Optical clocks based on ultra-narrow three-photon resonances in alkaline earth atoms

Tao Hong, Claire Cramer, Warren Nagourney, E. N. Fortson

Department of Physics, University of Washington, Seattle, Washington 98195.

Abstract

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. The method has the unique feature that the width and rate of the clock transition can 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. The three beams can be mixed to produce the optical frequency corresponding to the $^3P_0 - ^1S_0$ clock interval.

PACS numbers: 06.30.Ft, 32.80.Qk, 42.50.Gy
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 $^3P_0$ 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 trapping 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 be continuously adjusted from the MHz level to sub-mHz without loss of signal amplitude. Furthermore, proper alignment of the three beams can eliminate Doppler and recoil effects without the point confinement of a lattice trap. Finally, the three beams can be mixed to produce the clock frequency corresponding to the $^3P_0 - ^1S_0$ interval.

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). Our general results will be 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 described by the following density matrix equation,

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

where $\rho(t)$ is the atomic density matrix. The summation 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 in the interaction picture is defined by

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

where $\Omega_p$, $\Omega_s$ and $\Omega_w$ (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, $\omega_p$, $\omega_s$ and $\omega_w$, and the atomic resonance frequencies, $\omega_{21}$, $\omega_{32}$ and $\omega_{34}$, respectively. The Liouvillian matrix $L [\rho(t)]$ describes relaxation by spontaneous decay, and is defined in Eq. (3). Because the ground state $|1\rangle$ and the metastable state $|4\rangle$ normally have very long coherent times (typically much larger than seconds), here we assume there is no decay from these states. Decay rates $\gamma_{32}$ and $\gamma_{34}$ give the decay from state $|3\rangle$ to states $|2\rangle$ and $|4\rangle$ respectively, and typically they have values of order $10^7 s^{-1}$ or larger. $\gamma_{21}$ 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 $\gamma$ denotes the atomic characteristic decay rate, and the relative value of $\gamma_{21}$ is appropriate for Yb. With Sr and Ca, the basic behavior will be similar, but some numerical details will be markedly different because $\gamma_{21}$ is much smaller.
where absorption, as shown by the solid line in Fig. 2. Here for illustration, we chose a sharp absorption peak appears due to the electromagnetically induced transparency and

\[ | \Omega | < | \Omega_s |, \gamma_21, \gamma_32, \gamma_34 < | \Omega_s |, \]

a very sharp absorption peak appears due to the electromagnetically induced transparency and absorption, 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 \( | \Delta | < | \Omega_w |^2 / 2(\Delta_s + \Delta_p) + i(\gamma_{34} + \gamma_{32}) | \)), Eq. (4) takes a simple form that exhibits most of the important features:

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

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 small as \( | \Omega_s | \) increases, as illustrated in Fig. 3.

Thus the line width 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 was 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

\[
\Delta_p^{Peak} = \Delta_w - \Delta_s - W(\Delta_w - \Delta_s) / \gamma_{21}.
\]
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-noise ratio of an error signal determines the line width of a laser locked to a frequency discriminator [6], probe light intensity is normally increased until the absorption rate shortens the coherence time of the transition and broadens the line. These same considerations apply here. As $|\Omega_p|^2$ increases, the probed sharp absorption peak will be broadened and, when $(\Delta_w - \Delta s)$ is finite, pushed away from the intrinsic three-photon resonance frequency, as shown in Fig. 4. Here 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. It is evident that a system composed of a large number of atoms, such as laser-cooled neutral atoms, is more ideal to realize this proposal, because the absorption rate will be increased and the low light intensity limitation can be balanced. In addition, to reduce technical noise, some alternative ways to detect the sharp resonance feature can be considered in experiments. For example, detecting the fluorescence instead of the absorption can be considered because fluorescence peaks due to decays from states $|2\rangle$ and $|3\rangle$ have good correspondences with the absorption peak.

The Doppler effect is always a major cause of shifts and broadening of sharp optical resonances even for cold atoms. Neutral atom frequency standards typically use the method of Ramsey interference to eliminate the first order Doppler effect, while trapped ion standards and the optical lattice proposal make use of Lamb-Dicke confinement. A third technique becomes available for three-photon transitions: Doppler-free alignment of the three laser beams [7]. If the light wave vectors satisfy the phase matching relation (17), 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-noise ratio. It is therefore equivalent to the Ramsey method and superior to conventional Doppler-free saturated absorption spectroscopy, in which only atoms with zero velocity contribute to the signal. Also, the condition on the wave vectors \( \mathbf{k_p} + \mathbf{k_s} - \mathbf{k_w} = 0 \) is relaxed in comparison with two-photon electro-magnetically induced transparency (EIT) \([8]\), and readily satisfied in Yb, Sr and Ca, as shown in Table [I]

\[
\mathbf{k_p} + \mathbf{k_s} - \mathbf{k_w} = 0
\]  

(7)

\[
|\mathbf{k_p} - \mathbf{k_s}| \leq \mathbf{k_w} \leq \mathbf{k_p} + \mathbf{k_s}
\]  

(8)

In practice, alignment of the three beams will not be perfect; however, 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 when atoms are confined in a region larger than the optical wavelength. 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.

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 EIT 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 the single stabilized laser will correct for both its own frequency fluctuations as well as those of the other two lasers according to the algebraic relationship given above. The mixing can be done either in two steps using two separate doubly-resonant build-up cavities [9] or in a single step (as a 4-wave mixing process) in a single triply-resonant cavity [9]. A more indirect method for stabilizing the lasers would be 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 radiofrequency 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 radiofrequency mixing and applied to one of the lasers using an optical modulator. The comb spacing would be stabilized using a fourth, “flywheel” laser; this would lock the frequency of the radiofrequency 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 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 be continuously adjusted from the MHz level to sub-mHz without loss of 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 without the point confinement of a lattice trap by a proper alignment of the three beams.

Tao Hong would like to thank Yu Zhu Wang for helpful discussion. This work was supported by the National Science Foundation, Grant No. PHY 0099535.

[1] S. A. Diddams et al., Science 293, 825 (2001).
[2] Laser Spectroscopy XII, edited by M. Inguscio, M. Allegrini, A. Sasso (World Scientific, Singapore, 1996); Proceedings of the Workshop on the Scientific Applications of Clocks in Space, edited by L. Maleki (Jet Propulsion Laboratory, Pasadena, 1997); J. D. Prestage, R. L. Tjoelker, L. Maleki, Phys. Rev. Lett. 74, 3511 (1995).
[3] H. Katori, in Proceedings of the 6th Symposium on Frequency Standards and Metrology, edited by P. Gill (World Scientific, Singapore, 2002), pp. 323-330; I. Courtillot et al., Phys. Rev. A 68, 030501 (2003); H. Katori et al., Phys. Rev. Lett. 91, 173005 (2003); M. Takamoto, H. Katori, ibid. 91, 223001 (2003); S. G. Porsev, A. Derevianko, E. N. Fortson, Phys. Rev. A 69, 021403 (2004).
[4] S. E. Harris, Phys. Today 50, No. 7, 36 (1997); D. E. Roberts, E. N. Fortson, Phys. Rev. Lett. 31 1539 (1973); A. M. Akulshin, S. Barreiro, A. Lezama, Phys. Rev. A 57, 2996 (1998); T. Hong et al., Opt. Commun. 214, 371 (2002).
[5] Tao Hong and E. N. Fortson, to be published.

[6] B. C. Young, F. C. Cruz, W. M. Itano, J. C. Bergquist, Phys. Rev. Lett. 82 3799 (1999); W. Allan, *Time and frequency (time domain) characterization, estimation and prediction of precision clocks and oscillators*, IEEE trans. UFFC, 34, (1987).

[7] G. Grynberg, F. Biraben, M. Bassini, and B. Cagnac, Phys. Rev. Lett. 37, 283 (1976).

[8] In a two-photon EIT system, the Doppler effect cannot be completely eliminated unless the two light fields are degenerate in frequency.

[9] Y. R. Shen, *The Principles of Nonlinear Optics* (New York, J. Wiley, 1984).

[10] Th. Udem et al., Opt. Lett., 24, 881 (1999); Th. Udem et al., Phys. Rev. Lett., 82, 3568 (1999); M. Niering et al., Phys. Rev. Lett., 84, 5496 (2000); S. A. Diddams et al., Phys. Rev. Lett., 84, 5102 (2000).
TABLE I: Atom and corresponding optical wavelength candidates for forming the scheme in Fig. 1.

| Atom | Probe | Strong | Weak | Clock |
|------|-------|--------|------|-------|
| Yb   | 556 nm| 649 nm | 680 nm| 578 nm|
| Sr   | 689 nm| 688 nm | 679 nm| 698 nm|
| Ca   | 657 nm| 612 nm | 610 nm| 659 nm|

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).
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 zoom-in of the sharp peak.

FIG. 3: Narrow absorption peaks under different $|\Omega_s|$: $2\gamma$ (dash-dotted line), $3\gamma$ (dashed line), or $4\gamma$ (solid line). Here $|\Omega_p| = 0.00001\gamma$, $|\Omega_w| = 0.01\gamma$, $\Delta_s = 0.05\gamma$, $\Delta_w = -0.05\gamma$.

FIG. 4: 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), $1 \times 10^{-8}\gamma^2$ (solid line).