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ION OPTICAL CLOCKS AND QUANTUM INFORMATION PROCESSING

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*Abstrac*t – **Techniques developed for quantum-information processing using trapped ions may be useful in future atomic clocks. Here, we summarize experiments at NIST that (1) use quantum gates to entangle ions and thereby improve the measurement signal-to-noise ratio in spectroscopy and (2) implement sympathetic cooling and quantum state transfer techniques, which might be used to increase the number of choices of ions used for clocks.**
***Keywords* - Atomic clocks, optical clocks, quantum information, trapped-ion frequency standards**

I. INTRODUCTION

Research on quantum information processing has been stimulated largely by the interest in developing quantum cryptographic systems and quantum computers. The goal of quantum computing is extremely daunting, and it is generally agreed that the realization of a useful quantum computer is decades away at best. In the meantime, it appears that the basic ideas behind quantum computing can be applied to less-ambitious goals. At NIST, we are investigating the use of quantum information processing in two trapped-ion clock applications. In the first, quantum-mechanical entanglement can provide a resource for increased measurement precision in spectroscopy. Given high-fidelity quantum logic operations, the time required to reach a certain measurement precision could be reduced by a factor equal to the number of ions that are entangled, when compared to the case of unentangled atoms. In the second application, we indicate how a "logic" ion, which is simultaneously trapped with a "clock" ion, can be used to provide the functions of cooling and state measurement. This may increase the number of choices of possible clock ions. We summarize progress and plans in these two areas, emphasizing future optical clocks.

II. ENTANGLEMENT-ENHANCED SPECTROSCOPY

A collection of (neutral or charged) atoms whose internal states are entangled in a specific way can improve the quantum-limited signal-to-noise ratio in spectroscopy. In principle, the time required to reach a certain measurement precision could be reduced by a factor equal to the number N of atoms entangled, as compared to the case of N unentangled atoms. This would clearly be an advantage for large N, as in neutral-atom clocks. It would still be a significant practical advantage in atomic-ion frequency standards where, to maintain high-accuracy, a relatively small number (N ≅ 10 – 100) of trapped ions may be optimum due to various experimental constraints, as in the $^{199}Hg^{+}$ microwave clock experiment at NIST [1].

In spectroscopic experiments on N two-level atoms, we can view the problem in the following way using the spin-1/2 analog for two-level atoms. (Equivalently, we can think of performing nuclear magnetic resonance spectroscopy on an ensemble of N protons.) The total angular momentum of the system is given by the "Bloch vector" $\mathbf{J} = \Sigma_i \mathbf{S}_i$, where $\mathbf{S}_i$ is the spin of the ith atom ($S_i = ½$). By choosing the quantization axis along the z direction, eigenstates of the ions are written $|\downarrow\rangle_i$ and $|\uparrow\rangle_i$ with $S_{zi}|\downarrow\rangle_i = -½|\downarrow\rangle_i$ and $S_{zi}|\uparrow\rangle_i = +½|\uparrow\rangle_i$, where $S_{zi} = ½\sigma_{zi}$ and σ_{zi} is the usual Pauli matrix for the ith ion. The goal is to measure ω_0, the resonant frequency of transitions between the $|\downarrow\rangle$ and $|\uparrow\rangle$ states, relative to the frequency ω_R of a reference oscillator.

We first prepare an initial state for the spins. For concreteness, we assume spectroscopy is performed by applying (classical) fields of frequency ω_R according to the method of separated fields by Ramsey [2]. We also assume that the maximum value of T_R, the time between Ramsey pulses, is limited by practical experimental constraints such as the desire to lock the local oscillator to the atoms within a reasonable duration of time. After applying these fields, we measure the final state populations; for example, the number $N_\downarrow$ of atoms in the $|\downarrow\rangle$ state. This can be accomplished with nearly 100 % efficiency through detection of laser fluorescence using Dehmelt's "electron-shelving" technique [3]. The signal-to-noise ratio (for repeated measurements at a particular value of ω_R) is fundamentally limited by the quantum measurement fluctuations or "projection noise" [4] in the number of atoms observed to be in the $|\downarrow\rangle$ state. These number fluctuations are equivalent to the fluctuations in repeated measurements of $\mathbf{J}_\perp$, the angular momentum in a particular direction perpendicular to $\mathbf{J}$ [4-6]. Because of this, the determination of signal-to-noise ratio in spectroscopy is equivalent to the determination of the orientation of $\mathbf{J}$ about a particular axis. If we can reduce the fluctuations of $\mathbf{J}_\perp$ along a particular direction by entangling the atoms, then we can increase the angular resolution in that direction and therefore improve signal-to-noise ratio in spectroscopy. This is indicated schematically in Fig. 1.

Spectroscopy is typically performed on atoms initially prepared in the (unentangled) "coherent" spin state [7] state $\psi = |\downarrow\rangle_1|\downarrow\rangle_2 \cdot\cdot |\downarrow\rangle_N) = |J = N/2, m_J = -N/2\rangle$ (all atoms initialized in their ground states). For this case, the imprecision in a determination of the frequency of the transition is limited by projection noise to the "standard quantum limit" for the Allen variance $\sigma_y(\tau) = 1/(\omega_0[NT_R\tau]^{½}$ where τ represents the total averaging time. This limit has been observed in spectroscopy of ions [4] and in cesium-fountain atomic clocks [8]. If the

atoms can be prepared in particular entangled states, it is possible to achieve $\sigma_y(\tau) < 1/(\omega_0[NT_R\tau]^{1/2})$. The challenge is how to actually generate the states useful for spectroscopy. One possibility is to entangle a phonon or electromagnetic field with the atoms. For example, coupling atoms to coherent or squeezed harmonic oscillator states can lead to "spin-squeezed" states. [5,9-11]. Collision interactions in a cold gas sample may eventually produce much larger squeezing [12].

Squeezed light has been observed to impart spin squeezing to a collection of (S_i = 5) cesium atoms [13]. Squeezing of an (S_i = 4) atomic sample through a quantum-nondemolition measurement of transmitted light has also been observed [14]. Squeezing in these experiments was not applied to measurement however. The trapped-ion quantum computation scheme of Cirac and Zoller [15] provides a way to generate any desired entangled state for ions; however, Sørensen and Mølmer have recently devised an efficient one-step method to create spin-squeezed states [16]. Spin squeezing of $^9Be^+$ ions was recently demonstrated [6] using this scheme.

The above example assumes that the (equivalent) angular momentum **J** is the experimental observable. If we consider

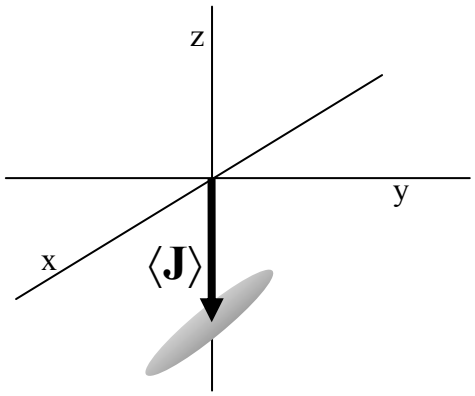

Figure 1. Spin squeezing. The net angular momentum vector (Bloch vector) ⟨**J**⟩ of a system of N spin-½ particles that points along the negative z axis in a frame that rotates at the spin precession frequency. The distance from the end of the Bloch vector to the surface of the spheroid is meant to indicate the square-root of the variance of the angular momentum measured along that direction. For the particular coherent spin state that points along the –z axis, ($\psi = |\downarrow\rangle_1|\downarrow\rangle_2 \cdots |\downarrow\rangle_N = |J = N/2, m_J = -N/2\rangle$), we have $\Delta J_z = 0$, $\Delta J_x = \Delta J_y = [J/2]^{1/2} = N^{1/2}/2$, where $\Delta J_i \equiv [\langle J_i^2\rangle - \langle J_i\rangle^2]^{1/2}$. These uncertainties would be indicated by a thin disk of radius $[J/2]^{1/2}$ in the x-y plane at the end of the angular momentum vector. When the net spin **J** is squeezed, the uncertainty in one direction is reduced below the value for a coherent spin state. In the figure, the spin is squeezed in the y direction so that the angular resolution of ⟨**J**⟩ about the x axis is increased.

other observables, it is possible to reach the Heisenberg limit of uncertainty in spectroscopy. Bollinger *et al.* [17] investigated theoretically the use of states having the form (after the first Ramsey pulse)

$$\psi = \tfrac{1}{\sqrt{2}}\left(|\downarrow\rangle_1|\downarrow\rangle_2 \cdots |\downarrow\rangle_N + |\uparrow\rangle_1|\uparrow\rangle_2 \cdots |\uparrow\rangle_N\right) \quad (1).$$

For this state, the total angular momentum vanishes along any axis, thereby precluding the measurement of **J** for determining ω_0 - ω_R. However, if after application of the second Ramsey pulse, we measure the parity operator $P \equiv \Pi_i\sigma_{zi}$, the resulting signal gives the exact Heisenberg limit for spectroscopy [17]. Therefore in principle, we could have

$$\sigma_y(\tau) = \frac{1}{\omega_0\sqrt{NT_R\tau}} \Rightarrow \frac{1}{\omega_0 N\sqrt{T_R\tau}}, \quad (2)$$

and the time required to reach a certain measurement precision would be reduced by N. Applying the method of Sørensen and Mølmer [16], an approximation to the state in Eq. (1) was generated for two and four $^9Be^+$ ions [18] and used in a spectroscopic experiment on two ions [6]. As another possible observable, we could choose to measure the variance operator [6].

III. Quantum logic for spectroscopy

An apparent limitation to modern high-accuracy atomic clocks is that they incorporate the functions of laser cooling, laser-fluorescence detection, and a good clock transition in the same atomic species. Some atoms have good clock transitions, but their laser cooling and detection transitions are impractical. Therefore, if we could provide the functions of cooling and detection some other way, more choices from the periodic table for potential clock atoms would become available. In the case of atomic ion clocks, we might be able to accomplish this in the following way.

In its simplest form, the idea is to simultaneously trap a "logic" ion and a "clock" ion. The logic ion would provide the functions of laser cooling and detection [19]. The logic ion is used to laser-cool all (harmonic) motional modes of both ions to their ground states. We assume that the clock ion is optically pumped to its initial state. After the clock radiation is applied, the clock ion is in general in a superposition state of the form $\alpha|\downarrow\rangle_C + \beta|\uparrow\rangle_C$, where the subscript C denotes the clock ion. With the use of coherent "mapping" pulses (optical sideband transitions), this state is first transferred onto a selected motional mode and subsequently onto the logic ion's internal state. Overall then, we carry out the mapping $\alpha|\downarrow\rangle_C + \beta|\uparrow\rangle_C \rightarrow \alpha|\downarrow\rangle_L + \beta|\uparrow\rangle_L$ where the subscript L denotes the logic ion. Finally we measure the logic ion, finding it to be either in the $|\downarrow\rangle_L$ state with probability $|\alpha|^2$ or in the $|\uparrow\rangle_L$ state with probability $|\beta|^2$

($|\alpha|^2 + |\beta|^2 = 1$). Each of these steps is a basic quantum logic operation and has been separately demonstrated in several experiments [20]. Although this technique requires several steps, they can all be accomplished reliably and on a time scale of order 1 ms. Therefore, the overhead in time will be negligible for precision clock experiments.

The group IIIA ions such as Tl^+ and In^+ have received considerable attention because, as Dehmelt pointed out [21], the $^1S_0 \rightarrow {}^3P_0$ transitions are free from static electric quadrupole shifts. These ions also have $^1S_0 \rightarrow {}^3P_1$ transitions at accessible wavelengths that can be used for cooling and detection, as has been demonstrated with In^+ where the $^1S_0 \rightarrow {}^3P_1$ transition ($\lambda \cong 231$ nm) has a linewidth of approximately 360 kHz [22,23]. If we transfer these functions to the logic ion, B^+, Al^+ and other ions that are difficult to laser cool might also be considered as clocks [19]. (The $^1S_0 \rightarrow {}^3P_1$ transition linewidths in B^+ and Al^+ are approximately 1.2 Hz and 320 Hz; which would therefore yield relatively weak cooling and poor detection sensitivities.) The logic ion might be $^9Be^+$ or Mg^+, but many other possibilities exist. Since the logic ion is assumed to be a different species, it is likely that laser cooling could be applied during application of the clock radiation without significantly affecting the clock levels. This would be important for very long clock interrogation times.

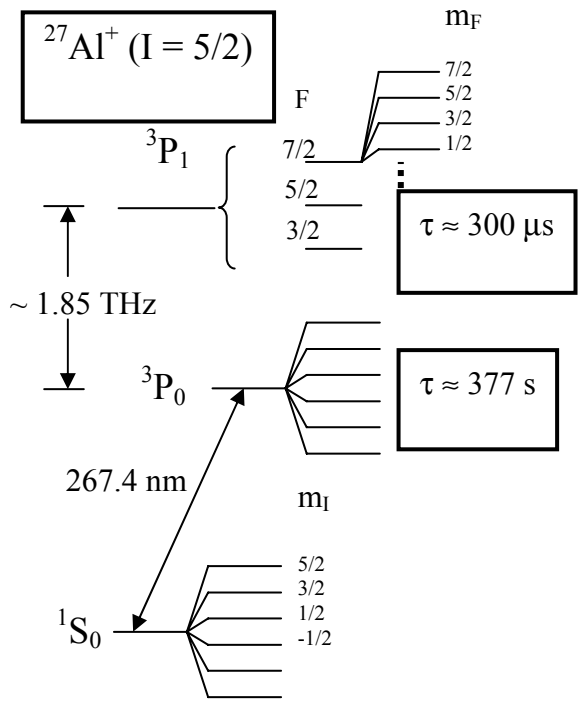

Figure 2. Partial energy level diagram for $^{27}Al^+$. The $^1S_0 \rightarrow {}^3P_0$ ($\Delta m_I = 0$) transitions could be used for an optical atomic clock.

IV. $^{27}Al^+$ Optical Clock

As an example, we consider $^{27}Al^+$ as the clock ion. ^{27}Al is the only stable isotope of aluminum and has nuclear spin I = 5/2. We show some of its relevant energy levels in Fig. 2. The $^1S_0 \rightarrow {}^3P_0$ transitions in $^{27}Al^+$ ($\lambda \cong 267.44$ nm, $\tau(^3P_0) = 377$ s [24]) would have features similar to the corresponding transitions in In^+ including a linear magnetic field dependence because of the difference in nuclear g-factors in the ground and excited states [21]. The corresponding Zeeman shift can be accounted for by measuring two $\Delta m_I = 0$ transitions with equal and opposite values of m_I [22]. Optical pumping to the $^1S_0$ ($m_I = \pm I$) states can be accomplished by driving the $^1S_0 \rightarrow {}^3P_1$ ($F = 7/2$, $m_F = \pm 7/2$) transition ($\tau(^3P_1) = 305$ μs [25]) with circularly polarized light.

We note that the magnetic-quadrupole-allowed $^1S_0 \rightarrow {}^3P_2$ transitions ($\lambda \cong 266.1$ nm, $\tau \cong 270$ s) could be driven by the fourth harmonic of a Nd-YAG laser. The $^3P_2$ hyperfine constant has been calculated to be 1125 MHz [26] so that several first-order field-independent transitions occur at magnetic fields around 100 G. Unfortunately, at the magnetic fields where these conditions hold, all $^3P_2$ states would have components with angular momentum $F > 1/2$, resulting in a static quadrupole shift as with many D-state levels. This would necessitate averaging the clock frequency for three equal-magnitude mutually-orthogonal B-field directions [27].

V. Project Status and Future Directions

With the use of entanglement, the experiment in [6] was able to demonstrate a frequency precision better than could possibly be obtained without the use of entanglement on the same number of ions. However, to be of practical use, the fidelity of entangling operations and number of ions must be increased significantly. Therefore, as part of an effort to demonstrate a scalable quantum computer, work is currently underway at NIST to improve the fidelity of quantum gates, of which some are applicable to generation of the states shown in Eq. (1) above [28]. These techniques, some of which do not require addressing of individual ions, might also be extended to large numbers of ions, as in the $^9Be^+$ Penning trap experiments [29].

Work is also underway to carry out ground state cooling of a $^{27}Al^+$ ion with a simultaneously stored $^9Be^+$ ion. We believe this should be reasonably straightforward because in a recent experiment [30], ground-state cooling of a single $^{24}Mg^+$ ion has been achieved through sympathetic cooling of a simultaneously stored $^9Be^+$ ion. Moreover, the technique for temperature measurements in [30] showed the feasibility of the mapping steps required to carry out the scheme described above. Therefore, we are currently developing a laser system to drive the relevant transitions for realizing an optical clock based on the $^1S_0 \rightarrow {}^3P_0$ transition in $^{27}Al^+$. Initial experiments will first attempt to carry out the measurement protocol on the $^1S_0 \rightarrow {}^3P_1$ transition.

Eventually, this scheme might be applied to driving electronic clock transitions in other ions such as B^+ [9] or perhaps nuclear transitions as suggested in [31].

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REFERENCES

[1] D.J. Berkeland, J.D. Miller, J.C. Bergquist, W.M. Itano, and D.J. Wineland, "Laser-cooled mercury ion frequency standard," Phys. Rev. Lett. **80**, 2089-2092 (1998).

[2] N. F. Ramsey, *Molecular Beams* (Oxford Univ. Press, London, 1963).

[3] For a summary, see: R. Blatt and P. Zoller, "Quantum jumps," Eur J. Phys. **9**, 250-279 (1988).

[4] W. M. Itano, *et al.*, "Quantum projection noise: population fluctuations in two-level systems," Phys. Rev. A **47**, 3554-3570 (1993).

[5] D. J. Wineland, J. J. Bollinger, W. M. Itano, and D. J. Heinzen, "Squeezed atomic states and projection noise in spectrosopy," Phys. Rev. A **50**, 67-88 (1994).

[6] V. Meyer *et al.,* "Experimental Demonstration of Entanglement-Enhanced Rotation Angle Estimation Using Trapped Ions," Phys. Rev. Lett. **86**, 5870-5873 (2001).

[7] F. T. Arecchi, E. Courtens, R. Gilmore, and H. Thomas, "Atomic coherent states in quantum optics," Phys. Rev. A **6**, 2211-2237 (1972).

[8] G. Santarelli *et al.*, "Quantum projection noise in an atomic fountain: a high stability cesium frequency standard," Phys. Rev. Lett. **82**, 4619-4922 (1999).

[9] D. J. Wineland *et al.*, "Squeezed atomic states and projection noise in spectrosopy," Phys. Rev. A **46**, R6797-R6800 (1992).

[10] A. Kuzmich, K. Mølmer, and E. Polzik, "Spin Squeezing in an Ensemble of Atoms Illuminated with Squeezed Light," Phys. Rev. Lett. **79**, 4782-4785 (1997).

[11] L. Vernac, M. Pinard, and E. Giacobino, "Spin squeezing in two-level systems" Phys. Rev. A **62**, 063812-1-9 (2000).

[12] A. Sørensen, L. –M. Duan, J. I. Cirac, and P. Zoller, "Many particle entanglement with Bose-Einstein-condensates," Nature **409**, 63-66 (2001).

[13] J. Hald, J. L. Sørensen, C. Schori, and E. S. Polzik, "Spin squeezed atoms: a macroscopic entangled ensemble created by light," Phys. Rev. Lett. **83**, 1319-1322 (1999).

[14] A. Kuzmich, L. Mandel, and N. P. Bigelow, "Generation of spin squeezing via continuous quantum nondemolition measurement," Phys. Rev. Lett. **85**, 1594-1597 (2000).

[15] J. I. Cirac and P. Zoller, "Quantum Computation with Cold, Trapped Ions," Phys. Rev. Lett. **74**, 4091-4094 (1995).

[16] A. Sørensen and K. Mølmer, "Entanglement and quantum computation with ions in thermal motion," Phys. Rev. A **62**, 022311-1 - 11 (2000).

[17] J. J. Bollinger, W. M. Itano, D. J. Wineland, and D. J. Heinzen, "Optimal frequency measurements with maximally correlated states," Phys. Rev. A **54**, R4649-4652 (1996).

[18] C. A. Sackett *et al.*, "Experimental Entanglement of four particles," *Nature* **404**, 256-259 (2000).

[19] D. J. Wineland, J. C. Bergquist, J. J. Bollinger, R. E. Drullinger, and W. M. Itano, "Quantum computers and atomic clocks," Proc. 6th Symposium on Frequency Standards & Metrology, ed. by P. Gill, World Scientific, Singapore (2002), pp 361-368.

[20] see for example: *Experimental Quantum Computation and Information,* Vol. 148 Int. School of Physics "Enrico Fermi," F. De Martini and C. Monroe, eds., IOS Press, Amsterdam 2002.

[21] H. G. Dehmelt, "Mono-ion oscillator as potential ultimate laser frequency standard," IEEE Trans. Instrum. Meas. **IM-31**, 83-87 (1982).

[22] Th. Becker *et al.*, "Indium single ion optical frequency standard," Proc. 2001 IEEE Int. Freq. Contr. Symp., IEEE Piscataway, NJ, Cat. No. 01CH37218, pp. 76-81.

[23] W. Nagourney, "Optical time and frequency standard based on single indium ion," Proc. 2001 IEEE Int. Freq. Contr. Symp., IEEE Piscataway, NJ, Cat. No. 01CH37218, pp. 82-84.

[24] T. Brage *et al.*, "Hyperfine induced transitions as diagnostics of isotopic composition and densities of low-density plasmas," Astrophys. J. **500**, 507-521 (1998).

[25] E. Träbert, A. Wolf, J. Linkemann, and X. Tordoir, "Measurement of the B^+ and Al^+ intercombination and Sc^{12+} forbidden transition rates at a heavy-ion storage ring," J. Phys. B.: At. Mol. Opt. Phys. **32**, 537-552 (1999).

[26] P. Jönsson, Lund Institute of Technology, private communication.

[27] W. M. Itano, "External-field shifts of the $^{199}Hg^+$ optical frequency standard," J. Res. Natl. Inst. of Stds. and Tech., **105**, 829-837 (2000).

[28] D. Leibfried *et al.*, "Experimental demonstration of a robust, high-fidelity geometric two ion-qubit phase gate," *Nature* **422**, 412-415 (2003).

[29] W. M. Itano *et al.*, "Bragg diffraction from crystallized ion plasmas, Science **279**, 686-689 (1998).

[30] M. Barrett *et al.*, "$^{24}Mg^+/^9Be^+$ sympathetic cooling for quantum logic," unpublished.

[31] E. Peik and Chr. Tamm, "Nuclear spectroscopy of the 3.5 eV transition in Th-229," Europhys. Lett. **61**, 181-186 (2003).