Decoherence in collective quantum memories for photons

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The influence of decoherence on the fidelity of quantum memories for photonic qubits is discussed. It is shown that despite the large entanglement of the collective storage states corresponding to single photons or nonclassical states of light the sensitivity to decoherence does not scale with the number of atoms. This is due to the existence of equivalence classes of storage states corresponding to states with the same number of dark-state polariton excitations but arbitrary excitations in other polariton modes. Several decoherence processes are discussed in detail: single-atom spin-flips and dephasing, atom loss and motion of atoms.

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I. INTRODUCTION

One of the essential ingredients for quantum information processing with photons as information carrier is a reliable quantum memory capable of a faithful storage of the quantum state of photons. They play a key role in network quantum computing, in long-distance, secure quantum communication and quantum teleportation. The application to teleportation is of particular interest because of its potentials for quantum information processing with linear optical elements. While photons are one of the most easy to handle information carriers, atoms or similar systems like quantum dots are reliable and long lived storage units. Furthermore Raman transitions provide a controllable and decoherence insensitive way of coupling between light and atoms. The conceptually simplest and for processing purposes best suited storage system for photonic qubits are individual atoms. Here coherent transfer techniques have been developed that allow a controlled transfer of quantum information from light to the atom and vice versa. However cavity QED settings in the strong-coupling regime are required to achieve reasonable fidelities for the state transfer. On the other hand if atomic ensembles are used rather than individual atoms no such requirements exists and coherent and reversible transfer techniques for individual photon wavepackets and cw light fields have been proposed and in part experimentally implemented.

The substantially alleviated requirements for the light-matter interface in the case of atomic ensembles are due to the enhanced coupling between collective many-atom states and the radiation field. The corresponding collective excitations of the ensemble are highly entangled many-particle states if nonclassical states of light are stored. So while classical information encoded, e.g. in single-particle Raman coherences, can be rather robust against decoherence processes this is not a priori clear for quantum correlations stored in the ensemble. In fact one might naively expect that the livetime of quantum correlations decreases with the number of atoms involved in the storage state in which case the system would be practically useless as a quantum memory. We therefore analyze in the present paper the influence of various decoherence mechanisms on the fidelity of the quantum memory. We show that each quantum state of the radiation field stored in the atomic ensemble corresponds to a whole class of many-particle states. It is due to the existence of these equivalence classes, which represent all states with the same number of excitations in specific quasi-particle modes, the dark-state polariton, and arbitrary excitations in other modes, that the quantum memory does not show an enhanced sensitivity to decoherence as compared to single-particle storage units.

In order to simplify the discussion we will here restrict ourselves to a quantum memory for a single-mode radiation field, realized for example in a weak-coupling resonator. In doing so we do not need to take into account decoherence effects on the longitudinal profile of a stored pulse arizing from atomic motion, which are however important in free-space configurations. First we reexamine the adiabatic transfer scheme of for quantum states of photon wavepackets to atomic ensembles in terms of quasi-particles (dark- and bright polaritons) in Sect. II. We will show that only specific quasi-particle modes are relevant for the storage in the adiabatic limit. In Sec. III we will discuss the effect of different decoherence mechanisms, individual random spin flips, dephasing of Raman coherences, loss of atoms, atomic motion and imperfect preparation. It will be shown that the decoherence rate of the stored quantum state does not depend on the number of atoms in all of these cases. This is because excitations of any quasi-particle mode other than the relevant dark-polariton mode do not matter in the adiabatic limit. They do matter, however, if non-adiabatic couplings are taken into account. We will therefore discuss the effect of decoherence in the presence of non-adiabatic couplings in Sec. VI.
II. DARK- AND BRIGHT-POLARITONS, EQUIVALENCE CLASSES OF STORAGE STATES

Let us consider an ensemble of $N$ 3-level atoms with internal states $|a\rangle$, $|b\rangle$ and $|c\rangle$ resonantly coupled to a single quantized mode of a resonator field with mode function $e^{i k_0 z}$ and a classical control field of Rabi-frequency $\Omega$ and mode function $e^{i k_1 r}$ as shown in Fig. 1. The dynamics of this system is described by a non-hermitian Hamiltonian $(E_b = \hbar \omega_b = 0)$:

$$H = \hbar \omega a^\dagger a + \hbar (\omega_a - i \gamma) \sum_{j=1}^N \sigma_{aa}^j + \hbar \omega_c \sum_{j=1}^N \sigma_{cc}^j +$$

$$+ \h g \sum_{j=1}^N \sum_{a} \sum_{\nu} a_{a} e^{i k_0 z_j} c_{\nu} e^{i k_1 r_j} \sigma_{ac}^j + \h.c. .$$

Here $\sigma_{\mu\nu}^j = |\mu\rangle_j \langle \nu|\rangle$ is the flip operator of the $i$th atom and the vacuum Rabi-frequency is assumed to be equal for all atoms. For the time being we disregard atomic motion and thus the phase factors $e^{i k_0 z_j}$ as well as $e^{i k_1 r_j}$, will be absorbed into the definition of the atomic states $|a\rangle_j$ and $|c\rangle_j$. We will however come back to the issue of atomic motion in section III. We also have introduced an imaginary part to the Hamiltonian to take into account losses from the excited state, e.g. via spontaneous emission. The complex Hamiltonian emerges from a Lindblad Liouville operator that includes the decay from the excited state and therefore out of the relevant subspace ${|a\rangle, |b\rangle, |c\rangle}$. The model does not take into account relaxation from the excited state back into the lower levels. Spontaneous emission into the resonator mode accompanied by a transition from $|a\rangle$ to $|b\rangle$ is of course automatically included in the model.

$$|a\rangle \quad g \quad \Omega(t) \quad |c\rangle$$

FIG. 1: 3-level atoms coupled to single quantized resonator mode and classical control field of (real) Rabi-frequency $\Omega(t)$; $g$-vacuum Rabi-frequency; the dashed line indicates spontaneous decay.

When all atoms are initially prepared in level $|b\rangle$ the only states coupled by the interaction are the totally symmetric Dicke-states [27] (after absorption of the spatial phase factors into the definition of the states)

$$|b\rangle_N = |b_1, b_2 \ldots b_N\rangle,$$ (2)

$$|a^1\rangle_N = \frac{1}{\sqrt{N}} \sum_{j=1}^N |b_1 \ldots a_j \ldots b_N\rangle,$$ (3)

$$|c^1\rangle_N = \frac{1}{\sqrt{N}} \sum_{j=1}^N |b_1 \ldots c_j \ldots b_N\rangle,$$ (4)

$$|c^2\rangle_N = \left(\frac{N}{2}\right)^{-1/2} \sum_{i<j=1}^N |b_1 \ldots c_i \ldots c_j \ldots b_N\rangle,$$ (5)

etc...

The couplings within the sub-systems corresponding to a single and a double excitation are shown in Fig. 2. The set of collective states can be separated into groups with a specific excitation number $n$ and atom number $N$. Due to the symmetry of the interaction there is no coupling between classes with different excitation. Decay out of the excited state and therefore out of the relevant sub-system couples only classes with different atom number.

![FIG. 2: Coupling of bare eigenstates of atom plus cavity system for at most two photons.](image)

In the following we will restrict ourselves to two-photon resonance, i.e. $\omega = \omega_a - \omega_c = \nu$. Furthermore for simplicity single-photon resonance is assumed as well. This is sufficient since here we are not interested in the fidelity of the transfer process itself. The influence of a finite two-photon detuning on the transfer process is discussed in detail in [28]. In the case of two-photon resonance, the interaction of the $N$-atom system with the quantized radiation mode has a family of dark-states, i.e. adiabatic eigenstates with vanishing component of the excited states $|a\rangle_j$

$$|D, n\rangle_N = \sum_{k=0}^n \xi_{nk}(-\sin \theta)^k (\cos \theta)^{n-k}|c^k, n - k\rangle_N,$$

$$\xi_{nk} = \sqrt{\frac{n!}{k!(n-k)!}}, \quad \tan \theta(t) = \frac{g \sqrt{N}}{\Omega(t)}.$$

It should be noted that although the dark states $|D, n\rangle_N$ are degenerate, there is no transition between them even if non-adiabatic corrections are taken into account due to the symmetry of the interaction Hamiltonian. Adiabatically rotating the mixing angle $\theta$ from 0 to $\pi/2$ leads to
a complete and reversible transfer of all photonic states to a collective atomic excitation if the maximum number of photons \( n \) is less than the number of atoms \( N \). If the initial quantum state of the single-mode light field is described by the density matrix \( \rho_f = \sum_\alpha \rho_{\alpha nm} |\alpha\rangle \langle \alpha| \), the transfer process generates a quantum state of collective excitations according to

\[
\sum_{n,m} \rho_{nm} |n\rangle \langle m| \otimes |b\rangle_N N \langle b| \downarrow \\
\sum_{n,m} \rho_{nm} |D,n\rangle_N N \langle D,m| \\
|0\rangle \langle 0| \otimes \sum_{n,m} \rho_{nm} |e^n\rangle_N N \langle e^m| .
\]

The dark states of the \( N \)-atom system can be identified as quasi-particle excitations of the so-called dark-state polaritons \( \Psi \) in the space of atoms and cavity mode [14]

\[
|D,n\rangle_N = \frac{1}{\sqrt{n!}} (\Psi^n |b,0\rangle_N ,
\]

where \( |b\rangle \) is the total ground state of the \( N \) atom system and \( |0\rangle \) the vacuum state of the cavity mode. The dark-state polariton defined as

\[
\Psi = \cos \theta(t) a - \sin \theta(t) \frac{1}{\sqrt{N}} \sum_{j=1}^N \sigma_j^b,
\]

is a superposition of the resonator mode and the collective spin corresponding to the ground-state transition \( |b\rangle \leftrightarrow |c\rangle \). Associated with the dark polariton is a bright polariton

\[
\Phi_0 = \sin \theta(t) a + \cos \theta(t) \frac{1}{\sqrt{N}} \sum_{j=1}^N \sigma_j^c.
\]

To obtain a complete set of operators in the space of the cavity mode and the \( N \) atoms in internal states \( |b\rangle \) and \( |c\rangle \) we also need to introduce the operators

\[
\Phi_l = \frac{1}{\sqrt{N}} \sum_{j=1}^N \sigma_j^{bc} \exp \left\{ 2\pi i \frac{l_j}{N} \right\} , \quad l = 1 \ldots N - 1,
\]

together with the hermitian adjoints \( \Psi^\dagger \) and \( \Phi_l^\dagger \). We will also refer to the \( \Phi_l^\dagger \)'s as bright polaritons. In the limit of small atomic excitations the polariton operators obey approximately bosonic commutation relations

\[
[\Psi, \Psi^\dagger] = \cos^2 \theta + \sin^2 \theta \frac{1}{N} \sum_{j=1}^N (\sigma_j^b - \sigma_j^c) = 1 + \mathcal{O}\left( \frac{n_c}{N} \right),
\]

\[
[\Phi_0, \Phi_0^\dagger] = \sin^2 \theta + \cos^2 \theta \frac{1}{N} \sum_{j=1}^N (\sigma_j^b - \sigma_j^c) = 1 + \mathcal{O}\left( \frac{n_c}{N} \right),
\]

where \( n_c = \sum_j \sigma_j^c \ll N \) is the total population in level \( |c\rangle \). Polariton operators of different type commute in lowest order of \( n_c/N \):

\[
[\Phi_l, \Phi_m^\dagger] = \delta_{lm} \frac{1}{N} \sum_{j=1}^N (\sigma_j^b - \sigma_j^c) = \delta_{lm} + \mathcal{O}\left( \frac{n_c}{N} \right),
\]

It should be noted that the dark and bright polariton operators are explicitly time dependent through the mixing angle \( \theta(t) \).

The collective storage state corresponding to a coherent state of light factorizes as can be seen quite easily

\[
e^{\alpha/2} \langle b,0\rangle_N = e^{\alpha a^\dagger} |b,0\rangle_N
\]

\[
\exp \left\{ \frac{\alpha}{\sqrt{N}} \sum j \sigma_j^b \right\} |b,0\rangle_N = \prod_j \left( 1 - \frac{\alpha \sigma_j^b}{\sqrt{N}} \right) |b,0\rangle_N.
\]

On the other hand, storage states corresponding to non-classical states of light such as Fock states are maximally entangled \( N \) particle states \( |e^n\rangle_N \) as can be seen from eq. (11). These states are known to be rather sensitive to decoherence processes. For example if for an initial state \( |e^n\rangle_N \) the atom number one undergoes a transition from level \( |b\rangle \) to an auxiliary state, say \( |d\rangle \), the resulting state is almost orthogonal to the original one

\[
|c_1, b_2 \ldots b_N \rangle + |b_1, c_2 \ldots b_N \rangle + \cdots |b_1, b_2 \ldots c_N \rangle
\]

\[
|c_1, b_2 \ldots b_N \rangle + |d_1, c_2 \ldots b_N \rangle + \cdots |d_1, b_2 \ldots c_N \rangle .
\]

If \( p \) denotes the probability of one atom to undergo a transition from \( |b\rangle \) to \( |d\rangle \) due to environmental interactions, the total probability \( P_{\text{error}} \) to end up in an orthogonal state scales as \( P_{\text{error}} \sim 1 - (1 - p)^N \sim pN \). Thus one might naively expect that for the storage of a single photon the collective quantum memory will have an \( N \) times enhanced sensitivity to decoherence as compared to a single-atom device. We will now show that this conclusion is generally not correct.

From the inverse relation

\[
a = \cos \theta(t) \Psi + \sin \theta(t) \Phi_0
\]

one recognizes that for the resonator mode only excitations of the dark polariton \( \Psi \) and the bright polariton \( \Phi_0 \) matter. Furthermore if after the storage of photon states in the atomic system the electromagnetic excitations are regenerated by rotating \( \theta \) back from \( \pi/2 \) to 0, only excitations in the dark polariton mode are relevant.
I.e. if $W$ denotes the total density operator of the combined atom-cavity system after the writing process, only the reduced density operator

$$\rho = \text{Tr}_\Phi \{ W \} \quad (19)$$

is relevant for the storage. Here $\text{Tr}_\Phi$ denotes the partial trace over all bright-polariton excitations. For this reason all states of the total system that have the same number of dark-polariton excitations but an arbitrary number of excitations in any bright-polariton mode are equivalent from the point of view of storage. I.e. there exist equivalence classes of storage states of the form

$$|D, n\rangle_N \equiv \left\{ \left( \Phi_1^\dagger \right)^k \left( \Phi_2^\dagger \right)^l \ldots \left( \Phi_l^\dagger \right)^n |b, 0\rangle_N \right\}. \quad (20)$$

It is important to note that any perturbation that acts only onto bright polariton modes does also not destroy superpositions of storage states, since $[\Phi_l, \Psi] = 0$:

$$\Phi_i^\dagger \sum_n \alpha_n |D, n\rangle_N = \Phi_i^\dagger \sum_n \frac{\alpha_n}{\sqrt{n!}} \left( \Psi^\dagger \right)^n |b, 0\rangle_N$$

$$= \sum_n \frac{\alpha_n}{\sqrt{n!}} \left( \Psi^\dagger \right)^n \left\{ \Phi_i^\dagger |b, 0\rangle_N \right\}. \quad (21)$$

Likewise all dark states with the same number of excitations $n$ but with different number of atoms $N \gg n$ are equivalent because in the adiabatic read-out process all dark-polariton operators corresponding to different $N$ have the same asymptotic mapping $\Psi \rightarrow a$ for $\theta \rightarrow 0$. This will be important later on when discussing the effect of atom losses from the system

$$|D, n\rangle_N \equiv |D, n\rangle_{N'}, \quad \text{if} \quad N, N' \gg n. \quad (22)$$

The importance of the equivalence classes stems from the fact that unwanted interactions with the environment which lead only to transitions within the equivalence classes and that do not destroy the relative phase between them do not affect the fidelity of the quantum memory.

We can now express the complex Hamiltonian $\hat{H}$ in terms of bright and dark polariton operators after adiabatically eliminating the excited state $|a\rangle$. Separating the oscillatory factor $e^{-i\omega t}$ by a canonical transformation and assuming two photon resonance, i.e. $\omega = \omega_c + \nu$ we arrive at

$$H = \hbar \omega \left( \Psi^\dagger \Psi + \sum_{l=0}^{N-1} \Phi_l^\dagger \Phi_l \right) - i \hbar \frac{\Omega^2(t)}{\gamma} \sum_{l=1}^{N-1} \Phi_l^\dagger \Phi_l \quad (23)$$

$$= \hbar \omega \left( \Psi^\dagger \Psi + \sum_{l=0}^{N-1} \Phi_l^\dagger \Phi_l \right) - i \hbar g^2 N \csc^2 \theta(t) \sum_{l=1}^{N-1} \Phi_l^\dagger \Phi_l. \quad (24)$$

We here see two important points. First of all the adiabatic dynamics does not couple different polariton modes. Secondly all bright polariton excitations $\Phi_l$, $l = 1, 2, \ldots, N - 1$ decay by optical pumping, i.e. by excitation to the excited state and successive spontaneous emission if $\theta \neq \pi/2$, while the dark polaritons $\Psi$ as well as the bright polaritons $\Phi_0$ are immune to spontaneous emission. Since the non-hermitian Hamiltonian is expressed in terms of explicitly time dependent variables it describes the dynamics only in the adiabatic limit $[17]

$$g \sqrt{N} T \gg 1, \quad (24)$$

where $T$ is a characteristic time of changes. If non-adiabatic corrections are taken into account, dark and bright polariton modes are coupled with a rate proportional to $\theta$.

### III. INFLUENCE OF DECOHERENCE ON STORAGE FIDELITY

#### A. Imperfect preparation

In the description of the storage process given in the last section we had assumed that every atom in the ensemble was prepared in the ground state $|b\rangle$. If the ensemble is large enough the probabibility to find an atom e.g. in state $|c\rangle$ will however be non-negligible, e.g. due to the interaction with a finite temperature reservoir. Naively one might expect that any atom left in state $|c\rangle$ after preparation of the ensemble would mimic a stored photon. This would require to make the initial probability to find an atom in state $|c\rangle$ small compared to unity, which is however not the case. It is rather sufficient that the initial probability of excitation of a dark-state polariton is small compared to unity. If we consider e.g. as initial state a thermal state of temperature $\beta = 1/k_BT$ ($\theta = \pi/2$)

$$\rho_0 = \frac{1}{Z} \exp \left\{ -\beta \hbar \omega_c \left( \hat{\Psi}^\dagger \hat{\Psi} + \sum_{l=1}^{N-1} \Phi_l^\dagger \Phi_l \right) \right\} \quad (25)$$

with $Z$ being the statistical sum, the mean number of initially excited dark-state polaritons is independent of $N$ and given by

$$\langle \hat{\Psi}^\dagger \hat{\Psi} \rangle = \frac{e^{-\beta \hbar \omega_c}}{1 - e^{-\beta \hbar \omega_c}} = \frac{1}{N} \sum_{i=1}^{N} \langle \sigma^c_{i} \rangle \quad (26)$$

Thus if the probability that an atom is initially in level $|c\rangle$ is small compared to unity, the number of initial dark-polariton excitations is small compared to unity as well.

#### B. Random spin flips and dephasing

On the level on individual atoms the storage occurs within the two-state system consisting of $|b\rangle$ and $|c\rangle$. If we assume that all other atomic states including $|a\rangle$ are energetically much higher, we may safely neglect decoherence processes involving the excitation of those states.
Then decoherence caused by individual and independent reservoir interactions can be described by the action of the two-level Pauli operators

\[ X_j = \sigma_{bc}^i + \sigma_{cb}^i, \quad Z_j = \left[ \sigma_{bc}^j, \sigma_{cb}^j \right], \quad Y_j = i\sigma_{bc}^j - i\sigma_{cb}^j. \]  

\( X_j \) describes a symmetric spin flip of the jth atom, \( Z_j \) a phase flip, and \( Y_j \) a combination of both. Any single-atom error can be expressed in terms of these and we will restrict the discussion here to the action of \( X_j \) (symmetric spin flip), \( X_j + iY_j \) (asymmetric spin flip) and \( Z_j \) (phase flip).

Inverting relations (27) one easily finds a representation of \( \sigma_{bc}^j \) in terms of polaritons

\[ \sigma_{bc}^j = \frac{1}{\sqrt{N}} \left( \sum_{l=1}^{N-1} \exp \left\{ -2\pi i \frac{l j}{N} \right\} \Phi_l - \Psi^\dagger \right) \]

\[ = \frac{1}{\sqrt{N}} \left( \sum_{l=1}^{N-1} \eta_{jl} \Phi_l^\dagger - \Psi^\dagger \right) = \frac{X_j + iY_j}{2}. \]  

(28)

Here and in the following \( \theta = \pi/2 \) is assumed, unless stated otherwise, which corresponds to the case of a completed transfer from the radiation field to the ensemble. Furthermore

\[ X_j = \frac{1}{\sqrt{N}} \left[ \sum_{l=1}^{N-1} \left( \eta_{jl} \Phi_l^\dagger + \eta_{jl} \Phi_l \right) - \Psi - \Psi^\dagger \right]. \]  

(29)

and

\[ Z_j = \frac{1}{N} \left[ \sum_{l=1}^{N-1} \eta_{jl} \Phi_l - \sum_{m=1}^{N-1} \eta_{jm} \Phi_m^\dagger - \Psi^\dagger \right]. \]  

(30)

One recognizes at this point that applying the approximate commutation relations \( [12] \), which have been obtained with the assumption \( \sigma_{bc}^i \approx 1 \) and \( \sigma_{cc}^j \approx 0 \), would here lead to \( Z_j = Y_j \). Thus care must be taken when using \( Z_j \).

1. **Spin flip from \( |b\rangle \rightarrow |c\rangle \)**

Consider a quantum memory initially in an ideal storage state \( W_0 \), i.e. without bright polariton excitations. Suppose an atom then undergoes a spin flip to the internal state \( |c\rangle \) if it is initially in state \( |b\rangle \). Such a spin flip process, which could mimic a stored photon, can be described by the positive map

\[ W_0 \rightarrow W_1 = \frac{\sigma_{bc}^i W_0 \sigma_{cb}^i}{\text{Tr}[\sigma_{bc}^j W_0 \sigma_{cb}^j]}. \]  

(31)

As noted in the previous section only the reduced density operator traced over the bright-polariton modes is of relevance for the storage process. Carrying out this trace yields

\[ \text{Tr}_\Phi \left( \sigma_{bc}^j W_0 \sigma_{cb}^j \right) = \frac{1}{N} \text{Tr}_\Phi \left[ \sum_{l,m=1}^{N-1} \eta_{jl} \Phi_l^\dagger W_0 \eta_{jm} \Phi_m \right] \]  

\[ + \frac{1}{N} \Psi^\dagger \rho_0 \Psi - \frac{1}{N} \text{Tr}_\Phi \left[ \sum_{l=1}^{N-1} \eta_{jl} \Phi_l^\dagger W_0 \Psi + h.a. \right]. \]  

(32)

where \( \rho_0 = \text{Tr}_\Phi \{ W_0 \} \). If we make use of the fact that the bright and dark polaritons commute in first order of \( 1/N \), we see that the last term in eq. (32) vanishes since there are no excitations of bright polaritons in the initial state \( W_0 \). For the same reason

\[ \text{Tr}_\Phi \{ \eta_{jl} \Phi_l^\dagger W_0 \eta_{jm}^\dagger \Phi_m \} = \rho_0 \delta_{lm}, \]  

(33)

and the first term in eq. (32) evaluates to \((1-1/N)\rho_0\). Thus we arrive at

\[ \text{Tr}_\Phi \left( \sigma_{bc}^j W_0 \sigma_{cb}^j \right) = \left( 1 - \frac{1}{N} \right) \rho_0 + \frac{1}{N} \Psi^\dagger \rho_0 \Psi, \]  

and

\[ \rho_1 = \text{Tr}_\Phi \{ W_1 \} = \left( 1 - \frac{1}{N} \right) \rho_0 + \frac{1}{N} \Psi^\dagger \rho_0 \Psi. \]  

(34)

One recognizes that the spin flip of an individual atom only causes an error of order \( 1/N \). This exactly compensates for the fact that the total spin-flip probability of the \( N \) atoms is \( N \) times the probability of a single atom.

From eq. (34) one can easily calculate the fidelity of the quantum memory after a single spin flip error, which for the case of an initial pure state \( \rho_0 = |\psi_0\rangle \langle \psi_0| \) is defined as

\[ f \left( |\psi_0\rangle \right) = \langle \psi_0 | \rho_1 | \psi_0 \rangle = \text{Tr} \{ \rho_1 \rho_0 \}. \]  

(35)

One finds e.g. for a stored Fock-state \( |n\rangle \) with \( n \ll N \)

\[ f_{b\rightarrow c}(|n\rangle) = \frac{1 - \frac{1}{N}}{1 - \frac{n+1}{N}} = 1 - \frac{n+1}{N} + O \left( \frac{1}{N^2} \right), \]  

(36)

while for a coherent state \( |\alpha\rangle \) holds

\[ f_{b\rightarrow c}(|\alpha\rangle) = \frac{1 - \frac{|\alpha|^2}{N}}{1 + \frac{|\alpha|^2}{N}} = 1 - \frac{|\alpha|^2}{N} + O \left( \frac{1}{N^2} \right). \]  

(37)

This reflects the general property of non-classical states to be more sensitive to decoherence than classical ones.

An alternative way of demonstrating that spin-flip errors do not depend on the number of atoms is to consider the Liouville operator \( \mathcal{L} \) describing uncorrelated spin flips with rate \( \Gamma \)

\[ \dot{W} = \mathcal{L} W = \sum_{j=1}^{N} \mathcal{L}_j W, \]  

(38)

\[ \mathcal{L}_j W = -\frac{\Gamma}{2} \left( \sigma_{bc}^j \sigma_{cb}^j W + W \sigma_{bc}^j \sigma_{cb}^j - 2\sigma_{bc}^j W \sigma_{cb}^j \right). \]  

(39)
Substituting expression (28) yields after tracing over the bright polariton excitations
\[ \mathcal{L}_\rho = -\frac{\Gamma}{2} \left( \Psi^\dagger \Psi \rho + \rho \Psi^\dagger \Psi - 2 \Psi^\dagger \rho \Psi \right). \] (40)

One recognizes that the decoherence rate of the reduced density operator of the ensemble of atoms due to spin flips is the same as for a single atom.

2. Symmetric spin flip

If instead of the asymmetric spin flip \( \sigma_{cb}^i = X_j + i Y_j \) a symmetric spin flip happens, eq. (34) attains the form
\[ W_0 \rightarrow W_1 = \frac{X_j W_0 X_j}{\text{Tr} \{X_j W_0 X_j\}}. \] (41)

We here have kept the normalization denominator although \( \text{Tr} \{X_j W_0 X_j\} = 1 \) because we want to make use of the approximate commutation relations between dark and bright polaritons which hold only to first order in \( 1/N \). Thus both, the numerator and denominator in (41) have to be expanded in the same way to keep the normalization. Carrying out the trace over the bright polaritons yields
\[
\text{Tr}_\Phi \left( X_j W_0 X_j \right) \\
= \frac{1}{N} \text{Tr}_\Phi \left[ \sum_{l,m}^{N-1} \left( \eta_{jl} \Phi_l^\dagger + \text{h.a.} \right) W_0 \left( \eta_{lm} \Phi_m^\dagger + \text{h.a.} \right) \right] \\
+ \frac{1}{N} (\Psi^\dagger + \Psi) \rho_0 (\Psi^\dagger + \Psi) \\
- \frac{1}{N} \text{Tr}_\Phi \left[ \sum_l^{N-1} \left( \eta_{jl} \Phi_l^\dagger + \eta_{j}^* \Phi_l \right) W_0 (\Psi + \Psi^\dagger) + \text{h.a.} \right].
\] (42)

Again the last term in eq. (42) vanishes since there are no excitations of bright polaritons in the initial state \( W_0 \) and in the first term only the combination
\[ \text{Tr}_\Phi \left\{ \eta_{jl} \Phi_l^\dagger W_0 \eta_{jm} \Phi_m \right\} = \rho_0 \delta_{lm} \] (43)
remains and this term evaluates to \( (1 - 1/N) \rho_0 \). This yields
\[ \text{Tr}_\Phi \left( X_j W_0 X_j \right) = \left( 1 - \frac{1}{N} \right) \rho_0 \\
+ \frac{1}{N} (\Psi^\dagger + \Psi) \rho_0 (\Psi^\dagger + \Psi), \] (44)
and we arrive at
\[ \rho_1 = \frac{(1 - \frac{1}{N}) \rho_0 + \frac{1}{N} (\Psi^\dagger + \Psi) \rho_0 (\Psi^\dagger + \Psi)}{1 - \frac{1}{N} + \frac{1}{N} (\Psi^\dagger + \Psi)^2}, \] (45)
which is similar to the case of an asymmetric spin-flip, eq. (34). Once again it is seen that the collective quantum memory does not have an enhanced sensitivity to spin flip errors as compared to a single-atom system.

The fidelity of the memory now reads for a stored Fock and coherent state
\[ f_{b+c}(\ket{n}) = 1 - \frac{2n + 1}{N} + O \left( \frac{1}{N^2} \right) \] (46)
\[ f_{b+c}(\ket{\alpha}) = 1 - \frac{1}{N} + O \left( \frac{1}{N^2} \right) \] (47)

3. Phase flip

If after the preparation of an ideal storage state an atom undergoes a phase flip the corresponding positive map would read
\[ W_0 \rightarrow W_1 = \frac{Z_j W_0 Z_j}{\text{Tr} \{Z_j W_0 Z_j\}}. \] (48)

This map is however not a good starting point of further discussions because the approximations used when introducing bosonic polariton operators lead to \( Z_j \equiv 1_j \). For this reason we follow a different approach and calculate the fidelity of the quantum memory directly. Consider an ideal storage state initially of the form
\[ \ket{\psi_0} = \sum_{n=0}^M c_n \ket{D, n} = \sum_{n=0}^M \frac{1}{\sqrt{n!}} (\Psi^\dagger)^n \ket{b, 0} \] (49)
where \( M \ll N \). If the \( k \)th atom undergoes a phase flip the state changes according to
\[ \ket{\psi_0} \rightarrow \ket{\psi_1} = \sum_{n=0}^M c_n \frac{1}{\sqrt{n!}} (\tilde{\Psi})^n \ket{b, 0} \] (50)
where
\[ \tilde{\Psi} = -\frac{1}{\sqrt{N}} \sum_j \sigma_{bc}^j = \Psi + \frac{2}{\sqrt{N}} \sigma_{bc}. \] (51)

Using eq. (28) this can be written in the form
\[ \tilde{\Psi} = \left( 1 - \frac{2}{N} \right) \Psi + \frac{2}{N} \sum_{l=1}^{N-1} \eta_{kl} \Phi_l. \] (52)

This yields in lowest order of \( 1/N \)
\[ \tilde{\Psi}^n = \left( 1 - \frac{2n}{N} \right) \Psi^n + \frac{2n}{N} \sum_l \eta_{kl} \Phi_l \Psi^{n-1} + O \left( \frac{1}{N^2} \right) \] (53)

Tracing over the bright polariton excitations leads to the reduced density operator
\[ \rho_1 = \text{Tr}_\Phi \left[ \ket{\psi_1}\bra{\psi_1} \right] \] (54)
\[ = \sum_{n, m=0}^M c^*_n c_m \left( 1 - \frac{2(n + m)}{N} \right) \ket{D, n}\bra{D, m} \]
and eventually to the fidelity
\[ f_{\text{deph}}(|\psi_0\rangle) = 1 - \frac{2\langle n \rangle}{N} + O\left(\frac{1}{N^2}\right), \tag{55} \]
where \( \langle n \rangle = \langle \Psi^\dagger \Psi \rangle \) is the average number of dark-state polaritons in the initial state. One recognizes that a phase flip of a single atom leads to a fidelity reduction which is of the order of \( 1/N \). The term \( 1/N \) again compensates for the fact that in an \( N \)-atom ensemble the likelihood that one arbitrary atom undergoes a phase flip is \( N \) times the probability of a phase flip for a single atom. It is interesting to note that the fidelity only depends on the average dark-state polariton number. I.e. dephasing affects in lowest order of \( 1/N \) classical and nonclassical states in a similar way.

**C. One-atom losses**

Another important source of errors in a collective quantum memory is the loss of an atom from the ensemble. As discussed in section II all storage states corresponding to the same dark-state excitations in ensembles of different atom number are equivalent as long as the atom number is large compared to the relevant number of stored photons. We now calculate the fidelity of the quantum memory after loss of one atom. We consider again an ideal initial storage state
\[ |\psi_0\rangle_N = \sum_{n=0}^{M} c_n |D, n\rangle_N, \tag{56} \]
where the subscript \( N \) denotes the total number of atoms in the ensemble and \( M \ll N \). The loss of an atom, which, without loss of generality, can be taken to be the \( N \)-th atom, can be described by the partial trace over the degrees of freedom of that atom
\[ W_1 = \text{Tr}_N \{|\psi_0\rangle\langle \psi_0|\}. \tag{57} \]
To carry out the trace let us first consider the case of a Fock state of \( n \) polaritons \(|D, n\rangle_N\)
\[ |D, n\rangle_N = \binom{N}{n}^{-1/2} \sum_{j_1 < \cdots < j_n} |b_1 \cdots c_{j_1} \cdots c_{j_n} \cdots b_N\rangle. \tag{58} \]
Tracing over the \( N \)th atom results into
\[ \text{Tr}_N \{|D, n\rangle_N \langle D, n|\} = \binom{N}{n}^{-1/2} \sum_{j_1 < \cdots < j_n} |b_1 \cdots c_{j_1} \cdots c_{j_n} \cdots b_N\rangle \langle \cdots | \tag{59} \]
\[ \begin{align*}
&= \binom{N}{n}^{-1/2} \binom{N-1}{n-1} \sum_{j_1 < \cdots < j_{n-1}} |b_1 \cdots c_{j_1} \cdots c_{j_{n-1}} b_{N-1}\rangle \langle \cdots | \\
&+ \binom{N}{n}^{-1/2} \binom{N-1}{n-1} \sum_{j_1 < \cdots < j_{n-1}} |b_1 \cdots c_{j_1} \cdots c_{j_{n-1}} b_{N-1}\rangle \langle \cdots |
\end{align*} \]
\[ = \frac{N-n}{N} |D, n\rangle_{N-1} \langle D, n| + \frac{n}{N} |D, n-1\rangle_{N-1} \langle D, n-1|, \tag{60} \]

Thus the fidelity of the quantum memory for a Fock state \(|n\rangle\) after loss of a single atom is given by
\[ f_{\text{loss}}(|n\rangle) = 1 - \frac{n}{N}. \tag{61} \]
The decrease of the fidelity again scales only as \( 1/N \). This result could of course have been expected as the \( n \) excitations are equally distributed over all atoms. Thus removing one reduces the stored information only by the amount \( n/N \). Generalizing the above result to nondiagonal elements leads after some calculation to
\[ \text{Tr}_N \{|D, n\rangle_N \langle D, m|\} = \binom{N-n}{N-m} \frac{|D, n\rangle_{N-1} \langle D, m|}{N} + \frac{\sqrt{nm}}{N} |D, n-1\rangle_{N-1} \langle D, m-1|. \tag{62} \]
Thus the fidelity after the loss of an atom reads for the case of a general state:
\[ f_{\text{loss}}(|\psi_0\rangle) = 1 - \frac{1}{N} \left( \langle \Psi^\dagger \Psi \rangle - \langle \Psi^\dagger \Psi \rangle \right) + O \left( \frac{1}{N^2} \right) \tag{63} \]
If the initial storage state corresponds e.g. to a coherent state, the second and third term in \( 63 \) compensate each other and the fidelity differs from unity only in order \( 1/N^2 \). Here again the robustness of classical states becomes apparent.

**D. Atomic motion**

Until now it has been assumed that the atoms used in the quantum memory are at a fixed position during the entire storage time. Since the coupling of the atoms to the quantum as well as control fields contains however a spatial phase, see eq. II, atomic motion results in an effective dephasing and will lead to a reduction of the fidelity. Recently Sun et al. have argued that inhomogeneities of the atom-light interaction strength or in the control field together with atomic motion lead to an increase of the characteristic decoherence rate by a factor \( \sqrt{N} \). We thus will analyze the effect of atomic motion in the following in more detail. To this end we will follow the approach of subsection III B 3 and describe the motion by the map of an initially ideal storage state \(|\psi_0\rangle\), \( |\psi_1\rangle \), according to
\[ |\psi_0\rangle \rightarrow |\psi_1\rangle = \sum_n c_n \frac{1}{n!} (\hat{\Psi}^\dagger (t))^n |b, 0\rangle, \tag{64} \]
where
\[ \hat{\Psi}^\dagger (t) = -\frac{1}{\sqrt{N}} \sum_j \sigma_j^c \exp \left\{ -i \Delta \tilde{k} \cdot \tilde{r}_j (t) \right\}, \tag{65} \]
with \( \tilde{r}_j (t) \) denoting the position of the \( j \)-th atom at time \( t \) and \( \Delta \tilde{k} = \tilde{k}_1 - \tilde{k}_0 \hat{e}_z \) is the wavevector difference between
control field and quantized mode. It should be noted that
is equivalent to a coupling field with inhomogeneous phase.

To reduce the effect of motion in an atomic vapor one could either reduce the temperature or use a buffer gas of sufficient density. In the latter case, which has been used in room temperature gas-cell experiments \[15\], the free motion is replaced by a diffusion. In the following we will restrict the discussion to this important case. We then can assume that the phase

$$\Delta \phi_j(t) \equiv \Delta \vec{k} \cdot \vec{r}_j(t)$$  \hspace{1cm} (66)

follows a Wiener diffusion process \[29\]:

$$\frac{d}{dt} \Delta \phi_j(t) = \mu_j(t),$$  \hspace{1cm} (67)

$$\mu_j(t) = 0,$$  \hspace{1cm} (68)

$$\mu_j(t) \mu_k(t') = D \delta_{jk} \delta(t-t')$$  \hspace{1cm} (69)

with \(D\) being a characteristic diffusion rate. We now want to show that the decrease in fidelity due to the phase diffusion is only determined by \(D\) and independent of the number of atoms \(N\). For this it is sufficient to consider an initial Fock state \(|n = 1\rangle\). Reexpressing the single-atom flip operators by collective ones yields

$$\Psi(t) = \frac{1}{\sqrt{N}} \sum_j e^{i \Delta \phi_j} \left[ - \sum_{i=1}^{N-1} \eta_{ji} \Phi_j + \Psi^\dagger \right].$$  \hspace{1cm} (70)

With this one finds for an initial Fock state \(W_0 = |D, 1\rangle \langle D, 1|\)

$$W_1(t) = \frac{\Psi^\dagger}{\sqrt{N}} \frac{\Phi_0}{\Phi_0^\dagger} \langle b, 0| \langle b, 0| \Psi \rangle, \hspace{1cm} (71)$$

where the overline denotes averaging over the phase diffusion process. From \(W_1(t)\) we can calculate the fidelity of the quantum memory by first tracing out the bright polaritons and then sandwiching with the original state \(|D, 1\rangle\). This yields

$$f_{\text{motion}}(1|1) = \langle D, 1| \text{Tr}_F \left( W_1(t) \right) |D, 1\rangle, \hspace{1cm} (72)$$

$$= \left( \frac{1}{N} \sum_j e^{i \Delta \phi_j} \right) \left( \frac{1}{N} \sum_k e^{-i \Delta \phi_k} \right) \hspace{1cm} \text{Tr}_F \left( \Psi \right) \hspace{1cm} \text{Tr}_F \left( \Psi \right) \hspace{1cm} \text{Tr}_F \left( \Psi \right) \hspace{1cm} \text{Tr}_F \left( \Psi \right)$$

$$= \frac{1}{N} + \frac{1}{N^2} \sum_{j \neq k} e^{i \Delta \phi_j} e^{-i \Delta \phi_k}$$

$$= \frac{1}{N} \left( 1 + (N-1)e^{-Dt} \right) \sim e^{-Dt}.$$  \hspace{1cm} (72)

A generalization to an arbitrary fock state \(|D, n\rangle\) leads to a fidelity decay proportional to \(\exp\{-nDt\}\). One recognizes that the atomic motion causes a decay of the fidelity with a rate given only by the single-atom diffusion rate \(D\). In contrast to the results of Sun, Yi and You \[24\] we find that there is no enhancement of the decay with increasing number of atoms, which is again due to the existence of equivalence classes.

IV. NON-ADIABATIC COUPLING AND DECOHERENCE

In section \[11\] it was shown that for the retrieval of a stored quantum state of light only the reduced density operator \(\rho_0\) is relevant. For this reason all states of the system which have the same number of dark state polariton excitations but an arbitrary number of excitations in the bright state polariton modes belong to the same equivalence class and lead to the same result, provided the read-out process is adiabatic. Due to decoherence a large number of bright state polaritons may be excited in the system after the storage period. Now the question arises what happens to these excitations if the read-out process is not adiabatic. Even if there is only a weak non-adiabatic coupling between bright and dark state polariton modes it may be sufficient to transfer some of the unwanted excitations into the dark-polariton mode. We will show in the following that only the bright-polariton mode \(\Phi_0\) can lead to non-adiabatic contributions to the read-out signal. For this we consider the (imaginary) interaction Hamiltonian eq. \[23\] in a rotating frame and add the coupling of the quantized resonator mode \(a\) to free-space modes \(b_k\):

$$H = -i \hbar g^2 N \gamma \cot^2 \theta(t) \sum_{l=1}^{N-1} \Phi_l \Phi_l^\dagger + h \sum_k \kappa a b_k + h a.$$  \hspace{1cm} (73)

The equation of motion for the dark-state polariton operator \(\Psi = \cos \theta(t) a - \sin \theta(t) \Sigma_{bc}\), with \(\Sigma_{bc} = \sum_{j} \sigma_{bc}^j / \sqrt{N}\) then reads

$$\dot{\Psi} = -\dot{\theta}(t) \sin \theta(t) a + \dot{\theta}(t) \Sigma_{bc} + i \hbar \left[ H, \Psi \right]$$

$$= -\dot{\theta}(t) \Phi_0 + i \kappa \cos \theta(t) \sum_k b_k. \hspace{1cm} (74)$$

Thus the dark-polariton is coupled to the outside modes \(b_k\) and the bright-polariton operator \(\Phi_0 = \sin \theta(t) a + \cos \theta(t) \Sigma_{bc}\) only. In a similar way one finds for \(\Phi_0\) and the \(b_k\)'s:

$$\dot{\Phi}_0 = \dot{\theta}(t) \Psi,$$  \hspace{1cm} (75)

$$\dot{b}_k = i \kappa a$$

$$= i \kappa \cos \theta(t) \Psi + i \kappa \sin \theta(t) \Phi_0. \hspace{1cm} (76)$$

One recognizes that \(\{\Psi, \Phi_0, b_k\}\) are a closed set of coupled operators even if non-adiabatic corrections in the read-out proportional to \(\dot{\theta}(t)\) are taken into account. Thus only decoherence induced excitations generated in \(\Phi_0\) will influence the read-out signal, all the other \(N-1\) bright polariton modes remain uncoupled from the storage systems.

V. SUMMARY

In the present paper we have studied the influence of individual decoherence processes on the fidelity of a quantum memory for photons based on ensembles of atoms.
Despite the fact that the atomic storage states corresponding to non-classical states of the radiation field are entangled many-particle states, the system shows no enhanced sensitivity to decoherence as compared e.g. to single-atom storage systems, if it is caused by a coupling of the atoms to individual and independent reservoirs. This is due to the existence of equivalence classes of storage states corresponding to the excitations of only one eigenmode of the system, the dark-state polariton. It was shown that all states with equal reduced density operator after tracing out the $N$ bright-polariton modes reproduce the same quantum state of light in the read-out process. For similar reasons no stringent requirements for preparation of the atomic system before the storage exist. It is sufficient that the number of atoms remaining in the storage level $|c⟩$ after preparation of the ensemble is small compared to the total number of atoms, which can easily be achieved by optical pumping. It was shown moreover that the loss of an atom from the sample causes only an error of the order of $1/N$. Motion of atoms during the storage time causes an effective dephasing and thus leads to a decrease in fidelity. It was shown, however, that the corresponding error is independent on the number of atoms, which is in contrast to the result of [12]. Finally since non-adiabatic effects only couple one of the bright polariton modes to the dark-polariton, the potentially large number of excitations in the $N$ bright-polariton modes caused by decoherence processes does not leak into the read-out signal in a significant amount even if the read-out process is not adiabatic.

The present paper proves that atomic ensembles are suitable systems for the storage of quantum states of the radiation field even in the presence of non-cooperative decoherence processes. It should be noted that this conclusion does however not apply to the quantum gate between stored photonic qubits based on dipole blockade proposed in [26] nor to the photon detection scheme suggested in [27].

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