Surface nanowire hosting a Tomonaga-Luttinger liquid with two-component spectral feature implying spin-charge separation

Yoshiyuki Ohtsubo,¹,²,³,☆ Jun-ichiro Kishi,² Kenta Hagiwara,²
Patrick Le Fèvre,³ François Bertran,³ Amina Taleb-Ibrahimi,³,⁴
Masaharu Matsunami,⁵ Kiyohisa Tanaka,⁵ and Shin-ichi Kimura¹,²,†

¹Graduate School of Frontier Biosciences,
Osaka University, Suita 565-0871, Japan
²Department of Physics, Graduate School of Science,
Osaka University, Toyonaka 560-0043, Japan
³Synchrotron SOLEIL, Saint-Aubin-BP 48, F-91192 Gif sur Yvette, France
⁴UR1/CNRSSynchrotron SOLEIL, Saint-Aubin, F-91192 Gif sur Yvette, France
⁵UVSOR Facility, Institute for Molecular Science, Okazaki 444-8585, Japan

(Dated: April 17, 2015)

Abstract

One-dimensional metallic surface state is created in atomic Bi-nanowires prepared on InSb(001) semiconductor substrate. Angle-resolved photoelectron spectroscopy (ARPES) reveals its spectral feature down to 8 K, showing the power-law scaling with binding energies around the Fermi level \(E_F\) as well as the temperature dependence of the photoelectron intensities. ARPES shows that the surface state band consists of two separate features dispersing with different Fermi velocity but touching each other at \(E_F\), suggesting a spin-charge separation. These results strongly suggest that a Tomonaga-Luttinger liquid is realized in the Bi-nanowire system on InSb(001).

PACS numbers: 71.20.-b, 73.20.At, 79.60.-i, 71.10.Pm
The Fermi liquid theory of ordinal three-dimensional (3D) metals breaks down in one-dimensional (1D) systems, producing various exotic quantum phases. Tomonaga-Luttinger liquid (TLL) \([1, 2]\) is the exactly solvable model of gapless 1D quantum system, characterized by the power-law scaling and spin-charge separation in low-energy excitation spectra \([3]\). So far, various 1D systems were studied as candidates of TLL and only a few of them showed metallic states with power-law spectral features, e.g. carbon nanotube \([4]\) and Lithium purple bronze \([5, 6]\). The other characteristic, spin-charge separation, is reported in some 3D materials with 1D electronic structures \([7–9]\).

On the surface of semiconductors, various self-assembled 1D atomic structures are known and have been regarded as suitable systems to study 1D metallic states \([10, 11]\). On such surface systems one can perform in-situ electron/hole doping by deposition of additional atoms and observation/control of local atomic structure by scanning probe techniques. These manipulation onto the surface 1D states could provide further insight into the 1D physics such as surface TLL which has attracted much attention during the past two decades. However, in spite of the variety of 1D surfaces, most of them do not behave as TLL at low temperature because of some difficulties, such as metal-insulator transitions driven by strong nesting \([12]\) and two-dimensional (2D) undulation of the surface electronic states \([13]\).

So far, the only candidate for surface TLL is the Au nanowire assembled on Ge(001) (Au/Ge(001)), showing power-law scaling in the low-energy spectral feature \([14, 15]\). However, even on Au/Ge(001), the direction of conduction path and the presence of the TLL phase itself is still under debate \([16–18]\). Moreover, the surface state of Au/Ge(001) crosses the Fermi level \((E_F)\) twice at each side of the surface Brillouin zone (SBZ), requiring additional consideration \([19]\) to apply the original TLL theory supposing single crossing at each side. Therefore, a new candidate for surface TLL, desirably with single \(E_F\) crossing, is awaited for the progress of surface-TLL research.

In this letter, we report on the case of a new candidate of surface TLL discovered on the Bi-induced 1D structure on InSb(001) (Bi/InSb(001)), studied here by angle-resolved photoelectron spectroscopy (ARPES). The Bi/InSb(001) surface state shows 1D dispersion with almost no undulation and stays metallic down to 8 K. The 1D state is the only metallic state of the sample, showing parabolic dispersion with its bottom at the center of SBZ. The photoelectron spectra in low-energy region obey the power-law scaling as a function of binding energy and sample temperature with the same power index \(\alpha = 0.75 \pm 0.10\).
Moreover, the surface-state band consists of two separate features dispersing with different Fermi velocities \( v_F \) but touching each other at \( E_F \), suggesting states due to spin-charge separation. These results strongly suggest that the 1D surface structure Bi/InSb(001) hosts surface TLL.

InSb(001) substrates were cleaned by repeated cycles of sputtering and annealing up to 680 K until the \( c(8 \times 2) \) low-energy electron diffraction (LEED) pattern was observed. Then, nominally 3 ML of Bi were evaporated at room temperature: 1 ML is defined as the atom density of bulk-truncated InSb(001) and the coverage was estimated using a quartz microbalance. A subsequent flash by direct current heating up to 680 K for \( \sim 10 \) seconds makes the \( p(1 \times 3) \) surface, indicated by the sharp and low-background LEED pattern shown in Fig. 1 (a). This surface does not show \( c(2 \times 6) \) pattern as reported in a previous work [20], where the post annealing was performed at \( \sim 470 \) K for 2 hours. Figure 1 (b) is the LEED pattern of the \( c(2 \times 6) \) surface reproduced according to the reference. Sometimes, we could find faint \( c(2 \times 6) \) spots on the \( p(1 \times 3) \) surface, possibly due to a small area of coexisting \( c(2 \times 6) \) phase. On the \( c(2 \times 6) \) surface, scanning tunneling microscopy showed 1D chain-like atomic structure along \([110]\) defined in Figure 1 [20]. Although the periodicity is not the same, it is natural to suppose similar 1D structure on the \( p(1 \times 3) \) surface. At least, most of surface 1D systems align along the smaller surface unit vector, \([110]\) in this case.

Figure 2 shows the surface electronic structure of the Bi/InSb(001) surface measured by ARPES.ARPES measurements were performed at the CASSIOPEE beamline of synchrotron SOLEIL and BL7U of UVSOR-III. The photon energies used range from 15 to 200 eV with linear and circular polarizations. The photoelectron kinetic energy at \( E_F \) and the overall energy resolution of the ARPES setups (\( \sim 20 \) meV) were carefully calibrated \textit{in-situ} from the photoelectron spectra on a Mo foil attached to the sample (see Figure 3 (a)).

Figure 2 (a) shows the energy contour around \( E_F \) measured with linearly polarized photons. \( k_x \) and \( k_y \) are defined to be parallel to \([110]\) and \([\overline{1}10]\), respectively. The linear dispersion along \( k_y \) clearly indicates an 1D metallic state on the surface. Figure 2 (a) also shows that this 1D state is the unique metallic state on the surface. As shown in the close-up view recorded with circularly polarized photons in Fig. 2 (b), the metallic state dispersion around \( E_F \) is exactly linear along \( k_y \) without any distortion along \( k_x \). The dispersion of the 1D state does not change with photon energies, indicating no dispersion along \( k_z \) as expected for a surface state. The absence of electronic states at \( k_x \sim 0.7 \) Å\(^{-1}\) suggests its origin from
the $p(1\times3)$ surface, not from the $c(2\times6)$ structure.

It should be noted that the 1D surface state appears with single domain, in contrast to the double-domain case of Au/Ge(001) \cite{15, 18}. It is due to a reduced symmetry of the InSb(001) surface as compared to (001) surfaces of group-IV semiconductors, like Si and Ge. While equivalent (001) atomic planes of Si/Ge appear with 90° rotation at each atomic step, resulting in two equivalent domains of the surface 1D structure, those of III-V semiconductors appear only at every two atomic steps without rotation. Thus, one can unambiguously determine the conduction path of the Bi/InSb(001) surface as parallel to [110], probably along the 1D chain direction on the surface.

Figures 2 (c-e) show the dispersion of the surface band on the Bi/InSb(001)-$p(1\times3)$ surface. As shown in (c) and (d), the surface state forms a single parabolic band with its bottom at $k_x = 0 \ \text{Å}^{-1}$. This surface state is the unique metallic state on the surface. This 1D metallic band crosses $E_F$ only once at each side of SBZ. While the bottom of the surface-state band shows small shift ($\sim 50 \ \text{meV}$) between $k_y = 0.0$ and $0.2 \ \text{Å}^{-1}$, $k_F$ shows no change accordingly. Indeed, no dispersion is observed in the $E-k_y$ mapping shown in Fig. 2 (e). The slight dispersion of the bottom of the surface-state band would be due to an overlap with bulk valence bands (the bulk band gap is only 0.17 eV at the center of Brillouin zone). As shown in Figure 2 (f), this 1D surface state does not appear on the $c(2\times6)$ surface. This might be due to a pairing between neighboring atomic chains on the $c(2\times6)$ surface, but this is to be investigated.

In order to examine the 1D nature of the surface state, we compared the angle-integrated photoelectron spectrum with that from ordinary metal (Mo foil attached to the sample), as shown in Fig. 3 (a). The angle integration was performed at $k_y = 0.2 \ \text{Å}^{-1}$ in order to avoid possible overlap of the bulk valence band. Figure 3 (a) clearly shows the suppression of the photoelectron intensity at $E_F$ comparing to that from the Mo foil, suggesting a deviation of the surface state from the Fermi-liquid framework. Indeed, down to binding energies around 0.1 eV, the spectrum can be very well fitted by the power-law function $I(E) \propto E^\alpha$ convolved with the Gaussian instrumental energy resolution ($\Delta E$). It gives a power index $\alpha = 0.77 \pm 0.05$ (a blue (solid) line on Fig. 3(a)). To be more precise, the broadening due to temperature should be taken into account, but at 8 K, it is negligible as compared to $\Delta E$ in this case.

The power-law scaling of TLL should show an universal power index with various pa-
rameters. Figure 3 (b) shows the temperature dependence of photoelectron intensities at $E_F$ in log scale. The intensity from 200 to 80 K decrease linearly with log $T$, indicating a power-law scaling of the spectral intensities, $I_{E=E_F}(T) \propto T^\alpha$. The change of slope below 80 K is due to the finite $\Delta E$. This broadening is already convolved in the fitting curve (blue solid line) giving a power index $\alpha = 0.73 \pm 0.10$, a value very close to that obtained by fitting the energy scale (Fig. 3 (a)). Note that $I_{E=E_F}(T)$ should be constant in the case of a Fermi-Dirac system. Moreover, the TLL spectral function taking the finite temperature into account (from Ref. [21]) well reproduces the angle-integrated spectra at rather high temperatures (see Fig. 3 (c) and (d)) within the variance of $\alpha = 0.77 \pm 0.05$. These results indicate that the 1D surface state on Bi/InSb(001) obeys an universal power-law scaling, in good agreement with that expected for TLL.

We also tried to capture the other prominent feature of TLL, namely spin-charge separation in low-energy region of excitation spectra. Figure 4 (a) shows the ARPES energy distribution curves (EDC) along $k_x$, in the same region as shown in Fig. 2 (d). As indicated by ticks, each spectrum consists of broad, but obviously separate two components. One branch is more intense with its bottom around 0.23 eV at $k_x = 0 \text{ Å}^{-1}$, and the other weak feature has its bottom at $\sim 0.09$ eV. For a better identification, we superimposed the energy positions of the two branches onto the second-derivative ARPES image (see Fig. 4 (b)). Two separate dispersions are also clearly seen in the second-derivative image. Each branch is nearly parabolic with different velocities at $E_F$, where they meet each other: dashed lines in Fig. 4 (b) are guides to the eye. Such dispersions agree exactly with what is expected from separated spinon and holon bands in a TLL [3]. The Fermi velocities for the two branches obtained from the dispersions, corresponding to spinon and holon excitations, are 1.4 and 3.6 eV Å, respectively.

At first glance, our observation of spin-charge separation with rather high power-law index, $\alpha=0.75$, may appear to disagree with previous results in which the spinon feature appeared only as broad leading edges of holon peaks [6]. Earlier work [22] claimed that the spinon feature becomes a very broad edge for high power index, typically $\alpha > 0.5$, and that it should make it difficult to observe the clear separation of spinon and holon peaks in photoelectron spectra. However, as a matter of fact, our result showing detectable spin-charge separation is in agreement with their statement. In Figure 4 (a), one can see that the spinon features in the TLL bands, dispersing at lower binding energies than the holon
ones, are also observed as broad humps, rather than as peaks, and their width is much larger than the instrumental energy resolution ($\Delta E \sim 20$ meV in this work). A broadening of the spinon/holon features is therefore also caused in the Bi/InSb(001) TLL state. A reason why one can observe it separately in this work would be the simple parabolic dispersion of the TLL band. Since it does not cross with any other bands, the bottom of spinon/holon bands, where the separation is the clearest, is easily observable.

In summary, a new candidate of surface TLL formed by Bi nanowires on a InSb(001) substrate has been discovered. A surface state shows an 1D dispersion with almost no undulation and stays metallic down to 8 K. The 1D state is the only metallic state of the sample and crosses $E_F$ only once at each side of SBZ, which perfectly matches the assumption made in the original TLL theory. The photoelectron spectra in low-energy region obey the power-law scaling with the binding energy as well as the sample temperature with the same power index $\alpha = 0.75 \pm 0.10$. Moreover, the surface-state band consists of two separate features dispersing with different $v_F$. They meet each other at $E_F$, suggesting spin-charge separation predicted in 1D electron systems. These results strongly suggest that the 1D surface structure Bi/InSb(001) hosts surface TLL and provides a fertile playground for further studies of low-dimensional physics.

The authors acknowledge Daniel Ragonnet and Françoise Deschamps for their support during the experiments on the CASSIOPÉE beamline at synchrotron SOLEIL. Part of ARPES experiments were performed under the UVSOR proposal No. 26-824. The present work was supported by the JSPS Grant-in-Aid for Research Activity Start-up (Grant No. 26887024).

* Electronic address: y.oh@fbs.osaka-u.ac.jp
† Electronic address: kimura@fbs.osaka-u.ac.jp
[1] S. Tomonaga, Prog. Theor. Phys. 5, 544 (1950).
[2] J. M. Luttinger, J. Math. Phys. 4, 1154 (1963).
[3] J. Voit, Rep. Prog. Phys. 57, 977 (1994).
[4] H. Ishii et al., Nature 426, 540 (2003).
[5] F. Wang et al., Phys. Rev. Lett. 96, 196493 (2006).
[6] L. Dudy et al., J. Phys.: Condens. Matt. 25, 014007 (2013).
[7] R. Claessen et al., Phys. Rev. Lett. 88, 096402 (2002).
[8] B. J. Kim et al., Nat. Phys. 2, 397 (2006).
[9] Y. Jompol et al., Science 325, 597 (2009).
[10] F. J. Himpsel et al., J. Phys.: Condens. Matt. 13, 11097 (2001).
[11] M. Grioni, S. Pons and E. Frantzeskakis, J. Phys.: Condens. Matt. 21, 023201 (2009).
[12] H. W. Yeom et al., Phys. Rev. Lett. 82, 4898 (1999).
[13] K. Yaji et al., Phys. Rev. B 87, 241413(R) (2013).
[14] C. Blumenstein et al., Nat. Phys. 7, 776 (2011).
[15] S. Meyer et al., Phys. Rev. B 90, 125409 (2014).
[16] K. Nakatsuji and F. Komori, Nat. Phys. 8, 174 (2012).
[17] J. Park et al., Phys. Rev. B 90, 165410 (2014).
[18] K. Yaji et al., submitted.
[19] N. Sedlmayr, P. Korell and J. Sirker, Phys. Rev. B 88, 195113 (2013).
[20] P. Laukkanen et al., Phys. Rev. B 81, 035310 (2010).
[21] D. Orgad, Philos. Mag. B 81, 377 (2001).
[22] J. W. Allen, Solid State Commun. 123, 469 (2002).
FIG. 1: (Color online). Low-energy electron diffraction patterns of Bi/InSb(001) with (a) \(p(1\times3)\) and (b) \(c(2\times6)\) periodicities taken at room temperature. (c) A schematic picture of \(p(1\times3)\) and \(c(2\times6)\) surface Brillouin zones.
FIG. 2: (Color online) (a, b) Constant energy contour around the Fermi level ($E_F$) taken at 8 K. Dashed and solid lines in (a) indicate boundaries of the $c(2\times6)$ and $p(1\times3)$ surface Brillouin zones, respectively. $k_x$ ($k_y$) is defined parallel (perpendicular) to [110]. ARPES intensities are integrated over 50 meV (a)/20 meV (b) windows centered at $E_F$. (c-e) The band dispersions around $E_F$ at 8 K with $h\nu = 15$ eV. (c) and (d) are measured along $k_x$ whereas (e) along $k_y$. (f) The same as (d) but from the $c(2\times6)$ surface.
FIG. 3: (Color online). (a) Photoelectron spectrum (red markers) integrated along $k_x$ ($k_y = 0.2$ Å$^{-1}$). A solid line between ±0.1 eV is the fitting with the power-law function ($E^a$) convolved with Gaussian broadening from experimental energy resolution ($\Delta E$). (b) Temperature dependence of the photoelectron intensity at $E_F$. The solid (blue) line is the fitting curve based on the power-law scaling ($T^{0.73}$) convolved with $\Delta E$ (20 meV). The horizontal dashed lines indicate the intensity at $E_F$ with $\Delta E$ at 0 K and the other line is $T^{0.73}$. (c, d) The same as (a) but taken at higher temperatures. Fitting curves are based on the finite temperature TLL spectral function presented in Ref. [21].
FIG. 4: (Color online). (a) ARPES energy distribution curves measured along \( k_x \) \((k_y = 0.2 \text{ Å}^{-1})\). Ticks indicate peak or shoulder positions on spectrum. (b) Second-derivative ARPES image measured with \( h\nu = 15 \text{ eV} \). Markers in the right side are the same positions as those in (a). Dashed lines are guides to the eyes for two separate dispersions.