Ultrastable Optical Clock with Neutral Atoms in an Engineered Light Shift Trap

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An ultrastable optical clock based on neutral atoms trapped in an optical lattice is proposed. Complete control over the light shift is achieved by employing the $5s^2\,^1S_0 \rightarrow 5s5p\,^3P_0$ transition of $^{87}\text{Sr}$ atoms as a “clock transition”. Calculations of ac multipole polarizabilities and dipole hyperpolarizabilities for the clock transition indicate that the contribution of the higher-order light shifts can be reduced to less than 1 mHz, allowing for a projected accuracy of better than $10^{-17}$.

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Careful elimination of perturbations on electronic states and of motional effects has been considered as a prerequisite for realizing an atom frequency standard [1]. A single ion trapped in an RF quadrupole field is one of the ideal systems that satisfy these requirements [2], as the trap prepares a quantum absorber completely at rest in free space for an extended time and its electric field vanishes at the center of the trap. Employing this scheme, quantum projection noise (QPN) limited spectroscopy [3] has been performed with an expected accuracy of $10^{-18}$ [1,2].

Despite its anticipated high accuracy, the stability of the single-ion based optical clock is severely limited by QPN; long averaging times are required to meet its ultimate accuracy [3]. The measure of the fractional instability is provided by the Allan variance, $\sigma_y(\tau) = \frac{1}{2} \sqrt{N \tau/\tau_m}$.

Assuming the transition line $Q \approx 1.6 \times 10^{14} \, \text{Hz}$ and a cycle time of $\tau_m \approx 0.1 \, \text{s}$, $4 \times 10^{7}$ measurement cycles are required for a single quantum absorber ($N = 1$) to reach $\sigma_y(\tau) = 10^{-18}$, corresponding to a total averaging time $\tau$ of a few months. For further increase of the stability, the averaging time increases quadratically and will become inordinately long.

One may think of increasing the number of quantum absorbers $N$ as employed in neutral atom based optical standards [4,5,8]. In this case, however, the atom-laser interaction time sets an upper bound for the $Q$-factor since an atom cloud in free space expands with finite velocity and is strongly accelerated by the gravity during the measurement. Hence the highest line $Q \approx 10^{12} \, \text{Hz}$ obtained for neutral atoms is 2 orders of magnitude smaller than that of a trapped ion. Furthermore, it has been pointed out that residual Doppler shifts arising from an imperfect waveform of the probe beam and atom-atom collisions during the measurement affect its ultimate accuracy [4,8].

In this Letter, we discuss the feasibility of an “optical lattice clock” [9], which utilizes millions of neutral atoms separately confined in an optical lattice [10] that is designed to adjust the dipole polarizabilities $\alpha_{E1}$ for the probed electronic states in order to cancel light field perturbations on the measured spectrum [11]. In striking contrast with conventional approaches toward frequency standards [1], the proposed scheme interrogates atoms while they are strongly perturbed by an external field. We will show that this perturbation can be canceled out to below $10^{-17}$ by carefully designing the light shift potentials. This scheme permits an exceptionally low instability of $\sigma_y(\tau) \approx 10^{-18}$ with an interrogation time of only $\tau = 1 \, \text{s}$, which may open up new applications of ultra precise metrology, such as the search for the time variation of fundamental constants [12] and the real time monitoring of the gravitational frequency shift.

Figure 1 illustrates the proposed scheme. Subwavelength confinement provided by the optical lattice localizes atoms in the Lamb-Dicke regime (LDR) [13], where the first order Doppler shift as well as the photon recoil shift disappears [14,15] and the second order Doppler shift can be made negligibly small by sideband-cooling atoms down to the vibrational ground state [3,16]. In addition, a 3-dimensional lattice with less than unity occupation could reduce the collisional frequency shifts [8,16]. Therefore this scheme simulates a system where millions of single-ion clocks operate simultaneously.

The transition frequency $\nu$ of atoms exposed to the lattice electric field of $E$ is described as,

$$
\nu = \nu^{(0)} - \frac{1}{4} \Delta \alpha(e, \omega) E^2 - \frac{1}{64} \Delta \gamma(e, \omega) E^4 - \ldots,
$$

where $\nu^{(0)}$ is the transition frequency between the unperturbed atomic states, $\Delta \alpha(e, \omega)$ and $\Delta \gamma(e, \omega)$ are the
FIG. 1: Simplified optical coupling scheme for $^{87}$Sr. (a) In the limit of large detunings $\delta_0$ of the coupling laser compared to the hyperfine splittings $\delta_{\text{hfs}}$, the squared transition dipole moment of the upper $J$ manifold can be simply added up, resulting in a quasi-scalar light-shift. (b) 3D optical lattice provides Lamb-Dicke confinement while it prevents atom-atom interactions.

The frequency $\nu_{\text{ac}}$ in the clock transition is estimated to be $-2.4$ Hz at $T = 293$ K. The uncertainty can be reduced to the $10$ mHz level by controlling the surrounding temperature variation $\Delta T \leq 0.5$ K. Alternatively, by operating the clock at $T = 77$ K, the blackbody shift is dramatically decreased to $10$ mHz because of its $T^4$ dependence.

Owing to the hyperfine interaction (HFI), the clock transition no longer consists of simple scalar states; therefore the tensor light shift arises. The fraction of the tensor shift in the total light shift is roughly proportional to $\delta_{\text{hfs}}/\delta$, i.e., the ratio of the hyperfine splitting of the coupled electronic state $\delta_{\text{hfs}}$ to the trapping laser detuning $\delta$ to that state. For the $^3P_0$, $^3D_1$, and $^1P_1$ state, as shown in Fig. 1(a), the ratio $\delta_{\text{hfs}}/\delta$ is $8.1 \times 10^{-5}$, $5.4 \times 10^{-6}$, and $2.1 \times 10^{-7}$, respectively. Therefore the tensor shift of the $^3P_0$ state can be 2 orders of magnitude larger than that of the $^1S_0$ state. In order to give an approximate estimate for this tensor shift, we calculated the polarization-dependent light shift in the $^3P_0$ state, taking into account the electric-dipole coupling to the $^3S_1$ and $^3D_1$ hyperfine manifolds that are energy shifted due to HFI. Both of these manifolds provide half ($\approx 65$ kHz) of the total light shift but dominate its tensor component: Although higher lying electronic states support another 50 % of the light shift, their contribution to the tensor shift can be less than 50 %, as the ratio $\delta_{\text{hfs}}/\delta$
monotonically decreases for larger $\delta$. The inset of Fig. 2 shows the result with the light polarization parameterized as $\mathbf{e} = \cos \theta \mathbf{e}_- + i \sin \theta \mathbf{e}_+$ ($\mathbf{e}_\pm$ represent the unit vector for $\sigma_\pm$ polarization), where a bias magnetic field of $B_0 = 30 \text{ mG}$ is applied to reduce the Raman coherences among the Zeeman substates. The $m = \pm 1/2$ state can be best used for the “clock” transition, as it exhibits the smallest polarization-dependence of less than 1 Hz. By employing these states, one could control the light shift within 1 mHz by defining $\theta$ within 1 mrad, even if one applied the linearly polarized trapping laser ($\theta = \pi/4$) where the gradient $d\delta_{ac}/d\theta$ (= 0.83 Hz/rad) is the largest.

In order to provide an estimate for the higher-order field contributions $O(\delta^4)$ described by ac hyperpolarizabilities, we have used the general theory \cite{21, 21, 22, 22} for calculating the light shift for the $5s^2 1S_0 \rightarrow 5s5p\,^3P_0$ transition on the basis of the Green’s function method in the Fues’ model potential approximation \cite{21}. These model potential calculations have reproduced Fig. 2 to within 3 % accuracy both for the amount of the ac Stark shifts as well as the intersection frequency $\omega_L (= 2\pi c/\lambda_L)$, confirming the validity of these two independent approaches.

To calculate the $M1$ and $E2$ contributions to the polarizability Eq. (4), the magnetic dipole and electric quadrupole atom-field interactions should be taken into account together with the electric dipole term in the amplitude,

$$\hat{V}(\mathbf{r}) = \hat{V}_{E1} + \hat{V}_{M1} + \hat{V}_{E2},$$

of the interaction Hamiltonian $\hat{H}(\mathbf{r},t) = \hat{V}(\mathbf{r})e^{-i\omega t} + \hat{V}^\dagger(\mathbf{r})e^{i\omega t}$ \cite{21}. The magnetic dipole polarizability for the ground state equals zero, while for the excited state $5s5p\,^3P_0$ its value is proportional to the squared fine-structure constant ($\alpha = 1/137$), to the splitting of the triplet states $E_{10} \equiv E_{5p\,^3P_1} - E_{5p\,^3P_0}$, and to the square of the wave functions overlapping integral. The quadrupole polarizabilities of both levels are on the order of $(\alpha \omega)^2$ and may also become considerable only in the closest vicinity of the resonance on the quadrupole-allowed transition. Numerical estimates with the frequency $\omega_L$ where $\Delta \alpha_{E1}(\omega_L) = 0$ gives $\alpha_{M1} \approx \alpha_{E2} \approx 10^{-7} \times \alpha_{E1}$ for both levels.

The hyperpolarizability $\gamma(\mathbf{e},\omega)$ is calculated starting from a formal expression for the fourth-order quasi-energy in terms of the field-free wave functions $|0\rangle$ and the reduced quasi-energy Green’s functions $G$ \cite{21, 22}:

$$\Delta E^{(4)} = -\frac{\sigma^4}{64} \gamma(\mathbf{e},\omega) = -\langle\langle 0|HGHGHGH|0\rangle\rangle + \langle\langle 0|HGH|0\rangle\rangle \langle\langle 0|H|H|0\rangle\rangle,$$

where the double brackets indicate the integration over the spatial variables and averaging over time. In Eq. (4) only the dipole term of Eq. (3) is taken into account in the interaction Hamiltonian $V(\mathbf{r})$.

After the time averaging and angular integration, the dipole dynamic polarizability and hyperpolarizability tensors may in general be resolved into 3 and 5 irreducible parts, of which only scalar terms $\alpha_p$ and $\gamma_q$ contribute in a state with the total momentum $J = 0$ \cite{22}. $\alpha_p$ and $\gamma_q$ are determined by linear combinations of frequency-dependent radial matrix elements of the second, third and fourth orders.

It is to note first that the scalar parts of $\gamma_0(\mathbf{e},\omega)$ are different for the linear and circular type of polarization, $\gamma_0(\mathbf{e},\omega) \neq \gamma_0(\omega)$, even for atoms in $S$-states, while all the terms of the polarizability in Eq. (2) are independent of $\mathbf{e}$ for a state with $J = 0$. Secondly, the number and type of singularities for the hyperpolarizability exceeds that for the polarizability, and the contribution of these singularities also depend on the polarization of the light field. E.g., for linearly polarized radiation, there are two-photon singularities of $\gamma_0(\omega)$ on the $J = 0$ states, while for the circular polarization such singularities cannot appear. So, the hyperpolarizability of the ground state with two equivalent electrons $5s^2 1S_0$ in Sr may be written as

$$\gamma'(\omega) = \gamma^c(\omega) + \frac{8}{9} \left[ \sigma_{101}(\omega, 2\omega, \omega) + \frac{3}{5} \Sigma_{121} \right]$$

$$- \frac{2}{5} \sigma_{121}(\omega, 2\omega, \omega)$$

$$\gamma^c(\omega) = \frac{8}{9} \left[ \Sigma_{101} + \frac{1}{5} \Sigma_{121} + \frac{6}{5} \sigma_{121}(\omega, 2\omega, \omega) \right]$$

$$- 2 \alpha_0(\omega) S_{-3}(\omega)$$

where $\alpha_0(\omega)$ is the polarizability and $S_{-3}$ is the so-called frequency-dependent oscillator strengths moment. The
following notations for the radial matrix elements and their combinations were used above:

\[
\begin{align*}
\Sigma_{l_1 l_2 l_3} &= \sigma_{l_1 l_2 l_3}(\omega, 0, \omega) + \sigma_{l_1 l_2 l_3}(\omega, 0, -\omega); \\
\sigma_{l_1 l_2 l_3}(\omega_1, \omega_2, \omega_3) &= R_{l_1 l_2 l_3}(\omega_1, \omega_2, \omega_3) + R_{l_1 l_2 l_3}(-\omega_1, -\omega_2, -\omega_3); \\
R_{l_1 l_2 l_3}(\omega_1, \omega_2, \omega_3) &= (0|rg_{l_1}^r g_{l_2}^r g_{l_3}^r r|0). \quad (6)
\end{align*}
\]

The arguments in Eqs. (6) indicate the frequency dependence in energy denominators [21]. Here, \(g_{l_3}^r\) is the radial Green’s function in the subspace of the jumping electron’s states with angular momentum \(l\).

In the vicinity of a one-photon resonance with frequency detuning \(\delta \ll \omega\), the product of the second-order and third-order matrix elements in Eq. (1), which corresponds to the term \(a_0 S_{-3}\) in Eq. (5), dominates the third-order poles of order \(\delta^{-3}\). The fourth-order matrix element in Eq. (4) has the second-order poles \((\delta^{-2})\). The two-photon \((2\omega)\) resonance singularity on the \(J = 0\) states in the radial matrix elements \(R_{l_1 l_2 l_3}(\omega, \omega, \omega)\) appears only for \(\gamma^1(\omega)\) and that on the \(J = 2\) states in \(R_{l_1 l_2 l_3}(\omega, 2\omega, \omega)\) appears both for \(\gamma^1(\omega)\) and \(\gamma^2(\omega)\).

We used the analytic Sturm-series representation of the Green’s function, corresponding to calculation of the infinite sums over the total atomic spectrum including the continuum. Finally, the radial integrals in Eq. (6) were presented in the form of absolutely converging series, well suited for the numerical computations. The numerical results for \(\gamma^1_{S_0}(\omega)\) and \(\gamma^2_{P_0}(\omega)\) at the intersection frequency of \(\omega_L\) are \(6.3 \cdot 10^6\) a.u. and \(2.7 \cdot 10^8\) a.u., respectively, which give fourth-order ac Stark shifts of \(\Delta E_{S_0}^{(4)}/\hbar \approx -5.3\cdot10^{-5}\) Hz and \(\Delta E_{P_0}^{(4)}/\hbar \approx -2.3\cdot10^{-3}\) Hz for the trapping laser intensity of 10 kW/cm². Therefore, the contribution of the higher order light shifts is as small as \(5 \times 10^{-18}\). Further elimination of this systematic error can be achieved by extrapolating the trapping laser intensity to zero in a quadratic way.

In summary, we have discussed the feasibility of precision spectroscopy of neutral atom ensembles confined in an optical lattice, by applying a light-shift cancellation technique on the \(^{1}S_0(F = 9/2, m_F = \pm 1/2) \rightarrow {}^3P_0(F = 9/2, m_F = \pm 1/2)\) clock transition of \(^{87}\)Sr that has a negligibly small tensor shift and a suitable transition moment to perform spectroscopy. Our calculation including the higher-order Stark shift confirmed that the measurement of the unperturbed atomic transition at 1 mHz level is feasible, allowing for the projected accuracy of better than \(10^{-17}\). Since this scheme is equivalent to millions of single-ion-clocks operated in parallel, excellent improvement of the stability \(\sqrt{\Delta R}\) can be expected. The theory and method for the numerical calculation of the hyperpolarizability tensor for arbitrary alkaline-earth atoms will be discussed in a forthcoming paper.

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Note Added: After submission of the paper, we have experimentally demonstrated the optical lattice clock and determined the intersection wavelength [25].

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[16] Assuming the proposed clock transition at \(\Delta = 698\) nm/\(2\pi\) with linewidth \(\Gamma_0\), the resonant dipole-dipole frequency shift \(\Gamma_0(\Delta/d)^3\) is on the order of \(10^{-5}\) Hz for atoms separated by the lattice period of \(d = 400\) nm.


