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Optical lattice clocks and frequency comparison

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Abstract. We consider designs of optical lattice clocks in view of the quantum statistics, relevant atomic spins, and atom-lattice interactions. The first two issues lead to two optimal constructions for the clock: a one-dimensional (1D) optical lattice loaded with spin-polarized fermions and a 3D optical lattice loaded with bosons. By taking atomic multipolar interactions with the lattice fields into account, an “atomic motion insensitive” wavelength is proposed to provide a precise definition of the “magic wavelength.” We then present a frequency comparison of these two optical lattice clocks: spin-polarized fermionic $^{87}$Sr and bosonic $^{88}$Sr prepared in 1D and 3D optical lattices, respectively. Synchronous interrogations of these two optical lattice clocks by the same probe laser allowed canceling out its frequency noise as a common mode noise to achieve a relative stability of $3 \times 10^{-17}$ for an averaging time of $\tau = 350$ s. The scheme, therefore, provides us with a powerful means to investigate intrinsic uncertainty of the clocks regardless of the probe laser stability. We discuss prospects of the synchronous operation of the clocks on the measurement of the geoid height difference and on the search of constancy of fundamental constants.

1. Introduction
To date, optical clocks based on singly trapped ions [1] and ultracold neutral atoms trapped in optical lattices that operate at the magic wavelength to eliminate light shift perturbation [2] are regarded as promising candidates for future atomic clocks. Since 2006, Sr-based “optical lattice clocks” have been evaluated close to the Cs clocks’ uncertainty limit internationally [3, 4]. To further reduce their uncertainty and instability, there remain essential experimental challenges. One is to find out better lattice geometries as well as interrogated atom species that bring out the potential performance of the clock scheme, taking into account the collisional frequency shift, the black body radiation shift, and the lattice induced light shifts. The other is to establish experimental schemes that fully utilize the advantage of a large number $N$ of atoms to improve the clock stability, which is currently hampered by the instability of the probe laser of typically $5 \times 10^{-16}$ at 1 s that is due to the thermal noise of a reference cavity [5].

In this paper, we first describe designs of optical lattice clocks in view of the quantum statistics and relevant atomic spins. This leads to two optimal configurations for the clock: a one-dimensional (1D) optical lattice loaded with spin-polarized fermions and a 3D optical lattice loaded with bosons [6]. In particular, we refer to an “atomic motion insensitive” wavelength that provides a precise definition [7] of a “magic wavelength” including atomic multipolar interactions.

We then present a frequency comparison of these two optical lattice clocks using fermionic $^{87}$Sr and bosonic $^{88}$Sr. Synchronous interrogations of the two clocks by the same probe laser allow canceling out its frequency noise as a common mode noise in evaluating the relative
Figure 1. (Color online) Energy levels for $^{88}\text{Sr}$ and $^{87}\text{Sr}$ atoms. Spin-polarized ultracold $^{87}\text{Sr}$ atoms were prepared by optical pumping on the $^1S_0 (F = 9/2) \rightarrow ^3P_1 (F = 9/2)$ transition at $\lambda = 689\text{ nm}$ with circularly polarized light. The first-order Zeeman shift and the vector light shift on the clock transition at $\lambda = 698\text{ nm}$ were eliminated by averaging the transition frequencies $f_{\pm}$. The lack of nuclear spin makes the energy structure of $^{88}\text{Sr}$ simpler.

stability of the clocks; therefore the scheme enables us to explore the intrinsic uncertainty of the clock regardless of the probe laser stability. On the other hand, in order to overcome the clock instability that is predominantly determined by the “thermal noise limit” of the probe laser, we mention a novel frequency stabilization scheme that does not employ the “electron shelving” technique for the clock state detection. We discuss possible impacts of these approaches.

2. Designing optical lattice clocks: Quantum statistics and optical lattice geometry

In addition to the removal of light shift perturbations, the control and prevention of atomic interactions are a serious concern in designing optical lattice clocks. The collisional frequency shift of atomic clocks operated with ultracold atoms is related to the mean field energy shift $\mathcal{E} = 4\pi\hbar^2 a g^{(2)}(0)/m$ of the relevant electronic state, with $a$ the $s$-wave scattering length, $n$ atomic density, and $m$ atomic mass. Here, $g^{(2)}(0)$ is the two-particle correlation function at zero distance, which is zero for identical fermions and $1 \leq g^{(2)}(0) \leq 2$ for distinguishable or bosonic atoms. Hence, collisional shifts are suppressed for ultracold fermions, while they are intrinsically unavoidable for bosons. The quantum statistical nature of atoms is determined by their total spins; that is, bosons have zero or integer spins and fermions have half-integer spins. In particular, for atoms with an even number of electrons that have a $J = 0$ state suitable for optical lattice clocks, their nuclear spins $I$ may be zero for bosons and $I \geq 1/2$ for fermions. Consequently, the total angular momentum $F = J + I$ of the clock states can be zero for bosonic atoms, but not for fermions, which causes coupling to the light polarization of the lattice field. Figure 1 shows the relevant energy levels for fermionic $^{87}\text{Sr}$ and bosonic $^{88}\text{Sr}$ isotopes of Sr that we employ in the following experiments.

We consider two lattice geometries. A one-dimensional (1D) (see Fig. 2(a)) or 2D lattice composed of a single electric field vector has spatially uniform light polarization. In contrast, a 3D lattice (see Fig. 2(b)) requires at least two electric field vectors; therefore, the synthesized
The lattice allows us to cancel out the vector light shift [10] by alternately interrogating the transition relative phases of the lattice lasers. In 1D optical lattice clocks with multiple atoms in each lattice, the clock transition (see Fig. 1) gives a trap depth of about 200 kHz. As the atoms are trapped near the site, the application of spin-polarized fermions [8, 9] may minimize the collisional frequency shift owing to their quantum-statistical properties. Figure 2(a) shows a schematic diagram for the “spin-polarized” 1D optical lattice clock [10], where the upward arrows correspond to spin-polarized fermionic atoms. The spatially uniform light field polarization of the 1D optical lattice allows us to cancel out the vector light shift [10] by alternately interrogating the transition frequencies \( f_\pm \), corresponding to two outer Zeeman components \( ^1S_0(F, \pm m_F) - ^3P_0(F, \pm m_F) \) of the clock transition (see Fig. 1). This vector light shift cancellation technique simultaneously cancels out the Zeeman shift, thereby realizing virtual spin-zero atoms.

The application of 3D optical lattices with a single (or less) atom in each lattice site is effective to suppress atomic collisions. However, light polarization inhomogeneity inevitable in 3D optical lattices makes a vector light shift for atoms with its angular momentum \( F \neq 0 \) problematic, as the “vector light shift cancellation” technique is no longer applicable. From this viewpoint, the 3D optical lattice clock is suitable for bosonic atoms with scalar clock states \( (J = 0) \) and has been demonstrated with bosonic \(^{88}\text{Sr} \) atoms [6]. Its systematic uncertainties were investigated at \( 3 \times 10^{-15} \) by referencing a spin-polarized 1D optical lattice clock with fermionic \(^{87}\text{Sr} \) atoms.

Finally in order to moderate the hyperpolarizability effects that cannot be eliminated, in particular, at the red-detuned magic wavelength where atoms are trapped at lattice intensity maxima, we investigated a blue-detuned lattice that confines atoms in the intensity minima of the electric field. For Sr, such wavelengths are found on the blue side of the \( 5s^2 \, ^1S_0 - 5s5p \, ^1P_1 \) transition at 461 nm; one such wavelength is found at \( \lambda_L \approx 390 \) nm on the blue side of the \( 5s5p^3 \, ^3P_0 - 5s6d \, ^3D_4 \) transition at 394 nm. For this magic wavelength, a laser intensity of \( I_L = 10 \text{ kW/cm}^2 \) gives a trap depth of about 200 kHz. As the atoms are trapped near the...
nodes of the standing wave, the effective light intensity that atoms experience is about one-
tenth of the intensity at the anti-nodes. Assuming a trap depth of 10 $\mu$K, the 4th-order light
shift is estimated to be 0.1 mHz, corresponding to a fractional uncertainty of 2 $\times$ 10$^{-19}$ [11].
The blue magic wavelength was experimentally determined to be 389.889(9) nm [11] employing
$^{87}$Sr atoms trapped in a 1D optical lattice operated at the (red-detuned) magic wavelength of
$\lambda_L = 813.4$ nm.

3. Multipolar interactions of atoms with lattice field and
atomic-motion-insensitive wavelength

Optical lattice clocks operated at the blue magic wavelength seem to be ideal in view of the
electric dipole (E1) interaction of atoms with the lattice laser field. However, when taking
atomic multipolar interactions into account, things are not that simple. Consider, for example,
the linearly polarized ($|e_x\rangle$) standing wave electric field $E = e_x E_0 \sin ky \cos \omega t$ with wave number
$k$ and frequency $\omega$, as shown in Fig. 2(c). Following the Maxwell equation $\nabla \times E = -\frac{1}{c} \partial B/\partial t$, with $c$ the speed of light, the corresponding magnetic field is given by
$B = -e_x E_0 \cos ky \sin \omega t$. This indicates that the electric and magnetic field amplitudes are one quarter of the wavelength
$\lambda/4 = \pi c/(2\omega)$ out of phase in space. Consequently, the magnetic dipole (M1) interaction is
largest at the nodes of the electric field. Furthermore, as the electric quadrupole (E2) interaction
is proportional to the gradient of the electric field, the E2 interaction is also largest at the nodes
of the electric field. Hence, the blue magic wavelength is not necessarily free of light shift due to
atomic multipolar interactions with the lattice field. The energy shift of atoms in optical lattices
is obtained by the second-order perturbation in the E1, M1, and E2 interactions that vary as
$V_{E1} \sin^2 ky$, $V_{M1} \cos^2 ky$, and $V_{E2} \cos^2 ky$. Consequently, it is no longer possible to perfectly
match the total light shift in two clock states, as the differential light shifts due to the M1 and
E2 interactions introduce an atomic-motion-dependent light shift because of their spatial
mismatch with the E1 interaction [12].

Although the contributions of the M1 and E2 interactions are 6-7 orders of magnitude smaller
than that of the E1 interaction in optical lattice clocks in Sr [2], they have a non-negligible
contribution in pursuing a 1 $\times$ 10$^{-18}$ level uncertainty. Therefore, a more precise definition of
the magic wavelength, including multipolar interactions, is necessary. Assuming the differential
polarizabilities of the E1, M1, and E2 interactions in the clock transition to be $\Delta \alpha_{E1}(\lambda_L)$,
$\Delta \alpha_{M1}(\lambda_L)$, and $\Delta \alpha_{E2}(\lambda_L)$, and the corresponding spatial distributions to be
$q_{E1}(r)$, $q_{M1}(r)$, and
$q_{E2}(r)$, the transition frequency of atoms in the optical lattices can be given by

$$\nu(\lambda_L) = \nu_0 - \frac{1}{2\hbar} [\Delta \alpha_{E1}(\lambda_L) q_{E1}(r) + \Delta \alpha_{M1}(\lambda_L) q_{M1}(r) + \Delta \alpha_{E2}(\lambda_L) q_{E2}(r)] E^2, \quad (1)$$

where the 4th- and higher-order terms and light polarization dependences are omitted. We have
shown that one can eliminate the atomic-motion-dependent light shift caused by the multipolar
interactions by choosing particular 3D optical lattice geometries that make $q_{M1}(r)$ or $q_{E2}(r)$
terms in phase or out of phase with respect to the spatial dependence of $q_{E1}(r)$ [7]. For example,
in the case of a 1D lattice with the E1 spatial dependence $q_{E1}(r) = \sin^2 ky = (1 - \cos^2 ky)$,
the corresponding M1 and E2 interactions can be expressed as $q_{M1}(r) = q_{E2}(r) = \cos^2 ky =
\Delta q - q_{E1}(r)$ with $\Delta q = 1$. Therefore, by taking $\Delta \alpha_{EM} \equiv \Delta \alpha_{E1} - \Delta \alpha_{M1} - \Delta \alpha_{E2}$
and $\Delta \alpha_0 \equiv \Delta \alpha_{M1} + \Delta \alpha_{E2}$, Eq. (1) can be rewritten as

$$\nu(\lambda_L) = \nu_0 - \frac{1}{2\hbar} \Delta \alpha_{EM}(\lambda_L) q_{E1}(r) E^2 - \frac{1}{2\hbar} \Delta \alpha_0(\lambda_L) \Delta q E^2, \quad (2)$$

where the second term on the right side varies in phase with the E1 interaction. This equation
suggests the precise definition of the magic wavelength to be an “atomic-motion insensitive”
wave length that satisfies $\Delta \alpha_{EM}(\lambda_L) = 0$. The last term provides a spatially constant offset of
Figure 3. (Color online) (a) Experimental setup for frequency comparison of 1D and 3D optical lattice clocks. A single laser with an RF frequency offset of ≈ 62 MHz probes the clock transitions of the two isotopes simultaneously. (b) Timing chart for the frequency comparison. A cycle time of $T_c = 1.4$ s is used to probe a shoulder of the Rabi excitation. Cancellation of the slowly varying residual magnetic fields is completed in $4T_c$. Thick vertical lines indicate a clock probing period of 100 ms.

typically 10 mHz or below and is solely dependent on the total laser intensity $\propto \Delta qE^2$ used to form the lattice. This offset frequency can be accurately determined by measuring the atomic vibrational frequencies in the lattice [7].

4. Frequency comparison of 1D and 3D optical lattice clocks

Optical lattice clocks [2] are expected to be highly stable, e.g., $\sigma_y(\tau) \sim 10^{-18}/\sqrt{\tau}$, if they operate at the quantum projection noise (QPN) limit of $\sigma_y(\tau) \sim (\Delta f/f_0)/\sqrt{N\tau}$, by taking advantage of the large number $N \sim 10^6$/s of atoms and moderately narrow linewidth $\Delta f = 1$ Hz. However, the frequency comparison of two optical lattice clocks [6] shows that their actual stabilities are essentially limited by those of the probe lasers due to the thermal noise of the reference cavity, e.g., $\sigma_y \sim 1 \times 10^{-15}$ for a 7.5-cm-long cavity made of ultralow expansion (ULE) glass [5] and by the Dick effect [13], which is attributed to the down-conversion of the high frequency ($n/T_c$ with $n$ an integer) components of the probe laser noise $\dot{\omega}(t)$ introduced by the discrete interrogation process with a cycle time $T_c$ of the clock operation.

When exciting the clock transition, the population fluctuation $\delta P$ of the excited state caused by the frequency noise $\delta \omega(t)$ is expressed using the sensitivity function $g(t)$ as in [13],

$$\delta P = \frac{1}{2} \int_0^{T_c} g(t) \delta \omega(t) dt, \quad (3)$$

where $T_c$ is a clock cycle time. The frequency servo translates a population fluctuation $\delta P$ into a frequency fluctuation of the probe laser, which degrades the stability of an atomic clock, known as the Dick effect.

We consider a frequency comparison of two optical lattice clocks that are simultaneously probed by a single laser, which allows the two clocks to be probed by the same frequency noise $\delta \omega(t)$. As long as the sensitivity functions $g(t)$ are the same for both clocks, their population fluctuations $\delta P$ should be equal; therefore the stability degradation due to $\delta P$ may be rejected as a common mode noise in evaluating the frequency difference of the two clocks. This allows an investigation of the relative stability of the two clocks that is only limited by the quantum projection noise of atoms regardless of the probe laser instability. Such a synchronous frequency comparison was previously demonstrated in the microwave frequency domain using Cs and Rb fountain clocks [14].
Figure 3(a) shows an experimental setup for frequency comparison of the 1D and 3D lattice clocks, loaded with $^{87}$Sr and $^{88}$Sr, respectively, as described in Sec. 2. After cooling and loading atoms in each optical lattice, we interrogated the two clock transitions synchronously with a 100 ms-long Rabi pulse. A cycle time of $T_c = 1.4$ s included the time spent for cooling and trapping of atoms in the lattices and for measuring the excitation probability by observing a shoulder of the Rabi spectrum, as depicted in Fig. 3(b). The clock laser frequencies $f_{87}$ and $f_{88}$ were stabilized to the clock transitions of $^{87}$Sr and $^{88}$Sr by steering the frequencies of acousto-optic modulators (AOMs) by respective digital servos. The beat note $\delta f = f_{88} - f_{87}$, which corresponds to their isotope shift $\delta f \approx 62$ MHz, was investigated to evaluate the stabilities and uncertainties of the two optical lattice clocks.

In a 1D lattice, we trapped $3 \times 10^3$ fermionic $^{87}$Sr atoms. They were spin-polarized in the $m_F = 9/2$ (or $-9/2$) stretched state in the $^1S_0$ ground state (see Fig. 1) to avoid atomic collisions via Pauli blocking [6, 8]. A bias magnetic field of 0.22 mT was applied to define the quantization axis. We excited the $^1S_0(F = 9/2, m_F = \pm 9/2) \rightarrow ^3P_0(F = 9/2, m_F = \pm 9/2)$ transitions with frequencies of $f_{\pm}$ successively and calculated $f_{87} = (f_+ + f_-)/2$ to eliminate the first order Zeeman and vector light shifts. For a 3D optical lattice clock, $2 \times 10^3$ bosonic $^{88}$Sr atoms were loaded into a single occupancy 3D lattice (see Fig. 2(b)). A bias magnetic field of $|B_m| = 2.36$ mT was applied to magnetically induce the $^1S_0 \rightarrow ^3P_0$ clock transition. A probe laser intensity of $I_p = 86$ mW/cm$^2$ was applied to compensate the relatively small magnetically-induced transition moment. These external fields introduced rather large frequency shifts; the second order Zeeman shift of $\Delta_B \approx -130$ Hz and the probe laser light shift of $\Delta_p \approx -1.5$ Hz, whose stabilities were evaluated with fractional uncertainties of $2 \times 10^{-17}$ and $1 \times 10^{-17}$, respectively. To eliminate the influence of a slowly varying environmental magnetic field $B_{\text{ext}}$ that affected the second order Zeeman shift as $\propto \pm B_m \cdot B_{\text{ext}}$, we reversed the direction of the bias magnetic field as $\pm B_m$ in successive measurements and calculated $f_{88} = (f_+ + f_-)/2$, where we note that the contribution of $\propto |B_{\text{ext}}|^2$ was negligibly small. Moreover, we installed fiber noise cancellation systems referencing the end mirrors of the 1D and 3D lattices to reduce the Doppler shifts arising from the relative motion between the probe lasers and atoms in the lattices.

Figure 4(a) shows the temporal response of the frequency difference $\delta f = f_{88} - f_{87}$ of the 1D-3D lattice clocks in the synchronous measurement. Thanks to the common mode noise rejection of the laser frequency noise, atomic frequency differences of a few times $10^{-16}$ are visible in tens of seconds. This will find useful applications in investigating possible perturbations on probed atomic transitions nearly in real time.

Figure 4(b) shows the relative stabilities of the 1D and 3D optical lattice clocks. The triangles show the relative stability of the two clocks measured for an asynchronous interrogation, where the 1D and 3D clocks operated alternately every 1 s [6]. The short-term stability is limited by that of the probe laser operated at the thermal noise limit of a reference cavity made of ULE glass. After an averaging time of a few tens of seconds, the Allan deviation starts to decrease due to the feedback control with a Nyquist frequency of $f_N = (\text{4 s})^{-1}$. The Allan deviation decreased with $\sigma_y(\tau) = 6 \times 10^{-15}/\sqrt{\tau}$ (green dashed line) and reached $1 \times 10^{-16}$ at $\tau = 2 \times 10^5$ s. In contrast, the relative stability measured for the synchronous interrogations is shown by the circles. The Allan deviation decreased with $\sigma_y(\tau) = 6 \times 10^{-16}/\sqrt{\tau}$ and reached $3 \times 10^{-17}$ for an averaging time of $\tau = 350$ s, which shows 10 times better stability than that of the asynchronous interrogation. It is noted that the stability in the synchronous measurement is approaching the QPN limit for $N = 1 \times 10^3$ and $\Delta f = 8$ Hz (blue dashed line). These parameters are similar to those used in the measurements.
Figure 4. (Color online) (a) Frequency difference $\delta f = f_{88} - f_{87}$ of the 1D-3D lattice clocks measured for the synchronous clock operation, where the isotope shift of $\approx 62$ MHz is subtracted. (b) Allan standard deviation calculated for $\delta f = f_{88} - f_{87}$ for sequential interrogations (triangles) and for synchronous interrogations (circles). In the synchronous interrogations, the Allan standard deviation decreased with $\sigma_\alpha(\tau) = 6 \times 10^{-16}\tau^{-1/2}$ and reached $3 \times 10^{-17}$ for an averaging time of $\tau = 350$ s. The blue dashed line shows the QPN limit for $N = 1 \times 10^3$ and $\Delta f = 8$ Hz.

5. Summary and outlook

We have discussed possible realizations of optical lattice clocks taking quantum statistics, the higher order and the multipolar atom-field interactions into account. Two configurations, 1D optical lattices loaded with spin-polarized fermions and 3D lattices loaded with bosons with a single atom in each lattice site, will allow an investigation of the clock scheme free from the collision shifts and the vector light shift. In addition, the concept of “atomic motion insensitive” wavelength is introduced to redefine the “magic wavelength” including the multipolar atom-field interactions. The “blue atomic-motion-insensitive” wavelength may provide neutral atoms with ideal traps for spectroscopy, as it closely simulates Paul traps for a single ion. We have demonstrated the advantage of employing a “large number of atoms” in optical lattice clocks by the synchronous operation of the 1D and 3D optical lattice clocks, which allowed us to explore a hitherto uninvestigated clock stability of $3 \times 10^{-17}$ with an averaging time as short as $\tau = 350$ s.

The synchronous frequency comparison scheme provides a convenient means to investigate systematic uncertainties of optical lattice clocks, such as the collision shift, the blackbody radiation shift, the higher order and the multipolar light shifts at fractional uncertainties of $1 \times 10^{-17}$ and beyond. The scheme is readily applicable to the frequency comparison of two clocks located at remote sites by sharing a local oscillator with the help of precise optical frequency link technologies [4, 15]. Assuming the achieved frequency stability shown in Fig. 4(b), the remote frequency comparison will allow an investigation of the geoid height difference of two sites with an uncertainty of 30 cm in minutes’ averaging time. On the other hand, the synchronous interrogation may significantly increase the stability in the frequency
comparison of optical lattice clocks made of different atomic species to investigate the constancy of fundamental constants [16] and the local position invariance [3]. In the case of optical lattice clocks employing Sr \((f_0 = 429 \text{ THz})\), Yb \((f_0 = 518 \text{ THz})\) and Hg \((f_0 = 1129 \text{ THz})\) atoms that are synchronously interrogated by probe lasers frequency-synthesized by an optical frequency comb driven by a single thermal-noise-limited local oscillator operated at \(5 \times 10^{16}\), relative stabilities of \(2.4 \times 10^{-17}/\sqrt{T}\) and \(1.5 \times 10^{16}/\sqrt{T}\) are expected for Sr/Yb and Sr/Hg frequency comparisons, where we assumed one million atoms and the excitation linewidth of \(\Delta f = 8 \text{ Hz}\).

In pursuit of a stability goal for the optical lattice clocks [2], e.g., \(1 \times 10^{18}/\sqrt{T}\), a novel frequency stabilization scheme needs to be developed. Considering the instability of the thermal noise (at 300 K) limited probe lasers that show a minimum of \(0.5 - 1 \times 10^{-15}\) independent of the averaging time in the range of ms to a few second [5], a servo-loop time constant much less than a second is beneficial to achieve better stability. In this direction, we are considering to make use of the transmitted probe laser through millions of lattice-trapped atoms, since the transmitted light carries full information of the atomic dipoles through the phase shift and absorption/emission that can be extracted as quadrature components using the FM spectroscopy technique. A detailed discussion will be given elsewhere.

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