REVIEW

National Science Review
3: 189–200, 2016
doi: 10.1093/nsr/nww013
Advance access publication 15 March 2016

PHYSICS

Special Topic: Cold Atoms

Precision measurement and frequency metrology with ultracold atoms

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ABSTRACT

Precision measurement and frequency metrology have pushed many scientific and technological frontiers in the field of atomic, molecular and optical physics. In this article, we provide a brief review on the recent development of optical atomic clocks, with an emphasis placed on the important inter-dependence between measurement precision and systematic effects. After presenting a general discussion on the motivation and techniques behind the development of optical lattice clocks, where the use of many atoms greatly enhances the measurement precision, we present the JILA strontium optical lattice clock as the leading system of frequency metrology with the lowest total uncertainty, and we describe other related research activities. We discuss key ingredients that have enabled the optical lattice clocks with ultracold atoms to reach the 18th digit in both precision and accuracy. Furthermore, we discuss extending the power of precision clock spectroscopy to study quantum many-body physics and to provide control for atomic quantum materials. In addition, we explore future research directions that have the potential to achieve even greater precision.

Keywords: ultracold atoms, optical atomic clocks, precision measurement, frequency metrology, many-body physics, quantum state control

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Received 28 December 2015; **Revised** 24 February 2016; **Accepted** 25 February 2016

INTRODUCTION: PRECISION FREQUENCY MEASUREMENT

Measurement is the foundation for physical science. In the field of atomic, molecular and optical physics, precision measurement has provided important tests of fundamental laws of physics, stimulated the development of new frontiers in science and technology, and seeded a wide range of revolutionary applications that bring important benefits to our society, including the realization and distribution of SI units [1], development of quantum-based measurements [2], and enhanced sensor technologies for medical science [3]. The initial work on cold atoms was strongly motivated by the scientific goal of achieving much higher measurement precision, which has in turn stimulated the ensuing rapid progress in research on cold atoms.

Improving measurement precision by orders of magnitude can lead to numerous discoveries in modern physics, including the test of constancy for the speed of light [4], early triumph of quantum electrodynamics via microwave spectroscopy of the fine structure of a hydrogen atom [5], and the recent exciting direct observation of gravitational waves [6]. Among all physical observables, frequency is the most accurately measured quantity and it hence deserves a special treatment [7,8]. The international standard (SI) of the second, our unit of time, is defined based on the microwave clock transition between two hyperfine ground states of cesium-133 atoms and this standard has been steadily improved over the past 50 years [9]. Over the last decade, however, tremendous progress has been made in achieving a total measurement uncertainty as low as 2×10^{-18} for frequency determination on the basis of optical atomic clocks [10]. In this article, we will discuss exclusively the precise and accurate measurement of optical frequencies: counting the number of optical cycles during a given time interval.

The most common form of a frequency standard or a clock comes in a passive configuration: it possesses a periodic oscillation that can be specifically initialized and then precisely probed. The periodic motion can be referred to as 'the pendulum', which

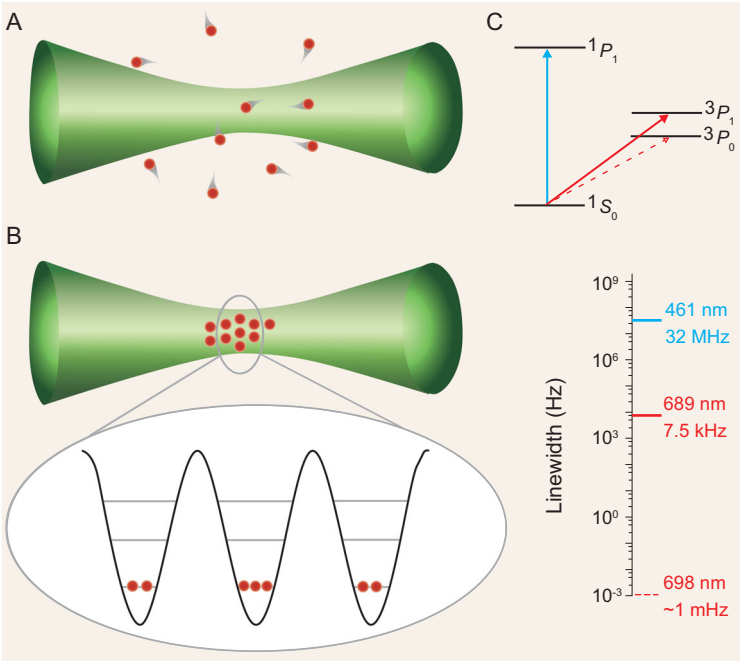

Figure 1. Decoupling the motional and electronic degrees of freedom. (A) Illustration of probing a hot and free atomic sample. The green Gaussian tube denotes the probe laser. When the atoms freely move around and their temperature is high, the Doppler shift, limited observation time, and various dephasing processes make it hard to perform precision measurements. (B) With cold and trapped atoms, the Doppler shift and dephasing is greatly reduced and the atoms can be probed for much longer time. Here, the atoms are tightly confined along the probe laser direction in the ground states of an optical lattice potential. This strong lattice confinement further decouples the residual motional disturbance from the internal, electronic degree of freedom (the clock transition). (C) Energy level diagram of strontium atoms. Three transitions are illustrated: thick blue line, a broad (32 MHz natural linewidth) blue transition at 461 nm for a first stage of magneto-optical trapping, cooling atoms to millikelvin temperatures; thin red line, a much narrower (7.5 kHz) red transition at 689 nm for a second stage of magneto-optical trapping, cooling atoms to about microkelvin temperatures; and dashed red line, an ultranarrow clock transition at 698 nm, which corresponds to one of the highest-quality-factor (the intrinsic $Q \sim 4 \times 10^{17}$) oscillators.

provides an ideally high-quality stable frequency reference. We drive this pendulum with the help of a local oscillator (LO), which is adjusted to be as close to the pendulum's natural frequency as possible. The LO should have good short-term stability to minimize the measurement noise, and the pendulum provides long-term frequency reproducibility and accuracy.

Historically, the pendulum has come in a variety of shapes and forms, such as celestial motions, macroscopic mechanical clocks, quartz crystal resonators [11] and, starting in the middle of the 20th century, microscopic atomic or molecular samples [9]. Neutral atoms, molecules, and their ion counterparts (henceforth atoms) hold promise for providing an ideal frequency reference because a properly prepared atom can be fully described with a two-level quantum system [12] and the intrinsic natural frequency of one atom is identical to any other atoms of the same species. However, to realize the full potential of an atomic frequency standard, the samples must be prepared with care; to increase the measurement precision and accuracy it became clear that atoms need to be cooled to sufficiently low temperatures.

Moving atoms limit spectroscopy investigations in several ways. First, the resonance frequency of each atom is shifted inhomogeneously via the Doppler effect. While advanced Doppler-insensitive spectroscopy methods have been developed [7,13], the remaining first order Doppler effect can still contribute to dephasing, while the second order effect is a significant concern for modern atomic clocks. Second, at room temperature atoms fly through a typical probing field in a rather short time ($<10^{-4}$ s), limiting the corresponding spectroscopic resolution. Third, the moving atoms often have complicated scenarios for collisions, resulting in the reduction of coherence time and the introduction of frequency shifts. These limitations are schematically illustrated in Fig. 1A.

The desire for better control of atomic motion for spectroscopy led to the development of laser cooling of atoms in the early 1980s [14,15]. As a result, atoms cooled to μK or lower temperatures ('ultracold atoms') have become widely available and precision measurements can be performed with the effect of atomic center-of-mass motion well accounted for in the spectroscopy budget, as illustrated in Fig. 1B. Working with optical atomic clocks today, we aim for the goal of preserving the longest possible coherence time between atomic states and an optical field. To accomplish this, we need to understand and control the atomic center-of-mass motion at the scale of an optical wavelength. Another challenge stems from the fact that optical probing of internal atomic states creates an inevitable back-action on the atomic center-of-mass motion. Such back-action limits measurement precision and control. Meeting these challenges requires preparing atoms at ultralow temperatures and confining them in specially engineered optical, radio frequency, or magnetic traps that decouple the atomic internal and external degrees of freedom for spectroscopy. Such a separation of internal and external dynamics is critical for precision measurement, frequency metrology, coherent manipulation of quantum systems, and quantum information science, as shown in the lower part of Fig. 1B.

Fortunately for the future prospects of precision measurement, there has been tremendous progress in research on ultracold atomic gases. Atoms can now be brought to move so slowly with a speed of mere 10^{-3}–10^{-2} m/s or be confined in external traps where atomic motions are cooled to the ground

state, such that the gas exhibits striking quantum behavior. Coherent matter waves in the form of quantum degenerate gases can now be routinely prepared, manipulated, and measured in the laboratory [16,17]. With important tools, such as the control of atomic interactions via Feshbach resonances and the introduction of optical lattices to regulate atomic motion, atomic systems are now prepared to take on challenging and outstanding problems including the study of strongly correlated quantum many-body systems and their dynamics. Here lie some tremendous opportunities to advance the field of precision measurement.

Using the geometry of atomic fountains based on ultracold cesium atoms, microwave atomic clocks continue to serve the scientific community by providing the SI definition of time at the 10^{-16} level. At the same time, atomic clock transitions are no longer confined to the microwave domain because optical frequency combs now provide a coherent bridge between optical and radio frequency spectral domains [7,8]. This coherent bridge allows us to explore optical transitions that have orders-of-magnitude-higher line quality. With laser cooling one can localize atoms within a fraction of optical wavelength inside a deeply bound trap, thus minimizing atomic motional effects on optical spectroscopy and state control. Furthermore, the quantized atomic motion and long interrogation times inside the trap permit high-resolution and precise investigations. Such capabilities have long been used in trapped-ion systems where charged ions are bound by external Lorentz forces with minimal perturbations to their electronic states. Indeed, the first optical atomic clock was demonstrated based on a single trapped $^{199}Hg^{+}$ ion [18]. It was soon followed by a single-ion optical clock based on an Al^{+} ion with a total uncertainty in fractional frequency of 8.6×10^{-18} [19], which was a record low at the time and more than one order of magnitude lower than that for the cesium clock (low numbers of 10^{-16}).

In this article, we will focus on the current champion for having the lowest total fractional frequency uncertainty, the JILA strontium optical lattice clock [20,21], and describe other related works and the context. Protocols for precision measurement have required large ensembles of neutral atoms to boost the measurement signal-to-noise ratio, explore atomic collective effects, or create a massively entangled system. Measurement precision and accuracy are illustrated schematically in Fig. 2. Typically, an enhanced measurement precision leads directly to enhanced accuracy; however, a carefully designed experimental system can help reduce systematic uncertainties from the first principles. For neutral atoms, external trapping potentials are created

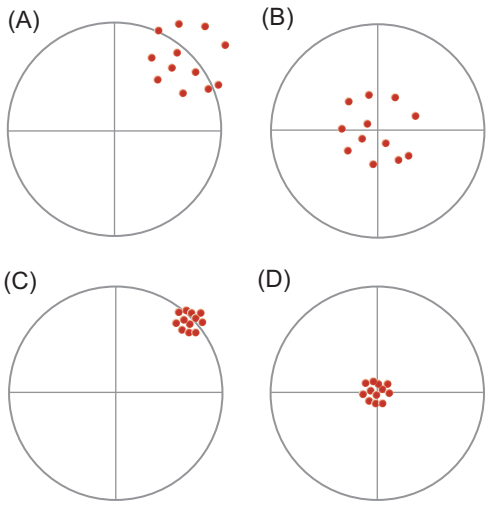

Figure 2. Illustration of measurement precision and accuracy (total uncertainty). (A) The measurement is neither precise nor accurate. (B) Accurate but not precise. (C) Precise but not accurate. (D) Both precise and accurate.

from spatially inhomogeneous energy shifts of the electronic states produced by an applied magnetic, electric, or optical field. It is thus crucial to construct atom traps that do not perturb the *in situ* precision spectroscopy of atomic transitions. Recently, our group became capable of controlling the quantum states of more than 1000 atoms so precisely that we achieved a more accurate and more precise atomic clock than any other existing atomic clocks. With the clock accuracy and stability both reaching the 10^{-18} level, our group has realized a single atomic clock with the top performance in both key ingredients necessary for a primary standard. This superior measurement precision has also enabled researchers to study complex quantum many-body systems and integrate them into the frontiers of precision metrology, where we seek to advance measurement beyond the standard quantum limit. Such advanced clocks will allow us to test the fundamental laws of nature and find applications among a wide range of technological frontiers.

The remaining sections are organized as follows. We first discuss the optical lattice clock based on fermionic strontium-87 (^{87}Sr) atoms, of which the 1 mHz-wide $^{1}S_0 \rightarrow {}^{3}P_0$ transition frequency has been recommended as a secondary representation of the SI second [22], and illustrate some key aspects that enable such a clock to reach 10^{-18}-level precision and total uncertainty. In the following section, we review some recent examples for extending the power of clock precision to study atomic interactions and quantum many-body physics. We also discuss using precision spectroscopy tools as precise control of the internal degrees of freedom of atoms,

as well as similar tools associated with other degrees of freedom. In the final section, we explore some research directions that have the potential to achieve greater precision in the future.

NEUTRAL ATOM ENSEMBLE OPTICAL LATTICE CLOCKS

Choosing nature's high-*Q* pendulum: alkaline earth(-like) elements

Important criteria for an atomic frequency reference include having a high quality factor (Q) for the clock transition with its center frequency, and having a minimal sensitivity to environmental perturbations. The neutral alkaline earth(-like) atoms are excellent clock species as they possess the $^1S_0 \rightarrow {}^3P_0$ transition (Fig. 1C). The superior property of $^1S_0 \rightarrow {}^3P_0$ as a clock transition was first discussed by Hans Dehmelt for Al^+ ions [23]. Because the electronic angular momentum is zero, the transition frequency is 10 000 times less sensitive to environmental perturbations such as magnetic field fluctuation (10^{-3} to 10^{-2} gauss) compared to alkali metal atoms such as cesium. The scalar nature of both electronic states also facilitates an easy characterization and more precise control of optical dipole traps that are used to confine atoms. On the other hand, a finite nuclear spin and the corresponding hyperfine interactions induce a tiny admixture of the bare $^3P_0$ state with higher lying P states, leading to a finite, but very narrow clock transition of 1 mHz natural linewidth for ^{87}Sr [24,25]. The resulting Q for the clock transition is 4×10^{17}, and yet the linewidth is still reasonably large that the best frequency-stabilized laser can efficiently interrogate this transition [24]. Of course, many alkaline earth(-like) atoms, such as Mg, Ca, Sr, Yb, Hg, Al^+, In^+, and others, share this attractive property of a high-quality clock transition. For example, ^{171}Yb has a Q value of about 5×10^{16}. However, ^{87}Sr exhibits one of the longest $^3P_0$ lifetimes. This will become an important factor in the near future when quantum entanglement becomes part of the metrology pursuit for improved precision [26,27]. In this article, we focus on the JILA Sr clock that has realized to date 2×10^{-18} total uncertainty, the lowest among all atomic clocks [21].

Decoupling the internal and external degrees of freedom: spectroscopy of strongly confined atoms in optical lattices

Even though two-stage laser cooling via convenient MHz- and kHz-wide transitions (Fig. 1C) very efficiently reduces the atomic temperature to about 1 μK [28,29], residual Doppler shift and other systematic effects limit the total uncertainty of free-space optical frequency standards to the 10^{-14} to 10^{-15} level [10]. If both the ground and excited clock states can be strongly confined in a single optical trap, and the trap provides the same AC Stark shifts for both states, then interrogating the clock transition inside the trap can potentially be free of trap perturbations. Optical lattices provide an excellent platform for strongly confining the atoms (Fig. 1B). Operating with a narrow clock transition, it is straightforward to reach the resolved-sideband regime [30,31] where the Doppler effects are relegated to motional sidebands far detuned from the carrier of the clock transition. By increasing the trap frequency above the single photon recoil frequency, one reaches the Lamb-Dicke regime [32] where excitation of the motional sideband is further suppressed because the strong optical potential, rather than the atom, takes up the recoil momentum from an absorbed photon [33]. One thus enters an ideal regime for optical spectroscopy where the probed transition frequency is free of Doppler and recoil effects [10].

The idea of working with atomic states of similar polarizabilities (or AC Stark shifts) in an optical trap was first discussed in 1994 [34]. The magic wavelength idea of exactly zero differential AC Stark shift was explicitly proposed in 1999; see section entitled 'A zero transition-shift FORT' in Ref. [35]. The first proposal on the use of the $^1S_0 \rightarrow {}^3P_0$ as a clock transition in neutral atoms trapped by an optical lattice at the magic wavelength was made in Ref. [36]. These concepts of a well-engineered trapping potential for clock operations [37] have led to rapid experimental progress in optical lattice clocks [38–40].

Measuring for 6 or 600 months: building a 10× more stable local oscillator

A good LO is an integral part of a clock that aims for top performance. Ideally, the coherence time of the LO should match that for the atomic quantum state superposition. This would allow one to take full advantage of the high quality factor of the atomic transition for the maximum measurement precision and clock stability. However, in practice the best laser with the highest frequency stability is still about a factor of 10 shorter in its coherence time than that for the Sr clock states, thus limiting the time duration for a single, coherent clock measurement. To make matters worse, atomic clocks are operated in a pulsed mode, where a certain amount of time has to be invested to laser cool atoms and trap them in an optical lattice for the initial state preparation. The actual clock operation when

the LO frequency is checked against the atomic resonance takes only a certain fraction of the total clock cycle. Thus, excessive noise from the LO will be introduced to the clock measurement through an aliasing process known as the Dick effect [41,42], further decreasing the clock stability.

For an optical lattice clock, the participation of thousands of atoms for measurement has the potential for unprecedented clock precision and stability as the fundamental limit to the clock state determination is set by quantum projection, which scales with the square root of the particle number. For example, it is anticipated that such a clock may one day deliver instability of 1×10^{-17} at 1 s. However, up till 2012 the lattice clock stability was basically the same as that for single trapped ion clocks. This clock instability (2×10^{-15} at 1 s; see [43]) was all due to the limited stability (1×10^{-15}) of the clock laser that introduced additional sampling noise arising from the Dick effect [44]. Thus, after Ludlow *et al.* demonstrated that the accuracy of the JILA Sr clock surpassed that of the Cs primary standard in 2008 [43], our group started making dramatic improvement to its stable laser technology. One of the approaches our group took was to construct a new ultrastable optical reference cavity based on a 40-cm long ultralow expansion glass spacer, striking a good balance between the thermal noise contribution and the vibration noise sensitivity for the net cavity fractional frequency stability [45,46]. This LO system eventually demonstrated an optical coherence time of 20 s [47] and has allowed us to extend the clock interrogation time to beyond 1 s, limited by the atomic interactions [48]. Some examples of the typical narrow lineshapes recorded in the JILA Sr clock are shown in Fig. 3.

The improved LO stability directly resulted in the demonstration of 1×10^{-17} fractional stability at 10^3 s in the comparison of two independent JILA Sr optical lattice clocks, corresponding to single-clock instability of $3.1 \times 10^{-16}/\sqrt{\tau}$, with τ being the averaging time in seconds [49]. This work was vastly improved upon the JILA clock results from 2008 [43], as shown in Fig. 4. It achieved clock stability near the fundamental quantum projection noise limit for 2000 independent atoms, fulfilling for the first time the promised superior clock stability for a multi-atom clock. Similar 1-s clock stability has also been achieved with a ytterbium (Yb) optical lattice clock and a long-term stability has reached 2×10^{-18} after averaging for 7 h [50]. The strontium [20] and ytterbium [50] clock stability is almost an order of magnitude better than that of the best single-ion clock [19], demonstrating the power of operating with 10^3 atoms. On the basis of the present JILA optical LO of 1×10^{-16} fractional

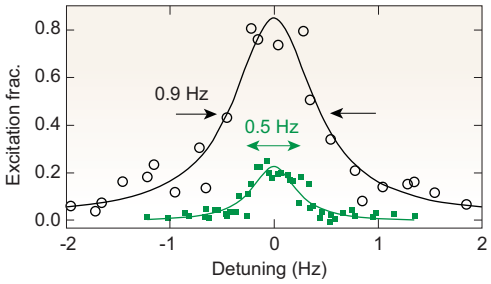

Figure 3. Exploring the atom-photon coherence in a strontium lattice clock (reprinted with permission from authors of Ref. [21]). With improved optical local oscillators, one needs to implement longer clock pulses and interrogate narrower lineshapes in order to optimize the clock stability. Here, the black circles and line show a typical line scan based on a 1-s interrogating pulse in the JILA Sr clock, leading to a record single-clock fraction instability of 2.2×10^{-16} at 1 s [21]. To explore the limit of atom-photon coherence in the system, the clock transition was probed with a very long (4-s) interrogating pulse (green squares) and a 0.5 Hz linewidth is observed. The fundamentally achievable linewidth is set by the Fourier width; the achieved linewidth and peak excitation fraction are affected by inter-atomic interactions.

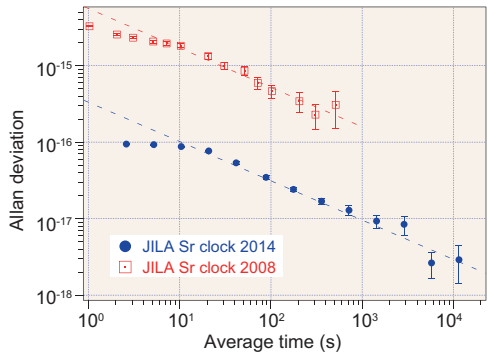

Figure 4. Recent measurements of optical clock stability. Vertical axis shows the Allan Deviation, a measure of the clock instability as a function of the averaging time. With improved optical LOs, the JILA strontium optical lattice clock with 2000 atoms recently achieved 1-s instability of 3.4×10^{-16} and extended this measurement precision to reach below 3×10^{-18} after averaging for multiple hours (blue circles) [20]. This is vastly improved upon the JILA clock results from 2008 (red squares showing a Sr-Ca clock comparison) [43]. The ytterbium lattice clock has achieved similar results [50]. This clock stability is almost an order of magnitude better than the best single-ion clock [19], showing the power of operating with many particles.

instability, it took our group 6 months to a year to perform a full clock systematic evaluation at the 2×10^{-18} level. With the previous-generation JILA optical LO of 1×10^{-15} instability, this accuracy goal would have taken an impractically long period (on the order of 600 months) to achieve. In fact, when it requires an averaging time of 10^6 s to

evaluate a particular systematic effect, it becomes so overwhelming that other fluctuating effects will make such a measurement impossible to accomplish.

Clearly, there is much to be desired for a continued improvement of optical LOs, which will continue to drive the clock performance to higher levels. Our group is undertaking intensive efforts to push the fractional frequency instability of the LO towards the level of a few times 10^{-17}. To suppress the dominant thermal Brownian noise in the optical reference cavity [46], our group is collaborating with PTB and Crystalline Mirror Solutions to employ new cavity material such as silicon crystal [51], novel crystalline mirror coating [52], and improved frequency modulation process [53] for the laser-cavity lock. As we continue to advance the coherence time of the optical LO, the Dick noise contribution will continue to fall, and this will allow us to employ an increasingly large number of atoms in a lattice such that the fundamental quantum projection noise limit is further reduced. As the clock stability is advanced by another factor of 10, it will take 100× less averaging time to reach a certain precision level where key systematic uncertainties can be evaluated and minimized for our clock to achieve higher accuracy.

Improving precision or accuracy: they advance in lockstep

For the accurate determination of any physical quantity, precise measurement represents only a necessary first step. For the Sr optical lattice clock to advance in accuracy, we must measure all systematic effects that are known to cause frequency shifts and account for these values in the final evaluation of the clock transition frequency corresponding to the intrinsic property of the Sr atom free of any external perturbations. Except for the atomic interaction-induced frequency shift, all systematic shifts are related to single-particle effects. The most dominant systematic effect is the blackbody radiation (BBR)-induced Stark shift arising from the thermal environment experienced by the atoms. Various other forms of external electric or magnetic fields give rise to several other important systematic uncertainties [10].

An effective approach to suppress the atomic interaction effect is to reduce the atomic density by either weakening the trapping confinement for the atoms or enlarging the trap volume [49]. The resultant atomic interaction frequency shift has been characterized to a few parts of 10^{-19}. The dominant BBR shift is controlled and characterized to high accuracy via precise determination of the Sr polarizability [21,54,55] and by the *in situ* measurement of the BBR radiation field around the atoms using radiation thermal sensors calibrated against the NIST International Temperature Scale. The entire clock chamber resides inside a BBR shielding enclosure to create a nearly homogeneous thermal environment for the atoms [20,21]. The thermally equilibrated enclosure is important for ensuring the thermal radiation closely follows the ideal BBR spectrum to reduce the corresponding frequency error to below 1×10^{-18}. Alternative efforts to reduce the BBR frequency shift uncertainty include (1) building an in-vacuum radiation shield that provides a well-characterized room-temperature BBR environment for the atoms in a ytterbium optical lattice clock [56], and (2) placing atoms in a mostly cryogenic environment such that both the BBR shift uncertainty and the shift itself are reduced [57]. Other systematic effects are efficiently determined through differential measurements via self-comparison with digital lock-in [49,58], building on excellent LO stability and the high precision nature of the lattice clock [20]. Table 1 presents the frequency shifts, uncertainties, and measuring methods for the JILA Sr clock of 2.1×10^{-18} total uncertainty. We note a recently published result on the single Yb^+ ion clock at PTB for a total systematic uncertainty of 3.2×10^{-18} [59]. The University of Tokyo group achieved a 7.1×10^{-18} uncertainty for their Sr optical lattice clock [57].

While a superior clock stability (precision) greatly eases the evaluation of clock systematics (accuracy), having all systematic effects sufficiently stable and under control is indispensable for building an accurate clock. The measurement precision improves with increasing averaging time τ only for certain types of noise. For example, the uncertainty improves as $1/\sqrt{\tau}$ under white frequency noise, but does not improve at all under $1/f$ flicker noise (f is the Fourier frequency for noise power spectral density) [60]. As we push the measurement uncertainty to the laboratory limit, we will likely encounter many systematic effects that may be too small to be noticed previously. These effects can sometimes be interwoven with each other and cause serious confusions. The important trick is then to achieve a sufficient level of measurement precision at a time scale shorter than the typical period at which a particular fluctuating systematic effect sets in. This will give us the capability to chase down individual systematic effects, improving the experimental conditions or implementing feedback loops.

When Nicholson *et al.* achieved clock instability of low 10^{-16} at 1 s, our group started hunting for varying systematic effects below 1×10^{-17} with averaging times of just a few tens of minutes. However, initially our group had a hard time averaging down the frequency difference between two JILA

Table 1. Frequency shifts, uncertainties, and measuring methods for the JILA Sr clock, with original data reported in Ref. [21] (reprinted with permission from the authors). The frequency shift, Δ_{Sr}, and the uncertainty of the shift, σ_{Sr}, are given in fractional frequency units multiplied by 10^{-18}. Uncertainties are quoted as 1σ standard errors.

Sources for shift	Δ_{Sr}	σ_{Sr}	Measuring method
Lattice Stark	−1.3	1.1	Trap modulation, digital lock-in [21,49,58]
BBR static	−4562.1	0.3	In-vacuum radiation thermometry [20,21]; measurement of Sr static polarizability [54]
BBR dynamic	−305.3	1.4	In-vacuum radiation thermometry [20,21]; measurement of Sr dynamic polarizability [21]
dc Stark	0.0	0.1	Applied DC field, digital lock-in [21,49,58]
Probe Stark	0.0	0.0	Intensity modulation, digital lock-in [21,49,58]
First-order Zeeman	−0.2	0.2	Magnetic field stabilization, digital lock-in [21,49,58]
Second-order Zeeman	−51.7	0.3	Magnetic field modulation, digital lock-in [21,49,58]
Density	−3.5	0.4	Density modulation, digital lock-in [21,49,58]
Line pulling + tunnelling	0.0	<0.1	Upper bound estimate based on lineshape [20,21]
Second-order Doppler	0.0	<0.1	Upper bound estimate based on optical phase stabilization [21]
Background gas	0.0	<0.6	Upper bound estimate based on vacuum pressure [21]
Servo offset	−0.5	0.4	Self-comparison with digital lock-in [21,49,58]
AOM phase chirp	0.6	0.4	Self-comparison with digital lock-in [21,49,58]
Total	−4924.0	2.1	

Sr clocks. The corresponding Allan deviation would stop following the $1/\sqrt{\tau}$ behavior when τ exceeded a few thousand seconds, indicating long-term fluctuations. Between the work reported in Ref. [49] and in Ref. [20], our group identified a drifting magnetic field possibly due to an ion pump near the main chamber of one of the JILA Sr clocks. The atoms themselves were then employed to function as a collocated magnetometer to sense the magnetic field and a feedback loop was implemented to zero the field during the clock operation. This improvement, together with control over other systematic effects, quickly led to a reduction of the long-term clock instability to 3×10^{-18}. The important message to remember is thus: improved clock precision often results in enhanced clock accuracy, simply because one can measure the important effects with a 'magnifying' lens. Precision and accuracy advance in lockstep.

USING PRECISION TOOLS TO MEASURE AND CONTROL ATOMIC QUANTUM MATERIALS

Understanding the interactions between atoms

Strongly interacting quantum many-body phenomena are among the most intriguing and rewarding research topics today. For quantum dynamics to manifestly dominate system evolution, it is important that the underlying interaction energies are larger than or at least comparable to competing single-particle energy scales such as the tunneling rate in an optical lattice [61] or the spin precession (Rabi) frequency in an optical lattice clock [48]. A prerequisite for in-depth studies of many-body physics is often to first precisely measure the relevant inter-particle interactions and then build our understandings at increasingly complex levels involving many particles.

The underlying spirit for measuring inter-atomic interactions is the same as that for measuring most other systematic shifts in a clock: one first prepares an atomic sample that is under well-characterized control such that the interaction parameter is intrinsically stable, and then measure it precisely. The only difference is that, instead of suppressing the interaction as in the case for making the best clock, one now has the freedom to enhance the atomic interaction via increased atomic density, and then measure the interaction parameter to a percent level or better. For example, in an optical lattice clock, the *p*-wave interaction between nuclear-spin-polarized Sr atoms at microkelvin temperatures is precisely determined via Ramsey spectroscopy [48,62].

At microkelvin or lower temperatures, atomic interactions arise predominantly from the *s*-wave and *p*-wave partial components. Owing to the strong decoupling between the electronic-orbital and nuclear-spin degrees of freedom [24,25], alkaline-earth atoms prepared in the two electronic clock states are predicted to exhibit nuclear spin (I) independence for their atomic collisional parameters. However, nuclear spin does play an important role by choosing *s*- or *p*-wave interaction through proper

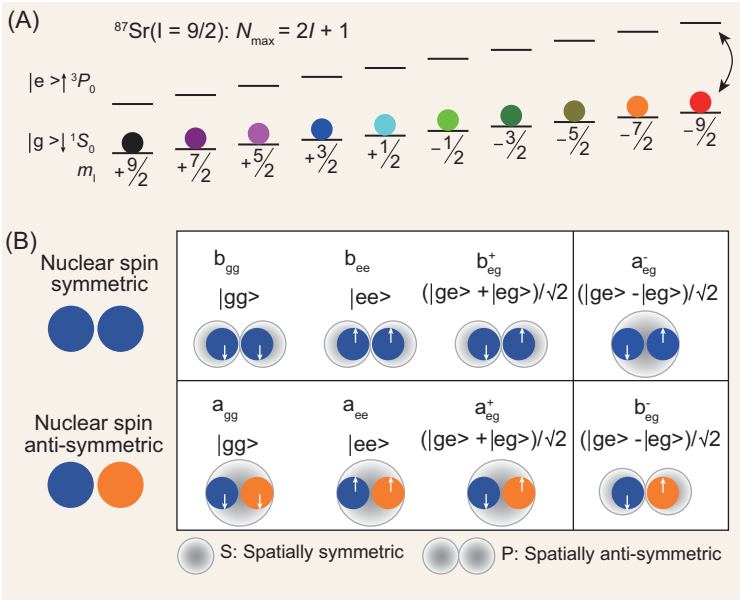

Figure 5. Diagram of SU(N)-symmetric interactions between ultracold strontium atoms. (A) Energy levels for the two lowest electronic states ($^1S_0$ and $^3P_0$) of ^{87}Sr atoms in a magnetic field, each with 10 nuclear spin states, depicted by colors. Atoms prepared in these 20 states are predicted to exhibit nuclear spin (I) independence for their inter-atomic collisional parameters. This property leads to a SU($N \leq 2I + 1$) symmetry for the interaction physics, as spectroscopically observed in recent experiments [67,68]. (B) Interactions between two fermionic atoms characterized by four s-wave (a) and four p-wave (b) elastic scattering parameters. The interactions are governed by the anti-symmetry of the overall quantum mechanical wave-function and the individual symmetries in motional states (bottom labels), nuclear spins (left labels), and electronic orbitals (white arrows).

symmetrization of the overall wavefunction as dictated by quantum statistics. This property directly leads to a SU($N \leq 2I + 1$) symmetry for the interaction physics [63–66], as shown in Fig. 5. This symmetry allows alkaline-earth atoms to emerge as a unique platform for testing many-body physics. Experimental observation of this symmetry, however, was not achieved until two essential conditions were met: first, a sufficiently stable laser must be available to provide a long coherence time that is commensurate with the small energy difference between various configurations; second, accurate control must be implemented for the nuclear spin distribution and ensemble parameters such as the temperature and total atom number. The first spectroscopic observation of SU(N)-symmetric interactions in Sr orbital magnetism [67] was enabled by the same optical LO that supports the record performance of the JILA Sr clock [20]. Based on the work reported in Refs. [48] and [67], all relevant s- and p-wave scattering parameters for ^{87}Sr were determined, providing firm knowledge for engineering these interactions to produce novel quantum states.

When the atomic sample is cooled to lower temperatures, the increased atomic density and interaction can reduce the required length of the optical coherence time. Using a LO of about 10^{-13} instability, spin-exchange interactions at the kHz level were observed in a quantum degenerate sample of fermionic ytterbium-173 atoms [68]. Subsequently, coherent interorbital spin-exchange dynamics was observed [69]. At the same time, working with relatively small (hertz-level) interaction and clock precision does have advantages: first, the inelastic processes occur much more slowly, allowing a large separation of time scales between coherent and dissipative processes; second, when the interaction is significantly smaller than single-particle energy scales such as the optical trap frequency and the optical lattice energy band-gap, the system remains in the linear regime. Complex and higher order processes such as mode-changing collisions are energetically suppressed and, accordingly, theory treatment becomes greatly simplified [62]. These are important considerations for reliably determining the interaction properties of ultracold atoms.

Exploring quantum many-body physics without quantum degeneracy

Once the interaction parameters are determined, they form the basis for implementing control and providing understandings for more complex observations. When the high coherence of the optical LO allows atoms to coherently interact with each other for a sufficiently long time, even the seemingly weakly interacting electronic orbitals in Sr develop strong many-body correlations that reach beyond mean-field physics [48,62,67]. In Refs. [48,67], a spin-orbital Hamiltonian, including the Sr clock states (electronic orbitals) and nuclear spins, is established based on the independently determined interaction parameters. A many-body treatment of this Hamiltonian accurately reproduced the dynamic evolution of atomic orbital coherence measured via Ramsey spectroscopy. Replacing operators with their mean values (mean-field treatment), however, leads to predictions inconsistent with observations. This experiment-theory collaboration demonstrates the emergence and development of quantum many-body correlations in atomic samples at microkelvin temperatures, well above the onset of quantum degeneracy for the atomic gas. Here, the quantum effects from atoms dominate over other 'classical' dephasing sources. First, the interrogated clock states (the orbital degree of freedom) are well protected from thermal (Doppler) perturbation by the strong optical lattice confinement. Second, the atomic states are manipulated with optical pulses from an optical LO whose intrinsic coherence time

is about 20 s, much longer than the experimental duration for dynamic evolutions (a few tens of millisecond to 1 s), enabling the many-body correlations to emerge clearly over classical laser noise in measurements of first-order expectation values of the electronic-orbital operator, such as the evolution of Ramsey fringe contrast.

Improving the measurement precision via further enhanced LO stability will allow us to observe more intriguing quantum effects. For example, higher order moments of the orbital operator are more sensitive to classical laser noise [48]. Hence, an improved LO can facilitate direct measurements of the quantum orbital noise distribution to probe higher order correlations and entanglement. With an improved LO, one can also design sophisticated experimental sequences that are specifically tailored to the spectral response of a particular quantum system, while discriminating against unwanted noise, thus making it easier or more robust to identify and then quantify the quantum nature of system evolution [70–72].

Providing stable and coherent control of ultracold atoms

Applying precision optical tools in ultracold atoms has been rewarding not only from the processes of measurement, but also from the prospects of creation and control. A prominent example is the first production of a quantum gas of polar molecules [73] via coherent Raman state transfer enabled by frequency comb-assisted laser phase stabilization across the visible spectrum. Ultrastable optical cavities have played a similar role for coherent state transfer processes in ultracold molecules [74—76] and in precision molecular ion spectroscopy in search for an electron electric dipole moment [77,78]. In Ref. [67], the spectral resolution available with an optical LO of 1×10^{-16} stability enables us to address individual nuclear spin states and prepare the desired nuclear spin configurations. These types of control on single-particle state manipulations are important ingredients for emerging quantum information science and technology based on ultracold atoms.

Relying on the ultranarrow clock transition of alkaline earth atoms, protocols have been proposed to induce novel synthetic spin-orbit coupling in cold strontium atoms with negligible spontaneous-emission-related heating [79]. In this scheme, spin-orbit coupling modifies the effective collisional energy scales that can be determined via Ramsey spectroscopy and mean-field analysis; these modified interactions can give rise to beyond-mean-field spin-orbit coupling dynamics. Furthermore, alkaline earth atoms hold promise for realizing strongly correlated quantum states via highly coherent atom-photon coupling such as optical flux lattices [80]. The capability to engineer stable, coherent interaction and other competing single-particle energy scales, while minimizing undesired heating or dissipative processes, distinguishes the precision spectroscopy tool described here for alkaline earth atoms from other tools for preparing many-body quantum states. How to improve our understanding of strongly correlated quantum states in a way that pushes the frontier of precision measurement science, and how to go beyond the standard quantum limit of a system consisting of many independent particles, are among the most exciting open physics questions. One might as well ponder the question whether we will be able to create a topologically protected quantum state that is robust against certain systematic effects, making a key physical quantity, such as the clock transition frequency, maximally insensitive to these systematic fluctuations, and thus creating an emerging quantum standard that is intrinsically accurate.

Achieving precision measurement and control in all degrees of freedom

So far we have only briefly described precision spectroscopy measurements that probe the internal degrees of freedom of atoms. Complementary tools have been independently developed for visualizing the external (motional) degrees of freedom. One such precision tool is the high-spatial-resolution imaging techniques, in particular, few-site or single-site imaging for 2D atomic samples [81—83]. Here, 'site' refers to a length scale, a typical optical lattice site spacing of around 0.5 μm. Precision *in situ* imaging provides detailed information on the real-space distribution of atoms (for both average values and correlations) in a trap [84], and complements the time-of-flight method that probes the sample in momentum space [85]. Furthermore, whenever a high-resolution imaging tool is capable of distinguishing two points very close in real space, it can be converted into a precision tool that tightly focuses a 'surgical knife' optical beam onto one atom without affecting the other, thus enabling spatially-resolved local manipulation of atoms [86,87].

Combining these precision tools in all degrees of freedom will provide the most reliable and comprehensive characterization and control of a quantum system. Indeed, even though interaction energies are precisely measured in clock spectroscopy experiments, in order to extract scattering parameters one must have the knowledge on the spatial

distribution of atoms [67]. Applying medium-resolution imaging together with RF and microwave spectroscopic tools has already led to the imaging of optical lattice sites with discrete atomic occupancy [88] and the probing of energy-momentum excitation spectrum [89]. The combination of the recently developed high-resolution imaging techniques and optical precision spectroscopic tools will continue to enable new discoveries based on ultracold atoms. Research along this line has already started in the JILA Sr laboratory.

TOWARDS A MORE COLLECTIVE AND NON-DESTRUCTIVE FUTURE

Many-particle optical lattice clocks, now leading the field of precision frequency metrology and clocks [20,21,57], benefit from the square root improvement of the measurement sensitivity with the number N of independent atoms, also known as the standard quantum limit (SQL). While much understanding has been gained about inter-atomic interactions, these interactions have not been utilized in a way that enables a many-particle clock to break the SQL. Quantum correlations among particles, however, allow higher measurement sensitivity to be achieved. Heisenberg's uncertainty principle suggests the maximum phase sensitivity in a clock measurement would scale as $1/N$, known as the Heisenberg limit. Heisenberg scaling for improved clock stability remains an experimental challenge. Overcoming the SQL by using correlated atomic samples to approach the Heisenberg limit can be achieved via two pathways: by preparing spin-squeezed atomic states and by producing maximally entangled states, which have both attracted numerous experimental and theoretical studies [10]. A key idea is that in order to count the total atom number in each clock state, it is preferable not to know which atom is in which state. When only the collective information is gathered with a detection system, the measurement of the state population collapses the atomic wave function into an entangled state, leading to a reduced measurement uncertainty for the relevant collective spin component that is the precessing quantity in a clock. Similar ideas have been proposed to pursue the quantum non-demolition detection of strongly correlated systems [90]. While the noise of the optical LO may prevent the clock stability from scaling according to the Heisenberg limit, partially entangled states can still generate a substantial gain in clock stability [26,27]. The advancement of entanglement-enhanced measurement and the continuous improvement of optical LOs constitute the two major routes to push the frontier of quantum metrology towards the 19th digit and beyond.

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