激光与光电子学进展

光学频率标准研究进展

赵国栋[1,2]，卢晓同[1*]，常宏[1,2**]

[1]中国科学院国家授时中心时间频率基准重点实验室，陕西 西安 710600；
[2]中国科学院大学天文与空间科学学院，北京 100049

摘要 光钟在近20年里发展迅速，稳定度和系统不确定度均比当前最好的微波原子钟高出两个量级，目前已有10个光学跃迁被国际计量局选定为二级秒定义并参与原子时的产生。本文介绍了光钟的工作原理和系统性能的评估，阐述了离子光钟和光晶格钟的最新研究进展，并总结了光钟绝对频率测量方法和进入二级秒定义的光频跃迁的测量结果。

关键词 光通信；光钟；光频标；秒定义；原子钟

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Research Progress of the Optical Frequency Standard

Zhao Guodong[1,2], Lu Xiaotong[1*], Chang Hong[1,2**]

[1]*Key Laboratory of Time and Frequency Primary Standards, National Time Service Center, Chinese Academy of Sciences, Xi'an* 710600, *Shaanxi, China*;
[2]*School of Astronomy and Space Science, University of Chinese Academy of Sciences, Beijing* 100049 *China*

Abstract Optical clocks have developed rapidly in the past 20 years, and their stability and systematic uncertainty are two orders of magnitude better than the current best microwave atomic clocks, and currently, there are 10 optical transitions which have been selected as the secondary representations of the definition of the second by the International Bureau of Metrology and participate in the generation of the international atomic time. This paper introduces the operational principle and evaluations of the performance of optical clocks, the latest research progress of ionic optical clocks and optical lattice clocks, elaborates the progress of the absolute frequency measurement of optical clocks, and summarizes the measurement results of the secondary representations of the definition of the second by optical frequency transitions.

Key words optical communications; optical clocks; optical frequency standard; definition of the second; atomic clock

1 引言

原子频标利用原子外层电子在两个能级间钟跃迁信号来锁定本地频率振荡器的频率，其概念最早由Maxwell于1873年提出。直到1955年英国国家物理实验室(NPL)的Essen等[1]首次实现了基于磁选态铯束原子钟，原子频标才得以实现。此后原子钟的精度不断提高，1967年，国际计量委员会(CIPM)通过决议将时间的基本单位“秒”定义为——位于海平面上的^{133}Cs原子基态的两个超精细能级在零磁场中跃迁振荡9192631770次所持续的时间为一个原子时秒。这是人类首次利用“自然标准”定义基本单位，开创了国际计量领域的新纪元。当前性能最好的微波原子钟的系统不确定度约为1×10^{-16}，其系统不确定度的进一步提升已经变得十分困难(主要受限于碰撞频移，分布腔相移和二阶塞曼频移)[2]。基于光频跃迁(比微波频率高5个数量级)的光钟有望实现10^{-19}量级甚至更低的系统不确定度，能大幅度提升原子频标的性能。

目前被研究得最多的光钟包括单离子光钟和基于大量中性原子的光晶格钟。随着激光冷却，窄线宽激光器和飞秒光学频率梳等技术的出现和发展[3-5]，光钟在近20年里取得了瞩目的研究成果。随着越来越多的光钟系统不确定度进入10^{-18}量级，CIPM已经将利用光学频率重定义时间的基本单位“秒”提上了日程。在2018年，CIPM就利用光钟作为“秒”定义的装置提出了5个具有里程碑意义的建议[6]。

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通信作者：*luxiaotong@ntsc.ac.cn；**changhong@ntsc.ac.cn

1）至少3台光钟（处于不同实验室或不同种类）的系统不确定度比当前最好的铯喷泉钟小约2个数量级。

2）里程碑1中至少有1台光钟与不同单位的光钟进行至少3次独立的频率比对测量（测量的不确定度小于5×10^{-18}），测量方式可以是与可搬运光钟比对，通过先进的链路比对或者（多种光钟间）频率比值的闭环测量。

3）里程碑1中的光钟与3台独立的铯主频标钟进行3次独立的频率比对，且测量结果从根本上受限于铯钟的不确定度（即光钟绝对频率的测量不确定度小于3×10^{-16}）。

4）光钟（或作为二级秒定义的光钟）可以定期参与国际原子时（TAI）的计算。

5）至少测量5种光钟间的光频比值，每组频率比值至少测量2次，且测量结果间的差异小于5×10^{-18}。

这5个里程碑式提议不仅要求光钟系统不确定度达到1×10^{-18}，还要求光钟的复现性优于5×10^{-18}且具备稳定报数的能力。而具备如此优异性能的光钟不仅能大幅度提高TAI的精度，还能用于一些前沿物理的研究，比如：通过测量光钟绝对频率随时间的变化来寻找精细结构常数随时间可能的微变，进而发现新的物理[7-8]；探测引力波和暗物质[9-10]；验证广义相对论预言的引力频移和洛伦兹变换对称性[11-13]；用于相对论测地学提高大地水平面的测量精度[14-15]；用于量子模拟[16-17]、量子计算[18]和量子存储[19]等。

尽管已有论文综述光钟的原理、关键技术和发展[20-21]，但目前还没有文献综述光钟近几年的发展且鲜有文献阐述光钟绝对频率测量。本文介绍了离子光钟和光晶格钟的工作原理及光钟性能的评估，包括：稳定度和系统不确定度；以时间节点为序，分别阐述了离子光钟和光晶格钟的研究进展；总结了光钟绝对频率测量的原理、方法和关键技术，以及所有参与二级秒定义的光频跃迁的测量结果。

2　光钟性能评估

光钟工作原理图1所示，原子（离子）源产生的热的原子（离子）经过激光冷却降低动能后被光晶格（离子阱）俘获。在完成态制备后，钟激光（其相位被参考至超稳光学腔上以提高频率稳定度）对原子（离子）进行钟跃迁探测以获得钟激光与钟跃迁共振频率间的频差，最后通过伺服系统纠正钟激光的频率使钟激光与原子跃迁保持共振。将光梳的重频和载波包络偏移频率锁定至氢钟（氢钟频率可溯源至国际原子）上后便可利用光学频率梳测量钟激光的频率，在修正系统误差和统计误差后就能推断钟跃迁的绝对频率。

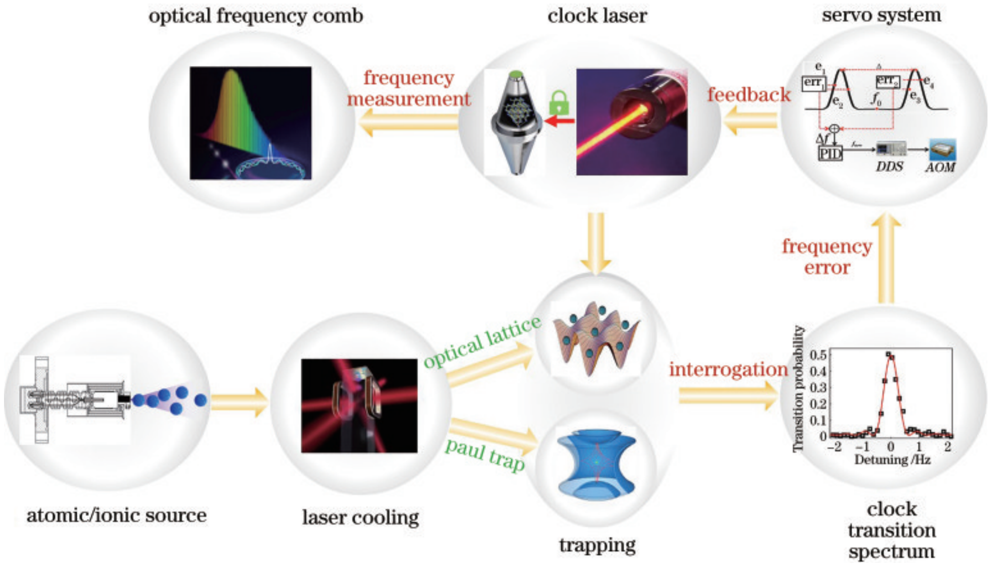

图1　光钟工作原理

Fig. 1　Operational principle of optical clocks

光钟性能评估主要包括稳定度和系统不确定度两个方面。稳定度从时域上表征了光钟输出频率的抖动，较高的稳定度意味着较高的自比对测量精度。系统不确定度则表征了光钟系统误差修正量的精度。由于电磁场、地球引力势和技术误差的存在，需要通过测量或结合数值计算的方式修正这些扰动对钟跃迁频率的影响，进而获得无外场扰动的、参考到地球平均海平面的钟跃迁频率。较高的稳定度则可以在有限的测量时间内获得更小的测量误差，有助于更为准确地测量系统误差，进而减小总的系统不确定度。

2.1　稳定度

影响光钟稳定度的因素主要包括Dick噪声和原子探测噪声[22]。Dick噪声来源于光钟周期性地钟跃迁探测，钟激光的随机噪声无法被平均至零，即噪声会导致钟跃迁谱线激发率发生变化并由伺服系统对这个噪声进行补偿。然而白噪声长期的平均值为0，不需

要被修正，这就意味着伺服系统根据激发率波动（白噪声导致的）对钟激光频率进行了错误地修正，最终导致钟激光的频率抖动变大，降低光钟的稳定度。Dick噪声是限制目前绝大多数光晶格钟稳定度的最主要因素。Dick噪声限制的稳定度[22]可以表示为

$$\sigma_{\mathrm{Dick}}(\tau)=\sqrt{\frac{1}{\tau}\frac{1}{|g_0|^2}\sum_{k=1}^{\infty}S_y(k/T)|g_k|^2}\,,\tag{1}$$

式中：τ为平均时间；T为钟反馈周期；$S_y(\cdot)$代表钟激光的单边噪声功率密度；g_k为频率敏感函数傅里叶展开后的系数。当钟激光的噪声主要为闪烁频率噪声（或者$1/f$噪声）时，式（1）可以被近似[23]表示为

$$\sigma_{\mathrm{Dick}}(\tau)=\frac{\sigma_{\mathrm{clock}}}{\sqrt{2\ln 2}}\sqrt{\frac{T}{\tau}}\sqrt{\sum_{k=1}^{\infty}\frac{1}{k}\frac{|g_k|^2}{|g_0|^2}}\,,\tag{2}$$

式中：σ_{clock}为钟激光的闪烁频率噪声极限，可通过分析钟运行过程的钟频率误差信号获得，即可以在不知道钟激光单边噪声功率密度情况下估计系统的Dick极限。

光钟利用“shelved optical electron amplifier”技术探测钟跃迁激发率[24]。对于单离子光钟而言，只需要重复探测处于基态的粒子数就能推断钟跃迁的激发概率，光晶格钟则需要分别探测处于基态和激发态的粒子数（获得激发态粒子数和总的粒子数）。光晶格钟通常采用破坏式探测方式，每个钟探测周期都需要花费大量的时间来制备冷原子，光晶格钟的稳度主要受限于Dick噪声。通过提升钟激光稳定度[25-26]、提高钟探测占空比[27]、采用“零死时间”运行模式[28]和非破坏性探测[29]等方式可以减小Dick噪声的影响。得益于深的离子阱，钟跃迁探测不会导致离子逃逸（但会增加离子温度），因此离子光钟不需要重复地制备离子，钟探测的占空比大，Dick噪声对其稳定度影响通常较小。然而，离子光钟参与钟跃迁探测的粒子数只有1个，这就导致其原子探测噪声很强。受限于原子探测噪声的稳定度[22,30]可以表示为

$$\sigma_{\mathrm{Det}}(\tau)=\frac{1}{2K_0 f_0}\sqrt{\frac{T_{\mathrm{C}}}{\tau}}\sqrt{\frac{1}{N}+\frac{1}{\gamma N}+\delta_{\mathrm{N}}^2}\,,\tag{3}$$

式中：K_0表示钟跃迁谱线在半高处的斜率（频率敏感度）；f_0为钟跃迁频率；T_{C}为钟探测周期；N表示参与钟探测的粒子数；γ对应每个粒子被探测到的光子数；δ_{N}为技术噪声。对于单离子光钟而言$N=1$，因此稳定度通常在$10^{-15}\,(\tau/\mathrm{s})^{-0.5}$量级。通过减小钟跃迁谱线的线宽（提升钟探测时间）可以增加K_0，进而提升稳定度。此外，离子晶体光钟可以同时俘获大量的离子，有望大幅度提升稳定度[31-32]。

2.2　不确定度

通过测量或者计算获得的系统各项频移量都会有一个不确定度，一般由测量或计算结果的1倍标准差来表征。由于目前秒定义的装置不是光钟，因此光钟无法用准确度来评估其性能，而系统所有频移修正量的B类不确定度则是当前唯一能反映光钟自评估精度的量。

光晶格钟利用囚禁在（光驻波场形成的）势阱里大量中性原子减小原子探测噪声，具备10^{-18}甚至更高秒稳定度的潜力。尽管将晶格光波长调节至“魔术波长”可以消除一阶交流斯塔克频移[33]，但剩余的高阶斯塔克频移在高精度光晶格钟系统评估中不容忽视[34]。而同一格点原子间相互作用也会导致与原子密度相关的频移[35]。对于光晶格钟而言，系统的主要频移项包括：黑体辐射频移、密度频移、晶格光交流斯塔克频移、二阶塞曼频移、直流斯塔克频移和钟激光交流斯塔克频移。其中，黑体辐射频移是绝大多数光晶格钟系统频移量和不确定度最大的项，也是限制$^{87}\mathrm{Sr}$和$^{171}\mathrm{Yb}$等光晶格钟系统不确定度进入10^{-19}量级的最主要因素[36-37]。这种情况下，研制基于$^{169}\mathrm{Tm}$[38]、$^{199}\mathrm{Hg}$[39]、$^{111}\mathrm{Cd}$[40]、$^{24}\mathrm{Mg}$[41]等元素的光晶格有望将室温下的黑体辐射频移不确定度降一个量级以上。此外，冷光钟也可以将黑体辐射频移总的不确定度降低至10^{-19}量级[42]，其通过移动光晶格技术将原子移动到一个低温腔体中进行钟跃迁探测，有效地降低了黑体辐射频移量及相应的不确定度。除了上述主要频移项外，还有许多效应会引入系统频移：背景气体碰撞、线牵引、隧穿效应、二阶多普勒频移、声光调制器相位啁啾和伺服误差。这些效应导致的频移量通常可以被控制在10^{-18}量级或者更小，但对于高精度的光晶格钟而言，仍需要严格地评估这些效应的影响[36-37]。

单离子光钟只有一个离子，因此原则上不存在密度频移，然而离子在势阱中的宏运动和微运动导致了二阶多普勒频移和交流斯塔克频移。此外，离子阱周围杂散电磁场和离子阱缺陷等因素会导致离子偏离鞍点，还会受到射频场导致的额外微运动频移。不同的离子光钟评估的主要系统频移项不一致，但一般都需要评估的系统频移项包括：宏运动频移、额外微运动频移、黑体辐射频移、二阶塞曼频移、电四极频移、钟激光交流斯塔克频移、背景气体碰撞频移、伺服误差和一阶多普勒频移[43-51]。常见的离子光钟里面，$^{40}\mathrm{Ca}^+$[43]、$^{88}\mathrm{Sr}^+$[44]和$^{171}\mathrm{Yb}^+$[45-46]光钟的主要系统频移项仍是黑体辐射频移，而$^{27}\mathrm{Al}^+$[47-48]、$^{199}\mathrm{Hg}^+$[49]和$^{115}\mathrm{In}^+$[50]光钟室温下的黑体频移很小，黑体辐射项的不确定度通常在10^{-19}量级。当前，较高的离子温度（mK量级）、离子阱缺陷和杂散电磁场导致的宏运动频移和额外微运动频移是限制高性能离子光钟（如$^{27}\mathrm{Al}^+$光钟）系统不确定度的最主要因素。

3　光钟研究进展

3.1　离子光钟

利用离子实现光学频率标准的想法最早由Dehmelt[51]于1973年提出。受限于当时光频测量困难、离子动能大和缺乏稳定光学本地振荡器等因素，离子

光钟的研制进展缓慢。随着光学频率梳、激光冷却和窄线宽激光器等技术的出现，2001年，美国国家标准标准局（NIST）报道了$^{199}\mathrm{Hg}^{+}$光钟和$7\times10^{-15}\ (\tau/\mathrm{s})^{-0.5}$的稳定度，基于单离子的光学频率标准才真正意义上被实现[52]。此后20年，单离子光钟的性能不断提升，离子种类和研究机构也日益增加。2006年，NIST将$^{199}\mathrm{Hg}^{+}$光钟的系统不确定度降低至7.2×10^{-17}，超越了最好的微波原子钟，并将钟跃迁绝对频率的测量不确定度减小至9.1×10^{-16}，是光学频率标准的重要进展[53]。2012年，NIST将$^{27}\mathrm{Al}^{+}$光钟的系统不确定减小至8.6×10^{-18}，首次将光钟的系统不确定度推进至10^{-18}量级[45]。2019年，通过新设计的离子阱将离子制备到接近振动基态，NIST将$^{27}\mathrm{Al}^{+}$光钟的系统不确定进一步减小至9.4×10^{-19}，将光钟的系统不确定度推进至10^{-19}量级[46]。$^{27}\mathrm{Al}^{+}$光钟需要用到量子逻辑谱进行钟跃迁探测[54]，而$^{199}\mathrm{Hg}^{+}$光钟的钟跃迁探测和用于激光冷却的光都在紫外波段。这些因素导致$^{27}\mathrm{Al}^{+}$和$^{199}\mathrm{Hg}^{+}$光钟的研究难度很大。因此，一些容易实现的别的元素的单离子光钟相继被实现，包括$^{171}\mathrm{Yb}^{+}$[43-44]、$^{88}\mathrm{Sr}^{+}$[42]、$^{40}\mathrm{Ca}^{+}$[41]、$^{117}\mathrm{In}^{+}$[48]。表1总结了当前国内外单离子光钟的系统不确定度和稳定度（其中，PTB为德国物理技术研究院，NICT为日本国家信息通信技术研究所，APM为中国科学院精密测量科学与技术创新研究院，NRC为加拿大国家研究委员会）。2019年，PTB通过两台$^{171}\mathrm{Yb}^{+}$光钟（系统不确定均约3×10^{-18}）长达半年的频率比对，在10^{-21}量级验证了洛伦兹对称性[13]；APM最近几年在$^{40}\mathrm{Ca}^{+}$离子光钟方面取得了一系列进展并于2021年通过液氮冷却降低系统黑体辐射频移，将$^{40}\mathrm{Ca}^{+}$离子光钟总的系统不确定度降低至3×10^{-18}，达到了世界先进水平[55]。APM还成功研制了$^{27}\mathrm{Al}^{+}$光钟（协同冷却离子为$^{40}\mathrm{Ca}^{+}$），系统不确定度为7.8×10^{-18}[56]。此外，华中科技大学也开展了$^{27}\mathrm{Al}^{+}$光钟的研制，并完成了钟跃迁谱线的探测[57]。

表1　国内外代表性单离子光钟系统不确定度和稳定度

Table 1　Systematic uncertainty and stability of representative single-ion optical clocks in the domestic and overseas

Type	Institution	Uncertainty	Stability /[$(\tau/\mathrm{s})^{-0.5}$]
$^{27}\mathrm{Al}^{+}$	NIST	9.4×10^{-19}[46]	1.2×10^{-15}[46]
$^{27}\mathrm{Al}^{+}$	APM	7.9×10^{-18}[47]	3.4×10^{-14}[13]
$^{171}\mathrm{Yb}^{+}$	PTB	2.7×10^{-18}[13]	1.4×10^{-15}[13]
$^{88}\mathrm{Sr}^{+}$	NRC	1.1×10^{-17}[61]	3×10^{-15}[62]
$^{88}\mathrm{Sr}^{+}$	NPL	4.9×10^{-17}[63]	2.2×10^{-14}[63]
$^{40}\mathrm{Ca}^{+}$	APM	3×10^{-18}[56]	3×10^{-15}[56]
$^{117}\mathrm{In}^{+}$	NICT	5×10^{-16}[48]	–
$^{199}\mathrm{Hg}^{+}$	NIST	1.9×10^{-17}[47]	3.9×10^{-15}[47]

除了上述元素的离子光钟，最近几年，黑体辐射频移极小的$^{176}\mathrm{Lu}^{+}$光钟也取得了重要研究进展[58]，2018年，新加坡大学量子技术中心的研究表明，$^{176}\mathrm{Lu}^{+}$光钟室温下（假设温度不确定度为5 K）的系统黑体辐射频移不确定度低至2×10^{-19}。2022年，加利福尼亚大学实现了对精细结构常数变化非常敏感的$^{226}\mathrm{Ra}^{+}$离子光钟[59]，其系统不确定度为9×10^{-16}，稳定度为$1.1\times10^{-13}\ (\tau/\mathrm{s})^{-0.5}$。同年，PTB首次实现了$\mathrm{Ar}^{13+}$高价离子光钟[60]，系统不确定度为$2.2\times10^{-17}$，稳定度为$3.2\times10^{-14}\ (\tau/\mathrm{s})^{-0.5}$。高价离子光钟对外界电磁场的扰动很不敏感，是高性能离子光钟的发展的新趋势，其在测量精细结构常数随时间可能的微变，寻找暗物质和寻找第五种假设的基本作用力等方面有着传统光钟不具备的优势。

3.2　光晶格钟

在2002年第六届频率标准与计量研讨会上，日本东京大学（UT）的Katori[64]提出了高精度光晶格钟的实现方式：利用光晶格将大量中性原子囚禁在Lamb-Dicke区域以消除光子反冲频移和一阶多普勒频移，且形成光晶格的晶格光工作在“魔术波长”以消除一阶光频移。2003年，Katori等[65]进一步指出以$^{87}\mathrm{Sr}$原子$5s^2\ ^1S_0\rightarrow 5s5p\ ^3P_0$为钟跃迁的光晶格钟可以将高阶光频移控制在$10^{-17}$以下，并于2005年率先实现了$^{87}\mathrm{Sr}$光晶格钟[66]。2006年，法国的巴黎天文台（LNE-SYRTE）和美国联合天体联合物理实验室（JILA）也实现了$^{87}\mathrm{Sr}$光晶格钟并测量了钟跃迁的绝对频率[67-68]，其结果与同年UT重测的结果一致[69]。此后光晶格钟的发展速度极快，其他原子种类的光晶格钟也相继被实现，如$^{88}\mathrm{Sr}$[70]、$^{171}\mathrm{Yb}$[71]和$^{199}\mathrm{Hg}$[72]。2008年，JILA将$^{87}\mathrm{Sr}$光晶格钟的系统不确定度减小至1.5×10^{-16}，且稳定度为3×10^{-16}（200 s平均时间），超越了当时性能最好的微波原子钟[73]。2013年，NIST将$^{171}\mathrm{Yb}$光晶格钟的稳定度提升至$3.2\times10^{-16}\ (\tau/\mathrm{s})^{-0.5}$；经过7000 s的平均，实现了$1.8\times10^{-18}$的频率稳定度，在世界上首次展示了$10^{-18}$量级的频率稳定度[74]。2014年，JILA实现了基于腔增益的$^{87}\mathrm{Sr}$光晶格钟，其系统不确定度为6.4×10^{-18}[75]；次年，他们更加精确地评估了黑体辐射频移和晶格光交流斯塔克频移，将系统不确定度降低至2×10^{-18}，且稳定度为$2.2\times10^{-16}\ (\tau/\mathrm{s})^{-0.5}$，成为了当时世界上性能最好的钟[76]。2017年，NIST的$^{171}\mathrm{Yb}$光晶格钟研究组通过交替探测两团冷原子的办法来消除Dick噪声，实现了“零死亡时间”光晶格钟，将稳定度提升至$6\times10^{-17}\ (\tau/\mathrm{s})^{-0.5}$[28]；他们提出“运行魔术波长”技术使一阶和二阶晶格光交流斯塔克频移相互抵消，可将晶格光交流斯塔克频移的不确定度降低至10^{-19}量级[77]。2018年，Ushijima等[78]提出“魔术光强”技术可将晶格光交流斯塔克频移不确定度降低至2×10^{-19}。NIST将$^{171}\mathrm{Yb}$光晶格钟的系统不确定度降低至1.4×10^{-18}，并经过10^5 s的平均将稳定度提升至3.2×10^{-19}[35]。2019年，JILA将钟激光参考到工作温度为124 K的低温单晶硅光学腔上，将钟激光的线宽和热

噪声极限稳定度分别降低至 7 mHz 和 4×10^{-17}，并通过两台光钟（分别为一维和三维^{87}Sr光晶格钟）完全异步的频率比对推断单台光钟的稳定度为 $4.8\times10^{-17}(\tau/\mathrm{s})^{-0.5}$，是目前稳定度最高的光钟[79]。2020年，Takamoto等[11]研制了两台可搬运^{87}Sr光晶格钟，且系统稳定度均约 5.5×10^{-18}；通过同步测量两台高度差约452 m的可搬运光晶格钟间的频差，他们将广义相对论引力频移修正系数降低至 $(1.4\pm9.1)\times10^{-5}$，其测量精度与利用两台高度差约8500 km的伽利略卫星的测量结果 $(0.19\pm2.48)\times10^{-5}$ 在同一量级。这是首次在地面上将引力频移修正系数的不确定度降低至 10^{-5} 量级，展示了光晶格钟的超高的测量精度。2021年，JILA通过原位测量技术在亚毫米量级精确验证了广义相对论指出的引力频移，其测量精度达到了惊人的 7.6×10^{-21}[12]。同年，NIST报道了^{87}Sr光晶格钟，^{171}Yb光晶格钟和^{27}Al^{+}光钟的相互的频率比对结果，将频率^{27}Al^{+}/^{171}Yb、^{27}Al^{+}/^{87}Sr和^{171}Yb/^{87}Sr的频率比值不确定度分别降低至 5.9×10^{-18}、8×10^{-18}和 6.8×10^{-18}，且将三种光钟的频率比值的闭合值测量不确定度降低至 6×10^{-19}[80]。2021年，中国科学院国家授时中心（NTSC）报道了其研制的空间站^{87}Sr光晶格钟原理样机[81]；并于2022年随梦天号实验舱发生升空。2022年，JILA提出"魔术阱深"技术大幅度降低（沿重力方向）浅光晶格里的密度频移[82]。表2列出了国内外代表性光晶格钟的研究进展（RIKEN为日本理化技术研究所，NIM为中国计量科学研究院，ECNU为华东师范大学）。

表2　国内外代表性光晶格原子钟的系统不确定度和稳定度

Table 2　Systematic uncertainty and stability of representative optical lattice clocks in the domestic and overseas

Type	Institution	Uncertainty	Stability /[$(\tau/\mathrm{s})^{-0.5}$]
^{87}Sr	JILA	2×10^{-18}[34]	4.8×10^{-17}[79]
^{171}Yb	NIST	1.4×10^{-18}[35]	6×10^{-17}[28]
^{88}Sr	PTB	2×10^{-17}[70]	1.6×10^{-16}[70]
^{87}Sr	PTB	1.5×10^{-17}[83]	4.1×10^{-16}[22]
^{199}Hg	RIKEN	7.5×10^{-17}[84]	2×10^{-15}[84]
^{199}Hg	LNE-SYRTE	1.7×10^{-16}[85]	3.4×10^{-15}[85]
^{171}Yb	ECNU	1.27×10^{-16}[86]	–
^{171}Yb	APM	–	4.6×10^{-16}[87]
^{87}Sr	NIM	7.2×10^{-18}[88]	1.18×10^{-15}[88]
^{87}Sr	NTSC	5.1×10^{-17}[89]	4.7×10^{-16}[89]

4　光钟的绝对频率测量与二级秒定义

当前时间的基本单位——"秒"是由^{133}Cs喷泉钟定义的，因此光钟的绝对频率需要溯源到^{133}Cs喷泉钟上。光钟绝对频率测量原理如图2所示，光钟与飞秒光学频率梳某一根梳齿进行拍频，而光梳的重复频率（f_{r}）和载波相位包络偏移（f_{ceo}）则参考一个标准频率源上。标准频率源可以是绝对频率已知的光钟、（作为主频标或者次级频标的）喷泉钟或者氢钟。同类型的光钟可以通过光学频率比对的方式直接测量两台钟的频差，然后推断另一台频率未知的光钟的绝对频率[90]。但不同类型光钟间的频率比对需要借助光学频率梳[80]，且通过光学频率梳将任意两台光钟的钟激光相

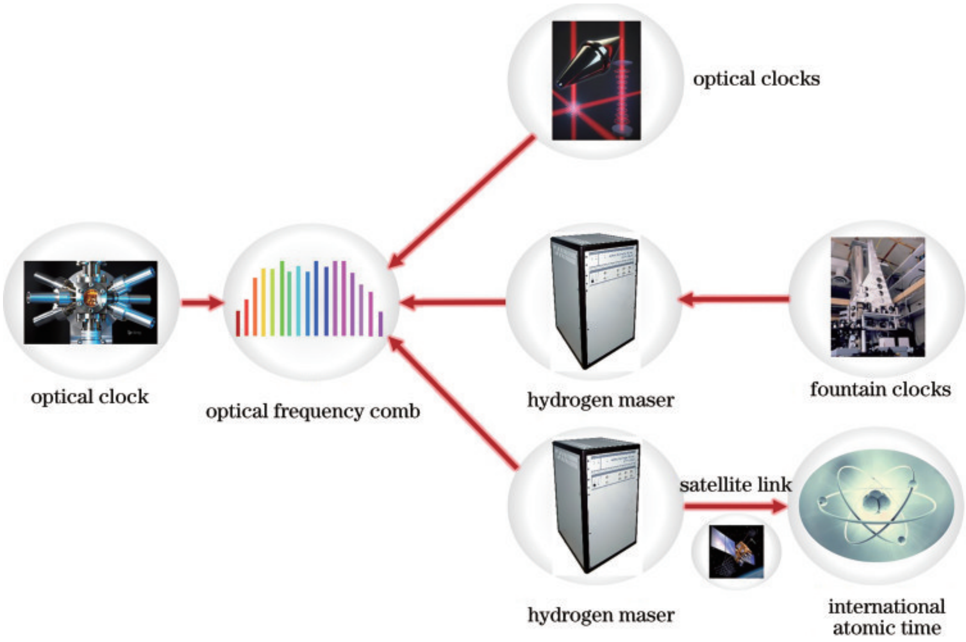

图2　光钟绝对频率测量原理

Fig. 2　Absolute frequency measurement principle of optical clock

位相互锁定，并利用同步测量技术提高测量精度[91]。将飞秒光学频率梳锁定到利用主频标或者二级频标喷泉钟驾驭的氢钟上就可以将光频溯源到喷泉钟上，而作为主频标或者二级频标喷泉钟的频率是已知的。光钟与主频标（即^{133}Cs喷泉钟）的比对且频率测量精度受限于主频标的系统不确定度是光频跃迁更替秒定义的前提条件，因此许多研究团队开展了相关的实验工作[14,83,92]。其中，2013年，LNE-SYRTE报道了他们两台锶光钟分别与3台独立的^{133}Cs喷泉钟（FO1、FO2和FOM）的频率比对结果[92]，将^{87}Sr光晶格钟的绝对频率不确定度降低至3.1×10^{-16}，测量精度受限于微波原子钟的系统不确定度。2020年，PTB报道了他们2017～2019年期间^{87}Sr光晶格钟与两台^{133}Cs喷泉钟（CSF1和CSF2）的频率比对结果[83]，将^{87}Sr光晶格钟的绝对频率不确定度降低至1.5×10^{-16}。

当没有喷泉钟和频率已知的光钟时，通过氢钟溯源TAI同样可以精确地测量光钟的绝对频率。这里的氢钟能够溯源本地协调世界时，而本地协调世界时通过卫星可溯源TAI，最后通过每个月发布的时间公报（Circular T）就可以将氢钟溯源到TAI和由主频标和二级频标定义的秒（SI Second）上。2012年，NRC的^{88}Sr^{+}光钟研究组首次展示了通过溯源TAI的方式测量光钟的绝对频率，此后这种技术被广泛地应用[147]。其中，2021年，韩国标准科学研究院（KIRSS）采用溯源TAI的方式测量了^{171}Yb光晶格钟的绝对频率[111]，通过长达1年的测量时间，他们将绝对频率的测量不确定度降低至2.6×10^{-16}。通过溯源TAI的方式测量光钟绝对频率的测量不确定度主要来自：卫星链路和氢钟“死时间”。其中，卫星链路不确定度的贡献[106]可以表示为

$$u\left[\frac{f_{\mathrm{UTC(k)}}}{f_{\mathrm{TAI}}}-1\right]=\frac{\sqrt{2}\times u_{\mathrm{Link}}}{86400\times5}\left(\frac{5}{T_{\mathrm{total}}}\right)^{0.9},\tag{4}$$

式中：$f_{\mathrm{UTC(k)}}$和f_{TAI}分别表示协调世界时在本地守时实验室的实现和国际原子时的频率；u_{Link}为卫星链路的A类不确定度；T_{total}表示总的绝对频率测量天数。显然，通过增加有效测量时间，可以有效减小卫星链路误差对绝对频率测量结果的影响。目前光晶格钟系统有效运行率通常在60%左右，导致光钟与氢钟比对不连续。分析氢钟“死时间”导致的不确定度的常用方式是：根据氢钟的噪声模型产生随机噪声，噪声持续的时间覆盖所有的测量时间（死时间+有效测量时间），然后分析有效测量时间内的噪声均值与总体均值的差异，重复这个过程并将它们的标准差作为氢钟“死时间”导致的统计不确定度[86,98,105]。TAI与秒定义（SI）差异的不确定度可以通过每月发布的Circular T获得。当绝对频率的测量周期无法完整对应Circular T发布的周期时，则需要具体计算测量过程中的TAI均值，将产生额外的不确定度。

除了主频标，其他能够参与TAI计算的频标被称为二级频标。目前总共有10个光跃迁频率被BIPM采纳为二级频标[154]，包括：^{87}Sr（$5s^{2}\ {}^{1}S_{0}\rightarrow5s5p\ {}^{3}P_{0}$）、^{88}Sr（$5s^{2}\ {}^{1}S_{0}\rightarrow5s5p\ {}^{3}P_{0}$）、^{88}Sr^{+}（$5s\ {}^{2}S_{1/2}\rightarrow4d\ {}^{2}D_{5/2}$）、^{171}Yb（$6s^{2}\ {}^{1}S_{0}\rightarrow6s6p\ {}^{3}P_{0}$）、^{171}Yb^{+}（$6s\ {}^{2}S_{1/2}\rightarrow5d\ {}^{2}D_{3/2}$）、^{171}Yb^{+}（$6s\ {}^{2}S_{1/2}-4f^{13}6s^{2}\ {}^{2}F_{7/2}$）、^{199}Hg（$6s^{2}\ {}^{1}S_{0}\rightarrow6s6p\ {}^{3}P_{0}$）、^{199}Hg^{+}（$5d^{10}6s\ {}^{2}S_{1/2}\rightarrow5d^{9}6s^{2}\ {}^{2}D_{5/2}$）、^{27}Al^{+}（$3s^{2}\ {}^{1}S_{0}\rightarrow3s3p\ {}^{3}P_{0}$）、^{40}Ca^{+}（$4s\ {}^{2}S_{1/2}\rightarrow3d\ {}^{2}D_{5/2}$）。图3总结了各个组对这10个二级频标绝对频率测量结果。随着越来越多的光钟进入二级秒定义且逐渐参与国际原子时的产生，利用光频标更替秒定义的进程正在稳步推进，2022年，第27次国际计量大会（CGPM）已经通过决议：将在2026年，第28次CGPM上讨论选择重定义秒的光频标元素种类，并在2030年，第29次CGPM上正式将光频标作为主频标[155]。

5　结束语

光钟已经实现了4.8×10^{-17}的秒级稳定度，9.4×10^{-19}的准确度，且两台光钟比对的频率复现性也达到了1×10^{-18}，各项指标均比最好的微波原子钟高出两个数量级。经过世界各国的共同努力，CIPM 2018提出的光钟作为秒定义的复现装置提出的5个建议中：第1条和第3条已经基本实现[13,46,34-35,83,92]；第4条建议则比较容易实现；第2条和第5条建议要求不同实验室间光钟的频率比对不确定度优于5×10^{-18}，对光钟本身的稳定度和频率比对链路要求都很高。目前不同实验室间（或不同类型）光钟频率比对的最小不确定度为5.9×10^{-18}[80]，已经非常接近预期目标。

广义相对论指出越靠近大质量天体，时钟就会走得越慢，即被广泛验证了其正确性的引力频移。由于地球引力势随时间变化，地面光钟的频率约以每年10^{-18}的相对速率变化[156]。由于导致引力频移的是地球重力势能，因此工作在微重力环境的空间光钟可大幅度降低引力频移，提升光钟长期的频率稳定性。高性能的空间光钟结合先进的卫星-卫星和卫星-地面链路可对卫星导航、深空探测、相对论测地学和基础物理研究等领域产生深远的影响[98,157]。国内外已经有不少单位提出了空间光钟研制计划。欧洲联盟在2007年就提出了空间光学原子钟（SOC）计划，旨在实现系统不确定度小于2×10^{-17}的空间（锶原子）光晶格钟[158]，并于2018年实现了空间^{88}Sr光晶格原子钟样机且系统的不确定度达到了2×10^{-17}，自比对稳定度为$4.1\times10^{-16}\ (\tau/\mathrm{s})^{-0.5}$。日本于2011年提出研制国际空间站的日本实验舱内的“多用途小型有效载荷柜（MSPR）”的计划，以建立高精度时频体系和进行一系列基础物理研究。RIKEN于2020年报道的可搬运^{87}Sr光晶格钟在体积和功耗方面已经达到了计划要求[11]，但离在轨运行和火箭发射要求还有一定的距离。在“十三五”期间，中国提出了“高精密时频”计划，其目标包括建立

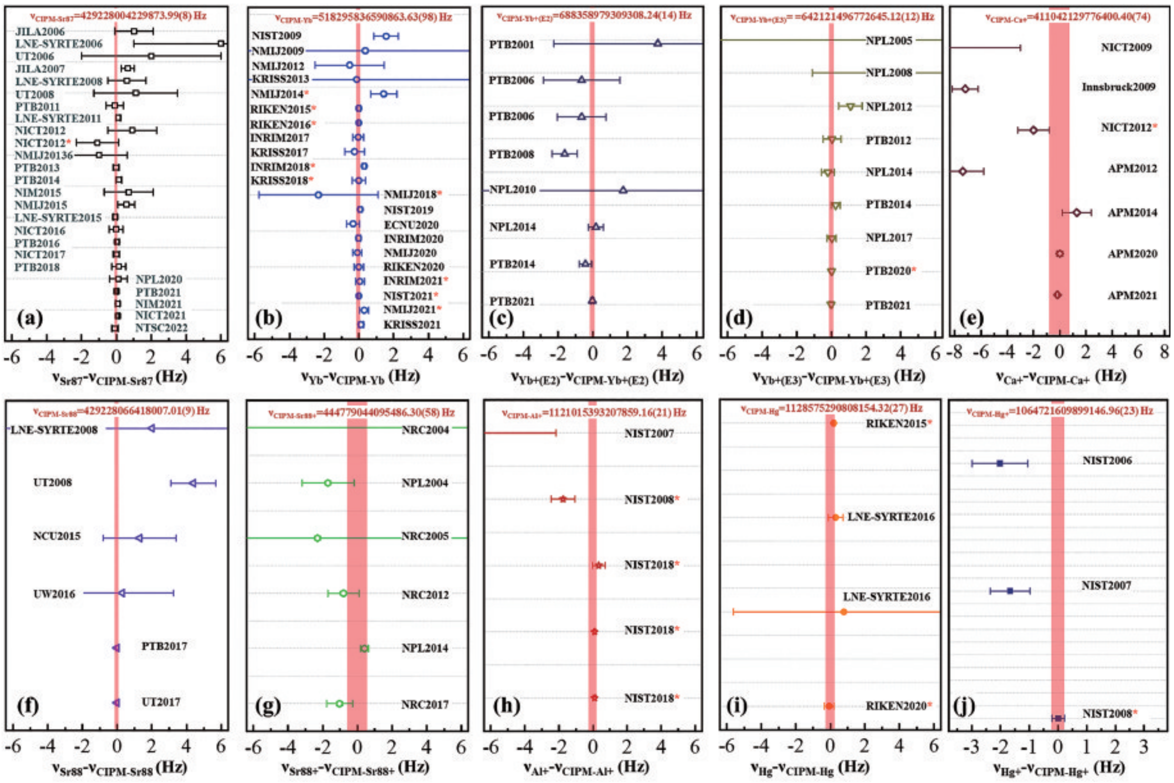

图 3 参与二级秒定义的光频跃迁的绝对频率测量[红色阴影区域表示 CIPM 2021 年推荐的频率值和相应的不确定度(不是图中数据加权平均的结果)]。(a)^{87}Sr 光晶格钟绝对频率测量[JILA[67,93]、LNE-SYRTE[68,92,94]、NICT[95-98]、日本计量研究院(NMIJ[99-100])、PTB[14,83,101-103]、NIM[104-105]、NPL[106]、UT[69, 107]、NTSC[89]];(b)^{171}Yb 光晶格钟绝对频率测量[NIST[71,80]、KRISS[14,108-111]、意大利国家计量研究院(INRIM[112-114])、RIKEN[84,91,115]、NMIJ[116-121]、ECNU[86]];(c)^{171}Yb^{+}(E2)光钟绝对频率测量(PTB[122-127]、NPL[7,128]);(d)^{171}Yb^{+}(E3)光钟绝对频率测量(PTB[8,43,127, 129]、NPL[7,130-133]);(e)^{40}Ca^{+}光钟绝对频率测量[NICT[96,134]、茵斯布鲁克大学(INNSBURK[135])、APM[136-1139]];(f)^{88}Sr 光晶格钟绝对频率测量[LNE-SYRTE[140]、UT[141-142]、哥白尼大学(NCU[143])、华沙大学(UW[144])、PTB[70]];(g)^{88}Sr^{+}光钟绝对频率测量(NRC[145-148]、NPL[63,149]);(h)^{27}Al^{+}光钟绝对频率测量(NIST[80,150-151]);(i)^{199}Hg 光晶格钟绝对频率测量(LNE-SYRTE[37,85]、RIKEN[84,152]);(j)^{199}Hg^{+}光钟绝对频率测量(NIST[47,53,153])(名称上带“*”号的表示其绝对频率是通过不同光钟频率比值测量获得)

Fig. 3 Absolute frequency measurements of the optical transitions adopted as the secondary representations of the definition of the second [red shaded areas represent CIPM 2021 recommended frequency values and their corresponding uncertainties (they are not the result of a weighted average of the data in the figures)]. (a) Absolute frequency measurements of the ^{87}Sr optical lattice clock [JILA [67,93], LNE-SYRTE[68,92,94], NICT[95-98], National Metrology Institute of Japan (NMIJ[99-100]), PTB[14,83,101-104], NIM [104-105], NPL[106], UT[69, 107], and NTSC[89]]; (b) absolute frequency measurements of the ^{171}Yb optical lattice clock [NIST[71,80], KRISS[14,108-111], Istituto Nazionale di Ricerca Metrologica (INRIM[112-114]), RIKEN[84,91,115], NMIJ[116-121], and ECNU[86]]; (c) absolute frequency measurements of the ^{171}Yb^{+} (E2) optical clock [PTB[122-127] and NPL [7,128]]; (d) absolute frequency measurements of the ^{171}Yb^{+} (E3) optical clock [PTB[8,43,127, 129] and NPL [7,130-133]]; (e) absolute frequency measurements of the ^{40}Ca^{+} optical clock [NICT[96,134], Universität Innsbruck (INNSBURK[135]), and APM[136-139]]; (f) absolute frequency measurements of the ^{88}Sr optical lattice clock [LNE-SYRTE[140], UT [141-142], Nicolaus Copernicus University (NCU[143]), University of Warsaw (UW[144]), and PTB[70]]; (g) absolute frequency measurements of the ^{88}Sr^{+} optical clock (NRC[145-148] and NPL[63,149]); (h) absolute frequency measurements of the ^{27}Al^{+} optical clock (NIST[80,150-151]); (i) absolute frequency measurements of the ^{199}Hg optical lattice clock (LNE-SYRTE[37,85] and RIKEN[84,152]); (j) absolute frequency measurements of the ^{199}Hg^{+} optical clock (NIST[47,53,153]) (names marked with an "*" indicates that their absolute frequencies are obtained by frequency ratio measurement between different optical clocks)

不确定度优于 10^{-18} 的空间时频体系,并在此基础上进行一系列空-地时频传递和基础物理实验。2022年,由NTSC主导研制的空间^{87}Sr光晶格原子钟已经成功发射进入中国空间站,是空间光钟研制的重要进展。美国也于2021年底美国国防部高级研究计划局提出“ROCkN”项目,其目的是研制可在车辆、飞机和卫星等平台上运行的光钟[159],且精度比现有最好微波原子钟精度高两个量级。随着可移动和空间光钟的发展以及高精度的空间和地面光钟网络的建立,光学频率标准将大大提高卫星导航的精度,并极大地促进基础科学研究和工程技术等领域的发展。

参 考 文 献

[1] Essen L, Parry J V L. An atomic standard of frequency and time interval: a Cæsium resonator[J]. Nature, 1955, 176(4476): 280-282.

[2] Heavner T P, Donley E A, Levi F, et al. First accuracy evaluation of NIST-F_2[J]. Metrologia, 2014, 51(3): 174-182.

[3] Chu S, Hollberg L, Bjorkholm J E, et al. Three-dimensional viscous confinement and cooling of atoms by resonance radiation pressure[J]. Physical Review Letters, 1985, 55(1): 48-51.

[4] Salomon C, Hils D, Hall J L. Laser stabilization at the millihertz level[J]. Journal of the Optical Society of America B, 1988, 5(8): 1576-1587.

[5] Udem T, Reichert J, Holzwarth R, et al. Absolute optical frequency measurement of the cesium D_1 line with a mode-locked laser[J]. Physical Review Letters, 1999, 82(18): 3568-3571.

[6] Riehle F, Gill P, Arias F, et al. The CIPM list of recommended frequency standard values: guidelines and procedures[J]. Metrologia, 2018, 55(2): 188-200.

[7] Godun R M, Nisbet-Jones P R, Jones J M, et al. Frequency ratio of two optical clock transitions in $^{171}Yb^+$ and constraints on the time variation of fundamental constants[J]. Physical Review Letters, 2014, 113(21): 210801.

[8] Huntemann N, Lipphardt B, Tamm C, et al. Improved limit on a temporal variation of m_p/m_e from comparisons of Yb^+ and Cs atomic clocks[J]. Physical Review Letters, 2014, 113(21): 210802.

[9] Kolkowitz S, Pikovski I, Langellier N, et al. Gravitational wave detection with optical lattice atomic clocks[J]. Physical Review D, 2016, 94(12): 124043.

[10] Wcisło P, Morzyński P, Bober M, et al. Experimental constraint on dark matter detection with optical atomic clocks[J]. Nature Astronomy, 2017, 1: 9.

[11] Takamoto M, Ushijima I, Ohmae N, et al. Test of general relativity by a pair of transportable optical lattice clocks[J]. Nature Photonics, 2020, 14(7): 411-415.

[12] Bothwell T, Kennedy C J, Aeppli A, et al. Resolving the gravitational redshift across a millimetre-scale atomic sample[J]. Nature, 2022, 602(7897): 420-424.

[13] Sanner C, Huntemann N, Lange R, et al. Optical clock comparison for Lorentz symmetry testing[J]. Nature, 2019, 567(7747): 204-208.

[14] Grotti J, Koller S, Vogt S, et al. Geodesy and metrology with a transportable optical clock[J]. Nature Physics, 2018, 14(5): 437-441.

[15] Takamoto M, Tanaka Y, Katori H. A perspective on the future of transportable optical lattice clocks[J]. Applied Physics Letters, 2022, 120(14): 140502.

[16] Yin M J, Wang T, Lu X T, et al. Rabi spectroscopy and sensitivity of a Floquet engineered optical lattice clock[J]. Chinese Physics Letters, 2021, 38(7): 073201.

[17] Lu X T, Wang T, Li T, et al. Doubly modulated optical lattice clock: interference and topology[J]. Physical Review Letters, 2021, 127(3): 033601.

[18] Daley A J, Boyd M M, Ye J, et al. Quantum computing with alkaline-Earth-metal atoms[J]. Physical Review Letters, 2008, 101(17): 170504.

[19] Nunn J, Dorner U, Michelberger P, et al. Quantum memory in an optical lattice[J]. Physical Review A, 2010, 82(2): 022327.

[20] Ludlow A D, Boyd M M, Ye J, et al. Optical atomic clocks[J]. Reviews of Modern Physics, 2015, 87(2): 637-701.

[21] Katori H. Optical lattice clocks and quantum metrology [J]. Nature Photonics, 2011, 5(4): 203-210.

[22] Al-Masoudi A, Dörscher S, Häfner S, et al. Noise and instability of an optical lattice clock [J]. Physical Review A, 2015, 92(6): 063814.

[23] Dick G J. Local oscillator induced instabilities in trapped ion frequency standards[C]//Proceedings of the Nineteenth Annual Precise Time and Time Interval (PTTI) Applications and Planning Meeting, December 1-3, 1987, Redondo, Beach, CA. [S.l.: s.n.], 1987.

[24] Nagourney W, Sandberg J, Dehmelt H. Shelved optical electron amplifier: observation of quantum jumps[J]. Physical Review Letters, 1986, 56(26): 2797-2799.

[25] Keller J, Ignatovich S, Webster S A, et al. Simple vibration-insensitive cavity for laser stabilization at the 10^{-16} level[J]. Applied Physics B, 2014, 116(1): 203-210.

[26] Häfner S, Falke S, Grebing C, et al. 8×10^{-17} fractional laser frequency instability with a long room-temperature cavity[J]. Optics Letters, 2015, 40(9): 2112-2115.

[27] Xia J J, Lu X T, Chang H. Interrogation of optical Ramsey spectrum and stability study of an ^{87}Sr optical lattice clock[J]. Chinese Physics B, 2022, 31(3): 034209.

[28] Schioppo M, Brown R C, McGrew W F, et al. Ultrastable optical clock with two cold-atom ensembles [J]. Nature Photonics, 2017, 11(1): 48-52.

[29] Vallet G, Bookjans E, Eismann U, et al. A noise-immune cavity-assisted non-destructive detection for an optical lattice clock in the quantum regime[J]. New Journal of Physics, 2017, 19(8): 083002.

[30] Wineland D J, Monroe C, Itano W M, et al. Experimental issues in coherent quantum-state manipulation of trapped atomic ions[J]. Journal of Research of the National Institute of Standards and Technology, 1998, 103(3): 259-328.

[31] Arnold K, Hajiyev E, Paez E, et al. Prospects for atomic clocks based on large ion crystals[J]. Physical Review A, 2015, 92(3): 032108.

[32] Aharon N, Spethmann N, Leroux I D, et al. Robust optical clock transitions in trapped ions using dynamical decoupling[J]. New Journal of Physics, 2019, 21(8): 083040.

[33] Courtillot I, Quessada A, Kovacich R P, et al. Clock transition for a future optical frequency standard with trapped atoms[J]. Physical Review A, 2003, 68(3): 030501.

[34] Westergaard P G, Lodewyck J, Lorini L, et al. Lattice-induced frequency shifts in Sr optical lattice clocks at the 10^{-17} level[J]. Physical Review Letters, 2011, 106(21):

210801.

[35] Rey A M, Gorshkov A V, Kraus C V, et al. Probing many-body interactions in an optical lattice clock[J]. Annals of Physics, 2014, 340(1): 311-351.

[36] Bothwell T, Kedar D, Oelker E, et al. JILA SrI optical lattice clock with uncertainty of 2.0×10^{-18}[J]. Metrologia, 2019, 56(6): 065004.

[37] McGrew W F, Zhang X, Fasano R J, et al. Atomic clock performance enabling geodesy below the centimetre level[J]. Nature, 2018, 564(7734): 87-90.

[38] Golovizin A, Fedorova E, Tregubov D, et al. Inner-shell clock transition in atomic thulium with a small blackbody radiation shift[J]. Nature Communications, 2019, 10: 1724.

[39] de Sarlo L, Favier M, Tyumenev R, et al. A mercury optical lattice clock at LNE-SYRTE[J]. Journal of Physics: Conference Series, 2016, 723: 012017.

[40] Yamaguchi A, Safronova M S, Gibble K, et al. Narrow-line cooling and determination of the magic wavelength of Cd[J]. Physical Review Letters, 2019, 123(11): 113201.

[41] Kulosa A P, Fim D, Zipfel K H, et al. Towards a Mg lattice clock: observation of the $^1S_0$-$^3P_0$ transition and determination of the magic wavelength[J]. Physical Review Letters, 2015, 115(24): 240801.

[42] Ushijima I, Takamoto M, Das M, et al. Cryogenic optical lattice clocks[J]. Nature Photonics, 2015, 9(3): 185-189.

[43] Gao K L. The $^{40}Ca^+$ ion optical clock[J]. National Science Review, 2020, 7(12): 1799-1801.

[44] Jian B, Dubé P, Madej A A. Evaluation of the blackbody radiation shift uncertainty for NRC's strontium ion clock[C]//2016 IEEE International Frequency Control Symposium (IFCS), May 9-12, 2016, New Orleans, LA, USA. New York: IEEE Press, 2016.

[45] Huntemann N, Okhapkin M, Lipphardt B, et al. High-accuracy optical clock based on the octupole transition in $^{171}Yb^+$ [J]. Physical Review Letters, 2012, 108(9): 090801.

[46] Huntemann N, Sanner C, Lipphardt B, et al. Single-ion atomic clock with 3×10^{-18} systematic uncertainty[J]. Physical Review Letters, 2016, 116(6): 063001.

[47] Chou C W, Hume D B, Koelemeij J C, et al. Frequency comparison of two high-accuracy Al^+ optical clocks[J]. Physical Review Letters, 2010, 104(7): 070802.

[48] Brewer S M, Chen J S, Hankin A M, et al. $^{27}Al^+$ quantum-logic clock with a systematic uncertainty below 10^{-18}[J]. Physical Review Letters, 2019, 123(3): 033201.

[49] Rosenband T, Hume D B, Schmidt P O, et al. Frequency ratio of Al^+ and Hg^+ single-ion optical clocks; metrology at the 17th decimal place[J]. Science, 2008, 319(5871): 1808-1812.

[50] Ohtsubo N, Li Y, Nemitz N, et al. Frequency ratio of an $^{115}In^+$ ion clock and a ^{87}Sr optical lattice clock[J]. Optics Letters, 2020, 45(21): 5950-5953.

[51] Dehmelt H G. Proposed 10^{-14} $\delta\nu/\nu$ laser fluorescence spectroscopy on Tl^+ mono-ion oscillator[J]. Bulletin of the American Physical Society, 1973, 18: 1521.

[52] Diddams S A, Udem T, Bergquist J C, et al. An optical clock based on a single trapped $^{199}Hg^+$ ion[J]. Science, 2001, 293(5531): 825-828.

[53] Oskay W H, Diddams S A, Donley E A, et al. Single-atom optical clock with high accuracy[J]. Physical Review Letters, 2006, 97(2): 020801.

[54] Schmidt P O, Rosenband T, Langer C, et al. Spectroscopy using quantum logic[J]. Science, 2005, 309 (5735): 749-752.

[55] Cui K F, Chao S J, Sun C L, et al. Evaluation of the systematic shifts of a $^{40}Ca^+$—$^{27}Al^+$ optical clock[J]. The European Physical Journal D, 2022, 76(8): 140.

[56] Huang Y, Zhang B L, Zeng M Y, et al. Liquid-nitrogen-cooled Ca^+ optical clock with systematic uncertainty of 3×10^{-18}[J]. Physical Review Applied, 2022, 17(3): 034041.

[57] Wei W Z, Hao P, Ma Z Y, et al. Measurement and suppression of magnetic field noise of trapped ion qubit [J]. Journal of Physics B: Atomic, Molecular and Optical Physics, 2022, 55(7): 075001.

[58] Arnold K J, Kaewuam R, Roy A, et al. Blackbody radiation shift assessment for a lutetium ion clock[J]. Nature Communications, 2018, 9: 1650.

[59] Holliman C A, Fan M, Contractor A, et al. Radium ion optical clock[J]. Physical Review Letters, 2022, 128(3): 033202.

[60] King S A, Spieß L J, Micke P, et al. An optical atomic clock based on a highly charged ion[J]. Nature, 2022, 611 (7934): 43-47.

[61] Dubé P, Jian B, Vutha A C. Advances in the uncertainty evaluation of a 88Sr single ion optical clock[C]//2018 European Frequency and Time Forum (EFTF), April 10-12, 2018, Turin, Italy. New York: IEEE Press, 2018: 315-317.

[62] Dubé P, Madej A A, Shiner A, et al. $^{88}Sr^+$ single-ion optical clock with a stability approaching the quantum projection noise limit[J]. Physical Review A, 2015, 92 (4): 042119.

[63] Barwood G P, Huang G, Klein H A, et al. Agreement between two $^{88}Sr^+$ optical clocks to 4 parts in 10^{17}[J]. Physical Review A, 2014, 89(5): 050501.

[64] Katori H. Spectroscopy of strontium atoms in the lamb-dicke confinement[M]//Gill P. Frequency standards and metrology. Singapore: World Scientific, 2002: 323-330.

[65] Katori H, Takamoto M. Ultrastable optical clock with neutral atoms in an engineered light shift trap[J]. Physical Review Letters, 2003, 91(17): 173005.

[66] Takamoto M, Hong F L, Higashi R, et al. An optical lattice clock[J]. Nature, 2005, 435(7040): 321-324.

[67] Ludlow A D, Boyd M M, Zelevinsky T, et al. Systematic study of the ^{87}Sr clock transition in an optical lattice[J]. Physical Review Letters, 2006, 96(3): 033003.

[68] Le Targat R, Baillard X, Fouché M, et al. Accurate optical lattice clock with ^{87}Sr atoms[J]. Physical Review Letters, 2006, 97(13): 130801.

[69] Takamoto M, Hong F L, Higashi R, et al. Improved frequency measurement of a one-dimensional optical lattice clock with a spin-polarized fermionic ^{87}Sr isotope [J]. Journal of the Physical Society of Japan, 2006, 75 (10): 104302.

[70] Origlia S, Pramod M S, Schiller S, et al. Towards an optical clock for space: compact, high-performance optical lattice clock based on bosonic atoms[J]. Physical Review A, 2018, 98(5): 053443.

[71] Lemke N D, Ludlow A D, Barber Z W, et al. Spin-1/2 optical lattice clock[J]. Physical Review Letters, 2009, 103(6): 063001.

[72] McFerran J J, Yi L, Mejri S, et al. Neutral atom frequency reference in the deep ultraviolet with fractional uncertainty=5.7×10^{-15}[J]. Physical Review Letters, 2012, 108(18): 183004.

[73] Ludlow A D, Zelevinsky T, Campbell G K, et al. Sr lattice clock at 1×10^{-16} fractional uncertainty by remote optical evaluation with a Ca clock[J]. Science, 2008, 319(5871): 1805-1808.

[74] Hinkley N, Sherman J A, Phillips N B, et al. An atomic clock with 10^{-18} instability[J]. Science, 2013, 341(6151): 1215-1218.

[75] Bloom B J, Nicholson T L, Williams J R, et al. An optical lattice clock with accuracy and stability at the 10^{-18} level[J]. Nature, 2014, 506(7486): 71-75.

[76] Nicholson T L, Campbell S L, Hutson R B, et al. Systematic evaluation of an atomic clock at 2×10^{-18} total uncertainty[J]. Nature Communications, 2015, 6: 6896.

[77] Brown R C, Phillips N B, Beloy K, et al. Hyperpolarizability and operational magic wavelength in an optical lattice clock[J]. Physical Review Letters, 2017, 119(25): 253001.

[78] Ushijima I, Takamoto M, Katori H. Operational magic intensity for Sr optical lattice clocks[J]. Physical Review Letters, 2018, 121(26): 263202.

[79] Oelker E, Hutson R B, Kennedy C J, et al. Demonstration of 4.8×10^{-17} stability at 1 s for two independent optical clocks[J]. Nature Photonics, 2019, 13(10): 714-719.

[80] Beloy K, Bodine M I, Bothwell T, et al. Frequency ratio measurements at 18-digit accuracy using an optical clock network[J]. Nature, 2021, 591(7851): 564-569.

[81] Guo F, Tan W, Zhou C H, et al. A proof-of-concept model of compact and high-performance ^{87}Sr optical lattice clock for space[J]. AIP Advances, 2021, 11(12): 125116.

[82] Aeppli A, Chu A J, Bothwell T, et al. Hamiltonian engineering of spin-orbit-coupled fermions in a Wannier-Stark optical lattice clock[J]. Science Advances, 2022, 8(41): eadc9242.

[83] Schwarz R, Dörscher S, Al-Masoudi A, et al. Long term measurement of the ^{87}Sr clock frequency at the limit of primary Cs clocks[J]. Physical Review Research, 2020, 2(3): 033242.

[84] Ohmae N, Bregolin F, Nemitz N, et al. Direct measurement of the frequency ratio for Hg and Yb optical lattice clocks and closure of the Hg/Yb/Sr loop[J]. Optics Express, 2020, 28(10): 15112-15121.

[85] Tyumenev R, Favier M, Bilicki S, et al. Comparing a mercury optical lattice clock with microwave and optical frequency standards[J]. New Journal of Physics, 2016, 18(11): 113002.

[86] Luo L M, Qiao H, Ai D, et al. Absolute frequency measurement of an Yb optical clock at the 10^{-16} level using international atomic time[J]. Metrologia, 2020, 57(6): 065017.

[87] Zhang A, Xiong Z X, Chen X T, et al. Ytterbium optical lattice clock with instability of order 10^{-18}[J]. Metrologia, 2022, 59(6): 065009.

[88] Lu B K, Sun Z, Yang T, et al. Improved evaluation of BBR and collisional frequency shifts of NIM-Sr2 with 7.2×10^{-18} total uncertainty[J]. Chinese Physics Letters, 2022, 39(8): 080601.

[89] Lu X T, Guo F, Wang Y B, et al. Absolute frequency measurement of the ^{87}Sr optical lattice clock at NTSC using International Atomic Time[EB/OL]. (2022-11-08)[2022-12-02]. https://arxiv.org/abs/2211.04096.

[90] Takamoto M, Takano T, Katori H. Frequency comparison of optical lattice clocks beyond the Dick limit[J]. Nature Photonics, 2011, 5(5): 288-292.

[91] Nemitz N, Ohkubo T, Takamoto M, et al. Frequency ratio of Yb and Sr clocks with 5×10^{-17} uncertainty at 150 seconds averaging time[J]. Nature Photonics, 2016, 10(4): 258-261.

[92] Le Targat R, Lorini L, Le Coq Y, et al. Experimental realization of an optical second with strontium lattice clocks[J]. Nature Communications, 2013, 4: 2109.

[93] Campbell G K, Ludlow A D, Blatt S, et al. The absolute frequency of the ^{87}Sr optical clock transition[J]. Metrologia, 2008, 45(5): 539-548.

[94] Lodewyck J, Bilicki S, Bookjans E, et al. Optical to microwave clock frequency ratios with a nearly continuous strontium optical lattice clock[J]. Metrologia, 2016, 53(4): 1123-1130.

[95] Yamaguchi A, Shiga N, Nagano S, et al. Stability transfer between two clock lasers operating at different wavelengths for absolute frequency measurement of clock transition in ^{87}Sr[J]. Applied Physics Express, 2012, 5(2): 022701.

[96] Matsubara K, Hachisu H, Li Y, et al. Direct comparison of a Ca^{+} single-ion clock against a Sr lattice clock to verify the absolute frequency measurement[J]. Optics Express, 2012, 20(20): 22034-22041.

[97] Hachisu H, Petit G, Ido T. Absolute frequency measurement with uncertainty below 1×10^{-15} using international atomic time[J]. Applied Physics B, 2017, 123(1): 34.

[98] Hachisu H, Petit G, Nakagawa F, et al. SI-traceable measurement of an optical frequency at the low 10^{-16} level without a local primary standard[J]. Optics Express, 2017, 25(8): 8511-8523.

[99] Akamatsu D, Inaba H, Hosaka K, et al. Spectroscopy and frequency measurement of the ^{87}Sr clock transition by laser linewidth transfer using an optical frequency comb[J]. Applied Physics Express, 2014, 7(1): 012401.

[100] Tanabe T, Akamatsu D, Kobayashi T, et al. Improved frequency measurement of the $^1S_0-^3P_0$ clock transition in ^{87}Sr using a Cs fountain clock as a transfer oscillator[J]. Journal of the Physical Society of Japan, 2015, 84(11): 115002.

[101] Falke S, Schnatz H, Winfred J S R V, et al. The ^{87}Sr optical frequency standard at PTB[J]. Metrologia, 2011, 48(5): 399-407.

[102] Grebing C, Al-Masoudi A, Dörscher S, et al. Realization of a timescale with an accurate optical lattice clock[J]. Optica, 2016, 3(6): 563-569.

[103] Falke S, Lemke N, Grebing C, et al. A strontium lattice clock with 3×10^{-17} inaccuracy and its frequency[J]. New Journal of Physics, 2014, 16(7): 073023.

[104] Lin Y G, Wang Q, Li Y, et al. First evaluation and frequency measurement of the strontium optical lattice clock at NIM[J]. Chinese Physics Letters, 2015, 32(9): 090601.

[105] Lin Y G, Wang Q, Meng F, et al. A ^{87}Sr optical lattice clock with 2.9×10^{-17} uncertainty and its absolute frequency measurement[J]. Metrologia, 2021, 58(3): 035010.

[106] Hobson R, Bowden W, Vianello A, et al. A strontium optical lattice clock with 1×10^{-17} uncertainty and measurement of its absolute frequency[J]. Metrologia, 2020, 57(6): 065026.

[107] Hong F L, Musha M, Takamoto M, et al. Measuring the frequency of a Sr optical lattice clock using a 120 km coherent optical transfer[J]. Optics Letters, 2009, 34(5): 692-694.

[108] Park C Y, Yu D H, Lee W K, et al. Absolute frequency measurement of $^{1}\mathrm{S}_0(F=1/2)-{}^{3}\mathrm{P}_0(F=1/2)$ transition of ^{171}Yb atoms in a one-dimensional optical lattice at KRISS [J]. Metrologia, 2013, 50(2): 119-128.

[109] Kim H, Heo M S, Lee W K, et al. Improved absolute frequency measurement of the ^{171}Yb optical lattice clock at KRISS relative to the SI second[J]. Japanese Journal of Applied Physics, 2017, 56(5): 050302.

[110] Fujieda M, Yang S H, Gotoh T, et al. Advanced satellite-based frequency transfer at the 10^{-16} level[J]. IEEE Transactions on Ultrasonics, Ferroelectrics, and Frequency Control, 2018, 65(6): 973-978.

[111] Kim H D, Heo M S, Park C Y, et al. Absolute frequency measurement of the ^{171}Yb optical lattice clock at KRISS using TAI for over a year[J]. Metrologia, 2021, 58(5): 055007.

[112] Pizzocaro M, Thoumany P, Rauf B, et al. Absolute frequency measurement of the $^{1}\mathrm{S}_0-{}^{3}\mathrm{P}_0$ transition of ^{171}Yb [J]. Metrologia, 2017, 54(1): 102-112.

[113] Pizzocaro M, Bregolin F, Barbieri P, et al. Absolute frequency measurement of the $^{1}\mathrm{S}_0-{}^{3}\mathrm{P}_0$ transition of ^{171}Yb with a link to international atomic time[J]. Metrologia, 2020, 57(3): 035007.

[114] Pizzocaro M, Sekido M, Takefuji K, et al. Intercontinental comparison of optical atomic clocks through very long baseline interferometry[J]. Nature Physics, 2021, 17(2): 223-227.

[115] Takamoto M, Ushijima I, Das M, et al. Frequency ratios of Sr, Yb, and Hg based optical lattice clocks and their applications[J]. Comptes Rendus Physique, 2015, 16(5): 489-498.

[116] Kohno T, Yasuda M, Hosaka K, et al. One-dimensional optical lattice clock with a fermionic ^{171}Yb isotope[J]. Applied Physics Express, 2009, 2(7): 072501.

[117] Yasuda M, Inaba H, Kohno T, et al. Improved absolute frequency measurement of the ^{171}Yb optical lattice clock towards a candidate for the redefinition of the second[J]. Applied Physics Express, 2012, 5(10): 102401.

[118] Akamatsu D, Yasuda M, Inaba H, et al. Frequency ratio measurement of ^{171}Yb and ^{87}Sr optical lattice clocks [J]. Optics Express, 2014, 22(7): 7898-7905.

[119] Akamatsu D, Kobayashi T, Hisai Y, et al. Dual-mode operation of an optical lattice clock using strontium and ytterbium atoms[J]. IEEE Transactions on Ultrasonics, Ferroelectrics, and Frequency Control, 2018, 65(6): 1069-1075.

[120] Kobayashi T, Akamatsu D, Hosaka K, et al. Demonstration of the nearly continuous operation of an ^{171}Yb optical lattice clock for half a year[J]. Metrologia, 2020, 57(6): 065021.

[121] Hisai Y, Akamatsu D, Kobayashi T, et al. Improved frequency ratio measurement with ^{87}Sr and ^{171}Yb optical lattice clocks at NMIJ[J]. Metrologia, 2021, 58(1): 015008.

[122] Stenger J, Tamm C, Haverkamp N, et al. Absolute frequency measurement of the 435.5-nm $^{171}\mathrm{Yb}^{+}$-clock transition with a Kerr-lens mode-locked femtosecond laser [J]. Optics Letters, 2001, 26(20): 1589-1591.

[123] Tamm C, Lipphardt B, Schnatz H, et al. $^{171}\mathrm{Yb}^{+}$ single-ion optical frequency standard at 688 THz[J]. IEEE Transactions on Instrumentation and Measurement, 2007, 56(2): 601-604.

[124] Sherstov I, Tamm C, Stein B, et al. $^{171}\mathrm{Yb}^{+}$ single-ion optical frequency standards[C]//2007 IEEE International Frequency Control Symposium Joint with the 21st European Frequency and Time Forum, May 29-June 1, 2007, Geneva, Switzerland. New York: IEEE Press, 2007: 405-406.

[125] Tamm C, Weyers S, Lipphardt B, et al. Stray-field-induced quadrupole shift and absolute frequency of the 688-THz $^{171}\mathrm{Yb}^{+}$ single-ion optical frequency standard[J]. Physical Review A, 2009, 80(4): 043403.

[126] Tamm C, Huntemann N, Lipphardt B, et al. Cs-based optical frequency measurement using cross-linked optical and microwave oscillators[J]. Physical Review A, 2014, 89(2): 023820.

[127] Lange R, Huntemann N, Rahm J M, et al. Improved limits for violations of local position invariance from atomic clock comparisons[J]. Physical Review Letters, 2021, 126(1): 011102.

[128] Webster S, Godun R, King S, et al. Frequency measurement of the $^{2}\mathrm{S}_{1/2}-{}^{2}\mathrm{D}_{3/2}$ electric quadrupole transition in a single $^{171}\mathrm{Yb}^{+}$ ion[J]. IEEE Transactions on Ultrasonics, Ferroelectrics, and Frequency Control, 2010, 57(3): 592-599.

[129] Dörscher S, Huntemann N, Schwarz R, et al. Optical frequency ratio of a $^{171}\mathrm{Yb}^{+}$ single-ion clock and a ^{87}Sr lattice clock[J]. Metrologia, 2021, 58(1): 015005.

[130] Hosaka K, Webster S A, Blythe P J, et al. An optical frequency standard based on the electric octupole transition in $^{171}\mathrm{Yb}^{+}$ [J]. IEEE Transactions on Instrumentation and Measurement, 2005, 54(2): 759-

762.

[131] Webster S A, Hosaka K, Gill P, et al. Precision frequency measurement of the $^2S_{1/2}-^2F_7/_2$ electric octupole transition in a single $^{171}Yb^+$ ion[C]//Conference on Precision Electromagnetic Measurements Digest, June 8-13, 2008, Broomfield, CO, USA. [S.l.: s.n.], 2008: 96-97.

[132] King S A, Godun R M, Webster S A, et al. Absolute frequency measurement of the $^2S_{1/2}-^2F_{7/2}$ electric octupole transition in a single ion of $^{171}Yb^+$ with 10^{-15} fractional uncertainty[J]. New Journal of Physics, 2012, 14(1): 013045.

[133] Baynham C F A, Godun R M, Jones J M, et al. Absolute frequency measurement of the $^2S_{1/2}-^2F_{7/2}$ optical clock transition in $^{171}Yb^+$ with an uncertainty of 4×10^{-16} using a frequency link to international atomic time [J]. Journal of Modern Optics, 2018, 65(5/6): 585-591.

[134] Matsubara K, Li Y, Nagano S, et al. Absolute frequency measurement of the 40Ca clock transition using a LD-based clock laser and UTC(NICT)[C]//2009 IEEE International Frequency Control Symposium Joint with the 22nd European Frequency and Time forum, April 20-24, 2009, Besancon, France. New York: IEEE Press, 2009: 751-755.

[135] Chwalla M, Benhelm J, Kim K, et al. Absolute frequency measurement of the $^{40}Ca^+$ 4s $^2S_{1/2}-$3d $^2D_{5/2}$ clock transition[J]. Physical Review Letters, 2009, 102 (2): 023002.

[136] Huang Y, Cao J, Liu P, et al. Hertz-level measurement of the $^{40}Ca^+$ 4s $^2S_{1/2}-$3d $^2D_{5/2}$ clock transition frequency with respect to the SI second through the Global Positioning System[J]. Physical Review A, 2012, 85(3): 030503.

[137] Huang Y, Guan H, Liu P, et al. Frequency comparison of two $^{40}Ca^+$ optical clocks with an uncertainty at the 10^{-17} level[J]. Physical Review Letters, 2016, 116(1): 013001.

[138] Huang Y, Zhang H Q, Gao K, et al. Geopotential measurement with a robust, transportable Ca^+ optical clock [J]. Physical Review A, 2020, 102(5): 050802.

[139] 张华青. 车载钙离子光钟的研制与应用研究[D]. 武汉: 中国科学院精密测量科学技术创新研究院, 2022: 124-125. Zhang H Q. Development and application of vehicular $^{40}Ca^+$ ion optical clock[D]. Wuhan: Innovation Academy for Precision Measurement Science and Technology, Chinese Academy of Science, 2022: 124-125.

[140] Baillard X, Fouché M, Le Targat R, et al. Accuracy evaluation of an optical lattice clock with bosonic atoms [J]. Optics Letters, 2007, 32(13): 1812-1814.

[141] Akatsuka T, Takamoto M, Katori H. Optical lattice clocks with non-interacting bosons and fermions[J]. Nature Physics, 2008, 4(12): 954-959.

[142] Takano T, Mizushima R, Katori H. Precise determination of the isotope shift of $^{88}Sr-^{87}Sr$ optical lattice clock by sharing perturbations[J]. Applied Physics Express, 2017, 10(7): 072801.

[143] Morzyński P, Bober M, Bartoszek-Bober D, et al. Absolute measurement of the $^1S_0-^3P_0$ clock transition in neutral ^{88}Sr over the 330 km-long stabilized fibre optic link [J]. Scientific Reports, 2015, 5: 17495.

[144] Radzewicz C, Bober M, Morzyński P, et al. Accuracy budget of the ^{88}Sr optical atomic clocks at KL FAMO[J]. Physica Scripta, 2016, 91(8): 084003.

[145] Madej A A, Bernard J E, Dubé P, and Marmet L. Absolute frequency of the $^{88}Sr^+$ 5s $^2S_{1/2}-$4d $^2D_{5/2}$ reference transition at 445 THz and evaluation of systematic shifts[J]. Physical Review A, 2004, 70(1): 012507.

[146] Dubé P, Madej A A, Bernard J E, et al. Electric quadrupole shift cancellation in single-ion optical frequency standards[J]. Physical Review Letters, 2005, 95(3): 033001.

[147] Madej A A, Dubé P, Zhou Z C, et al. $^{88}Sr^+$ 445-THz single-ion reference at the 10^{-17} level via control and cancellation of systematic uncertainties and its measurement against the SI second[J]. Physical Review Letters, 2012, 109(20): 203002.

[148] Dubé P, Bernard J E, Gertsvolf M. Absolute frequency measurement of the $^{88}Sr^+$ clock transition using a GPS link to the SI second[J]. Metrologia, 2017, 54(3): 290-298.

[149] Margolis H S, Barwood G P, Huang G, et al. Hertz-level measurement of the optical clock frequency in a single $^{88}Sr^+$ ion[J]. Science, 2004, 306(5700): 1355-1358.

[150] Rosenband T, Schmidt P, Hume D, et al. Observation of the $^1S_0\rightarrow{}^3P_0$ clock transition in $^{27}Al^+$ [J]. Physical Review Letters, 2007, 98(22): 220801.

[151] Leopardi H, Beloy K, Bothwell T, et al. Measurement of the $^{27}Al^+$ and ^{87}Sr absolute optical frequencies[J]. Metrologia, 2021, 58(1): 015017.

[152] Yamanaka K, Ohmae N, Ushijima I, et al. Frequency ratio of ^{199}Hg and ^{87}Sr optical lattice clocks beyond the SI limit[J]. Physical Review Letters, 2015, 114(23): 230801.

[153] Stalnaker J E, Diddams S A, Fortier T M, et al. Optical-to-microwave frequency comparison with fractional uncertainty of 10^{-15}[J]. Applied Physics B, 2007, 89(2): 167-176.

[154] Recommended values of standard frequencies[EB/OL]. [2022-10-02]. https://www.bipm.org/en/publications/mises-en-pratique/standard-frequencies-second.

[155] CGPM. 27th CGPM: key outcomes and documents[EB/OL]. [2022-10-02]. https://www.bipm.org/en/cgpm-2022/documents.

[156] Schiller S, Görlitz A, Nevsky A, et al. Optical clocks in space[J]. Nuclear Physics B-Proceedings Supplements, 2007, 166: 300-302.

[157] Koyama Y. The use of very long baseline interferometry for time and frequency metrology[J]. MAPAN, 2012, 27 (1): 23-30.

[158] Bongs K, Singh Y, Smith L, et al. Development of a strontium optical lattice clock for the SOC mission on the ISS[J]. Comptes Rendus Physique, 2015, 16(5): 553-564.

[159] DARPA's ROCkN program aims to create portable optical atomic clocks[EB/OL]. [2022-10-02]. https://www.inceptivemind.com/darpas-rockn-program-aims-create-portable-optical-atomic-clocks/23132/.