# Anisotropy in Conductance of a Quasi-One-Dimensional Metallic Surface State Measured by a Square Micro-Four-Point Probe Method

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We have devised a "square micro-four-point probe method" using an independently driven ultrahigh-vacuum four-tip scanning tunneling microscope, and succeeded for the first time to directly measure anisotropic electrical conductance of a single-atomic layer on a solid surface. A quasi-one-dimensional metal of a single-domain Si(111)$4 \times 1$-In had a surface-state conductance along the metallic atom chains ($\sigma_{\parallel}$) to be $7.2(\pm 0.6) \times 10^{-4}$ S/□ at room temperature, which was larger than that in the perpendicular direction ($\sigma_{\perp}$) by ~60 times. The $\sigma_{\parallel}$ was consistently interpreted by a Boltzmann equation with the anisotropic surface-state band dispersion, while the $\sigma_{\perp}$ was dominated by a surface-space-charge-layer conductance.

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Low-dimensional metals represent a fascinating class of materials, showing exotic properties such as non-Fermi liquid behaviors. The transport phenomena of such metals naturally provide intriguing and challenging issues in condensed matter physics. While highly anisotropic bulk crystals have been the targets for the past decades, a metallic single-atomic layer or arrays of metallic atom chains on semiconductor crystals have recently been focused intensively, which has been triggered by direct observations of a Peierls transition and possible Luttinger-liquid behavior [1–5]. Thus, crystal surfaces with atomically controlled structures are a new promising playground for studying low-dimensional transport phenomena, with expectations of advancing from mesoscopic to nanoscale transport physics [6–8].

However, there have been no unambiguous, direct, and quantitative measurements of electronic transport through single-atomic layers on crystal surfaces (*surface-state conductance*). Isotropic or anisotropic two-dimensional (2D) conductance is typically measured by a two- or four-point probe method with van der Pauw (Montgomery) arrangements in which the electrodes are placed at edges of a sample [9,10]. Since the electrode spacing is in macroscopic scales, the methods are able only to detect conductance of metal thin films on insulating substrates [11] or that of single-atomic layers with daring subtraction of substrate contributions, leaving quite large ambiguity [12,13].

Recently, we have developed microscopic four-point probe (4PP) methods and have reported possible direct detection of surface-state conductance [14–16]. Microscopic 4PP measurements with $\mu$m probe spacing are more surface sensitive than macroscopic 4PP ones, because the measuring current flows mainly near the surface. Furthermore, by placing the micro-4PP at the center of a macroscopic sample surface (~ mm–cm in size), the sample can be regarded as infinitely large, so that the measured resistance can be analytically calculated by solving the Poisson equation [17]: Resistances of an infinite 2D layer/sheet measured with linear 4PP [Fig. 1(a)] and square 4PP [Fig. 1(b)] arrangements with equidistant probe spacings should be

$$R_{\mathrm{linear}} = \Delta V/I = \frac{1}{\pi\sqrt{\sigma_x \sigma_y}} \ln 2, \tag{1}$$

$$R_{\mathrm{square}} = \Delta V/I = \frac{1}{2\pi\sqrt{\sigma_x \sigma_y}} \ln\left(1 + \frac{\sigma_y}{\sigma_x}\right), \tag{2}$$

respectively. $\Delta V$ is voltage drops measured by an inner pair of probes in linear 4PP [16] or by a pair of any adjacent probes in square 4PP. $I$ is the measuring current flowing through another pair of probes. $\sigma_x$ and $\sigma_y$ are conductances along $x$ and $y$ directions, respectively, with the sample surface being on the $xy$ plane. Then one can notice an important fact from Eqs. (1) and (2). Even if the probes are rotated by 90° with respect to the sample surface having anisotropic conductance, the linear 4PP measurement gives the same value of resistance; $\sigma_x$ and $\sigma_y$ are just exchanged with each other in Eq. (1). This is true for any angles of rotation of linear 4PP, meaning that $R_{\mathrm{linear}}$ always gives a geometric mean of $\sigma_x$ and $\sigma_y$ only. The square 4PP, however, gives different values of resistance by exchanging $\sigma_x$ and $\sigma_y$ in Eq. (2) when the square

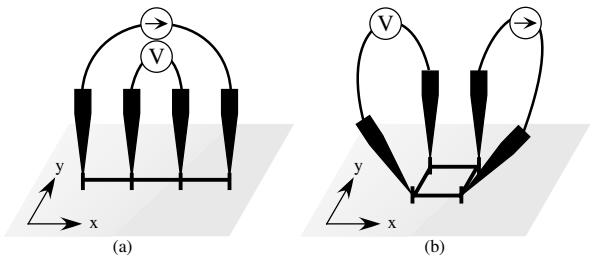


FIG. 1. Schematic drawings of the four-point probe method in (a) linear and (b) square arrangements. The probes are equidistant from each other. Current and voltage probes are shown.

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is rotated by 90° or another combination of current and voltage probes is selected. *Thus, anisotropy in conductance of infinite 2D sheets can be measured only by the square 4PP method, and $\sigma_x$ and $\sigma_y$ are obtained separately from the two values of measured resistance.*

In this Letter, we report the first measurements of anisotropic surface-state conductance of a single-domain Si(111)4 × 1-In surface (inset of Fig. 2), by the square micro-4PP method using an independently driven four-tip scanning tunneling microscope (STM) [14]. The surface-state band structure of this phase shows a quasi-one-dimensional (1D) metallic character, and its Fermi surface (Fermi line) is determined by angle-resolved photoemission spectroscopy (ARPES) [1,2,18]. Its atomic arrangement is a massive array of metallic quantum wires composed of four lines of In atom chains running along the $[10\bar{1}]$ direction, each wire being separated by a Si-atom chain in between the metallic wires [19–21]. In order to improve the measurement precision in the present work, the 4PP in square arrangement was rotated in a step of 15° with respect to the metallic chains, and all data were fitted by an analytical solution of the Poisson equation to obtain the conductance parallel to the metallic chains ($\sigma_{\parallel}$) and that in the perpendicular direction ($\sigma_{\perp}$) separately.

The 4 × 1-In surface has nowadays attracted great interest because of its Peierls transition [1,2], a possibility of non-Fermi-liquid behaviors, and relating conductivity changes [12]. There have never existed quantitative measurements of anisotropic surface-state conductance.

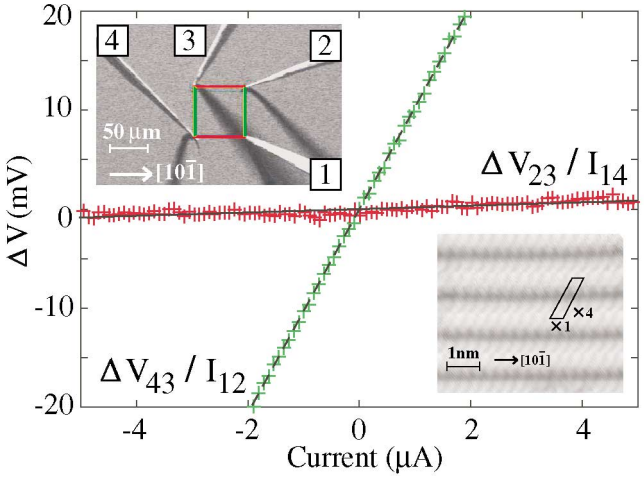


FIG. 2 (color). *I-V* curves of a single-domain Si(111)4 × 1-In surface measured by square micro-4PP method with the probe spacing (a side of the square) of 60 $\mu$m. The red and green colors correspond to the measurements of $\Delta V_{23}/I_{14}$ and $\Delta V_{43}/I_{12}$, respectively. The upper left inset is a SEM images of the probes. The In chains were parallel to the $[10\bar{1}]$ direction, horizontal direction in this image, which was determined by RHEED. The lower right inset is a STM image of the 4 × 1-In surface, separately taken by another single-tip STM.

The measurements were performed at room temperature by our four-tip STM system installed in an ultrahigh-vacuum scanning electron microscope (UHV-SEM) [14]. Each probe, made of a tungsten tip, was independently driven with piezoelectric actuators and a scanner in *xyz* directions to achieve precise positioning in nanometer scales. The SEM was used for observing the tips for positioning, as well as the sample surface together with scanning reflection-high-energy electron diffraction (RHEED) capability. Its details are described elsewhere [14,15]. The four-tip probes can be made to contact to the sample surface in arbitrary arrangements, with marginal damage by a tunneling current approach and minute direct contact, which was checked by SEM. Ohmic conductance measurements were confirmed by both linearity of current($I$)-voltage($V$) curves and resistance relation derived from Green's reciprocal theorem among a different combination of current-source and voltage-sensing probes. The 4 × 1-In surface was prepared by In deposition onto a well-cleaned Si(111)7 × 7 surface at 400 °C with the help of *in situ* RHEED observation. A vicinal Si wafer ($15 \times 3 \times 0.525$ mm$^3$, P-doped, bulk resistivity $\rho = 1 \sim 10\ \Omega$ cm) with 1.8° miscut from the (111) axis was used to grow a single-domain 4 × 1 phase [18].

Figure 2 shows *I-V* curves measured by the square micro-4PP method with 60 $\mu$m probe spacing. $\Delta V_{ij}$ is a voltage drop measured between probe $i$ and probe $j$, with current flowing from probe $k$ to probe $l$, $I_{kl}$ (see the upper left inset). Two values of resistance, $\Delta V_{23}/I_{14} = 170\ \Omega$ and $\Delta V_{43}/I_{12} = 10.3$ kΩ, were obtained from the gradients of the respective *I-V* curves, just by changing the combination of current and voltage probes. A line linking probe 1(2) with probe 4(3) is parallel to the metallic In chains, while that connecting probe 1(3) and probe 2(4) is perpendicular to the chains. The results indicate that an exchange of $\sigma_{\perp}$ and $\sigma_{\parallel}$, corresponding to $\sigma_x$ and $\sigma_y$ in Eq. (2), results in ~60 times difference in the measured resistance. This clearly demonstrates a detection of the anisotropy in conductance, and also means a direct detection of surface-state conductance (because the substrate conductance is isotropic). By the way, $\Delta V_{42}/I_{13}$ was measured to be 9.83 kΩ. This indicates $\Delta V_{23}/I_{14} + \Delta V_{42}/I_{13} \sim \Delta V_{43}/I_{12}$, realizing Green's reciprocal theorem and confirming the Ohmic conductivity measurements.

In contrast, the resistance measured by the linear micro-4PP method was essentially the same, irrespective of the probe orientation with respect to the In chains. Figure 3 gives *I-V* curves measured with the linear 4PP aligned parallel and perpendicular to the In chains. The SEM images of the probes are shown in the insets, where the In chains run in the horizontal direction. The gradients of the *I-V* curves give almost the same resistances, 4.4 and 5.3 kΩ within experimental error. Other orientations of the probes gave the same results. Thus, the distinction between Figs. 2 and 3 clearly demonstrates the expectation from Eqs. (1) and (2).

Although, in principle, two values of resistance obtained by the square 4PP in Fig. 2, combined with Eq. (2), allow one to derive $\sigma_{\parallel}$ and $\sigma_{\perp}$ separately, the 4PP square was rotated around its center, by repositioning each tip, and the square micro-4PP measurements were performed every 15° rotation to improve the measurement precision. The results are shown in Fig. 4, where SEM images of the probes with some rotation angles are also shown. The rotation angle, $\theta$, is defined by a line formed by two current probes and the In-chain direction. At each rotational position, two values of resistance, $\Delta V_{23}/I_{14}$ and $\Delta V_{43}/I_{12}$, were measured. The results shown here were acquired from an optimally prepared $4 \times 1$-In surface.

The Poisson equation gives an analytical form for resistance as a function of $\theta$ measured by this "rotational square micro-4PP method":

$$\frac{\Delta V}{I} = \frac{1}{2\pi\sqrt{\sigma_x \sigma_y}} \times \ln \sqrt{\frac{(\sigma_x/\sigma_y + 1)^2 - 4\cos^2\theta \sin^2\theta (\sigma_x/\sigma_y - 1)^2}{(\sin^2\theta + \sigma_x/\sigma_y \cos^2\theta)^2}}. \quad (3)$$

Equation (2) is derived by inserting $\theta = 90°$ in Eq. (3). By fitting Eq. (3) with the experimental data in Fig. 4, $\sigma_x$ and $\sigma_y$, corresponding to $\sigma_{\parallel}$ and $\sigma_{\perp}$, were determined to be $7.2(\pm 0.6) \times 10^{-4}$ and $1.2(\pm 0.1) \times 10^{-5}$ S/□, respectively, its anisotropy $\sigma_{\parallel}/\sigma_{\perp}$ being ~60. The errors come from the fitting process and are thought to originate from deviation of the actual probe positions from the vertices of the square. Negative values of $\Delta V/I$ at some $\theta$ come from deformed electrostatic potential contours due to the high anisotropy in conductance.

We did the similar measurements with different probe spacings. The Poisson equation tells that the conductance measured by the linear or square 4PP method should be inversely proportional to the probe spacing when the measured region and resulting current flow are three dimensional [14,15,17], but that it is independent of the probe spacing in 2D cases as shown in Eqs. (1) and (2). Through measurements with the probe spacing of 30 ~ 120 $\mu$m, the conductance as well as the anisotropy were found to be invariant with the probe spacing, indicating a 2D nature of the observed conductance.

We also performed similar measurements for the $1 \times$ 1-In (or $\sqrt{31} \times \sqrt{31}$-In) phase that was formed with different In coverages and annealing temperatures on the same substrate crystal [22]. The phase is known to have isotropic 2D metallic surface states [23]. The anisotropy in conductance for this phase was found to be 1.1 ~ 1.8, much smaller than that of the $4 \times 1$-In phase. Such a small anisotropy for the isotropic surface may come from aligned steps on the vicinal substrate. Therefore, we can safely say that the highly anisotropic conductance for the $4 \times 1$ phase is intrinsic in its surface-state bands.

Now, we discuss quantitatively the origins of the anisotropic conductance of the $4 \times 1$-In phase. In general, the measured conductance of a semiconductor crystal is a

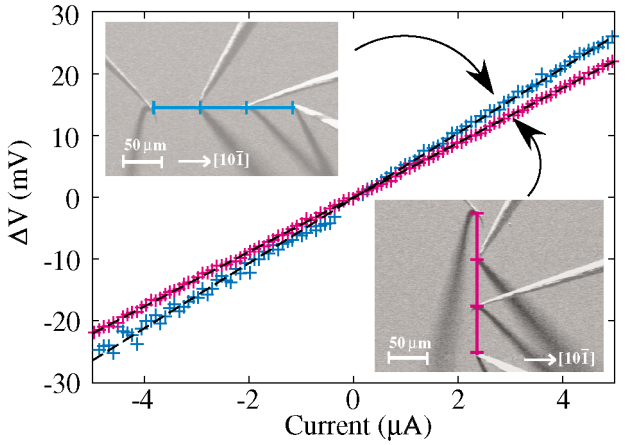


FIG. 3 (color). *I-V* curves of a single-domain $4 \times 1$-In surface measured by linear micro-4PP method with 60 $\mu$m probe spacing. The blue and purple colors correspond to the measurements parallel and perpendicular to the In chains (horizontal direction in these SEM images), respectively.

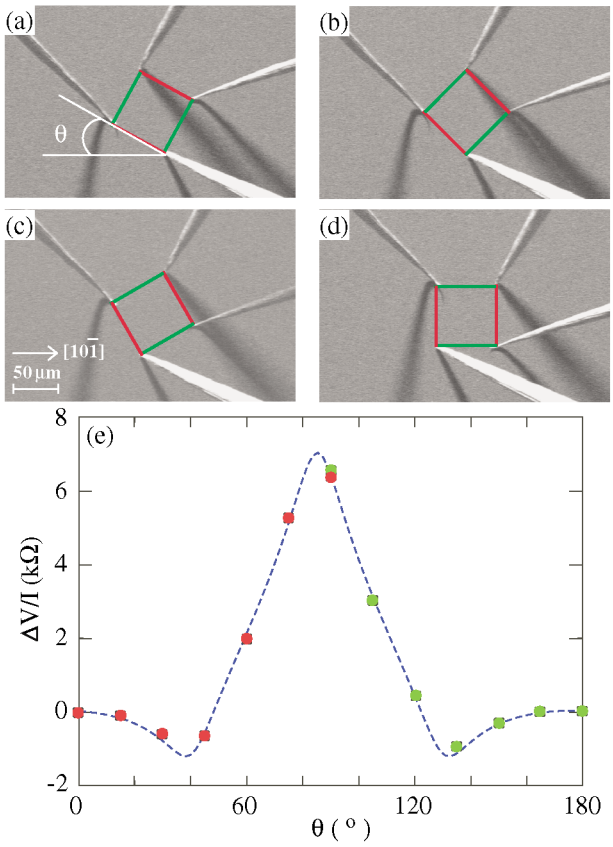


FIG. 4 (color). SEM images of rotational square micro-4PP measurements with 60 $\mu$m probe spacing, at (a) $\theta = 30°$, (b) 45°, (c) 60°, and (d) 90°. (e) Angle dependence of the measured resistance $\Delta V/I$ of the single-domain $4 \times 1$-In surface. Experimental data are fitted by Eq. (3).

sum of contributions from three current channels, $\sigma = \sigma_{ss} + \sigma_{sc} + \sigma_b$: (i) surface-state bands on the topmost atomic layers ($\sigma_{ss}$); (ii) bulk-state bands in a surface space-charge layer beneath the surface ($\sigma_{sc}$); and (iii) bulk-state bands in the inner crystal ($\sigma_b$) [15]. According to the previous and our ARPES measurements of this surface [18], the bulk-valence-band maximum at surface is located at $0.12(\pm 0.01)$ eV below Fermi level, indicating a formation of an inversion layer for an $n$-type Si wafer and an existence of $p/n$ junction between the surface-space-charge layer and the underlying bulk [24]. The $p/n$ junction ensures that the measuring current does not penetrate into the bulk [25]. So we can ignore the bulk current channel ($\sigma_b$). This is consistent with the result of probe-spacing dependence mentioned above that the measured conductance has a 2D nature, because $\sigma_{sc}$ of the inversion layer as well as surface-state conductance $\sigma_{ss}$ are 2D due to current confinement between the surface and the $p/n$ junction [14,15]. It is noted here that the inversion layer makes the measurements further surface sensitive by eliminating the bulk conductance.

By solving a Poisson equation for the space-charge layer for the present $n$-type wafer [24] with known values of surface and bulk Fermi-level positions and bulk resistivity $\rho$, the thickness of the inversion layer was calculated to be 160 nm (for $\rho = 1\ \Omega\,\mathrm{cm}$) and 490 nm (for $\rho = 10\ \Omega\,\mathrm{cm}$). The conductance of the inversion layer, $\sigma_{sc}$, is then calculated to be $1.2(\pm 0.4) \times 10^{-5}$ S/□ (for $\rho = 1\ \Omega\,\mathrm{cm}$) and $1.9(\pm 0.5) \times 10^{-5}$ S/□ (for $\rho = 10\ \Omega\,\mathrm{cm}$). The $\sigma_{sc}$ is now turned out to be similar to $\sigma_\perp$, but about 60 times smaller than $\sigma_\parallel$. Thus, it is obvious that $\sigma_\parallel$ is mainly a surface-state conductance, $\sigma_{ss}$, while $\sigma_\perp$ is dominated by the space-charge-layer conductance $\sigma_{sc}$. In fact, the In-coverage dependence of $\sigma_\perp$ was quite similar to that of the band bending, but $\sigma_\parallel$ changed in a different way. The details will be reported elsewhere. Thus, the interchain conductance ($\sigma_\perp$) is mainly through the surface space-charge layer.

Next we discuss the $\sigma_\parallel$ using a Boltzmann picture with the known Fermi surface of the $4 \times 1$-In surface states [1,18]. A relation between the 2D conductance and Fermi surface derived from Boltzmann equation is given as follows, with an approximation that the carrier relaxation time $\tau$ is independent of electron wave vector [26]:

$$\sigma_{ij} = \frac{e^2\tau}{2\pi^2\hbar} \int \frac{v_{ki} v_{kj} dk_F}{|v_k|}, \tag{4}$$

where $k_F$ and $v_{ki}$ are Fermi wave vector and Fermi velocity vector along the $i$ direction, the latter being obtained from the surface-state band dispersion, $v_k = \nabla_k E_k/\hbar$. Inserting the corresponding parameters reported by the previous ARPES results, the conductance tensor was calculated, and finally we obtained $\sigma_\parallel (= \sigma_{xx}) = 1.4 \times 10^{11} \times \tau_\parallel [\mathrm{S}/\square]$ and $\sigma_\perp (= \sigma_{yy}) = 7.2 \times 109 \times \tau_\perp [\mathrm{S}/\square]$, where $\tau_i$ is relaxation time in the $i$ direction. By comparing this $\sigma_\parallel$ with the experimentally obtained one, the relaxation time along the In chains is $\sigma_\parallel = 5.2(\pm 0.8) \times 10^{-15}$ sec. This value is similar to that in bulk In crystals and smaller than that of typical metal by nearly an order of magnitude, calculated by the Drude model [27]. The mean-free path along the In chains, estimated from this relaxation time and the group velocity at the Fermi level, is about 5 nm. By assuming $\tau_\parallel = \tau_\perp$ crudely, one can derive the anisotropy in surface-state conductance to be 19, which is reasonable with the experimental one.

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