



Quantum Griffiths Singularity in 2D Superconductors and Log-Periodic Quantum Oscillations in Ultra-quantum Topological Materials

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Part I: From a new 2D superconducting phase to the observation of quantum Griffiths Singularity

- 1. Physical Review Letters 114, 107003 (2015) (Editors' Suggestion)
- 2. Science 350, 542-545 (2015) (with a perspective paper: Science 350, 509)
- 3. Physical Review B 94, 144517 (2016)
- 4. Nano Letters (2017), DOI: 10.1021/acs.nanolett.7b03026



Part I: Quantum Phase Transition in 2D Crystalline Superconductors——From a new 2D superconducting phase to the observation of quantum Griffiths Singularity

Highly crystalline 2D superconductors

Yu Saito¹, Tsutomu Nojima² and Yoshihiro Iwasa^{1,3}

Abstract | Recent advances in materials fabrication have enabled the manufacturing of ordered 2D electron systems, such as heterogeneous interfaces, atomic layers grown by molecular beam epitaxy, exfoliated thin flakes and field-effect devices. These 2D electron systems are highly crystalline, and some of them, despite their single-layer thickness, exhibit a sheet resistance more than an order of magnitude lower than that of conventional amorphous or granular thin films. In this Review, we explore recent developments in the field of highly crystalline 2D superconductors and highlight the unprecedented physical properties of these systems. In particular, we explore the quantum metallic state (or possible metallic ground state), the quantum Griffiths phase observed in out-of-plane magnetic fields and the superconducting state maintained in anomalously large in-plane magnetic fields. These phenomena are examined in the context of weakened disorder and/or broken spatial inversion symmetry. We conclude with a discussion of how these unconventional properties make highly crystalline 2D systems promising platforms for the exploration of new quantum physics and high-temperature superconductors.

Nature Reviews Materials 2, 16094 (2017)

1. Detection of a new superconducting phase in 2D limit: a twoatom layer Ga film grown on semiconducting GaN(0001) Physical Review Letters 114, 107003 (2015) (Editors' Suggestion)

Acknowledgments

Collaborators:

Tsinghua University



Prof. Qi-Kun Xue Prof. Xucun Ma

Other collaborators: Jun-Ping Peng, Can-Li Song, Ying Xing, Qinghua Zhang, Jiaqi Guan, Zhi Li, Yanfei Zhao, Shuaihua Ji, Lili Wang, Ke He, Xi Chen, Lin Gu, Langsheng Ling, Mingliang Tian, Lian Li, X. C. Xie, Jianping Liu, Hui Yang

Peking University



Dr. Yi Sun





Two-atom layer Ga film grown on GaN(0001) by MBE



The superconductivity of two-atom layer Ga film on GaN(0001) detected by *in situ* STM



Dr. Huimin Zhang



Dr. Wei Li



The superconductivity of two-atom layer Ga film on GaN(0001) detected by *ex situ* transport and magnetization measurements.

Peking University



Dr. Yi Sun



(a) **Evidences for "2D" superconductivity:**

(a)

 10^{0}

10-1

10-2

10-3

 $\alpha =$

0.1

10 K

100

10

1 I(mA) 0.0

3.9

4.2

4.5

T (K)

4.8

5.1

5.4

V.S



BKT-like behavior

12 (

B"B



Therefore, a new superconducting phase in 2D limit is discovered.

Two-atom layer Ga on GaN vs. stable bulk α phase Ga:

Tc: 5.4 K vs. 1.08 k
 Hexagonal (graphene-like) vs. orthorhombic (completely different lattice constant)

A "man-made" or "artificial" 2D crystal or 2D superconductor!

Physical Review Letters 114, 107003 (2015)

Summary



- 1. By *in situ* STM/STS and *ex situ* transport and magnetization measurements, we discover a new 2D superconducting phase with a transition temperature up to 5.4 K in 2 ML (0.552nm) crystalline Ga films grown on wide band-gap semiconductor GaN.
- 2. It is the first conventional crystalline superconductor in 2D limit showing Tc enhancement and ex situ superconductivity. The observed superconductivity in atomic-scale thin films with relatively high T_c and H_c demonstrates the feasibility in developing dissipationless quantum electronic devices based on wide band-gap semiconductors.
- 3. Our result demonstrates a pathway for exploring atomic-scale 2D superconductors by surface and interface engineering in a broad range of metal-semiconductor heterostructures, which benefit from present semiconductor technology and ultrathin film fabrication technique.

Physical Review Letters 114, 107003 (2015) (Editors' Suggestion)

Part I: From a new 2D superconducting phase to the observation of Griffiths Singularity

2. Quantum Griffiths Singularity of Superconductor-Metal Transition in Ga Thin Films

Science 350, 542-545 (2015) (with a perspective paper: <u>Science 350, 509</u>)

Major Collaborators:



Xi Lin



X. C. Xie

Xucun Ma

Ying Xing Huimin Zhang Hai-Long Fu Haiwen Liu

All Authors:

Peking University: Ms Ying Xing, Mr. Hai-Long Fu, Dr. Haiwen Liu, Dr. Yi Sun, Prof. Fa Wang, Prof. Xi Lin, Prof. X. C. Xie, Prof. Jian Wang Tsinghua University and IOP: Dr. Hui-Min Zhang, Mr. Jun-Ping Peng, Prof. Xu-Cun Ma, Prof. Qi-Kun Xue



STM topographic images and STS results of 3 ML Ga film. (A) Constant current topographic STM images (3.0 V, 0.05 nA, 200×200 nm²) of Ga film on GaN substrate. Wheel-rut structure is shown in the inset of (A). (B) Typical tunneling conductance spectra (setpoint: 10 mV, 0.1 nA) at 3.00 K. The red curve is the fitting result by BCS theory, which yields a superconducting gap of Δ = 0.935 meV at 3.00 K and the broading factor Γ = 0.198 meV. (**C**) A series of normalized tunneling conductance spectra (setpoint: 10 mV, 0.1 nA) at various temperatures. Curves are shifted vertically for clarity. (**D**) The superconducting gap as a function of temperature. The blue dots show the measured gap, and the red curve



ng by the BCS function. (E) A ographic image (10 mV, 0.1 70 K. (F) A series of dI/dV along an 8 nm line shown in the spatial distribution of ng gaps and coherence peaks. ited vertically for clarity.



Transport properties of 3 ML Ga film: Si/Ga/GaN



Magnetic field induced superconductor-metal transition: quantum phase transition

Superconductor-insulator or superconductor-metal transition (SIT or SMT)



Magnetoresistance isotherms at ultralow temperatures.







Ying Xing Hai-Long Fu

Variation of the $R_s(T)$ values at various magnetic fields. ΔT is the temperature width of R_s plateau. The inset is an example of how to determine the R_s plateau width. The uncertainty of homemade circuit in this measurement is 0.015%.

Determination of QPT from measurement at nonzero temperature (Finite Size Scaling)

Phenomenological scaling law:

$$\frac{R_{\rm S}}{R_{\rm X}} = F\left(\frac{B - B_{\rm X}}{T^{1/z\nu}}\right)$$

$$R_{\rm S}(\delta,t) = R_{\rm X}F(\delta t)$$

Where F is an arbitrary function with

F(0) = 1 (at critical point)



Exponent *zv* obtained by scaling

(A)(C)(E)(G) The sheet resistance as a function of magnetic field close to the S-M transition boundary at different temperatures. (B)(D)(F)(H) Normalized $R_{\rm s}$ as a function of the scaling variable |B- $B_{\chi}|(T/T_{0})^{-1/2v}$. The bottom left insets are the temperature behaviours of the scaling parameter $t (t = (T/T_0)^{-1}$ 1/zv



The activated quantum scaling behavior: exponent *zv* as a function of magnetic field *B*.



Novel discovery: previous studies show the exponent is a constant.

<u>Ying Xing et al., Science 350, 542-545 (2015)</u></u>



Transport measurements and analysis on another 3 ML Ga film in different dilution refrigerator further confirm our observation.

More than 40 years ago, Robert B. Griffiths predicted that phase transitions can be dramatically changed by disorder effect and in particular the dynamical critical exponent can diverge. In the last 40 years, this theory has been applied to quantum phase transitions and developed into the theory of "quantum Griffiths singularity". However, the major signature of the theory, the divergence of dynamical critical exponent, is very difficult to observe in experiments.

The discovery of quantum Griffiths singularity of superconductor-metal transition in Gathin filmsScience 350, 542-545 (2015)



Science The World's Leading Journal of Original Scientific Research, Global News, and Commentary Science Home Current Issue Science Express Science Products My Science About the Journal Previous Issues Home > Science Magazine > 30 October 2015 > Markovic, 350 (6260): 509 Science 30 October 2015: < Prev | Table of Contents | Next > Article Views Vol. 350 no. 6260 p. 509 8 Read Full Text to Comment (0) DOI: 10.1126/science.aad4136 Summary Full Text PERSPECTIVE

> SUPERCONDUCTIVITY Randomness rules

Nina Markovic

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Phase transitions are perfect examples of physica predictions. Different types of phase transitions, ra treated within the same theoretical framework. Thi of the system, such as resistivity, heat capacity, or predictions can be strongly affected by random dis present in all real physical systems. In some syste no impurities. Such rare regions may be in a phas dramatically alter the nature of the transition, caus in the vicinity of the transition. These infinities are variety of systems, but they are not easily observe report the first experimental evidence of a Griffiths dimensional (2D) superconducting system.

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The editors suggest the following Related Resource

In Science Magazine

REPORT

Quantum Griffiths singularity of superconduct Ying Xing, Hui-Min Zhang, Hai-Long Fu, Haiwen I Qi-Kun Xue, Jian Wang, and X. C. Xie Science 30 October 2015: 542-545.Published onl »Abstract »Full Text »Full Text (PDF) » St

A perspective paper: <u>Science 350, 509</u>

The observation of the quantum Griffiths singularity in a 2D superconductor offers a new perspective on the previous studies of the superconductor-insulator and metal-insulator transitions. Although these transitions have been studied for decades, there are lingering unexplained inconsistencies between the critical exponents found in different physical systems, lack of universal behavior, and substantial uncertainty in locating the quantum regime. Some of these issues can now be possibly attributed to the effects of quenched disorder, thus recovering the elegance of being able to treat different systems within the same theoretical framework.

Discovery of quantum Griffiths singularity in 2D superconductors: potential universal behavior for superconductor-metal transition

S. Observation of quantum Griffiths singularity at superconducting LaAlO3/SrTiO3(110) interface Physical Review B 94, 144517 (2016)

Major Collaborators:



Jiacai Nie Shengchun Shen Beijing Normal University



Xi Lin Ying Xing Pengjie Wang Peking University

All authors:

<u>Shengchun Shen</u>, <u>Ying Xing</u>, <u>Pengjie Wang</u>, <u>Haiwen Liu</u>, <u>Hai-Long Fu</u>, <u>Yangwei Zhang</u>, <u>Lin He</u>, <u>X. C. Xie</u>, <u>Xi Lin</u>, <u>Jiacai Nie</u>, <u>Jian Wang</u>

Superconducting LaAlO3/SrTiO3(110) Interface





Quantum Griffiths singularity: Universal behavior for superconductormetal transition in 2D superconductors?



Answer: Quantum Griffiths singularity is detected at superconducting oxides interface.

Physical Review B 94, 144517 (2016)

The Gating effect on quantum Griffiths singularity:



FIG. 4. The quantum Griffiths singularity for $V_G = 20$ V. (a) The R(T) at zero magnetic field with $T_c^{zero}=0.109$ K, and the inset shows the definition of T_c^{onset} , with a value of 0.696 K. (b) The isotherms R(B) measured at different T. Zoom-in view of cross region is shown in the inset. (c) The isotherms R(B) measured at different T ranged from 0.020 K to 0.300 K. The inset provides the crossing points B_c as a function of T, which are determined from the cross point of every adjacent two R(B) curves. (d) The B dependence of zv values reveals the activated scaling behavior.

Hysteretic behavior at LAO/STO(110):



ever predicted by Gang Chen PRL 2013.

For $V_G = 60 V$

It is reminiscent of coexistence of superconductivity and ferromagnetism at polarized LAO/STO(001) interface.



ARTICLE

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DOI: 10.1038/ncomms1959

Evidence for charge-vortex duality at the LaAlO₃/SrTiO₃ interface

M.M. Mehta¹, D.A. Dikin¹, C.W. Bark², S. Ryu², C.M. Folkman², C.B. Eom² & V. Chandrasekhar¹

Figure 1 | MR in the superconducting regime. (a) Parallel field MR as a function of different rates in the superconducting regime at $V_g = 80$ V. Data for only one field sweep direction are shown for clarity. Arrow indicates the direction of field sweep. The inset shows the MR for forward and backward field sweep directions at a sweep rate of 300μ T s⁻¹. (b) Schematic of the system at different parallel field values. The top layer is the ferromagnet, the superconductor is shown through a gradient as extending some distance into the STO. In this simplified picture, magnetization reversal occurs by means of domain wall propagation in the ferromagnet. The perpendicular component of the field due to the domain wall induces vortices in the superconductor. (c) Perpendicular field MR for fields swept from negative to positive values for different field sweep rates for $V_g = 80$ V. (d) Schematic of the magnetization state of the system at different perpendicular fields. Owing to the shape anisotropy of the system, the majority of the moments lie in plane, but the external magnetic field orients the direction of the perpendicular component of the magnetization of the domain wall. It should be emphasized that the magnetization configuration in the real system during reversal is definitely far more complicated, but would still give rise to a perpendicular component of the magnetic field. All data were taken at T = 50 mK.

Summary

- 1. Quantum Griffiths Singularity has been observed at another 2D superconducting system: superconducting LaAlO3/SrTiO3(110) Interface;
- 2. Gating capability, potential coexistence of SC and FM may offer more modulation methods for detecting and analyzing quantum Griffiths singularity at 2D superconductors;
- 3. Quantum Griffiths singularity could be a universal behavior for superconductor-metal transitions.

Hints of quantum Griffiths singularity in other 2D superconductors from other groups





Multiple quantum criticality in a two-dimensional superconductor

J. Biscaras¹, N. Bergeal¹, S. Hurand¹, C. Feuillet-Palma¹, A. Rastogi², R. C. Budhani^{2,3}, M. Grilli⁴, S. Caprara⁴ and J. Lesueur^{1*}



Two-stage magnetic-field-tuned superconductorinsulator transition in underdoped La_{2-x}Sr_xCuO₄

Xiaoyan Shi^{1†}, Ping V. Lin¹, T. Sasagawa², V. Dobrosavljević¹ and Dragana Popović^{1*}

Further confirmation of quantum Griffiths singularity in superconductors from other groups

Iwasa group at Tokyo University found quantum Griffiths singularity like what we observed in Ga films by liquid gating measurement .

APS March Meeting 2016 Volume 61, Number 2 Monday–Friday, March 14–18, 2016; Baltimore, Maryland

Session S15: 2D Materials: Superconductivity and Correlations II 11:15 AM-2:15 PM, Thursday, March 17, 2016 Room: 314

Sponsoring Unit: DMP Chair: James Eckstein, UIUC

Abstract ID: BAPS.2016.MAR.S15.3

Abstract: S15.00003 : Griffiths singularity of quantum phase transition in ion-gated ZrNCI 12:03 PM-12:15 PM

Preview Abstract

MathJax On | Off + Abstract +

Authors:

Yu Saito (The University of Tokyo)

Tsutomu Nojima (Tohoku University)

Yoshihiro Iwasa (The University of Tokyo)

One of the most important topics in 2D superconductors *Nature Reviews Materials 2, 16094 (2017)*

Recent technological advances of thin films fabrication, especially mechanical exfoliation, led to discoveries of less-disordered highly-crystalline two-dimensional (2D) superconductors; atomically thin NbSe2 and ion-gated 2D materials, which show intrinsic properties of 2D superconductors with minimal disorder; for example, metallic ground state [1,2], and unconventional 2D Ising superconductivity due to pure spin-valley locking effect [3-5]. In this talk, we focus on magnetotransport properties of an ionic-liquid gated ZrNCI, which exhibited Griffiths singularity-like behavior in superconductor-metal-insulator transition induced by magnetic fields at low carrier concentrations. The overall behavior is quite similar to the recent results of superconducting Ga thin films, in which quantum Griffiths singularity was observed in vortex-glass state [6]. We will discuss the relationship between Griffiths singularity and quantum tunneling or flux flow of vortices phase (vortex liquid) in our system. [1] Y. Saito et al. Science 350, 409 (2015). [2] A. W. Tsen et al. arXiv 1507.08639 [3] Y. Saito et al. Nature Phys. doi: 10.1038/nphys3580. (arXiv:1506.04146). [4] X. Xi et al. arXiv:1507.08731. [5] J. M. Lu et al. arXiv:1506.07620. [6] Y. Xing et al. Science 350, 542 (2015).

Highly crystalline 2D superconductors

Yu Saito¹, Tsutomu Nojima² and Yoshihiro Iwasa^{1,3}

Abstract | Recent advances in materials fabrication have enabled the manufacturing of ordered 2D electron systems, such as heterogeneous interfaces, atomic layers grown by molecular beam epitaxy, exfoliated thin flakes and field-effect devices. These 2D electron systems are highly crystalline, and some of them, despite their single-layer thickness, exhibit a sheet resistance more than an order of magnitude lower than that of conventional amorphous or granular thin films. In this Review, we explore recent developments in the field of highly crystalline 2D superconductors and highlight the unprecedented physical properties of these systems. In particular, we explore the quantum metallic state (or possible metallic ground state), the quantum Griffiths phase observed in out-of-plane magnetic fields and the superconducting state maintained in anomalously large in-plane magnetic fields. These phenomena are examined in the context of weakened disorder and/or broken spatial inversion symmetry. We conclude with a discussion of how these unconventional properties make highly crystalline 2D systems promising platforms for the exploration of new quantum physics and high-temperature superconductors.

Nature Reviews Materials 2, 16094 (2017)

Three most important topics in 2D superconductors: Quantum metallic state, Quantum Griffiths Singularity, superconducting state maintained in huge in-plane magnetic field

Can them co-exist in one 2D superconductor?

4. Coexistence of Ising Superconductivity and Quantum Phase Transition in Macro-Size Monolayer NbSe₂

In collaboration with Prof. Shuaihua Ji etc



Nano Letters (2017), DOI: 10.1021/acs.nanolett.7b03026

Layered transition metal dichalcogenides NbSe₂



PRB 16, 801 (1977)

Nat. Phys. 12,92(2016)

J. Phys, Chem. Solids 26, 1029 (1965)



crossing bands from three (for bulk) to one

in the single-layer limit.

- Van der Waals coupling
- T_{CDW}~ 33 K (bulk);
- T_{sc}~7.2 K (bulk);
- Anisotropic s-wave superconductor (bulk).

Layered transition metal dichalcogenides NbSe₂



The superconductivity weakens in the monolayer limit.

The CDW order is strongly enhanced in monolayer limit.

Nat. Phys. 12,139(2016)

Nat. Nanotech. 10,765(2015)



STM image of monolayer $NbSe_2$ film grown on epitaxial bilayer graphene (BLG) on 6H-SiC(0001) substrate



 $18~\mathrm{nm} imes 18~\mathrm{nm}$

1.9 μm \times 1.9 μm

Substrate: bi-layer graphene with N-doped 6H-SiC(0001) substrates

Sample: MBE method, Shuaihua Ji (TSU)



Transport results in 1 ML NbSe₂



Zero resistance was not observed in previous monolayer flakes.



- Van der Waals interaction between SiC, BLG, and NbSe₂;
- The growth rate of 2.5 MLs per hour;
- Se capping layer with the thickness of 20 nm.

Nano Letters (2017), DOI: 10.1021/acs.nanolett.7b03026



Transport results in 1 ML NbSe₂



Parallel field



H-T phase diagram for 1 ML NbSe₂ **Ising Superconductivity**



T/Tc down to 0.13

Fulde-Ferrell-Larkin-Ovchinnikov state ? B_{c2}/B_{p} : 1.5~2.5 times

Ising superconductivity in gated MoS₂





- **B**_{eff} ~ from intrinsic spin-orbit coupling
- Spins of the pairing electrons are strongly pinned by B_{eff} in a direction orthogonal to an external magnetic field;
- The superconductivity can be strongly protective.



B_{eff}: Zeeman-like effective magnetic field



Spin-Orbit locking are dominated by the K (K') pockets

Science 350,1353(2015)

Magnetic field induced superconductor-metal transition



Ultralow-*T* transport: Xi Lin Group(ICQM)

Quantum Griffiths Singularity



Conclusion for Monolayer NbSe₂

- The macro-size atomically flat monolayer NbSe₂ films were successfully grown on bilayer graphene/SiC by MBE method, T_c^{onset} is above 6 K and zero resistance Tc around 2.4 K, higher than previous reports on NbSe₂ monolayers.
- In non-centrosymmetric monolayer NbSe₂, direct high magnetic field and low temperature measurement show $B_{c//}(0)$ is at least 5.09 times of the Pauli paramagnetic limit, consistent with Zeeman protected Ising superconducting mechanism.
- The magnetic field driven SMT is detected in monolayer $NbSe_2$ and the signature of the quantum Griffiths singularity is observed by ultralow temperature measurements.
- Monolayer NbSe₂ could be a new platform to create topological superconductivity and Majorana Fermions.

Nano Letters (2017), DOI: 10.1021/acs.nanolett.7b03026

Highly crystalline 2D superconductors

Yu Saito¹, Tsutomu Nojima² and Yoshihiro Iwasa^{1,3}

Abstract | Recent advances in materials fabrication have enabled the manufacturing of ordered 2D electron systems, such as heterogeneous interfaces, atomic layers grown by molecular beam epitaxy, exfoliated thin flakes and field-effect devices. These 2D electron systems are highly crystalline, and some of them, despite their single-layer thickness, exhibit a sheet resistance more than an order of magnitude lower than that of conventional amorphous or granular thin films. In this Review, we explore recent developments in the field of highly crystalline 2D superconductors and highlight the unprecedented physical properties of these systems. In particular, we explore the quantum metallic state (or possible metallic ground state), the quantum Griffiths phase observed in out-of-plane magnetic fields and the superconducting state maintained in anomalously large in-plane magnetic fields. These phenomena are examined in the context of weakened disorder and/or broken spatial inversion symmetry. We conclude with a discussion of how these unconventional properties make highly crystalline 2D systems promising platforms for the exploration of new quantum physics and high-temperature superconductors.

Nature Reviews Materials 2, 16094 (2017)

Quantum metal?

Outline

- Part I. Quantum Griffiths singularity for superconductor-metal transition : new quantum phase transition found in 2D superconductors
 - 1. Detection of a New Superconducting Phase in 2D limit
 - **Physical Review Letters 114, 107003 (2015) (Editors' Suggestion)**
 - 2. Quantum Griffiths Singularity of Superconductor-Metal Transition in Ga Thin Films
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 - *Physical Review B* 94, 144517 (2016)
 - 4. Coexistence of Quantum Griffiths Singularity and Ising superconductivity
 - Nano Letters (2017), DOI: 10.1021/acs.nanolett.7b03026
 - Perspective: to detect quantum Griffiths singularity for superconductormetal transition in disorder-controlled 2D superconducting systems and high Tc superconductors

Summary for 2D Crystalline Superconductors

Perspective of next generation of superconductors: superconductivity at 2D crystalline materials

1. Superconductivity at 2D limit;

e.g. Monolayer Pb and In on Si substrates

Prof. Qi-Kun Xue at Tsinghua University: Nature Physics 6, 104 (2010) **Prof. Hasegawa at University of Tokyo :** PRL 110, 237001 (2013) **Tomonobu Nakayama group at National Institute for Materials Science:** PRL107,207001 (2011)

2. Interface and high Tc superconductors;

e.g. Crystalline FeSe films on STO by interface engineering Iron-based compound revives search for room-temperature superconductors. 474 | NATURE | VOL 501 | 26 SEPTEMBER 2013

3. Quantum phase and phase transitions;

e.g. Quantum Griffiths Singularity in Crystalline Ga films, LAO/STO(112), NbSe₂ films, ZrNCl etc Disorder is important.

4. Potential topological superconductors.

Updated: Discovery of unconventional superconductivity on topological semimetals induced by hard point contact

- 1. Physical Review X 5, 031037 (2015)
- 2. Nature Materials 15, 38-42 (2016)
- 3. Science Bulletin 62, 425 (2017)
- 4. npj Quantum Materials 1, 16005 (2016)





Superconductivity on Dirac Semimetal Cd3As2 A new method to detect topological superconductivity! Further theoretical investigations are highly desired.

Superconductivity on Weyl Semimetal TaAs



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THANK YOU !