多电子动力学的强场调控与阿秒探测

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摘要 作为一种新型的超短极紫外/软X射线相干光源, 阿秒脉冲推动了物质科学的新发展. 它使得人们可以深入物质内部, 对各种原子尺度的微观过程, 以电子运动的自然时间尺度($1\text{ as}\sim 10^{-18}\text{ s}$)实现前所未有的时空分辨和超快调控. 阿秒脉冲的产生和应用与强激光脉冲驱动的物质内电子亚周期的超快动力学密切相关. 对其的研究, 有助于在时域、相位、纠缠等方面理解电子关联如何影响物质性质和动力学, 回答光是如何被原子、分子或者固体吸收的, 光电离时间、隧穿时间、电子迁移时间是否存在等基本科学问题. 虽然未来阿秒脉冲光源发展仍有待进一步增大光强和压缩脉宽, 但是目前结合超强的激光脉冲和超短的阿秒光源, 有可能在新的能量域、时间域、空间域实现电子动力学的强场调控和阿秒探测, 为未来拍赫兹光电子学、阿秒瞬态光谱等新的物质科学与信息技术奠定基础. 本文将回顾强场和超快物理在实验、理论、技术和应用上带来的推动, 聚焦电子和多电子动力学的强场效应和时域探测, 介绍部分相关领域的发展.

关键词 阿秒脉冲, 高次谐波, 强场, 隧穿电离, 电子关联

过去的30年见证了强场物理的快速发展和单阿秒脉冲的产生. 阿秒脉冲的脉宽已经短至可以与氢原子基态电子对应的经典圆周运动的开普勒周期相比拟. 超短强激光脉冲技术的产生与发展, 使得我们对光与物质相互作用的认识超越传统的非线性光学, 深入到以时域研究为特征、以亚光学周期电子/载流子动力学为焦点的新阶段. 对强电磁场驱动原子分子发生电离以及各种后续超快过程的研究, 催生了太赫兹辐射、阿秒光脉冲和阿秒电子脉冲产生的新手段, 促进了强场物理和阿秒物理的产生与发展^[1~6]. 与此同时, 对强场电子动力学实现量子调控的需求, 引发了飞秒激光波形精密控制和电子波包强场调控技术的进步^[7]. 阿秒脉冲的应用和强场电子动力学的调控, 使得在原子空间尺度和阿秒时间精度探测和控制原子、分子甚至固体内各种微观超快动力学成为可能, 为拍赫兹光电子学、阿秒测量学、超快光

学太赫兹技术奠定了基础^[8~13].

20世纪80年代左右, 实验发现强激光脉冲作用下原子分子会吸收多个光子导致多光子电离. 特别是原子分子吸收的光子数可以远大于电离所需要的最少光子数, 导致光电子能谱表现出一系列的间隔一个光子能量的峰, 即所谓的阈上电离(above-threshold ionization, ATI)^[14~16]. 同时, 人们发现伴随强场电离, 强激光驱动的气相原子分子会发出频率为驱动光频率奇数倍的相干辐射, 即高次谐波产生(high harmonic generation, HHG)^[17~20]. 分子的光解离过程也可以吸收多个光子发生阈上解离(above-threshold dissociation, ATD), 实验测量得到的解离碎片动能谱成分立峰分布^[21]. 借助于电子-离子符合探测技术^[22], 近期发现, 电离电子和解离碎片总能谱间隔单个光子能量, 揭示了分子内电子-核多个自由度整体吸收并分配多光子能量的本质^[23]. 这

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些新颖的物理现象呼唤对光与物质相互作用过程的新理解. 当强激光的电场接近甚至超过原子内的库仑场场强(1个原子单位的光强对应 $3.5 \times 10^{16} \text{ W/cm}^2$), 光对原子分子的作用已经不能再被当作微扰, 基于传统微扰近似的非线性光学理论失效, 要求发展非微扰的能够处理极端非线性新图像和新理论. 近自由的光电子在离开原子实之前强烈地振荡, 在一个光学周期里的平均动能(即有质动力能)正比于激光波长的平方和光强. 比如, 在光强为 10^{14} W/cm^2 下, 对应近红外激光(800 nm)的有质动力能约为6 eV; 对应远红外(1600 nm)则约为24 eV, 已经远大于大多数气相分子的第一电离能. 在太赫兹波段(0.3 mm), 有质动力能甚至达到0.8 MeV, 已经几乎可以产生正负电子对^[24]. 通过把电离和后续的电子动力学有机联系, 发展出了再散射图像^[25-27]. 在亚周期时间尺度内, 隧穿电离产生的近自由光电子被光场驱动与原子实碰撞, 发生散射、捕获或者辐射复合等, 导致高次谐波产生、ATI光电子产生、二次电离、里德堡态捕获、太赫兹辐射等各种复杂的物理现象^[28]. 飞秒强激光作用下的各种极端非线性过程对激光脉冲的时域电场波形极其敏感, 要求对脉冲的波形精确控制, 带动了载波包络相位稳定的少周期脉冲源、时域偏振门、多色控制门等技术的发展和应用^[29,30]. 通过激光波形剪裁, 控制电子的跃迁、转移、输运等, 有助于实现物质波形的调控, 发展基于电子波作为载体的拍赫兹信息技术具备了可行性^[9,11]. 目前, 通过强光与气相、液相、固相等物质的相互作用, 已具备产生太赫兹和阿秒脉冲等具有重要应用价值的新型光源的多种手段^[31]. 由于高次谐波辐射过程在时域上表现为每半个激光周期发生一次辐射的阿秒脉冲链, 通过波形的精确剪裁可以控制只发生单次辐射, 从而制备短于百阿秒的单个阿秒脉冲^[32-36]. 利用阿秒脉冲可以在阿秒时间分辨精度分辨电子轨道^[37]、电子能带^[38]、核与晶格振动^[39]、自旋轨道耦合^[40]等对光电离时间的影响, 深入研究电子及其关联行为^[41-43], 由对价电子的探测与操控发展为对内壳层和紧束缚以及高离化态离子的电子的探测与操控, 对物理、化学、生物、材料以及信息、能源等都具有重要科学意义和应用价值. 本文聚焦电子动力学的强场调控与阿秒探测, 从本课题组的研究角度评述相关工作和现状.

1 自电离态的时域探测

阿秒时域测量的第一个重要实验^[44]是对原子内壳

层电子及其关联动力学的时间分辨. 如图1所示, 利用中心光子能量约100 eV、脉宽900 as的阿秒脉冲电离气相氪原子(Kr). 阿秒脉冲敲掉一个内壳层电子, 制备了一个M壳层电子空穴. 由于电子关联, 空穴态不稳定, 会发生自发的俄歇衰变, 产生第二个电子-俄歇电子. 在光电离的同时, 施加与阿秒脉冲同步的弱红外激光脉冲, 导致光电子能谱和俄歇电子能谱出现调制. 通过改变二者时间延迟, 利用阿秒条纹相机技术(同样的方法也被用于阿秒脉冲脉宽诊断)^[45-48], 可以在时域上以阿秒精度分辨俄歇动力学过程. 实验发现, 氪原子M壳层电子空穴态寿命约为8 fs.

针对实验的理论解释, 我们与其他小组几乎同时建立了一个普适的时域量子理论^[49,50], 特别是我们首次给出了Fano线型的时域表示^[49].

$$F_{\text{Fano}}(t) = \frac{\Gamma}{2}(q - i)e^{iE_r t - \Gamma t/2} + i\delta(t), \quad (1)$$

其中 Γ 对应Fano共振的线宽, E_r 对应共振的能量, q 是Fano线型参数. 利用该理论可以更一般地处理激光协同下阿秒脉冲诱导的自电离过程. 自电离态是嵌入在连续态的一个束缚态, 通常由于电子关联导致二者的耦合, 使得自电离态不稳定, 进而自发电离发射电子. 注意到, 存在产生光电子的两个量子通道——经过共振中间态的自电离和直接阿秒光电离(二者之间的权重与 q 参数直接相关). 其中, 前者是在红外光脉宽内一

图1 (网络版彩色)阿秒脉冲光电离Kr诱导俄歇衰变过程的时域探测示意图. 根据文献^[44]绘制

Figure 1 (Color online) Photoionization of Kr by attosecond pulses and the subsequent Auger decay. Adapted from Ref. ^[44]

个较长的时间段内持续发生,而后者主要发生在很短的阿秒脉冲宽度内,因而红外光对这两个量子通道的调制不同,导致二者的干涉即Fano线型随红外光与阿秒脉冲的时间延迟而变化。如果阿秒脉冲宽很长,光电子能谱分布会在共振峰外发展出一个相差一个光子能量的边带峰,反映阿秒和红外光子同时参与的多光子干涉。进一步实验^[51]表明,利用阿秒脉冲可以实现中间瞬态过程及其耦合的时间分辨,为多电子关联等复杂动力学过程的深入研究开辟了道路。同时,该理论也指向了一个重要的问题,即Fano线型在满足测不准关系的频率域和时域上是如何形成的^[52~55],为利用阿秒瞬态吸收技术探测电子波包相干性及电子关联提供了支持。如图2所示,XUV(extreme ultra-violet)阿秒脉冲制备的自电离态与基态形成的偶极振荡随时间衰减,其辐射与XUV之间的作用可以利用XUV吸收谱进行探测。在某个延迟下施加红外光会电离抽运自电离态,终结自电离态的自发衰减和对应的辐射,导致吸收谱被调制。当延迟足够大时(对应探测时间无穷长),整个自电离衰减过程参与辐射,吸收谱呈现出Fano线型。在较小延迟时,只有有限时间段的自电离过程参与吸收谱的贡献,导致吸收谱较宽。随时间延迟增大,主峰旁波纹增多的同时也在降低,与之相反,主峰不断增高,最终形成Fano线型。

2 强场电离的泡利效应和反屏蔽效应

红外强激光电离原子通常是首先剥离价电子。由于单个光子能量远低于第一电离能,难以穿透外壳层电子的屏蔽,因此通常采用冻结原子实的单活跃电子近似处理闭壳层原子分子(如氖原子Ne、氙原子Xe、氮气分子 N_2)的强场电离^[56,57]。通过把针对原子情形的隧穿电离理论拓展到分子体系,发展的分子ADK(Amosov-Delone-Krainov)理论取得了很大的成功^[58]。该理论正确解释了电离能接近的氮气分子与氩原子(Ar)的电离速率比较接近,而与氙原子电离能比较接近的氧气分子(O_2),其电离速率则小很多。分子ADK理论通过考虑电子云密度分布的分波对称性,给出依赖分子取向的电离速率,为分子电离、库仑爆炸、分子解离等实验提供了重要参考^[59,60],得到了广泛应用。但是实际上对于开壳层原子和具有几何构型的分子而言,外壳层电子对外场的屏蔽强度实际上是随取向变化的^[61,62]。同时,由于分子特殊的几何构型,不同壳层之间的激发能通常也比较小,导致单活跃电子近似与实

图2 (网络版彩色)利用阿秒瞬态吸收探测Fano线型随时间的演化。其中, E 表示偏离共振中心 E_r 的能量差,单位是 $\Gamma/2$;时间 t 的单位是 $1/\Gamma$, $q=2$ 。根据文献^[52]绘制

Figure 2 (Color online) Attosecond transient absorbance of a Fano resonance showing the time-dependent formation of the Fano profile where E is measured from E_r in unit of $\Gamma/2$, t is in unit of $1/\Gamma$ and $q=2$. Adapted from Ref. ^[52]

际物理存在较大差距。对于多电子体系,电离电子受到原子核和其他电子的总作用势。为把隧穿电离理论推广到多电子复杂体系,可以从多电子理论出发推导出单电子的有效理论^[61]。通过假设电子隧穿时原子实绝热变化,引入极化势,较好地解释了实验测得的过渡金属的电离饱和强度。但是,这种冻结原子实并引入平均极化势的近似处理是否普遍成立,有待求解多体体系与强光相互作用的动力学方程进行检验。另外,即便在单活跃电子近似下,根据泡利所提出的电子不可分辨性,实验仍然有可能测到可观的效应。由于原则上我们无法区分被电离电子,在随后的电子与原子实复合发射高次谐波过程中,我们也无法区分电子回到哪个轨道。考虑到这种不可分辨性,我们发展了初步的多电子框架下的高次谐波产生理论,对分子轨道层析成像实验结果给出了成功的解释^[63~66]。

很长一段时间内,人们对线型极性分子的强场电离感到迷惑。对一氧化碳分子(CO),价电子多分布于碳原子附近。根据分子ADK理论,电子密度较大的方向电离速率较大,即当电场从碳原子指向氧原子(电场与永久偶极矩同向,即夹角为 0°)时,电离速率较大。实验发现确实如此^[67],但是电场反方向时的电离速率却远大于分子ADK理论结果。采取冻结原子实近似的单电子严格计算结果同样与实验结果相反。根据极性分子存在永久偶极矩,人们进一步引入电场与偶极矩平行和反平行时电离能的差异,但是仍然与实验结果不符。通过发展包含全部电子的含时Hartree-Fock程序包^[68,69],

我们针对一氧化碳分子与强激光的作用进行了计算, 所得结果与实验结果吻合很好. 我们发现, 在只考虑最高占据轨道(highest occupied molecular orbital, HOMO)的情况下引入原子实的瞬时极化势^[70], 所得结果与全电子计算结果接近(图3). 这表明虽然内壳层电子对电离的直接贡献较弱, 但其动态极化对外壳层电子在激光场驱动下的动力学有重要影响. 这种极化不能简单地用平均效应(比如电离能随取向变化的静态理论)来处理. 这种动态极化效应可以被理解作为一种动态的反屏蔽效应: 激光场极化原子实导致内壳层电子对原子实的库仑势屏蔽减弱, 强化了对价电子的束缚^[71,72]. 该研究首次表明原子实的动态极化对多电子体系的强场电离具有重要的动力学影响, 意味着对分子极化轨道、动态轨道进行成像的可能, 为进一步发展多电子隧穿^[73]和多通道高次谐波产生理论提供支持.

为了分析电子动态关联对高次谐波的影响, 利用含时Hartree-Fock方法从头计算了一氧化碳分子的高次谐波发射^[74]. 研究发现, 原子实动态极化效应不仅在一氧化碳分子强场隧穿电离中发挥重要作用, 对于HHG, 隧穿电离之后的传播以及再散射过程都受到动态极化的影响. 在复合时, 电子看到的是被激光场和极化场同时作用所变形的轨道(图4). 因此, 原子实的动态极化效应将调制电离概率, 调整电子电离到复合的时间, 使电子复合时的分子轨道产生变形. 该工作揭示了虽然原子实极化带来HOMO的变形近似绝热, 内壳层的电离和激发可以忽略不计, 但其动态极化带来的对HOMO的相互作用被编码在HOMO所发射的谐波谱中, 对谐波的探测有可能实现对动态轨道的成像, 推动多电子体系高次谐波产生的实验和理论研究.

3 电子动力学的多谱段探测与调控

超短激光脉冲与物质作用, 发射高次谐波的同时也会辐射太赫兹波, 频率区间覆盖近6个量级^[75]. 驱动光频率几十甚至上百倍的谐波产生与电子的阿秒动力学直接相关, 而频率是驱动光数1/100的太赫兹则应该对应皮秒尺度的电子动力学. 强光作用气体原子产生太赫兹辐射的机理长时间存在争议. 实验发现, 把基频脉冲和二倍频脉冲同时作用电离气体时, 产生的太赫兹强度提高3个量级以上^[76,77]. 在一个光学周期内改变双色场的时间延迟, 太赫兹波的强度会随之周期性地变化. 只要时间延迟300多阿秒, 太赫兹辐射可以由最强变为最弱甚至消失. 初期理论认为, 双色场产生太赫

图3 (网络版彩色)激光光强 $4 \times 10^{14} \text{ W/cm}^2$ 作用下理论计算及实验测量所得一氧化碳分子电离概率. 其中, SAO代表单轨道近似. SAO+P代表在单轨道近似下引入动态极化势, 所得结果与全电子TDHF计算结果(Full)类似, 并与实验数据^[67]吻合, 表明内壳层电子通过极化对价电子产生反屏蔽, 抑制分子取向 180° (电场指向氧原子)时的电离概率^[70]

Figure 3 (Color online) Calculated and measured ionization probabilities of CO at the laser intensity of $4 \times 10^{14} \text{ W/cm}^2$, where SAO represents the single-active orbital approximation and SAO+P represents the single-active orbital approximation with the core polarization potential. Full curves represent the full time-dependent HF calculation (Full). The agreement with experimental results^[67] demonstrates the antiscreening effects on the valence electrons by the polarized atomic core which suppress the ionization probability at alignment of 180° (the electric field pointing to the oxygen atom)^[70]

图4 (网络版彩色)一氧化碳分子最高占据轨道在电离和复合过程中的动态极化^[74]

Figure 4 (Color online) Dynamical polarization of the highest occupied molecular orbital of CO during the ionization and recombination^[74]

兹机制是四波混频^[76]. 但是, 中性气体或者等离子体的三阶非线性极化率都太小, 与实验不符. 后期理论认为, 太赫兹的产生与从中性气体到等离子体的光电离过程密不可分, 不同时刻电离的电子波包相干运动产生的光电流导致太赫兹辐射^[78]. 这两个理论对双色场相位延迟的依赖相差 0.5π , 对应时间约300 as. 显然, 对双色场电离气体产生太赫兹背后的机制理解取决于对强场

下电子动力学过程的深入分析和阿秒控制。实验上我们基于阿秒钟技术^[12]，利用偏振平行的双色场(800 nm、25 fs红外脉冲和它的倍频场)对氩原子电离动力学进行控制。虽然其中倍频场很微弱，但是体系的对称性被破坏，相邻半周期内辐射的相位差被强调制，导致偶次谐波产生和高强度太赫兹辐射。通过同时监测二者随双色场相位延迟的变化^[12,79]，我们开发高次谐波和太赫兹光谱同步探测与调控的HATS(high-harmonic and terahertz spectroscopy)技术，利用不同阶次谐波发光时刻的阿秒变化，精确获得双色场绝对相位延迟，时间分辨达到70 as(图5)。分析发现，在亚周期的电子动力学过程中，库仑势起着透镜的重要作用。由于库仑聚焦和散焦效应，直接电离离开原子的电子和再散射并被聚焦的电子对太赫兹辐射的贡献不同。双色场调制了二者的相对贡献，产生了宏观的电流辐射太赫兹。这表明再散射过程是太赫兹波和高次谐波产生背后的统一物理图像，其中太赫兹辐射对应电子与原子实的软散射，而高次谐波对应电子与原子实的硬碰撞，从而统一了四波混频和光电流模型，对于未来发展高效强太赫兹源产生手段以及结合太赫兹与阿秒辐射对动力学过程^[80,81]进行刻画有着重要启发。进一步研究表明，强场电离产生的低能电子与太赫兹存在关联^[82,83]。通过应用三维经典轨迹蒙特卡罗方法研究强激光作用下的阈值上电离，并通过把电子出射能谱与电离时刻对应的隧穿坐标联系，确认了库仑势对电子轨迹和末态动量的影响。结果表明，通过控制双色场相位延迟，可以调制电子能谱，而且不同能段调制不同。电子谱、电子产量以及光电流的调制表明了它们之间存在有机联系，支持了我们所提出的双色场产生太赫兹的机制主要源于电子与库仑场的软散射。该机制对于少周期激光脉冲产生太赫兹同样成立，但是产生的光电流及对应的太赫兹辐射对载波包络相位会有较强的依赖。

基于我们发展的HATS技术，通过用双色场电离准直分子，对角向高次谐波与太赫兹进行同步测量，实现了单次实验就可以测量分子角分辨光电离截面的全光探测方法^[81]。以往的实验方案中，通过探测高次谐波辐射强度得到复合截面。但是，由于高次谐波辐射强度依赖于电离强度和复合截面，其中不同角度的电离强度通过理论模型进行估计，不可避免地引入由模型带来的不确定性。通过对离子产额进行独立测量获取角向电离强度，则存在由于离子探测和谐波探测实验条件

图5 (网络版彩色)太赫兹和谐波辐射随基频和倍频脉冲间相位延迟的调制^[12]
Figure 5 (Color online) Yields of THz and high harmonic as functions of the phase delay between the fundamental and its second harmonic pulses^[12]

所需气体密度不同而带来的参数不一致性问题。在HATS技术方案中，针对分子各种取向对太赫兹和高次谐波同步探测，避免了以上所提到的问题。我们的理论研究表明，双色场产生的太赫兹电场强度正比于分子在不同角度下的隧穿电离概率^[80]。因此，可以利用太赫兹对电离强度进行标定，单次实验即可得到光复合微分截面(图6)。由于光电离截面承载分子的轨道结构以及相位等信息，通过分析不同谐波能量的微分截面，特别是角向极小值随谐波能量的变化，可以得到氮气分子轨道不同分波之间的相干信息，与单组态计算基本吻合。截面最小处反映了光电离/复合中分子双中心量子干涉效应，其与理论的细微差异有可能源自多电子效应，有待深入研究。该工作不仅实验演示了测量光电离截面的一种新方法，同时为实现强场下分子中电子波包的相干控制提供了可能方案。

4 电子空穴相干性的强场调控和阿秒探测

阿秒光电离或者强场隧穿电离在产生光电子的同时，在原子实内制备了空穴。X射线自由电子激光^[84,85]或高次谐波^[46,86]等相干X射线光源目前已经可以达到飞秒或阿秒的脉宽，足以制备空心原子和诱导随后的奇异电子关联行为，如级联的俄歇过程^[87-90]和电离引起的吸收饱和^[91,92]等。另一方面，强红外激光则会从外壳层开始剥离电子，并持续地驱动电子和对应的空穴。

图6 (网络版彩色)取向氮分子的太赫兹与谐波同步探测(a)及所得角向光电离截面实验与理论预言的比较(b)^[81]
 Figure 6 (Color online) The simultaneous measurement of THz and high harmonics for aligned N_2 (a) and the comparison between experimental results and calculational results for photo-recombination cross sections (b)^[81]

各种多体效应, 如动态极化^[70,73]等对电子和空穴的关联动力学及电子相干性有重要的影响。当内壳电子被红外激光进一步激发并参与超快动力学时, 多电子信息会编码到高次谐波谱。空穴反映了离子由于电子关联^[42,93~95]或者自旋轨道耦合等导致的阿秒尺度瞬态激发^[96~100]。电子空穴的关联动力学及其所导致的量子相干性与电离动力学的快慢直接相关^[101~104]。对应的重要问题是, 在电离过程中空穴是如何演化的, 电子-空穴相干性是如何弛豫的, 电子间的多体相互作用^[105,106]以及电子-核^[123,107]、自旋-轨道等多自由度的耦合在电荷迁移、超导、化学反应和生命过程中扮演何种角色。阿秒条纹相机技术和阿秒瞬态光谱技术为实时观测和操控电子关联提供了重要支持^[87~89,101,102,108~110]。最新研究表明, 不可逆的隧穿电离过程和可逆的电子布居过程共同导致不同自旋态电离时间差异^[100], 对应快速电子剥离可能存在一个普适的原子阿秒响应函数^[106]。

相比之下, 目前对阿秒脉冲驱动下红外光场制备的电子-空穴动力学尚缺乏研究^[111~113]。我们提出结合阿秒脉冲与强红外激光利用阿秒瞬态整形的发射谱^[114,115]技术, 有可能同时对内壳层和价壳层电子空穴动力学进行相干探测和控制, 理解电子重排动力学和电子相干性如何在电子-空穴对或多个通道之间演化和转移^[116]。如图7所示为闭壳层原子与强红外激光脉冲和单阿秒脉冲作用, 其中阿秒脉冲的中心频率对应内壳层和价壳层的跃迁能。在没有红外脉冲的情况下, 由于泡利排斥原理, 直接吸收阿秒光子从内壳层到价

图7 (网络版彩色)闭壳层原子Ne与阿秒脉冲和红外光场同时作用的动力学示意图^[116]
 Figure 7 (Color online) Dynamics during the interaction of an attosecond pulse and an infrared pulse with close-shell atoms Ne^[116]

壳层的偶极跃迁是禁戒的。但是, 一旦红外光场从价壳层电离原子, 就会在价壳层留下与光电子关联的空穴, 同时打开了内壳层到价壳层的跃迁通道, 这样就形成了一个阿秒吸收的控制门。随后阿秒光子的共振吸收则把空穴转移到内壳层。尚未离开原子实并与价壳层空穴耦合的光电子转而与芯空穴耦合。从电离到两个空穴的相干传递导致高次谐波辐射的多个路径(图7): 谐波可以通过复合到价壳层空穴或芯空穴中辐射, 或者通过价芯共振辐射。其中通过价芯共振辐射路径伴随光电子在对应价壳层空穴和芯空穴的不同连续态之间的转移。由于多个通道的干涉, 理论计算发现, 谐波

图 8 (网络版彩色)阿秒脉冲和红外脉冲同时作用氖原子辐射的谐波谱^[116]. (a) 150 as; (b) 500 as; (c) 1 fs
Figure 8 (Color online) Harmonic spectra from neon subjected to the attosecond and IR pulses^[116]. (a) 150 as; (b) 500 as; (c) 1 fs

频谱中表现出明显的由于芯价跃迁的共振峰和复合到内壳层空穴带来的谐波截止频率扩展(图8). 共振峰附近的谐波辐射强度随阿秒与红外脉冲时间延迟的变化表明, 每当激光场强取局部最大值时, 信号就达到局部最大, 原因在于, 红外光场最强时, 价壳层电离产生的空穴占据数最大, 导致芯价跃迁最强. 但是随着时间延迟, 信号并非单调递增, 而是存在调制. 这源于电离过程存在着相干性, 而这种相干性被进一步转移到内壳层空穴-光电子对. 阿秒脉宽越短, 调制越大, 表明利用超短的阿秒脉冲可以探测电离及光跃迁过程的阿秒时间电子相干性.

5 总结与展望

多电子动力学是阿秒物理的重要研究内容, 也是阿秒瞬态技术可以大放异彩的重要领域. 超强和超快光脉冲技术的结合, 提供了对多电子动力学操控和探测的重要手段. 本文主要从原子分子物理角度, 对其中的部分课题进行了简单评述. 显然, 这些技术同样可以而且正在应用到液相和固相凝聚态物理以及纳米科学等. 可以预期, 阿秒脉冲技术的发展和运用将使我们在更短的时间精度和更高的空间分辨上对微观电子动力学及其关联有新的认识, 物质科学的新革命正在到来.

参考文献

- 1 Agostini P, Di Mauro L F. The physics of attosecond light pulses. *Rep Prog Phys*, 2004, 67: 1563
- 2 Krausz F, Ivanov M. Attosecond physics. *Rev Mod Phys*, 2009, 81: 163–234
- 3 Pabst S. Atomic and molecular dynamics triggered by ultrashort light pulses on the atto-to picosecond time scale. *Eur Phys J-Spec Top*, 2013, 211: 1–71
- 4 Calegari F, Sansone G, Stagira S, et al. Advances in attosecond science. *J Phys B-At Mol Opt Phys*, 2016, 49: 062001
- 5 Young L, Ueda K, Gühr M, et al. Roadmap of ultrafast X-ray atomic and molecular physics. *J Phys B-At Mol Opt Phys*, 2018, 51: 032003
- 6 Lindroth E, Calegari F, Young L, et al. Challenges and opportunities in attosecond and XFEL science. *Nat Rev Phys*, 2019, 1: 107–111
- 7 Baltuška A, Udem T, Uiberacker M, et al. Attosecond control of electronic processes by intense light fields. *Nature*, 2003, 421: 611–615
- 8 Kim K T, Zhang C, Shiner A D, et al. Petahertz optical oscilloscope. *Nat Photonics*, 2013, 7: 958–962
- 9 Garg M, Zhan M, Luu T T, et al. Multi-petahertz electronic metrology. *Nature*, 2016, 538: 359–363
- 10 Ramasesha K, Leone S R, Neumark D M. Real-time probing of electron dynamics using attosecond time-resolved spectroscopy. *Annu Rev Phys Chem*, 2016, 67: 41–63

- 11 Krausz F, Stockman M I. Attosecond metrology: From electron capture to future signal processing. *Nat Photonics*, 2014, 8: 205–213
- 12 Zhang D, Lü Z, Meng C, et al. Synchronizing terahertz wave generation with attosecond bursts. *Phys Rev Lett*, 2012, 109: 243002
- 13 Reimann J, Schlauderer S, Schmid C P, et al. Subcycle observation of lightwave-driven Dirac currents in a topological surface band. *Nature*, 2018, 562: 396–400
- 14 Agostini P, Fabre F, Mainfray G, et al. Free-free transitions following six-photon ionization of xenon atoms. *Phys Rev Lett*, 1979, 42: 1127–1130
- 15 Freeman R R, Bucksbaum P H, Milchberg H, et al. Above-threshold ionization with subpicosecond laser pulses. *Phys Rev Lett*, 1987, 59: 1092–1095
- 16 Paulus G G, Becker W, Nicklich W, et al. Rescattering effects in above-threshold ionization: A classical model. *J Phys B-At Mol Opt Phys*, 1994, 27: L703–L708
- 17 McPherson A, Gibson G, Jara H, et al. Studies of multiphoton production of vacuum-ultraviolet radiation in the rare gases. *J Opt Soc Am B*, 1987, 4: 595–601
- 18 Ferray M, L'Huillier A, Li X F, et al. Multiple-harmonic conversion of 1064 nm radiation in rare gases. *J Phys B-At Mol Opt Phys*, 1988, 21: L31–L35
- 19 L'Huillier A, Lewenstein M, Salières P, et al. High-order harmonic-generation cutoff. *Phys Rev A*, 1993, 48: R3433–R3436
- 20 Chang Z, Rundquist A, Wang H, et al. Generation of coherent soft X rays at 2.7 nm using high harmonics. *Phys Rev Lett*, 1999, 79: 1795–1795
- 21 Posthumus J H. The dynamics of small molecules in intense laser fields. *Rep Prog Phys*, 2004, 67: 623–665
- 22 Lu P F, Wang J P, Li H, et al. High-order above-threshold dissociation of molecules. *Proc Natl Acad Sci USA*, 2018, 115: E3323
- 23 Wu J, Kunitski M, Pitzer M, et al. Electron-nuclear energy sharing in above-threshold multiphoton dissociative ionization of H₂. *Phys Rev Lett*, 2013, 111: 023002
- 24 Mourou G A, Tajima T, Bulanov S V. Optics in the relativistic regime. *Rev Mod Phys*, 2006, 78: 309–371
- 25 Corkum P B. Plasma perspective on strong field multiphoton ionization. *Phys Rev Lett*, 1993, 71: 1994–1997
- 26 Piraux B, L'Huillier A, Rzażewski K. Super-intense laser-atom physics. *Nato Sci*, 1993, doi: 10.1007/978-1-4615-7963-2
- 27 Schafer K J, Yang B, Dimauro L F, et al. Above threshold ionization beyond the high harmonic cutoff. *Phys Rev Lett*, 1993, 70: 1599–1602
- 28 Lewenstein M, Balcou P, Ivanov M Y, et al. Theory of high-harmonic generation by low-frequency laser fields. *Phys Rev A*, 1994, 49: 2117–2132
- 29 Kienberger R, Hentschel M, Uiberacker M, et al. Steering attosecond electron wave packets with light. *Science*, 2002, 297: 1144–1148
- 30 Feng X, Gilbertson S, Mashiko H, et al. Generation of isolated attosecond pulses with 20 to 28 femtosecond lasers. *Phys Rev Lett*, 2009, 103: 183901
- 31 Ghimire S, Reis D A. High-harmonic generation from solids. *Nat Phys*, 2018, 15: 10–16
- 32 Zhao K, Zhang Q, Chini M, et al. Tailoring a 67 attosecond pulse through advantageous phase-mismatch. *Opt Lett*, 2012, 37: 3891
- 33 Kim K T, Zhang C, Ruchon T, et al. Photonic streaking of attosecond pulse trains. *Nat Photonics*, 2013, 7: 651–656
- 34 Chini M, Zhao K, Chang Z. The generation, characterization and applications of broadband isolated attosecond pulses. *Nat Photonics*, 2014, 8: 178–186
- 35 Li J, Ren X, Yin Y, et al. 53-attosecond X-ray pulses reach the carbon K-edge. *Nat Commun*, 2017, 8: 186
- 36 Wang X, Wang L, Xiao F, et al. Generation of 88 as isolated attosecond pulses with double optical gating. *Chin Phys Lett*, 2020, 37: 023201
- 37 Klünder K, Dahlström J, Gisselbrecht M, et al. Probing single-photon ionization on the attosecond time scale. *Phys Rev Lett*, 2011, 106: 143002
- 38 Cavalieri A L, Müller N, Uphues T, et al. Attosecond spectroscopy in condensed matter. *Nature*, 2007, 449: 1029–1032
- 39 Haessler S, Fabre B, Higuier J, et al. Phase-resolved attosecond near-threshold photoionization of molecular nitrogen. *Phys Rev A*, 2015, 80: 011404
- 40 Jordan I, Huppert M, Pabst S, et al. Spin-orbit delays in photoemission. *Phys Rev A*, 2017, 95: 013404
- 41 Leone S R, McCurdy C W, Joachim B, et al. What will it take to observe processes in 'real time'? *Nat Photonics*, 2014, 8: 162–166
- 42 Ossiander M, Siegrist F, Shirvanyan V, et al. Attosecond correlation dynamics. *Nat Phys*, 2017, 13: 280–285
- 43 Silva R E F, Jiménez-Galán Á, Amorim B, et al. Topological strong-field physics on sub-laser-cycle timescale. *Nat Photonics*, 2019, 13: 849–854
- 44 Drescher M, Hentschel M, Kienberger R, et al. Time-resolved atomic inner-shell spectroscopy. *Nature*, 2002, 419: 803–807
- 45 Drescher M, Hentschel M, Kienberger R, et al. X-ray pulses approaching the attosecond frontier. *Science*, 2001, 291: 1923–1927
- 46 Hentschel M, Kienberger R, Spielmann C, et al. Attosecond metrology. *Nature*, 2001, 414: 509–513
- 47 Itatani J, Quéré F, Yudin G L, et al. Attosecond streak camera. *Phys Rev Lett*, 2002, 88: 173903
- 48 Kitzler M, Milosevic N, Scrinzi A, et al. Quantum theory of attosecond XUV pulse measurement by laser dressed photoionization. *Phys Rev Lett*, 2002, 88: 173904
- 49 Zhao Z X, Lin C D. Theory of laser-assisted autoionization by attosecond light pulses. *Phys Rev A*, 2005, 71: 362–368
- 50 Wickenhauser M, Burgdörfer J, Krausz F, et al. Time resolved Fano resonances. *Phys Rev Lett*, 2005, 94: 023002
- 51 Wang H, Chini M, Chen S, et al. Attosecond time-resolved autoionization of argon. *Phys Rev Lett*, 2010, 105: 143002

- 52 Kaldun A, Blättermann A, Stooß V, et al. Observing the ultrafast buildup of a Fano resonance in the time domain. *Science*, 2016, 354: 738–741
- 53 Gruson V, Barreau L, Jiménez-Galan Á, et al. Attosecond dynamics through a Fano resonance: Monitoring the birth of a photoelectron. *Science*, 2016, 354: 734–738
- 54 Busto D, Barreau L, Isinger M, et al. Time-frequency representation of autoionization dynamics in helium. *J Phys B-At Mol Opt Phys*, 2018, 51: 044002
- 55 Desrier A, Maquet A, Taieb R, et al. Ionization dynamics through a Fano resonance: Time-domain interpretation of spectral amplitudes. *Phys Rev A*, 2018, 98: 053406
- 56 Keldysh L V. Ionization in the field of a strong electromagnetic wave. *Sov Phys JETP*, 1965, 20: 1307
- 57 Ammosov M V, Delone N B, Krainov V P. Tunnel ionization of complex atoms and of atomic ions in an alternating electromagnetic field. *Sov Phys JETP*, 1986, 64: 1191
- 58 Tong X M, Zhao Z X, Lin C D. Theory of molecular tunneling ionization. *Phys Rev A*, 2002, 66: 033402
- 59 Zhao Z X, Tong X M, Lin C D. Alignment-dependent ionization probability of molecules in a double-pulse laser field. *Phys Rev A*, 2003, 67: 043404
- 60 Lin C D, Tong X M, Zhao Z X. Effects of orbital symmetries on the ionization rates of aligned molecules by short intense laser pulses. *J Mod Opt*, 2006, 53: 21–33
- 61 Zhao Z X, Brabec T. Tunnel ionization of open-shell atoms. *J Phys B-At Mol Opt Phys*, 2006, 39: L345–L351
- 62 Zhao Z, Brabec T. Tunnel ionization in complex systems. *J Mod Opt*, 2007, 54: 981–997
- 63 Patchkovskii S, Zhao Z, Brabec T, et al. High harmonic generation and molecular orbital tomography in multielectron systems: Beyond the single active electron approximation. *Phys Rev Lett*, 2006, 97: 123003
- 64 Zhao Z X, Yuan J M, Brabec T. Multielectron signatures in the polarization of high-order harmonic radiation. *Phys Rev A*, 2007, 76: 239–253
- 65 Patchkovskii S, Zhao Z, Brabec T, et al. High harmonic generation and molecular orbital tomography in multielectron systems. *J Chem Phys*, 2007, 126: 114306
- 66 Zhao Z, Wu J, Yuan J. Toward tomographic imaging of singlet and triplet states using high-order harmonic generation. *J Phys B-At Mol Opt Phys*, 2008, 41: 155601
- 67 Wu J, Schmidt L P H, Kunitski M, et al. Multiorbital tunneling ionization of the CO molecule. *Phys Rev Lett*, 2014, 108: 183001
- 68 Zhang B, Yuan J, Zhao Z. DMTDHF: A full dimensional time-dependent Hartree-Fock program for diatomic molecules in strong laser fields. *Comput Phys Commun*, 2015, 194: 84–96
- 69 Hu S, Zhao Z X, Chen J, et al. Ionization dynamics of C_2H_2 in intense laser fields: Time-dependent Hartree-Fock approach. *Phys Rev A*, 2015, 92: 053409
- 70 Zhang B, Yuan J, Zhao Z. Dynamic core polarization in strong-field ionization of CO molecules. *Phys Rev Lett*, 2013, 111: 163001
- 71 Rapp J, Bauer D. Effects of inner electrons on atomic strong-field-ionization dynamics. *Phys Rev A*, 2014, 89: 033401
- 72 Bauer J H, Mota-Furtado F, O'Mahony P F, et al. Ionization and excitation of the excited hydrogen atom in strong circularly polarized laser fields. *Phys Rev A*, 2014, 90: 063402
- 73 Zhao Z X, Yuan J M. Dynamical core polarization of two-active-electron systems in strong laser fields. *Phys Rev A*, 2014, 89: 023404
- 74 Zhang B, Yuan J M, Zhao Z X. Dynamic orbitals in high-order harmonic generation from CO molecules. *Phys Rev A*, 2014, 90: 035402
- 75 Zhou Z, Zhang D, Zhao Z, et al. Terahertz emission of atoms driven by ultrashort laser pulses. *Phys Rev A*, 2009, 79: 063413
- 76 Cook D J, Hochstrasser R M. Intense terahertz pulses by four-wave rectification in air. *Opt Lett*, 2000, 25: 1210–1212
- 77 Dai J, Karpowicz N, Zhang X C. Coherent polarization control of terahertz waves generated from two-color laser-induced gas plasma. *Phys Rev Lett*, 2009, 103: 023001
- 78 Kim K Y, Glowina J H, Taylor A J, et al. Terahertz emission from ultrafast ionizing air in symmetry-broken laser fields. *Opt Express*, 2007, 15: 4577–4584
- 79 Lü Z H, Zhang D W, Meng C S, et al. Attosecond synchronization of terahertz wave and high-harmonics. *J Phys B-At Mol Opt Phys*, 2013, 46: 155602
- 80 Chen W, Huang Y, Meng C, et al. Theoretical study of terahertz generation from atoms and aligned molecules driven by two-color laser fields. *Phys Rev A*, 2015, 92: 033410
- 81 Huang Y, Meng C, Wang X, et al. Joint measurements of terahertz wave generation and high-harmonic generation from aligned nitrogen molecules reveal angle-resolved molecular structures. *Phys Rev Lett*, 2015, 115: 123002
- 82 Liu J, Chen W, Zhang B, et al. Trajectory-based analysis of low-energy electrons and photocurrents generated in strong-field ionization. *Phys Rev A*, 2014, 90: 063420
- 83 Liu J, Fu Y, Chen W, et al. Offset angles of photocurrents generated in few-cycle circularly polarized laser fields. *J Phys B-At Mol Opt Phys*, 2017, 50: 055602

- 84 Emma P, Akre R, Arthur J, et al. First lasing and operation of an angstrom-wavelength free-electron laser. *Nat Photonics*, 2010, 4: 641–647
- 85 Rohringer N, Ryan D, London R A, et al. Atomic inner-shell X-ray laser at 1.46 nanometres pumped by an X-ray free-electron laser. *Nature*, 2012, 481: 488–491
- 86 Paul P M. Observation of a train of attosecond pulses from high harmonic generation. *Science*, 2011, 292: 1689
- 87 Young L, Kanter E P, Krässig B, et al. Femtosecond electronic response of atoms to ultra-intense X-rays. *Nature*, 2010, 466: 56–61
- 88 Erk B, Boll R, Trippel S, et al. Imaging charge transfer in iodomethane upon X-ray photoabsorption. *Science*, 2014, 345: 288–291
- 89 McFarland B K, Farrell J P, Miyabe S, et al. Ultrafast X-ray auger probing of photoexcited molecular dynamics. *Nat Commun*, 2014, 5: 4235
- 90 Li Z, Vendrell O, Santra R. Ultrafast charge transfer of a valence double hole in glycine driven exclusively by nuclear motion. *Phys Rev Lett*, 2015, 115: 143002
- 91 Rackstraw D, Ciricosta O, Vinko S, et al. Saturable absorption of an X-ray free-electron-laser heated solid-density aluminum plasma. *Phys Rev Lett*, 2015, 114: 015003
- 92 Fukuzawa H, Son S K, Motomura K, et al. Deep inner-shell multiphoton ionization by intense X-ray free-electron laser pulses. *Phys Rev Lett*, 2013, 110: 173005
- 93 Sansone G, Pfeifer T, Simeonidis K, et al. Electron correlation in real time. *ChemPhysChem*, 2012, 13: 661–680
- 94 Isinger M, Squibb R J, Busto D, et al. Photoionization in the time and frequency domain. *Science*, 2017, 358: 893–896
- 95 Silva R E F, Blinov I V, Rubtsov A N, et al. High-harmonic spectroscopy of ultrafast many-body dynamics in strongly correlated systems. *Nat Photonics*, 2018, 12: 266–270
- 96 Rottke H, Ludwig J, Sandner W. 'Short' pulse MPI of xenon: The ionization channel. *J Phys B-At Mol Opt Phys*, 1996, 29: 1479–1487
- 97 Young L, Arms D A, Dufresne E M, et al. X-ray microprobe of orbital alignment in strong-field ionized atoms. *Phys Rev Lett*, 2006, 97: 083601
- 98 Bryan W A, Stebbings S L, McKenna J, et al. Atomic excitation during recollision-free ultrafast multi-electron tunnel ionization. *Nat Phys*, 2006, 2: 379–383
- 99 Rohringer N, Santra R. Multichannel coherence in strong-field ionization. *Phys Rev A*, 2009, 79: 053402
- 100 Fechner L, Camus N, Ullrich J, et al. Strong-field tunneling from a coherent superposition of electronic states. *Phys Rev Lett*, 2014, 112: 213001
- 101 Goulielmakis E, Loh Z H, Wirth A, et al. Real-time observation of valence electron motion. *Nature*, 2010, 466: 739–743
- 102 Ott C, Kaldun A, Argenti L, et al. Reconstruction and control of a time-dependent two-electron wave packet. *Nature*, 2014, 516: 374–378
- 103 Sabbar M, Heuser S, Boge R, et al. Resonance effects in photoemission time delays. *Phys Rev Lett*, 2015, 115: 133001
- 104 Sabbar M, Timmers H, Chen Y J, et al. State-resolved attosecond reversible and irreversible dynamics in strong optical fields. *Nat Phys*, 2017, 13: 472–478
- 105 Cederbaum L S, Zobeley J. Ultrafast charge migration by electron correlation. *Chem Phys Lett*, 1999, 307: 205–210
- 106 Breidbach J, Cederbaum L S. Universal attosecond response to the removal of an electron. *Phys Rev Lett*, 2015, 94: 033901
- 107 Ferré, A, Boguslavskiy A E, Dagan M, et al. Multi-channel electronic and vibrational dynamics in polyatomic, high-order harmonic generation. *Nat Comm*, 2014, 6: 5952
- 108 Schultze M, Fiess M, Karpowicz N, et al. Delay in photoemission. *Science*, 2010, 328: 1658–1662
- 109 Vrakking M J J. Attosecond imaging. *Phys Chem Chem Phys*, 2014, 16: 2775–2789
- 110 Kobayashi Y, Reduzzi M, Chang K F, et al. Selectivity of electronic coherence and attosecond ionization delays in strong-field double ionization. *Phys Rev Lett*, 2018, 120: 146–151
- 111 Mukamel S, Healton D, Zhang Y, et al. Multidimensional attosecond resonant X-ray spectroscopy of molecules: Lessons from the optical regime. *Annu Rev Phys Chem*, 2013, 64: 101–127
- 112 Leeuwenburgh J, Cooper B, Averbukh V, et al. High-order harmonic generation spectroscopy of correlation-driven electron hole dynamics. *Phys Rev Lett*, 2013, 111: 123002
- 113 Leeuwenburgh J, Cooper B, Averbukh V, et al. Reconstruction of correlation-driven electron-hole dynamics by high-harmonic-generation spectroscopy. *Phys Rev A*, 2014, 90: 033426
- 114 Beaulieu S, Bloch E, Barreau L, et al. Phase-resolved two-dimensional spectroscopy of electronic wave packets by laser-induced XUV free induction decay. *Phys Rev A*, 2017, 95: 041401
- 115 Cao W, Warrick E R, Neumark D M, et al. Attosecond transient absorption of argon atoms in the vacuum ultraviolet region: Line energy shifts versus coherent population transfer. *New J Phys*, 2016, 18: 013041
- 116 Zhao J, Yuan J M, Zhao Z X. Strong-field ionization inducing multi-electron-hole coherence probed by attosecond pulses. 2015, arxiv: 1510.07947

Summary for “多电子动力学的强场调控与阿秒探测”

Strong-field control and attosecond probing of multielectron dynamics

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The past thirty years witnessed rapid developing of strong field physics that leads to the first generation of attosecond (10^{-18} s) pulses. The duration of available pulses has recently been reduced to less than 100 as, approaching the Kepler period of a classical electron revolving around the nucleus. For the first time, electron motions inside atoms, molecules and solids can be traced, probed and even steered in real time. It pushed the study on ultrafast dynamic processes into a new frontier, giving birth to attosecond physics. The development of attosecond pulses provides unprecedented coherent ultrashort XUV (extreme ultra-violet) of soft X-ray sources that enable imaging the atomic structure and resolving the related quantum dynamics for matter science. In particular, it allows the probing and monitoring of electron dynamics in the natural time scale of attoseconds with atomic spatial resolution. The generation and applications of attosecond pulses are directly related to the control and manipulation of the sub-cycle electron dynamics. Recent studies on the correlated multielectron dynamics have provided insight into the optical and dynamical properties of matter in the perspectives of time, phase and entanglement. It deepens our understanding on some of the fundamental questions, e.g., how the single or multiple photons are absorbed and how short are the quantum processes such as photoionization, tunneling ionization and charge migration. Although currently attosecond pulses are limited by the flux and pulse duration, the combination of attosecond pulses with intense infrared laser pulses paves new ways of controlling and probing electron dynamics in the domains of energy, time and space that lays the foundation for petahertz optoelectronics and attosecond pulse based transient spectroscopy. In this review, we will first briefly summarize the advancement of strong field and ultrafast physics including the relevant experimental findings, the theoretical explorations, the related technologies and the possible applications. Then we focus on the sub-cycle electron or multielectron dynamics and their manifestation on the ionization and coherent radiation properties. We start with the time behavior of Fano resonance and how it can be probed in time domain. In section 2, the correlated electron dynamics is discussed in term of anti-screening effect and Pauli effect that influence strong field ionization and high harmonic generation processes. In section 3, the coherent emission with frequency from terahertz to soft XUV and their joint measurement are discussed by emphasizing the generation mechanism of terahertz wave in two-color laser pulses. The developed high-harmonic and terahertz wave spectroscopy (HATS) is shown capable of imaging the molecular structure and dynamics. In section 4, the time-resolving of electron and hole dynamics and their coherence is proposed based on attosecond transient absorption technique. The coherence of ionization and the correlated electron dynamics is hinted crucial for electron-hole interaction. The topics are very limited and mainly selected from the perspectives of our group.

attosecond pulse, high harmonic generation, strong-field, multielectron, correlation

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