Gate-controlled ground state crossover in a two-dimensional superconductor

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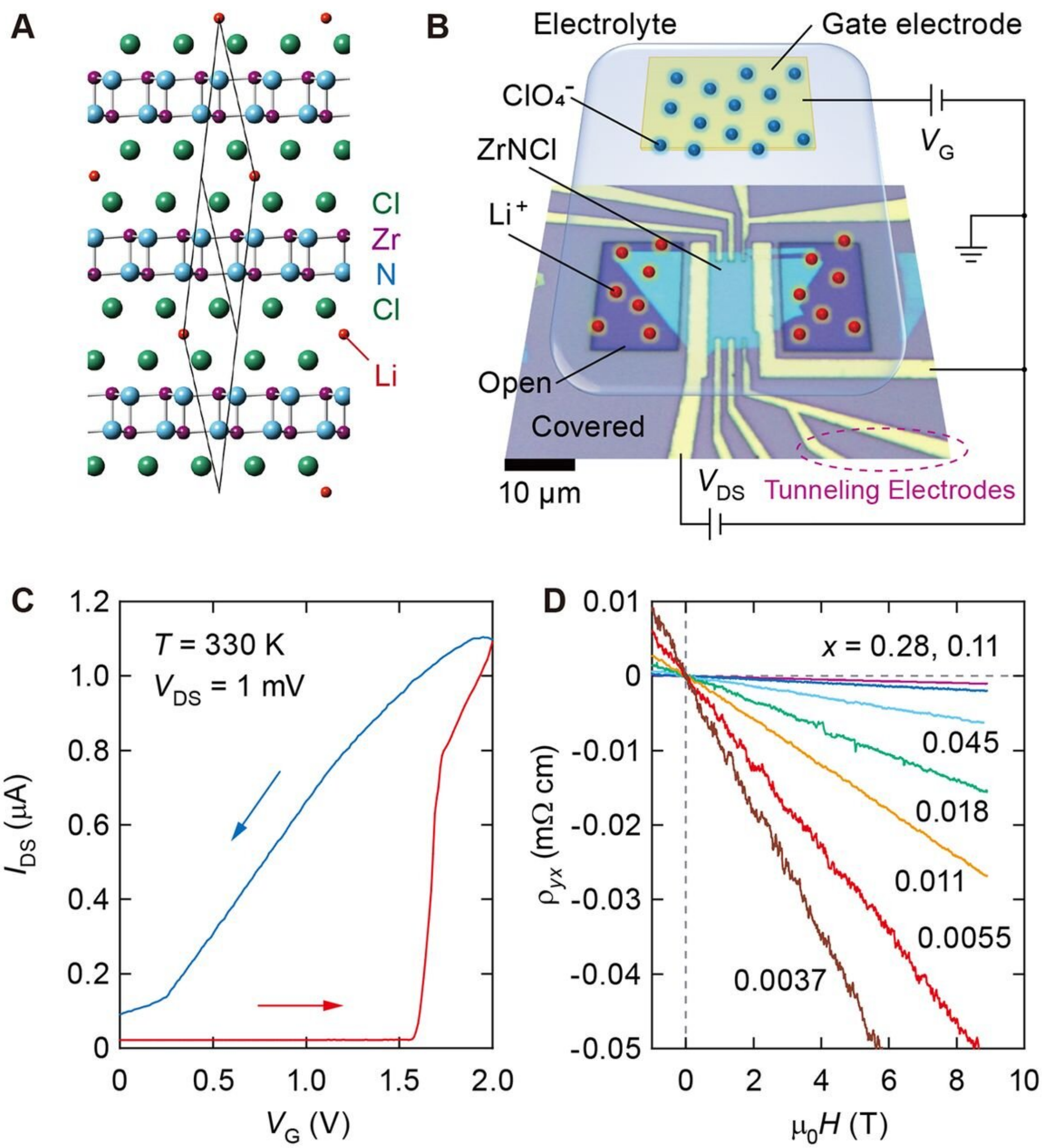
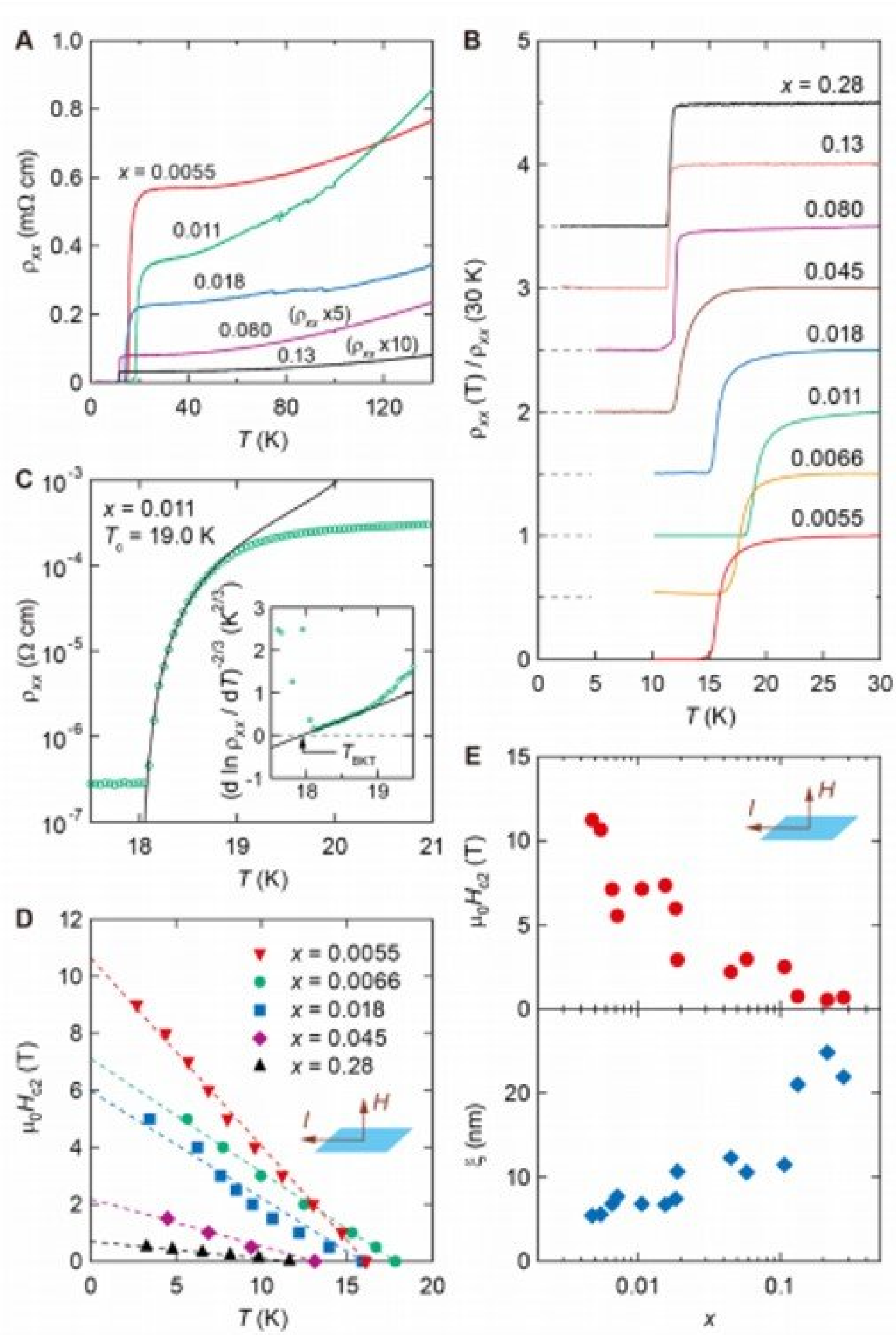


(A) Side view of LixZrNCl crystal structure. Solid lines represent the rhombohedral unit cell. (B) Schematic illustration of the ionic-gating device based on a real optical micrograph picture of a ZrNCl single crystal flake and patterned electrodes. Narrow contacts are prepared for the tunneling spectroscopy measurements. PMMA covers the whole device except for the outer area of the flake and the gate electrode. The electrolyte containing LiClO4 is dropped on the device. Gate voltage VG is applied to the electrolyte, and lithium cations and ClO4 anions move oppositely. Lithium cations intercalate from the sides of the flake. (C) Source-drain current IDS of the device in intercalation operation. During the forward sweep of VG (red), IDS increases steeply, whereas the change of IDS is gradual in the backward scan (blue). VG is swept at a speed of 10 mV/sec. (D) Antisymmetrized transverse resistivity at 150 K for various values of the Li content x. The linear slope is used to determine x. Credit: Science, doi: 10.1126/science.abb9860

In the paired fermion systems, the Bardeen-Cooper-Schrieffer (BCS) superfluidity and Bose-Einstein condensation (BEC) are two extreme limits of the ground state. In a new report in *Science*, Yuji Nakagawa and a team of scientists in applied physics, quantum electronics, emergent matter science and materials research in Japan, reported crossover behavior from the BCS limit to the BEC limit by varying the carrier density in a 2D superconductor electron-doped, layered material ZrNCl containing intercalated layered nitride. The team showed how the ratio of the superconducting transition temperature and Fermi temperature in the low carrier density limit was consistent with the theoretical upper bound expected in the BCS-BEC crossover regime. The results indicated how the gate-doped semiconductor provided an ideal platform for the 2D BCS-BEC crossover without additional complexities as those noted in other solid-state systems.

The BCS-BEC crossover

The phenomenon of fermion pairing, and condensation are fundamental to a variety of systems including neuron stars to superconductors and ultracold atomic gases. Two limiting cases for fermion condensation are described by two distinct theories known as the Bardeen-Cooper-Schrieffer (BCS) theory for which physicist John Bardeen et al. received the Nobel prize in 1972, and the Bose-Einstein condensation (BEC) developed by physicists Satyendra Nath Bose and Albert Einstein in 1924. The BCS theory details the superfluidity in the weak-coupling or high-density limit where individual fermions directly condense to a coherent state of fermion pairs - a type of condensation typically observed in the superconductivity of electrons. The latter often occurred during the strong-coupling, low-density limit. At first, fermion pairs behave as bosons and then they undergo the BEC to the superfluid state in a phenomenon seen in fermionic gases. The two limits are connected continuously through an intermediate regime known as the BCS-BEC crossover.

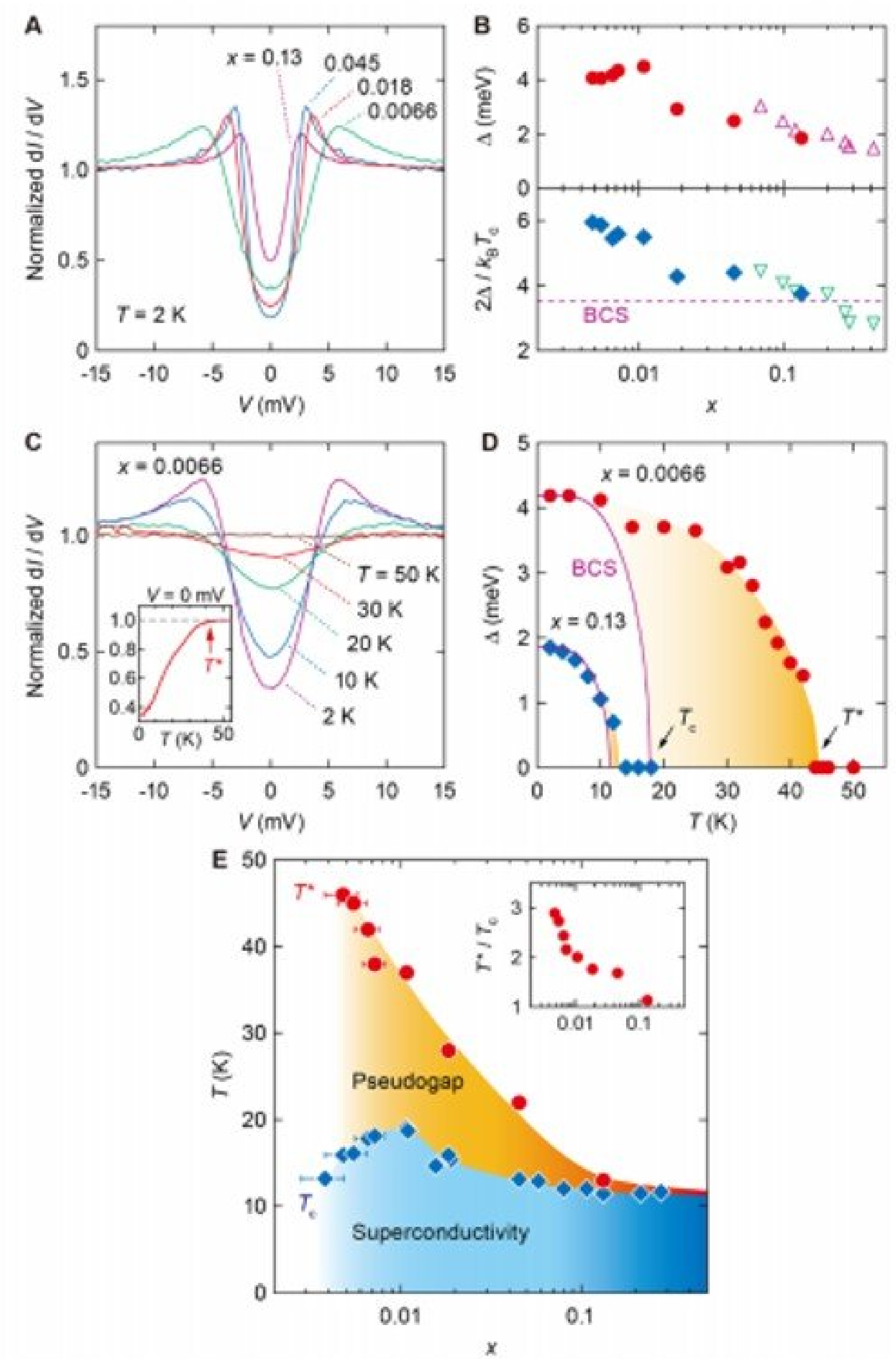


Transport properties of LixZrNCl. (A) Temperature dependence of resistivity at different doping levels. The resistivities at x = 0.080 and 0.13 are multiplied by 5 and 10, respectively. (B) Resistivity normalized at 30 K. Each curve is shifted by 0.5, and gray dashed lines indicate zero lines. (C) Resistivity at x = 0.011 showing the BKT transition. The black line is the fit to the Halperin-Nelson formula. Inset: resistivity plotted on a [d(ln ρ)/dT]–2/3 scale. (D) Out-of-plane upper critical field Hc2 as a function of temperature. Dashed lines are linear extrapolations to 0 K for each doping levels. (E) Doping dependence of Hc2 at 0 K in (D) (top) and in-plane coherence length ξ (bottom). Credit: Science, doi: 10.1126/science.abb9860

Experimental settings

Physicists use ultracold atomic gases and superconductors as favorable experimental settings to observe the BCS-BEC crossover by controlling the coupling strength between the constituent fermions in a quasi-continuous manner. In ultracold atomic gasses the coupling strength can be highly modulated using Feshbach resonances sweeping across the crossover regime from the BEC limit. Researchers can control the carrier density and the coupling strength to enter the crossover regime from the BCS limit within superconductors. In superconductors, the dimensionless coupling strength can be determined using the superconducting gap and Fermi energy measured from the bottom of the conduction band. As the ratio between the superconducting gap and the Fermi energy increased via enhanced pairing interactions or reduced carrier density, the system entered the BCS-BEC crossover regime, accompanied by enhanced ratios of superconducting critical temperature and Fermi temperature. For example, niobium (Nb) and aluminum (Al) reside deeply within the BCS limit, while more exotic superconductors including iron-based semiconductors are located close to the BCS-BEC crossover regime. The coupling strengths are however not high enough to reach the BEC limit beyond the crossover regime due to complex activities such as low carrier density, strong electron correlation effects and magnetic ordering that cloud the phenomena. As a result, physicists remain to clearly demonstrate the BCS-BEC crossover during the study of superconductors. In this work, Nakagawa et al. studied the superconductor Li_xZrNCl – a lithium intercalated layered nitride to understand the phenomena.

Investigating the superconductor

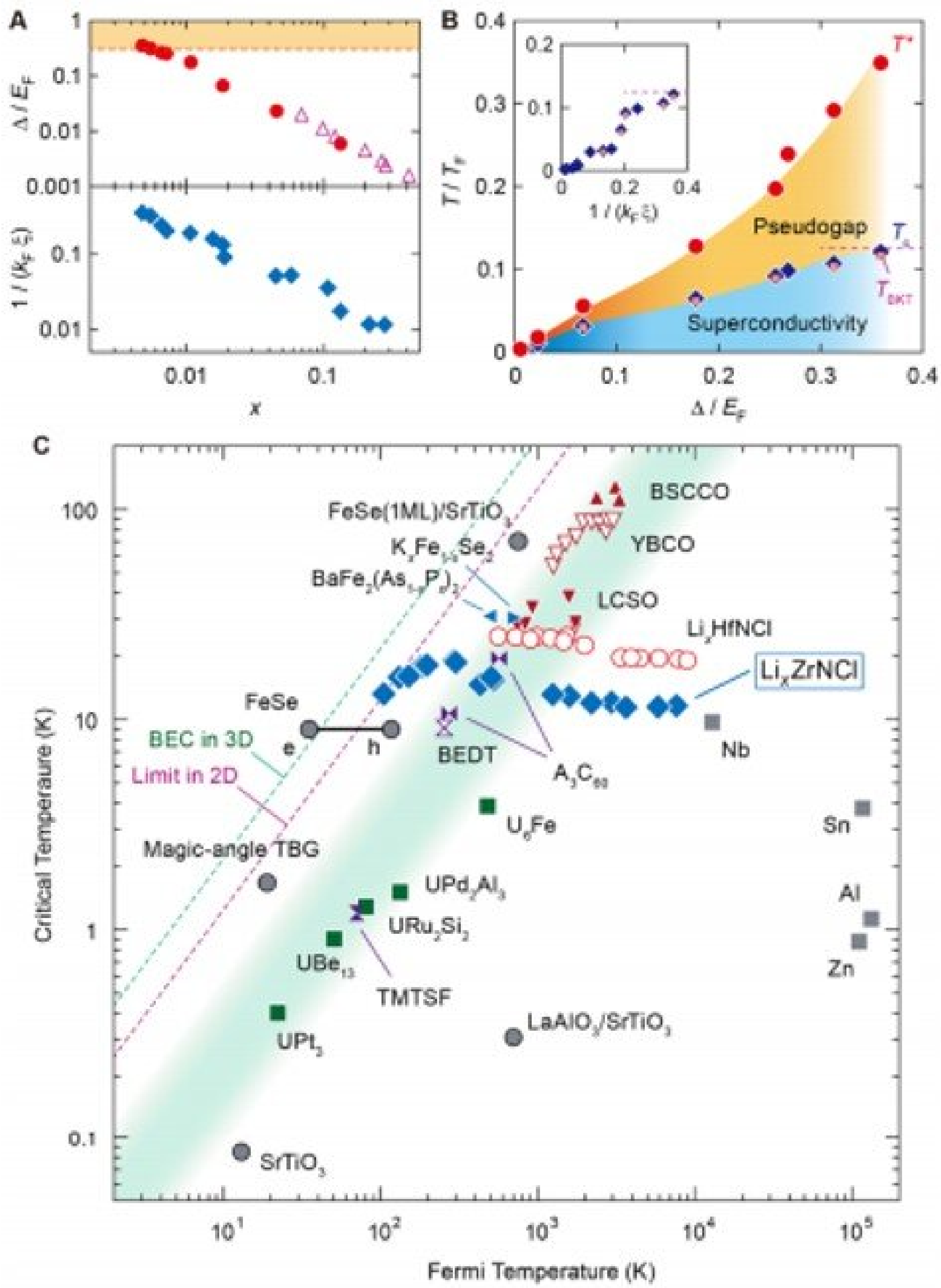


Tunneling spectroscopy of LixZrNCl. (A) Symmetrized and normalized tunneling spectra at 2 K. At each doping level, spectra at 55 K are used for the normalization to remove the biasand x-dependent background after the subtraction of channel resistivity (15, 27). (B) Doping dependence of superconducting gap Δ (top) and its ratio to the critical temperature Tc (bottom). The BCS theory predicts 2Δ/kBTc = 3.52 (dashed line). Open symbols are measured values in polycrystalline samples (29). (C) Tunneling spectra at x = 0.0066 for different temperatures normalized at 55 K without symmetrization. Inset: temperature scan of zero-bias-conductance (ZBC), dI/dV at V = 0. Gap-opening temperature T* is determined by a 1% drop of ZBC. (D) Δ at x = 0.0066 (circles) and 0.13 (diamonds) as a function of temperature. Solid lines indicate the BCStype gap function with Tc determined by the resistive transition. (E) Phase diagram of LixZrNCl. The temperature regime between Tc and T* represents the pseudogap state. The error of carrier density is estimated by measurements in multiple Hall probes. Inset: the ratio between T* and Tc. Credit: Science, doi: 10.1126/science.abb9860.

In the Li_xZrNCl superconductor, lithium supplied electrons to the double honeycomb ZrN layer, which formed a band insulator in the absence of doping. Researchers had previously conducted single-crystal measurements of pristine ZrNCl using ionic gating methods. In recent work, Nakagawa et al. introduced a modified device structure and noted a dimensional crossover from anisotropic three-dimensional (3D) to 2D superconductors by decreasing the carrier density. In this work, the team detailed the superconductivity behavior of Li_xZrNCl in an even lower carrier density regime. The scientists used an ionic-gating device structure and prepared narrow electrodes for tunneling spectroscopy on the channel region between the source and drain electrodes and covered the device with a poly (methyl methacrylate) (PMMA) resist. During gate voltage (V_G) applications, the team traced the intercalation process through the measurement of source-drain current. The resistive transition in the highly doped regime was sharp, while it substantially broadened in the lightly doped regime to represent a dimensional crossover from anisotropic 3D to 2D superconductors.

The dimensional crossover

The 3D to 2D dimensional crossover of the superconductor occurred due to reduced carrier density to therefore form a unique and unexpected phenomenon to enable the crossover. The team credited the feature to the rhombohedral stacking of ZrNCl layers, where the unit contained three layers. Using density functional theory calculations, they confirmed the experimental outcomes. During the cooling process, the scientists performed tunneling spectroscopy, where the decreasing carrier density corresponded to stronger coupling. Nakagawa et al. also discussed the pseudo-gap states in several materials and compared them with the present system. The Li_xZrNCl material offered a simpler test bed since its band-insulator was free from electron correlation effects, magnetic orders and density waves. The team credited the pseudo gap state observed in Li_xZrNCl to pre-formed pair formation during the BCS-BEC crossover phenomenon. They then highlighted a bulk study, where NMR measurements on polycrystalline Li_xZrNCl samples showed a pseudogap state on the high doping side of the superconducting dome.



The BCS-BEC crossover in superconducting LixZrNCl. (A) Doping dependence of the ratio between superconducting gap and Fermi energy (Δ/EF) (top) and the ratio between interparticle distance and coherence length (1/kFξ) (bottom). The orange area represents the BCS-BEC crossover regime (22). Open triangles are measured values from specific heat measurement (29). (B) The phase diagram of the BCS-BEC crossover. Gap opening temperature T*, critical temperature Tc and critical temperature of BKT transition TBKT are normalized by Fermi temperature TF and plotted as functions of Δ/EF with red spheres, dark blue diamonds, and pink squares, respectively. The dashed line represents the theoretically predicted upper bound, TBKT/TF = 0.125. Inset: Tc/TF and TBKT/TF as functions of 1/kFξ. (C) Uemura plot: Critical temperature versus Fermi temperature is plotted for various superconductors. As x is decreased, LixZrNCl departs from the BCS limit, arriving at the crossover region having traversed the shaded area, where most of the unconventional superconductors are located (8). The dashed line denoted as "BEC in 3D" represents the critical temperature in the BEC limit in 3D Fermi gas systems, Tc = 0.218 TF (2). The other dashed line, denoted as "Limit in 2D", corresponds to the general upper limit of TBKT = 0.125 TF in all 2D fermionic systems. Credit: Science, doi: 10.1126/science.abb9860.

Outlook

In this way, Yuji Nakagawa and colleagues showed 2D BCS-BEC crossover by systematically tuning the coupling strength of superconductors in Li_xZrNCl samples. The team realized the 2D BCS-BEC crossover due to the dimensional crossover from the anisotropic 3D to 2D by reducing the carrier density of the samples. They compared this crossover to arrays of 2D clouds of Fermi gases, wherein too dimensionality was affected by the coupling strength. Additional studies on the phenomenon will help advance the understanding of fermion condensation physics.

More information: 1. Nagakawa, Y. et al. Gate-controlled BCS-BEC crossover in a two-dimensional superconductor, *Science*, 10.1126/science.abb9860
2. Greiner M. et al. Emergence of a molecular Bose-Einstein condensate from a Fermi gas. *Nature*, DOI: 10.1038/nature02199.

3. Gaebler J. P. et al. Observation of pseudogap behaviour in a strongly interacting Fermi gas. *Nature Physics*, doi.org/10.1038/nphys1709