Quantum engineering of atomic phase-shifts in optical clocks

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Quantum engineering of time-separated Raman laser pulses in three-level systems is presented to produce an ultra-narrow, optical-clock transition free from light shifts and with a significantly reduced sensitivity to laser parameter fluctuations. Based on a quantum artificial complex-wave-function analytical model, and supported by a full density matrix simulation including a possible residual effect of spontaneous emission from the intermediate state, atomic phase-shifts associated to Ramsey and Hyper-Ramsey two-photon spectroscopy in optical clocks are derived. Various common-mode Raman frequency detunings are found where the frequency shifts from off-resonant states are canceled, while strongly reducing their uncertainties at the 10^{-18} level of accuracy.

The control and even the cancelation of systematic frequency shifts inherent in atom-light interactions are important tasks for high precision measurement in optical lattice clocks exploiting high quality factors from ultranarrow transitions [1]. For instance, optical clocks avoid a dephasing of the clock states through carefully designed optical traps producing controlled ac Stark shifts of those states [2]. Engineering the phase-shifts which dephase a wave-function oscillation at its natural Bohr frequency is also strongly relevant to a wide range of quantum matter experiments using trapped ions, neutral atoms, and cold molecules [3]. The standard approach for reducing those phase-shifts is the decrease of the probe laser intensity. Continuous progress in the manipulation of the laser-atom/molecule interaction has opened a new direction, quantum state engineering, in the quantum control of atomic/molecular systems [4]. For ultracold atoms, quantum engineering leads to the quantum simulation of Hamiltonians describing different physical systems, as in the synthesis of magnetic fields which exploit the coupling between internal and external atomic states [7–9]. Elsewhere, highly coherent and precisely controlled optical lattice clocks are explored for quantum simulation of many-body spin systems [10] and optical-clock systems have been proposed to probe the many-body atomic correlation functions [11].

The present work employs the quantum engineering of atom-laser interactions to produce a perfect cancelation of the frequency shifts of a given atomic/molecular level scheme. Laser pulse sequences shaped in duration, intensity and phase are modeled with a synthesized Hamiltonian, where the non-Hermitian evolution of the atomic wave-function is modified by ac Stark shifts from levels both internal and external to the probed system. This approach enables the time-dependent solution of the atomic phase-shift to be computed in analytical form, making possible a detailed exploration of laser parameters which cancel the frequency shifts. In so doing, we can easily identify ways to control these shifts to a challenging 10^{-18} level of optical clock accuracy [12].

Our approach is based on a non hermitian evolution of the atomic wavefunction whose phase is modified by the ac Stark shifts from levels internal and external to the probed system. The atomic time dependent phase-shifts are derived from such a model in a simple analytical form. This quantum engineering method allows us to explore various experimental conditions for optical clock based on alkaline earth atoms, but it can be applied to any other systems dealing with careful control of the wavefunction phase-shift.

For two-photon optical clocks, the ac Stark shifts [13–14] are induced by a large number of off-resonant driven transitions, making their suppression very difficult to realize in a simple manner. Cancelation of the frequency shifts in those optical clocks using pulsed Electromagnetically Induced Transparency and Raman (EIT-Raman) spectroscopic interrogation was explored in [15–16]. Alternatively, frequency-shifts of one-photon clock transitions were compensated and the uncertainty of the frequency measurement strongly reduced using so-called Hyper-Ramsey spectroscopy with two Ramsey pulses of different areas, frequencies and phases [17]. Refs. [18–19] pointed out that a laser frequency step applied during that pulse sequence cancels the residual light-shift and an additional echo pulse compensates both the dephasing and the uncontrolled variations of the pulse area, as tested in a recent experiment on a single trapped ^{171}Yb^+ ion clock [20]. Within our quantum engineering
approach we introduce here generalized Hyper-Raman-Ramsey (HRR) techniques for two-photon optical clocks and derive precise conditions required to operate at the $10^{-18}$ level of accuracy and stability. To reach this level of performance, the following criteria are sought: i) the clock phase shift from ac Stark effects is eliminated, ii) the shift cancelation is stable against fluctuations in the laser parameters; iii) the Ramsey fringe contrast is maximized; iv) that contrast is obtained at the unperturbed clock frequency. We satisfy simultaneously all these targets with the generalized three-level HRR techniques. In analogy with quantum simulations using ultracold atoms [22], our synthesized frequency shift may be described through an effective Hamiltonian determining the final atomic state at the end of the full pulse sequence.

We examine three-level optical bosonic clocks for $^{88}$Sr and $^{174}$Yb with the level structure of Fig. 1(a). The doubly forbidden optical clock transition is driven by a two-photon transition between atomic states $|1\rangle \equiv |1\rangle_{S0}$ and $|2\rangle \equiv |3\rangle_{P0}$ via the $|3\rangle \equiv |3\rangle_{P1}$ excited state. The $|3\rangle \rightarrow |2\rangle$ transition is driven by laser induced magnetic coupling [22]. Our quantum engineering approach could also be applied to the magnetically induced spectroscopy scheme where a static magnetic-field induces a weakly allowed transition between two same-parity states as proposed in [23] and demonstrated for $^{174}$Yb [24] and for $^{88}$Sr [25, 26]. There, the $|3\rangle \rightarrow |2\rangle$ transition is driven by static magnetic mixing. The present work explores different laser pulsed excitation schemes, as shown generally in Fig. 1(a), with a free-evolution time $T$ and (eventually) an echo pulse duration $\tau_m^{\text{echo}}$ with a laser phase reversal taking place between the initial and final interrogation times, $\tau$ and $\tau_m$, respectively. We probe the clock transi-

## Table I: Complex two-photon wave-function parameters.

| $\Delta_{LS}$ | Two-photon |
|---------------|------------|
| $\Delta_{ed}$ | $\Delta_{LS} + (\Omega_2^2 - \Omega_1^2) \frac{2\gamma_n}{\Delta + \gamma_n}$ |
| $\Delta_{eff}$ | $\Delta_{LS} - i(\Omega_2^2 - \Omega_1^2) \frac{2\gamma_n}{\Delta + \gamma_n}$ - $\gamma_0^2/2$ |
| $\Omega_{eff}$ | $2\Omega_1\Omega_2 \frac{\Delta - \gamma_n}{\Delta + \gamma_n}$ |

![Fig. 1](image1.png)

**Fig. 1:** (color online) (a) Three-level atomic system configuration for spectroscopy of a forbidden atomic transition in $^{174}$Yb and $^{88}$Sr optical lattice clocks. Atomic parameters are described in text. (b) EIT-Raman laser pulses in a Ramsey-type (on,off,on) sequence with interrogation times $\tau$, $\tau_m^{\text{echo}}$ (if inserting an echo pulse) and $\tau_m$, respectively, and free evolution at the clock frequency for a time $T$.

![Fig. 2](image2.png)

**Fig. 2:** (color online) $^{88}$Sr transition probabilities measured on $^3P_0$ clock state (left panels) and $\delta\nu$ frequency-shifts versus the common mode detuning $\Delta_0$ (right panels) for different operating parameters all satisfying the primary condition $\nu(\Delta_0^m) = 0$. Rabi frequencies $\Omega_1 = 20\sqrt{\pi\Delta_0^m/3}$ and $\Omega_2 = \Omega_1/100$, free evolution time $T = 3$ s, pulse duration $\tau = 0.1875 = 3/16$ s. In the right panel, the frequency-shifts are shown with a relative $\pm 10\%$ pulse area variation (solid and dashed lines). All curves computed from Eqs. 2 and 3. (a) Ramsey spectroscopy of the clock transition based on the $\pi/2 - T - \pi/2$ laser pulse sequence. (b) HRR scheme with the $\pi/2 - T - \pi/2$ pulse sequence including a phase reversal during the second pulse. In this case, a plateau is obtained around a magic common mode Raman detuning $\Delta_0^m/2\pi \sim 11.2$ GHz, with all conditions from Eqs. 1 satisfied.
tion using a population detection of levels $^1S_0$ or $^3P_0$ as initially proposed in ref [13].

The light shift compensation is based on control of the Bloch vector phase in the equatorial plane before the last pulse in the HRR sequence. A properly oriented vector combined with a well chosen pulse area produces full occupation of the final state, and this condition is ideally realized within the standard Ramsey sequence. However, in presence of light shift and relaxation processes, the wave-function is not properly aligned and the final pulse cannot produce the required state. Those imperfections may be compensated by a final pulse with a tuned pulse area, while the incorrect phase of the wave-function is controlled by an echo pulse, and eventually by a laser phase reversal.

Within our model, the Rabi frequencies $\Omega_i$ ($i = 1, 2$) are defined by electric and magnetic dipole couplings. The laser detunings are introduced as $\Delta_1 = \Delta_0 - \eta_1$ and $\Delta_2 = \Delta_0 - \delta - \eta_2$, where $\Delta_0$ is the common mode frequency detuning from the excited state, $\delta$ is the Raman clock frequency detuning, $\eta_i$, the light-shift correction induced by external levels for the $(i = 1, 2)$ corresponding transition, produces a correction $\Delta_{\text{ext}} = \eta_1 - \eta_2$ to the field-free clock transition. That transition also experiences an internal shift leading to the $\Delta_{LS}$ total shift listed in the first line of the Table [1]. The effective complex Rabi frequency $\Omega_{eff}$ of Table [1] determines the two-photon coupling between $[1]$ and $[2]$. The spontaneous emission rate from the intermediate state $[3]$ is given by $\Gamma$, optical relaxations are $\gamma_1 = \gamma_2 = \Gamma/2$ and a decoherence $\gamma_c$ for the lower level superposition can also be introduced. The numerical solution of the standard equations for the unitary trace density matrix used in ref [22] allows us to examine with high accuracy the linesshapes and shifts of the clock transition as investigated in [13, 17, 18, 20]. The external light shift contribution are derived from the dynamic polarizability calculations presented in the Supplemental Material.

A key point for clock precision is the quantum control of the $\Delta_{LS}$ shift, with the frequency shift of the central Ramsey clock fringe approximatively given by $\delta \nu \sim \Delta_{LS} \times \tau/T$ for $\tau \approx \tau_m$ [28]. We show that all the light shift contributions to the clock transition, from internal and external states, can be canceled by operating the excitation lasers at a ‘magic’ common mode detuning $\Delta_m$. The optical clock applications of a synthesized shift require simultaneous cancelation and stability of the phase shift at the magic detuning, as expressed by the conditions

$$\delta \nu(\Delta_m^0) = 0; \quad \left[ \frac{\partial \delta \nu}{\partial \Delta } \right]_{\Delta_m^0} = 0; \quad \left[ \frac{\partial^2 \delta \nu}{\partial \Delta^2 } \right]_{\Delta_m^0} = 0. \quad (1)$$

The magic detuning is derived within an effective wavefunction model based on a non-hermitic Hamiltonian, with a small decoherence term $\delta - i \gamma_c/2$ inside the Raman detuning condition and a term $-i \Gamma/2$ associated with spontaneous emission from the intermediate state inside the common-mode detuning $\Delta_m$.

For a two pulse sequence with pulse durations $\tau, \tau_m$ separated by dark time $T$, the $P_{\alpha\alpha}(\tau, T, \tau_m)$ final occupation of the state can be calculated, as in the Supplemental Material, and the $\Phi_{\alpha\alpha}$ atomic phase shift extracted from the probability expression. We report here the analytical expression for the phase shift $\Phi_3 = \Phi^{(3)P_0}$ determining the optical lattice clock shift measured in the two-photon HRR spectroscopy. When $T >> \tau, \tau_m$, the associated $\delta \nu(\Delta_0)$ frequency shift measured on the central fringe is

$$\delta \nu(\Delta_0) \approx -\frac{\Phi^{(3)P_0}}{2\pi(T + (\tau + \tau_m echo + \tau_m)/2)}. \quad (2)$$

In the $\Delta_0 \gg \Gamma$ regime, the role of spontaneous emission is strongly reduced and the population transfer to the excited state is negligible. In this case, the clock phase-shift depends only on pulse area. The full expression of the phase-shift corresponding to a composite sequence of three different laser pulse areas $\theta_i, \theta_j, \theta_k$ including a $\pi$ phase reversion during the second pulse is:

$$\Phi^{(3)P_0} = \arctan \left[ \frac{\tan[\theta_j] + \tan[\theta_k]}{(\tan[\theta_j] - \tan[\theta_k]) + 2 \tan[\theta_j] \tan[\theta_k]} + \frac{\omega^2 + (\Omega_{eff}^2 - \Delta_{eff}^2) \tan[\theta_j] \tan[\theta_k]}{(\tan[\theta_j] - \tan[\theta_k]) + 2 \Delta_{eff}^2 \tan[\theta_j] \tan[\theta_k]} \right]$$

$$(3)$$

where pulse areas are defined by $\tau_{i,j,k} = \omega \tau_{m echo}, \tau_m$ and $\omega^2 = \Delta_{eff}^2 + \Omega_{eff}^2$. When the second pulse area is $\theta_j \to 0$ and $\theta_i \neq \theta_k$, we recover the full expression of the HRR phase-shift expression derived perturbatively in [17] for a two-level system:

$$\Phi^{(3)P_0} = \arctan \left[ \frac{\Delta_{eff} \tan[\theta_k] + \tan[\theta_j]}{1 - \left( \frac{\Delta_{eff}}{\omega} \right)^2 \tan[\theta_j] \tan[\theta_k]} \right] \quad (4)$$

For the well-known Ramsey configuration [28] where $\theta_i =$
\[ \theta_k = \theta, \text{ that shift is} \]
\[
\Phi(3P_0) = \arctan \left[ -\frac{2\Delta m}{1 - \left( \frac{\Delta m}{\omega} \right)^2 \tan[\theta]} \right] 
\]
\approx -2 \arctan \left[ \frac{\Delta_{eff}}{\omega} \tan[\theta] \right] 
\tag{5}

From a geometrical point of view, this Ramsey phase-shift is exactly two times the Eulerian angle accumulated by a Bloch vector projection of rotating components in the complex plane using a two dimensional Cauchy-Klein representation of the spin 1/2 rotational group \cite{29, 30}.

Different examples of shift cancelation with a fringe contrast nearly equal to one in the absence of spontaneous emission are presented in the left panels of Fig. 2 neglecting the spontaneous emission. Results for the synthesized \( \delta \nu(\Delta_0) \) dependence to change in selected laser parameters are presented in the right panels of that figure and in Fig. 3 including spontaneous emission for different laser pulsed excitation schemes. The Ramsey sequence of Fig. 2(a) with \( \tau_m = \tau \ll T \) and \( \tau_m^{echo} \rightarrow 0 \) produces a quasi linear \( \delta \nu \) dependence on \( \Delta_0 = 0 \) condition of Eqs. (1) is satisfied. The HRR sequence of Fig. 2(a) produces a \( \delta \nu(\Delta_0) \) cubic dependence providing an excellent compensation and stability of the phase shift. The HRR+Phase scheme generalizing the two-level one of \cite{17}, where the laser pulse sequence includes a phase reversal during the second pulse scheme produces a synthesized plateau against fluctuations in the laser frequency around the magic detuning, as in Fig. 2(c). Note that the associated fringes still have the maximum contrast.

Our synthesized light-shift approach allows the realization of shift cancelation with insensitivity to fluctuations of the laser intensity and/or pulse duration. For instance the HRR+Phase scheme suffers from an unstable shift compensation as a function of pulse area. That dependence of the synthesized phase shift on pulse area is strongly eliminated within the synthesized phase shift approach using an additional echo pulse with phase rever-

\[ \Omega_{eff} \rightarrow -\Omega_{eff} \] phase shift is introduced within the first part of the second pulse divided in two parts with areas \( \pi \) and \( \pi/2 \), respectively. A very high stability against laser intensity was verified. Magic detunings for other HRR interrogation parameters are in Table II.

Figs. 2 and 3 examine clock interrogation with \( \Delta_0^m \) frequency detunings in the GHz range, therefore in a regime where the approximations of \( \Gamma \) and \( \gamma_c = 0 \) should be good enough. However a density matrix numerical simulation pointed out that the spontaneous emission rate from the excited state cannot be totally neglected, as in \cite{19} for a two-level system. The dashed line HRR results of Fig. 3(a) and (b) point out that at the magic detuning the slope \( \partial \delta \nu / \partial \Delta_0 \) is now different from zero, significantly compromising the insensitivity of the cancelation to laser fluctuations. The relaxation introduces into the

| \( ^{88}\text{Sr} \) | \( \Delta_{ext}/2\pi \) (GHz) | \( \langle \tau, \tau_m \rangle \) (s) | \( I_1 \) (mW/cm\(^2\)) | \( I_2 \) (W/cm\(^2\)) |
|-----|----------------|------------------|--------------|----------------|
|     |                |                  | 0.066        | 0.016          |
|     |                |                  | 4.2          | 42.5           |
|     |                |                  | 160          | 640            |
|     |                |                  | 2.35         | 37.4           |
|     |                |                  | 57.2         | 229            |
|     |                |                  | 37.4         | 229            |

| \( ^{174}\text{Yb} \) | \( \Delta_{ext}/2\pi \) (GHz) | \( \langle \tau, \tau_m \rangle \) (s) | \( I_1 \) (mW/cm\(^2\)) | \( I_2 \) (W/cm\(^2\)) |
|----------------|----------------|------------------|--------------|----------------|
|     |                |                  | 0.78          | 0.2            |
|     |                |                  | 12.5          | 122            |
|     |                |                  | 122           | 487            |
|     |                |                  | 1.3           | 0.32           |
|     |                |                  | 5.2           | 8.1            |
|     |                |                  | 8.1           | 32.3           |

TABLE II: Raman interaction parameters including laser intensities \( I_1, I_2 \) and pulse durations \( \langle \tau, \tau_m \rangle \), at fixed \( T = 5 \) s, for \( ^{88}\text{Sr} \) and \( ^{174}\text{Yb} \) two-photon interrogation leading to the reported \( \Delta_0^m \) magic detuning.

FIG. 3: (Color online) Fractional frequency-shift (in \( 10^{-18} \) unit), based on a \( \pi/2 - T - 3\pi/2 \) pulse laser sequence, in the presence of spontaneous emission relaxation. All conditions from Eqs. (1) are fulfilled at the mHz level, by adding between the preparation and detection pulses, an echo pulse of duration \( \tau_m^{echo} = 2\tau \), reversing the sign of one Rabi frequency. (a) \( ^{88}\text{Sr} \) clock frequency-shift around \( \Delta_0^m = 11.2 \) GHz with \( \Gamma/2\pi = 32 \) MHz and (b) \( ^{174}\text{Yb} \) clock frequency-shift around \( \Delta_0^m = 326.7 \) GHz with \( \Gamma/2\pi = 28.9 \) MHz. The analyses based on density matrix results (solid dots • and stars *) are reproduced by the complex wavefunction model (solid and dashed lines). Rabi frequencies as in Fig. 2 free evolution time \( T = 5 \) s.
\( \omega \) effective frequency a phase-shift modifying the coherent evolution of the atomic wavefunction. The insensitivity is recovered by applying an HRR+Echo sequence with a finely tuned length of the third detection pulse, \( \pi + 1.07 \pi/2 \) for \(^{88}\)Sr at magic detuning \( \Delta_0^{m} = 11.2 \) GHz, and \( \pi + 1.003\pi/2 \) for \(^{174}\)Yb at \( \Delta_0^{m} = 326.7 \) GHz, as shown by stars/dots and continuous lines in Figs. 3(a) and 3(b), respectively. Note that including relaxation, both models, wavefunction and density matrix results are fully consistent for high common mode detunings, for instance in the prediction of the required third pulse area.

Our wavefunction approach is very efficient for deriving the clock shift under different operating conditions and determining the laser parameters for the optical clock shift cancelation at the mHz level. We present its application to a Hyper-Ramsey-Ramsey spectroscopic method, based on two pulses with areas \( \pi/2 \) and \( 3\pi/2 \), that cancels the light-shift and efficiently suppresses the sensitivity to laser field fluctuations without the additional frequency step on the laser frequency required in refs [18–20]. Synthesized phase-shifts are important for high precision measurement using Ramsey spectroscopy [31, 32]. Hyper-Ramsey probing schemes based on magic wavelengths [33] and any Raman-Ramsey or two-photon excitation strongly detuned from the intermediate excited state [34]. The light-insensitive two-photon approach could be applied also to atom interferometry [35, 36] and other type of Doppler-free two-photon configurations [37, 38].

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