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光频梳在精密测量中的应用*

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摘 要:光学频率梳在过去17年间极大地促进了精密科学领域的发展。光学频率梳首先建立了微波频率与光学频率的直接联系,为时频领域的相关研究带来了突破性的进展;在此基础上,利用光学频率梳稳定的频域梳齿特性极大地推动了激光精密计量与测试技术的进步,同时光学频率梳良好的时域相干性也为实现高速、宽带的高精密分子光谱探测提供了前所未有的手段。简述了光学频率梳的产生于发展,综述了光学频率梳在激光频率测定、绝对距离测量和精密吸收光谱探测、高速非线性光谱与光谱成像及高精度时间频率传递应用中的进展。这也说明了光学频率梳作为一种具有优秀时频特性的激光器必将继续推动精密科学领域的发展。

关键词: 光频梳;精密计量;精密吸收光谱;超快光谱
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Precision measurement and spectroscopic applications of femtosecond optical frequency combs

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Abstract: Over the past seventeen years, notable advances have occurred in a diverse range of scientific areas following the development of femtosecond optical frequency combs. Frequency comb first establishes a direct link between microwave and optical frequencies, thus enabling the breakthrough of time- and frequency-domain research. Frequency stabilization of comb has led to a revolution in frequency metrology and precision measurement. In addition, well-defined temporal coherence across the optical spectrum makes it a unique tool for molecular spectroscopic applications, simultaneously providing high speed measurement, high spectral resolution and broad spectral coverage. This tutorial review provides an introduction to femtosecond optical frequency combs, covering their principles of operation and applications and advance in frequency metrology, absolute distance measurement, precision absorption spectroscopy, high speed nonlinear spectroscopy and microscopy and high precision time and frequency transfer. In this way it aims to demonstrate their potential as a spectroscopic tool that could play a very significant role in future advances in the precision sciences.

Keywords: optical frequency combs; precision measurement; precision absorption spectroscopy; ultrafast spectroscopy

0 引 言

在光频梳出现之前,谐波频率链可实现标准不确定度优于1 kHz的绝对光学频率测量[1]。但复杂的结构和高昂的成本限制了此系统的广泛应用。如何能简单有效地精确测量光学频率成为激光精密测量领域,乃至国际计量界的难题。早在1978年,Eckstein J. N. 等人[2]曾提出利用锁模激光的宽带相干特性测量激光频率的设想。这一想法在20年后才最终得以实现[3-5]:锁模激光器可以实现其纵模频率间隔等于脉冲的重复频率,且相干带宽内一致性可以达到10^{-16}量级[6]。至此,在频率域内,一把相对于频率零点浮动的“尺子”便呈现出来。然而,“尺子”的浮动依然无法实现简单、直接的绝对光学频率

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测量。2000 年,Jones D. J. 等人[7]利用 f-2f 自参考锁定方法将这把浮动的尺子完全固定下来[7],至此,光频梳实现了射频频率与光学频率之间的相干传递。Hänsch T. W.[8]和 Hall J. L.[9]也因在光频梳方面的卓越贡献获得了 2005 年诺贝尔物理学奖。

虽然光频梳概念的提出最初仅为了精确测定光学频率,但其频率上线性数学关系与相干传递为现阶段的精密激光测量技术带来了全方位的提升[10-11]。

在国内,本人领导的干涉与光谱课题组较早地开展光频梳在精密测量中的应用研究并取得了一些研究成果。本文结合近 10 年的研究积累,综述该研究领域的现状及进展。

1 绝对光学频率精确标定

光频梳是由上万根频谱域的梳齿所组成的,每一根梳齿的频率表达式为:

$$\nu = n \cdot f_{rep} + f_{ceo} \tag{1}$$

式中:n 为纵模序数,f_{rep} 是光频梳纵模的频率间隔,即重复频率。f_{ceo} 是光频梳纵模相对于频率零点的偏差,即偏置频率。f_{rep} 和 f_{ceo} 均为射频频率。因此,光频梳的出现首先解决了直接精确获得光学频率的难题。光频梳的纵模充当了“频率测尺”的刻度,待测激光与光频梳的拍频则可精确给出相对于“频率刻度”的偏差,如图 1 所示。可以看出,利用光频梳测量光学频率分为两个部分。首先,测量激光频率的整数部分,即待测激光频率处于光频梳频谱的第 n 个纵模附近。目前一般商用激光波长计均可达到 10^{-8} 量级,满足确定唯一整数 n 的粗测精度。其次,测量激光频率的小数部分,即所述的“频率刻度”偏差。若待测激光与光频梳位于同一光学波段,则光学拍频可直接被光电探测器获得,通过频率计可读出拍频率[12]。利用式(2)可以计算出待测激光的绝对频率。

$$\nu = n \cdot f_{rep} + f_{ceo} + f_{beat} \tag{2}$$

式中:f_{beat} 光学拍频频率。通常,光频梳的输出光谱限制在几个特定的光学波段,并且光谱带宽也非常受限。当测量其他波段的激光频率时,需要采用光学频率转换,如倍频[13-14]、和频[15-16]、差频[17-18]、光频梳拓谱技术[19-22]等,将待测激光或光频梳转换至相同波段。

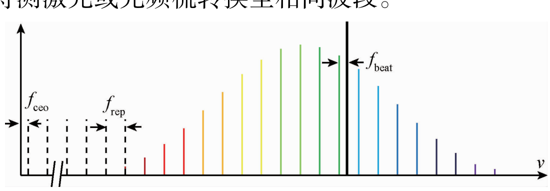


图 1 利用光频梳实现精确绝对光学频率标定

Fig. 1 Precision optical frequency measurement using frequency comb

除了采用激光波长计粗测与频率计精测相结合的方式,也可通过调节光频梳的可变频率参量,记录 n、f_{rep}、f_{ceo} 和 f_{beat} 的变化。不同的系统设置参数组成了多元线性方程组,可计算出待测激光频率[23]。

2 可溯源的几何尺度测量

考虑到频率、时间、速度、距离之间的物理关系,光频梳的“频率测尺”特性可以转化为“长度测尺”,并在最近十年得到了极大的重视和发展,涌现出了一系列新的理论和技术。按照测量对象的不同,光频梳距离测量可分为两大类:绝对距离测量和位移测量。利用光频梳的绝对距离测量解决了传统时间飞行法的低精度时间分辨[24]和合成波长法的初值辅助[25-26]问题;利用光频梳校准的位移测量则解决了测量溯源性问题[27-28]。

2.1 光频梳频标合成波长法绝对距离测量

光频梳作为可溯源的频率标准可为传统合成波长法中的可调谐激光器提供准确的频率参考[29-42]。如图 2 所示,实际测量中,将可调谐激光器通过拍频鉴相的方式锁定至光频梳,作为测量光源。可调谐激光器频率由式(2)确定。通过将 f_{rep}、f_{ceo} 和 f_{beat} 锁定至微波基准,实现对可调谐激光器频率的精确锁定。频率锁定后,调节 f_{rep} 产生合成波长,进行绝对距离测量。这种方法锁定的可调谐激光频率秒稳阿伦均方差达到 10^{-12} 量级[39],在此基础上本课题组的吴学健等人将距离测量标准不确定度减小到了 5 nm[41],可为 Avogadro 常数精确测定和“kg”单位的重新定义奠定基础。

使用光频梳作为频率标准的合成波长法提高了合成波长法的测量精度,并且测量结果溯源至频率基准。尽管在精度和溯源性上有所提升,但是以光频梳作为频率标准的合成波长法延续了传统合成波长法的不足:合成波长链的建立依赖初值,多波长分时测量过程中要求测量目标位置基本不变,测量时间长。因此,这种方法不适用于快速、初值未知的大尺寸绝对距离测量。

2.2 模间拍频合成波长法绝对距离测量

2000 年 Minoshima K 提出利用光频梳各纵模之间的拍频产生合成波长链可实现绝对距离测量[43-50]。在射频谱上模间拍频表现为一系列间隔为重复频率 f_{rep} 的拍频分量,即 f_{rep},$2f_{rep}$,…,nf_{rep}。由于这些拍频相位稳定,因此可使用不同的拍频相互组合形成合成波长链进行绝对距离测量。此方法在 240 m 的距离上实现了标准差 50 μm 的绝对距离测量[43]。但由于使用的拍频频率低,拍波波长长,对于确定的相位细分能力,仅依靠纵模间拍频形成的合成波长链测量精度有限。为进一步提高测量精度,使用声光移频外差干涉的方式,将合成波长的最后一级精化到光波波长,实现亚微米的距离分辨[46]。

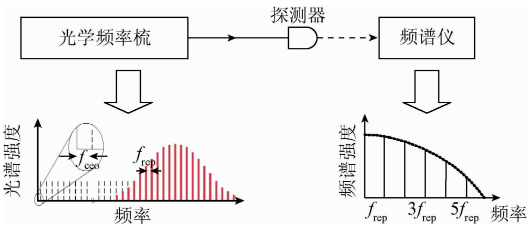


图 2　光频梳纵模间拍频合成波长产生原理

Fig. 2　Absolute distance measurement based on synthetic wavelength method using intermode beats

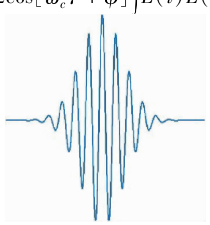
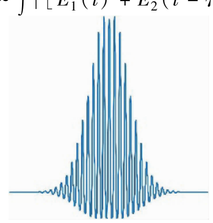
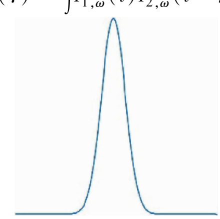
模间拍频合成波长法利用光频梳分立频谱的特性,直接产生合成波长链。此外,所使用的不同波长的拍波对目标进行并行测量,相比于可调谐激光器的分时测量缩短了测量时间。然而这种方法仍然不能克服合成波长法对初值的依赖性,不适用于待测距离未知的绝对距离测量。

2.3　脉冲对准时间飞行法绝对距离测量

在飞秒脉冲测量中,当飞秒脉冲之间的相对延迟 $\tau=0$ 时,则两脉冲的相关信号便出现极大。如果利用光频梳脉冲的相干特性,使干涉仪中参考臂与测量臂之间的飞秒脉冲进行互相关测量,根据相关信号的峰值位置判断光脉冲的时域位置,便可计算出空间距离信息[51]。表 1 所示为 3 种典型的飞秒脉冲相关信号。其中,一阶自相关属于脉冲之间的干涉现象,低功率情况下可由光电探测器直接获得;二阶自相关则需要利用非线性光学晶体来获得,对光脉冲的功率有一定要求。

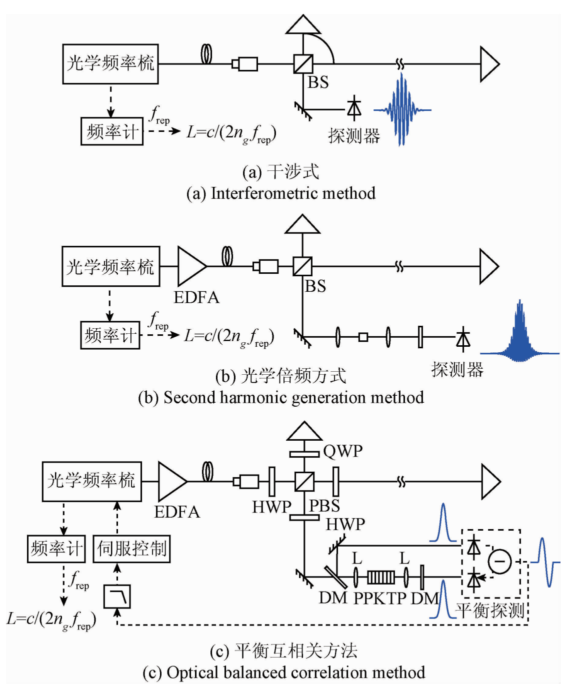
表 1　典型的飞秒脉冲相关信号

Table 1　Autocorrelation of femtosecond pulse

	一阶自相关	二阶电场自相关	二阶强度自相关(平衡互相关)
脉冲要求	具有相同偏振分量	相同线偏振光	正交线偏振光
数学表示	$I(\tau)\propto 2\cos[\omega_c\tau+\phi]\int E(t)E(t-\tau)\mathrm{d}t$	$I_{2\omega}(\tau)\propto\int\lvert[E_1(t)+E_2(t-\tau)]^2\rvert^2\mathrm{d}t$	$I_{2\omega}(\tau)\propto\int I_{1,\omega}(t)I_{2,\omega}(t-\tau)\mathrm{d}t$
信号波形			
距离测量文献	文献[52-60]	文献[61]	文献[62-64]

由时间飞行法原理,仅关心相关信号的峰值位置。一阶自相关信号与二阶电场自相关信号中的载波增加了额外的信号包络拟合步骤,距离测量装置分别如图 3(a)和 3(b)所示。因此,二阶强度自相关信号可简化峰值提取过程。使用平衡探测器对倍频信号进行差分探测,可提高信噪比,将峰值检测转变为过零差分信号[63-64],方便伺服单元锁定,如图 3(c)所示。

本文的研究结果证明,基于光频梳脉冲对准的时间飞行法可将绝对距离测量精度提升至亚微米。利用参考脉冲对测量脉冲进行光学采样克服了传统时间飞行法中电子器件响应速度对时间分辨能力的限制,满足对位置未知的远距离物体高精度绝对距离测量的需求。但无论基于哪种方式的脉冲对准方法,只有当参考脉冲和测量脉冲在时域上重合时才有互相关信号产生。受光频梳脉冲重复频率 f_r 的限制,参考臂与测量臂的臂长差被限制在一系列分立值 $m\cdot c/(2n_g f_{rep})$,m 为正整数,c 为真空中的光速,n_g 为群折射率。因此,从原理上看,这种测量方法不能实现连续的距离测量。



(a) 干涉式
(a) Interferometric method

(b) 光学倍频方式
(b) Second harmonic generation method

(c) 平衡互相关方法
(c) Optical balanced correlation method

图 3　脉冲对准时间飞行法绝对距离测量

Fig. 3　Absolute distance measurement based on time of Flight method

为克服这一困难,可使用参考臂机械扫描或重复频率扫描进行脉冲对准,弥补量程中的中断区域[60]。对于机械扫描,这种对准方式相当于将测量结果溯源到参考臂导轨的位移精度,而不是频率基准,降低了结果的可信度。此外,使用机械扫描的方式实现无死区的脉冲对准,受导轨运动副多自由度误差限制[65-66],也会降低机械扫描脉冲对准测量的精度。对于重复频率调节,虽然避免了机械调节带来的误差,但受光频梳重复频率调节范围的限制,只能在参考臂测量臂的臂长差较大的情况下才能实现无死区的脉冲对准。对于 100 MHz 量级重复频率的光频梳,要求臂长差至少为数十米。

2.4　光频梳色散干涉法绝对距离测量

为了解决脉冲重合对距离测量量程的限制,一些研究小组采用光频梳色散干涉法[67-73]在探测时将光谱中的各纵模按照频率在空间分离,在电荷耦合元件(charge-coupled device, CCD)不同像元上得到一系列频率不同的连续光信号,实际测量中可以直接使用光谱仪代替分光器件和 CCD[67]。通过干涉方法求解这一连续信号的相位,并利用波长和相位的关系得到最终的待测距离,如图 4 所示。这种方法利用色散的方式解除了各纵模之间的相位锁定关系,将脉冲探测转化为直流干涉信号的探测,因此不受时域脉冲重合要求的限制,可在连续范围内实现绝对距离测量。测量中,色散干涉的表达式为:

$$I(\omega) \propto \left[2 + 2\cos\left(\omega \frac{L}{c}\right)\right] \cdot |A(\omega - \omega_c)|^2 \qquad (3)$$

式中:c/L 为干涉信号周期,ω_c 为载波角频率。计算距离时,首先对测得的干涉光谱进行傅里叶变换。由于函数 $[2 + 2\cos(\omega L/c)]$ 的傅里叶变换包含 $-\omega L/c$、0、$\omega L/c$ 3 个频率分量,通过带通滤波选出 $\omega L/c$ 频率分量并对其进行傅里叶逆变换。对逆变换得到的函数通过虚部与实部的除法求出相位 $\omega L/c$,最后通过直线拟合求解斜率 L/c,得到待测距离[76]。

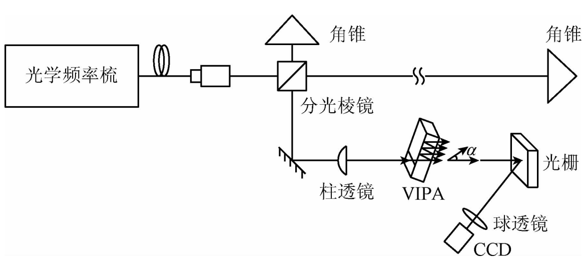


图 4　光频梳色散干涉法绝对距离测量

Fig. 4　Absolute distance measurement based on frequency comb spectral interferometry

使用色散干涉法进行绝对距离测量,将周期脉冲信号转化为不同频率的连续光干涉,有效解决了时间飞行法中脉冲对准对量程的限制。理论上,在实现对光谱单纵模分辨的情况下,光频梳色散干涉法的固有量程为 $c/2f_{rep}$,并可以通过改变重复频率的方式实现量程拓展[73]。但受色散元件分光能力限制,实际应用中很难实现对单纵模的频率分辨。通常需要使用法布里-珀罗标准具对光谱滤波才能实现单纵模的分辨。光谱滤波降低了脉冲功率,不适合大尺寸绝对距离测量。

2.5　光梳法绝对距离测量

2009 年,美国国家标准技术研究所(national institute of standards and technology, NIST)提出利用两台重复频率略有差异的光频梳作为光源进行绝对距离测量[74]。重频差引起脉冲之间的时域线性扫描,保证了对任意时刻脉冲的光学采样,从而解决了脉冲重合引起的量程受限问题,测量装置如图 5 所示。参考脉冲和测量脉冲重叠引起的测量死区可使用偏振分光方式而消除[75]。双光梳干涉式绝对距离测量结合时间飞行法和干涉相位细分技术,可将绝对距离测量标准不确定度减小至 5 nm。高精度严重依赖两台激光器之间的相干性,需要使用稳频窄线宽激光器在光学波段建立相位锁定关系,而不是通常使用的微波频率基准[76-80],这对于两台激光器的相干性提出了要求。

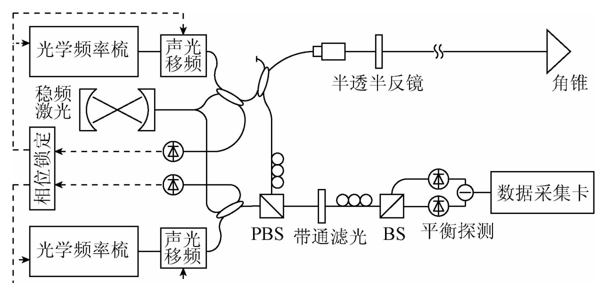


图 5　双光梳干涉式绝对距离测量

Fig. 5　Dual comb ranging via electronic-field interferometry

为了降低双光梳苛刻的相干性要求,本课题组 Zhang H. Y. 等人[81-84]和 Shi H 等人[85]采用第 2 类相位匹配倍频峰值提取技术可避免干涉相位不稳定的影响。虽然采集信号不包含干涉相位信息,但峰值稳定性依然能保证小于 100 nm 的绝对距离测量不确定度[81,84],这给依赖于激光器相干性的双光梳距离测量技术带来了新的突破。

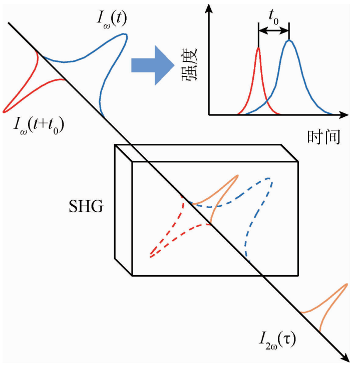


图 6　第 2 类相位匹配倍频

Fig. 6　Type II second harmonic generation of femtosecond pulses

2.6　精密位移测量

传统精密位移测量主要利用双频激光干涉仪[86],结合干涉相位细分技术,实现大尺寸、亚纳米分辨[87-88]。另外,法布里-珀罗扫描干涉仪在原理上不会引入双频激光干涉仪中的非线性误差,也被广泛应用于微纳尺度精密位移测量当中[89-90]。

直接利用国际度量衡委员会(international committee for weights and measures, CIPM)推荐的稳频激光光源[91]或将测量激光器的频率与 CIPM 推荐的稳频激光频率建立联系可获得具有计量学意义的位移测量结果[87-88]。不过,这些稳频激光仅包含几个分立的频率值,给溯源性测量带来不便。参照光频梳绝对光学频率标定,本课题组 Zhu M. H. 等人[92-93]利用光频梳的宽带相干特性作为中间频率过渡,可将精密位移测量直接溯源至频率基准。同时如果将位移变化转换为光频梳的频率变化,则可以实现亚纳米位移精密控制[94],如图 7 所示,这又扩展了双光梳在几何量测量上的应用。

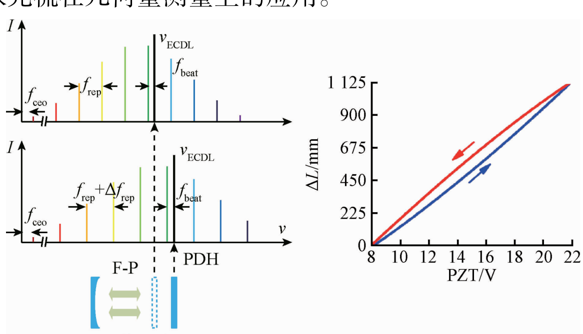


图 7　利用光频梳参考的精密位移控制

Fig. 7　Comb-calibrated precision displacement control

3　高精度空气折射率测量与补偿

目前,空气折射率测量主要包括基于抽充气[95-96]、等效合成波长[97-101]的直接光学干涉测量,以及基于 Edlén 经验公式[102]的间接测量。测量结果的计量学意义不足,且仅为单点测量。

光频梳在推动绝对距离测量应用飞速进展的同时,也促进了空气折射率的精密测量。本课题组 Yang L. J. 等人[103]采用光频梳色散干涉法的气体折射率测量可在 15 ms 内实现 1.2×10^{-8} 的标准不确定度,如图 8 所示。针对大尺寸距离测量中环境条件不唯一性,双色激光绝对距离测量方法的提出解决了这一问题[104-107]。此方法在距离测量的同时实现了传播路径上的折射率测量,可有效补偿空气折射率的影响,节省了传感器的分布式布置。不过,双色法是建立在 Edlén 公式的基础上,因此距离测量不确定度不优于 10^{-8} 量级。

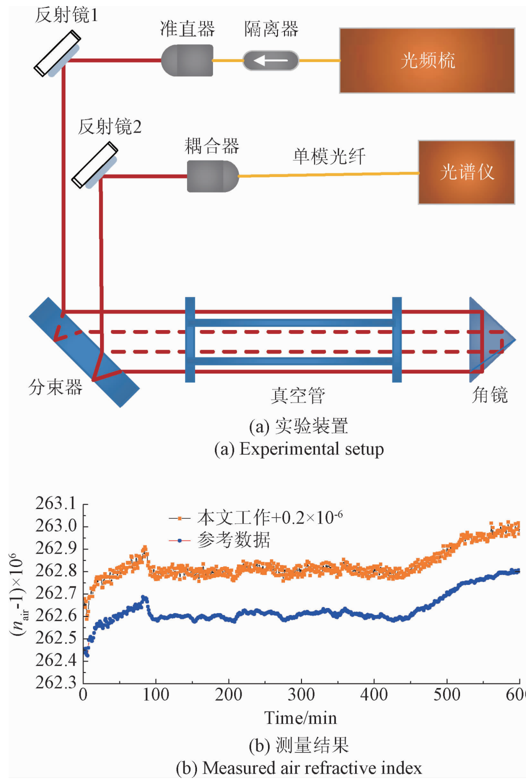


图 8　光频梳色散干涉法的气体折射率测量

Fig. 8　Air refractive index measurement via comb-based spectral interferometry

4　高分辨率气体分子吸收光谱探测

传统吸收光谱测量装置及仪器包括棱镜、光栅、傅里叶变换光谱仪和单频激光光谱。这几种光谱测量装置在光谱分辨率,探测灵敏度,频率测量精度及测量速度上均与目前高分辨率、高灵敏度及快速遥测的迫切需求相去甚远。以光频梳为主体的吸收光谱测量有望成为今后精密光谱测量的主力,本课题组成为国内首先开展双光梳吸收气体光谱测量的单位。

首先,光频梳作为一把频率“测尺”,可直接提供单频激光光谱的绝对光学频率参考[14-15,18-19,108-112]。由此,谱线频率测量精度至少提高至 kHz 量级,兼具溯源性。结合成熟的单频激光光谱测量技术,光谱探测灵敏度也非常高,适合谱线数据的精确测定。不过,利用单频激光光谱技术获得宽带吸收光谱信息,需要串行逐线探测频带内各谱线,大大增加测量时间。

其次,光频梳作为宽带相干激光光源,可与高精细腔或多次反射气体池配合使用,有利于提高探测灵敏度。如图 9 所示,探测后端与传统分光装置,如光栅[113-114]、虚像相位阵列(virtual image phase array, VIPA)[115-118]、扫描干涉仪[119-121]以及傅里叶变换光谱仪[122-127],相结合,从而使传统技术重新焕发生机。不过光谱分辨率、谱线测量精度均受到分光装置的限制。

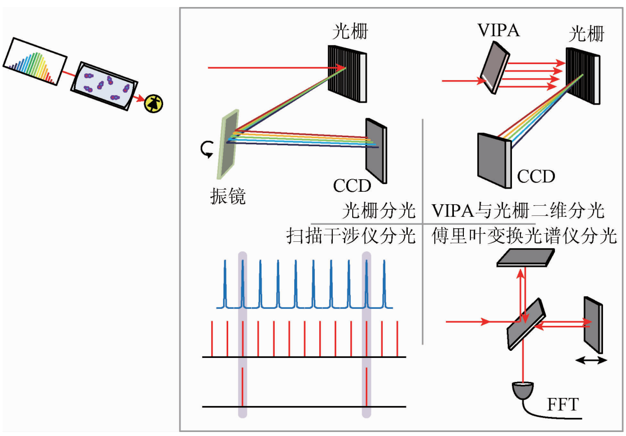


图 9 光频梳作为宽带相干光源的光谱应用

Fig. 9 Precision molecular spectroscopy using frequency comb as a broadband coherent source

光频梳的时间频率特性在精密吸收光谱中最充分的发挥莫过于双光梳光谱测量技术[128-129]。目前最优的光谱分辨率和谱线频率不确定度均为 100 kHz 量级[130-131],达到了前所未有的高度。两台重复频率不尽相同的光频梳在时域内可以产生出类似傅里叶变换红外光谱的测量效果,使激光脉冲扫描完全取代了机械扫描,干涉光谱数据刷新率达到 kHz 量级。对应地,在频谱域内将光频波段测量转换至射频波段。由于属于独立激光干涉,因此需要光频梳之间具有稳定的相位锁定关[129]。频率控制是直接有效的方法[130-143]。频率参考的选择直接影响光谱测量性能。本课题组 Yang H. L. 等人[144-145]研究发现锁定于光学频率参考的系统较直接锁定至射频基准的系统具有更高的光谱测量性能。随着激光器技术的发展,谐振腔复用双光梳光源[146-148]、电学调制光频梳[149-153]、半导体微腔光频梳[154-156]均可输出高相干性异步光学脉冲,从而利于获得分辨率,高信噪比光谱。但这些新兴双光梳系统的光谱带宽仍无法与锁模激光器相比。无独有偶,既然双光梳系统的频率稳定性问题无法避免,借鉴自适应原理,在测量中同时采集畸变的干涉光谱信息和光频梳频率变化信息,通过硬件或软件补偿畸变,同样可恢复出无畸变光谱信息[131,157-159]。此类系统中无绝对频率参考,因此光谱标定需要借助光谱数据库。

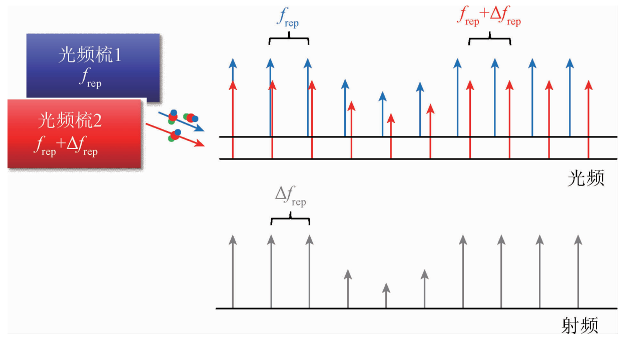
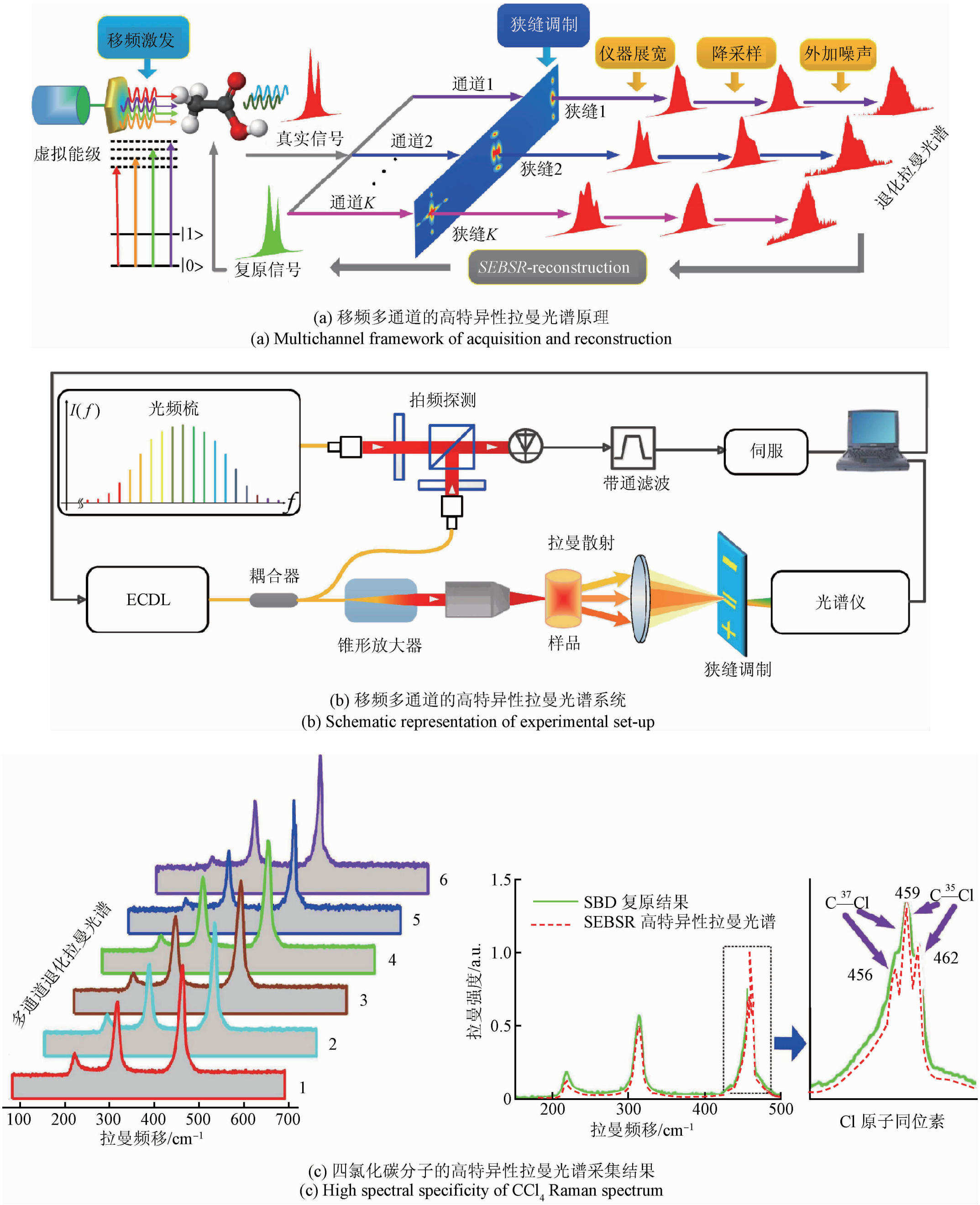


图 10 双光梳气体吸收光谱技术

Fig. 10 Dual-comb molecular spectroscopy

5 高特异性拉曼光谱

除了获得高精度的分子吸收光谱之外,光频梳在频谱域的“梳齿”特性还为获得高特异性的分子振动光谱提供了可能。本课题组陈琨等人在 2015 年提出了一种利用光频梳锁定可调谐半导体激光器进行多通道移频激发探测的高特异性拉曼光谱探测方案,如图 11 所示。所谓光谱特异性是指把光谱特征、谱带分解成为分离的成分的进而区分不同化学成分与分子结构的能力。高的光谱特异性能实现光谱特征、谱带更明显的分离与提取,具备更强的区分不同化学成分与识别分子精细结构的能力。高的光谱特异性主要包含 3 个方面的内涵:高分辨率、高信噪比和高精度。一方面,光频梳作为频谱的“标尺”可以对可调谐激光器的激发波长做校准,从而保证拉曼光谱的高精度;更为重要的是,频域内的等间隔梳齿分布特性可以在实现半导体激光器波长锁定的同时实现对波长的精确调谐,从而构建起实现对分子进行多通道移频激发的拉曼光谱探测系统。多通道移频激发的拉曼光谱探测有助于解决仪器卷积与探测器欠采样引起的谱带重叠、细节丢失和光谱畸变引起的分辨退化问题,从而实现光谱信息量的增加和拉曼信号的超分辨提取;同时还具有对外界各种噪声和扰动的具有稳健抵抗能力的拉曼光谱数据恢复与提取方法,能够获取极低信噪比条件下拉曼信号。实现一种高特异性的拉曼光谱探测[160-162]。利用这一系统可以成功实现对 CCl_4 中的氯原子的两种天然同位素(^{37}Cl 和 ^{35}Cl)精细拉曼结构的探测,如图 11(c)所示。



(a) 移频多通道的高特异性拉曼光谱原理
(a) Multichannel framework of acquisition and reconstruction

(b) 移频多通道的高特异性拉曼光谱系统
(b) Schematic representation of experimental set-up

(c) 四氯化碳分子的高特异性拉曼光谱采集结果
(c) High spectral specificity of CCl_4 Raman spectrum

图 11　基于光学频率梳的高特异性拉曼光谱技术

Fig. 11　Frequency comb based Raman spectroscopy with high spectral specificity

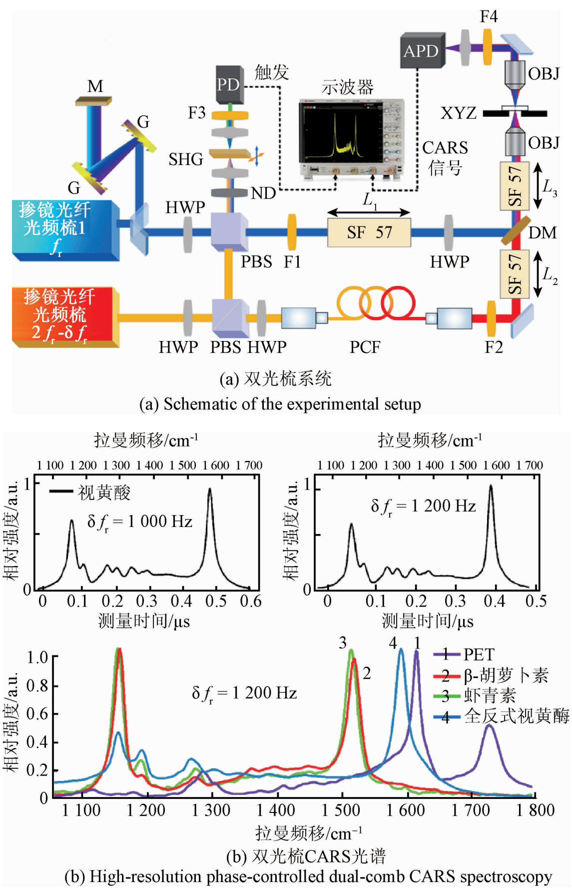
6　高速相干拉曼光谱及其显微成像

光频梳的时间频率特性除了在线性的分子吸收光谱中展现出了独特的优势之外,近几年来非线性的双光梳光谱技术开始得到发展。对于分子内部的能级分布而言,其振动能级反映了分子独特的“指纹”信息,具有高的分子特异性。利用飞秒脉冲光源可以实现对分子内部的振动能级的探测,这种光谱的产生依赖于三阶非线性效应,称之为相干拉曼光谱。光频梳的时域相干特性以及重频不一致的双光梳系统为实现高速相干拉曼光谱探测提供了全新的途径。

2014 年德国马克斯普朗克研究所的 Hansch T 研究组利用冲击脉冲探测原理,利用两台具有重频差的钛蓝宝石光频梳实现了超快的相干反斯克托斯拉曼散射(coherent anti-stokes Raman scattering, CARS)光谱采

集[163]。其单一 CARS 光谱的测量时间可以低至 15 μs;这一技术使得宽带 CARS 光谱的测量速度相对于传统技术提高了 1 000 倍,可以极大地推动无标记的 CARS 显微成像技术的应用。

在此之后,本课题组 Chen K 等人[164]发展了一种新的基于脉冲相位控制的高速宽带双光梳 CARS 光谱与显微成像技术,利用的双光梳时域高速脉冲扫描以及光谱聚焦激发的方式实现了对富含丰富化学信息的分子指纹振动区的探测,成为国内首先研究双光梳非线性光谱以及非线性显微成像的课题组。这一技术可以在获得了高的光谱分辨率的同时、将单一宽带光谱的采集时间进一步缩短到 500 ns,相对于传统 CARS 技术而言速度提高了 10^5 倍[165-167],这也是目前对于单条宽带 CARS 光谱已报道的最快测量速度,显示了基于相位控制的双光梳 CARS 在测量速度方面的巨大优势,如图 12 所示。图 13(d)~(f)所示为选择不同拉曼峰作为成像衬度的 CARS 显微图像,双光梳 CARS 技术利用单点探测器可以完成对所有光谱分量的采集,避免了时域扰动,更利于快速动态探测;该技术的另一显著优点在于能够保持稳定的光谱信噪比,这对于进一步提高采集速度具有重要意义。



(a) 双光梳系统
(a) Schematic of the experimental setup
(b) 双光梳CARS光谱
(b) High-resolution phase-controlled dual-comb CARS spectroscopy

图 12 基于脉冲相位控制的双光梳 CRAS 光谱技术
Fig. 12 Phase-controlled dual-comb CARS

与此同时,陈琨等人将双光梳 CARS 探测技术应用于显微成像中,实现了 kHz 量级的像素刷新速度;高速的宽带的 CARS 光谱特性对多目标分子探测与成像,多组分分析以及提高图像的特异性和对比度都具有直接的帮助,如图 13 所示。结合高速成像与宽带探测的优势,双光梳 CARS 技术真正实现了高速 CARS 宽带光谱三维显微成像,进一步推动了无标记的非线性显微成像的发展与应用。高重复频率的光学频率梳可以显著提高双光梳 CARS 光谱采集速度以及光谱分辨率;随着重复频率为 1 GHz 的光频梳的商业化应用,相位控制的双光梳 CARS 技术的单像素刷新率能够达到了百 kHz 量级;为真正实现复杂生物系统中的动力学过程探测,包括新陈代谢、物质转化等细胞生命活动提供了最有潜力的手段。

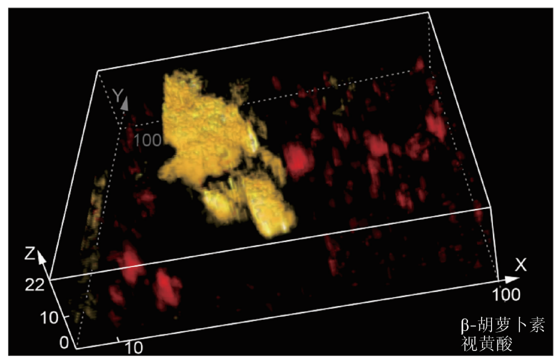


图 13 双光梳 CARS 对 RA 与 β-胡萝卜素的混合体系的三维成像探测
Fig. 13 Three-dimensional dual-comb CARS imaging of the mixture of RA and β-carotene

7 频率基准自由空间的精确传递

目前远距离时间频率传递主要采用 GPS 载波相位观测和卫星双向时间频率传递[168],同步误差小于 10^{-15}/d。考虑到有望成为未来下一代时间频率基准的光钟的稳定度和不确定度均可达到 10^{-18} 量级[169-170],因此微波波段的时间频率传递无法满足光钟的传递精度。

光纤相位噪声抑制技术[171]的提出激发了光学频率相干传递技术的发展[172-175],频率传递精度在 100 s 积分时间便优于 10^{-19}[175],极大地抑制了光纤传输链路部分的相位噪声,如图 14 所示。虽然光学频率相干传递无法实现时间同步,但却实现了远距离光钟的稳定度比对[176-177],光频梳在这其中起到了频率相干综合的作用。通过射频调制的光学载波技术可实现时间与频率的同时传递[178-179],不过,传递精度与光钟稳定性相比仍比较低。

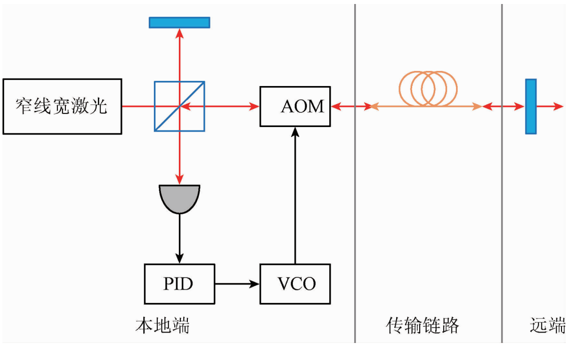


图 14　光纤相位噪声抑制简化示意图

Fig. 14　Simplified schematic of fiber noise cancellation

由于光频梳直接建立了射频频率与光学频率之间的联系,光频梳输出的飞秒脉冲便是时间和频率的“天然”传播载体。飞秒脉冲同步技术主要包括时序探测和时序延迟[180],如图 15 所示。时序探测通过探测参考光路与传输光路脉冲重复频率的高次谐波,通过鉴相方式,将时序误差反馈至光学延迟装置[181-183]。由于时序误差在射频波段检出,光电转换产生的附加噪声极大地限制传输噪声底,同步误差通常为 10^{-13} 量级。时序延迟技术采用飞秒脉冲平衡互相干技术检测传输链路中的时序误差,将时序误差的探测灵敏度提高至阿秒水平[184-188]。其探测原理与前面提到的脉冲对准绝对距离测量一致。同时,采用基于光纤 sagnac 干涉仪的光学-微波相位探测便可将脉冲时序再精确地传递至微波源[189-191]。时序探测和时序延迟均利用光纤链路进行时序传递。由于光纤的色散效应,传输链路需要预先进行色散管理,对于小范围的时序同步尚可满足,例如自由电子激光、粒子加速器和相位阵列天线等,但对于地域尺度的时间频率传递则显得异常复杂。

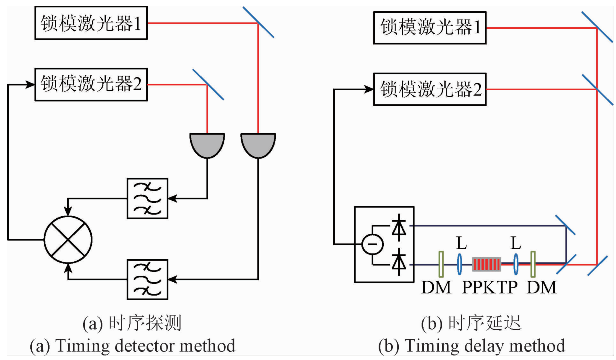


图 15　锁模激光器同步简化示意图

Fig. 15　Synchronization of femtosecond lasers

2013 年,美国 NIST 实现了基于双光梳系统的双向自由空间时间频率传递。此系统在 2 km 自由空间路径的传递误差达到 1 fs,1 000 s 平均的频率稳定性为 10^{-18} 量级[192]。目前,此研究组的已实现了 12 km 自由空间传输[193-195],如图 16 所示。非线性异步光学采样绝对距离测量系统同样可以实现自由空间的频率传递,且秒稳频率传递误差小于 1.3×10^{-16}[84]。

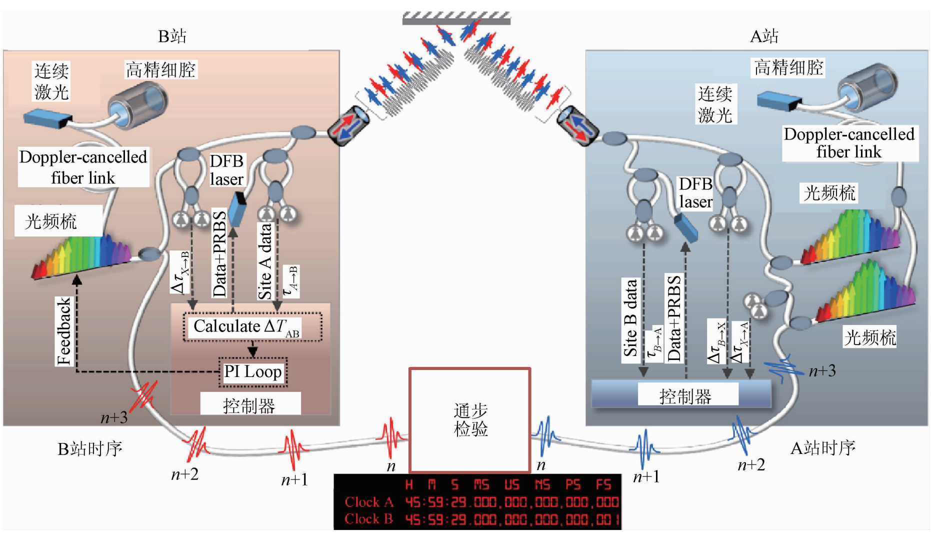


图 16　基于光频梳的双向自由空间时间频率传递[195]

Fig. 16　Two-way time and frequency transfer via frequency combs[195]

8　结　　论

光频梳作为微波频率和光学频率的桥梁,可使激光精密计量与测试技术直接溯源至时间频率基准。由于优异的时间频率特性,光频梳在激光频率测定、绝对距离测量、精密吸收光谱探测、高速非线性光谱与成像和高精度时间频率传递等方面展现出无可比拟的优势。不只如

此，光频梳在阿秒科学、低噪声微波产生、天文光谱观测、光钟、无标记显微成像等前沿领域已发挥出至关重要的作用，必将为测量与光谱技术的发展带来新跨越。作为一种具有优秀时频特性的飞秒激光器，光频梳的潜在应用还将得到不断的拓展；随着光频梳的发展与应用在国际研究中的日趋广泛，以及越来越多的国内研究小组开展相关工作，相信光频梳继续推进对精密世界以及高速动态过程的认识。

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