Optical Clock with Millihertz Linewidth Based on a Phase-Matching Effect

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We propose a new scheme for the optical frequency standard based on the phase-matching effect of the nonadiabatic interaction of two quasimonochromatic fields with the states $^1S_0$, $^1P_1$, and $^3P_0$ of atoms ^{88}Sr, which are trapped in an optical lattice with magic wavelength. After establishing the phase correlation between two laser fields by the nonadiabatic process, the final linewidth for the difference frequency field, which can be generated by a nonlinear optical crystal, is about 1 mHz.

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With the first optical lattice clock appearing [1], optical clocks [2–9] entered a new realm. By using the laser with magic wavelength to form the optical lattice and trap atoms in the Lamb-Dicke regime [10,11], the final Fourier-limited linewidth for the optical clock is about 1.5 Hz [2], which is the narrowest linewidth yet reported for neutral-atom-based optical clock. Several other novelty schemes may bring the opportunity for the realization of a higher-accuracy clock, such as electromagnetically induced transparency (EIT) optical clock with expected accuracy better than 2×10^{-17} [9], the three-photon resonance optical clock with the capability of continuously adjusting the linewidth of clock transition from MHz to sub mHz [8], and the magnetic-field-induced-spectroscopy clock [6,7]. However, if one wants to achieve a clock transition linewidth narrower than 1 Hz as proposed in [6–9], an available coherent laser with linewidth of millihertz is a prerequisite. The fact is that the linewidth of the best laser obtained to date in the optical frequency standard is about 0.2 Hz [12–14], which is the limitation at present and only several groups can achieve this level. The most effective method used to improve the coherence of the laser is the Pound-Drever-Hall (PDH) technique [15]. However, this technique is limited by the Brownian-motion noise of the supercavity [16]. It is very difficult to remove the 0.2 Hz limitation. To realize a laser light source with millihertz linewidth, such as that predicted by the Schawlow-Townes formula is a decades long dream for scientists.

In this Letter, we propose a new mechanism for an optical clock and light source, which is based on the phase-matching effect of the nonadiabatic interaction of two fields with the three-level Λ-type atomic system [17,18] of the neutral atom ^{88}Sr. This scheme provides the possibility to get a high-accuracy optical clock with the expected linewidth of about 1 mHz, which is much better than the best optical clock at present. Even only used as a source of light, its linewidth is 2 orders of magnitude better than the 0.2 Hz laser. Our proposal is suitable for any alkaline earth atoms, as an example, the Λ-type atomic system of ^{88}Sr as in [9] is considered here.

The atomic level configuration is shown in Fig. 1. Here, the upper state $|3\rangle$ is $^1P_1$ and the two lower states $|1\rangle$ and $|2\rangle$ are $^1S_0$ and $^3P_0$. The spontaneous decay rates of $|3\rangle$ to $|1\rangle$ and $|2\rangle$ via electric dipole transition (E1) and magnetic-dipole transition (M1) one-photon transitions are $\gamma_1 = 2\pi \times 32$ MHz and $\gamma_2 = 2\pi \times 0.823$ mHz, for the latter the coupling matrix element $|\langle 3|\mathrm{M1}|2\rangle| = 0.022\mu_B$ [9]. The coherence damping rates between the upper state $|3\rangle$ and the two lower state $|1\rangle$ and $|2\rangle$ are both $\Gamma/2$, where $\Gamma = \gamma_1 + \gamma_2$. Since the spontaneous decay rate from $|2\rangle$ to $|1\rangle$ is $2\pi \times 5.5 \times 10^{-12}$ Hz [19], the decoherent rate γ_0 between $|2\rangle$ and $|1\rangle$ is determined by the lattice laser, background gas collision, and the blackbody radiation line broadening, which can be controlled about 2 mHz [9,20,21]. The small γ_0 here is important that it could cause the phase-matching effect in the nonadiabatic regime, which could lead the narrow linewidth of the spectral density for the difference frequency. Two quasimonochromatic fields can be written as $E_1(z, t) = E_1^o(z, t)e^{i\phi_1(z,t)}$ and $E_2(z, t) = E_2^o(z, t)e^{i\phi_2(z,t)}$, whose wavelengths are 461 nm

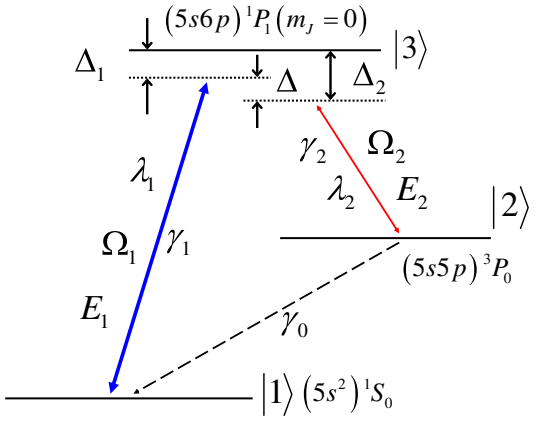

FIG. 1 (color online). The three-level Λ-type atomic system of ^{88}Sr. The spontaneous decay rates from state 3 to states 1 and 2 are $\gamma_1 = 2\pi \times 32$ MHz and $\gamma_2 = 2\pi \times 0.823$ mHz. $\Delta_{1,2}$ denote the detunings from the one-photon resonance corresponding to the fields $E_{1,2}$ with the relative detuning $\Delta = \Delta_1 - \Delta_2$ from the two-photon resonance.

for $E_1(z, t)$ with Rabi frequency $\Omega_1(t, z) = D_{13}E_1(z, t)/\hbar$ and 1354 nm for $E_2(z, t)$ with Rabi frequency $\Omega_2(t, z) = M_{23}B_2(z, t)/\hbar$, where $B_2(z, t) = E_2(z, t)e^{i\pi/2}/c$, coupling with atoms ^{88}Sr that are trapped in optical lattice. D_{13} and M_{23} are the dipole moment and magnet-dipole moment of $|1\rangle - |3\rangle$ and $|2\rangle - |3\rangle$ transition.

The interaction of a Λ-type atomic system with two resonant fields derives the atom into a coherent superposition of the two lower levels, which is called the dark state and is decoupled with fields. That is to say one certain field state corresponds to one certain atomic dark state. Therefore, the field information could be stored by atomic states. This interaction has two extreme regimes: one is the adiabatic regime and the other is the nonadiabatic regime. In the former one, since the phase diffusion between two fields is smaller than γ_0 [22], the atomic states, such as the population distribution, could follow any fluctuations of the fields [17,18]. There is no externally generated coherence. In this scheme, the field fluctuations will drive the atom adiabatically into a new dark state [17], which is again decoupled with fields. However, in the latter regime, the phase diffusion between two lasers is larger than γ_0, and atomic state could not follow the fluctuations of fields. Thus one could not treat the field-atom interaction in adiabatic limit. In the nonadiabatic regime, atoms could hold the field information, such as phases, frequencies and amplitudes, for a moment. That is to say atoms could always drag the field changing, which could suppress the field fluctuations. Therefore, due to the coherence between two lower atomic states, a strong coherence could be built up between two fields resulting in pulse matching [23] and phase matching [17,18]. The phase correlation between two fields could strongly suppress the phase-difference fluctuations and the spectrum of difference frequency field could be much improved [17,24,25]. This phase-matching effect is proposed to be a new optical clock mechanism in this Letter.

First, we consider the simple resonance case, in which the one-photon resonance detunings $\Delta_{1,2}$ are both zero. Because of the strong coherence damping rate between the upper state with two lower states, atomic variables σ_{13} and σ_{23} could be on steady states quickly (Here $\sigma_{ij} = \langle i|\rho|j\rangle$ is the density matrix variable). We could treat them in adiabatic regime and they could be expressed as

$$\begin{aligned}\sigma_{13} &= i\frac{\Omega_2}{\Gamma}\sigma_{12} - i\frac{\Omega_1}{\Gamma}n_{13},\\ \sigma_{23} &= i\frac{\Omega_1}{\Gamma}\sigma_{21} + i\frac{\Omega_2}{\Gamma}n_{23},\end{aligned} \tag{1}$$

where $n_{13} = \sigma_{11} - \sigma_{33}$ and $n_{23} = \sigma_{22} - \sigma_{33}$ are the population differences. Here, we should emphasize that all atomic variables and Rabi frequencies are the function of z and t. For expressing simply, we omit (z, t). However, due to the so small decoherent rate γ_0 between two lower atomic states that one could not treat the dynamic of atomic variable σ_{12} in adiabatic way. It should be treated by nonadiabatic way

$$\frac{d}{dt}\sigma_{12} = -\gamma_0\sigma_{12} - i\Omega_1\sigma_{32} + i\Omega_2^*\sigma_{13}. \tag{2}$$

In order to discuss the dynamic of difference frequency, it is convenient to consider the difference frequency function $E(t, z) = E_1(z, t)E_2^*(z, t)$. The dynamic equations for its amplitude and phase parts, $E^o(z, t) = E_1^o(z, t)E_2^o(z, t)$ and $\phi(z, t) = \phi_1(z, t) - \phi_2(z, t)$, could be expressed as

$$\frac{d}{dt}\phi = -\frac{2(\zeta_1|\Omega_2|^2 + \zeta_2|\Omega_1|^2)|\sigma_{12}|}{\Gamma E^o(t, z)}\sin(\varphi - \phi), \tag{3}$$

$$\begin{aligned}\frac{d}{dt}E^o = &-\frac{2}{\Gamma}(\zeta_1 n_{13} + \zeta_2 n_{23})E^o - \frac{2\beta}{\Gamma}(\zeta_1|\Omega_2|^2\\ &+ \zeta_2|\Omega_1|^2)|\sigma_{12}|\cos(\varphi - \phi),\end{aligned} \tag{4}$$

Here $\beta = \frac{\hbar^2 c}{D_{13}M_{23}}$, $\zeta_{1,2} = \frac{3\lambda_{1,2}^2 n\gamma_{1,2}c}{8\pi}$, n is the atomic density and φ is the phase of σ_{12}. From above equations, one could see that due to the difference phase noise fluctuation the intensity of difference frequency also could not be considered in adiabatic limit as done in [17,18]. The correlation function for the difference frequency function, $\langle E(t, z)E(t + \tau, z)\rangle$, can be written as

$$\frac{d}{d\tau}\langle E(z, t)E(z, t + \tau)\rangle = -\frac{4}{\Gamma}(\zeta_1 n_{13} + \zeta_2 n_{23})\langle E(z, t)E(z, t + \tau)\rangle - \frac{4\beta}{\Gamma}(\zeta_1|\Omega_2|^2 + \zeta_2|\Omega_1|^2)\langle E(z, t)\sigma_{12}(z, t + \tau)\rangle, \tag{5}$$

where the function $\langle E(t, z)\sigma_{12}(t + \tau, z)\rangle$ is governed by

$$\frac{d}{d\tau}\langle E(t, z)\sigma_{12}(t + \tau, z)\rangle = -\Gamma_g\langle E(t, z)\sigma_{12}(t + \tau, z)\rangle - \frac{2}{\Gamma\beta}(n_{13} + n_{23})\langle E(t, z)E(t + \tau, z)\rangle. \tag{6}$$

Since atoms are trapped in optical lattice, their velocities are zero. The time differential operator in Eq. (6) is only to time t, not to space z. However, in Eqs. (3)–(5), fields change strongly with space and the time differential operator should be $d/dt = \partial/\partial t + c\partial/\partial z$. The power spectral density $S(\omega, z)$ of the difference frequency is given by the Fourier transformation

$$S(\omega, z) = \int\langle E(t, z)E(t + \tau, z)\rangle e^{i\omega\tau}d\tau. \tag{7}$$

With the help of Eqs. (3)–(7), one could get the spectral density for difference frequency changing with interacting length,

$$\frac{\partial}{\partial z}S(\omega, z) = -(\kappa + i\tilde{\kappa})S(\omega, z), \tag{8}$$

where

$$\kappa(\omega) = \frac{4(\zeta_1 n_{11} + \zeta_2 n_{22})}{\Gamma c} - \frac{8}{\Gamma^2 \Gamma_g c}(\zeta_2|\Omega_1|^2 + \zeta_1|\Omega_2|^2) \times \frac{\Gamma_g^2}{(\omega - \omega_0)^2 + \Gamma_g^2} \tag{9}$$

is the absorption coefficient. Here $\Gamma_g = \gamma_0 + \frac{2}{\Gamma}(|\Omega_1|^2 + |\Omega_2|^2)$ and $\omega_0 = \omega_1 - \omega_2$ is the center frequency of the difference frequency field. The imaginary coefficient $\tilde{\kappa}$ is not important here and we do not consider it. As one could see, the atomic system here is no longer the reference but a frequency filter, which only allows the frequency satisfying the $|\omega - \omega_0| < \Gamma_g$ pass. If the initial spectral density of the difference frequency is Lorentzian in shape with linewidth γ and intensity I, we have

$$S(\omega, z) = I\frac{\gamma^2}{\gamma^2 + (\omega - \omega_0)^2}\exp\left[-\int_0^z (\kappa + i\tilde{\kappa})dz'\right]. \tag{10}$$

As seen from Eq. (8), in the nonadiabatic regime, the spectral density for the difference frequency is narrowed with the field-atom interacting distance. With enough interacting length, the final linewidth is Γ_g, which has the form of the EIT linewidth. However, with narrowing of the spectral density, the field powers will also decrease. The corresponding absorption coefficients of two noncorrelated lasers are

$$\kappa_1 = \frac{4\zeta_1 n_{11}}{\Gamma c} \quad \text{and} \quad \kappa_2 = \frac{4\zeta_2 n_{22}}{\Gamma c}. \tag{11}$$

Since atoms mostly accumulate in level 1, one could assume $n_{11} \approx 1$ and $n_2 \approx 0$. Therefore, the intensity of laser 2 will be almost unchanged.

Here we assume that the initial linewidths of two lasers are both 1 Hz. Thus the linewidth of their difference frequency spectral density is 1.414 Hz because of the uncorrelation between two lasers. In order to get the linewidth of mHz, the characteristic linewidth Γ_g, which has the form of the usual EIT linewidth, should be closed to mHz. Thus we assume that the initial intensities of the lasers 1 and 2 are 0.83 pW/cm^2 and 1.3 mW/cm^2 for each. The corresponding Rabi frequencies are both $2\pi \times 100$ Hz and $\Gamma_g = 2\pi \times 2.2$ mHz. The interacting length here is 20 μm, with the atomic density of 10^{12} cm^{-3}. Thus the initial absorption coefficients are $\kappa_1 = 0.1\ \mu\text{m}^{-1}$, $\kappa(\omega = \omega_0) = 0.07\ \mu\text{m}^{-1}$ and κ_2 is very small and could be neglected. After passing the atomic system, the intensity of laser 1 reduces to 0.11 pW/cm^2 with the corresponding Rabi frequency 36 Hz, and the linewidth of spectral density for the difference frequency is reduced to 2 mHz. Figure 2 shows the spectral density for the difference frequency changing with the interacting way. The remaining power is 13% of the initial value. However, the biggest part is contained outside the 2 mHz linewidth. The remaining power of laser 1 in the 2 mHz linwidth is about 84 photons/(s · mm^2).

So far we have only considered the resonant coupling case. In the nonresonant case, the absorption coefficient $\kappa(\omega, z)$ decreases rapidly with Raman detuning Δ, which is not what we want. The absorption coefficient changing with Raman detuning Δ is similar to Eq. (7) with Γ_g replaced by $\sqrt{\Delta^2 + \Gamma_g^2}$. As one could see due to the existence of Raman detuning Δ, the unabsorbed band of the atom filter largely expands. Therefore the final linewidth for difference frequency and the absorption coefficient are strongly influenced by Raman detuning Δ.

Comparing with [8,9], here we are not limited to the interaction of the lasers with atoms in the adiabatic regime as used in [8,9], but the nonadiabatic regime is considered. Both schemes in [8,9] need a prerequisite laser with linewidth narrower than 1 mHz. However, the state-of-the-art laser linewidth is 0.2 Hz, which does not satisfy the requirement. Our scheme proposed here could solve this problem. In our scheme, two lasers with linewidths of $\gamma = 2\pi \times 1$ Hz will be phase matching via nonadiabatic interaction with atoms, resulting in millihertz linewidth of difference frequency between two lasers. Atoms here could be seen as a filter, which has a low-pass feature and could eliminate the phase-difference fluctuation with frequencies larger than Γ_g.

Because of the narrow linewidth of difference frequency, it could be applied in frequency standard. The layout for the new optical clock scheme is shown in Fig. 3. Two

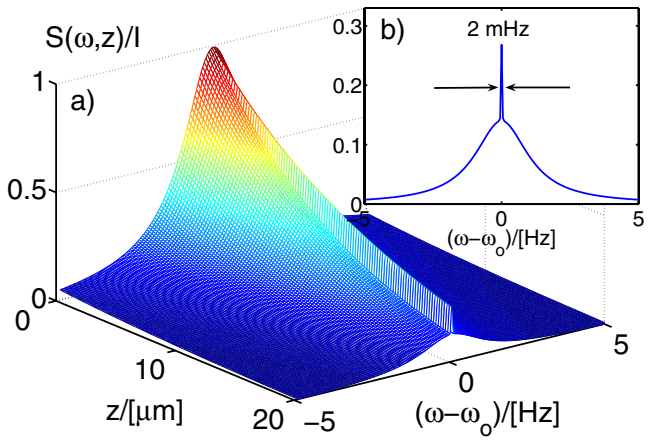

FIG. 2 (color online). (a) The line shape of the power spectrum for the difference frequency field changing with interacting length. At the beginning, the power spectrum has a Lorentzian shape with linewidth of 1.4 Hz. (b) The line shape of the output field. The linewidth of the peak is 2 mHz.

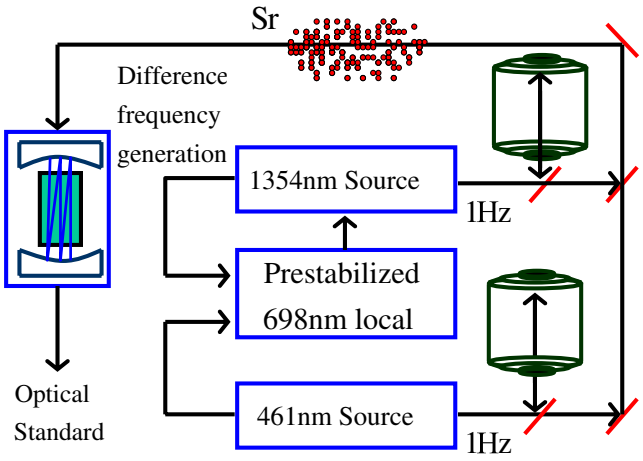

FIG. 3 (color online). Experimental layout. Two lasers are prestablized by two supercavities, and nonadiabatically interact with ^{88}Sr atoms that are trapped in magic optical lattice. The bichromatic field passes difference frequency crystal, from which an optical frequency standard at 698 nm with 2 mHz linewidth is output.

lasers, at 461 and 1354 nm, are prestabilized by two supercavities resulting in 1 Hz linewidths for both lasers. As discussed in [9], they are both prestablized by a 698 nm local laser source. Moreover, the 698 nm laser is also locked to the atomic transition line by another atomic system with an accuracy of 1 Hz (not shown in Fig. 3). Both lasers pass and couple with the ^{88}Sr atoms, which are trapped in the magic optical lattice with a wavelength of 813.5 nm [2,5,9]. Because of the small intensity of the output fields, difference frequency generation may be a challenge. We suggest a method that the nonlinear crystal is put into a F-P cavity, as shown in Fig. 3. The difference frequency of the two laser sources will be an optical clock. This optical clock works continuously and does not need modulation. Here we want to stress again that due to the nonadiabatic process, fast phase-difference fluctuation is absorbed by atomic system. The atoms can be thought as a kind of ''quantum narrow-band filter''. This filter only lets the difference frequency in range $|\omega - \omega_0| < \Gamma_g$ pass atomic system since two fields decoupled from atom within adiabatic regime as shown in Eqs. (8) and (9), and the other difference frequency parts are absorbed by atoms in nonadiabatic regime. As the transmitted difference frequency field is with a spectrum filtered via ''quantum narrow-band filter'', it resembles a narrow-band thermal field with large residual amplitude fluctuations. Therefore, besides the low power, the optical phase may not be followed easily and the counting of the frequency might be difficult. Thus it would require an additional effect to use the output field as an optical clock or narrow linewidth light source. However, because of the rapid development of the science and technology, these difficulties could be ultimately overcome.

In summary, we have proposed a novel optical clock mechanism based on an optical scheme via the phase-matching effect in a Λ-type atomic system. By the nonadiabatic interacting process of lasers with atoms, one can get a difference frequency field with the expected linewidth of 2 mHz. We do not require the condition that the contribution from phase diffusion of fields to the effective decay rate should be smaller than the decoherence decay rate between two lower levels. Results show that this mechanism can provide a possible new way to realize an optical clock with a millihertz linewidth, which is a formidable limitation to any present optical clock.

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Note added.—Recently, we found that this spectral narrowing phenomena with microwave difference frequency is experimentally demonstrated [25].

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