985 \text{ Å} \times 985 \text{ Å}$. Acquisition parameters are $f_0 = 159.337 \text{ kHz}$, $k = 27.7 \text{ N/m}$, $A = 14 \text{ nm}$ and $\Delta f = -8.0 \text{ Hz}$.

layer atoms to the adatoms: the height difference between the first layer atoms and the second layer atoms is 0.62 Å for the ($\sqrt{3} \times \sqrt{3}$) phase and 0.81 Å for the (1×1) phase. As a result, the height difference of the adatom layer between the (1×1) and the ($\sqrt{3} \times \sqrt{3}$) phases is estimated to be 1.2 Å . Nevertheless, the height difference measured by NC-AFM is larger than the theoretically predicted value. Although the reason of the difference is not understood, we expect that our experimental results and further theoretical investigations can clarify the unknown structure of the (1×1) phase.

Figure 3(b) shows a work function image, which was simultaneously measured with a NC-AFM topography [Fig. 3(a)]. The line profiles of Figs. 3(a) and (b) are shown in Figs. 3(c) and (d), respectively. Fig. 3(d) shows that while the two ($\sqrt{3} \times \sqrt{3}$) regions on different Si(111) terraces have same work function, the work function on the (1×1) phase is about 200 meV higher than that on the ($\sqrt{3} \times \sqrt{3}$) phase. By several KPFM measurements, it is determined that the work function on the (1×1) phase is $201 \pm 16 \text{ meV}$ higher than that on the ($\sqrt{3} \times \sqrt{3}$) phase. It could be explained by effective electric dipole moments on the (1×1) phase. Electron transfer from a Si of the first layer to a Pb adatom on the (1×1) phase generates the electric dipole moment directed from the Si(111) substrate to the Pb adatoms. This dipole moment increases the work function on the (1×1) region: the sur-

face electric potential on the region is positively shifted. In contrast, it was reported that the work function is reduced by the electric dipole moment caused by the electron transfer from the adatom to the substrate on the Na-deposited $\text{TiO}_2(110)$ surface [34]. The dipole moment on the Pb/Si(111)-(1×1) surface was firstly suggested by Custance *et al.* [30]. They reported that when the voltage pulse at negative sample bias with respect to the STM tip was applied on the ($\sqrt{3} \times \sqrt{3}$) region near a phase boundary, the (1×1) region has expanded to the location where the voltage pulse was applied. This phase transformation under the STM tip were attributed to the coupling between the electric field induced by the tip and the dipole moments on the (1×1) phase. We also observed some strikes due to the fluctuation of the boundary between the two phases using NC-AFM as shown in the arrows in Fig. 2(d). Since the CPD was compensated in our experiments, our results show that the fluctuation can be also induced by the interaction force without the electric field.

We also succeeded in obtaining atomically resolved NC-AFM images of both the (1×1) and ($\sqrt{3} \times \sqrt{3}$) phases as shown in Fig. 4. The CPD was compensated in each phase by applying the DC voltage to the sample with respect to the grounded tip. In this atomic scale imaging, KPFM was not used to prevent from the possible artifact previously reported, which is induced by the modulation of the cantilever motion in the short-range interac-

FIG. 4: The atomically resolved NC-AFM images of (a) the Pb/Si(111)-($\sqrt{3} \times \sqrt{3}$) surface and (b) the Pb/Si(111)-(1×1) surface acquired with the same tip at same acquisition parameters. The image size is $87 \text{ \AA} \times 87 \text{ \AA}$. Acquisition parameters are $f_0 = 163.891 \text{ kHz}$, $k = 32.5 \text{ N/m}$, $A = 11 \text{ nm}$, and $\Delta f = -7.7 \text{ Hz}$. (c, d) The NC-AFM image obtained with the same tip at $\Delta f = -14.0 \text{ Hz}$ and $\Delta f = -18.2 \text{ Hz}$, respectively. The state of the tip apex was changed during scanning downward on the dotted line in Fig. 4(d). The DC bias voltage applying the sample with respect to the grounded tip was -373 mV for the ($\sqrt{3} \times \sqrt{3}$) phase and -203 mV for the (1×1) phase.

tion regime due to the AC electric field [35]. To compare the corrugation between the two phases, after imaging the ($\sqrt{3} \times \sqrt{3}$) region [Fig. 4(a)], we imaged the (1×1) region [Fig. 4(b)] at the same tip condition and the same acquisition parameters. The set point of the frequency shift (Δf) was set to -7.7 Hz . Then, by imaging the ($\sqrt{3} \times \sqrt{3}$) region again (not shown), it was confirmed that the tip condition did not change during imaging. Then, at a closer tip-surface distance ($\Delta f = -14.0 \text{ Hz}$), the (1×1) image was imaged as shown in Fig. 4(c). At last, during scanning the surface at $\Delta f = -18.2 \text{ Hz}$, the tip condition was changed as shown by the dotted line in Fig. 4(d). While the atomic corrugation on the ($\sqrt{3} \times \sqrt{3}$) phase [Fig. 4(a)] is $0.5 \text{ \AA}$, that on the (1×1) phase [Fig. 4(b)] is only $0.1 \text{ \AA}$ under the same conditions: the atomic corrugation on the (1×1) is much smaller than that on the ($\sqrt{3} \times \sqrt{3}$) phase under the same acquisition parameters. With decreasing the tip-surface distance, the corrugation on the (1×1)

phase increases and reaches $0.3 \text{ \AA}$ for Fig. 4(d). However, the tip-surface distance was too close to stably obtain the image and the tip apex was finally changed. Using different cantilevers, high resolution NC-AFM images of the (1×1) phase as shown in Fig. 5(a) can be obtained. The line profile [Fig. 5(b)] shows that the atomic corrugation is estimated to be $0.3 \text{ \AA}$. One reason why the corrugation of the (1×1) phase is much smaller than that on the ($\sqrt{3} \times \sqrt{3}$) phase under the same acquisition parameters is the higher conductivity of the (1×1) phase: the low charge corrugation as the metal could provide the small corrugation of NC-AFM images. Another possible reason can be attributed to the overlap of the force field of the neighboring Pb adatoms by shorter inter-atomic distances ($3.8 \text{ \AA}$) on the (1×1) phase. Even measuring semiconducting Si(100)-(1×2) surface [36], the line profile in the parallel direction to the dimer row shows the small corrugation due to the short inter-atomic distance ($3.8 \text{ \AA}$).

FIG. 5: (a) A high resolution NC-AFM image of the Pb/Si(111)-(1 × 1) surface. The image size is 43 Å × 43 Å. Acquisition parameters are $f_0 = 159.772$ Hz, $k = 28.1$ N/m, $A = 16$ nm and $\Delta f = -8.8$ Hz. (b) The line profile of Fig. 5(a).

Although it is not clear whether the extended electronic states on the (1 × 1) phase contribute the small corrugation of the NC-AFM images or not, measurements of the short-range interaction force by the site-specific force spectroscopy [3–5] and the theoretical calculations will clarify the NC-AFM imaging mechanism of metallic surfaces.

IV. CONCLUSION

We have explored the Pb/Si(111)-(1 × 1) phase and ($\sqrt{3} \times \sqrt{3}$) pure phase at room temperature using NC-AFM. The topographic height difference between the two phases, which is carefully obtained by scanning at the onset of the short-range interaction force with compensating the CPD by KPFM, is larger than the theoretically expected value. The measured work function on the (1 × 1) phase is 201 ± 16 meV higher than that on

the ($\sqrt{3} \times \sqrt{3}$) pure phase, which could be attributed to the electric dipole moment on the (1 × 1) phase. Our atomically resolved NC-AFM images of the two phases obtained at same tip condition and same acquisition parameters show that the (1 × 1) phase has smaller atomic corrugation than the ($\sqrt{3} \times \sqrt{3}$) phase. We expect that the structure model of the (1 × 1) phase and the NC-AFM imaging mechanism of metallic surfaces can be clarified by comparison between our experimental information and theoretical calculation.

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- [1] S. Morita, R. Wiesendanger, E. Meyer, Noncontact Atomic Force Microscopy (Springer-Verlag, Berlin, 2002).
 - [2] F. J. Giessibl, Rev. Mod. Phys. **75**, 949 (2003).
 - [3] M. A. Lantz, H. J. Hug, R. Hoffmann, P. J. A. van Schendel, P. Kappenberger, S. Martin, A. Baratoff, and H.-J. Güntherodt, Science **291**, 2580 (2001).
 - [4] M. Abe, Y. Sugimoto, O. Custance and S. Morita, Appl. Phys. Lett. **87**, 173503 (2005).
 - [5] Y. Sugimoto, P. Pou, O. Custance, P. Jelinek, S. Morita, R. Perez, and M. Abe, Phys. Rev. B **73**, 205329 (2006).
 - [6] Y. Sugimoto, P. Pou, M. Abe, P. Jelinek, R. Perez, S. Morita, and O. Custance, Nature **446** (2007), in press.
 - [7] N. Oyabu, O. Custance, Y. Insook, Y. Sugawara, and S. Morita, Phys. Rev. Lett. **90**, 176102 (2003).
 - [8] N. Oyabu, Y. Sugimoto, M. Abe, O. Custance, and S. Morita, Nanotechnology **16**, S112 (2005).
 - [9] Y. Sugimoto, M. Abe, S. Hirayama, N. Oyabu, O. Custance, and S. Morita, Nature Mater., **4**, 156 (2005).
 - [10] Y. Sugimoto, P. Jelinek, P. Pou, M. Abe, S. Morita, R. Perez, and O. Custance, Phys. Rev. Lett., in press.
 - [11] T. Eguchi, Y. Fujikawa, K. Akiyama, T. An, M. Ono, T. Hashimoto, Y. Morikawa, K. Terakura, T. Sakurai, M. G. Lagally and Y. Hasegawa, Phys. Rev. Lett. **93**, 266102 (2004).
 - [12] Y. Sugimoto, M. Abe, S. Hirayama, and S. Morita, Nanotechnology **17**, 4235 (2006).
 - [13] Y. Sugimoto, M. Abe, S. Konoshita, and S. Morita, Nanotechnology **18**, 084012 (2007).
 - [14] Ch. Loppacher, M. Bammerlin, M. Guggisberg, S. Schär, R. Bennewitz, A. Baratoff, E. Meyer, and H.-J. Güntherodt, Phys. Rev. B **62**, 16944 (2000).
 - [15] S. Orisaka, T. Minobe, T. Uchihashi, Y. Sugawara, and S. Morita, Appl. Surf. Sci. **140**, 243 (1999).
 - [16] P. Dieska, I. Stich, and R. Perez, Phys. Rev. Lett. **91**, 216401 (2006).
 - [17] G. Le Lay, M. Abraham, A. Kahn, K. Hricovini, and J. E. Bonnet, Phys. Scr. **T35**, 261 (1991).
 - [18] L. Seehofer, G. Falkenberg, D. Daboul, and R. L. Johnson,

- Phys. Rev. B **51**, 13503 (1995).
- [19] K. Horikoshi, X. Tong, T. Nagao, and S. Hasegawa, Phys. Rev. B **60**, 13287 (1999).
- [20] O. Custance, J. M. Gomez-Rodriguez, A. M. Baro, L. Jure, P. Mallet, and J.-Y. Veullen, Surf. Sci. **482-485**, 1399 (2001).
- [21] I. Brihuega, O. Custance, Rubén Pérez, and J. M. Gómez-Rodríguez, Phys. Rev. Lett. **94**, 046101 (2005).
- [22] J. Slezak, P. Mutombo, and V. Chab, Phys. Rev. B **60**, 13328 (1999).
- [23] E. Ganz, I.-S. Hwang, F. Xiong, S. K. Theiss, J. Golovchenko, Surf. Sci. **257**, 259 (1991).
- [24] X. Tong, K. Horikoshi, S. Hasegawa, Phys. Rev. B **60**, 5653 (1999).
- [25] I.-S. Hwang, R.E. Martinez, C. Liu, J.A. Golovchenko, Phys. Rev. B **51**, 10193 (1995).
- [26] L. Seehofer, G. Falkenberg, D. Daboul, and R.L. Johnson, Phys. Rev. B **51**, 13503 (1995).
- [27] I.-S. Hwang, S.-H. Chang, C.-K. Fang, L.-J. Chen, and T.T. Tsong, Phys. Rev. Lett. **94**, 045505 (2005).
- [28] M. Abe, Y. Sugimoto, and S. Morita, Nanotechnology **16**, S68 (2005).
- [29] T.-L. Chan, C. Z. Wang, M. Hupalo, M. C. Tringides, Z.-Y. Lu and K. M. Ho, Phys. Rev. B **68**, 045410 (2003).
- [30] O. Custance, I. Brihuega, J.-Y. Veullen, J. M. Gomez-Rodriguez and A. M. Baro, Surf. Sci. **482-485**, 878 (2001).
- [31] T. R. Albrecht, P. Grtter, D. Horne, and D. Rugar, J. Appl. Phys. **69**, 668 (1991).
- [32] S. Kitamura, M. Iwatsuki, Appl. Phys. Lett. **72**, 3154 (1998).
- [33] K. Okamoto, Y. Sugawara, and S. Morita, Appl. Surf. Sci. **188**, 381 (2002).
- [34] A. Sasahara, H. Uetsuka, and H. Onishi, Jpn. J. Appl. Phys. **43**, 4647 (2004).
- [35] K. Okamoto, Y. Sugawara, and S. Morita, Jpn. J. Appl. Phys. **42**, 7163 (2003).
- [36] K. Yokoyama, T. Ochi, A. Yoshimoto, Y. Sugawara, and S. Morita, Jpn. J. Appl. Phys. **39**, L113 (2000).