Homogeneous and heterogeneous nucleations in the surface phase transition: Si(111)4 × 1-In

Hyungjoon Shim, Youjin Jeon, Jonghoon Yeo and Geunseop Lee
Department of Physics, Inha University, Incheon 402-751, Korea
E-mail: glee@inha.ac.kr

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Abstract
Homogeneous and heterogeneous nucleations in a reduced-dimensional system undergoing a first-order structural phase transition were examined by using low electron energy diffraction and scanning tunneling microscopy. The high-temperature 4 × 1 phase of a Si(111)-In surface was supercooled at temperatures below the transition temperature ($T_c$) and evolved slowly into a low-temperature 8 × 2 phase with time. The transition rate decreased significantly as the temperature approached $T_c$. The kinetics of the observed homogeneous nucleation was analyzed by classical nucleation theory. The introduction of oxygen atoms reduced the hysteresis and accelerated nucleation significantly, showing that the $T_c$-raising oxygen impurity plays the role of a nucleation seed for heterogeneous nucleation.

1. Introduction
Phase transitions in solids have been a subject of fundamental research in statistical physics and condensed-matter physics. Over the past few decades, the observations of both structural and electronic transitions in a number of metal/semiconductor surfaces have renewed interest in the phase transitions in reduced dimensions [1–8]. A study of the phase transitions at the surfaces has fundamental importance in understanding novel physics, which is richer than in the bulk, because of the enhanced electronic correlation and electron–phonon coupling [9]. A Si(111)-In surface composed of an array of indium atomic chains is a prototypical example undergoing a symmetry-breaking (4 × 1-to-8 × 2) structural phase transition, accompanied by a metal-insulator transition at approximately 120–130 K [3]. Interesting phenomena, such as phase separation [10–13] and topological soliton excitations [11, 14–16], have also been reported in this system.

Extensive studies of the Si(111)-In surface have focused on the nature of the phase transition, including the driving mechanism [3, 17–20] and transition order [11, 12, 19, 21]. Two different views exist on the nature of the transition. One is a charge density wave (CDW) transition scenario that the high-temperature 4 × 1 phase has a static structure and transforms to the CDW with an 8 × 2 structure at low temperature [3, 19, 22] (see figure 1(a)). This was attributed to the Peierls instability (or Fermi-surface nesting) inherent in the quasi-one-dimensionality of the system [23]. Another view contends that the 8 × 2 phase is stabilized by simple energy lowering caused by lattice distortions, and the 4 × 1 phase is an incoherent dynamical fluctuation of the 8 × 2 phase [17, 18, 20]. Both views, despite their differences, assume a continuous transition [11, 24].

On the other hand, there are a number of experimental findings in favor of the first-order nature of the phase transition [12, 21, 25]. Recently, observations of the long-lived electronically excited 4 × 1 phase at 20 K [26] and the thermal hysteresis in the transition [27] have shown the existence of an energy barrier inherent to the first-order transition. Therefore, this transition is expected to reveal interesting phenomena pertinent to the first-order transition, such as supercooling and homogeneous/inhomogeneous nucleations. Thus far, these phenomena has not been explored.

In this work, we investigated the homogeneous and heterogeneous nucleations of a low-temperature 8 × 2 phase in a supercooled 4 × 1 phase of the Si(111)-In surface. The 4 × 1-to-8 × 2 transition at supercooling temperatures proceeded slowly with time. This homogeneous transition rate decreased significantly as the supercooling temperature approached the genuine transition temperature ($T_c$). Its temporal and temperature...
dependence was analyzed reasonably by employing the classical nucleation theory (CNT). In addition, the introduction of oxygen impurities reduced the hysteresis and expedited nucleation of the $8 \times 2$ phase by reducing the activation barrier to the transition. It indicates that the oxygen impurities act on the nucleation seeds for heterogeneous nucleation. In comparison, indium adatoms did not induce nucleation.

2. Experimentals

The experiments were performed by using low energy electron diffraction (LEED) and scanning tunneling microscopy (STM) in separate ultrahigh vacuum chambers with base pressures below $1.2 \times 10^{-10}$ Torr. The Si (111)$4 \times 1$-In surface was prepared by depositing a monolayer of In onto a Si(111) substrate held at 700–750 K. For the LEED measurements, the sample temperature was adjusted by dropping liquid nitrogen (LN2) continuously into cryostat for cooling and stopping its supply for heating. The temperature was monitored by a thermocouple attached to a sample holder connected to the cold finger, and calibrated carefully with the true sample temperature. The cooling and heating rates of the sample were controlled with $0.5–2$ K min$^{-1}$. LEED patterns were recorded continuously using a charge-coupled device camera at different sample temperatures. For the STM measurements, the sample temperature was adjusted by the amount of LN2 supplied to the cryostat and by an ohmic heater at the cold finger. For the isothermal measurements in both LEED and STM, the temperatures were maintained to be stable within 0.2 K. Oxygen impurities were introduced on the surface by backfilling the chamber with oxygen gas through a variable leak valve.

3. Results and discussion

Figure 1(b) shows the changes in the LEED intensities of the $4 \times$ spots, $8 \times$ spots, and $\times 2$ streaks through a structural phase transition during a cycle of cooling and heating the sample\(^1\). The LEED intensities were obtained by integrating the intensities of the regions surrounded by the circles for the spots and the rectangle for the streak shown in figure 1(a), with a background intensity (determined by a mean intensity on the boundary) subtracted. The Debye–Waller effect was not corrected. As the temperature was varied, steep and hysteretic

\(^1\) The effect of the LEED beam on the transition was tested by moving the beam spot over the sample. It was found to be absent or negligible, if any.
changes in the diffraction intensity were observed for the superstructure diffraction beams (both 8× spots and ∼2 streaks) from the broken-symmetry phase. The intensity-temperature curves of the 8× and ∼2 beams coincide with each other almost perfectly when their intensities are properly scaled. For convenience, the temperatures, where the superstructure diffraction intensities begin to grow during cooling and vanish during heating, are labeled as \( T_{c,\text{cooling}} \) and \( T_{c,\text{heating}} \), respectively. The genuine transition temperature in a strict thermodynamic sense, \( T_c \), where both phases have equal free energies, should lie between the two temperatures \( T_{c,\text{cooling}} < T_c < T_{c,\text{heating}} \). The hysteresis width, \( \delta T = T_{c,\text{heating}} - T_{c,\text{cooling}} \), is about 8 K \( (T_{c,\text{cooling}} = 123 \text{ K and } T_{c,\text{heating}} = 131 \text{ K}) \) for the pristine surface in figure 1(b). The hysteretic changes showed little differences when the cooling and heating speeds were varied from 0.5–2 K min\(^{-1}\). This observation agrees well with a previous study reporting the hysteresis in the 4× 1-to-8× 2 transition and thus proving that the transition was first-order \( [27] \).

Figure 1(b) further shows that similar hysteresis is also observable for the 4× spots as the intensity is changed conversely to the superstructure diffraction \( [28] \).

The structural phase transition between the 4× 1 and the 8× 2 phases was reported to be influenced significantly by impurities at the surface \( [29-33] \). Previously, the onset temperature during cooling, \( T_{c,\text{cooling}} \), was measured. With the extrinsic defects on the 4× 1 surface, \( T_{c,\text{cooling}} \) was found to be decreased in many cases (H \( [30, 32] \), Na \( [29] \), In \( [33] \)) but increased exceptionally in the case of O \( [30, 31] \). Figure 1(c) compares the temperature dependence of the 8× diffraction intensity during the cooling-heating cycle for the pristine surface and surfaces with the representative impurities, In (lowering the \( T_{c,\text{cooling}} \) \( [33] \)) and O (raising the \( T_{c,\text{cooling}} \) \( [30, 31] \)). Both \( T_{c,\text{cooling}} \) and \( T_{c,\text{heating}} \) were shifted by the introduction of impurities. These shifts suggest that the genuine transition temperature, \( T_c \), should also be altered: \( T_c \) was reduced by In adsorption and increased by O adsorption. In addition, the hysteresis was also affected differently. In the case of the O-added surface, the hysteresis was reduced significantly. In comparison, the In-added surface showed a marginal increase. The results suggest that the energy barrier between the 4× 1 and the 8× 2 phases near the transition is reduced upon adsorption. In addition, the hysteresis was also affected differently. In the case of the O-added surface, the hysteresis was reduced significantly. In comparison, the In-added surface showed a marginal increase. The results suggest that the energy barrier between the 4× 1 and the 8× 2 phases near the transition is reduced upon adsorption.

Hysteresis in the first-order transition is caused by the slow kinetics relative to the fast change in temperature during the measurement. Therefore, the pristine surface remaining as the 4× 1 phase at temperatures between \( T_{c,\text{cooling}} \) \( (123 \text{ K}) \) and \( T_{c} \) (to be determined) during cooling was supercooled. On the other hand, this supercooled 4× 1 surface will eventually turn into a 8× 2 surface. Figure 2(a) shows such temporal evolution for the supercooled surfaces. The changes in the 8× LEED intensities were monitored over a long time (more than three hours) for the surface maintained at different supercooling temperatures. The rates of the 4× 1-to-8× 2 transition (determined by the initial slopes) showed a strong decrease with the supercooling temperature approaching \( T_{c,\text{heating}} \).

The first-order transitions with hysteresis are found frequently in materials undergoing liquid–solid or solid–solid structural transformations \( [34] \). An analogy of the 4× 1-to-8× 2 surface transition to liquid–solid structural transformation was assumed for a detailed analysis of the transition kinetics. The kinetics of the phase transformation is governed by two kinetic factors; nucleation and growth. According to the classical theory of crystallization (liquid–solid transformation) kinetics, the fraction of the transformed phase \( \alpha (t) \) under isothermal conditions can be described by the Johnson–Mehl–Avrami–Kolmogorov (JMAK) formulae \( [35-37] \), which is given by the following:

\[
X(t) = 1 - e^{-Kt^n},
\]

where the coefficient, \( K \), includes information of both the nucleation and growth rates, and the Avrami coefficient, \( n \), depends on the nucleation and growth mechanisms as well as the dimensionality of the system. In the initial stage of the transition \( (small \ t) \), the JMAK formulae reduces to \( X(t) = Kt^n \). The application of the JMAK formulae to the data in figure 2(a) suggests that \( n = 1 \) for the 4× 1-to-8× 2 surface transition. Figure 2(b) presents the values of the transition rate \( K \) \( (the \ initial \ slopes \ of \ the \ intensity-temperature \ data \ in \ figure \ 2(a)) \) for different supercooling temperatures. Although this transition rate includes the information of both nucleation and growth, its temperature-dependence is dominated by the nucleation rate. This is because the nucleation rate as a function of \( 1/(T_c - T) \) varies significantly in the narrow supercooling temperature range \( (123–131 \text{ K}) \) of the present experiment. In contrast, the growth rate changes slowly as a function of \( 1/T \).

An attempt was made to fit the initial 4× 1-to-8× 2 transition rate in figure 2(b) in the framework of the CNT for the solid-phase formation in the ambient liquid phase in the structural transition \( [38] \). The temperature dependence of the transition rate \( K(T) \), being dominated by the homogeneous nucleation rate \( I(T) \), is given by

\[
K(T) \sim I(T) = I_0 e^{-W(T)/T},
\]

where \( I_0 \) is a preexponential term, which depends weakly on temperature, and \( W(T) \) is the activation barrier of nucleation, i.e., the work needed to form a critical nucleus which can grow spontaneously to form the new phase. Normally, \( W(T) \) is temperature-dependent and given by \( W(T) = W_0 [T_c/(T_c - T)]^\alpha \) (\( \alpha \) is a dimension-
dependent exponent: $\alpha = d - 1$ for the system with the generalized dimensionality, $d$. See appendix.) In the structural transformation between the liquid and solid in the bulk, $T_c$ is the melting temperature and $\alpha = 2$. In the present case, the best fit of the data to the formulae resulted in the values of $T_c = 130.5 \pm 1.5$ K and $\alpha = 0.35 \pm 0.07$. The value of $T_c$ is reasonable in that it lies between the $T_c^{\text{cooling}}$ and $T_c^{\text{heating}}$, being quite close to the $T_c^{\text{heating}}$. This suggests that the $8 \times 2$ phase is barely superheated. This is presumably because the intrinsic defects (mostly In vacancies) act as heterogeneous nuclei for the $8 \times 2$-to-$4 \times 1$ transformation during heating. A value of $\alpha < 1$ suggests that the dimensionality of the transition is between one and two. This is also reasonable because the two-dimensional $4 \times 1$-to-$8 \times 2$ transition is highly anisotropic, i.e. quasi-one dimensional.

From the fitting parameters\(^2\), the activation barrier of nucleation $W(T)$ was calculated and is plotted in figure 2(c). The temperature dependence of this kinetic barrier height obtained for the $4 \times 1$-to-$8 \times 2$ transition clearly shows the rapid increase as the supercooling temperature approaches the $T_c$. This analysis shows that the $8 \times 2$ phase is barely superheated. This is presumably because the intrinsic defects (mostly In vacancies) act as heterogeneous nuclei for the $8 \times 2$-to-$4 \times 1$ transformation during heating. A value of $\alpha < 1$ suggests that the dimensionality of the transition is between one and two. This is also reasonable because the two-dimensional $4 \times 1$-to-$8 \times 2$ transition is highly anisotropic, i.e. quasi-one dimensional.

The nucleation kinetics, and the transition kinetics as well, at the supercooled temperature can be enhanced by introducing impurities. A well-known, prototypical example is the water-to-ice transition where supercooled pure water freezes immediately when a small amount of tiny particles are added. Similar impurity-induced heterogeneous nucleation in the first-order transition occurs for the supercooled $4 \times 1$ surface. Figure 3 shows the change in the intensity of the $8 \times 8$ LEED spot measured as a function of the amounts of impurities, oxygen and indium, introduced on a supercooled $4 \times 1$ surface maintained at 128 K. When the surface was exposed to oxygen gas at this supercooling temperature (inside the hysteresis region of the pristine surface), the $8 \times 2$ LEED spots developed abruptly. The $8 \times 2$ LEED intensity grew with the increasing O dose until it was saturated. In contrast, introduction of the In impurities did not develop an $8 \times 2$ LEED intensity. This clearly shows that the oxygen impurity plays the role of a nucleation seed, which reduces the activation barrier height and expedites the nucleation kinetics [26, 39, 40], whereas the In impurity does not. The O-induced decrease in the barrier height of nucleation can be also confirmed by the significant increase in the transition rate of the O-adsorbed surface compared to that of the pristine surface (see figures 2(a) and (b)). The barrier height of nucleation at 124.5 K showed an approximately 10% reduction by the O adsorption of the 0.2 L.

\(^2\) The fitting parameters $W_0$, $T_c$, and $\alpha$ were obtained from the fitting to the logarithm of the formula (2) for the better convergence.
The contrasting roles of oxygen and indium in the nucleation kinetics is in line with their contrasting effects on the transition temperature \( T_c \); \( T_c \)-raising oxygen versus \( T_c \)-reducing indium \([30, 33]\).

The adsorbed O as a nucleation seed can be visualized by monitoring the O-induced nucleation of the 8 × 2 phase in the supercooled 4 × 1 surface in real space, providing deeper insights. Figure 4 shows a series of STM images taken over the same area at 127 K while the supercooled surface was exposed to oxygen gas. The pristine surface, prepared to be free of In adatoms, typically contains many vacancy defects (inset in figure 4(a)). At this supercooling temperature, the surface remained in the 4 × 1 phase for an extended time (more than two hours) without forming an 8 × 2 phase. This indicates that vacancy defects do not induce nucleation of the 8 × 2 phase at the supercooling temperature. On the other hand, once the dosing of oxygen gas started, the surface exhibited immediate formation and evolution of the 8 × 2 areas as the number of O defects (bright and dark features...
shown in the inset in figure 4(b) [41] increased. Figures 4(b)–(e) show that the 8 × 2 domains were formed over the area where the adsorbed O features were close to each other. They grew both in size and number with increasing O adsorption. This clearly reveals the enhanced heterogeneous nucleation kinetics, demonstrating the role of oxygen inducing nucleation and condensation of the 8 × 2 phase.

The results obtained from the LEED and STM studies reveal the kinetics of the 4 × 1-to-8 × 2 transition of the Si(111)-In surface. In the mean-field theory of the phase transition, the whole system is assumed to be uniform and take a specific phase at each stage of the transition. In reality, however, the first-order phase transition proceeds through nucleation and growth. Therefore, the system becomes intrinsically non-uniform and consists of nuclei of one phase (8 × 2) immersed in the other phase (4 × 1) unless it is in thermodynamic equilibrium. The kinetics of the first-order 4 × 1-to-8 × 2 transition can be described by the change in the total free energy of the non-uniform system due to the formation of 8 × 2 nuclei in an otherwise uniform 4 × 1 phase.

Figure 5(a) shows the free-energy change (ΔG) as a function of the average size (r) of the 8 × 2 nuclei generated in the otherwise uniform 4 × 1 phase. (b) Energetics schematically describing both the thermodynamic free-energy difference (ΔGℓ = G8×2ℓ − G4×1ℓ) between uniform 4 × 1 (r = 0) and 8 × 2 (r → ∞) phases and the kinetic barrier (W for r*) separating them: a pristine surface versus an O-adsorbed surface at T < Tc.

4. Conclusions

This study examined the homogeneous and impurity-induced heterogeneous nucleation kinetics of the 8 × 2 phase in the supercooled 4 × 1 phase of the Si(111)-In surface. For a pristine 4 × 1 surface, the kinetics of the

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3 The low-temperature STM images reveal that the oxygen impurities are not hopping but displaced from the symmetric sites along the chain direction in concert with the developing 8 × 2 ordering. The details will be published elsewhere.
temperature-dependent rate of the transition into the 8 × 2 phase was found to be well captured by the classical theory of homogeneous nucleation. In addition, oxygen impurities expedited the 4 × 1-to-8 × 2 transition by acting as nucleation seeds for heterogeneous nucleation.

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Appendix. Nucleation kinetics of phase transformation in d-dimension

Consider a d-dimensional system in a high-temperature phase, phase-I (e.g., liquid or 4 × 1 phase) cooled to a supercooling temperature \( T^\text{cooling} < T < T_c \). By employing the same approach of the CNT developed for a three-dimensional bulk system [38], the temperature dependence of the activation energy barrier to form a nucleus of a low-temperature phase, phase-II (e.g., solid or 8 × 2 phase) with a critical size (radius, \( r^* \)) was derived. The formation of a phase-II nucleus leads to a change in free energy \( (G) \),

\[
\Delta G = -V|\Delta G_\text{II}| + S_\gamma,
\]

where \( V \) is the volume of the phase-II nucleus, \( S_\gamma \) is the phase-II/phase-I interfacial area, \( \gamma (>0) \) is the phase-II/phase-I interfacial energy per unit area, and \( \Delta G_\text{II} = G_\text{II} - G_\text{I} (<0) \) is the free energy per unit volume at \( T < T_c \) between the two phases. For an isotropic nucleus with radius \( r \) in d-dimension, \( V = A r^d, S = B r^{d-1} \), where \( A \) and \( B \) are coefficients. Then

\[
\Delta G = -A r^d \Delta G_\text{II} + B r^{d-1} \gamma.
\]

The condition, \( \frac{d(\Delta G)}{dr} = 0 \) at a critical size \( r = r^* \), leads to

\[
r^* = \frac{d - 1}{d} \frac{B \gamma}{A |\Delta G_\text{II}|},
\]

and

\[
W(T) = \Delta G^* = \frac{(d - 1)r^{d-1}}{d A |\Delta G_\text{II}|} \frac{(B \gamma)^d}{r^{d-1}},
\]

where \( W(T) \) is a activation barrier to nucleation at temperature \( T \). Since \( |\Delta G_\text{II}| \) is proportional to supercooling \( (\Delta T = T_c - T) \) below the transition temperature, the temperature dependence of \( W(T) \) is given by

\[
W(T) \sim \frac{1}{(|\Delta G_\text{II}|)^{d-1}} \sim \frac{T_c}{(T_c - T)^\alpha} \quad (\alpha = d - 1).
\]

Therefore, \( \alpha = 2, 1, \) and \( 0 \) for the three, two, and one dimensional systems, respectively. For the 4 × 1-to-8 × 2 transition, which is a two-dimensional transition in the highly anisotropic, quasi-one dimensional system, values of \( \alpha \) between 0 and 1 are expected.

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