

“西光所建所六十周年暨《光子学报》创刊五十周年”专辑

引用格式: JIN Zuanming, GUO Yingyu, JI Bingyu, et al. Development of Ultrafast Spin-based Terahertz Photonics(Invited)[J]. Acta Photonica Sinica, 2022, 51(7):0751410

金钻明,郭颖钰,季秉煜,等. 超快太赫兹自旋光电子学研究进展(特邀)[J]. 光子学报, 2022, 51(7):0751410

超快太赫兹自旋光电子学研究进展(特邀)

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摘 要: 太赫兹科学技术在光谱、成像、传感、生物医药、安全检测等方面展现出了巨大的应用潜力和价值。基于新材料和新机理, 研发高效、超宽带和低成本的太赫兹光子学器件是太赫兹科学技术的重要挑战。近年来的研究表明, 太赫兹光子学和超快自旋电子学深度交叉, 获得了很大的关注。本文对超快太赫兹自旋光电子学所研究的物理机理和器件设计应用进行讨论。在物理机理研究方面, 阐明了太赫兹脉冲为研究超快自旋电子学提供强大工具, 实现了太赫兹驱动自旋波, 探测自旋输运和超快磁测量。在器件设计与应用方面, 介绍了基于自旋的新型太赫兹光子学器件, 包括自旋太赫兹辐射源的优化方法, 自旋太赫兹调制器的工作原理, 自旋太赫兹探测器的设计方案。超快太赫兹自旋光电子学不仅有助于人们理解宏观自旋电子学现象背后的微观物理机制, 而且有望实现高效的太赫兹光子学器件和光谱学应用。

关键词: 太赫兹; 自旋电子学; 超快光谱; 太赫兹产生和调控

中图分类号: TN241

文献标识码: A

doi: 10.3788/gzxb20225107.0751410

0 引言

太赫兹波指的是 0.1~10 THz (1 THz=10¹² Hz) 的电磁辐射, 介于微波毫米波与光学红外线之间, 处于电子学向光子学的过渡区域。太赫兹波具有良好的时间和空间相干性, 光子能量低, 传输容量大, 穿透力强, 覆盖许多模式的本征频率^[1-2]。这些特点使得太赫兹波在许多重要应用领域, 如通信传感、精密光谱、雷达成像、生物医学检测、国防安检等, 有着巨大的科学和应用价值^[3-5]。太赫兹科学技术是物理学、光学和电子学等学科的研究焦点^[6-13]。以太赫兹安检、太赫兹成像和太赫兹光谱分析为主的太赫兹技术有望成为极具竞争力的新兴产业。

典型的相干太赫兹宽频辐射脉冲宽度小于 1 ps, 表现出恒定的载波-包络相位。电场强度可超过 1 MV/cm, 相应的磁场强度为 0.33 T^[14]。基于抽运-探测技术, 可以构建太赫兹时域光谱^[15], 太赫兹发射光谱^[16], 光抽运-太赫兹探测光谱^[17-18], 太赫兹抽运-光探测光谱^[19]和太赫兹抽运-太赫兹探测光谱^[20]等。利用太赫兹时域光谱, 可有效研究太赫兹电磁脉冲与准粒子(比如磁子、声子、等离子激元)以及带内电子跃迁的相互作用^[14-20]。时间分辨的太赫兹光谱技术通过无接触的方式, 准确提取二维材料^[21]、钙钛矿太阳能材料^[22]、超导体^[23]、半金属^[24]在内的各种量子材料的载流子寿命、迁移率、浓度和复电导率等参数。实现不同材料中光诱导激发态(包括激子跃迁^[25], 热极化子等^[26])时间演化过程的可视化。

基金项目: 国家自然科学基金(Nos. 61988102, 61975110, 61735010), 111 项目(No. D18014), 上海市科委国际联合实验室项目(No. 17590750300), 上海市科委重点项目(No. YDZX20193100004960), 上海市科学技术委员会科技创新行动计划项目(No. 21JC1402600), 上海市青年科技启明星计划(No. 18QA1401700), 上海市教育委员会和上海市教育发展基金会“晨光计划”(No. 16CG45), 上海高校青年东方学者计划(No. QD2015020)

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收稿日期: 2022-05-19; **录用日期:** 2022-06-13

<http://www.photon.ac.cn>

上述关于太赫兹光谱的应用中,宽频太赫兹相干辐射主要是通过超快光子学的方法产生,包括:光整流^[27-28],光电导天线^[29-30],和空气等离子体^[31-34]等。基于光整流效应太赫兹辐射的频谱宽度受限于晶体本征吸收(自由载流子和相干声子)以及相位匹配条件。光电导天线可产生毫瓦级太赫兹辐射,但器件的激光损伤阈值低,使用寿命较短。空气等离子体的太赫兹宽频辐射需要使用放大级飞秒激光脉冲,脉冲的低频分量缺失,稳定性较差,难以实现太赫兹光谱系统的小型化。基于上述超快光子学太赫兹辐射源的技术特点和瓶颈,亟需探索新的材料结构和新的物理机制,实现新型的太赫兹辐射源、调制器和探测器等功能器件的研发。

近20年来,科学家利用飞秒激光对电子的自旋超快动力学进行了广泛而深入的研究。研究飞秒时间尺度上自旋波和自旋流的激发、调控和探测,形成了超快自旋电子学^[35-40]。值得注意的是,法国科学家BEAUREPAIRE E等发现超快退磁过程中伴随着太赫兹波的产生^[41]。德国科学家KAMPFRATH T等证实太赫兹脉冲是自旋波激发与相干调控的有效手段^[42]。越来越多的理论和实验工作表明,超快自旋电子学与太赫兹光子学密切相关^[43],形成新的研究领域—超快太赫兹自旋光子学,研究框架如图1所示。一方面,太赫兹脉冲为研究超快自旋电子学提供强大工具,可实现太赫兹驱动自旋波,探测自旋输运和超快磁测量。另一方面,通过探索太赫兹自旋电子学效应,逐步实现新型的基于超快电子自旋的太赫兹光子学器件,包括:太赫兹辐射源,调制器和探测器。

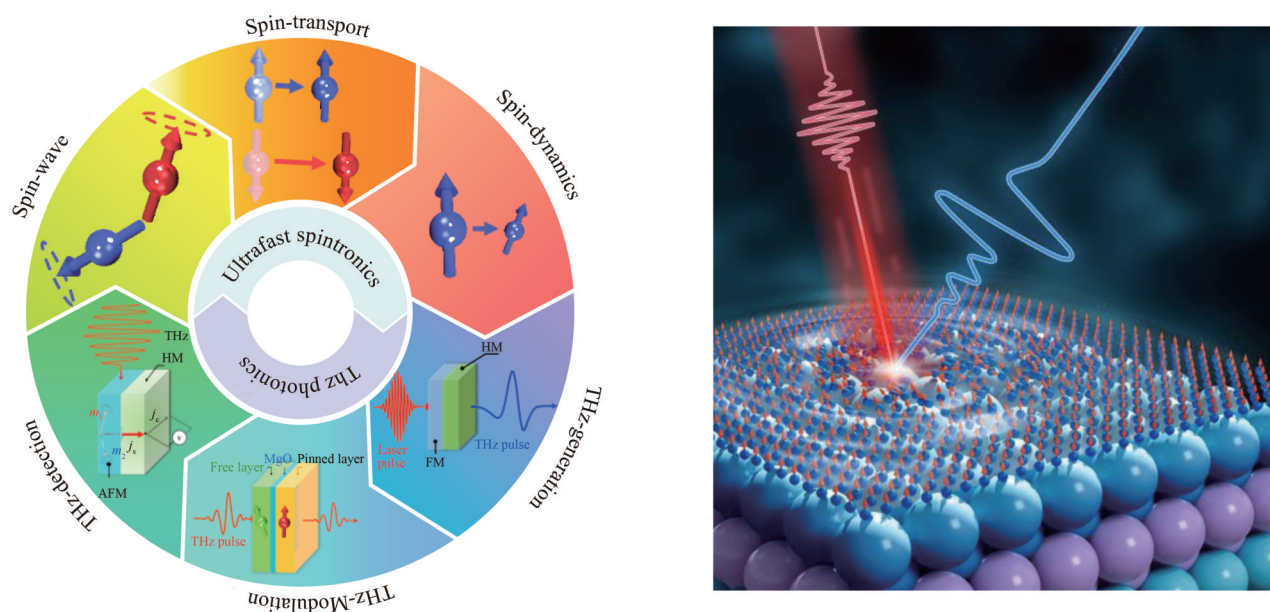


图1 超快太赫兹自旋光子学的研究框架与示意图

Fig.1 Research framework and diagram of ultrafast spin-based terahertz photonics

1 物理机理研究

超快太赫兹自旋光子学研究的物理机理主要包括:太赫兹驱动自旋波、太赫兹探测自旋输运和太赫兹超快磁测量。

1.1 太赫兹驱动自旋波

自旋波 (spin waves) 是磁性材料中磁矩进动的集体激发,其量子化的准粒子称为磁子 (magnon)。对于铁磁体,典型的自旋波动力学在 GHz 频率范围,随着外加磁场的增加而增加。Kittel 模型下的磁共振在 20 T 时达到太赫兹频率。而在反铁磁体中,相邻且方向相反的自旋之间的交换场强度可达数百特斯拉。反铁磁自旋波的本征频率与各向异性场和交换场乘积的平方根成正比,处在 THz 频率范围。研究太赫兹自旋波的产生和调控是开发和利用反铁磁材料的重要基础^[44]。原则上,太赫兹脉冲激发的任何自由度,比如电子、自旋和晶格都可以作为驱动电子自旋的转矩。其中,最直接的是作用于样品宏观磁化矢量 \mathbf{M} 上的太赫兹塞曼转矩 (Zeeman torque), $T \propto \mathbf{M} \times \mathbf{B}_{\text{THz}}$, 与 THz 脉冲的磁场分量 B_{THz} 成正比^[45-46], 如图 2(a) 所示。

如图 2(b) 所示,线偏振的太赫兹脉冲可以选择性地、非热激发稀土正铁氧体 SmDyFeO_3 中的准铁磁

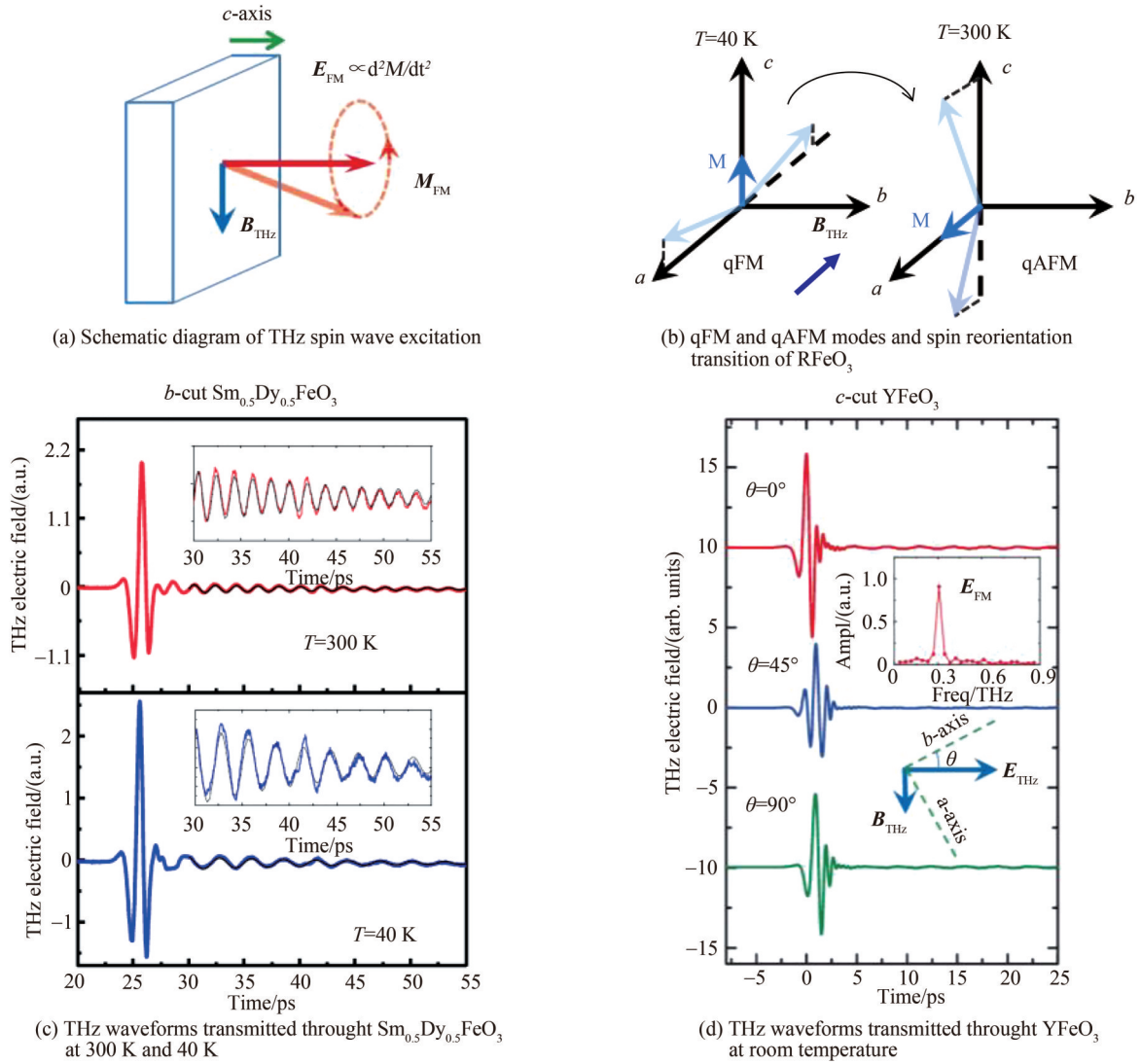


图2 太赫兹自旋波激发与自旋重取向的光谱实验结果^[49,54]

Fig. 2 Terahertz spin wave excitation and spin reorientation transition of $RFeO_3$ ^[49,54]

(quasi FM-mode, qFM)和准反铁磁(quasi AFM-mode, qAMF)共振模式。当 B_{THz} 平行于 M ,激发qAFM模式;当 B_{THz} 垂直于 M ,激发qFM模式。太赫兹脉冲通过瞬态塞曼转矩诱导反铁磁体NiO和稀土正铁氧体 $RFeO_3$ ($R=Y, Nd, Pr, Dy, Er, Ho$)的自旋波^[46-50]。通过探测自旋进动产生的自由感应衰减信号(Free Induction Decay, FID)检测太赫兹自旋波的共振频率、偏振态和衰减时间常数。稀土正铁氧体表现出天然的太赫兹超材料特征^[51-52]。除了利用两个具有一定延迟时间的太赫兹脉冲实现自旋波的相干控制^[53],也可以利用 $YFeO_3$ 介电常数的各向异性,以单个太赫兹脉冲实现自旋波的相干操控^[54]。通过反铁磁自旋波频率随温度的变化,可以得到 $RFeO_3$ 的各向异性常数^[55]和自旋重取向温区^[56]等。相比于飞秒激光脉冲的电场与自旋的间接耦合,太赫兹脉冲磁场所诱导的塞曼转矩直接与自旋相互作用,在样品中产生的热效应较小。

此外,利用太赫兹辐射可以研究电子自旋如何与其他自由度的耦合,如图3所示。在 $TmFeO_3$ 单晶中观测到太赫兹光子与反铁磁共振模式的反交叉现象,实验表明 $TmFeO_3$ 单晶的法布里-珀罗谐振腔可以实现太赫兹光子与反铁磁自旋波的强耦合—太赫兹磁子-极化激元(图3(a))^[57]。 $Er_xY_{1-x}FeO_3$ 单晶在强磁场、极低温及不同掺杂浓度下的太赫兹光谱实验证实, Er^{3+} 电子顺磁共振(Electron Paramagnetic Resonance, EPR)可与 Fe^{3+} 磁振子模式强耦合(图3(b)), Fe^{3+} 和 Er^{3+} 耦合率与 Er^{3+} 稀土离子的浓度依赖关系满足迪克标度模型^[58]。通过测量 $YFeO_3$ 单晶的自旋共振频率随外加磁场的变化,发现了 $YFeO_3$ 的qFM和qAFM模式的反交叉行为(图3(c))。当切角为 58° ,磁场为30 T时,得到最大的耦合强度。磁子-磁子耦合为没有光子参与的奇异量子光学真空现象提供研究平台^[59]。

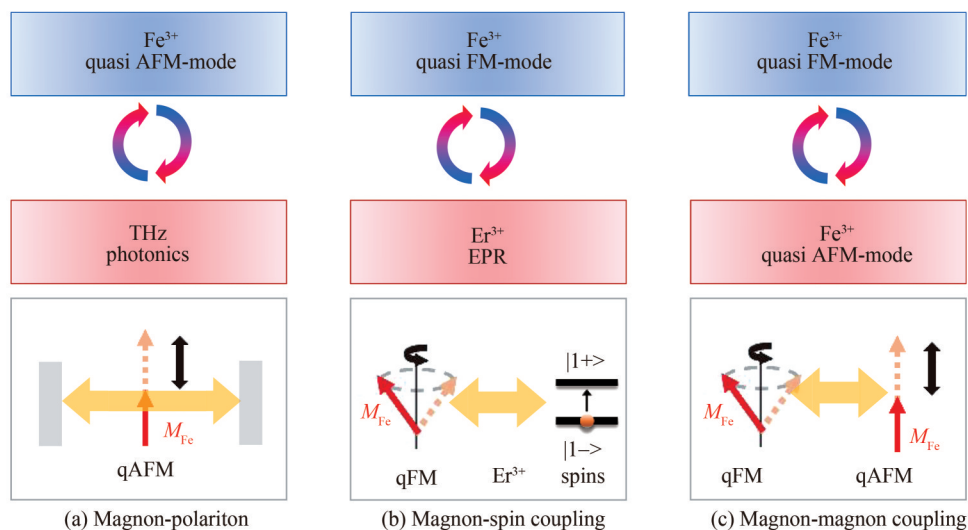


图3 磁子-极化激元,磁子-自旋耦合和磁子-磁子耦合示意图

Fig. 3 Schematic illustrations of magnon-polariton, magnon-spin coupling, and magnon-magnon coupling

除了太赫兹塞曼转矩外,太赫兹脉冲的电场分量对电磁子(electromagnons)提供有效的转矩^[60]。太赫兹电场驱动的电场可以导致CuMnAs的Néel磁矢量发生转换^[61]。利用太赫兹脉冲的电场分量实现的耦合机制还包括:由于激发电子跃迁引起的磁各向异性变化^[62],声子诱导的自旋-轨道调制^[63]和交换耦合^[64]。基于反铁磁TmFeO₃的自旋与定制天线结构,通过局域增强的太赫兹电场强耦合,时间和光谱的指纹特性表明,太赫兹电脉冲可以实现自旋在势垒间的超快相干转换^[65]。

1.2 太赫兹探测自旋输运

当太赫兹电场振荡在磁性材料中诱导瞬态电流,通过分析太赫兹电磁场的衰减和相位延迟研究自旋的输运过程,包括:太赫兹巨磁阻效应(THz Giant Magnetoresistance, GMR),太赫兹隧穿磁阻效应(THz Tunneling Magnetoresistance, TMR)和太赫兹各向异性磁阻效应(THz Anisotropic Magnetoresistance, AMR),如图4(a),图5(a)和5(b)所示。

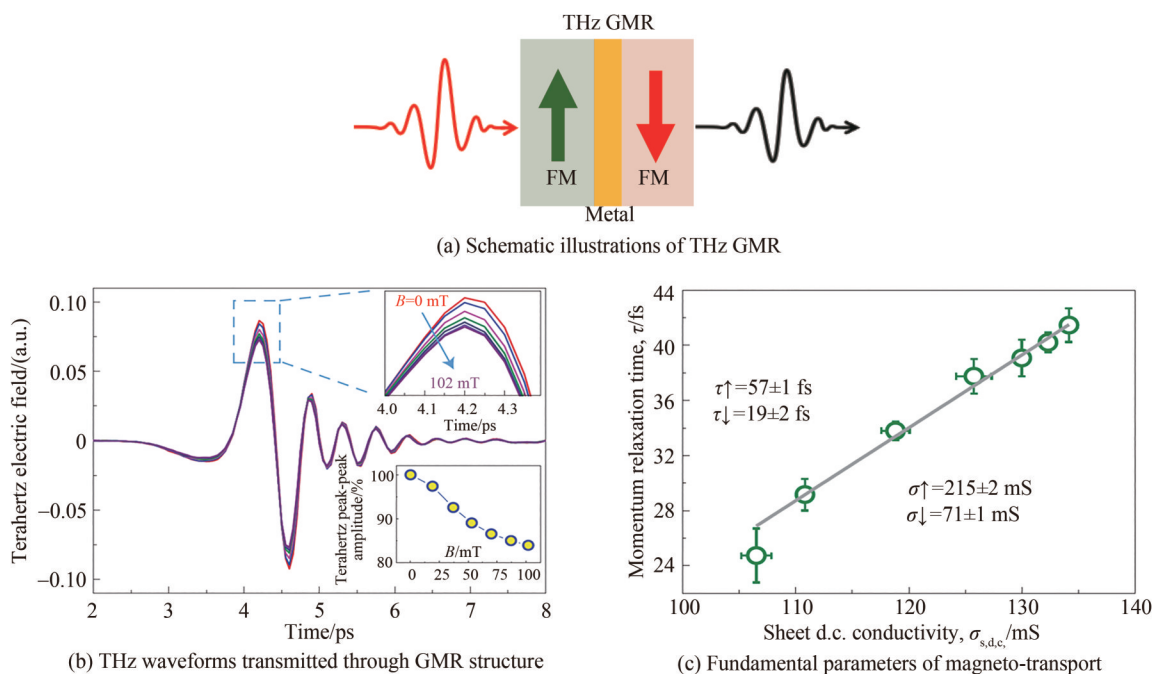


图4 太赫兹巨磁阻效应的原理验证实验^[67]

Fig.4 Principle verification experiment of THz GMR^[67]

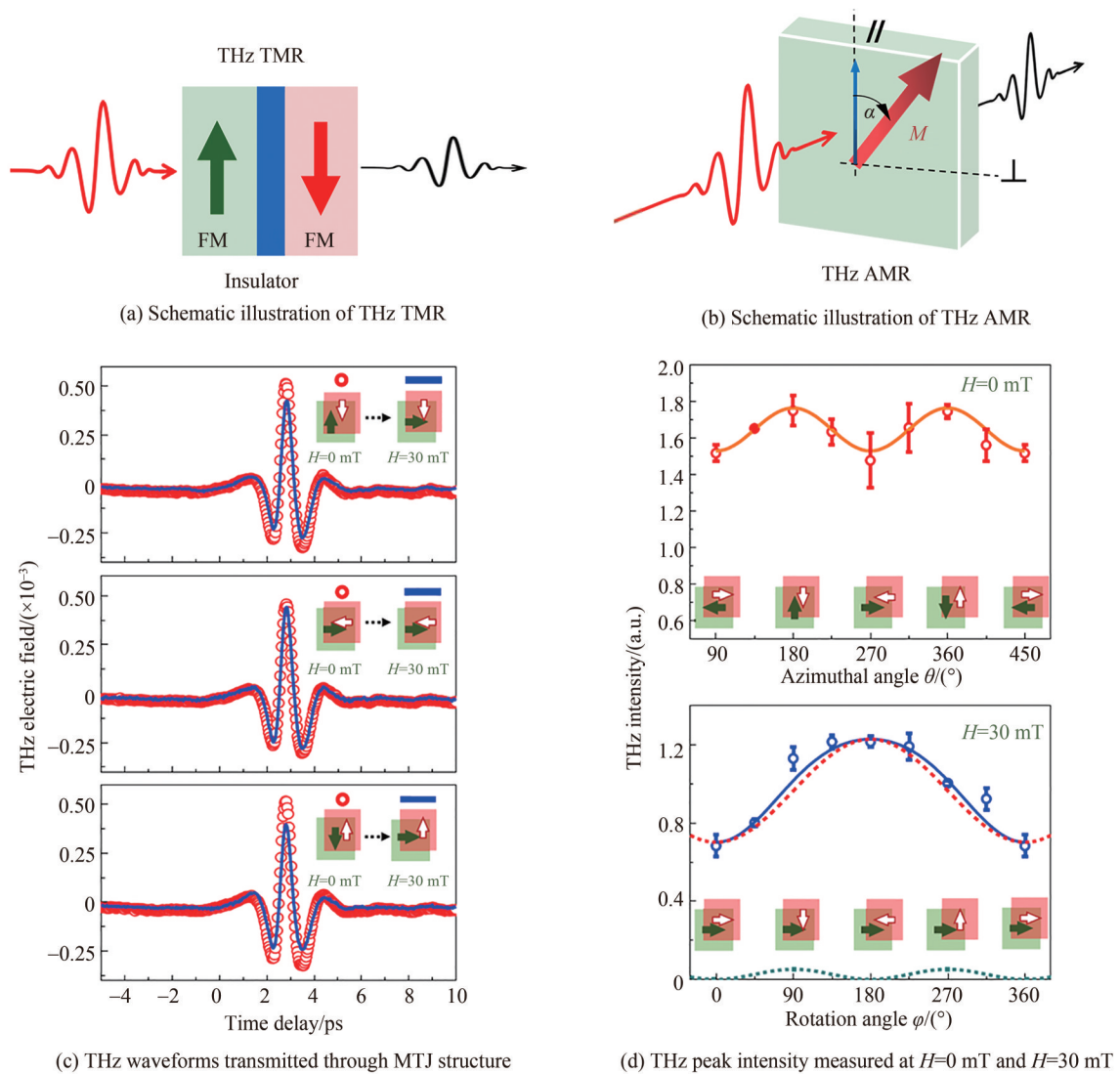


图5 太赫兹隧穿磁阻效应和各向异性磁阻效应^[68]
 Fig.5 Principle verification experiments of THz TMR and AMR^[68]

GMR是指铁磁和非铁金属相间的多层纳米结构中,两层磁性材料(FM)磁化方向相反情况下的电阻值,明显大于磁化方向相同时的电阻值^[66]。1935年MOTT N F提出的“双流”模型成功解释了巨磁电阻效应。GMR起源于电子的散射率,取决于自旋方向和磁性材料的磁化方向。静态磁电阻率测量部分反映了非对称的电子自旋输运,但无法避免电接触效应。超快光谱中使用的飞秒激光会加热电子,很难实现GMR结构中Mott电导率的本征测量。也就是说,现有的实验手段很难获得自旋分辨的载流子浓度和动量散射时间等基本参数。太赫兹时域光谱作为一种非接触式的测量手段,太赫兹波的振荡周期对应于电子-电子散射时间尺度。由于光子能量为meV量级,太赫兹波探测的是费米面附近非热电子的电导率。图4(b)表示太赫兹脉冲经过GMR结构的时域光谱。当外加磁场的强度增加至100 mT时,GMR结构使太赫兹波电场的透过率下降约20%。太赫兹波的吸收正比于样品的电导率,样品的电导率随磁场强度的增加而增加。通过Drude模型拟合,得到样品复电导率的实部 $\sigma_{s,dc}$ 和电子动量散射时间 τ 与磁场的依赖关系。 $\sigma_{s,dc}$ 和 τ 都随磁场的增加而增加。结合Drude模型和Mott双流模型,对不同磁场下的 $\sigma(\omega)$ 进行全局拟合,最终得到了自旋依赖的面电导率。如图4(c)所示,自旋向上和自旋向下电子的面电导率存在3倍的关系。这是由于自旋向上电子的动量散射时间约为自旋向下电子的3倍。在费米面处,自旋向上和自旋向下电子的浓度几乎相等^[67]。

磁隧道结(Magnetic Tunnel Junction, MTJ)一般为铁磁层/非磁绝缘层/铁磁层(FM/I/FM)的三明治结构。产生TMR效应的物理机理是自旋相关的电子隧穿效应。在硅片上设计MTJ,室温下只需弱磁场

(30 mT)驱动,通过旋转样品调控太赫兹波激发皮秒时间尺度上自旋流的隧穿几率(图5(c)),实现了太赫兹TMR原理验证实验。太赫兹脉冲的振幅调制深度达到60%(图5(d))。该磁性超晶格薄膜的总厚度仅为77.45 nm,为目前已有报道的太赫兹磁调制器件厚度的近千分之一,这对太赫兹调制器件的集成化、小型化起到关键作用^[68]。

除了GMR和TMR,AMR是磁性材料中最基本的自旋-轨道相互作用引起的输运现象。AMR描述各向异性的电荷电导率,依赖于电流和磁化强度的相对方向。它的起源通常被认为来自于“外在效应”,即自旋依赖的电子散射,忽略了散射无关的“内在效应”。超宽带的太赫兹探测范围(从直流到28 THz)涵盖了Co, Ni, Ni₈₁Fe₁₉和Ni₅₀Fe₅₀等多晶铁磁薄膜中的扩散型和弹道型的带内电子输运。基于玻尔兹曼输运理论分析太赫兹透过率数据,成功区分了AMR的“外部效应”和“内部效应”,Ni, Ni₈₁Fe₁₉和Ni₅₀Fe₅₀样品的AMR主要表现为外部效应,然而六边形结构的Co微晶薄膜有比较大的内在AMR贡献^[69]。此外,AMR的“内外效应”具有不同的温度依赖关系^[70]。

1.3 太赫兹超快磁测量

飞秒激光诱导超快(亚皮秒)退磁通常伴随着磁矩(magnetic moment)的瞬变,在Ni^[41]、Co₂MnSn^[71]等铁磁薄膜中产生太赫兹脉冲辐射。太赫兹发射的极性取决于样品的磁化方向。研究的难点在于,通过远场的太赫兹电磁脉冲发射信号,准确重构超快自旋动力学过程。

如图6(a)的光路图所示,采用太赫兹发射光谱技术首次实现了单晶Fe薄膜的超快磁测量^[72]。实验样品为生长在MgO衬底上的10 nm的单晶Fe薄膜,并在Fe薄膜上再覆盖一层12 nm厚的MgO薄膜。图6(b)为该样品测得的太赫兹电光取样(EO)信号。通过实验光路的传递函数和探测函数,从太赫兹发射EO信号,重构出磁动力学 $M(t)$,如图6(c)中的实线所示。实验结果与理论计算(虚线)结果一致,表明太赫兹发

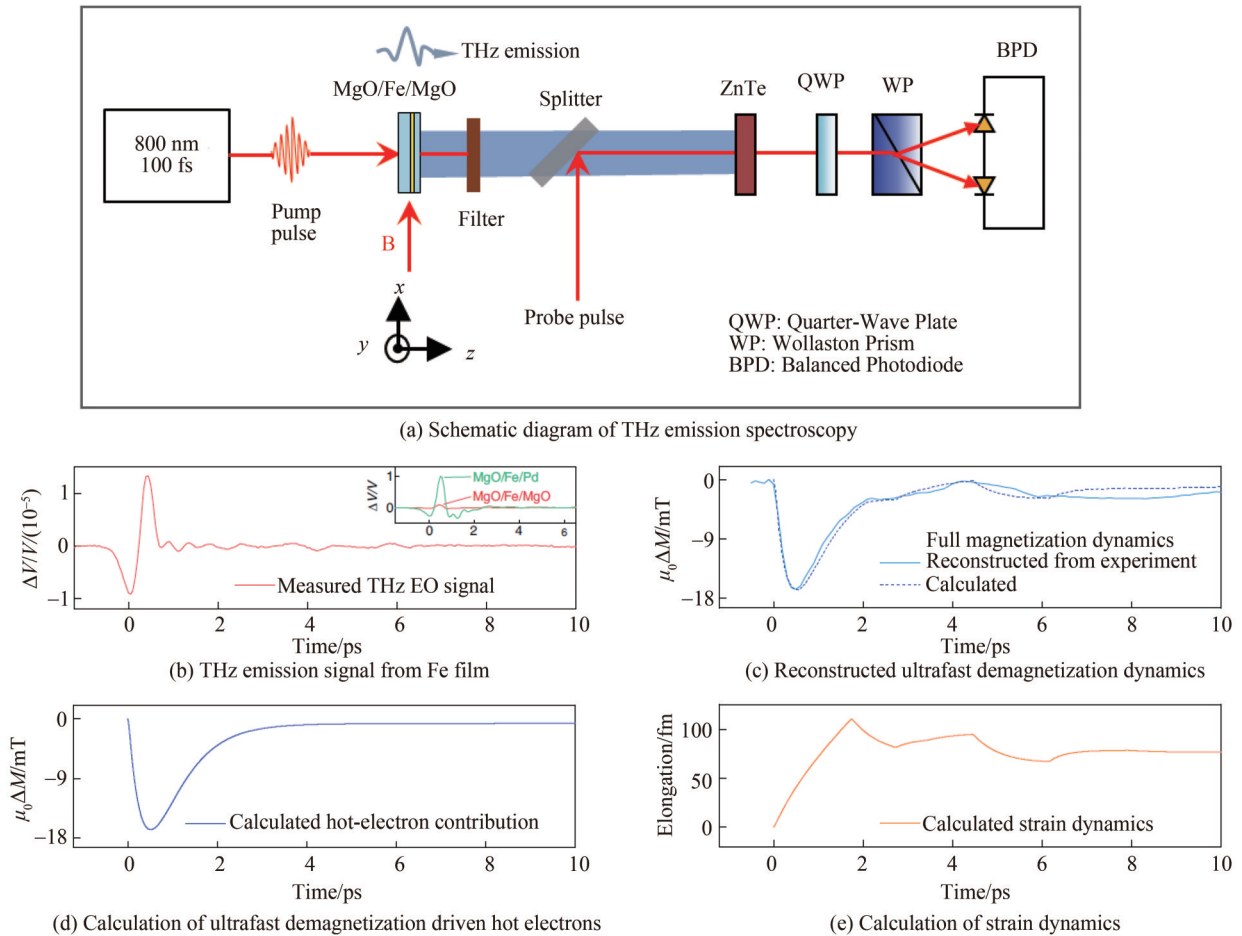


图6 太赫兹发射光谱示意图与飞秒激光瞬态退磁动力学^[72]
Fig.6 Schematic diagram of THz emission spectroscopy and ultrafast demagnetization dynamics^[72]

射光谱结合信号重构方法可有效获得铁磁薄膜的超快退磁及弛豫动力学。如图6(d)所示,超快退磁信号的时间尺度约为500 fs,恢复时间约为1.7 ps。如图6(e)所示,磁恢复过程中存在几个皮秒的磁扰动信号。基于理论分析,该信号起源于激光激发相干声学声子诱导的磁弹效应。激光直接诱导退磁信号的大小约是声波驱动退磁信号的7倍。研究结果表明,区别于传统的光、电或磁驱动,声学脉冲也能非热控制超快自旋动力学。

值得一提的是,在铁磁/非磁重金属(FM/HM)异质结构中,KAMPFRATH T等利用飞秒激光注入自旋流,并通过逆自旋霍尔效应(ISHE)转换成横向电荷流,辐射出较强的太赫兹相干脉冲^[73]。在这一过程中,自旋流的极化方向与铁磁层的磁化方向一致。如图7所示,对产生的太赫兹脉冲进行重构,得到Fe/Pt, γ -Fe₂O₃/Pt, Fe₃O₄/Pt等不同异质结的瞬态自旋流动力学 $j_s(t)$ 。Fe₃O₄/Pt中的自旋流可以通过Fe/Pt和 γ -Fe₂O₃/Pt中的自旋流线性叠加而成^[74]。此外,可以通过圆偏振激光的螺旋度和外部磁场控制自旋光电流的方向^[75]。利用太赫兹发射光谱,研究发现除ISHE效应之外,在Ag/Bi界面上存在逆Rashba-Edelstein效应(Inverse Rashba-Edelstein Effect, IREE),并实现了超快自旋流-电荷流转换^[76-78]。在YIG/Pt异质结中,基于超快自旋泽贝克效应(Spin Seebeck effect)实现了超快自旋流-电荷流转换^[79]。我们通过对YIG/Pt异质结构进行高温退火处理后再原位生长Pt层,优化后的YIG/Pt界面有效提高了自旋流-电荷流的转换效率^[80]。此外,太赫兹发射光谱还能对不同自旋-电荷转换效率的材料进行高通量筛选^[81]。

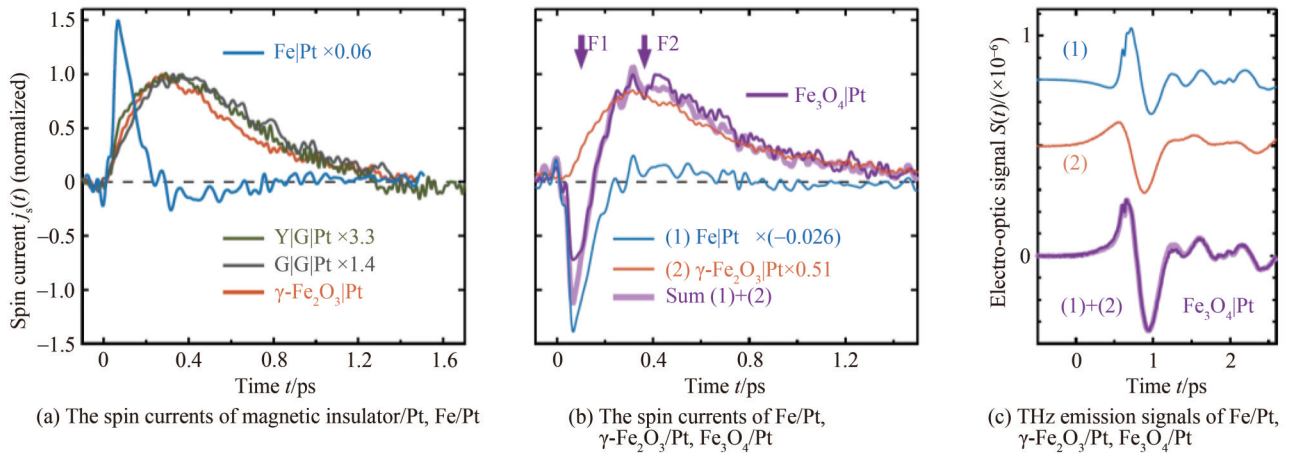


图7 磁性绝缘体/Pt和Fe/Pt异质结的自旋流及其太赫兹发射光谱^[74]

Fig.7 The spin currents and THz emission signals of magnetic insulator/Pt and Fe/Pt^[74]

2 器件设计与应用

超快太赫兹自旋光电子学的核心器件主要包括:自旋太赫兹辐射源、自旋太赫兹调制器和自旋太赫兹探测器。

2.1 自旋太赫兹辐射源

为进一步提升太赫兹的辐射效率,国内外科研人员提出多种自旋太赫兹辐射源的优化方案。

首先是提高自旋流-电荷流的转换效率。通常用自旋霍尔角描述材料的电荷-自旋相互转换能力。如图8所示,选取自旋霍尔角大的非磁性重金属作为太赫兹发射层^[82-84]。一方面,通过掺杂,退火等方法增强铁磁层中自旋流的光注入^[85-86]。另一方面,通过优化铁磁和非磁性金属层的厚度^[87-88]和铁磁/重金属层的界面^[89-90],可以有效提高太赫兹辐射强度。

在双面抛光的熔融SiO₂衬底上,通过磁控溅射在超真空腔中(1×10^{-8} Torr, 1 Torr=133 Pa)生长了纳米级厚度的三层膜异质结构HM₁/FM/HM₂。非磁性金属层HM₁为Pt, HM₂为W或者Ta。HM₁和HM₂之间夹一层铁磁Co₂₀Fe₆₀B₂₀。溅射中使用高纯的W, Pt, Ta和Co₂₀Fe₆₀B₂₀靶材来制备样品。利用台阶仪定标溅射速率分别为0.034, 0.050, 0.042和0.022 nm/s。通过改变溅射时间控制样品厚度。如图9所示,利用三层异质结构W(4 nm)/Co₂₀Fe₆₀B₂₀(4 nm)/Pt(4 nm)和Ta(4 nm)/Co₂₀Fe₆₀B₂₀(4 nm)/Pt(4 nm),可有效提高太赫兹辐射强度^[82,91]。最近,人们将目光投向更复杂的异质结构。磁性层材料包括:哈斯勒合金^[92-93]、非共线反

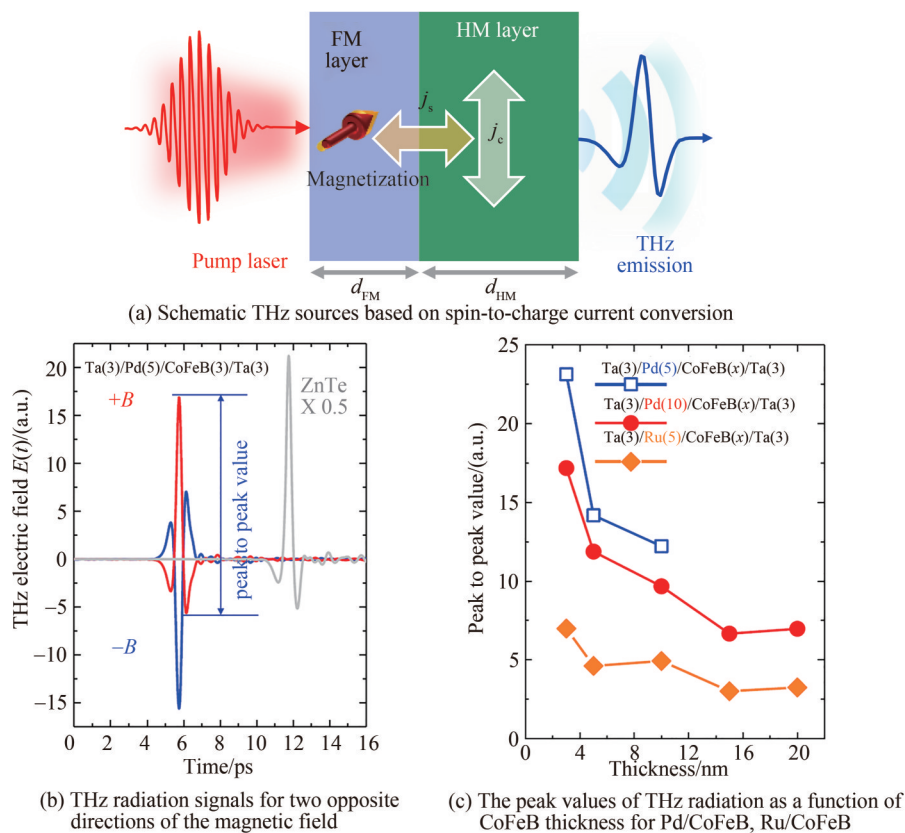


图 8 基于超快自旋流-电荷流转换的太赫兹辐射源优化^[83]
Fig. 8 THz generation based on spin-to-charge current conversion^[83]

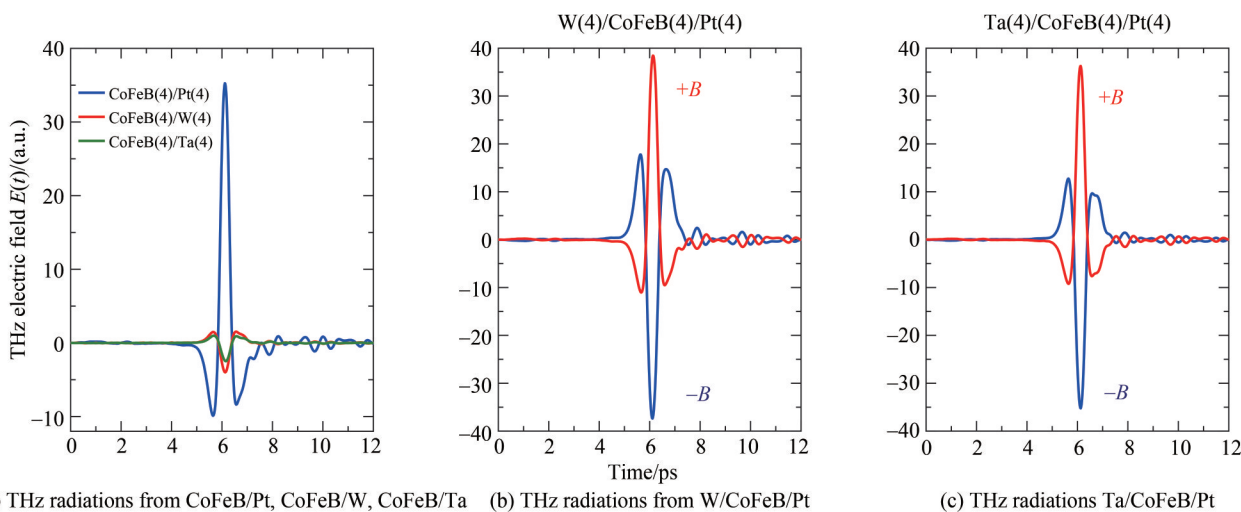


图 9 W/CoFeB/Pt 和 Ta/CoFeB/Pt 异质结构的太赫兹脉冲辐射^[91]
Fig. 9 THz radiations from W/CoFeB/Pt and Ta/CoFeB/Pt heterostructures^[91]

铁磁 (Mn_3Sn)^[94], 反铁磁绝缘体 (NiO)^[95], 二维铁磁体 ($CrSiTe_3$, Fe_3GeTe_2)^[96-97] 等。太赫兹发射层材料包括: 拓扑绝缘体 (Bi_2Se_3)^[98], 二维半导体 (MoS_2)^[99] 和 Mn_2Au ^[100] 等。

其次是提高入射激光脉冲的能量利用率。一方面, 可以增大三层异质结发射器的辐照面积, 提高太赫兹辐射的峰值电场强度^[101]。另一方面, 可以利用金属-介质光子晶体结构^[102] 或者光学介质谐振腔 (TiO_2/SiO_2 周期结构)^[103] 提升对激光的吸收率, 从而提高太赫兹辐射效率。最后是提高太赫兹的输出耦合效率。将自旋太赫兹辐射源与超半球硅透镜组合, 使衬底和空气界面耦合出更强的太赫兹辐射^[87]。可以将异质结构与天线结构相结合^[104-106], 或者制备成具有电极的条带结构^[107] 提高太赫兹辐射强度。

相比于传统的宽频太赫兹辐射源,纳米尺度的磁性异质结器件不仅高效稳定,制作成本低,而且具有独特的优势。第一,磁性薄膜异质结在脉冲激光激发下可以高效产生宽频相干太赫兹辐射。在脉冲宽度约为150 fs的激光脉冲激发下,产生的太赫兹辐射有效覆盖1~3 THz频谱范围。进一步降低入射激光脉冲的宽度可以拓宽太赫兹发射的频谱范围,甚至覆盖1~30 THz^[82]。第二,自旋流的极化方向和太赫兹辐射的偏振方向都可以通过外加磁场灵活调控,这是光电天线、半导体晶体等无法实现的。第三,自旋太赫兹辐射源适用于多种不同的光激发条件^[108],从纳焦级能量的飞秒振荡器到毫焦级能量的飞秒激光放大器都可以作为激发光源。气体分子、原子和非线性晶体的太赫兹辐射源通常需要毫焦级的脉冲能量才能有效产生太赫兹辐射。第四,基于纳米厚度金属薄膜的制作工艺十分成熟,可以将磁性异质结加工成不同的超构表面,从而实现宽频太赫兹辐射源与光学参量调控技术相集成。

2.2 自旋太赫兹调控器

太赫兹电磁波的高效调控是太赫兹技术发展和应用的关键因素之一。自旋太赫兹辐射源具有很强的调控能力。不仅可以通过改变FM/HM异质结中的HM的自旋霍尔角实现太赫兹辐射波形调控^[73],而且可以设计多层的合成磁结构调控太赫兹的波形和相位^[109-110]。利用几十 mT 量级的小磁场就足以改变自旋阀结构中自由层的磁化方向,从而改变两层铁磁材料的相对磁化方向。使太赫兹发射或透射从高功率状态快速切换到低功率状态^[68,111]。

如图 10(a)所示,利用飞秒微纳加工技术制作微米尺度的线栅结构或块状 FM/HM 异质结(图 10(b))。

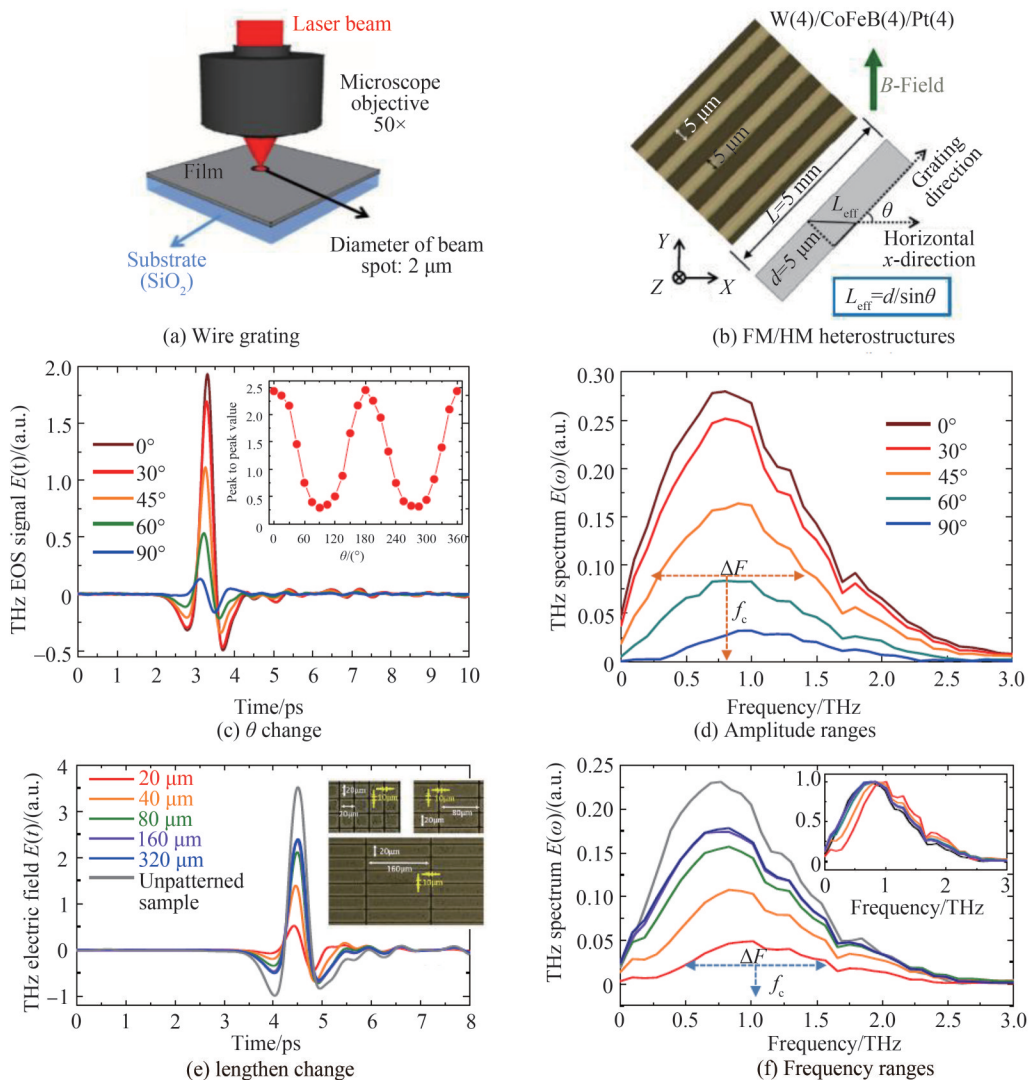


图 10 微纳结构自旋太赫兹发射源示意图与太赫兹发射光谱^[112-113]
Fig.10 THz emission signals from patterned spintronic THz emitters^[112-113]

通过改变线栅与外加磁场的夹角(图 10(c))或块状结构的长度(图 10(e)),可有效调控太赫兹辐射的振幅和频谱范围^[112-113],如图 10(d)和 10(f)所示。此外,可以通过改变磁场的空间分布^[114-115],和利用激光脉冲的光热效应^[116],实现太赫兹脉冲波形调控。

如前所述,通过外加磁场可以灵活调控太赫兹辐射的偏振方向。除此之外,自旋太赫兹辐射源还可以与液晶^[117]、超材料^[118]、铁电体^[119]、多铁材料^[120]相结合,以及级联多个自旋太赫兹器件^[121]等方案,实现太赫兹辐射的偏振调控。通过改变外加磁场与线栅结构的夹角,可以实现太赫兹脉冲从线偏振态到左旋或右旋圆偏振态的任意转换^[122]。最近,通过时间分辨的磁场实现了自旋太赫兹发射脉冲的偏振调制率达到 10 kHz^[123]。

2.3 自旋太赫兹探测器

如 1.1 节所述,反铁磁共振处于太赫兹频率范围,因此反铁磁体是实现太赫兹频率信号共振检测的一种可能方案。理论上,已经证明反铁磁体可以作为太赫兹频率探测器的工作介质^[124-125]。工作原理如图 11(a)所示,入射太赫兹波的磁场分量产生 Zeeman 转矩,作用于反铁磁体并诱导 Néel 矢量进动,基于自旋泵浦效应(spin-pumping)产生自旋流。自旋流注入相邻的 HM 层,基于 ISHE 产生电荷流和直流电压信号。最近,在反铁磁 Cr_2O_3 和 MnF_2 中实现了自旋泵浦实验。无外加磁场时,反铁磁体 Cr_2O_3 和 MnF_2 的太赫兹共振频率分别为 0.163 THz 和 0.245 THz。实验结果表明,AFM/HM 异质结构有望成为无源的自旋太赫兹探测器^[126-127]。理论研究进一步表明,可以通过改变施加于反铁磁体上的直流偏置磁场调谐 AFM/HM 探测器的共振探测频率。此外,增加外加磁场和减小 AFM 层的厚度都有望提高探测器的灵敏度^[128]。

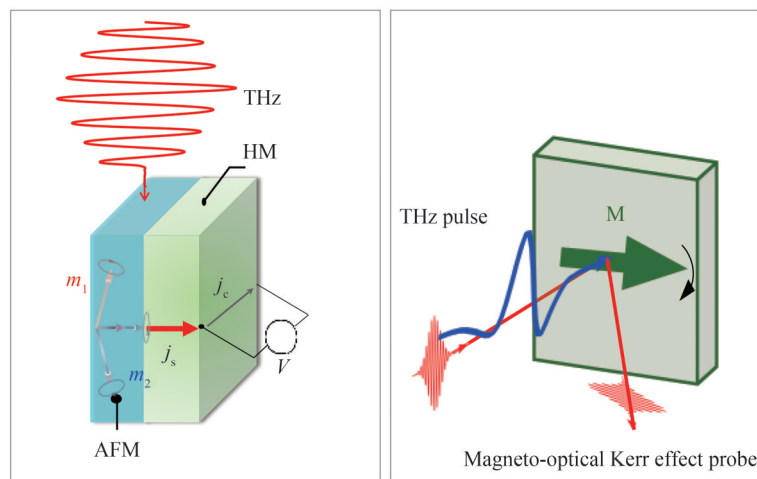


图 11 基于太赫兹自旋泵浦和太赫兹抽运-磁光探测原理的太赫兹探测示意图

Fig. 11 Principle of spintronic THz detector based on THz spin-pumping and THz pump magneto-optical Kerr probe

如图 11(b)所示,利用太赫兹抽运-磁光探测光谱证实了单周期太赫兹脉冲与铁磁性 Co 薄膜磁化之间存在相位锁定耦合^[129]。具体地,单周期线偏振太赫兹脉冲的磁场分量的强度为 0.4 T,以偏离法线 20° 入射 10 nm 的 Co 薄膜。利用一个 50 fs 的探测光脉冲,通过 MOKE 时间分辨测量其磁动力学。时间分辨的 MOKE 信号和太赫兹脉冲波形几乎相同。实验中,太赫兹场与磁化强度的耦合不依赖于共振激发或自旋波。在没有共振激发和能量耗散的情况下,由太赫兹电磁场激发的时间分辨磁化动力学紧紧地与太赫兹脉冲的相位锁定在一起。当太赫兹脉冲磁场分量的极性反转,MOKE 信号随之反转。MOKE 信号的幅度与太赫兹脉冲的磁场分量成正比关系。该研究成果为基于自旋的宽频太赫兹相干探测提供了一种新思路^[129-130]。

3 总结与展望

尽管超快太赫兹自旋光电子学的研究取得了显著进展^[131],在太赫兹宽谱测量^[132]、磁结构检测成像^[133]、太赫兹超分辨成像^[134-135]、可编码太赫兹辐射源^[136]、太赫兹矢量光束^[137-138]等领域实现初步的应用。然而,自旋太赫兹辐射源强度进一步提升的速度减慢;自旋太赫兹调制器的调制频谱较窄,磁隧道结太赫兹调制的

速度较慢;自旋太赫兹探测器尚处于理论和实验验证阶段。为了解决这些问题,超快太赫兹自旋光电子学研究更为复杂的材料和结构体系,将包含更多的物理、材料和光学等学科交叉。

值得一提的是,拓扑绝缘体作为一种新型二维材料,具有奇特的电子结构和自旋结构,被认为是未来实现超低功耗磁随机存储器等自旋电子器件和量子计算的候选材料。拓扑绝缘体的体态是有能隙的半导体或者绝缘体,表面态则为无能隙的金属态。拓扑绝缘体表面态电子的自旋高度极化,强自旋轨道耦合效应导致拓扑绝缘体的自旋-动量锁定特性。在圆偏振光激发下,具有中心对称破缺结构的拓扑绝缘体表面态存在一个与激发光的圆偏振特性无关的位移电流和与圆偏振特性依赖的注入电流,他们分别来自于拓扑绝缘体表面态的线光生伏打效应和圆光生伏打效应。一方面,使用四分之一波片的转角改变入射飞秒激光的偏振态,可以有效控制拓扑绝缘体 Bi_2Se_3 , Sb_2Te_3 , Bi_2Te_3 等表面态中的超快非线性光电流^[139-141]。另一方面,基于拓扑绝缘体强的自旋-轨道耦合效应,有望替代重金属层,实现高效的太赫兹发射。因此,拓扑绝缘体/铁磁异质结有望实现任意波形可调谐的拓扑自旋太赫兹发射器^[142-144]。

超快太赫兹自旋光电子学研究是一个新兴的研究领域,获得了国内外研究人员的关注,并取得了一系列研究成果。本文主要介绍了超快太赫兹自旋光电子研究原理、方案与应用。一方面,太赫兹脉冲为研究超快自旋电子学提供强大工具,可实现太赫兹驱动自旋波,探测自旋输运和超快磁测量。另一方面,通过研究太赫兹自旋电子学效应,逐步实现了自旋太赫兹辐射源,调制器和探测器等多种太赫兹光子学器件。研究成果不仅有望提升自旋电子学中的超快磁记录和读写的速度,而且能为设计出更多高效的太赫兹光子学器件提供参考。

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Development of Ultrafast Spin-based Terahertz Photonics(Invited)

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Abstract: Terahertz (THz) radiation is generally defined as the region of the electromagnetic spectrum in the range of 0.1 to 10 THz, between the millimeter and infrared frequencies. THz radiation is important from both scientific and application point of view. THz science and technology has been an active research area for a wide variety of applications: such as spectroscopy, imaging and sensing, biology and medical sciences, and security evaluation. The development of efficient, ultra-broadband, and low-cost THz photonic devices requires new materials and mechanisms, which is the key challenge for the field of THz science and technology. The discovery of THz electromagnetic pulse emission from ultrafast demagnetization by femtosecond laser pulses gave insight into the microscopic interactions that connect the ultrafast spintronics and the THz photonics.

Based on our experimental observations, this paper reviews the recent developments and applications, the current understanding of the physical processes, and the perspectives of ultrafast spin-based THz photonics.

Firstly, ultrashort THz pulses have been demonstrated as a promising tool to investigate the ultrafast spintronics. We review the fundamental physical processes and properties including THz-driven spin waves, THz spin transport probing, and ultrafast THz magnetometry. 1) The THz pulses are used to excite and control the antiferromagnetic spin waves in rare-earth orthoferrites with the THz time-domain spectroscopy. In addition, we observe the magnon-polariton, magnon-spin coupling, and magnon-magnon coupling in the condensed matter systems. 2) We demonstrate the magnetic modulation of THz waves, along with heat- and contact-free giant magnetoresistance, tunneling magnetoresistance and anisotropic magnetoresistance readout using ultrafast THz signals. We directly determine the spin-dependent densities and momentum scattering times of conduction electrons. The various magnetic configurations between the parallel state and antiparallel state of the magnetizations of the ferromagnetic layers in the magnetic tunnel junctions have the effect of changing the conductivity, making a functional modulation of the propagating THz electromagnetic fields. 3) We demonstrate a method of ultrafast THz magnetometry, which indicates the sub-picosecond demagnetization dynamics in a laser-excited iron film. The measurements reveal the contributions originating from magnetization quenching and acoustically-driven modulation of the exchange interaction. In addition, the ultrafast photoinduced spin transport can be extracted from the THz emission signals. We observe the transition of laser-induced THz spin currents from torque-mediated to conduction-electron-mediated transport in ferromagnetic/non-magnetic heterostructures.

Secondly, by exploring the ultrafast THz spintronic effects, new applications in THz photonic devices emerge, including spintronic THz emitters, THz modulators and THz detectors. 1) The ferromagnetic/non-magnetic heterostructure under the excitation of femtosecond laser has proved to be a potential candidate for high-efficiency THz emission. The ultrafast spin-charge conversion based on the Inverse Spin Hall Effect (ISHE) is used to generate broadband THz radiation. We summarize the efforts that have been made to improve the performance of spintronics-based THz emitters. Up to date, the efficiency of spintronics-based THz emission has been enhanced to reach the same level of millimeter-thick ZnTe crystal. 2) The combined spintronic and photonic heterostructures are exploited to realize active modulation of THz radiation. In addition, it is demonstrated that the THz radiation can be mediated coherently through the charge current induced by the ISHE and the built-in transient current quasi-simultaneously created within the patterned heterostructures. 3) Using the ISHE, an antiferromagnet/heavy metal bilayer is

theoretically promising for the realization of a resonant, compact, and tunable THz detector. In addition, a coherent and phase-locked coupling between a single-cycle THz transient and the magnetization of cobalt films suggests new opportunities for THz pulse detection.

Finally, a brief summary and outlook are given. Looking to the future, we introduce the applications of ultrafast spin-based THz photonics, such as ultra-broadband measurements, magnetic structure detection and imaging, and THz near-field microscopy. In addition, topological materials bear a large potential for efficient spin-to-charge conversion due to the inherent spin-momentum locking. The topological insulator/ferromagnetic heterostructures are expected to present a high-performance THz radiation. In addition, the topological spintronic THz emitter will show a potential to generate arbitrary THz waveforms. One can anticipate that the research scope of ultrafast spin-based THz photonics will successfully be used to understand the fundamental physics in new materials and give rise to high-efficient THz photonic devices and spectroscopy applications. We hope that our work will stimulate more fundamental and technological developments in this new research field.

Key words: Terahertz; Spintronics; Ultrafast spectroscopy; Terahertz generation and modulation

OCIS Codes: 040.2235; 160.3820; 300.6530; 300.6495

Foundation item: National Natural Science Foundation of China (Nos. 61988102, 61975110, 61735010), the 111 Project (No. D18014), the International Joint Lab Program of the Science and Technology Commission Shanghai Municipality, China (No. 17590750300), the Key Project of the Science and Technology Commission Shanghai Municipality, China (No. YDZX20193100004960), Science and Technology Commission of Shanghai Municipality (No. 21JC1402600), the Shanghai Rising-Star Program of the Science and Technology Commission of Shanghai Municipality, China (No. 18QA1401700), the Chenguang Project of Shanghai Educational Development Foundation, China (No. 16CG45), the Young Eastern Scholar Project of Shanghai Municipal Education Commission, China (No. QD2015020)