An N-atom Collective State Atomic Clock with Root-N Fold Increase in Effective Frequency and Root-N Fold Reduction in Fringe Width

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Abstract

We describe a collective state atomic clock with Ramsey fringes narrowed by a factor of $\sqrt{N}$ compared to a conventional clock, N being the number of non-interacting atoms, without violating the uncertainty relation. This narrowing is explained as being due to interferences among the collective states, representing an effective $\sqrt{N}$ fold increase in the clock frequency, without entanglement. The detection process, which measures a collective state, can be used to increase the quantum efficiency of detection significantly, yielding a net improvement in stability by as much as a factor of 10.

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It is well known that the width of the fringes, observed as a function of the detuning, in a pulsed excitation of an atomic transition is limited by the inverse of the interaction time. This effect is routinely observed in systems such as microwave or Raman atomic clocks [1-5]. It is also well known that the effective interaction time can be extended by employing Ramsey’s technique of separated field excitations [6]. In that case, the transit time limited linewidth is determined by the inverse of the time delay between the two fields. The temporal profile of the field envelope seen by the atoms is a pair of square pulses, each with a duration $T_1$, separated by $T_2$. For a conventional clock (CC), the Ramsey technique produces a sync function with a width of $\sim T_1^{-1}$, modulated by a sinusoid with a fringe width of $\sim T_2^{-1}$, all centered at the carrier frequency.

The width of these fringes can be reduced by making use of entanglement, as demonstrated by Wineland et al. using trapped ions [7]. Consider, for example, a situation where the use of entanglement allows one to couple the ground state of three particles to a state where all three particles are in the excited state, representing a collective excitation. This corresponds to an effective increase in the transition frequency by a factor of three. As such, the detuning for a single atom gets tripled for this collective excitation, so that the width of the Ramsey fringe gets reduced by a factor of three. However, realizing such a scheme for a large number of particles is beyond the capability of current technology.

Here, we describe a scheme that produces Ramsey fringes that are narrower by a factor of more than $10^3$ for parameters that are readily accessible, without making use of entanglement. While the concept can be applied to other types of atomic clocks, as described later, the specific experiment we propose is an optically off-resonant Raman atomic clock using ensembles of $N$ cold atoms. The clock transition is detected by measuring one of the collective states rather than measuring individual atomic states. The fringes observed as a function of the Raman (i.e. two photon) detuning is found to be $\sim \sqrt{N}$ times narrower than the transit time limited width that would be seen by measuring individual atomic states. For the current state of the art of trapped atoms, the value of $N$ can easily exceed $10^6$, so that a reduction of fringe width by a factor of more than $10^3$ is feasible.

The reduction in the width of the fringe, especially by such a large factor, strongly violates the conventional transit time limit of spectroscopic resolution. However, we show in [8], via a detailed analysis of the standard quantum limit and the Heisenberg limit, that, indeed, this violation of the conventional transit-time limit is allowed, and is within the constraint
of the more fundamental uncertainty principle of quantum mechanics. We also show that under certain conditions, frequency fluctuation of the CSAC can be significantly smaller, by as much as a factor of ten, than that for a conventional clock employing the same transition and same atomic flux. The ultra-narrow resonances produced in this process may also open up the possibility of exploring novel ways of implementing spin-squeezing techniques for further improvement in clock stability [9–12].

The optically off-resonant Raman atomic clock employs three hyperfine energy levels in a Λ scheme depicted in Fig. 1 (a). The ground states $|1\rangle$ and $|2\rangle$ of this atom interact with an excited state $|3\rangle$ via two coherent electromagnetic light fields of frequencies $\omega_1$ and $\omega_2$, respectively, detuned from resonance by $\delta_1$ and $\delta_2$, respectively. The Hamiltonian after the dipole approximation, rotating wave approximation, and rotating wave transformation can be expressed as [13]:

$$H = \hbar \left[ (\delta \sigma_{11} - \delta \sigma_{22} - 2\Delta \sigma_{33}) - (\Omega_1 \sigma_{13} + \Omega_2 \sigma_{23} + h.c.) \right]$$

where $\sigma_{\mu\nu} = |\mu\rangle\langle\nu|$, $\delta \equiv \delta_1 - \delta_2$ is the two photon detuning, $\Delta \equiv (\delta_1 + \delta_2)/2$ is the average detuning, and $\Omega_{1,2}$ are the Rabi frequencies. Here, we have also assumed a phase transformation applied to the Hamiltonian so that $\Omega_{1,2}$ are real. We assume next that $\Delta \gg \Gamma, \Omega_1, \text{ and } \Omega_2$ (where $\Gamma$ is the decay rate of state $|3\rangle$) so that the effect of $\Gamma$ can be neglected, and state $|3\rangle$ can be eliminated adiabatically [14, 15]. We assume further that $\Omega_1 = \Omega_2$ so that the light shift for states $|1\rangle$ and $|2\rangle$ are matched. Under these conditions, the Hamiltonian of the reduced two level system can be expressed as:

$$H_{\text{red}} = \frac{\hbar}{2} \left[ (\delta \sigma_{11} - \delta \sigma_{22} - 2\Delta \sigma_{33}) - (\Omega \sigma_{13} + \delta \sigma_{33} + h.c.) \right]$$

where $\sigma_z = (\sigma_{11} - \sigma_{22})$ and $\sigma_x = (\sigma_{12} + \sigma_{21})$. The quantum state for this effective two level system is given by $|\psi(t')\rangle = W_{\text{eff}}^t|\psi(t')\rangle$ where $|\psi(t')\rangle = \tilde{c}_1(t')|1\rangle + \tilde{c}_2(t')|2\rangle$, and the propagation operator is given by [16]

$$W_{\text{eff}}^t = e^i\frac{\delta t}{2} \begin{pmatrix} \cos \phi + i \frac{\delta}{\Omega'} \sin \phi & -i \frac{\Omega}{\Omega'} \sin \phi \\ -i \frac{\Omega}{\Omega'} \sin \phi & \cos \phi + i \frac{\delta}{\Omega'} \sin \phi \end{pmatrix}$$

where $\phi = \Omega' t/2$, and $\Omega' \equiv \sqrt{\Omega^2 + \delta^2}$ is the generalized Rabi frequency.

When this system is excited by two pulses of duration $T_1$, separated in time by $T_2$, we have $\Omega_1(t) = \Omega_2(t) = \Omega_0[U(t) - U(t - T_1) + U(t - (T_1 + T_2)) - U(t - (2T_1 + T_2))]$ where $U(t)$ is the Heaviside step function. When $\delta \ll \Omega$ and the width of the pulse is chosen to be
\( \Omega T_1 = \pi/2 \), each pulse acts on the system as a propagation operator \( W^0_{\pi/2} = (I - i\sigma_x)/\sqrt{2} \). 

While the system is between \( t = T_1 \) and \( t = T_1 + T_2 \) where no interaction is present, the propagation operator can be expressed as \( W^0_{\delta T_2} = \sigma_{11} + e^{i\delta T_2} \sigma_{22} \). After passing through the three zones, the state of the atom that was originally in state \(|1\rangle\) is \(|\psi\rangle = W^0_{\pi/2} W^0_{\delta T_2} W^0_{\pi/2} |1\rangle = -i e^{i\theta} (\sin \theta |1\rangle + \cos \theta |2\rangle) \) where \( \theta = \delta T_2/2 \) is the dephasing angle. The probability of the atom being in state \(|2\rangle\) is \( P_2 \equiv |\langle 2|\psi\rangle|^2 = (\cos \theta)^2 \).

The discussion can be generalized to \( N \) atoms that are all excited by the same field. We assume that there are no overlaps between the wavefunctions of the atoms and there is no interaction among them \( |\psi\rangle \). The evolution of each atom under these assumptions can be described individually, and the total quantum state is simply the outer (tensor) product of individual quantum states \(|\psi_i\rangle\). However, the interaction can also be described equivalently using a basis of collective states \(|\psi_{\text{col}}\rangle\) \(|\psi_{\text{col}}\rangle\). The Hilbert space of \( N \) two level atoms is spanned by \( 2^N \) states. Thus, when transformed to the collective state basis, there are also \( 2^N \) collective states. For identical Rabi frequencies and resonant frequencies, however, only the generalized symmetric states \(|\psi_{\text{sym}}\rangle\), of which there are only \((N + 1)\), are relevant, and the rest of the \((2^N - N - 1)\) states become decoupled. The case where inhomogeneity of the Rabi frequencies and different Doppler shifts experienced by different atoms are taken into account is presented in \( \text{[20]} \). We also note that if different atoms see different phase factors from the excitation fields, these factors can be absorbed into the definition of the generalized symmetric states \(|\psi_{\text{sym}}\rangle\). The simplified symmetric states, known as the conventional Dicke states \(|\psi_{\text{Dicke}}\rangle\), represent the case where it is assumed that the mean separation between the atoms is much less than the wavelength corresponding to the two level transition (which, for the co-propagating off resonant Raman excitation, is \( \sim (k_1 - k_2)^{-1} \)). While this constraint is not necessary for the concept proposed here \(|\psi_{\text{sym}}\rangle\), it is easier to describe the process initially under this constraint. The observables computed remain correct when this constraint is not met. Some of these Dicke states are as follows: \(|E_0\rangle \equiv |11\ldots 1\rangle\), \(|E_1\rangle \equiv \sum_{i=1}^{N} |11\ldots 2_i\ldots 1\rangle/\sqrt{N}\), \(|E_2\rangle \equiv \sum_{i,j, i \neq j} |11\ldots 2_i\ldots 2_j\ldots 1\rangle/\sqrt{NC_2}\), \(|E_3\rangle \equiv \sum_{i,j,k} |11\ldots 2_i\ldots 2_j\ldots 2_k\ldots 1\rangle/\sqrt{NC_3}\), and \(|E_N\rangle \equiv |222\ldots 2\rangle\) where \( NC_n = N!/n!(N-n)! \).

For instance, \(|E_2\rangle\) is the Dicke state with two atoms in \(|2\rangle\) and the rest in \(|1\rangle\). Any two atoms can be in \(|2\rangle\) with equal probability, with \( NC_2 = N(N-1)/2 \) such possible combinations.

The Hamiltonian in the basis of the symmetric collective states is \( H = \sum_{k=0}^{N} [-\hbar \delta |E_k\rangle \langle E_k|] + \sum_{k=0}^{N-1} [\hbar \Omega_{k+1} |E_k\rangle \langle E_{k+1}| + H.c.] \) where \( \Omega_{k+1} = \sqrt{N-k} \sqrt{k+1} \Omega \) is the Rabi frequency be-
between the collective states $[17, 18]$. The states are separated by $\hbar \delta$ in energy and couple at different rates. For instance, $\Omega_1 = \Omega_N = \sqrt{N} \Omega$, $\Omega_2 = \Omega_{N-1} = \sqrt{2(N-1)} \Omega$ and so forth. The middle states have the strongest coupling rate of $\Omega_{N/2} = N \Omega$ and the end states couple most weakly.

The final state of the system at the end of the second $\pi/2$ pulse can be derived by using either the collective state picture or, equivalently, the single atom picture. For a large value of $N$, carrying out the calculation in the collective states basis is numerically cumbersome and analytically intractable. However, we can find the state trivially by using the single atom picture and then determining the coefficients of the collective states by simple projection, given the definition of the $(N+1)$ generalized symmetric collective states. As such, the final state of the system is $|\psi\rangle = \prod_{i=1}^{N}(W_0^{\pi/2}W_0^{\delta T_2}W_0^{\pi/2})_i|1_i\rangle$. In the basis of the generalized symmetric collective states, this becomes:

$$|\psi\rangle = (-ie^{i\theta})^N \sum_{k=0}^{N} \sqrt{N C_k} (\sin \theta)^{N-k} (\cos \theta)^k |\tilde{E}_k\rangle$$

The population of the state $|\tilde{E}_N\rangle$ at the end of the separated field experiment is

$$P_N^C \equiv |\langle \tilde{E}_N|\psi\rangle|^2 = (\cos \theta)^{2N}$$

which is simply $(P_2)^N$. This quantity, $P_N^C$, represents the probability of finding the whole system in the state $|E_N\rangle$ whereas $P_2$ represents the probability of finding each atom in state $|2\rangle$. In a conventional experiment, the population of atoms in state $|2\rangle$ is measured, for example, by collecting fluorescence produced by coupling $|2\rangle$ to an auxiliary state. The resulting signal is proportional to $P_2$, independent of the number of atoms. The experiment that we propose, to be described shortly, produces a signal that is proportional to $P_N^C$.

When (4) is plotted for various values of $N$ (Fig. 1 (b)), it is evident that the linewidth of the fringe as a function of $\theta$ decreases as $N$ increases. The value of the linewidth, defined as the full width half maximum (FWHM), is given by $\Gamma(N) = 2 \arccos (2^{-1/2N})$. The derivative of $[\Gamma(1)/\Gamma(N)]^2$ with respect to $N$, for large $N$, approaches the value of $0.8899 + O(N^{-3/2})$, which we have verified with a linear fit to $[\Gamma(1)/\Gamma(N)]^2$. To a good approximation, $\Gamma(N)/\Gamma(1) \approx \sqrt{N}$. Noting that $\theta = \delta T_2/2$, $\Gamma(1) \simeq \pi/T_2$ is understood to be the transit time limited linewidth. Then $\Gamma(N) = \Gamma(1)/\sqrt{N} = \pi/(T_2 \sqrt{N})$ is a violation of the transit time limit, which is discussed in [8], along with the physical interpretation of what occurs in the collective atomic clock system.
Figure 1. (a) The three level atomic system (b) Population of state $|E_N\rangle$ at the end of the Ramsey pulse sequence as a function of $\delta$. Note the narrowing of linewidth as $N$ increases.

Before proceeding further, we describe the experimental approach that can be used to measure $P_N^C$, as summarized in Fig. 2. For concreteness, and without loss of generality, we consider $^{85}\text{Rb}$ as the atomic species. We start by trapping atoms in a magneto-optical trap (MOT) cooled down to the Doppler cooling limit of $T_D = \hbar \Gamma/(2k_B) = 146 \, \mu\text{K}$ [21]. After capturing about $10^6$ atoms in a cloud with a diameter of $\sim 1 \, \text{mm}$ [22, 23], the trapping magnetic field and the repump beams are turned off while the trapping beam is kept on for $\sim 100 \, \mu\text{s}$ to pump nearly all the atoms to the $F = 2$ state. A bias magnetic field of $\sim 1 \, \text{G}$, generated with a pair of Helmholtz coils, is turned on in the $\hat{z}$ direction. While the atoms are in free fall, we turn on a pair of co-propagating right circularly polarized ($\sigma_+$) Raman beams in the $\hat{z}$ direction. One of these beams is tuned to be $\sim 1.5 \, \text{GHz}$ red detuned from the $F = 2 \rightarrow F' = 3$ transition (D2 manifold), and the other is tuned to be $\sim 1.5 \, \text{GHz}$ red detuned from the $F = 3 \rightarrow F' = 3$ transition (D2 manifold). The second Raman beam is generated from the first one via an acousto-optic modulator (AOM), for example. The AOM is driven by a highly stable frequency synthesizer (FS), which is tuned close to $\sim 3.034 \, \text{GHz}$ corresponding to the frequency difference between the $F = 2$ and $F = 3$ states in the $5S_{1/2}$ manifold.

These beams would excite off-resonant Raman transitions between $F = 2$, $m_F = m$ and $F = 3$, $m_F = m$ levels, for $m = \pm 2, \pm 1, 0$. With $g_F = -1/3$ for $F = 2$ and $g_F = 1/3$ for $F = 3$, each transition will be shifted by $\delta_z = -m(g_{F=3} - g_{F=2})\mu_B B/\hbar = -0.94 \, [\text{MHz/Gauss}] \cdot mB$ where $B$ is the applied magnetic field expressed in the unit of Gauss. The signals from the five transitions are resolved if the linewidth of the Raman transition is less than $\delta_z$. The $m = 0$ transition is the most insensitive to the external magnetic field and its fluctuations, making it ideal for building a stable collective atomic clock. Hence, the energy levels $|1\rangle$
and $|2\rangle$ from the previously discussed $\Lambda$ scheme would correspond to hyperfine ground states $F = 2$, $m_F = 0$ and $F = 3$, $m_F = 0$ respectively. The $\sigma_+$ Raman transitions occur through the excited states $F' = 2$, $m_{F'} = 1$ and $F' = 3$, $m_{F'} = 1$. The resulting four level system can be reduced to a two level system in the same manner as the $\Lambda$ system by adiabatically eliminating the excited states together. The resulting two level system has a coupling rate that is the sum of the two Raman Rabi frequencies, one involving the $F' = 2$, $m_{F'} = 1$ state, and the other involving the $F' = 3$, $m_{F'} = 1$ state. The laser power at $\omega_1$ and $\omega_2$ are adjusted to ensure that the light shifts of levels $|1\rangle$ and $|2\rangle$ are matched.

In the first interaction zone, the co-propagating Raman beams interact with the atomic ensemble for a duration of $\Omega T_1 = \pi/2$. After waiting for a time $T_2$, chosen such that $T_2 \gg T_1$, we pulse the Raman beams again, in place, to interact with the atomic ensemble for another duration $\Omega T_1 = \pi/2$. The Raman beams can be pulsed in place as long as the width of the beams is much larger than that of the free-falling, thermally expanding atomic cloud.

After these excitations, we probe the population in one of the collective states, $|E_N\rangle$, where all the individual atoms are in state $|2\rangle$, by a method of zero photon detection. For illustrative purposes, let us consider first a situation where the atomic ensemble is contained in a single mode cavity with mode volume $V$, cavity decay rate $\gamma_c$, and wavevector $k_2 = \omega_2/c$. The cavity is coupled to the atomic transition $|2\rangle \rightarrow |3\rangle$ with coupling rate $g_c = |e\langle r\rangle|E/\hbar$, where $|e\langle r\rangle|$ is the dipole moment of the atom and the field of the cavity is $E = \sqrt{2\hbar\omega_2/(\varepsilon_0 V)}$. If we then send a probe beam, an off-resonant classical laser pulse with frequency $\omega_1$, the presence of the cavity will allow Raman transitions to occur between the collective states $|E_k\rangle$ and $|E_{k+1}\rangle$ with the coupling rates $\Omega'_{k+1} = \sqrt{N-k}\sqrt{k+1}\Omega'$ where $\Omega' = \Omega_1g_c/2\Delta$. The schematic of the interaction is shown in Fig. 3.
Figure 3. Interaction between the collective states in the bad cavity limit, which is an irreversible process.

In the bad cavity limit where $\gamma_c \gg \sqrt{N}\Omega'$, the Raman transitions will still occur. However, the atomic system will not reabsorb the photon that has been emitted during the process, such that the transition from $|E_k\rangle$ to $|E_{k+1}\rangle$ will occur, but not vice versa. The electric field of such a photon is $E = \sqrt{2\hbar\omega_2/(\epsilon_0 Ac\tau)}$, where $A$ is the cross sectional area of the atomic ensemble, $c$ is the speed of light, and $\tau$ is the duration of the photon. This limit applies in our case, which has no cavity. In this limit, the stimulated Raman scattering is an irreversible process that can be modeled as a decay with an effective decay rate that is singular to each $|E_j\rangle$ state. The decay rate from state $|E_1\rangle$ is $\gamma_0 = 4NL|g_e\Omega_1|^2/(\Delta^2 c) = N\gamma_{sa}$ where $\gamma_{sa} = 16L\Omega'^2/c$ [24] is the decay rate for a single atom. The value of $g_e$ is given by $|e\langle r\rangle| \cdot E$. The effective decay rates for the other states can be calculated following the same logic as $\gamma_j = (j + 1)(N - j)\gamma_{sa}$.

The photons scattered through this process are emitted in the direction of propagation of the probe beam. The beam consisting of the probe and the emitted photons is sent to a high speed detector, which produces a dc voltage as well as a signal at the beat frequency of $\sim 3.034$ GHz. The phase of this beat frequency signal is unknown. As such, the total signal is sent in two different paths, one to be multiplied by the FS signal and another to be multiplied by the FS signal shifted in phase by 90 degrees. Each of these signals is squared, then combined and sent through a low pass filter (LPF) to extract the dc voltage that is proportional to the number of scattered photons. A voltage reading above a predetermined threshold value will indicate the presence of emitted photons during the interrogation period. The interrogation period is set to $\gamma_0 T = 10$ where $\gamma_0 = \gamma_{N-1} = N\gamma_{sa}$ is the slowest decay rate, to ensure that even the longest lived state has a chance to decay completely. If no photon emission occurs and the voltage reads below the threshold, this indicates that the atoms are all in $|2\rangle$ and the collective state of the system is $|E_N\rangle$. For any other collective state, at least one photon will be emitted. For a given value of $\delta$, this process is repeated $m$
times (where the choice of \( m \) would depend on the temporal granularity of interest). The fraction of events corresponding to detection of no photons would represent the signal for this value of \( \delta \). The process is now repeated for a different value of \( \delta \), thus enabling one to produce the clock signal as a function of \( \delta \). Usual techniques of modulating the detuning and demodulating the signal can be used to produce the error signal for stabilizing the FS, thus realizing the CSAC.

In order to compare the performance of the CSAC to that of the comparable CC, we examine the stability of the clocks by investigating the fluctuation that has both quantum mechanical and classical components, or \( \delta f |_{total} = (\Delta S_{QM} + \Delta S_{class})/(\partial S/\partial f) \), where \( S(f) \) is the signal and \( f \) is the detuning of the clock away from its center value. Because the signal depends on the frequency, the fluctuations in a clock are not necessarily constant, and there isn’t a single value of signal to noise ratio (SNR) to compare unless we compare the two clocks at a particular value of the frequency. Instead, the fluctuations must be compared as a function of \( f \) for completeness. In [20], we discuss the quantum fluctuation due to quantum projection noise, \( \Delta P = \sqrt{P(1 - P)} \) [9], where \( P \) is the population of the state to be measured, the classical noise in the long term regime, and the effects of detector efficiency and the collection efficiency. The ratios of the frequency fluctuations in the CC to the frequency fluctuations in the CSAC show that the two clocks perform comparably around the signal at \( f = 0 \) if the clocks have perfect collection efficiency. However, the traditional clock suffers from collection efficiency issues that the collective clock is immune to. For the CC, a resonant beam probes the clock state, generating spontaneously emitted photons. The collection efficiency of such system is limited by the solid angle of the detection system. On the other hand, the CSAC collects the fluorescence of photons through coherent Raman scattering, which enables large collection efficiency that can be close to unity. As such, for the same number of atoms detected per unit time, the CSAC is expected to perform better than the CC by as much as a factor of 10.

In the particular implementation of the CSAC considered here, we have used off-resonant Raman transition. However, effects such as differential light shifts can limit the stability of such a clock. The ground states can also be coupled directly by using a microwave pulse, which has the advantage of being free from differential light shifts. Thus, the CSAC can also be realized by using a traveling wave microwave pulse sequence for the separated Ramsey field experiment [25], as long as the detection pulse remains the same.
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[1] M. Niering, R. Holzwarth, J. Reichert, P. Pokasov, T. Udem, M. Weitz, T. W. Hänsch, P. Lemonde, G. Santarelli, M. Abgrall, P. Laurent, C. Salomon, and A. Clairon, Phys. Rev. Lett. 84, 5496 (2000).

[2] G. Santarelli, P. Laurent, P. Lemonde, A. Clairon, A. G. Mann, S. Chang, A. N. Luiten, and C. Salomon, Phys. Rev. Lett. 82, 4619 (1999).

[3] E. Arimondo (Elsevier, 1996) pp. 257 – 354.

[4] F.-X. Esnault, E. Blanshan, E. N. Ivanov, R. E. Scholten, J. Kitching, and E. A. Donley, Phys. Rev. A 88, 042120 (2013).

[5] G. Wilpers, T. Binnewies, C. Degenhardt, U. Sterr, J. Helmcke, and F. Riehle, Phys. Rev. Lett. 89, 230801 (2002).

[6] N. F. Ramsey, Phys. Rev. 78, 695 (1950).

[7] P. O. Schmidt, T. Rosenband, C. Langer, W. M. Itano, J. C. Bergquist, and D. J. Wineland, Science 309, 749 (2005).

[8] See Supplemental Material 1 for more detailed discussion on the physical interpretation of linewidth reduction and its relevance to the transit time limit.

[9] W. M. Itano, J. C. Bergquist, J. J. Bollinger, J. M. Gilligan, D. J. Heinzen, F. L. Moore, M. G. Raizen, and D. J. Wineland, Phys. Rev. A 47, 3554 (1993).

[10] M. Kitagawa and M. Ueda, Phys. Rev. A 47, 5138 (1993).

[11] J. Hald, J. L. Sørensen, C. Schori, and E. S. Polzik, Phys. Rev. Lett. 83, 1319 (1999).

[12] A. Kuzmich, L. Mandel, and N. P. Bigelow, Phys. Rev. Lett. 85, 1594 (2000).

[13] M. S. Shahriar, P. R. Hemmer, D. P. Katz, A. Lee, and M. G. Prentiss, Phys. Rev. A 55, 2272 (1997).

[14] P. R. Hemmer, M. S. Shahriar, V. D. Natoli, and S. Ezekiel, J. Opt. Soc. Am. B 6, 1519 (1989).

[15] M. Kasevich and S. Chu, Phys. Rev. Lett. 67, 181 (1991).

[16] M. O. Scully and M. S. Zubairy, Quantum Optics (Cambridge University Press, Cambridge,
1997).

[17] R. H. Dicke, Phys. Rev. 93, 99 (1954).

[18] R. Sarkar, M. E. Kim, R. Fang, Y. Tu, and M. S. Shahriar, (2014), arXiv:1408.2296.

[19] F. T. Arecchi, E. Courtens, R. Gilmore, and H. Thomas, Phys. Rev. A 6, 2211 (1972).

[20] See Supplemental Material 2 for detailed analyses on the effects of parameter imperfections, noise, detector efficiency, and collection efficiency.

[21] C. Foot, Atomic Physics (Oxford University Press, New York, 2008).

[22] W. Ketterle and N. J. Van Druten, Adv. At. Mol. Opt. Phys. 37, 181 (1996).

[23] K. Kowalski, V. Cao Long, K. Dinh Xuan, M. Glódz, B. Nguyen Huy, and J. Szonert, Comput. Meth. Sci. Technol. Spec. SI (2), 129 (2010).

[24] L.-M. Duan, M. Lukin, J. I. Cirac, and P. Zoller, Nature 414, 413 (2001).

[25] P. R. Hemmer, M. S. Shahriar, M. Prentiss, D. P. Katz, K. Berggren, J. Mervis, and N. P. Bigelow, Phys. Rev. Lett. 68, 3148 (1992).
As we have shown in the main body, the fact that the linewidth in a CSAC is narrower by a factor of $\sqrt{N}$ can be proven mathematically. However, it is instructive to discuss the physical mechanism that leads to this narrowing. Furthermore, it is also important to address the issue of why the violation of the conventional notion of the transit time limit does not contradict the fundamental laws of quantum mechanics.

**PHYSICAL INTERPRETATION OF LINE NARROWING**

We consider a simple picture of an oscillator and a probe in order to understand the physical explanation for why the linewidth of a CSAC narrows by $\sqrt{N}$. A clock is essentially an oscillator oscillating at frequency $\omega$. In order to ascertain that the oscillator has not drifted, the oscillator frequency is mapped into light and interacts with a two level atom, with the ground state $|1\rangle$ and the excited state $|2\rangle$, and a transition frequency $\omega_0$. If $\omega$ does not match $\omega_0$, an error signal proportional to $\delta = \omega - \omega_0$ is produced to correct for
the difference. Now consider for a moment that we can create a two state superposition of $N$ atoms such that they are all either in the ground state or the excited state. In other words, $|\psi\rangle = C_0 |E_0\rangle + C_N |E_N\rangle$ where $|E_0\rangle = |111...11\rangle$ and $|E_N\rangle = |222...22\rangle$. The energy difference between these two states is $N\omega_0$. The oscillator frequency is still $\omega$, but when a light field with $N$ photons is compared with such a two level system, the difference in energy is $N\delta = N\omega - N\omega_0$. If it were possible to produce an error signal that is proportional to this energy difference without degrading the effective signal to noise ratio (or, more accurately, the ratio of noise to the SVS, as discussed in Supplementary Material 1), the resulting clock would be $N$-fold more accurate.

However, this clean two level superposition of collective states is virtually impossible to achieve with a collection of $N$ atoms and a single field since there is no electric dipole moment to excite the $|E_N\rangle$ state directly from the $|E_0\rangle$ state. What occurs instead is that all the states between these get excited as well, as illustrated in Figure 1. If we consider only the excitations from state $|E_0\rangle$, there are $N$ possible transitions that can occur, so that the error signal includes the set of all the possible detunings, $\delta, 2\delta, 3\delta, ... N\delta$. In other words, there are effectively $N$ different sensors running at the same time. All the other states also act as sensors as they interact with the others. It turns out, as we have proven mathematically in the main body of the paper, that the error signal becomes proportional to $\sqrt{N\delta}$, corresponding to an effective detuning of $\sqrt{N\delta}$. Thus, in effect, the clock transition frequency is enhanced by a factor of $\sqrt{N}$.

In the Ramsey fringe experiment, the error signal that is generated occurs as a result of the phase difference between the interacting states. A detailed picture can be viewed in
Consider first a single two level atom, initially in state $|1\rangle_A$, going through the Ramsey fields. In the Jaynes-Cummings model, when a field with $m$ photons interacts with an atom, the $\pi/2$-pulse will produce the quantum state $|\psi\rangle = |1\rangle_A|m\rangle_\nu - i|2\rangle_A|m-1\rangle_\nu$. The energy of state $|2\rangle_A|m-1\rangle_\nu$ is lower than that of state $|1\rangle_A|m\rangle_\nu$ by $\hbar\delta$. In the second zone, these two composite states evolve freely for a time $T_2$ and accumulate different phases. State $|1\rangle_A$, with energy 0 remains the same, whereas $|2\rangle_A$ with energy $\omega_0$ evolves as $e^{i\omega_0 T_2}$. The field with $m$ photons evolve as $e^{im\omega T_2}$ whereas the field with $m-1$ photons evolve as $e^{i(m-1)\omega T_2}$. Thus, the quantum state of the total system at the end of the dark zone is \begin{equation}
 |\psi\rangle = e^{im\omega T_2}|1\rangle_A|m\rangle_\nu - ie^{i\omega_0 T_2}e^{i(m-1)\omega T_2}|2\rangle_A|m-1\rangle_\nu \end{equation}

The net accumulated phase difference in the two states is $e^{i\delta T_2}$. The third zone where another $\pi/2$-pulse occurs produces interference between the two states, so that when interrogation occurs, the signal produced is in the form of Ramsey fringes that oscillate at frequency $\delta$. Therefore, the energy difference between the two composite states determines the oscillation frequency of the Ramsey fringes. Alternatively, if one were to plot the signal as a function of the dark zone time, $T_2$, the width of the fringe is given by the inverse of this energy difference. If the same calculation is carried out now for a two state system where the ground state is $|E_0\rangle_A|m\rangle_\nu$ and the excited state is $|E_N\rangle_A|m-N\rangle_\nu$, where $|E_0\rangle_A$ and $|E_N\rangle_A$ are the collective states of $N$ atoms, then the energy difference is $N\delta$ and the width of the fringe as a function of $T_2$ would be $1/(N\delta)$ and the width of the Ramsey fringe as a function of $\delta$ will be $(T_2^{-1}/N)$.

As mentioned earlier, such a two level system of collective states for a large value of $N$ is virtually impossible to realize. Instead, for $N$ atoms, the first Ramsey zone produces a superposition of all the states from $|E_0\rangle_A$ to $|E_N\rangle_A$. In the second zone, each of the collective states $|E_k\rangle_A$ accumulates a phase factor of $e^{i(\delta T_2)k}$ with respect to the state $|E_0\rangle_A$. When the atoms pass through the third zone, each of these collective states interferes with one another and contributes to the total population of $|E_N\rangle_A$. It is the collection of these interferences among all the collective states that produces the narrowed linewidth.

We have verified this interpretation explicitly for two atoms. The collective states in this case are (where the subscript $A$ has been dropped) $|E_0\rangle$, $|E_1\rangle$, and $|E_2\rangle$. After they accumulate different phases in the second zone, each of them contributes to the final state $|E_2\rangle$ by amount $\chi_0 = 1/4$, $\chi_1 = e^{i\delta T}/2$, and $\chi_2 = e^{2i\delta T}/4$ respectively. The total signal
Figure 2. Two atom Ramsey fringe experiment; collective states in Ramsey fringe experiment

\[
\begin{align*}
\delta f & \rightarrow |1\rangle \\
|1\rangle & \rightarrow |1\rangle e^{im\omega T_2} |2\rangle e^{i(m-1)\omega T_2} \\
|2\rangle & \rightarrow |2\rangle e^{im\omega T_2} |1\rangle e^{i(m-1)\omega T_2} \\
N\delta & \rightarrow |E_0\rangle \\
|E_0\rangle & \rightarrow |E_0\rangle e^{i(m-N)\omega T_2} \\
|E_0\rangle & \rightarrow |E_0\rangle e^{i(m-N)\omega T_2}
\end{align*}
\]

is \( S_{col} = |\langle E_2|E_2\rangle|^2 = \cos^4 (\delta T_2/2) \). This comes about because \( S_{col} = |\chi_0 + \chi_1 + \chi_2|^2 = |\chi_0 + \chi_1|^2 + |\chi_1 + \chi_2|^2 + |\chi_0 + \chi_2|^2 - (\chi_0^2 + \chi_1^2 + \chi_2^2) \). In other words, it is as though \(|E_0\rangle\) and \(|E_1\rangle\) interfered together to produce Ramsey fringes at frequency \( \delta \), \(|E_1\rangle\) and \(|E_2\rangle\) interfered together to produce Ramsey fringes at frequency \( \delta \), and \(|E_0\rangle\) and \(|E_2\rangle\) interfered together to produce Ramsey fringes at frequency \( 2\delta \); the signal observed is the addition of all these Ramsey fringes minus an overall factor (see Figure 3), which is due to the fact that the actual process is a simultaneous interference between the three states.

VIOLATION OF THE CONVENTIONAL NOTION OF THE TRANSIT TIME LIMIT

The narrowing of the CSAC fringe as given by \( \Gamma(N) = \Gamma(1)/\sqrt{N} = \pi/(T_2\sqrt{N}) \) violates the conventional transit time limit, which constrains the fringe width to be at least \( \sim 1/T_2 \). This is a manifestation of the uncertainty relation \( \Delta f \cdot \Delta t \geq 1 \), which apparently follows from the Heisenberg uncertainty principle of \( \Delta E \cdot \Delta t \geq \hbar \). However, when we properly define \( \Delta f \) as the uncertainty in the fringe width – in the case of the Ramsey technique considered here – and \( \Delta t \) as the total observation time, we can derive the uncertainty relations more systematically and show that despite the fact that the conventional transit time limit is violated, the Heisenberg uncertainty principle is not violated.

First, consider a single atom that undergoes the Ramsey fringe experiment. The uncertainty in the fringe width is \( \Delta f = (1/T_2) \), where \( T_2 \) is the separation period between the two \( \pi/2 \) pulses. When the experiment is repeated \( m \) times, it is as though the separation period expands \( m \)-fold, so that the effective observation time is in fact \( \Delta t = mT_2 \), and the uncertainty in the fringe width is \( \Delta f = (1/T_2)/\sqrt{m} \) in the standard quantum limit (SQL).
Figure 3. In a two atom ensemble, each of the three collective states interfere with one another to produce different Ramsey fringes (a)-(c). The overall envelope is not drawn. The sum of these interferences gives the narrowing of the fringe linewidth as seen in (d). In (d), the dotted curve represents the signal from a single atom and the solid curve the signal from two atoms for comparison.

and $\Delta f = (1/T_2)/m$ in the Heisenberg limit (HL). Hence, the product $\Delta f \cdot \Delta t$ yields $\sqrt{m}$ in the SQL and 1 in the HL. Note that as $m \to 1$, the SQL approaches the HL, which is the more vigorous and fundamental limit.

Next, consider $N$ atoms in the same Ramsey fringe experiment during a single trial. Since each atom, in its individual state, is considered separately from the rest, having $N$ atoms is equivalent to running $N$ trials simultaneously. The effective observation time in this case is $\Delta t = NT_2$, and the uncertainties in the fringe width are $\Delta f = (1/T_2)/\sqrt{N}$ in the SQL and $\Delta f = (1/T_2)/N$ in the HL. Moreover, if the experiment is repeated $m$ times, the effective observation time increases to $\Delta t = mNT_2$, and the uncertainties in the fringe width are $\Delta f = (1/T_2)/\sqrt{mN}$ in the SQL and $\Delta f = (1/T_2)/(mN)$ in the HL. Thus, we find that the uncertainty relations for $N$ atoms and $m$ trials are $\Delta f \cdot \Delta t = \sqrt{mN}$ in the SQL and $\Delta f \cdot \Delta t = 1$ in the HL.
Consider next the CSAC case, containing \( N \) atoms, and repeated \( m \) times. As we have shown in the body of the paper, the frequency fluctuation in the CSAC is \( \Delta f = \frac{1}{(T_2 \sqrt{mN})} \) for ideal detection efficiency. It may not be obvious what the effective observation time is for this case. However, given the fact that, under ideal detection efficiency, the CSAC is equivalent to the case of \( N \) atoms repeated \( m \) times, we are led to conclude that the effective observation time is \( \Delta t = T_2 mN \). As such, we get \( \Delta f \cdot \Delta t = \sqrt{mN} \), which is the SQL in this case. In the HL, we could get \( \Delta f \cdot \Delta t = 1 \). Thus, we see that when the frequency uncertainty and the observation times are interpreted properly, the CSAC signal does not violate the fundamental quantum limit.

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An N-atom Collective State Atomic Clock with Root-N Fold Increase in Effective Frequency and Root-N Fold Reduction in Fringe Width - Supplemental Material 2: Effects of Parameter Imperfections, Noise, Detector Efficiency, and Collection Efficiency

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As noted in the body of the paper, under ideal conditions, the signal for the collective state atomic clock (CSAC) is proportional to $S_{col} = \prod_{i=1}^{N} P_{2i} = (P_2)^N = \cos^{2N}(\delta T_2/2)$ with $P_2 = \cos(\delta T_2/2)$. However, various non-idealities can cause a significant modification to this signal. Furthermore, it may not be obvious as to what the minimum detectable frequency fluctuation would be, due to the effects of fundamental quantum noise and other sources of fluctuations. We address these issues in this supplement.

The first section examines inhomogeneities in experimental parameters that affect the signal. The atoms in an ensemble, however cold, are not stationary but have a distribution of velocities. For the corresponding conventional clock (CC), this leads to a broadening of the Ramsey fringes and a modest reduction in the peak amplitudes. In contrast, for the CSAC, this effect reduces the peak amplitude very significantly, while the width of the fringes remains essentially unchanged. Another effect of importance is that fields are not uniform across the width of the atomic ensemble. Hence, the overall signal is affected strongly by the ratio of the Gaussian beam width to the width of the atomic ensemble. The second
section compares the effect of quantum and classical noise in the CC and in the CSAC. For simplicity, this section assumes that none of the inhomogeneities in the first section exist and that the ideal signal can be generated. Finally, the third section examines the effects of the interrogation time and the detector efficiency, assuming that the ideal signal can be generated.

**EFFECTS OF VELOCITY DISTRIBUTION AND FIELD INHOMOGENEITY**

A two level atomic system $|\psi\rangle$ interacts with light fields and evolves as $|\psi(t' + t)\rangle = W_{\Omega t}^{\delta t}|\psi(t')\rangle$, where

$$W_{\Omega t}^{\delta t} = e^{i\delta t/2} \begin{pmatrix} \cos \phi - i \frac{\delta}{\Omega} \sin \phi & -i \frac{\Omega}{\Omega'} \sin \phi \\ -i \frac{\Omega}{\Omega'} \sin \phi & \cos \phi + i \frac{\delta}{\Omega} \sin \phi \end{pmatrix}$$

(1)

The two levels in the proposed scheme are, for example, the hyperfine ground states of an alkali atom such as $^{85}$Rb. After the $\pi/2$-dark-$\pi/2$ sequence, the system is in state $|\psi\rangle = W_{\Omega t}^{\delta t} W_{0}^{\delta T_{2}} |1\rangle$. Unlike in the main body of the paper, we here do not make the approximation that $\delta \ll \Omega$. Then the signal we expect to see for a single atom is proportional to $P_{2} = |\langle 2 | \psi \rangle|^{2} = |\langle 2 | W_{\Omega t}^{\delta t} W_{0}^{\delta T_{2}} |1\rangle|^{2}$, and the collective state signal is

$$S_{col} = \prod_{i=1}^{N} |\langle 2 | W_{\Omega t}^{\delta t} W_{0}^{\delta T_{2}} W_{\Omega t}^{\delta t} W_{0}^{\delta T_{2}} |1\rangle|^{2} = |\langle 2 | W_{\Omega t}^{\delta t} W_{0}^{\delta T_{2}} W_{\Omega t}^{\delta t} W_{0}^{\delta T_{2}} |1\rangle|^{2N}$$

(2)

We assume that the density of atoms in the trap is fixed at $3 \times 10^{7}$ mm$^{-3}$, so that the width of the atomic ensemble, which has a Gaussian spatial distribution, varies with the number of atoms. With $N = 10^{6}$ atoms in the trap, the size of the MOT is 0.2 mm.

When an atom with velocity $v$ interacts with a field with frequency $\omega$ propagating in the direction of the atom, the frequency of the field is shifted by $\delta_{D} = v\omega/c$. The Maxwell Boltzmann velocity distribution is $\rho_{MB}(v,T) = \sqrt{m_{a}/(2\pi kT)} exp^{-m_{a}v^{2}/(2kT)}$ where $m_{a}$ is the atomic mass and $T$ is the temperature. We assume the temperature to be given by the Doppler cooling limit, so that $T_{MOT} = \Gamma_{Rb} h/(2k) = 146$ $\mu$K for $^{85}$Rb. The average velocity is then $v_{av} \sim 19.0$ cm/s, with a corresponding Doppler shift of $\delta_{D_{av}} = 2.0$ Hz. Under these conditions, the signal is

$$S_{Dop} = \prod_{v' = -v_{av}}^{5v_{av}} |\langle 2 | W_{\pi/2}^{(\delta+\delta_{D}(v'))} W_{0}^{\delta_{D}(v')} |1\rangle|^{2} \rho_{MB}(v',T_{MOT})$$

(3)
where we take into account velocities that are up to five times the $v_{av}$. Plotted in Figure 1 are the signals $S_{col}$ and $S_{Dop}$ for various $N$ values, with $T_2 = 10^{-4}$ s and $\Omega = 5 \cdot 10^6$ s$^{-1}$. The Doppler effect decreases the overall signal while having virtually no effect on its width. It decreases exponentially as $N$ increases. However, for the given choice of temperature and $N = 10^6$, this effect is negligible on the signal so that $S_{Dop} \simeq S_{col}$. We can therefore eliminate the Doppler effect as an experimental impediment. 

Consider next the effect of the inhomogeneity in the laser field amplitude. We assume that the atomic ensemble has a Gaussian spread with a width of $\omega_A$: $\rho_N(\gamma) = \rho_0 e^{-\left(\gamma^2/\omega_A^2\right)}$. Each of the two laser fields that produce the Raman-Rabi excitation is also assumed to have a Gaussian profile with a width of $\omega_L > \omega_A$. Since the Raman-Rabi frequency is proportional to the product of the Rabi frequencies for each of these lasers, it follows that the Raman-Rabi frequency is also a Gaussian with a width of $\omega_L$: $\Omega(\gamma) = \Omega_0 e^{-\left(\gamma^2/\omega_L^2\right)}$. The peak value of $\Omega$ (i.e., $\Omega_0$) is chosen so that the atoms at the center ($r = 0$) experience a
Figure 2. Collective state signal at the end of the Ramsey field experiment for various Gaussian beam widths. \( N = 10^6 \) atoms; \( \Omega = 5 \cdot 10^6 \); \( T_2 = 10^{-4} \) s. Dashed line plots the ideal signal, \( S_{\text{col}} \). Solid line shows the reduced signal, \( S_\Omega \), where the effect of inhomogeneity in intensity of the laser beam is taken into account.

The signals for various ratios of \( w_L/w_A \) are plotted in Figure 2. To ensure that the peak signal amplitude is at 80 \% of the ideal signal value, \( \omega_L \) must be at least \( \sim 20 \omega_A \). However, this is not a demanding constraint if there are \( N = 10^6 \) trapped atoms. With the typical density of \( 10^7 \text{ mm}^{-3} \), the width of the Gaussian beam needs to be only 4 mm.

**EFFECTS OF QUANTUM AND CLASSICAL NOISE**

In order for the CSAC to be useful, it must perform at least as well as, or better than, the CC, and for that, we must compare the two clocks' stability in the short term and the
long term regimes. The stability of a clock can be measured by investigating the frequency fluctuation that has both quantum mechanical and classical components. Before comparing the stabilities of the CSAC and the CC, it is instructive first to review briefly the stability of a CC.

For concreteness, we consider an off-resonant Raman-Ramsey clock as the CC. The population of the detected state $|2\rangle$ at the end of the second pulse is given by $P_2 = \cos^2(fT_2/2)$, where $T_2$ is the separation period of the two $\pi/2$-pulses and $f$ is the deviation of the clock frequency away from its ideal value, expressed in radial units (i.e. rad/s rather than Hz). The signal is detected by probing the desired state for a duration of time. If $\tilde{N}$ is the number of atoms per unit time and $\tau$ is the interrogation period, the net signal is $S_{sa} = \tilde{N}\tau P_2 = \tilde{N}\tau \cos^2(fT_2/2)$. For the sake of comparison, we allow the number of atoms per trial in the CSAC signal, $N$, multiplied by the number of trials, $m$, to equal $\tilde{N}\tau$. Therefore, we can write $S_{sa} = mN \cos^2(fT_2/2)$. The quantum mechanical variance of this quantity is $\Delta S_{QM,sa} = (\sqrt{mN}/2) \sin(fT_2)$, where the derivation is made by noting that the fluctuations in $mN$ is $\sqrt{mN}$ [1], and the projection noise in a single two level atomic system is $\Delta P_2 = \sqrt{P_2(1-P_2)}$ [1]. (It should be noted that the fluctuation in $mN$ is also a manifestation of this projection noise, as discussed in detail in [1].) When the probability of finding the population in this state is unity or nil, the projection noise vanishes; on the other hand, it is largest at $P_2 = 1/2$. Calculating the slope from the signal, we find that $\partial S_{sa}/\partial f = -[mN/(2\gamma_{sa})] \sin(fT_2)$, where $\gamma_{sa} = 1/T_2$ is the linewidth.

Assuming perfect quantum efficiency for the detection process, the frequency fluctuation can be written as $\delta f|_{total} = (\Delta S_{QM} + \Delta S_{class})/(\partial S/\partial f)|$, which can be regarded as noise ($\Delta S$), both quantum and classical, over the Spectral Variation of Signal ($\partial S/\partial f$), or SVS. In what follows, we consider first the effect of quantum noise only. Thus, the quantum frequency fluctuation (QFF) for a CC can be expressed as

$$\partial f_{QM,CC} \equiv \frac{\Delta S_{QM,sa}}{(\partial S_{sa}/\partial f)} = \frac{\gamma_{sa}}{\sqrt{mN}} \quad (5)$$

It should be noted that while both $\Delta S_{QM}$ and $(\partial S/\partial f)$ depend on $f$, their ratio is a constant, which is merely an accident due to the fact that the signal is cosinusoidal. However, this accidental cancellation has led to an apparently simple perception of the QFF as being simply the ratio of the linewidth $(\gamma_{sa})$ to the SNR, where the SNR is understood to be $\sqrt{mN}$. This expression for the SNR, in turn, follows from thinking about the signal as
being \( S' = mN \) and noise \( N' \) as being \( \sqrt{mN} \), so that \( \text{SNR} \equiv S'/N' = \sqrt{mN} \). However, it should be clear from the discussion above that the signal is not given by \( mN \), and noise is not given by \( \sqrt{mN} \); rather, they both depend on \( f \).

In cases where frequency fluctuation is not a constant (as will be the case for the CSAC), we can no longer measure the stability of the clock in terms of a constant \( \gamma/\text{SNR} \). Instead, it is necessary to carry out the full calculation of the frequency fluctuation as a function of frequency. Thus, we will adopt the convention that the net frequency fluctuation, \( \delta f \), should be thought of as the ratio of the noise to the SVS. This approach should be adopted universally for all metrological devices. Of course, for devices where the relevant quantity is not the frequency, the definition should be adapted accordingly. For example, in an interferometer that measures phase, the relevant quantity can be expressed as follows: net phase fluctuation is the ratio of the noise to the Angular Variation of Signal (AVS).

Following this convention, we can now examine the net frequency fluctuation of the CSAC and compare it to that of the CC. We will first compare their quantum fluctuations, which is relevant in the short term regime, and then the classical fluctuations, which dominates the long term regime. The collective state signal for \( m \) trials is \( S_{\text{col}} = mP_N^C = m \cos^2N \left( fT_2/2 \right) \) and the projection noise is \( \Delta P_N^C = \sqrt{P_N^C(1 - P_N^C)} \) for a single trial and \( \Delta P_N^C = \sqrt{m}\sqrt{P_N^C(1 - P_N^C)} \) for \( m \) trials, so that the total quantum mechanical noise in the signal is

\[
\Delta S_{QM,\text{col}} = \sqrt{m\cos^N \left( fT_2/2 \right)} \sqrt{1 - \cos^2N \left( fT_2/2 \right)}
\]

and the SVS is

\[
\partial S_{\text{col}}/\partial f = -(mN/\gamma_{sa}) \sin \left( fT_2/2 \right) \cos^{2N-1} \left( fT_2/2 \right)
\]

Therefore, the frequency fluctuation in the CSAC due solely to quantum noise can be expressed as:

\[
\delta f_{QM,\text{CSAC}} = \left| \frac{\gamma_{sa}}{N\sqrt{m}} \sqrt{\frac{1 - P_N^C}{P_N^C}} \cot \left( \frac{fT_2}{2} \right) \right|
\]

where \( P_N^C \) is a function of \( f \). Thus, unlike in the case of the CC, the frequency fluctuation is not a constant, and depends strongly on \( f \).

We consider first the limiting case of \( f \to 0 \). Using Taylor expansion, it is easy to see that

\[
\delta f_{QM,\text{CSAC}} \simeq \frac{\gamma_{sa}}{\sqrt{mN}}
\]
Figure 3. (left) Ratio of the QFF in the CC to the QFF in the CSAC, for $T_2 = 10^{-4}$ s, $m = 1000$ and $N = 10^6$. It should be noted that the fluctuation in the CC is independent of $f$ while that of the CSAC varies significantly with $f$. (right) Ratio of the SVS of the CSAC to the SVS of the CC for $T_2 = 10^{-4}$ s, $m = 1000$ and $N = 10^6$. The vertical lines in the plots show where the FWHM are.

which is the same as that of the CC, given in Eq. (5). This can be understood physically by noting that while the fringe width becomes much narrower for the CSAC, the SNR also decreases due to the fact that a single observation is made for all $N$ atoms in a given trial.

The QFF for the CSAC, given in Eq. (8), is smallest as $f \rightarrow 0$ and increases as $f$ moves away from resonance. The ratio of the QFF for the CC, given in Eq. (5), to that of the CSAC, given in Eq. (8), is plotted as a function of $f$ in the left side of Figure 3 for $T_2 = 10^{-4}$ s, $m = 1000$ and $N = 10^6$. Here, the vertical bars indicate the FWHM of the CSAC signal. It is clear from this plot that the QFF for the CSAC increases significantly as we move away from resonance. However, since a servo will keep the value of $f$ confined to be close to zero, the frequency stability of the CSAC, under quantum noise limited operation, should be very close to that of the CC, assuming that all the other factors remain the same.

The classical frequency fluctuation (CFF), $\partial f|_{\text{class}} = \Delta S_{\text{class}}/(\partial S/\partial f)$, is the limiting factor in the long term stability. While the quantum fluctuation is dominated by quantum projection noise, the classical noise is dominated by noise in the electronics employed to generate the clock signal. Since the pieces of equipment used in the development of both the CSAC and CC suffer from similar noise issues, the variance $\Delta S$ is of the same order of magnitude for both clocks. On the other hand, the SVS, $(\partial S/\partial f)$, is not the same, as was
shown previously. The ratio of the SVS of the CSAC to the SVS of the CC is
\[
\frac{\partial S_{\text{col}}}{\partial f} / \frac{\partial S_{\text{sa}}}{\partial f} = \frac{\cos^2 N \left( \frac{fT_2}{2} \right)}{\cos^2 \left( \frac{fT_2}{2} \right)} = \frac{P^C_N}{P_2}
\]  
\[\text{(10)}\]
and is plotted in Figure 3 (right). With \(\Delta S_{\text{class, col}} \sim \Delta S_{\text{class, sa}}\), the ratio of the CFF of the CSAC to the CFF of the CC can be written
\[
\frac{\delta f_{\text{class, CSAC}}}{\delta f_{\text{class, CC}}} \approx \frac{\cos^2 \left( \frac{fT_2}{2} \right)}{\cos^2 N \left( \frac{fT_2}{2} \right)}
\]  
\[\text{(11)}\]
Similar to the ratio of the two clocks in QFF, Eq. (11) is smallest as \(f \to 0\) and increases as \(f\) moves away from resonance. Thus, with respect to both quantum and classical sources of noise, the CSAC must be operated near \(f \approx 0\) for optimal performance.

We have investigated the effects of quantum and classical noise by deriving the expression for fluctuation in frequency. However, as was shown in the first section, the signal is also a function of other experimental variables; and in general, the fluctuations in any of these can be expressed as
\[
\partial A \equiv \left| \frac{\Delta S_{QM}(A) + \Delta S_{\text{class}}(A)}{\partial S(A)/\partial A} \right|
\]  
\[\text{(12)}\]
where \(A\) is the variable whose fluctuation is of interest, and the signal \(S\) is expressed in terms of \(A\).

**EFFECT OF DETECTOR EFFICIENCY**

We recall briefly that in the CSAC detection scheme, a laser with a frequency corresponding to one leg of the Raman transition interacts with the atoms, which are in the quantum state \(|\psi\rangle = c_N |E_N\rangle + \sum_{j=0}^{N-1} c_j |E_j\rangle\). Interaction between this field, the atoms, and the free space vacuum modes on the other leg would lead to production of photons unless \(c_N = 1\) and \(c_j = 0\) for all \(j\). These photons are detected using a heterodyning technique, as described in the main body of the paper. The voltage output of the heterodyning system is proportional to the amplitude of the electric field corresponding to the photons.

In general, one or more photons are produced as \(|E_j\rangle\) decays to \(|E_{j+1}\rangle\) and subsequent states. The time needed for these photons to be produced depends on the vacuum and probe field induced Raman transition rates between \(|E_j\rangle\) and \(|E_{j+1}\rangle\). If one assumes perfect efficiency for detecting each of these photons, and waits for a time long compared to the inverse of the weakest of these transition rates, then the detection of no photons implies
that the system is in state $|E_N\rangle$. In practice, we can choose a small threshold voltage at the output of the heterodyning system as an indicator of null detection. Thus, any signal below this threshold would be viewed as detection of the quantum system in the $|E_N\rangle$ state, and all signals above this threshold would be discarded. The number of events below this threshold for $m$ trials carried out with all the parameters of the experiment unchanged, is the derived signal for the CSAC. After collecting data for all the values of detuning that is of interest, the result would ideally yield the plot of the CSAC signal $S_{col} = |c_N|^2$, averaged over $m$ trials. However, with a fractional detector efficiency and finite detection period, the signal would deviate from the ideal result.

Consider first the effect of the detection period. Given the decay rate of the off-resonant Raman process, $\gamma_j = (j + 1)(N - j)\gamma_{sa}$ as described in the main body, the probability that $|E_j\rangle$ will produce zero photons during the measurement period $\tau$ is $P_{0,j} = e^{-\gamma_j \tau}$. Thus, the total probability of zero photon emission (which should vanish ideally for any $c_j \neq 0$) is given by $P_0 = \sum_{j=0}^{N-1} |c_j|^2 e^{-\gamma_j \tau}$. The collective state signal, $S_{col}$, is the total probability of finding zero photons during $\tau$, and can be expressed as $S_{col} = |c_N|^2 + \sum_{j=0}^{N-1} |c_j|^2 e^{-\gamma_j \tau}$. Noting that $\gamma_N = 0$, we can rewrite this compactly as $S_{col} = \sum_{j=0}^{N} |c_j|^2 e^{-\gamma_j \tau}$. The lower and upper bounds of $S_{col}$ can be established by considering the strongest and the weakest effective decay rates. The strongest decay rate occurs for the middle state, $\gamma_{N/2} = (N/2)(N/2 + 1) \approx (N^2/4)\gamma_{sa}$, where $N \gg 1$ approximation has been made. With the substitution of the largest decay rate for each $|E_j\rangle$ into the equation for $S_{col}$, the lower bound is set by

$$S_{LB} = |c_N|^2 + (1 - |c_N|^2) e^{-N^2/4\gamma_{sa} \tau} \quad (13)$$

Likewise, with the substitution of the weakest decay rate for each $|E_j\rangle$, $\gamma_0 = \gamma_{N-1} = N\gamma_{sa}$, into $S_{col}$, the upper bound is set by

$$S_{UB} = |c_N|^2 + (1 - |c_N|^2) e^{-N\gamma_{sa} \tau} \quad (14)$$

The signal produced in time $\tau$ will then lie somewhere between the lower and the upper bounds.

Consider next the effect of non-ideal detection efficiency of the heterodyning scheme. To be concrete, let us define as $\eta$ the efficiency of detecting a single photon. In practice, this parameter will depend on a combination of factors, including the quantum efficiency of the high-speed photodetector and the overlap between the probe laser mode and the mode of the
emitted photon. For the CSAC, it should be noted that we are interested in knowing only whether one or more photons have been detected, and not in the actual number of photons. When more photons are emitted, the detector will have a better chance of observing a non-zero signal, and hence distinguish zero photon emission from the rest with more certainty. For example, if three photons are emitted during the interrogation time, then four different outcomes are possible:

- All three photons are detected, with probability \( \eta^3 \);
- Two of the photons are detected, with probability \( \eta^2(1 - \eta) \); this can occur for any two of the photons, so the multiplicity is 3;
- One photon is detected, with probability \( \eta(1 - \eta)^2 \) and multiplicity of 3.
- No photons are detected, with probability \( \epsilon^3 \) where \( \epsilon \equiv 1 - \eta \)

The sum of these probabilities is 1. The probability that at least 1 photon is detected is thus \( 1 - \epsilon^3 \). For any state \( j \neq N \), the probability of detecting at least 1 photon is therefore \( 1 - \epsilon^{N-j} \).

Moreover, we must also consider how the effective detection efficiency is influenced by the fact that the collective states decay at different rates. Specifically, the \( j \)th level for \( j < N \) might produce \( N - j \) photons, \( N - j - 1 \) photons, down to no photons, depending on the length of the measurement time and the effective decay rate. If the system is in the state \( |E_{N-3}\rangle \), for example, it can produce up to 3 photons but with probabilities that change over the course of the detection period. For a given time \( \tau \), \( |E_{N-3}\rangle \) evolves into a sum of the states \( |E_{N-3}\rangle \rightarrow \sum_{k=N-3}^{N} a_{jk}(\tau)|E_k\rangle \), where the coefficient \( a_{jk}(\tau) \) depends on the effective decay rate that is specific to each state, and changes as the states evolve in time. The detector efficiency can be inserted to show the true probability of detecting a non-zero signal, keeping in mind that no photon is produced if the ensemble remains in state \( |E_{N-3}\rangle \), produces 1 photon by evolving to state \( |E_{N-2}\rangle \), and so on. Then the probability of at least one photon being produced during a period of \( \tau \) is

\[
P_{N-3} = \sum_{k=N-3}^{N} (1 - \epsilon^{k-N+3}) |a_{jk}(\tau)|^2
\]
Thus, the total probability of detecting at least one photon is:

\[ P = \sum_{j=0}^{N-1} |c_j|^2 \sum_{k=j}^{N} \left( 1 - \varepsilon^{k-j} \right) |\alpha_{jk}(\tau)|^2 \]  

The probability of seeing no photon is

\[ S_{col} = 1 - P = 1 - \sum_{j=0}^{N-1} |c_j|^2 \sum_{k=j}^{N} \left( 1 - \varepsilon^{k-j} \right) |\alpha_{jk}(\tau)|^2 \]  

The numerical analysis for a large number of atoms is tedious and scales as at least \((N - 1)!\) for the CSAC. However, we can take the worst case scenario to serve as the upper bound for the signal. The worst case occurs when only a single photon is produced as a result of \(|E_j\rangle\) decaying to only the \(|E_{j+1}\rangle\) state, so that the index of the second summation stops at \(k = j + 1\). In this case, we can write \(|a_{j,j+1}(\tau)| = (1 - e^{-\gamma_j \tau})\) and the signal becomes

\[ S_{col} = |c_N|^2 + \varepsilon \left( 1 - |c_N|^2 \right) + \eta \sum_{j=0}^{N-1} |c_j|^2 e^{-\gamma_j \tau} \]  

Now, using the approach we employed in arriving at equations Eq. \((13)\) and Eq. \((14)\), we now consider the strongest and the weakest decay rates for single photon production to arrive at the lower and upper bounds of the zero photon count signal:

\[ S_{LB} = 1 - \eta \left( 1 - |c_N|^2 \right) \left( 1 - e^{-\frac{N^2}{T_2} \gamma_{sa} \tau} \right) \]  

\[ S_{UB} = 1 - \eta \left( 1 - |c_N|^2 \right) \left( 1 - e^{-N \gamma_{sa} \tau} \right) \]  

Figures in 4 are plots of the ideal signal (under infinite detection time and \(\eta = 1\)), the lower bound, and the upper bound for various values of \(\tau\) and \(\eta\) for \(N = 10^6\), \(T_2 = 10^{-4}\) s, and \(\gamma_{sa} = 10^5\) s\(^{-1}\). As can be seen, the detector efficiency and measurement time do not affect the peak value of the amplitude. As the signal trails off for non-zero detuning, however, the difference increases. The decrease in \(\eta\) affects both \(S_{UP}\) and \(S_{LB}\) similarly, whereas the effect of the decrease in \(\tau\) is more evident in \(S_{UB}\). With the given parameters, the interrogation period of \(\tau = 10^{-5}\) s and detector efficiency of \(\eta = 0.99\) yields almost ideal signal. A somewhat lower value of \(\eta\) (e.g. 0.77) still yields a signal that is nearly ideal near zero detuning, which is the desired operating regime for the CSAC, as pointed out earlier.

If we set \(\gamma_{sa} \tau = 1\), the signal depends on \(\eta\) as

\[ S_{col} \simeq 1 - \eta \left[ 1 - \cos^{2N} \left( fT_2/2 \right) \right] \]
Figure 4. Plot of the ideal signal (dashed blue line), the upper bound (red line), and the lower bound (green line) for $N = 10^6$, $T_2 = 10^{-4}$ s, and $\gamma_{sa} = 10^5$ s$^{-1}$. Note that in (c) and (d), the upper and lower bounds are virtually indistinguishable.

for large $N$ and $m = 1$. Hence, we can calculate the QFF for the CSAC to see how it depends on the detector efficiency, and how it compares to the CC. For the CC, it is straightforward to show that with $S_{sa} = \eta N \cos^2 (fT_2/2)$, the quantum mechanical noise in the signal is $\Delta S_{sa} = \sqrt{\eta N} \cos (fT_2/2) \sin (fT_2/2)$ and the SVS is $|\partial S_{sa}/\partial \delta| = (\eta N/\gamma_{sa}) \cos (fT_2/2) \sin (fT_2/2)$, so that the QFF is $\delta f_{Q,CC} = \gamma_{sa}/\sqrt{\eta N}$. It is also straightforward to calculate the QFF for the CSAC. The total quantum mechanical noise in the CSAC signal in Eq. (21) is:

$$\Delta S_{Q,\text{col}} = \sqrt{\eta} \cos^N (fT_2/2) \sqrt{1 - \cos^{2N}(fT_2/2)}$$

and the SVS is

$$\partial S_{\text{col}}/\partial f = -(\eta N/\gamma_{sa}) \sin (fT_2/2) \cos^{2N-1}(fT_2/2)$$

Thus, the QFF in the CSAC is:

$$\delta f_{Q,\text{CSAC}} = \left| \frac{\gamma_{sa}}{N \sqrt{\eta}} \sqrt{\frac{1 - P_N^c}{P_N^c}} \cot \left( \frac{fT_2}{2} \right) \right|$$
which approaches $\gamma_{sa}/\sqrt{\eta N}$ as $f \to 0$. Assuming that the detector efficiencies of the CSAC and the CC can be essentially the same, they do not affect the ratio of the two QFFs.

**EFFECT OF COLLECTION EFFICIENCY**

We consider next the effect of the collection efficiency, $\beta$. The signal, for both the CSAC and CC, is directly proportional to $\beta$. Thus, it is easy to see, using Eqs. (5) and (8) of this supplement, that

$$
\zeta \equiv \frac{\delta f_{QM,CSAC}}{\delta f_{QM,CC}} = \left[ \frac{1}{\sqrt{N}} \sqrt{1 - \frac{P_C}{P_N}} \cot \left( \frac{fT_2}{2} \right) \right] \sqrt{\frac{\beta_{CC}}{\beta_{CSAC}}} \tag{25}
$$

where $\beta_{CC}$ ($\beta_{CSAC}$) is the collection efficiency of the CC (CSAC).

As noted above, the quantity written in the square bracket in Eq. (25) approaches unity as $f \to 0$. Thus, in this limit, we see that the ratio of the QFF for the CSAC to that of the CC would depend on the ratio of the collection efficiencies of the detection processes. As discussed in the main body, the coherent stimulated Raman scattering based detection method used for the CSAC process has a collection efficiency that is close to unity, or $\beta_{CSAC} \simeq 1$. As for the CC, the fluorescence is collected from the spontaneous emission process, which emits photons in a dipolar radiation pattern. We can estimate typical values of $\beta_{CC}$ by considering, for example, a CC that makes use of cold atoms released from a MOT. For a lens placed at a distance of 5 cm, with a diameter of 2.5 cm, ignoring the dipolar pattern of radiation for simplicity, and assuming it to be uniform in all directions, this system yields a value of $\beta_{CC} \simeq r^2/(4d^2) = 1/16$ corresponding to $\zeta \sim 0.25$. In a typical CC, various geometric constraints make it difficult to achieve a value of $\beta_{CC}$ much larger than this. In fact, in cases where the total volume occupied by the CC has to be constrained in order to meet the user requirements, the value of $\beta_{CC}$ is typically 1%, which would correspond to $\zeta \sim 0.1$. Thus, the near unity collection efficiency of the CSAC can lead to an improvement of the clock stability by as much as a factor of 10.

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[1] W. Itano, J. Bergquist, J. Bollinger, J. Gilligan, D. Heinzen, F. Moore, M. Raizen, and D. Wineland, Phys. Rev. A **47**, 3554 (1993).