Fundamental limitations in spin-ensemble quantum memories for cavity fields

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Inhomogeneously broadened spin ensembles play an important role in present-day implementation of hybrid quantum processing architectures. When coupled to a resonator such an ensemble may serve as a multi-mode quantum memory for the resonator field, and by employing spin-refocusing techniques the quantum memory time can be extended to the coherence time of individual spins in the ensemble. In the present paper we investigate such a memory protocol capable of storing an unknown resonator-field state, and we examine separately the various constituents of the protocol: the storage and read-out part, the memory hold time with the spin ensemble and resonator field decoupled, and the parts employing spin refocusing techniques. Using both analytical and numerical methods we derive how the obtainable memory performance scales with various physical parameters.

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I. INTRODUCTION

During the past decade various physical ensemble systems have been utilized as quantum memories for propagating optical fields. The first experimental demonstrations typically employed alkali-vapors [1–4], while rare-earth-metal ions in solids have later been used [5,6]. In the solid-state implementations the transition frequencies are inhomogeneously broadened due to variations in the local environment, which on the one hand allows multi-mode performance but on the other hand presents a challenge: The retrieval of the stored quantum information requires the dephasing caused by the frequency inhomogeneity to be reversed. To this end, atomic frequency-comb (AFC) techniques [7] and controlled reversible inhomogeneous broadening (CRIB) [10,11] were employed in Refs. [7] and [8], respectively. In both examples, effectively homogeneous subsets of the inhomogeneously broadened ensemble were prepared by hole-burning techniques—thus sacrificing optical depth of the material to achieve coherence. For classical light pulses, these preparation steps can be avoided using certain spin-refocusing techniques [12,13], essentially based on the Hahn echo [14]. However, these have been shown to be inapplicable at the quantum level due to noise generated by excited-state absorbers [12,16]. Nonetheless, a simple scheme using two π-pulses has recently been proposed which uses a silencing mechanism to prevent emission from the excited-state ensemble after the first π-pulse while allowing a faithful read-out from the non-inverted ensemble after the second π-pulse [17]. This revival-of-silenced-echo (ROSE) protocol plays an important role in the present paper.

The key feature of the above-mentioned ensemble approaches is the collective dipole-moment enhancement, which enables a sufficiently strong free-space light-matter interaction. This is in stark contrast to cavity quantum electrodynamics (CQED) where the radiation field is enhanced by a high-finesse cavity in order to interact efficiently with a single matter particle [18]. In an intermediate regime proposals exist to use inhomogeneously broadened ensembles coupled to cavities of moderate finesse as quantum memories for propagating optical fields [19,20]. The enhancement by a cavity ensures sufficient optical depth in the AFC and CRIB quantum memory schemes mentioned above.

In the microwave regime, the strong-coupling regime has recently been reached between superconducting coplanar waveguide resonators and, on the one hand, ensembles of electronic spins [21,22], and on the other hand, superconducting Joseph-junction qubits [24]. Due to the tunability of such resonators it is possible to construct hybrid quantum systems with the cavity acting as a “quantum bus” between a “processor” and a “memory” unit [25,28]. While preselection of spectral portions of the spin ensemble constitutes a means to mitigate the effects of inhomogeneous broadening [29], the tunability of the cavity also enables the implementation of the ROSE protocol [30,31]. Noting that electron-spin degrees of freedom may even be transferred to nuclear-magnetic degrees of freedom [32], the superconducting CQED has brought back the modern concepts of spin-refocusing quantum memories to their origin of ESR and NMR.

In Ref. [30] we described how the ROSE protocol can be implemented between a tunable microwave-resonator quantum bus and a spin-ensemble quantum memory, and the feasibility of the protocol in its entirety was assessed for the special case using nitrogen-vacancy centers in diamond as the memory unit. The present paper is targeted on isolating and assessing the effect of the individual constituents of the protocol; the spin-cavity transfer mechanism, the silencing mechanism, the π-pulse process, and the role of decoherence. Hence, our work is intended to present a firm base for developing and optimizing specific quantum-memory protocols for intra-cavity fields using.
The paper is arranged as follows: In Sec. III the basic equations and assumptions are presented for the spin ensemble system coupled to a cavity, and in Sec. III it is briefly reviewed how this system enables a quantum memory protocol. The actual examination of the quantum memory performance begins in Sec. IV with an account for the storage and read-out part of the protocol, in Sec. V the decoupling of the spin ensemble from the cavity is examined, and in Sec. VI the effect of refocusing mechanisms is investigated. The paper is concluded with a brief discussion in Sec. VII, and we present a number of mathematical derivations in Appendixes A–E.

II. PHYSICAL MODELING AND EQUATIONS OF MOTION

The physical system under consideration is shown schematically in Fig. 1(a). The field $\hat{a}_c$ in a one-sided cavity is coupled to a spin ensemble, and the frequency $\omega_j$ of the $j$th spin is assumed to be static but inhomogeneously broadened around a central spin frequency $\omega_s$ with a Lorentzian distribution:

$$f(\omega) = \frac{w/2\pi}{(\omega - \omega_s)^2 + \frac{w^2}{4}},$$  \hspace{1cm} (1)

where $w$ is the full width at half maximum (FWHM). In the frame rotating at $\omega_s$ the free evolution of the spin ensemble and the cavity field is governed by the Hamiltonian (taking $\hbar = 1$):

$$\hat{H}_0 = \Delta_{cs}\hat{a}_c^\dagger\hat{a}_c + \sum_j \frac{\Delta_j}{2}\hat{\sigma}_z^{(j)},$$ \hspace{1cm} (2)

where $\Delta_{cs} = \omega_c - \omega_s$ is the detuning of the cavity resonance frequency $\omega_c$, and $\Delta_j = \omega_j - \omega_s$. The coupling between the spin ensemble and the cavity field is governed by the interaction Hamiltonian:

$$\hat{H}_1 = \sum_j g_j(\hat{\sigma}_+^{(j)}\hat{a}_c + \hat{\sigma}_-^{(j)}\hat{a}_c^\dagger),$$ \hspace{1cm} (3)

where $g_j$ is the coupling strength of the $j$th spin and $\hat{\sigma}_+^{(j)}$, $\hat{\sigma}_-^{(j)}$, and $\hat{\sigma}_z^{(j)}$ are Pauli operators. The cavity field can be driven by a coherent-state field $\beta$ with the associated Hamiltonian:

$$\hat{H}_{ext} = i\sqrt{2\kappa}(\beta\hat{a}_c^\dagger - \beta^\ast\hat{a}_c),$$ \hspace{1cm} (4)

where $\kappa$ is the field-decay rate of the cavity and $\beta$ is normalized such that $|\beta|^2$ is the number of photons incident on the cavity per second. Decay processes are handled in the Markov approximation, e.g. by the master equation: $\frac{d\hat{\rho}}{dt} = -i[\hat{H}, \hat{\rho}] + \sum_{\alpha}\mathcal{L}[\hat{c}_\alpha]\hat{\rho}$, where $\hat{H}$ is the total Hamiltonian and the Lindblad part is given by $\mathcal{L}[\hat{c}_\alpha]\hat{\rho} = -\frac{1}{2}\hat{c}_\alpha\hat{c}_\alpha^\dagger\hat{\rho} - \frac{1}{2}\hat{\rho}\hat{c}_\alpha^\dagger\hat{c}_\alpha + \hat{c}_\alpha^\dagger\hat{c}_\alpha\hat{\rho}$. The cavity leakage gives rise to a Lindblad term with $\hat{c}_\xi = \sqrt{2\kappa}\hat{a}_c$, and for each individual spin a collision-like dephasing with characteristic waiting time $\tau$ is modeled by $\hat{c}_j = \frac{1}{\sqrt{2\tau}}\hat{\sigma}_z^{(j)}$.

This manuscript applies the above formalism in two regimes: (i) In a quantum-memory protocol the exchange of information between the cavity field and the spin ensemble occurs in the linear regime with $\langle\hat{\sigma}_z^{(j)}\rangle \approx -1$, which can be described in the Holstein-Primakoff approximation [33]. For the specific choice of Lorentzian inhomogeneous broadening the dynamical evolution may often be described exactly or approximately by analytical expressions (the reason for this is discussed in appendix A), which enhances the physical understanding of the processes involved. (ii) In order to employ spin-refocusing techniques in the quantum-memory protocol, the spin ensemble must be subjected to $\pi$-pulses which involves a non-linear regime of the dynamical evolution. This can only be handled numerically; we shall employ a method which divides the inhomogeneous spin ensemble into homogeneous sub-ensembles, $\mathcal{M}_1, \mathcal{M}_2, \ldots, \mathcal{M}_M$:

$$\hat{S}_x^{(m)} = \sum_{j \in \mathcal{M}_m} \hat{\sigma}_x^{(j)} \quad \hat{S}_y^{(m)} = \sum_{j \in \mathcal{M}_m} \hat{\sigma}_y^{(j)} \quad \hat{S}_z^{(m)} = \sum_{j \in \mathcal{M}_m} \hat{\sigma}_z^{(j)},$$ \hspace{1cm} (5)

and which accounts for the quantum state through the first and second moments of the physical variables (details can be found in Ref. [30]). Comments on the accuracy of the numerical procedure is presented in Appendix D.

FIG. 1. (a) The physical system under consideration: A spin ensemble is placed within a one-sided cavity, which may be driven externally by the field $\beta$ through a mirror with field-decay rate $\kappa$. (b) The quantum-memory protocol consists of storage and retrieval parts with a strongly and resonantly coupled spin-cavity system (shaded area), $\pi$-pulses for inverting the spin ensemble in a resonant but weakly coupled regime (hatched area), and parts (white) with effective spin-cavity decoupling. Due to the two $\pi$-pulses, a silenced spin echo occurs in the middle of the sequence. For later reference $T_i$ denotes the duration of the $i$th part of the sequence.
III. A SPIN-ENSEMBLE QUANTUM MEMORY: THE BASIC IDEA

The fundamental idea behind the spin-ensemble quantum memory relies on the specific interaction Hamiltonian \( \hat{H}_1 = g_{\text{ens}}(\hat{a}_c \hat{b}^\dagger + \hat{a}^\dagger_c \hat{b}) \), which can be rewritten as: \( \hat{H}_1 = g_{\text{ens}}(\hat{a}_c \hat{b}^\dagger + \hat{a}^\dagger_c \hat{b}) \), where the collective-spin-mode annihilation operator is given by:

\[
\hat{b} = \sum_j g_j \hat{a}_j^{(j)},
\]

and \( g_{\text{ens}} = [\sum_j g_j^2]^{1/2} \) is the ensemble coupling constant scaling as \( \sqrt{N} \) times the individual-spin coupling strength. The creation operator \( \hat{b}^\dagger \) is the hermitian conjugate of \( \hat{b} \), and the specific choice of \( g_{\text{ens}} \) ensures the commutation relation \( [\hat{b}, \hat{b}^\dagger] = 1 \) in the linear regime with \( \langle \hat{a}_j^{(j)} \rangle \approx -1 \). On resonance \( (\Delta_{\text{ens}} = 0) \), and in the absence of inhomogeneities and decay mechanisms \( (\Delta_j = \kappa = \tau^{-1} = 0) \), the evolution is governed solely by \( \hat{H}_1 \) leading to the following equations of motion in the Heisenberg picture:

\[
\begin{align*}
\dot{\hat{a}}_c(t) &= \cos(g_{\text{ens}} t) \hat{a}_c(0) - i \sin(g_{\text{ens}} t) \hat{b}(0), \\
\dot{\hat{b}}(t) &= -i \sin(g_{\text{ens}} t) \hat{a}_c(0) + \cos(g_{\text{ens}} t) \hat{b}(0).
\end{align*}
\]

At time \( T_{\text{swap}} = \frac{\pi}{2g_{\text{ens}}} \) the cavity-field and spin-ensemble quantum states are swapped. In a practical realization, however, there are several challenges to address: (i) The inhomogeneity in spin-resonance frequencies gives rise to a separate phase evolution for the individual spins, \( \hat{a}_j^{(j)}(t) = \hat{a}_j^{(j)}(0)e^{-i\Delta_j t} \), and the spin-ensemble excitation quickly disappears from the symmetric mode \( \hat{b} \) coupled to the cavity. (ii) This dephasing mechanism can be counter-acted by spin-refocusing techniques, which however involves that the entire spin ensemble must be transferred to the excited state with possible instabilities as a concern [34]. (iii) A mechanism must be devised in order to effectively switch on and off the basic interaction of Eq. (7). (iv) The impact of decay processes (modeled by \( \kappa \) and \( \tau \)) and their interplay with the ensemble characteristics \( (g_{\text{ens}} \text{ and } \omega_s) \) must be understood in order to devise the best parameter regime. (v) The spin-refocusing mechanisms may be driven by an external field \( \beta \) through the cavity, and the feasibility of this process in terms of energy or power must be accounted for.

The process in Eq. (7) has been demonstrated experimentally in nitrogen-vacancy (NV) centers in diamond coupled to a co-planar micro-wave cavity [27]. In that work it was not attempted to employ spin-refocusing techniques, and inhomogeneous broadening, indeed, presented the main limitation of the quantum memory.

A. A quantum memory protocol enabled by spin-refocusing techniques

In this manuscript it is assumed that an initial state of the cavity field is given, and our task is to transfer this state to the spin ensemble and retrieve it again after a specified memory time, \( T_{\text{mem}} \). To reach this goal we employ a quantum-memory protocol, the most important constituents of which have been shown schematically in Fig. 1(b). First, by a storage process the cavity field must be transferred to the spin ensemble. During this process the cavity leakage should be minimized, i.e. the cavity-\( Q \)-parameter must be high in order that the cavity-field decay rate is much slower than the characteristic transfer rate. The same applies for the retrieval process at the end of the protocol. Second, in order to employ spin-refocusing techniques the spin ensemble must reside in an inverted state (roughly) half of the memory time and near the ground state for the remaining time, and two \( \pi \)-pulses are applied in order to facilitate this. To prevent superradiant processes during these pulses and while the spins are excited, the cavity must be in a low-\( Q \) mode to ensure stability [34]. Third, in between storage, retrieval, and \( \pi \)-pulses the spin-cavity should evolve freely for durations adjusted by the requirement that the spin-refocusing process matches the desired memory time. In these periods the spin-cavity system should be decoupled to prevent leakage of the stored quantum state and to prevent generation of excess noise. We shall decouple the spin-cavity system effectively by detuning the cavity frequency from the spin-resonance frequency. Finally, between the main parts of the protocol shown in Fig. 1(b) there are short periods of time during which the cavity parameters, \( \kappa \) and \( \omega_s \), are adjusted. In this work they are treated as infinitely fast and are thus disregarded. See Ref. [30] for further details of a practical memory protocol and of its experimental feasibility.

According to Eq. (7) an ideal memory swaps the state of a cavity field into and back from the spin ensemble oscillator according to: \( X_c^{\text{out}} = -X_c^{\text{in}} \) and \( P_c^{\text{out}} = -P_c^{\text{in}} \), where

\[
\dot{X}_c = \frac{\hat{a}_c + \hat{a}_c^\dagger}{\sqrt{2}}, \quad \dot{P}_c = -i(\hat{a}_c - \hat{a}_c^\dagger),
\]

fulfill the commutation relation \([X_c, P_c] = i\). For this reason we define the gain \( G \) in terms of the mean values of the actual process as:

\[
X_c^{\text{out}} = -G X_c^{\text{in}}, \quad P_c^{\text{out}} = -G P_c^{\text{in}}.
\]

We also note that the minimum uncertainty state of the cavity field fulfills \( \langle \delta X_c^2 \rangle = \langle \delta P_c^2 \rangle = \frac{1}{2} \), which holds in particular for all coherent states. Hence, when applying coherent input states to the quantum memory, the variance of the output state quadratures \( \sigma^2 \equiv \langle \delta X_c^{\text{out}} \rangle = \langle \delta P_c^{\text{out}} \rangle \geq \frac{1}{2} \) is a measure of the added noise from the memory protocol.

Due to the operator nature of Eq. (7) the quantum memory should work ideally for any quantum state, and
the performance of the memory protocol is fully characterized by its impact on coherent states due to their (over)completeness. As will be quantitatively justified in Sec. VI B the protocol is well described in practice by a linear input-output relation, and in this case the transformation of the first and second moments of $X_c$ and $P_c$, parametrized through $G$ and $\sigma^2$, is sufficient for characterizing the quantum memory performance \[32\]. We note that the protocol may give rise to phase rotations and asymmetries in the above input-output relations, and in this case the above definitions of $G$ and $\sigma^2$ must be generalized, which is the subject of Appendix B.

IV. SWAPPING BETWEEN THE CAVITY FIELD AND SPIN COMPONENTS

This section considers the impact of inhomogeneous broadening and decay mechanisms on the otherwise idealized spin-cavity evolution of Eq. (7). We start with a discussion of mean values while second moments are covered in the end of the section.

For the storage part of the quantum memory protocol, the relevant initial state is the ground state for the spin ensemble, $\langle \hat{\sigma}_z^{(j)}(0) \rangle = -1$ and $\langle \hat{a}_c^{(j)}(0) \rangle = 0$, i.e. $\langle \hat{b}(0) \rangle = 0$, while we take the cavity-field to be in a coherent state of amplitude $\langle \hat{a}_c(0) \rangle = \alpha$. With the cavity coupled resonantly to the spin ensemble ($\Delta_{cs} = 0$) the subsequent evolution follows Eqs. (C1) and (C2) as exemplified by the solid curves in Fig. 2(a). In comparison to the idealized behavior of Eq. (7) we now observe an exponentially decaying envelope function $\exp(-\frac{1}{2}[\kappa + \Gamma]t)$, where $\Gamma = w + \frac{1}{2\tau}$, and also a slight slow-down of the oscillatory rate $g_{ens} \rightarrow g'_{ens}$, with $g'_{ens}$ being stated after Eq. (C2). The swapping time $T_{swap}$ is defined by the requirement $\langle \hat{a}_c(T_{swap}) \rangle = 0$, which is given analytically by Eq. (C3).

The properties of the spin state immediately after the resonant swap process, $t = T_{swap}$, is given not only by the specific spin-mode $\hat{b}$—due to the spin-frequency inhomogeneity the stored information is distributed among other spin modes already during the swapping part, and the exact details of this distribution will eventually affect the quantum-memory fidelity. The spin-state mean values at $t = T_{swap}$ can in principle be calculated by formal integration, $\langle \hat{\sigma}_z^{(j)}(T_{swap}) \rangle = -ig_j \int_0^{T_{swap}} e^{-(\gamma_\perp + i\Delta_j)(T_{swap} - t')\langle \hat{a}_c(t') \rangle}dt'$ using Eq. (C1) where $\gamma_\perp = \tau^{-1}$. It is more instructive, however, to examine the free evolution of spins for $t \geq T_{swap}$, and to this end we artificially decouple the spin-cavity system at $t = T_{swap}$ by setting $g_j \rightarrow 0$. The subsequent evolution of $\langle \hat{a}_c \rangle$ and $\langle \hat{b} \rangle$ is exemplified in Fig. 2(a) with dashed curves. While $\langle \hat{a}_c \rangle$ remains constant at zero, the spin mode $\langle \hat{b} \rangle$ decays due to the dephasing from inhomogeneous broadening. However, if we at $t = T_{swap}$ also impose an ideal inversion process (around the y-axis), $\hat{\sigma}_z^{(j)} \rightarrow -\hat{\sigma}_z^{(j)}$ and $\hat{b} \rightarrow -\hat{b}$, the subsequent evolution will eventually affect the quantum-memory fidelity.

**FIG. 2.** (a) The solid curves show the evolution of $\langle \hat{a}_c \rangle$ (blue) and $\langle \hat{b} \rangle$ (red) versus time for a coupled spin-cavity system with $g_{ens} = 2.5\Gamma$. The dashed lines show the free evolution if $g_{ens}$ is changed to zero at $t = T_{swap}$, and the dotted lines show the same but with a perfect inversion process included at $t = T_{swap}$ (the blue dashed and dotted lines coincide). (b) The solid curves show the phase $\phi_j$ of individual spin components for four different frequency classes with $\Delta_j$ taking the values (counting from the horizontal axis and away): $w$ (red), $2w$ (green), $3w$ (blue), and $4w$ (cyan). The inversion process implies $\phi_j \rightarrow -\phi_j$ at $t = T_{swap}$ with the dashed lines representing the free evolution in the absence of the inversion process and the dotted lines (for the two lowest values of $\Delta_j$) the hypothetical free evolution of duration $T_{focus}$ back in time to the effective point of origin. In panel (c) the thick solid line (red) shows the phase $\phi_j$ versus $\Delta_j$ at $t = T_{swap}$. The thin gray curve corresponds to the four cases examined in (b). The dashed line represents a fit of $T_{focus}$ to the function $\phi_j = \frac{\pi}{2} + \Delta_j T_{focus}$ in the linear regime near $\Delta_j = 0$, and the dotted lines follow $\phi_j = \Delta_j T_{swap}$ and $\phi_j = \Delta_j T_{swap} + \pi$. The gray curve represents (on an arbitrary vertical scale) the inhomogeneous distribution of Eq. (1) used in this example.
where the phases are defined as $\langle \hat{\phi}^{(j)} \rangle \equiv |\langle \hat{\phi}^{(j)} \rangle| e^{-i\phi_j}$. The solid curves represent the scenario of decoupling ($g_j \to 0$) and ideal inversion immediately after the swapping process at $t = T_{\text{swap}}$, and the spin echo occurs when the phases refocus at $\phi_j - \frac{\pi}{2} \approx 0$. However, as is evident from the zoom-in panel, the different classes are refocused at slightly different times, which eventually leads to a slight degradation in quantum-memory performance. To elucidate this phenomenon even further, the accumulated phase $\phi_j$ during the swap is shown versus $\Delta_j$ in Fig. 2(c). Clearly, for small values of $|\Delta_j|$ there is a linear dependence and we define the slope to be $T_{\text{locus}}$ (i.e. the duration for refocusing in this linear regime) being represented by the dashed line. This slope depends on a non-trivial way on the inhomogeneous distribution, the coupling constants, and the decay processes; however, a crude estimate $T_{\text{locus}} \approx \frac{2}{T_{\text{swap}}}$ can be made, see Eq. (C5). For large values of $|\Delta_j|$ another slope equal to $T_{\text{swap}}$ occurs (dotted lines in Fig. 2(c), see Eq. (C4)), and the actual phase is seen to change smoothly between these two regimes, which are distinguished by $|\Delta_j|$ being either smaller or larger than the characteristic rate $T_{\text{swap}}^{-1}$ of the cavity-field variations. The above observations resemble the role of aberrations in geometric optics, where e.g. rays far from the center of lenses give rise to imperfect imaging properties. In our case the magnitude of imperfections is governed by the width of the linear regime compared to the inhomogeneous frequency distribution (thin gray curve in Fig. 2(c)) of spins storing the information—we shall be more quantitative on this below.

In order to calculate the impact of the storage and retrieval part on a quantum-memory protocol we employ the following idealized scenario: (1) The storage and retrieval parts have durations (referring to Fig. 1(b)) $T_1 = T_2 = T_{\text{swap}}$ defined by the condition $\langle \hat{a}_c(T_{\text{swap}}) \rangle = 0$. (2) For the decoupled parts, and during the $\pi$-pulses, we set $g_j = 0$ such that the decoupling is ideal. (3) The inversion pulses are perfect and infinitely fast. (4) The duration of the three decoupling parts are adjusted such that $T_2 = T_0$, $\sum_{i=1}^2 T_i = T_{\text{mem}}$, and $2T_{\text{locus}} + \sum_{i=2}^6 T_i = 2T_4$ (the latter ensures an even amount of time spent in the inverted and non-inverted states between the effective focus points). The choice of equal swapping times and focusing times for the storage and retrieval parts is not obvious and will be commented on below. We note that for $\gamma = 0$ the decoupling parts just serve as delay with no degradation in memory performance.

Now, consider Fig. 3(a) which shows the gain degradation versus $g_{\text{ens}}/\Gamma$. Evidently, the performance improves when this ratio increases; for larger $g_{\text{ens}}$ the swap process occurs faster, and in turn a larger fraction of the spin ensemble remains in the linear region (i.e. their phase is determined by the coupling to the field and not by their own frequency during the swap). For increasing $g_{\text{ens}}/\Gamma$ the central linear regime in Fig. 2(c) grows in comparison to the width of the Lorentzian distribution.

Next, for a fixed ratio $g_{\text{ens}}/\Gamma = 2.5$ leading to the gain $G_0 = 0.997$ in Fig. 3(a), we wish to examine the impact of non-zero $\kappa$ and $\gamma_{\perp}$. From the functional behavior of Eqs. (C1) and (C2), i.e. from the envelope function $e^{-\frac{\Gamma}{2}(\kappa + \gamma_{\perp})t}$ in the homogeneous case, we make a naive guess that during the total storage and retrieval time, $2T_{\text{swap}}$, the gain degradation amounts to $e^{-\frac{\Gamma}{2}(\kappa + \gamma_{\perp})T_{\text{swap}}}$ while only $\gamma_{\perp}$ plays a role in between according to $e^{-\gamma_{\perp}T_{\text{mem}} - 2T_{\text{swap}}}$.

Hence, we expect $\tilde{G} = G_0 e^{-\kappa T_{\text{swap}} + \gamma_{\perp}T_{\text{mem}} - 2T_{\text{swap}}}$, which is examined by the numerical results presented in Fig. 3(b+c). In panel (b) the cavity leakage is absent, $\kappa = 0$, while both $\gamma_{\perp}$ and $T_{\text{mem}}$ are varied. In panel (c) the spin decoherence is turned off, $\gamma_{\perp} = 0$, while $\kappa$ is varied. In both panels the solid curve represents the above expectation and corresponds to the numerical results to a good approximation. For this reason we wish to maintain the above naive but simple expectation as a convenient rule of thumb.

The above considerations, however, do not represent the optimum solution, which must in practice be calculated numerically. To illuminate the underlying problems, consider first the swapping time, $T_{\text{swap}}$, which we took as equal for the storage and retrieval parts. In the presence of decay mechanisms ($\kappa$ and $\gamma_{\perp}$, not both zero) there is actually an asymmetry in the storage and retrieval part: The perfect retrieval consists of a cavity field $\langle \hat{a}_c(T_{\text{mem}}) \rangle = -G_0$ traced backward in time until $\langle \hat{a}_c(T_{\text{mem}} - T_{\text{rev}}) \rangle = 0$, but the reversed direction of
time leads to an exponentially increasing behavior due to \( \kappa \) and \( \gamma_\perp \), and hence the “reverse swapping time”, \( T_{\text{rev}}^{\text{focus}} \), given by Eq. (C3) with sign changes on \( \kappa \) and \( \gamma_\perp \), is not exactly equal to the forward swap time \( T_{\text{swap}} \). Second, in the same manner the focusing time is also changed, \( T_{\text{focus}} \to T_{\text{focus}}^{\text{rev}} \), and we may wish to test the implications by the following protocol: (1) The storage and retrieval times are set to \( T_1 = T_{\text{swap}} \) and \( T_7 = T_{\text{swap}} \), (2) the decoupling periods are adjusted according to \( T_{\text{focus}} + T_{\text{focus}}^{\text{rev}} + \sum_{i=2}^{6} T_i = 2T_4 \) instead of the choice made above. The phases \( \phi_j \) of individual spin components then follow the red solid line in Fig. 3(d) after the initial storage, and the time-reversed evolution of the perfect retrieval amounts to the green dashed line (the example here corresponds to the parameters of the right-most data point in Fig. 3(c), i.e. \( \gamma_\perp = 0 \) and \( \kappa \neq 0 \)). Now, the mathematical implications of the refocusing mechanism is to change the slope of the red solid line reaching the black dotted curve. The particular usage of \( T_{\text{focus}} \) and \( T_{\text{focus}}^{\text{rev}} \) ensures a perfect match of slopes in the central part with small \( |\Delta_j| \); however, the entire frequency range cannot be matched, and it is intuitively clear that slight changes in the slope of either the dashed or dotted curves in Fig. 3(d) might improve the performance. The gain resulting from the scenario of panel (d) is in fact 2 \% lower than the value found in panel (c). Although the choices behind the result of Fig. 3(a,b,c) are not reached by a true optimization, it is simple, pragmatic, and works very well. For this reason we maintain this choice for the remaining of the manuscript.

Turning to the second moments, we expect both the cavity field and the spin components to remain in their minimum uncertainty state. Excess noise is generated due to our inability to predict whether excitations reside in the cavity or in the spin ensemble. For a non-inverted ensemble there is no energy available to facilitate any unknown distribution of excitations (the energy represented by the input quantum field is weak and also insufficient in this respect), and hence we expect no excess noise generated in the storage and retrieval processes. For an inverted state we know that excess noise may be generated \[34\]: however, for the specific case discussed here with \( g_j = 0 \) during periods of inversion, there is no exchange of energy between the spin ensemble and the cavity field, and in turn our knowledge of the excitations is not degraded. In total, no excess noise is expected in the idealized protocol, and in particular, the storage and retrieval processes do not generate noise. We have confirmed this by numerical simulations.

V. SPIN-CAVITY DECOUPLING BY FREQUENCY DETUNING

The decoupling of the spin ensemble from the cavity field during parts of the quantum memory protocol, see Fig. 1(b), is necessary for the following reasons: (i) The primary echo occurring half-way through the protocol should be silenced \[17\], i.e. the energy represented by the spin excitation must not leak to the cavity. (ii) During periods of spin-inversion ensemble the spin-cavity coupling generates excess noise \[34\], and a proper decoupling prevents this excess noise from interfering with the particular spin-mode holding the stored quantum state. (iii) The standard Hahn-spin-echo scheme employed here is based on the idea that the individual free spin evolution, \( \sigma_-(t) = \sigma_-(0)e^{-i\Delta t} \), can be reversed by appropriate spin-inversion processes such that the total phase accumulated by each spin is independent on \( \Delta \).

We note that there are physical implementations, in which a direct, on-demand decoupling, \( g_j \to 0 \), is possible, e.g. in atomic lambda systems with the cavity field coupled to the spin ensemble by a Raman process \[36\]. However, in this manuscript the decoupling mechanism is based on detuning the cavity frequency from the spin-resonance frequency, and the present section describes quantitatively the implications of this detuning, \( \Delta_{cs} \), being finite. Physically, the spin-ensemble oscillator remains coupled to the cavity-field oscillator. Thus, in connection to point (ii) above, a small probability remains for exchanging energy between the two oscillators, and hence an increased noise in the spin and cavity variables. In connection to point (iii), the spin-ensemble oscillator may induce a small cavity field, which in turn presents a slight back action on the otherwise free spin evolution. This is equivalent to the ac Stark effect on electrical dipoles, and the effect is discussed mathematically in Appendix E.

In order to isolate and investigate the impact of the effective spin-cavity decoupling on the quantum memory protocol, we numerically simulate the following scenario: The initial coherent state is prepared directly in the spin-ensemble oscillator, \( \langle \hat{a}_c(0) \rangle = 0 \) and \( \langle \hat{b}(0) \rangle = b_0 \), i.e. the swapping procedures (parts 1 and 7 in Fig. 1(b)) discussed in Sec. 1V are absent. In addition, the spin-inversion processes (parts 3 and 5 in Fig. 1(b)) are ideal and infinitely fast. Hence, we essentially model a pure spin-refocusing process with \( T_2 \equiv T_6 \equiv T, T_4 = 2T \), and \( T_{\text{mem}} = 4T \) but with the cavity playing a “spectator role”. During parts 2 and 6 the detuning is \( \Delta_{cs} \), and during part 4 it is either of \( \Delta_{cs}' = \pm \Delta_{cs} \). For simplicity we keep \( \kappa \) constant in the entire period.

An example of numerical simulations for the above protocol with \( \Delta_{cs}' = \Delta_{cs} \) is shown in Fig. 4(a) for mean values of the spin and in Fig. 4(b) for the relative excess spin noise, \( \text{RESN} = \frac{1}{2N} \langle \delta S^x_{c} + \delta S^y_{c} \rangle - 1 \) (we remind that the coherent-state variance is \( \langle \delta S^2_{c} \rangle = \langle \delta S^2_{c} \rangle = N \)). With the protocol gain \( G \) and phase shift \( \theta \) defined by \( \langle S_-(T_{\text{mem}}) \rangle = \langle S_-(0) \rangle \cdot G e^{-i\theta} \), we see by the zoom-in of the inset in panel (a) that the gain is slightly below unity at \( t = T_{\text{mem}} \). In this example the spin decoherence rate is absent, \( \gamma_\perp = 0 \), and the fact that \( G < 1 \) shows that the spin-cavity decoupling is not completely ideal. It is possible to predict approximately the behavior of the mean values since the large spin-cavity detuning, \( \Delta_{cs} \gg g_\text{ens} \), allows adiabatic elimination of the cavity field from the
FIG. 4. All panels: $\gamma_\perp = 0$ and $g_{\text{ens}} = 2.5 \Gamma$. (a) The transverse-spin-component mean value versus time for the spin-refocusing protocol of Sec. V with $\kappa/w = 0.075$ and $\Delta_{\text{cs}}/w = 20$. Red circles: Numerical simulation. Black solid line: Adiabatic model of Eqs. (15)-(17). (b) For the same parameters as panel (a) the solid line shows the relative excess spin noise (RESN) versus time. The red dotted line denotes the steady-state value of Eq. (12). (c) $1 - G$ versus spin-cavity detuning $\Delta_{\text{cs}}$ for $\kappa/w$ taking the values of 0.075 (red circles), 0.75 (green crosses), and 7.5 (blue diamonds). Open symbols correspond to $\Delta_{\text{cs}}' = \Delta_{\text{cs}}$ while the closed symbols (circles only) are obtained with $\Delta_{\text{cs}}' = -\Delta_{\text{cs}}$. Solid lines show the prediction of Eq. (10) while the red dotted line corresponds to $1 - G = g^2_{\text{ens}}/\Delta^2_{\text{cs}}$. (d) With the same symbols as panel (c) the memory phase shift versus $\Delta_{\text{cs}}$. Solid lines correspond to Eq. (11). (e) The RESN at the mid point, $t = 2T$, versus $\Delta_{\text{cs}}$ with symbols of panel (c). The open and closed circles are superimposed. Solid lines are theoretical according to Eq. (12). (f) The relation between the RESN at the mid- and end-points of the protocol. All data points were obtained for $\tilde{C} < 0.06$ and shown with the symbols of panel (c).

and the gain and phase shift are expected to be:

$$G \approx e^{-\gamma_\perp T_{\text{mem}} \left(1 - \frac{g^2_{\text{ens}}(\Delta_{\text{cs}} + \Delta'_{\text{cs}})/w}{\kappa^2 + \Delta^2_{\text{cs}}}\right)^2},$$

$$\theta \approx -2\arctan\left[\frac{g^2_{\text{ens}}(\Delta_{\text{cs}} + \Delta'_{\text{cs}})/w}{\kappa^2 + \Delta^2_{\text{cs}}}\right].$$

In Fig. 4(c) the numerically determined gain (open symbols with $\Delta'_{\text{cs}} = \Delta_{\text{cs}}$) is compared to the above expression (solid lines), and the agreement is seen to be quite good (the symbols being slightly higher than the solid lines). We note that Eq. (10) predicts unity gain when $\Delta_{\text{cs}} = -\Delta_{\text{cs}}$, which is not confirmed by the numerical calculations (closed symbols in Fig. 4(c)). The difference between the open red symbols and the corresponding solid line matches quite well the residual gain imperfection represented by the closed symbols, which are seen to scale as $g^2_{\text{ens}}/\Delta^2_{\text{cs}}$ (dotted red line). We note that in steady state $\langle \hat{a}_c \rangle = \frac{i g_{\text{ens}} \langle b \rangle}{\kappa + \Delta_{\text{cs}}}$ such that the relative energy leakage to the cavity at $t = T_{\text{mem}}$, being of the order $g^2_{\text{ens}}/\Delta^2_{\text{cs}}$, is missing from the spin degree of freedom. This effect was not covered by the adiabatic theory of appendix D. We also examine the memory phases shift $\theta$ in Fig. 4(d), which is seen to agree very well with the prediction of Eq. (11).

Now, turning to the variance of the spin components, all individual spins are in the same state ($\langle \hat{\sigma}_x^{(j)} \rangle = 1$, $\langle \hat{\sigma}_y^{(j)} \rangle = 0$) after the first ideal inversion pulse at $t = T$ if we consider the initial quantum state as the vacuum state, $b_0 = 0$ (a weak non-zero value does not change this picture). The subsequent evolution can then in principle be computed analytically by an off-resonant version of the calculations in appendix D, which in steady state leads to the following expression for the RESN:

$$\text{RESN}(2T) = \frac{2\kappa \tilde{C}}{(\kappa + \Gamma)(1 - \tilde{C})},$$

where $\tilde{C} = C(1 + \Delta'^2_{\text{cs}})/(\kappa + \Gamma)^{-1}$ generalizes the cooperativity parameter $C = \frac{\tilde{C}}{2\kappa}$, derived for a resonant spin-cavity interaction $[34]$. The above expression is seen to agree very well with the numerically calculated mid-point $(t = 2T)$ excess noise as shown in Fig. 4(e). After the second inversion process at $t = 3T$ we cannot expect the new steady-state value of spin variances to be calculated from the simplified homogeneous case as in appendix D since at $t = 3T$ the state of individual spins is correlated to $\Delta_j$. However, due to the refocusing effect around $t = 3T$ the new steady-state spin variance during $3T < t < 4T$ must be affected by the spin variance recorded into the spin memory during $2T < t < 3T$. The RESN at $t = 4T$ depends in a complicated way on the RESN at $t = 2T$, but in the limit of efficient spin-cavity decoupling ($\tilde{C} \ll 1$) the numerical simulations show that to a good approximation the two spin variances fulfill: $\text{RESN}(4T) = \text{RESN}(2T)^{2\kappa T/\kappa^2}$, see Fig. 4(f).
To conclude this section, the effective spin-cavity decoupling by detuning leaves the spin ensemble to evolve almost freely provided that the cavity-induced phase shift from various periods of finite macroscopic spin polarization is balanced (e.g., $\Delta_{\text{cs}} > 0 \text{ around the peaks at } t = 0 \text{ and } t = 4T,$ and $\Delta_{\text{cs}} < 0 \text{ around the peak at } t = 2T$ in Fig. 4(a) as discussed above). In this spin-refocusing protocol, a small residual effect on the gain scales as $1 - \tilde{G} \approx \frac{\sigma_{\text{cs}}^2}{2\Delta_{\text{cs}}}$ and a small excess variance is present, originating from the inverted spin ensemble and scaling as $\text{RESN}(T_{\text{mem}}) = 2\sigma^2 - 1 \approx \frac{4\kappa g^2}{\Delta_{\text{cs}}} \text{ when } \tilde{C} \ll 1$ and $\Delta_{\text{cs}} \gg \kappa + \Gamma$.

VI. EXTERNALLY APPLIED INVERSION PULSES

We now turn to the investigation of how the inversion pulses affect the performance of the memory protocol. Referring to Fig. 4(b), we maintain for the storage pulses affect the performance of the memory protocol. (i) hyperbolic secant pulses $\text{(37)}$ for the cavity field, and (ii) rotations “by hand” of the entire spin ensemble. The different strategies have been shown schematically in Fig. 5(a). As a starting point we neglect spin dephasing ($\tau = \infty$).

The hyperbolic secant pulses are widely used and perform well for inhomogeneous distributions of light-matter couplings $\text{(38)}$. In this case the cavity field varies as: $a_c(t) = a_{c,\text{max}} \text{sech}(\beta_{\text{sec}}(t - t_{\text{sec}}))^{1+\mu}$, where $a_{c,\text{max}}$ is the maximum amplitude of the cavity field, $\beta_{\text{sec}}^{-1}$ is the characteristic duration of the pulse, $t_{\text{sec}}$ is the center time of the pulse, and $\mu$ determines the shape of the inversion frequency profile (the higher $\mu$, the closer the profile resembles a top-hat distribution of width $\mu\beta_{\text{sec}}$). The maximum cavity field gives rise to a maximum Rabi frequency $\chi_{\text{max}} = 2g_{a,\text{max}}$ (with $g$-being the coupling parameter from the interaction Hamiltonian $\text{(3)}$ assumed to be equal for all spins), and when $\chi_{\text{max}} \geq \mu\beta_{\text{sec}}$ the inversion is known to work well. We will vary the Rabi frequency across this threshold value and hence deduce the effect of insufficient driving. In addition, various inversion bandwidths $\mu\beta_{\text{sec}}$ will be examined. We note that the externally applied field $\beta$ must be tailored to account for cavity filtering and for the reaction field of the spin dipoles $\text{(31)}$. The external driving is truncated to a finite duration of $\approx 16/\beta_{\text{sec}}$, during which the cavity is tuned to low-$Q$ mode ($\kappa = 7.5\omega$). The inversion bandwidths examined are $3\omega$, $6\omega$, and $9\omega$, for which the resulting average gain $\tilde{G}$ and relative excess variance $2\sigma^2 - 1$ have been plotted in Fig. 5(b,c). The varying strength of the external driving $\beta$ leads to varying probabilities $\rho_{\text{exc}}^\text{end}$ for a spin to be excited at the end of the protocol, which is conveniently used as horizontal axis in these figures. Evidently, for insufficient driving (leading to large excitation probabilities $\rho_{\text{exc}}^\text{end}$, the protocol performs poorly both in terms of gain and variance. Conversely, when the external driving is sufficient, both $\tilde{G}$ and $2\sigma^2$ become quite close to their ideal values of unity—the asymptotic values symbolized by the horizontal dashed lines will be discussed shortly. For the case of $\mu\beta_{\text{sec}} = 3\omega$ (red circles in Fig. 5) this asymptotic value is significantly higher than in the remaining examples, which we attribute to an insufficient frequency-bandwidth of the inversion pulse (i.e. a non-negligible fraction of spins are poorly inverted despite a large driving strength).
Now, to examine whether the above results are general or unique to the hyperbolic secant pulses, we employ a series of inversion pulses where the entire spin ensemble is simply rotated abruptly by an angle equal or close to the ideal value of \( \pi \). The cavity is maintained in low-\( Q \) mode for a duration \( \approx 12 / w \) around the abrupt spin rotation. We examine the cases where the first, second, and both inversion pulses are non-perfect. In all three cases, a non-ideal rotation angle \( \neq \pi \) leads to a finite excitation probability \( p_{exc}^{\text{end}} \) at the end of the protocol. The results have been added to Fig. 5 and it is indeed seen that all the simulation results fall approximately on the same curve. In other words, the final excitation probability \( p_{exc}^{\text{end}} \) defines the performance to a large extent. The horizontal dashed lines correspond to the case where both rotation angles have been set to \( \pi \), leading essentially to \( p_{exc}^{\text{end}} = 0 \). The reason that \( G < 1 \) and \( 2 \sigma^2 > 1 \) despite the ideal \( \pi \)-pulse inversions was discussed in Sec. V.

A. The effect of dephasing during inversion pulses

Our next step is to choose a finite dephasing time \( \tau < \infty \) for the spin coherence. According to the discussions in Sec. [V] we would naively expect the gain to scale roughly as \( G \propto e^{-\gamma(T_{\text{mem}} - T_{\text{swap}})} \), but in the following we shall see that the inversion pulses impose some corrections to this picture. The impact on the noise variance will also be examined. As a starting point, the inversion strategy of hyperbolic secant pulses (the first three strategies in Fig. 5(a)) are compared for various values of \( \gamma \) and \( T_{\text{mem}} \) using a sufficient external driving \( \chi_{max}^{\text{ext}} = \mu \beta_{\text{sech}} \). The performance of the gain parameter \( G \) is shown in Fig. 6(a), and the simulation results can be fitted to the function \( G = G_0 e^{-\gamma(T_{\text{mem}} - T_0)} \); however, with a fitting parameter \( T_0 \), which does not match the naive guess of \( T_{\text{swap}} \) but instead varies with \( \beta_{\text{sech}} \) as illustrated in Fig. 6(b): The smaller the \( \beta_{\text{sech}} \), the longer the inversion process, and in turn the longer the fitted \( T_0 \). The black triangles in Fig. 6(b) corresponds to an infinitely fast hyperbolic secant pulse and is calculated using two ideal \( \pi \)-rotations as outlined by the sixth strategy in Fig. 5(a). This relation between \( T_0 \) and \( \beta_{\text{sech}} \) is also shown with black circles in Fig. 5(c), and we confirm the naive guess \( T_0 \approx T_{\text{swap}} \) for very fast inversion pulses only. Physically, the stored information resides partly as population degrees of freedom during the inversion process, which in turn has a shielding effect from the dephasing processes: The longer the duration of the inversion pulses, the shorter becomes the effective time \( T_{\text{mem}} - T_0 \) of dephasing—the durations used in the simulations have been illustrated in Fig. 5(f).

From the above discussion one may think that longer inversion pulses may be slightly advantageous since the dephasing is partly turned off. However, this effect comes at a price when considering the noise properties of the memory protocol. While the simulations show that the noise variance does not depend on \( T_{\text{mem}} \) (additional waiting time does not contribute additional noise), it does depend on both \( \gamma \) and \( \beta_{\text{sech}} \) as shown in Fig. 6(c). In general, dephasing processes may counteract noise generation [it dampens both mean values and second moments in the equations of motion], which is indeed seen by the black triangles in Fig. 6(c) for infinitely fast inversion pulses. However, turning to hyperbolic secant pulses there is an additional noise generation, which grows with increasing \( \gamma \). This noise generation is more pronounced when the characteristic duration \( \beta_{\text{sech}}^{-1} \) of the inversion pulses is longer. In other words, while the damping of the gain parameter \( G \) is reduced during the inversion pulse, there is an accompanying noise generation at the same time. We note that increasing \( \gamma \) will de-

![Fig. 6](image-url)
crease the quality of the inversion process in terms of population—the hyperbolic secant pulses simply perform worse in presence of dephasing. The effect of imperfect inversion was investigated in Fig. 3(c), showing approximately a monotonous connection between $P_{\text{exc}}$ and $2\sigma^2 = 1$. However, this effect cannot alone explain the increased noise as is evident from Fig. 5(d); although $P_{\text{exc}}$ increases slightly when $\gamma_\perp$ grows, the increase in the relative excess noise variance $2\sigma^2 - 1$ (symbols) is markedly larger than the population effect corresponding to the guides-to-the-eye from Fig. 5(c), which are reproduced in Fig. 6(d). Hence, the increase in noise caused by dephasing must arise during the inversion process. We observe from Fig. 6(e), the relative gain of these data is comparable to the relative noise increase [equal to $\frac{1}{2\sigma^2} \frac{\partial^2 (2\sigma^2)}{\partial (\gamma_\perp/w)^2} = \theta T_0$ shown by black circles] is comparable to the relative noise increase [equal to $\frac{1}{2\sigma^2} \frac{\partial^2 (2\sigma^2)}{\partial (\gamma_\perp/w)^2} = \theta T_0$ shown by red squares].

B. Asymmetric input-output relations

As our final discussion of inversion pulses, we consider the actual input-output map of the memory protocol. This map was introduced in simple terms in Eq. (9), but it turns out that a generalization is required when inversion pulses are of insufficient strength. Consider first Fig. 7(a) which shows the mean value and standard deviation of $\hat{X}_c$ and $\hat{P}_c$ for both input and output states in a simulation series using hyperbolic secant pulses of sufficient strength and bandwidth. Each gray circle denote by its center the mean value of $\hat{X}_c$ and $\hat{P}_c$, and its radius corresponds to the standard deviations $\delta(\hat{X}_c^2)^{1/2} = \delta(\hat{P}_c^2)^{1/2}$. The black circles denote the same for the output state, and apart from a tiny phase rotation $\theta$ (caused by the cavity-induced phase shifts being not completely compensated) the gray and black circles are essentially equal, i.e. $G \approx 1$ and no extra noise is added. In contrast, the example given in Fig. 7(b) shows a more complicated input-output map, which mathematically follows the parametrization of Eq. (11). In addition to an overall phase shift induced by the spin-cavity coupling, this generalized map allows for describing the situation in which the gain parameter is not symmetric in $\hat{X}_c$, $\hat{P}_c$-space but instead depends on the phase of the input state. We note that the inversion pulses are encoded with a specific phase, which breaks the symmetry. The input-output map is decomposed into two main axes; a major axis of gain $G_1$ and variance $\sigma_1^2$ and a minor axis of gain $G_2$ and variance $\sigma_2^2$. The asymmetric transformation of mean values is seen by the elliptic shape of the black dashed envelope curve in Fig. 7(b) whereas the asymmetric output variances $\sigma_1^2 \neq \sigma_2^2$ is shown by the black circles actually being oval shaped.

The asymmetric gain and variances of the input-output map are shown as a function of $p_{\text{exc}}$ in Figs. 7(c,d). The different symbols correspond to those defined in Fig. 5(a), and the average gain $G$ and variance $\sigma^2$ of these data points were shown previously in Fig. 5(b,c) with the solid blue curves also reproduced. We observe that for inversion processes of high quality [leading to a small $p_{\text{exc}}$] the asymmetry disappears. Also, if either of the two inversion pulses is perfect (i.e. following the fourth or fifth strategy in Fig. 5(a)), the asymmetry also disappears. In Fig. 7(c,d) this materializes as the cyan tip-up triangles and the magenta hexagons falling on the solid blue line, which represents the behavior of the average values $G$ and $\sigma^2$.

The typical relationship between gain and variance is shown in Fig. 7(e). Apart from the “symmetric cases” of cyan tip-up triangles and magenta hexagrams, the data points seem to follow a general trend. A very low gain parameter and a gain parameter above unity lead to excess noise. For $G \lesssim 1$ there is a regime where one quadra-
ture is squeezed and the output state has a minor-axis variance of $\sigma_2^2 < \frac{1}{2}$.

Knowing the gain and variance parameters of a linear input-output relation for $\hat{X}_c$ and $\hat{P}_c$, it is possible to calculate the fidelity $F_q$ for a qubit encoded into the $|0\rangle$, $|1\rangle$ Fock states of the cavity field [32]. For the asymmetric case discussed here, $F_q$ is given by Eq. (12), and the infidelity $1 - F_q$ resulting from all the simulations discussed above is shown in Fig. 7(f). In addition, the solid blue curve is based on the guide-to-the-eye from Fig. 5(b,c) using a symmetric formula for $F_q$ (i.e. with $\mathcal{G}_1 = \mathcal{G}_2 \equiv \mathcal{G}$ and $\sigma_1^2 = \sigma_2^2 \equiv \sigma^2$). While Eq. (12) presents corrections to such a symmetrized formula the average parameters $\mathcal{G}$ and $\sigma^2$ govern the fidelity of the memory protocol quite accurately. The asymmetry does not in itself present a serious problem for the memory protocol apart from the fact that the asymmetry only arises when the inversion pulses are insufficient in strength.

VII. DISCUSSION

We have investigated the fundamental limitations of a quantum memory protocol, which uses a spin ensemble for storage and retrieval of a cavity-field quantum state. The quantum memory performance is fully characterized by the input-output relation of cavity-field mean values and variances, parametrized by the mean-value gain $\mathcal{G}$ and the output state variance $\sigma^2$. In particular, we have examined how the various parts of the protocol—the storage and read-out, the waiting times with the spin ensemble decoupled from the cavity, and the externally driven inversion pulses—affect the obtainable gain and variance, and the effect of decoherence mechanisms was also investigated.

We note that the results of the present manuscript, derived for a single mode of the cavity field, can be extended immediately to the multi-mode case, as was also shown in Ref. [30]. The only change in the multi-mode case is a longer $T_{\text{mem}}$ required to accommodate all the individual pulses.

Our analysis focused primarily on the impact of an inhomogeneous spin-frequency distribution. The inhomogeneity arises naturally from the ensemble nature of the spins and is essential for the multi-mode capability of the protocol. In some practical realizations, e.g. planar wave guide resonators coupled to nitrogen-vacancy centers in diamond [27], there is also an inhomogeneous distribution of coupling strengths $g$ for the spin-cavity interaction. Such an inhomogeneity will in general not affect our conclusions related to the linear regime of the spin-cavity interaction. However, when inversion pulses are applied, there may be spin classes which experience insufficient driving while others are subjected to a nearly perfect inversion process. Such scenarios can also be examined numerically by our formalism, and we refer to Ref. [30] for a specific example with inhomogeneous coupling strengths.

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Appendix A: Special properties of the Lorentzian inhomogeneous spin-frequency distribution

Under the Holstein-Primakoff approximation, and under the assumption that the initial state of each spin is uncorrelated to its resonance frequency, the behavior of the Lorentzian broadened spin ensemble corresponds exactly to that of homogeneous broadening. To show this, assume in the following $\hat{\sigma}^{(j)} \approx -1$ (the argument also holds for $\hat{\sigma}^{(j)} \approx 1$) and consider the Heisenberg-Langevin equations:

$$\frac{\partial \hat{a}_c}{\partial t} = - (\kappa + i \Delta_{cs}) \hat{a}_c - i \sum_{j=1}^{N} g_j \hat{\sigma}^{(j)} + \hat{f}_a, \quad (A1)$$

$$\frac{\partial \hat{\sigma}^{(j)}}{\partial t} = - (\gamma_\perp + i \Delta_j) \hat{\sigma}^{(j)} - ig_j \hat{a}_c + \hat{f}_j, \quad (A2)$$

where $\hat{f}_a$ and $\hat{f}_j$ are Langevin-noise operators accounting for the coupling to the environment (we assume that the environment coupling experienced by the $j$th spin does not depend on its resonance frequency $\Delta_j$). A formal integration of the spin operators leads to:

$$\hat{\sigma}^{(j)}(t) = \hat{\sigma}^{(j)}(0) e^{-(\gamma_\perp + i \Delta_j) t}$$

$$+ \int_0^t e^{-(\gamma_\perp + i \Delta_j)(t-t')} [-ig_j \hat{a}_c(t') + \hat{f}_j(t')] dt'. \quad (A3)$$

Next, our assumption that $\hat{\sigma}^{(j)}(0)$ is not correlated to $\Delta_j$ allows for integration over the inhomogeneous ensemble. In the harmonic-oscillator picture of Eq. (1) we find:

$$\hat{b}(t) = \hat{b}(0) e^{-(\gamma_\perp + \frac{\Delta}{2}) t}$$

$$+ \int_0^t e^{-(\gamma_\perp + \frac{\Delta}{2})(t-t')} [-ig_{\text{ens}} \hat{a}_c(t') + \sum_j \frac{g_j}{g_{\text{ens}}}] \hat{f}_j dt', \quad (A4)$$

which was derived using the residue theorem being convenient and applicable for the Lorentzian distribution (1). Taking the derivative of the above leads to the coupled equations (with $\hat{b} = \sum_j \frac{g_j}{g_{\text{ens}}} \hat{f}_j$):

$$\frac{\partial \hat{a}_c}{\partial t} = - (\kappa + i \Delta_{cs}) \hat{a}_c - ig_{\text{ens}} \hat{b} + \hat{f}_a, \quad (A5)$$

$$\frac{\partial \hat{b}}{\partial t} = - (\gamma_\perp + \frac{\Delta}{2}) \hat{b} - ig_{\text{ens}} \hat{a}_c + \hat{f}_b. \quad (A6)$$

These are identical to the equations for a homogeneously broadened sample ($\Delta_j = 0$ for all $j$) provided that we replace $\gamma_\perp \to \Gamma = \gamma_\perp + \frac{\Delta}{2}$. 

11
Appendix B: Gain, variance, and qubit fidelity

The definition of $\mathcal{G}$ and $\sigma^2$ in Sec. [III A] is generalized in the following. We note that phase shifts (e.g. induced by the off-resonant spin-cavity interaction) may occur in the input-output relations as marked by the angle $\theta$ in Fig. 7(a). In addition, as shown in Fig. 7(b), the gain and variance may depend on the phase of the input state since the inversion pulses may break the symmetry. We cover both of these scenarios by the input-output mean-value relation:

$$
\begin{bmatrix}
-X_c^{\text{out}} \\
-P_c^{\text{out}}
\end{bmatrix} = \mathbf{R}(\theta_1) \begin{bmatrix}
\mathcal{G}_1 & 0 \\
0 & \mathcal{G}_2
\end{bmatrix} \mathbf{R}(-\theta_0) \begin{bmatrix}
X_c^{\text{in}} \\
-P_c^{\text{in}}
\end{bmatrix},
$$

(1)

where $\mathbf{R}(\theta) = \begin{bmatrix}
\cos \theta & -\sin \theta \\
\sin \theta & \cos \theta
\end{bmatrix}$ accounts for a counter-clockwise rotation of angle $\theta$ in the $(X_c, P_c)$-coordinate system (corresponding to multiplying $a_c$ by $e^{i\theta}$). In the symmetric case, $\mathcal{G}_1 = \mathcal{G}_2 \equiv \mathcal{G}$, the transformation (1) reduces to a rotation by the angle $\theta = \theta_1 - \theta_0$, being the angle shown in Fig. 7(a). For the asymmetric case of Fig. 7(b) the angle $\theta = \theta_1 - \theta_0 = 2.3^\circ$ accounts for the fact that for input states located on the $X_c$- or $P_c$-axis, the output states are rotated slightly from the main axes shown as solid lines in Fig. 7(b). The angle between the horizontal $X_c$-axis and the major axis in this figure is $\theta_1 = 84.2^\circ$.

The variance of the output state may depend on the quadrature phase. Defining $\hat{X}_c(\theta) = X_c \cos \theta + P_c \sin \theta$, we must have $\langle \delta \hat{X}_c(\theta)^2 \rangle = \langle \delta X_c^2 \rangle \cos^2 \theta + \langle \delta P_c^2 \rangle \sin^2 \theta + \langle \delta X_c \hat{P}_c + \hat{P}_c \hat{X}_c \rangle \sin \theta \cos \theta \equiv \langle \delta X_c(\theta)^2 \rangle \frac{\sin 2\theta}{2}$, and $\langle \delta X_c(\theta)^2 \rangle^{1/2}$ corresponds to the angle-dependent radius of the black, oval shapes in Fig. 7(b). In all our simulations the main axes of these oval shapes coincide with the main axes of the mean value transformation (solid lines in Fig. 7(b)), and we define the variance $\sigma_1^2$ and $\sigma_2^2$ for these main axes. We will generally define $\mathcal{G}$ and $\sigma^2$ as the average values, $\mathcal{G} \equiv \frac{1}{2}(\mathcal{G}_1 + \mathcal{G}_2)$ and $\sigma^2 \equiv \frac{1}{2}(\sigma_1^2 + \sigma_2^2)$, as was done e.g. in Fig. 5(b,c).

For a qubit encoded into the |0⟩ and |1⟩ Fock states of the cavity field, the quantum memory fidelity is given by:

$$
F_q = \frac{1}{6} \left\{ \frac{3}{(\sigma_1^2 + \frac{1}{2})(\sigma_2^2 + \frac{1}{2})} + \frac{\mathcal{G}_1}{\sigma_1^2 + \frac{1}{2}} + \frac{\mathcal{G}_2}{\sigma_2^2 + \frac{1}{2}} \frac{G_1^2(\sigma_2^2 - 1) - G_2^2(\sigma_1^2 - 1)}{(\sigma_1^2 + \frac{1}{2})^2 - (\sigma_2^2 + \frac{1}{2})^2} \\
- \frac{G_1^2(\sigma_2^2 - 1) - G_2^2(\sigma_1^2 - 1)}{2(\sigma_1^2 + \frac{1}{2})(\sigma_2^2 + \frac{1}{2})} \right\}.
$$

(B2)

The derivation of this formula will be presented elsewhere [33].

Appendix C: The cavity-to-spin swapping procedure

The dynamical evolution of the spin- and cavity-field mean values during a resonant transfer of information is discussed mathematically in this appendix. For a Lorentzian inhomogeneous distribution we consider the mean values of Eqs. [A5] and [A6] on resonance ($\Delta cs = 0$), and given an initial cavity field $\langle \hat{a}_c(0) \rangle = \alpha$ coupled to the vacuum spin state $\langle \hat{b}(0) \rangle = 0$, the evolution becomes:

$$
\langle \hat{a}_c(t) \rangle = \alpha e^{-\frac{\Delta cs t}{2G_{\text{ens}}} \sin(g_{\text{ens}}t)} - \frac{\kappa - \Gamma}{2g_{\text{ens}}} \sin(g_{\text{ens}}t),
$$

(C1)

$$
\langle \hat{b}(t) \rangle = -i \alpha g_{\text{ens}} e^{-\frac{\Delta cs t}{2G_{\text{ens}}} \sin(g_{\text{ens}}t)}.
$$

(C2)

where $g_{\text{ens}} = g_{\text{ens}} \sqrt{1 - \frac{(\kappa - \Gamma)^2}{4g_{\text{ens}}^2}}$ is assumed real to obtain an oscillatory solution. The initial excitation residing in the cavity is transferred completely to the spin ensemble at the time $t = T_{\text{swap}}$ given by:

$$
T_{\text{swap}} = \pi \frac{2G_{\text{ens}}}{2g_{\text{ens}}} \left( 1 - \frac{2}{\pi} \arctan \left( \frac{\kappa - \Gamma}{2g_{\text{ens}}} \right) \right).
$$

(C3)

The evolution of the jth spin from its initial state $\langle \hat{a}_c^{(j)}(0) \rangle = 0$ is given by formal integration of the expectation value of Eq. (A2): 

$$
\langle \hat{a}_c^{(j)}(t) \rangle = -ig_j \int_0^t e^{-\gamma_\perp + i\Delta_j(t-t')} \langle \hat{a}_c^{(j)}(t') \rangle dt'.
$$

However, the complexity of Eq. (C1) prevents a simple analytical formula, and we shall consider limiting cases. First, in the limit $|\Delta_j/T_{\text{swap}}| \ll 1$ the rate of change in $\langle \hat{a}_c(t) \rangle$ is much slower than $\Delta_j$, and by partial integration we find (neglecting $\gamma_\perp$ and using $\frac{\partial \langle \hat{a}_c \rangle}{\partial \Delta_j} \ll |\Delta_j|$):

$$
\langle \hat{a}_c^{(j)}(T_{\text{swap}}) \rangle \approx g_j e^{-i\Delta_j/T_{\text{swap}}} \frac{\Delta_j}{\Delta_j},
$$

(C4)

yielding the phase relation of the dotted lines in Fig. 2(c). In the contrary case, $|\Delta_j/T_{\text{swap}}| \ll 1$, we make a crude approximation of $\langle \hat{a}_c(t) \rangle = \alpha(1 - e^{-\Delta_j t/T_{\text{swap}}})$ maintaining $\langle \hat{a}_c(0) \rangle = \alpha$ and $\langle \hat{a}_c(T_{\text{swap}}) \rangle = 0$, which leads to (neglecting $\gamma_\perp$):

$$
\langle \hat{a}_c^{(j)}(T_{\text{swap}}) \rangle \approx -ig_j \alpha T_{\text{swap}} \frac{2}{3} \left( 1 - i\Delta_j \frac{2T_{\text{swap}}}{3} \right),
$$

(C5)

translating into the phase $\phi_j \approx \frac{\pi}{2} + \frac{2}{3} T_{\text{swap}} \Delta_j$.

Appendix D: Requirement for numerical simulations

The formal equivalence between Lorentzian and homogeneous broadening gives rise to analytical expressions for the physical observables under specific circumstances. For instance, if the initial state at $t = 0$ is the perfectly inverted state, $\langle \hat{a}_c^{(j)} \rangle = 1$ and $\langle \hat{a}_c^{(j)} \rangle = \langle \hat{g}_x^{(j)} \rangle = 0$, with the cavity field in the coherent state of amplitude $\langle \hat{a}_c \rangle = \alpha$ on
mines the quantum-noise limit of the spin components, amount to \[34\]: pronounced for large values of \(\Delta\). The variance is over-estimated, most is approached for increasing \(\Delta\) increases with \(\lambda\) where \(2\Delta\) resonance with the spins, \(\Delta_{cs} = 0\), the evolution becomes [34]:

\[
a_c(t) = i (\lambda_+ + \Gamma)e^{\lambda_+ t} - (\lambda_- + \Gamma)e^{\lambda_- t}, \quad (D1)
\]

\[
S^{\text{eff}}(t) = i g N \alpha e^{\lambda_+ t} - e^{\lambda_- t}, \quad (D2)
\]

where \(2\lambda_0 = \lambda_+ + \lambda_-\) and identical equations hold for \(\langle \delta P_c^2 \rangle\) and \(\langle \delta S_z^2 \rangle\). The steady-state values at \(t = \infty\) amount to [34]:

\[
\langle \delta \hat{X}_c^2 \rangle = \left\langle \delta \hat{X}_c^2(\infty) \right\rangle = \frac{g_{\text{ens}}^2}{(\kappa - \Gamma)^2 + 4g_{\text{ens}}^2}, \quad \langle \delta \hat{S}_x^2 \rangle = \left\langle \delta \hat{S}_x^2(\infty) \right\rangle = \frac{2N \cdot g_{\text{ens}}^2}{(\kappa - \Gamma)^2 + 4g_{\text{ens}}^2}, \quad (D6)
\]

\[
\langle \delta \hat{S}_z^2 \rangle = \left\langle \delta \hat{S}_z^2(\infty) \right\rangle = N \cdot \frac{1 + C \kappa^2 + C}{1 - C}, \quad (D7)
\]

We note that these steady-state can be generalized to non-zero values of \(\Delta_{cs}\) by adding the term \([\Delta_{cs}/(\kappa + \Gamma)]^2\) in both the numerator and denominator. In turn, this leads to the result of Eq. [12].

The above analytical results present an important test base for numerical simulations. In this article we divide the shape function \(f(\Delta)\) into finite sub-ensembles each of frequency width \(d\Delta\) and subjected to a cut-off \(-\Delta_{cut} \leq \Delta \leq \Delta_{cut}\). In the following we estimate the numerical effects of the finite \(d\Delta\) and \(\Delta_{cut}\).

Numerical simulations of the scenario discussed around Eqs. [11] and [12] has been shown in Fig. [3]a) when \(\Delta_{cut}/\Gamma\) is varied between 20 and 100. Clearly, since the cut-off limits the frequency bandwidth, the mean-value decay of the transverse spin component \(S_\perp\) becomes slower. We also note that the relative error increases with \(C\), but in any case the simulations converge toward the analytical curve for increasing cut-off frequencies. For the same values of \(\Delta_{cut}\) the transverse-spin-component variance is shown in Fig. [3]b) showing a similar trend. The variance is over-estimated, most pronounced for large values of \(C\), but the correct value is approached for increasing \(\Delta_{cut}\). The above simulations were performed for \(\gamma_\perp = 0\), \(g_{\text{ens}} = 2.5\Gamma\), and number of spins \(N = 6.25 \times 10^{10}\). The latter determines the quantum-noise limit of the spin components, \(\langle S_x^2 \rangle = \langle S_y^2 \rangle = N\), such that the standard deviation becomes \(\sqrt{N} = 2.5 \times 10^5\). For numerical calculations to be faithful, one should ensure that spurious effects of the sub-ensemble discretization is well below this limit. In Fig. [3]c) it is exemplified how an increased \(\Delta_{cut}\) decreases the magnitude of such effects, and furthermore, that the finite frequency spacing \(d\Delta\) causes an artificial revival at \(t = 2\pi/d\Delta\). In practical spin-echo calculations it is required that \(d\Delta < 2\pi/T_{\text{mem}}\) to avoid these revivals.

### Appendix E: Phase shifts induced by a detuned cavity

This appendix calculates analytically the impact of a detuned cavity on the spin ensemble. The initial state is taken as a coherent spin state, a homogeneous distribution of coupling strengths is assumed, and the spin-cavity detuning is large, \(\Delta_{cs} \gg \kappa, g_{\text{ens}}\). The latter allows the cavity field to be adiabatically eliminated from the dynamical equations leading to the effective spin Hamiltonian [39]:

\[
\hat{H} = \frac{1}{2} \sum_{j=1}^{N} \left( \Delta_j \hat{\sigma}_z^{(j)} - \frac{g_{\text{ens}}^2 \Delta_{cs} \hat{S}_x \hat{S}_z}{\kappa^2 + \Delta_{cs}^2} \right), \quad (E1)
\]

and the cavity leakage is translated into a correlated spin decay with \(\hat{c} = \sqrt{\gamma_{\text{mem}}^2} \hat{S}_z\), \(\gamma_{\text{mem}} = \frac{2g_{\text{ens}}^2}{\kappa^2 + \Delta_{cs}^2}\), in the language of the Lindblad part of the master equation, \(\mathcal{L}[\hat{\rho}] = -\frac{1}{\kappa} \hat{\rho} \hat{c}^\dagger \hat{c} \hat{\rho} - \frac{1}{\kappa} \hat{c}^\dagger \hat{c} \hat{\rho} \hat{\rho} \hat{c}^\dagger \hat{c}\). In the Holstein-Primakoff approximation the spin-ensemble evolution is then governed by:

\[
\frac{\partial \langle \hat{\sigma}_z^{(j)} \rangle}{\partial t} = -[\gamma_\perp + i \Delta_j] \langle \hat{\sigma}_z^{(j)} \rangle - i \gamma_{\text{mem}} \hat{S}_z, \quad (E2)
\]
Inverted ensemble ($p = -1$) has the solution:

$$\langle \hat{S}_- (t) \rangle = \langle \hat{S}_- (0) \rangle e^{-t^{\frac{\kappa}{2}} e^{i\zeta t}}.$$  \hfill (E3)

In turn, by formal integration of Eq. (E2), the evolution of the individual spins can be deduced:

$$\langle \hat{\sigma}_- (t) \rangle = \langle \hat{\sigma}_- (0) \rangle e^{-\gamma_\perp + 2\Delta_j + t + i\zeta} e^{-i\Delta_j t} \left[ 1 - \frac{\zeta + i\Delta_j}{\Delta_j} \right].$$  \hfill (E4)

where it is assumed that the initial mean value, $\langle \hat{S}_- \rangle$, has vanished due to the frequency inhomogeneity, $t \gg \omega^{-1}$. The second term of the effective Hamiltonian (E2) is only in effect while $\langle \hat{S}_- \rangle$ is not negligible, i.e. during the spin-dephasing process, by which the second term in the square brackets above is acquired. We observe that in addition to the free evolution, $e^{-i\Delta_j t}$, the spins acquire a complex phase which varies non-linearly with $\Delta_j$ and hence complicates the spin-refocusing procedure.

In order to calculate the performance of such a refocusing procedure, let the spin state evolve until $t = T$, at which time a perfect and infinitely fast inversion process around the $x$-axis is performed. Mathematically, this corresponds to taking the conjugate of the complex spin variables of Eqs. (E3) and (E4) evaluated at the time $T$. At this time we also allow for changing the cavity parameters such that $\zeta$ attains a new value, $\zeta'$, for the subsequent evolution. Repeating the above procedure for the time range $T \leq t \leq 2T$ with $p = 1$ leads to the ensemble-spin component (assuming $T \gg \omega^{-1}$):

$$\langle \hat{S}_- (t) \rangle = \frac{\langle \hat{S}_- (0) \rangle e^{-\gamma_\perp t} e^{i(\frac{\pi}{2} + i\zeta')} (t + 2T)}{1 + \frac{i(\zeta + \zeta')}{\omega}}.$$  \hfill (E5)

For completeness, and in order to follow the two-π-pulse protocol of Sec. VII we proceed the evolution of the state in the time range $2T \leq t \leq 3T$ maintaining $\zeta'$:

$$\langle \hat{S}_- (t) \rangle = \frac{\langle \hat{S}_- (0) \rangle e^{-\gamma_\perp t} e^{-i(\frac{\pi}{2} + i\zeta')} (t + 2T)}{1 + \frac{i(\zeta + \zeta')}{\omega}}.$$  \hfill (E6)

and after a perfect inversion at $t = 3T$ we revert back to the original $\zeta'$ and calculate for $3T \leq t \leq 4T$:

$$\langle \hat{S}_- (t) \rangle = \frac{\langle \hat{S}_- (0) \rangle e^{-\gamma_\perp t} e^{-i(\frac{\pi}{2} + i\zeta')} (t + 4T)}{1 + \frac{i(\zeta + \zeta')}{\omega}}.$$  \hfill (E7)

Evaluated at $t = 4T$ this leads to the predictions of Eqs. (10) and (11).

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