Diffusion induced decoherence of stored optical vortices

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We study the coherence properties of optical vortices stored in atomic ensembles. In the presence of thermal diffusion, the topological nature of stored optical vortices is found not to guarantee slow decoherence. Instead the stored vortex state has decoherence surprisingly larger than the stored Gaussian mode. Generally, the less phase gradient, the more robust for stored coherence against diffusion. Furthermore, calculation of coherence factor shows that the center of stored vortex becomes completely incoherent once diffusion begins and, when reading laser is applied, the optical intensity at the center of the vortex becomes nonzero. Its implication for quantum information is discussed. Comparison of classical diffusion and quantum diffusion is also presented.

Introduction Photons can carry orbital angular momentum (OAM), which can be created 1, manipulated 2 and detected 3. The OAM states are associated with vortices of a helical phase structure \( e^{i m \varphi} \). Each vortex is a topological defect, characterized by a winding number \( m \), obtained from the \( 2 \pi m \) phase twist around the vortex. The light with OAM is exemplified by the most commonly known Laguerre-Gaussian (LG) modes \( \text{LG}_{p}^{m} \) with \( p \) the number of nodes in radial direction 4, 5. Entanglement through the OAM degree of freedom has been demonstrated for photon pairs from parametric down conversion 6. Since the winding number can be any integers, the OAM states have been proposed theoretically for multiple-alphabet quantum cryptography with higher information density and a higher margin of security 4, 8. Other proposals, such as quantum coin tossing and violation of Bell inequalities, using the OAM states have been reviewed by Ref. 9.

One of the key challenges for optically based quantum information (QI) processing is the difficulty of storing optical fields. It has been demonstrated 10, 11, 12 that a superposition of the OAM states can be stored in a non-rotating BEC in terms of vortex states of the condensate. Meanwhile, the OAM states can also be stored in “hot” atomic ensembles using slow light techniques 13, 14, 15, 16, 17. The information of photonic states, namely the amplitude and phase, is continuously transformed into Raman coherence, i.e., spin-density waves, of the atomic ensemble, which can be later retrieved. In practice, processes such as inhomogeneous magnetic field 18 and/or thermal diffusion can lead to the decay of the Raman coherence. Then, to what extent can the optical vortex states be stored coherently? Are they going to be more robust than the Gaussian state? The answers to these questions will determine how the OAM are used in QI.

Generally speaking, the topological structure of vortices makes it a good candidate for QI 19, 20, 21 because it is stable against continuous deformations which cannot cause it to decay or to “unwind”. Actual studies of such robustness against various processes are of course necessary. In particular, one needs to study the robustness in the presence of diffusion 22 for the stored optical vortex state, which is crucial for applications such as quantum repeaters 23, 24 and multiple beam splitters for generating entangled single photons 25. Towards answering this question, Pugatch, et al. 17 showed that in the presence of diffusion, the dark center of a stored OAM mode is well preserved, and the dark center of a Gaussian mode generated by blocking its center disappears after a short time of diffusion. They attributed the stable dark center of the stored OAM mode to the robustness of the topological nature of vortex in the presence of vortex.

In this Letter, we provide a careful study for the robustness of the stored vortex states (exemplified by \( p = 0 \) unless otherwise stated) in the presence of diffusion. We find that the stable dark center of the vortex states is not directly associated with the topological robustness. And the vortex states are actually more vulnerable to diffusion than the Gaussian state. This is because: (1) the diffusion is a global process during the storage and it can destroy the topological order; (2) the readout process only measures a specific output modes corresponding to Raman coherence but not the full state; (3) for vortex states, the Raman coherence interferes destructively, while the full quantum state evolves differently.

Our basic assumption is that there is no spin exchange or interaction between atoms, the evolution of coherence obeys diffusion equation \( \dot{\rho} = D \nabla^2 \rho \), where \( \rho \) and the \( D \) are the density matrix and the diffusion coefficient, respectively. We consider a three-level lambda system as generally used for light storage 13, 14: the weak probe laser is applied between the ground state \( |1 \rangle \) and the excited state \( |3 \rangle \), and the pump laser is addressing the transition between \( |3 \rangle \) and another ground state \( |2 \rangle \).

Comparison between the decay for vortex states and vortex-free states Using the propagator of the diffusion equation, the expected atomic coherence of a LG mode after diffusion of duration \( t \) is 17

\[
\rho_{12}(\vec{r}, t) = \frac{(-g/Q)}{\sqrt{s(t)^{m+1}}} A_m(r, \sqrt{s(t)w_0}) e^{-im\varphi} \quad (1)
\]

with \( g \) the vacuum Rabi frequency for the probe field,
the radial profile of a LG mode shows that different angular momentum states decay faster than p modes. 

After some time of diffusion is just the Gaussian mode. 

The scaling factor $\sqrt{s(t)}$ in $A_m$ of Eq. (11) means that the functional forms of coherence are preserved for both the LG mode and the Gaussian mode. This implies the functional form stability of stored coherence against diffusion does not require topological defects. Indeed, the disappearance of the dark center of the center-blocked Gaussian mode after diffusion demonstrates as the stability of the Gaussian mode: diffusion tries to restore the nonzero intensity at its center. Of course, the restoration can also be understood by decomposing the center-blocked Gaussian mode to LG modes and noting that $p \neq 0$ modes decay faster than $p = 0$ mode and what is left after some time of diffusion is just the Gaussian mode.

From the above discussion, it is clear that for the LG light ($m \neq 0$), the coherence at the center $r = 0$ stays zero during diffusion. However, as we show now, population at $r = 0$ does not stay zero. To simplify the discussion, we assume, at time $t = 0$, the populations of atoms are $\rho_{11}(\vec{r}, t = 0) = 1$ and $\rho_{22}(\vec{r}, t = 0) = |\rho_{12}(\vec{r}, t = 0)|^2$, which is a good assumption for strong pump and weak probe lasers as usually used in light-storage experiments. At time $t$ after diffusion, we have $\rho_{11}(\vec{r}, t) = 1$ and

$$\rho_{22}(r, t) = \frac{4e^{-\frac{2r^2}{w_0^2}}}{\pi} \frac{P (32D^2t^2 + r^2w_0^2 + 4Dt w_0^2)}{8Dt + w_0^2}$$

for $m = 1$ vortex state and

$$\rho_{22}(r, t) = \frac{2e^{-\frac{2r^2}{w_0^2}}}{\pi} \frac{P}{(8Dt + w_0^2)}$$

for a Gaussian mode. The evolutions of populations are plotted in Fig. (1) and Fig. (2). While it seems that coherence only diffuses outwards in Fig. (1), Eq. (2) and Fig. (1) clearly show that diffusion goes in all directions as it should be. The outwards moving coherence during diffusion is because the interference cancels the inwards diffusing coherence. In contrast, the population does not interfere with each other and thus the diffusion towards the center is clearly seen. Indeed, the population at the center quickly approaches a global maximum as time increases (Fig. (1)). We also note that integration of $\rho_{22}$ over the whole space is conserved during the diffusion.

**Diffusion induced decoherence** We have seen that diffusion for coherence and population obeys different equations (Eq. (11)). This brings phase decoherence for the stored coherence. To characterize the decoherence, we define a coherence factor $f = \frac{\rho_{11}^2 + \rho_{22}^2}{\rho_{11}^2 + \rho_{22}^2 + \rho_{12}^2 + \rho_{21}^2}$, where $\eta$ is an infinitesimal number. $f$ is a linear function with $f = 1$ for a pure state and $f = 0$ for a completely mixed state. Thus, $f$ is a good parameter to describe the (local) coherent property. As a specific example, we plot the coherence factor $f$ in Fig. (1) and Fig. (2). Figure (1): shows that right after diffusion begins, coherence factor $f(r = 0)$ or the stored vortex drops from 1 to 0 because at $r = 0$, the population becomes nonzero when diffusion starts while its coherence keeps zero. We note that such sudden changes within an infinitesimal time are very uncommon in physical processes. As diffusion time increases, $f(r)$ drops faster for $r$ that do not have much initial inhomogeneous coherence, and $f(r)$ approaches zero at very large time. This latter result holds for a Gaussian mode as well (shown in Fig. (2)). We also note that at $r$ where the diffusion of stored inhomogenous coherence and population has not arrived at, the coherence factor $f$ stays at 1 because all population is in $|1\rangle$ and it is a pure state. But the weight $\rho_{22}$, justified by its non-negative and conserved integration, of these coherence factors for the retrieved light approaches zero.

Here is how coherence factor $f$ may be obtained from the experiments. When the reading pulse is applied, the incoherent part will be retrieved as fluorescence in all directions. Collecting both the fluorescence and the coherent emission then allows extracting $f$. Of course, setting the detector at the forward direction as generally used, e.g., in [27], can only collect a finite ratio of fluorescence, while the coherent part is collected by the detector [27].
What we want to emphasize is that incoherence makes intensity at the center of a retrieved vortex nonzero, prohibited by a coherent vortex state. As diffusion time increases, nonzero intensity at the center builds up. Therefore, generation of mixed state makes additional loss of retrieved fidelity compared with vortex-free case. Diffusion of the population makes visibility decrease and finally kill the vortex. Of course, this part of reduction of fidelity can be alleviated by using spatial filtering of the optical mode to prevent spontaneously emitted photons from going into the detectors. In this case, the retrieval probability is just the fidelity \( F = 1/s(t)^{m+1} \).

The different collection efficiency of fluorescence photons and coherent photons by a forward set detector helps explain why the visibility of the center-blocked Gaussian mode disappears very quickly, while the hole of vortex disappears very slowly \[17\]. The homogeneous phase of the center-blocked Gaussian mode makes nonzero coherence inside the hole after diffusion and thus the disappearance of hole once the coherence is read. This is in contrast with a stored vortex, whose coherence at the center remains zero all the time. Its nonzero intensity at the center comes only from incoherence of the center. However, were most of fluorescence photons collected by the detector, the dark center of a vortex would disappear quicker than that of the Gaussian mode. It is the spatial filtering of the optical mode that helps to overcome the fluorescence from the center.

**Dependence of decay on order of phase singularity** We now come back to the decay of coherence. We have noted that the coherence of stored LG modes decays according to a power law \( F = 1/s(t)^{m+1} \). The larger the order of phase singularity, given by \( m \), is, the faster the coherence decays. An additional example of exponential decay rate \( 2Dk^2 \) due to diffusion of a plane wave \( e^{-ikx} \), which is faster than the power law decay, also corroborates this idea, because a plane wave has infinity number of phase singularities. Incidentally, the limit of \( F \) for large \( m \) does
not go to exponential decay of a plane wave is because the amplitude of coherence in LG modes is not a constant, different from the case of a plane wave. We also note that the larger $k$ of a plane wave, the larger decay rate, which is because larger $k$ means the pattern have higher spatial frequency and the diffusion becomes easier to cancel the coherence. Such diffusion of a plane wave happens if the pump and probe lasers have different wave vectors.

A few more remarks on the different decay behaviors are in order. First, as a direct application of our discussion, the less phase gradient, the better for stored coherence against diffusion. For example, diffusion of stored general $LG_p^m$ with both winding number and number of radial nodes being non-zero $\mathbf{2}$ induces faster decay for $p > 0$ than $p = 0$. Furthermore, we note that although the number of nodes in the coherence does not change with diffusion time, the positions for the off-center radial nodes change, which is different from the center one. The non-moving position of the center node comes from geometric symmetry of the vortex. Since the decay rate depends on both $|m|$ and $p$ for the mode $LG_p^m$, if OAM states are to be used as bases for quantum information, a preferred basis to reduce loss of entanglement for quantum information is actually two modes with same $p$ but opposite $m$. Second, since diffusion induced decay of the total intensity is not exponential for Gaussian and LG modes, the exponential decay at a rate $20,000 s^{-1}$ of Ref. [17] is not determined by diffusion.

The classical diffusion vs the quantum diffusion So far, we have only discussed decoherence induced by the classical diffusion associated with the inhomogeneous distribution of coherence. We note that decoherence can also happen in a homogeneous system as a result of the quantum diffusion. The quantum diffusion in light storage system happens when pump and probe lasers couple different momentum states, which introduces decoherence [28]. But this decoherence is reversible, say by photon echo techniques, in contrast with the classical diffusion. This is because the quantum diffusion is described by the complex Schrodinger equation with $i$ in it, while classical diffusion does not. Finally, we note that when the same momentum states are coupled by choosing pump and probe lasers to have the same wave vectors, the quantum diffusion disappears [28]. Incidentally, inhomogeneous magnetic fields also induce quantum diffusion.

Discussion Our results indicate that diffusion actually introduces more decoherence in a stored vortex mode than a stored Gaussian mode, which may have important implication for quantum information. Of course we, however, do not rule out that in other processes, such as a quantum gate operation, vortex states are possibly much better than the topological-free state. Nor did we discuss diffusion-free systems such as BEC [29] and bound excitons in semiconductors [30, 31].

Conclusion We found that during diffusion, the coherence of stored vortex states decays faster than that of Gaussian states. This is surprising because vortex states are associated with topological properties, and are presumably considered as more stable than topological-defect-free states. The underlying reason is that diffusion is a non-local process. More generally, the less phase gradient of a stored coherence, the better for it against diffusion. Furthermore, calculation of coherence factor showed that the center of stored vortex becomes completely incoherent once diffusion begins, and when reading laser is applied, the optical intensity at the center of the vortex builds up. It’s implication for quantum information was discussed. Finally, we compared the classical diffusion and the quantum diffusion.

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