

A metallic room-temperature d-wave altermagnet

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[Ques] Altermagnetism is a recently discovered unconventional magnetic phase that is characterized by time-reversal symmetry breaking and spin-split band structures in materials with zero net magnetization. Recently, spin-polarized band structures and a vanishing net magnetization were observed in semiconductors MnTe and MnTe₂, confirming this unconventional magnetic order. Metallic altermagnets offer advantages for exploring physical phenomena related to low-energy quasiparticle excitations and for applications in spintronics because the finite electrical conductivity of metals allows direct manipulation of the spin current through the electric field.

[关键问题] 如何在零净磁化的金属体系中实现室温稳定的 d 波交替磁性，并验证其自旋劈裂与自旋动量锁定的本征特性。

[Sum] 研究者通过合成高质量 KV₃Se₂O 单晶，并结合核磁共振（NMR）、角分辨光电子能谱（ARPES/自旋 ARPES）和第一性原理计算，首次确凿地证明该材料是一种具有 d 波对称性自旋劈裂的金属性室温交替磁体。

[亮点 1] 该工作首次在金属材料中实现了室温稳定的交替磁序。尽管整体磁化为零，但其能带呈现清晰自旋劈裂。多种实验手段相互验证，确认其磁有序温度远高于室温，展示了交替磁性在实际器件应用中的巨大潜力。

[亮点 2] 研究揭示 KV₃Se₂O 具有 d 波对称性的自旋动量锁定与高达 1.6 eV 的巨大自旋劈裂，费米面展现强各向异性的自旋极化特征。该性质意味着材料可支持高度自旋极化电流，为设计高效率的自旋流源和低功耗自旋电子学器件提供新方向。

[思考] 未来可探索该 d 波交替磁体是否能在与非常规超导体或拓扑材料的界面中产生新奇的量子态，如拓扑超导、马约拉纳零能模等。此外，也可研究交替磁序与自

旋密度波之间的耦合机制，理解其竞争或协同关系是否可诱导新的量子相，如非常规配对超导或激子凝聚。

[拓展阅读 1] 什么是 d 波交替磁性？

[拓展阅读 1] 交替磁性是一类在整体上无净磁化，但能带出现自旋劈裂的新型磁序，与传统铁磁或反铁磁均不同。其关键在于自旋极化随动量呈现特定对称性分布，而 d 波交替磁性则意味着自旋劈裂随晶体动量方向呈现 d 波型（如 $\cos 2\theta$ ）变化。KV₃Se₂O 中，自旋极化不仅存在，而且具有显著的角向各向异性，这不同于铁磁的各向同性，也区别于依赖自旋轨道耦合的非磁性体系自旋纹理。d 波交替磁体的意义在于，它在没有净磁场的情况下产生巨大自旋极化，极利于实现自旋注入、高自旋流效率及低能耗操作，是新兴自旋电子学的理想材料平台。

[拓展阅读 2] 为什么零净磁化仍能产生自旋劈裂？

[拓展阅读 2] 传统观念认为能带自旋劈裂需依赖铁磁序或强自旋轨道耦合。但交替磁体打破了这一限制：虽然其总磁化为零，但空间中不同动量点的自旋极化呈“反向成对”分布，从而在能带层面形成可观测的自旋劈裂。KV₃Se₂O 的晶格结构、磁轨道排列与交换作用使其在金属性体系中稳定形成这种动量空间的自旋“交错”分布，这种新型对称性破缺方式为材料提供了零磁化下的巨大自旋流来源。理解这类机制有助于推动低磁噪声、高频响应以及可集成化的交替磁材料设计。

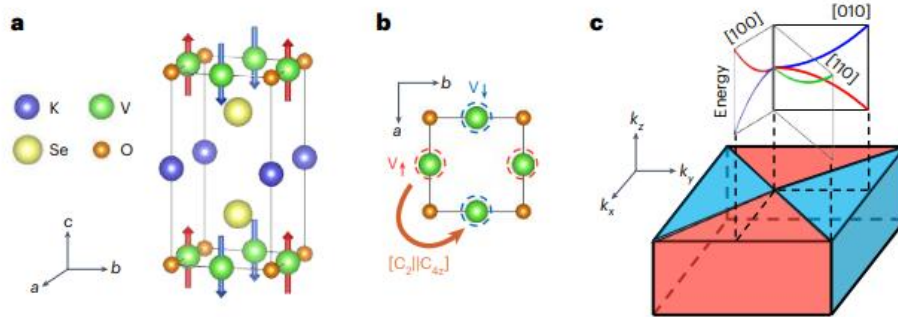
[introduction] Altermagnetism describes a new type of long-range magnetic order beyond conventional ferromagnetism and antiferromagnetism^{1,2,3,4,5}. Historically, altermagnets were classified as antiferromagnets, as both exhibit zero net magnetization. Recent theoretical studies have found that non-relativistic spin-splitting can exist in certain magnets with zero net magnetization, even without spin-orbit coupling (SOC), leading to the concept of altermagnetism^{1,2,3,4,5,6,7,8,9,10,11}. In the language of symmetry, an enhanced symmetry group called a spin space group^{12,13,14,15,16,17,18,19} classifies magnetic order by the allowed symmetry operations connecting opposite-spin sublattices. In conventional antiferromagnets, opposite-spin sublattices can be connected by inversion or translation operation, whereas ferromagnets have only one spin sublattice, which does not require any symmetry operation. By contrast, altermagnets essentially require rotation or a mirror operation to connect opposite-spin sublattices^{1,2,6,7}, resulting in momentum-

dependent spin-splitting in electronic band structures. The same symmetry operation connects opposite-spin sub-bands in reciprocal space^{1,2,6}. Unlike the spin-splitting induced by the Rashba–Dresselhaus interaction^{20,21} in non-magnets lacking inversion symmetry and the Zeeman interaction in ferromagnets, the non-relativistic symmetry operation acts as a crucial aspect in the momentum-dependent spin-splitting in altermagnets^{1,2}. The combination of spin-split band structures and zero net magnetization in altermagnets has immense application potential, ranging from spintronics to quantum information processing^{22,23,24,25,26}.

Although symmetry analyses of spin structures have predicted numerous altermagnet candidates^{1,2}, only a few have been experimentally verified. Recent spin- and angle-resolved photoemission spectroscopy (SARPES) experiments have revealed momentum-dependent spin-splitting in the band structures of MnTe (ref. 27) and MnTe₂ (ref. 28), which is solid evidence for the existence of the altermagnetic phase. These two materials are semiconductors with a global bandgap at the Fermi level (E_F)^{12,13,29,30}, which limits their potential applications in spintronics. The metallic candidates CrSb and RuO₂ have attracted widespread attention^{31,32,33,34,35,36,37,38,39,40,41}. Owing to the C₃ symmetry of the spin sublattices in CrSb and MnTe, opposite-spin channels have the same group velocities in all directions, leading to an unpolarized current. Despite the observation of the anomalous Hall effect and spin current and torque in RuO₂ (refs. 37,38,39,40), there remains controversy about whether it is non-magnetic or magnetic^{41,42,43}.

In this work, we identified a metallic altermagnet KV₂Se₂O with a magnetic ordering temperature well above room temperature. The momentum-dependent spin-splitting across E_F reached 1.6 eV, one of the highest values reported for altermagnets. This altermagnetic order is characterized by d-wave exchange splitting, which is a magnetic counterpart to unconventional d-wave superconductivity. The extreme anisotropy of the spin-polarized Fermi surfaces (FSs) with C₂ symmetry results in notable differences in the group velocities of opposite-spin channels. This enables the generation of a highly polarized electric current and a giant spin current, which are crucial for achieving high-performance spintronic devices. Another spin-density-wave (SDW) order arises from perfect FS nesting below 100 K, which is a unique opportunity for studying the interplay

of many-body effects coupled with unconventional magnetism.



Multiferroic nematic d-wave altermagnetism driven by orbital-order on the honeycomb lattice

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[Ques] Altermagnets provide promising platforms for unconventional magnetism, whose controllability could enable a new generation of spintronic devices. While various bulk altermagnets have been discovered, altermagnetism in two-dimensional van der Waals materials remains elusive.

[关键问题] 如何在二维范德华材料中利用轨道序实现无需晶体畸变的 d 波交替磁性，并与铁电极化形成内在耦合。

[Sum] 本文通过第一性原理计算、模型哈密顿量与对称性分析，首次理论预测了单层 VCl_3 是一种由轨道序驱动的多铁性 d 波交替磁体，揭示了其独特的电子机制、自旋劈裂与铁电极化之间的强耦合。

[亮点 1] 研究提出了由电子相互作用导致的轨道有序可直接驱动交替磁性与铁电性，无需传统结构畸变或自旋轨道耦合。该机制突破了现有多铁性交替磁体的设计框架，为在强关联、低维范德华材料中构筑新型电控磁性系统提供了根本性新途径。

[亮点 2] 工作预测在蜂窝晶格的 VCl_3 单层中形成具有 C_2 对称性的 d 波向列交替磁序，其自旋劈裂在 Γ -M 方向可达约 0.2 eV。材料同时具有节点线结构与谷自由度，使其在无自旋轨道耦合下仍展现谷自旋电子学潜力，适用于低功耗可操控的自旋电-谷电子器件。

[思考] 未来可探索是否能利用电场同时翻转轨道序与铁电极化，从而同步改变自旋劈裂方向，实现“轨道—电—自旋”的联动调控。进一步，在 VCl_3 与金属交替磁体（如 $\text{KV}_3\text{Se}_2\text{O}$ ）的异质结构中，两种不同来源的 d 波交替磁性是否会产生增强、自发错配或新型界面态，值得系统研究。

[拓展阅读 1] 什么是轨道序驱动的交流磁性？

[拓展阅读 1] 在许多磁性材料中，自旋有序与晶格结构密切相关，而轨道序往往只是辅助角色。然而在单层 VCl_3 中，电子间的库仑排斥促使 d 轨道出现特定的占据不均匀分布，从而形成轨道序。轨道序反过来重塑了交换作用路径，使得自旋在动量空间按照 d 波方式发生交错排列，最终导致交替磁性产生。与传统磁性的区别在于：其无需晶体结构畸变，也不依赖强自旋轨道耦合，而是纯电子关联驱动的结果。这种机制拓展了交替磁性的物理来源，为二维强关联材料的磁性工程提供全新视角。

[拓展阅读 2] 多铁性与交替磁性的耦合为何重要？

[拓展阅读 2] 多铁性材料同时具有磁极化与电极化，可实现电控磁性或磁控电性，是低能耗信息器件的核心材料平台。在 VCl_3 中，轨道序不仅导致 d 波交替磁性，也打破空间反演对称性，从而自发产生面内铁电极化。由于两者具有共同的电子起源，它们之间的耦合强度远高于传统多铁材料。这意味着通过电场可直接操控轨道序，进而影响自旋劈裂方向，实现非易失性多态存储、可编程自旋极化甚至多比特逻辑构型，极具应用潜力。

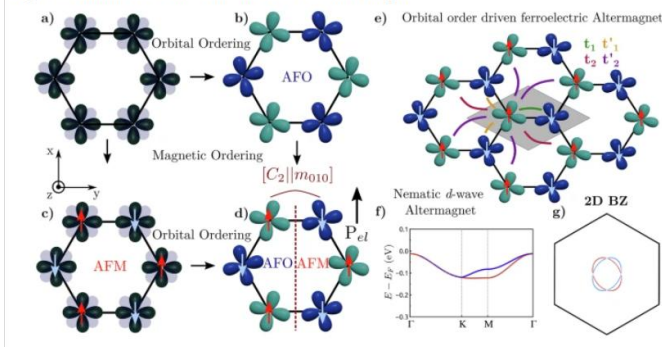
[introduction] Multiferroic materials, defined by the coexistence and the possible coupling of multiple ferroic orders¹, offer a unique platform for functional properties, enabling the control of multiple degrees of freedom (charge, spin, lattice) through external factors. This potential is further enhanced by the remarkable tunability of two-dimensional (2D) van der Waals materials through strain, stacking, and twisting, enabling the creation of exotic phenomena such as unconventional superconductivity^{2,3,4}, heavy fermion Kondo systems^{5,6}, quantum spin liquid states^{7,8,9,10}, and 2D multiferroicity^{11,12,13,14,15,16,17}.

Altermagnetism has emerged as a fascinating new class of antiferromagnetic order, characterized by non-relativistic spin-splitting of electronic bands in momentum space, even in materials composed of light elements^{18,19,20,21,22,23}, while maintaining zero net magnetization. This unique property combines features of both ferromagnetism and antiferromagnetism in a single material, offering unique opportunities for spintronics and quantum materials research. Clearly, the possibility to manipulate the spin properties of an altermagnet with electricity is particularly attractive because electric fields are much easier to manipulate and integrate into modern electronic devices than magnetic fields. Electrical tuning is potentially also faster (subnanosecond) and could use less energy, two

crucial properties for the development of high-speed, low-power spintronic devices. Recently, a novel ferroelectric switchable altermagnetism (FSA) has been theoretically proposed, where the reversal of ferroelectric polarization is coupled to the switching of altermagnetic (AM) spin splitting^{18,24,25,26}.

Altermagnetism arises in fully compensated magnets where the joint parity and time-reversal, with τ being the fractional lattice translation, are broken. In such systems, the distinct spin sublattices are linked by rotational or mirror symmetry operations. Since the AM spin splitting is a non-relativistic property that manifests without the need for spin-orbit coupling (SOC), the symmetry-theoretical framework should adopt spin space group (SSG)^{27,28,29} although alternative theoretical approaches have been also proposed^{30,31}. An interesting case arises when the symmetry breaking is driven not by a structural distortion generating an effective crystal field on the magnetic ions, but rather by electronic degrees of freedom. This phenomenon occurs in orbitally ordered systems, where an electronic instability leads to the formation of inequivalent sublattices with distinct orbital occupancy patterns^{17,32,33,34}. The emergence of the orbitally-ordered (OO) state can induce the AM phase via electronic symmetry breaking, unveiling a hidden AM phase that remains undetectable through conventional spin-space group analysis³⁵.

Fig. 1: Orbital-order driven ferroelectric altermagnet.



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