

**Orientation-to-alignment conversion and spin squeezing**S. M. Rochester,<sup>1,2</sup> M. P. Ledbetter,<sup>2</sup> T. Zigdon,<sup>2</sup> A. D. Wilson-Gordon,<sup>3</sup> and D. Budker<sup>2,4,\*</sup><sup>1</sup>*Rochester Scientific, El Cerrito, California 94530-1757, USA*<sup>2</sup>*Department of Physics, University of California at Berkeley, Berkeley, California 94720-7300, USA*<sup>3</sup>*Department of Chemistry, Bar-Ilan University, Ramat Gan 52900, Israel*<sup>4</sup>*Nuclear Science Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA*

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The relationship between orientation-to-alignment conversion (a form of atomic polarization evolution induced by an electric field) and the phenomenon of spin squeezing is demonstrated. A “stretched” state of an atom or molecule with maximum angular-momentum projection along the quantization axis possesses orientation and is a quantum-mechanical minimum-uncertainty state, where the product of the equal uncertainties of the angular-momentum projections on two orthogonal directions transverse to the quantization axis is the minimum allowed by the uncertainty relation. Application of an electric field for a short time induces orientation-to-alignment conversion and produces a spin-squeezed state, in which the quantum state essentially remains a minimum-uncertainty state, but the uncertainties of the angular-momentum projections on the orthogonal directions are unequal. This property can be visualized using the angular-momentum probability surfaces, where the radius of the surface is given by the probability of measuring the maximum angular-momentum projection in that direction. Brief remarks are also given concerning collective-spin squeezing and quantum nondemolition measurements.

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**I. INTRODUCTION**

Since the pioneering work of Kitagawa and Ueda [1], the concept of spin squeezing, or the redistribution of uncertainties from one spin component to another, has drawn significant attention [2]. Reducing the uncertainty in a particular spin component to be measured at the expense of others can, in principle, allow measurements at the fundamental Heisenberg limit of uncertainty, which scales as  $1/F$  for the relative uncertainty of a measurement of the projection of an effective angular momentum  $F$ , rather than at the standard quantum limit, which scales as  $1/\sqrt{F}$ .

One area in which spin squeezing is of practical interest is optical magnetometry, i.e., the idea of gaining sensitivity via spin squeezing is an attractive one. Unfortunately, this application is not as straightforward as it may seem and, in fact, there is no sensitivity gain in a rather broad class of situations [3]. Nevertheless, as has been shown recently, squeezing can lead to increase in bandwidth [4,5], and can increase sensitivity in cases involving nonexponential relaxation [6].

In this paper, we discuss two aspects of spin squeezing relevant to atomic magnetometry. The first concerns the relationship between spin squeezing and a type of polarization evolution known as alignment-to-orientation conversion (AOC), which occurs when an electric field is applied to a polarized atomic ensemble. Here, “orientation” refers to the rank-one atomic polarization moment having a preferred direction, and “alignment” to the rank-two polarization moment with a preferred axis but no preferred direction. Alignment-to-orientation conversion is an important mechanism for atomic magnetometry, occurring, for example, in nonlinear Faraday rotation [7]. It also occurs in other areas such as nuclear

quadrupole resonance (NQR) [8], and has been extensively studied for many years.

Here, we point out that there is a close relationship between AOC and spin squeezing: when an atom in a stretched state is placed in an orthogonal electric field, spin squeezing is caused as a result of, in this case, the inverse process of orientation-to-alignment conversion (OAC). We quantify the amount of squeezing that can be obtained, and illustrate the process using a polarization visualization technique. The electric field needed to produce the squeezing can be either dc or off-resonant ac. In fact, the latter has already been used to generate spin squeezing in the ground state of alkali-metal atoms (see Refs. [9,10]; this case is analyzed in Appendix C).

Note that there is an essential difference between the squeezing produced by AOC and that discussed in Ref. [1]: in the latter case, the squeezing is produced by an operator acting on the collective spin of the ensemble, while the electric-field Hamiltonian that induces AOC acts on the individual spin of each atom. From a practical standpoint, it is much more desirable to squeeze the collective spin, rather than the individual spins, because the effective angular momentum participating in the scaling discussed above can be made very large.

We then discuss a second aspect of spin squeezing that has been demonstrated in the context of optical magnetometry: collective spin squeezing via a quantum nondemolition (QND) interaction. Here, we remark on the origin of increased noise in the unobserved spin quadrature that ensures compliance with the uncertainty relations for orthogonal spin projections.

**II. SPIN SQUEEZING BY INTERACTION WITH THE ELECTRIC FIELD**

Measurements involving quantum systems are fundamentally limited by uncertainty relations derived from the commutation relations of quantum-mechanical operators. A textbook

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example is a state of a system (such as an atom or a molecule; we will henceforward refer to an atom) with total angular momentum  $F$  prepared in a “stretched” state with a fixed projection  $m$  on a chosen quantization axis ( $z$ ) such that  $|m| = F$ , i.e., the state  $|F, m = F\rangle$ . A state stretched along another direction can be written by rotating  $|F, F\rangle$  using the quantum-mechanical rotation operator. For the state  $|F, F\rangle_{\hat{x}}$  stretched along  $\hat{x}$ , this gives [1]

$$|F, F\rangle_{\hat{x}} = 2^{-F} \sum_{k=0}^{2F} \binom{2F}{k}^{\frac{1}{2}} |F, F - k\rangle, \quad (1)$$

where the binomial coefficients are given by

$$\binom{n}{k} = \frac{n!}{k!(n-k)!}. \quad (2)$$

If an experiment is performed that measures the  $x$  projection of  $|F, F\rangle_{\hat{x}}$ , the outcome is always the same ( $F_x = F\hbar$ ). On the other hand, measuring the projection on an orthogonal axis, say  $y$ , one measures zero on average,  $\langle F_y \rangle = 0$ , but each specific measurement can yield any result such that  $-F \leq F_y/\hbar \leq F$ , with a similar result for the projection on  $z$ . The uncertainty relation for the angular-momentum projections reads as

$$\Delta F_y \Delta F_z \geq \frac{\hbar^2}{2} F, \quad (3)$$

where the uncertainty in  $\Delta F_y$  is defined according to

$$\Delta F_y = \sqrt{\langle F_y^2 \rangle - \langle F_y \rangle^2}, \quad (4)$$

and similarly for  $\Delta F_z$ . Explicitly calculating the uncertainties  $\Delta F_y$  and  $\Delta F_z$  using the appropriate quantum-mechanical operators, we find that, as expected from symmetry, these uncertainties are equal, and that their values realize the equality in the expression (3), which means that the stretched state is a *minimum-uncertainty state*.

One way of visualizing the state is using angular-momentum probability surfaces (AMPS) [11–13]. The radius of the surface in a given direction is proportional to the probability to measure the maximum angular-momentum projection ( $=F$ ) in that direction. This corresponds to the quasiprobability distribution plotted in Ref. [1] and is the analog, for spin states, of the  $Q$  function of quantum optics. The AMPS for  $|F, F\rangle_{\hat{x}}$  is shown in the upper-left plot of Fig. 1. It is clearly pointing in the  $x$  direction, and is symmetric about  $\hat{x}$ .

We next consider the evolution of the stretched state under the influence of an electric field. Let us assume that the field is applied along the quantization axis  $z$ . For simplicity, we assume that the electric field is either dc or linearly polarized off-resonant ac (the two cases are essentially equivalent [9,10]). The Hamiltonian of the system in the presence of the electric field is

$$H_E = -\frac{1}{2}\alpha_0 E^2 - \frac{1}{2}\alpha_2 E_z^2 \frac{3F_z^2 - \mathbf{F}^2}{\hbar^2 F(2F-1)}, \quad (5)$$

where  $\alpha_0$  is the scalar polarizability of the state, and  $\alpha_2$  is the tensor polarizability. (The vector polarizability does not contribute to the Hamiltonian, as we are considering a linearly

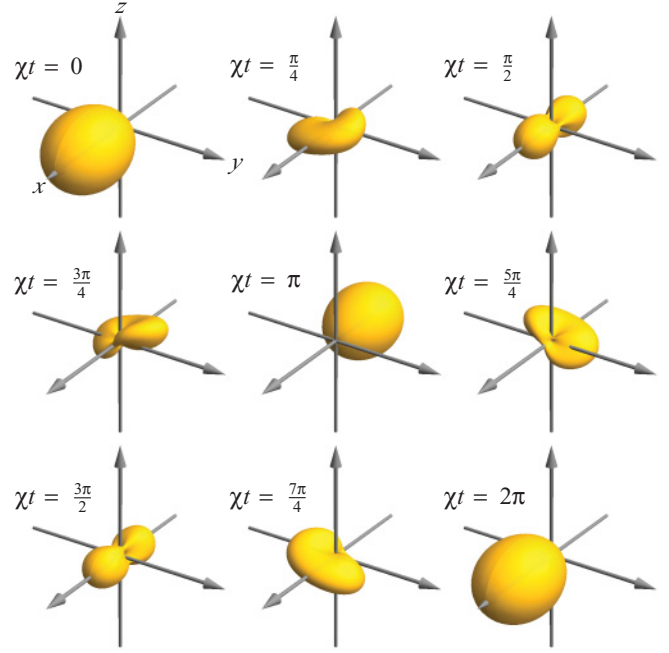


FIG. 1. (Color online) Angular-momentum probability surfaces (see text) showing an  $F = 2$  atomic state initially stretched along  $\hat{x}$  and evolving in the presence of an electric field along  $\hat{z}$ . A complete cycle of orientation-to-alignment conversion is shown.

polarized electric field.) We neglect the scalar polarizability term and the part of the tensor polarizability that is independent of  $F_z$  since they result only in a common shift of the Zeeman sublevels of the ground or excited hyperfine states, and so do not lead to evolution of the Zeeman polarization. We therefore consider the following Hamiltonian for each spin in the ensemble:

$$H_E = \chi F_z^2 / \hbar, \quad (6)$$

where we have defined

$$\chi = -\frac{3}{2\hbar F(2F-1)} \alpha_2 E_z^2. \quad (7)$$

This Hamiltonian has the same form as the one presented by Kitagawa and Ueda [1] for “one-axis twisting.” However, there is an essential difference in that  $F_z$  in Ref. [1] refers to the collective spin of an ensemble of particles, whereas we are considering a single atom or, equivalently, an ensemble of uncorrelated atoms. The Hamiltonian  $H_E$  generates the unitary transformation

$$U(t) = e^{-i\chi t F_z^2 / \hbar^2}. \quad (8)$$

The evolution of  $|F, F\rangle_{\hat{x}}$  in the Schrödinger picture according to the evolution operator  $U(t)$  is illustrated in Fig. 1 for the case of  $F = 2$ . The state oscillates between having a preferred direction, indicating that it possesses orientation (for example, at  $\chi t = 0$ ), and having a preferred axis, indicating that it possesses alignment (for example, at  $\chi t = \pi/2$ ); we therefore refer to this kind of evolution as OAC or AOC.

We note that the evolution of polarized atomic and molecular states in the presence of an electric field has been extensively studied in the literature (see, for example, [11,13,14] and references therein). However, it has not been broadly recognized that spin squeezing is naturally associated with this evolution. In fact, we will now see that, as the process of OAC begins, the uncertainty of a spin measurement along a particular axis perpendicular to  $x$  is reduced, while that along the orthogonal axis is increased, so that the state, to first order, remains a minimum-uncertainty state. To show this, we explicitly calculate the uncertainty in the measurement of  $F_y$  as a function of time; a rotation about the  $x$  axis allows us to find the axis of optimal squeezing.

Following Ref. [1], we analyze the means and variances of the operators  $F_x(t), F_y(t)$  in the Heisenberg picture using the raising and lowering operators

$$F_{\pm}(t) = F_x(t) \pm iF_y(t), \quad (9)$$

which evolve in the Heisenberg picture as

$$\begin{aligned} F_+(t) &= U(t)^\dagger F_+(0) U(t) \\ &= e^{i\chi t F_z^2/\hbar^2} F_+(0) e^{-i\chi t F_z^2/\hbar^2} \\ &= F_+(0) e^{2i\chi t(F_z/\hbar + \frac{1}{2})}, \end{aligned} \quad (10)$$

$$F_-(t) = [F_+(t)]^\dagger = e^{-2i\chi t(F_z/\hbar + \frac{1}{2})} F_-(0), \quad (11)$$

where  $F_-(0) = F_+^\dagger(0)$ . The details of the derivation of Eqs. (10) and (11) are presented in Appendix A. The components of  $F_x(t), F_y(t)$  are now given by

$$\begin{aligned} F_x(t) &= \frac{1}{2}[F_+(t) + F_-(t)] \\ &= \frac{1}{2}[F_+(0)e^{2i\chi t(F_z/\hbar + \frac{1}{2})} + e^{-2i\chi t(F_z/\hbar + \frac{1}{2})} F_-(0)], \end{aligned} \quad (12)$$

$$\begin{aligned} F_y(t) &= \frac{1}{2i}[F_+(t) - F_-(t)] \\ &= \frac{1}{2i}[F_+(0)e^{2i\chi t(F_z/\hbar + \frac{1}{2})} - e^{-2i\chi t(F_z/\hbar + \frac{1}{2})} F_-(0)]. \end{aligned} \quad (13)$$

In Appendix B, we present a detailed calculation of the expectation value  $\langle F_x \rangle$  [Eq. (15a)]. The means and variances of the other components of the angular momentum can be calculated in a similar manner. In order to analyze the mean values and variances of the spin projections along all directions transverse to  $x$ , it is convenient to write  $F_y(t)$  in a coordinate frame obtained from the original one by rotating about  $\hat{x}$  by an angle  $\nu$ , according to the unitary transformation

$$F_{y,\nu} = e^{i\nu F_x(t)/\hbar} F_y e^{-i\nu F_x(t)/\hbar}. \quad (14)$$

The expectation values of the components of the angular momentum become

$$\langle F_x \rangle = \hbar F (\cos \chi t)^{2F-1}, \quad (15a)$$

$$\langle F_{y,\nu} \rangle = 0, \quad (15b)$$

$$\langle F_{z,\nu} \rangle = 0, \quad (15c)$$

while the variances are given by

$$(\Delta F_x)^2 = \hbar^2 \frac{F}{2} \left[ 2F(1 - \cos^2(2F-1)\chi t) - \left(F - \frac{1}{2}\right) A \right], \quad (16a)$$

$$\begin{aligned} (\Delta F_{y,\nu})^2 &= \hbar^2 \frac{F}{2} \left\{ 1 + \frac{1}{2} \left(F - \frac{1}{2}\right) \right. \\ &\quad \left. \times [A + \sqrt{A^2 + B^2} \cos(2\nu + 2\delta)] \right\}, \end{aligned} \quad (16b)$$

$$\begin{aligned} (\Delta F_{z,\nu})^2 &= \hbar^2 \frac{F}{2} \left\{ 1 + \frac{1}{2} \left(F - \frac{1}{2}\right) \right. \\ &\quad \left. \times [A - \sqrt{A^2 + B^2} \cos(2\nu + 2\delta)] \right\}, \end{aligned} \quad (16c)$$

where  $A = 1 - \cos^{2F-2} 2\chi t$ ,  $B = 4 \sin \chi t \cos^{2F-2} \chi t$ , and  $\delta = \frac{1}{2} \arctan \frac{B}{A}$ . According to Eqs. (16),  $\Delta F_{y,\nu}$  is minimized and  $\Delta F_{z,\nu}$  is maximized when  $\cos(2\nu + 2\delta) = -1$ , i.e.,  $\nu = \frac{\pi}{2} - \delta$ . This determines the axis with the best squeezing at a given time.

Considering the evolution shortly after the application of the electric field, we find that, to first order, the state remains a minimum-uncertainty state. However, the uncertainties in  $F_y$  and  $F_z$  are no longer equal [Eqs. (16)]; therefore, we have generated a *spin-squeezed state* (SSS). This is indicated by AMPS plots observed along the  $x$  axis (Fig. 2). The utility of SSS is that, in principle, they allow an improved sensitivity in certain appropriately designed measurements. [The fundamental quantum-mechanical limit on the uncertainty of a measurement of  $F_x$  is not  $\hbar\sqrt{F}/2$  as implied by the uncertainty relation (3) for the case of  $\Delta F_x = \Delta F_y$  (the standard quantum limit), but the Heisenberg limit Bollinger [15,16] of  $\hbar/\sqrt{2}$ .]

Wineland *et al.* [17] defined a squeezing parameter to indicate sensitivity to rotations of the angular-momentum states. Considering squeezing along the  $y$  axis rotated by an angle  $\nu$  about  $\hat{x}$ , the squeezing parameter  $\xi_R$  is the uncertainty in the rotation of the spin,  $\Delta F_{y,\nu}/|\langle F_x \rangle|$ , normalized to the uncertainty  $1/\sqrt{2F}$  expected in the standard quantum limit, i.e., the uncertainty obtained using a stretched state:

$$\xi_R = \sqrt{2F} \frac{\Delta F_{y,\nu}}{|\langle F_x \rangle|}. \quad (17)$$

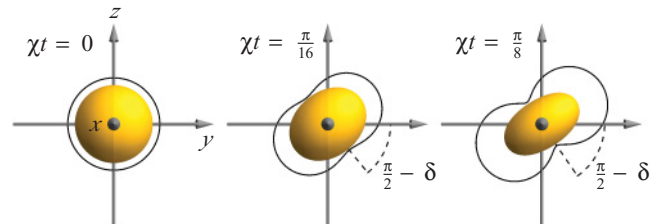


FIG. 2. (Color online) The initial part of the evolution shown in Fig. 1 as viewed from the  $x$  direction, showing the process of squeezing. The solid line is a polar plot of  $\Delta F_{y,\nu}$  as a function of azimuth, with the angle  $\nu = \pi/2 - \delta$  of the minimum-uncertainty axis indicated by the dashed line.

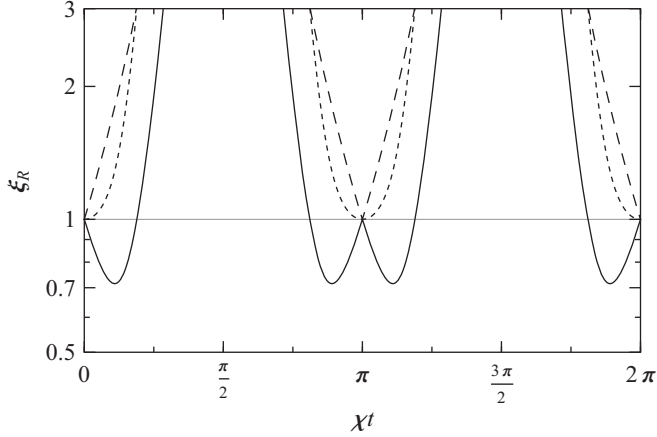


FIG. 3. The squeezing parameter  $\xi_R$  for  $F = 2$  as a function of time. The solid curve shows  $\xi_R$  for the most strongly squeezed axis ( $\nu = \frac{\pi}{2} - \delta$ ), while the dashed curve gives  $\xi_R$  for the orthogonal “antisqueezing” axis. A value  $\xi_R < 1$  (below the gray line) indicates a squeezed state. The dotted curve shows the product of the squeezing parameters for the two orthogonal axes, with a value of 1 indicating a minimum-uncertainty state.

Substituting from Eqs. (15a) and (16b), we find the squeezing along the minimum-uncertainty axis to be

$$\xi_R = \frac{\sqrt{1 + \frac{1}{2}(F - \frac{1}{2})(A - \sqrt{A^2 + B^2})}}{|(\cos \chi t)^{2F-1}|}. \quad (18)$$

In Fig. 3, we plot the squeezing parameter for the minimum-uncertainty axis as a function of time for  $F = 2$ . We observe that  $\xi_R$  initially decreases below unity, indicating a squeezed state, but subsequently tends toward infinity at  $t = \frac{\pi}{2} + k\pi$ , with integer  $k$ , because  $|\langle F_x \rangle|$  tends to zero. From Eq. (15a), we see that this is true for any value of  $F$ . If the electric field inducing OAC is turned off at the time corresponding to the minimum value of  $\xi_R$ , we obtain squeezing along a fixed axis, the direction of which can be found from Fig. 4. (Note that in some cases it is desirable to have the squeezing axis

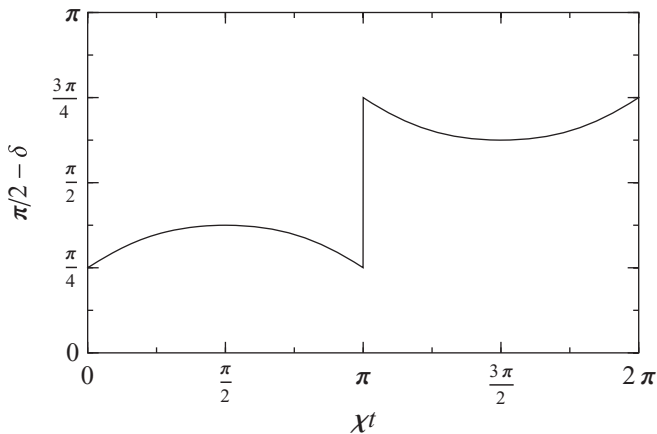


FIG. 4. The angle  $\nu = \frac{\pi}{2} - \delta$  of the minimum uncertainty axis as a function of time for a state with  $F = 2$ . The  $\pi/2$  shift halfway through the cycle corresponds to the mirror symmetry seen between the first and second halves of the cycle in Fig. 1.

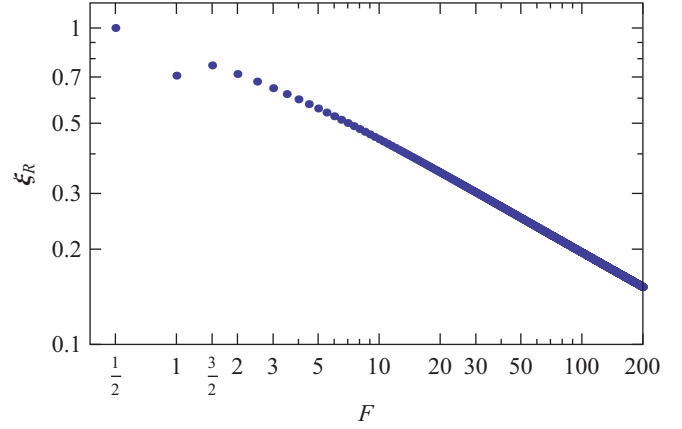


FIG. 5. (Color online) Logarithmic plot of the minimum value of  $\xi_R$  with respect to  $t$  as a function of  $F$  when  $\nu = \frac{\pi}{2} - \delta$ . The dependence quickly approaches a power law ( $\xi_R \propto F^{-1/3}$ ). Note that the case of  $F = 1$  has an apparently anomalously small value of  $\xi_R$ . This is a special case in which the optimum squeezing parameter is achieved as  $\chi t$  approaches  $\pi/2$ , when the spin projection  $\langle F_x \rangle$  goes to zero.

rotate in time; this could be accomplished here by subsequent application of a magnetic field.)

In Fig. 5, we plot the minimum of  $\xi_R$  with respect to  $t$  as a function of  $F$ . This plot differs from the corresponding plot in Ref. [1] because we use a different definition of the squeezing parameter. To find the asymptotic behavior of  $\xi_R$  for large  $F$ , we note that as  $F$  increases, the time  $\chi t$  at which the squeezing is minimized decreases faster than  $1/\sqrt{F}$ , but slower than  $1/F$ . Thus, for  $F \gg 1$ , we can assume that, near the minimum, the parameters  $\gamma = 1/(\chi t F)$  and  $\beta = \chi^2 t^2 F$  are both small. Writing the square of the squeezing parameter (18) in terms of  $\gamma$  and  $\beta$ , we expand to second order and find

$$\xi_R^2 \approx \frac{2\beta^2}{3} + \frac{\gamma^2}{4}. \quad (19)$$

Substituting back for  $F$  and  $t$ , we minimize with respect to  $t$  and find that at the time

$$\chi t_{\min} \approx \frac{3^{1/6}}{2^{2/3}} F^{-2/3}, \quad (20)$$

the minimum squeezing parameter is given by

$$\xi_R^{\min} \approx \frac{3^{1/3}}{2^{5/6}} F^{-1/3}, \quad (21)$$

equivalent to the result found in Ref. [18]. Relatively large values of  $F$  available for single atoms are  $F = 4$  in the ground state of Cs (see Appendix C for a calculation of squeezing in this system) and  $F = 12.5$  in a metastable state of Dy [19]. Even higher values of  $F$  are attainable in Rydberg atoms and in molecules with large rotational excitation. Very large effective values of  $F$  can be achieved in a somewhat different situation in which squeezing is done on a correlated ensemble, as discussed below.

The polarization evolution due to the Hamiltonian (6) is termed “single-axis twisting” by Kitagawa and Ueda [1] because the effect on the angular-momentum probability distribution can be visualized as resulting from a twisting



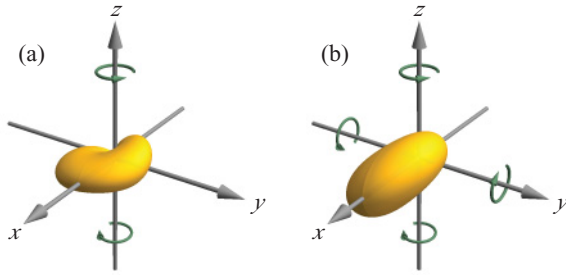


FIG. 6. (Color online) Comparison of AMPS for the (a) “single-axis twisting” and (b) “two-axis countertwisting” Hamiltonians at times equal to  $1/8$  of the respective quantum-beat periods for an  $F = 2$  state initially stretched along  $\hat{x}$ . Twisting about the  $z$  axis introduces “swirliness” in (a); simultaneous twisting about the  $y$  axis (b) cancels this effect.

motion about the  $z$  axis [Fig. 6(a)]. For the purposes of generating squeezed states, this type of evolution has some drawbacks resulting from the asymmetry of the evolution with respect to the  $z$  and  $y$  axes. First, as we have seen in Fig. 4, the optimal squeezing axis changes as a function of time. Second, the distortion in the probability distribution introduced by the twisting motion, described as “swirliness” in Ref. [1], limits the maximum squeezing that can be obtained. These effects can be obviated by creating a more symmetric Hamiltonian in which twisting is performed about two orthogonal axes (“two-axis countertwisting”), as shown in Fig. 6(b). Here, we have plotted the effect of a Hamiltonian of the form  $H = \chi(F_z^2 - F_y^2)/\hbar$ . Interaction described by such a Hamiltonian can be achieved by the use of two incoherent fields, such as a light field along with a magnetic field inducing the nonlinear Zeeman effect [10], two light fields of different frequencies, or a light field and a static electric field (see Ref. [2] and references therein). This removes the swirliness, fixes the squeezing axis, and allows the maximum amount of squeezing to be attained. We do not further consider the two-axis Hamiltonian here, as the simpler Hamiltonian (6) illustrates the principle under discussion.

The value for spin squeezing found here differs from that reported by Kitagawa and Ueda [1] because we use a form of the squeezing parameter, normalized to the expectation value of  $F_x$ , that is appropriate for measurements of rotation. There is a second, more fundamental difference between the two results, however, stemming from the definition of the interaction Hamiltonian. While the interaction Hamiltonian of Kitagawa and Ueda [1] relates to an ensemble with quantum correlations among the individual spins [20], our  $H_E$  (6) operates on each individual spin in the ensemble without creating correlation. This difference shows up clearly in the case in which all the atoms are in a  $F = 1/2$  state. For an individual  $F = 1/2$  atom, there is no differential shift induced by  $H_E$ , and, in any case, the only possible polarized state is a stretched state (the highest-rank polarization possible is orientation). Thus, no squeezing is possible in this case. (For the common experimental case of alkali-metal atoms, with electronic spin  $J = 1/2$ , the additional nuclear spin, resulting in higher total angular momentum, allows spin squeezing [10].) However, for  $N$  spin- $1/2$  atoms in the case in which

the Hamiltonian acts on the collective spin  $F_z^{(\text{tot})} = \sum_i F_z^{(i)}$ , the correlation between the particles can result in squeezing as for a fictitious particle with  $F = N/2$ . This means that very large effective values of  $F$  can be obtained, which would not be feasible for an uncorrelated ensemble for which  $F$  is the angular momentum of each polarized atom. Some brief remarks on methods of producing squeezing of a correlated ensemble are given in the next section.

### III. REMARKS ON ENSEMBLE SQUEEZING

Various techniques have been introduced in recent years to achieve collective-spin squeezing in atomic ensembles. These include methods based on collective interactions in Bose-Einstein condensates (see Ref. [21] and references therein), employing collective interactions between atoms via an optical cavity (see Ref. [22] and references therein), or methods based on generating squeezing by performing so-called QND measurements on an ensemble [23–26]. The latter technique has attracted particular attention in the context of atomic magnetometry (see Ref. [27] and references therein). We note here that spin squeezing does not normally lead to significant improvement of an optimized magnetometer measuring quasistatic magnetic fields [3]; however, QND techniques can extend the sensitivity to ac magnetic fields [4,5], and can improve the sensitivity for cases involving nonexponential relaxation [6].

Let us briefly discuss quantum nondemolition measurements, which minimally perturb the spin component being measured [28–30]. For example, consider an ensemble of spin- $1/2$  atoms polarized in an unknown direction perpendicular to the  $y$  axis. In order to determine the angle between the atomic polarization direction and the  $x$  axis, linearly polarized probe light propagating along the  $x$  axis can be employed. The polarization of the probe light will be rotated due to circular birefringence depending on the projection of the atomic spin along  $\hat{x}$ . If the probe light is detuned from atomic resonance, the rate of atomic transitions is low, so that the atomic polarization is not destroyed. A continuous measurement produces a more and more precise measurement of the spin projection along the  $x$  axis, indicating that the interaction with the probe light is squeezing the atomic state. This means that the spin projection in the orthogonal direction must be becoming more uncertain in order to preserve the uncertainty relation. What is the mechanism for this antisqueezing? The linearly polarized probe light can be thought of as being composed of a superposition of left- and right-circularly polarized light, each of which produces ac Stark shifts that mimic the effect of a magnetic field directed along the  $x$  axis. In the absence of noise, these two fictitious magnetic fields nominally cancel, but the effect of polarization noise (resulting from photon shot noise) in the light beam is to produce an effective fluctuating magnetic field along  $\hat{x}$ . The resulting fluctuating spin precession causes a spread of the atomic state along the  $y$  axis, preserving the uncertainty relation. Note that this process becomes more complicated for states with  $F > 1/2$  due to the evolution of the internal degrees of freedom of the atoms, as well as the coherence between the atoms (see Ref. [27] and references therein). This is discussed in more detail in Ref. [31]; see also Ref. [32].

#### IV. CONCLUSION

We have shown that the process of orientation-to-alignment conversion, as induced by an electric field, is intimately connected with spin squeezing. Following Kitagawa and Ueda [1], we quantified the amount of squeezing obtained by this mechanism for a state of angular momentum  $F$ . While the squeezing is improved for higher angular momentum, the scalability of this approach is limited by the available values of  $F$  found in usable atomic and molecular states. Alternative methods involving correlated atomic ensembles can achieve much higher effective angular momenta; we briefly discussed one such approach, using a quantum nondemolition measurement.

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#### APPENDIX A: CALCULATION OF $F_+(t)$

In order to determine the expression for the evolution of the raising operator  $F_+(t)$  given in Eq. (10), we consider the matrix elements of  $F_+(t)$  between any two eigenstates  $|F, m\rangle$

of the Hamiltonian, so that

$$\begin{aligned} \langle F, m' | F_+(t) | F, m \rangle &= \langle F, m' | e^{i\chi t F_z^2/\hbar^2} F_+(0) e^{-i\chi t F_z^2/\hbar^2} | F, m \rangle \\ &= e^{i\chi t m^2} \langle F, m' | F_+(0) | F, m \rangle e^{-i\chi t m^2}. \end{aligned} \quad (\text{A1})$$

The only nonzero matrix elements of the raising operator are those for which  $m' = m + 1$ . For these matrix elements, we have

$$\begin{aligned} \langle F, m + 1 | F_+(t) | F, m \rangle &= e^{i\chi t (m+1)^2} \langle F, m + 1 | F_+(0) | F, m \rangle e^{-i\chi t m^2} \\ &= \langle F, m + 1 | F_+(0) e^{i2\chi t (m+\frac{1}{2})} | F, m \rangle. \end{aligned} \quad (\text{A2})$$

The energy eigenstates form a complete set, and so this equation can be written in the operator form given in Eq. (10).

#### APPENDIX B: CALCULATION OF $\langle F_x(t) \rangle$

Here, we derive the expectation value (15a) of the operator  $F_x(t)$  [Eq. (12)] given the initial state  $|F, F\rangle_{\hat{x}}$  [Eq. (1)]:

$$\begin{aligned} \langle F_x(t) \rangle &= \langle F, F |_{\hat{x}} \frac{1}{2} [F_+(0) e^{2i\chi t (F_z/\hbar + \frac{1}{2})} \\ &\quad + e^{-2i\chi t (F_z/\hbar + \frac{1}{2})} F_-(t)] | F, F \rangle_{\hat{x}} \\ &= \text{Re} [\langle F, F |_{\hat{x}} e^{-i2\chi t (F_z/\hbar + \frac{1}{2})} F_-(t) | F, F \rangle_{\hat{x}}]. \end{aligned} \quad (\text{B1})$$

We substitute Eqs. (1) and (11) into Eq. (B1) and use the formula for the action of the  $F_-(0)$  operator on the eigenstates  $|F, m\rangle$ :

$$F_- |F, m\rangle = \hbar \sqrt{F(F+1) - m(m-1)} |F, m-1\rangle. \quad (\text{B2})$$

This results in

$$\langle F_x(t) \rangle = \hbar \sum_{k, k'=0}^{2F} \text{Re} \left[ \langle F, F - k' | 2^{-2F} \binom{2F}{k'}^{\frac{1}{2}} e^{-i\mu(F_z/\hbar + \frac{1}{2})} \sqrt{F(F+1) - (F-k)(F-k-1)} \binom{2F}{k}^{\frac{1}{2}} | F, F - k - 1 \rangle \right], \quad (\text{B3})$$

with  $\mu = 2\chi t$ . Terms in the sum are nonzero only when  $F - k' = F - k - 1$ , i.e.,  $k' = k + 1$ . Thus, we find

$$\begin{aligned} \langle F_x(t) \rangle &= \hbar \text{Re} \left[ \langle F, F - k - 1 | 2^{-2F} \sum_{k=0}^{2F} \sqrt{\binom{2F}{k+1} \binom{2F}{k}} \sqrt{(2F-k)(k+1)} e^{-i\mu(F-k-1+\frac{1}{2})} | F, F - k - 1 \rangle \right] \\ &= \hbar \text{Re} \left[ 2^{-2F} \sum_{k=0}^{2F} \sqrt{\frac{2F!}{(2F-k-1)!(k+1)!} \frac{2F!}{(2F-k)!k!}} (2F-k)(k+1) e^{-i\mu(F-k-1+\frac{1}{2})} \right] \\ &= \hbar \text{Re} \left[ 2^{-2F} \sum_{k=0}^{2F} \sqrt{\frac{2F(2F-1)!}{(2F-k-1)!(k+1)k!} \frac{2F(2F-1)!}{(2F-k)(2F-k-1)!k!}} (2F-k)(k+1) e^{-i\frac{\mu}{2}(2F-2k-1)} \right] \\ &= \hbar \text{Re} \left[ \frac{2F}{2^{2F}} \sum_{k=0}^{2F} \binom{2F-1}{k} e^{-i\frac{\mu}{2}(2F-k-1)} e^{i\frac{\mu}{2}k} \right] = \hbar \text{Re} \left[ \frac{F}{2^{2F-1}} (e^{-i\frac{\mu}{2}} + e^{i\frac{\mu}{2}})^{2F-1} \right] \\ &= \frac{\hbar F}{2^{2F-1}} \left( 2 \cos \frac{\mu}{2} \right)^{2F-1}, \end{aligned} \quad (\text{B4})$$

where we have used the binomial formula

$$(x + y)^n = \sum_{j=0}^n \binom{n}{j} x^{n-j} y^j. \quad (\text{B5})$$

Simplifying, we obtain Eq. (15a).

### APPENDIX C: SQUEEZING DUE TO ORIENTATION-TO-ALIGNMENT CONVERSION IN CESIUM

Here, we apply the general expressions for squeezing due to orientation-to-alignment conversion presented in the text for the specific case of Cs atoms in the presence of a uniform off-resonant light field. We assume stationary atoms (for example, in a far-off-resonant optical trap) that are initially in the  $F = 4$  hyperfine ground state and prepared in a stretched state as assumed in the text. We apply a  $z$ -polarized light field to the atoms detuned by  $\Delta$  from the  $D_1 F = 4 \rightarrow F' = 3$  or  $F' = 4$  transition, where  $\Delta$  is greater than the natural width but is much smaller than the splitting between hyperfine-structure levels, so that the ground-state level shift is predominantly due to the ac Stark effect arising from the interaction of the light with the near-resonant transition.

The optimum value of the squeezing parameter for  $F = 4$  is  $\xi_R = 0.6$ , as can be found from Fig. 5. The time dependence of the squeezing is given in terms of the quantum-beat frequency  $\chi$ , which is found from the part of the Stark Hamiltonian that is proportional to  $F_z^2$  [see Eq. (6)]. This term can be obtained by writing

$$\hbar\chi = \langle F, m = 1 | H_{\text{eff}} | F, m = 1 \rangle - \langle F, m = 0 | H_{\text{eff}} | F, m = 0 \rangle, \quad (\text{C1})$$

where  $H_{\text{eff}}$  is the effective ground-state Hamiltonian describing ac Stark shifts induced by mixing with the upper state. For light tuned near the  $F = 4 \rightarrow F' = 4$  transition, the second term is zero, and the first term is found from second-order perturbation theory as

$$\langle F, m = 1 | H_{\text{eff}} | F, m = 1 \rangle = \frac{|\langle F, m = 1 | d_z E | F', m = 1 \rangle|^2}{4\Delta}. \quad (\text{C2})$$

Evaluating the dipole matrix element in terms of the reduced matrix element (see, for example, Ref. [33] for a discussion of dipole matrix elements in the presence of hfs), we find

$$\chi = \frac{\langle J = 1/2 || d || J' = 1/2 \rangle^2 E^2}{384\hbar^2 \Delta}; \quad (\text{C3})$$

the value of the reduced dipole matrix element for the  $D_1$  transition is  $\langle J = 1/2 || d || J' = 1/2 \rangle = 3.2 ea_0$ . For light tuned near the  $F = 4 \rightarrow F' = 3$  transition, both terms of Eq. (C1) are nonzero, but the magnitude of  $\chi$  works out to be the same. From the formula (17) for the squeezing parameter, it can be shown that the optimal value for  $F = 4$  occurs at the time when  $\chi t = 0.036 \times 2\pi$ .

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