Atomic displacements and electronic properties for 4×1-8×2 phase transition of In-adsorbed Si(111) surface

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Abstract. The In-adsorbed Si(111) 4×1 reconstructed surface is known to undergo a phase transition to an 8×2 structure at approximately 130 K. This structural transition accompanies a metal-insulator transition known as the Peierls transition or charge density wave (CDW) transition. We construct a continuous displacement model and discuss the surface electronic states using a tight-binding model based on the first principles calculations. We performed Monte Carlo simulations, estimated the atomic displacement, and calculated the surface electronic properties around the transition temperatures, in terms of the density of states projected into the reciprocal space (RDOS). The results show that the nature of the transition can be understood as the order-disorder transition of an Ising system.

1. Introduction
The In-adsorbed Si(111) 4×1 reconstructed surface is known to undergo a phase transition to an 8×2 structure at approximately 130 K [1]. The In adatoms form quasi one-dimensional chains, and this structural phase transition accompanies the metal-insulator transition known as the Peierls transition or the charge density wave (CDW) transition [2] - [7]. We previously discussed the properties of this transition with a simple Ising model [8]. In this paper, we extend the Ising model to a continuous displacement model to check the validity of the Ising model analyses and describe other models for discussing the details of this phase transition, particularly the surface electronic properties.

2. Theoretical Models and Monte Carlo simulation
2.1. Displacement model
We select a 4×1 cell as the unit cell and introduce two order parameters in the unit cell. The order parameters take continuous values, and can be approximately considered to represent the displacement of In atoms along the atomic chain. The total energy can be expanded with the order parameters, by optimising the surface structure for given displacements of the In adatoms. Such expansion gives an effective Hamiltonian [9] as

$$H_{\text{lat}} = \sum_{\ell,m,\nu} \left( \varepsilon_2 u_{\ell,m,\nu}^2 + \varepsilon_4 u_{\ell,m,\nu}^4 + J u_{\ell,m,\nu} u_{\ell+1,m,\nu} \right)$$
\[ + K \sum_{\ell, m} \left( u_{\ell, m, 0} u_{\ell, m, 1} u_{\ell, m+1, 0} u_{\ell, m+1, 1} + u_{\ell, m, 0} u_{\ell-1, m, 1} u_{\ell, m+1, 0} u_{\ell-1, m+1, 1} \right) , \]  

where \( u_{\ell, m, \nu} \) is the \( \nu \)-th order parameter in cell \((\ell, m)\), \( \ell \) is the index along the chain and \( m \) is the index across the chain, as shown in figure 1. Here, \( \nu \) takes either value, 0 or 1.

2.2. Tight-binding model

Once the positions of the In atoms are determined in terms of the order parameter, the surface electronic states of the system are calculated based on a tight-binding model. The two localised surface electronic states induced by the two adsorbed In atoms in the unit cell give rise to two electronic states of the system. The Hamiltonian of these surface electronic states is given by

\[ \mathcal{H}_{\text{ele}} = \sum_{\ell, m, \gamma} \epsilon^{(\gamma)}_{\ell, m} c^{(\gamma)\dagger}_{\ell, m} c^{(\gamma)}_{\ell, m} + \sum_{\ell, \ell', m', \gamma} \left( t^{(\gamma)}_{\ell, \ell', m, m'} c^{(\gamma)\dagger}_{\ell, m} c^{(\gamma)}_{\ell', m'} + \text{h.c.} \right) , \]

where \( c^{(\gamma)}_{\ell, m} \) and \( c^{(\gamma)\dagger}_{\ell, m} \) are the annihilation and creation operators for state \( \gamma \) in the cell \((\ell, m)\), respectively. The indices, \( \ell \) and \( m \) are the same as in the above-mentioned displacement model. \( \epsilon^{(\gamma)} \) is the site energy for state \( \gamma \), and \( t^{(\gamma)}_{\ell, \ell', m, m'} \) is the transfer integral between states \( \gamma \) at cells \((\ell, m)\) and \((\ell', m')\).

We consider the transfer integrals up to the first neighbour in the In atomic chain and across the In atomic chain, so that the transfer integrals are given as

\[ t^{(\gamma)}_{\ell, \ell', m, m'} = -t^{(\gamma)}_{\text{inter}} \delta_{\ell', \ell} \delta_{m', m \pm 1} - t^{(\gamma)}_{\text{intra}} (\ell, m, \{ u_{\ell, m, \nu} \}) \delta_{\ell', \ell \pm 1} \delta_{m', m} , \]

where \( t^{(\gamma)}_{\text{inter}} \) is the interchain transfer and \( t^{(\gamma)}_{\text{intra}} (\ell, m, \{ u_{\ell, m, \nu} \}) \) is the intrachain transfer integral for state \( \gamma \), respectively. The intrachain transfer depends on the order parameters as

\[ t^{(\gamma)}_{\text{intra}} (\ell, m, \{ u_{\ell, m, \nu} \}) = t^{(\gamma)}_{0} - \tau (\sigma_{\ell+1, m} - \sigma_{\ell, m}) , \]

through the quantity \( \sigma_{\ell, m} \) defined as \( \sigma_{\ell, m} = 2u_{\ell, m, 0} - (u_{\ell, m, 1} + u_{\ell-1, m, 1}) \), where \( t^{(\gamma)}_{0} \) and \( \tau^{(\gamma)} \) are constants. Parameter \( \tau^{(\gamma)} \) determines the nature of the Peierls transition.

2.3. Monte Carlo simulation

| Table 1. Parameters used in computer simulation. Unit is meV. |
|-----------------|----------------|--------------------|----------------|----------------|----------------|-----------------|-----------------|
| \( \varepsilon_{2} u^{2}_{0} \) | \( \varepsilon_{4} u^{4}_{0} \) | \( J_{u} u^{2}_{0} \) | \( K u^{2}_{0} \) | \( \epsilon^{(0)} \) | \( \epsilon^{(1)} \) | \( t^{(0)}_{0} \) | \( t^{(\gamma)}_{\text{inter}} \) |
| -50.0 | 40.0 | 30.0 | 0.285 | -180 | 150 | 425 | 100 | -50 |

We performed Monte Carlo simulations to clarify structural and electronic properties related to the phase transition using the above two models with parameters listed in table 1. The parameters included in the displacement model are chosen to reproduce the transition temperature, the anisotropy of the system [8], and the total energy difference between the 4×1 and 4×2 ordered structures optimized by the first principles calculation performed by Gonzáles
et al.[10] The parameters in the tight-binding model are also chosen to reproduce the two surface electronic bands obtained by the first principles calculation [10].

The averages of properties are taken over 100,000 and 1,000 sample systems generated by the Metropolis algorithm after neglecting 100,000 and 50,000 sample systems for structural and electronic properties, respectively. The sample system contains $300 \times 30$ cells for calculating structural properties and $32 \times 8$ cells for calculating electronic properties. Note that there are two order parameters or two localised electronic states in the unit cell.

3. Calculation results

![Figure 1](image1.png)  ![Figure 2](image2.png)

**Figure 1.** Sketch of Si(111)4×1-In surface system. Rectangle is 4×1 unit cell. Arrows indicate displacement denoted by order parameters.

**Figure 2.** Distribution function of order parameter at several temperatures.

3.1. Structural properties

Figure 2 shows the temperature dependence of the distribution function of the order parameter. The distribution has two peaks even at high temperatures around the positions corresponding to the bi-stable positions subsisting in the model. This result indicates that the Ising model provides a good approach to this system.

3.2. Surface electronic properties

To discuss the electronic states in the reciprocal space for disordered structures, for which the wave number is not a good quantum number, we introduce the reciprocal-space projected density of states (RDOS) [11]. The RDOS provides information that can be compared with the angle resolved ultraviolet photoemission spectra measured in the reciprocal space. Figure 3 demonstrates the obtained RDOS for several temperatures. At high temperatures, the In displacement is mostly random, and the RDOS shows two blurry bands of metallic character. At the transition temperature, gaps begin to open at the Fermi level on the zone boundary of the first Brillouin zone of the 4×2 structure. At low temperatures, the surface is mostly ordered to the 8×2 structure, and the RDOS shows four sharp bands of insulating character with clear gaps. These results agree with the experimentally observed metal-insulator transition of this system.

4. Conclusion and summary

We performed Monte Carlo simulations with a displacement model and a tight binding model based on the first principles calculation. The simulation results show the distribution of the
displacements reflecting the bi-stability due to the Peierls instability. The nature of this transition can be understood as a kind of order-disorder phase transition occurring in a system with a strong Ising character. Although atomic displacement is not generally considered in the CDW study, the positions of atoms are strongly coupled with electronic states, so that the distribution of atomic displacement should be sufficiently considered in the experimental and theoretical analyses of diffraction and spectroscopy. In this paper, we demonstrate the temperature dependence of the surface electronic states with the RDOS. They indicate a metal-insulator transition comparable to the photoemission experiments. The same approach can also analyse other quantities, including surface conductivity and the Patterson map. Those analyses will be presented in a separate paper.

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