Variable-Temperature Micro-Four-Point Probe Method for Surface Electrical Conductivity Measurements in Ultrahigh Vacuum

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We have developed a ultrahigh-vacuum system for electrical conductivity measurements with high surface sensitivity by using monolithic micro-four-point probe method (probe spacing being ~ μm) at temperatures ranging from ~10 K to 400 K, combined with simultaneous structure analysis by reflection-high-energy electron diffraction (RHEED). This apparatus enables direct measurements of electrical conductivity at the topmost atomic layers on crystal surfaces as a function of temperature. Then, the surface transport properties are unambiguously correlated with surface structures, especially with temperature-induced surface phase transitions. The apparatus including the electrical measurement system is described in detail, together with some typical data from Si(111)-7 × 7 clean surface. [DOI: 10.1380/ejsnt.2003.50]

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I. INTRODUCTION

For measuring electrical conductivity with high surface sensitivity, we have recently developed four-point probe methods with microscopic probe spacing (~μm)[1-10]. The measuring current fed through a pair of the four probes penetrates into the specimen as deep as the probe spacing roughly, which means a relatively higher surface sensitivity as the probe spacing is reduced. Especially, the probe spacing is as small as the Debye length of a semiconductor, the current mainly flows through the surface space-charge layer. Simultaneously, we can expect a larger fraction of the current passing through the surface states, electronic states characteristic to the topmost atomic layers of crystals. This will lead to an opportunity of exploring low-dimensional or nanometer-scale transport physics in a variety of surface states [11, 12].

We have developed two types of machines for the micro-four-point probe method, independently-driven four-tip scanning tunneling microscope (4T-STM) [1-5] and monolithic micro-four-point probes[3, 4, 6-10]. Both of them work under scanning electron microscope in ultrahigh vacuum (UHV), enabling precise probe positioning on aimed areas on the specimen surface. But the conductivity measurements can be done only at room temperature (RT), which limits the physics we can discuss from the data.

The present Technical Note reports our newly developed UHV system for the monolithic micro-four-point probes with temperature variation ranging from ~10 K to 400 K. This machine enables direct measurements of surface electrical conductivity at temperature-induced surface phase transitions as well as detailed analysis of carrier scattering mechanism at the surface layers of crystals. We believe that this opens up an opportunity for a new type of low-dimensional or nanometer-scale transport physics using a variety of surface states on crystals, especially on semiconductors.

II. MICRO-FOUR-POINT PROBES

Figure 1 (a) is a scanning electron microscope (SEM) image of a monolithic micro-four-point probe, which has been developed at Mikkroelektronik Centret of Technical University of Denmark[7], and now commercially available[8]. Such probes with 4, 8, 10, 20, 30 and 60 μm probe spaces were made using silicon-based micro-fabrication technology following a procedure similar to that used to fabricate micro-cantilevers for atomic-force microscopy. The probes consist of four sharpened silicon oxide cantilevers, in line and equidistant from each other, extending from a silicon support chip. The silicon oxide pads on the chip are undercut, so that deposition of metal onto the chip results in conducting paths that are insulated from the support chip. The composition and thickness of metal layer, composed of Ti/Au alloy, are controlled to make the cantilevers strain-free. Leakage current from the metal layer to Si substrate is less than 10 pA at RT, and much smaller at lower temperatures. The probes are very flexible (the spring constant being 0.4-100 N/m) and easily bent, which makes the contact with sample surfaces straightforward even when the surface plane is not aligned with the probes. The contact pressure in our typical measurements was 10^{-8} ~10^{-7} N, estimated from the probe advancement (~0.1μm) towards the sample from the contact point.

The probe chip is mounted on a ceramic plate and wire-bonded to Au electrodes with Al wires as shown in Fig. 1(b). The plate is set on a probe holder (Fig. 1(c)) that has electrical contacts from the external circuit to the probe. The probe holder is introduced with a transfer rod from a load-lock chamber into a main UHV chamber without breaking vacuum.

Figure 1(d) is a schematic illustration of the side view of the probe plate contacting a sample surface. The angle between the probe and the sample surface is set to be 30°, so that the cantilevers bend to make the direct contacts. Figure 1(e) is a grazing-incidence SEM image of the probe contacting a sample surface, which was taken in another UHV-SEM system[6, 9].

In a new UHV chamber for temperature-variable micro-four-point probe method described here, we have em-

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FIG. 1: A monolithic micro-four-point probe. (a) A SEM image of the chip. (b) The chip mounted on a ceramic plate. (c) The plate mounted on a holder. (d) A schematic illustration of the side view of the probe contacting a sample surface. (e) A grazing-incidence SEM image of the cantilevers contacting a sample surface.

ployed an optical microscope to observe the probe from the outside of the chamber. Figure 2 shows the optical microscope images of a micro-four-point probe contacting a sample Si crystal in the new chamber. The sample surface faces downward, and the probe approaches from the bottom. Four cantilevers are separately observed in the inset (the probe spacing being 10 µm in this case). Such an observation by a microscope is indispensable for controlling the probe approaching and making gentle contacts to the sample, because the cantilevers are so thin and weak that they are easily broken by clash with the sample.

III. UHV SYSTEM

Figure 3 shows the design drawings of the UHV system, and Fig. 4 is its photo. The system consists of two UHV chambers, main and load-lock ones, separated by a gate valve from each other. The main chamber, evacuated by a combination pump, contains sample/probe stages at the bottom of Liq. N2/He tank, RHEED system, and two evaporators. The sample and micro-four-point probe mounted on their holders are introduced with a transfer rod from the load-lock chamber. The system is set on air cushions for vibration damping. A binocular optical microscope (Nikon SMZ645) is employed to observe the probe through a view port during approaching toward the sample surface (Fig. 2).

Figure 5 illustrates the section around the sample/probe stages in the main chamber. The sample holder is set facing downward on a stage that enables lateral shift (XY directions with ±3 mm travel distance) and azimuth (θ) rotation of ±90° round the center, by using piezoelectric actuators. The θ rotation enables the cantilevers aligning in particular crystal orientations of the sample. The crystal orientation and surface structure of the sample can be determined by simultaneous RHEED observations; its electron beam passes through in a direction perpendicular to the screen. The probe holder is set on another stage below that enables vertical shift (Z direction) by a piezoelectric actuator for the probe approaching to and retracting from the sample. With this mechanism for fine positioning, we
FIG. 2: A micro-four-point probe contacting a sample surface (a Si wafer) on the variable-temperature stage in the UHV system, seen with an optical microscope through a view port. The inset is a magnified image.

can make the probe contact to the sample very softly on aimed areas on the surface. Actually, no damage was detected at the contact points by SEM in another UHV chamber [6].

The sample and probe stages are thermally connected to the bottom of a Liq. N$_2$/He tank, and surrounded by radiation shields in three fold. Thus we can cool down the probe as well as the sample. The stages can be thermally isolated from the tank by a heat switch in-between when heating the sample. There are small holes on the radiation shields for material deposition from evaporators, sample/probe transfer from the load-lock chamber, RHEED electron beam, and access by the optical microscope. These holes can be closed by rotating the outer radiation shield to reach the lowest temperature. A small resistive heater is attached on the sample stage, which enables slow heating of the whole stage (0.5 K/min at most). Thermocouples are attached at several places on the stages and on the sample holder as well. Since the sample holder has electrical connections to the both ends of the sample, we can heat the sample resistively by direct current through it, up to 1250°C.

Thus, with this system, we can prepare the sample surface on the stage by heating and material deposition in situ, and also confirm the surface structure by RHEED. And the conductivity measurements by the micro-four-point probe as well as RHEED observation can be done as a function of temperature ranging from $\sim$10 K to 400 K.

IV. ELECTRICAL MEASUREMENT SYSTEM

Since the specimen is grounded for RHEED observation and thermal connection directly to the Liq. N$_2$/He tank, the electrical measurement system is isolated from the chamber ground by using an insulating-type DA/AD converter (emaISA-A25, 16 bit with a sampling frequency of 100kHz, Adtek system science Co.) in a PC. The system is shown in Fig.6. The insulating DA/AD converter enables low-noise measurements with marginal leakage current. The expected resistance to be measured ranges from $10^2\Omega$ to $10^5\Omega$, with current of 1 nA$\sim$100 µA. The measuring current is produced by amplifiers (INA114 (Burr-Brown) and OP07) with a conversion of 10 µA/1V. The voltage drop is amplified by 20 times with INA114.

We tested this measurement system with a dummy resistance of 163 kΩ, confirming that the digitized current output from the DA converter was in a unit of 3.1 nA, and that the measured voltage contained noise of about ±50µV, which was much larger than the resolution of the AD converter (1.6µV). This noise came from the wiring in the whole system. From this data, we can say that the measurable resistance ranges from 10 mV/3.1-nA$\sim$3 MΩ to 100-µV/100-µA$\sim$1Ω with our system. This corresponds to the 2D sheet conductivity of $6 \times 10^{-2} \sim 2 \times 10^{9}\mu$S/square.
V. RESULTS

Figure 7 shows RHEED patterns taken by the present UHV system. The sample was a vicinal Si(111) crystal oriented 1.8° off from (111) surface towards [112] azimuth. The electron beam irradiated the surface in the step-up direction along [112] azimuth. A regularly stepped 7×7 clean substrate was prepared according to a method in Ref. [13] to minimize step bunches and kink density. The flash heating up to 1250°C was done for cleaning the surface at the sample stage in the main chamber. Figure 7(a) is its RHEED pattern at RT, in which each super-spot is slightly elongated along the vertical direction in this image due to narrow terraces of the 7×7 structure along the beam direction.

Indium of 1 ML was deposited on this 7×7 surface with resistive heating at 450°C. Then, a well-ordered, regularly stepped, and single-domain Si(111)-4×1-In surface superstructure was prepared at RT as shown in Fig. 7(b), where the In atomic chains were parallel to the steps. This was checked by scanning tunneling microscopy (STM) in the other UHV chamber separately.

The 4×1-In surface have recently attracted much attention due to a phase transition below RT. The surface consists of a massive array of metallic nanowires composed of four In atomic chains [14], and has quasi-one-dimensional metallic surface-state bands[15]. Their Fermi surfaces (Fermi lines actually) nearly bisect the 4×1-surface Brillouin zone [15, 16], leading to a formation of 1D charge density wave (CDW) having an energy gap 2∆ at Fermi level (E_F), due to Peierls instability[17]. Actually, this surface is known to transform into a 8×2' structure below 130K, and show an energy gap at E_F as revealed by photoemission spectroscopy (PES) [16, 18]. The 1D Fermi-surface nesting gives the intra-chain periodicity doubling (×2'), while the phase locking between the neighboring CDW chains give the inter-chain periodicity doubling (×8)[16].

When this surface was cooled below RT, the RHEED pattern changed as shown in Fig. 7(c) at 150 K and (d) at 90 K. The 4×1 pattern (b) was observed between RT and ~200 K, while weak half-order streaks (indicated by black arrowheads in Fig. 7(c)) appeared between 200 K and 130K, indicating a 4×2' phase (Fig. 7(c)).

http://www.sssj.org/ejssnt
FIG. 6: The electrical measurement system for micro-four-point probe method. The generation of measuring current and voltage-drop measurement are carried out by a single board of DA and AD converters in a personal computer.

FIG. 7: RHEED patterns taken from a vicinal Si(111) crystal surface (1.8° off towards [112] azimuth from <111> direction), with electron beam incidence in the step-up direction along [112] azimuth. (a) A clean 7×7 structure at RT. (b) 4×1-In structure at RT. (c) 4×′2′-In structure at 150 K, and (d) 8×′2′-In structure at 90 K. The unit cells of the respective structures in the reciprocal lattice are indicated by yellow quadrangles.

means a periodicity doubling along the In atomic chains as expected, while there is no correlation of this doubling between the neighboring chains. Below 130 K, it changed into the 8×′2′ structure (Fig. 7(d)), indicating a periodicity doubling across the In chains (the 1/8-th fractional-order spots are indicated by red arrowheads). This is due to finite inter-chain coupling by the finite band dispersion of surface states across the chains or simply by the Coulomb interaction between the neighboring charge-density waves [16]. The observed phase transition was consistent with the previous reports [16, 19]. By using a micro-four-point probes with 8μm probe spacing, we found a drastic change in electrical resistance at this phase transition; the resistance for an n-type crystal showed almost constant values above 130 K, while it began to rise steeply with cooling further. It increased eventually by three orders of magnitude at 90 K. This dramatic change in resistance precisely corresponds to formation of the 8×′2′ phase, and means the metal-insulator transition in the surface states. In other words, we have succeeded in directly measuring the surface-state conductance. The details are described elsewhere [20].

Here we show the results of resistance measurements for the Si(111)-7×7 clean surface on an n-type Si crystal with resistivity of 2~15Ωcm at RT. This surface did not show any change in RHEED by cooling down to 75 K. Figure 8 shows the I-V curves measured during slow cooling from 350 K to 75 K. The gradients of the curves near \( I = 0 \)A steadily increases with cooling, meaning that the measured resistance monotonously increases with temperature lowering, from 30 kΩ at 350 K to 3 MΩ at 75 K, as shown in Fig. 9. This result looks quite strange if we consider the electrical conduction through a bulk Si crystal. According to textbooks on semiconductor physics[21], this temperature range is known as a so-called 'saturation region', in which all donors are ionized so that the carrier concentration is almost constant, while the carrier mobility increases with cooling due to reduction of phonon scattering. As a result, the resistance should decreases with temperature lowering, which is opposite to our present result in Fig. 9. This means that we
FIG. 8: *I-V* curves measured from a Si(111)-7 × 7 clean surface as a function of temperature.

FIG. 9: Resistance obtained from the gradient of *I-V* curves at zero current in Fig. 8, measured from a Si(111)-7 × 7 clean surface as a function of temperature with a micro-four-point probe of 10 µm spacing.

have mainly measured the surface region that has different transport properties from the bulk region. According to the detailed analysis of the data [22], we have proposed that the electronic transport here is a kind of hopping conduction through the surface space-charge layer as well as the surface states. The details will be reported elsewhere [22].

VI. SUMMARY

We have shown our newly developed UHV system in which electrical conductivity measurements with monolithic micro-four-point probes and structure analysis with RHEED can be simultaneously carried out as a function of temperature between ~10 K and 400 K. We have demonstrated the performance with resistance data of a Si(111)-7 × 7 clean surface measured with a micro-four-point probe of 10 µm probe spacing, which shows an unusual temperature dependence. This means that the data reflect the transport property of surface layers, which is quite different from the bulk one, though we cannot yet separate the surface-state transport from the surface-space-charge-layer transport with the present data only. Under some special condition e. g., with an inversion layer beneath the surface that electrically isolates the surface layer from the underlying bulk region, the measurement becomes more surface-sensitive enough for detecting the surface-state conductivity at low temperatures, which will be reported elsewhere[20].

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[1] I. Shiraki, F. Tanabe, R. Hobara, T. Nagao, and S. Hasegawa, Surf. Sci. 493, 633 (2001).
[2] S. Hasegawa, I. Shiraki, F. Tanabe, and R. Hobara, Current Appl. Phys. 2, 465 (2002).
[3] S. Hasegawa and F. Grey, Surface Science 500, 84 (2002).
[4] S. Hasegawa, I. Shiraki, F. Tanabe, R. Hobara, T. Kanagawa, T. Tanikawa, I. Matsuda, C. L. Petersen, T. M. Hansen, P. Bøggild, and F. Grey, Surf. Rev. Lett., (2003), in press.
[5] T. Kanagawa, T. Tanikawa, I. Matsuda, and S. Hasegawa, Phys. Rev. Lett., in press.
[6] I. Shiraki, T. Nagao, S. Hasegawa, C. L. Petersen, P. Bøggild, T. M. Hansen, and F. Grey, Surf. Rev. Lett. 7, 533 (2000).
[7] C. L. Petersen, F. Grey, I. Shiraki, and S. Hasegawa, Appl. Phys. Lett. 77, 3782 (2000).
[8] Monolithic micro-four-point probes are commercially available; http://www.capres.com/
[9] S. Hasegawa, I. Shiraki, T. Tanikawa, C. L. Petersen, T. M. Hansen, P. Bøggild, and F. Grey, J. Phys.: Cond. Matter 14, 8379 (2002).
[10] T. M. Hansen, K. Stokbro, O. Hansen, T. Hassenkam, I. Shiraki, S. Hasegawa, and P. Bøggild, Rev. Sci. Inst. 74, No. 8 (August, 2003), in press.
[11] S. Hasegawa, J. Phys.:Condens. Matter 12, R463 (2000).
[12] S. Hasegawa, X. Tong, S. Takeda, N. Sato, and T. Nagao, Prog. Surf. Sci. 60, 89 (1999).
[13] J.-L. Lin, D. Y. Petrovykh, J. Viernow, F. K. Men, D. J. Seo, and F. J. Himpsel, J. Appl. Phys. 84, 255 (1998).
[14] O. Bunk, G. Falkenberg, J. H. Zeysing, L. Lottermoser, and R. I. Johnson, M. Nielsen, F. Berg-Rasmussen, J. Baker, and R. Feidenhans'l, Phys. Rev. B59, 12228 (1999)
[15] T. Abukawa, M. Sasaki, F. Hisamatsu, T. Goto, T. Kinoshita, A. Kakizaki and S. Kono, Surf. Sci., 325 33 (1995).
[16] H. W. Yeom, S. Takeda, E. Rotenberg, I. Matsuda, K. Horikoshi, J. Schaefer, C. M. Lee, S. D. Kevan, T. Ohta, T. Nagao, and S. Hasegawa, Phys. Rev. Lett. 82, 4898 (1999).
[17] G. Grüner, Density Waves in Solids (Addison-Wesley, 1994).
[18] H. W. Yeom, K. Horikoshi, H. M. Zhang, K. Ono, and R. I. G. Uhrberg, Phys. Rev. B 65, 241307 (2002).
[19] C. Kumpf, O. Bunk, J. H. Zeysing, Y. Su, M. Nielsen, R. L. Johnson, R. Feidenhans'l, and K. Bechgaard, Phys. Rev. Lett., 85 4916 (2000).
[20] T. Tanikawa, I. Matsuda, T. Kanagawa, and S. Hasegawa, Phys. Rev. Lett., submitted.
[21] For example, S. M. Sze, *Physics of Semiconductor Devices*, (John Wily and Sons, NY, 1981).
[22] T. Tanikawa, K. Yoo, I. Matsuda, S. Hasegawa, and Y. Hasegawa, Phys. Rev. B, submitted.