High-Accuracy Optical Clock via Three-Level Coherence in Neutral Bosonic ^{88}Sr

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An optical atomic clock scheme is proposed that utilizes two lasers to establish coherent coupling between the $5s^2\,{}^1S_0$ ground state of ^{88}Sr and the first excited state, $5s5p\,{}^3P_0$. The coupling is mediated by the broad $5s5p\,{}^1P_1$ state, exploiting the phenomenon of electromagnetically induced transparency. The effective linewidth of the clock transition can be chosen at will by adjusting the laser intensity. By trapping the ^{88}Sr atoms in an optical lattice, long interaction times with the two lasers are ensured; Doppler and recoil effects are eliminated. Based on a careful analysis of systematic errors, a clock accuracy of better than 2×10^{-17} is expected.

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Armed with superior resonance quality factors, optical atomic clocks based on single trapped ions [1–3] or a collection of laser cooled neutral atoms [4,5] are expected to outperform microwave-based atomic clocks in the near future, with potential stability $\sim 1 \times 10^{-16}$ at 1 s and accuracy below 1×10^{-17}. While a large number of quantum absorbers in a neutral atom system provide an advantage in the enhanced short-term frequency stability, atomic motion during the probe phase seriously limits the attainable accuracy. A competitive proposal is to localize neutral cold atoms spatially in a Lamb-Dicke regime, while the trapping potential is designed such that its presence does not influence the clock transition frequency [6]. This scheme can be potentially realized using a far-off-resonance dipole trap operating at a wavelength where the ground and the excited states of the clock transition experience exactly the same ac Stark shifts. In particular, if the atoms are confined near the ground vibrational levels in an optical lattice, frequency shifts associated with Doppler, collision, and recoil will be reduced to negligible levels.

To realize such a scheme, it is essential that the polarizabilities of the two clock states are matched to high accuracy at the "magic" wavelength [7]. States with scalar polarizabilities are preferred, to avoid the problem of the complex and sometimes uncontrolled light polarization inside an optical lattice. The fermionic isotope, ^{87}Sr, offers a nearly satisfactory solution [7,8]. The nuclear magnetic dipole moment of ^{87}Sr makes the $5s^2\,{}^1S_0 - 5s5p\,{}^3P_0$ transition weakly dipole allowed, with a predicted linewidth of about 1 mHz [7,9,10]. The two clock states have small electronic angular momenta (due to hyperfine mixing) and their polarizabilities are nearly scalar. However, the hyperfine structure can cause a clock frequency shift (possibly as large as 10 Hz) by fluctuations of the lattice light polarization [7]. The large nuclear spin ($I = 9/2$) also brings complexity in state preparation and field control.

In this Letter, we propose a scheme for an optical clock that is based on states of true scalar nature, namely, $5s^2\,{}^1S_0 - 5s5p\,{}^3P_0$ of ^{88}Sr (with $I = 0$). A direct transition between these two states is, of course, completely forbidden, and the clock scheme is based instead on three-level quantum coherence established by two probe laser frequencies. A scheme similar to what is discussed here was investigated for trapped ions, with a measured 75 kHz coherence linewidth and a projected clock accuracy in the 10^{-15} range [11]. Recently, for alkaline-earth atoms a scheme based on four levels and three probe lasers has emerged [12], although an estimate of its expected accuracy is not yet available. The analysis we performed here indicates that a high-accuracy optical clock based on a multilevel scheme requires rather complex control of light shifts for several intermediate levels. Furthermore, a comparison of the requirements on optical frequency mixing and laser phase-locking indicates a more straightforward implementation for the present scheme.

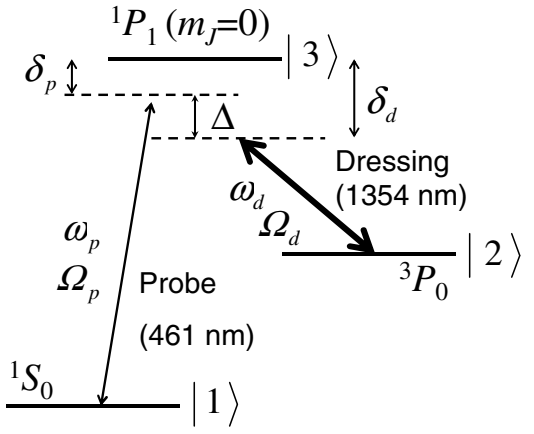

FIG. 1. Three-level coherence in neutral ^{88}Sr. The two clock levels are $5s^2\,{}^1S_0$ and $5s5p\,{}^3P_0$. By applying the probe and the dressing laser as shown, a coherent coupling is established between the clock levels, mediated by the $5s5p\,{}^1P_1$ level.

Our proposal rests on the three-level lambda-type scheme shown in Fig. 1. A promising candidate in this context is Sr [6–8,13], which we consider here in detail. A similar scheme is conceivable for bosonic isotopes of other alkaline-earth species (including Yb). Since the ground state ($5s^2\,{}^1S_0$)—subsequently denoted by the state vector $|1\rangle$—and the first excited state ($5s5p\,{}^3P_0$)—$|2\rangle$—of ^{88}Sr are characterized by a total angular momentum of $J = 0$, electric and magnetic one-photon transition matrix elements between these two states vanish to any multipole order. $|2\rangle$ decays radiatively via $E1M1$ two-photon emission after a lifetime of a few thousand years [9]. For practical purposes, it is therefore legitimate to set the decay width of $|2\rangle$ equal to zero. The $5s5p\,{}^1P_1$ state, which is referred to as $|3\rangle$ in our scheme, can be reached from $|1\rangle$ via $E1$ one-photon absorption using a *probe* laser at a wavelength of 461 nm (frequency ω_p). One $M1$ photon from a *dressing* laser at 1354 nm (frequency ω_d) is needed to resonantly couple $|2\rangle$ and $|3\rangle$. The latter decays primarily to $|1\rangle$. The decay width is $\gamma = 2\pi \times 32$ MHz.

Assuming linear polarization of both the probe and the dressing lasers, and assuming that the electric component of the probe laser (amplitude $\mathcal{E}_0$) and the magnetic component of the dressing laser (amplitude $\mathcal{B}_0$) are aligned along the same axis, level 3 is characterized by $m_J = 0$ for our purposes. Let $\Omega_p = \langle 3|D_0|1\rangle\mathcal{E}_0/\hbar$ denote the Rabi frequency associated with electric-dipole (D_0) coupling between levels 1 and 3. Similarly, $\Omega_d = \langle 3|M_0|2\rangle\mathcal{B}_0/\hbar$ is the Rabi frequency referring to the dressing process (M_0, magnetic dipole). Ω_p and Ω_d may be taken to be real.

We first analyze the dressed-atom problem within the subspace of $|2\rangle$ and $|3\rangle$ in the absence of the probe laser. The equations of motion in the interaction picture read

$$i\dot{c}_2(t) = \frac{\Omega_d}{2} e^{i\delta_d t} c_3(t), \tag{1}$$

$$i\dot{c}_3(t) = \frac{\Omega_d}{2} e^{-i\delta_d t} c_2(t) - i\frac{\gamma}{2} c_3(t), \tag{2}$$

where $c_2(t)$ and $c_3(t)$ represent probability amplitudes and the rotating-wave approximation has been applied. The dressing laser detuning is $\delta_d = \omega_d - (\omega_3 - \omega_2)$ [ω_i stands for the unperturbed frequency of $|i\rangle$, $i = 1, 2, 3$].

Because of the action of Ω_d, some $|3\rangle$ character is admixed to $|2\rangle$ (and vice versa), so that a long-lived dressed state, $|\tilde{2}\rangle$, emerges whose decay rate can be adjusted through the dressing laser intensity. It is easily seen that to leading order with respect to $\Omega_d/(\delta_d + i\gamma/2)$

$$|\tilde{2}\rangle = \{|2\rangle e^{-i\omega_2 t} + b|3\rangle e^{-i(\omega_3+\delta_d)t}\} e^{-i\beta t}, \tag{3}$$

$$b = \frac{1}{2}\frac{\Omega_d}{\delta_d + i\gamma/2}, \qquad \beta = \frac{1}{2}\Omega_d b. \tag{4}$$

It follows from the equation for β that for small detuning the decay rate of $|\tilde{2}\rangle$ is $\tilde{\gamma} = \Omega_d^2/\gamma$. The dressed state deriving primarily from $|3\rangle$ decays with a rate that is virtually identical to γ.

Using the atomic structure code described in Ref. [9], which accurately treats electron correlation and spin-orbit effects in the valence shell, we obtain for the magnetic coupling matrix element $|\langle 3|M_0|2\rangle| = 0.022\mu_B$. Hence, if an effective width $\tilde{\gamma}$ of, say, 1 mHz is desired (similar to the ^{87}Sr case), a dressing laser intensity of 3.9 mW/cm^2 must be chosen and is used for numerical results presented in this Letter. (By decreasing the intensity, the effective linewidth can be made even narrower.) Under these circumstances, the parameter b in Eqs. (3) and (4), which describes the degree of admixture of the $|3\rangle$ character in the long-lived dressed state, has a magnitude of $<10^{-5}$. In other words, the dressed states in this scheme are hardly distinguishable from bare atomic states. We exploit this fact later when estimating level shifts from sources not included in the simple three-level picture of Fig. 1.

Since there is no direct coupling between $|1\rangle$ and $|2\rangle$, it is apparent from Eq. (3) that the transition associated with the long-lived dressed state can be probed from the ground state only by tuning ω_p in the vicinity of $\omega_3 + \delta_d - \omega_1$, i.e., in the vicinity of the broad absorption profile of level 3. Hence, $\delta_p = \omega_p - (\omega_3 - \omega_1) \sim \delta_d$. The long-lived and the short-lived states are excited in a coherent fashion by the probe laser. They interfere and give rise to a narrow dip structure in the absorption profile—an effect known as *electromagnetically induced transparency* (EIT) [14]. This dip is more than 10^{10} times narrower ($\tilde{\gamma} = 2\pi \times 1$ mHz) than the bare absorption profile of level 3. The broad background is therefore essentially constant across the narrow structure.

This quantum interference may be treated, to a first approximation, within a wave function-based approach, but it fails to take into consideration the repopulation of $|1\rangle$ via spontaneous emission from $|3\rangle$. More importantly, the loss of coherence between levels 1 and 2 cannot be described. Decoherence is caused by lattice-induced spontaneous emission, atomic tunneling between adjacent sites in the optical lattice and ensuing collisions between the atoms. Another factor included is the loss of phase coherence between the probe and the dressing laser. Utilizing standard density-matrix theory [15], the absorption rate from the ground state is

$$W_{\text{abs}} = \frac{\Omega_p^2}{\gamma} \frac{\Delta^2 + \gamma_c\{\gamma_c + \tilde{\gamma}/2\}}{[2\Delta(\Delta + \delta_d)/\gamma - \{\gamma_c + \tilde{\gamma}/2\}]^2 + [\Delta + 2\gamma_c(\Delta + \delta_d)/\gamma]^2}, \tag{5}$$

where $\Delta = (\omega_p - \omega_d) - (\omega_2 - \omega_1)$, and γ_c stands for the loss rate of coherence between $|1\rangle$ and $|2\rangle$.

If the dressing laser is off, or if $\gamma_c \gg \tilde{\gamma}$, the simple Lorentzian profile connected with the transition from $|1\rangle$ to $|3\rangle$ is recovered. Otherwise, if we disregard γ_c for a moment, the absorption rate vanishes at $\Delta = 0$. A deep, narrow dip occurs in the absorption profile [16], and the width of the EIT dip for $\gamma_c = 0$ is

$$\Delta_{\mathrm{FWHM}} = \tilde{\gamma}\left\{1 - 8\frac{\delta_d^4}{\gamma^4} + O\left(\frac{\delta_d^6}{\gamma^6}\right)\right\}. \quad (6)$$

This demonstrates that not only the position of the EIT dip, but also its width, is very insensitive with respect to fluctuations of δ_d. A variation of the dressing laser frequency by as much as 100 kHz induces a relative change of the EIT width of less than 10^{-7}. The effect is illustrated in Fig. 2(a) ($\gamma_c = 0$). Hence, by measuring W_{abs} as a function of $\omega_p - \omega_d$, the clock frequency $\omega_2 - \omega_1$ can be determined with high accuracy by tracking the position of the dip. Neither ω_p nor ω_d must be particularly stable, and only the difference needs to be stabilized.

Also shown in Fig. 2(a) is the absorption profile near $\Delta = 0$ for relatively large detuning ($\delta_d = 5 \times \gamma$). Overall, the signal amplitude is drastically reduced in this case. There is a narrow absorption peak in the immediate vicinity of $\Delta = 0$, but, as can be seen in Fig. 2(b), this peak all but disappears as soon as decoherence becomes appreciable. [We have chosen $\gamma_c = \tilde{\gamma}$ in Fig. 2(b).] On the other hand, for small detuning the EIT dip is still clearly visible, even though it is not as deep as in the absence of decoherence and it has become broader. The position of the EIT minimum and the insensitivity of the EIT width with respect to laser frequency (δ_d or δ_p) fluctuations of 100 kHz or so are essentially unaffected.

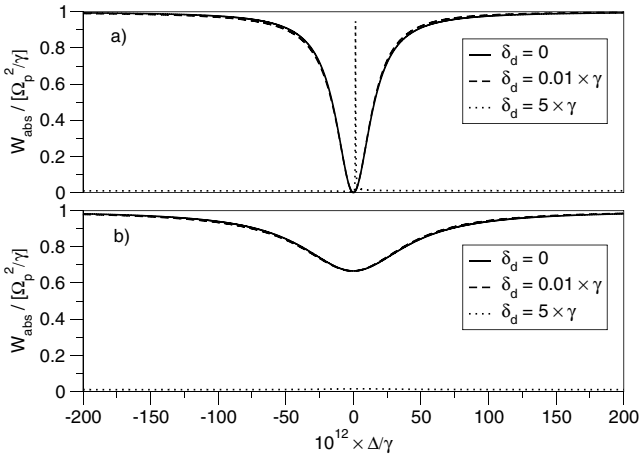

FIG. 2. Absorption rate from the ground state of ^{88}Sr in the presence of the two laser frequencies indicated in Fig. 1. The laser-induced width, $\tilde{\gamma}$, is $2\pi \times 1$ mHz. (a) Loss of phase coherence between levels 1 and 2 is neglected, $\gamma_c = 0$. Note that the curves for $\delta_d = 0$ and $\delta_d = 0.01 \times \gamma = 2\pi \times 320$ kHz are virtually indistinguishable. (b) Macroscopic decoherence and laser-induced decay are comparable, $\gamma_c = \tilde{\gamma}$.

The discussion has so far covered only resonant couplings within the span of $|1\rangle$, $|2\rangle$, and $|3\rangle$. In order to evaluate the potential accuracy of the proposed clock scheme, we need to consider the ac Stark shifts due to nonresonant electric-dipole coupling of $|1\rangle$, $|2\rangle$, and $|3\rangle$ to states inside and outside the three-level subspace. Our calculation of these shifts employs accurate experimental and theoretical data on excitation energies and oscillator strengths of atomic Sr [6,17,18]. For a probe laser intensity of 10 μW/cm^2, levels 1 and 2 are ac Stark shifted by less than 1 mHz. The dressing laser, operating at an intensity of 3.9 mW/cm^2, causes an ac Stark shift of levels 1 and 2 of -41 and -20 mHz, respectively. Thus, the clock frequency is systematically shifted by about $+21$ mHz. Since the intensity of the dressing laser can be stabilized to better than 1%, we conclude that the ac Stark effect due to the probe and dressing lasers can be experimentally characterized at the sub-mHz level.

To allow for a sufficiently long interrogation time and eliminate systematic frequency shifts associated with atomic motion, the atoms will be confined in a Lamb-Dicke regime in an optical lattice at the magic wavelength for the clock transition, as shown in Fig. 3(a). The lattice trapping field causes, to leading order, no net frequency shift of the clock transition as the level shifts of 1 and 2 [150 kHz well depth in Fig. 3(a), under a lattice laser intensity of 10 kW/cm^2] are exactly matched. (The studies in Ref. [7] suggest that higher-order ac Stark effects are negligible at this lattice laser intensity.) Level 3 in our scheme, however, is shifted relative to levels 1 and 2, and this shift depends on the local intensity an atom experiences in the lattice. We calculate a relative shift of -250 kHz (maximum in magnitude), at a lattice laser intensity of 10 kW/cm^2 and a magic wavelength of 813.5 nm [8]. Figure 3(a) shows the lattice-induced shift of level 3 in comparison to γ. This shift is similar—in effect and in magnitude—to the drift of the probe and dressing laser frequencies. It has, therefore, an equally negligible effect on the properties of the EIT dip.

To make the final estimate of the clock accuracy, the level shifts shown in Fig. 3(a) contain the ac Stark shifts due to the simultaneous presence of ω_p and ω_d laser fields as discussed above. The lattice field induces a spontaneous emission limited linewidth of the clock transition at $\sim$15 mHz. The detuning and decoherence (laser frequency variations, atomic motion inside lattice, spontaneous emission, etc.) related changes in the EIT linewidth may lead to a small shift in the experimental search of the line center, but the shift magnitude is estimated to be under 10^{-18}. The blackbody radiation induced clock frequency shift is estimated to be -0.54 Hz at 300 K, with a temperature coefficient of -7.5 mHz/K (the associated linewidth broadening is on the order of 1 mHz and is thus negligible). Finally, the strict scalar nature of the clock transition reduces the light polarization and stray magnetic field-

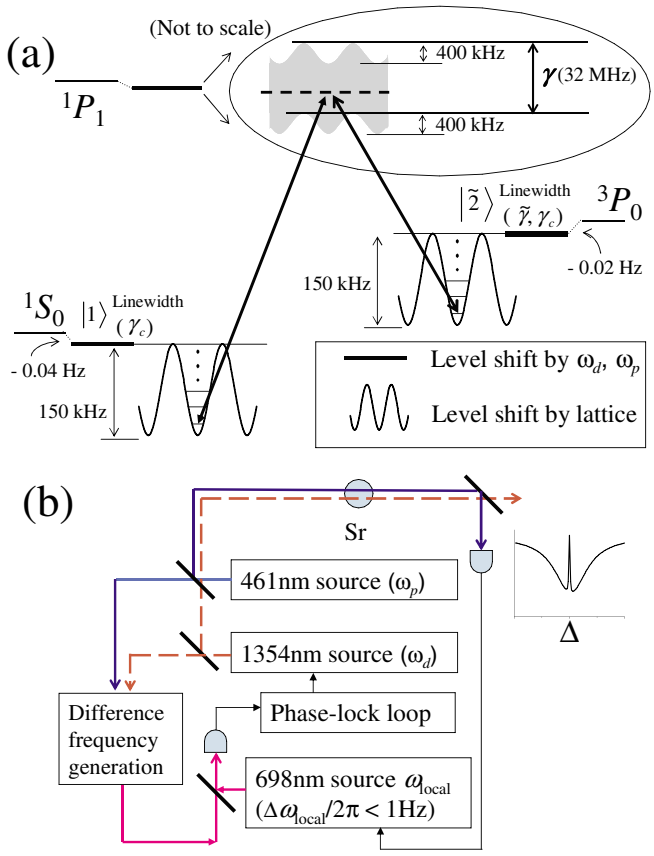

FIG. 3 (color online). (a) Lattice-confined atoms for use in the EIT clock scheme. The ac Stark shift due to the dressing laser on the clock transition is shown, as are the level shifts associated with the lattice trapping field. The expanded view of the $^1P_1$ state compares the lattice-induced level shift versus the linewidth γ. Atoms are cooled to the Lamb-Dicke regime, predominantly occupying the ground vibrational level in the lattice. (b) Experimental implementation of the EIT-based optical clock. A prestabilized narrow-linewidth local oscillator at 698 nm establishes a tight frequency track between the probe and dressing lasers. This enables a probe of the narrow EIT resonance, which in turn provides a long-term clock reference for the 698 nm local oscillator.

induced frequency shift below 10^{-18}. In view of the available control accuracy of the laboratory temperature and the dressing laser intensity, the overall accuracy limit is estimated to be smaller than 8 mHz, or 2×10^{-17}.

This EIT-based clock scheme is implemented as shown in Fig. 3(b). A narrow-linewidth (< 1 Hz) laser at 698 nm (ω_{local}) is developed by prestabilizing ω_{local} to a stable, passive optical cavity. The difference frequency, $\omega_p - \omega_d$, between the probe laser and the dressing laser, generated by a nonlinear optical crystal, is phase locked to the 698 nm local oscillator. One of the two lasers, such as ω_d, as shown in Fig. 3(b), can be used as the slave to accomplish the optical phase lock loop. Thus, $\omega_p - \omega_d = \omega_{\text{local}}$. This is prestabilization, implemented before ω_p and ω_d interact with the atoms. ω_p and ω_d may fluctuate on the order of 100 kHz, but their difference fluctuates <1 Hz.

The frequency of ω_{local} can be precisely scanned with reference to a stable cavity mode, which, of course, has an inevitable slow frequency drift due to the material nature of the reference. However, precise scanning of $\omega_p - \omega_d$ through the EIT resonance enables one to activate a slow feedback loop to stabilize $\omega_p - \omega_d$ to the value of $\omega_2 - \omega_1$; i.e., the quantum resonance corrects the long-term drift of the cavity resonance according to the clock transition frequency. The feedback is applied directly on the 698 nm local oscillator (ω_{local}) to ensure $\Delta = 0$.

The tunability of the linewidth, the exquisite insensitivity with respect to light polarization in the optical lattice, the straightforward state control, and the small, controllable ac Stark shifts promise that our EIT-based clock scheme using bosonic ^{88}Sr will represent a practical and robust approach for optical atomic clocks with superior short-term stability and competitive accuracy.

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