Layer-cake 2-D superconductivity: Developing clean 2-D superconductivity in a bulk van der Waals superlattice

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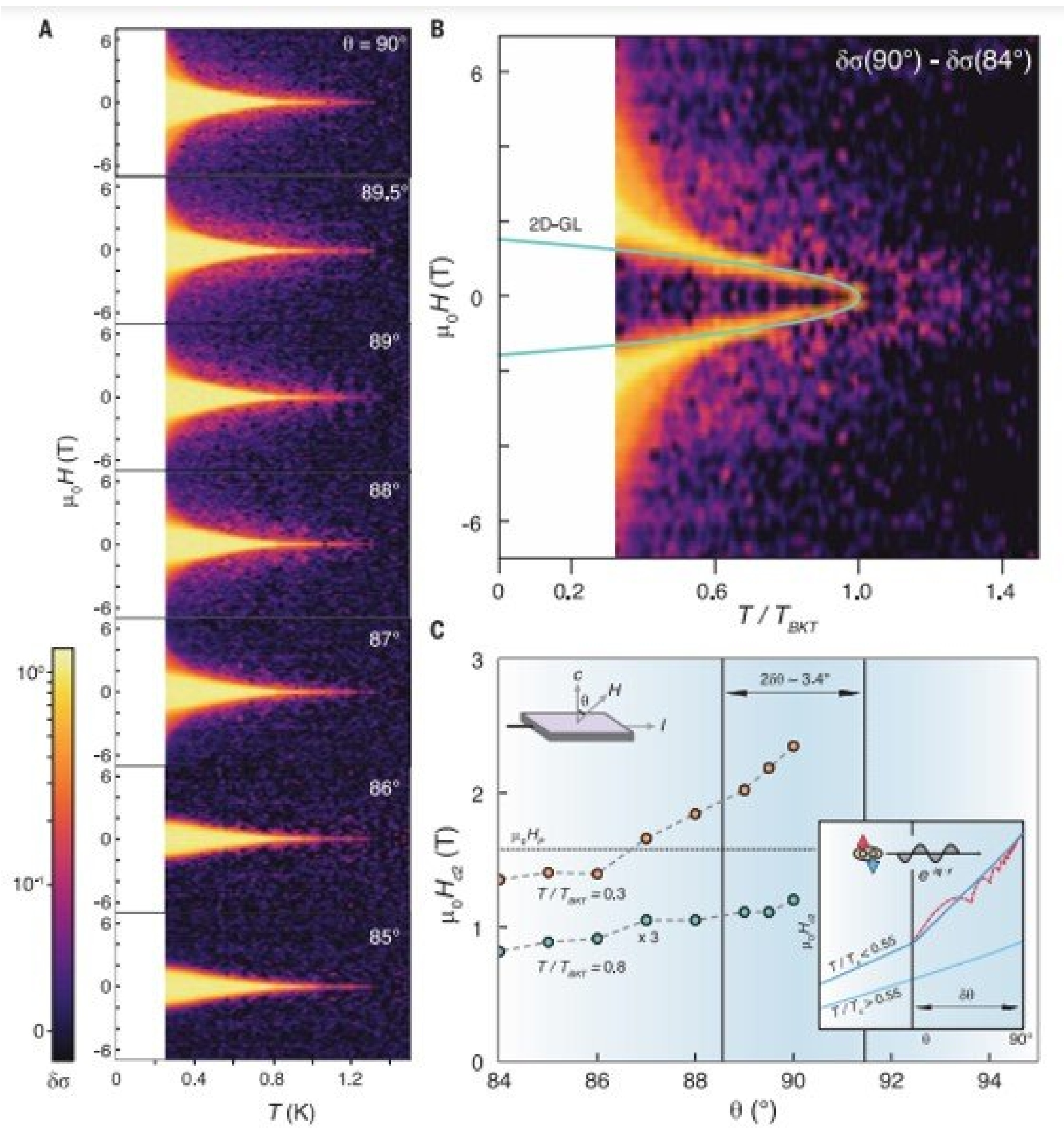


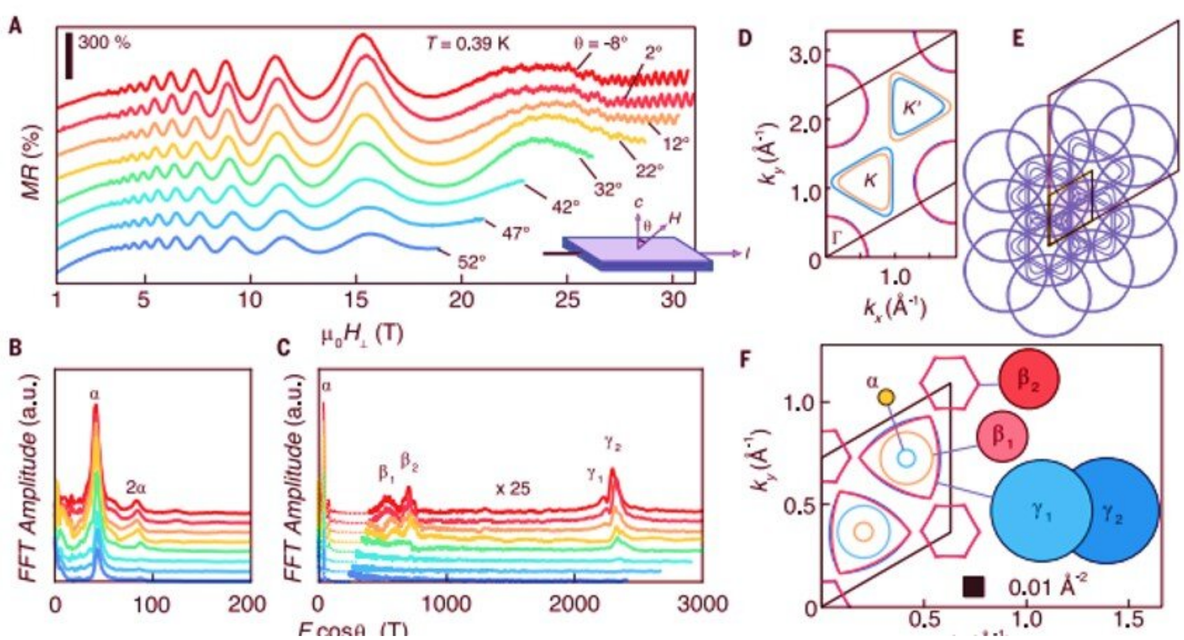
Superconducting phase diagram of Ba6Nb11S28. (A) Excess conductivity relative to the normal state ds(m0H, T) for field angles q near the ab-plane (q = 90°). (B) Difference between ds(m0H, T) for q = 90° and q = 84°. The temperature axis is normalized to TBKT. The green curve represents the 2D Ginzburg-Landau (2D-GL) model of m0Hc2. (C) Angular dependence of m0Hc2 at T/TBKT = 0.3 (orange) and m0Hc2 at T/TBKT = 0.8 (green, magnified by a factor of 3). Inset: Schematic depiction of m0Hc2 in a clean 2D system where an enhancement is expected within a critical region |q – 90°| HP (dark blue solid line). Theoretical studies of 2D FFLO superconductors further predict a cascade of magnetic vortex states that appear as a corrugation of m0Hc2(q) within this regime (37) (red dashed line). Credit: Science, doi: 10.1126/science.aaz6643

Materials science has had a profound historical impact on humanity since the advent of the Iron and Bronze ages. Presently, materials scientists are intrigued by a class of materials known as quantum materials, whose electronic or magnetic behavior cannot be explained by classical physics. Discoveries in the field of quantum materials are followed by a surge of research to uncover new physics or quantum information in science. In a new report now published on *Science*, A. Devarakonda and a team of scientists in physics at the Massachusetts Institute of Technology, Harvard University and the Riken Center for Emergent Matter Science in the U.S. and Japan reported the synthesis of a highly interesting novel quantum material.

The construct may allow physicists to study obscure quantum effects that have hitherto remained unknown. In this study, the team developed a bulk superlattice containing the transition metal dichalcogenide (TMD) superconductor 2H-niobium disulfide (2H-NbS_2, 2H phase) to generate enhanced two-dimensional (2-D), high-electronic quality and clean-limit inorganic superconductivity.

Superconductivity

Superconductivity can hypothetically allow high-speed applications without power loss and contribute to the development of concepts such as levitating express trains. Researchers can partly realize such applications at present using materials that superconduct at high enough temperatures and use 2-D materials to simplify problems, while highlighting the physics behind superconductivity. Early experimental works with granular aluminum (Al) and amorphous bismuth (Bi) films showed 2-D superconductivity by precisely controlling the superconducting layer thickness, which were later used in ground-breaking studies. These include the Berezinskii-Kosterlitz-Thouless (BKT) transition; an early example describing the topological transition, for which physicist received the Nobel prize in physics in 2016.

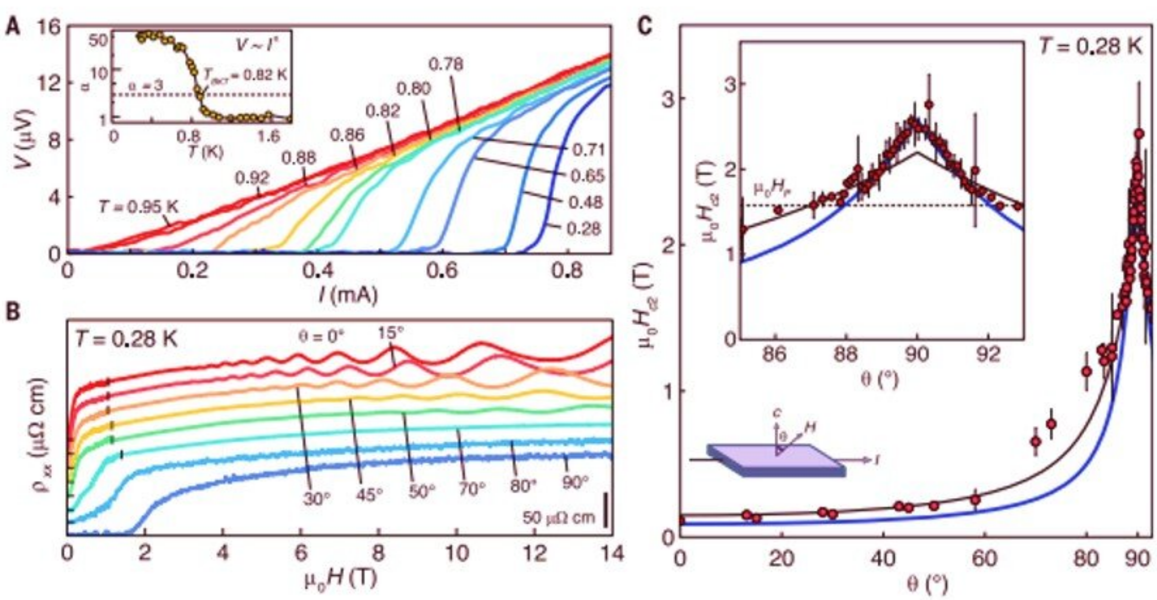


Quantum oscillations and electronic structure of Ba6Nb11S28. (A) Magnetoresistance as a function of perpendicular field at temperature T = 0.39 K for different field rotation angles q (geometry defined as shown in the inset). Curves are vertically offset by 150% of MR for clarity. (B and C) Low-frequency range (B) and full range (C) of quantum oscillation amplitude FFT as a function of perpendicular frequency F cos(q). The FFT amplitudes for the higher-frequency pockets are multiplied by 25. (D) DFT calculation of monolayer H-NbS2 Fermi surfaces including spin-orbit coupling (17). (E) Depiction of zone-folding scheme involving the 3 × 3 superstructure imposed by the Ba3NbS5 block layer where the reduced Brillouin zone is enclosed by the bold line. (F) Electronic structure of zonefolded monolayer H-NbS2 with experimentally observed Fermi surface cross-sectional areas drawn to scale as solid circles. The black box corresponds to 0.01 Å–2. Credit: Science, doi: 10.1126/science.aaz6643

In parallel, scientists have also studied anisotropic bulk superconductivity to understand the superconducting state in the context of 2-D superconductivity, which include transition metal dichalcogenides (TMDs), i.e. atomically thin semiconductors of the type MX_2 where M is a transition metal and X is a chalcogen atom (group 16 elements in the periodic table). Recent advancements in materials engineering have also shown the possibility to exfoliate van der Waals (vdW) layered materials to allow atomically thin 2-D superconductors to be readily accessible. However, such flakes of exfoliation can degrade, reducing the sample quality. Devarakonda et al. therefore used high quality 2H-NbS_2 (2H-niobium disulfide) monolayers in this work with a clean-limit 2-D superconductor exhibiting a BKT (Berezinskii-Kosterlitz-Thouless) transition. They then further synthesized a single-crystal material; $Ba_6Nb_{11}S_{28}$ using high-quality H-NbS_2 monolayers and Ba_3NbS_5 block layers, within which the TMD layers were strongly decoupled.

Developing and characterizing the layer-cake

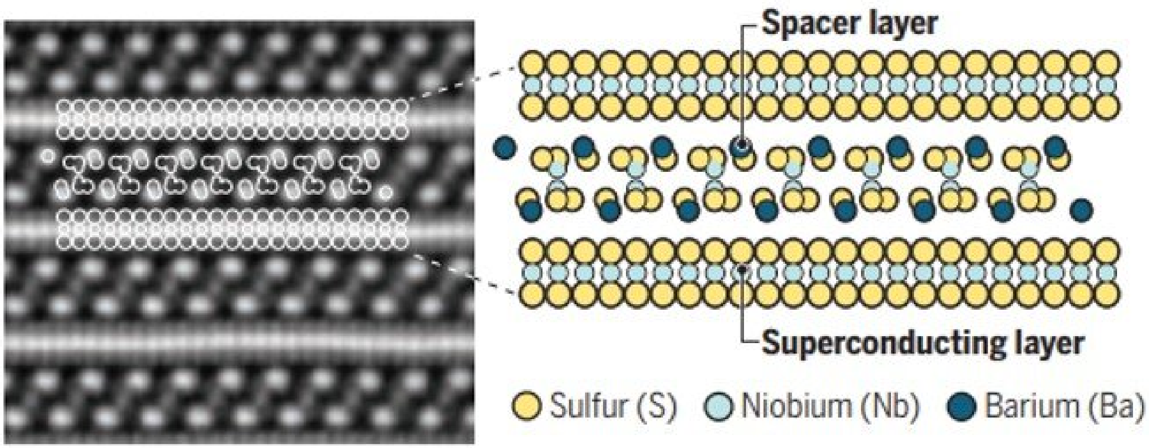
Devarakonda et al. therefore made the resulting material ($Ba_6Nb_{11}S_{28}$) as pure as possible to study the pure physics of 2-D superconductivity. The discovery of clean 2-D superconductivity in $Ba_6Nb_{11}S_{28}$ will open the door to better understand 2-D superconductivity associated with quantum phenomena. The material contained alternating layers of the 2-D superconductor NbS_2 and an electronically uninteresting spacer layer Ba_3NbS_5 – much like a layer cake with a thin layer of chocolate (i.e. NbS_2) between thicker layers of cake (i.e. the spacer layer). The layering protected the NbS_2 layer from cracking or air/moisture exposure to allow much cleaner 2-D superconductivity. The team used high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) to investigate the resulting structure. The material displayed a clean-limit 2-D superconductor exhibiting a BKT transition at T_{BKT} = 0.82 K and prominent 2-D Shubnikov-de Haas (SdH) quantum oscillations; a macroscopic manifestation of the inherent quantum nature of matter.



2D superconductivity and Pauli limit breaking in Ba6Nb11S28. (A) Current-voltage characteristics I(V) from T = 0.95 K to T = 0.28 K. The inset shows the evolution of the power law V º I a ; the horizontal line marks a = 3. (B) Longitudinal resistivity as a function of field m0H for different values of q. Curves are vertically offset by 20 mW·cm for clarity (horizontal lines). Vertical ticks separate regions measured with low current (7 mA) and higher current (70 mA) to avoid suppression of superconductivity by Joule heating. For q = 80° and 90°, only low current is used. (C) Angular dependence of upper critical field m0Hc2 measured at T = 0.28 K with fits to the 2D-Tinkham model, computed using data in the range |q – 90°| 1.7° (black curve), respectively. The inset shows a detailed view near q = 90° where an enhancement of m0Hc2(q) is observed across the Pauli limit m0HP. Credit: Science, doi: 10.1126/science.aaz6643

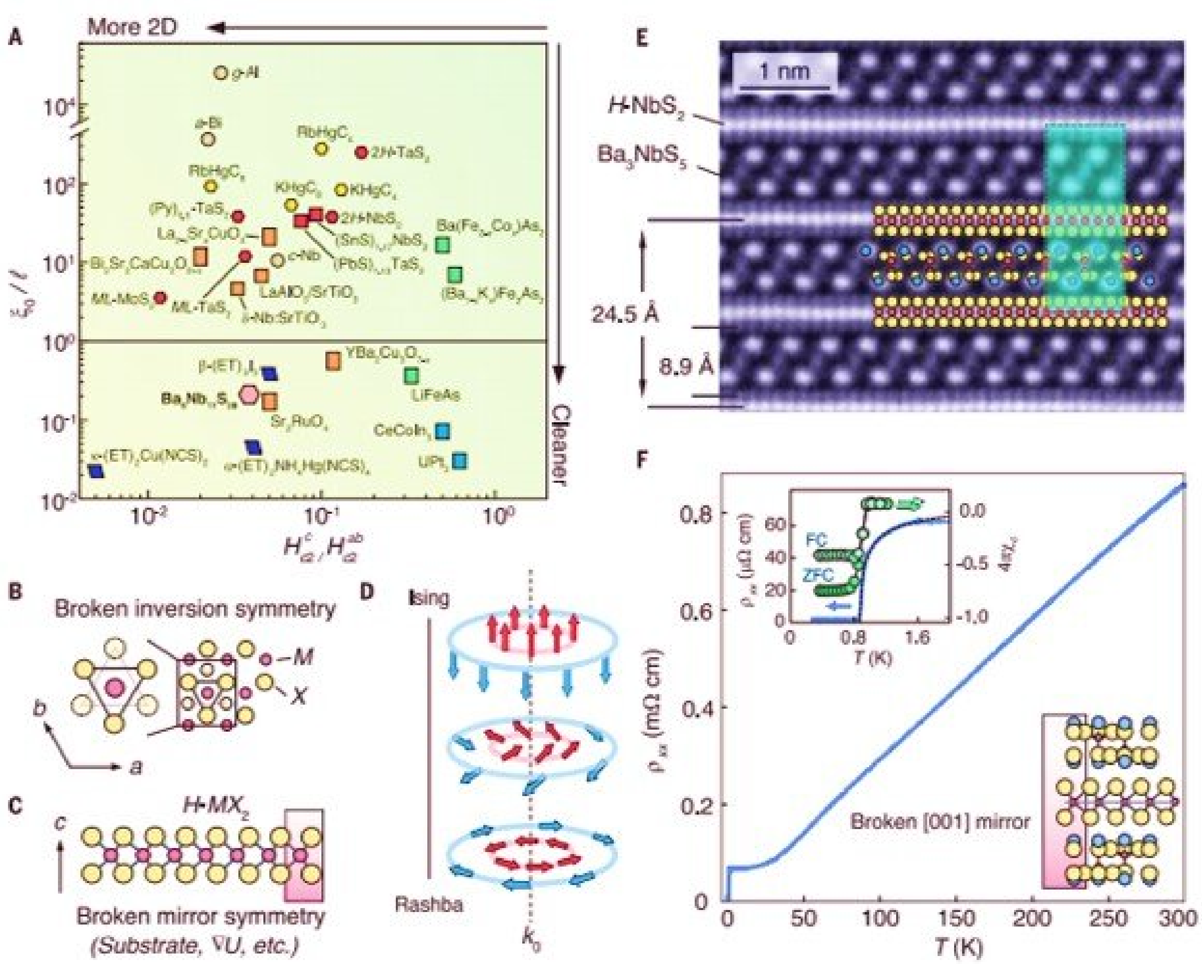
Properties of the new quantum material

Using magnetotransport measurements, Devarakonda et al. showed the cleanliness of the material and further evidence for the 2-D electronic architecture. The results were qualitatively different from the starting material 2H-NbS_2, which maintained warped and elliptical Fermi surfaces in its electronic structure. Although quantum oscillations had not yet been reported in 2H-NbS_2, the team noted the onset of Shubnikov-de Haas (SdH) quantum oscillations in $Ba_6Nb_{11}S_{28}$ within magnetic fields between 2 and 3 Tesla to indicate quantum mobilities. The scientists analyzed the quantum oscillations and low-field magnetoresistance of $Ba_6Nb_{11}S_{28}$, which placed the materials in the clean limit of superconductivity.



Alternating layers of superconducting NbS2 and a Ba3NbS5 spacer allow high electron mobility in the NbS2 while also protecting it. This creates a “layer cake”– like structure that permits clean superconducting behavior. Credit: Science, doi: 10.1126/science.abd4225

They also recorded the current/voltage characteristics of the material across the superconducting transition. In addition to the observed BKT transition, the work showed the appearance of a clean, 2-D superconducting state with enhanced stability, which Devarakonda et al. credited to the high purity of the NbS_2 layers in $Ba_6Nb_{11}S_{28}$. Since the bulk $Ba_6Nb_{11}S_{28}$ material already displayed 2-D physics, the research team originally proposed the now known process of inserting the spacer layers instead of fabricating exfoliated nanodevices detailed in previous work. The team also noted the scope to functionalize the spacer layer by introducing magnetic constituents. In this way, the large electronic mean free path (average distance traveled by a moving particle) of $Ba_6Nb_{11}S_{28}$ allowed clean-limit super-conductivity to potentially realize unconventional phases as predicted in monolayer superconductors.



2D superconductivity and Ba6Nb11S28. (A) Survey of superconducting materials characterized by anisotropy of the upper critical field Hc c2=Hab c2 and ratio of the Pippard coherence length to mean free path. The boundary between the clean and dirty limits is shown as a horizontal line. (B) Crystal structure of H-MX2 projected onto the ab-plane. Lack of inversion symmetry is illustrated by the missing chalcogen (X) inversion partners (dashed circles). (C) The ab-plane mirror symmetry in monolayer H-MX2 can be broken by substrates or local fields (∇U). (D) Depiction of momentum space spin-orbit texture for monolayer H-MX2 with varying degrees of Ising and Rashba coupling. (E) HAADF-STEM image of Ba6Nb11S28 taken along the axis (scale bar, 1 nm). A simulation of the model structure is overlaid with one unit cell shaded in green. Ba, Nb, and S atoms are depicted as blue, red, and yellow circles, respectively. (F) Resistivity as a function of temperature in Ba6Nb11S28 showing the superconducting transition. Upper inset: Magnified view of the transition) and magnetic susceptibility 4pcc measured with zero field cooling (ZFC) and field cooling (FC). Lower inset: H-NbS2 layer and mirror symmetry–breaking Ba3NbS5 block layers. Credit: Science, doi: 10.1126/science.aaz6643

Impact of the new quantum material

The inherent beauty of the material remains in the natural growth of the heterostructure, which much like a layer-cake naturally separated during the process of synthesis. This allowed the synthetic process to be much less labor-intensive compared to the manual addition of each layer. The ease of synthesis may allow different types of layered materials to be developed where the 2-D layers are naturally protected by their environment. The technique can yield different types of quantum materials aside from superconductors, including topological insulators suited for quantum computing. The new discovery allows a simpler, alternative approach to the existing process of exfoliated nanodevice fabrication. A. Devarakonda and colleagues envision extending this strategy to other materials beyond the $Ba_6Nb_{11}S_{28}$ detailed here.

More information: Devarakonda A. Clean 2D superconductivity in a bulk van der Waals superlattice, *Science*, 10.1126/science.aaz6643

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Yu Saito et al. Superconductivity protected by spin–valley locking in ion-gated MoS2, *Nature Physics* (2015). DOI: 10.1038/nphys3580