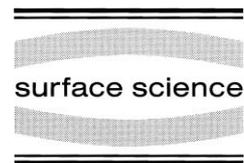




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Epitaxial growth of Cu onto Si(111) surfaces at low temperature

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Abstract

Epitaxial growth of Cu onto the Si(111) surface was studied using reflection high-energy electron diffraction. To understand the mechanism of epitaxy, its dependence on substrate temperature and deposition rate was systematically investigated. The measured Cu–Cu atomic distance of the surface layers deposited at room temperature (RT) and 160 K is smaller than the lattice constant 2.56 Å of Cu(111) up to 10 and 3 monolayers (ML), respectively. This change is attributed to the silicide formation. The change in the intensity oscillations when the temperature is decreased from RT to 160 K was seen to be identical to intensity oscillation changes observed while increasing the deposition rate to 50 ML min⁻¹ and holding the substrate at RT. The intensity oscillations are irregular up to 8 ML, and become regular >8 ML. The regular period after 8 ML is smaller than the thickness of the Cu(111) layer. A model for growth of Cu on the Si(111)-(7×7) surface at 160 K is proposed. The changes in periodicity are due to the silicide, defect formation, and partially two-dimensional nanocrystal layers. © 1998 Elsevier Science B.V. All rights reserved.

Keywords: Copper; Epitaxy; Electron–solid diffraction; Metallic films; Metal–semiconductor interfaces; Metal–semiconductor nonmagnetic thin film structures; Molecular beam epitaxy; Reflection high-energy electron diffraction (RHEED); Silicon; Single crystal epitaxy; Single crystal surfaces; Solid phase epitaxy; Surface defects; Superlattices

1. Introduction

Epitaxial growth of Cu on the Si(111)-(7×7) surface at room temperature (RT) was studied using low energy electron diffraction (LEED) and Auger electron spectroscopy (AES) by Daugy et al. and Kermmann et al. [1,2]. The AES results of experimentation by Daugy et al. indicated that Cu grew by the layer-by-layer growth mode at least over 12 monolayers (ML) [1]. However, the AES results of experimentation by Kermmann et al. showed a quasi layer-by-layer growth for the first monolayer and either formation of three-

dimensional islands or intermixing of Cu and Si atoms, or the other diffusion effects with increasing coverage [2]. Photoemission spectroscopy (PES) measurements showed that the Cu–Si intermixing was up to 12 ML with a composition gradient [3]. Using scanning tunneling microscopy (STM), Tosch and Neddermeyer observed the formation of clusters on the faulted halves of the (7×7) unit mesh in the dimer-adatom-stacking-fault (DAS) model at the thickness of 1/9 ML [4–6]. The defects on the surface were observed slightly. Cu atoms were below Si adatoms at this coverage. After deposition of 3 ML, irregular three-dimensional islands due to the Cu silicide formation were observed, and the surface showed to be disordered at this stage. The growth apparently deviated from the layer-by-layer growth. Yasue

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et al. observed that at the low coverage of 0.05 ML, Cu atoms locate on adatoms as well as rest atoms and some atoms form small clusters [7]. At 0.76 ML triangular clusters were seen to saturate almost the whole center of both the faulted and the unfaulted halves of the DAS model on the (7×7) surface, and additional irregularly shaped islands were observed at this stage of growth. At 2.2 ML the growth proceeds in an island growth.

Jalochowski et al. observed a number of the reflection high-energy electron diffraction (RHEED) intensity oscillations when the metals Au, Ag and Pb were deposited onto the Si(111)- (7×7) surface at 95 K [8–10]. They concluded that at such low temperature, the growth occurred by the layer-by-layer growth mode. In the STM observations, however, Ag growth on the Si(111)- (7×7) at 80–100 K showed two-dimensional layer-like growth [11]. Recently, RHEED intensity oscillations during growth of Cu onto the Si(111)- (7×7) at low temperature using RHEED were also observed [12]. For temperatures from 123 to 373 K, Cu growth occurred conclusively by the layer-by-layer-like growth, and the interface was confirmed to be intermixed. At 123 K such intermixing layers with the α -Cu(111) phase that contained Si <10% was measured up to 8 ML. Above 223 K this intermixing with the η -Cu₃Si phase was confirmed up to 60 ML.

We studied Ag epitaxial growth on the Si(111) surface at low temperature [13]. For the growth of Ag on the Si(111)- (7×7) surface, the measured intensity oscillations during growth of Ag showed to be irregular up to 6 ML and became regular above 6 ML. The regular period after growth of 6 ML was smaller than the thickness of the Ag(111) layer. The growth was not completely characterized by layer-by-layer growth. We concluded that the changes in periodicity were attributed to island and defect formation. These results motivated us to study growth of Cu on Si(111)- (7×7) in order to understand the growth mechanism in metals/Si(111) interface at low temperature, since the Cu/Si(111) system is different from Ag. The Cu and Si interaction is relatively strong. A p–d hybridization in the Cu–Si intermixing results in silicide formation, even at 100 K [14].

The incommensurate (5×5) surface with the silicide composition $>250^\circ\text{C}$ was also confirmed [1,15].

We report the RHEED studies of the Cu growth on the Si(111) surface at low temperature. The measured Cu–Cu atomic distance of the surface layer deposited at RT and 160 K is smaller than the lattice constant 2.56 Å of Cu(111) up to 10 and 3 ML, respectively. This change is attributed to the silicide formation. The change in the intensity oscillations when the temperature is decreased from RT to 160 K was seen to be identical to intensity oscillation changes observed while increasing deposition rate to 50 ML min⁻¹ and holding the substrate at RT. The intensity oscillations are irregular up to 8 ML, and become regular >8 ML. The regular period beyond 8 ML is smaller than the thickness of the Cu(111) layer. The changes in periodicity are due to the silicide, defect formation, and partially two-dimensional nanocrystal layers.

2. Experimental methods

An ultrahigh vacuum (UHV) chamber equipped with a RHEED system was used in this study. The residual gas pressure of the UHV chamber was $<5 \times 10^{-10}$ Torr except during Cu deposition, at which time the average vacuum was $\sim 3 \times 10^{-9}$ Torr. The electron beam energy of RHEED was 15 keV. The substrates were p-type Si(111) wafers with a resistivity of 50 Ωcm and the size was $25 \times 4 \times 0.4$ mm³. Cu was deposited onto the (7×7) structure using an alumina-coated basket and molecular beam epitaxy (MBE). Deposition rates were monitored with a quartz-crystal oscillator. The total uncertainty of the quartz-crystal oscillator was estimated to be $\sim 15\%$. This error was calibrated using the completion of the $(\sqrt{3} \times \sqrt{3})$ -Ag structure formed on the Si(111)- (7×7) surface at 500°C, since the critical coverage of the $(\sqrt{3} \times \sqrt{3})$ -Ag structure was concluded experimentally corresponding to 1 ML [16]. A monolayer in this paper corresponds to ca 7.84×10^{14} cm⁻² Si atoms on the Si(111)

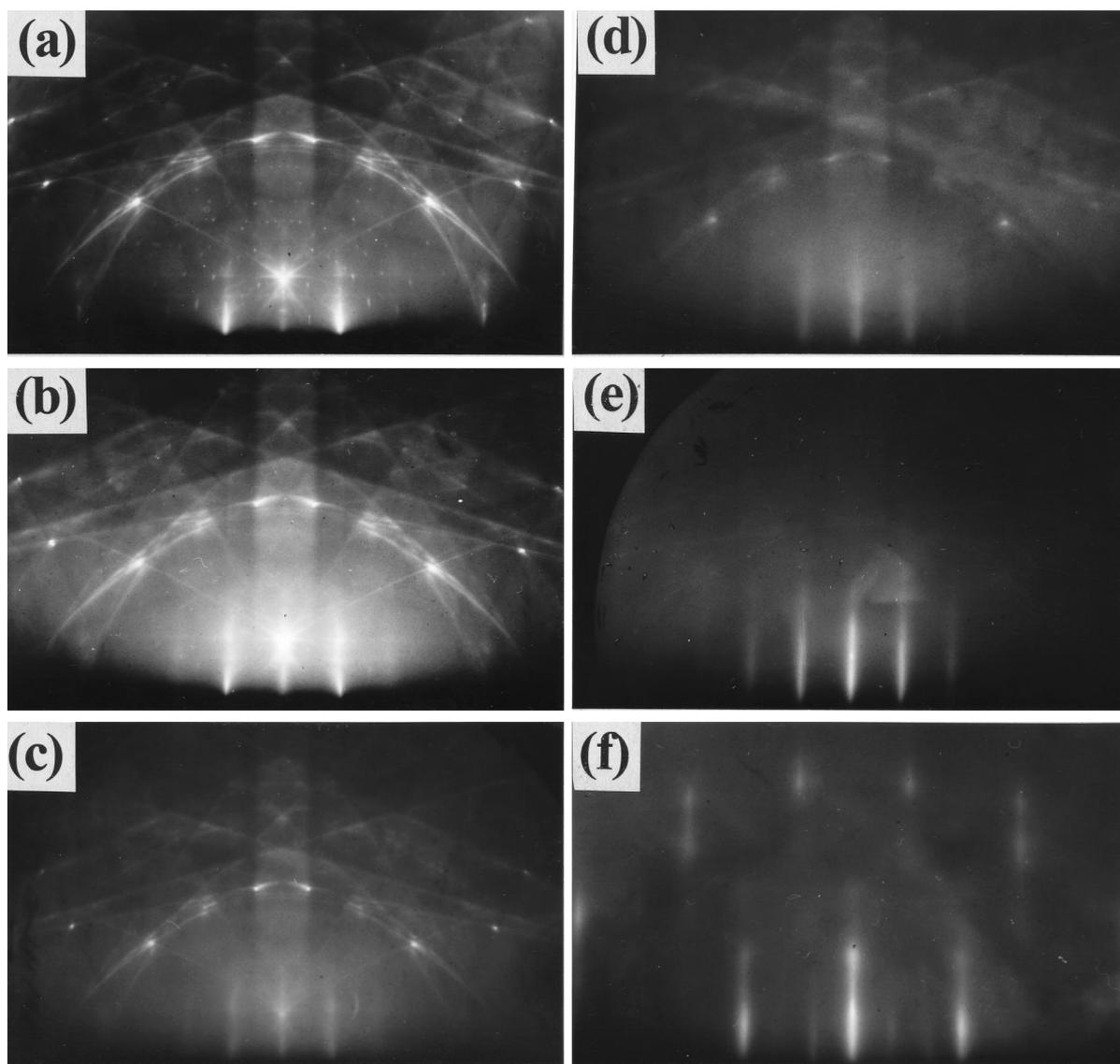


Fig. 1. RHEED patterns after Cu growth onto the Si(111)-(7 \times 7) surface at RT. The growth rate is 2.4 ML min⁻¹. Deposited thicknesses are (a) 1 ML, (b) 1.5 ML, (c) 2 ML, (d) 4 ML, (e–f) 10 ML. (a–e) [211] incidence, (f) [110] incidence, (g) the spacing changes of two peaks in intensity profiled during the growth, corresponding to two reflections due to Cu layers. The profiles are obtained from linescans across the reflections due to Cu layers in the RHEED patterns.

surface. Therefore, the thickness of one atomic layer of Cu(111) is 2.24 ML.

A clear (7 \times 7) RHEED pattern of the surface was observed by annealing the sample to 1200°C several times, and heating at 700°C for 2 h in order

to avoid the survival of Cu at the surface which may dissolve into Si after annealing [15]. We measured the RHEED specular beam intensity at a small incident angle $<0.6^\circ$, under the out-of-phase condition [17–22].

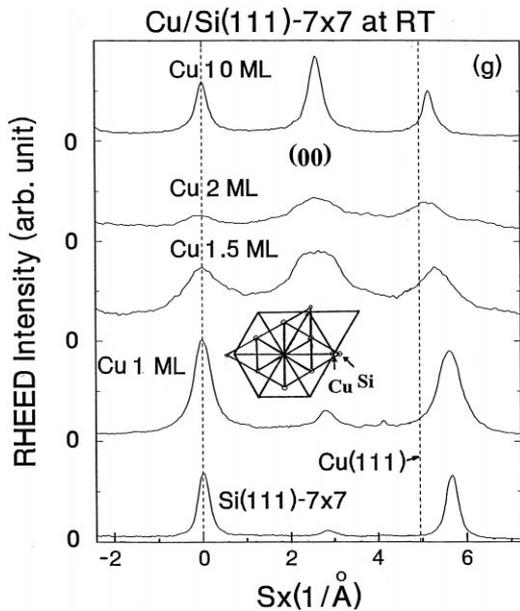


Fig. 1. (continued)

3. Experimental results

3.1. RHEED observations during Cu growth on Si(111) surface

Fig. 1a–e shows the RHEED patterns at RT and a deposition rate of 2.4 ML min^{-1} with the thicknesses of 1, 1.5, 2, 4 and 10 ML of Cu, respectively. The patterns are taken of the [211] incidence of the electron beam. Fig. 1f shows a pattern taken of the [110] incidence with the same thickness of 10 ML as shown in Fig. 1e. In Fig. 1a–b, the (7×7) superlattice reflections and the fundamental reflections of Si become weak. In Fig. 1c–d, only the fundamental reflections of epitaxial layers of Cu can be observed. These reflections are seen to be blurry. This means that the surface atoms become lateral disordered when compared to the (7×7) structure. After further deposition, Fig. 1e–f show that the reflections are long and sharp streaks, indicating that very thin crystals of Cu grow epitaxially onto the surface [23,24]. The epitaxial orientations are $(111)_{\text{Cu}} // (111)_{\text{Si}}$ and $[111]_{\text{Cu}} // [112]_{\text{Si}}$, respectively. Fig. 1g demonstrates the spacing changes of the two peaks reflected by Cu layers at 0 and ca

4.92 \AA^{-1} during the growth, corresponding to the spacing of two reciprocal lattice units $(2 \times 2\pi) / 2.56 \text{ \AA} = 4.92 \text{ \AA}^{-1}$ of the deposited Cu layer. The intensity profiles are linescans across the streaks reflected by Cu, obtained from the above patterns – (7×7) and deposited surfaces with the thickness of 1, 1.5, 2 and 10 ML, respectively. Each beam profile includes three peaks, and all the beam profiles are symmetric with respect to the peak at ca 3 \AA^{-1} , which corresponds to the specular beam. In this figure, we shift the beam profiles to choose the origin of the S_x axis at the position of the left peak in order to indicate easily the spacing change. S_x is the component of the momentum transfer along the linescan direction. The insert exhibits the reciprocal lattice structure of the Cu(111) layer on the Si(111) surface. The dashed lines show the spacing of two reciprocal lattice units of the Cu(111) layer. As can be seen in Fig. 1, the spacing between the two peaks due to reflections by Cu is larger than that due to Cu(111). The Cu–Cu atomic distances of the surface, thus, are $< 2.56 \text{ \AA}$ of the lattice constant of the Cu(111) surface, and are contracted by 13.4, 7.0, 3.5, 5.1 and 4.3% for the thicknesses of 1, 1.5, 2, 4 and 10 ML, respectively. These results show the Si–Cu intermixing at RT. However, this change in the atomic distance was not observed in the case of Ag [13].

Fig. 2a is a RHEED pattern of the clean (7×7) structure at 160 K. Fig. 2b–f are the RHEED patterns at 160 K with thicknesses of 1, 2, 3, 5 and 6 ML, respectively. The patterns are similar to those seen at RT. At low coverage, as shown in Fig. 2c–d, the (7×7) structure completely disappears. The reflections from the (1×1) -like structure are blurry, this strongly shows the disorder of surface atoms at this stage. For relatively high coverage, the reflection streaks from Cu layers become sharp and the Cu layers grow epitaxially the same as for the case at RT. Fig. 2g demonstrates the spacing changes of streaks reflected by the thin crystals of Cu during the growth obtained in the same way as in Fig. 1g. The measured Cu–Cu atomic distances are also contracted by 6.4, 5.0, 5.0, -1 and -1% for 1, 2, 3, 4 and 5 ML, respectively. These values below 3 ML are close to 2.47 \AA of the lattice constant of the $\eta\text{-Cu}_3\text{Si}$ phase.

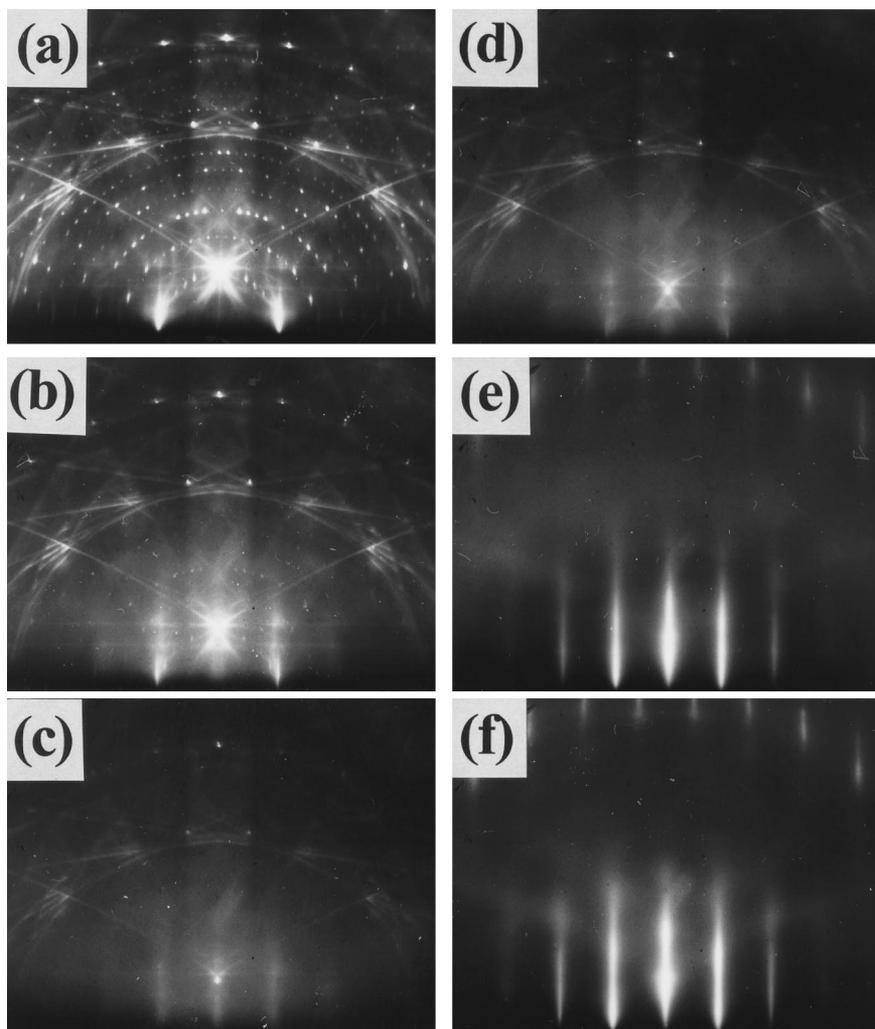


Fig. 2. (a) A RHEED pattern of the (7×7) structure at 160 K. (b–f) RHEED patterns after growth of Cu at 160 K with the thicknesses of 1, 2, 3, 5 and 6 ML, respectively. The patterns are similar to those seen at RT. (g) The spacing changes of two peaks of intensity profiles layers during the growth, corresponding to two reflections due to Cu layers. The profiles are obtained from line scans across the reflections due to Cu layers in the RHEED patterns.

This result shows that the Cu–Si intermixing also occurs at 160 K. After growth of 20 ML at 160 K, the RHEED patterns were observed as shown in Fig. 3a–b. These patterns indicate the blurry reflections from two-dimensional crystals of Cu. The measured Cu–Cu atomic distance from these patterns is near the lattice constant of Cu(111). This means that the epitaxially formed layers correspond to the Cu(111) layers. By analyzing the reflection intensity profile, the typical size of such

2D crystals is estimated to be 80 \AA [19–21]. This indicates that the formed layers consist of nanocrystals.

3.2. The dependence of temperature and deposition rate of RHEED intensity oscillation

Fig. 4 shows the measured intensity changes at various substrate temperatures from RT to 250°C with a deposition rate of 2.4 ML min^{-1} . At RT

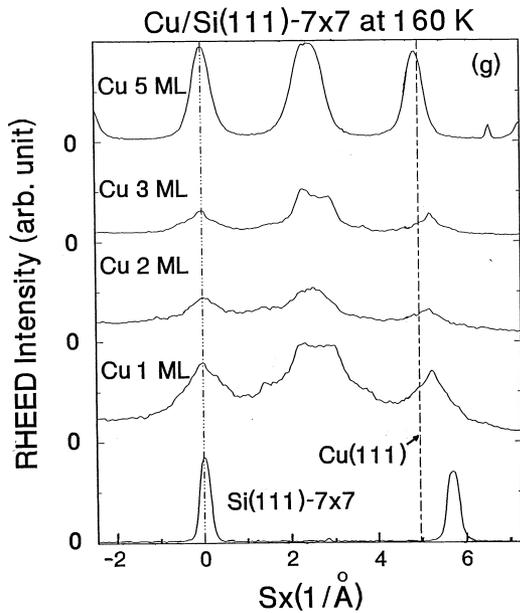


Fig. 2. (continued)

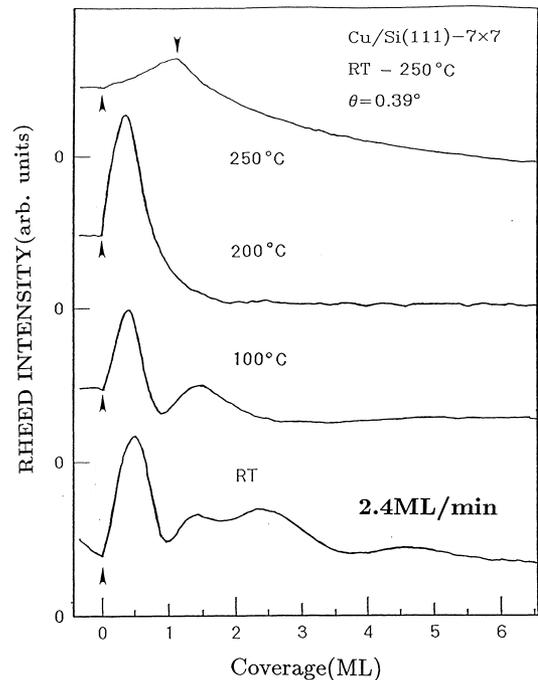
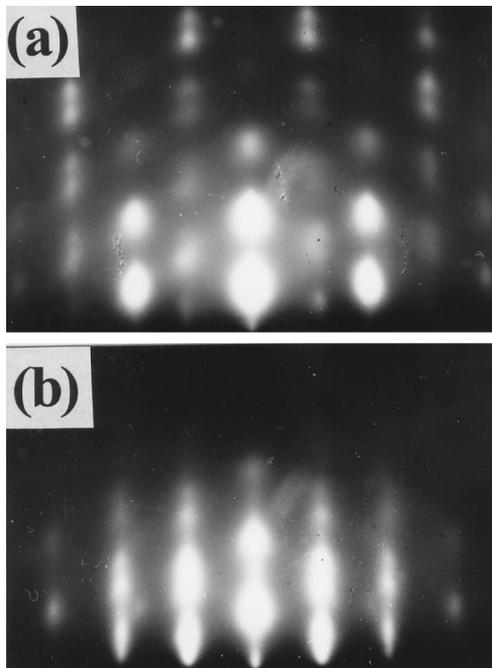
Fig. 4. RHEED intensity changes during growth of Cu onto the Si(111)-(7×7) surface at various substrate temperatures (RT–250°C) with a deposition rate of 2.4 ML min⁻¹.

Fig. 3. RHEED patterns after growth of 20 ML at 160 K. (a) [211] incidence. (b) [110] incidence.

the intensity shows four peaks where the first peak corresponds to 1 ML of growth of Cu, and the following other peaks show different periods. At 100°C the intensity shows two peaks, and the shape of the first peak is similar to that seen at RT, the second peak is seen up to 2.5 ML. At 200°C, only one peak is observed, and the peak disappears completely near 2.0 ML. At 250°C, the intensity peak appears at nearly 1.3 ML and the change is different from the cases below 250°C. The observed RHEED pattern changes to be the incommensurate (5×5) structure (I-5×5) [15]. The observed peak appears at 1.3 ML, and corresponds to the critical coverage of the (I-5×5) structure [1]. With further deposition, the reflections from three-dimensional-islands co-existing with the (I-5×5) structure could be observed >4 ML of growth in the RHEED transmission pattern at a relatively small incident angle of the electron beam. We conclude, thus, that the Cu grows in the form of the (I-5×5) surface structure plus three-dimensional-islands >250°C, for exam-

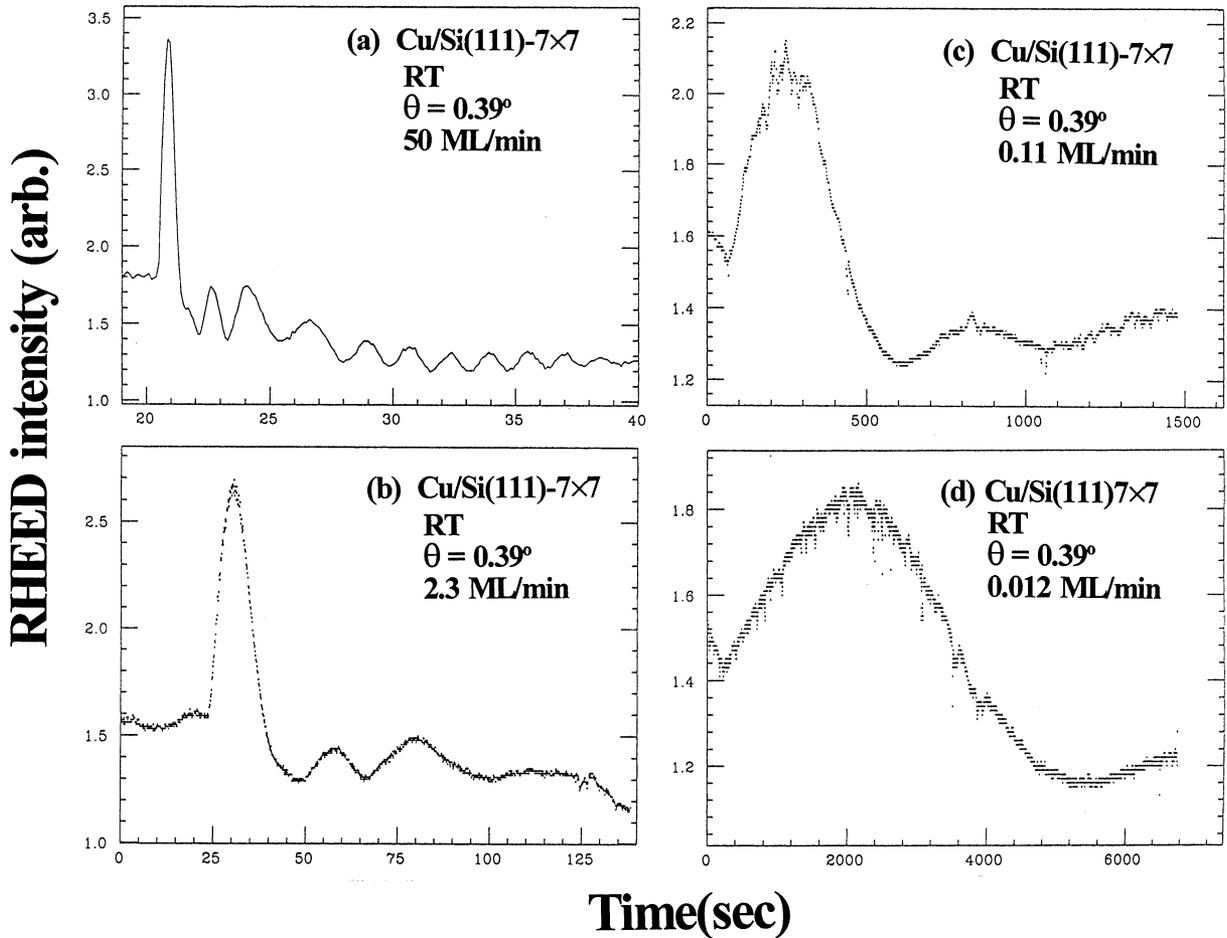


Fig. 5. The RHEED intensity changes with the different deposition rates at RT. (a) 50 ML min⁻¹, (b) 2.3 ML min⁻¹, (c) 0.11 ML min⁻¹, (d) 0.012 ML min⁻¹.

ple, Stransky–Krastanov (S–K) mode. This is consistent with the AES results, where Cu silicide islands were confirmed to form >1.3 ML when the substrate temperature is >250°C [1].

Fig. 5 shows the RHEED intensity changes at RT with the different deposition rates. At a high rate of 50 ML min⁻¹, more than ten oscillation peaks can be seen in Fig. 5a. The first peak corresponds to 1 ML, and the intensity oscillations are irregular between the second and fifth peak, and become regular after the fifth peak where the peak period corresponds to 1 ML. As shown in Fig. 5b–c. At the deposition rates of 2.3 and 0.11 ML min⁻¹, the peak number of oscillations

decreases with decreasing deposition rate. At the very low rate of 0.012 ML min⁻¹, we still observe a single peak in Fig. 5d, indicating that for the growth of the first layer, the two-dimensional nuclei forms on the terrace even at this low rate. Since the RHEED intensity oscillation is associated with the formation of the two-dimensional nuclei on the terrace of steps.

Fig. 6a shows RHEED intensity oscillations at RT, 220 K, and 160 K. Curve A is the same data with that seen at RT in Fig. 4. With decreasing temperature, the intensity oscillations become weak between 2 and 5 ML. After 6 ML, the oscillations with the period of 1 ML reappear clearly.

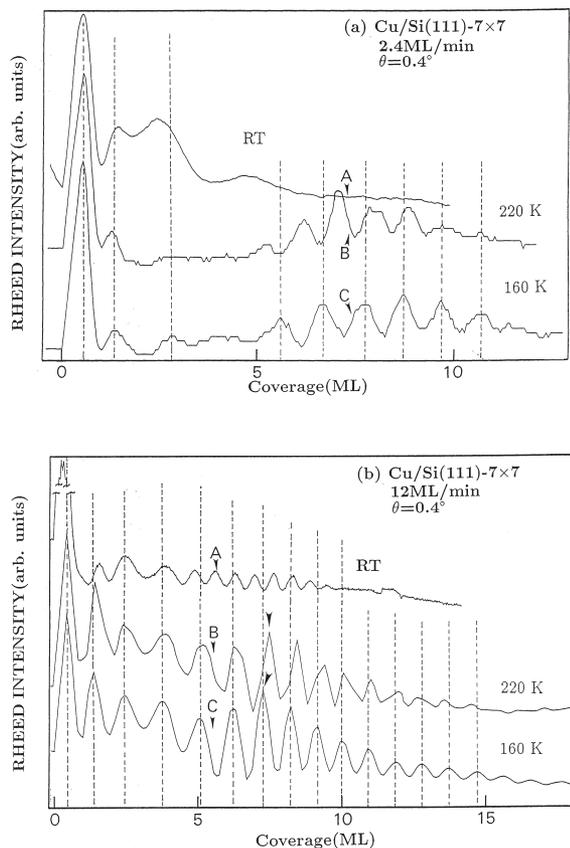


Fig. 6. (a) RHEED intensity oscillations during Cu deposition onto the Si(111)-(7 × 7) surface at RT, 220 and 160 K with the different deposition rates; (a) 2.4 ML min⁻¹. A is the same data with that at RT in Fig. 4. (b) 12 ML min⁻¹.

Fig. 6b shows the measured intensity oscillations at a deposition rate of 12 ML min⁻¹. The oscillations are clearly present, even at RT > 10 peaks are observed. In particular, the oscillations below 8 ML at 220 and 160 K are irregular. Above 8 ML, the oscillations become regular with the period of 1.0 ML. This period is smaller than the thickness 2.24 ML of Cu(111). As shown by arrows in curves B and C at 160 and 220 K, there is a phase shift at the peak position. At RT, the peak periods after 3 ML are very small, and the oscillations disappear quickly.

Fig. 7 shows the intensity changes by an interruption during the growth. Fig. 7a is the same data with C in Fig. 6b. We stopped the deposition at the top of the second peak (arrow) by a shutter

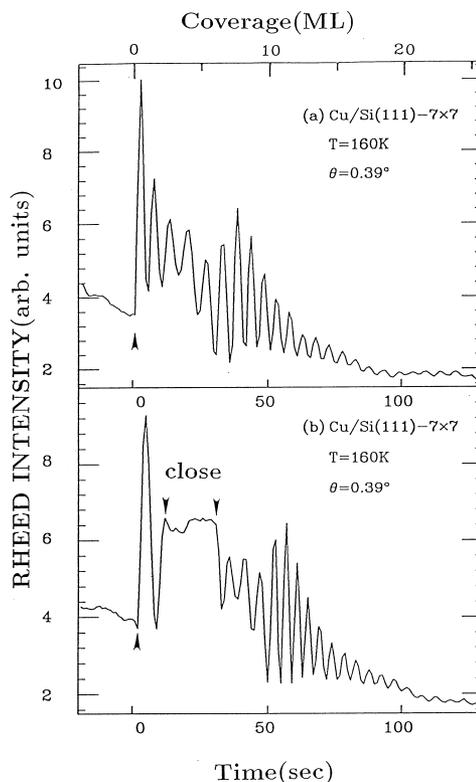


Fig. 7. RHEED intensity changes after the interruption during growth of Cu. (a) Without the interruption. The same data with C in Fig. 6b. (b) With interruption.

for ~20 s, then restarted the deposition. The measured oscillations are shown in Fig. 7b. The oscillations indicate that the interruption does not affect the intensity oscillations. This means that the diffusion of atoms is not significant during the interruption in comparison with the Ag growth. In the case of Ag, a detectable change in oscillations after the interruption was measured. The diffusion of Cu atoms at 160 K seems to be smaller than that of Ag atoms because of the silicide formation. The Cu–Si interaction in the silicide is relatively strong.

Fig. 8a is a RHEED intensity oscillation during growth of Cu onto the Si(111)-(7 × 7) at 120 K at a deposition rate of 15 ML min⁻¹. Fig. 8b is an enlarged figure only for the initial stage, the irregular oscillations can be clearly seen. The intensity decreases first when starting the deposition. The

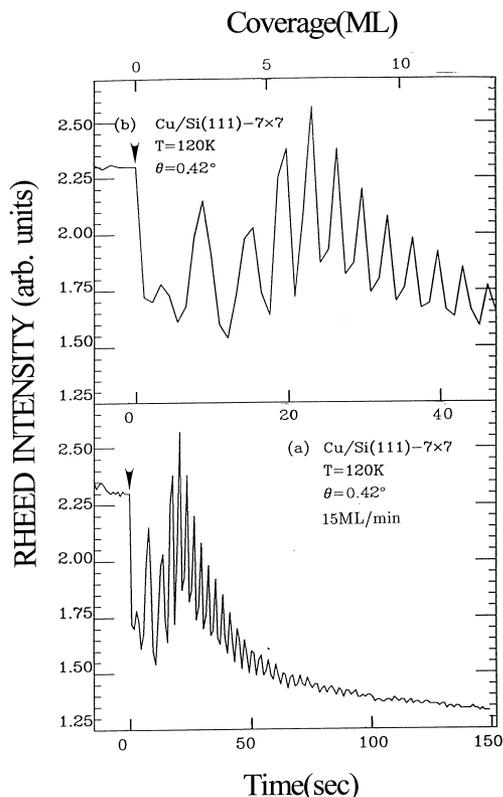


Fig. 8. (a) A RHEED intensity oscillation during growth of Cu onto the Si(111)-(7 × 7) surface at 120 K with a deposition rate of 15 ML min⁻¹. (b) An enlarged figure of (a).

oscillations are irregular up to the fifth peak, and >46 peaks are observed. Above 6 ML, the periods become regular. In this case, we observed that the intensity first decreases, and that there is about a 1/2 ML phase difference compared to that seen in Fig. 6b. This initial decrease in the intensity is in agreement with a kinematics model. However, we observed that the intensity first rises upon dosing in Fig. 6, which is not able to be explained only by considering the kinematics model. This discrepancy is attributed to the various dynamic effects with the different incident angles [25,26]. The dynamic calculations for the homoepitaxial growth of Si on a Si(111) surface showed that the intensity possibly first decreases or increases as a function of incident angles. In our measurements, the incident angle of the electron beam was set $<0.5^\circ$ at which the dynamic calculations exhibit that the

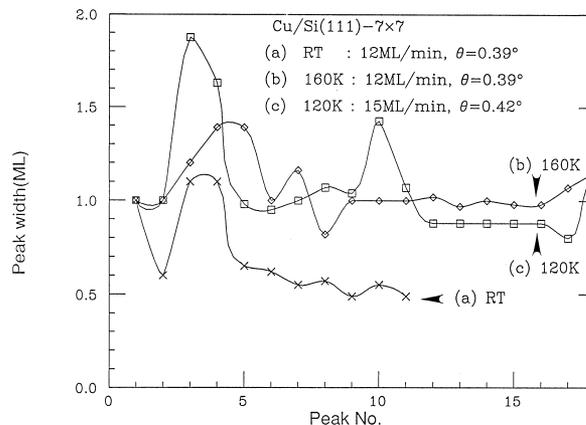


Fig. 9. A change of periods against the peak numbers of intensity oscillations at RT, 160 and 120 K.

each period of the oscillation contains double peaks with a very sharp minimum at the start of each period [25,26]. As can be seen in Figs. 7 and 8, there is actually a slight asymmetry for some measured peaks including a small peak shoulder at start of the peaks, which might correspond to that sharp minimum due to the dynamic effects at the incident angle $<0.5^\circ$. One of the reasons why we can hardly see the minimum peak at the start might be related to our data gathering resolution not being as fast as the dropping rate of that minimum peak. However, the theoretical calculations performed show that such dynamic effects do not change the oscillation period that always occurs in 1 ML [25,26]. Therefore, in our experiment the peak period of oscillations corresponds to the completion of layers no matter how the intensity first increases or decreases. Thus, by carefully analyzing the oscillation spacing, the surface morphology can be understood. Silicide formation containing a various composition of Cu would essentially cause the change in the layer thickness. The oscillation spacing should exhibit such a change in the thickness.

Fig. 9 shows a relation of the peak numbers and the observed oscillation periods at RT, 160 (Fig. 6b) and 120 K (Fig. 8). The periods are always irregular between the third and sixth peak, and become regular after the sixth peak. The period of each peak after the sixth peak corresponds to 1 ML, with the exception of the case at

RT. However, the period of each peak is smaller than the 2.24 ML thickness of Cu(111).

4. Discussion

4.1. Silicide formation

The Cu/Si(111) system at RT was investigated by AES, PES and STM [1–5,7]. AES indicated that Si atoms have a solubility of ca 30% into Cu layers for low coverage, and the possibility for either the formation of three-dimensional islands or a stable fcc α -Cu(111) phase with the amount of Si below 10% for low coverage, which has almost the same lattice constant as the Cu(111). PES qualitatively showed that the Cu–Si silicide forms within 12 ML even at RT. Both AES and PES consistently showed the existence of the composition gradient at the Si–Cu interface at least within 5 ML. STM investigations showed that by the deposition of 3 ML the Si surface was covered entirely with irregularly shaped three-dimensional islands, which were supposed to be in connection with the Cu silicide formation [4,5]. On the contrary, during the growth of Cu at low temperature, AES also showed the occurrence of the intermixing interface of Cu–Si even at 100 K [14]. From further AES investigations to this interface at 123 K, the Cu–Si intermixing was measured up to 8 ML where the Si Auger signal became weaker after 4 ML and disappeared at 8 ML [12], while such an intermixing at RT was probed up to 60 ML.

In our experiment, the measured Cu–Cu atomic distances during growth are smaller than the bulk value 2.56 Å of the Cu(111) surface, and are contracted by 13.4, 7.0, 3.5, 5.1 and 4.3% for the thicknesses of 1, 1.5, 2, 4 and 10 ML, respectively. In the same way, the measured atomic distances at 160 K are contracted by 6.4, 5.0, 5.0, –1 and –1% for the thicknesses of 1, 2, 3, 4 and 5 ML, respectively. Taking the silicide formation into account, we conclude that the changes in the Cu–Cu atomic distance are due to the silicide formation [1,3]. In our case, the measured atomic distance at 160 K is close to the lattice constant 2.47 Å of the η -Cu₃Si phase. However, AES results

suggested that the Cu–Si intermixing corresponds to the α -Cu(111) phase containing <10% of Si.

4.2. Irregular periods in RHEED intensity oscillations and a growth mode of Cu/Si(111)

When the deposition rate is increased to 50 ML min⁻¹, as can be seen in Fig. 5a, a number of intensity oscillations are observed, even at RT. This could be understood by considering that collisions between surface atoms might become frequent and the formation of two-dimensional nuclei on terraces might be increased when increasing the deposition rate. Therefore, the change in the intensity oscillations while increasing the deposition rate to 50 ML min⁻¹ and holding the substrate at RT was seen to be identical to intensity oscillation changes observed when the temperature is decreased from RT to 160 K.

As can be seen in Fig. 6a–b, the oscillations obtained at 220 and 160 K are irregular in their period as well as peak height within 8 ML, and beyond 8 ML, the oscillations become regular. At the stage of the first 8 ML, silicide formation occurs conclusively, which was not seen in the Ag/Si(111) interface. This indicates that the growth of Cu on Si(111) is fully or near fully different from Ag growth. Upon recent theoretical considerations as mentioned above, dynamic effects may not have a significant contribution to the RHEED intensity period. Hence, we conclude that the observed irregular oscillations in our experiment are predominantly due to silicide and the possible defect formation. Based on such a conclusion, we propose a growth model of Cu on the Si(111)-(7 × 7) surface as shown in Fig. 10.

Fig. 10a–b are schematics of the growth for 1–8 ML where the layers consist of a silicide, including a number of defects. At this stage of the growth, AES experimentally indicated that the silicide layers are within a gradient of Si up to 8 ML [12]. In addition, a number of defects might be generated in the grown silicide layers and the top layer of Si(111). STM images clearly showed that the Si(111)-(7 × 7) surface could be significantly generating a large number of defects even though only contaminated by a sub-monolayer of Cu [4,5]. These generated defects might be devel-

Cu/Si(111)-7×7 160 K

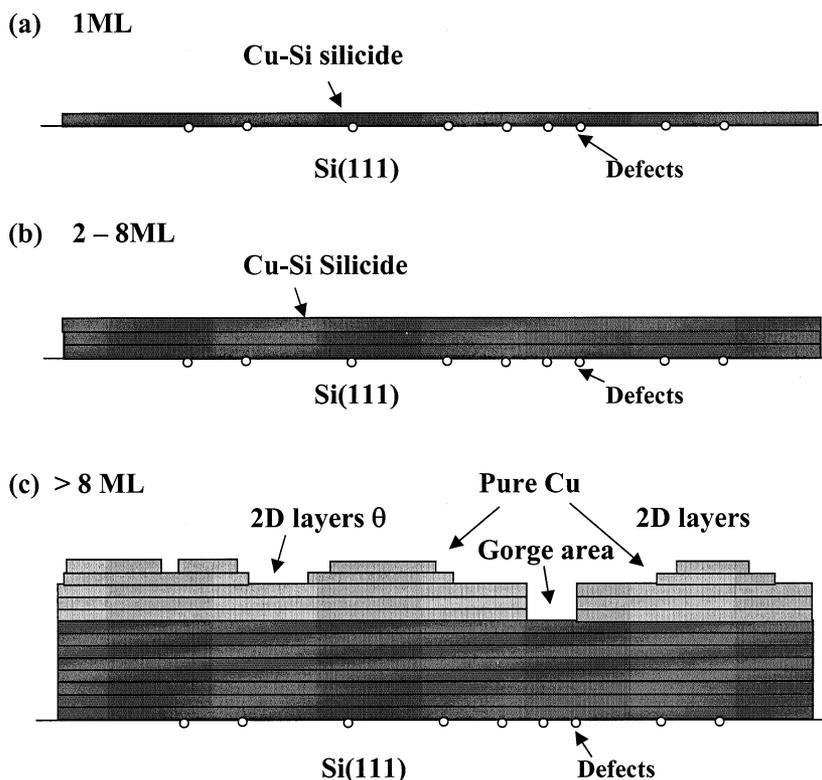


Fig. 10. A proposed growth model of Cu on the Si(111)-(7×7) surface at 160 K.

oped in the formed silicide layers during the growth. For instance, as shown in Fig. 9, the maximum irregular periods at 160 and 120 K are ca 1.5 and 1.9 ML. The real thickness, because of a dissolved composition of 25% Si, would actually be 1.9 and 2.4 ML, when 1.5 and 1.9 ML were measured. This real thickness is smaller than or equal to the thickness of 2.4 ML of η -Cu₃Si, indicating the possible occurrence of defects in the layers during growth. One or more such silicide phases and defect formation may cause any manner of strange looking “oscillations”.

Fig. 10c shows a growth model beyond 8 ML growth of Cu. As can be seen in Fig. 6b, the oscillations at this stage become regular. We assume that the formed Cu layers consist of almost pure Cu(111), because AES does not manifest further silicide formation at this stage either. In

our experiment the observed period is 1 ± 0.15 ML (the estimated experimental error for thickness is $\pm 15\%$), smaller than the 2.24 ML thickness of one atomic layer of Cu(111). This means the growth is not an ideal layer-by-layer growth. Also the relatively large difference between the measured period of oscillations and the ideal thickness of Cu(111) could not be explained only by considering the defect formation as we assumed in the Ag case, where the measured difference is only ca 17% [13]. The growth then might be accompanied with a partly two-dimensional-layer, a gorge area between partially two-dimensional layers in which the Cu atoms do not epitaxially grow, and generation of a number of defects. As mentioned in section 3.1, such partially layers are two-dimensional nanocrystals of Cu(111). In addition, the growth of Cu layers occurs in a multilayer growth

as shown in Fig. 10c, since the calculations for RHEED intensity oscillations show damping of intensities due to the multilayer growth [19–22]. For an ideal layer-by-layer (two-dimensional) growth, the RHEED intensity in a kinematics approximation oscillates as [19,20,27]

$$\frac{I}{I_0} = (1 - 2\theta_0)^2,$$

θ_0 is the ideal coverage of a layer. The intensity for such an ideal two-dimensional-layer growth oscillates with the coverage, that is, 1 ML. In contrast, for a three-dimensional-island growth, the intensity will be exponentially decreased and written by [19,20,27]

$$\frac{I}{I_0} = \exp(-4\theta_0),$$

θ_0 is the total dose of grown Cu expressed in monolayers. The intensity will exponentially decrease with the growth. If the growth is not either the ideal two-dimensional-layer or three-dimensional-island growth, it is possible to have an intermediate or transition growth mode, a partially two-dimensional-layer to have occurred. The RHEED intensity, therefore, might be a result of intensity oscillations with the real coverage θ ($\theta < \theta_0$) for a partly layer growth, and the intensity also might be damping due to the multilayer growth [19–22], which corresponds to an exponential function. As a sum of the oscillating and exponential functions, the intensity will vary by following a damping oscillation with the real coverage θ . The oscillation period will eventually be smaller than that of an ideal layer of Cu, 2.24 ML. In our proposed model, we suppose there is ca 45% of grown Cu involved in the gorge area and defect formation, and the remaining 55% of grown Cu contributing to the two-dimensional nanocrystal layer growth. In this case the estimated oscillation period will be $55\% \times 2.24 = 1.23$ ML, which is very close to 1 ± 0.15 ML of the period that we measured in our experiment. Hence we can qualitatively explain why the oscillation period is less than that of the ideal Cu(111) layer.

5. Summary

In conclusion, the measured Cu–Cu atomic distances of the surface layer deposited at RT and 160 K are smaller than the lattice constant of 2.56 Å up to 10 and 3 ML, respectively. This change is attributed to the silicide formation. The change in the intensity oscillations when the temperature is decreased from RT to 160 K was seen to be identical to intensity oscillation changes observed while increasing the deposition rate to 50 ML min⁻¹ and holding substrate at RT. The intensity oscillations are irregular up to 8 ML, and become regular >8 ML. The regular period after 8 ML is smaller than the thickness of the Cu(111) layer. A model for growth of Cu on Si(111)-(7 × 7) surface at 160 K is proposed. The changes in periodicity are due to the silicide, defect formation and partially two-dimensional nanocrystal layer growth.

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