Prospects for a Millihertz-Linewidth Laser

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We propose a new light source based on having alkaline-earth atoms in an optical lattice collectively emit photons on an ultranarrow clock transition into the mode of a high Q resonator. The resultant optical radiation has an extremely narrow linewidth in the mHz range, even smaller than that of the clock transition itself due to collective effects. A power level of order 10^{-12} W is possible, sufficient for phase locking a slave optical local oscillator. Realizing this light source has the potential to improve the stability of the best clocks by 2 orders of magnitude.

Time and frequencies are the quantities that we can measure with the highest accuracy by far. From this fact derives the importance of clocks and frequency standards for many applications in technology and fundamental science. Some applications directly relying on atomic clocks are GPS, synchronization of data and communication networks, precise measurements of the gravitational potential of Earth, radio astronomy, tests of theories of gravity, and tests of the fundamental laws of physics.

With the advent of octave spanning optical frequency combs [1,2] it has become feasible to use atomic transitions in the optical domain to build atomic clocks. Optical clocks based on ions [3] and ultracold neutral atoms confined in optical lattices [4] have recently demonstrated a precision of about 1 part in 10^{15} at 1 s and a total fractional uncertainty of 10^{-16} [4] or below [3], surpassing the primary cesium microwave standards [5,6].

The state-of-the-art optical atomic clocks do not achieve the full stability that is in principle afforded by the atomic transitions on which they are founded. These transitions could have natural line Qs of order 10^{18}, exceeding the fractional stability of the clocks by a factor of ~ 100. The main obstacle that prevents us from reaping the full benefit of the ultranarrow clock transitions is the linewidth of the lasers used to interrogate these transitions. These lasers are stabilized against carefully designed passive high-Q cavities and achieve linewidths <1 Hz, making them the most stable coherent sources of radiation. It is mainly the thermal noise of the reference cavity mirrors that prevent a further linewidth reduction [7] and substantially reducing this noise is hard [8].

An elegant solution to these problems would be to directly extract light emitted from the ultranarrow clock transition [9]. That light could then be used as an optical phase reference, circumventing the need for an ultrastable reference cavity. Unfortunately, the fluorescence light emitted on a clock transition is too weak for practical applications. For instance, for 10^6 fully inverted ^{87}Sr atoms the power of the spontaneously emitted light is of the order of 10^{-16} W.

The key observation that motivates this work is that if we could coerce the ensemble of atoms to emit the energy stored in them collectively rather than individually, the resulting power of order 10^{-12} W would be large enough to be technologically relevant. We show that such collective emission of photons can indeed be accomplished if the atoms are located inside a high-Q cavity. The collective interaction between atoms and cavity fields [10–13] as well as lasers based on emission from microscopic atom samples [14–17] are of great theoretical and experimental interest in quantum optics.

Such a laser would operate in a strikingly different regime from conventional lasers: the cavity relaxation rate exceeds the atomic relaxation rates by many orders of magnitude. This system is thus an extreme case of a bad-cavity laser. Such bad-cavity lasers have been studied in the past [18–20], although in those papers the separation of time scales was not nearly as large as is considered here. In the microwave domain masers are prime examples of a bad-cavity laser. Also in the previous systems the cavity field contains macroscopic numbers of photons while in our system the field occupation number can be $\ll 1$. The extremity of the current system makes it necessary that we revisit the foundations of the theory to obtain reliable predictions. We will focus on the example of ^{87}Sr confined in a lattice potential to make the presentation clearer and more concise. Our results however are general and can be easily translated to other alkaline-earth-like atomic systems with similar structure.

The general setup is shown schematically in Fig. 1. We consider N ultracold two level atoms with transition frequency ω_a confined in an optical lattice. e and g correspond to the two clock levels, $^3P_0$ and $^1S_0$ in ^{87}Sr. The atoms have a spontaneous decay rate γ and we model all inhomogeneous processes by an effective relaxation rate T_2^{-1} for the atomic dipole. The laser transition is coupled to a single mode of a high Q-optical resonator with resonance frequency ω_c and linewidth κ. We assume that the atoms are held in place by the external optical lattice potential exactly in such a way that all atoms couple maximally and
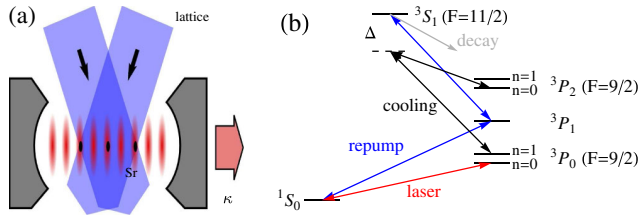
FIG. 1 (color online).   (a) Experimental scheme and (b) the level structure. The ultranarrow $^3P_0$-$^1S_0$ transition indicated by the red arrow is the laser transition. The repumping lasers are indicated by the blue arrows, spontaneous decay into the ^3P states by the gray arrow, and the Raman sideband cooling lasers by the black arrows. The n quantum numbers indicate the vibrational levels of $^3P_0$ and $^3P_2$.

with the same phase to the specific cavity mode. Repumping lasers resonantly drive transitions from the $^1S_0$ to the $^3P_1$ transition and from there to the $^3S_1$ state. The repump lasers optically pump the atoms into the $^3P_0$ and $^3P_2$ states. A Raman transition from $^3P_0$ to $^3P_2$ via $^3S_1$ is used to implement sideband cooling to the vibrational ground state and to optically pump all atoms to the $^3P_0$ state, thus providing inversion for the laser transition. We summarize all these repump steps by an effective repump rate w. We have estimated ac Stark shifts incurred by these repump and cooling lasers and we find that for repump rates up to around $w \sim 10^3$ s^{-1} the ac Stark shift induced fluctuations of the atomic transition frequency is negligible at the mHz level, if the laser intensity is stabilized to 1%. With the continuous cooling in place we are justified in assuming that the atoms are in the ground state of the lattice.

A discussion of the relative and absolute magnitude of the different rate constants is in order. For ^{87}Sr, the linewidth of the doubly forbidden intercombination line is $\gamma \approx 0.01$ s^{-1}. The inhomogeneous lifetime in the most recent generation of lattice clock experiments has been pushed to $T_2 \sim 1$ s [21]. The effective repump rate w can be widely tuned from 0 to values of order 10^4 s^{-1} and beyond. The cavity parameters enter the physics through the cooperativity parameter $C = \Omega^2/(\kappa\gamma)$, which is independent of the length of the cavity. For definiteness we consider a cavity with an effective mode volume $V_{\text{eff}} = (1 \text{ mm})\pi(50 \text{ } \mu\text{m})^2$. Because of the extremely weak dipole matrix element of the inter combination transition of order $10^{-5} e a_0$, with e the electron charge and a_0 the Bohr radius, this leads to a vacuum Rabi frequency of order $\Omega \sim 37$ s^{-1}. In this setup the cavity decay rate is by far the largest time scale. For a finesse of $\mathcal{F} = 10^6$ and the above cavity parameters the cavity linewidth is $\kappa = 9.4 \times 10^5$ s^{-1}, and thus $C \approx 0.15$.

The coupled atom-cavity system can be described by the Hamiltonian

$$\hat{H} = \frac{\hbar\omega_a}{2}\sum_{j=1}^{N}\hat{\sigma}_j^z + \hbar\omega_c\hat{a}^\dagger\hat{a} + \frac{\hbar\Omega}{2}\sum_{j=1}^{N}(\hat{a}^\dagger\hat{\sigma}_j^- + \text{H.c.}). \quad (1)$$

In this formula, $\Omega = \hbar^{-1}\sqrt{\hbar\omega_c/(2\epsilon_0 V_{\text{eff}})}$, ϵ_0 the vacuum

permittivity, and $\hat{a}$ and $\hat{a}^\dagger$ are bosonic annihilation and creation operators for photons in the laser mode. We have introduced Pauli matrices $\hat{\sigma}_j^z = |e_j\rangle\langle e_j| - |g_j\rangle\langle g_j|$ and $\hat{\sigma}_j^- = (\hat{\sigma}_j^+)^\dagger = |g_j\rangle\langle e_j|$ for the jth atom.

We take the various decay processes into account by means of the usual Born-Markov master equation for the reduced atom-cavity density matrix $\hat{\rho}$, $d\hat{\rho}/dt = (i\hbar)^{-1} \times [\hat{H}, \hat{\rho}] + \mathcal{L}[\hat{\rho}]$, with the Liouvillian $\mathcal{L}[\hat{\rho}] = \mathcal{L}_{\text{cavity}}[\hat{\rho}] + \mathcal{L}_{\text{spont.}}[\hat{\rho}] + \mathcal{L}_{\text{inhom.}}[\hat{\rho}] + \mathcal{L}_{\text{repump}}[\hat{\rho}]$. The Liouvillian for cavity decay is $\mathcal{L}_{\text{cavity}}[\hat{\rho}] = -\kappa/2(\hat{a}^\dagger\hat{a}\hat{\rho} + \hat{\rho}\hat{a}^\dagger\hat{a} - 2\hat{a}\hat{\rho}\hat{a}^\dagger)$, the spontaneous decay of the atoms is described by $\mathcal{L}_{\text{spont.}}[\hat{\rho}] = -\gamma/2\sum_{j=1}^{N}(\hat{\sigma}_j^+\hat{\sigma}_j^-\hat{\rho} + \hat{\rho}\hat{\sigma}_j^+\hat{\sigma}_j^- - 2\hat{\sigma}_j^-\hat{\rho}\hat{\sigma}_j^+)$, and the Liouvillian for the inhomogeneous lifetime is $\mathcal{L}_{\text{inhom.}}[\hat{\rho}] = 1/(2T_2)\sum_{j=1}^{N}(\hat{\sigma}_j^z\hat{\rho}\hat{\sigma}_j^z - \hat{\rho})$. The Liouvillian for the repumping is identical to that for the spontaneous decay with the replacements $\gamma \rightarrow w$, $\hat{\sigma}_j^- \rightarrow \hat{\sigma}_j^+$, and $\hat{\sigma}_j^+ \rightarrow \sigma_j^-$.

An important aspect of this system that is born out by the master equation is that the coupling of the atoms to the light field is completely collective. The emission of photons into the cavity acts to correlate the atoms with each other similar to the case of ideal small sample superradiance [22,23], leading to the buildup of a collective dipole. The locking of the phases of the dipoles of different atoms gives rise to a macroscopic dipole that radiates more strongly than independent atoms. The macroscopic dipole is also more robust against noise from decay processes and repumping, leading to a reduced linewidth.

We have verified that the system settles to steady state much faster than the anticipated total operation time, provided that the repump rate is not too close to the laser threshold derived below. For the representative example parameters used below, the relaxation oscillations decay after a time $\lesssim 1$ s while the total operation time will typically be >1 min. We therefore focus entirely on the steady state behavior in this Letter. To find the steady state we introduce a cumulant expansion to second order for the expectation values of system observables [24]. We denote raw expectation values by $\langle\ldots\rangle$ and cumulant expectation values by $\langle\ldots\rangle_c$. In our model all expectation values, cumulant or raw, are completely symmetric with respect to exchange of particles, for instance $\langle\hat{\sigma}_j^z\rangle_c = \langle\hat{\sigma}_1^z\rangle_c$ and $\langle\hat{a}^\dagger\hat{\sigma}_j^-\rangle_c = \langle\hat{a}^\dagger\hat{\sigma}_1^-\rangle_c$ for all j, $\langle\hat{\sigma}_i^+\hat{\sigma}_j^-\rangle_c = \langle\hat{\sigma}_1^+\hat{\sigma}_2^-\rangle_c$ for all $i \neq j$, etc. In this formalism where we explicitly keep track of higher order correlations, the total phase invariance of the system is not broken and we have $\langle\hat{a}\rangle = \langle\hat{a}^\dagger\rangle = \langle\hat{\sigma}_1^\pm\rangle = 0$. The only nonzero cumulant of the first order is then the inversion $\langle\hat{\sigma}_1^z\rangle_c = \langle\hat{\sigma}_1^z\rangle$. This cumulant couples to second order cumulants through the atom-field coupling,

$$\frac{d\langle\hat{\sigma}_1^z\rangle_c}{dt} = -(w + \gamma)(\langle\hat{\sigma}_1^z\rangle_c - d_0)$$
$$+ i\Omega(\langle\hat{a}^\dagger\hat{\sigma}_1^-\rangle_c - \langle\hat{a}\hat{\sigma}_1^+\rangle_c), \quad (2)$$

where $d_0 = (w - \gamma)/(w + \gamma)$. The atom-field coherence $\langle\hat{a}^\dagger\hat{\sigma}_1^-\rangle_c$ evolves according to

$$\frac{d\langle\hat{a}^{\dagger}\hat{\sigma}_1^-\rangle_c}{dt} = -\left(\frac{w+\gamma}{2} + \frac{1}{T_2} + \frac{\kappa}{2} - i\delta\right)\langle\hat{a}^{\dagger}\hat{\sigma}_1^-\rangle_c$$

$$+ \frac{i\Omega}{2}\left[\langle\hat{a}^{\dagger}\hat{a}\hat{\sigma}_1^z\rangle_c + \langle\hat{a}^{\dagger}\hat{a}\rangle_c\langle\hat{\sigma}_1^z\rangle_c\right.$$

$$\left. + \frac{\langle\hat{\sigma}_1^z\rangle_c + 1}{2} + (N-1)\langle\hat{\sigma}_1^+\hat{\sigma}_2^-\rangle_c\right], \quad (3)$$

where $\delta = \omega_c - \omega_a$. We have estimated the third order cumulant in square brackets to be much smaller than the other terms and therefore neglect it. The second and third terms represent the exchange of energy between cavity field and a single atom. They are noncollective in nature. The last term describes the coupling of the atom-field coherence to the collective spin-spin correlations which locks the relative phase between atoms and field to the phase of the macroscopic atomic dipole.

The spin-spin correlations evolve according to

$$\frac{d\langle\hat{\sigma}_1^+\hat{\sigma}_2^-\rangle_c}{dt} = -(w + \gamma + 2T_2^{-1})\langle\hat{\sigma}_1^+\hat{\sigma}_2^-\rangle_c$$

$$+ \frac{\Omega\langle\hat{\sigma}_1^z\rangle_c}{2i}[\langle\hat{a}^{\dagger}\hat{\sigma}_1^-\rangle_c - \langle\hat{\sigma}_1^+\hat{a}\rangle_c], \quad (4)$$

where we have dropped the small third order cumulants of the type $\langle\hat{a}^{\dagger}\hat{\sigma}_1^z\hat{\sigma}_2^-\rangle_c$. To close the set of equations we also need the equation for the mean photon number,

$$\frac{d\langle\hat{a}^{\dagger}\hat{a}\rangle_c}{dt} = -\kappa\langle\hat{a}^{\dagger}\hat{a}\rangle_c + \frac{N\Omega}{2i}(\langle\hat{a}^{\dagger}\hat{\sigma}_1^-\rangle_c - \langle\hat{\sigma}_1^+\hat{a}\rangle_c). \quad (5)$$

We consider the steady state of this system for the case $\delta = 0$ by setting the time derivatives in Eqns. (2)–(5) to zero. The resulting algebraic equations can be solved exactly. Simple approximate results can be obtained in certain limits on which we will focus in our discussion to explain the underlying physics. The numerical results reproduced in the figures are based on the exact solutions and agree well with the approximate treatment.

Our first goal is to understand the role of the collective effects. Neglecting all decay constants but κ in Eq. (3), keeping only the collective term proportional to $\langle\hat{\sigma}_1^+\hat{\sigma}_2^-\rangle_c$, and approximating $N - 1 \approx N$ we find for the atom-field coherence in steady state $\langle\hat{a}^{\dagger}\hat{\sigma}_1^-\rangle = iN\Omega\kappa^{-1}\langle\hat{\sigma}_1^+\hat{\sigma}_2^-\rangle_c$. Inserting this result into the steady state equation for the inversion determines the saturated inversion and plugging that and $\langle\hat{a}^{\dagger}\hat{\sigma}_1^-\rangle$ into the steady state equation for the spin-spin correlations yields the central equation

$$0 = \langle\hat{\sigma}_1^+\hat{\sigma}_2^-\rangle_c\left(-\Gamma + d_0 N\gamma C - 2\frac{N^2\gamma^2 C^2}{w+\gamma}\langle\hat{\sigma}_1^+\hat{\sigma}_2^-\rangle_c\right), \quad (6)$$

where $\Gamma = \gamma + w + 2/T_2$ is the total relaxation rate of the atomic dipole. The solution for $\langle\hat{\sigma}_1^+\hat{\sigma}_2^-\rangle_c$ corresponding to the term in parenthesis is the physically stable solution. The laser threshold is the pump rate at which the gain $d_0 N\gamma C$ overcomes the losses Γ. In the limit $\Gamma/(\gamma NC) \rightarrow 0$ this condition turns into $w > \gamma$. At threshold the pump overcomes the atomic losses which is in contrast to con-

ventional lasers where threshold is obtained when the pump overcomes the cavity losses.

At threshold the spin-spin correlations change sign signifying the onset of collective behavior. Interestingly, the spin-spin correlations change sign again at a larger pump rate above which the atoms return to normal noncollective emission. This upper threshold comes about because d_0 eventually saturates at 1 while the pump induced noise grows with w. Setting $d_0 = 1$ and neglecting all atomic noise sources other than w we find for the maximum pump rate

$$w_{\text{max}} = NC\gamma. \quad (7)$$

Above this threshold the pump noise destroys the coherences between different spins faster than the collective interaction induced by the light field can establish them.

A minimum number of particles is necessary to have collective behavior. Below this critical number, $\langle\hat{\sigma}_1^+\hat{\sigma}_2^-\rangle_c$ is never positive and hence the spin-spin correlations are never collective. The critical particle number can be estimated from Eq. (6). Assuming $T_2\gamma \ll 1$, we find $N_{\text{crit}} = 2/(C\gamma T_2)$. Physically, this equation means that there must be enough particles for the system to be in the collective strong coupling regime.

The collective versus noncollective behavior is nicely illustrated by the outcoupled laser power as a function of w and N (Fig. 2). Above threshold the outcoupled power rapidly increases until the collective dipole is destroyed at the second threshold w_{max}.

Equation (6) also allows us to determine the maximum spin-spin correlation of $\langle\hat{\sigma}_1^+\hat{\sigma}_2^-\rangle_c = 1/8$ which is obtained for the pump rate $w_{\text{opt}} = NC\gamma/2$. At this pump rate the laser power reaches its maximum of

$$P_{\text{max}} = \hbar\omega_a N^2 C\gamma/8. \quad (8)$$

The scaling of that power with the square of the number of atoms underlines the collective nature of the emission.
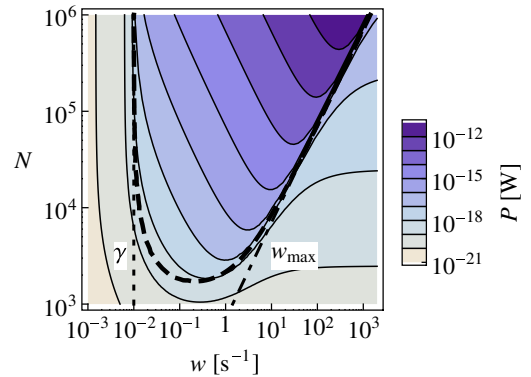

FIG. 2 (color online). Power as a function of pump rate w and atom number N. The rapid buildup of power above threshold $w \sim \gamma$ can be seen as well as the decrease of emitted power for too strong a pump. The dashed line shows the boundary of the region of collective emission as determined by the zero of the term in parenthesis in Eq. (6). Here, $\gamma = 0.01$ s^{-1}, $1/T_2 = 1$ s^{-1}, $\Omega = 37$ s^{-1}, and $\kappa = 9.4 \times 10^5$ s^{-1}.
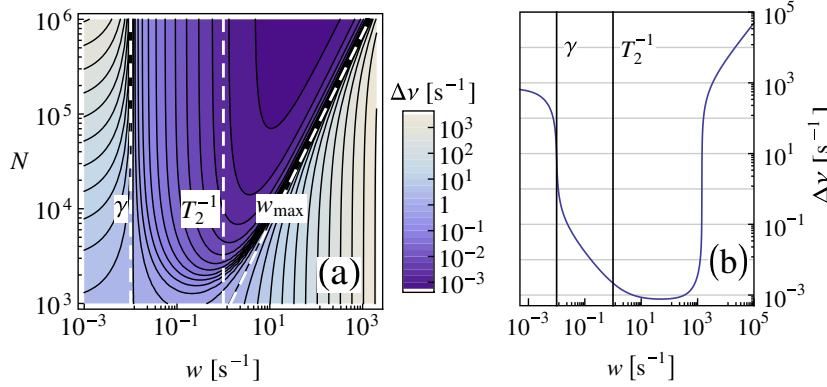
FIG. 3 (color online). (a) Linewidth vs w and N. The white dashed lines indicate (from left to right) the spontaneous decay rate γ, the inhomogeneous relaxation rate $1/T_2$, and the maximum pump rate w_{max} [Eq. (7)]. Parameters as in Fig. 2. (b) is a cut through (a) for $N = 10^6$ atoms.

Remarkably, this power is only a factor of 2 smaller than the power expected for perfect superradiant emission from the maximally collective Dicke state at zero inversion. Figure 2 shows that an outcoupled power of order 10^{-12} W is possible with 10^6 atoms.

From the perspective of potential applications the most striking feature of this laser is its ultranarrow linewidth. To find the spectrum we use the quantum regression theorem to find the equations of motion for the two time correlation function of the light field $\langle \hat{a}^\dagger(t)\hat{a}(0)\rangle$. This correlation function is coupled to the atom-field correlation function $\langle \hat{\sigma}^+(t)\hat{a}(0)\rangle$. Factorizing $\langle \hat{\sigma}^z(t)\hat{a}^\dagger(t)\hat{a}(0)\rangle \approx \langle \hat{\sigma}^z(t)\rangle \times \langle \hat{a}^\dagger(t)\hat{a}(0)\rangle$ we arrive at the closed set of equations

$$\frac{d}{dt}\begin{bmatrix} \langle \hat{a}^\dagger(t)\hat{a}(0)\rangle \\ \langle \hat{\sigma}^+(t)\hat{a}(0)\rangle \end{bmatrix} = \begin{bmatrix} -\kappa/2 & \frac{iN\Omega}{2} \\ -\frac{i\Omega\langle\hat{\sigma}^z\rangle_c}{2} & -\Gamma/2 \end{bmatrix}\begin{bmatrix} \langle \hat{a}^\dagger(t)\hat{a}(0)\rangle \\ \langle \hat{\sigma}^+(t)\hat{a}(0)\rangle \end{bmatrix}. \tag{9}$$

The initial conditions are the steady state solutions discussed above. Laplace transforming the solutions yields the spectrum which is Lorentzian with linewidth $\Delta\nu$ for $\omega_a = \omega_c$.

For our example parameters $\Delta\nu$ is shown in Fig. 3. The leftmost dashed line in that figure is γ corresponding to the threshold for collective behavior. When the pump strength w passes through that threshold the linewidth gets rapidly smaller with increasing w. When w reaches $1/T_2$, indicated by the second dashed line, essentially all atoms are phase locked together. From that point on the pump noise due to w grows in proportion to the size of the collective spin vector. Therefore the linewidth is approximately constant. Making similar approximations as in the steady state calculations we obtain the estimate for the minimum laser linewidth, $\Delta\nu = C\gamma$. That estimate agrees well with our numerical results. It is important to note that the parameters for achieving the maximum outcoupled power Eq. (8) and the minimum linewidth are compatible with each other. For our example parameters the estimate yields a linewidth smaller than the homogeneous linewidth of the atomic clock transition. We have studied the dependence of the laser frequency on the atom-cavity detuning and we find that this narrow linewidth can be observed if the cavity resonance frequency is stable at the 1 kHz level which is relatively easy to achieve experimentally. When w in-

creases beyond w_{max}, indicated by the third dashed line, the collective dipole is destroyed and the linewidth increases rapidly until it is eventually given by w.

Future research is targeted at fully understanding the recoil effects and the detailed nature of the joint atomic and field state.

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