

二维反铁磁材料的磁光研究进展

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摘要 磁性材料独有的磁响应特性增强了传统电荷型器件的磁控能力,而超薄甚至可单原子层剥离的二维磁性材料在低维尺度下增强了电子自旋与电荷、晶格的相互作用并带来了非凡的物性。在自旋电子学中,反铁磁材料不仅展现了高频、低耗、抗串扰的优势,其与超导、磁相变等现象联系的关联电子态还疏通了人们利用光电子学探索低维磁性及其机理的道路。近年来,得益于额外的磁自由度、二维光电效应、反铁磁与铁磁/非磁材料的功能化异质结,激光驱动的二维反铁磁材料成为研究热点。综合稳态和瞬态的显微光学手段,回顾总结了二维反铁磁材料中的各类磁光效应和元激发准粒子研究,包括激光与物质相互作用中的载流子、激子、声子、磁子及其耦合态效应。随着可见光到太赫兹成像和其他配套技术的发展,二维反铁磁材料的检测和调控难题正逐步得到解决,高极化度和低阻尼输运的微纳自旋应用有望被植入到光伏、光信息处理领域并发挥重要作用。

关键词 材料; 二维反铁磁体; 自旋; 磁光效应; 元激发准粒子

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1 引言

以电子电荷属性为基础的半导体微电子学是现代信息技术的基石,带来了小型机和超大规模集成电路等诸多革命性成就。然而,在半导体器件进一步微纳化的过程中,由于器件功耗的增加和量子效应等限制,摩尔定律不再适用,仅通过操纵和优化电子的电荷属性已经无法从本质上提升电子计算机的存储和运算能力。于是,人们将目光转向了新的电子自由度——自旋。电子的自旋性质与材料的原子、分子磁矩及集体的宏观磁性有紧密联系,21世纪以来,基于自旋操纵的磁电阻器件获得了巨大成功,自旋电子学继而催生了自旋阀^[1]、磁隧道结^[2]、自旋晶体管^[3]、自旋发光器件^[4]、自旋霍尔器件^[5]、自旋激光器^[6]等一系列具备非易失性和低焦耳热优势的前沿应用。

早在上千年前,物质的磁性就已经被观测并得到实际应用,如中国古代所使用的指南针,但由于自然界大多数材料的磁导率相对较低,磁应用高度受限于新型材料的发掘力度,近现代一些微纳磁读写和存储器件则对此提出了更高的要求。考虑到磁性材料的光学吸收长度一般在几十纳米量级,为保证调控和集成的便利性,在工艺允许的情况下,许多三维块体磁性材料的厚度已经降低到了准二维情形下的纳米量级。在更

极限的情况下,研究者们也一直在探寻类似石墨烯的具有原子极限厚度的 van der Waals 层状二维磁性材料。然而,长期以来受 Mermin-Wagner 自旋热涨落定理的约束,普遍认为当磁性材料的尺寸从三维降低到二维时,其块体状态下的本征磁性很难继续维持。尽管在自旋注入、离子插层和化学掺杂等外界因素辅助下的二维材料展现出了额外磁性^[7-9],直到 2017 年 Gong 等^[10]和 Huang 等^[11]相继发现了单层 Cr₂Ge₂Te₆ 和 CrI₃ 中的本征磁性,之后二维磁性材料才真正走入大众视野。这类二维磁性的存在根源于磁各向异性,即磁各向异性为自旋设置了一个首选方向,保护长程磁序不受自旋热涨落的影响。二维磁性材料的发现天然地为电子磁矩的对称性调谐和高效灵活控制提供了包括机械应变、外场调节、化学修饰和异质结组装在内的多种方案^[12]。这些磁调控手段也将为量子计算^[13]、低维超导^[14]、拓扑磁性^[15]带来全新的研究契机。

在磁性材料家族中,反铁磁(AFM)材料是一类特殊的存在,其内部虽然具有磁有序性,但因为相邻电子磁矩的反平行排列,对外不显示磁性^[16]。最初,由于铁磁/反铁磁堆叠界面间的交换偏置作用,反铁磁薄膜常被用作磁电阻存储器结构中的钉扎层。随后的研究发现,反铁磁材料具备丰富的物理特性和巨大的应用潜力:第一,反铁磁材料自旋进动频率共振在太赫兹波

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段,相对于铁磁材料而言,具备更大的自旋存储密度;第二,反铁磁材料的杂散场几乎为零,在抵抗外场干扰的同时展示出良好的自旋输运能力,契合于超紧凑的微型设备^[17];第三,反铁磁材料可以根据外界物理场(如温度、应力和磁场等)的变化引发特殊的磁相变,形成反铁磁-铁磁或反铁磁-亚铁磁等丰富的磁相,在特定体系下还会出现自旋液体及自旋玻璃态^[18-19],表现出磁电、磁力、磁热、磁光等多种物理特性^[20]。此外,相比铁磁材料,反铁磁材料可以更广泛地以绝缘体、金属、半金属、半导体或超导体的形式存在,并产生了大量实例,与强关联电子系统(莫特绝缘体、电荷转移型绝缘体)的联系还蕴藏着高温超导等有趣的物理机制^[14, 21]。特别地,在二维状态下,各类元激发准粒子及其耦合态是帮助人们表征和调控反铁磁磁性的重要研究对象,其中电荷-自旋-轨道-晶格自由度的耦合意味着外界物理场能够极大地影响材料的非平衡状态,产生许多尚未探索过的物质相^[22]。

区别于电学或磁学方法,基于磁光效应的全光探测方法利用了光电磁场与物质磁序和内部磁化场作用后光场参数会发生变化的特性^[23]。光的激发作用同样会影响材料的磁态,诱导的净磁化自旋流能够发挥逻辑处理和计算的作用^[24]。同时,光学方案具有显微、高速、非接触、高灵敏度、高时空和能量分辨的多种优势,并有利于观察反铁磁材料在极端物理条件下的各类磁响应^[25]。本文从光学表征和调控的角度出发对近年来二维反铁磁材料领域的研究进展进行综述。在第 2 节介绍了包括三卤化铬、过渡金属磷硫化物等典型反铁磁材料的基本结构、性质和分类方法,在第 3 节和第 4 节分别从宏观磁光效应和微观元激发准粒子的角度出发介绍了激光与二维反铁磁材料的相互作用^[16, 25-27]。随着应用能力的提升和应用场景的拓展,二维反铁磁材料为新型磁器件的构建和升级带来了更多可能。

2 典型反铁磁材料的结构、性质和分类

磁性材料中表现出的宏观磁矩通常源于过渡金属阳离子 3d 轨道电子的自旋和轨道角动量,其邻近交换作用是 Weiss 分子场形成的根本原因,使自旋力矩平行或反平行排列并形成长程磁序^[16],其交换积分是区分铁磁性和反铁磁性的关键因素^[28]。如图 1 所示,两个原子(i 和 j)之间的磁矩通常会产​​生交换能,可以用海森堡自旋哈密顿量(H)来描述,即 $H = -\sum_{i,j} JS_i S_j$,其中 S_i 和 S_j 是自旋算符或磁矩方向的单位矢量, J 是自旋 S_i 和 S_j 之间的交换积分,包含铁磁的直接交换、双交换和反铁磁的非相邻原子的超交换作用等。当材料具有很高的电导率,而趋近于金属性时,原子间的作用趋于直接交换作用,反之则为双交换或超交换作用^[29]。在具体材料中,广义力学量 H 还可能包括磁各向异性、自旋-轨道耦合和磁场作用能,并表现出一些特殊

的磁耦合作用,如 Dzyaloshinsky-Moriya (DM)、Kitaev 相互作用^[30]。当 $J > 0$ 时,相邻原子磁矩同向排列,磁矩倾向铁磁排布;当 $J < 0$ 时,相邻原子磁矩反向排列,磁矩倾向反铁磁排布^[31]。从微观角度出发,认为反铁磁体系中有一对相互抵消的自旋链,即两个自旋子晶格,磁化强度分别为 M_1 和 M_2 ,则磁化强度(M)和代表反铁磁性的 Néel 矢量(L)可以分别定义为 $M = M_1 + M_2$ 和 $L = M_1 - M_2$ ^[32]。在特殊情况下,DM 相互作用使两个反铁磁自旋子晶格呈较小的倾斜,产生了微弱的非共线分量和较弱的铁磁性,该状态称为倾斜型反铁磁态。这类属性的材料可通过与铁磁材料类似的探测方法进行表征^[33-34]。但对于通常意义上 $|L| = 0$ 的共线反铁磁材料,其内部两个自旋子晶格的反向磁矩互相抵消,从而不存在易被捕捉的净磁矩。按磁序的耦合方式,可将反铁磁材料分为层内反铁磁耦合和层间反铁磁耦合两类。更详细地,可以分为 A 型(层内原子为铁磁耦合,层间原子为反铁磁耦合)、C 型(层内原子为反铁磁耦合,层间原子为铁磁耦合)和 G 型(层内和层间原子均为反铁磁耦合)。A 型二维反铁磁材料主要包括三卤化铬 CrX_3 ($X = \text{Cl}, \text{I}$)^[11, 35-36]、过渡金属二卤化物 MX_2 ($M = \text{Fe}, \text{Co}, \text{Ni}; X = \text{Cl}, \text{Br}, \text{I}$)^[37]、过渡金属磷硫化物 CrPS_4 ^[38]、过渡金属铋碲化物 MnBi_2Te_4 ^[39]、过渡金属氧卤化物 CrSBr ^[40-41], C 型二维反铁磁材料以过渡金属磷硫化物 MPX_3 ($M = \text{Fe}, \text{Mn}, \text{Ni}, \text{Co}, \text{Cd}, \text{Zn}; X = \text{S}, \text{Se}$) 为代表^[42], G 型反铁磁材料则是以某些金属氧化物为主^[43]。

2.1 三卤化铬

常见的三卤化铬 CrX_3 包含 CrCl_3 、 CrBr_3 、 CrI_3 三种化合物,它们具有相同的电子和晶体结构,在低温下表现出磁有序性,且在单层极限下都能维持铁磁性,在常温下空间群为 $C2/m$ 单斜相,其少层材料在低温下转变为 $R3$ 菱方相^[44-45],如图 2(a) 和图 2(b) 所示。不同于 CrBr_3 的层间铁磁耦合,少层 CrI_3 和 CrCl_3 分别为 A 型面外和面内反铁磁耦合^[36]。 CrCl_3 单斜相的钉扎作用限制了原子间的磁交换作用,因而少层材料展现出更大的反铁磁耦合强度和反铁磁-铁磁转变磁场^[46]。当配位卤素原子从 Cl 到 Br 再到 I 时,不同 Cr 原子间距离的增加减弱了直接交换作用,此时配位体的电负性也逐渐降低,与 Cr 更倾向于共价键连接,加强了超交换和自旋-轨道耦合作用,影响了材料的磁各向异性并使磁有序温度提高^[47]。与层间反铁磁耦合基态共存的是, CrI_3 扭角堆叠的双层可以形成 Moiré 超晶格,从而形成由扭角调控的奇异磁态^[48-49]。以拓扑非平凡的 skyrmion 晶相为例,其稳定存在于小扭角堆叠的 CrI_3 和 CrBr_3 中已得到了理论证明^[50]。

2.2 过渡金属磷硫化物

块体 MPX_3 的磁性早在 19 世纪 90 年代就已经被发现,但直到近些年来,人们才通过机械剥离的方法

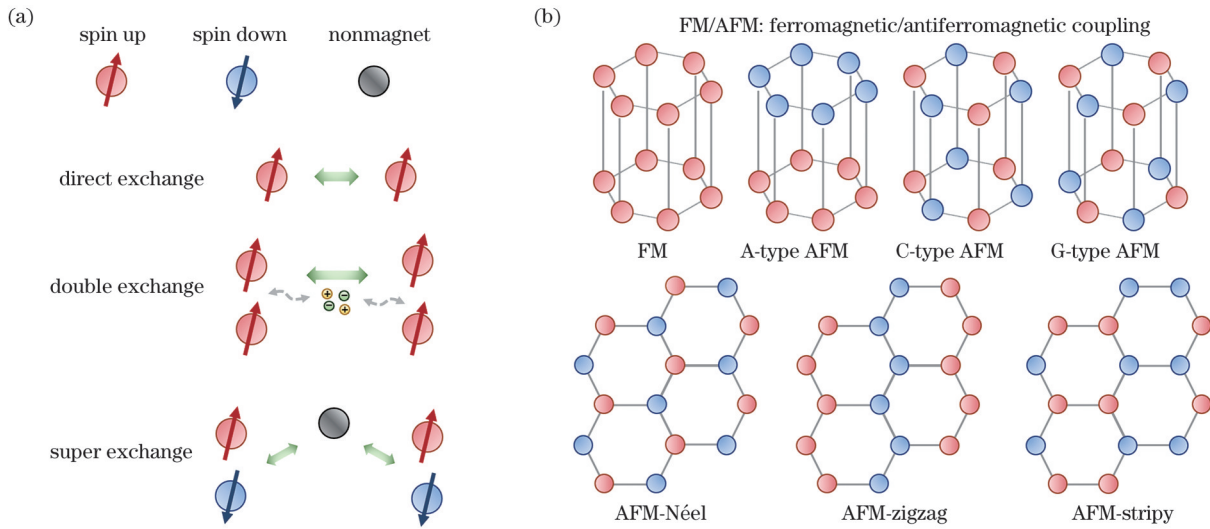


图 1 磁有序的形成和分类。(a)原子磁矩之间的直接交换作用、双交换作用、超交换作用示意图;(b)按照磁性原子层内/层间的铁磁/反铁磁耦合作用对磁性材料的分类

Fig. 1 Formation and classification of magnetic ordering. (a) Schematic of direct exchange, double exchange and super exchange between atomic magnetic moments; (b) classification of magnets according to ferromagnetic/antiferromagnetic coupling within/between magnetic atomic layers

获取 MPX_3 的二维少层结构并研究了其磁性^[51]。如图 2(c)和图 2(d)所示,少层和块体 MPX_3 分别表现为空间群为 $C2/m(X=S)$ 或 $R\bar{3}(X=Se)$ 的单斜相,而单层 MPX_3 中除了单斜相外,还可能附带不同的结构相^[42]。在单个晶胞内, $[P_2X_6]^{4-}$ 单元位于排列成蜂窝形状的 6 个 M 原子的中心,每个金属离子与 6 个硫族原子相连^[42]。原子组分的差异直接影响了层间距离 (d),其规律性与 M 原子的半径变化 (r) 一致,即 $d(MnPS_3) > d(FePS_3) > d(NiPS_3)$ 和 $r(Mn) > r(Fe) >$

$r(Ni)$ ^[42]。M 原子也强烈影响了材料的磁性、电子性质和层间耦合性质, $MnPS_3$ 、 $NiPS_3$ 和 $FePS_3$ 分别表现出海森堡型、XY 或 XXZ 型和伊辛型反铁磁性^[52]。由于相对较弱的层间耦合, $MnPS_3$ 也可近似为准二维伊辛型反铁磁体,具有面外各向异性,不同于 $NiPS_3$ 、 $MnPS_3$ 的面内各向异性^[16]。 MPX_3 的基态磁对称性由第一、第二和第三近邻交换作用的相对强度决定,可表现为锯齿型反铁磁 ($FePS_3$ 等) 和 Néel 型反铁磁性 ($MnPS_3$ 等)。

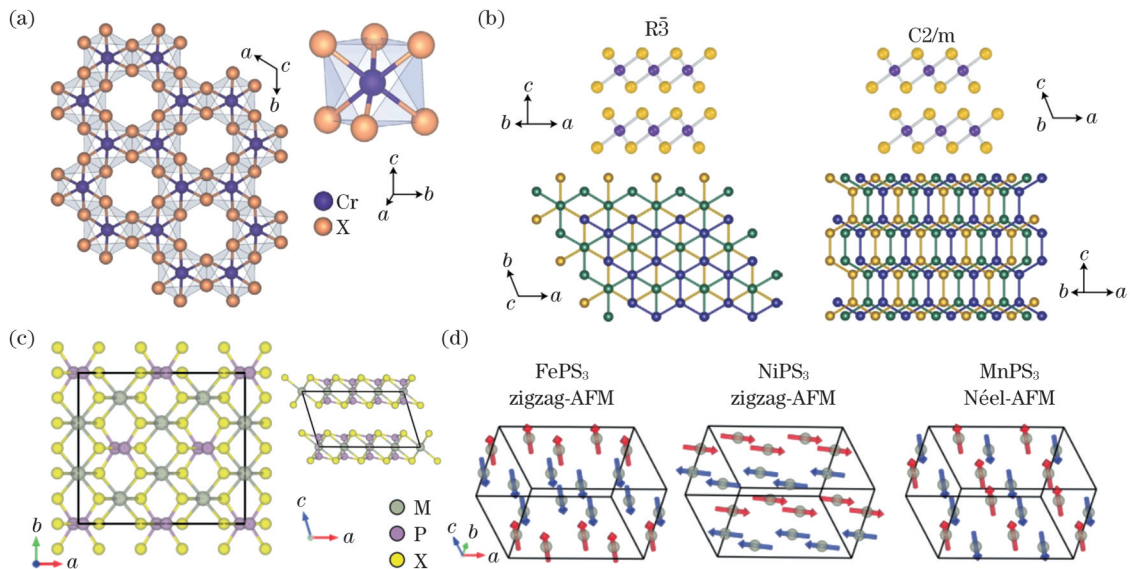


图 2 两类反铁磁材料的结构示意图。(a)三卤化铬 CrX_3 的八面体蜂窝构型^[35]; (b) CrX_3 菱形和单斜相^[44]; (c) 过渡金属磷硫化物 MPX_3 的六角蜂窝构型^[21]; (d) $FePS_3$ 、 $NiPS_3$ 、 $MnPS_3$ 的基态磁结构^[21]

Fig. 2 Structural diagrams of two types of antiferromagnetic materials. (a) Octahedral honeycomb configuration of chromium trihalide CrX_3 ^[35]; (b) rhombic and monoclinic phases of CrX_3 ^[44]; (c) hexagonal honeycomb configuration of transition metal phosphorus sulfur compound MPX_3 ^[21]; (d) ground-state magnetic structures of $FePS_3$, $NiPS_3$, and $MnPS_3$ ^[21]

2.3 其他反铁磁材料

除了上述两类,另一些二维反铁磁材料也成为了学界的新宠。最近,过渡金氧卤化物 CrSBr 因层间反铁磁耦合、三轴磁各向异性、可见到近红外带隙、激子(束缚电子-空穴对)荧光等特性而受到了广泛关注^[40-41, 53]。CrOCl 则拥有独特的 stripy 型面内反铁磁构型和磁弹性耦合性质,允许通过电场或应力作用对磁结构进行操纵^[54]。Zeng 等^[55]通过改进的化学气相传输法可控合成了高质量厘米级 FeOCl 单晶,通过机械剥离得到的二维 FeOCl 表现出强烈的面内光学和电学各向异性。Kulish 等^[56]预测了一系列多样的二维过渡金属二卤化物,在单层情况下,FeX₂、CoX₂和 NiX₂更偏向铁磁耦合基态,而 VX₂、CrX₂和 MnX₂更偏向于反铁磁基态,这种区分取决于近邻原子 d-d 轨道电子的直接交换和卤素原子 p 轨道介导的超交换作用之间的竞争。由于平面内铁磁和反铁磁态的共存, NiI₂ 等多层磁性材料中每个单层内相邻原子的自旋指向以螺旋形式排列,形成螺旋磁态^[37]。三卤化钒化合物 VX₃(X=Cl, Br, I)与三卤化铬在结构上有许多相似性,以 VI₃ 为例,该材料在经历临界温度以下的结构相变后表现为铁磁性^[57-58],而 VBr₃ 则为本征的反铁磁性^[59]。此结构中每个 V 原子都有两个未配对的 d

电子,使得该类材料非常容易通过掺杂、应力等手段实现铁磁-反铁磁相的切换^[60]。另一类三卤化物 RuCl₃ 在略高于 Néel 温度的状态下可以形成独特的 Kitaev 自旋液体态^[61-62],其反常光谱响应揭示了电荷-自旋之间的耦合作用和激子效应的存在^[63]。此外,有理论研究预测了单层 RuCl₃ 中光对磁有序性的有效调节,光生自由载流子和激子可以使该反铁磁材料相变为稳定的铁磁相^[64]。在拓扑磁性方面, A 型反铁磁 MnBi₂Te₄ 材料具有 0.2 eV 的本征带隙,当层间反铁磁态被外磁场完全极化为铁磁态时,会发生拓扑量子相变^[65]。除了 van der Waals 层状材料外, NiO、Cr₂O₃ 等过渡金属氧化物^[66-68]和稀土正铁氧体 DyFeO₃、TmFeO₃ 等^[69-70]是较早被研究的块体反铁磁材料。在另一类锰族化合物 Mn₃X(X=Sn, Ge)非共线反铁磁材料中, Mn 离子在晶格中以 120° 的逆三角序形成 Kagome 构型,这种几何阻挫自旋系统不仅表现出非常大的反常霍尔效应,还能够带来相当大的可调磁光效应^[71]。上述一些典型二维层状反铁磁材料及基本性质的总结如表 1 所示,其中 1L 表示单层, 2L 表示双层。A 型反铁磁材料的单层具有本征的铁磁性,其磁相变温度称为居里温度,不同于反铁磁材料的 Néel 温度。

表 1 典型的二维层状反铁磁材料及基本性质

Table 1 Typical two-dimensional layered antiferromagnets and basic properties

AFM coupling type	Chemical type	Material	Symmetry	Bandgap /eV	Magnetic orientation	Critical temperature /K	Ref.
Interlayer A-type	Halide	CrI ₃	Monoclinic C2/m	1.5	Vertical	45 (1L, 2L), 61 (bulk)	[11, 35]
		CrCl ₃	Monoclinic C2/m	3.0	Parallel	10 (1L), 17 (bulk)	[36]
	Chalcogenide	CrSBr	Orthorhombic Pmmn	1.8 (1L), 1.5 (bulk)	Parallel	132 (2L), 150 (bulk)	[38, 72]
		CrPS ₄	Monoclinic C2/m	2.28 (1L), 1.4 (bulk)	Vertical	50 (1L), 36 (bulk)	[39]
		MnBi ₂ Te ₄	Trigonal R3/m	0.2	Vertical	20 (few-layer), 25 (bulk)	[40-41]
Intralayer C/G-type	Chalcogenide	NiPS ₃	Monoclinic C2/m AFM-zigzag	1.6	Parallel	130 (2L), 150 (bulk)	[42, 73]
		FePS ₃		1.5	Vertical	117	[21, 74]
	Halide	MnPS ₃	Monoclinic C2/m AFM-Néel	3.0	Vertical	78 (bulk)	[42, 75]
		MnPSe ₃	Trigonal R3 AFM-Néel	2.5	Parallel	70 (bulk)	[42]
		RuCl ₃	Monoclinic C2/m AFM-zigzag	1.0-1.9	Parallel	15	[61-62]
	CrOCl	Orthorhombic Pmmn AFM-stripy	2.38 (1L), 1.41 (bulk)	Vertical	160 (1L), 14 (bulk)	[54, 76]	

3 二维反铁磁材料的磁光效应

二维反铁磁材料的工程化调控取决于材料的结

构、磁相和能带性质,这些属性可通过各类磁光效应,以光场强度、能量、相位、偏振等参数改变的形式来体现。磁光效应源于材料内部电荷-自旋耦合、能带的交

换分裂和自旋轨道耦合之间的相互作用^[77],不仅可用于磁序检测,还可用于光学调制和数据存取等多个方面^[78]。除了广为人知的法拉第效应、磁光克尔效应(MOKE)、塞曼效应、磁二向色性、Voigt 和 Cotton-Mouton 效应以外,磁光现象还包括光与物质相互作用导致物质磁性改变而引起的各种光学现象。如图 3 所示,按照光的作用机理和收集方式,反铁磁材料中的磁

光现象包括:磁矩诱导的旋光效应,晶格结构和磁序引起的非弹性散射^[79],自旋-晶格对称性相关的非线性光响应^[80],自旋-电荷耦合相关的光致发光^[35],受激振荡的自旋共振发射^[81],偏振脉冲光引发的逆磁光效应等^[23]。通过这些磁光特性可以对二维反铁磁材料进行精细表征,甚至进一步记录外场调控下内部磁畴的变化。

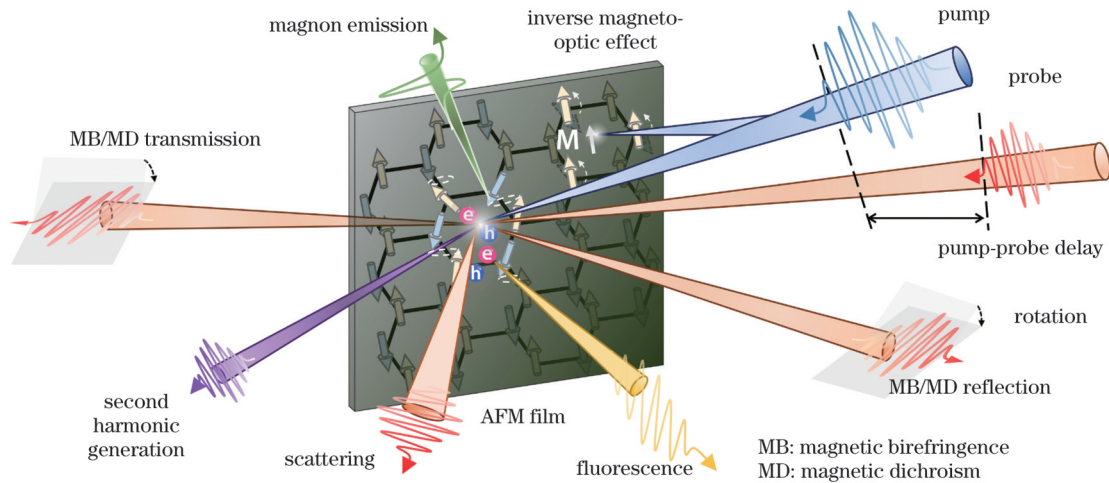


图 3 反铁磁薄膜中的各类磁光现象

Fig. 3 Multiple magneto-optical phenomena in antiferromagnetic thin film

3.1 稳态磁光效应

对于块状磁性材料,利用常规的振动磁强计、中子散射、磁共振和微波技术能够探测到磁序,并成像不同区域样品的磁畴属性,利用反常霍尔和隧穿磁阻效应等电学运输的方式也能够较为全面地演示材料的磁化方向^[82]。但对于以机械剥离方式获得的二维层状磁性材料来说,由于微米量级的小尺寸和灵敏度的缺乏,微观磁序很难通过这些非局域的检测手段直接确定。X 射线光谱是适合表征反铁磁序的有效工具之一,X 射线的光子能量与过渡金属原子内壳层 p 电子到外层 d 电子的跃迁能量共振,利用 p-d 电子跃迁所引起的光谱偏振态变化,就能够判断出样品微观磁序的排列状态。然而,X 射线光谱的实验条件非常苛刻,与中子散射方法类似,需要用到大型同步辐射装置^[83],且难以实现理想的时间分辨。

可见光到太赫兹波段的光学检测方法是对上述工具的有力补充。1845 年,磁光法拉第效应被发现^[84],透过磁性材料的光的偏振状态因外加磁场而表现出单向旋光特性,这使得法拉第效应被广泛地应用于光隔离器等非互易器件中。随后,1876 年,MOKE 被观测到^[23],经磁铁表面反射的光的偏振状态也出现旋转。根据磁化强度与材料表面和光入射面的相对关系,MOKE 具有极向、纵向和横向三种配置。人们利用 MOKE 能够直接观测材料的磁序、磁畴结构、磁相变行为、磁各向异性和自旋态密度等一系列特征。从光的

传播角度看,如图 4(a)所示,在各向同性铁磁材料中,当外部光激励改变了介电常数时,材料对左旋和右旋圆偏振光响应的简并性会被打破,手性相关的反射、吸收振幅差引起了椭偏率的变化,相位差引起了偏振面的旋转,这两种偏振变化分别对应介电张量实部和虚部的响应^[85],最终表现出磁圆双折射(MCB)或磁圆二向色效应(MCD)^[86]。由于偏振变化量正比于铁磁材料的磁化强度,这两种磁圆效应也称为线性磁光效应。但事实上,对于内部净磁矩完全补偿为零的共线反铁磁材料而言,时间和空间反演对称性都得到了保留,材料对圆偏振光产生的线性磁光效应可以忽略,即较难通过上述线性磁光效应的探测直接确定磁序^[28]。此时,外部激励引发的相对介电常数变化与磁化强度的平方成正比,可以考虑材料对线偏振光产生的二阶磁光效应,即常见的磁线双折射(MLB,又称为 Voigt 或 Cotton-Mouton)效应或磁线二向色(MLD)效应^[87]。相对于线性磁光效应,二阶磁光效应的成因涉及到晶体和磁结构对称性等多个方面,且强度要弱得多^[83]。

磁序为材料在晶格层面上的对称性提供了额外的自由度,可利用对称性破缺的性质来解决上述磁光耦合效率不足的问题。如图 4(b)~(e)所示,Zhang 等^[21]基于锯齿型反铁磁材料磁对称性的破缺,在双层 FePS₃ 和多层 NiPS₃ 中分别发现了巨 MLD 效应,显著高于其他二维反铁磁材料。具体地,反铁磁序对材料旋转对称性的破坏引发了电子能带的各向异性,使得电子更容易沿晶格的锯齿轴

而不是自旋方向激发,区别于依赖样品磁化指向的 Voigt 效应。除了材料的本征二向色性外,通过对光学微腔的利用,FePS₃中的自旋-电荷耦合强度得到了显著调控,达到了接近 1 的线二向色度,并且利用

微腔的尺寸能够调谐 MLD 的光谱特性^[88]。类似的还有各向异性的反铁磁半导体 CrSBr 和绝缘体 CrOCl,它们都具有低对称度的正交结构,在低温的磁基态下表现出高度的偏振敏感特性和 MLD^[89-90]。

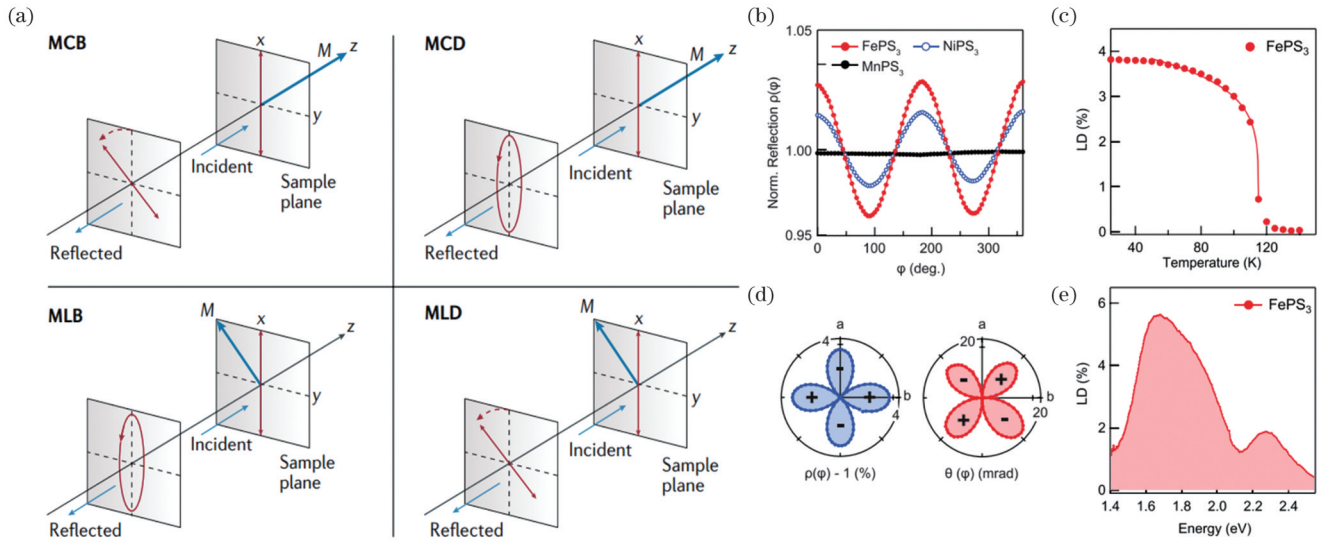


图 4 磁光效应中的双折射和二向色性。(a)磁圆双折射(MCB)、磁圆二向色性(MCD)、磁线双折射(MLB)、磁线二向色性(MLD)效应对反射光偏振的影响^[28];(b)FePS₃、NiPS₃和 MnPS₃的MLD对比^[21];(c)FePS₃中MLD的温度相关性^[21];(d)反射光的各向异性和偏振旋转^[21];(e)MLD的光谱响应^[21]

Fig. 4 Birefringence and dichroism in magneto optic effect. (a) Influence of magnetic circular birefringence (MCB), magnetic circular dichroism (MCD), magnetic linear birefringence (MLB), and magnetic linear dichroism (MLD) on polarization of reflected light^[28]; (b) MLD comparison of FePS₃, NiPS₃, and MnPS₃^[21]; (c) temperature dependence of MLD in FePS₃^[21]; (d) anisotropy and polarization rotation of reflected light^[21]; (e) spectral response of MLD^[21]

除了引起偏振改变的磁光效应外,强激光的照射还能诱导材料内部电场极化或电流的产生,结合反铁磁材料的磁态发生光电流、二次谐波、磁光散射等非线性过程^[80,91-92]。这些效应经常与电子系统的玻色子激发产生联系^[93],在常规磁光效应很弱甚至消失的情况下,其对反铁磁材料的激发态物理研究以及相关器件应用非常重要。反铁磁序在打破材料空间反演对称性的同时可保留时空组合反演对称性。Zhang等^[91]通过理论计算揭示了双层CrI₃中线偏振光激发的磁光电流效应,能带中的共振光学跃迁产生光电导,通过对磁相变的控制,光电流的方向和幅值能够被显著调节。如图5所示,Song等^[94]报道了石墨烯/多层CrI₃异质结中的自旋光伏效应,多层CrI₃的层间磁序直接影响了异质结在垂直方向上的光电流和隧穿电流的大小,光电流的圆偏振度还显示出对磁态和光子能量的相关性,可以通过磁场扫描完成调控,并证明了磁光电流作为磁序探测工具的有效性,类似地,电荷与磁场的强相关性还能影响光电流响应的频率相关性和偏振手性^[95]。除此之外,MnPS₃、FePS₃等二维反铁磁材料还展现出了优异的光电响应,非常适合于高灵敏度的光电探测,覆盖波段从紫外延伸到近红外^[96-97]。在非线性光学方面,如图6(a)和图6(b)所示,不同于FePS₃和NiPS₃的锯齿形磁序,MnPS₃的

Néel型反铁磁序同时打破了时间和空间反演对称性,使之能直接通过二次谐波进行成像^[80],通过提取偏振分辨的二次谐波可以将其与仅由晶格反演对称性破缺带来的信号区分开,并清晰地观测到多层MnPS₃中的长程反铁磁序和单层样品中磁序的缺失^[98]。在A型反铁磁双层CrI₃中,层间反铁磁耦合同时打破了时间和空间反演对称性,产生了较大的二次谐波,其偏振相关性进一步揭示了单斜堆积顺序和层状反铁磁性的微观起源^[99],类似的还有由CrSBr和CrOCl低对称性磁结构带来的高偏振度二次谐波发射^[89-90]。拉曼光谱表征的准弹性散射是研究声子、磁子的常用工具^[79],如图6(c)和图6(d)所示,NiPS₃在低温下被发现存在二维XXZ到XY型反铁磁的磁相变,其反铁磁序可存在于双层以上样品,但在单层中被显著抑制^[73]。偏振分辨的拉曼光谱也被用于研究反铁磁体MnPS₃和CrCl₃等材料的各向异性拉曼模式^[44,92]。Wang等^[100]通过电子拉曼散射检测了少量NiPS₃中对应于d-d轨道的跃迁吸收并研究了其中的光子散射模式,自旋和d电子之间的关联证明了磁各向异性的起源。拉曼光谱还与时间分辨的MOKE结合,用于FePS₃和MnPS₃的热导率测试^[101]。相对于MOKE等测试手段来说,基于拉曼光谱的方法缺乏对微观磁序的直接观察,但其对反铁磁材料的

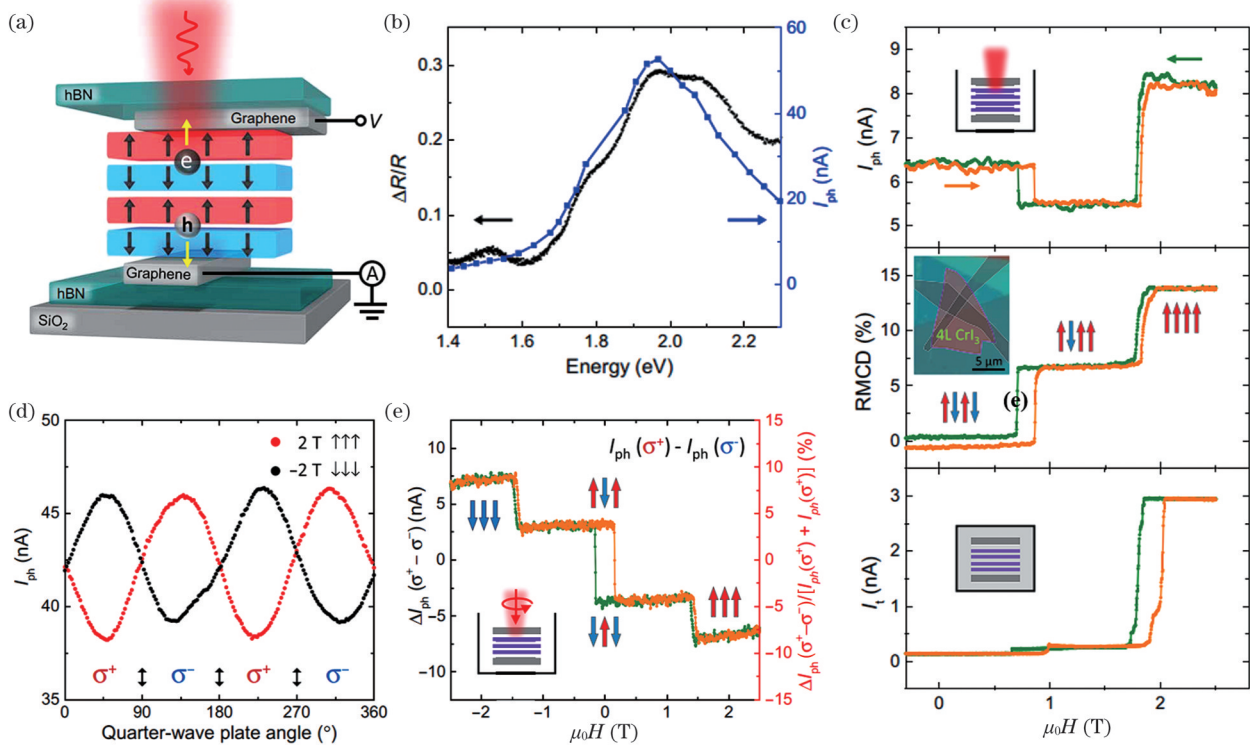


图 5 CrI₃中的磁光电流效应。(a)四层CrI₃-石墨烯器件示意图^[94];(b)CrI₃的差分反射谱和光电流的谱响应^[94];(c)层间磁序(中)对光电流(上)和隧穿电流(下)的调控作用^[94];(d)三层CrI₃中光电流的圆偏振响应特性^[94];(e)铁磁/反铁磁态对光电流圆偏振度的调控^[94]

Fig. 5 Magnetic photocurrent effect in CrI₃. (a) Schematic of four-layer CrI₃ graphene device^[94]; (b) differential reflection spectrum of CrI₃ and spectral response of photocurrent^[94]; (c) regulation of interlayer magnetic order (middle) on photocurrent (above) and tunneling current (below)^[94]; (d) circular polarization response characteristics of photocurrent in trilayer CrI₃^[94]; (e) regulation of circular-polarized degree of photocurrent by ferromagnetic/antiferromagnetic states^[94]

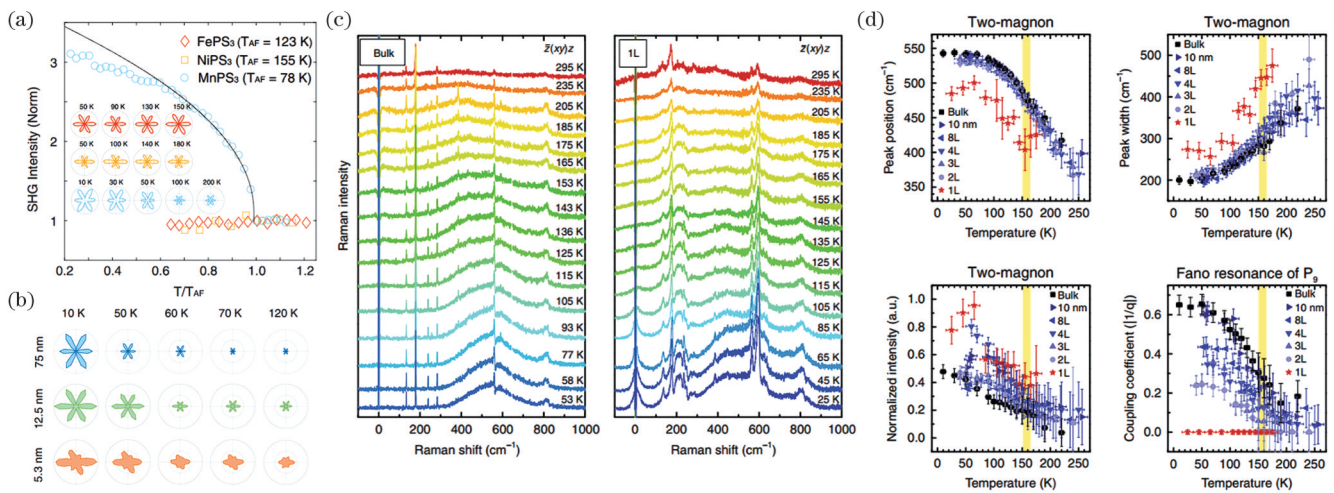


图 6 MPX₃的磁光二次谐波和拉曼光谱。(a)FePS₃、NiPS₃、MnPS₃中二次谐波发射的温度相关性^[80];(b)不同厚度MnPS₃二次谐波发射的温度和偏振相关性^[80];(c)块体和单层NiPS₃的拉曼散射光谱^[73];(d)温度和样品厚度对磁振荡的影响^[73]

Fig. 6 Magneto-optic second harmonic and Raman spectra of MPX₃. (a) Temperature dependences of second harmonic emissions in FePS₃, NiPS₃ and MnPS₃^[80]; (b) temperature and polarization dependences of second harmonic emissions in MnPS₃ with different thicknesses^[80]; (c) Raman scattering spectra of bulk and monolayer NiPS₃^[73]; (d) effects of temperature and sample thickness on magnetic oscillation^[73]

基本磁激发、层间磁耦合和零磁场上临界温度附近的磁态等信息具有更简易直观的描述能力。

3.2 反铁磁序的调控及探测

对于反铁磁材料的磁性操控,常见的手段包括磁

场、电场(包括自旋矩)、温度场、应力调控(磁各向异性或磁结构相变)、被动调控(离子掺杂、界面工程等)和光场(包括热激发和自旋共振)六个方面^[102-103]。依赖磁光效应开展的磁序探测与调控过程相辅相成,共

同促进了反铁磁材料的应用研究。

基于磁圆二向色性测试, Song 等^[104]测试了外磁场对 2~4 层反铁磁 CrI₃ 的调控作用, 当垂直磁场逐渐增大时, 双层 CrI₃ 逐渐变为层间铁磁耦合(用 ↑↑ 表示), 3 层和 4 层 CrI₃ 还会出现未完全抵消的部分反铁磁态(如 ↑↑↓ 和 ↑↑↑↓ 等)。自旋极化电流或自旋霍尔效应相关的自旋转矩允许通过电学方法读写反铁磁材料的磁态, 这对于层内强反铁磁耦合的材料来说非常重要^[17]。而对于层间弱耦合的 A 型反铁磁材料来说, 通过静电场就能够旋转子晶格的自旋方向, 从而实现了对磁性的电控制, 这是通过改变材料内部的电极化方向实现的, 相对于外加磁场的调控方式来说具有效益方面的优势^[93]。如图 7(a)~(c) 所示, Huang 等^[105]利用静电场调控了双层 CrI₃ 中的磁性, 在磁相变附近的恒定磁场下, 反铁磁和铁磁状态的切换可以完全由电压控制, 同时临界磁场也受到电压的调控。此外, 利用 MOKE 显微镜, 调谐过程中 CrI₃ 的磁态能够被精细成像。Song 等^[106]使用隧穿电流和扫描 MCD 显微镜测量演示了应力调控对二维 CrI₃ 磁序的影响; 在 3 层 CrI₃ 中, 压力改变了层间磁序的排列, 使两个反铁磁和一个铁磁态三态共存, 这为设计二维磁相和功能化器件提供了充分的可能。类似的压力调控工作也通过 CrI₃ 的

偏振和光谱响应得到了验证^[78,107]。在双层 CrI₃ 中, León 等^[108]发现应力的拉伸使得材料的反铁磁态更加稳定, 而应力的压缩则使得材料转为铁磁态, 其内在因素来自层间诱导位移所带来的磁序变化。如图 7(d) 和图 7(e) 所示, 结合偏振分辨的二次谐波成像, Ni 等^[109]检测了 XY 型反铁磁 MnPS₃ 中由应力控制的面内/面外 Néel 矢量方向的切换, 相对于热调控手段, 应力调控的 Néel 矢量旋转不受晶格对称性限制。在磁有序温度以下, 应力会导致 NiPS₃ 和 FePS₃ 的绝缘体-金属相变^[110-111], 在抑制长程磁有序的同时形成了可室温存在的短程磁有序, 而压力驱动甚至导致了 FePS₃ 的低温超导性^[112]。在被动调控方面, 通过将各种有机阳离子插入层状材料的 van der Waals 间隙, 可以在不明显引入缺陷和杂质的情况下有效地调节电子掺杂浓度, 并因此改变磁序, 如 NiPS₃^[113], 其在较低浓度下显示出反铁磁到铁磁的转变, 继续提高掺杂浓度则使得材料回归反铁磁态。Ma 等^[114]通过在 MoSSe 上沉积 Cr 原子, 合成了独特的单原子层 MoCrSSe 反铁磁材料, 其反铁磁基态的电子带隙约为 0.4 eV, 适用于红外光电探测, 当转变为铁磁状态后, 材料会发生导电的半导体-金属相变, 具有磁开关应用的潜力。

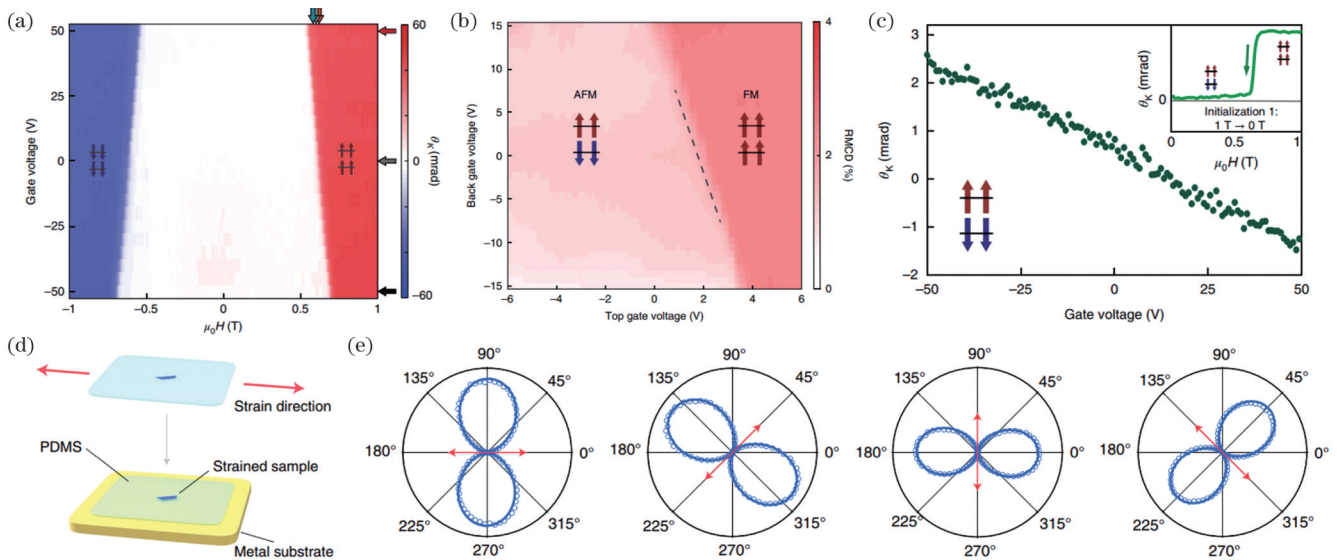


图 7 反铁磁序的电学和压力调控。(a) CrI₃ 磁态的电场和磁场调控, 由 MOKE 信号体现^[105]; (b) 稳态磁场下通过背栅和顶栅电压控制的铁磁/反铁磁态, 由 MCD 信号体现^[105]; (c) 零磁场下的 MOKE 信号与栅极电压的线性相关性^[105]; (d) MnPS₃ 的面内应力控制示意图^[109]; (e) 应力方向控制的 Néel 矢量, 由偏振分辨的二次谐波信号体现^[109]

Fig. 7 Electrical and pressure regulations of antiferromagnetic order. (a) Electric and magnetic field regulations of CrI₃ magnetic state reflected by MOKE signal^[105]; (b) ferromagnetic/antiferromagnetic state regulated by back and top gate voltages reflected by MCD signal under steady-state magnetic field^[105]; (c) linear correlation between MOKE signal and gate voltage under zero magnetic field^[105]; (d) schematic of in-plane stress control of MnPS₃^[109]; (e) Néel vector orientated by stress direction, reflected by polarization-resolved second harmonic signal^[109]

铁磁-反铁磁异质界面的交换偏置也是一种磁场调控方式。Zhu 等^[115]报道了机械剥离 CrCl₃/Fe₃GeTe₂ 异质结在低温下超过 50 mT 的交换偏置场, 并且可以通过温度和异质结厚度进行调控。类似地,

FePS₃/Fe₃GeTe₂ 的异质结界面则改变了 Fe₃GeTe₂ 表面的自旋结构, 使 Fe₃GeTe₂ 的矫顽场提高了一倍, 居里温度也得到了 30 K 以上的提升^[116], 这种临界温度的提升有利于实现二维磁性的常温化应用^[117]。在

$\text{Fe}_3\text{GeTe}_2/\text{MnPS}_3$ 异质结中,通过 MOKE 和 MCD 也观察到了矫顽场的增强、居里温度的提高和较大的交换偏置效应^[118]。异质结中不同材料的生长或堆叠顺序会影响离子之间的磁交换作用,影响界面处的磁有序^[119]。在单层 WSe_2 /少层 CrI_3 异质结堆叠结构中,利用少层 CrI_3 各层磁化可精确调控单层 WSe_2 的能谷激子特性,利用这种磁近邻效应的关联作用,Zhong 等^[120]通过探测 WSe_2 激子荧光的圆偏振性,对 CrI_3 的铁磁和反铁磁畴进行了二维成像。反过来可以进一步通过磁场调控和其他手段(如电场、绝缘插层)调控激子荧光的圆偏振性^[121-122]。另一方面,反铁磁内部磁序的改变会诱导层间激子-磁子耦合,在 $\text{MoSe}_2/\text{MnPS}_3$ 异质结中,当温度维持在 Néel 温度以下时, MoSe_2 的激子发光峰会因为层间耦合作用而蓝移,并且可以通过磁序和堆叠角度来控制这种层间耦合的强度^[123]。除了层内激子之外, VI_3/CrI_3 异质结中的层间激子也被报道具有磁调控特征,在具有超过 20 meV 的能谷劈裂的同时具备较长的能谷寿命^[124]。在理论方面,Yang 等^[125]利用第一性原理计算了双层 CrI_3 和 $\text{CrBr}_3/\text{CrI}_3$ 异质结在层间铁磁/反铁磁耦合下的 MOKE 响应,在不同铁磁/反铁磁配置情况下,利用层间自旋态的反转能够切换 MOKE 角的转角方向,这种基于对称性分析的结论适用于更一般的二维磁异质结。

3.3 超快激光的控磁作用

将磁电子器件嵌入已建立的半导体电子学中的一个关键需求是以尽可能高的运行速度实现自旋信号与电荷信号的相互切换。与非磁材料的研究类似,光与磁性材料的相互作用也发生在超短的时间尺度内,超短激光脉冲可以通过驱动电子或热激发的方式实现对磁化状态的控制和探测。与连续激光的作用不同,飞秒脉冲激光由于其固有的高功率和短持续时间特性,能使材料的磁性在超短的时间尺度内发生巨大改变,产生瞬时的线性和非线性磁光效应,因而飞秒脉冲诱导的自旋动力学成为了研究无静态磁性的反铁磁材料的强有力工具。更重要的是,原子磁矩的角动量转移效率决定了自旋操纵的速度^[126],反铁磁序的变化可以直接通过自旋子系统内部角动量的重新分布来实现,不需要将多余角动量转移到其他子系统(如晶格),避免了额外的延迟和损耗,从而在速度和效率上优于铁磁材料,在磁存储和接近光频率的快速计算方面具有很大潜力^[86]。

在块体磁性材料研究中,由脉冲激光引起的退磁过程最早于 1996 年在铁磁金属镍薄膜中发现^[128],飞秒脉冲使得铁磁序快速淬灭,电子、自旋和晶格在时间弛豫动力学中依次占据主导作用。经研究,飞秒激光与铁磁材料的作用主要包含飞秒尺度下的超

快退磁、皮秒尺度下的磁化恢复和纳秒尺度下的自旋进动三个阶段。研究者们通过在传统电子-晶格双温模型中加入自旋温度,构建了唯象的电子-自旋-晶格三温模型。脉冲光激发后,电子、自旋和声子系统先后升温,能量在不同子系统间传输,三个子系统温度的不匹配及产生的能量交换扰动了磁有序性^[128-129]。但即便如此,飞秒激光退磁的机制,包括电子-声子散射、电子-磁子散射、库仑交换散射、电磁耦合和超扩散自旋输运等因素的主次之分一直存在争议^[130]。超快激光可以同时带来相干和非相干作用^[131],其中某些特殊的机制,如惯性驱动的自旋开关、完全相干无损的自旋波激发等,仅在反铁磁材料中得到验证^[25],而更一般意义上的非相干热诱导退磁机制可使材料的本征净磁化强度迅速淬灭,同时磁各向异性也可能快速变化,宏观表现为自旋重定向^[132]及超快磁相变^[66-67]等。另一方面,逆磁光效应(逆法拉第效应或逆 Cotton-Mouton 效应)也是典型的非热相干过程^[25, 68],这是由于反铁磁材料净磁矩的激发是在无光吸收的情况下完成的,利用脉冲偏振光激发产生的磁场脉冲可以实现对自旋方向的定向控制。

超快激光能显著改变反铁磁材料的磁光响应。如图 8(a)~(c)所示,Zhao 等^[71]在 40 nm 厚的 Mn_3Sn 薄膜中发现了超快激光调制的巨大 Voigt 效应,其反射光偏振变化比瞬态 MOKE 转角大一个数量级以上,也远大于晶体结构和应力因素等引起的变化;同时,该材料具有在高于常温的条件下完成自旋操纵的优势。在 CuMnAs 薄膜中,超短光脉冲诱导的自旋重定向大大提升了电阻率变化程度,能够比拟传统铁磁多层膜中的巨磁阻效应^[133],且能通过光学手段进行时间分辨的实时检测。除了材料的内部激发外,外部传感同样能达到控磁效果。Schlauderer 等^[70]通过外置天线的强太赫兹脉冲耦合改变了反铁磁 TmFeO_3 薄膜中的磁各向异性,在满足势垒能量的情况下实现了相干自旋开关。与反铁磁氧化物中激光触发的绝缘-金属态超快切换类似^[67],Belvin 等^[81]通过时域太赫兹光谱的电导率监测发现了 NiPS_3 中由超快激光控制的瞬时反铁磁金属态,如图 8(d)~(f)所示,具体表现为激光产生的光生激子分解为可迁移的载流子,电导率增加的同时伴随着反铁磁磁矩的自旋进动,其优势在于 NiPS_3 高相干度激子解离产生的自由载流子是未热化的,不会因为声子-自旋这一弛豫通道而扰动长程磁序。Zhang 等^[134]探测了超快激光在块体 FePS_3 中诱导的退磁过程,其动力学寿命与体系温度呈强相关性,并在 Néel 温度附近表现出发散行为,揭示了在飞秒时间尺度上的相干操纵磁各向异性的方法。

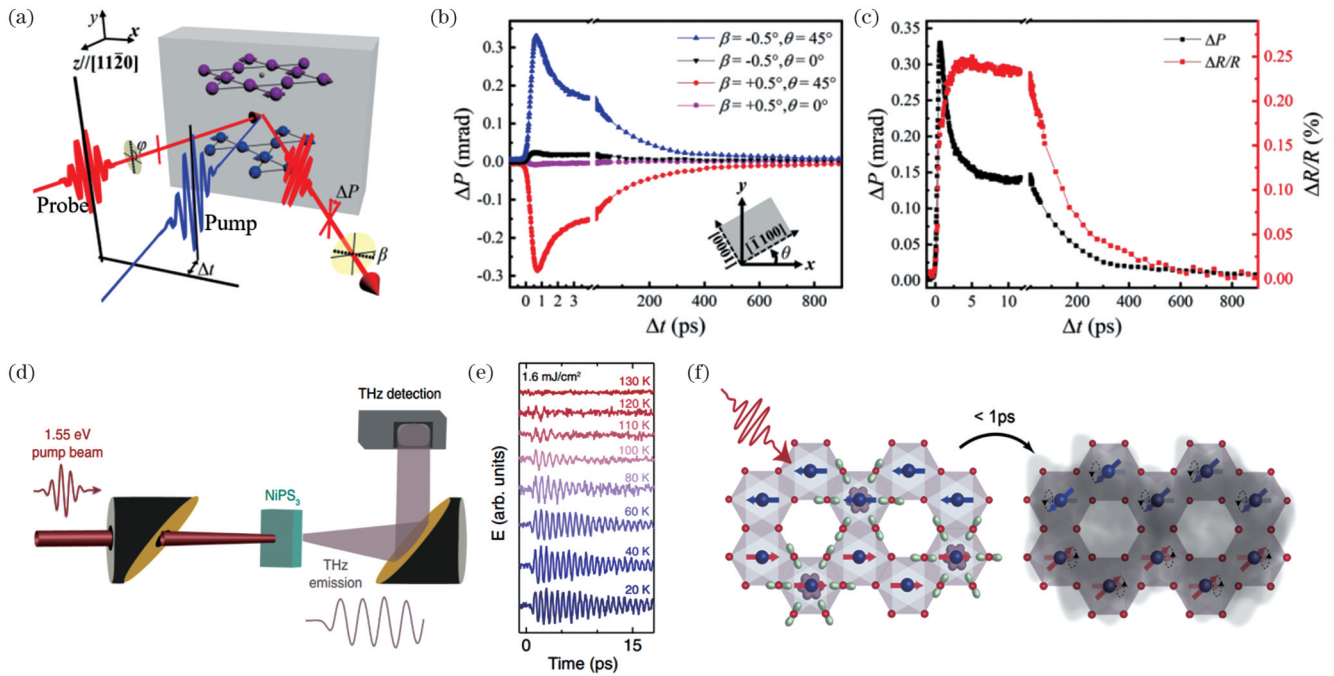


图 8 超快激光的控磁作用。(a)激光诱导 Mn_3Sn 反铁磁序淬灭及探测过程示意图^[71]；(b)室温下不同入射角配置的自旋动力学^[71]；(c) Mn_3Sn 自旋动力学和瞬态吸收信号的对比^[71]；(d) NiPS_3 太赫兹发射测量的实验装置示意图^[81]；(e)不同温度下相干磁子振荡带来的时间演化太赫兹电场^[81]；(f)激光在 NiPS_3 中诱导的瞬态反铁磁金属态^[81]

Fig. 8 Magnetic control effect of ultrafast laser. (a) Schematic of laser-induced Mn_3Sn antiferromagnetic order quenching and detection^[71]; (b) spin dynamics with different incident angle configurations at room temperature^[71]; (c) comparison of spin dynamics and transient absorption signals in Mn_3Sn ^[71]; (d) schematic of experimental configuration for detection of NiPS_3 terahertz emission^[81]; (e) time-evolution terahertz electric fields caused by coherent magnon oscillation at different temperatures^[81]; (f) laser-induced transient antiferromagnetic metal state in NiPS_3 ^[81]

4 二维反铁磁体系中的元激发准粒子研究

在光与物质相互作用系统中,微观准粒子是集体激发和能量振荡的量子化描述,是连接宏观和微观量子现象的纽带,在凝聚态物理学中占有重要地位。在磁性材料中,外界扰动可能使材料磁矩在自旋有序的平衡位置附近发生振荡。由于原子磁矩之间的交换耦合作用,这种振荡可形成传导波,并被量子化为磁子。类似光子和声子,磁子是一种服从玻色-爱因斯坦分布的准粒子,它能够与光子、电子和声子等其他准粒子相互耦合,引发一系列新奇现象,相关的各类稳态和瞬态光谱研究得到了广泛关注。

4.1 磁子及其输运

对于磁性材料来说,激光对自旋电子系统的集体激发产生了磁子^[86]。一方面,磁子振荡过程反映了自旋系统的激发态能量变化,其频率能通过外界物理场进行调谐;另一方面,作为自旋角动量在实空间传输的载体,磁子能够完成对信息的处理和传输^[24, 135-137],其频率决定了自旋电子器件的开关速度^[24]。不同于反铁磁材料两个子晶格的交换作用,由于弱磁各向异性^[126],基于铁磁材料的器件的运行频率在 GHz 量级,通常应用于电信和雷达系统中,而具备更高运行频率

和带宽的反铁磁材料则对太赫兹应用大有裨益^[138-139]。

单层铁磁 CrI_3 在实现高频自旋波方面具有两个优点: Ising 型自旋相互作用产生的强磁各向异性以及蜂窝形晶格内两个 Cr^{3+} 子晶格之间的强交换耦合^[140]。 Cr^{3+} 离子对应两个自旋波分支模式,分别为同相进动的声学 and 异相进动的光学模式,最高频率达到了太赫兹量级。如图 9(a)~(c)所示,通过在反铁磁双层 CrI_3 中引入非磁单层 WSe_2 ,形成了异质结堆叠结构,Zhang 等^[141]在实验中打破了双层 CrI_3 的层间对称性,这不仅利于磁子的偏振光学检测,其异质结间的电荷转移过程还提升了 CrI_3 的光吸收和磁子激发效率。凭借栅压控制的静电掺杂方式,该团队还在数十 GHz 的范围内实现了对 CrI_3 磁子模式的调谐^[141]。在小于电子带隙的能量内,很多反铁磁材料还存在多个表征 d-d 轨道跃迁的激发能级^[22, 142],这些多能级跃迁与相干态磁子的激发密切相关。如图 9(d)所示,在与 Ni 离子轨道共振的基础上,Afanasiev 等^[143]使用波长调谐的超短光脉冲激发了 NiPS_3 中轨道能量相关的多个磁子模式,证明了磁各向异性与电子轨道跃迁的相关性,跃迁电子、相干磁子和声子等准粒子能普遍地与磁各向异性场相互耦合,其中的太赫兹电场分量能够反过来驱动磁子,并在时域中被直接观测到^[144]。光激发带来的电荷输

运过程同样会影响磁子模式,在 FePS₃ 与拓扑绝缘体 Bi₂Te₃ 和铁磁半导体 Cr₂Ge₂Te₆ 的堆叠结构中,拉曼光

谱表明磁子模式的频移与异质结界面电荷转移行为具有较强的相关性^[145]。

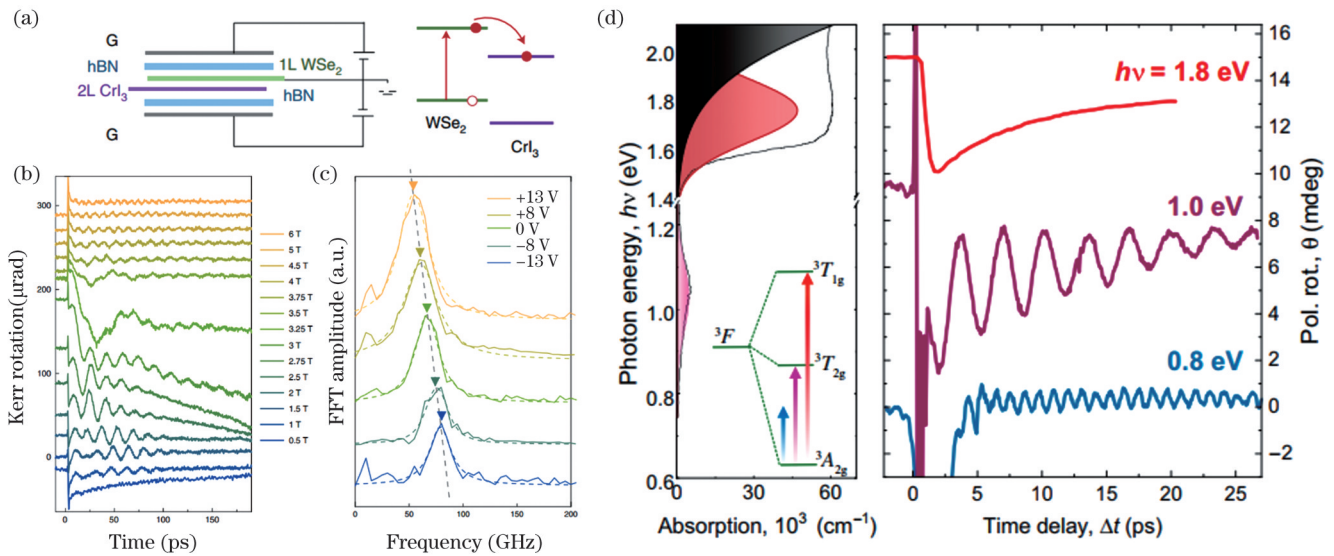


图 9 超快激光诱导的磁子动力学。(a) 双层 CrI₃/单层 WSe₂ 异质结电控器件构筑及层间电荷转移过程示意^[141]; (b) 不同磁场下的磁子动力学过程^[141]; (c) 受栅极电压调控的磁子频率^[141]; (d) NiPS₃ 中不同能级光吸收跃迁对应的磁子动力学^[143]
 Fig. 9 Magnon dynamics induced by ultrafast laser. (a) Construction of electric control devices with bilayer CrI₃/monolayer WSe₂ heterostructure and schematic of interlayer charge transfer process^[141]; (b) magnon dynamics under different magnetic fields^[141]; (c) magnon frequency regulated by gate voltage^[141]; (d) magnon dynamics corresponding to optical absorption transition at different energy levels in NiPS₃^[143]

超快纳米级磁逻辑电路的发展需要产生频率尽可能高、波长尽可能短的相干自旋波^[14]。在反铁磁自旋输运方面,成熟的研究对象为块体过渡金属氧化物^[146]。在该类材料中,反铁磁性和电荷绝缘性共存,使得角动量可以仅通过磁子传输而不产生电荷驱动效应,由此带来的低能量耗散使得磁子可在微米范围内传播并具有接近极限的磁操作速度。Xing 等^[147]报道了二维 MnPS₃ 中长达几微米的磁子输运,随着材料厚度的增加和测试温度的降低,表面散射作用减小,磁子寿命和输运距离增加。在该工作中,基于自旋霍尔效应的电学测试被当作测量磁子输运的常规手段^[148],但在一定程度上缺乏对磁子输运过程的动态观察。最近,CrSBr 中磁子-激子耦合行为引发了一定的关注,相关工作使得相干磁子输运的全光探测成为可能。具体地,Bae 等^[149]通过空间分辨的瞬态吸收光谱对光生激子进行了成像,由于磁子-激子耦合,磁子的时域振荡直接体现在瞬态吸收动力学中,其空间分辨的测试结果进一步反映了相干磁子在二维平面上的传播,具有长达 10 ns 的相干时间(比块体 DyFeO₃ 长 2 个数量级^[150])和高达 7 μm 的相干传输长度。

4.2 二维激子效应

低光子能量的太赫兹发射经常与载流子的带内跃迁和晶格振动相关,而较高能量的可见和近红外光发射则与带间复合相关。在非磁的低维半导体和某些低维绝缘体中,激子发射是作为带间复合的一种而普遍存在的。就磁性材料而言,很多偏向金属性的材料的

磁性由巡游电子主导,并没表现出明显的激子效应,因而较早的磁-激子相关研究是围绕磁性材料(提供磁性)和非磁半导体材料(提供激子)的异质结磁近邻效应展开的^[151]。近年来越来越多的工作表明,在磁性半导体和绝缘体中,激子是可以作为本征属性而存在的,由于磁结构诱导的光学各向异性和自旋轨道耦合作用,载流子和激子的光致发光效应与材料的磁态密切相关。

在单层 CrI₃ 的铁磁性被发现后,研究者们把其分子轨道中高度局域化的光激发与磁性激子联系起来。如图 10(a) 所示,Seyler 等^[35]通过温度、外磁场调控下的荧光和反射光谱测试,揭示了单层 CrI₃ 在 1.1 eV 能量处的激子特征,线宽为 180 meV,并且具有圆偏振特性。激子的发射模式和圆偏振度与磁序的调控情况紧密相关,施加的面外磁场诱导的磁化翻转能够改变荧光的圆偏振方向,在双层 CrI₃ 中,当层间磁序转变为反铁磁耦合时,荧光的圆偏振度消失^[35]。如图 10(b)~(d) 所示,CrI₃ 激子也具有极化子特性,光激发产生的电荷转移型激子和纵向光学声子之间发生强耦合作用,在拉曼光谱中出现了多阶激子-声子耦合态,长程磁序的出现将电子-声子耦合强度提高了约 50%^[152]。Wu 等^[153]基于第一性原理计算进一步揭示了单层 CrI₃ 中的激子效应可极大增强材料的磁光响应,并理论预测了磁光信号与光频率和衬底状态的相关性。与 CrI₃ 相比,CrBr₃ 和 CrCl₃ 具有更局域化的激子波函数,这意味着增大的激子结合能以及在实空间中更短的电子-空穴距离^[36]。通过对深能级激子的详细分析,认为

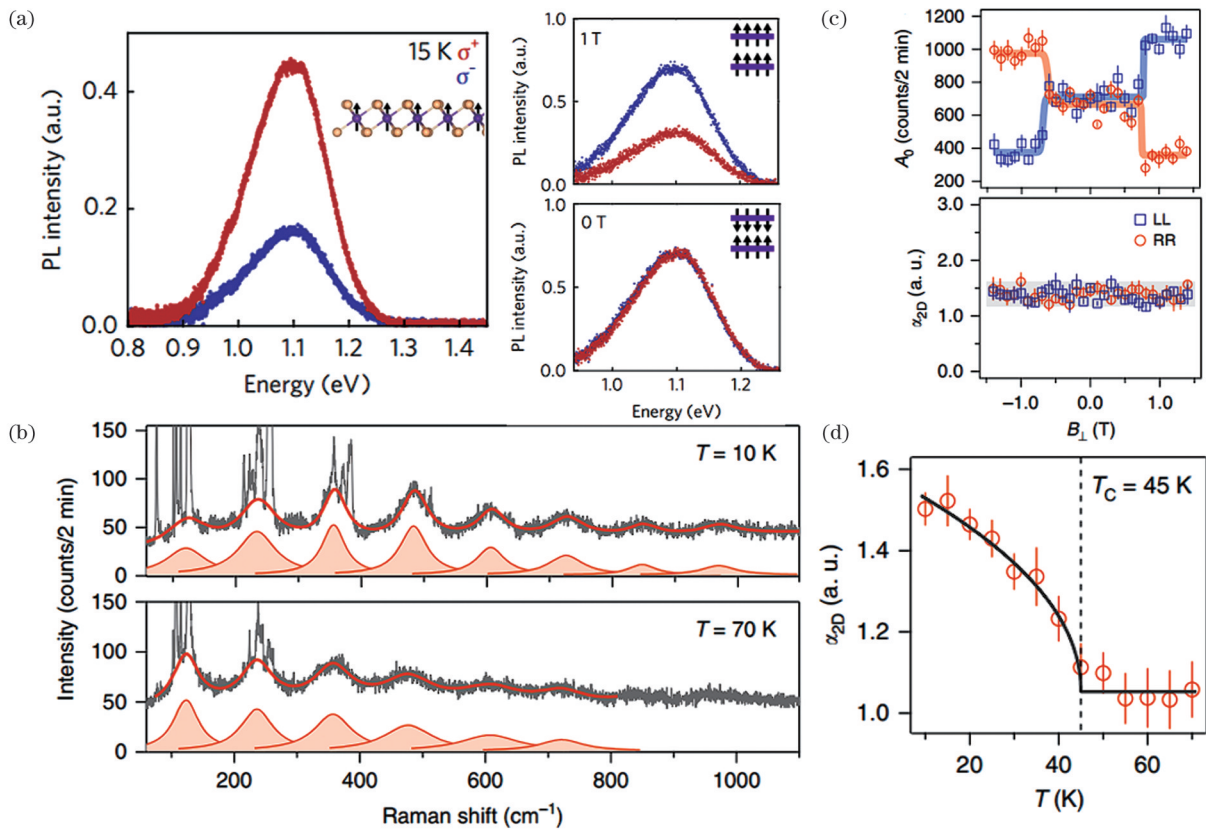


图 10 CrI₃中激子的光学特性。(a)单层和双层CrI₃的具有圆偏振特性的激子发射^[35];(b)不同温度下与激子相关的声子修饰电子态,表现为周期性的拉曼模式^[152];(c)拉曼模式振幅(上)和电子-声子耦合强度(下)的磁场相关性^[152];(d)电子-声子耦合强度在磁临界温度附近的转变情况^[152]

Fig. 10 Optical properties of excitons in CrI₃. (a) Excitonic emission with circular polarization characteristics of monolayer and bilayer CrI₃^[35]; (b) phonon modified electronic states related to excitons at different temperatures reflected by periodic Raman modes^[152]; (c) magnetic field correlation between Raman mode amplitude (above) and electron-phonon coupling intensity (below)^[152]; (d) transition of electron-phonon coupling intensity near magnetic critical temperature^[152]

CrCl₃中的激子更接近Frenkel型,CrI₃中的激子具有混合的Frenkel-Wannier特征^[154]。

以NiPS₃为代表的过渡金属磷硫化物在Néel温度以下的异常吸收光谱表明了强电荷-自旋耦合^[155],其中的激子被证明为Wannier型^[156]。2020年,Kang等^[22]将少层NiPS₃中的窄线宽稳态光发射归因于Zhang-Rice态激子从单重态到三重态的跃迁,在可见光和X射线光谱中发现了块体NiPS₃激子约0.4 meV的极窄发射线宽[图11(a)],并证明了这种多体相干态激子的层数相关性。当NiPS₃厚度减小到双层时,体系的磁有序性逐渐受到抑制^[73],激子荧光强度也逐渐减弱,最终在单层样品中完全消失。进一步地,Ho等^[142]通过热调制反射光谱发现了NiPS₃的A和B两类激子模式,明确了低温下A激子的多能级精细特征。类似于光吸收中的二向色性,在NiPS₃被线偏振光激发后,激子发光仍然保持了高度的各向异性,Hwangbo等^[156]将这种激子发光的各向异性归因于材料内部的锯齿形反铁磁序,同时发现了激子-声子耦合的多声子束缚态和可能存在的强激子-磁子耦合作

用。最近,NiPS₃中这种多声子束缚态被证明起源于局域化的d-d轨道电子跃迁^[157]。如图11(b)所示,通过温度、磁场和激发光偏振调控,Wang等^[32]实现了对多层NiPS₃激子荧光的偏振调节,其发射偏振与Néel矢量方向高度一致,并且表现出高度的线偏振特性。类似于NiPS₃,最近在反铁磁体NiI₂中也发现了Zhang-Rice态激子的窄线宽发光^[158],线宽达到5 meV,由于激子峰与材料的铁电螺旋相直接相关,利用反演对称性的破缺可以有力地调控NiI₂磁激子。另一方面,Ni原子组成的三角形晶格使该材料成为实现二维量子纠缠态的理想候选^[158]。此外,MnPS₃中的大激子结合能^[159]和CrPS₄中的磁耦合态激子^[160]也得到了报道。如图11(c)、(d)所示,在磁有序温度以下时,CrSBr激子表现出窄线宽特性,与CrI₃的层间磁态类似,双层反铁磁CrSBr中的电子和空穴轨道与层间磁序直接耦合。对于双层以上材料,增加的材料厚度对应于增强的层间耦合现象,当通过磁场控制磁序并将反铁磁基态转换到铁磁激发态时,激子跃迁能量会发生明显变化^[40]。

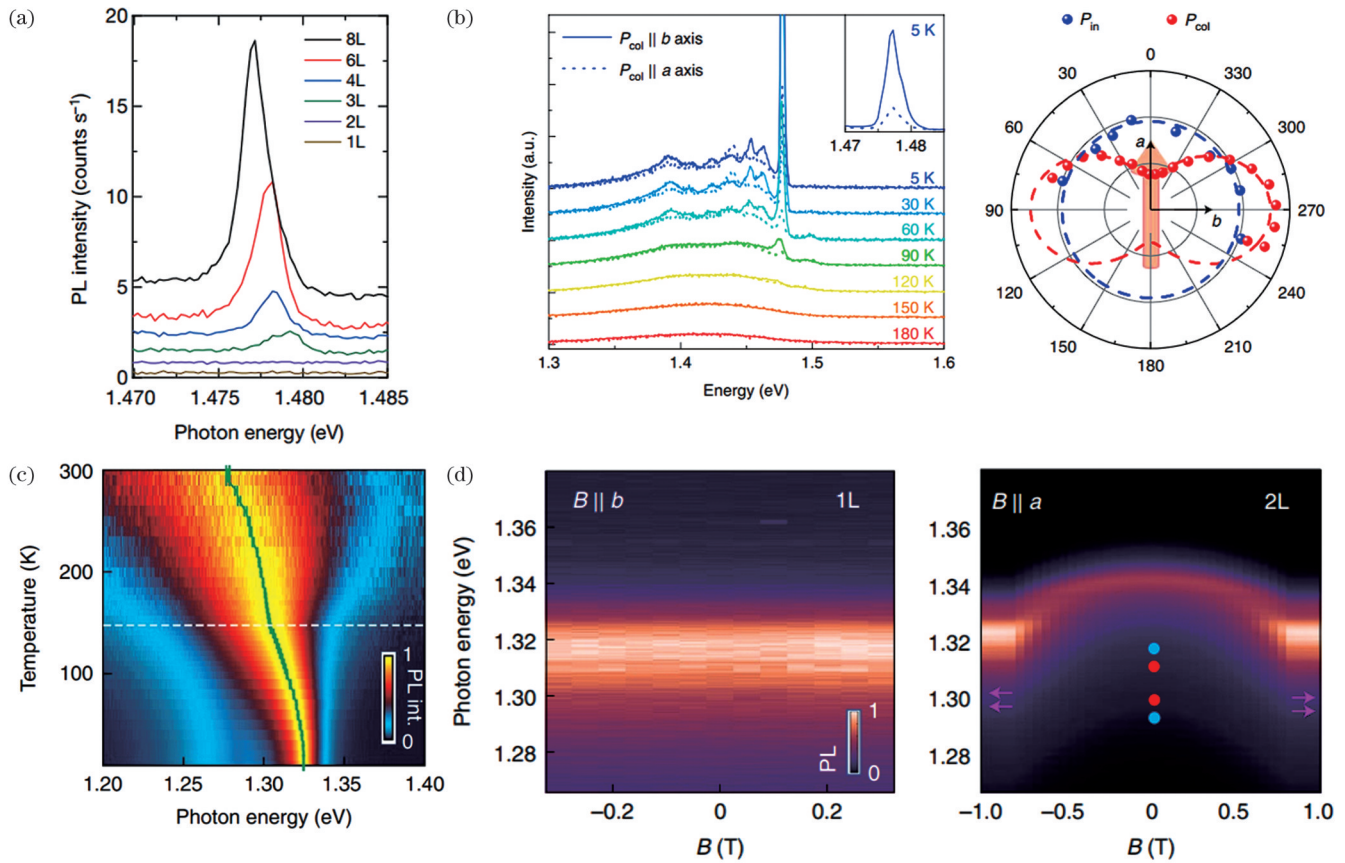


图 11 两类二维反铁磁材料的激子效应。(a)不同厚度 NiPS₃ 激子的窄线宽荧光发射特征^[22];(b)多层 NiPS₃ 激子荧光的温度和偏振相关性^[32];(c)CrSBr 激子荧光的温度相关性^[40];(d)单层和双层 CrSBr 荧光的磁场相关性^[40]

Fig. 11 Exciton effects of two types of two-dimensional antiferromagnets. (a) Narrow linewidth fluorescence emission characteristics of NiPS₃ excitons with different thicknesses^[22]; (b) temperature and polarization dependences of multilayer NiPS₃ exciton fluorescence^[32]; (c) temperature dependence of exciton fluorescence in CrSBr^[40]; (d) correlation between fluorescence and magnetic field in monolayer and bilayer CrSBr^[40]

4.3 磁子-极化子耦合态

磁子和声子是磁有序系统中两种相关的低能激发,由于能量位置接近,一些材料中的声子振荡模式被证明与长程磁序密切相关。对于许多反铁磁材料来说,虽然激光的光子能量远离磁子的本征能量,但通过各种机制产生的超快声子已被证明为驱动和控制自旋动力学的有力工具,其中光激发的声子模式可以完全相干地操纵磁序^[144]。类似于电荷与光子的极化子耦合态^[161],磁子和声子也能够通过相互作用耦合形成磁子极化子,并同样在光谱中表现为色散反交叉的磁子和声子曲线。磁子与声子的杂化可为反铁磁材料的磁光控制提供一个新的研究平台。

在理论研究中,FePS₃会出现低温下的额外声子模式,MnPS₃则表现出显著的自旋-声子耦合^[162]。对应地,六角晶格二维反铁磁体中磁子-声子极化子的激发得到了理论证明^[163],相关拓扑磁子和手性声子的可能应用也被提出。如图 12(a)、(b)所示,Liu 等^[164]实验报道了这种磁子-声子耦合准粒子的形成,他们借助高达 30 T 的磁场对二维反铁磁 FePS₃ 的磁子能量进行直接调谐,塞曼频移使磁子在能量最近的位置与声子

发生共振并产生强耦合作用。微观耦合模型和偏振分辨测试结果表明,磁子将自旋角动量传递给声子,并通过强耦合产生声子自旋,证明了二维反铁磁半导体作为集成光电、声子和磁器件枢纽系统的潜力^[164]。Vaclavkova 等^[165]利用多波段拉曼光谱发现了二维 FePS₃ 中的磁子极化子,通过外加磁场调控,磁子峰的低能分支分别与三个声子模式产生反交叉,标志着强耦合的出现。基于远红外透射光谱,Zhang 等^[166]在 FePS₃ 中得到了巨大的磁子极化子耦合强度,最大值可达 0.5 meV。如图 12(c)、(d)所示,通过近共振泵浦手段,Sun 等^[167]在相对较小的磁场 (9 T) 下实现了 FePS₃ 中的磁子-声子强耦合,耦合特征比非共振泵浦条件下更显著,并且磁子-声子的耦合模式相对纯声子模式出现了不同的偏振特征。类似地,Mai 等^[168]利用温度相关的拉曼光谱确定了层状 MnPSe₃ 中成对激发的磁子与声子之间的杂化态。

在 A 型反铁磁材料 CrCl₃ 中,由于较弱的层间磁耦合,较低的磁子频率与铁磁材料的磁振荡频率接近,有利于微波波段的耦合。MacNeill 等^[169]报道了 CrCl₃ 在微波波段的宽带吸收光谱,在打破旋转对称性后,两个

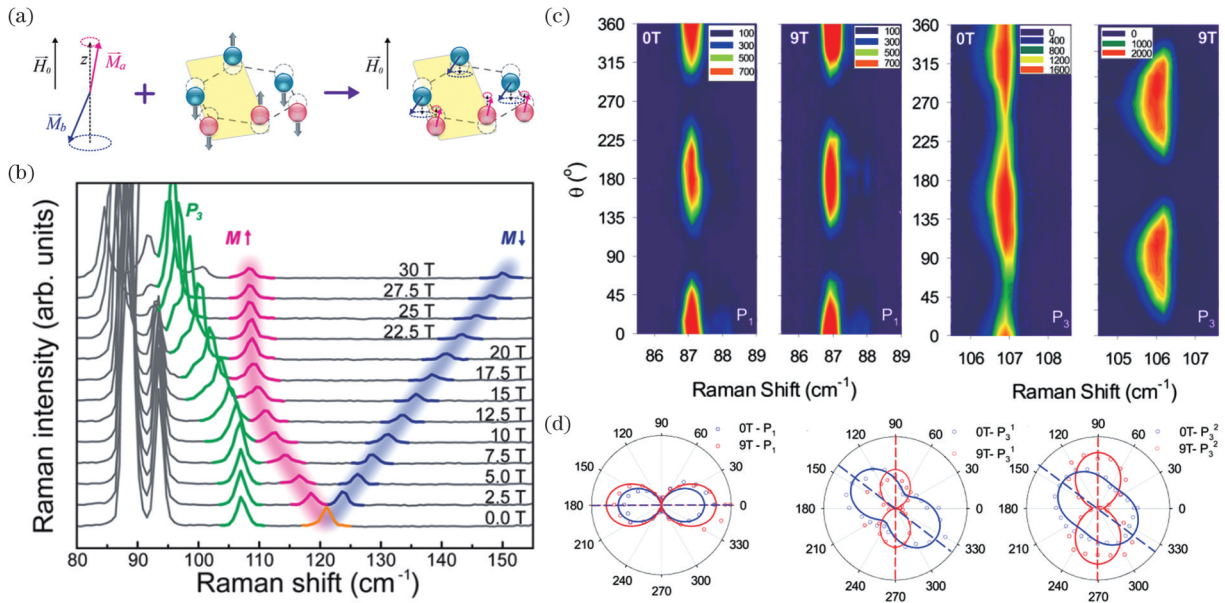


图 12 FePS₃中的强磁子-声子耦合。(a)经典模型中的磁子、声子和磁子-声子强耦合模式示意图^[164]；(b)FePS₃在0~30 T 磁场中的拉曼光谱和磁子-声子强耦合的反交叉特性^[164]；(c)磁场调控下纯声子模式(左)和磁子-声子耦合模式(右)的偏振特性对比；(d)对应图 12(c)的强度等值线图^[167]

Fig. 12 Strong magnon-phonon coupling in FePS₃. (a) Schematics of magnon, phonon and magnon-phonon strong coupling modes in classical model^[164]; (b) Raman spectra of FePS₃ and anti-cross characteristics of strong magnon-phonon coupling in 0~30 T magnetic field^[164]; (c) comparison of polarization characteristics between pure phonon mode (left) and magnon-phonon coupling mode (right) controlled by magnetic field^[167]; (d) intensity contour maps corresponding to Fig. 12(c)^[167]

共振模式之间可以诱导具有大的可调谐能隙的强磁子-磁子耦合。进一步地, Zhang 等^[170]研究了 CrCl₃ 磁子与平面超导腔体模式的耦合作用, 通过磁场控制的自旋翻转和 CrCl₃ 在腔中位置的改变, 发现磁子的光学模和声学模都表现出反交叉特性, 在不同的微波磁场分量和外加磁场的激励下, 两者可分别与腔模耦合, 并且声模的耦合强度大于光模的耦合强度。对于追求输运距离和时间相干性的磁子器件来说, 直接观察磁子-声子相互作用对于理解磁子的退相干路径和相关的能量传递过程具有重要意义^[168]。通常来说, 磁子和声子之间的相互作用可能会对激发的磁子能量造成不利影响, 使其快速耗散, 导致散射速率的增加和磁子寿命的降低, 最终限制磁子扩散距离。但是, 对于非相干的情况, 由于一些材料较低的声阻尼, 磁子和声学声子之间的杂化可能反而能够增强宏观尺度上的磁子输运^[171]。另一方面, 处于亚 meV 能量位置的相干磁子同样可以与能量大出 3 个数量级以上的激子发生耦合作用, 例如 CuB₂O₄ 荧光光谱中归因于磁子辅助复合的多条激子伴线^[172]。此外, 磁子-激子耦合态也在反铁磁/非磁异质结界面中被发现, 如反铁磁磁子通过界面交换作用对激子施加的动态能谷调控^[173]。

5 总结与展望

反铁磁材料的独特优势及反铁磁自旋光电子学的发展将毫无疑问地推动自旋电子学相关领域的发展。一方面, 对磁光效应的探索更有助于反铁磁材料应用

场景的延伸, 如新型磁光效应中的磁手性二向色性, 非偏振光和手性介质结合对光同样具有非互易表现^[174-175], 不仅能应用在反铁磁序的探测中, 还对非偏光信号的调制有应用价值。另一方面, 光学技术本身的更新迭代使得在更加微观、多维和超快尺度上探测磁序变得容易, 例如单自旋扫描显微镜、自旋极化扫描隧道显微镜、光镊、近场分辨光谱等技术^[176-178]。可以预见, 基于磁光效应开展的反铁磁材料相关研究会集中于更加复杂和极端的体系中, 如具备非共线、螺旋磁性、多铁性、拓扑磁性等磁性质的材料, 原子层厚度的二维反铁磁材料及其异质结的引入将使得这些复杂磁态展现出更加奇异的特性。举例来说, 通过控制多铁性反铁磁材料内部的磁序, 可以有效地调控层间电子耦合, 便于通过光学手段探测、操纵、发射和跟踪自旋信息; 由于自旋动量锁定, 拓扑反铁磁材料能被应用在高能效自旋电子学中, 带来巨大的光学非线性系数、高效的自旋滤波、高电荷流-自旋流转换效率、高电子迁移率和长自旋扩散长度等特性^[179], 拓宽磁学、光子学和其他交叉学科的研究范围。

由于量子限域效应, 二维材料相对于传统块体材料提供了一个更好的光与物质相互作用的研究平台^[180]。虽然就目前来说, 磁性材料在二维条件下写入和读取磁状态的能力有待提高, 并仍需借鉴传统块体材料在超快磁序控制和自旋输运等方面取得的研究成果^[102, 146], 但相对于很多尚未完善的物理机制, 二维反铁磁材料已经展现出了许多应用方面的优势。例如,

除了某些材料表现出的各向异性磁阻^[181], CrSBr 通过磁序与磁输运之间的耦合作用表现出了巨负磁阻响应, 是磁性金属材料的 10 倍和稀磁半导体的 2 倍以上^[41], 少层 CrI₃ 表现出的巨磁隧穿电阻^[104, 182]也远远大于基于 MgO 势垒的铁磁隧穿结^[2]。反铁磁自旋电子学的关键是利用和控制好磁性, 即 Néel 矢量或自旋排列的变化^[16, 183], 其应用朝着快速度、高密度和低损耗的方向发展, 并且能够在脱离外部磁场的情况下完成自旋切换^[16-17, 27]; 而除了自身性质的改变外, 二维反铁磁材料与磁性超表面的结合可能加强对磁手性和光学偏振的操控能力^[184], 与微腔的结合则能使光子、声子、磁子以及超导量子发生准粒子相干作用, 带来的更少的微粒散射和更长的自旋相干时间是磁孤子^[185]、自旋超流体^[186]等现象的先决条件, 有助于二维反铁磁材料在量子信息处理和传感中发挥更重要的作用^[13]。诚然, 在二维反铁磁材料的应用推广方面, 仍有许多问题需要解决, 其中很关键的一点是, 除了二维化导致的热扰动增强使得材料磁序稳定性对低温的要求更高外, 维度降低所增强的自旋散射作用在一定程度上阻碍了高极化度磁输运过程, 并且超大的表面积体积比使得这些超薄层状材料更容易在自然情况下遭受掺杂、氧化甚至降解, 这些问题需要通过新材料的发掘以及制备工艺的提高来解决。目前一些理论研究已经预测了能够在室温下稳定工作的二维反铁磁材料的特性, 如 FeAs 和 Cr₂Br₃S₃ 等的高温磁相^[187-188]、CrTe₂ 的室温可调谐磁光效应^[189]。

参 考 文 献

- Jedema F J, Filip A T, van Wees B J. Electrical spin injection and accumulation at room temperature in an all-metal mesoscopic spin valve[J]. *Nature*, 2001, 410(6826): 345-348.
- Parkin S S P, Kaiser C, Panchula A, et al. Giant tunnelling magnetoresistance at room temperature with MgO (100) tunnel barriers[J]. *Nature Materials*, 2004, 3(12): 862-867.
- Chuang P, Ho S C, Smith L W, et al. All-electric all-semiconductor spin field-effect transistors[J]. *Nature Nanotechnology*, 2015, 10(1): 35-39.
- Kim Y H, Zhai Y X, Lu H P, et al. Chiral-induced spin selectivity enables a room-temperature spin light-emitting diode[J]. *Science*, 2021, 371(6534): 1129-1133.
- Jungwirth T, Wunderlich J, Olejnik K. Spin Hall effect devices[J]. *Nature Materials*, 2012, 11(5): 382-390.
- 黄于, 周沛, 杨一功, 等. 自旋激光器的动力学特性及应用研究进展[J]. *强激光与粒子束*, 2021, 33(11): 111006. Huang Y, Zhou P, Yang Y G, et al. Progress in research of dynamic properties and applications of spin-lasers[J]. *High Power Laser and Particle Beams*, 2021, 33(11): 111006.
- Han W, Kawakami R K, Gmitra M, et al. Graphene spintronics [J]. *Nature Nanotechnology*, 2014, 9(10): 794-807.
- Tongay S, Varoosfaderani S S, Appleton B R, et al. Magnetic properties of MoS₂: existence of ferromagnetism[J]. *Applied Physics Letters*, 2012, 101(12): 123105.
- Guguchia Z, Kerelsky A, Edelberg D, et al. Magnetism in semiconducting molybdenum dichalcogenides[J]. *Science Advances*, 2018, 4(12): eaat3672.
- Gong C, Li L, Li Z L, et al. Discovery of intrinsic ferromagnetism in two-dimensional van der Waals crystals[J]. *Nature*, 2017, 546(7657): 265-269.
- Huang B, Clark G, Navarro-Moratalla E, et al. Layer-dependent ferromagnetism in a van der Waals crystal down to the monolayer limit[J]. *Nature*, 2017, 546(7657): 270-273.
- Hellman F, Hoffmann A, Tserkovnyak Y, et al. Interface-induced phenomena in magnetism[J]. *Reviews of Modern Physics*, 2017, 89(2): 025006.
- Lachance-Quirion D, Tabuchi Y, Gloppe A, et al. Hybrid quantum systems based on magnonics[J]. *Applied Physics Express*, 2019, 12(7): 070101.
- Brataas A, van Wees B, Klein O, et al. Spin insulatronics[J]. *Physics Reports*, 2020, 885: 1-27.
- Šmejkal L, Mokrousov Y, Yan B H, et al. Topological antiferromagnetic spintronics[J]. *Nature Physics*, 2018, 14(3): 242-251.
- Baltz V, Manchon A, Tsoi M, et al. Antiferromagnetic spintronics [J]. *Reviews of Modern Physics*, 2018, 90(1): 015005.
- Železný J, Wadley P, Olejnik K, et al. Spin transport and spin torque in antiferromagnetic devices[J]. *Nature Physics*, 2018, 14(3): 220-228.
- Maniv E, Murphy R A, Haley S C, et al. Exchange bias due to coupling between coexisting antiferromagnetic and spin-glass orders [J]. *Nature Physics*, 2021, 17(4): 525-530.
- Savary L, Balents L. Quantum spin liquids: a review[J]. *Reports on Progress in Physics*, 2017, 80(1): 016502.
- 罗健, 张小伟, 代波. 反应磁控溅射氧化镍薄膜的自旋塞贝克效应[J]. *人工晶体学报*, 2021, 50(9): 1668-1674.
- Luo J, Zhang X W, Dai B. Spin seebeck effect of nickel oxide thin films prepared by reactive magnetron sputtering[J]. *Journal of Synthetic Crystals*, 2021, 50(9): 1668-1674.
- Zhang Q, Hwangbo K, Wang C, et al. Observation of giant optical linear dichroism in a zigzag antiferromagnet FePS₃[J]. *Nano Letters*, 2021, 21(16): 6938-6945.
- Kang S, Kim K, Kim B H, et al. Coherent many-body exciton in van der Waals antiferromagnet NiPS₃[J]. *Nature*, 2020, 583(7818): 785-789.
- Lan T S, Ding B F, Liu B L. Magneto-optic effect of two-dimensional materials and related applications[J]. *Nano Select*, 2020, 1(3): 298-310.
- Chumak A V, Vasyuchka V I, Serga A A, et al. Magnon spintronics[J]. *Nature Physics*, 2015, 11(6): 453-461.
- Němec P, Fiebig M, Kampfrath T, et al. Antiferromagnetic optospintronics[J]. *Nature Physics*, 2018, 14(3): 229-241.
- Jungwirth T, Marti X, Wadley P, et al. Antiferromagnetic spintronics[J]. *Nature Nanotechnology*, 2016, 11(3): 231-241.
- Gomonay E V, Loktev V M. Spintronics of antiferromagnetic systems (review article)[J]. *Low Temperature Physics*, 2014, 40(1): 17-35.
- Mak K F, Shan J, Ralph D C. Probing and controlling magnetic states in 2D layered magnetic materials[J]. *Nature Reviews Physics*, 2019, 1(11): 646-661.
- Kurebayashi H, Garcia J H, Khan S, et al. Magnetism, symmetry and spin transport in van der Waals layered systems[J]. *Nature Reviews Physics*, 2022, 4(3): 150-166.
- Gibertini M, Koperski M, Morpurgo A F, et al. Magnetic 2D materials and heterostructures[J]. *Nature Nanotechnology*, 2019, 14(5): 408-419.
- Rahman S, Torres J F, Khan A R, et al. Recent developments in van der Waals antiferromagnetic 2D materials: synthesis, characterization, and device implementation[J]. *ACS Nano*, 2021, 15(11): 17175-17213.
- Wang X Z, Cao J, Lu Z G, et al. Spin-induced linear polarization of photoluminescence in antiferromagnetic van der Waals crystals [J]. *Nature Materials*, 2021, 20(7): 964-970.
- Higo T, Man H Y, Gopman D B, et al. Large magneto-optical Kerr effect and imaging of magnetic octupole domains in an

- antiferromagnetic metal[J]. *Nature Photonics*, 2018, 12(2): 73-78.
- [34] Wu M X, Isshiki H, Chen T S, et al. Magneto-optical Kerr effect in a non-collinear antiferromagnet Mn_3Ge [J]. *Applied Physics Letters*, 2020, 116(13): 132408.
- [35] Seyler K L, Zhong D, Klein D R, et al. Ligand-field helical luminescence in a 2D ferromagnetic insulator[J]. *Nature Physics*, 2018, 14(3): 277-281.
- [36] Molina-Sánchez A, Catarina G, Sangalli D, et al. Magneto-optical response of chromium trihalide monolayers: chemical trends[J]. *Journal of Materials Chemistry C*, 2020, 8(26): 8856-8863.
- [37] McGuire M. Crystal and magnetic structures in layered, transition metal dihalides and trihalides[J]. *Crystals*, 2017, 7(5): 121.
- [38] Kim S, Lee J, Lee C G, et al. Polarized Raman spectra and complex Raman tensors of antiferromagnetic semiconductor CrPS_4 [J]. *The Journal of Physical Chemistry C*, 2021, 125(4): 2691-2698.
- [39] Yan J Q, Zhang Q, Heitmann T, et al. Crystal growth and magnetic structure of MnBi_2Te_4 [J]. *Physical Review Materials*, 2019, 3(6): 064202.
- [40] Wilson N P, Lee K, Cenker J, et al. Interlayer electronic coupling on demand in a 2D magnetic semiconductor[J]. *Nature Materials*, 2021, 20(12): 1657-1662.
- [41] Telford E J, Dismukes A H, Lee K, et al. Layered antiferromagnetism induces large negative magnetoresistance in the van der Waals semiconductor CrSBr [J]. *Advanced Materials*, 2020, 32(37): 2003240.
- [42] Du K Z, Wang X Z, Liu Y, et al. Weak van der Waals stacking, wide-range band gap, and Raman study on ultrathin layers of metal phosphorus trichalcogenides[J]. *ACS Nano*, 2016, 10(2): 1738-1743.
- [43] Gu M Q, Rondinelli J M. Nonlinear phononic control and emergent magnetism in Mott insulating titanates[J]. *Physical Review B*, 2018, 98(2): 024102.
- [44] Klein D R, MacNeill D, Song Q, et al. Enhancement of interlayer exchange in an ultrathin two-dimensional magnet[J]. *Nature Physics*, 2019, 15(12): 1255-1260.
- [45] Guo K, Deng B W, Liu Z, et al. Layer dependence of stacking order in nonencapsulated few-layer CrI_3 [J]. *Science China Materials*, 2020, 63(3): 413-420.
- [46] Serri M, Cucinotta G, Poggini L, et al. Enhancement of the magnetic coupling in exfoliated CrCl_3 crystals observed by low-temperature magnetic force microscopy and X-ray magnetic circular dichroism[J]. *Advanced Materials*, 2020, 32(24): 2000566.
- [47] McGuire M A, Clark G, Santosh K C, et al. Magnetic behavior and spin-lattice coupling in cleavable van der Waals layered CrCl_3 crystals[J]. *Physical Review Materials*, 2017, 1(1): 014001.
- [48] Song T C, Sun Q C, Anderson E, et al. Direct visualization of magnetic domains and moiré magnetism in twisted 2D magnets[J]. *Science*, 2021, 374(6571): 1140-1144.
- [49] Xu Y, Ray A, Shao Y T, et al. Coexisting ferromagnetic-antiferromagnetic state in twisted bilayer CrI_3 [J]. *Nature Nanotechnology*, 2022, 17(2): 143-147.
- [50] Akram M, LaBollita H, Dey D, et al. Moiré skyrmions and chiral magnetic phases in twisted CrX_3 ($X = \text{I}, \text{Br}, \text{and Cl}$) bilayers[J]. *Nano Letters*, 2021, 21(15): 6633-6639.
- [51] Coak M J, Jarvis D M, Hamidov H, et al. Tuning dimensionality in van-der-Waals antiferromagnetic Mott insulators TMPS_3 [J]. *Journal of Physics: Condensed Matter*, 2020, 32(12): 124003.
- [52] Basnet R, Wegner A, Pandey K, et al. Highly sensitive spin-flop transition in antiferromagnetic van der Waals material MPS_3 ($M = \text{Ni}$ and Mn)[J]. *Physical Review Materials*, 2021, 5(6): 064413.
- [53] Yang K, Wang G Y, Liu L, et al. Triaxial magnetic anisotropy in the two-dimensional ferromagnetic semiconductor CrSBr [J]. *Physical Review B*, 2021, 104(14): 144416.
- [54] Gu P F, Sun Y J, Wang C, et al. Magnetic phase transitions and magnetoelastic coupling in a two-dimensional stripy antiferromagnet [J]. *Nano Letters*, 2022, 22(3): 1233-1241.
- [55] Zeng Y, Gu P F, Zhao Z J, et al. 2D FeOCl : a highly in-plane anisotropic antiferromagnetic semiconductor synthesized via temperature-oscillation chemical vapor transport[J]. *Advanced Materials*, 2022, 34(14): 2108847.
- [56] Kulish V V, Huang W. Single-layer metal halides MX_2 ($X = \text{Cl}, \text{Br}, \text{I}$): stability and tunable magnetism from first principles and Monte Carlo simulations[J]. *Journal of Materials Chemistry C*, 2017, 5(34): 8734-8741.
- [57] Kong T, Stolze K, Timmons E I, et al. VI_3 -a new layered ferromagnetic semiconductor[J]. *Advanced Materials*, 2019, 31(17): 1808074.
- [58] Tian S J, Zhang J F, Li C H, et al. Ferromagnetic van der Waals crystal VI_3 [J]. *Journal of the American Chemical Society*, 2019, 141(13): 5326-5333.
- [59] Kong T, Guo S, Ni D R, et al. Crystal structure and magnetic properties of the layered van der Waals compound VBr_3 [J]. *Physical Review Materials*, 2019, 3(8): 084419.
- [60] Zhao S, Wan W H, Ge Y F, et al. Prediction of chalcogen-doped VCl_3 monolayers as 2D ferromagnetic semiconductors with enhanced optical absorption[J]. *Annalen Der Physik*, 2021, 533(6): 2100064.
- [61] Pai Y Y, Marvinney C E, Feldman M A, et al. Magnetostriction of α - RuCl_3 flakes in the zigzag phase[J]. *The Journal of Physical Chemistry C*, 2021, 125(46): 25687-25694.
- [62] Kim H S, Kee H Y. Crystal structure and magnetism in α - RuCl_3 : an *ab initio* study[J]. *Physical Review B*, 2016, 93(15): 155143.
- [63] Sandilands L J, Sohn C H, Park H J, et al. Optical probe of Heisenberg-Kitaev magnetism in α - RuCl_3 [J]. *Physical Review B*, 2016, 94(19): 195156.
- [64] Tian Y Z, Gao W W, Henriksen E A, et al. Optically driven magnetic phase transition of monolayer RuCl_3 [J]. *Nano Letters*, 2019, 19(11): 7673-7680.
- [65] Li J H, Wang C, Zhang Z T, et al. Magnetically controllable topological quantum phase transitions in the antiferromagnetic topological insulator MnBi_2Te_4 [J]. *Physical Review B*, 2019, 100(12): 121103.
- [66] Duong N P, Satoh T, Fiebig M. Ultrafast manipulation of antiferromagnetism of NiO [J]. *Physical Review Letters*, 2004, 93(11): 117402.
- [67] Fiebig M, Duong N P, Satoh T, et al. Ultrafast magnetization dynamics of antiferromagnetic compounds[J]. *Journal of Physics D: Applied Physics*, 2008, 41(16): 164005.
- [68] Satoh T, Cho S J, Iida R, et al. Spin oscillations in antiferromagnetic NiO triggered by circularly polarized light[J]. *Physical Review Letters*, 2010, 105(7): 077402.
- [69] Afanasiev D, Hortensius J R, Ivanov B A, et al. Ultrafast control of magnetic interactions via light-driven phonons[J]. *Nature Materials*, 2021, 20(5): 607-611.
- [70] Schlauderer S, Lange C, Baierl S, et al. Temporal and spectral fingerprints of ultrafast all-coherent spin switching[J]. *Nature*, 2019, 569(7756): 383-387.
- [71] Zhao H C, Xia H, Hu S, et al. Large ultrafast-modulated Voigt effect in noncollinear antiferromagnet Mn_3Sn [J]. *Nature Communications*, 2021, 12: 5266.
- [72] Chen Q, Ding Q Y, Wang Y T, et al. Electronic and magnetic properties of a two-dimensional transition metal phosphorous chalcogenide TMPS_4 [J]. *The Journal of Physical Chemistry C*, 2020, 124(22): 12075-12080.
- [73] Kim K, Lim S Y, Lee J U, et al. Suppression of magnetic ordering in XXZ-type antiferromagnetic monolayer NiPS_3 [J]. *Nature Communications*, 2019, 10: 345.
- [74] Lançon D, Walker H C, Ressouche E, et al. Magnetic structure and magnon dynamics of the quasi-two-dimensional antiferromagnet FePS_3 [J]. *Physical Review B*, 2016, 94(21): 214407.
- [75] Sun Y J, Tan Q H, Liu X L, et al. Probing the magnetic ordering of antiferromagnetic MnPS_3 by Raman spectroscopy[J]. *The Journal of Physical Chemistry Letters*, 2019, 10(11): 3087-3093.

- [76] 肖寒, 弭孟娟, 王以林. 二维磁性材料及多场调控研究进展[J]. 物理学报, 2021, 70(12): 127503.
- Xiao H, Mi M J, Wang Y L. Recent development in two-dimensional magnetic materials and multi-field control of magnetism[J]. Acta Physica Sinica, 2021, 70(12): 127503.
- [77] Feng W X, Hanke J P, Zhou X D, et al. Topological magneto-optical effects and their quantization in noncoplanar antiferromagnets[J]. Nature Communications, 2020, 11: 118.
- [78] Huang B, Cenker J, Zhang X O, et al. Tuning inelastic light scattering via symmetry control in the two-dimensional magnet CrI₃ [J]. Nature Nanotechnology, 2020, 15(3): 212-216.
- [79] Sun Y J, Pang S M, Zhang J. Review of Raman spectroscopy of two-dimensional magnetic van der Waals materials[J]. Chinese Physics B, 2021, 30(11): 117104.
- [80] Chu H, Roh C J, Island J O, et al. Linear magnetoelectric phase in ultrathin MnPS₃ probed by optical second harmonic generation [J]. Physical Review Letters, 2020, 124(2): 027601.
- [81] Belvin C A, Baldini E, Ozel I O, et al. Exciton-driven antiferromagnetic metal in a correlated van der Waals insulator[J]. Nature Communications, 2021, 12: 4837.
- [82] Matsuda T, Kanda N, Higo T, et al. Room-temperature terahertz anomalous Hall effect in Weyl antiferromagnet Mn₃Sn thin films[J]. Nature Communications, 2020, 11: 909.
- [83] Mertins H C, Valencia S, Gaupp A, et al. Magneto-optical polarization spectroscopy with soft X-rays[J]. Applied Physics A, 2005, 80(5): 1011-1020.
- [84] Zhang W H, Qi Q Q, Zhou J, et al. Mimicking faraday rotation to sort the orbital angular momentum of light[J]. Physical Review Letters, 2014, 112(15): 153601.
- [85] Argyres P N. Theory of the faraday and kerr effects in ferromagnetics[J]. Physical Review, 1955, 97(2): 334-345.
- [86] Siegrist F, Gessner J A, Osslander M, et al. Light-wave dynamic control of magnetism[J]. Nature, 2019, 571(7764): 240-244.
- [87] Xu J, Zhou C, Jia M W, et al. Imaging antiferromagnetic domains in nickel oxide thin films by optical birefringence effect[J]. Physical Review B, 2019, 100(13): 134413.
- [88] Zhang H Q, Ni Z L, Stevens C E, et al. Cavity-enhanced linear dichroism in a van der Waals antiferromagnet[J]. Nature Photonics, 2022, 16(4): 311-317.
- [89] Zhang T L, Wang Y M, Li H X, et al. Magnetism and optical anisotropy in van der Waals antiferromagnetic insulator CrOCl[J]. ACS Nano, 2019, 13(10): 11353-11362.
- [90] Lee K, Dismukes A H, Telford E J, et al. Magnetic order and symmetry in the 2D semiconductor CrSBr[J]. Nano Letters, 2021, 21(8): 3511-3517.
- [91] Zhang Y, Holder T, Ishizuka H, et al. Switchable magnetic bulk photovoltaic effect in the two-dimensional magnet CrI₃[J]. Nature Communications, 2019, 10: 3783.
- [92] Long G, Zhang T, Cai X B, et al. Isolation and characterization of few-layer manganese thiophosphite[J]. ACS Nano, 2017, 11(11): 11330-11336.
- [93] Morimoto T, Nagaosa N. Shift current from electromagnon excitations in multiferroics[J]. Physical Review B, 2019, 100(23): 235138.
- [94] Song T C, Anderson E, Tu M W Y, et al. Spin photovoltaic effect in magnetic van der Waals heterostructures[J]. Science Advances, 2021, 7(36): eabg8094.
- [95] Watanabe H, Yanase Y. Chiral photocurrent in parity-violating magnet and enhanced response in topological antiferromagnet[J]. Physical Review X, 2021, 11(1): 011001.
- [96] Kumar R, Jenjeti R N, Austeria M P, et al. Bulk and few-layer MnPS₃: a new candidate for field effect transistors and UV photodetectors[J]. Journal of Materials Chemistry C, 2019, 7(2): 324-329.
- [97] Xu T F, Luo M, Shen N M, et al. Ternary 2D layered material FePSe₃ and near-infrared photodetector[J]. Advanced Electronic Materials, 2021, 7(8): 2100207.
- [98] Ni Z L, Zhang H Q, Hopper D A, et al. Direct imaging of antiferromagnetic domains and anomalous layer-dependent mirror symmetry breaking in atomically thin MnPS₃[J]. Physical Review Letters, 2021, 127(18): 187201.
- [99] Sun Z Y, Yi Y F, Song T C, et al. Giant nonreciprocal second-harmonic generation from antiferromagnetic bilayer CrI₃[J]. Nature, 2019, 572(7770): 497-501.
- [100] Wang X Z, Cao J, Li H, et al. Electronic Raman scattering in the 2D antiferromagnet NiPS₃[J]. Science Advances, 2022, 8(2): eab17707.
- [101] Kargar F, Coleman E A, Ghosh S, et al. Phonon and thermal properties of quasi-two-dimensional FePS₃ and MnPS₃ antiferromagnetic semiconductors[J]. ACS Nano, 2020, 14(2): 2424-2435.
- [102] Song C, You Y F, Chen X Z, et al. How to manipulate magnetic states of antiferromagnets[J]. Nanotechnology, 2018, 29(11): 112001.
- [103] 蒋小红, 秦泗晨, 幸子越, 等. 二维磁性材料的物性研究及性能调控[J]. 物理学报, 2021, 70(12): 127801.
- Jiang X H, Qin S C, Xing Z Y, et al. Study on physical properties and magnetism controlling of two-dimensional magnetic materials [J]. Acta Physica Sinica, 2021, 70(12): 127801.
- [104] Song T C, Cai X H, Tu M W Y, et al. Giant tunneling magnetoresistance in spin-filter van der Waals heterostructures[J]. Science, 2018, 360(6394): 1214-1218.
- [105] Huang B, Clark G, Klein D R, et al. Electrical control of 2D magnetism in bilayer CrI₃[J]. Nature Nanotechnology, 2018, 13(7): 544-548.
- [106] Song T C, Fei Z Y, Yankowitz M, et al. Switching 2D magnetic states via pressure tuning of layer stacking[J]. Nature Materials, 2019, 18(12): 1298-1302.
- [107] Li T X, Jiang S W, Sivasdas N, et al. Pressure-controlled interlayer magnetism in atomically thin CrI₃[J]. Nature Materials, 2019, 18(12): 1303-1308.
- [108] León A M, González J W, Mejía-López J, et al. Strain-induced phase transition in CrI₃ bilayers[J]. 2D Materials, 2020, 7(3): 035008.
- [109] Ni Z L, Haglund A V, Wang H, et al. Imaging the Néel vector switching in the monolayer antiferromagnet MnPSe₃ with strain-controlled Ising order[J]. Nature Nanotechnology, 2021, 16(7): 782-787.
- [110] Matsuoka T, Haglund A, Xue R, et al. Pressure-induced insulator-metal transition in two-dimensional Mott insulator NiPS₃ [J]. Journal of the Physical Society of Japan, 2021, 90(12): 124706.
- [111] Coak M J, Jarvis D M, Hamidov H, et al. Emergent magnetic phases in pressure-tuned van der Waals antiferromagnet FePS₃[J]. Physical Review X, 2021, 11(1): 011024.
- [112] Wang Y G, Ying J J, Zhou Z Y, et al. Emergent superconductivity in an iron-based honeycomb lattice initiated by pressure-driven spin-crossover[J]. Nature Communications, 2018, 9: 1914.
- [113] 何聪丽, 许洪军, 汤建, 等. 基于二维材料的自旋-轨道矩研究进展[J]. 物理学报, 2021, 70(12): 127501.
- He C L, Xu H J, Tang J, et al. Research progress of spin-orbit torques based on two-dimensional materials[J]. Acta Physica Sinica, 2021, 70(12): 127501.
- [114] Ma X C, Tian Y, Zhao P, et al. Janus MoCrSSe monolayer: a strong two dimensional polar antiferromagnet[J]. Applied Surface Science, 2022, 581: 152420.
- [115] Zhu R, Zhang W, Shen W, et al. Exchange bias in van der Waals CrCl₃/Fe₃GeTe₂ heterostructures[J]. Nano Letters, 2020, 20(7): 5030-5035.
- [116] Zhang L M, Huang X Y, Dai H W, et al. Proximity-coupling-induced significant enhancement of coercive field and curie temperature in 2D van der Waals heterostructures[J]. Advanced Materials, 2020, 32(38): 2002032.
- [117] Liu S S, Yang K, Liu W Q, et al. Two-dimensional ferromagnetic superlattices[J]. National Science Review, 2019, 7(4): 745-754.

- [118] Dai H W, Cheng H, Cai M H, et al. Enhancement of the coercive field and exchange bias effect in $\text{Fe}_3\text{GeTe}_2/\text{MnPX}_3$ ($X = \text{S}$ and Se) van der Waals heterostructures[J]. *ACS Applied Materials & Interfaces*, 2021, 13(20): 24314-24320.
- [119] Yang C Y, Pan L, Grutter A J, et al. Termination switching of antiferromagnetic proximity effect in topological insulator[J]. *Science Advances*, 2020, 6(33): eaaz8463.
- [120] Zhong D, Seyler K L, Linpeng X Y, et al. Layer-resolved magnetic proximity effect in van der Waals heterostructures[J]. *Nature Nanotechnology*, 2020, 15(3): 187-191.
- [121] Zollner K, Faria P E, Jr, Fabian J. Proximity exchange effects in MoSe_2 and WSe_2 heterostructures with CrI_3 : twist angle, layer, and gate dependence[J]. *Physical Review B*, 2019, 100(8): 085128.
- [122] Ciorciaro L, Kroner M, Watanabe K, et al. Observation of magnetic proximity effect using resonant optical spectroscopy of an electrically tunable $\text{MoSe}_2/\text{CrBr}_3$ heterostructure[J]. *Physical Review Letters*, 2020, 124(19): 197401.
- [123] Onga M, Sugita Y, Ideue T, et al. Antiferromagnet-semiconductor van der Waals heterostructures: interlayer interplay of exciton with magnetic ordering[J]. *Nano Letters*, 2020, 20(6): 4625-4630.
- [124] Subhan F, Hong J S. Large valley splitting and enhancement of curie temperature in a two-dimensional VI_3/CrI_3 heterostructure[J]. *The Journal of Physical Chemistry C*, 2020, 124(13): 7156-7162.
- [125] Yang K, Hu W T, Wu H, et al. Magneto-optical kerr switching properties of $(\text{CrI}_3)_2$ and $(\text{CrBr}_3/\text{CrI}_3)$ bilayers[J]. *ACS Applied Electronic Materials*, 2020, 2(5): 1373-1380.
- [126] Kirilyuk A, Kimel A V, Rasing T. Ultrafast optical manipulation of magnetic order[J]. *Reviews of Modern Physics*, 2010, 82(3): 2731-2784.
- [127] Tauchert S R, Volkov M, Ehberger D, et al. Polarized phonons carry angular momentum in ultrafast demagnetization[J]. *Nature*, 2022, 602(7895): 73-77.
- [128] Beaurepaire E, Merle J C, Daunois A, et al. Ultrafast spin dynamics in ferromagnetic nickel[J]. *Physical Review Letters*, 1996, 76(22): 4250-4253.
- [129] 任乃飞, 许美玲, 顾佳方, 等. 飞秒激光作用下铁磁薄膜的热化动力学分析[J]. *中国激光*, 2010, 37(8): 2057-2062.
- Ren N F, Xu M L, Gu J F, et al. Analysis of thermalization dynamics on ferromagnetic thin film excited by femtosecond laser[J]. *Chinese Journal of Lasers*, 2010, 37(8): 2057-2062.
- [130] 苏玉伦, 尉正行, 程亮, 等. 基于超快自旋-电荷转换的太赫兹辐射源[J]. *物理学报*, 2020, 69(20): 204202.
- Su Y L, Wei Z X, Cheng L, et al. Terahertz emitters based on ultrafast spin-to-charge conversion[J]. *Acta Physica Sinica*, 2020, 69(20): 204202.
- [131] Radu I, Vahaplar K, Stamm C, et al. Transient ferromagnetic-like state mediating ultrafast reversal of antiferromagnetically coupled spins[J]. *Nature*, 2011, 472(7342): 205-208.
- [132] Mikhaylovskiy R V, Hendry E, Kruglyak V V, et al. Terahertz emission spectroscopy of laser-induced spin dynamics in TmFeO_3 and ErFeO_3 orthoferrites[J]. *Physical Review B*, 2014, 90(18): 184405.
- [133] Kašpar Z, Surýnek M, Zubáč J, et al. Quenching of an antiferromagnet into high resistivity states using electrical or ultrashort optical pulses[J]. *Nature Electronics*, 2021, 4(1): 30-37.
- [134] Zhang X X, Jiang S W, Lee J, et al. Spin dynamics slowdown near the antiferromagnetic critical point in atomically thin FePS_3 [J]. *Nano Letters*, 2021, 21(12): 5045-5052.
- [135] Bozhko D A, Vasyuchka V I, Chumak A V, et al. Magnon-phonon interactions in magnon spintronics (Review article)[J]. *Low Temperature Physics*, 2020, 46(4): 383-399.
- [136] Cenker J, Huang B, Suri N, et al. Direct observation of two-dimensional magnons in atomically thin CrI_3 [J]. *Nature Physics*, 2021, 17(1): 20-25.
- [137] McCreary A, Simpson J R, Mai T T, et al. Quasi-two-dimensional magnon identification in antiferromagnetic FePS_3 via magneto-Raman spectroscopy[J]. *Physical Review B*, 2020, 101(6): 064416.
- [138] Ivanov B A. Spin dynamics of antiferromagnets under action of femtosecond laser pulses (Review article)[J]. *Low Temperature Physics*, 2014, 40(2): 91-105.
- [139] Rubano A, Satoh T, Kimel A, et al. Influence of laser pulse shaping on the ultrafast dynamics in antiferromagnetic NiO [J]. *Physical Review B*, 2010, 82(17): 174431.
- [140] Jin W C, Kim H H, Ye Z P, et al. Raman fingerprint of two terahertz spin wave branches in a two-dimensional honeycomb Ising ferromagnet[J]. *Nature Communications*, 2018, 9: 5122.
- [141] Zhang X X, Li L Z, Weber D, et al. Gate-tunable spin waves in antiferromagnetic atomic bilayers[J]. *Nature Materials*, 2020, 19(8): 838-842.
- [142] Ho C H, Hsu T Y, Muhimma L C. The band-edge excitons observed in few-layer NiPS_3 [J]. *npj 2D Materials and Applications*, 2021, 5: 8.
- [143] Afanasiev D, Hortensius J R, Matthiesen M, et al. Controlling the anisotropy of a van der Waals antiferromagnet with light[J]. *Science Advances*, 2021, 7(23): eabf3096.
- [144] Nova T F, Cartella A, Cantaluppi A, et al. An effective magnetic field from optically driven phonons[J]. *Nature Physics*, 2017, 13(2): 132-136.
- [145] Ghosh A, Palit M, Maity S, et al. Spin-phonon coupling and magnon scattering in few-layer antiferromagnetic FePS_3 [J]. *Physical Review B*, 2021, 103(6): 064431.
- [146] Lee K, Lee D K, Yang D S, et al. Superluminal-like magnon propagation in antiferromagnetic NiO at nanoscale distances[J]. *Nature Nanotechnology*, 2021, 16(12): 1337-1341.
- [147] Xing W Y, Qiu L Y, Wang X R, et al. Magnon transport in quasi-two-dimensional van der Waals antiferromagnets[J]. *Physical Review X*, 2019, 9(1): 011026.
- [148] Cornelissen L J, Liu J, Duine R A, et al. Long-distance transport of magnon spin information in a magnetic insulator at room temperature[J]. *Nature Physics*, 2015, 11(12): 1022-1026.
- [149] Bae Y J, Wang J, Xu J W, et al. Exciton-coupled coherent magnons in a 2D semiconductor[J]. *Nature*, 2022, 609(7926): 282-286.
- [150] Hortensius J R, Afanasiev D, Matthiesen M, et al. Coherent spin-wave transport in an antiferromagnet[J]. *Nature Physics*, 2021, 17(9): 1001-1006.
- [151] Zhao C, Norden T, Zhang P Y, et al. Enhanced valley splitting in monolayer WSe_2 due to magnetic exchange field[J]. *Nature Nanotechnology*, 2017, 12(8): 757-762.
- [152] Jin W C, Kim H H, Ye Z P, et al. Observation of the polaronic character of excitons in a two-dimensional semiconducting magnet CrI_3 [J]. *Nature Communications*, 2020, 11: 4780.
- [153] Wu M, Li Z L, Cao T, et al. Physical origin of giant excitonic and magneto-optical responses in two-dimensional ferromagnetic insulators[J]. *Nature Communications*, 2019, 10: 2371.
- [154] Acharya S, Pashov D, Rudenko A N, et al. Real- and momentum-space description of the excitons in bulk and monolayer chromium tri-halides[J]. *npj 2D Materials and Applications*, 2022, 6: 33.
- [155] Kim S Y, Kim T Y, Sandilands L J, et al. Charge-spin correlation in van der Waals antiferromagnet NiPS_3 [J]. *Physical Review Letters*, 2018, 120(13): 136402.
- [156] Hwangbo K, Zhang Q, Jiang Q N, et al. Highly anisotropic excitons and multiple phonon bound states in a van der Waals antiferromagnetic insulator[J]. *Nature Nanotechnology*, 2021, 16(6): 655-660.
- [157] Ergecen E, Ilyas B, Mao D, et al. Magnetically brightened dark electron-phonon bound states in a van der Waals antiferromagnet[J]. *Nature Communications*, 2022, 13: 98.
- [158] Son S, Lee Y J, Kim J H, et al. Multiferroic-enabled magnetic-excitons in 2D quantum-entangled van der Waals antiferromagnet NiI_2 [J]. *Advanced Materials*, 2022, 34(10): 2109144.
- [159] Birowska M, Faria P E, Jr, Fabian J, et al. Large exciton binding energies in MnPS_3 as a case study of a van der Waals layered

- magnet[J]. *Physical Review B*, 2021, 103(12): L121108.
- [160] Gu P F, Tan Q H, Wan Y, et al. Photoluminescent quantum interference in a van der Waals magnet preserved by symmetry breaking[J]. *ACS Nano*, 2020, 14(1): 1003-1010.
- [161] Tang Y X, Zhang Y B, Ouyang H, et al. Ultrafast response of a hybrid device based on strongly coupled monolayer WS_2 and photonic crystals: the effect of photoinduced coulombic screening [J]. *Laser & Photonics Reviews*, 2020, 14(4): 1900419.
- [162] Wang Y M, Zhang J F, Li C H, et al. Raman scattering study of magnetic layered MPS_3 crystals ($M = Mn, Fe, Ni$) [J]. *Chinese Physics B*, 2019, 28(5): 056301.
- [163] Ma B W, Fiete G A. Antiferromagnetic insulators with tunable magnon-polaron Chern numbers induced by in-plane optical phonons[J]. *Physical Review B*, 2022, 105(10): L100402.
- [164] Liu S, Granados Del Águila A, Bhowmick D, et al. Direct observation of magnon-phonon strong coupling in two-dimensional antiferromagnet at high magnetic fields[J]. *Physical Review Letters*, 2021, 127(9): 097401.
- [165] Vaclavkova D, Palit M, Wyzula J, et al. Magnon polarons in the van der Waals antiferromagnet $FePS_3$ [J]. *Physical Review B*, 2021, 104(13): 134437.
- [166] Zhang Q, Ozerov M, Boström E V, et al. Coherent strong-coupling of terahertz magnons and phonons in a van der Waals antiferromagnetic insulator [EB/OL]. (2021-08-26) [2022-02-05]. <https://arxiv.org/abs/2108.11619>.
- [167] Sun Y J, Lai J M, Pang S M, et al. Magneto-Raman study of magnon-phonon coupling in two-dimensional Ising antiferromagnetic $FePS_3$ [J]. *The Journal of Physical Chemistry Letters*, 2022, 13(6): 1533-1539.
- [168] Mai T T, Garrity K F, McCreary A, et al. Magnon-phonon hybridization in 2D antiferromagnet $MnPS_3$ [J]. *Science Advances*, 2021, 7(44): eabj3106.
- [169] MacNeill D, Hou J T, Klein D R, et al. Gigahertz frequency antiferromagnetic resonance and strong magnon-magnon coupling in the layered crystal $CrCl_3$ [J]. *Physical Review Letters*, 2019, 123(4): 047204.
- [170] Zhang Q, Xue J S, Sun Y T, et al. Coupling of microwave photons to optical and acoustic magnon modes in the layered antiferromagnetic insulator $CrCl_3$ [J]. *Physical Review B*, 2021, 104(9): 094303.
- [171] Rückriegel A, Duine R A. Long-range phonon spin transport in ferromagnet-nonmagnetic insulator heterostructures [J]. *Physical Review Letters*, 2020, 124(11): 117201.
- [172] Kudlacik D, Ivanov V Y, Yakovlev D R, et al. Exciton and exciton-magnon photoluminescence in the antiferromagnet CuB_2O_4 [J]. *Physical Review B*, 2020, 102(3): 035128.
- [173] Gloppe A, Onga M, Hisatomi R, et al. Magnon-exciton proximity coupling at a van der Waals heterointerface [J]. *Physical Review B*, 2022, 105(12): L121403.
- [174] Sato T, Abe N, Kimura S, et al. Magnetochiral dichroism in a collinear antiferromagnet with No magnetization [J]. *Physical Review Letters*, 2020, 124(21): 217402.
- [175] Atzori M, Santanni F, Breslavetz I, et al. Magnetic anisotropy drives magnetochiral dichroism in a chiral molecular helix probed with visible light [J]. *Journal of the American Chemical Society*, 2020, 142(32): 13908-13916.
- [176] Chen W, Sun Z Y, Wang Z J, et al. Direct observation of van der Waals stacking-dependent interlayer magnetism [J]. *Science*, 2019, 366(6468): 983-987.
- [177] Thiel L, Wang Z, Tschudin M A, et al. Probing magnetism in 2D materials at the nanoscale with single-spin microscopy [J]. *Science*, 2019, 364(6444): 973-976.
- [178] Scholl P, Schuler M, Williams H J, et al. Quantum simulation of 2D antiferromagnets with hundreds of Rydberg atoms [J]. *Nature*, 2021, 595(7866): 233-238.
- [179] Cheong S W, Fiebig M, Wu W D, et al. Seeing is believing: visualization of antiferromagnetic domains [J]. *npj Quantum Materials*, 2020, 5: 3.
- [180] 马志军, 魏荣妃, 胡忠亮, 等. 2D材料和准2D材料的非线性光学特性及应用 [J]. *中国激光*, 2017, 44(7): 0703002.
- Ma Z J, Wei R F, Hu Z L, et al. 2D materials and quasi-2D materials: nonlinear optical properties and corresponding applications [J]. *Chinese Journal of Lasers*, 2017, 44(7): 0703002.
- [181] Fina I, Marti X, Yi D, et al. Anisotropic magnetoresistance in an antiferromagnetic semiconductor [J]. *Nature Communications*, 2014, 5: 4671.
- [182] Wang Z, Gutiérrez-Lezama I, Ubrig N, et al. Very large tunneling magnetoresistance in layered magnetic semiconductor CrI_3 [J]. *Nature Communications*, 2018, 9: 2516.
- [183] Gomonay O, Baltz V, Brataas A, et al. Antiferromagnetic spin textures and dynamics [J]. *Nature Physics*, 2018, 14(3): 213-216.
- [184] Fan F, Zhao D, Tan Z Y, et al. Magnetically induced terahertz birefringence and chirality manipulation in transverse-magnetized metasurface [J]. *Advanced Optical Materials*, 2021, 9(24): 2101097.
- [185] Galkina E G, Ivanov B A. Dynamic solitons in antiferromagnets (review article) [J]. *Low Temperature Physics*, 2018, 44(7): 618-633.
- [186] Qaiumzadeh A, Skarsvåg H, Holmqvist C, et al. Spin superfluidity in biaxial antiferromagnetic insulators [J]. *Physical Review Letters*, 2017, 118(13): 137201.
- [187] Jiao Y L, Wu W K, Ma F X, et al. Room temperature ferromagnetism and antiferromagnetism in two-dimensional iron arsenides [J]. *Nanoscale*, 2019, 11(35): 16508-16514.
- [188] Wu D X, Zhuo Z W, Lü H F, et al. Two-dimensional $Cr_2X_3S_3$ ($X = Br, I$) Janus semiconductor with intrinsic room-temperature magnetism [J]. *The Journal of Physical Chemistry Letters*, 2021, 12(11): 2905-2911.
- [189] Yang X X, Zhou X D, Feng W X, et al. Tunable magneto-optical effect, anomalous Hall effect, and anomalous Nernst effect in the two-dimensional room-temperature ferromagnet $1T-CrTe_2$ [J]. *Physical Review B*, 2021, 103(2): 024436.

Progress of Research on Two-dimensional Antiferromagnets with Magneto-optic Properties

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Abstract

Significance Semiconductor microelectronics, which are based on the charge properties of electrons, have brought about revolutionary advancements to modern information technology. However, necessary improvements in the computing and information-processing capabilities of devices cannot only be achieved by manipulating and optimizing the charge properties of electrons. The manipulation of electronic spins, one of the most basic characteristics of magnetic materials, has enabled magnetic devices to become considerably successful, resulting in a series of cutting-edge applications with the advantages of nonvolatility and low-calorie requirements.

Antiferromagnets, magnetic materials with the internal magnetic moment offset to zero, exhibit fascinating physical properties and have high application potential. First, the spin precession frequency resonant at the terahertz band of antiferromagnets has a higher spin storage density than that of ferromagnets. Second, the stray field of antiferromagnets is almost zero, which provides a strong ability to resist external interference. Third, antiferromagnets are often accompanied by many complex electronic states that give rise to some novel effects. However, owing to the characteristics of weak or zero net magnetic moments, the detection and regulation of antiferromagnetic materials have been challenging for a long time.

In recent years, ultrathin and monolayer-exfoliated two-dimensional materials have provided new opportunities for research on antiferromagnets. The low-dimensional scale has improved the interaction among the lattice, electronic spin, and charge, which not only gives rise to complex and rich magnetic states but also paves the way for exploring low-dimensional magnetism and its applications by using interdisciplinary research fields such as optoelectronics. Research based on optical means has many advantages such as enabling microscopic-level, high-speed, noncontact, high-sensitivity analyses with a high space-time and high-energy resolution. In addition, it is conducive to the observation of various magnetic responses of antiferromagnets under extreme physical conditions. The developments of magneto-optical principles and optical detection technology have resulted in the use of various laser spectrum and polarization detection schemes to solve the challenges presented by antiferromagnetic material research. Therefore, it is necessary to summarize research advances on two-dimensional antiferromagnets in the existing magneto-optical field.

Progress This paper reviews the progress in the field of magneto-optical research pertaining to antiferromagnets from the perspectives of optical characterization and regulation. After introduction of the basic principles, the main research objects of the interactions between laser and two-dimensional antiferromagnets are extended to the forms of the macroscopic magneto-optical effect and microscopic elementary excitation quasiparticle, respectively.

After a brief introduction in Section 1, the basic structure, properties, and classification of typical van der Waals antiferromagnets, including chromium trihalide and transition metal phosphorus sulfide (Fig. 2), are introduced in Section 2, along with some other common materials (Table 1). Starting from different magnetic coupling characteristics (interlayer or intralayer antiferromagnetic coupling), the magnetic ordering of these antiferromagnetic materials in combination with the molecular configuration and chemical composition are reviewed.

In Section 3, multiple magneto-optical effects are discussed for magnetic thin films (Fig. 3). In addition to the well-known Faraday effect, magneto-optical Kerr effect, Zeeman effect, magnetic dichroism, and Voigt and Cotton-Mouton effects, various specific magnetic changes in matters resulting from the interactions between light and matters are also included. For example, two-dimensional FePS₃ exhibits giant linear dichroism because the destruction of the rotational symmetry by the antiferromagnetic order renders the electron energy band anisotropic (Fig. 4). The spin photovoltaic effect is demonstrated in multilayer CrI₃, the interlayer magnetic order directly affects the magnitudes of the photocurrent and tunneling current in the direction vertical to the heterojunction. The circular polarization of the photocurrent also reveals a correlation between the magnetic state and the photon energy (Fig. 5). In terms of spectral detection, the second-

harmonic emission and scattering spectra that are closely related to the magnetic order are also reviewed (Fig. 6). Subsequently, the ability of the magneto-optical effect to clearly reflect the regulation of two-dimensional antiferromagnets by means of magnetic field, electric field, temperature field, stress regulation (Fig. 7), passive regulation, and ultrafast lasers is discussed (Fig. 8).

In the spectral research presented in Section 4, as a link between macro- and micro-quantum phenomena, elementary excitation quasiparticles are shown to greatly facilitate related research on low-dimensional condensed matter physics. Under antiferromagnetic conditions, the disturbance of the magnetic order may affect the properties and mutual coupling of various quasiparticles in the material. The magnons produced by the collective excitation of the laser to the spintronic system facilitate the loading of electrical information, and their frequency determines the switching speed of the spintronic devices. For applications, the energy of the magnons can be adjusted using an all-optical method and an electrostatic doping method controlled by the gate voltage (Fig. 9). On the other hand, research on the transport of magnons in two-dimensional antiferromagnetic materials has also been conducted and these transport phenomena could potentially be studied by all-optical imaging. Research on the same Bosonic excitons in two-dimensional antiferromagnets is another important research topic. The Frenkel-like excitons in CrI_3 have circular-polarized and polaronic characteristics (Fig. 10), while the Wannier-like excitons in NiPS_3 prefer linear polarization and high coherence in the emission spectra (Fig. 11). For another low-energy excited Bosonic phonon, strong coupling magnon-phonon states are formed through the tuning effect of a large magnetic field (Fig. 12). The dispersive anti-cross feature of this polaron state is clearly reflected in the spectra, which can provide a new research platform for the magneto-optical control of antiferromagnetic materials.

Section 5 summarizes the prospects for further research and the application of two-dimensional antiferromagnets in magneto-optical and related fields.

Conclusions and Prospects Compared with ferromagnets, the unique advantages of antiferromagnets in magneto-optical research are undoubtedly expected to accelerate research in spintronics-related fields. Research on antiferromagnets based on the magneto-optical effect is foreseen to focus on more complex and extreme systems in non-collinear, helical, topological, multiferroic, spin-liquid magnetic states, etc. The control of these magnetic states would enable interlayer electronic coupling to be effectively adjusted to facilitate the detection, manipulation, emission, and tracking of spin information through optical means. This approach could introduce huge optical nonlinearity, efficient spin filtering, high conversion efficiency between charge current and spin current, high electron mobility, long spin diffusion length, and other characteristics, and expand the research scope of magnetism, photonics, and other interdisciplinary disciplines. Although the ability to conduct antiferromagnetic writing and reading under two-dimensional conditions would also need to be fundamentally improved, future breakthroughs in two-dimensional antiferromagnetic opto-spintronics can certainly be anticipated.

Key words materials; two-dimensional antiferromagnet; spin; magneto-optic effect; elementary excitation quasiparticle