

一种新磁性的发现——交错磁性

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几千年来, 磁铁的神秘力量一直吸引着人们的眼光, 中华民族在其认识历程中留下了浓墨重彩的一笔。早在五千年前的先秦时期, 勤劳的中国人民在采矿过程中发现了天然磁铁矿。《山海经》将其记作“慈石”, 并形象地把磁石吸铁比喻为慈母爱子, 这表明古人对磁铁性能的认识已相当深入。除了最早发现磁石外, 中国人民还在应用磁石方面走在了世界前列。据《三辅黄图》记载, 荆轲刺秦失败后, 秦王嬴政下令用磁石建造阿房宫大门, 以阻止刺客携带铁质暗器进入宫殿, 这可以说是世界上最早的安检门。中国还是世界公认发明指南针的国家。战国时期的史书《鬼谷子》中就提到了使用“司南”来辨别方位的办法。古代中国人民将天然磁石磨成勺状, 放置在光滑平面上, 在地球磁场的作用下, 其勺柄端指向南方, 这就是现代指南针的雏形。到了北宋时期, 勇敢的中国航海家们开始利用指南针导航, 从而结束了依赖“星相导航”的历史, 这对全球航海技术和社会发展都产生了深远影响。此后, 随着指南针经由阿拉伯人传入欧洲, 大航海时代开启, 世界文明出现了跨时代转折。

在自然界尺度的另一端——微观世界中, 基本粒子的“自旋”属性同样令人着迷。一个世纪前, 斯特恩-盖拉赫实验证明了电子不仅带有电荷, 还有自旋这一内禀属性, 揭开了探索电子自旋奥秘的序幕。自那时起, 人们对磁性和自旋的探索从未停止过, 相关研究也屡次获得诺贝尔奖的认可。其中, 法国科学家路易·奈尔(Louis Néel)因对反铁磁的预测和开创性研究获得了1970年的诺贝尔物理学奖。反铁磁性(antiferromagnetism)的发现补充了之前已知的几种磁性类型——抗磁性(diamagnetism)、顺磁性(paramagnetism)和铁磁性(ferromagnetism), 共同构成当今我们对磁性材料理解的基础框架。上述对磁性材料的分类是建立在相邻磁矩共线排列的基础上的。然而, 在过去二十年里, 科学家也开始广泛关注相邻磁矩非共线排列的材料体系, 例如螺旋磁性(spin spiral)、磁斯格明子(skyrmion)、自旋冰(spin ice)以及笼目磁体(kagome magnet)等。尽管这类复杂结构尚未得到充分解析(本文暂不展开讨论), 但相关研究拓展了我们对磁性世界的认知。



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自从奈尔先生发现反铁磁性以来, 人们普遍认为铁磁(包括亚铁磁)和反铁磁是共线磁体的主要形式。在铁磁体中, 相邻磁矩呈平行排列, 导致在实空间具有宏观磁化, 而在倒易空间则表现为能带自旋劈裂(图1(a))。与此相反, 反铁磁体的相邻磁矩呈反平行排列, 导致在实空间磁矩相互抵消, 而倒易空间的能带为自旋简并(图1(b))。反铁磁体中磁矩的严格补偿需要对称性来加以保护。奈尔先生最早提出了一种相邻磁矩反平行排列的晶格结构, 其中相反的磁亚晶格通过平移对称操作(translation, t)而相互关联。换句话说, 当对该晶格施加时间反演(time reversal, T)操作时, 得到的磁结构与初始态的区别仅为空间上平移了特定的晶格常数, 从而保证了体系的宏观磁化为零。另一方面, 由于薛定谔方程在平移操作下不变, 体系中自旋向上和自旋向下的电子能谱完全相同, 导致倒易空间中的能带自旋简并。因此, 宏观磁化的存在曾被认为是时间反演对称性破缺的必要条件。

传统的磁学观点认为, 铁磁体与反铁磁体的特征是互相排斥, 泾渭分明的。然而, 2019~2022年期间, 多个研究团队相继从理论上提出了一种新的共线磁结构——交错磁体(altermagnet), 打破了这近一百年来的认知^[1~11]。2022年5月, 我们与美国康奈尔大学的D. Ralph教授团队同期独立报道了交错自旋劈裂效应驱动的非常规自旋力矩行为, 被国际同行认为是交错磁体的原创性实验^[2](initial experiments), 后续的谱学和输运实验进一步证实了交错磁性的存在。

在交错磁体中, 相邻磁矩呈反平行排列, 从而在实空间

引用格式: 白桦, 宋成. 一种新磁性的发现——交错磁性. 科学通报, 2025, 70: 4156–4164

Bai H, Song C. A new type of magnetism emerges—altermagnetism (in Chinese). Chin Sci Bull, 2025, 70: 4156–4164, doi: [10.1360/TB-2024-1381](https://doi.org/10.1360/TB-2024-1381)

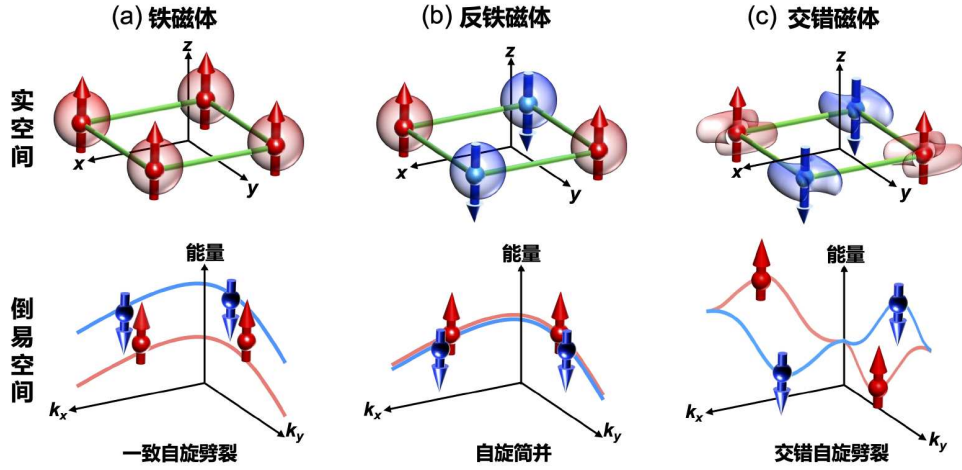


图 1 (网络版彩色)铁磁体、反铁磁体和交错磁体在实空间和倒易空间中的示意图. (a) 铁磁体实空间磁矩平行排列, 倒空间存在一致自旋劈裂. (b) 反铁磁体实空间相邻磁矩反平行排列, 倒空间自旋简并. (c) 交错磁体实空间相邻磁矩反平行排列, 但具有各向异性自旋密度, 倒空间存在交错自旋劈裂

Figure 1 (Color online) Schematics of a ferromagnet, an antiferromagnet, and an altermagnet in real and reciprocal spaces. (a) Ferromagnet exhibits parallel moments in real space and the unidirectional spin splitting in reciprocal space. (b) Antiferromagnet exhibits antiparallel moments in real space and spin degeneracy in reciprocal space. (c) Altermagnet exhibits antiparallel moments and the anisotropic spin density in real space, as well as the alternating spin splitting in reciprocal space

磁矩相互补偿, 导致净磁矩为零, 表现出类似于反铁磁体的特征. 从倒易空间看, 交错磁体中自旋向上和自旋向下的能带交错地劈裂, 表现出类似于铁磁体的特征(图1(c)). 这种看似矛盾的特征集合使得交错磁体融合了铁磁体与反铁磁体的双重优势: 一方面, 由于自旋能带劈裂, 交错磁体在理论上具有显著的自旋极化流和隧穿电阻效应, 相比反铁磁体, 有望实现更高效的信息读写; 另一方面, 净磁矩为零意味着交错磁体不会产生杂散场, 且对外不显示磁性, 相比铁磁体在密度和高稳定性信息存储方面有显著优势. 此外, 由于相邻磁矩间具有强烈的反铁磁耦合, 交错磁体的本征磁动力学频率高达THz量级(与反铁磁体量级相同), 显著高于铁磁体(GHz量级), 这使得交错磁体在实现高速信息读写和处理方面以及太赫兹通讯领域具有潜力. 交错磁体的概念一经提出, 引起了广泛关注, 并催生了一系列揭示新奇物理现象的理论与实验研究, 展现了其在构筑新型自旋电子器件方面的巨大潜力.

既然交错磁体有如此多的优势, 为什么直到最近才被发现呢? 这就要从共线磁体的对称性讲起. 自奈尔先生提出反铁磁性的概念以后, 物理学家们认识到, 除了平移对称性, 反铁磁中相反的磁亚晶格还可以通过其他对称性进行关联, 典型的就空间反演对称性(inversion, P). 可以证明, 如果一类磁体的空间和时间反演的联合对称性(PT)保持, 那么其倒易空间的能带必然具有自旋简并性^[12]. 反之, 若一类磁体满足 PT 和 tT 对称性破缺, 例如相反磁亚晶格通过旋转对称性(rotation, C)关联, 那么其倒易空间的自旋简并性可能在某些位置被打破. 此时, 对称性决定了该体系在实空间和倒易空间中

存在交错排列的自旋极化(即动量相关的能带自旋劈裂或称为对称性连接的自旋-动量锁定), 这是交错磁体的一个重要特征.

对于磁亚晶格反平行排列的共线磁体, 可以通过分析其晶体对称性来识别它是否为交错磁体. 例如, 在典型中心反演对称的NiAs型晶体结构中, 若磁性原子处于As所在位置, 而非磁原子处于Ni的位置, 体系 PT 和 tT 对称性保持, 因而具有反铁磁的特征; 而当磁性原子处于Ni所在位置时, 体系 PT 和 tT 对称性破缺(如 α -MnTe和CrSb), 因而表现为交错磁体的特征^[13](图2(a)). 类似地, 在中心反演对称体系中, 若调换初基晶胞中某一对磁亚晶格的位置, 反铁磁体在理论上也可能转变为交错磁体(图2(b)).

对称性分析可以初步判断体系是否为交错磁体, 但要确认交错磁性的存在, 仍然需要依赖第一性原理计算和实验证实. 角分辨光电子能谱(angle-resolved photoemission spectroscopy, ARPES)技术广泛用于探测材料的能带结构, 因此, 它是探测交错磁性的重要手段之一^[14]. 实验结果表明, 在不同的交错磁体候选材料中, 都能够观察到自旋能带劈裂的现象. 以 α -MnTe为例, 软X射线-ARPES (soft X-ray ARPES)的测试结果表明, 沿倒易空间 Γ -K-K方向存在显著的自旋能带劈裂^[15], 这一实验结果与理论计算结果吻合得很好(图3(a)), 支撑了 α -MnTe中存在交错磁性. 自旋分辨的ARPES技术(spin-resolved ARPES)能够区分自旋向上和自旋向下的能带^[16], 因而可以更清晰地揭示交错磁体自旋劈裂的对称性(图3(b)), 并进一步证实交错磁性的存在. 值得注意的是, 除了 α -MnTe^[17-20]之外, ARPES实验结果表明其他交错磁体候选材

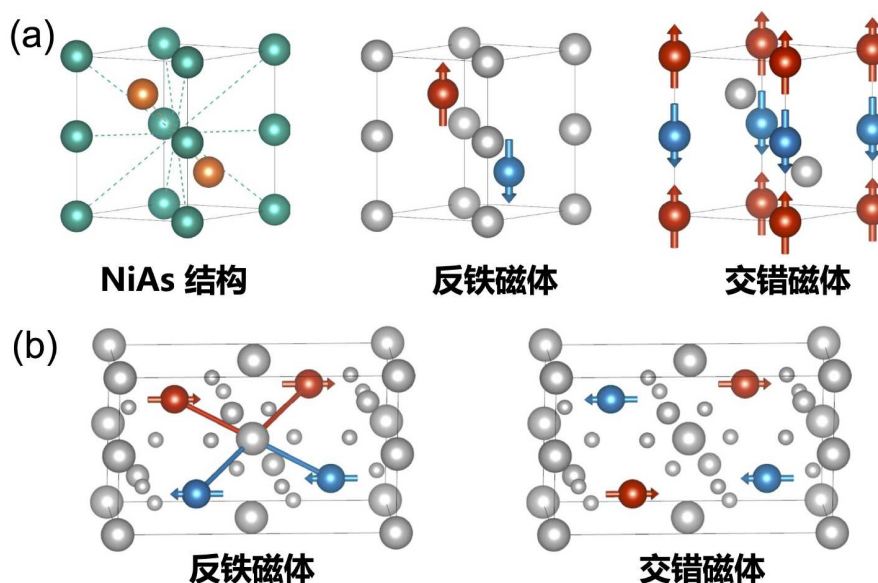


图 2 (网络版彩色)通过晶体对称性分辨交错磁体^[13]. (a) 根据磁性原子和非磁原子的位置识别交错磁体和反铁磁体. (b) 根据相反的磁亚晶格中原子的位置识别交错磁体和反铁磁体

Figure 2 (Color online) Identification of altermagnets through crystal symmetry^[13]. (a) Distinguishment of altermagnets and antiferromagnets based on positions of magnetic and non-magnetic atoms. (b) Distinguishment of altermagnets and antiferromagnets based on positions of opposite spin sublattices

料如CrSb^[21~26]、RuO₂^[27,28]、Rb掺杂V₂Te₂O^[29]和K掺杂V₂Se₂O^[30]等,也表现出交错磁性的显著特征.

X射线磁圆二色谱(X-ray magnetic circular dichroism, XMCD)是另一类可以用来探测交错磁体中时间反演对称性破缺的谱学技术,且具有元素可分辨的优势. 尽管XMCD不能直接解析倒易空间的能带结构,但结合第一性原理计算后,可以判断体系中是否存在交错排列的自旋极化^[27,31,32],为交错磁性的存在提供佐证. 更为重要的是, XMCD结合光发射电子显微镜(photoemission electron microscopy, PEEM)技术,能够对交错磁体实现磁畴成像^[33],为深入研究交错磁性提供了更为直接的探测手段. 事实上,除了磁圆二色谱外, X射线吸收谱(X-ray absorption spectroscopy, XAS)还有另一类典型的磁圆二色谱(X-ray magnetic linear dichroism, XMLD)测试构型,可以用来探测自旋排列的轴向. 通常来说,铁磁具有时间反演对称性破缺,因而可以利用XMCD探测其磁矩方向^[34]. 对于反铁磁而言, XMCD往往难以得到有效的信号,而XMLD则常用于表征其90°磁畴^[35]. 交错磁体兼具铁磁体与反铁磁体的特征, XMCD和XMLD均可反映其磁矩排列状态,但二者的探测内容有所区别. 以 α -MnTe为例,其磁晶各向异性轴为(0001)面内 $\langle 1\bar{1}00 \rangle$ 的等效晶轴,因而奈尔矢量存在6个等效状态. XMLD可以探测奈尔矢量所排布的轴向, XMCD则可以判断处于所在轴的0°还是180°态. 因此,结合XMLD和XMCD的PEEM结果可以获得 α -MnTe中空间分辨的奈尔矢量排布(图3(c)),这在常规的共线反铁磁体中难以实现.

磁性材料中自旋与电荷的相互作用及其输运行为是自旋电子学的核心研究内容之一,也是其器件应用的关键之一. 在交错磁体中,交错自旋能带劈裂会引发一系列时间反演对称破缺的非平庸磁响应,包括反常霍尔效应^[36~42](anomalous Hall effect, AHE)、反常能斯特效应^[43,44](anomalous Nernst effect, ANE)、磁光克尔效应^[41](magneto-optical Kerr effect, MOKE)、非相对论的自旋-电荷转换^[45~53](non-relativistic spin-charge conversion)和巨磁电阻(giant magnetoresistance, GMR)/隧穿磁电阻效应^[54~59](tunneling magnetoresistance, TMR)等. 其中, AHE、ANE和MOKE效应源于倒易空间中的贝里曲率热点,因此需要考虑自旋轨道耦合的影响. 而非相对论的自旋-电荷转换过程、GMR/TMR等是交错自旋劈裂的直接结果,不需要考虑自旋轨道耦合. 这些物理过程不仅揭示了交错磁体在多物理场(电学、热学和光学)激励下的磁响应(图4(a)~(c)),展现了交错磁体的独特物理特性,也为构筑基于交错磁体的自旋电子器件奠定了理论基础.

上述依赖于时间反演对称破缺的磁响应(AHE、ANE、MOKE和TMR等)为探测交错磁体的奈尔矢量提供了可行的方案. 另一方面,如何实现对奈尔矢量的高效操控同样至关重要,这直接关系到基于交错磁体的自旋器件能否实现低功耗的信息写入和数据处理. 电流诱导的自旋轨道力矩效应为操控交错磁性提供了契机^[60,61]. 利用自旋轨道力矩实现奈尔矢量的180°确定性翻转,对磁存储的二进制写入十分重要,但同时极具挑战. 沿电流方向施加辅助磁场是诱使“确定性”

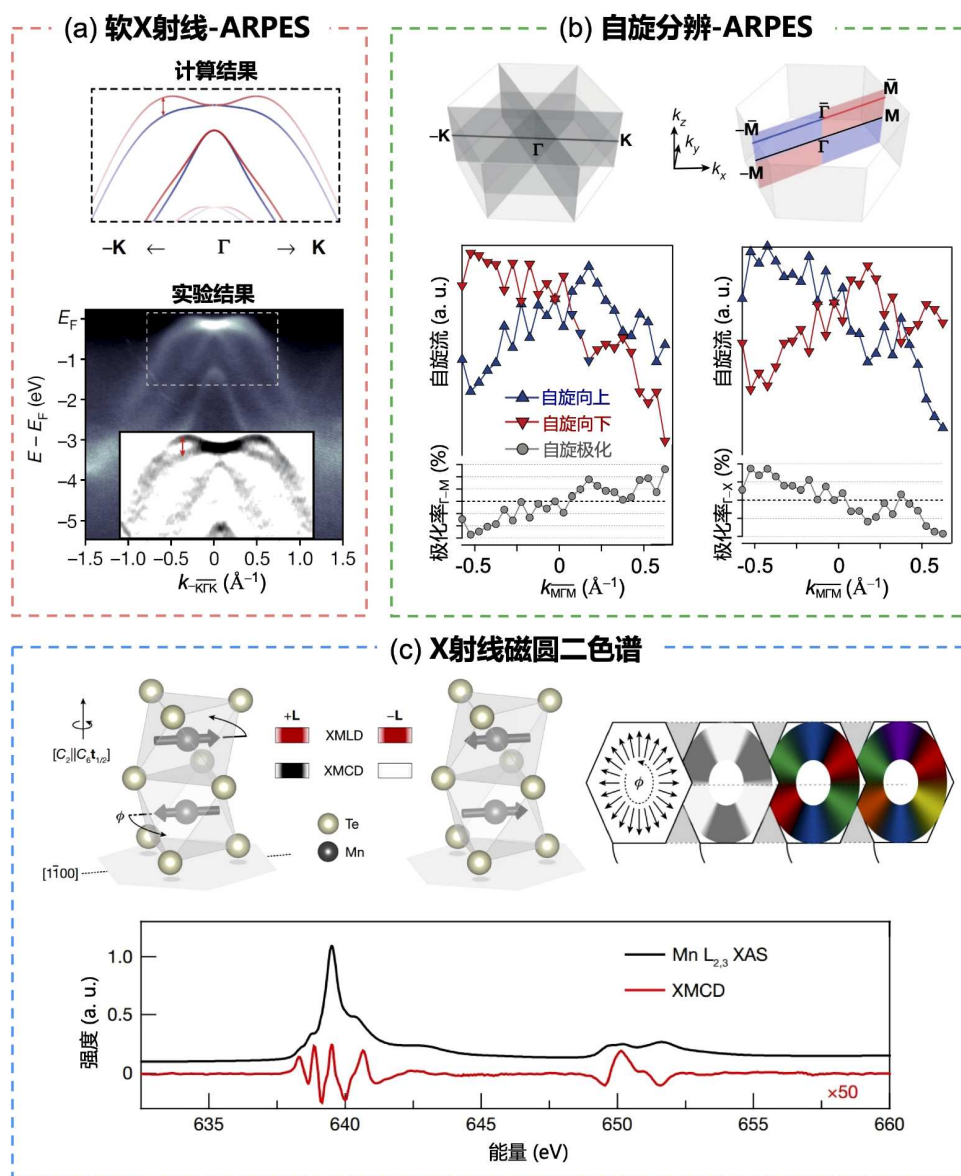


图 3 (网络版彩色)谱学技术探测交错磁性^[15,33]. (a) 软X射线-ARPES探测 α -MnTe的能带劈裂. (b) 自旋分辨-ARPES探测 α -MnTe自旋劈裂的对称性. (c) X射线磁圆二色谱探测 α -MnTe的时间反演对称性破缺

Figure 3 (Color online) The probe of altermagnetism via spectroscopic techniques^[15,33]. (a) The probe of electronic band splitting in α -MnTe using soft X-ray ARPES. (b) The probe of symmetry of spin-split bands in α -MnTe using angle-resolved ARPES. (c) The probe of time-reversal symmetry breaking in α -MnTe using XMCD

的可行方案, 在磁场和自旋轨道力矩协同作用下, 奈尔矢量实现 180° 循环翻转^[41](图4(d)). 此外, 由于交错磁体的序参量“交错磁序”包含奈尔矢量和晶体对称性两个维度, 引入应变调制晶体对称性为调控交错磁性提供了新的维度^[62,63]. 值得注意的是, 在应变和自旋轨道力矩的协同作用下, 可以在零磁场下对交错磁体奈尔矢量的 180° 确定性翻转^[62], 这不仅体现出交错磁体的构效关系, 也为构筑高密度、超快、低功耗磁随机存储器提供了新的方向.

交错磁体中的自旋-电荷转换能够产生非常规的面外自旋极化流, 从而实现对工业界青睐的垂直磁记录层的零磁场磁化翻转(图4(e))^[48,64]. 其逆过程可将面外自旋转换为电荷流读出(图4(f)), 这一物理过程可用于构建可调的太赫兹发射器^[50]. 从倒易空间看, 交错磁体具有动量相关的能带自旋劈裂, 因而若构筑隧道结构型后, 在不同动量位置的电子具有不同的隧穿概率, 理论上表现出类似铁磁的显著隧穿磁电阻效应(TMR比值 $>100\%$)^[54,55], 可用于磁存储器的信息读取.

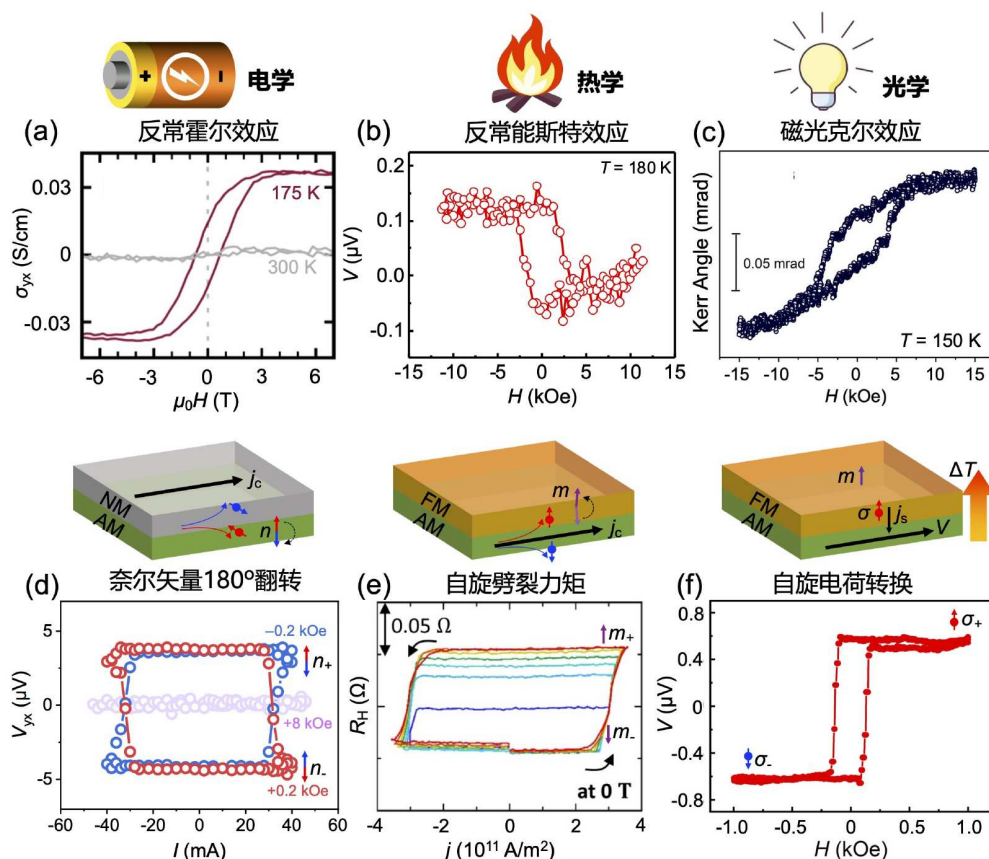


图4 (网络版彩色)交错磁体的自旋相关现象^[38,41,44,48,49]. (a) 反常霍尔效应. (b) 反常能斯特效应. (c) 磁光克尔效应. (d) 奈尔矢量180°翻转. (e) 自旋劈裂力矩. (f) 自旋电荷转换

Figure 4 (Color online) Spin-related phenomena in altermagnets^[38,41,44,48,49]. (a) Anomalous Hall effect. (b) Anomalous Nernst effect. (c) Magneto-optical Kerr effect. (d) 180° switching of Néel vector. (e) Spin splitting torque. (f) Spin-charge conversion

基于对称性分析已初步识别了超过200个交错磁体的候选材料^[65], 包括金属^[21]、半导体^[15]、绝缘体^[66]和超导体^[67,68], 以及二维范德华材料^[69]和有机材料^[4]等. 具体而言, 交错磁体例如 α -MnTe允许室温(交错)磁性半导体性共存, 这是磁性半导体领域长期追求的目标. 交错磁绝缘体例如 α -Fe₂O₃具有磁子能带劈裂的特性^[70], 这将进一步增进对自旋波领域的认知. 交错磁性同样允许与超导性共存, 为构筑零场下具有超导二极管效应的约瑟夫森结提供了新的途径^[71,72]. 范德华材料和有机材料与交错磁性的结合将推动可堆垛/柔性交错磁体器件的发展. 动量空间中, 交错磁体可具有2/4/6等偶数个等能面, 对应的磁密度具有 $d/g/i$ 波对称性. 理论工作指出, d 波对称性的交错磁体具有各向异性的自旋电导率, 因而允许存在巨磁阻效应, 而在具有更高阶对称性的交错磁体中是不被允许的^[55]. 作为对照, 隧穿磁电阻效应依赖于动量相关的导电通道, 而并非依赖于平均化的自旋电导率, 因而理论上在任意对称性的交错磁体中均被允许^[55]. 值得注意的是, 尽管交错磁体的理论相关研究已经取得诸多进

展, 但实验研究尚处于起步阶段. 相比于传统的铁磁和反铁磁材料, 交错磁体对于晶体质量和单畴的要求更为严格, 这也成为了限制交错磁性实验探究的严峻挑战^[33]. 面对这一难题, 优化材料生长工艺和探索新的交错磁体材料将成为该领域重要的发展方向.

交错磁体的发现是近年来材料科学和凝聚态物理领域的一个重要突破, 入选了Science 2024年度十大科学突破^[65]. 它挑战了传统的铁磁和反铁磁理论, 提出了一种新的磁性状态. 除了自旋电子学的细分领域外, 交错磁体的发现有望为凝聚态物理的其他研究领域带来新的研究活力, 例如超导^[67,68]、拓扑学^[73,74]和超快光学^[75,76]等. 尽管目前我们对于交错磁体的理解仍处于早期阶段, 而且实验中发现的交错磁体材料仍较为局限, 但随着理论和实验研究的不断深入, 交错磁体将在自旋电子学材料与器件、量子信息技术以及其他信息功能材料等领域中发挥越来越重要的作用. 未来更多的实验和理论研究将进一步揭示交错磁体中蕴含的新现象、新机制、新物理, 推动相关技术的进步和应用的发展.

致谢 感谢国家自然科学基金(52225106, 12241404)和国家重点研发计划(2022YFA1402603)资助。

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Summary for “一种新磁性的发现——交错磁性”

A new type of magnetism emerges—altermagnetism

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Altermagnets are characterized by non-relativistic alternating spin splitting in the band structure and collinear compensated magnetic moments in real space. They combine the advantages of both ferromagnetic and antiferromagnetic order, exhibiting time-reversal symmetry-breaking magneto responses, vanishing stray fields, and high-frequency spin dynamics. Consequently, altermagnets hold great potential for various research fields, especially for developing spintronic devices such as high-density magnetic memories and terahertz nano-oscillators. Furthermore, altermagnetism is found in a broad spectrum of materials, including metals, semiconductors, insulators, and superconductors, thereby stimulating widespread interest in functional material research. In this paper, we provide an overview of recent experimental progress in altermagnets, focusing particularly on observations of lifted spin degeneracy via spectroscopic techniques and the resultant spin transport phenomena.

The non-relativistic alternating lifting of spin degeneracy is a pivotal aspect of altermagnets. Substantial effort has been dedicated to directly probing the spin splitting with altermagnetic symmetry in reciprocal space. Angle-resolved photoemission spectroscopy (ARPES) is a powerful technique for investigating the electronic energy bands of materials, making it well-suited for probing spin splitting in altermagnets. Pioneering measurements were recently achieved in altermagnet candidates, such as α -MnTe and CrSb, using soft X-ray ARPES. Besides, spin-resolved ARPES can directly distinguish spin-up and spin-down bands at lower photon energies, which has been employed for various altermagnets, including layered Rb-doped V_2Te_2O and K-doped V_2Se_2O . Another crucial technique in element-specific spectroscopy and microscopy is X-ray magnetic circular dichroism (XMCD). Characteristic XMCD data of altermagnets have been theoretically calculated and experimentally explored. Although band splitting cannot be directly resolved by this method, the broken time-reversal symmetry and the presence of alternating spin polarization can be indirectly inferred in conjunction with theoretical models. The XMCD technology sparked a natural interest in utilizing photoelectron emission microscopy to image altermagnetic domains. It can also be detected in angle-resolved photoemission spectra, offering a direct way to observe time-reversal symmetry breaking in the band structure.

Functionalizing the interaction between spin and charge constitutes a core focus of spintronics, and it is explored under various external stimuli such as magnetic or electric fields, optical or thermal excitation, and strain. The spin-split band structure of altermagnets enables time-reversal symmetry-breaking responses, such as anomalous Hall and Nernst effect, magneto-optical Kerr effect, giant/tunneling magnetoresistance, non-relativistic charge-spin conversion, and unconventional piezomagnetism. The modulation of altermagnetism by multiple methods, such as electric current and strain, is also discussed.

Overall, exploring altermagnetic candidates in various material systems is highly significant, as it opens new horizons in various fields, including spintronics, magnonics, ultrafast photonics, phononics, superconductivity, topology, and multiferroicity. Specifically, altermagnetic semiconductors such as α -MnTe permit the coexistence of high-temperature magnetism and semiconducting properties, a long-pursued goal in dilute magnetic semiconductor research. For altermagnetic insulators, the chiral-split magnon bands would advance the research of spin waves. Altermagnetism is also proposed to be compatible with spin-triplet superconductors, providing a platform for exploring unconventional superconductors with zero stray fields and developing field-free superconducting diode effects. Moreover, experimental investigations of organic and van der Waals altermagnetic candidates would pave the way to flexible and easily stackable spintronic devices.

spintronics, altermagnet, spin splitting, time-reversal symmetry breakingdoi: [10.1360/TB-2024-1381](https://doi.org/10.1360/TB-2024-1381)