Optimal frequency measurements with maximally correlated states

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We show how maximally correlated states of N two-level particles can be used in spectroscopy to yield a frequency uncertainty equal to $(NT)^{-1}$, where T is the time of a single measurement. From the time-energy uncertainty relation we show that this is the best precision possible. We rephrase these results in the language of particle interferometry and obtain a state and detection operator which can be used to achieve a phase uncertainty exactly equal to the $1/N$ Heisenberg limit, where N is the number of particles used in the measurement. [S1050-2947(96)50712-2]

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Quantum limits to noise in spectroscopy [1–4] and interferometry [5–13] have been a subject of fundamental, and to an increasing degree, practical interest. This is especially true for spectroscopy on trapped atoms or ions where the number of particles N is fixed and kept small to reduce undesired perturbations. Naively, the uncertainty of a spectroscopic or interferometric measurement is limited by counting statistics to be inversely proportional to $N^{1/2}$. However, a number of proposals have shown that by introducing quantum correlations between the particles, the measurement uncertainty can be reduced so that it scales inversely with N rather than $N^{1/2}$ [2–13]. None of the ideas discussed so far, however, have realized the fundamental limit for quantum noise in the measurement of atomic or interferometric phase, which we show to be precisely equal to $1/N$. In the present work we realize this fundamental quantum limit for any N with an approach that examines a different type of correlation and measures a different operator than previously considered.

We start by considering the spectroscopy of N two-level particles. In an equivalent spin representation [1,14], let $|J=N/2, M_J=-N/2\rangle$ denote the state where all the particles are in the ground (spin-down) state $|g\rangle$ (with energy E_g) and $|N/2,N/2\rangle$ denote the state where all the particles are in the excited (spin-up) state $|e\rangle$ (with energy E_e). We show how the state

$$|\Psi_M\rangle \equiv \{|N/2,N/2\rangle + |N/2,-N/2\rangle\}/\sqrt{2} \qquad (1)$$

can be used to measure $\omega_0=(E_e-E_g)/\hbar$ with a frequency uncertainty equal to $(NT)^{-1}$, where T is the time of a single measurement. This state is "maximally correlated" in the sense that a measurement of any one atom's energy eigenstate determines the state of all of the others. It is an N-particle version of the two-particle states discussed in the Einstein-Podolsky-Rosen experiments [15]. The use of $|\Psi_M\rangle$ requires measuring a different operator than customary in spectroscopy. We find a measurement operator which yields a $1/(NT)$ uncertainty and discuss how this measurement operator and $|\Psi_M\rangle$ can be realized with small numbers of trapped ions. Our arguments are phrased in the language of spectroscopy by the Ramsey technique of separated oscil-

lating fields [16]. The Ramsey technique is formally equivalent to Mach-Zehnder interferometry [4,6]. Therefore, after obtaining our results for optimal frequency measurements, we rephrase them in terms of interferometry.

References [2] and [4] discuss the basic idea of using correlated or squeezed spin states to improve the precision in spectroscopy for the Ramsey technique of separated oscillating fields. We briefly review the idea here and start by considering the case of N uncorrelated particles where each particle is initially prepared in its ground state $|g\rangle$. The initial state of the composite system for this case is equivalent to the $|J=N/2, M_J=-N/2\rangle$ state of a $J=N/2$ spin. This initial ($t=0$) state has $\langle J_z\rangle_0=-N/2$, $\langle J_x\rangle_0=\langle J_y\rangle_0=0$, and $\Delta J_x(0)=\Delta J_y(0)=N^{1/2}/2$. [Here $\langle A\rangle_t$ denotes the expectation value of an operator A at time t and $\Delta A(t) \equiv \langle \Delta A^2\rangle_t^{1/2}$, where $\Delta A^2 \equiv A^2-\langle A\rangle^2$.] The Hamiltonian for the equivalent spin system is $H=-\vec{\mu}\cdot\vec{B}$, where $\vec{\mu}=\mu_0\vec{J}$ is the magnetic moment of the composite system and $\vec{B}$ is the applied field. Here $\vec{B}=B_0\hat{z}+\vec{B}_1$, where $B_0=-\hbar\omega_0/\mu_0$ (we assume $\mu_0<0$) and $\vec{B}_1$ is an applied field used to perform spectroscopy. We assume that $\vec{B}_1$ is perpendicular to and rotates about the z axis according to $\vec{B}_1=B_1[-\hat{x}\sin\omega t +\hat{y}\cos\omega t]$. In the Ramsey technique [16], B_1 is applied (is nonzero) for two periods of length $t_{\pi/2}=\pi/(2\Omega_R)$, where $\Omega_R=|\mu_0B_1|/\hbar$ is the Rabi frequency, separated by a period of length T during which $B_1=0$. It is convenient to describe the Ramsey technique in a frame of reference rotating with $\vec{B}_1$. With the assumption $\Omega_R\gg|\omega_0-\omega|$, the first pulse rotates the spin vector about $\vec{B}_1$ (the y axis in the rotating frame) by 90°. The spin vector then precesses about the z axis during the field-free period, acquiring an angle $(\omega_0-\omega)T$ relative to its initial direction (the $-x$ axis) in the rotating frame. This angle could be read out by measuring, for example, J_x in the rotating frame. Experimentally this is done by applying the second $\pi/2$ pulse, which rotates the spin vector by 90° about the $\vec{B}_1$ axis, and then measuring the number of atoms in $|e\rangle$. This final measurement is equivalent to measuring J_z. We obtain

$$\langle J_z\rangle_{t_f}=(N/2)\cos(\omega_0-\omega)T, \qquad (2)$$

where $t_f = 2t_{\pi/2} + T$. Throughout this paper we assume $T \gg t_{\pi/2}$ so that $t_f \simeq T$.

Measurements of J_z as a function of ω, along with Eq. (2), can be used to estimate the frequency ω_0. [We assume that ω_0 is sufficiently well known that ω can be chosen on the central fringe of Eq. (2). This is true for atomic frequency standards and clocks.] Because of the statistical nature of quantum mechanics, the number of particles which are observed to make a transition to $|e\rangle$ from measurement to measurement will fluctuate by $\Delta J_z(t_f)$ [1,2,4]. This produces an uncertainty in the estimate of ω_0 of $|\Delta\omega| \equiv \Delta J_z(t_f)/|\partial\langle J_z\rangle_{t_f}/\partial\omega|$ [4]. For the initial $|N/2,-N/2\rangle$ state of uncorrelated particles, we obtain $|\Delta\omega(\text{uncorrelated})| = \Delta J_y(0)/[T|\langle J_z\rangle_0|] = N^{-1/2}T^{-1}$ independent of ω. [Experimental measurements are usually made with $\omega \sim \omega_0 \pm \pi/(2T)$, where Eq. (2) has its steepest slope. This minimizes the contribution of any added noise.] With correlations between the internal states of the particles it is possible to start with a state $\langle\vec{J}\rangle = \hat{z}\langle J_z\rangle_0$ such that $\Delta J_y(0) < N^{1/2}/2$. Such ''spin-squeezed'' states can be used to improve the resolution in Ramsey spectroscopy if $\Delta J_y(0)/|\langle J_z\rangle_0| < (2J)^{-1/2} = N^{-1/2}$.

Some correlated states, such as $|\Psi_M\rangle$ in Eq. (1), have a mean spin vector $\langle\vec{J}\rangle = 0$. In this case the previous description of the Ramsey technique in terms of the precession of a mean spin vector is inadequate. In order to motivate how the maximally correlated state $|\Psi_M\rangle$ can improve the precision of Ramsey spectroscopy, recall that for a single particle the precession angle $(\omega_0 - \omega)T$ that is measured in the Ramsey method is, in the rotating frame, just the phase factor $e^{-i(\omega_0 - \omega)T}$ that the excited state $|e\rangle$ acquires relative to the ground state $|g\rangle$ during the freed precession period T. Consequently, it may be possible to improve the precision of Ramsey spectroscopy with a state which, when rotated by the first $\pi/2$ pulse, is a coherent superposition of two energy eigenstates whose energies differ by more than $\hbar(\omega_0 - \omega)$. For N two-level particles the eigenstates $|N/2,-N/2\rangle$ and $|N/2,N/2\rangle$ provide the largest energy difference, with an accumulated phase difference over the free precession period which is N times greater than for a single particle. However, because $\langle\Psi_M|\vec{J}|\Psi_M\rangle = 0$, some operator other than J_z must be detected after the final $\pi/2$ pulse.

Consider the operator $\widetilde{O} = \Pi_{i=1}^{N}\sigma_{z_i}$, where σ_{z_i} is the z Pauli spin matrix for the ith atom. For $J = N/2$ this operator is diagonal in the $|J,M_J\rangle$ basis with eigenvalues $(-1)^{J-M_J}$. It can be detected by measuring the number of particles in either the spin-up or spin-down state. Experimentally this can be done with nearly 100% efficiency using electron shelving and quantum jump detection [1]. If N_g particles are measured in the spin-down state (the ground state), the result of this measurement is assigned the value $(-1)^{N_g}$ [17]. Suppose the initial state is $\exp\{i\pi J_y/2\}|\Psi_M\rangle$, so that at the end of the first $\pi/2$ pulse the state $|\Psi_M\rangle$ is created. We want to calculate $\langle\widetilde{O}\rangle_{t_f} \equiv \langle\Psi_f|\widetilde{O}|\Psi_f\rangle$, where

$$|\Psi_f\rangle = e^{-i(\pi/2)J_y}e^{-i(\omega_0-\omega)TJ_z}|\Psi_M\rangle. \tag{3}$$

Let $|e\rangle_i$ and $|g\rangle_i$ denote the excited and ground states of the

ith atom and $S_{y_i} = \sigma_{y_i}/2$, where σ_{y_i} is the y Pauli spin matrix for the ith atom. The state $|\Psi_f\rangle$ can be written

$$|\Psi_f\rangle = \frac{1}{\sqrt{2}}\left\{ e^{-iN\phi}\prod_{i=1}^{N} e^{-i(\pi/2)S_{y_i}}|e\rangle_i + e^{iN\phi}\prod_{i=1}^{N} e^{-i(\pi/2)S_{y_i}}|g\rangle_i \right\}, \tag{4}$$

where $\phi = (\omega_0 - \omega)T/2$. With $\exp[-i(\pi/2)S_{y_i}] = (1 - S_{+_i} + S_{-_i})/\sqrt{2}$, Eq. (4) can be rewritten as

$$|\Psi_f\rangle = \frac{1}{2^{(N+1)/2}}\left\{ e^{-iN\phi}\prod_{i=1}^{N} (|e\rangle_i + |g\rangle_i) + e^{iN\phi}\prod_{i=1}^{N} (-|e\rangle_i + |g\rangle_i) \right\}. \tag{5}$$

Explicit computation then yields $\langle\widetilde{O}\rangle_{t_f} = (-1)^N\cos[N(\omega_0 - \omega)T]$ and, because $\widetilde{O}^2 = 1$, $\langle\Delta\widetilde{O}^2\rangle_{t_f} = \sin^2[N(\omega_0 - \omega)T]$. Note that $\langle\widetilde{O}\rangle_{t_f}$ has the same form as that of a single, two-level system with frequency interval $N\omega_0$. The state $|\Psi_M\rangle$ can therefore be used in spectroscopy with a frequency uncertainty $|\Delta\omega| = \Delta\widetilde{O}(t_f)/|\partial\langle\widetilde{O}\rangle_{t_f}/\partial\omega| = (NT)^{-1}$ independent of ω.

The Ramsey method measures ω_0 by measuring the free precession of N identical two-level particles; that is, ω_0 is measured by observing the free time evolution of the system. With the state $|\Psi_M\rangle$ a frequency uncertainty of $(NT)^{-1}$ is obtained. We show that this is the best precision that can be obtained on N identical two-level particles which undergo free time evoloution for a period of length T. This follows from an application of the time-energy uncertainty principle

$$\delta t^2\langle\Delta H^2\rangle \geq \hbar^2/4, \tag{6}$$

where $\langle\Delta H^2\rangle$ is the variance of the Hamiltonian and δt^2 is the variance in estimating time from a measurement on the system. (Measurements of an operator A can be used to determine time with an uncertainty $\Delta A/|d\langle A\rangle/dt|$. See Ref. [18] for a simple proof of the time-energy uncertainty relation and Ref. [19] for additional rigorous discussions.) For the system of N identical two-level particles, Eq. (6) can be reexpressed in terms of dimensionless quantities

$$\delta\varphi^2\langle\Delta h^2\rangle \geq \tfrac{1}{4}, \tag{7}$$

where $\varphi = \omega_0 t$ and $h = \Sigma_{i=1}^{N}\{\tfrac{1}{2}|e\rangle_{ii}\langle e| - \tfrac{1}{2}|g\rangle_{ii}\langle g|\}$. Note that we are here considering the full 2^N-dimensional Hilbert space and not just the $J = N/2$ subspace discussed earlier. We can establish an upper limit $\langle\Delta h^2\rangle \leq N^2/4$ from $\langle\Delta h^2\rangle = \langle h^2\rangle - \langle h\rangle^2 \leq \langle h^2\rangle$ and $\langle h^2\rangle \leq N^2/4$. The last inequality follows because the maximum eigenvalue of h^2 is $N^2/4$. These inequalities and Eq. (7) imply $\delta\varphi \geq N^{-1}$. An uncertainty $\delta\varphi$ in determining φ after a free time evolution of duration T results in an uncertainty $\delta\omega_0 = \delta\varphi/T$ in the determination of ω_0. From the previous discussion, $\delta\omega_0$ must satisfy

$$\delta\omega_0 \geq \frac{1}{NT}. \tag{8}$$

Ramsey spectroscopy is formally equivalent to Mach-Zehnder interferometry. In Mach-Zehnder interferometry, schemes [5–13] which use nonclassical input states to approach the $1/N$ Heisenberg limit for large N have been proposed [20]. Examples include the illumination of one of the input ports by a squeezed vacuum [5] where experiment has shown improvement over the shot-noise limit [8], the use of correlated input states [6,7,9], and the use of two Fock states containing equal numbers of particles as inputs [12]. Reference [13] also considers the dual Fock input state, but with a phase measurement scheme that has been optimized according to quantum information theory. Most of these cases show an asymptotic phase sensitivity proportional to $1/N$. In general, the constant of proportionality is >1. By rephrasing our results for spectroscopy in terms of Mach-Zehnder interferometry, we obtain an input state and detection scheme that achieve a sensitivity *equal* to $1/N$ even for small N. The state after the first beam splitter that is formally equivalent to $|\Psi_M\rangle$ is $|\Psi_M\rangle_{\mathrm{int}} = \{|N\rangle_a|0\rangle_b + |0\rangle_a|N\rangle_b\}/\sqrt{2}$, where a and b denote the modes of the two arms of the interferometer, $|N\rangle$ denotes the state with N particles, and $|0\rangle$ denotes the vacuum. (The particles can be bosons or fermions [7].) The operator $\tilde{O}$ can be detected by measuring the number of particles $N_{b'}$ in the b' output mode of the second beam splitter. The result of such a measurement is assigned the value $(-1)^{N_{b'}}$.

Cirac and Zoller [21] have recently described a method for preparing general quantum states of a string of N laser-cooled ions in a linear rf trap. The method uses a well-focused laser beam to couple the internal states of individual ions with a mode of the ion string. The mode is assumed to be a quantized harmonic oscillator and is initially prepared in the $|n=0\rangle$ state. Reference [21] discusses the steps needed to prepare the state $|\Psi_M\rangle$. Coupling the internal state of an individual ion with a mode of the ion string without perturbing the state of the neighboring ions may be difficult. Typical ion spacings are approximately 10 μm [22] and, because the frequency differences between the modes increase with decreasing ion separation, small ion spacings are desirable.

Here we discuss a different method for making $|\Psi_M\rangle$ that does *not* require interacting with individual ions. This method refines and extends the techniques discussed in Refs. [2] and [4]. In these references, ions in a linear rf trap were assumed to be prepared in either one of the $|N/2,\pm N/2\rangle$ states. This was followed by a resonant coupling of the internal states of all the ions (with identical interaction strength) with a center-of-mass (c.m.) mode. This coupling can take the form $\hbar\Omega(J_+a + J_-a^\dagger)$ or $\hbar\Omega(J_+a^\dagger + J_-a)$, where $a^\dagger$ (a) is the raising (lowering) operator for the c.m. mode, and J_+ (J_-) is the raising (lowering) operator for the $J=N/2$ ladder of atomic states. If the c.m. mode is initially prepared in a coherent state or a squeezed state, Refs. [2] and [4] show that correlated atomic states can be made. In order to make the state $|\Psi_M\rangle$, we consider the second-order sideband interactions

$$H_2' = \hbar\Omega'(J_+a^\dagger b + J_-ab^\dagger),$$

$$H_2'' = \hbar\Omega''(J_+ab^\dagger + J_-a^\dagger b), \tag{9}$$

$$H_3 = \hbar\Omega_3(b^\dagger c + bc^\dagger),$$

where b and c denote the lowering operator for the second and third c.m. modes. Suppose the system is initially prepared in the state $|J=N/2,-J\rangle|0\rangle_a|1\rangle_b|0\rangle_c$. Application of a $\pi/2$ pulse with H_2' generates the coherent superposition

$$\{|J,-J\rangle|0\rangle_a|1\rangle_b|0\rangle_c + |J,-J+1\rangle|1\rangle_a|0\rangle_b|0\rangle_c\}/\sqrt{2}. \tag{10}$$

We now ''shelve'' the first term of Eq. (10) with a π pulse of the H_3 interaction. This swaps the wave functions of the b and c c.m. modes with the result that the second term in Eq. (10) remains unchanged but the first term becomes $|J,-J\rangle|0\rangle_a|0\rangle_b|1\rangle_c$. A π pulse with H_2'' can now be used to increase M_J by 1 in the second term of Eq. (10) without affecting the first term. This is then followed by a π pulse of H_2', which further increases M_J by 1. In this manner, by alternating H_2' and H_2'' π pulses, M_J in the second term of Eq. (10) can be increased to $J-1$ with an $n=1$ Fock state in one of the a or b modes and $n=0$ states in the other c.m. modes. Suppose this term is $|J,J-1\rangle|1\rangle_a|0\rangle_b|0\rangle_c$. (A similar argument follows if the $n=1$ Fock state is in the b mode.) Application of another π pulse with H_3 results in the state

$$\{|J,-J\rangle|0\rangle_a|1\rangle_b|0\rangle_c + |J,J-1\rangle|1\rangle_a|0\rangle_b|0\rangle_c\}/\sqrt{2}. \tag{11}$$

A π pulse with H_2'' now results in the desired state $|\Psi_M\rangle|0\rangle_a|1\rangle_b|0\rangle_c$.

Realization of the above scheme appears feasible with a string of ions in a linear rf trap. The second-order sideband interactions H_2' and H_2'' can be realized for the two c.m. modes corresponding to motion orthogonal to the ion string axis. For example, if ω_0 is a ground-state hyperfine transition, then H_2' and H_2'' can be realized by stimulated Raman transitions tuned to $\omega_0 + \omega_a - \omega_b$ and $\omega_0 - \omega_a + \omega_b$, respectively. (We assume $\omega_a \neq \omega_b$ and require that the laser beam waists be large compared to the ion string.) Parametric mode coupling has been used in mass spectroscopy experiments [23] to generate H_3 and exchange the states of two c.m. modes (in a classical regime). It has also been discussed in the quantum regime [24]. Preparation of $n=1$ Fock states has been realized with a single trapped ion [25]. By preparing a state where one of the ions is shelved in an auxiliary level, these single-ion techniques can be used to create an $n=1$ Fock state for the c.m. mode of a string of ions. Alternately, a coupled trap [24] could possibly be used to transfer an $n=1$ Fock state from a single trapped ion to the c.m. mode of a string of ions.

Current proposals for accurate microwave frequency standards based on trapped ions include linear ion traps with a small number ($N<50$) of trapped ions [26]. Therefore the preparation of $|\Psi_M\rangle$ and its use in frequency metrology is of important practical interest even for small N. Models for the decay of quantum coherence predict that the coherence in $|\Psi_M\rangle$ may decay up to N^2 times faster than for a single ion

[27]. In one experiment, a lower limit of 10 min has been obtained for the coherence time of an individual ion [28]; coherence times more than an order of magnitude longer than this are anticipated [29]. Therefore, for $N < 50$, coherence times for $|\Psi_M\rangle$ on the order of 10 s or longer appear feasible in trapped ion experiments. This is comparable to measurement times used in current trapped ion experiments (where the length of the measurement time can be limited by the local oscillator stability). In addition, it is long enough to provide a means to study the decoherence of a large quantum system.

In summary, we have shown how the maximally correlated state $|\Psi_M\rangle$ can be used in spectroscopy to yield a frequency uncertainty equal to $(NT)^{-1}$. This is the least uncertainty that can be achieved by observing the free time evolution of N two-level atoms. Preparation of $|\Psi_M\rangle$ and its use in frequency metrology appear feasible for small numbers of ions in a linear rf trap.

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