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Superconducting single-atomic-layer Tl-Pb compounds on Ge(111) and Si (111) surfaces



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ABSTRACT

The set of known single-atomic-layer (SAL) superconductors has been limited so far by a few examples, including Si(111)-hex- $\sqrt{7} \times \sqrt{3}$ -In reconstruction, Si(111)-SIC-Pb and -HIC-Pb phases, and Si(111) $\sqrt{3} \times \sqrt{3}$ -(Tl, Pb) compound phase. In the present study where transport properties of SAL compounds of Tl-Pb with different composition ratios on Ge(111) and Si(111) substrates have been studied *in situ* in ultrahigh vacuum, the list of SAL superconductors has been enlarged by three new members, Ge(111) $\sqrt{3} \times \sqrt{3}$ (Tl, Pb), Ge(111)3×3-(Tl, Pb), and Si(111)4×4-(Tl, Pb) systems which show superconducting transition at the critical temperatures of 2.03 K, 0.83 K and 0.79 K, respectively. Bearing in mind that the atomic arrangements and electronic band structures of these SAL compounds of Tl-Pb have already been established, the obtained data set is believed to constitute a solid basis for the prospective theoretical investigations on the nature of SAL superconductivity. In addition, all the new systems demonstrate a noticeable Rashba-type spin splitting in the metallic surface-state bands and, therefore, they might be promising materials for future superconducting spintronics as well as for basics research of exotic superconductivity.

1. Introduction

It has been believed for a long time that superconductivity, a fascinating ability of some materials to exhibit zero resistance at low temperature, is a property of bulk materials and should be suppressed in extremely thin films, since large thermal and quantum fluctuations, proximity effects and scattering at surfaces should disturb the coherent motion of Cooper pairs. However, in their seminal work [1], Zhang et al. demonstrated that a single atomic layer (SAL) was quite enough to retain superconductivity. The goal was reached with metal-adsorbed silicon surface reconstructions. Since then, the SAL superconductors have attracted considerable interest, but up to now there are only a few examples known, including Pb/Si(111), so-called SIC and HIC (stripped and hexagonal incommensurate) phases [1-3], quasi-hexagonal Si (111)-hex- $\sqrt{7} \times \sqrt{3}$ -In reconstruction [2] (the other superconducting quasi-rectangular Si(111)-rec- $\sqrt{7} \times \sqrt{3}$ -In reconstruction [1,4,5] actually contains two-atom-layer thick In [6,7]), and a monolayer compound Si(111) $\sqrt{3} \times \sqrt{3}$ -(Tl, Pb) reconstruction [8]. Thus, finding new

SAL superconductors is required to extend variety of material systems for SAL superconductors, which may lead to finding exotic superconductivity, higher critical temperature T_c , and applications to superconducting electronics.

A possible search direction might reside in modification of the known superconducting films. In principle, this can be done in three possible ways, namely, (i) by adding appropriate adsorbates onto SALs, (ii) by changing a substrate for growth of SALs, and (iii) by varying the SAL composition if the SAL is a compound. Regarding item (i), possibility for both increasing and decreasing T_C of the In/Si(111) reconstruction by overgrowth of metal-phthalocyanine molecular monolayers was demonstrated in Ref. [9]. As for items (ii) and (iii), the family of Tl-Pb compounds on Si(111) and Ge(111) surfaces provides an unique possibility to explore effects of changing the substrate and composition ratio.

In the present paper, we report on the results of the low-temperature transport measurements on the Ge(111) $\sqrt{3} \times \sqrt{3}$ -(Tl, Pb), Ge(111) 3×3 -(Tl, Pb), and Si(111) 4×4 -(Tl, Pb) systems and compare them with

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the known data reported for the Si(111) $\sqrt{3} \times \sqrt{3}$ -(Tl, Pb) system [8]. All systems were found to be SAL superconductors characterized by different T_C .

2. Experimental details

In situ electrical resistivity measurements were performed with an ultra-high-vacuum four-point-probe (UHV-4PP) system [10], consisting of four copper wires with 100 μ m in diameter and the probe spacing of 200 μ m, which were attached to a low-temperature scanning tunneling microscopy (STM) head. The sample and the 4PP were cooled down to 0.8 K, and a magnetic field up to 7 T could be applied perpendicular to the sample surface. The system was also equipped with reflection high-energy electron diffraction (RHEED) facility for monitoring sample structures during growth of SALs as well as the surface structures of substrates.

3. Results

3.1. Formation, composition and structure of Tl-Pb compounds

To set the stage, let us start with describing the formation procedures, compositions and atomic arrangements of the Tl-Pb compounds under investigation. In all cases, the growth was started with preparation of the 1×1 -Tl surface reconstruction which was the same for both Si(111) and Ge(111) substrates. It contains 1.0 ML (monolayer, corresponding to 7.8×10^{14} cm⁻² for Si(111) and 7.2×10^{14} cm⁻² for Ge (111)) of Tl atoms occupying every T₄ site on the bulk-truncated substrate surfaces [11-13]). The 1×1-Tl surfaces were prepared either by adsorbing Tl onto the atomically clean $Ge(111)c(2 \times 8)$ surface held at about 150 °C or onto the atomically clean Si(111)7×7 surface held at about 300 °C. The Si(111) $\sqrt{3} \times \sqrt{3}$ -(Tl, Pb) and Ge(111) $\sqrt{3} \times \sqrt{3}$ (Tl, Pb) compound reconstructions were obtained by depositing 1/3 ML of Pb onto the 1×1 -Tl surfaces at room temperature (RT). Both reconstructions have the same atomic arrangement [14,15] with very similar structural parameters (see Fig. 1 (a) and Table 1). They are built of 1.0 ML of Tl atoms arranged into the honeycomb network of chained trimers (Kagome lattice) and 1/3 ML of Pb atoms occupying the honeycomb units. The T₁ sites are those where the Tl trimers are centered and Pb atoms reside.

When Pb atoms are further deposited, the other phases of Tl-Pb compounds, Ge(111)3×3-(Tl,Pb) [15] and Si(111)4×4-(Tl, Pb) [16], appear as a result of partial substitution of Tl by Pb atoms. The detailed descriptions of their formation processes, evaluation of their composition and atomic arrangement are given elsewhere [15,16]. In brief, the

Table 1

Structural parameters of $\sqrt{3} \times \sqrt{3}$ -(Tl, Pb) SALs on Si(111) and Ge(111) substrates [14,15]. *d*(Tl-Tl) is the side length of the Tl-atom trimer, *h*(Tl(Pb)-Ge (Si)) is the distance between Tl(Pb)-atom layer and the top-layer Ge(Si)-atom layer, and *h*(Pb-Tl) is the height difference between Tl- and Pb-atom layers. The unit is Å for all values.

Substrate	d(T1-T1)	h(Tl-Ge(Si))	h(Pb-Ge(Si))	h(Pb-Tl)
Ge(111)	3.43	2.52	2.84	0.31
Si(111)	3.34	2.50	2.85	0.34

Ge(111)3×3-(Tl, Pb) compound (Fig. 1 (b)) contains 6 Tl atoms and 7 Pb atoms per 3 × 3 unit cell (i.e., ~0.67 ML of Tl and ~0.78 ML of Pb) and its structure can be visualized as hexagons of Pb atoms surrounded by Tl trimers. The Si(111)4×4-(Tl, Pb) compound phase (Fig. 1 (c)) contains 9 Tl atoms and 12 Pb atoms per 4 × 4 unit cell (i.e., ~0.56 ML of Tl and 0.75 ML of Pb) and its structure consists of hexagonal arrays of Pb atoms surrounded by triangular arrays of 6 Tl atoms and Tl trimers.

In this experiment, in order to obtain high-quality Ge(111) 3×3 -(Tl, Pb) and Si(111) 4×4 -(Tl, Pb) superstructures, slightly more amounts of Pb atoms (~0.8 ML) than the nominal coverages for the respective superstructures were deposited onto the 1×1 -Tl surfaces.

3.2. Superconducting properties

3.2.1. $Ge(111)\sqrt{3} \times \sqrt{3}$ -(Tl, Pb)

The results of the transport measurements on the Ge(111) $\sqrt{3} \times \sqrt{3}$ -(Tl, Pb) at low temperature are summarized in Figs. 2 and 3. Fig. 2 (a) shows the temperature dependence of the sheet resistivity which evidences the transition to the superconducting state at around 2.0 K, where the resistance reaches less than the detection limit. Accurate fitting of the experimental data to the Almazov-Larkin-Maki-Thompson (ALMT) correction [2] for including superconducting fluctuation above the transition, yields the critical temperature (T_c) of 2.03 K. It has been recognized that superconducting transitions in the 2D systems can exhibit signatures that are consistent with Berezinskii-Kosterlitz-Thouless (BKT) mechanism [17,18] where the dissipation (resistance) is related to binding-unbinding of vortex-anti-vortex pairs due to phase fluctuation in the Cooper pair wavefunction, in particular, proved for the Si $(111)\sqrt{3} \times \sqrt{3}$ -(Tl, Pb) system [8]. Fig. 2 (b) shows that the data for the present Ge(111) $\sqrt{3} \times \sqrt{3}$ -(Tl, Pb) system are also consistent with this behavior and yield $T_{BKT} = 1.97$ K, according to the Halperin-Nelson equation with T_c being fixed at 2.03 K. Remind that the transport measurements on the previous Si(111) $\sqrt{3} \times \sqrt{3}$ -(Tl,Pb) system yield $T_c = 2.25 \text{ K}$ and $T_{BKT} = 2.24 \text{ K}$ [8], slightly higher than those for the



Fig. 1. Structural ball-and-stick models (upper; plan view, lower; sectional view) of the (a) $Ge(111)\sqrt{3} \times \sqrt{3}$ -(Tl, Pb), (b) $Ge(111)3 \times 3$ -(Tl, Pb), and (c) $Si(111)4 \times 4$ -(Tl, Pb) compound reconstructions, respectively. Tl atoms are shown by purple balls, Pb atoms by green balls, and Ge or Si atoms by blue (T₁ site), yellow (T₄ site) and white (H₃ site) balls. The reconstruction unit cells are outlined by the red rhombuses.



Fig. 2. (a) Temperature dependence of the sheet resistivity for Ge(111) $\sqrt{3} \times \sqrt{3}$ -(Tl,Pb) system. The red solid line is a result of the least-square fit to the Almazov-Larkin-Maki-Thompson correction [2] with $T_c = 2.03$ K. (b) Detailed temperature dependence of the sheet resistance below T_c . The red solid line depicts the expected BKT transition behavior, to obtain $T_{BKT} = 1.97$ K.

present Ge(111) substrate.

Fig. 3 (a) shows the sheet resistivity of the Ge(111) $\sqrt{3} \times \sqrt{3}$ -(Tl, Pb) system as a function of temperature under different magnetic fields, while the results of the magnetoresistance measurements at different

temperatures are shown in Fig. 3 (b). The data are summarized in Fig. 3 (c) showing the temperature dependence of the upper critical field (H_{c2}) at which the sheet resistivity is a half of the normal-state resistivity. Evaluation of the obtained data in the framework of Ginzburg-Landau



Fig. 3. Change of the sheet resistivity of the Ge(111) $\sqrt{3} \times \sqrt{3}$ -(Tl, Pb) system (a) with temperature under different magnetic fields and (b) with magnetic field at different temperatures. (c) Relation between the critical temperature T_c and the upper critical field H_{c2} , extracted from (a) (red squares) and (b) (blue circles) by simply defining the transition at the middles of the resistivity curves. (d) Result of the Ullah-Dorsey scaling theory analysis by using the data in (a). $\Delta\sigma$ in the vertical axis shows the conductivity derivation from that at T_c . The horizontal axis shows the temperature deviation from T_c . T_c under each magnetic field was determined so that all curves in (a) collapse into universal curves. (e) Relation between T_c and H_{c2} defined from the Ullah-Dorsey scaling theory.

(GL) theory yields the GL coherence length at zero temperature $\xi_{GL}(0)$ for the Ge(111) $\sqrt{3} \times \sqrt{3}$ -(Tl, Pb) as ~18.5 nm.

However, the present result in Fig. 3 (c) shows slight upturn at lower temperature regime (< 1.3 K), while the GL theory predicts a linear relation in the $H_{c2} - T_c$ diagram. This means that our determination of T_c and H_{c2} described above is too crude, because the resistivity curves in Fig. 3 (a) and (b) are deformed under different magnetic fields and at different temperatures, which we should take into account for determining T_c and H_{c2} .

In the 2D superconductors, it is known that excess conductance is generated by the fluctuation of superconducting order parameter. Ullah and Dorsey calculated this effect by considering the fluctuation up to second-order contributions to the free-energy expansion using a Hartree approximation [19]. The excess conductivities under different magnetic fields are scaled with a universal relation [20,21]. Variable transformation of $R_{\text{sheet}}(T)$ under different magnetic fields (Fig. 3 (a)) results in the universal relation as plotted in Fig. 3 (d) by tuning T_c as a fitting parameter for each curve; the upper branch corresponds to the portions at $T < T_c$ while the lower branch is for $T > T_c$. The T_c values obtained by this scaling analysis is used for plotting the interpolated $H_{c2} - T_{c}$ relation as in Fig. 3 (e), which obeys a linear relation expected from the GL theory. Evaluation of the obtained data in the framework of GL theory yields the GL coherence length at zero temperature $\xi_{GL}(0)$ for the $Ge(111)\sqrt{3} \times \sqrt{3}$ -(Tl, Pb) as ~ 15.1 ± 0.3 nm and the upper critical field $H_{c2}^{\rm MF}(0)$ as ~ 1.44 ± 0.03 T.

3.2.2. Ge(111)3×3-(Tl, Pb)

The results of the transport measurements on the Ge(111)3 × 3-(Tl, Pb) system are summarized in Fig. 4. Fig. 4 (a) shows a temperature dependence of the sheet resistivity where a red curve is the fitting result by ALMT correction which yields the critical temperature $T_c = 0.83$ K. Since this temperature is close to the limit accessible in our experimental system, the zero resistance is not achieved. The same shortage comes out also in the resistance measurements with applied magnetic field (Fig. 4 (b) and (c)). As a result, they provide only qualitative information that the superconducting transition becomes broader and shifts to the lower temperature with increasing the magnetic field, and do not allow determination of quantitative characteristics such as the GL coherence length and the BKT transition temperature $T_{\rm BKT}$.

3.2.3. Si(111)4×4-(Tl, Pb)

Fig. 5 summarizes the results on the transport properties of the Si (111) 4×4 -(Tl, Pb) system. One can see that this is also a SAL superconductor with evaluated transition temperature of 0.79 K (Fig. 5 (a)). The critical temperature is yielded by fitting the results including the ALMT correction. Fig. 5 (b) and (c) summarize the data of the transport measurements with applied magnetic field. Similar to the case of Ge (111) 3×3 -(Tl, Pb) above, this data set also suffers from the fact that the transition temperature is below the temperature limit accessible in

the present experiment. Nevertheless, one can notice interesting features in Fig. 5 (b) in the region close to the low temperature limit, namely with increasing the magnetic field, a superconductor-insulator transition takes place and there is also a certain indication on the resistivity saturation towards the lowest temperature with finite residual values for R(T) curves measured at magnetic field of 0.05, 0.10 and 0.15 T. This behavior is reminiscent of that observed on the doubleatomic Tl layer on Si(111) [22], which was interpreted as a magneticfield-induced superconductor-insulator transition intermediated by a quantum-metal state [23]. It is worth noting, however, that measurements conducted at further lower temperatures are apparently required to prove this suggestion. Fig. 5 (c) shows magnetoresistance curves at different temperatures. All curves cross at a point around 0.24 T above which the resistivity decreases with cooling while the resistivity increases with cooling below the field. It indicates again a magnetic-fieldinduced superconductor-insulator transition.

4. Discussion

Table 2 summarizes the obtained superconducting parameters of various phases of Tl-Pb SAL compounds, supplemented with the data for the relevant Si(111) $\sqrt{3} \times \sqrt{3}$ -(Tl, Pb) and Si(111)-SIC-Pb systems in the literature. Let us compare first the $\sqrt{3} \times \sqrt{3}$ -(Tl, Pb) SALs on Si (111) and Ge(111) which have almost identical atomic arrangements but different hosting substrates. One can see that both are superconductors with similar critical temperatures, which means that superconducting properties are associated mainly with the Tl-Pb metal layer, without significant influence from the substrates. Nevertheless, the T_c is apparently lower on Ge(111) than on Si(111), 2.03 K versus 2.25 K. There are several possible reasons for this difference. First, one could suppose some coupling of the substrate phonons with the electrons of the metal atomic layer. This might be thought as a certain analog of the known isotopic effect in bulk superconductors where the critical temperature varies inversely with the square root of the atomic mass [24]. Bearing in mind that the mass ratio of Ge and Si atoms is ~2.6, the observed difference of T_c between on Ge and on Si is much smaller than this expectation. The other possible reason might be associated with the difference in electronic band structure of the two systems. In case of the Si(111) $\sqrt{3} \times \sqrt{3}$ -(Tl, Pb), the projection of the Si bulk valence band lies below the Fermi level E_{F} , while for the Ge $(111)\sqrt{3} \times \sqrt{3}$ -(Tl, Pb), the maximum of the bulk valence band is above E_{F} [15]. Then, additional hole doping into the Tl-Pb layer from the Ge substrate might suppress its superconducting properties. One could also remind the difference in the lattice constants of Si and Ge crystals, which naturally produces different strain in the Tl-Pb layer and interface, a possible factor affecting the superconducting properties.

The data for the Si(111) 4 × 4-(Tl,Pb) and Ge(111) 3 × 3-(Tl,Pb) compounds demonstrate that increasing the Pb content in the Tl-Pb compound layer leads to the lowering of T_c with respect to those for the



Fig. 4. Change of the sheet resistivity of Ge(111)3×3-(Tl, Pb) system (a) with temperature at zero magnetic field and (b) under different magnetic fields, and (c) with magnetic field at different temperatures.



Fig. 5. Change of the sheet resistivity of the Si(111)4 \times 4-(Tl, Pb)) system (a) with temperature at zero magnetic field and (b) under different magnetic fields, and (c) with magnetic field at different temperatures.

Table 2

The amounts of Tl atoms Θ_{Tl} and Pb atoms Θ_{Pb} and the superconducting parameters, the critical temperature T_c and the GL coherence length at zero temperature $\xi_{\text{GL}}(0)$, for various phases of Tl-Pb compounds on the Si(111) and Ge(111) surfaces, together with those of the Si(111)-SIC-Pb phase given for reference.

System Θ_{T1} (M	L) Θ_{Pb} (ML)	T_c (K)	$\xi_{\rm GL}(0)$ (nm)	Ref.
$\begin{array}{ccc} Si(111)\text{-SIC-Pb} & 0 \\ Si(111)\sqrt{3} \times \sqrt{3}\text{-(Tl, Pb)} & 1.0 \\ Ge(111)\sqrt{3} \times \sqrt{3}\text{-(Tl, Pb)} & 1.0 \\ Si(111)4 \times 4\text{-(Tl, Pb)} & 0.56 \\ Ge(111)3 \times 3\text{-(Tl, Pb)} & 0.67 \end{array}$	1.3 0.33 0.33 0.75 0.78	1.1 2.25 2.03 0.79 0.82	74 ± 6 22.2 ± 0.3 15.1 ± 0.3 -	[2] [8] present work Present work Present work

 $\sqrt{3} \times \sqrt{3}$ -(Tl,Pb) compounds on both substrates. A possible reason might be associated with the specific atomic structures of these compounds. One can notice a kind of phase separation in their atomic arrangements, namely they are built of arrays solely of Pb atoms surrounded by boundary regions occupied solely by Tl. Bearing this in mind, one could remind that T_c for the pure Pb films thicker than 5 ML is as high as ~6.0 -6.7 K [17,25], but it drops rapidly for the thinner Pb films and is only 1.1 K for the SAL Si(111)-SIC-Pb phase according to the transport measurements [2]. The SAL Si(111)1 × 1-Tl reconstruction demonstrates a weak metallicity at RT [26,27] and undergoes a metal-insulator transition upon cooling to ~230 K, without superconducting transition [22]. Thus, decay of the Tl-Pb compound into the separate arrays of Pb and Tl atoms could be a reason for the poorer superconducting properties of these systems.

5. Conclusions

In conclusion, low-temperature transport measurements on the SAL Tl-Pb compounds on Ge(111) and Si(111), including Ge(111) $\sqrt{3} \times \sqrt{3}$ -(Tl, Pb), Ge(111) 3 × 3-(Tl, Pb), and Si(111) 4 × 4-(Tl, Pb) systems have been all revealed to be superconductors with the transition temperatures of 2.03 K, 0.83 K and 0.79 K, respectively. Together with the known information on their atomic arrangements and electronic band structures, the present results constitute a valuable data set which is awaiting its theoretical analysis for elucidating peculiarities of superconductivity in single-atomic-layer films. Additional merit that enhances scientific and technological significance of the Tl-Pb compounds resides in occurrence of the Rashba-type spin splitting of their metallic surface-state bands.

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