Optical Clocks Based on Ultranarrow Three-Photon Resonances in Alkaline Earth Atoms

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A sharp resonance line that appears in three-photon transitions between the $^1S_0$ and $^3P_0$ states of alkaline earth and Yb atoms is proposed as an optical frequency standard. This proposal permits the use of the even isotopes, in which the clock transition is narrower than in proposed clocks using the odd isotopes and the energy interval is not affected by external magnetic fields or the polarization of trapping light. With this method, the width and the rate of the clock transition can, in principle, be continuously adjusted from the MHz level to sub-mHz without loss of signal amplitude by varying the intensities of the three optical beams. Doppler and recoil effects can be eliminated by proper alignment of the three optical beams or by point confinement in a lattice trap. Light-shift effects on the clock accuracy can be limited to below a part in 10^{18}.

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Atomic clocks using optical transitions with high intrinsic line Q offer unprecedented opportunities for improved metrology standards [1] and tests of fundamental physics [2]. Recently, much attention has focused on using the forbidden $^1S_0 \rightarrow {}^3P_0$ transitions in alkaline earth and Yb atoms, which become weakly allowed in the odd isotopes through the hyperfine interaction of the nuclear spin [3]. The atoms can be confined in an optical lattice trap to eliminate the first-order Doppler and recoil effects, and the trap wavelength can be set to the "magic" value at which the ground and excited states undergo the same light shift, leaving the clock transition unshifted [3].

Here we discuss the alternative of basing a clock on the sharp lines that appear in three-photon transitions between the $^1S_0$ and the $^3P_0$ states. This method permits the use of the even isotopes, in which the clock transition is narrower than in the odd isotopes and the energy interval is not affected by external magnetic fields or the polarization of trap light at the magic wavelength. The three-photon scheme offers other interesting options. By varying the intensities of the three optical beams, the rate, and hence the width, of the clock transition can, in principle, be continuously adjusted from the MHz level to sub-mHz without loss of signal amplitude. Furthermore, Doppler and recoil effects can be eliminated by point confinement in a lattice trap at the magic wavelength or by proper alignment of the three optical beams. Finally, the three beams can be produced using lasers with a moderate amount of frequency noise, and a spectrally narrow beam at the exact clock frequency can be generated by mixing the three beams in nonlinear crystals and applying standard laser stabilization techniques to just one of the lasers.

The proposed frequency standard consists of four atomic energy levels interacting with three light fields: a strong coupling field, a weak coupling field, and a probe field, as shown in Fig. 1(a). We assume precise relative phase coherence among these fields; later we discuss how the stabilization mentioned above can establish this coherence. Our general results are applicable to any of the alkaline earth atoms and Yb, but, as an example, the four levels for the specific case of Yb are shown in Fig. 1(b). Relevant transitions and wavelengths for Ca, Sr, and Yb are shown in Table I. The idea is based on the concept of electromagnetically induced transparency and absorption (EITA) [4]. The three light fields connect the state $|1\rangle$ (the $^1S_0$ ground state) to the state $|4\rangle$ (the metastable $^3P_0$ state) via two short-lived intermediate states, $|2\rangle$ and $|3\rangle$ ($^3P_1$ and $^3S_1$), but the width and position of the three-photon EITA features are determined by the narrow initial and final states and not the relatively broad intermediate states. In the case of the even isotopes trapped in an optical dipole trap [3], the ground and the metastable state both have zero total angular momentum, so their energy difference is unaffected by external magnetic fields or trapping light polarization. Here, for simplicity, we assume only one sublevel in each intermediate state ($^3P_1$ and $^3S_1$) participates, and ignore any effect of polarization of the light fields. We also ignore other decays of $^3S_1$, including those to the metastable $^3P_2$ state, from which atoms can be removed by a separate laser.

Under the rotating wave approximation, the four-level atomic system coupled to the three light fields can be

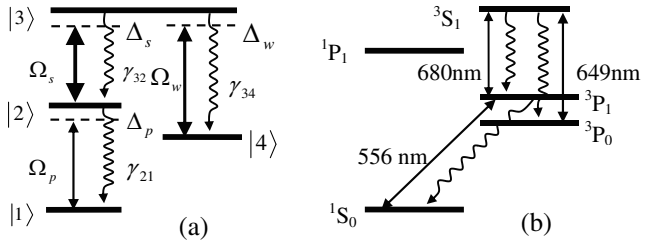

FIG. 1. (a) Energy level structure and optical couplings of the four-level atomic system for making an atomic optical frequency standard. (b) Specific case of Yb as an example for the scheme in (a).

described by the following density matrix equation:

$$\frac{d\rho(t)}{dt} = -\frac{i}{\hbar}[H, \rho(t)] + L[\rho(t)], \tag{1}$$

where $\rho(t)$ is the atomic density matrix. The sum of the diagonal elements satisfies the probability normalization, i.e., $\rho_{11} + \rho_{22} + \rho_{33} + \rho_{44} = 1$.

The matrix for the system Hamiltonian is defined by

$$H = \hbar \begin{bmatrix} 0 & -\Omega_p/2 & 0 & 0 \\ -\Omega_p^*/2 & -\Delta_p & -\Omega_s/2 & 0 \\ 0 & -\Omega_s^*/2 & -\Delta_s - \Delta_p & -\Omega_w/2 \\ 0 & 0 & -\Omega_w^*/2 & \Delta_w - \Delta_s - \Delta_p \end{bmatrix}, \tag{2}$$

where Ω_p, Ω_s, and Ω_w (see Fig. 1) are the complex Rabi frequencies associated with the couplings of the probe field, the strong coupling field, and the weak coupling field to atomic transitions $|1\rangle \rightarrow |2\rangle$, $|2\rangle \rightarrow |3\rangle$, and $|3\rangle \rightarrow |4\rangle$, respectively. $\Delta_p = \omega_p - \omega_{21}$, $\Delta_s = \omega_s - \omega_{32}$, and $\Delta_w = \omega_w - \omega_{34}$ are the detunings between the field frequencies, ω_p, ω_s, and ω_w, and the atomic resonance frequencies, ω_{21}, ω_{32}, and ω_{34}, respectively. The Liouvillian matrix $L[\rho(t)]$, defined in Eq. (3), describes relaxation by spontaneous decay. Because the ground state $|1\rangle$ and the metastable state $|4\rangle$ normally have very long coherence times (typically much larger than seconds), here we assume there is no decay from these states. Decay rates γ_{32} and γ_{34} give the decay from state $|3\rangle$ to states $|2\rangle$ and $|4\rangle$, respectively, and typically have values of order 10^7 s^{-1} or larger. γ_{21} is the rate from $|2\rangle$ to $|1\rangle$ (the intercombination transition $^3P_1 \rightarrow {}^1S_0$), and the value ranges from about 10^6 s^{-1} in Yb to about 10^3 s^{-1} in Ca. In the following numerical calculations, we choose $\gamma_{32} = \gamma_{34} = 10\gamma_{21} = \gamma$, where γ denotes the atomic characteristic decay rate, and the relative value of γ_{21} is appropriate for Yb. With Sr and Ca, the basic behavior is similar, but some numerical details are markedly different because γ_{21} is much smaller.

TABLE I. Atom and corresponding optical wavelength candidates for forming the scheme in Fig. 1.

Atom	Probe ($^1S_0$-$^3P_1$)	Strong ($^3P_1$-$^3S_1$)	Weak ($^3S_1$-$^3P_0$)	Clock
Yb	556 nm	680 nm	649 nm	578 nm
Sr	689 nm	688 nm	679 nm	698 nm
Ca	657 nm	612 nm	610 nm	659 nm

$$L[\rho(t)] = \begin{bmatrix} \gamma_{21}\rho_{22} & -\gamma_{21}\rho_{12}/2 & -(\gamma_{32}+\gamma_{34})\rho_{13}/2 & 0 \\ -\gamma_{21}\rho_{21}/2 & -\gamma_{21}\rho_{22} + \gamma_{32}\rho_{33} & -(\gamma_{21}+\gamma_{32}+\gamma_{34})\rho_{23}/2 & -\gamma_{21}\rho_{24}/2 \\ -(\gamma_{32}+\gamma_{34})\rho_{31}/2 & -(\gamma_{21}+\gamma_{32}+\gamma_{34})\rho_{32}/2 & -(\gamma_{32}+\gamma_{34})\rho_{33} & -(\gamma_{32}+\gamma_{34})\rho_{34}/2 \\ 0 & -\gamma_{21}\rho_{42}/2 & -(\gamma_{32}+\gamma_{34})\rho_{43}/2 & \gamma_{34}\rho_{33} \end{bmatrix}. \tag{3}$$

Considering the system in steady state ($d\rho/dt = 0$) and retaining the probe field only to first order in $|\Omega_p|^2$, we obtain the following expression for the absorption rate of the probe light per atom:

$$\mathrm{Im}(\Omega_p\rho_{21}) = \mathrm{Im}\left(\frac{|\Omega_p|^2\rho_{11}}{-2\Delta_p - i\gamma_{21} + M}\right), \tag{4}$$

where

$$M = \frac{|\Omega_s|^2}{2(\Delta_s + \Delta_p) + i(\gamma_{34} + \gamma_{32}) + \frac{|\Omega_w|^2}{2(\Delta_w - \Delta_s - \Delta_p)}}.$$

When $|\Omega_p| \ll |\Omega_w|$, γ_{21}, γ_{32}, $\gamma_{34} < |\Omega_s|$, a very sharp absorption peak appears due to EITA, as shown by the solid line in Fig. 2. Here, for illustration, we chose $|\Omega_p| = 0.0001\gamma$, $|\Omega_w| = 0.01\gamma$, $|\Omega_s| = 3\gamma$, and $\Delta_s = \Delta_w = 0$. The sharp peak is much narrower than the normal single-photon absorption peak, shown by the dashed line in Fig. 2 with $|\Omega_w| = |\Omega_s| = 0$.

Close to the sharp absorption peak, when the three-photon detuning $\Delta \equiv \Delta_s + \Delta_p - \Delta_w$ is very small [i.e., when $|\Delta| \ll |\Omega_w|^2/|2(\Delta_s + \Delta_p) + i(\gamma_{34} + \gamma_{32})|$], Eq. (4) takes a simple form that exhibits most of the important features:

$$\mathrm{Im}(\Omega_p\rho_{21}) = \rho_{11}|\Omega_p|^2\{\gamma_{21}[1 + 4(\Delta - \bar{\Delta})^2/W^2]\}^{-1}, \tag{5}$$

where $W = \gamma_{21}|\Omega_w|^2/|\Omega_s|^2$ is the full width of the three-photon resonance and $\bar{\Delta} = -\Delta_p|\Omega_w|^2/|\Omega_s|^2$ is the shift in the resonance peak from $\Delta = 0$ due to individual photon mistunings. Both this width and shift become arbitrarily

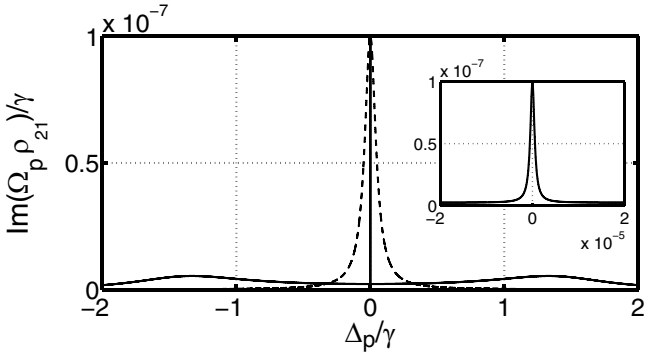

FIG. 2. Absorption rate per atom of the probe light field under the condition of electromagnetically induced transparency and absorption (solid line) or the normal condition of no coupling light fields (dashed line). The inset shows a magnification of the sharp peak.

small as $|\Omega_s|$ increases, as illustrated in Fig. 3(a). Thus the linewidth of the resonance can be very narrow and give a very high Q value. Also, the height of the peak in Eq. (5) clearly equals the full single-photon absorption rate $\rho_{11}|\Omega_p|^2/\gamma_{21}$, as is shown in Fig. 2.

Under the further assumption that $|\Omega_s/\Omega_w|^2 \gg 1$, the position of the sharp absorption peak in Eq. (5) may be written in terms of the probe laser frequency as

$$\Delta_p^{\text{peak}} = \Delta_w - \Delta_s - W(\Delta_w - \Delta_s)/\gamma_{21}. \tag{6}$$

This shows that the shift in the peak position from the atomic intrinsic three-photon resonance frequency is less than the linewidth W provided the detuning of the individual fields is controlled well enough that $|\Delta_s - \Delta_w| < \gamma_{21}$. In this case, if the probe laser frequency is locked to the narrow peak, although the individual frequencies of the strong and weak coupling laser fields might still fluctuate, the algebraic sum of the three laser frequencies is locked very close to the three-photon resonance, i.e., $\omega_p + \omega_s - \omega_w \approx \omega_{21} + \omega_{32} - \omega_{34}$. External magnetic fields or optical trapping fields can shift the intermediate states relative to the $|1\rangle$ and $|4\rangle$ states, and thereby shift the three-photon resonance peak a small amount according to Eq. (6). A more detailed analysis of such effects, as well as the effect of polarization of the individual optical fields will appear elsewhere [5].

Because the signal-to-noise ratio of an error signal determines the linewidth of a laser locked to a frequency discriminator [6], probe light intensity is normally increased until the absorption rate broadens the line. These same considerations apply here. As $|\Omega_p|^2$ increases, the probed sharp absorption peak is broadened and, when $(\Delta_w - \Delta_s)$ is finite, pushed away from the intrinsic three-photon resonance frequency, as shown in Fig. 3(b). Here

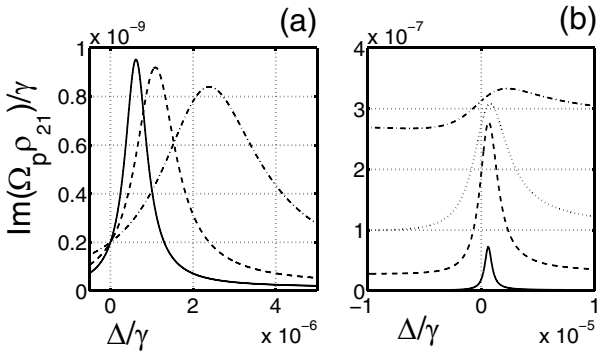

FIG. 3. (a) Narrow absorption peaks under different $|\Omega_s|$: 2γ (dash-dotted line), 3γ (dashed line), or 4γ (solid line), where $|\Omega_p| = 0.000\,01\gamma$, $|\Omega_w| = 0.01\gamma$, $\Delta_s = 0.05\gamma$, and $\Delta_w = -0.05\gamma$. (b) Narrow absorption peaks under different probe light intensities: $|\Omega_p|^2 = (4 \times 10^{-6})\gamma^2$ (dash-dotted line), $(1 \times 10^{-6})\gamma^2$ (dotted line), $(2.5 \times 10^{-7})\gamma^2$ (dashed line), and $(1 \times 10^{-8})\gamma^2$ (solid line), where $|\Omega_s| = 4\gamma$, $|\Omega_w| = 0.01\gamma$, $\Delta_s = 0.05\gamma$, and $\Delta_w = -0.05\gamma$.

Eq. (1) is solved numerically, thereby reflecting the nonlinear effect of $|\Omega_p|^2$. Thus, in practice, we have to make a proper trade-off between the distortion of the sharp absorption peak and the signal magnitude. To control the shifts shown in Eq. (6) and Fig. 3(b), the detuning $\Delta_w - \Delta_s$ can be held close to zero with sufficient accuracy by referencing the individual laser frequencies to their atomic transitions.

Numerically, in the case of Yb, a strong coupling laser intensity of 4 mW/cm^2 together with weak coupling and probe laser intensities of 1 nW/cm^2 and 10 pW/cm^2 would yield a sharp EITA resonance of width $W = 2\pi(2 \times 10^{-2}$ Hz) and peak contrast of 10^{-2} s^{-1} in the photon absorption rate per atom, amounting to 20% of the background absorption rate. A detuning of one of the lasers by 2 kHz from its transition resonance would shift the narrow clock peak by only a part in 10^{18}. Assuming 10^6 atoms, the linewidth and signal strength would yield a purely statistical precision better than a part in 10^{18} in 100 s.

We must also consider the off-resonant couplings of the three laser fields. The only significant effect comes from the off-resonant light shift of the clock levels, $\delta\omega_{\text{OR}}$, caused by the strong coupling laser field. We have calculated this shift in Yb using results from Porsev *et al.* [3]; for the strong coupling laser wavelength of 680 nm and intensity 4 mW/cm^2 used in the estimates above, we find $\delta\omega_{\text{OR}} = 2\pi(1 \times 10^{-2}$ Hz). It should not be difficult to measure this shift and control it to 5%, in order to maintain the part in 10^{18} accuracy quoted above. These estimates show the kind of potential inherent in this proposed clock scheme.

The Doppler effect is always a major cause of shifts and broadening of sharp optical resonances even for cold atoms. Frequency standards with free atoms typically use the method of Ramsey interference to eliminate the first-order Doppler effect, while trapped ion standards and neutral atom optical lattices make use of Lamb-Dicke confinement. Lamb-Dicke confinement in an optical lattice at the magic wavelength should work quite well for our three-photon proposal as well. When a high power confinement laser is undesirable, as perhaps in space, a third technique is available for three-photon transitions: Doppler-free alignment of the three laser beams [7]. If the light wave vectors satisfy the phase matching relation:

$$\mathbf{k}_p + \mathbf{k}_s - \mathbf{k}_w = 0, \tag{7}$$

the Doppler frequency shift is zero regardless of the atomic velocity. Atoms with arbitrary velocities can therefore contribute to the probe signal effectively and hence enhance the signal-to-noise ratio. It is therefore equivalent to the Ramsey method and superior to conventional Doppler-free saturated absorption spectroscopy, in which only atoms with a zero velocity component along the beam contribute to the signal. Also, the condition imposed by Eq. (7) on the wave vectors, $|k_p - k_s| \leq k_w \leq k_p + k_s$, is

not as restrictive as two-photon electromagnetically induced transparency (EIT) [8], and readily satisfied in Yb, Sr, and Ca, as shown in Table I. An additional advantage of this Doppler-free alignment is that there is no net momentum transfer in the three-photon transition process from light fields to the atom; thus there is no recoil energy shift of the resonance.

In practice, alignment of the three beams is not perfect, although in the Ca clock [1] Doppler misalignment using Ramsey interference is not yet the limiting factor at the current 10^{-15} level of accuracy and further improvement should be possible with colder atoms. Partial cancellation of the Doppler effect could still be very useful because it would correspondingly increase the size of the Lamb-Dicke region, thereby eliminating the first-order Doppler effect even when atoms are confined in a region larger than the optical wavelength.

A practical issue with the three-photon technique is how to combine the three laser frequencies to make an optical frequency standard. To begin with, it is necessary for the sum of the laser frequencies, $\omega_p + \omega_s - \omega_w$, to have a very small jitter, since this is the effective frequency of the sharp EITA peak that constitutes the clock reference. The complexity of independently stabilizing each laser to its own optical cavity can be avoided by using nonlinear techniques to directly generate a beam at the clock frequency $\omega_{\text{clock}} = \omega_p + \omega_s - \omega_w$; this beam can be frequency locked to a stable optical cavity by applying the correction to only *one* laser. The corrections applied to this single laser will compensate for both its own frequency fluctuations and those of the other two lasers according to the algebraic relationship for ω_{clock}, thereby imposing the necessary phase coherence among the three lasers for EITA. The optical cavity would then be referenced to the atoms via the sharp EITA peak. The mixing to obtain ω_{clock} can be done either in two steps using two separate doubly resonant build-up cavities [9] or in a single step (as a four-wave mixing process) in a single triply resonant cavity [9]. A more indirect method for stabilizing the lasers is to compare the three lasers to the nearest components of a comb [10] generated by a femtosecond laser whose repetition rate is stabilized to a radio frequency source. If $\Delta\omega_p$, $\Delta\omega_s$, and $\Delta\omega_w$ are the three beat notes for the lasers, the quantity $\Delta\omega_{\text{clock}} = \Delta\omega_p \pm \Delta\omega_s \pm \Delta\omega_w$ could be generated by radio frequency mixing and applied to one of the lasers using an optical modulator. The comb spacing would be stabilized using a fourth, ''flywheel'' laser, to lock the frequency of the radio frequency source.

In conclusion, we have set forth the scheme for an optical frequency standard based on the remarkably sharp resonance line that appears in three-photon transitions between the $^1S_0$ and $^3P_0$ states of alkaline earth and Yb atoms. The scheme has an advantage of permitting the use of the even isotopes, in which the clock transition is narrower than in the odd isotopes and should not be shifted by external magnetic fields or polarized trapping light. Based on the electromagnetically induced transparency and absorption, the width of the clock resonance in the scheme can, in principle, be continuously adjusted from the MHz level to sub-mHz without a loss of the signal amplitude by varying the intensities of the three optical beams. We believe that this unique feature will be very useful in locking lasers to the transition. Furthermore, Doppler and recoil effects can be eliminated by the point confinement of a lattice trap or by a proper alignment of the three beams.

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