·强激光物理与技术·



液体薄膜靶在激光驱动辐射源和 激光离子加速中的应用

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摘 要: 流动的无支撑液体薄膜在各个领域有着广泛应用。超强激光作用在这样的薄膜上,可产生涵盖 太赫兹到伽马射线的高亮度次级辐射及高能的离子,并具有高重频、低成本、可连续工作等显著优势。概述了 液体薄膜靶的制备和表征方法,阐明了液体薄膜靶相对于传统靶材的特性和优势,并对其在激光驱动辐射源和 激光离子加速中的应用做出了总结和展望。

Application of liquid film targets in laser-driven radiation sources and laser ion acceleration

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Abstract: Flowing unsupported liquid thin films are widely used in various fields. Energetic ions and high brightness secondary radiation covering THz to gamma rays will be generated when super intense laser acting on such a film, with the significant advantages of high repetition-rate, low cost and continuous operation. Methods for preparing liquid films include impinging jets, converging nozzle, gas-dynamic nozzle and wire-guided jet, each with its own characteristics, which can be measured by optical diagnosis. White light interference and monochromatic light interferometry are introduced, and an example is given to illustrate these two methods. This paper provides an overview of the preparation and characterization of liquid thin film target, and summarizes its applications and prospect in laser-driven radiation sources as well as laser ion acceleration.

Key words: liquid film, laser-driven radiation source, laser-drive ion acceleration, high repetition-rate

处于流动状态的液体在一定条件下可形成厚度在 μm 到 nm 尺度、无需衬底或容器而独立存在的薄膜。其中 的液体分子不断更替,受到扰动或破坏后可快速地恢复初始状态。这样的液体薄膜在 20 世纪 50 年代就被用于火 箭发动机的推进器部分,实现燃料与氧化剂的雾化及掺混^[1],也曾被作为可饱和吸收体用于染料激光器实现超短 脉冲输出^[2]。超薄的液体膜也是优良的样品载体,可用于测量不同液体或溶液的吸收光谱^[3-5],以及与水辐解、氢键 强化相关的超快过程研究^[69]等。

近年来,人们发现超短超强激光与固体薄膜靶材相互作用能够产生涵盖太赫兹到伽马射线的极宽波段内的高 亮度辐射^[10-11],以及高能的质子、重离子^[12]。激光驱动的辐射源和离子源具有尺寸小、脉宽短、瞬态流强高等特 点,在高分辨成像、聚变快点火、质子放疗等^[13-14]领域都有着巨大的应用潜力。液体薄膜的密度、厚度等主要参数 与固体薄膜靶接近,在很多情况下完全可替代后者,而使用液体薄膜作为激光打靶的靶材,具有高重频、低成本、

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可连续工作等显著优势。因此,基于液体薄膜靶的激光驱动辐射产生和离子加速引起了越来越多的关注。本文将 介绍液体薄膜靶的制备、表征方法,并对其在激光驱动辐射源和激光离子加速中的应用做出综述和展望。

1 液体薄膜靶的制备方法

最早关于液体薄膜制备方法的报道见于 1833 年 Savart 的工作^[15]。他们使用两束高速液体射流以一定的角度对撞, 在正交的方向上形成液膜。其原理为:两束液柱在相向方向 的动量相互抵消,在正交方向上形成辐射状动量,并在表面 张力的作用下形成闭合液膜,如图 1 所示。由于成膜因素复 杂,自 1960 年代起,许多理论模型被用于对该方法进行定量 描述^[16-21],研究者发现:射流直径越小,液膜越薄;对撞夹角越 大,液膜越薄且长宽比越小;高粘度流体会使得成膜更厚更 均匀^[22];当两束液柱错开而非对心对撞时,液膜会更薄且有 一定偏转角度^[23-24],如图 1(d)所示。2018 年, Morrison 等人利 用双柱错位对撞法制备了最薄 450 nm 的液膜^[25],这是该方法 目前报道的最薄厚度。双柱对撞法因其较高的成膜质量和 调控自由度而得到了广泛的应用,诸如航天发动机^[2]、纳米 颗粒合成^[26]、化学反应器^[27]、废水处理^[28]等场合。

除了双柱对撞法,另一种制备液体薄膜靶的方法是使用 收束型喷嘴,原理与双柱对撞法类似。不同的是,其"对撞" 的过程是在喷嘴内部完成。该方法在1950年代就有过报道^[29], 并曾被用于染料激光器^[2]。2017年, Galinis等人使用双光子



3D 打印的精密方形收束喷嘴制备了稳定的液膜^[30]; 2018 年, Ha 等人研究了液膜尺寸及厚度对方形喷嘴长宽、收束 角度和流体物性的依赖关系^[31]; 2022 年, Crissman 等人用玻璃收束喷嘴得到了最薄 250 nm 的液膜^[32]。相比双柱对 撞法,收束喷嘴法装置简易,无需对准,状态稳定,具有较高的可移植性,但调节的自由度较低,仅能通过改变流量 来控制液膜。

2018年,在收束喷嘴的基础上,Koralek等人提出了基于微流控芯片制备液体薄膜靶的方案^[4]。他们通过两个高速气体通道对液体射流进行吹夹而形成液膜,这种方法能够实现厚度小于 20 nm 的超薄液膜,使其在近两年的超快过程和物质结构的探测中得到了应用^[6-9]。2021年,Belšak等人通过流体模拟研究了气体流量、吹夹角度和液体物性对成膜的影响^[33]。气体吹夹法在保持简易和稳定的同时增加了气相的调节自由度,相比收束喷嘴法更为灵活。但此方法不适用于高真空条件下,这限制了其应用范围。

另一种简易的制备液体薄膜的方法是使用两根金属丝导引,让液体在重力作用下落帘式流过形成液膜^[34]。这 种方法适用于低粘度液体或溶液,对流路系统要求较低,制备形成的液体薄膜厚度通常在 10 μm 以上。表1总结 了上述4种常见液膜制备方法的特点。

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Table 1	Characteristics	s of the common	ı liquid film pr	eparation n	neth	ods	
	表 1	常见液膜制备	百法的特点				

method	minimum thickness/nm	other features		
impinging jets	450	high adjustability, complex device, low stability		
converging nozzle	250	simple device, high stability, low adjustability		
gas-dynamic nozzle	$\sim \! 20$	simple device, high stability, cannot be used in vacuum		
wire-guided jet	\sim 5000	simple device, low repetition rate capability		

2 液体薄膜靶的表征

对于薄膜各项性能指标的表征是薄膜制备后不可或缺的环节。在激光打靶实验中,薄膜厚度及均匀性对于某些应用如激光离子加速有着至关重要的影响^[10-13]。由于液体薄膜无支撑基底且不能承受应力,所以常采用光学的

方法测量膜厚。常见的方法有反射光谱法、单色光干涉法等。

反射光谱法是利用薄膜的白光干涉来测量膜厚。当使用白光照射薄膜时,薄膜两个表面之间的光的干涉会导致不同波长反射光的反射率不同,光谱呈周期性振荡。具体而言,当一束宽谱光以*θ*角入射厚度为*h*的薄膜,其材料 折射率为*n*(*λ*),设反射光谱的两个反射极大峰位分别为*λ*_i和*λ*_j,若其间还有(*j*-*i*-1)个极大值,则膜厚的表达式为

$$h = \left[\frac{2\sqrt{n^2(\lambda_i) - \sin^2\theta}}{(j-i)\lambda_i} - \frac{2\sqrt{n^2(\lambda_j) - \sin^2\theta}}{(j-i)\lambda_j} \right]^{-1}$$
(1)

单色光干涉法采用一束单色平行光照明膜区, 在反射方向观察膜面的干涉条纹, 从而获得整个膜区的相对厚度分布。干涉图样中的明暗条纹可以直观地给出液膜的等厚线, 方便判断液膜的整体均匀性和厚度变化率, 也可以通过相位恢复和解缠等算法获得厚度的分布信息。单色光干涉法结合反射光谱法就可以便捷地测量液膜的厚度空间分布, 而无需逐点扫描。图 2(a)为北京大学激光加速器实验室采用双柱对撞法制备的液体靶, 图 2(b)为液膜面的单色光干涉图样, 其中标注点的反射光谱测量结果如图 2(c)所示。根据测量结果, 选择λ_i = 536.506 nm, λ_{i+4} = 691.879 nm, 可计算得到膜厚为3.34 μm。





除以上方法外,还有一些其它测量液膜靶厚度的方法。例如,Wang等利用脉冲光通过水膜后的延时进行自相 关测量得到膜厚^[33];Koralek等利用水或某种溶液对不同波长光的吸收率曲线,通过测量吸收光谱得到液膜的厚度^[4]; Zhong等通过液膜中产生的阿秒脉冲干涉测量液膜厚度,具有很好的实时性和真空兼容性^[36]。

此外,液膜附近的真空度、高重频打靶时靶体的稳定性、靶面法向角度测控等也是需要研究的重要内容。 2018年, Morrison等人表征液膜在激光扰动后自发恢复时间为70 μs,由此估算最大可支持重频超过10 kHz^[25];2019年, George等人使用侧向显微镜测量液膜法向振动在2 μm内,远小于激光的瑞利长度^[37];2020年, Snyder等人测量 其实验中液膜附近最低能达到的背景压力为4.27 Pa,并研究了背压对离子加速效果的影响^[38]。

3 液体薄膜靶在激光驱动辐射源和激光离子加速中的应用

将强激光聚焦于微米尺度后,在焦斑处与薄膜靶材相互作用可以在极短的时空尺度内产生各种辐射和高能离

子,这为紧凑型辐射源和粒子加速器提供了新的方案。由于每次相互作用后焦斑附近的靶材会被电离为等离子体 而完全损坏,要实现连续稳定输出束流或辐射,就需要源源不断地提供靶材。现有的基于固体靶的方案难以实现 在 kHz 级的高重频下长时间连续打靶^[38-42]。近年来,一些研究者将液体薄膜靶用于激光打靶实验中,取得了许多 意义重大的成果。

3.1 液体薄膜靶产生太赫兹辐射

强太赫兹辐射源对于基础研究和实际应用都有着重要的意义,利用超强激光与气体靶和固体薄膜靶作用可产 生超短超强的太赫兹辐射^[43]。2017年,Qi等人利用边框导引法制备了大约177μm厚的液膜,克服了液体对太赫兹 的强烈吸收问题,首次利用激光与水薄膜靶相互作用产生太赫兹辐射^[44]。他们使用两个相距4mm,直径170μm 的铝丝作为引导边框,制备的液膜厚度在一定范围内可以由流量来调节,最终在1kHz的重频下得到了稳定的宽 谱太赫兹辐射。在实验中他们还测量了辐射强度与激光脉宽和偏振的依赖关系,为进一步提高激光驱动太赫兹源 的亮度奠定了基础。

此后,越来越多的工作将液体薄膜用于激光驱动太赫兹源的研究。2018年,Qi和Yiwen等人使用收束喷嘴法 替代了边框导引法获得了120μm厚的液膜,这种薄膜流速更快,可支持重频更高,能够承受更高激光功率,并研究 了在非对称光场激发下的太赫兹辐射^[4546];2019年,Anton等人利用三种不同参数的收束喷嘴得到的百微米厚的液 膜研究了太赫兹辐射与液体种类、浓度、脉冲宽度之间的关系^[47];2020年,Huang等人使用金属收束喷嘴制备了厚 度小于20μm的水膜,在双脉冲激发下同时得到太赫兹辐射和X-射线^[4849];2020年,Li等人改进边框引导法,使用 两根直径不同的铝丝制备出厚度变化的楔形水膜,进一步证实太赫兹辐射受等离子体密度、等离子体空间分布的影响^[50]; 2021年,Zhao等人使用更细的边框引导制备90μm左右的液膜,用于太赫兹激发下的液体超快动力学探测^[51-53]; 2021年,Ponomareva等人使用文献^{[21}中的收束喷嘴制备的液膜研究了双脉冲激发下预等离子体对太赫兹辐射的增 益作用^[54-56]。此外,液氢、液氮、液态金属等靶材也已经被证实可以用于激光驱动的液体太赫兹辐射源^[57]。

3.2 液体薄膜靶产生高次谐波-X射线

强激光与薄膜靶材的非线性相互作用可以产生最高光子能量到 keV 量级的高次谐波。基于高次谐波的极紫 外/X 射线源、阿秒光源在很多领域都有重要的应用^[58]。

从液体薄膜中产生高次谐波不仅可以提高光源平均亮度,也可用于探索液体中的电子结构。2018年,Luu等人^[59]使用 50 μm 直径液柱对撞制备的 1.2~1.9 μm 厚度水膜和几种醇膜,在 1 kHz 重频下产生了超过 20 eV 的高次谐波,并发现其光谱对能带宽度和带隙非常敏感,表明液相高次谐波光谱学是研究液体中电子结构和亚飞秒电子散射过程的潜在途径。2019年,该团队又在少周期激光(7 fs)激发下得到高次谐波光源可以成为高相干和完全偏振测量^[60],发现谐波偏振态和液体性质与流速无关,证实了激光驱动的高次谐波光源可以成为高相干和完全偏振的新型光源。2021年,Yang等人也用类似的液膜系统在中红外脉冲激光(3.3 μm)下测量到高次谐波^[61]。帝国理工学院的团队利用文献 [30] 的液膜证实高次谐波来源于液体表面而非周围的蒸汽或液体内部,得到比 Luu 等高一倍的谐波截止能量^[62]。2022年,武汉物理与数学研究所的团队建立统计共振模型一定程度上揭示了液体产生高次谐波的机制,成功解释了上述实验中测量到的现象,这为激光驱动的高次谐波光源和液体中的电子动力学探测提供了更好的基础^[64-64]。

3.3 液体薄膜靶产生高能离子

除了各种辐射以外,超强激光与薄膜靶材相互作用还能加速出高能离子,常见的加速机制有靶背法向鞘层场 加速(Target Normal Sheath Acceleration,简记为 TNSA)和辐射压加速(Radiation Pressure Acceleration,简记为 RPA) 等。近年来通过 RPA-TNSA 级联混合加速机制大幅提高了超短脉冲激光加速重离子、超重离子的能量^[65-66]。包含 激光加速系统、离子收集与选能系统的激光加速器,已经实现了在应用端处 1% 能散的质子束流^[67]。TNSA 机制要 求靶材不能太厚,否则热电子无法穿透靶体形成鞘层电场,靶材厚度通常在微米量级。RPA 机制有着更高的激光 能量转换效率,同时存在稳相机制^[68],有可能产生品质更好的离子束流,但也要求靶材的厚度在 100 nm 以下。

使用固体薄膜作为靶材加速离子,最高工作频率也就在 Hz 量级^[69]。使用液体薄膜作为靶材可使平均流强上 升 2~3 个数量级^[70]。同时,用液体薄膜靶替代固体靶,可以有效减少重频打靶时的碎片积累和降低制靶成本,并 更好地对靶材厚度等参数进行调节,提高激光离子加速的可控性和稳定性。2018年, Morrison等人^[25]利用两股直 径 30 μm、流速 24 m/s 的乙二醇液柱错位相碰的方案形成液膜,最薄处为 450 nm。他们使用单发能量为 5 mJ 的飞 秒激光在 1 kHz 的高频打靶条件下产生了最高能量为 2.3 MeV 的质子。2022年, Puyuelo-Valdes 等人在高真空度实 验条件下(10⁻² Pa)实现了对撞水膜靶的激光离子加速,其水膜厚度在1μm 左右,质子截止能量达到 3.5 MeV^[71]。

液体靶用于激光离子加速时, 靶周围的真空度、靶厚度、位置稳定性等因素都会直接影响离子的能量。未来 需要进一步探索利用液体靶产生数十甚至上百兆电子伏特高能质子的方案。制备厚度小于 100 nm 的超薄液膜靶 以进入 RPA 加速区域是一个重要的方向。

3.4 液体薄膜靶作为等离子镜光学元件

液体薄膜不仅可以作为粒子与辐射源,还可用做等离子镜。传统的等离子镜是镀了增透膜的电介质镜,是一种用于改善激光时域对比度的消耗性光学元件,高重频打靶时其代价是十分昂贵的。使用液体薄膜靶作为等离子镜能够极大降低成本。液体薄膜靶自身对激光的吸收较弱,厚度合适时就能形成增透膜的效果以透过预脉冲,而当光强很强的主脉冲到达时,液体被电离为等离子体,将主脉冲反射,起到分离主、预脉冲,改善激光时间对比度的作用^[72]。已有一些工作实现了Hz级的准液体等离子镜^[73-75],2019年,George等人首次报道了液体薄膜在kHz重频下的激光时域对比度改善^[37],同时发现:液膜越薄,则能够支持越大的激光谱宽,即更短的脉冲;而为了使预脉冲强度不超过材料损伤阈值,在等离子镜表面的光斑通常较大,这对液膜的大小和均匀性也提出了更高的要求。因而面向等离子体镜应用的更大更均匀的高质量液膜靶等离子镜,也是液体薄膜靶的重要发展方向。

4 总 论

液体薄膜靶的制备技术经过了几十年的发展,已经比较成熟,其在激光驱动辐射源和激光离子加速中的应用 方兴未艾,中子源也可能在不久的将来实现^[76]。无窗无碎片的液体薄膜靶因其快速更新的重频性、灵活的可控性 及材料可循环的经济性和连续工作能力,有望代替固体薄膜成为应用的主流靶材及消耗性光学元件。超薄、超 大、均匀的液体薄膜靶的制备是未来需要攻克的难关,建立在线的膜厚、稳定性诊断系统对于应用也至关重要。 随着这些技术的进步与完善,液体薄膜可在激光驱动的辐射源和离子加速应用中发挥更加重要的作用。

目前,北京大学激光加速器团队在国家重点研发专项"拍瓦激光加速器及应用示范"的支持下,正努力攻关研制面向肿瘤治疗的激光驱动质子放疗装置。液体薄膜靶是实现激光加速器长时间连续稳定运行的关键靶材之一,将发挥重要作用。

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