Laser spectroscopy and quantum optics: application towards future atomic clocks

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Abstract

From atomic fountains to ion traps, recent techniques in trapping and isolating atoms and ions from environmental perturbations has provided access to their extremely narrow natural linewidths. Combined with techniques of laser frequency and amplitude stabilization, future atomic clocks based on all optical interrogation are becoming feasible. This paper will discuss the requirements for such frequency/time standards and review some of the more promising clock proposals as well as their fundamental limitations.

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I. INTRODUCTION

Future atomic frequency standards will no doubt benefit from the overlapping fields of laser spectroscopy and quantum optics. With the advent of tunable dye lasers in the early 70's optical spectroscopy made giant steps forward in sensitivity and resolution that has brought the two fields closer together than ever. Quantum optics serves not only to define the fundamental limitations on the precision with which an atom can be probed but also provides the spectroscopist with a whole new array of tricks with which they can use light to study and manipulate matter. In return, the tools and precision gained in laser spectroscopy has enabled many of the former gedanken experiments of quantum optics to become realized in the laboratory to test the very foundations of quantum mechanics [1].

Frequency standards require high stability and accuracy. Periodic systems are therefore sought to measure time which have a high Q [2] and mean resonant frequency that does not drift over time. The response of an atom to an electromagnetic field meets both criteria. The narrow resonance of atomic transitions provides Q factors up to 10^9 ensuring high

FIGURES

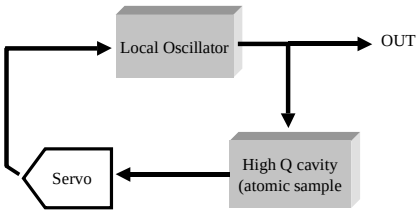

FIG. 1. Schematic for feedback in atomic frequency standards.

stability, while the presumed constancy of quantum mechanical laws ensures a high accuracy.

Atomic frequency standards are usually composed of a local oscillator (LO), a high Q resonator, and a servo as shown in Fig. 1. The local oscillator provides energy for the resonator (atoms) by driving them at their resonant frequency. The deviations of the LO from the atomic resonance are detected and sent to a servo which corrects the frequency of the LO, thus ensuring its medium and long term stability. As the servo system cannot respond instantaneously, the short term stability of the local oscillator must be at least as good as that of the atomic resonance. Atomic transition frequencies are used from the radio and microwave regimes ($\approx 10^6 - 10^9$) up to the optical regime ($\approx 10^{14}$). It is only up to the microwave regime, however, that frequencies can be directly counted with present day electronics. Because of this, the current definition of the second is based on the ground state hyperfine transition in the cesium atom at 9.192 631 770 GHz [3]. Currently, optical frequencies can only be measured indirectly via a long chain of nonlinear optical and electronic mixers. Such frequency chains are difficult to build and are usually tailored for a specific transition. Because of this and the loss in accuracy within the chain, radio and microwave frequency standards are usually referred to as time standards (atomic clocks) while optical frequency standards are sometimes referred to as wavelength standards as their wavelength can accurately define length scales using a wavemeter and the presently defined definition for the speed of light (299 792 458 m/sec) through the simple relation $\lambda = c/\nu$. Currently the most accurately measured "optical" frequency standard is the methane stabilized He-Ne laser measured at 88 376 182 599 937 (23) Hz (3.39 μm) relative to the cesium atomic clock. This frequency can then be linked to other optical transitions to measure their absolute frequency.

There are countless proposals and demonstrations for improvements in both time and length standards. In this paper we will review some of the more promising and interesting proposals,

discussing their ultimate potential and limitations. In section 2 we will review definitions of stability and accuracy of frequency sources. Section 3 will address factors which limit the stability and accuracy of high resolution spectroscopy due to perturbations of our quantum time keepers. In section 4 we will survey different proposals and current experiments which seek to improve current frequency standards by eliminating (or minimizing) the perturbations outlined in section 3.

II. CHARACTERIZATION OF TIME AND FREQUENCY: STABILITY VS. ACCURACY

As mentioned previously, the figures of merit for an atomic clock are its stability and accuracy. Several techniques have been developed to quantify and measure these quantities. We will introduce the most often quoted and important measures. The frequency *stability* of an oscillator is usually expressed in terms of the two-sample Allan variance [9] given by:

$$\sigma_y^2(\tau) = \frac{1}{2(K-1)} \sum_{k=1}^{K-1} \frac{(\langle\omega_k\rangle_\tau - \langle\omega_{k+1}\rangle_\tau)^2}{\omega_o^2}, \tag{1}$$

where ω_o is the angular frequency and $\langle\omega_k\rangle_\tau$ is the average frequency for the kth measurement over a time interval τ. The quantity $\sigma_y(\tau)$ is a measure of the fluctuation between successive measurements of the frequency for a given averaging time τ. Figure 2 shows an example of the Allan variance plotted for a Hydrogen maser clock and a power stabilized cryogenic sapphire oscillator developed as a local oscillator for driving future atomic clocks [6]. The plot shows the characteristic $1/\sqrt{\tau}$ roll-off of the Allan variance for "white" noise sources; that is, for sources of frequency noise that exhibit a flat spectrum in the frequency domain such as shot-noise limited performance.

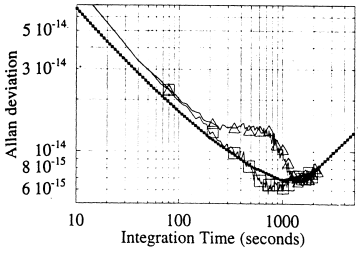

FIG. 2. Allan variance comparing stability of H-maser to stabilized sapphire oscillator [6].

As such white-noise is considered completely random, longer integration times naturally average out the frequency fluctuations so that the variance improves as a function of τ. We can also see from Fig. 2 that at some τ, the variance changes slope. At this point longer averaging times produce

greater instabilities. This is usually attributed to a long term drift of the measured resonance. Situations may also arise when noise reductions won by longer averaging times are just annulled by changes in the mean value (this is common in most high-performance frequency sources). This domain is referred to as "flicker of frequency" or flicker-noise and shows a τ^0 dependance. Although not completely understood, the flicker of frequency domain is usually attributed to time-dependent changes of systematic offsets (We will discuss the effects of systematic offsets shortly). The Allan variance is an extremely powerful measure of an oscillators stability. In the white-noise domain, the coefficient of the $\tau^{-\frac{1}{2}}$ slope is related to the linewidth of the resonance. For an ideal frequency-servo controlled system, the full-width at half maximum of this linewidth is related to the Allan variance scaled to 1 second by [9]:

$$\delta\nu_{FWHM} = 2\pi^2(\nu\sigma_1)^2 \tag{2}$$

where $\sigma_y(\tau) = \sigma_1/\sqrt{(\tau)}$. The coefficient σ_1 is a commonly quoted measure of a systems stability up to the time when the slope rolls off.

A frequency source characterized by a very good short term stability may not necessarily be accurate. The *accuracy* of a frequency source is related to the *repeatability* of the measured mean frequency for several independent systems. If the mean frequency is shifted by *systematic* offsets, it may show excellent stability, but an inaccurate frequency. The determination of an atomic frequency standard's accuracy is therefore generally more difficult. The source must first exhibit good stability over a sufficiently long integration time such that the uncertainty in its stability is minimized and a measured value for the mean frequency can be obtained. Then the arduous task of labeling and characterizing all possible systematic offsets is performed. This "error budget" must include any effects which could possibly shift the resonance. They can be characterized by theoretically modelling the effects and subsequently varying the relevant parameter(s) which produces them and extrapolating the effect at zero perturbations. (We will list some of the more common effects and their characterization in an upcoming section). When the uncertainty due to all systematic effects is taken into account, a final expression for the oscillators relative uncertainty can be expressed by $\delta\nu/\nu_o$. This is demonstrated in the comparison of the hydrogen maser and cesium atomic standards. Although the hydrogen maser shows superior stability for short and medium time scales, its overall accuracy is degraded by systematic uncertainties due to frequency-shifting collisions with the walls of its teflon coated cell. Thus the cesium standard, which exhibits higher accuracy is used. The most accurate primary standard in the United States is NIST-7, located in Boulder Colorado, with a recorded uncertainty of less than 1 part in 10^{14}.

III. SPECTROSCOPY OF REAL ATOMIC SYSTEMS

Although individual atoms provides an ideal frequency reference, in practice they become strongly perturbed by each other, their environment, and their relative motion. Although we cannot present an exhaustive list here, we will discuss some of the more significant linewidth broadening and shifting effects. Perturbations to the atom which cause a broadening of the line effects the stability of the atomic frequency standard. The most obvious and prominent effect is Doppler broadening due to the average velocity distribution of the atoms, $\sqrt{\langle \nu^2 \rangle}$. This results in a Doppler broadened transition due to thermal excitation at a temperature T of $\delta\nu \approx \nu_o/c\sqrt{8kTln2/m}$ where k is Boltzman's constant and m is the mass of the atom. Another prominent effect is broadening due to the finite interaction time of the light with the atom, or "transit-time" broadening. Because of the finite interaction time, the atom "sees" not a single frequency (which is defined from t= $+\infty$ to$-\infty$) but a quasi-monochromatic frequency given by the Fourier transform of the field over the interaction time. Assuming a Gaussian beam profile for the laser beam waist, one finds a linewidth broadening contribution of $\delta\nu \approx 0.4\tau^{-1}$ where τ is the interaction time [10]. A final important broadening mechanism to mention is collisional broadening. Elastic and inelastic collisions of the atom with itself or other background species can randomly perturb the phase of the atom. This effect depends in great detail on the particular atomic system used and the experimental setup, but generally is proportional to the atomic density N (for collisions with partners) and the mean velocity v of the atoms. Collisions can also induce a *shift* of the resonant frequency and therefore are also a cause of systematic effects which limit the accuracy of an atomic frequency standard. In general most of the broadening mechanisms not only affect the stability of the standard (due to symmetric broadening of the transition) but also their accuracy due to higher order effects which cause the broadening to be asymmetric.

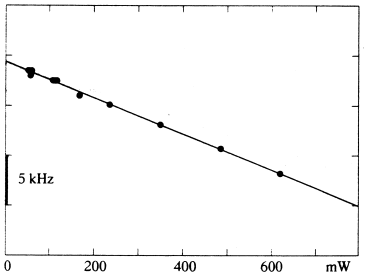

FIG. 3. Frequency shift of the two-photon line versus incident light power [11].

There are numerous other systematic effects which can act to shift the mean resonance of an atomic transition. We will name only a few here that will come into the discussion in the next section. The second order Doppler effect (often times referred to as time dilation effects in the literature) is proportional to the averaged velocity squared of the atomic system and is problematic for standards based on atomic beams. It's origin can be seen by considering the relativistic-energy conservation demands when a two-level atom absorbs a photon of energy $\hbar\omega_{ik} = E_k - E_i$ [10]:

$$\hbar\omega_{ik} = \sqrt{p_k^2c^2 + (Mc^2 + e_k)^2} - \sqrt{p_i^2c^2 + (Mc^2 + E_i)^2} \tag{3}$$

where M is the mass of the particle, c is the speed of light, $p_{i(k)}$ and $E_{i(k)}$ is the momentum and energy of the particle before (after) the absorption of the photon. Expanding this expression into a power series of $(1/c^n)$ and solving for the resonant absorbtion frequency one finds

$$\omega_{ik} = \omega_o + \mathbf{k}\cdot\mathbf{v_i} - \omega_o\frac{v_i^2}{2c^2} + \frac{\hbar\omega_o^2}{2Mc^2} + ... \tag{4}$$

In this expression, ω_o is the resonant frequency of the atom in its rest frame. The second term is the linear Doppler shift due to the motion of the atom in the direction of the photon where the momentum of the photon is $p = \hbar k$ and $\mathbf{v_i}$ is the velocity of the atom before the absorption. The third term expresses the second order Doppler shift. As this term is quadratic in the atoms velocity it is independent of the atoms direction and cannot be eliminated or canceled in standard "Doppler -free" geometries. This term therefore represents a shift of the atomic resonance. (The fourth term shown express the recoil energy of the atom due to the absorption of the photon.) The linear ($\Delta\nu \propto E$) and quadratic ($\Delta\nu \propto E^2$) Stark effect resulting from the perturbation of the atomic Hamiltonian by an electric field manifests itself in both the microwave and optical regimes in a variety of different ways. One example is in the optical regime, where the AC stark shift is sometimes refered to as the light shift. To measure the absolute frequency of an atomic transition, the shift of the resonant frequency due to the probing field must be taken into account. Figure 3 shows the measurement of this light shift in an experiment measuring the $5S_{1/2}$ to $5D_{3/2}$ two-photon transition in ^{85}Rb [11]. The linearity of the measured fit with varying power allowed the spectroscopist to extrapolate the uncertainty in the absolute frequency at zero power to less than 1kHz, and hence this systematic effect was not a limiting factor in the measurement [4]. The list of systematic errors goes on and on depending on the creativity of the researcher thinking up any and all possibilities. In the next section we will look at current atomic frequency standards and some promising future proposals. Each system has its own limiting factors effecting its stability and accuracy.

IV. CURRENT AND FUTURE ATOMIC FREQUENCY STANDARDS

Many of the current experiments in frequency metrology are based on variations of present frequency standards. We will therefore begin by briefly reviewing the problems overcome by the present day operational atomic clocks and their current limitations.

A. Optically pumped cesium frequency standards

Current time standards are based on RF spectroscopy of atomic hyperfine splittings, in particular cesium. The largest perturbation to an atoms linewidth is linear Doppler broadening. An atomic beam provides a well collimated beam of atoms whose velocity distribution *perpendicular* to their direction is very small. By probing the atomic hyperfine transition at 90 degree's to the beam direction, the first order Doppler contribution can be greatly reduced. The next instability arises from the reduced interaction time of the RF local oscillator with the quickly moving atoms. Norman Ramsey's ingenious idea of separated fields [12] greatly reduced this broadening and is a standard technique used today in most atomic frequency standards, including many high resolution optical frequency measurements [13]. The experiment consists of completely inverting the population in a 2-level system by the application of two $\pi/2$ pulses separated in time. We assume the initial state of the atomic beam is prepared in the ground state such that the total state of the system can be written as a product state in the form $|\Psi\rangle = |0\rangle \otimes |0\rangle \otimes ....$ In the first interaction region an RF $\pi/2$ pulse is applied such that the system is prepared in an equal distribution of its ground and excited state. The single atomic wavefunction is now expressed as

$$|\Psi\rangle_{atom} \rightarrow \frac{1}{\sqrt{2}}(|0\rangle + i|1\rangle) \tag{5}$$

The atom now posses a nonzero dipole moment $\langle d(t)\rangle$ that precesses in the field-free region at its eigenfrequency ω_o. When it enters the second Ramsey zone at time T later, the atomic wavefunction will have accumulated a phase angle $\Delta\varphi = \omega_o T$. During this same time, the phase of the LO will have changed by ωT. The relative phase between the dipole and field is therefore $(\omega_o - \omega)T$. The interaction between the field and the dipole in the second zone will depend upon this relative phase. The probability that the atom can be found in state $|1\rangle$ will be given by

$$P = (1 + cos([\omega_o - \omega]T)/2 \tag{6}$$

If the frequency of the probing field is equal to that of the atomic resonance, the system will simply have received a π pulse and be completely inverted. Small frequency differences will result

in large phase differences given sufficiently long T, resulting in incomplete inversion. Monitoring the population of the atomic system after the second interaction zone allows one to determine the detuning of the LO and adjust it accordingly.

Current cesium frequency standards use a combination of RF and optical spectroscopy based on Ramsey's technique of separated zones [14]. See Fig. 4.

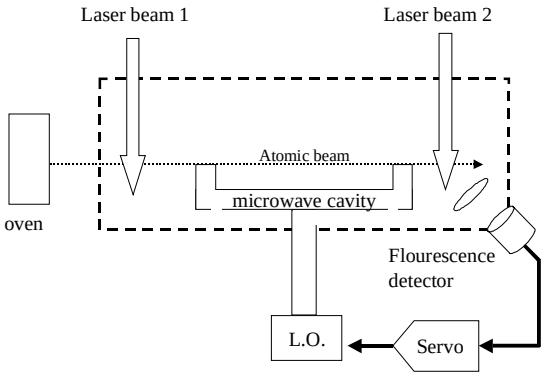

FIG. 4. Schematic of an optically pumped cesium standard utilizing a Ramsey zone geometry.

An atomic beam of cesium atoms is initially prepared in the ground state hyperfine level F=3 by optically pumping the atoms with the first laser beam . A high Q local oscillator provides the Ramsey $\pi/2$ pulses and the final state of the system is optically detected by driving the F=3 ground state transition to an upper atomic state and collecting the radiated fluorescence. If maximum fluorescence is not detected, the atomic system has not been completely inverted and the local oscillator is adjusted to keep it in phase with the atomic transition. Of course there are many complicated details involved in determining the final performance of this system. The *stability* of this frequency standard is ultimately limited by the length of the atomic beam (ie the time between Ramsey pulses) as this determines its minimum linewidth. There are many practical difficulties in extending the length of the atomic beam arbitrarily such as non-uniformities in axial magnetic fields leading to end-to-end cavity phase shifts [15]. Current limitations on stability in these systems is about $10^{-13}\tau^{-1/2}$. Atomic frequency standards based on thermal atomic beams are ultimately limited in *accuracy* by second order Doppler broadening at the 10^{-14} level of uncertainty due to the uncertainty in the atomic velocity distribution [15]. In the following sections we will look at future atomic clocks that will take us beyond these levels of instability and uncertainty.

B. Zacharias's fountain

In the early 1950's J.R. Zacharias proposed the use of gravity to slow down atomic beams by directing them upwards [16]. The slow atoms in the tail of the thermal Boltzman distribution were expected to follow a ballistic trajectory, creating a "fountain" of atoms. This would have the benefit of longer interaction times when used with a Ramsey separated zone geometry for the rising and falling atoms, as well as slower velocities. Although he built an apparatus 9 meters long, the experiment was not successful [17]. Since then new techniques in optically cooling and manipulating atoms have been developed [18] that have allowed such fountains to be realized.

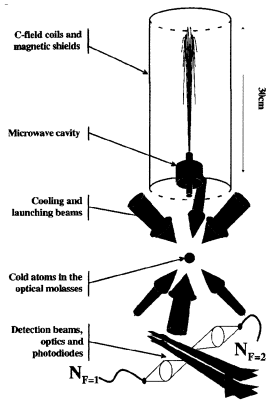

FIG. 5. Atomic fountain schematic [19].

Although each design varies slightly, the basic components involved in an atomic fountain are shown in Fig. 5. Initially, a collimated atomic beam is sent in beneath the fountain apparatus. A laser beam is directed into the atomic beam and tuned to the "red" side of the atomic resonance such that the atomic transitions are Doppler shifted into resonance. The atomic beam is then slowed down and eventually stopped by the laser due to the radiation pressure. The atoms are halted in the region of an optical molasses and/or a magneto-optical trap (MOT) [18]. Large atomic densities of relatively cold ($\approx \mu K$) atoms can be built up in this way. The atoms are then launched upward by another laser beam. The launch velocity is on the order of a few meters/sec. In the present example, the atoms reach an apogee 0.3 meters above the trap [19]. Ideally the atoms could be excited optically as well as with microwaves [5]. In this case a microwave cavity applies a $\pi/2$ pulse to the atoms on their way up and down, corresponding to a 0.5 sec. interaction time. As the atoms fall back down, two laser beams are used to measure the final state populations from the induced fluorescence. The measured Ramsey fringes versus detuning of the microwave oscillator are shown in Fig. 6. The central fringe in the figure shows a width of only 1.2 Hz FWHM. The

local oscillator is stabilized to the peak of this narrow fringe as outlined in the previous section. The envelope modulating the Ramsey fringes is due to dephasing between the microwave field and the oscillating atomic dipole. This is often times refered to as the Rabi envelope as it is caused by transit time broadening of the resonance due to the finite interaction time of the atom with the field in the excitation regions.

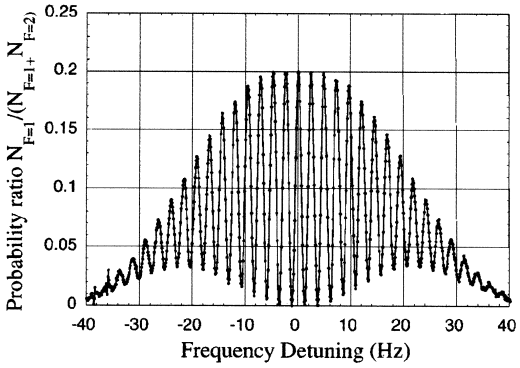

FIG. 6. Ramsey fringes observed from atoms detected in the F=1 and F=2 hyperfine levels of ^{87}Rb [19].

Atomic fountains all but eliminated the effects of second order Doppler shifts that limited thermal beams. They also offer greater stability due to the greatly increased interaction times. As the frequency standards increase in stability and accuracy, the role of quantum mechanics becomes more vital in determining their ultimate limitations. It has just recently been reported that a cesium atomic fountain has reached the quantum mechanically predicted stability limit [20]. This limit is imposed by the statistical fluctuations in the measurements of the state populations during each cycle of the fountain. That is to say, the measurement of an atom's internal state after it has traversed through the atomic fountain can only be described statistically, and cannot be predicted for any given measurement with complete certainty. This is commonly referred to as the shot noise limit in optical and electronic systems. As a fundamental source of noise for population measurements with a fixed number of atoms or ions, this effect has been named *quantum projection noise* as it can be interpreted to arise from the random projection of the atomic state vector into one of the eigenstates of the measurement process [21]. When the measurement of the atomic states is limited in this way, the frequency stability is predicted to be given by [22]

$$\sigma_y(\tau) = \frac{1}{\omega_o \sqrt{N T_R}} \tau^{-1/2}. \tag{7}$$

This expression assumes a Ramsey separated field geometry with time T_R separating the pulses, ω_o is the angular frequency of the atomic transition, and N is the number of atoms. By varying

the number of atoms in the fountain, the predicted $N^{-1/2}$ law was confirmed at the 6% level. The measured short-term frequency stability for this system was measured for N=6 $\times$ 10^5 to be $4 \times 10^{-14}\tau^{-1/2}$. This is currently the best reported frequency stability for any primary frequency standard.

Although the stability of the cesium fountain reported was a record, it is limited in its accuracy primarily due to frequency shift induced cold collisions and the blackbody radiation induced AC Stark shifts. As the collisional shift is proportional to the atomic density, this results in a trade-off between using large atomic densities to increase the stability on short time scales according to Eq.7 and low atomic densities to minimize collisional shifts and improve long term accuracy. It has recently been demonstrated that an atomic fountain utilizing ^{87}Rb, which is predicted to have a much smaller cross-section for collisions, exhibits lower collisional shifts and may lead to a relative accuracy in the 10^{-17} range.

C. Ion and neutral traps for atomic frequency standards

Although the prospects for atomic fountains look promising, the ultimate frequency standards may eventually come from optical and magnetic traps of neutral atoms and ions. The most attractive feature of these types of standards is the ability to interact with the atom or ion for long times leading to narrower linewidths and thus enhancing the potential short term stability of frequency standards. These schemes have potential for both optical and microwave frequency standards. Recent work in laser cooling and trapping of neutral atoms [18] enables a fairly large number of atoms ($\approx 10^7$) to be cooled to sub mK temperatures and confined for long periods of time, thus allowing longer interaction times. The reduced thermal velocity of these cold atoms reduces Doppler broadening and shifting effects. Ion standards simply use electric potentials to trap ions in a linear string. Although there are far fewer ions stored in such traps compared to neutral atomic traps, they can be stored for much longer times. A current mercury ion standard has demonstrated a fractional frequency instability of $3.3 \times 10^{-13}\tau^{-1/2}$ for measurement times $\tau < 2$ hours for the 40.5 Ghz ground-state hyperfine transition. It's current accuracy of 3.4×10^{-15} is limited by a number of different systematic effects, in particular the uncertainty in the Zeeman shift due to the fields used to trap the ions. As the number of ions in these traps is very low, quantum projection noise is expected to be a limiting factor in future ion standards. One way in which the quantum limited stability of Eq. 7 may be beat is by using maximally correlated entangeled states [24]. As a simple example, consider the case were the total state of the N-ion system after the first Ramsey pulse is in the entangled state

$$|\Psi\rangle = \frac{1}{\sqrt{2}}(|00...0\rangle + |11..1\rangle) \tag{8}$$

as opposed to a product state of the individual atomic wavefunctions given in Eq. 5. After free evolution in the interaction free region, the total state of the system, in the frame rotating with the driving frequency ω is

$$|\Psi\rangle = \frac{1}{\sqrt{2}}(|(00...0\rangle + e^{iN(\omega-\omega_o)t}|11..1\rangle) \tag{9}$$

If the first ion is disentagled from the rest, the probability of finding it in state $|1\rangle$ is now

$$P_n = \frac{1}{2}\{1 + cos(N(\omega - \omega_o)T)\}. \tag{10}$$

The oscillation frequency of the Ramsey fringes will now be increased by a factor of N, likewise the width of the central fringe will be narrower by a factor of N and the quantum limited stability of Eq. 7 will go as $1/N$. Although this simple model demonstrates the potential use of quantum entanglement to circumvent the limitations imposed by shot noise, it neglects the real world effects of decoherence. A more complete description of the problem which includes the effects of decoherence can be found in Ref [25]. Here it is found that because a maximally entangled state also decoheres at a rate proportional to N (the number of ions in the experiment), the precision gained from the entanglement is exactly cancelled by the effects of decoherence on the signal to noise ratio. The authors demonstrate that the best resolution is obtained by preparing the initial system in a partially entangled state, which will not be as fragile to the effects of decoherence as the maximally entangled state. Although current ion standards have not yet reached this quantum limited stability regime, such "squeezed atomic states" may eventually provide a means to further improve the frequency stability of time standards which are based on a small number of atoms or ions.

V. CONCLUSION

We have breifly reviewed the requirements for atomic frequency standards and given examples of the more promising future standards, namely those based on atomic fountains and ion traps which isolate the atoms and ions from perturbations which broaden and shift their resonance. There are many more proposals which we certainly cannot cover in this review. All proposed methods, however, have in common the requirements outlined in this paper.

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[2] The number of oscillations a resonator makes until its energy diminishes to a fraction of its initial value.

[3] There is no uncertainty associated with this number as the second is defined by the time taken for the cesium atom to undergo this many oscillations.

[4] In this experiment the 2-photon transition was measured with an uncertainty of 1.3×10^{-11}, the best measurement of this transition to date.

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