Two-dimensional superconductivity in monolayer iron-based superconductor $CsCa_2Fe_4As_4F_2$

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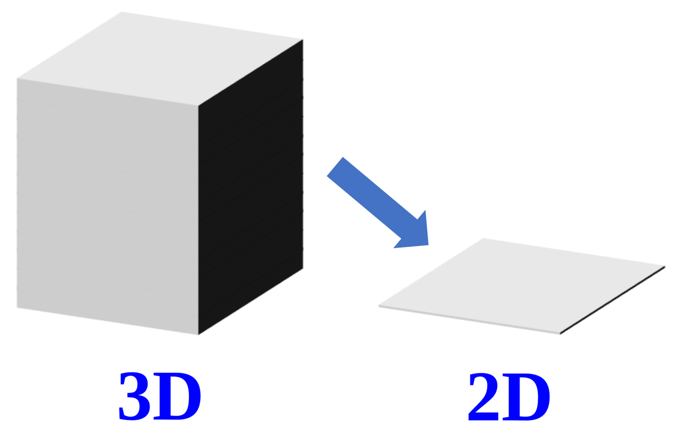
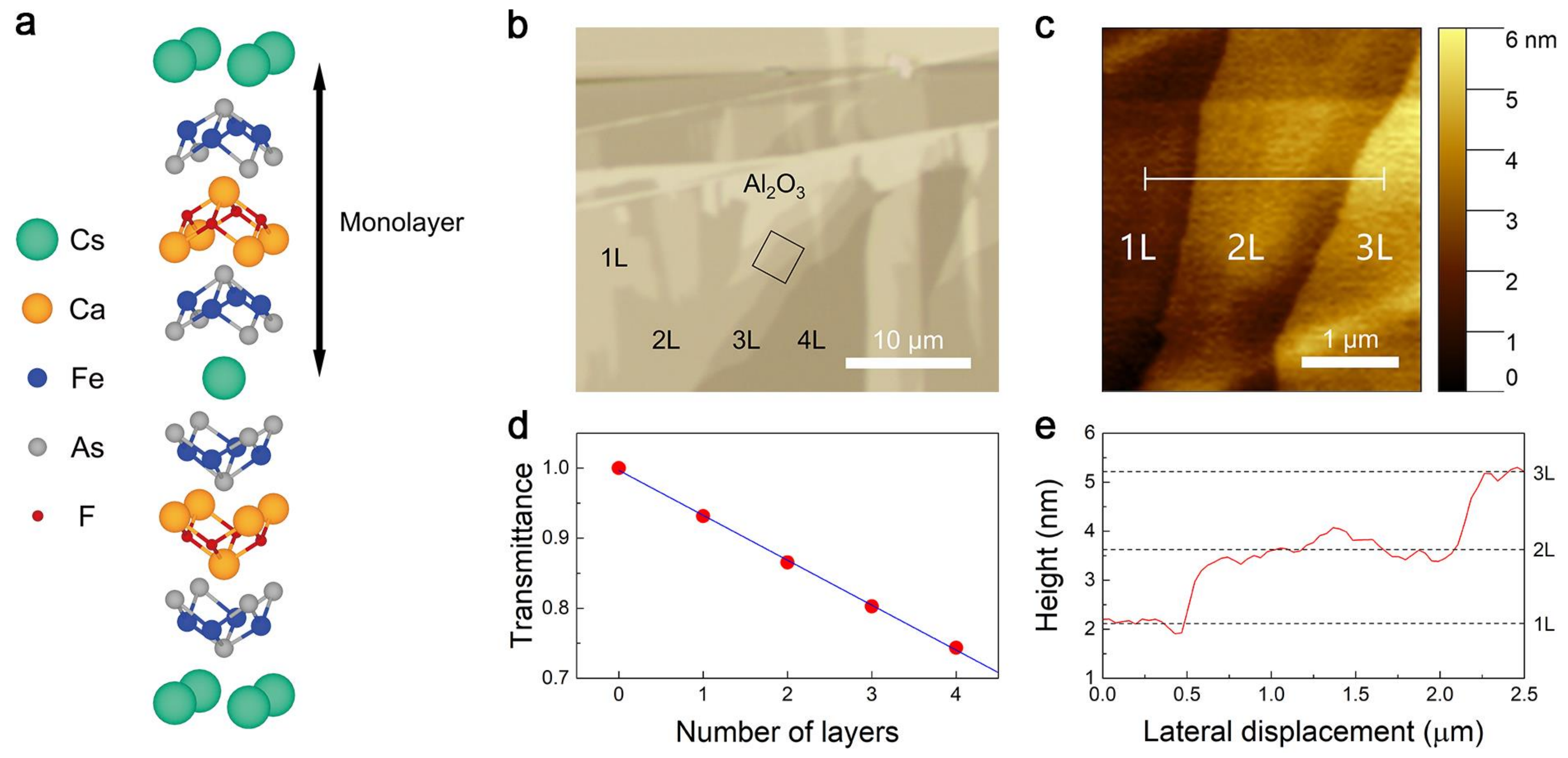
Introduction

Approaching the two-dimensional (2D) limit is considered as a significant method to study the mechanism of high-temperature superconductivity, particularly for cuprate and iron-based high-temperature superconductors that share a layered lattice structure. Rather than the great successes in the monolayer cuprate superconductor $Bi_2Sr_2CaCu_2O_{8+\delta}$ and the monolayer iron-based superconductor FeSe, study on the iron-arsenic superconductors approaching 2D limit is still elusive. Here, we successfully fabricate ultrathin flakes of iron-based superconductor $CsCa_2Fe_4As_4F_2$ (Cs-12442) down to monolayer, and we apply the electrical transport measurements to explore the superconductivity.

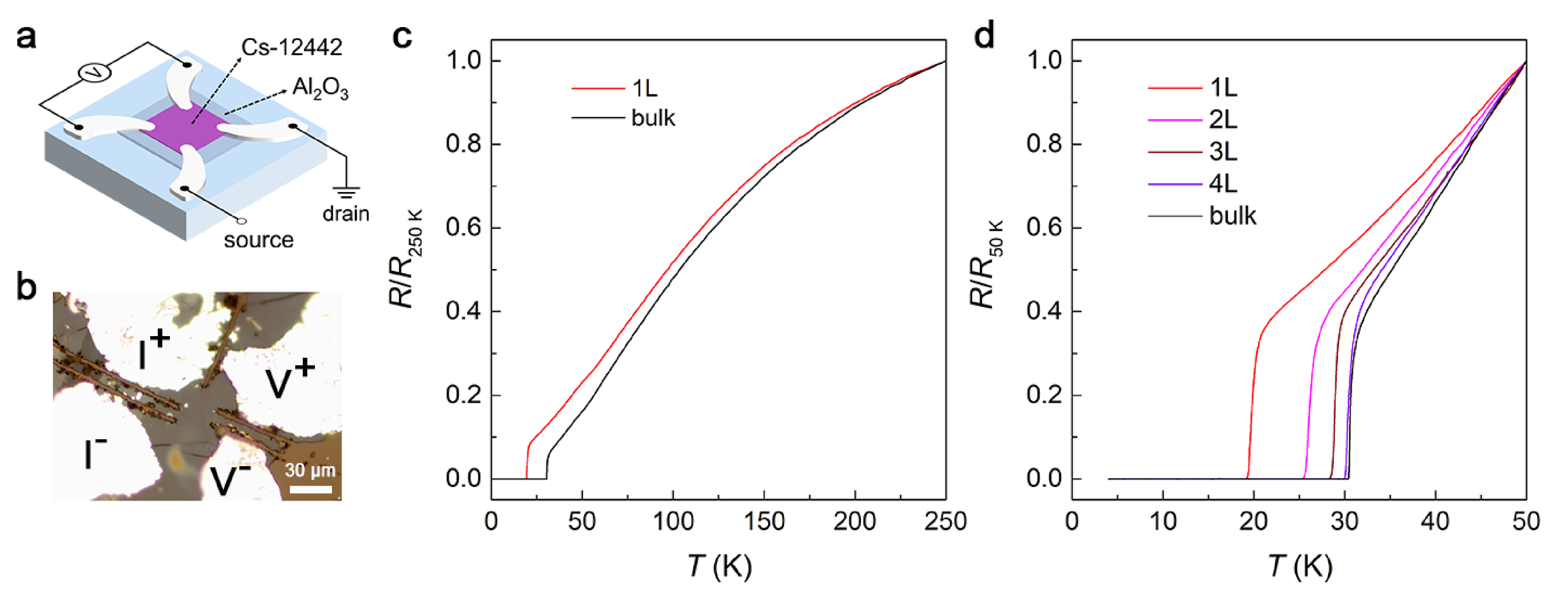


Exfoliation and characterization of monolayer Cs-12442



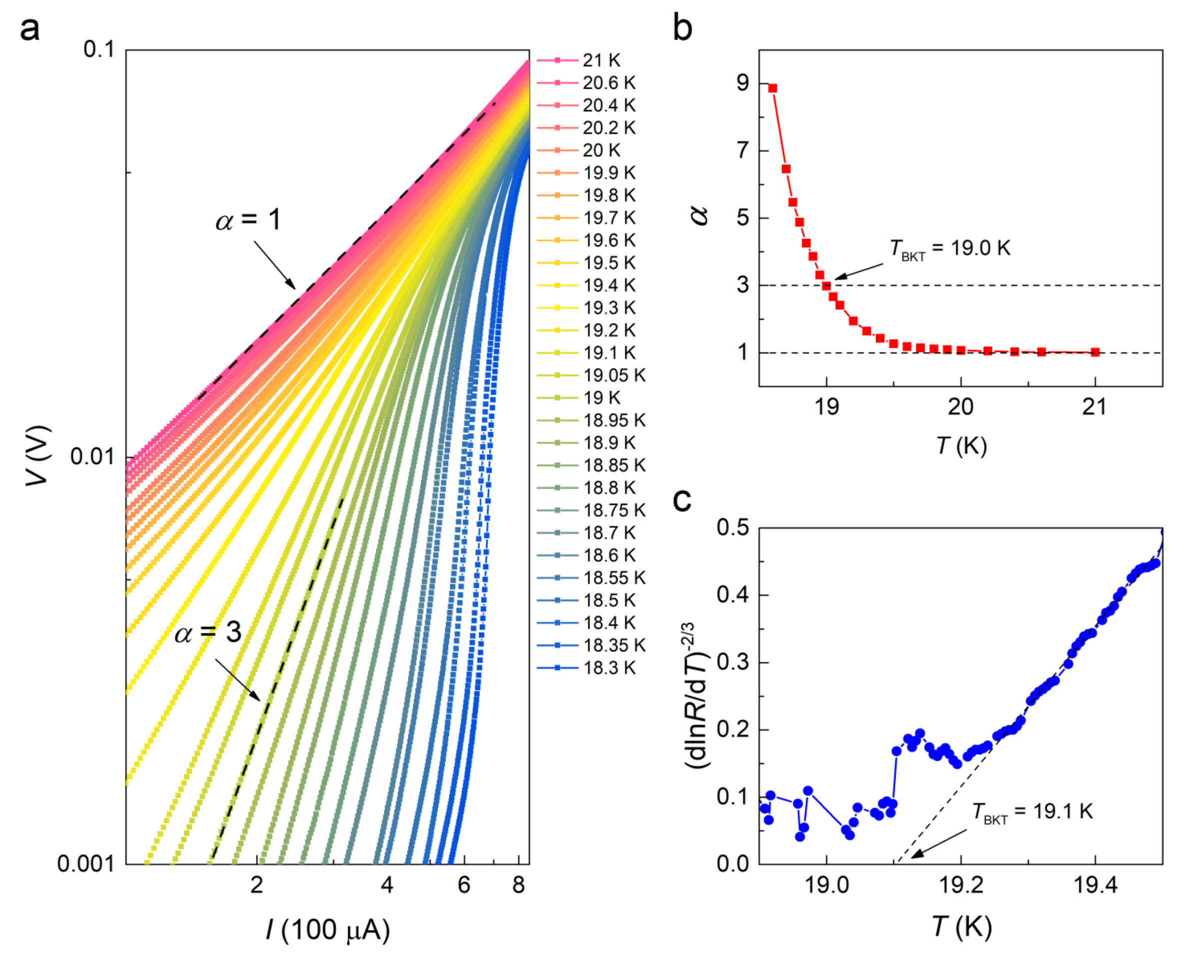
Experimentally obtaining large area monolayer Cs-12442 via conventional mechanical exfoliation turns out to be quite difficult. To overcome this challenge, the Al_2O_3-assisted exfoliation method is applied to our work. Here, the optical transmittance and the atomic force microscope (AFM) topography are combined together to precisely determine the layer numbers. The height difference between contiguous number of layers is approximately 1.6 nm, which is consistent with the thickness of a half unit cell of Cs-12442[1]. Therefore, we define one half unit cell of Cs-12442 as a "monolayer" (1L).

Electrical transport measurement of monolayer Cs-12442



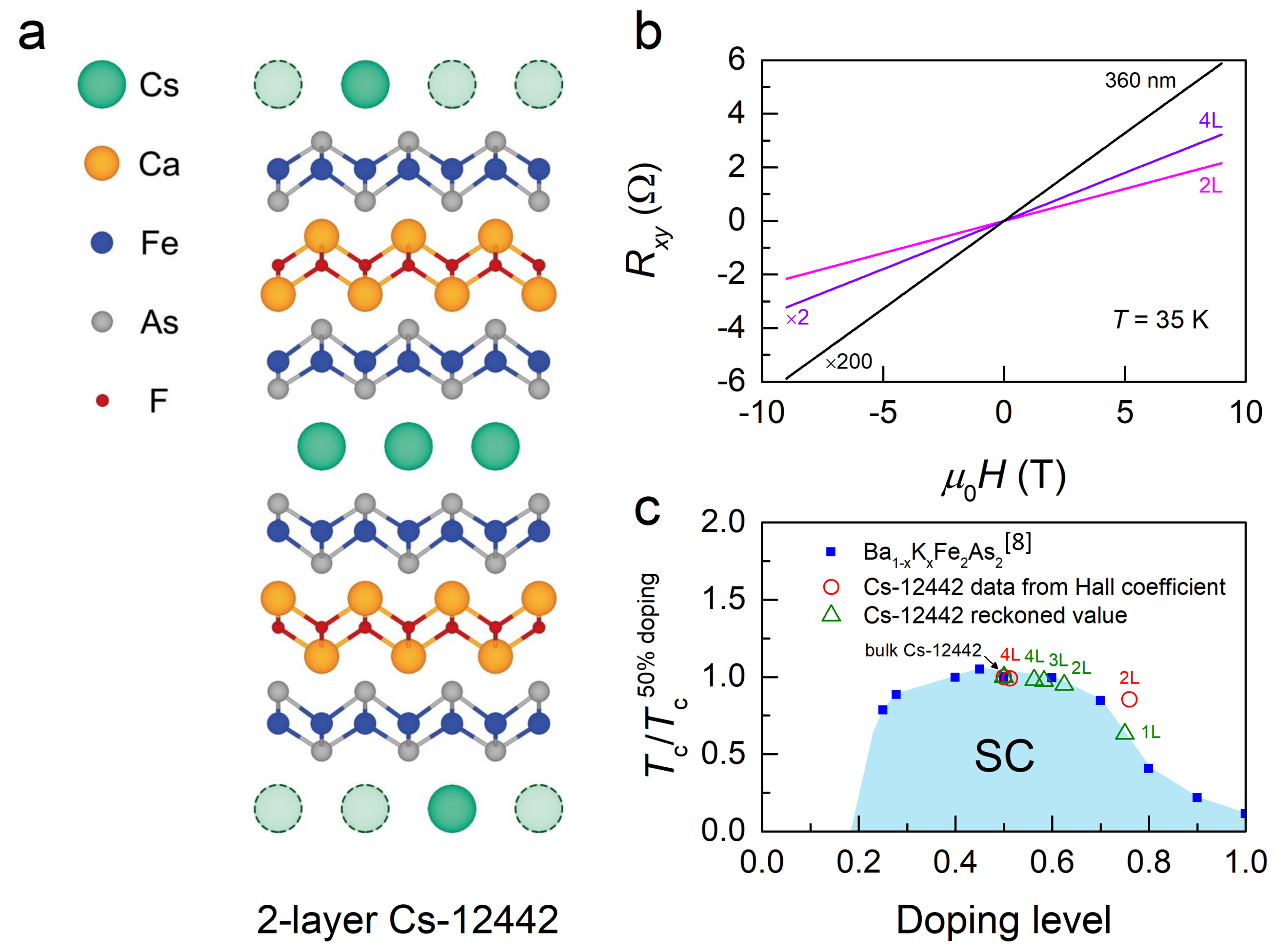
The four-probe method is applied to measure the sample resistance. As the film thickness decreases, the superconducting transition temperature (T_c) initially maintains unchanged (about 30 K) until 4 layers, and then decreases to 19.6 K for the monolayer.

Probing 2D superconductivity in monolayer Cs-12442

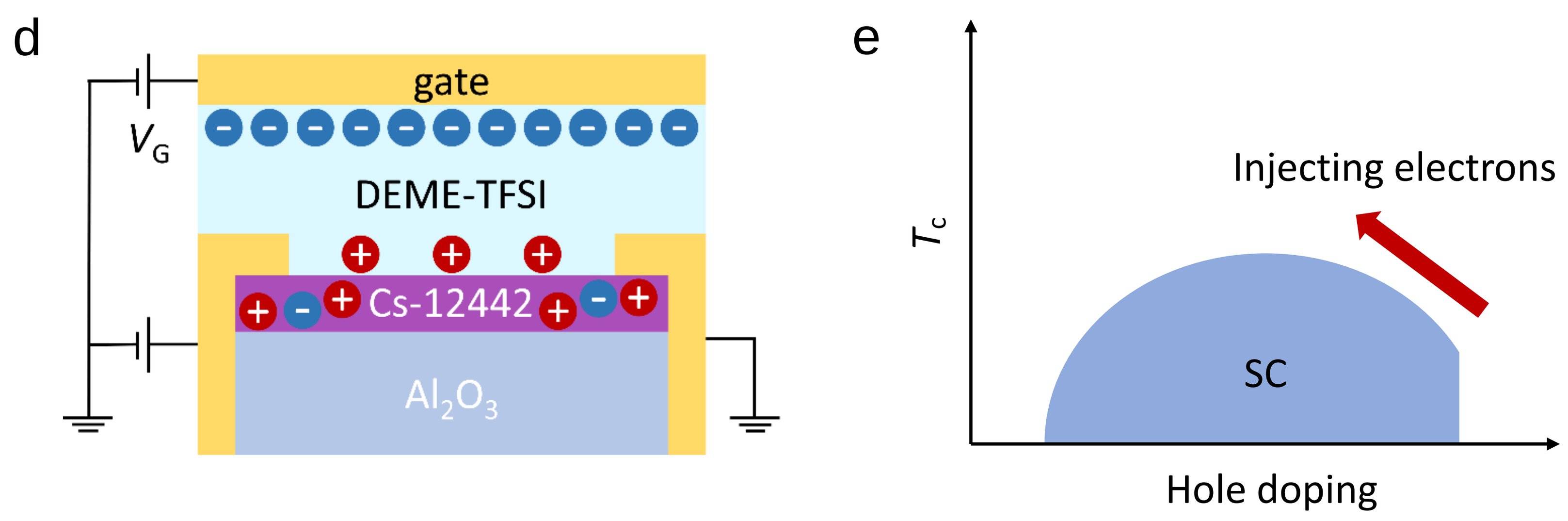


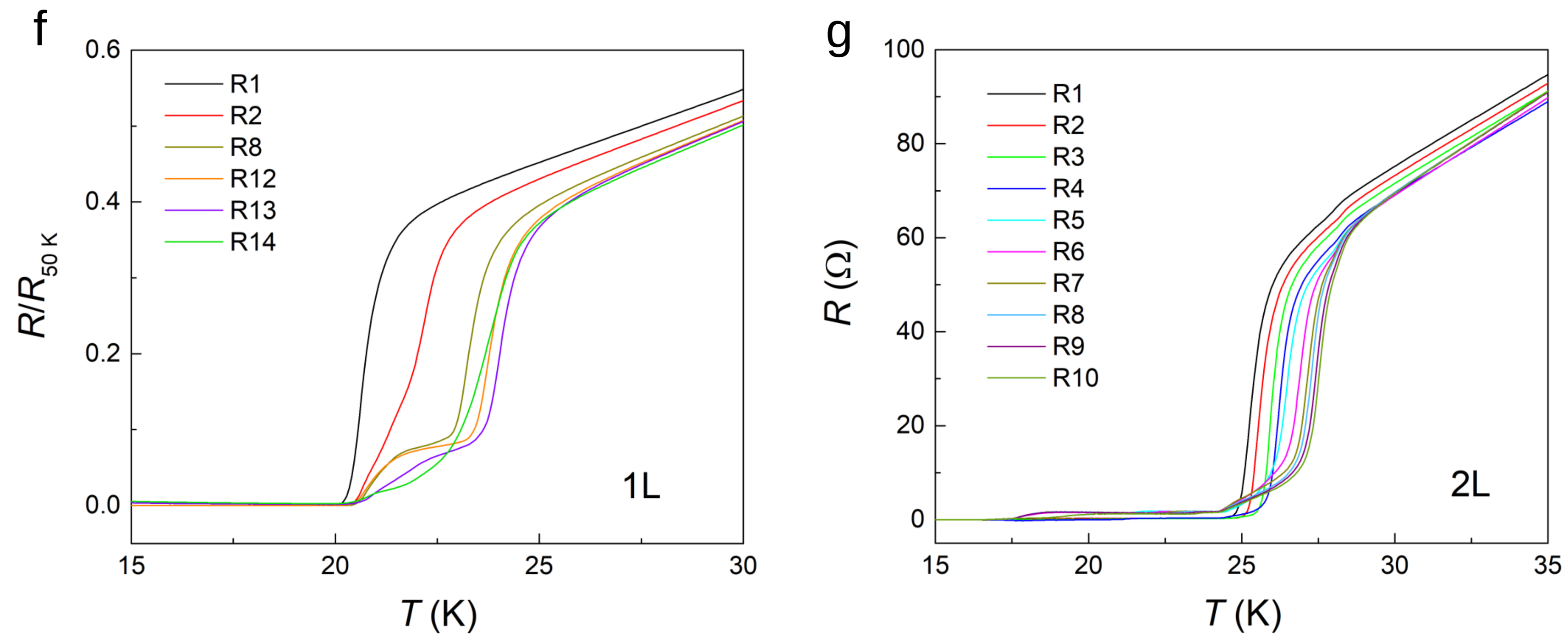
The signature of Berezinskii-Kosterlitz-Thouless (BKT) transition, which can be revealed from *I-V* characteristic curves, is regarded as evidence for 2D superconductivity[2, 3]. For the BKT transition, the current-induced Lorentz force causes vortex-antivortex pairs unbinding, resulting in a $V \sim I^{\alpha}$ behavior with $\alpha(T_{BKT}) = 3$. Besides, the *R-T* curve follows a typical BKT-like behavior with $R(T) = R_0\exp[-b(T/T_{BKT} - 1)^{-1/2}]$, when the temperature approaches T_{BKT}[4]. T_{BKT} obtained from both two methods shows to be close to T_c^{zero} (19.2 K), indicating the 2D superconductivity in monolayer Cs-12442.

Hole doping effect in Cs-12442 ultrathin flakes



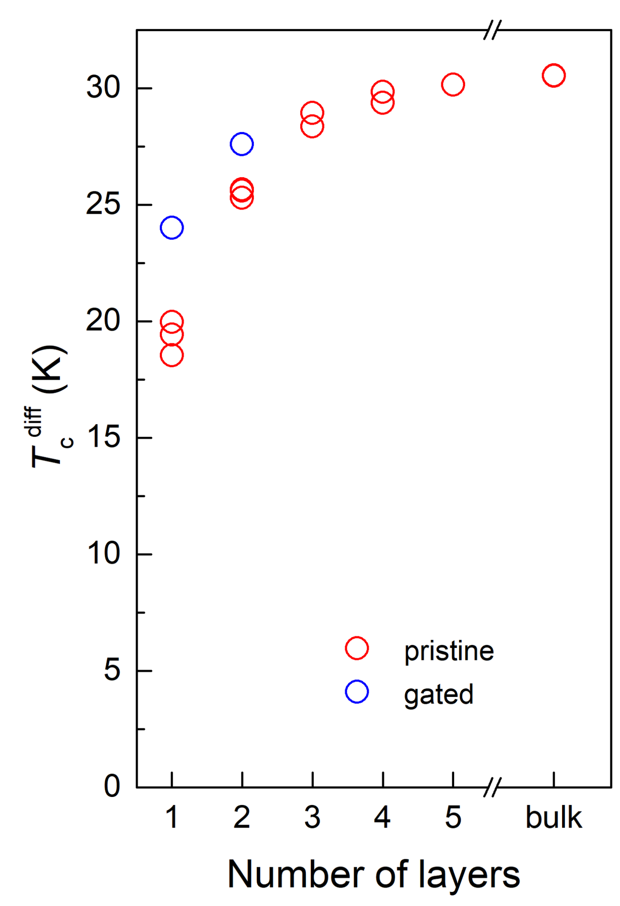
The cleavage in the 12442-type superconductors mainly appears along the alkali-metal atoms plane[5]. The Cs atoms left on the exfoliated surface can escape out or be oxidized easily, leading to more hole doping[6, 7], especially for the ultrathin flakes. The Hall measurements are applied to trace the carrier density, and an increase of hole doping is observed in Cs-12442 ultrathin flakes, indicating that the doping level of Cs-12442 ultrathin flakes deviate from the optimized level, resulting the reduction of T_c.





To further confirm the hole doping effect in the Cs-12442 ultrathin flakes, the ionic liquid gating is applied to modulate the carrier density. Under a positive gate voltage, electrons are injected into the sample, and an enhancement of T_c occurs both in the monolayer sample and the 2-layer sample. Here, the hole density in the gated sample is reduced towards the optimized level, thus the enhanced T_c confirms the hole doping effect.

Summary



- The monolayer Cs-12442 is successfully obtained by the Al_2O_3-assisted exfoliation method.
- With decreasing flake thickness, T_c initially maintains unchanged (about 30 K) until 4 layers, and then decreases to 19.6 K for the monolayer.
- Evidence of the 2D superconductivity in the monolayer is revealed by the signature of BKT transition.
- The reduction of T_c is mainly attributed to the change of doping, resulting from the loss of surface Cs atoms during exfoliation, indicated by Hall measurements and ionic liquid gating experiments.
- Compared with the bulk crystal, a reduction of T_c still exists in the gated monolayer and 2-layer samples, thus, we cannot completely deny the interlayer coupling effect. Yet the impact of it on T_c is far less significant than people imagine from our experiments.

References

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