

### Spin squeezing and reduced quantum noise in spectroscopy

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We investigate the quantum-mechanical noise in spectroscopic experiments on ensembles of  $N$  two-level (or spin- $\frac{1}{2}$ ) systems where transitions are detected by measuring changes in state population. By preparing correlated states, here called squeezed spin states, we can increase the signal-to-noise ratio in spectroscopy (by approximately  $N^{1/2}$  in certain cases) over that found in experiments using uncorrelated states. Possible experimental demonstrations of this enhancement are discussed.

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Squeezed states of the electromagnetic field have received much attention for over a decade [1] and have now been applied to fundamental metrology such as the reduction of noise in optical interferometers below the standard quantum limit [2]. Related to this are theoretical investigations of noise reduction in fermion interferometers with correlated input states [3,4]. In this paper we investigate an application of squeezing to the reduction of quantum-mechanical noise in spectroscopic experiments on ensembles of two-level systems. We also suggest how this squeezing might be realized in an experiment.

We will be specifically interested in experiments where transitions are detected by measuring the changes in population of one of the two levels. This is to be contrasted with experiments which detect the radiation transmitted through an absorber; these experiments benefit from squeezed radiation [5]. We first show that preparation of correlated input states (squeezed spin states) improves, in a fundamental way, the signal-to-noise ratio (SNR) in any spectroscopic experiment which is limited by the quantum-mechanical uncertainty in the measurement of the level populations. We then discuss, as an example, a possible experimental realization of this improvement using laser-cooled stored ions, where the SNR is currently limited by quantum fluctuations in measured populations [6].

Since the dynamics of a two-level system interacting with radiation are the same as the dynamics of a spin- $\frac{1}{2}$  particle in a magnetic field [7] we will discuss only the latter case. We begin by assuming an ensemble of  $N$  spin  $S = \frac{1}{2}$  systems each with magnetic moment  $\mu = \mu_0 \mathbf{S}$  and whose direct dipole-dipole interaction can be neglected. Each spin interacts with an externally applied magnetic field  $B_0 \hat{z}$  ( $B_0 = \text{const}$ ) through the Hamiltonian  $H_0 = -\mu \cdot (B_0 \hat{z}) = \hbar \omega_0 S_z$  where  $\omega_0$  is the precession frequency of each spin about the  $\hat{z}$  axis. We assume  $N$  is fixed and the relaxation negligible [6]. Suppose all the spins are initially prepared at time  $t=0$  in the  $|-\frac{1}{2}\rangle$  state. In this case, the spin wave function for the ensemble can be represented by the Dicke state  $|J, M = -J\rangle$  where  $\mathbf{J} = \sum_i \mathbf{S}_i$  ( $\mathbf{S}_i$  is the spin of the  $i$ th particle),  $J = N/2$ , and  $M = \langle J_z \rangle$  [8]. For this state,  $\Delta J_x(t=0) = \Delta J_y(0)$

$= (J/2)^{1/2}$  and  $\Delta J_z(0) = 0$ , where  $\Delta A$  is the square root of the variance of operator  $A$ ,  $(\Delta A)^2 \equiv \langle A^2 \rangle - \langle A \rangle^2$ . In Fig. 1(a), the lower cone shows, pictorially, a more general initial state with  $\langle \mathbf{J} \rangle = \langle J_z \rangle \hat{z}$ ,  $\langle J_z \rangle < 0$ , and  $\Delta J_y < \Delta J_x$  in a reference frame which corotates about the  $\hat{z}$  axis with the spin precession frequency  $\omega_0$ . In this rotating frame, the field  $B_0$  transforms to zero and the state remains stationary [9].

We perform spectroscopy (here, essentially nuclear magnetic or electron spin resonance) using the Ramsey method of separated oscillatory fields [9]. Description in terms of the Ramsey method leads to mathematical simplification, but the improvement of SNR is quite general. The Ramsey method consists of applying an oscillat-

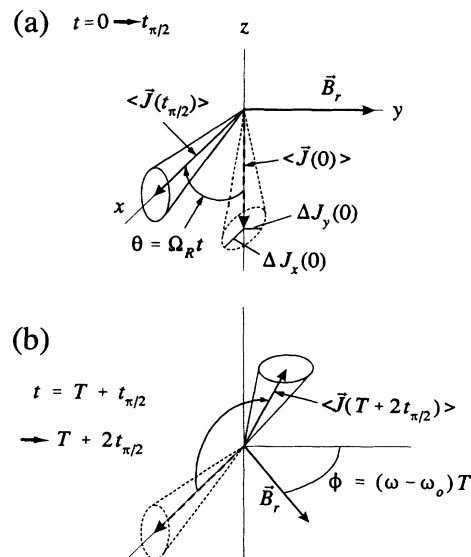


FIG. 1. Pictorial representation of the Ramsey method of spectroscopy in a frame corotating with the spin precession frequency. In (a), the lower cone represents the initial spin state for the Ramsey spectroscopy described in the text. After application of the second  $\pi/2$  pulse [upper cone in (b)], we detect the number of spins in the  $|+\frac{1}{2}\rangle$  state.

ing classical field  $\hat{y}B_1(t)\cos(\omega t)$  where  $\omega \approx \omega_0$ . This field can be decomposed into two fields which rotate about the  $\hat{z}$  axis; one of these fields  $\mathbf{B}_r$  ( $|\mathbf{B}_r| = B_1/2$ ) rotates in the same direction as the spin precession and the other can usually be neglected as is assumed here [9]. We will assume that  $B_1$  is nonzero and constant with value  $B_{10}$  from time  $t=0$  to  $t=t_{\pi/2}$  such that  $\Omega_R t_{\pi/2} = \pi/2$  and  $|\omega - \omega_0|t_{\pi/2} \ll 1$ , where  $\Omega_R \equiv \mu_0 B_{10}/(2\hbar)$  is the Rabi frequency. At  $t=0$  in the rotating frame,  $\mathbf{B}_r$  lies along the  $y$  axis as shown in Fig. 1(a). The lower cone of Fig. 1(a) rotates about  $\mathbf{B}_r$  and preserves its size and shape so that after time  $t_{\pi/2}$ , the cone lies along the  $x$  axis. At  $t=t_{\pi/2}$ ,  $B_1$  is reduced to zero and the cone remains stationary along  $x$ . After a time  $T$  ( $\gg t_{\pi/2}$ ),  $B_1$  is made equal to  $B_{10}$  again for a time  $t_{\pi/2}$ . If  $\omega \neq \omega_0$ ,  $\mathbf{B}_r$  is now at an angle  $\phi = (\omega - \omega_0)T$  with respect to its original direction and the cone precesses about  $\mathbf{B}_r$  as shown in Fig. 1(b). At time  $t_f \equiv T + 2t_{\pi/2}$ , the number of particles in the  $|+\frac{1}{2}\rangle$  state is measured. The expected number of particles  $\langle N_+ \rangle$  in the  $|+\frac{1}{2}\rangle$  state, where  $N_+ = J + J_z$ , is given by the Ramsey resonance curve

$$\langle N_+ \rangle = J - \langle J_z(0) \rangle \cos(\omega - \omega_0)T. \quad (1)$$

The discussions of correlated-particle interferometer experiments [3,4] parallel the description of the Ramsey method.

With  $N$  constant we eliminate noise due to fluctuations in  $N$  as found in an atomic beam experiment. However, successive measurements of  $N_+$  for a particular value of  $\omega - \omega_0$  fluctuate by  $\Delta N_+$  due to fluctuations in the number of spins measured to be in the  $|+\frac{1}{2}\rangle$  state. This produces apparent fluctuations in the center frequency of the Ramsey curve, given by  $|\Delta\omega| \equiv \Delta N_+ / |\partial \langle N_+ \rangle / \partial \omega| = \Delta J_z(t_f) / |\partial \langle J_z(t_f) \rangle / \partial \omega|$ . It is these fluctuations we desire to make small. If the initial state is the  $|J, -J\rangle$  Dicke state, we find  $|\Delta\omega|_{DS} = 1/TN^{1/2}$ . The somewhat surprising independence of  $|\Delta\omega|_{DS}$  on  $\omega$  occurs because the quantum noise is proportional to the slope of  $\langle N_+ \rangle$ . In the presence of added noise it is advantageous to operate at frequencies  $\omega$  where  $|\partial \langle N_+ \rangle / \partial \omega|$  is maximized in which case  $\langle N_+ \rangle = J = N/2$ . This condition on  $\omega$  is assumed in the remaining discussion. The frequency noise  $|\Delta\omega|_{DS}$  has been observed in experiments on trapped ions [6]. By use of suitably prepared ‘‘squeezed’’ initial spin states, which show correlations between the individual spin- $\frac{1}{2}$  particles, it should be possible to achieve  $|\Delta\omega| < |\Delta\omega|_{DS}$ . Hence, we define squeezing in Ramsey spectroscopy as  $\xi_R < 1$  where

$$\xi_R \equiv |\Delta\omega| / |\Delta\omega|_{DS} = [2J]^{1/2} \Delta J_y(0) / |\langle J_z(0) \rangle|. \quad (2)$$

Spin squeezing has been defined in other ways. From the commutation relations for angular momentum, the corresponding uncertainty relation can be given by  $\Delta J_x \Delta J_y \geq |\langle J_z \rangle / 2|$  and the expressions which follow from cyclic permutations. From this expression, it is natural to define squeezed states [10] as those where  $\Delta J_i / |\langle J_j \rangle / 2|^{1/2} < 1$  for some  $i \neq j \in \{x, y, z\}$ . This squeezing is present in Bloch states [8], which are obtained by rotating the  $|J, \pm J\rangle$  Dicke states [10]. For example, during the first Ramsey pulse [Fig. 1(a)],  $\Delta J_x / |\langle J_z \rangle / 2|^{1/2} = (\cos\theta)^{1/2}$

$\leq 1$ . However, these rotated  $|J, \pm J\rangle$  Dicke states have  $\xi_R = 1$  and do not improve the spectroscopy we describe here. Spin squeezing might also be defined as follows [4]: Let  $\Delta J_{\perp}$  denote the smallest uncertainty of a spin component perpendicular to mean spin vector  $\langle \mathbf{J} \rangle$ . The spin is squeezed if  $\xi_{spin} < 1$  where

$$\xi_{spin} \equiv \Delta J_{\perp} / |\langle \mathbf{J} \rangle / 2|^{1/2}. \quad (3)$$

For the Bloch states,  $\xi_{spin} = 1$ . A squeezed spin state with  $\xi_{spin} < 1$  can be rotated so that  $\langle \mathbf{J} \rangle = \langle J_z \rangle \hat{z}$  and  $\Delta J_{\perp} = \Delta J_y$  and can then be used in Ramsey spectroscopy with  $\xi_R = (J / |\langle \mathbf{J} \rangle|)^{1/2} \xi_{spin}$ .

We now examine possible methods to prepare the initial squeezed spin state. For a single particle ( $J = \frac{1}{2}$ ),  $\xi_{spin}, \xi_R \geq 1$ . For  $J \geq 1$ , states with  $\xi_{spin}, \xi_R \leq 1$  can be prepared using an interaction proportional to the square of angular momentum operators [4]. Because we were unable to find a physical interaction of this type for our problem, we have investigated an interaction of the form  $-\mu_0 J_x z B' \cos(\omega_m t_p)$ . Here,  $B'$  has units of a field gradient ( $\partial B_x / \partial z$ ),  $z = z_0(a^\dagger + a)$  is the quantum-mechanical amplitude of a harmonic oscillator of frequency  $\omega_z$ ,  $z_0$  is its zero-point amplitude [ $z_0 = \Delta z$  (coherent state)],  $a^\dagger$  and  $a$  are raising and lowering operators,  $t_p$  is the time during the preparation period, and we assume  $\omega_z \neq \omega_0$ . For a suitable choice of  $\omega_m$ , this interaction gives rise to a resonant coupling between  $\mathbf{J}$  and  $z$ . In the interaction picture and in the rotating-wave approximation it has the form

$$H_1 = -\hbar \Omega (J_+ a + J_- a^\dagger), \quad (4a)$$

for  $\omega_m = \omega_0 - \omega_z$  and

$$H_2 = -\hbar \Omega (J_+ a^\dagger + J_- a), \quad (4b)$$

for  $\omega_m = \omega_0 + \omega_z$  where  $J_+ \equiv J_x + iJ_y$ ,  $J_- = (J_+)^\dagger$ , and  $\Omega \equiv \mu_0 B' z_0 / 4\hbar$ .

The Heisenberg equations of motion for  $a$  and  $J_-$  can be solved analytically for Eqs. (4) when  $dJ_z/dt_p = 0$ . If the initial spin state is the  $|J, -J\rangle$  Dicke state, this amounts to the small-angle approximation  $J_z = -J\mathbf{I}$  where  $\mathbf{I}$  is the identity operator [11]. For the interaction  $H_1$ , we find

$$\xi_{spin}^2 = \xi_R^2 = \cos^2 \Omega_N t_p + \xi_z^2(t_p=0) \sin^2 \Omega_N t_p, \quad (5)$$

where  $\Omega_N^2 \equiv N \Omega^2$  and  $\xi_z(t_p=0) \equiv \Delta z(t_p=0) / \Delta z$  (coherent state) is the initial squeezing of the harmonic oscillator. Therefore if  $\xi_z(t_p=0) < 1$ , this squeezing is transferred to the spins (in a time  $\pi/2\Omega_N$ ) like wave-function exchange between coupled harmonic oscillators [12]. Because of the limited validity of the small-angle approximation, we have numerically evaluated Schrödinger’s equation to compute  $\xi_{spin}$  and  $\xi_R$  for two special cases shown in Figs. 2 and 3 assuming an initial  $|J, -J\rangle$  Dicke state. For our implementation of squeezing [Eqs. (4)], these figures show the improvement in SNR which can be obtained over the quantum limit obtained with uncorrelated input states.

This squeezing can, in principle, be generated by the interaction of an ensemble of atoms with a single-mode cavity field [1,11,13]. It can be viewed as the complement of

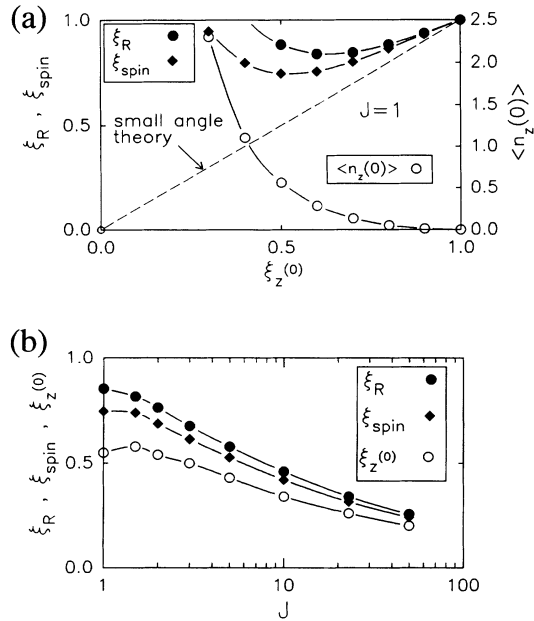


FIG. 2. Squeezed spin state preparation assuming the interaction of Eq. (4a) and assuming the harmonic oscillator is initially in the squeezed vacuum state where  $\langle z(t_p=0) \rangle = \langle \dot{z}(t_p=0) \rangle = 0$ ,  $\xi_z(t_p=0) < 1$ . In (a), we plot the first minima (as a function of preparation time  $t_p$ ) of  $\xi_{\text{spin}}$ , the resulting value of  $\xi_R$ , and the corresponding value of  $\langle n_z(t_p=0) \rangle$  ( $\equiv \langle a^\dagger(0)a(0) \rangle$ ) vs  $\xi_z(t_p=0)$  for the case  $J=1$ . As expected, the small-angle theory is valid for  $\xi_z(t_p=0) \rightarrow 1$ , ( $\langle n_z(0) \rangle \rightarrow 0$ ). In (b), we show the first minimum values (as a function of preparation time  $t_p$ ) of  $\xi_{\text{spin}}$  which have also been minimized with respect to  $\xi_z(t_p=0)$ , the resulting value of  $\xi_R$ , and the corresponding values of  $\xi_z(t_p=0)$  vs  $J$ .

the squeezing of a single-mode radiation field by coupling to an ensemble of spins [11,13]. It might also be generated in an ensemble of trapped ions where the harmonic oscillator corresponds to the center-of-mass (c.m.) oscillation in a particular ( $z$ ) direction. For simplicity, we consider  $N$  ions trapped along the axis of a linear rf trap [14]. The two-level system is the Zeeman doublet for an unpaired electron on each ion.  $B'$  is a gradient field  $\partial B_x / \partial z$  generated by current  $I_y \hat{y}$  in two wires (which could double as trap electrodes) situated as the positions  $z = \pm z_T$  relative to the ions. We assume the Coulomb interaction between the ions is large enough that the frequencies of the ions' internal modes are significantly different from  $\omega_z$ . The c.m. mode is first laser cooled to the zero-point state [15]. A "squeezed vacuum" state of the c.m. mode (appropriate for Fig. 2) could be obtained by suddenly changing the ions' well depth or driving the  $z$  oscillation parametrically at  $2\omega_z$  [12]. A coherent state of nonzero amplitude (appropriate for Fig. 3) could be created by suddenly changing the center position of the well or driving the zero-point state with a classical resonant excitation [12]. When  $\mu_0$  is equal to two Bohr magnetons, we find  $\Omega/2\pi \approx 2I_y z_T^{-2} \times (M\omega_z/2\pi)^{-1/2}$  where  $I_y$ ,  $z_T$ ,  $M$ , and  $\omega_z/2\pi$  are expressed in amperes, centimeters, atomic mass units, and megahertz, respectively. For  $I_y = 0.1$  A,  $z_T = 0.01$  cm,  $M = 24$  u ( $^{24}\text{Mg}^+$ ), and  $\omega_z/2\pi = 1$  MHz, we

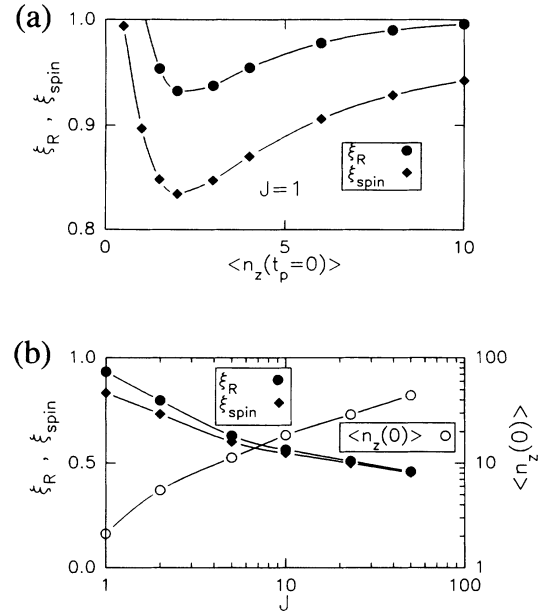


FIG. 3. Squeezed spin state preparation assuming the interaction of Eq. (4b) and assuming the harmonic oscillator is initially prepared in a coherent state [ $\xi_z(t_p=0) = 1$ ] with  $\langle z(t_p=0) \rangle \neq 0$  and  $\langle \dot{z}(t_p=0) \rangle = 0$ . In (a) we show the first minima of  $\xi_{\text{spin}}$  (as a function of  $t_p$ ), and the resulting values of  $\xi_R$  vs  $\langle n_z(t_p=0) \rangle$  for  $J=1$ . In (b), we show the first minimum values (as a function of preparation time  $t_p$ ) of  $\xi_{\text{spin}}$  which have also been minimized with respect to  $\langle n_z(t_p=0) \rangle$ , the resulting values of  $\xi_R$ , and the corresponding values of  $\xi_z(t_p=0)$  vs  $J$ . We find the first minima of  $\xi_{\text{spin}}$  (as a function of  $t_p$ ) occur after  $\langle J_z \rangle$  has reached its maximum value and the spin vector is moving back toward the negative  $z$  axis. In this case the squeezed state must be rotated back to the negative  $z$  axis before applying the Ramsey fields.

find  $\Omega/2\pi \approx 400$  Hz. The interesting cooling method proposed by Harde [16] uses the same parametric coupling described here. For Ramsey spectroscopy, the relative phase between the squeezing and Ramsey fields must be reproducible from measurement to measurement.

When  $N$  is even, there exist correlated states where  $\xi_R = (N/2 + 1)^{-1/2}$  (see also Ref. [3]) in which case the uncertainty in the measurement of  $\omega_0$  will be approximately proportional to  $N^{-1}$  rather than the usual  $N^{-1/2}$ . For example, for  $N=2$  ( $J=1$ ), the  $|J=1, M=0\rangle$  Dicke state is maximally squeezed ( $\langle J_z \rangle = 0$  for this state, but  $\xi_R \rightarrow 2^{-1/2}$  as  $|\langle J_z \rangle| \rightarrow 0$ ). It can be used in Ramsey spectroscopy by detecting  $J_z^2$  rather than  $J_z$  [17]. For this state the correlations between spins is clear. If one spin is measured to be in the  $+\frac{1}{2}$  ( $-\frac{1}{2}$ ) state, the other spin will be found in the  $-\frac{1}{2}$  ( $+\frac{1}{2}$ ) state. Starting from the  $|n_z=0\rangle |J=1, M=-1\rangle$  state, we can prepare this squeezed state by making  $\omega_z$  anharmonic to break the degeneracy of the  $\delta n_z = 1$  transitions. With a classical field we drive to the  $|1\rangle |1, -1\rangle$  state and then turn the anharmonicity off. We then apply the interaction of Eq. (4a) to drive to the  $|0\rangle |1, 0\rangle$  squeezed state.

In summary, it should be possible to improve the signal-to-noise ratio in spectroscopic experiments which

detect the changes of state population over the case where uncorrelated particles are used. It might also be possible to demonstrate spin squeezing in individual atoms or ions with  $J \geq 1$  such as in the Zeeman sublevels of a particular hyperfine state [17]. The relatively large value of  $\Omega$  suggests that cavity-QED problems in the strong-coupling (weak relaxation) regime might be studied in the ion system. The correlated states described here could be used in Bell's inequality experiments. A possibility is the creation of multiparticle correlated states which strongly violate

Bell's inequality [18]. The individual particle states could be detected with nearly 100% efficiency [6].

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