文章编号:1001-5078(2024)08-1179-09　　·综述与评论·

纳秒可见光全固态拉曼激光器的研究进展

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摘　要:纳秒脉冲可见波段激光在遥感、相干雷达系统、精密加工、激光清洗、液态染料激光器泵浦等领域广泛应用,全固态激光器以其结构紧凑、占用空间小、成本低、寿命长等特点备受青睐。近年来,纳秒可见光全固态激光器发展迅速,本文简述了传统激光二极管泵浦掺 Nd^{3+}、Yb^{3+} 等稀土离子晶体输出近红外光的二阶非线性频率变换产生纳秒脉冲可见光固体激光器的技术水平。从非线性频率变换角度介绍了几种出光波段丰富的纳秒脉冲可见光全固态拉曼激光器的技术特点,重点介绍了拉曼频移、拉曼混频和金刚石拉曼,对其性能特点和技术瓶颈进行了综述,并在最后做了总结和展望。

关键词:拉曼激光;拉曼频移;拉曼混频;金刚石拉曼
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Research progress of nanosecond visible all-solid-state Raman lasers

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Abstract:Nanosecond pulsed visible laser is widely used in remote sensing,coherent radar system,precision machining,laser cleaning,and liquid dye laser pumping and so on. All-solid-state laser is favored for its compact structure,small volume,low-cost,long life and so on. In recent years,nanosecond visible all-solid-state lasers have been developing rapidly. In this paper,the state of the art of nanosecond pulsed visible solid-state lasers generated by second-order nonlinear frequency conversion of near-infrared light output from Nd^{3+},Yb^{3+} and other rare-earth ion-doped crystals pumped by conventional laser diodes is briefly described. The technical characteristics of several nanosecond pulse visible all-solid-state Raman lasers with abundant outgoing wavelength bands are introduced from the perspective of nonlinear frequency conversion,focusing on Raman frequency shifting,Raman frequency mixing,and diamond Raman,with an overview of their performance characteristics and technical bottlenecks,and concluding with a summary and outlook.

Keywords:Raman laser;Raman frequency shift;Raman frequency mixing;diamond Raman

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1 引 言

可见光波段大致为400～780 nm,该波段激光可直接被人眼看到,不仅在日常生活中不可或缺,其在高新科技如显微镜、高端材料制备、激光信标、激光通信等领域也广泛应用。这其中以纳秒脉冲形式运转的可见光激光器兼具高重复频率、窄脉宽、窄线宽、高单脉冲能量的特点备受关注,其在遥感、相干雷达系统、精密加工、激光清洗、液态染料激光器泵浦等领域具有不可替代的独特优势。由于组成全固态激光器各要素均为固体形态,结构紧凑、占用空间小、成本低、寿命长,因此纳秒脉冲可见光全固态激光器受到科技工作者的广泛关注。目前产生纳秒脉冲可见光的全固态激光器传统工作方式为采用激光二极管泵浦掺Nd^{3+}、Yb^{3+}等稀土离子晶体输出近红外光的二阶非线性频率变换(倍频、和频等)。在单脉冲能量方面,2016年,美国劳伦斯利弗莫尔国家实验室(LLNL)的HAPLS项目采用二极管泵浦板条Nd:glass晶体并经过LBO晶体倍频,获得单脉冲能量44 J,脉宽22 ns的527 nm绿光输出[1]。2017年,美国尖端光电公司的XU团队[2]采用二极管泵浦Nd:YLF的主震荡功率放大后倍频,获得单脉冲能量4 J,脉宽13 ns的527 nm绿光输出。高平均功率方面,2015年,Trumpf公司型号为TruMicro 7240的碟片激光器产生300 W@515 nm,脉宽小于30 ns@5 kHz～100 kHz的激光出射,实验室验证功率达到800 W[3]。倍频效率方面,2023年,中科院固体激光实验室Liu团队[4]采用LBO晶体将经双通放大的1064 nm基频光倍频,实现67.4 W的532 nm绿光输出,倍频效率75.4 %,这是迄今为止532 nm激光的最高倍频效率。和频方面,2023年,Yu[5]等报道了通过将1064 nm和1319 nm基频光和频,腔外通过LBO晶体倍频产生重复频率500 Hz,单脉冲能量10.4 mJ,脉宽62 ns,波长589 nm的单频黄光。虽然传统方式产生纳秒可见光的固体激光器综合性能优良,但是其出光波段单一,无法满足差异化的应用需求。为此,本文从近年来发展较迅速的拉曼技术路线综述了出光波段较丰富的纳秒脉冲可见光全固态拉曼激光器的研究进展。

2 纳秒脉冲可见光全固态拉曼激光器

全固态激光器产生可见光的方式除了前文提到的传统方式外,采用三阶非线性频率变换方式的受激拉曼散射也可产生可见激光。本文主要介绍晶体拉曼里面的拉曼频移、拉曼混频和金刚石拉曼。

1928年,印度物理学家拉曼发现拉曼散射效应,时至今日,人们已经在掺杂稀土元素光纤(SiO_4、GeO、As_2S_3)、晶体(WO_4、VO_4、NO_3)、金刚石(C)等介质中实现受激拉曼散射。由于大部可见光波段通过成熟激光晶体下转换倍频方法无法直接获取,人们考虑利用级联拉曼转换的方式获得,结合其他混频过程,有能力突破传统固体激光器在对应波长的功率限制,为进一步拓展多波长激光应用创造途径。表1列出了几种典型拉曼晶体特性,不难看出,拉曼晶体不仅具有拉曼频移量大、热导率高等显著优势,而且拉曼晶体中高度对称排列的原子和分子使得拉曼晶体抑制谱线加宽能力、抗干扰能力强。受激拉曼散射增益具有无空间烧孔特性,通过简单的腔型设计,即可实现稳定单纵模运转[6]。其中,可见光光纤拉曼激光器单脉冲能量过低(在纳焦水平)[7],本文未做展开,主要介绍晶体拉曼和金刚石拉曼。

表1 拉曼增益介质参数比对

Tab.1 The parameter comparison of different Raman medium

parameters	Optical fiber(SiO_4, GeO, As_2S_3)	Crystal(WO_4, VO_4, NO_3)	Diamond
Thermal conductivity/($W \cdot m^{-1} \cdot K^{-1}$)	1	5～10	2000
Transmission spectrum range/μm	<2.50	0.30～5	>0.23
Raman gain coefficient/($cm \cdot GW^{-1}$)	0.01～0.10	1～10	10
Raman shift/cm^{-1}	400～600	400～1000	1332.30

2.1 晶体拉曼

拉曼晶体主要有钨酸盐、钒酸盐、硝酸盐等,目前主要有两种方法实现可见光出射。第一种方法是用短波长激光泵浦后进行拉曼频移,直接产生;第二

种方法是应用红外 LD 泵浦，先将近红外基频光进行拉曼频移，然后进行混频。

2.1.1 拉曼频移

拉曼频移类似于离子能级下转换，应用短波激光泵浦，能通过级联方式输出各阶斯托克斯光和反斯托克斯光，但由于各阶激光波长间隔十几纳米（晶体拉曼频移较小），受限于腔镜镀膜工艺，导致多阶拉曼激光同时振荡输出，较难实现单一波长高功率输出。为实现单一波长高效率出射，除提升输出镜镀膜工艺外，也可在激光出口处插入分束棱镜，提取目标波长，但不能提升特定波长的转换效率[8]，如见图1所示。1997年，美国汉普顿大学的 Chuan[9]团队采用重复频率为30 Hz，脉宽为8 ns的532 nm激光泵浦 $Ba(NO_3)_2$ 拉曼晶体，实现 1^{st} stokes 563 nm 处 25 mJ 能量输出，2^{nd} Stokes 599 nm 处 32 mJ 能量输出，光光转换效率分别为40 %和51.2 %。2004年澳大利亚麦考瑞大学R. P. Mildren[10]等用532 nm激光泵浦 $KGd(WO_4)_2$ 拉曼晶体，泵浦光重频5 kHz，脉宽10 ns，功率1 W，得到最高输出功率396 mW，光光转换效率39.6 %，出射激光参数见表2。两年后的2006年[11]，该团队通过优化拉曼谐振腔结构，将泵浦光功率提升到2.4 W，得到了1.5 W@588 nm的拉曼激光输出，光光转换效率提升到64 %，斜率效率达到78 %。2007年，白俄罗斯国家科学院的 Vodchits[8]团队应用脉宽为105 ns，重频为1 kHz，功率为3.4 W的准连续532 nm激光泵浦 $Ba(NO_3)_2$ 拉曼晶体，得到共六阶级联拉曼激光出射，最大光光转换效率35.3 %@598.7 nm，详细数据见表3。2012年，西北大学的 Bai[12]团队应用重频1 kHz，脉宽10.1 ns，功率5.02 W的绿光泵浦 KGW 晶体，在与拉曼晶体偏振匹配的工况下，分别获得了两种波长激光出射，2.58 W @579.54 nm，光光转换效率51.4 %，3.18 W @588.33 nm，光光转换效率63.3 %。

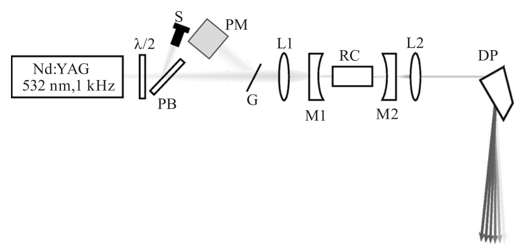


图1 532 nm激光泵浦 $Ba(NO_3)_2$ 拉曼激光器[8]

Fig. 1 $Ba(NO_3)_2$ Raman laser pumped by 532 nm[8]

表2 532 nm激光泵浦 $KGd(WO_4)_2$ 拉曼晶体

Tab. 2 $KGd(WO_4)_2$ Raman laser pumped by 532 nm

parameters	1^{st} Stokes		2^{nd} Stokes		3^{rd} Stokes		4^{th} Stokes	
Wavelength/nm	555	559	579	589	606	622	636	658
Power/mW	314	386	245	396	155	215	89	192

表3 532 nm激光泵浦 $Ba(NO_3)_2$ 拉曼晶体

Tab. 3 $Ba(NO_3)_2$ Raman laser pumped by 532 nm

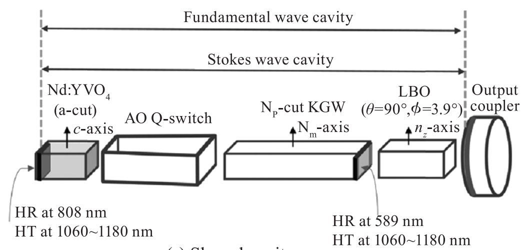
parameters	1^{st} Stokes	2^{nd} Stokes	3^{rd} Stokes	4^{th} Stokes	5^{th} Stokes	6^{th} Stokes
wavelength/nm	563.4	598.7	638.7	684.5	737.3	799.0
power/W	0.69	1.19	0.60	0.22	0.40	0.19

此方法产生可见光无需混频过程，其光光转换效率较高，接近量子效率。受限于量子缺陷，高功率泵浦拉曼晶体存在热效应，泵浦功率较低，导致出射功率在五瓦以下，少见更高功率的相关报道。

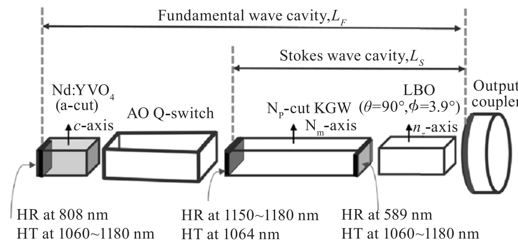
2.1.2 拉曼混频

近红外拉曼激光混频多采用成熟的红外 LD 泵浦，是获得可见光波段的另一种重要手段。1998年，H. M. Park[13]等首次报道了 $LiIO_3$/LBO 腔内倍频的580 nm纳秒拉曼黄光激光器，输出功率0.55 W。第二年，该团队将黄光功率提高到了1.2 W[14]。2001年，Kaminskii[15]等发现 YVO_4 和 $GdVO_4$ 晶体优良的拉曼特性，并提出应用Nd：YVO_4 和Nd：$GdVO_4$作为自拉曼晶体产生可见光和近红外波段激光，实现了自拉曼激光出射。2021年，台湾国立阳明交通大学的 Hsiao 团队[16]通过808 nm LD 泵浦Nd：YVO_4晶体主动声光调 Q，在重频400 kHz

下，实现最高15.1 W的589 nm黄光输出，如见图2所示，这是迄今为止基于拉曼混频单波长调Q激光出射的最高功率。2021年，温州大学段延敏[17]团队从改善自拉曼晶体热效应，综合考虑基频激光性能和提高拉曼变频性能设计了 YVO_4/Nd：YVO_4/YVO_4 三段式键合晶体提升拉曼转换效率和输出功率得到1.63 W的657 nm激光输出，脉宽11.5 ns，并通过实验验证了拉曼过程光束净化效应和脉宽压窄特性[18-19]。次年，该团队[20]通过808 nm LD泵浦双端键合Nd：YVO_4晶体并混频，实现LBO晶体温度调控波长的离散波长可见光输出，如图3所示。表4按时间顺序罗列了近30年纳秒脉冲可见光拉曼激光器的科研成果。



(a) Shared cavity



(b) Separate cavity

图2　基于Nd：YVO_4的声光调Q腔内拉曼混频589 nm黄光激光器[16]

Fig. 2 Experimental configurations of the cavities for generating the yellow laser at 589 nm in an AO Q-switched Nd：YVO_4 laser with intracavity SRS and SHG.[16]

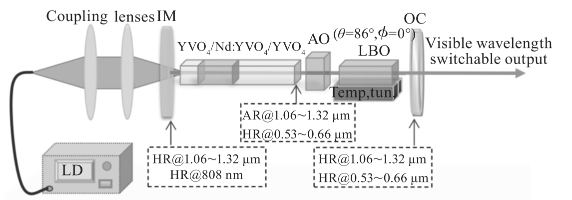


图3　CPM LBO晶体级联Nd：YVO_4自拉曼混频激光器实验装置[20]

Fig. 3 Experimental arrangement for selective frequency mixing in a cascaded Nd：YVO_4 self-Raman laser with a CPM LBO crystal[20]

表4　近年来拉曼混频研究成果

Tab. 4 The achievements of Raman frequency mixing in recent years

year	Wavelength /nm	Output power /W	Single pulse energy/μJ	LD-Visible conversion efficiency/%	Repetition frequency /kHz	Pulse width /ns
1998[13]	580	0.55	140.0	18.00	4.00	30.0
1999[14]	578	1.20	120.0	33.00	10.00	30.0
2007[21]	590	3.14	314.0	3.20	10.00	15.0
2009[22]	588	5.70	100.0	24.20	60.00	16.0
2009[23]	588	7.93	70.0	30.00	110.00	18.0
2009[24]	588	5.70	95.0	24.20	60.00	16.0
2010[25]	590	8.30	550.0	6.57	15.00	20.0
2013[26]	559	3.55	118.0	11.00	30.00	25.0
2016[27]	589	4.20	126.0	3.30	300.00Hz*/33.30	13.6
2018[28]	559	0.81	13.5	8.00	60.00	19.0
	588	1.73	28.8	17.00	60.00	20.0
2019[29]	540	0.42	20.9	6.96	20.00	47.2
	567	0.27	13.3	4.42	20.00	4.8
	597	0.36	18.1	6.01	20.00	5.6
2019[30]	567	0.31	15.3	5.01	20.00	4.7
	597	0.51	25.4	8.45	20.00	4.1
2020[31]	559	2.03	34.0	10.40	60.00	12.2
	588	3.43	57.0	17.60	60.00	12.7
2020[32]	597	0.59	39.0	9.80	15.00	3.0
2020[33]	* *	0.09	8870.0	7.40	0.01	7.7
2020[34]	588	13.70	1370.0	5.88	10.00	12.5
2020[35]	588	7.60	69.1	18.10	110.00	11.0
	588	4.10	410.0	9.76	10.00	8.0

续表4 近年来拉曼混频研究成果

Tab.4 The achievements of Raman frequency mixing in recent years

year	Wavelength /nm	Output power /W	Single pulse energy/μJ	LD-Visible conversion efficiency/%	Repetition frequency /kHz	Pulse width /ns
2020[36]	540	0.80	59.9	6.66	13.40	37.6
	567	0.43	39.5	3.58	10.90	2.4
	597	0.47	40.7	3.92	11.60	2.6
2020[37]	588	8.80	44.0	34.00	200.00	8.0
2020[38]	540	0.80	80.0	13.40	10.00	33.9
	567	0.34	34.0	5.70	10.00	4.2
	597	0.46	46.0	7.70	10.00	4.7
	631	0.19	19.0	3.20	10.00	4.4
	668	0.33	32.6	5.50	10.00	4.6
2021[39]	578	0.18	28.2	1.92	6.50	6.5
2021[40]	588	0.66	41.3	3.80	16.00	2.8
2021[16]	589	15.10	37.8	37.75	400.00	10.2
2021[17]	657	1.63	27.0	11.50	60.00	11.5
2022[41]	589	0.78	19.0	4.88	41.00	3.6
2022[20]	588	2.20	36.7	14.19	60.00	20.0
	620	1.31	21.8	8.45	60.00	10.9
	657	1.58	26.3	10.19	60.00	13.8
2023[42]	588	2.85	57.0	5.75	50.00	3.0
2023[43]	589	1.10	110.0	11.40	10.00	0.9
2023[44]	579	1.00	100.0	2.50	10.00	1.3
	589	0.72	72.0	1.80	10.00	1.5

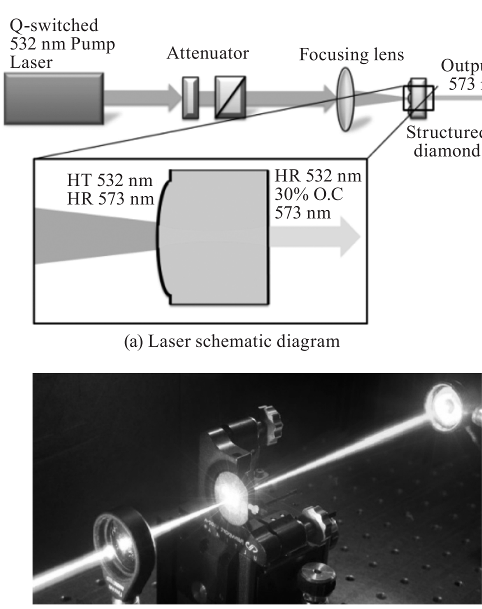
注:*脉冲包络频率 **multiwavelengths @588 nm,584 nm,594 nm,580 nm,589 nm

以上激光器分别尝试了内腔式、自拉曼、外腔式等腔型,在优化拉曼晶体尺寸参数、改善晶体热效应、提升晶体拉曼性能等方面做了大量的工作,将输出功率提升过十瓦量级,并且在保持着强劲的上升势头。

2.2 金刚石拉曼

为了克服热效应的影响,人们尝试将传统的激光晶体从棒状塑造成板条、碟片、光纤等形状,以减轻热效应。而金刚石以其优于常规YAG晶体140倍的超高热导率[45],天然的具备散热优势,并且其拥有低的热膨胀系数、高度的化学惰性及优异的光学性能。伴随着化学气相沉积(CVD)制备工艺的提高,人造金刚石的光学品质得到快速提升,光学级的金刚石晶体也因此表现出优异的拉曼性能,极大地克服了传统粒子数反转激光器的热效应、以及波长和输出功率难以兼顾的难题。目前,人们已经利用金刚石实现了波段从深紫外到可见光的拉曼转换[46-48],准连续模式功率已达千瓦量级[49],纳秒脉冲功率也已达数十瓦水平。2004年,俄罗斯科学院的Kaminskii[50]等人在CVD金刚石中观察到了受激拉曼散射现象,随后各国科技工作者相继开展金刚石拉曼激光器研究。2007年,Kaminskii[51]团队应用532 nm激光泵浦,第一次发现单晶金刚石的可见光拉曼出射。2015年,英国斯凯莱德大学的Kemp课题组[52]通过脉宽1.5 ns、重频10 kHz、波长532 nm的高重频激光分别泵浦微透镜和单片形式金刚石,如图4所示,产生总功率为134 mW和96 mW的三阶斯托克斯激光573 nm、620 nm、676 nm,光光转换效率84 %和59 %。金刚石晶体从传统的块状拓展至片状形式,获得了转换效率的提升。两年后的2017年[53],课题组通过优化谐振腔结构,将微透镜和单片两种腔

型激光器的输出功率提升 7 倍和 300 倍,单片式最大功率达 31 W,这是迄今调研的最高功率记录。2018 年,俄罗斯普罗霍罗夫物理研究所的 Pashinin 团队[54]应用脉冲 Yb∶YAG 激光器泵浦金刚石并对一阶斯托克斯激光倍频,产生重频1500 Hz、脉宽 10 ns、功率 0.22 W 的 597 nm 激光,实现黄光波段出射,受限于泵浦激光功率,输出黄光仍存在很大提升空间。2021 年,中科院理化技术研究所的 Tu[55]团队采用重频为 10 kHz、脉宽 11 ns、功率 22 W 的 532 nm 激光泵浦金刚石,得到 572.5 nm 拉曼频移光,输出功率 9.2 W,斜效率 60.8 %,如图 5 所示。通过替换输出镜镀膜波段[56],实现 1.95 W 的 620 nm二阶斯托克斯激光出射,斜率效率为 22.8 %。2022 年,空间激光通讯与检测技术重点实验室的 Chen[57]等应用 Nd∶YAG 出射1064 nm 激光泵浦金刚石的腔内倍频技术实现 2 kHz、750 mW、5 ns 的 620 nm 红光出射,光光转换效率为 11 %,提供了一种实现脉冲红光的新方法。2023 年,河北工业大学的 Ding 团队[58]研制了一台脉冲 532 nm 绿光泵浦的五阶斯托克斯级联金刚石拉曼激光器,如图 5 所示,通过将一阶 Stokes 黄橙光(573 nm)锁定在振荡器中,实现了 620 nm、676 nm 、743 nm 和 824 nm 的级联拉曼激光输出,总峰值功率达 70.7 kW,总转换效率为 31.5 %,实现了可控多波长输出。

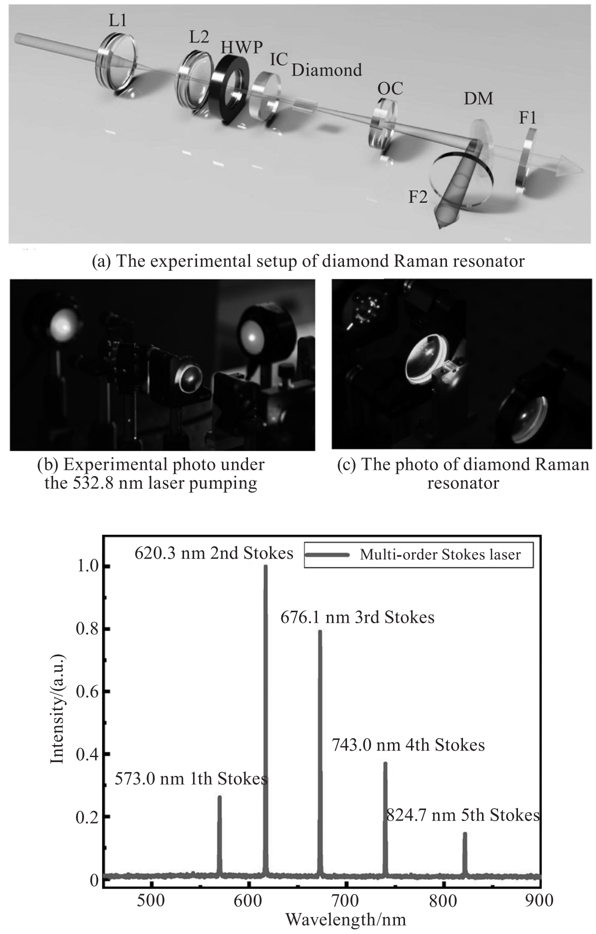


(a) Laser schematic diagram

(b) Laser experimental device diagram

图 4　单片式金刚石拉曼激光器[52]

Fig. 4 Monolithic diamond Raman laser[52]



(a) The experimental setup of diamond Raman resonator

(b) Experimental photo under the 532.8 nm laser pumping

(c) The photo of diamond Raman resonator

图 5　金刚石拉曼激光器原理图与实验结果[55]

Fig. 5 Schematic diagram and experimental results of diamond Raman laser[55]

3　结　论

本文介绍了拉曼频移激光器、拉曼混频激光器、金刚石拉曼激光器在纳秒可见光领域的研究进展,如图 6 所示。相比于传统的激光二极管泵浦掺 Nd^{3+}、Yb^{3+} 等稀土离子晶体输出近红外光的二阶非线性频率变换方式,这三种方式出光波段丰富,但是出光性能存在差距。拉曼激光器出光波段理论上可覆盖整个可见光波段,这依赖于丰富的拉曼介质和泵浦光波长。输出功率方面,受限于量子缺陷,晶体拉曼频移激光器输出功率接近五瓦量级,应用较少,多见于金刚石拉曼。晶体拉曼混频激光器输出功率过十瓦量级,且腔型丰富,可离散调谐,比较受科研工作者的追捧。金刚石拉曼激光器凭借出色的热传导性能,适用于拉曼频移、拉曼混频等多种形式。通过优化金刚石介质尺寸,输出功率已达数十瓦量级,在提升泵浦光功率的前提下,输出功率仍存在很大的提升空间,有望成为高功率纳秒可见光拉曼激光器的佼佼者。

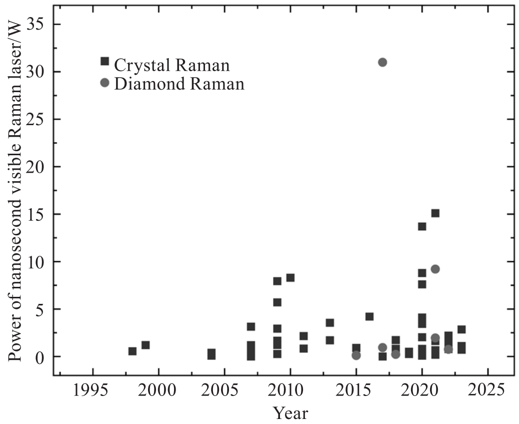


图6 纳秒可见光全固态拉曼激光器的输出功率历程图

Fig. 6 Output power evolution of nanosecond visible all-solid-state Raman lasers

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