

Nuclear spin squeezing by continuous quantum non-demolition measurement: a theoretical study

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Abstract

We propose to take advantage of the very weak coupling of the ground-state helium-3 nuclear spin to its environment to produce very long-lived macroscopic quantum states, here nuclear spin squeezed states, in a gas cell at room temperature. To perform a quantum non-demolition measurement of a transverse component of the previously polarized collective nuclear spin, a discharge is temporarily switched on in the gas, which populates helium-3 metastable state. The collective spin corresponding to the $F = 1/2$ metastable level then hybridizes slightly with the one in the ground state by metastability exchange collisions. To access the nuclear spin fluctuations, one continuously measures the light field leaking out of an optical cavity, where it has interacted dispersively with the metastable state collective spin. In a model of three coupled collective spins (nuclear, metastable and Stokes for light) in the Primakoff approximation, and for two measurement schemes, we calculate the moments of the collective nuclear spin squeezed component I_z conditioned on the optical signal averaged over the observation time t . In the photon counting scheme, we find that the squeezed observable is I_z^2 rather than I_z . In the homodyne detection scheme, we analytically solve the stochastic equation for the state of the system conditioned on the measurement; the conditional expectation value of I_z depends linearly on the signal and the conditional variance of I_z does not depend on it. The conditional variance decreases as $(\Gamma_{\text{sq}} t)^{-1}$, where the squeezing rate Γ_{sq} , which we calculate explicitly, depends linearly on the light intensity in the cavity at weak atom-field coupling and saturates at strong coupling to the ground state metastability exchange effective rate, proportional to the metastable atom density. Finally, we take into account the de-excitation of metastable atoms at the walls, which induces nuclear spin decoherence with an effective rate γ_α . It imposes a limit $\propto (\gamma_\alpha/\Gamma_{\text{sq}})^{1/2}$ on the conditional variance reached in a time $\propto (\gamma_\alpha/\Gamma_{\text{sq}})^{-1/2}$. A multilingual version is available on the open archive HAL at <https://hal.archives-ouvertes.fr/hal-03083577>.

Keywords: spin squeezing ; helium 3 ; nuclear spin ; quantum metrology ; stochastic wave functions

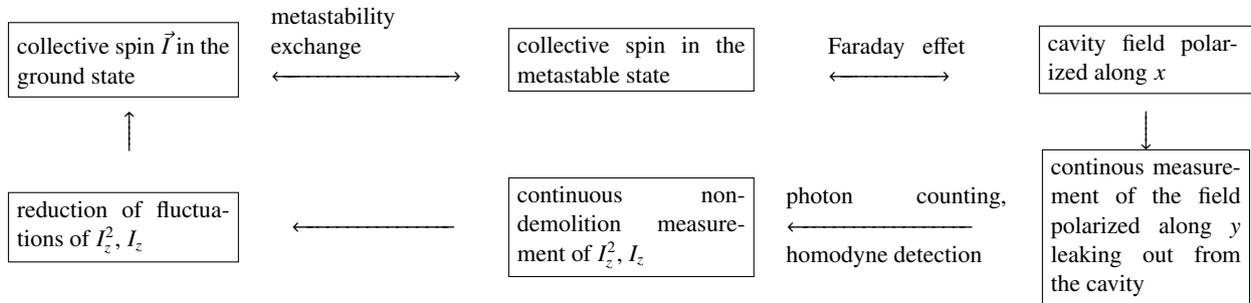
1. Introduction

Helium-3 in its ground state enjoys the remarkable property of having a purely nuclear spin 1/2, perfectly isolated from the outside world even in an environment as hostile to quantum coherences as a gas of helium in a centimetric cell at room temperature and a pressure of the order of a millibar. By well-mastered nuclear polarization techniques, reaching a polarization of 90 %, we can then routinely prepare (for example for lung imaging by nuclear magnetic resonance [1]) a giant collective nuclear spin with an extremely long lifetime. Recently, a coherence time T_2 larger than 60 hours was measured in ultra-precise magnetometry devices [2], that seems limited only by the longitudinal decay time T_1 due to collisions with the cell walls.¹ These numbers make the macroscopic nuclear spin in a room temperature gas an *ideal* system for the production, the study and the use of entangled states, and therefore a competitor of cold atomic gases and Bose-Einstein condensates in metrology and quantum information processing [4]. Already in 2005, we suggested that the nuclear spins of helium-3 could give rise to quantum memories [5] or to non-local quantum states [6] with very long lifetimes. Since then, experimental breakthroughs have been made in the field of

1. Times T_1 of several hundred hours can even be obtained [3].

spin squeezing, notably by means of non-demolition quantum measurements (QND) in atomic alkali gases interacting with the electromagnetic field [4, 7, 8, 9], which recently made it possible to obtain a squeezed spin state with a lifetime of one second in the hyperfine ground state of rubidium under metrological conditions [10]. We recall that, in analogy with the squeezed states of light in quantum optics, a (here collective) spin \vec{I} is said to be squeezed if it admits reduced fluctuations with respect to the standard quantum limit in a direction z orthogonal to that x of the mean spin²; this leads to increased accuracy in pointing the mean spin in that direction, hence in its precession frequency around the y axis, in any metrological device such as an atomic clock or a magnetometer [11, 12] that is sufficiently free from technical noise to reach the standard quantum limit [13].

Transposing to the nuclear spin of helium-3 the technique of squeezing by QND measurement used for the hyperfine spins of alkalis, represents a real challenge, however, due to the specificity of the nuclear spin: its weak coupling to the environment. The singlet ground state of helium-3, separated in energy by about 20 eV from the first excited state, is not directly accessible by laser. However, by means of an oscillating discharge, a small fraction of the gas atoms, on the order of 10^{-6} , can be brought into the metastable triplet state, an excellent starting point for near infrared optical transitions. The orientation of the nuclear spins is then obtained through an indirect process, metastability exchange optical pumping [3]. Initially, the angular momentum is transferred by laser-matter interaction from photons to metastable atoms, a priori to their electron spin (the only one to be strongly coupled to the laser field) but a posteriori also to their nuclear spin thanks to hyperfine coupling. Secondly, we take advantage of the metastability exchange collisions between metastable and ground state atoms to orient the nuclear spins in the ground state, with a time scale of the order of a second, limited by the low density of the atoms in the metastable state. Even though the metastability exchange collision can transfer quantum correlations (see references [5, 6] and our section 3.2), we cannot expect that a single measurement on a small fraction of the atoms (10^{-6}) projects the whole system into a squeezed state. The solution we propose is to perform a continuous QND measurement amplified by a resonant optical cavity in which the cell is placed. Indeed, although the metastable atoms individually have a relatively short lifetime (they lose their quantum correlations and fall back into the ground state in each collision with the cell walls), a continuous destructive measurement of the light leaking out of the cavity after interaction with the metastable atoms amounts to performing a continuous QND measurement on the collective nuclear spin in the ground state, which prepares it into the desired squeezed state without affecting its lifetime.³ As we will see, our procedure for reducing spin fluctuations is more precisely summarized by the diagram:



This work gives a detailed theoretical description of the squeezing mechanism and its limits; a more detailed feasibility study taking into account the experimentally accessible values of the parameters is carried out in reference [14]. Very recently, similar ideas have been put forward in a different physical system, the alkali-rare gas mixture [15, 16]. We are confident that quantum manipulation of long-lived nuclear spins is promised rapid development, opening up new perspectives for basic research and applications, in particular in magnetometry [9, 17, 18, 19, 20].

2. The standard quantum noise corresponds to $\Delta I_y^{\text{st}} = \Delta I_z^{\text{st}} = (|\langle I_x \rangle|/2)^{1/2}$ (our spins being dimensionless here) i.e. to the equality in the case of cylindrical symmetry of the fluctuations around the mean spin in the Heisenberg inequality $\Delta I_y \Delta I_z \geq |\langle I_x \rangle|/2$. A more detailed discussion shows that, in the considered geometry, the noise-to-signal ratio in a precession frequency measurement is in fact proportional to $\xi = (2I)^{1/2} \Delta I_z / |\langle I_x \rangle|$ (this naturally brings in the uncertainty angle on the spin direction in the xz plane) rather than to the naively expected $\Delta I_z / \Delta I_z^{\text{st}}$ ratio. In our proposed scheme, we degrade ξ^2 by a factor ≈ 2 using a partially polarized nuclear spin state $\langle I_x \rangle / I < 1$, see figure 3, but we aim to gain much more with spin squeezing.

3. This type of measurement thus differs from the instantaneous measurement of quantum mechanics textbooks, which projects the state of the system into an eigenspace of the measured observable according to the von Neumann postulate.

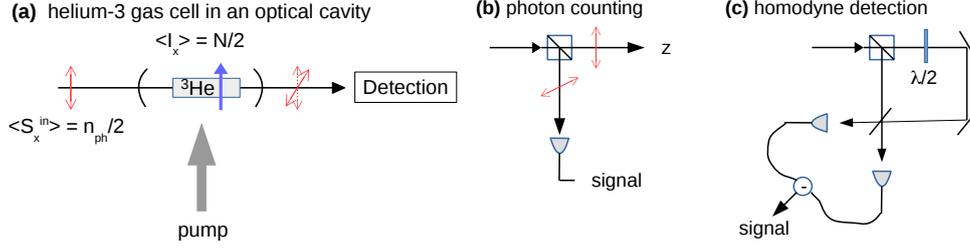


Figure 1 – Overview of the set-up. (a) A centimetric glass cell is filled with a helium-3 gas at room temperature and placed in an optical cavity with axis z (horizontal axis in the figure). The Stokes spin of light and the atomic spins (nuclear and metastable) are linearly polarized along x (vertical axis in the figure). The cavity mode polarized along y , initially empty, is populated by the Faraday effect due to the quantum fluctuations of the metastable spins along z during its propagation in the gas. It is measured continuously outside the cavity by one of the following two methods: (b) the photons leaking out of the cavity polarized along y are separated from those polarized along x by a polarizing beam-splitter and then detected in the photon counting regime. (c) Outside the cavity, a homodyne detection of one quadrature of the outgoing field polarized along y is carried out, using as local oscillator the outgoing field polarized along x (whose polarization has been previously rotated with a half-wave plate to bring it along y).

2. Overview and semi-classical description

The considered physical system is shown in figure 1. A cell filled with a partially polarized gas of a few mbar of pure helium-3 atoms is placed inside an optical cavity. While the majority of atoms remain in the singlet ground state 1^1S of helium, a weak discharge brings a tiny fraction of the atoms, usually $\approx 10^{-6}$, into the metastable triplet state 2^3S . On the one hand, the cavity is driven by a laser beam propagating along the cavity axis z and linearly polarized in the direction x , which is also the direction of polarization of the atomic sample, to excite the $2^3S - 2^3P$ transition with a large frequency detuning; on the other hand, atoms in the metastable state 2^3S (of electronic and nuclear hyperfine spin) are coupled to atoms in the ground state (of purely nuclear spin) by metastability exchange collisions; remarkably, although each exchange collision is individually incoherent, this leads to a well-defined macroscopic coupling between the corresponding collective spins [21, 22]. As the Faraday interaction with the metastable atoms causes the polarization of the light initially directed along x to rotate slightly around the z axis, by an angle proportional to the component of the collective metastable spins along z as we will see, a continuous destructive measurement of the polarization component along y of the field leaking out of the cavity (i) by counting photons as indicated in figure 1b or (ii) by homodyne detection as in figure figure 1c, ultimately performs a non-demolition continuous quantum measurement of the collective nuclear spin along z of helium-3 atoms in the ground state.

In the rest of this section, by a semi-classical treatment of the spin fluctuations around their expectation values in the stationary state, we reduce our complex physical system to the simpler one of three coupled collective spins, of which section 3 will give a quantum description.

The relevant atomic structure of the ^3He atom and the transitions excited by the cavity field are shown in figure 2. We call \vec{I} the collective nuclear spin in the ground state, \vec{J} and \vec{K} the collective spins associated with the hyperfine manifolds $F = 3/2$ and $F = 1/2$ in the metastable state. For light propagating along z , we introduce the Stokes spin [23] built from the creation and annihilation operators of a photon in the linearly polarized cavity modes along x and y :⁴

$$S_x = \frac{1}{2} (c_x^\dagger c_x - c_y^\dagger c_y) \quad ; \quad S_y = \frac{1}{2} (c_x^\dagger c_y + c_y^\dagger c_x) \quad ; \quad S_z = \frac{1}{2i} (c_x^\dagger c_y - c_y^\dagger c_x). \quad (1)$$

We assume for simplicity that the cell is uniformly illuminated by the cavity mode. Within the limit of a large detuning and a weak saturation of the atomic transition by the field, the excited state 2^3P can be eliminated adiabatically and

4. Equivalently, we can build the Stokes spin \vec{S} using annihilation operators in circularly polarized modes, $c_1 = \frac{1}{\sqrt{2}}(c_x - ic_y)$, $c_2 = \frac{1}{\sqrt{2}}(c_x + ic_y)$ [24], in which case $S_z = \frac{1}{2} (c_1^\dagger c_1 - c_2^\dagger c_2)$.

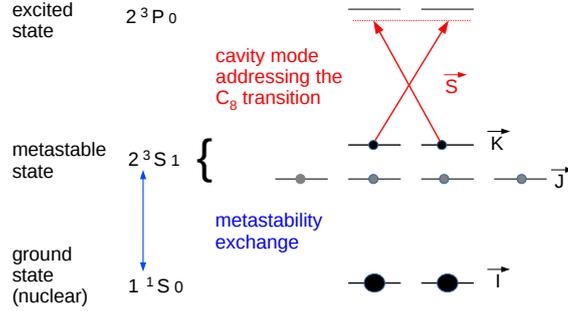


Figure 2 – Relevant energy levels of the ^3He atoms (the Zeeman sublevels correspond to the choice of z as the quantization axis, the atoms being polarized along x). The cavity mode polarized along x excites the transition C_8 between the level $F = 1/2$ of the metastable state 2^3S_1 and the highest energy level $F = 1/2$ of the excited state 2^3P , with a negative frequency detuning much larger in absolute value than the Doppler half-width of the excited state (of the order of 1 GHz), so that the velocity class resonant with the laser is almost empty, but much smaller than the 6.74 GHz hyperfine splitting in the metastable state (and a fortiori than the fine splitting $2^3P_1 - 2^3P_0$ of 29.6 GHz in the excited state), so that the metastable level $F = 3/2$ is very weakly affected by the laser. (Note: The frequency separation does not make it possible to largely satisfy these two constraints, and we cannot exclude that the coupling of $F = 3/2$ to the field has a small effect on the squeezing dynamics; we neglect it here but we could take it into account with a more complete than our minimal model Hamiltonian (2), like that of reference [23]). The six sublevels of the 2^3S_1 metastable state are coupled to the two (purely nuclear) sublevels of the 1^1S_0 ground state by metastability exchange collisions.

the interaction Hamiltonian between the metastable spin \vec{K} and the Stokes spin \vec{S} takes the Faraday form [23]:

$$H = \hbar\chi K_z S_z \quad (2)$$

which is none other than the lightshift operator of Zeeman sublevels in the metastable level $F = 1/2$, as we can clearly see from the form of S_z in footnote 4. The coupled nonlinear equations describing the evolution of the mean spins are given in Appendix A, see equations (A.1)-(A.3). Besides the evolution due to the Faraday Hamiltonian (2) and to the metastability exchange collisions, they include the contribution of the usual Liouvillian terms in the quantum master equation describing the injection of a polarized coherent field along x in the cavity and the losses due to the output mirror, whose combined effect leads to $\langle S_x \rangle = n_{\text{ph}}/2$ in the stationary state in the absence of atoms, n_{ph} being the average number of photons in the polarized mode along x . These equations are then linearized around a partially polarized stationary solution (A.8)-(A.9), and the fluctuations of the spin \vec{J} and the collective alignment tensor in $F = 3/2$ are eliminated adiabatically⁵ to obtain coupled equations for the fluctuations of the three collective spins \vec{I} , \vec{K} and \vec{S} , whose stationary mean values are given by:

$$\langle \vec{I} \rangle_s = \frac{N}{2} \vec{u}_x \quad ; \quad \langle \vec{K} \rangle_s = \frac{n}{2} \vec{u}_x \quad ; \quad \langle \vec{S} \rangle_s = \frac{n_{\text{ph}}}{2} \vec{u}_x \quad (3)$$

Here \vec{u}_x is the unit vector along x , N and n are the effective numbers of ground-state and metastable atoms participating in the dynamics of the collective spins. As we show in Appendix A, these effective numbers are renormalized with

5. We think that this non-mathematically controlled approximation is reasonable for the proposed experiment, because the spin \vec{J} is not directly coupled to light so it is not directly affected by the continuous field measurement. On the other hand, by eliminating in the same way the fluctuations of the spin \vec{K} , directly coupled to the field, one would commit a non-negligible error on the spin squeezing dynamics in the case of the detection by photon counting (amounting to omitting the double jump C_d in the quantum master equation (37) and the rate Γ_0 in the average number of photons counted (45)) therefore strongly underestimating the number of photodetections required to achieve a given squeezing level), but a negligible error in the case of homodyne detection, as we have verified on the one-mode model in section 3.4.

respect to the total true numbers N_{cell} and n_{cell} in the cell, by polarization dependent factors:

$$N = \eta N_{\text{cell}} \quad ; \quad n = \left(\frac{1 - \eta^2}{3 + \eta^2} \right) \eta n_{\text{cell}} \quad (4)$$

where $\eta \in [0, 1]$ is the nuclear polarization,⁶ and the semi-classical equations for the fluctuations of the three collective spins are:

$$\frac{d}{dt} \delta S_z = -\frac{\kappa}{2} \delta S_z \quad ; \quad \frac{d}{dt} \delta S_y = -\frac{\kappa}{2} \delta S_y + \chi \langle S_x \rangle_s \delta K_z \quad (5)$$

$$\frac{d}{dt} \delta I_z = -\gamma_f \delta I_z + \gamma_m \delta K_z \quad ; \quad \frac{d}{dt} \delta I_y = -\gamma_f \delta I_y + \gamma_m \delta K_y \quad (6)$$

$$\frac{d}{dt} \delta K_z = -\gamma_m \delta K_z + \gamma_f \delta I_z \quad ; \quad \frac{d}{dt} \delta K_y = -\gamma_m \delta K_y + \gamma_f \delta I_y + \chi \langle K_x \rangle_s \delta S_z \quad (7)$$

Here, κ is the cavity loss rate, γ_m and γ_f are the effective metastability exchange rates in the metastable state and in the ground state. The latter depend on the nuclear polarization as below and in figure 3a, and are in the same ratio as the effective atom numbers N and n (4) forming the collective spins:

$$\gamma_f = \frac{4 + \eta^2}{8 - \eta^2} \frac{1 - \eta^2}{3 + \eta^2} \frac{1}{T} \quad ; \quad \gamma_m = \frac{4 + \eta^2}{8 - \eta^2} \frac{1}{\tau} \quad ; \quad \frac{\gamma_m}{\gamma_f} = \frac{N}{n} \gg 1 \quad (8)$$

the individual metastability exchange collisions rates $1/T$ and $1/\tau$ experienced by an atom in the ground state and in the metastable state being proportional to n_{cell} and N_{cell} respectively, with the same proportionality constant. In figure 3b, we also show the nuclear polarization dependence of the effective Faraday coupling Ω_a (26) between light and the nuclear spin hybridized by the metastable, which controls the spin squeezing rate in (32).

3. Quantum description

In section 2, we have seen that we can model our complex physical system as three coupled collective spins (3): the nuclear spin \vec{I} in the ground state, the spin \vec{K} in the hyperfine level $F = 1/2$ of the metastable state and the Stokes spin \vec{S} of the cavity field. In this section, we present the full quantum treatment of this model. After having introduced the Primakoff approximation, we move on to the quantum description of the metastability exchange which couples the nuclear and metastable spins.

3.1. Primakoff approximation and metrological gain due to squeezing

Initially, the collective nuclear spin \vec{I} , the collective metastable spin \vec{K} and the Stokes spin \vec{S} of light are polarized along x , and will remain so throughout the experimental procedure. In the Holstein-Primakoff approximation, which treats the macroscopic spin components along x as classical variables, the remaining y and z components, orthogonal to the mean spins, behave like the quadrature operators (Hermitian and antihermitian parts of annihilation operators, therefore canonically conjugated, $[X, P] = i/2$) of three bosonic modes a, b, c :⁷

$$\frac{I_y}{\sqrt{N}} \stackrel{\text{Primakoff}}{\simeq} X_a = \frac{a + a^\dagger}{2} \quad ; \quad \frac{K_y}{\sqrt{n}} \stackrel{\text{Primakoff}}{\simeq} X_b = \frac{b + b^\dagger}{2} \quad ; \quad \frac{S_y}{\sqrt{n_{\text{ph}}}} \stackrel{\text{Primakoff}}{\simeq} X_c = \frac{c + c^\dagger}{2} \quad (9)$$

$$\frac{I_z}{\sqrt{N}} \stackrel{\text{Primakoff}}{\simeq} P_a = \frac{a - a^\dagger}{2i} \quad ; \quad \frac{K_z}{\sqrt{n}} \stackrel{\text{Primakoff}}{\simeq} P_b = \frac{b - b^\dagger}{2i} \quad ; \quad \frac{S_z}{\sqrt{n_{\text{ph}}}} \stackrel{\text{Primakoff}}{\simeq} P_c = \frac{c - c^\dagger}{2i} \quad (10)$$

6. Note that $n = 0$ in the fully polarized case $\eta = 1$. Indeed, the entire population of the metastable state is then in the extreme Zeeman sublevel $m_x = 3/2$ of the hyperfine state $F = 3/2$ and the manifold $F = 1/2$ is empty.

7. If we consider a large spin \vec{S} fully polarized along x , we can approximate the spin component in this direction by a classical variable, by setting $\hat{S}_x \simeq \langle \hat{S}_x \rangle$ so that $[\hat{S}_y/\sqrt{2\langle \hat{S}_x \rangle}, \hat{S}_z/\sqrt{2\langle \hat{S}_x \rangle}] \simeq i/2$.

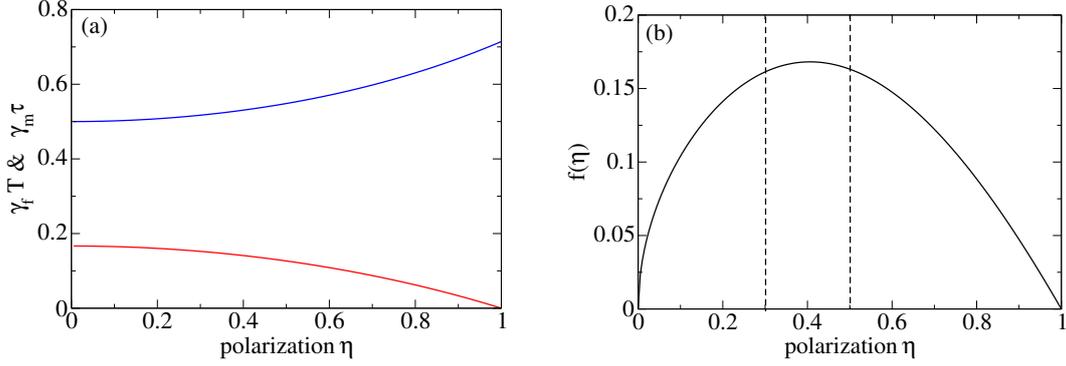


Figure 3 – (a) Effective metastability exchange rates γ_f (lower red curve) and γ_m (upper blue curve) as given by equation (8) in the ground and metastable states, as functions of nuclear polarization η , normalized by the metastability exchange collision rates $1/T$ and $1/\tau$ experienced by a ground-state and a metastable atom in the gas. (b) Nuclear polarization dependence of the Faraday frequency Ω_α entering in the rate (32) at which excitations are created by Faraday coupling in the hybridized nuclear bosonic mode, and in the spin squeezing rates (49) and (95), in the limit $\gamma_f \ll \gamma_m$; more precisely, we plot the factor $f(\eta) = \sqrt{\eta \frac{1-\eta^2}{3+\eta^2}}$ such that $\Omega_\alpha \simeq \Omega(\gamma_f/\gamma_m)^{1/2} = \chi \sqrt{n_{\text{ph}} n_{\text{cell}}} \sqrt{\frac{n_{\text{cell}}}{N_{\text{cell}}}} f(\eta)$. When the polarization varies between 0.3 and 0.5 (vertical dashed lines), $f(\eta)$ deviates by 4 % from its maximum $\simeq 0.17$ reached in $\eta = 0.42$. It is therefore advantageous to place oneself close to this value of η to reduce the temporal drift of Ω_α due to a slight damping of the nuclear polarization during spin squeezing (indeed, the optical pumping process is then interrupted and the lifetime of the polarization is reduced by the presence of the discharge, it becomes of the order of γ_α^{-1} , where γ_α is the induced decoherence rate of section 4.2.4).

We used the mean values (3) in the normalization. Let us make the link with the exact bosonic representation (1) of Stokes' spin, writing:

$$\frac{S_y}{\sqrt{n_{\text{ph}}}} - i \frac{S_z}{\sqrt{n_{\text{ph}}}} = \frac{1}{\sqrt{n_{\text{ph}}}} c_y^\dagger c_x \stackrel{\text{Primakoff}}{\simeq} c_y^\dagger \quad ; \quad \frac{S_y}{\sqrt{n_{\text{ph}}}} + i \frac{S_z}{\sqrt{n_{\text{ph}}}} = \frac{1}{\sqrt{n_{\text{ph}}}} c_x^\dagger c_y \stackrel{\text{Primakoff}}{\simeq} c_y \quad (11)$$

This shows that the creation operator c^\dagger in (9)-(10), identified with c_y^\dagger in Primakoff's approximation, transfers a photon from the highly populated coherent state cavity mode polarized along x into the initially empty cavity mode polarized along y . In Primakoff's approximation, the atom-field Faraday coupling Hamiltonian (2) is written:

$$H = \hbar \Omega P_b P_c \quad \text{with} \quad \Omega = \chi \sqrt{m_{\text{ph}}}. \quad (12)$$

As χ does not depend on the field strength in the cavity, Ω^2 is proportional to its intensity. Finally, let's write in terms of Primakoff variables the parameter ξ of reference [11] quantifying the level of spin squeezing usable in a magnetometer (the metrological gain is the higher as ξ is the lower, see our footnote 2), z being the maximal squeezing transverse direction to the average spin and the collective nuclear spin being of quantum number $I = N_{\text{cell}}/2$:

$$\xi^2 \equiv \frac{2I \text{Var}_{\text{signal}}(I_z)}{\langle I_x \rangle^2} = \frac{4 \text{Var}_{\text{signal}}(P_a)}{\eta} \quad (13)$$

The nuclear polarization η being fixed, and a non-demolition quantum measurement being carried out continuously on the nuclear spin, one must try to minimize the variance of P_a conditioned on the measurement signal to be defined, by making it decrease as much as possible below its initial value $1/4$.

3.2. Quantum master equation for metastability exchange

Let us consider in this subsection the evolution of the system due to metastability exchange only ($\chi = 0$). In a quantum treatment, the classical equations (6)-(7) become stochastic equations including quantum fluctuations. In

Primakoff's approximation, this gives for the quadratures X in the metastable and fundamental state:

$$dX_a = -\gamma_f X_a dt + \sqrt{\gamma_m \gamma_f} X_b dt + dX_a^{\text{stoch}} \quad ; \quad dX_b = -\gamma_m X_b dt + \sqrt{\gamma_m \gamma_f} X_a dt + dX_b^{\text{stoch}} \quad (14)$$

where we used the third equality of equation (8). Langevin noises dX_i^{stoch} , with $i \in \{a, b\}$, have zero mean, are independent random variables at different times, and have variances and equal-time covariances calculated in reference [5]:

$$\langle dX_i^{\text{stoch}} dX_j^{\text{stoch}} \rangle = D_{ij} dt \quad \text{with} \quad \underline{D} = \frac{1}{2} \begin{pmatrix} \gamma_f & -\sqrt{\gamma_m \gamma_f} \\ -\sqrt{\gamma_m \gamma_f} & \gamma_m \end{pmatrix} \quad (15)$$

We have equations of the same form as (14) for the quadratures P_i , with other Langevin noises dP_i^{stoch} , with the same covariance matrix as equation (15) between them but with a covariance matrix with the noises dX_i^{stoch} given by

$$\langle dX_i^{\text{stoch}} dP_j^{\text{stoch}} \rangle = \mathcal{D}_{ij} dt \quad \text{with} \quad \underline{D} = i \underline{D} \quad (16)$$

For calculating the mean values and variances of atomic observables, this stochastic formulation is equivalent to a quantum master equation for the atomic density operator ρ_{at} of the two bosonic modes a and b :

$$\frac{d\rho_{\text{at}}}{dt} = C \rho_{\text{at}} C^\dagger - \frac{1}{2} \{C^\dagger C, \rho_{\text{at}}\} \quad \text{with} \quad C = \sqrt{2\gamma_f} a - \sqrt{2\gamma_m} b \quad (17)$$

Indeed, the Langevin stochastic representation of the quantum master equation (17) for any operator A is written

$$dA = \frac{dt}{2} \{C^\dagger [A, C] - [A, C^\dagger] C\} + dA^{\text{stoch}} \quad \text{where} \quad dA^{\text{stoch}} = [C^\dagger, A] dB + dB^\dagger [A, C] \quad (18)$$

and dB is a Markovian stochastic operator with zero mean, with an equal-time covariance matrix

$$\langle dB dB^\dagger \rangle = dt \quad ; \quad \langle dB dB \rangle = \langle dB^\dagger dB^\dagger \rangle = \langle dB^\dagger dB \rangle = 0 \quad (19)$$

To be complete, let us sketch another reasoning, which avoids quantum Langevin noises. It suffices to admit that the equations of evolution on the means $\langle X_i \rangle$ and $\langle P_i \rangle$ taken from (6)-(7) derive from a quantum master equation of the Lindblad form (52). Since these equations are linear, the jump operators C_m surrounding ρ_{at} in the quantum master equation are linear combinations of a and b , and we recover (17).

3.3. Three-mode quantum master equation

The complete evolution, including the atom-field coupling of Hermitian Hamiltonian (12), metastability exchange and cavity losses, is described by the quantum master equation ⁸

$$\frac{d\rho}{dt} = \frac{1}{i\hbar} [H, \rho] + \kappa \left(c \rho c^\dagger - \frac{1}{2} \{c^\dagger c, \rho\} \right) + C \rho C^\dagger - \frac{1}{2} \{C^\dagger C, \rho\} \quad (20)$$

8. We neglect here the internal evolution of the atomic modes (spin precession) by supposing that the Zeeman sublevels are degenerate in the ground state and in the metastable level $F = 1/2$, that is that the external magnetic field is zero, $\vec{B} = \vec{0}$. This simplifying assumption calls for the following comments. (i) In the experiment, we plan to impose a guiding field along x of the order of μT to avoid a "wild" precession of the mean spin around a residual magnetic field of unknown direction [14]. From a theoretical point of view, we then place ourselves in the frame rotating around x at the corresponding nuclear spin Larmor frequency $\omega_L^{(x)}$ to eliminate this guiding field [in principle it would be necessary to compensate for the difference between the metastable and ground-state Larmor frequencies, for example by means of a fictitious magnetic field created by a light shift, but this precaution seems superfluous because the metastable Larmor frequency remains small compared to the effective metastability exchange rate γ_m]; in principle, the cavity must also be rotated at the angular speed $\omega_L^{(x)}$ so that it is stationary in the rotating frame; if the cavity remains fixed in the laboratory frame, we can switch on the Faraday coupling and perform a measurement of the field leaving the cavity no longer continuously but stroboscopically (each time the spin component to be squeezed merges with optical axis) [7]. (ii) If there exists during the squeezing phase a small parasitic static magnetic field (in addition to the guiding field) in the yz plane, at an angle ϕ with y , this field rotates at the angular frequency $-\omega_L^{(x)}$ in the rotating frame; in the stochastic equation of the one-mode model (58) for homodyne detection, this adds a contribution of Hermitian Hamiltonian $H_\alpha = \sqrt{N} \hbar \omega_L^{(y)} (\Omega_\beta / \Omega) [\cos(\phi - \omega_L^{(x)} t) X_\alpha + \sin(\phi - \omega_L^{(x)} t) P_\alpha]$ where $\omega_L^{(y)} \ll \omega_L^{(x)}$ is the Larmor angular frequency of the nuclear spin in the parasitic field and Ω_β / Ω is given by equation (26). The P_α term is absorbed in a phase change of ansatz (59). The X_α term does not change the parameter u of ansatz (59), and hence the variance of P_α in a realization, but adds an oscillating deterministic part to \bar{P}_α , namely $(\sqrt{N} \omega_L^{(y)} / 2 \omega_L^{(x)}) (\Omega_\beta / \Omega) [\sin(\phi - \omega_L^{(x)} t) - \sin \phi]$ and, according to (65), a deterministic part $\sqrt{\Gamma_{\text{ex}}} (\Omega_\beta / \Omega) (\sqrt{N} \omega_L^{(y)} / 2 \omega_L^{(x)}) [\cos(\phi - \omega_L^{(x)} t) - \cos \phi] / (\omega_L^{(x)} t) - \sin \phi$ to the integrated signal $\sigma(t)$. The $\sin \phi$ term in the signal can be canceled by

where C is the jump operator for metastability exchange (17), κ is the cavity loss rate, γ_m and γ_f are the effective metastability exchange rates in the ground state and in the metastable state.

Initially, the three modes are in vacuum state corresponding to a polarized state for the three spins. For this initial state, the first moments of the quadratures remain zero, and one can obtain a closed system of equations for the second moments. We find that the quadratures P maintain constant variances and zero covariances in the three modes,

$$\langle P_a^2 \rangle(t) = \langle P_b^2 \rangle(t) = \langle P_c^2 \rangle(t) = \frac{1}{4} \quad ; \quad \langle P_a P_b \rangle(t) = \langle P_a P_c \rangle(t) = \langle P_b P_c \rangle(t) = 0 \quad (21)$$

that the variance $\langle X_c^2 \rangle$ remains bounded and the covariances $\langle X_a X_c \rangle$ and $\langle X_b X_c \rangle$ remain zero, while the variances and covariance of the quadratures X_a and X_b , and therefore the number of excitations in the atomic modes,⁹ diverge linearly in time, at least as long as the Primakoff approximation is applicable. We give here explicitly only long-time behaviors:

$$\begin{aligned} \langle X_a^2 \rangle(t) &\underset{t \rightarrow +\infty}{=} \frac{\gamma_m \gamma_f}{(\gamma_m + \gamma_f)^2} \frac{\Omega^2 t}{4\kappa} + O(1) & \langle X_b^2 \rangle(t) &\underset{t \rightarrow +\infty}{=} \frac{\gamma_f^2}{(\gamma_m + \gamma_f)^2} \frac{\Omega^2 t}{4\kappa} + O(1) \\ \langle X_a X_b \rangle(t) &\underset{t \rightarrow +\infty}{=} \frac{\gamma_m^{1/2} \gamma_f^{3/2}}{(\gamma_m + \gamma_f)^2} \frac{\Omega^2 t}{4\kappa} + O(1) & \langle X_c^2 \rangle(t) &\underset{t \rightarrow +\infty}{=} \frac{1}{4} \left(\frac{\Omega}{2\kappa} \right)^2 \left(1 - \frac{2\gamma_m}{\kappa + 2(\gamma_m + \gamma_f)} \right) \end{aligned} \quad (22)$$

3.4. One-mode model

In this subsection, we derive a one-mode quantum master equation describing the slow evolution of the nuclear spin within the limit

$$\Gamma_{\text{ex}} \ll \gamma_f < \gamma_m \quad \text{and} \quad \Gamma_{\text{ex}} \ll \kappa \quad (23)$$

where Γ_{ex} is the rate at which excitations are created under the effect of Faraday coupling in the hybridized nuclear bosonic mode α defined below (it suffices to know here that $\Gamma_{\text{ex}} \propto \Omega^2$ so that (23) is a weak Faraday coupling limit $\Omega \rightarrow 0$). To this end, it is convenient to introduce the bosonic annihilation operators into a cleverly rotated basis, by means of the following linear combinations of the operators a and b :

$$\alpha = \sqrt{\frac{\gamma_m}{\gamma_m + \gamma_f}} a + \sqrt{\frac{\gamma_f}{\gamma_m + \gamma_f}} b \quad ; \quad \beta = \sqrt{\frac{\gamma_m}{\gamma_m + \gamma_f}} b - \sqrt{\frac{\gamma_f}{\gamma_m + \gamma_f}} a \quad (24)$$

α and β indeed correspond to the eigenmodes of the metastability exchange part of the three-mode quantum master equation (20) (in practice, we have $\gamma_m \gg \gamma_f$, see equation (8), so that the mode β corresponds to the metastable spin slightly hybridized with the spin of the ground state, and α to the nuclear spin slightly hybridized with the metastable spin). While the α mode undergoes a time divergence of its average number of excitations (hence the possibility of defining a rate Γ_{ex}), the β mode is strongly damped and tends towards a stationary value (see the results (21) and (22), which show that $\langle P_\beta^2 \rangle = 1/4$ and $\langle X_\beta^2 \rangle = O(1)$ where $X_\beta = (\beta + \beta^\dagger)/2$), which will allow to eliminate it adiabatically, just like the cavity field. In this new basis, the three-mode master equation (20) takes the form

$$\boxed{\frac{d\rho}{dt} = \frac{1}{i\hbar} [H, \rho] + \kappa \left(c\rho c^\dagger - \frac{1}{2} \{c^\dagger c, \rho\} \right) + \gamma_\beta \left(\beta\rho\beta^\dagger - \frac{1}{2} \{\beta^\dagger\beta, \rho\} \right)} \quad (25)$$

choosing the initial time “ $t = 0$ ” of the squeezing procedure; the other contributions cancel out for a duration t of the whole experiment multiple of $2\pi/\omega_L^{(x)}$. (iii) If we want to use the spin squeezed state to measure a magnetic field, we turn off the discharge and the guiding field and we arrange that the field to be measured \vec{B}_{mes} is oriented along y . The collective nuclear spin then precesses in the xz plane by an angle which must be measured to access \vec{B}_{mes} , and the initial direction of squeezing z is precisely that which is necessary for reducing the angular uncertainty on the spin. Another strategy consists in starting from an ordinary polarized state, unsqueezed, of the nuclear spin and in carrying out, discharge on but guiding field off, the continuous measurement of the light leaking out of the cavity in the presence of $B_{\text{mes}}\vec{t}_y$; in the one-mode model with homodyne detection of section 4.2.2, we then recover the magnetometry proposals of references [17, 18].

9. For the initial state considered, we have at all times $\langle X_a \rangle = 0$ and $\langle X_a^2 \rangle - \frac{1}{4} = \langle a^\dagger a \rangle$, where $\langle a^\dagger a \rangle$ is the average number of excitations in the nuclear spin mode, so that $\text{Var } X_a = \langle a^\dagger a \rangle + \frac{1}{4}$; one has indeed $a^\dagger a + 1/2 = X_a^2 + P_a^2$. The same relations hold for the other two modes.

where $\gamma_\beta \equiv 2(\gamma_m + \gamma_f)$ and, noting $P_\alpha = (\alpha - \alpha^\dagger)/2i$ and $P_\beta = (\beta - \beta^\dagger)/2i$ the P quadratures of the new modes,

$$H = \hbar(\Omega_\alpha P_\alpha + \Omega_\beta P_\beta)P_c \quad \text{with} \quad \Omega_\alpha \equiv \Omega \sqrt{\frac{\gamma_f}{\gamma_m + \gamma_f}} \quad \text{and} \quad \Omega_\beta \equiv \Omega \sqrt{\frac{\gamma_m}{\gamma_m + \gamma_f}} \quad (26)$$

Reference [25] explains in all generality how to perform an adiabatic elimination at the level of the master equation. Here we prefer to carry it out, as in reference [26], in the weak Faraday coupling limit $\Omega \rightarrow 0$ in the Monte Carlo wave function formalism [27, 28], where the density operator solution of the quantum master equation (25) is obtained by averaging pure states over independent stochastic realizations, each realization corresponding to the deterministic evolution of an unnormalized state vector $|\psi(t)\rangle$ under the action of the effective non-Hermitian Hamiltonian

$$H_{\text{eff}} = H - \frac{i\hbar}{2} (\kappa c^\dagger c + \gamma_\beta \beta^\dagger \beta) \quad (27)$$

interrupted randomly by quantum jumps (discontinuous evolutions $|\psi\rangle \rightarrow C|\psi\rangle$) of jump operators

$$C_c = \sqrt{\kappa}c \quad \text{and} \quad C_\beta = \sqrt{\gamma_\beta}\beta. \quad (28)$$

In the absence of the coherent coupling Ω in (26) the hybridized metastable mode and the cavity mode remain in the initial empty state. To first order in Ω , this state is coupled to states with an excitation in the cavity (by the action of P_c) and with zero or one excitation in the mode of the hybridized metastable (by the action of P_α or P_β). We can then truncate the Monte Carlo state vector $|\psi\rangle$ in the Fock basis $\{|n_\alpha\rangle_{\text{background}}|n_\beta\rangle_{\text{meta}}|n_c\rangle_{\text{cav}}\}$ as follows

$$|\psi\rangle = |\psi_\alpha^{00}\rangle|0\rangle|0\rangle + |\psi_\alpha^{01}\rangle|0\rangle|1\rangle + |\psi_\alpha^{11}\rangle|1\rangle|1\rangle \quad (29)$$

committing an error of norm $O(\Omega^2)$. Under the effect of the effective Hamiltonian (27), the fast components $|\psi_\alpha^{01}\rangle$ and $|\psi_\alpha^{11}\rangle$ exponentially join an adiabatic following regime of the slow component $|\psi_\alpha^{00}\rangle$ with rates $\kappa/2$ or $(\kappa + \gamma_\beta)/2$. Hence their adiabatic elimination within the limit (23)¹⁰

$$|\psi_\alpha^{11}\rangle_{\text{adiab}} \simeq \frac{i\Omega_\beta}{2(\kappa + \gamma_\beta)} |\psi_\alpha^{00}\rangle \quad \text{and} \quad |\psi_\alpha^{01}\rangle_{\text{adiab}} \simeq \frac{\Omega_\alpha}{\kappa} P_\alpha |\psi_\alpha^{00}\rangle \quad (30)$$

We put the expressions of $|\psi_\alpha^{11}\rangle_{\text{adiab}}$, $|\psi_\alpha^{01}\rangle_{\text{adiab}}$ in the Hamiltonian evolution equation of $|\psi_\alpha^{00}\rangle$ to obtain

$$i\hbar \frac{d}{dt} |\psi_\alpha^{00}\rangle = -\frac{i\hbar}{2} (\Gamma_{\text{ex}} P_\alpha^2 + \Gamma_0) |\psi_\alpha^{00}\rangle \equiv H_{\text{eff}}^{00} |\psi_\alpha^{00}\rangle \quad (31)$$

where we have introduced the rates

$$\Gamma_{\text{ex}} = \frac{\Omega_\alpha^2}{\kappa} \quad \text{and} \quad \Gamma_0 = \frac{\Omega_\beta^2}{4(\kappa + \gamma_\beta)} \quad (32)$$

By studying the effect of the cavity jump operator C_c and the metastability exchange jump operator C_β on the state vector (29), we can interpret the effective Hamiltonian of equation (31). (i) Let us first consider the effect of a cavity

10. In adiabatic following, the occupation probabilities of the excited components are $\langle \psi_\alpha^{11} | \psi_\alpha^{11} \rangle_{\text{adiab}} / \langle \psi | \psi \rangle = [\Omega_\beta^2 / 4(\kappa + \gamma_\beta)^2] \langle \psi_\alpha^{00} | \psi_\alpha^{00} \rangle / \langle \psi | \psi \rangle$ and $\langle \psi_\alpha^{01} | \psi_\alpha^{01} \rangle_{\text{adiab}} / \langle \psi | \psi \rangle = (\Gamma_{\text{ex}} / \kappa) \langle \psi_\alpha^{00} | P_\alpha^2 | \psi_\alpha^{00} \rangle / \langle \psi | \psi \rangle$ where we used (32). Within the limit (23), we can easily verify that they are $\ll 1$, so that almost all the population is in the component $|\psi_\alpha^{00}\rangle|0\rangle|0\rangle$ as it should be, which will allow us to replace $\langle \psi | \psi \rangle$ by $\langle \psi_\alpha^{00} | \psi_\alpha^{00} \rangle$. We also verify that another condition for the validity of adiabatic elimination, namely the slowness of the evolution of the hybridized nuclear spin α with respect to the fast variables, which reads here $\Gamma_{\text{ex}}, \Gamma_0 \ll \kappa, \kappa + \gamma_\beta$, is satisfied. However, these considerations do not allow us to show that the condition $\Gamma_{\text{ex}} \ll \gamma_f$ is necessary (unless $\kappa \ll \gamma_\beta$). To see it in general terms, we push to the order Ω^4 the computation of the effective Hamiltonian $H_{\text{eff}}^{00} = PH_{\text{eff}}P + PHQ(zQ - QH_{\text{eff}}Q)^{-1}QHP$ in the subspace $n_\beta = n_c = 0$ onto which P projects (here $Q = \mathbb{1} - P$ and $z = O(\Omega^2)$). Qualitatively, at this order, by action of H_α then of H_β on $|\psi_\alpha^{00}\rangle|0\rangle|0\rangle$ (with the obvious notation $H = H_\alpha + H_\beta$), we virtually create an excitation β alone, relaxing at the rate $\gamma_\beta/2$, hence the additional adiabaticity condition $\Gamma_0 \ll \gamma_\beta$; joined to $\Gamma_0 \ll \kappa$ and $\gamma_f < \gamma_m$, it implies $\Gamma_{\text{ex}} \ll \gamma_f$ since $\Gamma_{\text{ex}}/\gamma_f = (\Gamma_0/\kappa + \Gamma_0/\gamma_\beta)(4\gamma_\beta/\gamma_m) < 16(\Gamma_0/\kappa + \Gamma_0/\gamma_\beta)$. Quantitatively, we find a correction to the coefficient of P_α^2 in H_{eff}^{00} of type $H_\alpha G_0 H_\beta G_0 H_\beta G_0 H_\alpha$ (G_0 is the resolvent of H_{eff} for $\Omega = 0$) of the form $\hbar \Gamma_{\text{ex}} \Omega_\beta^2 / \gamma_\beta \kappa$, which must be negligible, which imposes $\Omega_\beta^2 / \gamma_\beta \kappa \ll 1$, i.e. $\Gamma_{\text{ex}} \ll \gamma_f$ taking into account $\gamma_f < \gamma_m$. The corrections to the scalar term are negligible as soon as $\Gamma_0 \ll \gamma_\beta, \kappa$, and the new term in P_α^4 which appears is negligible compared to $\hbar \Gamma_{\text{ex}} P_\alpha^2$ for $P_\alpha = O(1)$ if $\Gamma_{\text{ex}} \ll \kappa$.

jump, which occurs at time t with a rate $\kappa(\langle\psi_\alpha^{11}|\psi_\alpha^{11}\rangle + \langle\psi_\alpha^{01}|\psi_\alpha^{01}\rangle)_{\text{adiab}}/\langle\psi_\alpha^{00}|\psi_\alpha^{00}\rangle$. Just after the jump, the state vector, initially in the adiabatic following regime, becomes

$$|\psi(t^+)\rangle = C_c|\psi(t^-)\rangle_{\text{adiab}} \propto |\psi_\alpha^{01}(t^-)\rangle_{\text{adiab}}|0\rangle|0\rangle + |\psi_\alpha^{11}(t^-)\rangle_{\text{adiab}}|1\rangle|0\rangle \quad (33)$$

It is the superposition of an unstable component $|1\rangle|0\rangle$ and of a stable component $|0\rangle|0\rangle$. With a probability $\langle\psi_\alpha^{11}|\psi_\alpha^{11}\rangle_{\text{adiab}}/(\langle\psi_\alpha^{01}|\psi_\alpha^{01}\rangle + \langle\psi_\alpha^{11}|\psi_\alpha^{11}\rangle)_{\text{adiab}}$ the cavity jump is then followed by a metastability exchange jump before the system state vector has time to reach its adiabatic value. In this case, we have a ‘‘double jump’’, which ultimately does not affect the component $|\psi_\alpha^{00}(t^-)\rangle$ since

$$C_\beta C_c |\psi(t^-)\rangle_{\text{adiab}} \propto |\psi_\alpha^{00}(t^-)\rangle|0\rangle|0\rangle \quad (34)$$

This process contributes to the scalar term (proportional to the identity) in the effective Hamiltonian of equation (31). With the complementary probability $\langle\psi_\alpha^{01}|\psi_\alpha^{01}\rangle_{\text{adiab}}/(\langle\psi_\alpha^{01}|\psi_\alpha^{01}\rangle + \langle\psi_\alpha^{11}|\psi_\alpha^{11}\rangle)_{\text{adiab}}$ the state vector returns to its adiabatic value before other jumps occur, and is slaved to $|\psi_\alpha^{(01)}(t^-)\rangle_{\text{adiab}} \propto P_\alpha |\psi_\alpha^{00}(t^-)\rangle$, that is, the slow component $|\psi_\alpha^{00}(t^-)\rangle$ has effectively undergone a single quantum jump with a jump operator proportional to P_α . This process corresponds to the first term, proportional to P_α^2 , in the effective Hamiltonian of equation (31). (ii) Suppose next that the jump at time t is a metastability exchange jump, which occurs with a rate $\gamma_\beta \langle\psi_\alpha^{11}|\psi_\alpha^{11}\rangle_{\text{adiab}}/\langle\psi_\alpha^{00}|\psi_\alpha^{00}\rangle$. We verify in this case that the state vector after the jump, $C_\beta |\psi(t^-)\rangle$, is entirely unstable and almost immediately undergoes a second jump, a cavity jump. The total effect corresponds here again to a double jump and to the action of a scalar operator on the slow component. We derive from this discussion the following single jump and double jump rates :

$$\Gamma_s = \frac{\kappa(\langle\psi_\alpha^{11}|\psi_\alpha^{11}\rangle + \langle\psi_\alpha^{01}|\psi_\alpha^{01}\rangle)_{\text{adiab}}}{\langle\psi_\alpha^{00}|\psi_\alpha^{00}\rangle} \frac{\langle\psi_\alpha^{01}|\psi_\alpha^{01}\rangle_{\text{adiab}}}{(\langle\psi_\alpha^{01}|\psi_\alpha^{01}\rangle + \langle\psi_\alpha^{11}|\psi_\alpha^{11}\rangle)_{\text{adiab}}} = \Gamma_{\text{ex}} \frac{\langle\psi_\alpha^{00}|P_\alpha^2|\psi_\alpha^{00}\rangle}{\langle\psi_\alpha^{00}|\psi_\alpha^{00}\rangle} \equiv \Gamma_{\text{ex}}\langle P_\alpha^2 \rangle \quad (35)$$

$$\Gamma_d = \frac{\kappa(\langle\psi_\alpha^{11}|\psi_\alpha^{11}\rangle + \langle\psi_\alpha^{01}|\psi_\alpha^{01}\rangle)_{\text{adiab}}}{\langle\psi_\alpha^{00}|\psi_\alpha^{00}\rangle} \frac{\langle\psi_\alpha^{11}|\psi_\alpha^{11}\rangle_{\text{adiab}}}{(\langle\psi_\alpha^{01}|\psi_\alpha^{01}\rangle + \langle\psi_\alpha^{11}|\psi_\alpha^{11}\rangle)_{\text{adiab}}} + \frac{\gamma_\beta \langle\psi_\alpha^{11}|\psi_\alpha^{11}\rangle_{\text{adiab}}}{\langle\psi_\alpha^{00}|\psi_\alpha^{00}\rangle} = \Gamma_0 \quad (36)$$

We finally obtain the one-mode quantum master equation describing the slow evolution of the density operator ρ_α of the bosonic mode α (hybridized but almost purely nuclear spin):

$$\boxed{\frac{d\rho_\alpha}{dt} = C_s \rho_\alpha C_s^\dagger - \frac{1}{2}\{C_s^\dagger C_s, \rho_\alpha\} + C_d \rho_\alpha C_d^\dagger - \frac{1}{2}\{C_d^\dagger C_d, \rho_\alpha\}} \quad (37)$$

in terms of two quantum jumps, the single jump (cavity only) C_s and the double jump (of cavity and metastability exchange in that order or in the other) C_d :

$$C_s = \sqrt{\Gamma_{\text{ex}}}\mathbb{P}_\alpha \quad ; \quad C_d = \sqrt{\Gamma_0}\mathbb{1} \quad (38)$$

By solving equation (37) for the empty initial state of α , we get:

$$\langle X_\alpha^2 \rangle = \frac{1}{4}(1 + \Gamma_{\text{ex}}t) \quad ; \quad \langle P_\alpha^2 \rangle = \frac{1}{4} \quad (39)$$

which effectively designates Γ_{ex} as a rate of creation of excitations in the α mode. Going back to the initial atomic basis (unrotated) and by limiting the state vector (29) to its first term, we recover equation (21) and the first three results of equation (22) of the three-mode model, yet valid at any Faraday coupling Ω , not necessarily infinitesimal. Finally, the average number of photons polarized along y leaking out of the cavity per unit of time, given in the one-mode model by $\Gamma_0 + \Gamma_{\text{ex}}/4$ as shown by equation (45), agrees with the exact value $\kappa\langle c^\dagger c \rangle_s$, where the mean stationary number of y -polarized photons in the cavity $\langle c^\dagger c \rangle_s = \langle X_c^2 \rangle_s - 1/4$ is the last result of (22).¹¹

11. On the other hand, the value of $\langle c^\dagger c \rangle_{\text{adiab}}$ in the adiabatic form (30) of the state vector does not represent this number. The solution of the paradox is due to the existence of the de-excitation path (ii), that of the annihilation in the first jump of the excitation $n_\beta = 1$ in the metastable mode immediately followed by the loss of a cavity photon. The true output rate of y -polarized photons is therefore $\kappa\langle c^\dagger c \rangle_{\text{adiab}} + \gamma_\beta \langle \beta^\dagger \beta \rangle_{\text{adiab}}$.

4. Continuous non-demolition quantum measurement of nuclear spin

The quantum averages calculated in section 3 correspond to the ensemble averages over an infinite number of realizations of the experiment. In this section we study what really interests us, the evolution of the system, in one or more given realizations, conditioned on the results of a continuous measurement on the y-polarized light leaking out of the cavity. For this, we return to the formulation in terms of Monte Carlo wave functions, as in section 3, where stochastic trajectories $|\psi(t)\rangle$ corresponding to a particular sequence of quantum jumps reconstruct the density operator of the system conditioned on measurement results [28]. The precise form of the Monte Carlo jump operators, which is not unique in the stochastic reformulation of a quantum master equation, is then determined by the particular measurements made.

4.1. Squeezing by photon counting

Suppose that we continuously and directly count (by photodetection) the number of y-polarized photons leaking out of the cavity (see figure 1b), as proposed in reference [29]. The jump operator associated with this measurement is \sqrt{kc} , so the three-mode quantum master equation (20) is already in the right form to analyze the evolution of the state vector $|\psi(t)\rangle$ conditioned on the measurement.

The same is true within the limit of a weak Faraday coupling, $\Omega \rightarrow 0$, which leads to the one-mode model of section 3.4. As the jump operators C_d and C_s of the quantum master equation (37) both correspond to the cavity loss of a y-polarized photon (remember, C_d results from a cavity jump immediately followed or preceded by a metastability exchange jump, and C_s from a single cavity jump), the measurement cannot distinguish between the two, and the density operator conditioned on a given number n of detected photons is obtained by averaging over realizations having this same *total* number n of jumps. An unnormalized Monte Carlo state vector having undergone such n jumps during t is written

$$|\psi(t)\rangle = e^{-\frac{i}{\hbar} H_{\text{eff}}^{00}(t-t_n)} C_{\epsilon_n} e^{-\frac{i}{\hbar} H_{\text{eff}}^{00}(t_n-t_{n-1})} C_{\epsilon_{n-1}} \dots C_{\epsilon_1} e^{-\frac{i}{\hbar} H_{\text{eff}}^{00} t_1} |\psi(0)\rangle \quad (40)$$

where $\epsilon_k \in \{s, d\}$ and t_k are the type and time of the k th jump, H_{eff}^{00} is the effective Hamiltonian (31) and we denote the state vector of the one-mode model $|\psi\rangle$ rather than $|\psi_\alpha^{00}\rangle$ to simplify. The quantum average of an observable O is obtained by averaging over all possible trajectories, therefore by summing over the number and type of jumps and by integrating over their times:

$$\langle O \rangle(t) = \sum_n \int_{0 < t_1 < t_2 < \dots < t_n < t} dt_1 dt_2 \dots dt_n \sum_{(\epsilon_k)_{1 \leq k \leq n} \in \{s, d\}^n} \langle \psi(t) | O | \psi(t) \rangle \quad (41)$$

where the squared norm of each unnormalized state vector $|\psi(t)\rangle$ automatically gives its probability density [30]. By taking $O = \mathbb{1}$, we deduce the probability that n jumps occurred in the time interval $[0, t]$:

$$\Pi_n(t) = \int_{0 < t_1 < t_2 < \dots < t_n < t} dt_1 dt_2 \dots dt_n \sum_{(\epsilon_k)_{1 \leq k \leq n} \in \{s, d\}^n} \langle \psi(t) | \psi(t) \rangle \quad (42)$$

To evaluate (42), we take advantage of the fact that all the jump operators in (40) and their Hermitian conjugates commute with each other and with H_{eff}^{00} .¹² By using the identities

$$\sum_{\epsilon_n = s, d} \dots \sum_{\epsilon_1 = s, d} (C_{\epsilon_n}^\dagger C_{\epsilon_n} \dots C_{\epsilon_1}^\dagger C_{\epsilon_1}) = \left(\sum_{\epsilon_n = s, d} C_{\epsilon_n}^\dagger C_{\epsilon_n} \right) \dots \left(\sum_{\epsilon_1 = s, d} C_{\epsilon_1}^\dagger C_{\epsilon_1} \right) = (\Gamma_{\text{ex}} P_\alpha^2 + \Gamma_0 \mathbb{1})^n \quad (43)$$

and by injecting a closure relation in the eigenbasis of P_α such as $P_\alpha |p_\alpha\rangle = p_\alpha |p_\alpha\rangle$, after having integrated over the times t_k as allowed by the telescopic product of the evolution operators, we obtain

$$\Pi_n(t) = \frac{t^n}{n!} \int_{-\infty}^{+\infty} dp_\alpha (\Gamma_{\text{ex}} p_\alpha^2 + \Gamma_0)^n e^{-\Gamma_{\text{ex}} p_\alpha^2 t} e^{-\Gamma_0 t} \Pi(p_\alpha, 0) = \binom{2n}{n} \frac{(\Gamma_{\text{ex}} t / 8)^n e^{-\Gamma_0 t}}{(1 + \Gamma_{\text{ex}} t / 2)^{n+1/2}} \Phi\left(-n, \frac{1}{2} - n; \Gamma_0 t + \frac{2\Gamma_0}{\Gamma_{\text{ex}}}\right) \quad (44)$$

¹². For this reason, keeping the information on the jump times does not allow to increase the efficiency of spin squeezing by post-selection. Indeed, the density operator $\rho_\alpha(t)|_{t_1, \dots, t_n}$ knowing that n jumps occurred at times t_1, \dots, t_n leads to the same probability distribution of P_α as the density operator $\rho_\alpha(t)|_n$ knowing only that n jumps occurred during $[0, t]$.

where $\Pi(p_\alpha, 0)$ is the initial probability distribution of p_α (a Gaussian with zero mean and variance $1/4$) and Φ is Kummer's hypergeometric confluent function ${}_1F_1$. We notice that (44) is in fact a Gaussian average on p_α of a Poisson distribution with parameter $\lambda = (\Gamma_{\text{ex}} p_\alpha^2 + \Gamma_0)t$. We deduce the mean and the variance of the number of photodetections during t :

$$\langle n \rangle = \left(\Gamma_0 + \frac{1}{4} \Gamma_{\text{ex}} \right) t \quad ; \quad \text{Var } n = \langle n \rangle + \frac{(\Gamma_{\text{ex}} t)^2}{8} \quad (45)$$

Still using equation (44), we access the probability distribution of p_α knowing that n photons were detected in the time interval $[0, t]$, an even function of p_α :

$$\Pi_t(p_\alpha | n) = \frac{1}{\Pi_n(t)} \frac{t^n}{n!} (\Gamma_{\text{ex}} p_\alpha^2 + \Gamma_0)^n e^{-\Gamma_{\text{ex}} p_\alpha^2 t} e^{-\Gamma_0 t} \Pi(p_\alpha, 0) \quad (46)$$

As expected, it is an even function of p_α , as photodetection only gives access to the outgoing y-polarized field intensity and cannot distinguish between opposite $\pm p_\alpha$ values of the P_α quadrature of the hybridized nuclear spin along z . This results in the squeezing of P_α^2 fluctuations rather than P_α , which we characterize by the conditional mean and variance of P_α^2 knowing that n photons were detected during t , deduced from (46):

$$\langle P_\alpha^2 \rangle_n = \frac{(n+1) \Pi_{n+1}(t)}{\Gamma_{\text{ex}} t \Pi_n(t)} - \frac{\Gamma_0}{\Gamma_{\text{ex}}} \quad ; \quad \text{Var}_n(P_\alpha^2) \equiv \langle P_\alpha^4 \rangle_n - \langle P_\alpha^2 \rangle_n^2 = \frac{(n+1)^2}{(\Gamma_{\text{ex}} t)^2} \left[\frac{(n+2) \Pi_{n+2}(t)}{(n+1) \Pi_n(t)} - \frac{\Pi_{n+1}^2(t)}{\Pi_n^2(t)} \right] \quad (47)$$

Finally, using equation (46), we find for $\Gamma_{\text{ex}} t \rightarrow +\infty$ that the probability distribution of p_α^2 conditioned on the number n of photodetections is peaked around a value p_0^2 with a conditional variance tending to zero¹³:

$$\boxed{p_0^2 - \frac{1}{4} = \frac{n - \langle n \rangle}{\Gamma_{\text{ex}} t} \quad \text{hence} \quad \langle P_\alpha^2 \rangle_n \underset{\Gamma_{\text{ex}} t \rightarrow +\infty}{\sim} p_0^2} \quad ; \quad \boxed{\text{Var}_n(P_\alpha^2) \underset{\Gamma_{\text{ex}} t \rightarrow +\infty}{\sim} \frac{n}{(\Gamma_{\text{ex}} t)^2} \rightarrow 0} \quad (48)$$

By replacing in this expression n by its mean value and taking into account the value $1/8$ of the variance of P_α^2 in the initial state, we end up with the nuclear spin squeezing rate by photon counting:

$$\boxed{\Gamma_{\text{sq}} = \frac{\Gamma_{\text{ex}}^2}{8(\Gamma_0 + \frac{1}{4} \Gamma_{\text{ex}})}} \quad (49)$$

Correspondingly, the conditional probability distribution of p_α has two peaks at $\pm p_0$ as visible on the Wigner function in figure 5b, obtained by numerical simulation of the conditional evolution of the system over long times in the one-mode model (37).¹⁴ This clearly shows that, in a single realization of the experiment, the continuous photodetection

13. According to equation (45), the right-hand side of the first equation in (48) is asymptotically of the order of unity for a typical photodetection sequence. This equation in fact only makes sense for p_0^2 positive therefore $n > \Gamma_0 t$; then, the equivalents (48) apply when the gap between the two peaks in $\Pi_t(p_\alpha | n)$ is much larger than their width, which imposes $2p_0^2 \gg n^{1/2}/\Gamma_{\text{ex}} t = (\Gamma_0 + \Gamma_{\text{ex}} p_0^2)^{1/2}/\Gamma_{\text{ex}} t^{1/2}$. To obtain them, we pose $n = \gamma t$ with $\gamma > \Gamma_0$, then we write (46) in the form $\exp[-tS(p_\alpha)]\Pi(p_\alpha, 0)$ and we quadratize $S(p_\alpha)$ around its minima.

14. The absence of fringes shows that we have prepared a statistical mixture rather than a coherent superposition of two squeezed states of the P_α quadrature. We find indeed from equation (41) that $\langle p_0 | \rho_n(t) | p_0 \rangle / \langle p_0 | \rho_n(t) | p_0 \rangle = [(\Gamma_0 - \Gamma_{\text{ex}} p_0^2)/(\Gamma_0 + \Gamma_{\text{ex}} p_0^2)]^n \simeq \exp(-2\Gamma_{\text{ex}} t p_0^2) \lll 1$ on figure 5b, $\rho_n(t)$ being the conditional density operator. The Laplace method gives at long times the conditional Wigner distribution $W_t(x_\alpha, p_\alpha | n) \sim \pi \Pi_t(p_\alpha | n) \exp[-2x_\alpha^2/\Gamma_{\text{ex}} t s(p_\alpha)] / \sqrt{\pi \Gamma_{\text{ex}} t s(p_\alpha)/2}$ with $s(p_\alpha) = [1 + (\Gamma_0 + \Gamma_{\text{ex}} p_0^2)/(\Gamma_0 + \Gamma_{\text{ex}} p_\alpha^2)]/2$. We notice that $s(\pm p_0) = 1$ and $s(p_\alpha) \simeq 1$ for $\Gamma_0 \gg \Gamma_{\text{ex}}$. While $W_t(0, 0|n)$ is damped exponentially in time (in the limit $\Gamma_{\text{ex}} \ll \Gamma_0$, it comes $W_t(0, 0|n) \sim (\Gamma_{\text{ex}}/\Gamma_0)^{1/2} p_0 \exp(2p_0^2) \exp(-4p_0^4 \Gamma_{\text{sq}} t)$), we also have $W_t(0, 0|n) = \langle 2\varepsilon \rangle_n$ where $\varepsilon = \pm 1$ is the parity of the Monte Carlo wave function $\psi(p_\alpha, t)$. In a numerical simulation, we therefore have only a slow decay $\langle \varepsilon \rangle_n \simeq 1/\sqrt{\mathcal{N}_n}$ where \mathcal{N}_n is the number of trajectories that have undergone n jumps during t ; this leads to unphysical fringes with negative values in the Wigner distribution near x_α axis. To minimize this effect and make it imperceptible at a not too high p_α resolution ($dp_\alpha = 0.044$ on figure 5b), we stop the Monte Carlo simulation at a stage where there is exactly the same number $\mathcal{N}_n/2$ of even and odd wave functions. To obtain a coherent superposition of squeezed states, one would have to perform an additional post-selection, restricting oneself to Monte Carlo realizations of wavefunction $\psi(p_\alpha, t)$ of fixed parity ε (having undergone an even number of single jumps if $\varepsilon = 1$, an odd number otherwise, the jump operator C_s changing the parity). In the corresponding conditional density operator, one then has $\langle p_0 | \rho_{n,\varepsilon}(t) | p_0 \rangle / \langle p_0 | \rho_{n,\varepsilon}(t) | p_0 \rangle = \overline{\psi(-p_0, t) \psi(p_0, t)^{n\varepsilon}} / \overline{\psi(p_0, t) \psi(p_0, t)^{n\varepsilon}} = \varepsilon$ without the two-peak structure of the P_α distribution being affected at long times because $\Pi_t(p_\alpha | n, \varepsilon) / \Pi_t(p_\alpha | n) \propto 1 + \varepsilon [(\Gamma_0 - \Gamma_{\text{ex}} p_\alpha^2)/(\Gamma_0 + \Gamma_{\text{ex}} p_\alpha^2)]^n \rightarrow 1$ when $n \rightarrow +\infty$ at fixed nonzero p_α . The Wigner distribution $W_t(x_\alpha, p_\alpha | n, \varepsilon)$ now exhibits positive and negative fringes of maximum amplitude 2 on the $p_\alpha = 0$ axis. This filtering technique also overcomes the decoherence mechanisms of section 4.2.4, because the Monte Carlo wavefunctions remain of well-defined parity after the action of the corresponding jump operator $\gamma_\alpha^{1/2} \alpha$. This idea of controlling decoherence through parity measurements is well known in cavity quantum electrodynamics [31].

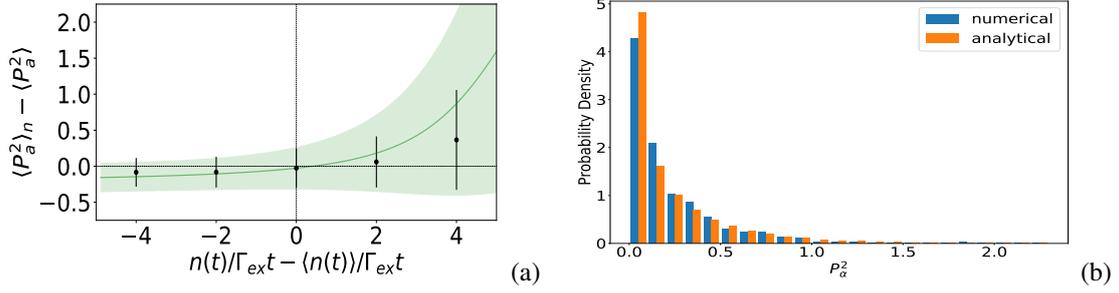


Figure 4 – Squeezing of P_a^2 by photon counting at short times: $\Gamma_{\text{ext}} t = 15$, where Γ_{ex} is the rate (32) of creation of excitations in the hybridized nuclear bosonic mode α . (a) Conditional mean and standard deviation of the squared nuclear spin quadrature P_a^2 knowing that n photons were detected in the time interval $[0, t]$, as functions of this number n . The standard deviation is plotted as a confidence interval. The unconditional mean $\langle P_a^2 \rangle = 1/4$ is independent of time, see equation (21). Black dots and error bars: numerical simulation of the 3-mode model with 3000 realizations; green line and colored area: analytical predictions taken from equations (44), (47) and (50) of the one-mode model. In practice, the black points are obtained after averaging over classes of values of n centered on these points (in a given class, the trajectories have close photodetection numbers n but independent histories for the metastability exchange jumps which the experimenter cannot access). 3-mode model parameters: $\Omega/\kappa = 1/3$, $\gamma_m/\kappa = 1/10$, $\gamma_f/\kappa = 1/1000$ (so that $\Gamma_{\text{ex}}/\kappa = 1/909$), $n_a^{\text{max}} = 64$, $n_b^{\text{max}} = n_c^{\text{max}} = 8$. This corresponds to $\Gamma_0/\Gamma_{\text{ex}} = 12\,500/601 \simeq 20.8$ and $\Gamma_{\text{sq}}/\Gamma_{\text{ex}} = 601/101\,202 \simeq 1/168$ where Γ_{sq} is the squeezing rate (49). (b) For the class centered on $n = \langle n(t) \rangle$, histogram of the conditional values of P_a^2 . Blue bars: numerical simulation of the three-mode model; orange bars: analytical predictions taken from equation (46) of the one-mode model.

of the y -polarized photons leaking out of the cavity makes more and more certain the value of P_a^2 , and therefore to a large extent of I_z^2 , the square of the component along z of the collective nuclear spin, as we clearly see by relating, in the limit $\Omega \rightarrow 0$, the conditional moments of P_a^2 , that is of I_z^2 to those of P_a^2 :

$$\langle P_a^2 \rangle_n = \frac{\gamma_m}{\gamma_f + \gamma_m} \langle P_a^2 \rangle_n + \frac{\gamma_f/4}{\gamma_f + \gamma_m} \quad ; \quad \text{Var}_n(P_a^2) = \frac{\gamma_m^2}{(\gamma_f + \gamma_m)^2} \text{Var}_n(P_a^2) + \frac{\gamma_f \gamma_m}{(\gamma_f + \gamma_m)^2} \langle P_a^2 \rangle_n + \frac{\gamma_f^2/8}{(\gamma_f + \gamma_m)^2} \quad (50)$$

Since the squeezing is on P_a^2 rather than P_a , the conditional angular distribution of the collective nuclear spin is bimodal (it has, like that of P_a , two well-separated peaks provided $\gamma_f/\gamma_m \ll (2p_0)^2$); these structures, which are narrower than the standard quantum limit, still allow for a more accurate angular pointing than in the unsqueezed state. We therefore redefine metrological gain (13) by replacing in the right-hand side of this equation the conditional variance of P_a by the square of the halfwidth δP_a of the peaks centered at $\pm P_{a,0}$ of the conditional probability distribution of P_a , then equating the center $P_{a,0}$ and the width $\delta(P_a^2)$ of the distribution of P_a^2 with the conditional mean and standard deviation of P_a^2 :

$$\xi^2 = \frac{4(\delta P_a)^2}{\eta} = \frac{(2P_{a,0}\delta P_a)^2}{\eta P_{a,0}^2} = \frac{[\delta(P_a^2)]^2}{\eta P_{a,0}^2} = \frac{\text{Var}_n(P_a^2)}{\eta \langle P_a^2 \rangle_n} \quad (51)$$

Note that this expression cannot be deduced from the method of moments explained in section II.B.6 of reference [4] taking P_a^2 as estimator, because the effect of the unitary transformation $\exp(2i\theta X_a)$ (in practice, a precession of the nuclear spin around a magnetic field along y) is not to shift the peak in the distribution of P_a^2 but to split it into two peaks centered at $(P_{a,0} \pm \theta)^2$.

Finally, we carry out a numerical verification of these analytical predictions in the three-mode model. In figure 4a, we plot the conditional mean of the square P_a^2 of the nuclear spin quadrature knowing that n photodetections occurred in the time interval $[0, t]$, with $\Gamma_{\text{ext}} t = 15$ (black dots), depending on this number n . The ensemble of realizations is divided into 5 classes corresponding to a number of photodetections falling within a given interval, and the black dots are obtained by averaging over the realizations in the same class. The numerical results are close to the analytical predictions taken from (47) and (50) and plotted in green, except in the extreme classes which include a too low number of realizations. On the other hand, the asymptotic analytical predictions (48), not shown, would be in

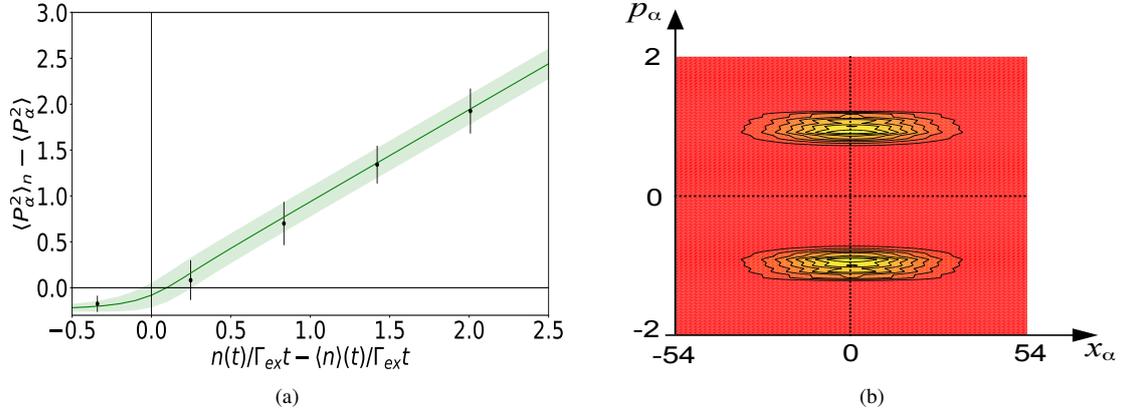


Figure 5 – Squeezing of P_α^2 by photon counting at long times, $\Gamma_{\text{ext}}t = 1000$, in the one-mode model (37) (this long time would make a simulation in the three-mode model more difficult). (a) Conditional mean and standard deviation of P_α^2 knowing that the number of photodetections n falls in a given class of values, similarly to figure 4a, for 2000 realizations. (b) Wigner distribution of the hybridized nuclear bosonic mode in the quadrature space (X_α, P_α) at $\Gamma_{\text{ext}}t = 1000$, obtained by averaging the dyads $|\psi(t)\rangle\langle\psi(t)|$ whose number of photodetections falls in the 3rd class of (a) (302 trajectories out of 5000 realizations). It has two peak lines but no interference fringes (see note 14).

disagreement with the simulations of the two models because the time $t = 15/\Gamma_{\text{ex}}$ is not long enough, it is much less than the squeezing time $1/\Gamma_{\text{sq}}$. In figure 4b, we plot the conditional probability distribution of P_α^2 corresponding to the central class of figure 4a; there is also good agreement between one-mode analytical calculation and three-mode numerical simulation. In figure 5, we are precisely exploring long times in the one-mode model, with $\Gamma_{\text{ext}}t = 1000$ that is $\Gamma_{\text{sq}}t \simeq 5.94$. Figure 5a, which is the equivalent of figure 4a, shows that $\langle P_\alpha^2 \rangle_n$ is then related to the number of photodetections n as in the analytical prediction (48), i.e. according to the internal bisector in the units of the figure, with a conditional standard deviation (48) roughly constant $\simeq (\Gamma_0 t)^{1/2} / \Gamma_{\text{ext}}t \simeq 1/(\Gamma_{\text{sq}}t)^{1/2}$ because Γ_0 is here $\gg \Gamma_{\text{ex}}$.

4.2. Squeezing by homodyne detection

We now assume that the y-polarized photons leaking out of the cavity are continuously measured by homodyne detection [32], as in figure 1c. We must first find the stochastic equations giving the evolution of the system state vector conditioned on homodyne detection, since the jump operators appearing naturally in (25) or (37) of the three-mode or one-mode quantum master equation are unsuitable. We then present some analytical results obtained in the one-mode model and then in the three-mode model, before briefly discussing the effect of the finite coherence time of metastable atoms. In order to obtain these results, we used the fact that, for the vacuum initial state considered here, the conditional state vector is given exactly at all times by a Gaussian ansatz [33], whatever the number of modes of the model, in presence or absence of decoherence.

4.2.1. Suitable stochastic formulation of the quantum master equation

A general quantum master equation of the Lindblad form [34]

$$\frac{d\rho}{dt} = \frac{1}{i\hbar} [H, \rho] + \sum_m C_m \rho C_m^\dagger - \frac{1}{2} \{C_m^\dagger C_m, \rho\} \quad (52)$$

with H the Hermitian part of the Hamiltonian and C_m the jump operators, can be rewritten in an equivalent way by adding an arbitrary constant to the jump operators and/or by mixing them by any linear unitary combination. In order to take into account a homodyne detection on the outgoing field, we form, from a jump operator C_m corresponding to a photodetection, the two ‘‘homodyne’’ jump operators $D_{m,\pm}$ [28]

$$D_{m,+} = \frac{\mu\mathbb{1} + C_m}{\sqrt{2}} \quad ; \quad D_{m,-} = \frac{\mu\mathbb{1} - C_m}{\sqrt{2}} \quad (53)$$

where μ^2 has the dimensions of a frequency. The measurement of the difference in the jump rates $D_+^\dagger D_+ - D_-^\dagger D_-$ then gives access to a quadrature of C_m . Thus, for μ real and C_m corresponding to the cavity jump operator C_c , see equation (28), the difference between the numbers of photons N_\pm detected during the short time interval Δt in the two output channels of figure 1c, which by definition constitutes the homodyne signal,

$$N_+ = (D_{c,+}^\dagger D_{c,+}) \Delta t \quad ; \quad N_- = (D_{c,-}^\dagger D_{c,-}) \Delta t \quad ; \quad \frac{N_+ - N_-}{2\mu} = \frac{c + c^\dagger}{2} \sqrt{\kappa} \Delta t \quad (54)$$

gives access to X_c ; it is indeed this quadrature of the field, conjugated to P_c therefore translated by a quantity proportional to P_b and to the time under the action of the Hamiltonian H (12), which provides information on P_a through metastability exchange collisions. In the case of the quantum master equation with 3 modes (25), one has to apply the doubling procedure (53) *a priori* only to the jump operator of the cavity. In practice, we will apply it also to the jump operator C_β , that is we will double by homodyning *all* the jump operators C_m , in order to avoid the discomfort of a hybrid representation mixing discrete quantum jumps and continuous stochastic evolution, see equation (55) to come. In the case of the one-mode quantum master equation (37), we need to “homodyne” the two jump operators C_s and C_d anyway, since each of them comes with the loss of a photon in a cavity, as explained in section 3.4.

Within the limit of a large amplitude of the local oscillator μ , we can act as if Δt were infinitesimal¹⁵ and represent the evolution of the Monte Carlo wave function, now normalized to unity, by a continuous nonlinear stochastic equation without quantum jumps [28, 35, 36] in Ito point of view:

$$\begin{aligned} d|\phi(t)\rangle = & -\frac{i}{\hbar} H|\phi(t)\rangle dt - \frac{1}{2} \sum_m \left(C_m^\dagger C_m - \langle \phi(t) | C_m + C_m^\dagger | \phi(t) \rangle C_m + \frac{1}{4} \langle \phi(t) | C_m + C_m^\dagger | \phi(t) \rangle^2 \right) |\phi(t)\rangle dt \\ & + \sum_m \left(C_m - \frac{1}{2} \langle \phi(t) | C_m + C_m^\dagger | \phi(t) \rangle \right) |\phi(t)\rangle d\zeta_m(t) \end{aligned} \quad (55)$$

where, to each jump operator C_m in the initial quantum master equation, we associate a continuous-time stochastic process $d\zeta_m(t)$, with real values, Gaussian, of zero mean, of variance dt , statistically independent of other processes and without memory. At the same level of approximation, the homodyne signal operator (54) is replaced by the sum of its average and a classical noise representing its fluctuations, which is none other than the corresponding $d\zeta_m$ [28]:

$$\frac{N_+ - N_-}{2\mu} = \frac{\sqrt{\kappa} \langle \phi | c + c^\dagger | \phi \rangle}{2} dt + \frac{1}{2} d\zeta_c \quad (56)$$

In practice, more than the homodyning history, that is the detailed time dependence of the homodyne detection signal, it is its time average over an interval of time $[0, t]$ which is easily accessible in an experiment and allows a post-selection of states (i.e. experimental realizations) in a set of non negligible statistical weight. We thus introduce the integrated signal having the dimension of the root of a frequency,

$$\sigma(t) \equiv \frac{N_+^{\text{tot}} - N_-^{\text{tot}}}{2\mu t} = \frac{1}{t} \int_0^t dt' \left[\sqrt{\kappa} \langle \phi(t') | X_c | \phi(t') \rangle + \frac{1}{2} \frac{d\zeta_c(t')}{dt'} \right] \quad (57)$$

and we will calculate in the following the mean and the variance of the quadrature P_a of the nuclear spin conditioned on σ .¹⁶

15. This approximation is valid for a time resolution, or a time step Δt , such that $\mu^{-2} \ll \Delta t \ll \kappa^{-1}$, where κ is in practice the fastest evolution rate in the system in the experiment.

16. Recall that the mean and the variance of an observable (here P_a) in a single realization $|\phi(t)\rangle$ of the stochastic equation, thus of the experiment, have in general no physical meaning, because there is no possible measurement giving the mean value of an observable in a single realization, on the contrary it is necessary to average over a large number of realizations of the experiment having evolved during t starting from the same initial pure case or density operator. A counter-example corresponds to the case where one can, by continuous measurements, trace back the time dependence of all the stochastic processes $d\zeta_m$; this is the case of the one-mode model subjected to a homodyne detection, the time dependence of the signal (56) fixing that of the single stochastic process $d\zeta_c$. This would then allow, in principle, to select, among a large number of realizations of the experiment, those leading to the chosen $|\phi(t)\rangle$ state, and to deduce the expectation values of observables in $|\phi(t)\rangle$; in practice, this would be unrealistic, given the infinitesimal statistical weight of the realizations to keep.

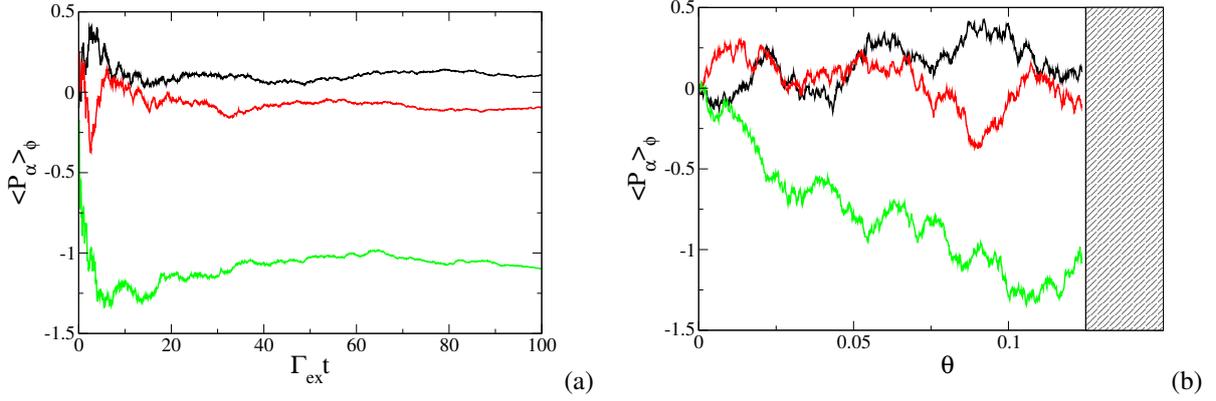


Figure 6 – In the case of squeezing by continuous homodyne detection: random walk (61) performed in the one-mode model by the quantum average of the quadrature P_α of the nuclear spin in a given realization of the experiment. (a) Quantum average as a function of true time t reduced by the rate (32) of creation of excitations for three realizations of the experiment; it is a stretched Brownian motion converging at long times towards a fixed but unpredictable value. (b) Idem as a function of the compact renormalized time θ (62); this time it is an ordinary Brownian motion but limited to $\theta \leq 1/8$.

4.2.2. Analytical results in the one-mode model

Let us explicitly write the stochastic equation (55) for the one-mode model (37):

$$d|\phi(t)\rangle = -\frac{dt}{2}\Gamma_{\text{ex}}[P_\alpha - \bar{P}_\alpha(t)]^2|\phi(t)\rangle + \sqrt{\Gamma_{\text{ex}}}\,d\zeta_s(t)[P_\alpha - \bar{P}_\alpha(t)]|\phi(t)\rangle \quad (58)$$

with $\bar{P}_\alpha(t) \equiv \langle\phi(t)|P_\alpha|\phi(t)\rangle$. The highlight is that the jump operator C_d proportional to the identity, which added noise in the photon counting detection scheme of section 4.1, gives no contribution and completely disappears in the homodyne case. Indeed, the photons emitted during these jumps come from the component $|1\rangle|1\rangle$ of the state vector (29) containing one excitation β , which makes them optically incoherent with the light field injected into the cavity, i.e. with the component $|0\rangle|0\rangle$ of (29), in the sense that $|1\rangle|1\rangle$ contributes to $\langle c^\dagger c \rangle$ but not to $\langle c + c^\dagger \rangle$. So only the stochastic process $d\zeta_s$ associated with the jump operator C_s remains. This process coincides with the one $d\zeta_c$ appearing in the homodyne detection signal (56), $d\zeta_s \equiv d\zeta_c$, a fact admitted here but which will be derived in section 4.2.3.

The stochastic equation (58) exhibits a linear noise term and a quadratic deterministic term in the operator P_α , real in Fourier space. For the initial state considered here, it is thus solved exactly by a Gaussian ansatz on the wave function in momentum representation, real and correctly normalized for the commutation relation $[X_\alpha, P_\alpha] = i/2$:

$$\langle p_\alpha|\phi(t)\rangle = [2\pi u(t)]^{1/4} \exp\{-u(t)[p_\alpha - \bar{P}_\alpha(t)]^2\} \quad (59)$$

On the other hand, the Gaussianity is lost in the squeezing by photodetection protocol of section 4.1. Using the Ito calculation,¹⁷ we find that u follows a deterministic evolution equation, to be integrated with the initial condition $u(0) = 1$:

$$du(t) = \Gamma_{\text{ex}} dt \quad \text{so that} \quad u(t) = 1 + \Gamma_{\text{ex}} t \quad \text{and} \quad \text{Var}_\phi P_\alpha(t) \equiv \frac{1}{4u(t)} = \frac{1}{4} \frac{1}{1 + \Gamma_{\text{ex}} t} \quad (60)$$

where we have also given the variance of P_α in the state $|\phi\rangle$. On the contrary, the equation for the mean value of P_α in $|\phi\rangle$ is purely stochastic, with a diffusion coefficient $D(t)$ depending on time and the initial condition $\bar{P}_\alpha(0) = 0$:

$$d\bar{P}_\alpha(t) = [2D(t)]^{1/2} d\zeta_s(t) \quad \text{with} \quad D(t) = \frac{\Gamma_{\text{ex}}}{8u(t)^2} = \frac{\Gamma_{\text{ex}}}{8(1 + \Gamma_{\text{ex}} t)^2} \quad (61)$$

17. We only keep the linear terms in dt or in noise, and we systematically replace the quadratic terms $d\zeta_s^2$ by their mean dt .

As $D(t)$ is of finite integral, $\bar{P}_\alpha(t)$ stabilizes asymptotically (at long times) at a fixed value on a single realization, as seen in figure 6, with a variance in the quantum state $\text{Var}_\phi P_\alpha$ tending to 0. This phenomenon of “stochastic convergence” towards an eigenstate of the measured observable (in this case P_α) is expected in the description of a quantum measurement by a diffusion equation of the state vector [35, 36, 37]. To show it here, we introduce a renormalized time θ in terms of which \bar{P}_α performs an ordinary Brownian motion with a unity diffusion coefficient, and we notice that this time is bounded:

$$\theta = \int_0^t dt' D(t') = \frac{\Gamma_{\text{ex}} t}{8(1 + \Gamma_{\text{ex}} t)} \xrightarrow{t \rightarrow +\infty} \theta_\infty = \frac{1}{8} \quad (62)$$

At the renormalized instant θ_∞ , \bar{P}_α follows a Gaussian law with zero mean and variance 1/4: \bar{P}_α has therefore the same asymptotic probability distribution ($t \rightarrow +\infty$) as that of the observable P_α in the initial quantum state of the nuclear spin.

We now come to the mean and the variance of P_α conditioned on the value \mathcal{S} of the time-integrated homodyning signal σ (57). Remarkably, we find that the conditional mean is always proportional to the signal, with a time-dependent proportionality coefficient, and that the conditional variance depends on time but not on the signal:

$$\langle P_\alpha \rangle_{\sigma=\mathcal{S}} = m(t) \frac{\mathcal{S}}{\sqrt{\Gamma_{\text{ex}}}} \quad \text{where} \quad m(t) = \frac{\Gamma_{\text{ex}} t}{1 + \Gamma_{\text{ex}} t} \quad ; \quad \text{Var}_{\sigma=\mathcal{S}}(P_\alpha) = \mathcal{V}(t) \quad \text{where} \quad \mathcal{V}(t) = \frac{1}{4(1 + \Gamma_{\text{ex}} t)} \quad (63)$$

These expressions denote Γ_{ex} as the nuclear spin homodyne squeezing rate in the one-mode model that is at weak Faraday coupling:

$$\Gamma_{\text{sq}} \underset{\Omega \rightarrow 0}{\sim} \Gamma_{\text{ex}} = \frac{\Omega^2 \gamma_f}{\kappa(\gamma_m + \gamma_f)} \quad (64)$$

In figure 7a, we plot $m(t)$ and $\mathcal{V}(t)$ as functions of the reduced time $\tau = \Gamma_{\text{ex}} t$. Just as the quantum variance in a single realization $\text{Var}_\phi P_\alpha$, with which it actually coincides, the conditional variance tends asymptotically towards zero as the inverse of time. In the conditional average, the coefficient $m(t)$ tends towards 1 at long times. To understand this, let's relate the integrated signal (57) to \bar{P}_α using adiabatic expressions (30) in the truncated state vector (29):

$$\sigma(t) = \frac{1}{t} \int_0^t dt' \left[\sqrt{\Gamma_{\text{ex}}} \bar{P}_\alpha(t') + \frac{1}{2} \frac{d\zeta_s(t')}{dt'} \right] \quad (65)$$

As $\bar{P}_\alpha(t)$ stabilizes asymptotically on a single realization, and the time average of the noise $d\zeta_s$ tends to zero like $1/t^{1/2}$, $\sigma(+\infty)$ directly gives the value of $\bar{P}_\alpha(+\infty)$ up to a constant factor $\sqrt{\Gamma_{\text{ex}}}$.

To derive the results (63), we first relate the conditional variance of the operator P_α to that of its quantum average in a realization \bar{P}_α as follows:

$$\begin{aligned} \text{Var}_{\sigma=\mathcal{S}}(P_\alpha) &\equiv \langle \langle \phi | P_\alpha^2 | \phi \rangle \rangle_{\sigma=\mathcal{S}} - \langle \langle \phi | P_\alpha | \phi \rangle \rangle_{\sigma=\mathcal{S}}^2 = \langle \langle \phi | P_\alpha^2 | \phi \rangle - \langle \phi | P_\alpha | \phi \rangle^2 \rangle_{\sigma=\mathcal{S}} + \langle \bar{P}_\alpha^2 \rangle_{\sigma=\mathcal{S}} - \langle \bar{P}_\alpha \rangle_{\sigma=\mathcal{S}}^2 \\ &= \langle \text{Var}_\phi P_\alpha \rangle_{\sigma=\mathcal{S}} + \text{Var}_{\sigma=\mathcal{S}}(\bar{P}_\alpha) = \frac{1}{4} \frac{1}{1 + \Gamma_{\text{ex}} t} + \text{Var}_{\sigma=\mathcal{S}}(\bar{P}_\alpha) \end{aligned} \quad (66)$$

where we have used expression (60) for the quantum variance of P_α in the state $|\phi\rangle$. It therefore remains to determine the conditional probability distribution of \bar{P}_α knowing that $\sigma = \mathcal{S}$,

$$P(\bar{P}_\alpha = p_\alpha | \sigma = \mathcal{S}) \equiv \frac{P(\bar{P}_\alpha = p_\alpha, \sigma = \mathcal{S})}{P(\sigma = \mathcal{S})} \quad (67)$$

The random variable $\bar{P}_\alpha(t)$, resulting from Brownian motion (61), has a Gaussian probability distribution; the same applies to the temporal integral of \bar{P}_α and to the noise $d\zeta_s$, therefore to the signal σ (65) which is their sum. As the variables \bar{P}_α and σ have zero means, their joint probability distribution is characterized by their covariance matrix, or

more directly by its inverse matrix, so that

$$\begin{aligned}
P(\bar{P}_\alpha = p_\alpha | \sigma = S) &= \frac{1}{2\pi \sqrt{\langle \bar{P}_\alpha^2 \rangle_{\text{stoch}} \langle \sigma^2 \rangle_{\text{stoch}} - \langle \sigma \bar{P}_\alpha \rangle_{\text{stoch}}^2}} \exp\left(-\frac{1}{2} \frac{p_\alpha^2 \langle \sigma^2 \rangle_{\text{stoch}} + S^2 \langle \bar{P}_\alpha^2 \rangle_{\text{stoch}} - 2p_\alpha S \langle \sigma \bar{P}_\alpha \rangle_{\text{stoch}}}{\langle \bar{P}_\alpha^2 \rangle_{\text{stoch}} \langle \sigma^2 \rangle_{\text{stoch}} - \langle \sigma \bar{P}_\alpha \rangle_{\text{stoch}}^2}\right) \\
&= \frac{1}{\sqrt{2\pi \langle \sigma^2 \rangle_{\text{stoch}}}} \exp\left(-\frac{S^2}{2 \langle \sigma^2 \rangle_{\text{stoch}}}\right) \\
&= \frac{1}{\sqrt{2\pi \left[\langle \bar{P}_\alpha^2 \rangle_{\text{stoch}} - \langle \sigma \bar{P}_\alpha \rangle_{\text{stoch}}^2 / \langle \sigma^2 \rangle_{\text{stoch}} \right]}} \exp\left[-\frac{1}{2} \frac{\left(p_\alpha - S \langle \sigma \bar{P}_\alpha \rangle_{\text{stoch}} / \langle \sigma^2 \rangle_{\text{stoch}}\right)^2}{\langle \bar{P}_\alpha^2 \rangle_{\text{stoch}} - \langle \sigma \bar{P}_\alpha \rangle_{\text{stoch}}^2 / \langle \sigma^2 \rangle_{\text{stoch}}}\right] \quad (68)
\end{aligned}$$

where $\langle \dots \rangle_{\text{stoch}}$ at time t is the average taken over all the realizations of the stochastic process $d\zeta_s(t')$ in the time interval $[0, t]$. We deduce that, in equations (63),

$$m(t) = \sqrt{\Gamma_{\text{ex}}} \frac{\langle \sigma(t) \bar{P}_\alpha(t) \rangle_{\text{stoch}}}{\langle \sigma^2(t) \rangle_{\text{stoch}}} \quad \text{and} \quad \mathcal{V}(t) = \frac{1}{4(1 + \Gamma_{\text{ex}} t)} + \frac{\langle \bar{P}_\alpha^2(t) \rangle_{\text{stoch}} - \frac{\langle \sigma(t) \bar{P}_\alpha(t) \rangle_{\text{stoch}}^2}{\langle \sigma^2(t) \rangle_{\text{stoch}}}}{4(1 + \Gamma_{\text{ex}} t)} \quad (69)$$

In order to determine their variances and covariance, we write $\sigma(t)$ and $\bar{P}_\alpha(t)$ as linear functionals of the stochastic process $d\zeta_s$ and we use the fact that the Langevin forces $d\zeta_s(t)/dt$ and $d\zeta_s(t')/dt'$ have a Dirac correlation function $\delta(t - t')$. Let us give the example of the first contribution to $\sigma(t)$:

$$\int_0^t dt'' \bar{P}_\alpha(t'') = \int_0^t dt'' \int_0^{t''} dt' [2D(t')]^{1/2} \frac{d\zeta_s(t')}{dt'} = \int_0^t dt' \int_{t'}^t dt'' [2D(t')]^{1/2} \frac{d\zeta_s(t')}{dt'} = \int_0^t dt' (t - t') [2D(t')]^{1/2} \frac{d\zeta_s(t')}{dt'} \quad (70)$$

where we changed the order of integration on t' and t'' then explicitly integrated on t'' . We end up with the expressions we are looking for (63), the simplicity of which follows from the fact that, in one realization of the experiment, we always have

$$\sigma(t) = \sqrt{\Gamma_{\text{ex}}} \frac{1 + \Gamma_{\text{ex}} t}{\Gamma_{\text{ex}} t} \bar{P}_\alpha(t) \quad (71)$$

Remarkably, the knowledge of the integrated signal $\sigma(t)$ in a realization of the experiment of duration t is sufficient to prepare the nuclear spin in a well-defined pure Gaussian state (59), with a parameter u given by equation (60) and a mean quadrature \bar{P}_α related to the signal by equation (71)

Finally, let us return to the quadrature P_α of the unhybridized nuclear spin, which is truly usable in the experiment once the discharge is switched-off in the cell, as shown by expression (13) of the metrological gain in a precession measurement. By inversion of transformation (24) and by limiting equation (29) to its first term (to the dominant order in Ω), it comes

$$\langle P_\alpha \rangle_{\sigma=S} = \left(\frac{\gamma_m}{\gamma_f + \gamma_m} \right)^{1/2} \langle P_\alpha \rangle_{\sigma=S} \quad \text{and} \quad \text{Var}_{\sigma=S}(P_\alpha) = \frac{\gamma_f}{4(\gamma_f + \gamma_m)} + \frac{\gamma_m}{\gamma_f + \gamma_m} \text{Var}_{\sigma=S}(P_\alpha) \quad (72)$$

The conditional variance of P_α at long times tends towards a nonzero value, although low in practice: this is the intrinsic limit of this nuclear spin squeezing scheme, which uses the metastable state of ^3He as an intermediate state.

4.2.3. Solution of the three-mode model

The study of spin squeezing in the one-mode model is limited to the regime (23) where the squeezing rate $\Gamma_{\text{sq}} \sim \Gamma_{\text{ex}}$ is the longest timescale in the system. However, it is crucial for applications to see how far we can speed up the squeezing process by increasing Γ_{ex} through, for example, the Faraday coupling Ω of metastable atoms to the cavity field. To this end, we obtain the analytical solution of the three-mode model by using the Gaussian character of the state vector which results, as for the one-mode model, from the initial state considered (the vacuum), from the linearity of the jump operators C_m and the quadraticity of the Hamiltonian H in the quadratures of the modes. The stochastic equation (55) therefore admits as an exact solution the Gaussian ansatz generalizing that of equation (59),

$$\langle p_\alpha, p_\beta, x_c | \phi(t) \rangle = \phi(\mathbf{q}, t) = [8\pi \det \underline{\underline{u}}(t)]^{1/4} \exp\left\{-[\mathbf{q} - \bar{\mathbf{q}}(t)] \cdot \underline{\underline{u}}(t) [\mathbf{q} - \bar{\mathbf{q}}(t)]\right\} \equiv e^{-S} \quad (73)$$

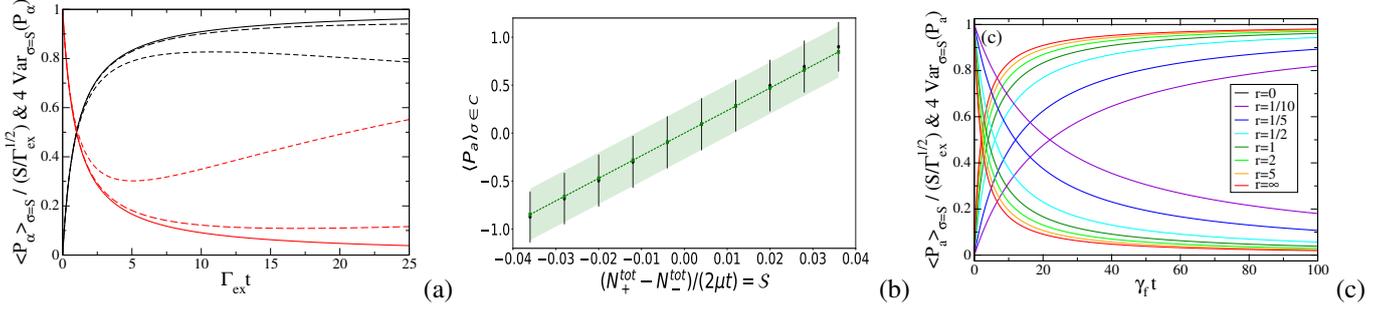


Figure 7 – Nuclear spin squeezing by continuous homodyne measurement. (a) In the one-mode model, mean (in black) and variance (multiplied by 4, in red) of the quadrature P_α of the hybridized nuclear spin conditioned on the integrated homodyne signal σ , as functions of the integration time t . Solid lines: analytical expressions (63). Dashed lines: expressions (102) and (103) in the presence of decoherence by de-excitation of metastable atoms at the cell walls (long dashed line: $\epsilon = 1/100$, short dashed line: $\epsilon = 1/10$, with $\epsilon = \gamma_\alpha/\Gamma_{\text{ex}}$ and γ_α the effective decoherence rate (98)). (b) In the three-mode model, for $\Gamma_{\text{ex}} = 5$, mean and standard deviation of the quadrature P_a of the nuclear spin conditioned on the signal σ belonging to a class C , the range of values of $\sigma/\sqrt{\Gamma_{\text{ex}}}$ having been divided into 10 classes of the same width (the values S of σ are in units of $\Gamma_{\text{ex}}^{1/2}$ on the horizontal axis). The standard deviation is plotted as a confidence interval. In black: numerical simulation of the stochastic equation (55) with 1079 realizations. Dashed green and colored area: exact results taken from relations (85), (86) and the analytical expression of the conditional probability distribution of \tilde{P}_a in terms of the variances and covariance (92) similarly to equation (68). The deviation between numerical and analytical results in the extreme classes is attributable to the low numbers of realizations falling in these classes. Parameter values: $\Omega/\kappa = 1/10$, $\gamma_m/\kappa = 1/10$, $\gamma_f/\kappa = 1/100$, $\Gamma_{\text{ex}}/\kappa = 1/1000$. (c) In the limit $\Gamma_{\text{ex}} \rightarrow 0$ at $\Gamma_{\text{ex}}/2\gamma_f$ fixed of the three-mode model, conditional mean and variance of P_a (94) as functions of reduced time $\gamma_f t$, for different values of the ratio $r = 2\Gamma_{\text{ex}}/\gamma_f$ (increasing curves: mean, decreasing curves: variance).

where \underline{u} is a real symmetric 3×3 matrix, $\bar{\mathbf{q}}$ is a real three-component vector, the coordinates $q_\alpha = p_\alpha$ and $q_\beta = p_\beta$ are in Fourier space (eigenbasis of the quadrature P) and the coordinate $q_c = x_c$ is in the “position” space (eigenbasis of the quadrature X). The only trick here was to choose as the metastability exchange jump operator $C_\beta = \sqrt{\gamma_\beta} i\beta$; this choice of phase, which of course does not change the quantum master equation (25), remains legitimate for the evolution conditioned on the homodyne detection of the field because the metastability jumps are not measured. In the mixed representation of the wave function (73), the Hamiltonian H is then purely imaginary and the jump operators are real, hence the real ansatz (73).¹⁸

To get the equations of motion on \underline{u} and $\bar{\mathbf{q}}$, we calculate in two different ways the relative variation $d\phi(\mathbf{q}, t)/\phi(\mathbf{q}, t)$ of the wave function, on the one hand by connecting it to the variation dS of the quantity S in (73), separated into a deterministic part dS_d and a noisy part dS_b , on the other hand by inserting ansatz (73) in the stochastic equation (55). By identifying the deterministic parts and the noisy parts of the two resulting forms, we obtain

$$-dS_b = \gamma_\beta^{1/2} \left(\frac{1}{2} \partial_{q_\beta} S - q_\beta + \bar{q}_\beta \right) d\zeta_\beta - \kappa^{1/2} \left(\frac{1}{2} \partial_{q_c} S - q_c + \bar{q}_c \right) d\zeta_c \quad (74)$$

$$-dS_d + \frac{1}{2} (dS_b)^2 = (\Omega_\alpha q_\alpha + \Omega_\beta q_\beta) \frac{dt}{2} \partial_{q_c} S - \frac{\gamma_\beta dt}{2} \left\{ q_\beta^2 - \frac{1}{2} + \frac{1}{4} \left[\partial_{q_\beta}^2 S - (\partial_{q_\beta} S)^2 \right] + \bar{q}_\beta (\partial_{q_\beta} S - 2q_\beta) + \bar{q}_\beta^2 \right\} \\ - \frac{\kappa dt}{2} \left\{ q_c^2 - \frac{1}{2} + \frac{1}{4} \left[\partial_{q_c}^2 S - (\partial_{q_c} S)^2 \right] + \bar{q}_c (\partial_{q_c} S - 2q_c) + \bar{q}_c^2 \right\} \quad (75)$$

It remains to insert in (75) the expression of dS_b taken from (74), by applying Ito’s rule of replacing the squares of the noises by their mean, then identifying the terms of degree 2 in $\mathbf{q} - \bar{\mathbf{q}}$ to obtain the purely deterministic equation linear

18. For example, $i\beta = i(X_\beta + iP_\beta)$ is represented in momentum by the real operator $-\partial_{p_\beta}/2 - p_\beta$, and $\beta^\dagger\beta$ by $-\partial_{p_\beta}^2/4 + p_\beta^2 - 1/2$.

on \underline{u} :¹⁹

$$\begin{aligned} du_{\alpha\alpha} &= -\Omega_\alpha dt u_{\alpha c} & du_{\alpha\beta} &= -\frac{dt}{2}(\gamma_\beta u_{\alpha\beta} + \Omega_\beta u_{\alpha c} + \Omega_\alpha u_{\beta c}) & du_{\alpha c} &= -\frac{dt}{2}(\kappa u_{\alpha c} + \Omega_\alpha u_{cc}) \\ du_{\beta\beta} &= -\Omega_\beta dt u_{\beta c} + \gamma_\beta dt(1 - u_{\beta\beta}) & du_{\beta c} &= -\frac{dt}{2}[(\gamma_\beta + \kappa)u_{\beta c} + \Omega_\beta u_{cc}] & du_{cc} &= \kappa dt(1 - u_{cc}) \end{aligned} \quad (76)$$

and the terms of degree 1 in $\mathbf{q} - \bar{\mathbf{q}}$ to obtain the stochastic linear equation for $\bar{\mathbf{q}}$:

$$d\bar{\mathbf{q}} = \frac{1}{2} \begin{pmatrix} 0 & 0 & 0 \\ 0 & -\gamma_\beta & 0 \\ \Omega_\alpha & \Omega_\beta & -\kappa \end{pmatrix} dt \bar{\mathbf{q}} + \frac{1}{2} [\text{Id} - \underline{c}(t)] \begin{pmatrix} 0 \\ \gamma_\beta^{1/2} d\zeta_\beta(t) \\ -\kappa^{1/2} d\zeta_c(t) \end{pmatrix} \quad (77)$$

Needless to say, $\bar{\mathbf{q}}$ is the vector of the quantum averages of the variables q in state vector (73);²⁰ in addition, we have introduced the notation \underline{c} for the inverse matrix of \underline{u} , which is none other than the quantum covariance matrix of q up to a numerical factor. We therefore have:

$$\langle \phi(t) | q_i | \phi(t) \rangle = \bar{q}_i(t) \quad \text{and} \quad \langle \phi(t) | q_i q_j | \phi(t) \rangle = \bar{q}_i(t) \bar{q}_j(t) + \frac{1}{4} c_{ij}(t) \quad \forall i, j \in \{\alpha, \beta, c\} \quad \text{with} \quad \underline{c}(t) = [\underline{u}(t)]^{-1} \quad (78)$$

The differential system (76) is easily integrated for the initial condition $\underline{u}(0) = \text{Id}$:

$$u_{\alpha\alpha}(t) = 1 + \frac{\Omega_\alpha^2 t}{\kappa} - \frac{2\Omega_\alpha^2}{\kappa^2} (1 - e^{-\kappa t/2}) \quad (79)$$

$$u_{\alpha\beta}(t) = \frac{\Omega_\alpha \Omega_\beta}{\gamma_\beta} \left(\frac{1}{\gamma_\beta + \kappa} + \frac{1}{\kappa} \right) (1 - e^{-\gamma_\beta t/2}) + \frac{\Omega_\alpha \Omega_\beta}{\kappa(\kappa - \gamma_\beta)} (e^{-\kappa t/2} - e^{-\gamma_\beta t/2}) + \frac{\Omega_\alpha \Omega_\beta}{\kappa(\gamma_\beta + \kappa)} (e^{-(\gamma_\beta + \kappa)t/2} - e^{-\gamma_\beta t/2}) \quad (80)$$

$$u_{\alpha c}(t) = -\frac{\Omega_\alpha}{\kappa} (1 - e^{-\kappa t/2}) \quad (81)$$

$$u_{\beta\beta}(t) = 1 + \frac{\Omega_\beta^2}{\gamma_\beta(\gamma_\beta + \kappa)} (1 - e^{-\gamma_\beta t}) - \frac{2\Omega_\beta^2}{\kappa^2 - \gamma_\beta^2} (e^{-\gamma_\beta t} - e^{-(\gamma_\beta + \kappa)t/2}) \quad (82)$$

$$u_{\beta c}(t) = -\frac{\Omega_\beta}{\gamma_\beta + \kappa} (1 - e^{-(\gamma_\beta + \kappa)t/2}) \quad (83)$$

$$u_{cc}(t) = 1 \quad (84)$$

It would have been different if we had taken as unknown the covariance matrix $\underline{c}(t)$, which obeys a Riccati nonlinear differential system [38]. Since $\bar{\mathbf{q}}$ describes a Brownian motion (partially damped because the friction matrix in (77) has eigenvalues 0, $\gamma_\beta/2$ and $\kappa/2$), and since the homodyne signal averaged over the time interval $[0, t]$ σ is deduced by integration, these random variables have a Gaussian statistic and we can reproduce the reasoning of section 4.2.2. We find for the conditional mean and variance of the quadrature P_a of the nuclear spin knowing that $\sigma = \mathcal{S}$ (this variance determines the metrological gain (13)):

$$\langle P_a \rangle_{\sigma=\mathcal{S}} = \frac{\langle \sigma(t) \bar{P}_a(t) \rangle_{\text{stoch}}}{\langle \sigma^2(t) \rangle_{\text{stoch}}} \mathcal{S} \quad (85)$$

$$\text{Var}_{\sigma=\mathcal{S}}(P_a) = \frac{1}{4} \left[\frac{\Omega_\beta^2}{\Omega^2} c_{\alpha\alpha}(t) + \frac{\Omega_\alpha^2}{\Omega^2} c_{\beta\beta}(t) - 2 \frac{\Omega_\alpha \Omega_\beta}{\Omega^2} c_{\alpha\beta}(t) \right] + \langle \bar{P}_a^2(t) \rangle_{\text{stoch}} - \frac{\langle \sigma(t) \bar{P}_a(t) \rangle_{\text{stoch}}^2}{\langle \sigma^2(t) \rangle_{\text{stoch}}} = \frac{1}{4} - \frac{\langle \sigma(t) \bar{P}_a(t) \rangle_{\text{stoch}}^2}{\langle \sigma^2(t) \rangle_{\text{stoch}}} \quad (86)$$

19. We notice that the quadratic terms in \underline{u} in the right-hand side of (75) cancel with those of $(dS_b)^2/2$ in the left-hand side.

20. We can therefore recover equation (77) from the stochastic equation deduced from (55) on the expectation value of an observable O , $d\langle O \rangle = (dt/i\hbar)\langle [O, H] \rangle + (dt/2) \sum_m \langle C_m^\dagger [O, C_m] + \text{h.c.} \rangle + \sum_m [\langle OC_m + \text{h.c.} \rangle - \langle C_m + C_m^\dagger \rangle \langle O \rangle] d\zeta_m$, where $\langle \dots \rangle$ is taken in state $|\phi(t)\rangle$, by specializing it to the cases $O = P_\alpha$, $O = P_\beta$ and $O = X_c$.

The expression in brackets in equation (86) is the matrix element of $\underline{c}(t)$ in the coordinate vector $(\Omega_\beta/\Omega, -\Omega_\alpha/\Omega, 0)$ of direction a in the rotated basis. The first term in the middle-hand side is therefore, up to a factor 4, the quantum variance of P_a in the stochastic state $\phi(t)$, depending on time but, let us recall, independent of the particular realization of $\phi(t)$. The simplified expression in the right-hand side follows from the property (21) on the unconditional mean $\langle P_a^2 \rangle(t) = 1/4$ and from the chain of equalities

$$\langle P_a^2 \rangle(t) = \langle \langle \phi(t) | P_a^2 | \phi(t) \rangle \rangle_{\text{stoch}} = \langle \langle \phi(t) | P_a^2 | \phi(t) \rangle - \langle \phi(t) | P_a | \phi(t) \rangle^2 + \langle \phi(t) | P_a | \phi(t) \rangle^2 \rangle_{\text{stoch}} = \langle \text{Var}_{\phi(t)} P_a \rangle_{\text{stoch}} + \langle \bar{P}_a^2(t) \rangle_{\text{stoch}} \quad (87)$$

To determine the variance and covariance of the random variables $\bar{P}_a(t)$ and $\sigma(t)$, it remains to calculate their amplitudes on the stochastic processes $d\zeta_\beta(t')$ and $d\zeta_c(t')$, formally integrating equation (77) by the method of variation of constants for \bar{P}_a and \bar{X}_c , and proceeding as in equation (70) for σ :

$$p_\beta(t, t') = -\frac{1}{2} \gamma_\beta^{1/2} \left\{ \frac{\Omega_\beta}{\Omega} c_{\alpha\beta}(t') + \frac{\Omega_\alpha}{\Omega} [1 - c_{\beta\beta}(t')] e^{-\gamma_\beta(t-t')/2} \right\} \quad (88)$$

$$p_c(t, t') = \frac{1}{2} \kappa^{1/2} \left\{ \frac{\Omega_\beta}{\Omega} c_{\alpha c}(t') - \frac{\Omega_\alpha}{\Omega} c_{\beta c}(t') e^{-\gamma_\beta(t-t')/2} \right\} \quad (89)$$

$$\sigma_\beta(t, t') = \frac{(\gamma_\beta \kappa)^{1/2}}{2t} \left\{ -c_{\alpha\beta}(t') [t - t' - f_\kappa(t - t')] \frac{\Omega_\alpha}{\kappa} + [1 - c_{\beta\beta}(t')] [f_{\gamma_\beta}(t - t') - f_\kappa(t - t')] \frac{\Omega_\beta}{\kappa - \gamma_\beta} - c_{\beta c}(t') f_\kappa(t - t') \right\} \quad (90)$$

$$\sigma_c(t, t') = \frac{1}{2t} - \frac{\kappa}{2t} \left\{ -c_{\alpha c}(t') [t - t' - f_\kappa(t - t')] \frac{\Omega_\alpha}{\kappa} - c_{\beta c}(t') [f_{\gamma_\beta}(t - t') - f_\kappa(t - t')] \frac{\Omega_\beta}{\kappa - \gamma_\beta} + [1 - c_{cc}(t')] f_\kappa(t - t') \right\} \quad (91)$$

where $f_\lambda(\tau) \equiv [1 - \exp(-\lambda\tau/2)]/(\lambda/2)$. We obtain:

$$\begin{aligned} \langle \sigma(t) \bar{P}_a(t) \rangle_{\text{stoch}} &= \int_0^t dt' [p_\beta(t, t') \sigma_\beta(t, t') + p_c(t, t') \sigma_c(t, t')] \quad ; \quad \langle \sigma^2(t) \rangle_{\text{stoch}} = \int_0^t dt' [\sigma_\beta^2(t, t') + \sigma_c^2(t, t')] \quad ; \\ \langle \bar{P}_a^2(t) \rangle_{\text{stoch}} &= \int_0^t dt' [p_\beta^2(t, t') + p_c^2(t, t')] \end{aligned} \quad (92)$$

We deduce from these results the long time limits²¹

$$\langle P_a \rangle_{\sigma=S} \xrightarrow{t \rightarrow +\infty} \left(\frac{\gamma_m}{\gamma_f + \gamma_m} \right)^{1/2} \frac{S}{\Gamma_{\text{ex}}^{1/2}} \quad ; \quad \text{Var}_{\sigma=S}(P_a) \xrightarrow{t \rightarrow +\infty} \frac{1}{4} \frac{\gamma_f}{\gamma_f + \gamma_m} \quad (93)$$

with which the predictions (72) of the one-mode model, however obtained within the weak coupling limit (23), are in perfect agreement.

As an application of our analytical solution of the three-mode model, let the rate Γ_{ex} tend to zero at fixed reduced time $\tau = \Gamma_{\text{ex}} t$ while maintaining (unlike the one-mode model) the ratio $\Gamma_{\text{ex}}/\gamma_f$ to a non-infinitesimal constant value. The physical motivation is clear: in the planned experiments [14], γ_f and Γ_{ex} are of the same order of magnitude but are really much smaller than γ_m and κ (by factors $\approx 10^{-6}$ and 10^{-9}). We find in this limit:²²

$$\boxed{\langle P_a \rangle_{\sigma=S} \sim \frac{\Gamma_{\text{sq}} t}{1 + \Gamma_{\text{sq}} t} \frac{S}{\Gamma_{\text{ex}}^{1/2}} \quad \text{and} \quad \text{Var}_{\sigma=S}(P_a) \sim \frac{1}{4} \frac{1}{1 + \Gamma_{\text{sq}} t}} \quad (94)$$

21. Let us give some results and intermediate considerations. (i) While $c_{\beta\beta}(t')$, $c_{\beta c}(t')$ and $c_{cc}(t')$ have a finite limit when $t' \rightarrow +\infty$ [we will need $c_{\beta\beta}(+\infty) = (1 + \rho)^{-1}$, $c_{\beta c}(+\infty) = \Omega_\beta / ((\gamma_\beta + \kappa)(1 + \rho))$ with $\rho = \Omega_\beta^2 \kappa / (\gamma_\beta(\gamma_\beta + \kappa)^2)$], $c_{\alpha\alpha}(t')$, $c_{\alpha\beta}(t')$ and $c_{\alpha c}(t')$ tend to zero as $1/t'$. (ii) In an integral over t' containing the exponential factor $\exp[-\gamma_\beta(t - t')/2]$ or its square, we can replace the function which multiplies it by its limit in $t' = +\infty$. (iii) For any uniformly bounded function $w(t, t')$, we can show for $v \in \{\beta, c\}$ that $\int_0^t dt' [(t - t') c_{\alpha v}(t') + w(t, t')]^2 / t^2 \rightarrow \int_0^{+\infty} dt' c_{\alpha v}^2(t')$. (iv) We then obtain the asymptotic limits $\langle P_a^2(t) \rangle_{\text{stoch}} \rightarrow (\Omega_\beta / 2\Omega)^2 I + (\Omega_\alpha / 2\Omega)^2 \rho / (1 + \rho)$, $\langle \sigma^2(t) \rangle_{\text{stoch}} \rightarrow (\Omega_\alpha^2 / 4\kappa) I$, $\langle \sigma(t) \bar{P}_a(t) \rangle_{\text{stoch}} \rightarrow (\Omega_\alpha \Omega_\beta / 4\Omega \kappa^{1/2}) I$ where $I \equiv \int_0^{+\infty} dt' [\gamma_\beta c_{\alpha\beta}^2(t') + \kappa c_{\alpha c}^2(t')]$. We thus deduce (93) from (85) and from the first equality in (86), without needing to know the value of I . We derive from the second equality in (86) the result $I = 1$, which we can also deduce from the equation of motion $dc_{\alpha\alpha}/dt = -\gamma_\beta c_{\alpha\beta}^2 - \kappa c_{\alpha c}^2$ integrated between $t = 0$ and $t = +\infty$.

22. In practice, it suffices to make Ω_α tend to zero at $\tau = \Gamma_{\text{ex}} t > 0$, Ω_β , γ_β and κ fixed. In particular, this makes all exponential transients disappear in equations (79)-(84). To simplify the calculations, it is useful to introduce the quantity $\rho = \Omega^2 \kappa / [2\gamma_m(\kappa + 2\gamma_m)^2]$ so that $\rho = (\Gamma_{\text{ex}} / 2\gamma_f)(1 + 2\gamma_m/\kappa)^{-2}$ in the limit $\gamma_f \rightarrow 0$.

where we have introduced the true or generalized squeezing rate

$$\Gamma_{\text{sq}} \equiv \left(\frac{1}{\Gamma_{\text{ex}}} + \frac{2}{\gamma_f} \right)^{-1} \quad (95)$$

We find the natural scaling of the signal by $\Gamma_{\text{ex}}^{1/2}$ already observed in the one-mode model and the same functional forms in time, but we lose all relation of proportionality of type (71) between integrated signal and quadrature average in a given realization, the conditional variance of \bar{P}_a now being $\neq 0$.²³ We represent in figure 7c the variation with reduced time $\gamma_f t$ of the conditional mean and variance (94) for different values of the ratio $r = 2\Gamma_{\text{ex}}/\gamma_f$. We notice that the squeezing process is all the faster as r is larger, and that it saturates to a limiting behavior. This was predictable, because Γ_{sq} is an increasing function of r with finite limit; at a fixed time, the conditional mean (in units of $S/\Gamma_{\text{ex}}^{1/2}$) is therefore an increasing function and the conditional variance a decreasing function of r , as seen in figure 7c. More precisely, in the weak coupling limit $\Omega \rightarrow 0$, where $r \rightarrow 0$, the generalized squeezing rate is equivalent to the rate Γ_{ex} of creation of excitations, in agreement with the one-mode model, and within the limit $r \rightarrow +\infty$, it saturates to the value $\gamma_f/2$. We cannot therefore squeeze faster than at the rate γ_f , which is not surprising: we cannot hope to reduce the fluctuations in nuclear spin before each atom in the ground state has undergone on average at least one metastability exchange collision, the effective rate γ_f being in practice of the same order of magnitude as the individual rate $1/T$ in equation (8) except in the case of extreme polarization, see figure 3a.

4.2.4. Effect of decoherence

To be complete, we take into account, in the homodyne squeezing scheme, the finite lifetime $(2\gamma_0)^{-1}$ of the metastable atoms, which de-excite when they reach the cell walls after diffusive motion in the gas. To this end, we add a jump operator $\sqrt{2\gamma_0}b$ to the three-mode quantum master equation (20). As the part other than Hermitian Hamiltonian remains quadratic in the quadratures of the modes, it can be put in reduced form by an appropriate rotation of the atomic modes, as we had already done in section 3.4: one simply has to expand (a, b) in the orthonormal eigenbasis of the rate matrix

$$\underline{\Gamma} = \begin{pmatrix} 2\gamma_f & -2\sqrt{\gamma_f\gamma_m} \\ -2\sqrt{\gamma_f\gamma_m} & 2(\gamma_0 + \gamma_m) \end{pmatrix} \quad (96)$$

with operator-valued coefficients α and β . The direction β remains that of the maximum eigenvalue γ_β of $\underline{\Gamma}$, and α that of the minimum eigenvalue γ_α , now nonzero. This leads to the quantum master equation

$$\frac{d\rho}{dt} = \frac{1}{i\hbar} \left[\hbar(\Omega_\alpha P_\alpha + \Omega_\beta P_\beta) P_c, \rho \right] + \kappa \left(c\rho c^\dagger - \frac{1}{2}\{c^\dagger c, \rho\} \right) + \gamma_\alpha \left(\alpha\rho\alpha^\dagger - \frac{1}{2}\{\alpha^\dagger\alpha, \rho\} \right) + \gamma_\beta \left(\beta\rho\beta^\dagger - \frac{1}{2}\{\beta^\dagger\beta, \rho\} \right) \quad (97)$$

The new expression for Faraday frequencies $\Omega_\alpha, \Omega_\beta$ and rates $\gamma_\alpha, \gamma_\beta$ can be found in Appendix B, which also gives the analytical expression of the mean and of the variance of the quadrature P_a of the nuclear spin conditioned on the integrated homodyne signal, in all generality. We restrict ourselves here to the physically useful limit $\gamma_0 \ll \gamma_m$ (we still have $\gamma_f < \gamma_m$). To lowest order in γ_0 , the coefficients $\Omega_\alpha, \Omega_\beta$ and γ_β remain unchanged, and we have

$$\gamma_\alpha \simeq \frac{2\gamma_0\gamma_f}{\gamma_m + \gamma_f} \quad (98)$$

which is the reduced rate of decoherence in the hybridized nuclear spin. Moreover, we place ourselves in the limit (23), with $\gamma_\alpha = O(\Gamma_{\text{ex}})$, which allows to evaluate the effect of decoherence using the one-mode model, which is obtained in the same way as in section 3.4. The stochastic equation (58) is completed as follows,

$$d|\phi(t)\rangle = -\frac{\Gamma_{\text{ex}} dt}{2} (P_\alpha - \bar{P}_\alpha)^2 |\phi(t)\rangle + \sqrt{\Gamma_{\text{ex}}} d\zeta_s(t) (P_\alpha - \bar{P}_\alpha) |\phi(t)\rangle - \frac{\gamma_\alpha dt}{2} (\alpha^\dagger \alpha + 2i\bar{P}_\alpha \alpha + \bar{P}_\alpha^2) |\phi(t)\rangle + \sqrt{\gamma_\alpha} d\zeta_\alpha(t) (i\alpha + \bar{P}_\alpha) |\phi(t)\rangle \quad (99)$$

23. We have indeed $\text{Var}_{\sigma=S}(\bar{P}_a) \sim \Gamma_{\text{ex}} t / [4(1 + \Gamma_{\text{ex}} t)] - \Gamma_{\text{sq}} t / [4(1 + \Gamma_{\text{sq}} t)]$.

We have taken care to choose $\gamma_\alpha^{1/2}i\alpha$ as the jump operator of the effective decoherence (the justification is the same as in section 4.2.3, decoherence jumps are not measured), which allows the equation to be solved by the same real Gaussian ansatz (59). This time we find²⁴

$$du = [\Gamma_{\text{ex}} + \gamma_\alpha(1-u)]dt \implies u(\tau) = 1 + \frac{1 - \exp(-\epsilon\tau)}{\epsilon} \quad (100)$$

$$d\bar{P}_\alpha = -\frac{1}{2}\gamma_\alpha\bar{P}_\alpha dt + \frac{\sqrt{\Gamma_{\text{ex}}}d\zeta_s + \sqrt{\gamma_\alpha}(u-1)d\zeta_\alpha}{2u} \quad (101)$$

where we have set $\tau = \Gamma_{\text{ex}}t$ and $\epsilon = \gamma_\alpha/\Gamma_{\text{ex}}$. The same Gaussianity arguments as in 4.2.2 section lead to the same dependencies in the signal \mathcal{S} of the conditional mean and variance,²⁵

$$\langle P_\alpha \rangle_{\sigma=\mathcal{S}} = m(\tau) \frac{\mathcal{S}}{\sqrt{\Gamma_{\text{ex}}}} \quad \text{with} \quad m(\tau) = \sqrt{\Gamma_{\text{ex}}} \frac{\langle \sigma(t) \bar{P}_\alpha(t) \rangle_{\text{stoch}}}{\langle \sigma^2(t) \rangle_{\text{stoch}}} \quad (102)$$

$$\text{Var}_{\sigma=\mathcal{S}}(P_\alpha) = \mathcal{V}(\tau) \quad \text{with} \quad \mathcal{V}(\tau) = \frac{1}{4} - \frac{\langle \sigma(t) \bar{P}_\alpha(t) \rangle_{\text{stoch}}^2}{\langle \sigma^2(t) \rangle_{\text{stoch}}} \quad (103)$$

and the variance and covariance taken over the stochastic processes $d\zeta_s \equiv d\zeta_c$ and $d\zeta_\alpha$,

$$\frac{\langle \sigma^2 \rangle_{\text{stoch}}}{\Gamma_{\text{ex}}} = \int_0^\tau \frac{d\tau'}{\tau^2} \left\{ \left[\frac{1}{2} + \frac{1 - e^{\epsilon(\tau'-\tau)/2}}{\epsilon u(\tau')} \right]^2 + \frac{[u(\tau') - 1]^2 [1 - e^{\epsilon(\tau'-\tau)/2}]^2}{u^2(\tau') \epsilon} \right\} = \frac{\epsilon\tau - 2(1 - e^{-\epsilon\tau/2})}{\epsilon^2\tau^2} + \frac{1}{4\tau} \quad (104)$$

$$\frac{\langle \sigma \bar{P}_\alpha \rangle_{\text{stoch}}}{\sqrt{\Gamma_{\text{ex}}}} = \int_0^\tau \frac{d\tau'}{\tau} \frac{e^{\epsilon(\tau'-\tau)/2}}{2u(\tau')} \left\{ \frac{1}{2} + \frac{1 - e^{\epsilon(\tau'-\tau)/2}}{\epsilon u(\tau')} + \frac{[u(\tau') - 1]^2 [1 - e^{\epsilon(\tau'-\tau)/2}]}{u(\tau')} \right\} = \frac{1 - e^{-\epsilon\tau/2}}{2\epsilon\tau} \quad (105)$$

These expressions allow to easily evaluate the effect of decoherence on spin squeezing through the metrological gain (13), see the dashed lines in figure 7a. For the practical case of a weak decoherence $\epsilon \ll 1$ and a time short compared to $1/\gamma_\alpha$, they can be expanded to first order in ϵ :

$$\boxed{m(\tau) = \frac{\tau}{1+\tau} - \epsilon \frac{(\tau+3)\tau^2}{12(\tau+1)^2} + O(\epsilon^2\tau^2) \quad ; \quad \mathcal{V}(\tau) = \frac{1}{4(\tau+1)} + \epsilon \frac{(\tau+3/2)\tau^2}{12(\tau+1)^2} + O(\epsilon^2\tau^2)} \quad (106)$$

We then deduces that the optimal squeezing on P_α is obtained at a time $t_{\text{opt}} \sim (3/\Gamma_{\text{ex}}\gamma_\alpha)^{1/2}$ and corresponds to a conditional variance $\mathcal{V}_{\text{opt}} \sim (\gamma_\alpha/12\Gamma_{\text{ex}})^{1/2}$. Note that in studies of spin squeezing of alkaline gases in cavities, we often introduce the cooperativity C of the coupled atom-field system, defined as the square of the coupling frequency divided by the decay rates of the coupled states [39]. In this sense, the cooperativity of the hybridized nuclear spin-field system is equal to

$$C \equiv \frac{\Omega_\alpha^2}{\kappa\gamma_\alpha} = \frac{\Gamma_{\text{ex}}}{\gamma_\alpha} \simeq \frac{\Omega^2}{2\gamma_0\kappa} \quad (107)$$

so that we recover the scaling law of power $-1/2$, usual in alkalis, relating the optimal spin variance to C [39]. More generally, the decoherence has a weak effect on the nuclear spin squeezing as long as we stay at short times in front of t_{opt} . The reader will find at the end of Appendix B an extension of these scaling laws beyond the one-mode model, i.e. for an arbitrary, not infinitesimal ratio $\Gamma_{\text{ex}}/\gamma_f$; this was retained in the abstract of the article. The link between \mathcal{V}_{opt} and cooperativity (107) is then broken.

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24. In the $\epsilon \ll 1$ regime, the long-time limit of the variance of P_α in a single realization depends strongly on the choice of phase in the effective-decoherence jump operator, which emphasizes the unphysical character of this variance (see note 16): if we take $\gamma_\alpha^{1/2}\alpha$ as the jump operator, we find that $\text{Var}_\phi P_\alpha \rightarrow \epsilon^{1/2}/4$ [14] instead of $\text{Var}_\phi P_\alpha \rightarrow 1/4\epsilon$ as in equation (100). More generally, the choice $\gamma_\alpha^{1/2} \exp(i\theta)\alpha$, $-\pi/2 < \theta < \pi/2$, leads to the Riccati equation for the parameter u of the (now complex) Gaussian ansatz $du = \Gamma_{\text{ex}}dt + \gamma_\alpha dt[\exp(2i\theta)u + [1 - \exp(2i\theta)]/2 - u^2[1 + \exp(2i\theta)]/2]$ so that $\text{Var}_\phi P_\alpha \rightarrow (1/4)\epsilon^{1/2} \sqrt{\cos\theta \cos(\theta/2)}$ in a time $\tau \sim 1/\sqrt{2\epsilon[1 + \exp(2i\theta)]}$; the power law in ϵ obtained for $\theta = 0$ is thus the rule, that obtained for $\theta = \pi/2$ is the exception.

25. We have simplified expression (103) using the identity $[4u(\tau)]^{-1} + \langle \bar{P}_\alpha^2 \rangle_{\text{stoch}} = 1/4$, which results as in equation (87) from the fact that the unconditional mean $\langle P_\alpha^2 \rangle = 1/4$, even in the presence of decoherence.

Appendix A. Semi-classical treatment and reduction to three coupled spins

Here we give the nonlinear equations that describe the dynamics of the system in semi-classical theory, and we linearize them for small fluctuations around a partially polarized stationary solution.

Nonlinear semi-classical equations. Starting from the considerations and notations of section 2, we take the average of the Heisenberg equations of motion in the quantum state of the system and perform the decorrelation approximation (called semi-classical in quantum optics) $\langle AB \rangle \simeq \langle A \rangle \langle B \rangle$ where A and B are two operators, to obtain the following nonlinear evolution equations for the expectation values of \vec{S} the Stokes spin of the cavity field, \vec{I} the collective nuclear spin in the ground state, \vec{J} and \vec{K} the collective spins associated with the manifolds $F = 3/2$ and $F = 1/2$ in the metastable state, and \vec{Q} the collective alignment tensor in $F = 3/2$, of Cartesian components $Q_{\alpha\beta}$:

$$\frac{d\langle S_x \rangle}{dt} = -\frac{\kappa}{2} \left(\langle S_x \rangle - \frac{n_{\text{ph}}}{2} \right) - \chi \langle K_z \rangle \langle S_y \rangle \quad \frac{d\langle S_y \rangle}{dt} = -\frac{\kappa}{2} \langle S_y \rangle + \chi \langle K_z \rangle \langle S_x \rangle \quad \frac{d\langle S_z \rangle}{dt} = -\frac{\kappa}{2} \langle S_z \rangle \quad (\text{A.1})$$

$$\frac{d\langle K_x \rangle}{dt} = \frac{d\langle K_x \rangle}{dt} \Big|_{\text{ME}} - \chi \langle K_y \rangle \langle S_z \rangle \quad \frac{d\langle K_y \rangle}{dt} = \frac{d\langle K_y \rangle}{dt} \Big|_{\text{ME}} + \chi \langle K_x \rangle \langle S_z \rangle \quad \frac{d\langle K_z \rangle}{dt} = \frac{d\langle K_z \rangle}{dt} \Big|_{\text{ME}} \quad (\text{A.2})$$

$$\frac{d\langle \vec{J} \rangle}{dt} = \frac{d\langle \vec{J} \rangle}{dt} \Big|_{\text{ME}} \quad \frac{d\langle Q_{\alpha\beta} \rangle}{dt} = \frac{d\langle Q_{\alpha\beta} \rangle}{dt} \Big|_{\text{ME}} \quad \frac{d\langle \vec{I} \rangle}{dt} = \frac{d\langle \vec{I} \rangle}{dt} \Big|_{\text{ME}} \quad (\text{A.3})$$

The terms proportional to the loss rate κ of the cavity output mirror make $\langle S_x \rangle$ relax towards its stationary value $\langle S_x \rangle_s = n_{\text{ph}}/2$ driven by the laser field polarized along x injected into the cavity, and the transverse means $\langle S_y \rangle$ and $\langle S_z \rangle$ towards zero. The terms proportional to the Faraday coupling χ between the cavity mode and the spin \vec{K} derive from the Hamiltonian (2). The contribution of metastability exchange collisions (ME) between ground-state and metastable atoms is deduced directly from the quantum master equation for the one-atom density operator of references [21, 22] by simple multiplication or division by the total number of ground-state atoms N_{cell} or metastable atoms n_{cell} in the cell.²⁶

$$\frac{d\langle \vec{K} \rangle}{dt} \Big|_{\text{ME}} = -\frac{7}{9\tau} \langle \vec{K} \rangle + \frac{1}{9\tau} \langle \vec{J} \rangle - \frac{1}{9\tau} \frac{n_{\text{cell}}}{N_{\text{cell}}} \langle \vec{I} \rangle - \frac{4}{3\tau} \frac{1}{N_{\text{cell}}} \langle \vec{Q} \rangle \cdot \langle \vec{I} \rangle \quad (\text{A.4})$$

$$\frac{d\langle \vec{J} \rangle}{dt} \Big|_{\text{ME}} = -\frac{4}{9\tau} \langle \vec{J} \rangle + \frac{10}{9\tau} \langle \vec{K} \rangle + \frac{10}{9\tau} \frac{n_{\text{cell}}}{N_{\text{cell}}} \langle \vec{I} \rangle + \frac{4}{3\tau} \frac{1}{N_{\text{cell}}} \langle \vec{Q} \rangle \cdot \langle \vec{I} \rangle \quad (\text{A.5})$$

$$\frac{d\langle Q_{\alpha\beta} \rangle}{dt} \Big|_{\text{ME}} = -\frac{2}{3\tau} \langle Q_{\alpha\beta} \rangle + \frac{1}{9\tau} \frac{1}{N_{\text{cell}}} \left(3 \frac{\langle I_\alpha \rangle \langle \Sigma_\beta \rangle + \langle I_\beta \rangle \langle \Sigma_\alpha \rangle}{2} - \delta_{\alpha\beta} \langle \vec{I} \rangle \cdot \langle \vec{\Sigma} \rangle \right) \quad (\text{A.6})$$

$$\frac{d\langle \vec{I} \rangle}{dt} \Big|_{\text{ME}} = -\frac{1}{T} \langle \vec{I} \rangle + \frac{1}{3T} \frac{N_{\text{cell}}}{n_{\text{cell}}} (\langle \vec{J} \rangle - \langle \vec{K} \rangle) \quad (\text{A.7})$$

where $\langle \vec{\Sigma} \rangle = \frac{2}{3} [\langle \vec{J} \rangle + 2\langle \vec{K} \rangle]$ is the expectation value of the electron spin in the metastable state. See equations (1.37b), (1.37a), (1.39) and (1.25) of reference [22] (taking into account a difference of a factor 6 in the definition of the alignment tensor), or to equations (VIII.30), (VIII.29), (VIII.32) and (VIII.15) of reference [21] (by adding a Kronecker factor $\delta_{\alpha\beta}$ omitted in (VIII.32)). Here $1/\tau$ and $1/T$, the individual metastability exchange collision rates for an atom in the metastable state and in the ground state, are in the ratio $T/\tau = N_{\text{cell}}/n_{\text{cell}}$ since, in one unit of time, an equal number of ground-state and metastable atoms have undergone an exchange collision [21, 22].

Partially polarized stationary solution. In a polarized stationary state of nuclear polarization $\eta \in [0, 1]$,

$$\langle I_x \rangle_s = \eta \frac{N_{\text{cell}}}{2} \quad ; \quad \langle I_y \rangle_s = \langle I_z \rangle_s = 0 \quad ; \quad \langle S_x \rangle_s = \frac{n_{\text{ph}}}{2} \quad ; \quad \langle S_y \rangle_s = \langle S_z \rangle_s = 0 \quad (\text{A.8})$$

²⁶ The collective expectation values are in fact related as follows to the one-atom expectation values $\langle \cdot \rangle_{\text{at}}$: $\langle \vec{I} \rangle = N_{\text{cell}} \langle \vec{I} \rangle_{\text{at}}$, $\langle \vec{J} \rangle = n_{\text{cell}} \langle \vec{J} \rangle_{\text{at}}$, $\langle \vec{K} \rangle = n_{\text{cell}} \langle \vec{K} \rangle_{\text{at}}$, $\langle \vec{Q} \rangle = n_{\text{cell}} \langle \vec{Q} \rangle_{\text{at}}$, $\langle \vec{\Sigma} \rangle = n_{\text{cell}} \langle \vec{\Sigma} \rangle_{\text{at}}$.

rotational invariance around x axis constrains the mean spins in the metastable state to be aligned along x , and the mean alignment tensor to be diagonal in the Cartesian basis, with equal eigenvalues in y and z directions. The system (A.1)-(A.3) thus admits a stationary solution where the only nonzero expectation values in the metastable state are:

$$\langle K_x \rangle_s = \frac{\eta}{2} \frac{1 - \eta^2}{3 + \eta^2} n_{\text{cell}} ; \quad \langle J_x \rangle_s = \eta \frac{5 + \eta^2}{3 + \eta^2} n_{\text{cell}} ; \quad \langle \Sigma_x \rangle_s = \frac{4\eta}{3 + \eta^2} n_{\text{cell}} ; \quad \langle Q_{yy} \rangle_s = \langle Q_{zz} \rangle_s = -\frac{1}{2} \langle Q_{xx} \rangle_s = -\frac{\eta}{12} \langle \Sigma_x \rangle_s \quad (\text{A.9})$$

Linearized semi-classical equations. We now linearize equations (A.1)-(A.3) for classical fluctuations around the stationary solution (A.8)-(A.9) by performing the substitution $\langle A \rangle \rightarrow \langle A \rangle_s + \delta A$ and treating δA to first order. By limiting ourselves to the subspace of transverse fluctuations, that is to say to the directions $\alpha = y, z$ orthogonal to the mean spins, we obtain a closed system:

$$\frac{d}{dt} \delta S_\alpha = -\frac{\kappa}{2} \delta S_\alpha + \chi \delta_{\alpha y} \langle S_x \rangle_s \delta K_z \quad (\text{A.10})$$

$$\frac{d}{dt} \delta K_\alpha = -\frac{7}{9\tau} \delta K_\alpha + \frac{1}{9\tau} \delta J_\alpha - \frac{2\eta}{3\tau} \delta Q_{\alpha x} - \frac{1}{9T} \left(1 + \frac{12}{n_{\text{cell}}} \langle Q_{\alpha\alpha} \rangle_s \right) \delta I_\alpha + \chi \delta_{\alpha y} \langle K_x \rangle_s \delta S_z \quad (\text{A.11})$$

$$\frac{d}{dt} \delta J_\alpha = -\frac{4}{9\tau} \delta J_\alpha + \frac{10}{9\tau} \delta K_\alpha + \frac{2\eta}{3\tau} \delta Q_{\alpha x} + \frac{10}{9T} \left(1 + \frac{6}{5n_{\text{cell}}} \langle Q_{\alpha\alpha} \rangle_s \right) \delta I_\alpha \quad (\text{A.12})$$

$$\frac{d}{dt} \delta Q_{\alpha x} = -\frac{2}{3\tau} \delta Q_{\alpha x} + \frac{\eta}{12\tau} \delta \Sigma_\alpha + \frac{1}{6T n_{\text{cell}}} \langle \Sigma_x \rangle_s \delta I_\alpha \quad (\text{A.13})$$

$$\frac{d}{dt} \delta I_\alpha = -\frac{1}{T} \delta I_\alpha + \frac{1}{3\tau} (\delta J_\alpha - \delta K_\alpha) \quad (\text{A.14})$$

Reduction to three coupled collective spins. By setting $\frac{d}{dt} \delta J_\alpha = 0$ in equation (A.12) and $\frac{d}{dt} \delta Q_{\alpha x} = 0$ in equation (A.13), we adiabatically eliminate the fluctuations of the collective spin \vec{J} and of the collective alignment tensor whose evolutions are governed by the metastability exchange only:

$$\delta J_\alpha^{\text{adiab}} = 2 \frac{10 + \eta^2}{8 - \eta^2} \delta K_\alpha + \frac{12\tau}{T} \frac{5 + 2\eta^2}{(3 + \eta^2)(8 - \eta^2)} \delta I_\alpha ; \quad \delta Q_{\alpha x}^{\text{adiab}} = \frac{3\eta}{8 - \eta^2} \delta K_\alpha + \frac{\tau}{T} \frac{\eta(13 + \eta^2)}{(3 + \eta^2)(8 - \eta^2)} \delta I_\alpha \quad (\text{A.15})$$

The transfer of adiabatic expressions (A.15) in equations (A.11) and (A.14) on δK_α and δI_α leads in the body of the article to the reduced system (5)-(7) coupling the fluctuations of the three spins (3), where γ_f and γ_m , the effective metastability exchange rates between the nuclear spin and the spin $F = 1/2$ of the metastable, are given by equation (8).

Appendix B. Solution of the three-mode model with decoherence for homodyne detection

Here we give the analytical solution of the three-mode model in the presence of decoherence, see the quantum master equation (97), for an evolution of the system conditioned on a continuous homodyne measurement of the component polarized along y of the field leaking out of the cavity. The value of the coefficients γ_α , γ_β , Ω_α and Ω_β , as well as the annihilation operators α and β , are deduced from a diagonalization of the rate matrix (96). The rates γ_α and γ_β are the eigenvalues in ascending order:

$$\gamma_{\alpha,\beta} = \gamma_m + \gamma_f + \gamma_0 \mp [(\gamma_m + \gamma_f + \gamma_0)^2 - 4\gamma_f\gamma_0]^{1/2} \quad (\text{B.1})$$

In terms of the Faraday frequencies Ω_α and Ω_β , the corresponding normalized eigenvectors are written as $(\Omega_\beta/\Omega, \Omega_\alpha/\Omega)$ and $(-\Omega_\alpha/\Omega, \Omega_\beta/\Omega)$, so that $\alpha = (\Omega_\beta a + \Omega_\alpha b)/\Omega$ and $\beta = (\Omega_\beta b - \Omega_\alpha a)/\Omega$ with

$$\Omega_\alpha = \frac{\Omega(\gamma_f - \gamma_\alpha/2)}{[\gamma_m\gamma_f + (\gamma_f - \gamma_\alpha/2)^2]^{1/2}} ; \quad \Omega_\beta = \frac{\Omega\sqrt{\gamma_m\gamma_f}}{[\gamma_m\gamma_f + (\gamma_f - \gamma_\alpha/2)^2]^{1/2}} \quad (\text{B.2})$$

with a choice of sign ensuring that $\alpha \rightarrow a$ and $\beta \rightarrow b$ when $\gamma_f \rightarrow 0$ and reproducing (26) when $\gamma_0 \rightarrow 0$. Since the jump operator $C_\alpha \propto \alpha$ describes unmeasured processes, we can, as we did for C_β , take it of the form $\sqrt{\gamma_\alpha} i \alpha$ and

reuse the real Gaussian ansatz (73) in order to solve the stochastic equation (55) on the state vector. In the evolution equation for matrix \underline{u} that appears in the ansatz, the indices α and β now play symmetrical roles and we obtain

$$\begin{aligned} du_{\alpha\alpha} &= -\Omega_\alpha dt u_{\alpha c} + \gamma_\alpha dt(1 - u_{\alpha\alpha}) & du_{\alpha\beta} &= -\frac{dt}{2}[(\gamma_\alpha + \gamma_\beta)u_{\alpha\beta} + \Omega_\beta u_{\alpha c} + \Omega_\alpha u_{\beta c}] & du_{\alpha c} &= -\frac{dt}{2}[(\gamma_\alpha + \kappa)u_{\alpha c} + \Omega_\alpha u_{cc}] \\ du_{\beta\beta} &= -\Omega_\beta dt u_{\beta c} + \gamma_\beta dt(1 - u_{\beta\beta}) & du_{\beta c} &= -\frac{dt}{2}[(\gamma_\beta + \kappa)u_{\beta c} + \Omega_\beta u_{cc}] & du_{cc} &= \kappa dt(1 - u_{cc}) \end{aligned} \quad (\text{B.3})$$

whose solution for the initial condition $\underline{u}(0) = \text{Id}$ is written

$$u_{\alpha\alpha}(t) = 1 + \frac{\Omega_\alpha^2}{\gamma_\alpha(\kappa + \gamma_\alpha)} (1 - e^{-\gamma_\alpha t}) - \frac{2\Omega_\alpha^2}{\kappa^2 - \gamma_\alpha^2} (e^{-\gamma_\alpha t} - e^{-(\kappa + \gamma_\alpha)t/2}) \quad (\text{B.4})$$

$$\begin{aligned} u_{\alpha\beta}(t) &= \frac{\Omega_\alpha \Omega_\beta}{\gamma_\alpha + \gamma_\beta} \left(\frac{1}{\kappa + \gamma_\alpha} + \frac{1}{\kappa + \gamma_\beta} \right) (1 - e^{-(\gamma_\alpha + \gamma_\beta)t/2}) + \frac{\Omega_\alpha \Omega_\beta}{(\kappa - \gamma_\beta)(\kappa + \gamma_\alpha)} (e^{-(\kappa + \gamma_\alpha)t/2} - e^{-(\gamma_\alpha + \gamma_\beta)t/2}) \\ &\quad + \frac{\Omega_\alpha \Omega_\beta}{(\kappa - \gamma_\alpha)(\kappa + \gamma_\beta)} (e^{-(\kappa + \gamma_\beta)t/2} - e^{-(\gamma_\alpha + \gamma_\beta)t/2}) \end{aligned} \quad (\text{B.5})$$

$$u_{\alpha c}(t) = -\frac{\Omega_\alpha}{\kappa + \gamma_\alpha} (1 - e^{-(\kappa + \gamma_\alpha)t/2}) \quad (\text{B.6})$$

$$u_{\beta\beta}(t) = 1 + \frac{\Omega_\beta^2}{\gamma_\beta(\kappa + \gamma_\beta)} (1 - e^{-\gamma_\beta t}) - \frac{2\Omega_\beta^2}{\kappa^2 - \gamma_\beta^2} (e^{-\gamma_\beta t} - e^{-(\kappa + \gamma_\beta)t/2}) \quad (\text{B.7})$$

$$u_{\beta c}(t) = -\frac{\Omega_\beta}{\kappa + \gamma_\beta} (1 - e^{-(\kappa + \gamma_\beta)t/2}) \quad (\text{B.8})$$

$$u_{cc}(t) = 1 \quad (\text{B.9})$$

The vector of coordinate averages $\bar{\mathbf{q}}$ appearing in ansatz (73) obeys the stochastic equation

$$d\bar{\mathbf{q}} = \frac{1}{2} \begin{pmatrix} -\gamma_\alpha & 0 & 0 \\ 0 & -\gamma_\beta & 0 \\ \Omega_\alpha & \Omega_\beta & -\kappa \end{pmatrix} dt \bar{\mathbf{q}} + \frac{1}{2} [\text{Id} - \underline{c}(t)] \begin{pmatrix} \gamma_\alpha^{1/2} d\zeta_\alpha(t) \\ \gamma_\beta^{1/2} d\zeta_\beta(t) \\ -\kappa^{1/2} d\zeta_c(t) \end{pmatrix} \quad (\text{B.10})$$

The unconditional expectation value $\langle P_a^2 \rangle$ always being equal to 1/4, the mean and the variance of P_a conditioned on the integrated homodyne signal are still given by equations (85) and (86), by generalizing the expressions (92) of the variances and covariance of the random variables $\bar{P}_a(t)$ and $\sigma(t)$ in the case of three independent stochastic processes $d\zeta_\alpha(t')$, $d\zeta_\beta(t')$ and $d\zeta_c(t')$ as follows:

$$\begin{aligned} \langle \sigma(t) \bar{P}_a(t) \rangle_{\text{stoch}} &= \int_0^t dt' \sum_{\nu \in \{\alpha, \beta, c\}} p_\nu(t, t') \sigma_\nu(t, t') \quad ; \quad \langle \sigma^2(t) \rangle_{\text{stoch}} = \int_0^t dt' \sum_{\nu \in \{\alpha, \beta, c\}} \sigma_\nu^2(t, t') \quad ; \\ \langle \bar{P}_a^2(t) \rangle_{\text{stoch}} &= \int_0^t dt' \sum_{\nu \in \{\alpha, \beta, c\}} p_\nu^2(t, t') \end{aligned} \quad (\text{B.11})$$

with the compact expressions of the corresponding amplitudes

$$p_\nu(t, t') = (-1)^{\delta_{\nu c}} \frac{\sqrt{\gamma_\nu}}{2\Omega} \left\{ \Omega_\beta e^{-\gamma_\alpha(t-t')/2} [\delta_{\alpha\nu} - c_{\alpha\nu}(t')] - \Omega_\alpha e^{-\gamma_\beta(t-t')/2} [\delta_{\beta\nu} - c_{\beta\nu}(t')] \right\} \quad (\text{B.12})$$

$$\sigma_\nu(t, t') = \frac{\delta_{\nu c}}{2t} + (-1)^{\delta_{\nu c}} \frac{\sqrt{\kappa\gamma_\nu}}{2t} \left\{ [\delta_{c\nu} - c_{c\nu}(t')] f_\kappa(t-t') + \sum_{\mu \in \{\alpha, \beta\}} \frac{\Omega_\mu}{\kappa - \gamma_\mu} [\delta_{\mu\nu} - c_{\mu\nu}(t')] [f_{\gamma_\mu}(t-t') - f_\kappa(t-t')] \right\} \quad (\text{B.13})$$

The index ν runs on the three values α, β, c and we set $\gamma_c = \kappa$. The δ function is that of Kronecker, and the f_λ function is the same as in equations (88)-(91).

The general solution that we have just presented includes the five rates $\gamma_\alpha, \Gamma_{\text{ex}} = \Omega_\alpha^2/\kappa, \gamma_f$ on the one hand, γ_β, κ on the other hand. The experimentally relevant regime is one where the last two are “infinitely” larger than the first three and only contribute through unobservable transient regimes. Mathematically, we reach this limit by making γ_f tend to zero with $\kappa, \gamma_m, \gamma_0$ and Ω fixed and with $\tau = \Gamma_{\text{ex}} t > 0$ fixed. Then the first three rates jointly tend towards zero, that is with finite-limit ratios $\Gamma_{\text{ex}}/\gamma_f \rightarrow \Omega^2 \gamma_m / [\kappa(\gamma_0 + \gamma_m)^2]$ and $\gamma_\alpha/\gamma_f \rightarrow 2\gamma_0/(\gamma_0 + \gamma_m)$, the rate γ_β reduces to $\gamma \equiv 2(\gamma_0 + \gamma_m)$ and the Faraday coupling Ω_β to Ω . All exponential transients disappear in the matrix elements (B.4)-(B.8) of \underline{u} except those relaxing at the rate γ_α . The amplitudes (B.12) and (B.13) on stochastic processes reduce to

$$\frac{p_\alpha(t, t')}{\sqrt{\Gamma_{\text{ex}}}} = \frac{u(\tau') - 1}{2u(\tau')} \sqrt{\epsilon} e^{-\epsilon(\tau-\tau')/2} \quad \frac{\sigma_\alpha(t, t')}{\Gamma_{\text{ex}}} = \frac{u(\tau') - 1}{\tau u(\tau')} \sqrt{\epsilon} \frac{1 - e^{-\epsilon(\tau-\tau')/2}}{\epsilon} \quad (\text{B.14})$$

$$\frac{p_\beta(t, t')}{\sqrt{\Gamma_{\text{ex}}}} = \frac{\sqrt{\rho}}{(1+\rho)u(\tau')} e^{-\epsilon(\tau-\tau')/2} \quad \frac{\sigma_\beta(t, t')}{\Gamma_{\text{ex}}} = \frac{\sqrt{\rho}}{(1+\rho)\tau} \left[\frac{2}{u(\tau')} \frac{1 - e^{-\epsilon(\tau-\tau')/2}}{\epsilon} + \rho + \frac{\gamma}{\kappa}(\rho - 1) \right] \quad (\text{B.15})$$

$$\frac{p_c(t, t')}{\sqrt{\Gamma_{\text{ex}}}} = \frac{(1-\rho)}{2(1+\rho)u(\tau')} e^{-\epsilon(\tau-\tau')/2} \quad \frac{\sigma_c(t, t')}{\Gamma_{\text{ex}}} = \frac{1}{\tau} \left[\frac{1}{2} + \frac{1-\rho}{1+\rho} \frac{1}{u(\tau')} \frac{1 - e^{-\epsilon(\tau-\tau')/2}}{\epsilon} + \frac{\rho}{1+\rho} \left(1 + \frac{2\gamma}{\kappa} \right) \right] \quad (\text{B.16})$$

where $\epsilon = \gamma_\alpha/\Gamma_{\text{ex}}$ as in section 4.2.4, the function $u(\tau)$ is given by equation (100) and the notation $\rho = \Omega^2 \kappa / [\gamma(\kappa + \gamma)^2]$ generalizes the one of footnote 22. Relations (85) and (86) remain valid, with the new expressions for the variance and covariance

$$\boxed{\frac{\langle \sigma^2 \rangle_{\text{stoch}}}{\Gamma_{\text{ex}}} = \frac{\epsilon\tau - 2(1 - e^{-\epsilon\tau/2})}{\epsilon^2 \tau^2} + \frac{\Gamma_{\text{ex}}}{4\tau \Gamma_{\text{sq}}^{\text{gen}}} \quad \text{and} \quad \frac{\langle \sigma \bar{P}_a \rangle_{\text{stoch}}}{\sqrt{\Gamma_{\text{ex}}}} = \frac{1 - e^{-\epsilon\tau/2}}{2\epsilon\tau}} \quad (\text{B.17})$$

and the generalized squeezing rate

$$\boxed{\Gamma_{\text{sq}}^{\text{gen}} = \left[\frac{1}{\Gamma_{\text{ex}}} + \frac{2(\gamma_0 + \gamma_m)}{\gamma_f \gamma_m} \right]^{-1}} \quad (\text{B.18})$$

which reproduce the variance and covariance (104) and (105) of the one-mode model with decoherence when $\Gamma_{\text{ex}}/\gamma_f \rightarrow 0$ and the spin squeezing rate (95) of the three-mode model without decoherence when $\gamma_0 \rightarrow 0$. The new results can be simplified within the useful limit of weak effective decoherence $\gamma_\alpha/\Gamma_{\text{ex}} \rightarrow 0$ by a order-one expansion in ϵ , which allows to generalize (106) as follows on the conditional mean and variance at a non-infinitesimal value of $\Gamma_{\text{ex}}/\gamma_f$:

$$m(t) = \frac{\Gamma_{\text{sq}}^{\text{gen}} t}{1 + \Gamma_{\text{sq}}^{\text{gen}} t} - \frac{\gamma_\alpha}{\Gamma_{\text{sq}}^{\text{gen}}} \frac{(3 + \Gamma_{\text{sq}}^{\text{gen}} t)(\Gamma_{\text{sq}}^{\text{gen}} t)^2}{12(1 + \Gamma_{\text{sq}}^{\text{gen}} t)^2} + O[(\gamma_\alpha t)^2] \quad (\text{B.19})$$

$$\mathcal{V}(t) = \frac{1}{4(1 + \Gamma_{\text{sq}}^{\text{gen}} t)} + \frac{\gamma_\alpha}{\Gamma_{\text{sq}}^{\text{gen}}} \frac{(\Gamma_{\text{sq}}^{\text{gen}} t + 3/2)(\Gamma_{\text{sq}}^{\text{gen}} t)^2}{12(1 + \Gamma_{\text{sq}}^{\text{gen}} t)^2} + O[(\gamma_\alpha t)^2] \quad (\text{B.20})$$

This generalization simply amounts to replace τ by $\Gamma_{\text{sq}}^{\text{gen}} t$ and ϵ by $\gamma_\alpha/\Gamma_{\text{sq}}^{\text{gen}}$ in the right-hand sides of (106).²⁷ The optimal squeezing on P_a is then obtained at a time $t_{\text{opt}} \sim (3/\Gamma_{\text{sq}}^{\text{gen}} \gamma_\alpha)^{1/2}$ and corresponds to a conditional variance $\text{Var}_{\sigma=S}^{\text{opt}}(P_a) \sim (\gamma_\alpha/12\Gamma_{\text{sq}}^{\text{gen}})^{1/2}$; the optimal metrological gain is deduced from this by equation (13).

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²⁷ It is in fact valid for all orders in ϵ since the proposed replacement does not change $\epsilon\tau$ (always equal to $\gamma_\alpha t$) and transforms equations (104) and (105) into equation (B.17).

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