Supplementary Material: Observation of Coulomb Gap and Enhanced Superconducting Gap in Nano-Sized Pb Islands Grown on $\mathbf{SrTiO_3}^*$

Yonghao Yuan(袁永浩)1.2, Xintong Wang(王心童)1.2, Canli Song(宋灿立)1.2, Lili Wang(王立莉)1.2, Ke He(何珂)1^{,2}, Xucun Ma(马旭村)^{1,2}, Hong Yao(姚宏)^{1,2,3},

Wei Li(李渭)'^{,2**}, Qi-Kun Xue(薛其坤)'^{,2,4**}

¹ State Key Laboratory of Low-Dimensional Quantum Physics, Department of Physics, *Tsinghua University, Beijing 100084* ²*Collaborative Innovation Center of Quantum Matter, Beijing 100084* 3 *Institute for Advanced Study, Tsinghua University, Beijing 100084* 4 *Beijing Academy of Quantum Information Sciences, Beijing 100193*

* Supported by the National Science Foundation (No. 11674191), Ministry of Science and Technology of China (No. 2016YFA0301002), and the Beijing Advanced Innovation Center for Future Chip (ICFC). W.L. was also supported by Beijing Young Talents Plan and the National Thousand-Young-Talents Program.

**Corresponding authors. Email: weili83@tsinghua.edu.cn; qkxue@mail.tsinghua.edu.cn

1. Lattice structure of Pb island

Figure S1 exhibits the atomic resolution image of Pb island grown on STO substrate. It presents a hexagonal lattice with lattice constant of 3.49 \AA , consistent with that of the (111) surface of FCC phase Pb.

Figure S1. Atomic resolution of Pb island on STO. A STM image taken on a Pb island (set point voltage $(V_s) = 45$ mV, tunneling current $(I_t) = 40$ pA). It shows a hexagonal lattice with lattice constant of 3.49 Å.

2. The relationship between superconducting gap and Pb island size

Figure S2a-c show the dependence of superconducting gap on Pb island size. The superconducting gap size ranges from 2.2 meV to 2.8 meV, which may originate from the shell effect in nano-sized superconductors.^[1] The gap size doesn't show significant dependence on island height (see Figure S2a). The gap size seems to decrease with island area (Figure S2b), but the uncertainty of the gap size make it challenge to make a definite conclusion. The existence of the Coulomb gap, which is related with the island area, may have an affect the investigation as well.

Figure S2. Dependence of superconducting gap on Pb island size. (a-c) The correlation between superconducting gap and Pb island height, area and volume, respectively.

3. Coulomb gap

Figure S3 shows the d*I*/d*V* spectra taken on different Pb islands, where their superconductivity is totally suppressed by applying high magnetic fields. Coulomb gaps are clearly revealed in these spectra but some of them also exhibit non-negligible background. In order to compare the zero-bias conductance (ZBC) and obtain the reliable gap sizes from these data, the d*I*/d*V* spectra are normalized. The background in each spectrum is simulated by a Bezier curve drawn from the data outside the gap energy (the red curves in Figure S3a1 n1). Then the background is divided from each spectrum and the normalized data are shown in Figure S3a2-n2. The ZBC values (*G0*) are obtained from the normalized data.

As mentioned in the main text, the spectrum on the smallest island in Figure 2b shows a harder Coulomb gap and starts to present Coulomb blockade feature (also see Figure S3a2). The Coulomb gap width in this spectrum is determined by the lowest energy peaks denoted by green arrows in Figure S3a2. Except for this spectrum, the normalized spectra all show Vshaped gap feature, which is the signature for Coulomb gap. Density of states (DOS) in Coulomb gaps are linearly depend on energy and can be tracked well by linear fits (green lines in Figure S3b2-n2). The Coulomb gap width is determined by the bias voltage at which the value of the fitting function equals to 1.

Figure S3. Normalization of d*I*/d*V* spectra and Coulomb gap. (a1-n1) d*I*/d*V* spectra taken on Pb islands with applying high magnetic fields ($V_s = 10$ mV, $I_t = 200$ pA). The blue circles and red curves denote the raw data and the background, respectively. (a2-n2) Normalized spectra. The green arrows in (a2) denote the lowest energy peaks. The green lines in (b2-n2) are the linear fitting lines for Coulomb gaps.

4. Linear dependence of DOS on energy inside Coulomb gap

Figure S4. Linear dependence of DOS inside Coulomb gap. (a, b) Data points inside the Coulomb gap, which are extracted from the spectra in Figure S3d2. The DOS (*G*) and energy (*E*) show good linear relationship.

To demonstrate the linear increase of DOS in Coulomb gap, as an example, the in-gap data in Figure S3d2 are extracted and plotted in Figure S4. Correlation coefficients between DOS (*G*) and energy (*E*) are -0.9961 for negative bias and 0.9958 for positive bias. The absolute values are both very close to 1, indicating *G* is linearly depend on *E*. This result is consistent with Coulomb gap in two-dimensional systems whose DOS obeys: $G \sim |E|$.^[2]

5. **Upper critical field** (H_{c2})

In the $R = 83.7$ nm Pb island, a vortex is observed under applied magnetic fields. The ZBC values taken away from the vortex are plotted in Figure S5 and the upper critical field is estimated by a linear fit. Due to the existence of Coulomb gap, the ZBC can't reach 1 even at very high fields. Therefore, the upper critical field should be corresponding to the ZBC value = the G_0 value in Coulomb gap (in this case, $G_0 = 0.92$). As shown in Figure S5, the corresponding upper critical field of this island is 0.30 T.

The upper critical field is sensitive to the size of Pb island, since H_{c2} is determined by G-L coherence length ζ_{GL} : $\mu_0 H_{c2} = \Phi_0 / 2\pi \zeta_{GL}^2$. ζ_{GL} is monotonically increased with quasiparticle mean free path *l*. Taking the size effect into account, *l* approximately obeys: $1/l = 1/l_0 + 1/R$, where l_0 is the original mean free path. Therefore, the upper critical fields in smaller islands are expected to be higher than those in larger islands. The magnetic field response on a smaller island $(R = 32.8 \text{ nm})$ indeed obey this law (mentioned in the main text).

Figure S5. ZBC values as a function of magnetic field. ZBC values are extracted from the dI/dV spectra taken from the off-vortices regions. The upper critical field ~ 0.3 T is estimated by a linear fit.

6. Transition temperature (T_c)

Figure S6a-f demonstrate the fitting results of the spectra in Figure 3c. The gap size is obtained based on Dynes model^[3] in each temperature (the 400 mK spectrum as well as its fitting curve has been shown in the main text Figure 1c. The gap size as a function of temperature is shown in Figure S6g. The fitting curve by BCS theory suggests that the T_c is 7.16 K, lower than that of bulk Pb. The fitting curve at low temperature obviously deviates from the data. The origin for the deviation is that the measured gap could be contributed by both superconducting gap and Coulomb gap, which provides more impact at low temperature. If the data points taken at 400 mK and 1.8 K are excluded, the fitting curve is more reliable (see Figure S6h). The corresponding T_c is 7.31 K, which is still not significantly higher than that of bulk Pb.

Figure S6. Estimation of transition temperature. (a-f) d*I*/d*V* spectra and fitting curves of the *R* $= 30.8$ nm Pb island at various temperatures. The data points are taken from the spectra in Figure 3c and denoted by red circles. The blues curves are the fitting results by Dynes model. (g) Superconducting gap size as a function of temperature. The red circles denote the gap size and the blue curve shows the fitting result by BCS theory. It suggests that the transition temperature T_c is 7.16 K. (h) The fitting result with same method but exclude the two data points at 400 mK and 1.8 K. The corresponding T_c is 7.31 K.

References

- [1] Bose S et al. 2010 *Nat. Mater.* **9** 550
- [2] Efros A and Shklovskii B 1975 *J. Phys. C: Solid State Phys.* **8** L49
- [3] Dynes R C et al. 1978 *Phys. Rev. Lett.* **41** 1509