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双层范德华磁性材料的理论研究进展

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摘要 二维范德华磁性材料因其独特的物理特性已成为下一代自旋电子器件的重要候选材料, 尤其在双层磁性材料中, 多样化的堆叠构型诱导出可调控的磁序和丰富的量子现象. 基于第一性原理计算, 研究人员能够系统预测和设计新型磁性堆叠结构. 随着人工智能的快速发展, 高通量计算与深度学习为双层磁性材料的探索开辟了新途径. 本文首先讨论了层间滑移与转角对磁序和物态的调控机制, 分析了层间耦合在磁电耦合、非共线自旋态及磁斯格明子形成中的关键作用. 随后, 介绍了高通量计算与深度学习在加速磁性材料堆叠设计中的关键应用. 最后, 对该领域面临的挑战进行了展望, 并提出了未来可能的发展方向.

关键词 二维磁性材料, 层间堆叠, 磁电耦合, 深度学习

随着信息技术的快速发展, 传统半导体技术逐渐逼近物理极限, 推动研究人员积极探索新材料和新的物理现象. 自旋电子学作为一种利用电子自旋自由度的新兴技术, 展现出实现低功耗高效信息处理的巨大潜力. 二维磁性材料凭借其独特的物理特性, 被视为新一代自旋电子器件的理想候选材料[1]. 自石墨烯成功剥离以来[2], 实现二维材料本征磁性的探索一直是研究热点. 2017年, CrI_3[3]和$Cr_2Ge_2Te_6$[4]两种二维磁体在实验上取得了重要突破, 随后更多二维范德华磁性材料被成功合成[5~9].

二维范德华磁性材料由原子层通过弱范德华力堆叠而成, 其独特的弱层间耦合赋予了原子层相对晶体排列的高度可调性, 从而形成多样化的堆叠构型. 随着合成方法和聚合物辅助转移技术的发展[10,11], 范德华层的按需堆叠成为可能, 展现出堆叠依赖的磁序[12,13]、反常谷霍尔效应[14]和莫尔超晶格中的非共线自旋[15,16]等新颖量子现象. 此外, 非极性单层通过特定堆叠可产生滑移铁电性, 为实现磁电耦合提供了新思路, 有望开拓新型关联物理和器件应用[17~19]. 这些研究表明, 堆叠序通过调控层间电荷与自旋重分布, 可以显著影响材料的拓扑特性、电子关联及磁性行为. 近年来, 层间滑移和转角这两个结构自由度的调控为探索和设计双层磁性体系中的新奇量子态提供了广阔平台.

随着理论计算方法的发展和计算性能的提升, 第一性原理计算在材料模拟和设计中发挥了重要作用[20,21]. 在研究双层磁性材料堆叠构型时, 第一性原理计算能够系统探索层间滑移和转角变化, 高效预测和设计具有新奇物态的堆叠结构. 近年来, 随着人工智能的快速发展, 数据驱动的科研范式为材料设计开辟了新途径. 高通量计算通过高效生成和分析大规模数据集, 突破了传统计算仅聚焦于少数化合物的局限. 同时, 深度学习的突破使得人工神经网络能够从大量计算数据中学习, 在无需调用第一性原理计算代码的情况下实现大规模材料体系的高效模拟[22,23]. 这些方法在双层磁性材料研究中的广泛应用加速了对材料性质的深入理解和新材料的发现.

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本文首先介绍双层范德华磁性材料在层间滑移和转角调控下展现的新奇量子态, 包括堆叠依赖的磁序、磁电耦合和莫尔超晶格中的非共线自旋态等. 随后, 结合高通量计算和深度学习加速的堆叠构型计算, 深入探讨人工智能在加速材料发现中的应用前景. 最后, 本文提出当前研究中面临的一些挑战, 并展望未来可能的发展方向.

1 双层磁性材料的堆叠调控

材料的磁性源于磁性原子轨道重叠所产生的磁矩之间的交换相互作用. 在研究二维磁性材料时, 为了准确描述这些复杂的相互作用机制, 采用有效自旋哈密顿量进行系统分析[24]. 自旋哈密顿量通过对描述晶格位点上电子行为的哈伯德模型进行折叠得到, 仅保留了自旋自由度[25]. 其一般形式可表示为

$$H_{\text{spin}} = -\sum_{i,j} \boldsymbol{S}_i \mathcal{J}_{ij} \boldsymbol{S}_j - \sum_i \boldsymbol{S}_i \mathcal{A}_i \boldsymbol{S}_i, \tag{1}$$

其中, $\boldsymbol{S}(S_x, S_y, S_z)$表示自旋向量, $\mathcal{A}$描述单离子各向异性, $\mathcal{J}$为交换相互作用矩阵, 包括各向同性海森堡相互作用、对称各向异性相互作用和反对称相互作用, 其中反对称部分对应Dzyaloshinskii–Moriya (DM)相互作用[26]. 这些相互作用参数可通过基于总能量[27,28]或微扰理论[29,30]的方法计算得到. 在双层磁性材料体系中, 堆叠构型显著影响层间交换耦合, 自旋与电荷之间的相互作用可能诱导出丰富的拓扑特性和关联现象. 本章将系统探讨层间滑移和转角对双层磁性材料物态的调控机制, 揭示其在诱导磁电耦合和非共线自旋等新奇量子态形成中的作用.

1.1 层间滑移

1.1.1 磁序转变

由于层间耦合对二维磁性材料的物理性质具有显著影响, 堆叠构型成为调控磁序的有效方法. 以CrI_3为例, 通过施加静水压力可实现其层间堆叠的机械调控, 诱导其从单斜相向菱面体相转变, 同时伴随反铁磁到铁磁的相变[12,13]. 如图1(a)所示, 双层CrI_3中, AB堆叠(菱面体相)倾向于层间铁磁耦合, 而AB′堆叠(单斜相)表现为层间反铁磁耦合. 这两种堆叠构型可通过层间滑移相互转换, 其磁序变化源于不同堆叠构型下层间交换相互作用的显著差异[31]. Cr原子的层间交换相互作用由I原子的p轨道杂化介导, 其磁序由反铁磁t_{2g}−t_{2g}轨道耦合与铁磁t_{2g}−e_g轨道耦合的竞争决定. 如图1(b)所示, AB堆叠构型中, 由于次近邻t_{2g}−e_g轨道耦合远多于最近邻t_{2g}−t_{2g}耦合, 层间交换表现为铁磁耦合. 当堆叠方式从菱面体相转变为单斜相时, 最近邻t_{2g}−t_{2g}耦合增多, 次近邻t_{2g}−e_g耦合减少, 导致层间反铁磁交换占主导地位. 最新研究揭示了CrI_3在磁序转变过程中展现出独特的拓扑性质[32]. 第一性原理计算和紧束缚模型分析表明, CrI_3单层和双层均为具有非零第二Stiefel-Whitney数(w_2=1)的二阶拓扑绝缘体, 这种拓扑态在铁磁和反铁磁构型转变中表现出鲁棒性. 此外, 理论预测表明, 通过180°旋转双层CrI_3的堆叠构型同样可诱导磁序转变[33].

其他二维材料体系也展现出类似的磁序调控特性[34~36]. 基于群论分析, 研究人员提出了从铁磁单层构建交错磁相双层的堆叠方法[37]. 以图1(c)所示的$NiZrI_6$为例, 通过直接堆叠引入面内C_2对称性, 实现了典型的交错磁性, 其中层间交换的各向异性进一步诱导自旋分裂. 在双层$PtCl_3$中, 层间位移能够诱导层内的铁磁-反铁磁相变[38]. 双层$MnBr_3$不仅表现出堆叠依赖的磁基态, 还展现出丰富的可调拓扑特性: 其铁磁双层表现出高陈数量子反常霍尔效应, 而在反铁磁双层中, 静电场或激光诱导的贝里曲率奇点引发了新型层霍尔效应[39].

1.1.2 滑移诱导的铁电性与磁电耦合

近年来, 二维材料中基于极性堆叠的滑移铁电现象引起了广泛关注[40~42]. 与传统铁电材料不同[43,44], 二维材料的滑移铁电性主要源于层间极化而非离子位移[45]. 这一机制要求堆叠模式同时打破镜像对称性和反演对称性, 形成可通过层间滑移切换的净极化. 外加电场能够有效驱动层间滑移, 实现高效且稳定的铁电极化翻转. 层间范德华力较弱, 导致切换势垒降低, 滑移铁电材料为实现多铁性和开发低能耗电子器件提供了理想平台.

基于群论的系统研究全面分析了所有可能的对称性, 提出多种堆叠铁电范式[46]. 在双层CrI_3中, 通过层间180°翻转并配合特定平移操作, 可打破镜像对称性和反演对称性, 诱导铁电极化. 该结构同时展现出面内和面外极化分量, 其铁电性和磁性可通过电场精确调控, 如图1(d)所示. 极性堆叠构型不仅产生铁电性, 还通过对称性破缺诱导出各向异性DM相互作用. 通过电场翻转铁电极化可实现DM相互作用的反转, 为反斯格明子的电场调控提供了新途径[47]. 在反铁磁双层VS_2中, 发

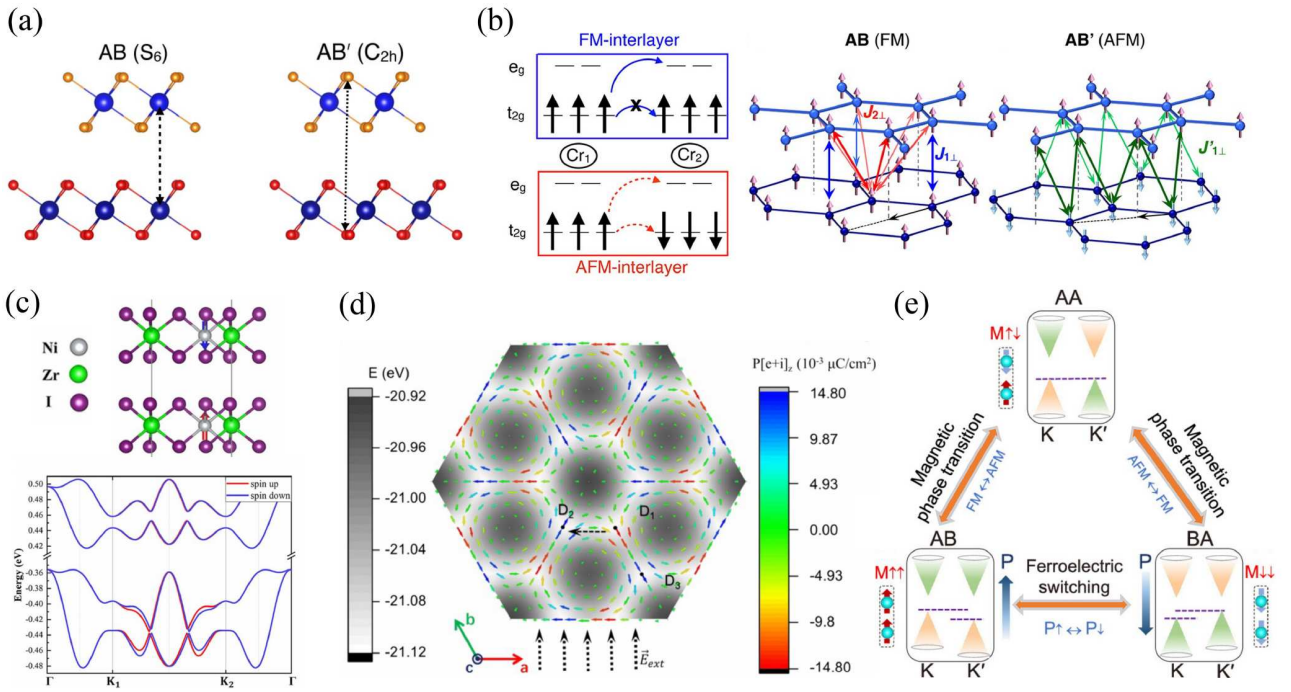

图 1 (网络版彩色)层间滑移对双层磁性材料的物态调控. (a) 双层CrI_3的AB堆叠和AB′堆叠晶体结构及其对应点群[31]. (b) 轨道依赖的层间超交换相互作用示意图: AB堆叠中Cr原子的层间最近邻相互作用($J_{1\perp}$)和次近邻相互作用($J_{2\perp}$), 以及AB′堆叠中的最近邻相互作用($J'_{1\perp}$)[31]. Copyright © 2018 American Chemical Society. (c) 直接堆叠双层$NiZrI_6$的晶体结构和能带结构[37]. Copyright © 2024 American Physical Society. (d) 双层CrI_3在不同平移下的极化强度与磁基态能量分布[46]. Copyright © 2023 American Physical Society. (e) 双层GdI_2中磁性、铁电与铁谷共存的多铁性机制示意图[34]. Copyright © 2024 American Chemical Society

Figure 1 (Color online) Interlayer sliding modulation of physical properties in bilayer magnetic materials. (a) Crystal structures and corresponding point groups of bilayer CrI_3 in AB-stacking and AB′-stacking[31]. (b) Schematic of orbital-dependent interlayer superexchange interactions: interlayer nearest-neighbor ($J_{1\perp}$) and second-neighbor ($J_{2\perp}$) interactions of Cr atoms in AB-stacking, and nearest-neighbor ($J'_{1\perp}$) interaction in AB′-stacking[31]. Copyright © 2018 American Chemical Society. (c) Crystal structure and band structure of direct-stacking bilayer $NiZrI_6$[37]. Copyright © 2024 American Physical Society. (d) Polarization and energy distribution of magnetic ground states for different translations in bilayer CrI_3[46]. Copyright © 2023 American Physical Society. (e) Schematic mechanism of coexisting magnetic, ferroelectric and ferrovalley multiferroic in bilayer GdI_2[34]. Copyright © 2024 American Chemical Society

现了铁谷介导的磁电耦合, 其铁电极化翻转伴随着层间相对滑移和磁性亚晶格的自旋翻转[19]. 理论预测表明, 双层GdI_2同时具有铁电性、铁磁性和谷极化, 可通过滑移铁电性实现磁序转变和谷极化的协同调控, 如图1(e)所示[34]. Liu等人[48]提出的双层FeS铁电模型展现了通过层间滑移实现面外电极化对量子反常层霍尔效应调控的可能. 在双层$MnBi_2Te_4$中, 层间滑移可同时调控其拓扑、磁性和铁电性质, 实现从铁磁绝缘体到陈绝缘体的转变, 其中电荷转移导致反铁磁基态与铁电相共存[49]. 此外, MnSe多层材料也展现出通过层间滑移调节磁相及其磁电耦合的潜力[50].

1.2 层间转角

1.2.1 莫尔超晶格

转角双层石墨烯中超导性和关联绝缘态的发现, 推动了对二维莫尔材料的广泛研究[51,52]. 莫尔超晶格是指当两个周期性晶格稍有错位时, 叠加形成的周期远大于原始晶格的独特结构[53~55]. 在范德华双层体系中, 小角度层间转角可诱导形成周期性莫尔超晶格. 这种超晶格结构对材料的能量景观产生周期性调制, 是理解二维转角材料量子行为的关键. 层间耦合形成的周期性莫尔势场显著影响电子行为, 其周期a_M由转角θ、两层晶格常数的失配δ和平均晶格常数a共同决定. 在小角度和小失配的情况下, 莫尔周期可表示为$a_M = \frac{a}{\sqrt{\theta^2+\delta^2}}$. 对于同质结构, 由于两层晶格常数相同, 计算可简化为仅与旋转角度θ相关[56,57]. 随着二维磁性材料研究的深入, 转角堆叠已成为探索新奇量子态和开发功能性器件的重要途径, 特别是在设计和调控二维转角磁体中的非共线自旋方面发挥着关键作用.

1.2.2 非共线自旋

非共线自旋是指磁矩取向不沿单一轴线排列的复杂磁性结构，包括斯格明子、螺旋磁性和磁多极子等多种新奇磁相[58~60]. 这类磁性结构因其在自旋电子学中的潜在应用价值而备受关注. 在转角体系中，局部莫尔堆叠引起的层间交换竞争被认为是形成非共线自旋态的关键机制. 体系的总自旋哈密顿量可表示为三项之和[61]:

$$H_{\text{spin}} = H_1 + H_2 + H_{\text{M}}, \tag{2}$$

其中，H_1和H_2分别代表两个二维磁体的自旋哈密顿量，H_{M}表示在转角界面处随莫尔周期变化的自旋哈密顿量. 为进一步探索转角双层体系的基态，研究人员采用Landau–Lifshitz–Gilbert (LLG)方程来模拟自旋动力学演化[62]:

$$\frac{\mathrm{d}\boldsymbol{m}}{\mathrm{d}t} = -\gamma \boldsymbol{m} \times \boldsymbol{B}^{\text{eff}} + \alpha \boldsymbol{m} \times \frac{\mathrm{d}\boldsymbol{m}}{\mathrm{d}t}, \tag{3}$$

其中，$\boldsymbol{m}$为磁化方向，$\boldsymbol{B}^{\text{eff}} = -\delta H_{\text{spin}} / \delta \boldsymbol{m}$为有效磁场，$\gamma$为回旋比，$\alpha$为Gilbert阻尼系数. 理论研究表明，原本具有共线自旋排列的二维磁体在转角堆叠形成的莫尔超晶格中可展现丰富的自旋构型[63~66]，其相图中出现多种非共线自旋态[66,67].

CrI_3因其层间堆叠依赖的磁序而引发了广泛研究兴趣. 如图2(a)所示，转角双层CrI_3的界面同时存在单斜和菱面体两种堆叠方式，在莫尔超晶格中形成周期性调制的层间交换耦合[16]. 这种交换耦合的竞争可能在铁磁和反铁磁畴的边界处诱导出现非共线自旋构型，

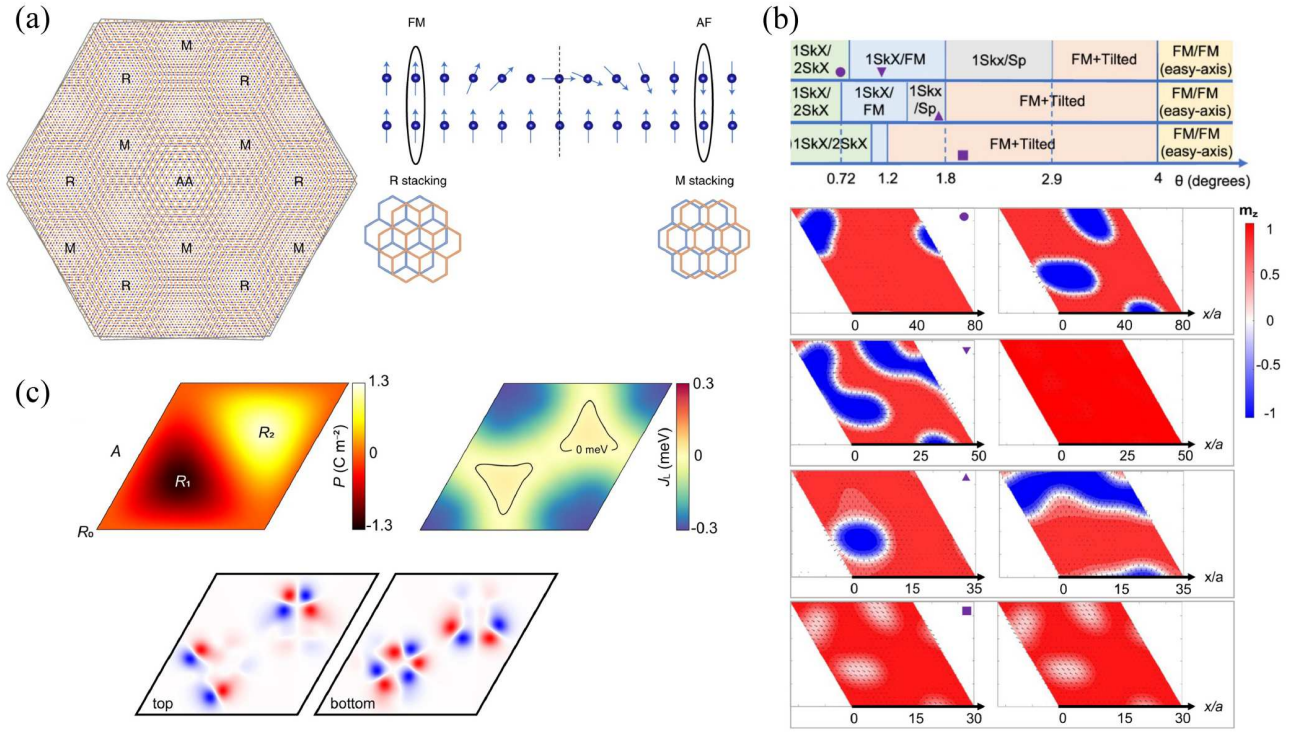

图 2 (网络版彩色)层间转角诱导的莫尔超晶格与非共线自旋. (a) 小转角双层CrI_3莫尔超晶格的结构示意图，展示了R型和M型堆叠区域之间的磁畴分布. 其中R、M和AA分别表示菱面体堆叠、单斜堆叠和直接堆叠构型[16]. Copyright © 2021 Springer Nature. (b) 转角双层CrI_3的相图及其磁化构型. 从上至下的相图分别对应D/J=0.2，0.1和0.05. 磁化结构从上到下依次展示了双斯格明子(1SkX/2SkX)、斯格明子/铁磁态(1SkX/FM)、斯格明子/螺旋磁态(1SkX/Sp)和铁磁+倾斜态. 左右面板分别对应上下两层的磁构型[69]. Copyright © 2021 American Chemical Society. (c) 3R型$LaBr_2$莫尔超晶格中的极化分布和层间交换相互作用分布，以及在转角θ=0.57°时的自旋构型[72]. Copyright © 2022 Springer Nature

Figure 2 (Color online) Interlayer twisting induced moiré superlattices and noncollinear spins. (a) Schematic illustration of small-angle twisted bilayer CrI_3 moiré superlattice, showing the magnetic domain wall formed between R- and M-stacking regions. R, M and AA represent rhombohedral, monoclinic and direct stacking, respectively[16]. Copyright © 2021 Springer Nature. (b) Phase diagram and magnetization textures of twisted bilayer CrI_3. Phase diagrams from top to bottom correspond to D/J=0.2, 0.1 and 0.05. Magnetization textures sequentially show skyrmion/skyrmion (1SkX/2SkX), skyrmion/ferromagnetic (1SkX/FM), skyrmion/spiral (1SkX/Sp) and ferromagnetic + tilted states from top to bottom. Left and right panels show the magnetic configurations of the top and bottom layers, respectively[69]. Copyright © 2021 American Chemical Society. (c) Distributions of the polarization and interlayer exchange interaction in 3R-type $LaBr_2$ moiré superlattices, along with the spin texture at twisting angle $\theta = 0.57°$[72]. Copyright © 2022 Springer Nature

突破传统的平行或反平行排列限制. 反演对称性破缺引起的DM相互作用可进一步稳定斯格明子[68~71], 图2(b)所示的相图展示了斯格明子与转角及DM相互作用的关联. 另外, 在小转角3R型堆叠的$LaBr_2$双层结构中, 研究人员发现了多铁性莫尔超晶格, 表现出强磁电耦合和磁双涡子[72]. 如图2(c)所示, 莫尔图案诱导的不均匀层间交换耦合形成了稳定的磁拓扑纹理, 并与交错的电极化畴实现耦合. 通过调节相对转角或施加层间应变, 可以有效调控层间交换耦合和空间对称性, 从而实现对这些拓扑磁性纹理的精确调控.

2 数据驱动的双层磁性材料研究

2.1 高通量计算

高通量计算材料设计作为材料科学的新兴领域, 突破了传统密度泛函理论(density functional theory, DFT)仅能处理少量化合物的局限. 通过建立自动化工作流程, 高通量计算能够高效生成、存储和分析数千种化合物, 显著加速了新材料的发现与设计进程[73,74]. 这一方法已在二维磁性材料的探索中取得了显著成果[75~77]. Pakdel等人[78]开展的研究凸显了高通量计算在探索二维范德华双层材料堆叠构型影响材料性质方面的显著优势. 如图3所示, 他们建立了系统的工作流程: 首先从二维材料数据库C2DB中筛选稳定的单层材料, 构建可能的同质双层结构; 随后计算层间距离和结合能, 结合滑移稳定性分析评估体系的稳定性; 最后对筛选出的稳定双层体系进行电、磁性质计算. 这一流程不仅系统筛选了稳定的双层磁性材料, 还深入揭示了堆叠构型对磁序的调控机制. 研究发现, $AgVP_2Se_6$和$FeTaTe_3$等37种磁性双层材料表现出堆叠构型依赖的磁态转变, 为滑移电子学器件的设计提供了重要材料基础. 其中, 镜像对称性破缺的单层材料由于内建偶极子诱导的电荷转移, 进一步调控层间耦合作用, 使双层堆叠展现出堆叠依赖的磁序, 这凸显了磁性Janus材料

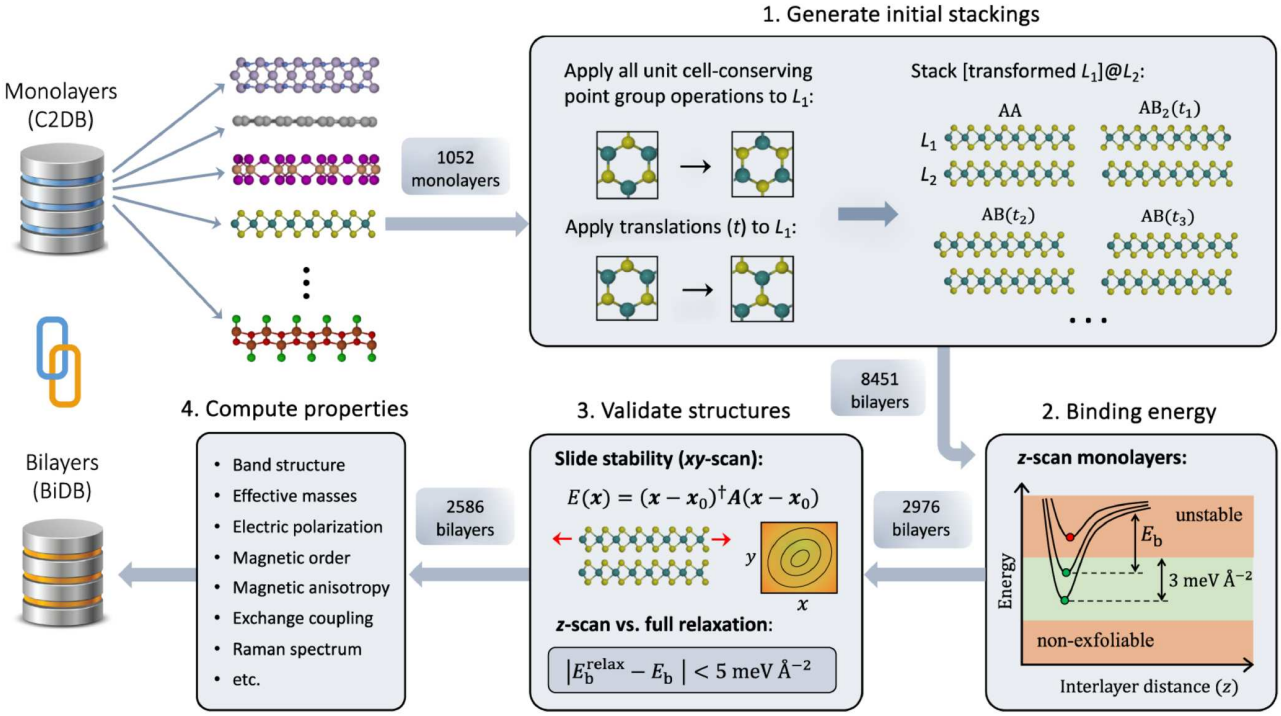

图 3 (网络版彩色)二维材料堆叠的高通量工作流. 首先导入1052个热力学稳定的单层结构, 基于AA堆叠构型通过点群对称操作和平移生成多种可能的堆叠构型. 经过结构重复性筛选后获得8451个双层结构. 通过计算层间结合能筛选出2976个热力学稳定的双层结构. 进一步经横向滑移测试和层间距离可靠性验证, 最终确定2586个稳定双层用于后续性质计算与分析[78]. Copyright © 2024 Springer Nature

Figure 3 (Color online) High-throughput workflow for two-dimensional material stacking. Starting from 1052 thermodynamically stable monolayers, various stacking configurations are generated through point group symmetry operations and translations based on AA stacking. Remove duplicate bilayers to obtain 8451 structures. Interlayer binding energy calculations identify 2976 thermodynamically stable bilayers. Further stability verification through lateral sliding tests and interlayer distance reliability checks yields 2586 stable bilayer structures for subsequent property calculations and analysis[78]. Copyright © 2024 Springer Nature

有望展现丰富可调的本征磁性. 此外, 该研究还筛选出了多种滑移铁电材料, 揭示了堆叠构型对层间极化的显著调控作用. 这些计算结果已存入范德华双层数据库BiDB, 以支持后续的深入研究和设计探索.

2.2 深度学习

转角双层磁体因其层间磁交换竞争诱导的非共线自旋态而受到广泛关注. 尽管第一性原理计算是研究这类体系的重要工具, 但其高计算成本限制了大规模结构研究中的应用. 深度学习的快速发展[22,23,79~81]为解决这一难题提供了新思路. 通过从大量计算数据中学习, 深度学习模型能够高效模拟大规模材料体系, 避免了直接进行第一性原理计算的需求. Zheng[68]开发的对称适应神经网络(symmetry-adapted artificial neural network, SANN)为研究转角磁性双层CrI_3开辟了新途径. 如图4(a)所示, 该研究采用锂离子封端的双层CrI_3团簇模型, 结合DFT和磁力理论(magnetic force theory, MFT)计算了不同转角和平移条件下的层间交换相互作用. SANN通过充分利用转角双层CrI_3的对称性丰富训练数据集, 构建了堆叠参数与层间磁交换相互作用的映射关系, 实现了对任意转角下层间交换的准确预测. 基于预测结果构建的自旋哈密顿量和LLG方程揭示, 在约60°转角时体系中出现有序分布的反铁磁域, 为磁斯格明子的稳定形成提供了理论依据. Li等人[82]发展的拓展深度学习DFT哈密顿量(extended deep learning DFT Hamiltonian, xDeepH)方法开创了探索复杂磁性体系电子结构的新范式. 如图4(b)所示, xDeepH通过深度等变神经网络框架, 将原子结构和磁结构信息映射为DFT哈密顿量(H_{DFT})的高效表示. 该方法结合了近视性原理与欧几里得和时间反演对称性的等变要求, 利用小规模磁性材料数据训练深度学习模型, 学习自旋-轨道H_{DFT}对原子和磁结构的依赖关系, 成功实现了对大尺度转角双层磁体H_{DFT}的预测. 在双层CrI_3固定60°转角的DFT数据集上训练后, xDeepH能够准确预测不同转角下的莫尔结构特性, 包括81.79°时的能带结构和63.48°时的磁斯格明子.

Lee等人[83]建立了哈密顿量参数与磁畴图像之间的双向映射. 如图4(c)所示, 他们设计的深度学习模型

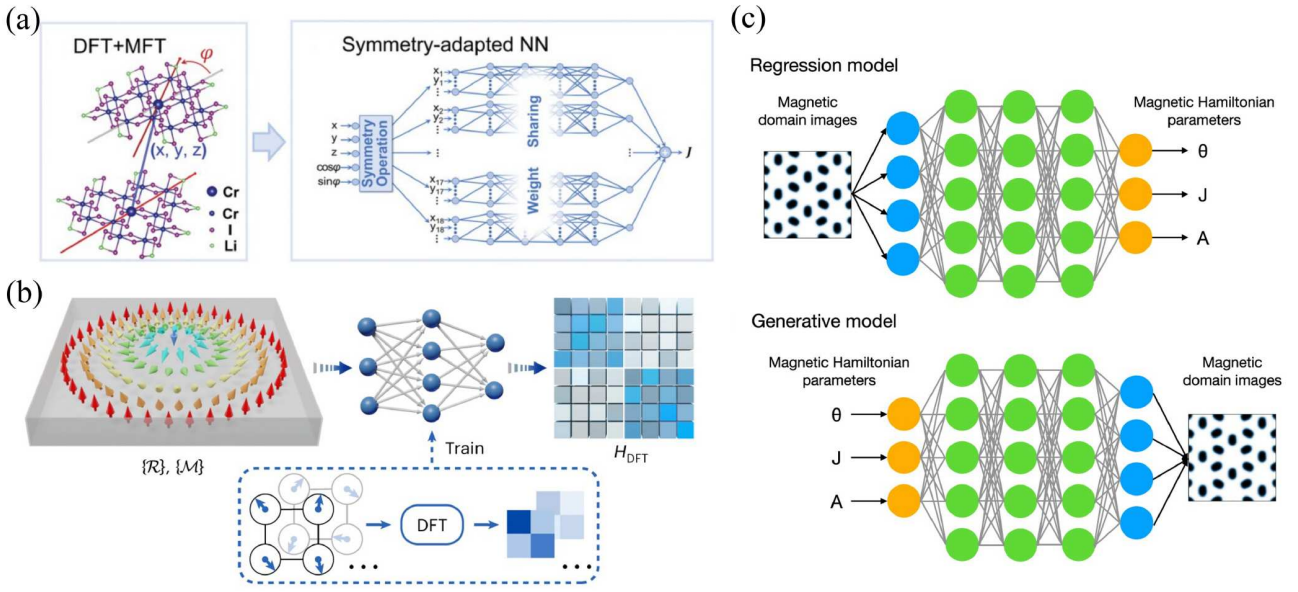

图 4 (网络版彩色)基于深度学习的双层磁体研究. (a) 锂离子封端的转角双层CrI_3团簇模型及SANN结构示意图[68]. Copyright © 2023 John Wiley & Sons. (b) xDeepH的工作流程: 等变神经网络将DFT哈密顿量(H_{DFT})表示为原子结构和磁结构的函数. 该模型在小尺寸结构(蓝色虚线框)的DFT数据上训练后, 可扩展应用于研究磁性超结构[82]. Copyright © 2023 Springer Nature. (c) 磁畴图像的深度学习模型示意图: 回归模型从磁畴图像中估算哈密顿量参数, 生成模型则基于给定参数预测磁畴图像[83]

Figure 4 (Color online) Deep learning research on bilayer magnets. (a) Li-terminated twisted bilayer CrI_3 cluster model and the structure of SANN[68]. Copyright © 2023 John Wiley & Sons. (b) Workflow of xDeepH: equivariant neural networks represent the DFT Hamiltonian (H_{DFT}) as a function of atomic structure and magnetic structure. The neural network models are trained on DFT data for small-size structures (blue dashed box) and applied to investigate magnetic superstructures[82]. Copyright © 2023 Springer Nature. (c) Schematic illustrating deep learning models for magnetic domain images: a regression model for estimating magnetic Hamiltonian parameters from input magnetic domain images and a generative model for producing predicted magnetic domain images based on input parameters[83]

既能通过回归方法从磁畴图像中估算哈密顿量参数(如转角、层内交换和单离子各向异性), 又能基于给定参数生成相应的磁畴图像. Yu等人[84]开发了自旋描述符用于构建复杂神经网络哈密顿量. 最近, Cai等人[85]提出的基于深度学习的NeuralMAG框架显著加速了微磁学模拟. 该方法采用U型神经网络计算退磁场, 通过编码器-解码器结构在多尺度上提取和累积局部自旋相互作用, 将计算复杂度从传统快速傅里叶变换方法的$O(N\log N)$降低到$O(N)$. 与其他神经网络方法不同, NeuralMAG专注于核心计算而非端到端近似, 展现出良好的通用性, 可广泛适用于不同尺寸、形状和材料参数的微磁学模拟. 此外, 多种考虑自旋自由度的机器学习原子间势函数[86~89]的发展, 为高效执行大规模自旋晶格动力学模拟奠定了基础.

3 总结与展望

二维范德华磁性材料因其独特的物理特性, 逐渐成为下一代自旋电子器件的理想候选材料, 为探索新奇量子态提供了理想的研究平台. 尤其是双层磁性材料体系, 通过层间耦合展现出高度可调的磁序及丰富的物理现象, 包括磁电耦合和斯格明子等. 本文系统地讨论了层间滑移对磁序、铁电性及磁电耦合的调控机制, 揭示了转角诱导的莫尔超晶格中非共线自旋态的丰富物理现象, 并重点介绍了高通量计算和深度学习在加速磁性材料堆叠设计中的创新应用.

双层范德华磁性材料凭借其丰富的堆叠自由度, 为实现多铁耦合提供了广阔的平台, 使同一材料体系中同时展现铁磁性、铁电性和铁谷等多种物理特性成为可能. 这类耦合现象引起了广泛关注, 为新型自旋电子器件的研发奠定了重要基础. 然而, 尽管二维磁性材料的研究较为深入, 铁电性的探索也在不断推进, 但在铁谷和拓扑等方面的研究仍需进一步加强. 此外, 目前已发现的二维多铁材料数量有限, 其物理性质及潜在应用前景仍处于初步探索阶段. 通过系统探索新型材料组合和堆叠方式, 有望实现更复杂的多铁耦合现象, 推动新型功能材料的研究与应用.

然而, 堆叠结构的多样性也为双层磁性材料的研究带来了巨大挑战. 传统的经验导向材料筛选方法在面对大量材料组合和堆叠构型时效率低下, 开发更高效的材料筛选工具成为当下需求. 建立系统完备的二维磁性堆叠数据库尤为关键, 这类数据库不仅需要涵盖多样化堆叠结构及其物理性质的全面信息, 还应具备灵活的数据更新与扩展能力, 以适应快速发展的研究需求. 在此基础上, 结合主动学习策略, 可以充分挖掘数据库中的已有计算结果, 通过智能识别和反馈机制优化研究路径, 快速筛选出最具潜力的材料组合. 高通量计算与机器学习的结合在揭示二维堆叠材料的构效关系中展现出巨大潜力[90,91]. 通过发展系统的数据库和可解释模型, 有望深入解析堆叠结构与物态之间的复杂关联, 这不仅能够为层间耦合机制提供新的物理视角, 还将为材料设计提供更加精准的理论指导. 此外, 深化深度学习在材料跨尺度模拟中的应用, 提升计算效率和预测准确性, 将为新型自旋电子器件的开发提供强有力的理论支持.

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Summary for “双层范德华磁性材料的理论研究进展”

Theoretical advances in bilayer van der Waals magnetic materials

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Two-dimensional (2D) magnetic materials have emerged as promising candidates for next-generation spintronic devices, owing to their distinct characteristics and potential for efficient low-power information processing. Since the successful exfoliation of graphene, extensive efforts have focused on realizing intrinsic magnetism in 2D materials. The experimental breakthrough occurred in 2017 with the discovery of intrinsic magnetism in monolayer CrI_3 and bilayer $Cr_2Ge_2Te_6$, leading to the synthesis of diverse 2D van der Waals (vdW) magnetic materials. These materials consist of atomically thin layers stacked together by weak vdW interactions. The weak interlayer coupling enables precise control over the relative crystal alignment between layers, resulting in diverse stacking configurations. Recent advances in synthesis methods and polymer-assisted transfer techniques have made on-demand stacking of vdW layers feasible, revealing novel physical phenomena such as stacking-dependent magnetic ordering and non-collinear spin textures in moiré superlattices. Moreover, sliding ferroelectricity, arising from polar stacking of nonpolar monolayers, provides alternative pathways for realizing magnetoelectric coupling, leading to novel correlated physics and device applications. These discoveries demonstrate that stacking order significantly influences the charge and spin redistribution between adjacent layers, thus affecting the topological properties, electronic correlations, and spin behaviors. Recent studies have focused on modulating the stacking order of 2D vdW magnetic materials, particularly through the manipulation of structural degrees of freedom such as interlayer sliding and twisting. These tunable variables provide a comprehensive platform for exploring and engineering novel quantum states in bilayer magnetic systems.

First-principles calculations based on fundamental quantum mechanical laws play a crucial role in materials simulation, particularly in predicting stacking configurations of bilayer magnetic materials through interlayer coupling analysis. The emergence of data-driven paradigms has transformed materials science research through the integration of high-throughput calculations and deep learning methods. High-throughput computational materials design enables efficient generation and analysis of extensive materials databases, overcoming traditional computational limitations. Simultaneously, advances in deep learning have enabled artificial neural networks to efficiently process large-scale datasets and establish structure-property relationships without requiring first-principles calculations. This approach, integrating first-principles calculations, high-throughput computation, and deep learning, significantly accelerates the discovery of novel bilayer magnetic materials with desired properties.

In this review, we first introduce the intriguing physical phenomena observed in bilayer vdW magnetic materials controlled through interlayer sliding and twisting, including stacking-dependent magnetism, magnetoelectric coupling, and the emergence of noncollinear spins in moiré superlattices. We then present recent progress in high-throughput computational design and deep learning applied to bilayer magnetic materials. Through these studies, we systematically describe the fundamental mechanisms of interlayer exchange coupling, stacking order modulated electronic and magnetic properties, and data-driven research in magnetic materials. Finally, we provide insights into future directions for material design and artificial intelligence-enabled research in 2D magnetic materials.

two-dimensional magnetic material, stacking order, magnetoelectric coupling, deep learning

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