Quantum Coherence between Two Atoms beyond $Q = 10^{15}$

C. W. Chou,* D. B. Hume, M. J. Thorpe, D. J. Wineland, and T. Rosenband

Time and Frequency Division, National Institute of Standards and Technology, Boulder, Colorado 80305, USA

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We place two atoms in quantum superposition states and observe coherent phase evolution for 3.4×10^{15} cycles. Correlation signals from the two atoms yield information about their relative phase even after the probe radiation has decohered. This technique allowed a frequency comparison of two $^{27}Al^{+}$ ions with fractional uncertainty $3.7^{+1.0}_{-0.8} \times 10^{-16}/\sqrt{\tau/\mathrm{s}}$. Two measures of the Q factor are reported: The Q factor derived from quantum coherence is $3.4^{+2.4}_{-1.1} \times 10^{16}$, and the spectroscopic Q factor for a Ramsey time of 3 s is 6.7×10^{15}. We demonstrate a method to detect the individual quantum states of two Al^{+} ions in a Mg^{+}-Al^{+}-Al^{+} linear ion chain without spatially resolving the ions.

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Coherent evolution of quantum superpositions follows directly from Schrödinger's equation, and is a hallmark of quantum mechanics. Quantum systems with a high degree of coherence are desirable for sensitive measurements and for studies in quantum control. Typically, quantum superposition states quickly decohere due to uncontrolled interactions between the system and its environment. However, through careful isolation and system preparation, quantum coherence has been observed in naturally occurring systems including photons and atoms, as well as in engineered macroscopic systems [1–4]. In order to observe the coherence time of a system, it must be compared to a reference system that is at least as coherent, a requirement that can be difficult to satisfy, particularly in systems with the highest degree of coherence. In atomic physics, quality (Q) factors as high as 1×10^{14} to 5×10^{14} [5–8] have been observed with laser spectroscopy, where the linewidths are often limited by laser noise rather than atomic decoherence. In this report we apply a recent spectroscopic technique [9] to directly observe atomic coherence beyond the laser limit and probe an atomic resonance with a Q factor above 10^{15}.

Historically, Mössbauer spectroscopy with γ rays has exhibited the highest relative coherence, as quantified by the spectroscopic Q factor (the ratio of oscillation frequency to observed resonance linewidth). Values as high as 8.3×10^{14} are observed [10] in the 93.3 keV radioactive decay of ^{67}Zn, limited by the nuclear lifetime of 13.4 μs. One crystal containing ^{67}Zn provided the probe radiation, while another served as the resonant absorber. The Mössbauer method might be extended to characterize optical transitions in atoms [11], but here we use a method based on Ramsey spectroscopy in which the phase fluctuations of the probe source are rejected as common-mode noise [9], enabling Ramsey times that exceed the probe coherence. Other experiments that compare pairs of microwave [12] or optical clocks [13] use a related technique to reduce Dick-effect noise [14,15].

In the experiment reported here, atomic superposition states evolve coherently for up to 5 s at a frequency of 1.12×10^{15} Hz. Following Chwalla *et al.* [9], a Ramsey pulse sequence [16] is simultaneously applied to two trapped $^{27}Al^{+}$ ions, labeled $i \in \{1, 2\}$ (see Fig. 1). The probe radiation for both ions is derived from the same source. Each ion is initialized in one of the two quantum states that make up the clock transition (clock states), which need not be the same for both ions. Immediately prior to the second $\pi/2$ pulse, a variable displacement $\mathbf{r_i}$ is applied to the ions. This Ramsey sequence induces a state change with probability $p_i = (1 + \cos\delta\phi_i)/2$, where $\delta\phi_i = \phi_L + \mathbf{k} \cdot \mathbf{r_i} - \phi_i$ is the difference between the phase accumulated by the laser ($\phi_L + \mathbf{k} \cdot \mathbf{r_i}$) and ion ($\phi_i$) during the free-evolution period T, and $\mathbf{k}$ is the laser beam wave vector, $\mathbf{k} = \hat{z}2\pi/(267\ \mathrm{nm})$. The correlation probability (the probability that both ions make a transition, or both do not make a transition) is then

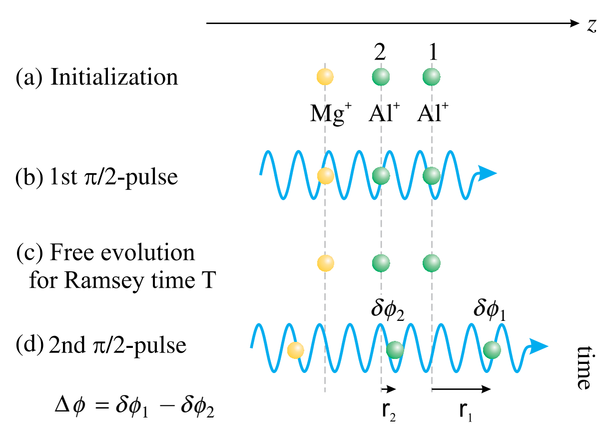

FIG. 1 (color online). Illustration of the protocol. (a) The detected states from the previous Ramsey experiment serve as the initial states for the current measurement. (b) First $\pi/2$ pulse, driven by a laser beam whose axis is along the axis of the ion array. (c) The clock state superpositions freely evolve. (d) Spacing adjustment at the end of the free-evolution period to vary the differential phase $\Delta\phi$, followed immediately by the second $\pi/2$ pulse. At the end of the sequence, ion states are detected to obtain the correlation.

$P = [2 + \cos(\delta\phi_1 - \delta\phi_2) + \cos(\delta\phi_1 + \delta\phi_2)]/4$. Here the relative phase, $\delta\phi_1 - \delta\phi_2$, is independent of ϕ_L, which is uniformly randomized over the interval $[0, 2\pi)$ by a pseudorandom number generator. Without knowledge of ϕ_L, the probability of correlated transitions is

$$P_c = \frac{1}{2\pi}\int_0^{2\pi} P d\phi_L = \frac{1}{2} + \frac{C}{2}\cos\Delta\phi, \qquad (1)$$

where $\Delta\phi = \phi_2 - \phi_1 + \mathbf{k}\cdot(\mathbf{r_1} - \mathbf{r_2})$ and $C \equiv P_{c,\max} - P_{c,\min} \leq \frac{1}{2}$ is the contrast.

The correlation signal P_c yields the differential phase of the two Al^+ "clock" ions, similar to the measurement of differential phase between source and absorber in Mössbauer spectroscopy. Its noise properties are equivalent to that of a single-ion Ramsey experiment with reduced contrast, and the statistical measurement uncertainty is determined by quantum projection noise [17]. When $|\Delta\phi|$ is kept near $\pi/2$, the statistical uncertainty of the ion-ion fractional frequency difference, or measurement instability, is $\sigma(\tau) \equiv \sigma_\nu/\nu = (2\pi\nu C\sqrt{T\tau})^{-1}$, where τ is the total measurement duration, σ_ν is the uncertainty in the measured frequency difference $(\phi_2 - \phi_1)/2\pi T$, and $\nu \approx 1.12$ PHz is the transition frequency. Importantly, the free-evolution period T is not limited by laser phase noise.

In the experiment, a linear Paul trap confines one $^{25}Mg^+$ ion and two Al^+ ions in an array [18,19] along the trap z axis (Fig. 1). The motional frequencies of a single Mg^+ in the trap are $\{f_x, f_y, f_z\} = \{5.13, 6.86, 3.00\}$ MHz. The ions are maintained in the order of Mg^+-Al^+-Al^+ (inter-ion spacing 2.69 μm) by periodically adjusting the trap conditions and verifying via Mg^+ spectroscopy the frequency of the "stretch" mode of motion, whose value is 5.1 MHz for the correct order [20].

The two Al^+ clock states, $|\downarrow\rangle \equiv |^1S_0, m_F = 5/2\rangle$ and $|\uparrow\rangle \equiv |^3P_0, m_F = 5/2\rangle$, are detected with an adaptive quantum nondemolition process [19]. The present implementation distinguishes all four states $|\downarrow_1\downarrow_2\rangle$, $|\downarrow_1\uparrow_2\rangle$, $|\uparrow_1\downarrow_2\rangle$, and $|\uparrow_1\uparrow_2\rangle$ by observing Mg^+ fluorescence after controlled interactions between the Al^+ and Mg^+ ions. Individual state detection relies on the two Al^+ ions having different amplitudes in several motional eigenmodes, which affects the state-mapping probability onto the Mg^+ ion. Information from several measurements is combined in a Bayesian process [19], to determine the most likely state of the two Al^+ ions with typically 99% probability in an average of 30 detection cycles (approximately 50 ms total duration). This technique allows individual state detection of two ions in the same trap, without the need for high spatial-resolution optics.

The Ramsey experiments have $\pi/2$-pulse durations of 1.2 ms and are carried out for various free-evolution periods T. For each T, $\Delta\phi_z \equiv \mathbf{k}\cdot(\mathbf{r_1} - \mathbf{r_2})$ is varied from 0 to beyond 2π by reducing the z-axis trap confinement and thereby increasing the ion spacing [21]. The duration required to shift $\mathbf{r_i}$ is approximately 10 ms [22]. Figure 2 shows the correlation signals for T between 0.1 and 5 s. Currently, collisions (approximately 0.7/minute) between the ions and background gas make it impractical to generate sufficient statistics for T greater than 5 s. The collisions result in changes of ion order and loss of ions due to chemical reactions.

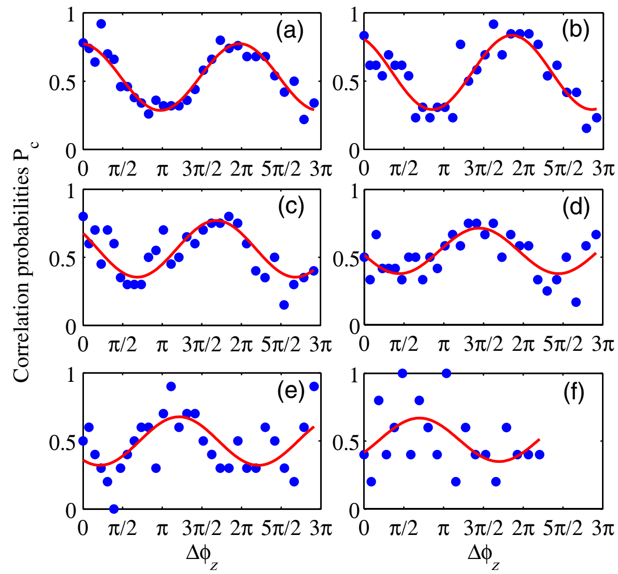

FIG. 2 (color online). Correlation probabilities P_c versus $\Delta\phi_z$ for various Ramsey times: (a) 0.1 s, 1500 probes; (b) 0.5 s, 600 probes;(c) 1 s, 600 probes; (d) 2 s, 360 probes; (e) 3 s, 300 probes; (f) 5 s, 100 probes. Dots: measurement outcomes; lines: maximum-likelihood fits to the fringes.

The phase difference $\phi_2 - \phi_1$, and thus the frequency difference, between the two Al^+ ions can be determined from the phases of the P_c fringes in Fig. 2. In the experiment, we apply a magnetic-field gradient of 1.32 ± 0.33 mT/m, as measured by monitoring the frequency of the $|F = 3, m_F = -3\rangle \rightarrow |F = 2, m_F = -2\rangle$ magnetic-field dependent transition in the $^{25}Mg^+$ $3s$ $S_{1/2}$ ground state hyperfine manifold, when the Mg^+ position along the trap axis is adjusted. This gradient induces a fractional frequency shift $(\nu_2 - \nu_1)/\nu = 1.32 \pm 0.33 \times 10^{-16}$ between the $|\downarrow\rangle \leftrightarrow |\uparrow\rangle$ transitions of the two Al^+ ions. The phases of the P_c fringes, determined by maximum-likelihood fits [23], increase linearly with T, as shown in Fig. 3(a). A linear fit has a slope of 0.84 ± 0.06 rad/s, corresponding to a measured shift of $1.19 \pm 0.08 \times 10^{-16}$, in agreement with the shift caused by the magnetic-field gradient. All reported uncertainties represent a 68% confidence interval.

We derive the contrast C from the maximum-likelihood fits to the data in Fig. 2. An exponential fit of C versus T yields a relative coherence time T_C of $9.7^{+6.9}_{-3.1}$ s, corresponding to a Q factor ($Q = \pi\nu T_C$ [24]) of $3.4^{+2.4}_{-1.1} \times 10^{16}$. A uniform prior distribution of T_C on the interval 0 to 25 s is assumed. The measured coherence time is compatible with the expected result, which is given by

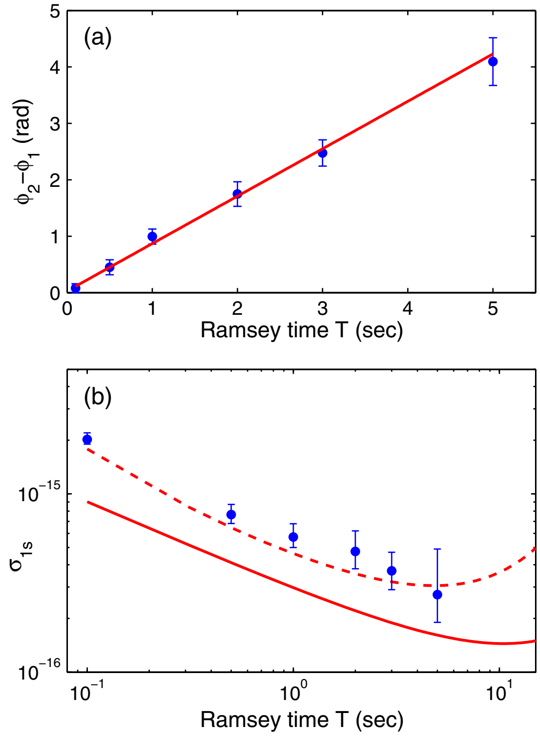

FIG. 3 (color online). (a) Differential phase $\phi_2 - \phi_1$ versus Ramsey time T. The solid line is a linear fit, with slope 0.84 ± 0.06 rad/s. (b) Measurement uncertainty extrapolated to 1 s averaging time as a function of Ramsey time. Dots: measurement results, where the uncertainties are derived from the uncertainties in the contrast C; solid line: theoretical lifetime-limited instability, where only phases corresponding to $\Delta\phi \approx \pm\pi/2$ are probed; dashed line: expected experimental instability, with $\Delta\phi$ uniformly distributed over $[0, 2\pi)$. The dashed line is derived from the measured coherence time of 9.7 s, and an approximate overhead of 100 ms per Ramsey measurement, which reduces the duty cycle.

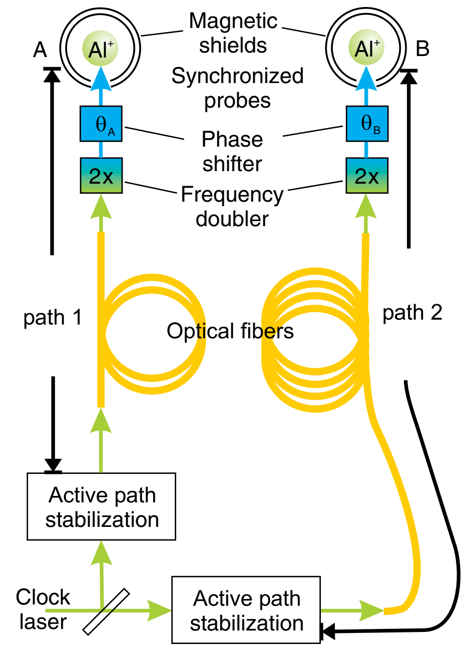

FIG. 4 (color online). Proposed frequency comparison of remote optical clocks, here based on Al^+ ions. Paths 1 and 2 that direct the clock laser light to the ions must be phase stabilized. Local frequency fluctuations, such as those caused by magnetic-field noise, should be minimized. The free-evolution periods are synchronized so that the atoms experience the same phase noise in the Ramsey pulses, the effect of which cancels in the protocol.

the lifetime $T' = 20.6 \pm 1.4$ s [25] of the Al^+ $|^3P_0\rangle$ state. When viewed in terms of Ramsey spectroscopy, for $T = 3$ s, the full-width-at-half maximum of the Ramsey signal corresponds to a Q factor of $2\nu T = 6.7 \times 10^{15}$.

The current protocol could significantly reduce the total duration of future high-precision measurements with atomic clocks. Figure 3(b) shows the measurement uncertainties extrapolated to 1 s ($\sigma_{1\,\mathrm{s}}$) versus Ramsey time T. The long-term statistical uncertainty is then $\sigma(\tau) = \sigma_{1\,\mathrm{s}}/\sqrt{\tau/\mathrm{s}}$ for a measurement duration τ. For $T = 3$ s, the frequency difference between the two Al^+ ions can be determined with a fractional uncertainty $\sigma = 1.1 \times 10^{-17}$ in a 1126 s measurement (900 s integrated free-evolution time), which can be extrapolated to infer a relative measurement uncertainty $\sigma_{1\,\mathrm{s}} = 3.7 \times 10^{-16}$. This result may be compared to a recent frequency difference measurement of two Al^+ clocks, where 65 000 s were required to reach the same uncertainty of 1.1×10^{-17} [18]. In general, the lifetime-limited contrast is $C = \frac{1}{2}\exp(-T/T')$, yielding an instability of $\sigma(\tau) = \exp(T/T')/(\pi\nu\sqrt{T\tau})$, which is shown for Al^+ in Fig. 3(b) (solid line). The optimal probe time of $T = T'/2$ yields $\sigma_{1\,\mathrm{s}} = 1.4 \times 10^{-16}$.

Although we have used the technique to measure two ions in the same trap, it may also be applied to clocks at different locations. A proposed frequency comparison of remote optical clocks is depicted in Fig. 4. The Ramsey free-evolution time T will initially be varied from a short duration to a long one, as shown in Fig. 3(a). This step allows a coarse determination of the difference-frequency, to know which fringe of the periodic Ramsey signal is being probed at long durations. As is common in atomic clocks, the main measurement proceeds via lock-in detection at the longest free-evolution time and with a phase difference that is modulated between $\Delta\phi \approx \pm\pi/2$ to maximize stability.

Note, however, that due to the requirement that ϕ_L be the same for both clocks, this technique is limited to comparisons between clocks operating at similar frequencies. Although the clocks need not be identical, the differential phase, $\Delta\phi$, must be known well enough to make phase errors of π unlikely. In order to retain control over the differential phase, the individual paths (paths 1 and 2 in Fig. 4) need to be phase-stabilized and the Ramsey pulses at the two locations need to be synchronized so that the two clocks experience the same laser phase noise. For ions with very long radiative lifetimes, the same technique could be used to compare two ion samples, each composed of maximally entangled states [26,27].

A similar approach can be taken in comparisons of two clocks composed of many unentangled atoms. The measurement protocol is again based on synchronized Ramsey pulses where the free-evolution time T exceeds the laser coherence time. The two clocks (labeled $X \in \{A, B\}$) measure transition probabilities $p_X = \frac{1}{2}[1 + \cos(\phi_X - \phi_L - \theta_X)]$, and the quantity of interest $\delta\phi_{AB} = \phi_A - \phi_B$ is determined from $\delta\phi_{AB} = \cos^{-1}(2p_A - 1) - \cos^{-1}(2p_B - 1) + \theta_A - \theta_B$, where θ_X are the controlled laser phase offsets at the two clocks. If we consider only atomic projection noise in p_A and p_B, this measurement has a variance of $\mathrm{var}(\delta\phi_{AB}) = \frac{1}{N_A} + \frac{1}{N_B}$, where N_A and N_B are the numbers of atoms in clocks A and B. The fractional frequency stability of the clock comparison is then $\sigma_y(\tau) = \sqrt{\mathrm{var}(\delta\phi_{AB})}/(2\pi\nu\sqrt{T\tau})$.

A complication is introduced by the fact that ϕ_L will be initially unknown, which leads to ambiguities in the trigonometric inversions from which $\delta\phi_{AB}$ is calculated. Such ambiguities will be absent in the majority of measurements, if an approximate value of $\delta\phi_{AB}$ can be determined through prior calibrations [with $\mathrm{var}(\delta\phi_{AB}) \ll 1$], and phase offsets θ_X are adjusted such that $\phi_A - \phi_B - (\theta_A - \theta_B) \approx \pi/2$. After this calibration procedure, p_A and p_B represent approximate quadratures of the laser-atom phase difference, and for most values of ϕ_L the trigonometric inversions are unambiguous. In such a measurement the Ramsey free-evolution time is no longer constrained by laser decoherence, and the Dick effect due to the probe source is absent. Therefore, more rapid frequency comparisons of similar-frequency many-atom optical clocks should also be possible.

Small values of $\sigma(\tau)$ in frequency comparisons are useful for evaluating and improving the performance of optical clocks and for metrological applications. For example, comparison of clocks in geographically distinct locations can be used to evaluate spatial and temporal variations in the geoid [7,13]. More generally, any physical process that leads to small, constant frequency shifts in an optical clock can be studied in this way. This includes relativistic effects as well as shifts caused by electric fields, magnetic fields and atom collisions. Our observation of a Q factor beyond 10^{15} and a frequency ratio measurement instability of $3.7 \times 10^{-16}/\sqrt{\tau/\mathrm{s}}$ highlights the intrinsic sensitivity of optical clocks as a metrological tool.

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*chinwen@nist.gov

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