Surface Electrical Conductivity Measurement System with Micro-Four-Point Probes at Sub-Kelvin Temperature under High Magnetic Field in Ultrahigh Vacuum

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We have constructed an ultrahigh vacuum system in which surface conductivity is measured in situ by micro-four-point probe method down to 0.8 K, combined with conventional surface preparation/analysis capability. In order to reduce the data scattering due to changes of the probe spacing, we have employed a dual configuration method in which the combinations with current/voltage probes are switched. The surface-state superconductivity of Si(111)-\(\sqrt{7} \times \sqrt{3}\)-In surface superstructure was confirmed with the critical temperature of 2.8 K. The critical magnetic field was 0.3-0.4 T in the surface-normal direction.

Keywords: Surface-state superconductivity; Micro-four-point probe; Surface electrical transport; Dual configuration method; In; Si(111)

I. INTRODUCTION

It is only for the last 15 years that the surface-state electronic transport has been measured and discussed as a new class of low-dimensional and symmetry-broken electronic transport systems [1, 2]. This is an inherent electrical conduction through the surface electronic states characteristic of the surface superstructures. Since the surface states on semiconductor substrates are in many cases formed within the band gap of the bulk bands of the substrate, the surface states and the bulk states are electronically decoupled from each other. Therefore, the carriers in the surface states can flow along the surface through the topmost atomic layers unless the carriers are not scattered inelastically into the bulk bands. But, in experiments, it is not easy at all to inject the carriers only into the surface states; by using direct-contact electrodes, the carriers are inevitably injected into the bulk states as well as into the surface states. As a result, the measured conductivity is usually a sum of the two parallel conductors, the surface-states and the bulk-states channels. The key point for measuring the surface-state conductivity is, therefore, to eliminate/reduce the contribution from the bulk conductivity of substrate which is in many cases large enough to easily hamper the detection of the surface-state conductivity.

Some researchers used high-resistivity (insulating) semiconductor substrates to reduce the substrate contribution to the measured conductivity [3–5], which is especially effective at low temperatures where the bulk carriers are frozen out. Another method to reduce the substrate bulk contribution is microscopic four-point probes (\(\mu\)4PP) in which the spacing between the probe electrodes is on the order of micrometers or sometimes nanometers [6, 7].

Two types of \(\mu\)4PP have been developed which operate in ultrahigh vacuum (UHV) and combined with sample preparation tools to make in-situ measurements possible. One is four-tip scanning tunneling microscopes (STM) in which four STM tips are independently driven as electrodes in arbitrary arrangements [8–15]. Another is monolithic \(\mu\)4PP in which four tiny probes are fabricated and fixed in line at the edge of Si chip [16–19] which are now commercially available [20]. Although the probe spacing and arrangement cannot be changed in the monolithic \(\mu\)4PP, it is much easier to operate than the four-tip STM. Many results have shown that the probe spacing on micrometer scale is effective enough to sensitively detect the surface-state conductivity on semiconductor substrates [21–28].

Cooling the sample down to liq.-\(\text{N}_2\) or liq.-\(\text{He}\) temperatures is effective not only to diminish the substrate conductivity, but also to understand the transport mechanism in the surface states. The lowest temperature at \(\mu\)4PP machines so far is around 10 K [27, 28]. This is sometimes not enough for observing quantum transport phenomena such as superconductivity and Kondo effect. Actually, the superconductivity of Si(111)-\(\sqrt{7} \times \sqrt{3}\)-In surface superstructure is recently reported with the critical temperature below 3 K [29, 30]. Therefore, we need a \(\mu\)4PP operating around 1 K.

Here we report the development of a monolithic \(\mu\)4PP machine operating in UHV in which the sample and probes are cooled down to 0.8 K using \(\text{He}\) only and the magnetic field up to 7 T is applied along the surface normal of the sample. The machine revealed the superconductivity of Si(111)-\(\sqrt{7} \times \sqrt{3}\)-In surface superstructure with the critical temperature of 2.8 K, consistent with the previous report [30] in which macroscopic electrodes were used. The magnetic field around 0.3 T was enough to destroy the superconductivity.

II. MICRO-FOUR-POINT-PROBE SYSTEM

Figure 1(a) is a photo and (b) is a design drawing of a measuring head for the \(\mu\)4PP constructed in the present research. The sample holder and the probe holder are indicated in blue and red, respectively. Figures 1(c) and (d) are their photos. The sample and the probe chip are edged with red lines in the photos. Figure 1(e) is a schematic illustration of the side view of the probe contacting the sample surface. The angle between the probe and the sample surface is set to be 30°, so that the probes bend by pushing it toward the sample, resulting in the direct contacts with all probes. The probes are very flexible.
FIG. 1: (a) A photo and (b) the design drawing of the measuring head for µ4PP. (c) A photo of the sample holder. The sample is edged with red lines. (d) A photo of the probe holder. The probe chip, mounted on a ceramic substrate, is edged with red lines. (e) A schematic illustration of the side view of the probe contacting the sample surface. (f) A schematic illustration of the top view of the probe chip mounted on a ceramic substrate. Four tiny probes extend from the left edge of the chip.

(three 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 details of the µ4PP chip is described in Ref. [17]. The probe holder is driven by a piezo-actuator with inertial-motion method. The probe chip is mounted on a ceramic substrate and wire-bonded to electrodes with metallic wires as shown in Figure 1(f). The substrate is set on a probe holder (Fig. 1(d)) that is electrically connected to the external circuit.

III. UHV SYSTEM

Figure 2 is the design drawing of UHV system, and Figure 3 is its photo. The system is composed of three chambers, a load-lock chamber, a preparation chamber, and a main chamber, separated from each other by gate valves. The load-lock chamber is pumped by a turbo-molecular pump. The preparation chamber and main chamber are pumped together by a combination pump to reach 10^{-10} Torr vacuum. The sample and probe holders are introduced from the load-lock chamber and transferred into the main chamber via the preparation chamber by using transfer rods. Thus, we can exchange the sample and probe without breaking vacuum of the preparation and main chambers.

The preparation chamber contains a sample stage on which the sample can be heated by direct current flowing through the sample and can be cooled down to about 100 K by liq. N_2. It is also equipped with reflection-high-energy electron diffraction (RHEED) system and five evaporators. Thus, the sample can be prepared by molecular beam epitaxy (MBE) technique with in-situ RHEED observation.

The main chamber, containing the measuring head (Fig. 1) at the bottom, is a long UHV pipe and inserted in a cryostat. A superconducting magnet is set at the bottom of the cryostat, which can apply the magnetic field up to 7 T in the direction perpendicular to the sample surface.

Figure 4 is a schematic illustration of the cooling system. The cooling procedure consists of two steps. The first step is, after introducing liq.^4He into a ‘1 K Pot’ reaching 4 K, to decompress the 1 K Pot by a rotary pump to reach around 2 K (yellow part in Fig. 4). Then ^3He gas, confined in the green part in Fig. 4, is liquefied in a ^3He Pot’. The second step is to use a sorption pump for decompressing the ^3He Pot. The sorption pump is cooled by ^4He vapor from the 1 K Pot, so that charcoal in the sorption pump adsorbs ^4He vapor in the ^3He Pot to make the pressure in the green part down to about 0.01 Torr. By this decompression, the ^3He Pot, together with the measuring head which is connected to the ^3He Pot, are cooled down to 0.3 K. Since, however, at the moment we do not have ^3He which is very expensive, we confined ^4He gas in the green part in Fig. 4. Then, we could reach 0.8 K by the procedure described above by using ^4He instead of ^3He. Since the heat of evaporation of liq. ^4He is smaller than that of liq. ^3He, the lowest temperature we can reach is slightly higher than in the ^3He case. The space between the outer liq. He tank (blue part in Fig. 4) and the measuring head is filled with ^4He gas when accelerating the cooling of measuring head and pots, while the space is evacuated to thermally isolate the measuring head from the surroundings when reaching and keeping the lowest temperature.
V. ELECTRICAL MEASUREMENT SYSTEM

The measured voltage and current data from each probe are amplified by a preamplifier shown in Fig. 5 which is placed outside the vacuum chamber. And via an AD/DA converter (USB-6343, National Instruments Corporation [32]), the data are stored in a PC. The voltage differences between any pair of probes of the four, which are six combinations of pairs in total, are always measured to ensure the consistency. By using switches Sw$_1$ and Sw$_2$ in the preamplifier of Fig. 5, we can set any probe as the current source or the voltage probe in four-point probe resistance measurements. This versatile switching of the probe role is very effective for ‘dual configuration measurements’ described below. The measuring current is swept in the rage of ±1 μA.

Auto-approach of probe: We use current monitoring method during the probe approach like in STM. With Sw$_3$ switching on, the sample is grounded. And by applying a bias voltage to all probes with Sw$_1$ switching on and Sw$_2$ off, the current flowing between the probes and sample is always monitored during the probe approach. Once the current is detected by one of the four probes, the probe motion is switched from auto-mode to manual mode to make soft direct contact for all probes. Since the probe holder is actuated only by a ‘course piezo’, the contact is not tunneling contact.

Dual configuration measurement: As mentioned above, the probes are so flexible that they are easily bent laterally as well as longitudinally by contact, resulting in unequal spacings between the probes. In the previous re-
ports [21–28], we assumed that the spacings between the probes were all equal to use a simple relation between the measured resistance $R$ and the sheet resistivity $R_S$: in case of the probe configuration shown by the left inset in Fig. 6,

$$R_{SA} = \frac{\pi}{\ln 2} R_A. \quad (1)$$

In case of the configuration shown by the right inset in Fig. 6,

$$R_{SB} = \frac{2\pi}{\ln(4/3)} R_B. \quad (2)$$

But this caused data scattering because the probe spacing changed from measurement to measurement; the probe spacings are not always equal. To correct this weak point, here we have used 'dual configuration method' [33, 34] in which two measurements are done by changing the combination of current-source probes and the voltage-pick-up probes. For example, the first measurement is done with the probes No. 1 and 4 as the current probes and the probes No. 2 and 3 as the voltage probes (the left inset in Fig. 6), resulting in the measured resistance $R_A$. Then the second measurements is done, without changing the probe position and pushing force, with the probe No. 1 and 2 as the current probes and the probe No. 3 and 4 as the voltage probes (the right inset in Fig. 6), obtaining the second value of resistance $R_B$. The sheet resistivity of the surface $R_S$ is obtained by an identity

$$\exp\left(-\frac{2\pi R_A}{R_S}\right) + \exp\left(-\frac{2\pi R_B}{R_S}\right) = 1 \quad (3)$$

This identity is satisfied with arbitrary probe spacings. $R_S$ is obtained by numerically solving this equation with a set of the measured data $R_A$ and $R_B$. As shown in Fig. 6, we actually confirmed that this dual configuration method is very effective to reduce the data scattering.

Because of these purposes mentioned above, we need to switch the role of each probe quickly. The CMOS analog switches $Sw_1$ and $Sw_2$ in Fig. 5 are indispensable for this.

Since the minimum current we can measure with our system is 31 pA and the maximum voltage is 10 mV, the maximum resistance we can measure is $\sim 300 \, M\Omega$. But, practically, it is $\sim 10 \, M\Omega/\square$ at most because it is difficult to detect the current at the auto-approach of the probes with higher-resistance samples due to the high-contact resistance which is in series with the sample resistance. The minimum resistance we can measure is, on one hand, estimated to be $\sim 5 \, \Omega (=10 \, \mu V/2 \, \mu A)$ because thermal noise in the circuit is about $\pm 5 \mu V$. Therefore, the 'zero resistance' of superconductivity in this paper means a resistance less than 5 $\Omega$.

**VI. RESULTS –SUPERCONDUCTIVITY–**

The sample, Si(111)-$\sqrt{7} \times \sqrt{7}$-In surface superstructure, was fabricated on a Si(111) wafer of n-type with the resistivity 1–10 $\Omega$-cm at room temperature [27, 29, 30, 35, 36]. Indium was deposited on the Si(111) substrate at room temperature, and then the sample was flash heated up to

![Fig. 6: Sheet resistance measured at room temperature for Bi(111) thin film (about 20 bilayer thick) grown on a Si(111)-7 $\times$ 7 substrate. The measurements were done 30 times by repeating cycles of detaching/contacting the probe from/to the sample surface. Up to the 21st measurement, the probe was retracted and approached by applying DC voltage to the piezo-actuator, resulting in the soft and reproducible contacts. After the 21st measurement, the probe chip was retracted by the inertial motion mode of piezo-actuator and approached again. Then, the probe spacing seemed to change. $R_{SA}$ (green symbols) and $R_{SB}$ (blue symbols) are the sheet resistances obtained with the probe configuration shown in the insets, respectively, by assuming the all probe spacings equal (Eqs. (1) and (2)), while $R_S$ (red symbols) is the sheet resistance obtained by the dual configuration method using Eq. (3). The data by the dual configuration remain almost constant while the single-configuration data are scattered significantly.](image-url)
As shown in Fig. 7, the resistance gradually decreased by cooling and steeply dropped around 3 K. The critical temperature of superconducting transition to zero resistance is 2.8 K, which is consistent with the previous report [30]. The gradual decrease in resistance above the critical temperature is explained by large superconducting fluctuation; Cooper pairs are locally formed, but not in a global coherent state. Scanning tunneling spectroscopy measurements probe the local property, indicating a slightly higher critical temperature, 3.18 K [29].

As shown in Fig. 8, the superconducting state is broken by applying magnetic field. The resistance begins to rise around 0.3 T, and the normal state is recovered around 0.45 T in this case. By increasing the magnetic field, vortices begin to penetrate through the superconducting surface and they move around causing resistance, and finally the superconductivity is totally broken at the upper critical field. Thus the system exhibits a behavior as a type-II superconductor though the bulk In is a type-I superconductor. The details of the superconducting properties are discussed elsewhere [37].

VII. SUMMARY

We have constructed a UHV system in which the surface conductivity is in situ measured by the μ4PP method down to 0.8 K under magnetic field up to 7 T. The superconductivity of Si(111)-√7×√3-In surface superstructure was confirmed with the critical temperature of 2.8 K, which was very well consistent with the previous report [30]. The critical field to break the superconductivity was around 0.3-0.4 T applied perpendicular to the surface. Thanks to the dual configuration method in four-point probe measurements, the data scattering was greatly reduced. The machine will be very useful to explore the surface-state superconductivity and quantum transport at very low temperature.

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[34] M. Yamada, Master Thesis (University of Tokyo, 2012).

