解説

Angle-resolved Photoemission Study of the Quantum Wires on Semiconductor Surfaces

Han Woong Yeom

Department of Applied Chemistry, the University of Tokyo

Angle resolved photoemission (ARP) for the surface studies can be in various forms of applications such as valence-band mapping, Fermi-surface mapping, angle-resolved high-resolution core-level measurements and photoelectron diffraction. Together with the other surface techniques such as STM and LEED, ARP using synchrotron radiation can provide a comprehensive atomic- and electronic-structure probes for the exotic nanostructures on semiconductor surfaces of current interests. ARP using synchrotron radiation (at Photon Factory, Max-Lab and Advanced Light Source) has been extensively applied to investigate the structural and, especially, the electronic properties of various well-ordered self-organized atomic-scale quantum wires on Si and SiC surfaces. The systems studied extend from the In (Al, Pb) quantum wires on the Si(001) surface and the In quantum wires on Si(111) to Si quantum wires on the SiC(001) surface. Although the In (Al, Pb) quantum wires on Si(001) and the Si quantum wires on SiC(001) show no strong 1D anisotropy in their electronic band structures, a clear 1D metallic band structure is observed on the In/Si(111) surface. The latter system further showed a metal-insulator phase transition of Peierls type, demonstrating the 1D physical property of the surface quantum-wire systems for the first time. The importance of the nanostructures on solid surfaces as brand new materials systems for low-dimensional condensed-matter physics studies and the crucial role of ARP using synchrotron radiation for the relevant studies are discussed.

1. Introduction

Semiconductor surfaces: the playground –Because of the lack of translational symmetry on the 'surface' of a crystal, the surface atoms form quasi two dimensional (2D) structures of their own, which are distinguished form those of 'bulk'. These quasi 2D systems, thus, have provided a variety of interesting physical phenomena, which cannot be encountered in bulk crystals[1-3]. Furthermore, the understanding of these surface phenomena has great importance in modern high-technology, for typical examples, the growth and process of electronic device materials and the catalysis in chemical engineering.

An 'ideal surface', which is formed by truncation of a bulk crystal, for example, of a covalent semiconductor with sp^3 hybridization, is left with numerous broken bonds of high free energy. The surface atoms try to rearrange themselves to reach a lower energy configuration. This rearrangement is called 'surface reconstruction' such as the 2×1 or $c(4\times2)$ structure on the Si(001) surface and the 7×7 structure on the Si(111) surface[4-8]. If extrinsic atoms, 'adsorbates', are put on a clean surface with a certain surface reconstruction, then the geometry of surface atoms can change further through bonding with adsorbates. These 'adsorbate-induced reconstructions' of surface atoms provide far more varieties of new 2D materials systems[4,5].

On the other hand, the surface reconstructions cause electronic structures specific to the surface layers. For instance, the broken bonds of surface atoms of ideal semiconductor surfaces, called 'dangling bonds (DB)', will cause new electronic states (localized bonds and also 2D extended states of 'surface-state bands') in the bulk band gap[1-3,5]. As easily expected, the surface reconstructions of clean surfaces and by adsorption will create different kinds of surface electronic orbitals, bonding or antibonding orbitals, which originate from the bonds (or interaction in general) between adsorbates and substrate atoms, between substrate surface atoms and between adsorbates themselves.

1D nanostructures on surfaces; new exotic 1D metals?– Among the new materials systems due to surface reconstructions with and without adsorbates, of particular recent interests are the 1D nanostructures (or quantum wires) formed mostly through the 'self organization' during various adsorption, growth, and reaction processes[9-40]. In general, the 1D solids, (especially the metallic 1D systems) feature exotic and attractive physical phenomena such as the instability of a metallic phase (Peierls instability), anomalous phonon dispersion (Kohn anomaly), spin or charge density wave (SDW or CDW), triplet superconductivity and non-Fermi-liquid behavior[41]. The most well-know example is Peierls instabil-

* Department of Applied Chemistry, the University of Tokyo, 1-1 Oho, Tsukuba-shi, Ibaraki 305-0801, Japan
TEL/FAX: 0298-64-5584 E-mail: yeom@sr.t.u-tokyo.ac.jp

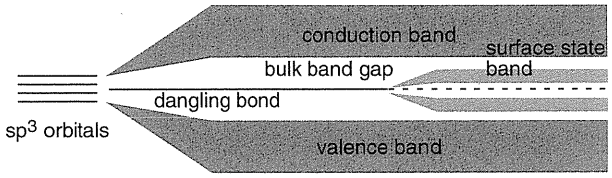

Figure 1. Schematics of surface reconstruction and surface-state band formation.

ity, where the electrons and holes near the Fermi level couple with a static lattice vibration leading to a spatial modulation of charges [charge density waves (CDW)] and to opening an electronic energy gap at Fermi level[41]. So far, only very limited number of materials have been shown to have 1D electronic properties such as a few organic conductors, Pt chain compounds (e.g. $K_2Pt(CN)_4Br_{0.3}\cdot 3.2H_2$), transition metal chalcogenides (e.g. $NbSe_3$) and transition metal bronzes (e.g. $K_{0.3}MoO_3$)[41].

In fact, surface systems are good candidates to find brand new 1D materials due to the inherent low dimensionality and due even to the possibility of atom manipulations by angstrom-resolution scanning-probe-microscopy techniques. Furthermore, in contrast to the known 1D materials, the surface 1D systems, if any, have no 3D coupling between the 1D chains, which may yield new physical properties through most probably the enhanced fluctuations. Not only elucidating the exotic 1D physical properties, but the study of atomic-scale quantum wires on semiconductor surfaces is expected to provide important building blocks for the near future nanometer-scale electronic and magnetic device technology.

Several examples of 1D nanostructures on metal and semiconductor surfaces–In recent years, indeed many atomic-scale 1D chain structures have been observed on various surfaces mostly through the scanning tunneling microscopy (STM) investigations. On metal surfaces, the famous examples extend from (1) the atoms on the edges of the regular array of steps on a vicinal Ni surface[9], (2) the 1D polymer-like reaction products with the $O_2(H_2)$ reaction on the Cu(110) and Ag(110) (Ni(110)) surfaces (so called the 'psuedomolecules, see **Fig. 2**)[10] and (3) the domain boundaries on a disordered FeSi alloy surfaces[11] to (4) the striped phase of an ordered surface alloy of Au/Cu(001)[12].

On semiconductor surfaces, another variety of 1D chains can be listed, such as (1) the missing dimer chains on the Ge/Si(001)[13] and the Bi/Si(001) surfaces[14], (2) the embedded Bi wires on Si(001) (see **Fig. 3**)[15], (3) Al, In, Ga, Pb, and Sn dimer chains on Si(001)[16-23], (4) the Si dimer wires on SiC(001)[24-31], (5) diamond-like C chains on SiC(001)[32,33], (6) the Ga atomic wires on H-terminated Si(001)[34], (7) the In (ref. 35–38) and Au chains on Si(111) (ref. 39) and (8) the C_2H_4 molecular chains on Si(001)[40]. The following parts of this review will concentrates on several such 1D quantum wires on semiconductor surfaces.

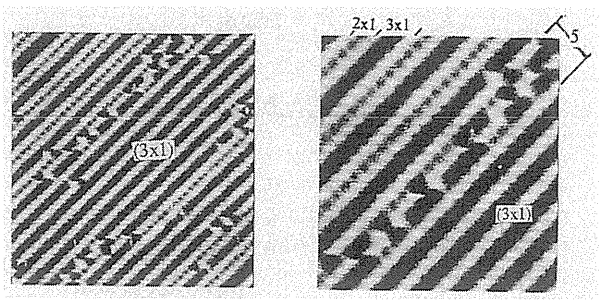

Figure 2. 1D 'psuedomolecules' on Ag(110) (STM images)[10].

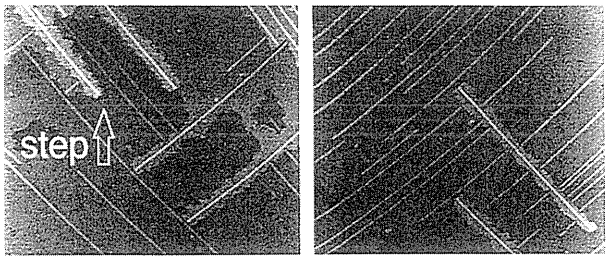

Figure 3. Bi–Si quantum wires on Si(001) (STM images)[15].

2. Methods of investigation; Photoelectron spectroscopy using synchrotron radiation for solid surfaces

The ways of approaching the 1D quantum wires on surfaces can be multifold; (1) investigating the ground state structures—atomic structures, chemical bondings and electronic band structures, (2) investigating the kinetics and dynamics to understand the mechanism of formation and (3) investigating the transport, magnetic and optical properties. In this review, mostly the first approach, especially for the chemical bondings and electronic band structures, will be discussed. Although STM is the unique probe to identify the quantum wires on surfaces, the detailed determination of the atomic structures, chemical bondings and electronic band structures is usually beyond STM's capability. Instead, powerful and direct experimental probes are provided by the modern diffraction and spectroscopy techniques using synchrotron radiation. That is, surface X-ray diffraction is one of the most powerful structural probe of a surface system and the most powerful way of investigating the chemical bondings and electronic band structures is synchrotron-radiation photoelectron spectroscopy (PES)[42].

Overview of PES–In PES techniques, basically the energy, momentum and spin of photoelectrons, the electrons ejected from the near surface layers by absorbing photons irradiated, are analyzed to determine the binding energy, Bloch wave vector and spin state of the initial electronic states. Due to the short mean free path of photoelectrons within the matter (5–100 Å), this technique is inherently surface sensitive. Several different kinds of surface-related information can be obtained in PES by fully utilizing syn-

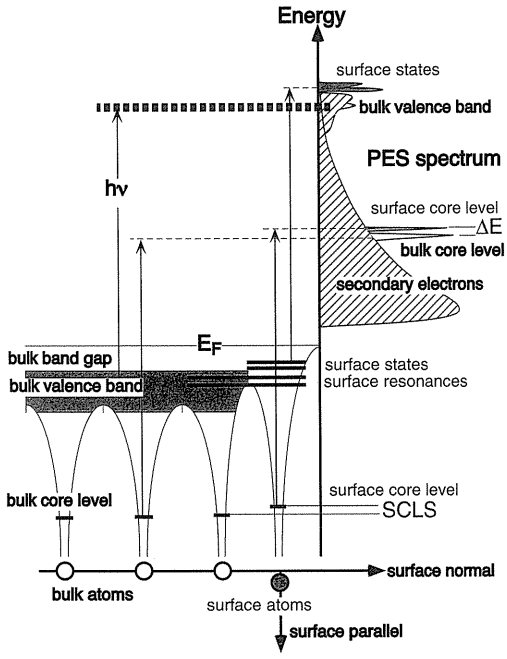

Figure 4. Overview of photoelectron spectroscopy for surfaces.

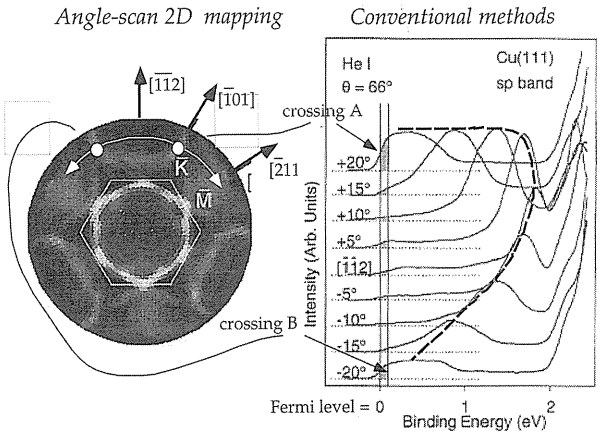

Figure 5. Fermi surface mapping through 2D angle-scan PES[44,45].

chrotron radiation.

Angle-resolved valence-band photoemission: ARPES and Fermi surface mapping–At first, angle-resolved measurements of photoelectrons from valence bands, called angle-resolved photoelectron spectroscopy (ARPES), allow the determination of 2D band structures of the surface states since the 2D in-plane electron momentum is conserved through the photoemission process[43,44]. This is far more obvious for the surface states on semiconductor surfaces, especially for the electronic states localized on the surface quantum wires, well inside the bulk band gap[5]. In case of the low-dimensional 'metallic systems', the detailed shape ('topology'), of the Fermi surfaces (contours) is of prime importance for their physical properties and the Fermi contours can directly be determined only by ARPES[41]. The most convenient and successful way of measuring the Fermi contours is by measuring the angle-resolved photoelectron intensity from the small energy window centered at Fermi level as a function of the 2D electron wave vector (i.e. photoelectron emission angle)[44,45].

Angle-resolved core-level photoemission: surface core levels and PED–At second, the measurements of core levels with sufficient energy resolution can resolve out the 'surface core levels' of surface layers- the energy-shifted core levels of surface atoms from those of bulk atoms. The core levels of adsorbates can be investigated to find different core level binding energies due to different kinds of chemical and structural environments of adsorbates[18,25,46]. That is, the surface core levels provide crucial information on the surface structures and surface chemical bondings, which guide the qualitative modeling of an unknown surface structure along with the morphological information obtained by STM. These surface core levels or surface core level shifts can now be calculated by *ab inito* density functional methods, which can be compared with the experiments to give important qualitative chemical, structural and sometimes dynamical information[47].

The angular (or energy) distribution of photoelectron intensities from a core level can further be measured. Such distribution reflects the surface structures through the scatterings of photoelectrons with surrounding atoms and the consequent interferences between the scattered and directly-emitted photoelectron waves [the phenomenon called photoelectron diffraction (PED)][48]. Detailed analyses of these angular or energy distributions, called angle-scan or energy-scan PED patterns, can lead to quantitatively determining the surface structures with 0.1 Å resolution[19,48].

Thus, together with the complementary surface techniques of LEED, RHEED and STM, we have a quantitative structural, chemical and electronic probe for a given surface system by PES. Especially, in contrast to the limitation of surface X-ray diffraction (and also other electron diffraction techniques) to the well ordered 2D systems, PED can be applied to the disordered systems with only local orderings and to chemically and structurally inhomogeneous and complex systems. This advantage makes the combined use of PES and PED using the high-flux and high-resolution synchrotron radiation an unique and powerful probe, in particular, for the surface nanostructures discussed below.

3. Recent examples of 1D nanostructures on semiconductor surfaces

Covalently bonded In (Al) chains on Si(001) and surface polymerization—As mentioned above, In, Al, Ga, Pb, and Sn adsorbates form 1D dimer chains on the Si(001) surface, which develop into a well ordered 2×2 phase at 0.5 ML[16,17]. These quantum wires (extends easily to a length of several 100 Å) attracted much recent attention as candidates for observing interesting 1D physical properties.[49]

Extensive experimental investigations, such as by STM, X-ray standing wave, tensor LEED[50], ion scattering have

been devoted in determination of the atomic structure of the In (Ga, Al, and Pb) quantum chains. We have studies the structure of the In quantum wires qualitatively by high-resolution core level PES[18] and X-ray photoelectron diffraction (XPD)[17] and further quantitatively by low-energy PED of In 4d using synchrotron radiation[19]. These studies consistently established the so called 'parallel symmetric dimer structure' (see **Fig. 8**), where the symmetric adsorbate dimers are formed between the Si dimers beneath.

Since the key for the understanding of the possible 1D properties lies on the behavior of electrons, the electronic band structures of In, Al, and Pb-induced 2×2 surfaces have been studied extensively[20-22]. For example, **Fig. 9** shows the surface-state band dispersions measured for the 2×2-In and -Al surfaces by ARPES. These series of studies all consistently showed that (1) the quantum wire phases are semiconducting with a rather large band gap (≥1 eV), (2)

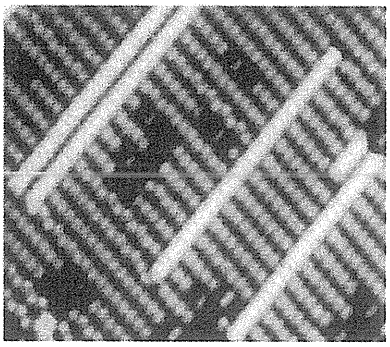

Figure 6. In quantum chains on Si(001) (STM images)[16].

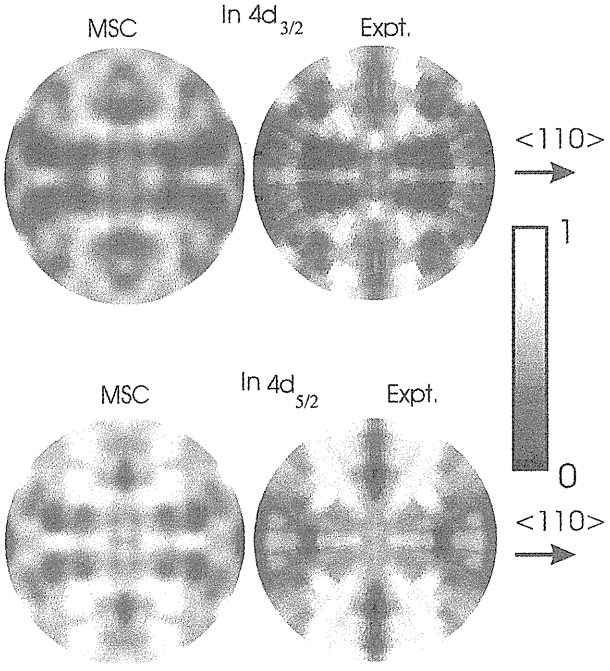

Figure 7. Simulated and experimentally measured In 4d angle-scan PED patterns of the Si(001) 2×2-In surface (data taken at BL-18A of Photon Factory)[19].

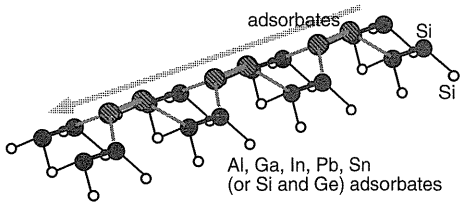

Figure 8. Atomic structure of In quantum chains on Si(001)[17,19,50].

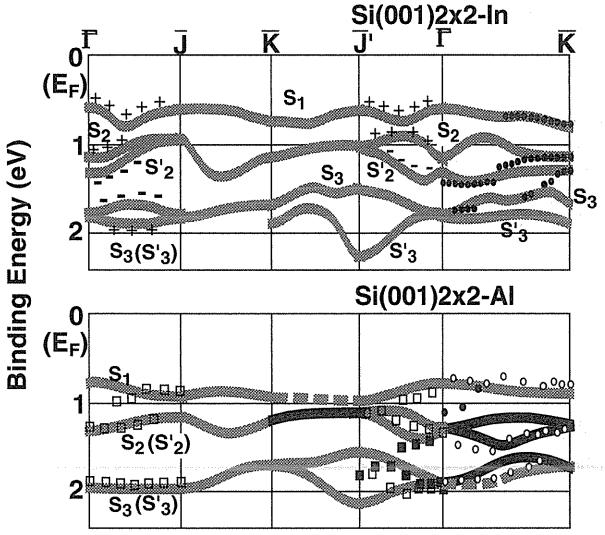

Figure 9. Experimental dispersions of the surface state bands for the 2×2-In and -Al surfaces (data taken at BL-18A of Photon Factory)[21,22].

there are five surface-state bands within the bulk band gap and (3) the surface band structures of 2×2-In, Ga, and Al (and even probably Pb) are essentially the same. Through detailed investigation of these surface state bands and through comparison to *ab initio* density-of-states calculations, it was clearly concluded that these five surface states correspond to the four back bonds to Si and one dimer bond of an adsorbate dimer[22].

Although an STM study on the In quantum wires has suggested Peierls paring as the mechanism of the dimer formation, the experimental and theoretical electronic structure studies consistently indicate that the dimers are covalently bonded with well-localized surface electrons[49]. This is also corroborated by the experimentally determined atomic structures where the adsorbate dimers have very optimal covalent bond lengths for both the adsorbate-adsorbate and adsorbate-Si bonds[19,50].

The ARPES results for the surface-state bands also show that there is no obvious 1D anisotropy in the band structure of the quantum wires in clear contrast to their 1D appearance in STM and to the strong anisotropy in their initial growth. *That is, in terms of the electronic structure, these quantum wires are not 1D materials systems.* The initial anisotropic growth (the quantum wire formation) was, then, successfully explained by 'surface polymerization', where

only the local bondings of adsorbates and substrates are important instead of the 1D electronic coupling[51].

Zero dimensional Si chains on SiC(001) and cellular fluctuation–One of the most recent example of 1D quantum wires is the highly-stable and highly-controllable quantum chains of Si on the 3C–SiC(001)[24] surface. These quantum wires appear on the Si-rich surface of the cubic SiC(001) surface[24,52] there are also Si-terminated[53,54] and C-terminated surfaces for this rather new CVD grown semiconductor crystal, which has promising and attractive features for the high-frequency, high-power and high-voltage device applications. The atomic scale 'wires' shown in the STM images[24] (**Fig. 10a**) are thought to consist of Si dimers[24–31]. The density of these 'quantum' wires, whose length is as long as $\sim 1\,\mu$m, can be controlled quite easily and the most dense phase of them is the 3×2 phase.

The local atomic structure of the Si quantum wires is currently under significant debate with three contradicting models with different Si surface coverages and even with different surface unit cell orientations (see **Fig. 11**)[25–31]. Although we have provided a strong support of one of these models (model *a*) through identifying three surface components of Si 2p core level from the 3×2 surface, there are still discrepancies with recent *ab initio* calculations. Further studies, in particular, more direct structural studies by PED, are underway.

Irrespective of the structural uncertainties of these unique quantum wires on SiC(001), the band structure of the electrons localized on these wires in the 3×2 phase was well characterized by an extensive ARPES study[27]. In this study, we found four different electronic bands related to the quantum wires within the band gap of the bulk SiC. The band structure measured shows, however, only very small dispersions for all the surface-state bands, which suggests that the electrons are well localized without a significant interaction even along the quantum wires. That is, each building block of the wires is rather independent of each other. Thus a Si quantum wire is not 1D in terms of its electrons but is rather 'zero dimensional'.

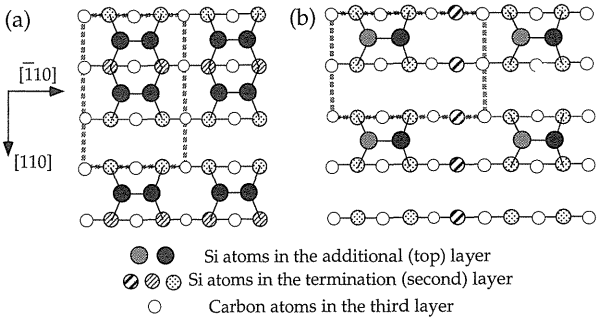

Figure 11. The proposed structure models of the 3C–SiC(001) 3×2 surface[25,26].

A peculiar 0D property shows up in detailed STM images of the 3×2 phase as shown in **Fig. 10b**[56]. At a certain condition, the 3×2 unit cells show random spatial fluctuations (in their structure or charge density). This fluctuation (or disorder) is not dynamic and is frozen even at room temperature. We believe that this fluctuation reflects the 0D character of the surface, which is, however, contradictory with its 1D appearance in STM[56]. The mechanism of 1D chain formation of this surface is under investigation now mainly using STM.

As demonstrated in the two above examples, the 1D arrangement of unit cells on surfaces is not a sufficient condition to have exotic 1D electronic systems. That is, we need to find a proper 1D metallic system, where the surface electrons couple indeed one-dimensionally to have partially filled bands. There have been few candidates of such 1D *metallic* systems on metal and semiconductor surfaces, and the following 4×1 phase on Si(111) induced by In adsorption is one of them. This system was recently shown to feature true 1D electronic behaviors, for the first time as a surface system, putting a significant milestone in this line of research.[38]

Metallic In chains on Si(111) and charge density wave–Initial growth of In on Si(111) results in a variety of 2D phases

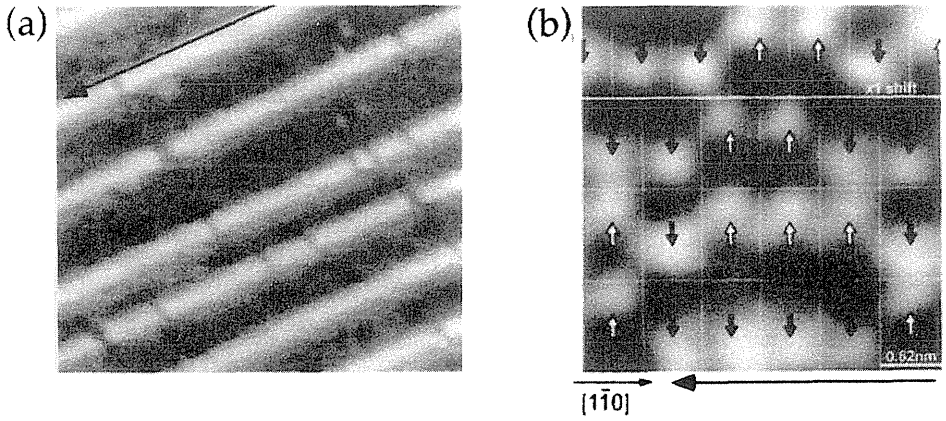

Figure 10. (a) Si quantum wires on 3C–SiC(001) (STM image) and (b) an empty state STM image of the 3×2 phase composed of the quantum wires[24,56].

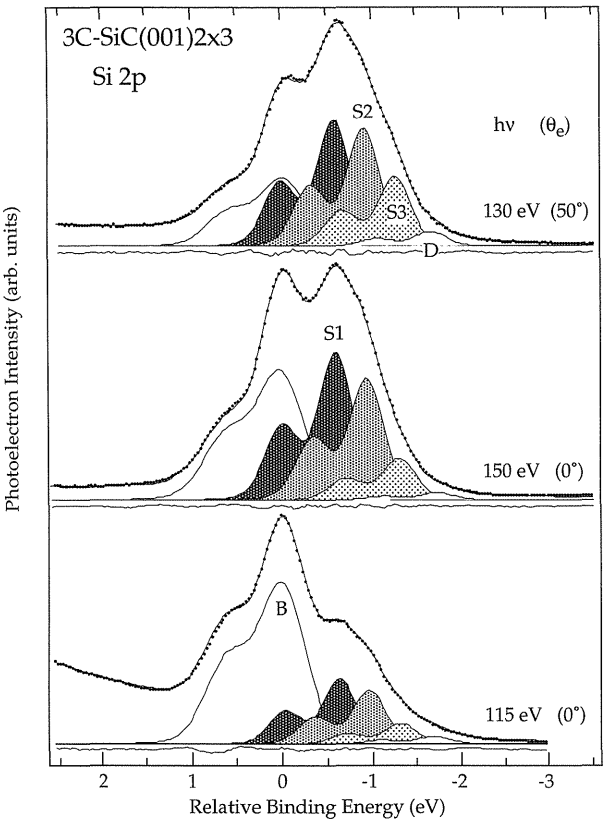

Figure 12. The Si 2p core levels of the 3C-SiC(001) 3×2 surface clearly showing three surface components (data taken at BL-22 of Max-Lab)[25,26].

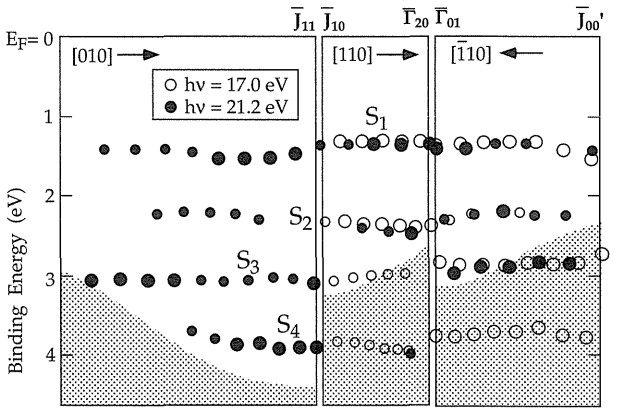

Figure 13. Experimental dispersion curves for the surface states of the 3×2 phase (data taken at BL-33 of Max-Lab)[27].

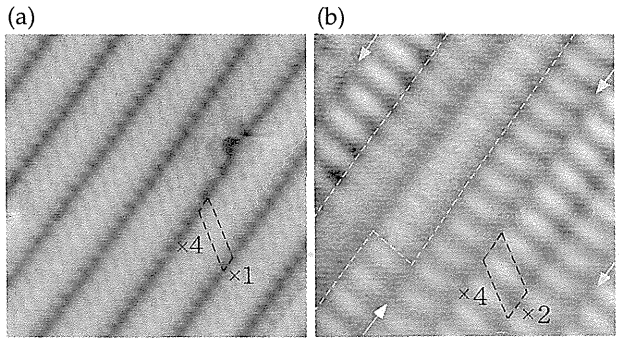

Figure 14. (a) The 4×1 phase at RT and (b) the 4×"2" phase at 65 K for In/Si(111); STM images.

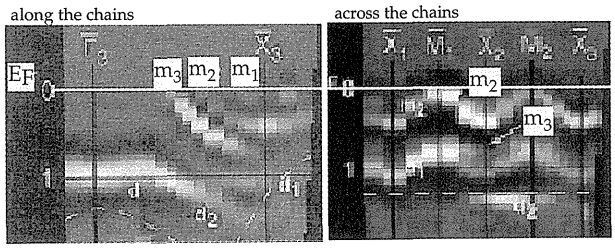

Figure 15. Surface state bands of the the 4×1 phase at RT for In/Si(111); the gray-scale E_k-$k_{//}$ diagram obtained by ARPES (data taken at BL-18A of Photon Factory)[38].

and one of them has a $4a_0 \times a_0$ (a_0=3.84 Å) periodicity (the 4×1-In surface hereafter)[4,35]. The 4×1-In surface has a linear chain structure along the ×1 direction, which appears as bright stripes spaced 13.3 Å apart in the real space images obtained by STM, whose structure is still uncertain[38].

The electronic structures of the 4×1-In surface have been studied by both angle-resolved inverse and direct PES which clearly showed that this surface is metallic with three partially filled electronic bands, m_1, m_2, m_3.

The Fermi contours of these metallic bands are determined in detail by 2D angle-scan PES. The experimentally determined Fermi contours are composed of straight or wavy lines centered along the $\bar{X}$-$\bar{M}$-$\bar{X}$ lines of the surface Brillouin zone (SBZ). Their degree of straightness provides a direct measure of the 1D character or anisotropy of each band, since dispersion normal to the chains implies two-dimensionality and also curved contours. In contrast to the substantially curved contours for m_1 and m_2, m_3's Fermi contour is perfectly straight suggesting nearly ideal 1D metallic character. Most importantly, this Fermi contour precisely bisect the (4×1) SBZ zone along the chains. Thus a charge density modulation coupled to a lattice vibration of wavelength $2a_0$ along the In chains could lead to a Peierls-like instability with a "nesting vector" of $2k_F=\pi/a_0$ (a_0=3.84 Å).

The instability of the 4×1 metallic phase is indeed observed when the 4×1-In surface is cooled down to a temperature below ~100 K. The RHEED and LEED patterns at <100 K shows extra ×2 periodicity streaks along the chains. These streaks indicate that the charge density and/or the lattice of the surface is perturbed by a $2a_0$ periodicity modulation along the linear chains, which, however, has a very poor correlation between the chains. Such streaks in diffraction patterns are a characteristic feature of 1D systems.

Accompanying this 4×1-4×"2" phase transition, the surface electronic structure exhibits drastic changes. The ARPES spectra measured along the linear chains (along $\bar{\Gamma}$-$\bar{X}$ of SBZ) at RT and at ~100 K found that the LT phase is almost semiconducting without any electronic states cross-

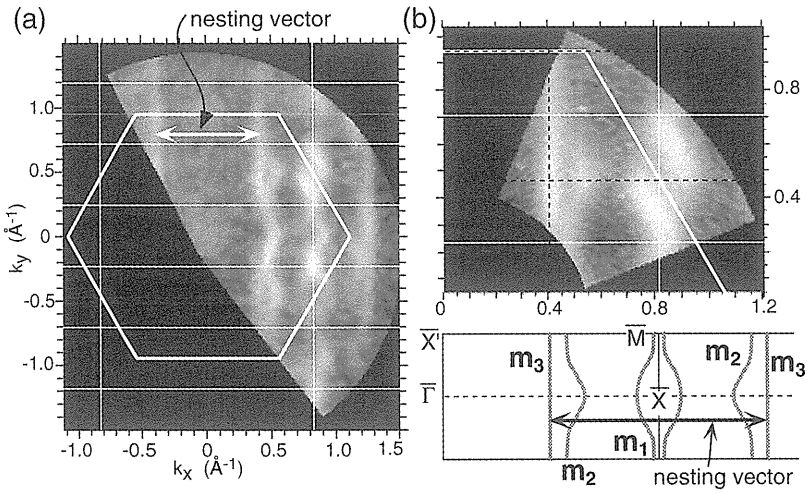

Figure 16. Experimental Fermi contours of 4×1-In (data taken at BL 7.0.1 of Advanced Light Source)[38].

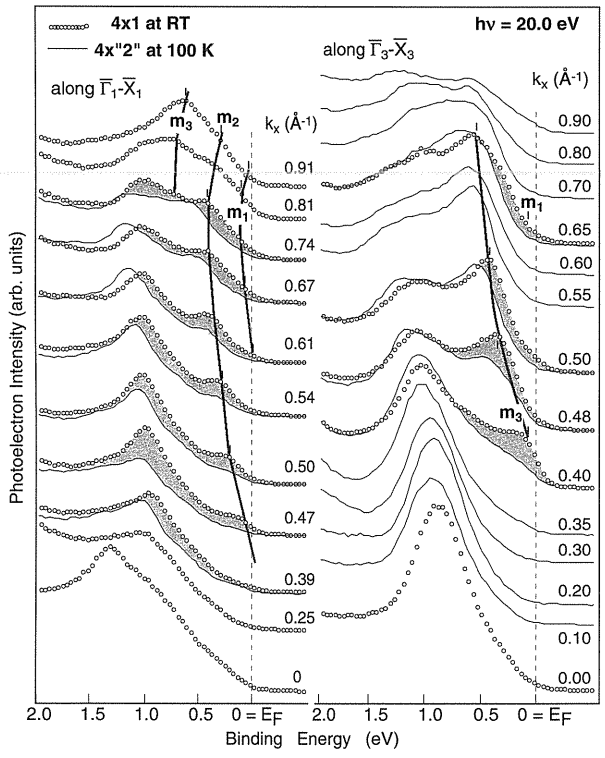

Figure 17. ARPES spectra for the 4×1 and 4×"2"-In surfaces (data taken at BL-7B of Photon Factory)[38].

ing the Fermi level. Thus, the transition from the 4×1 to the 4×"2" phase is obviously a metal-nonmetal transition as expected from Peierls instability[38].

Further, the charge density modulation induced by Peierls instability can be observed directly by STM, which maps the local surface charge densities[38]. The STM image of the 4×"2"-In surface taken at ~65 K shows a remarkable difference from that of RT featuring a pronounced $2a_0$-periodicity modulation of the surface charge along the linear chains. There is frequent phase mismatching of the ×2 modulation between the neighboring chains as shown in **Fig. 14**(b). This manifests the 1D nature of the charge density modulation observed.

For the known bulk quasi-1D materials systems, the finite interchain interactions ultimately produce 3D ground states. In the present case of a surface system, an evidence for finite 2D interchain coupling is observed in STM and electron diffraction[38]. The lack of 3D interaction in the present system may result in a more fundamental differences between the surface 1D materials and other bulk 1D materials through, most probably, the enhanced fluctuations. We note that the ratio of the estimated band gap (50~150 meV) to the transition temperature times the Boltzmann constant is ~10 and is thus much larger than simple mean field theory would predict. This ratio is one of the largest among the known 1D CDW systems and fluctuations in 1D is known to suppress the phase transition temperature much lower than predicted by the simple mean field theory.

It is now very clear that we have found a unique 1D CDW transition on solid surfaces, which is composed of quantum wires 'self-organized' on a 2D lattice. Up to now, the real-space observation of the CDW in quasi-1D materials was extremely limited to the very recent report on bulk organic materials[57] although CDW has been frequently imaged in 2D systems. However due to the great advantage of a well-ordered surface system for STM imaging, the present observation has far more clarity and enables more detailed study of the spatial fluctuations of CDW, an essential aspect of 1D systems. The direct real space observation should provide a unique test case of the microscopic theories for CDW ground states, fluctuations, and excitations in 1D. Such STM studies are right now underway. As shown above, extensive ARPES studies have led to the prediction of the CDW phase transition on the Si(111)4×1-In surface and have clarified the mechanism of this phase transition by observing the band gap and the Fermi surfaces[38]. Higher-reso-

lution quantification of the property and the size of the CDW gap is now underway along with the examination of the Luttinger liquid behavior and the dynamical behavior of the Fermi surfaces during the phase transition[58].

4. Conclusions

The recent studies of self-organized quantum wires on semiconductor surfaces extends the horizon of the low-dimensional materials science significantly. Many other interesting one-dimensional systems could be found from the anisotropically grown adsorbate systems on various solid surfaces. It can also be suggested that the developing technique of atom manipulation with STM can tailor various types of artificial one-dimensional systems on solid surfaces. These new and interesting materials systems are expected to provide not only the exotic one-dimensional physical properties, but important building blocks for the nanometer-scale electronic and magnetic device technology. The detailed structure studies and, especially, the electronic structure studies are of prime importance in finding really 1D materials on surfaces, which can be successfully performed in a comprehensive way by the modern PES/PED techniques using the 3rd generation synchrotron radiation. In parallel to the development of the probes to the transport, optical and magnetic properties of such surface 1D materials systems, it is an urgent task to establish an advanced and dedicated undulator beamline/end-station for the studies of in-situ grown nanostructures on surfaces in a Japanese synchrotron radiation source.

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きいわーど

自己組織化量子細線

原子，分子などの構造ユニットが多彩な相互作用で自発的につながって空間的に閉じた方向へ変化し，秩序体を作る物理的，化学的，生物学的過程を自己組織化という。自己組織化は様様な例があるが半導体表面における吸着，反応，成長過程で現われるナノー構造の自発的結成とその組織化も，ナノー電子/磁気デバイスの可能性と量子物性の発現のため，最近大きく注目されている一例である。主なナノー構造としては自己組織化した量子ドットと自己組織化した量子細線があげられ，細線の幅としては～3 Åから～1000 Åまでさまさまな系が報告されている。

パイアルスー不安定性

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