Enhancing the capacity and performance of collective atomic quantum memory

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Present schemes involving the quantum non-demolition interaction between atomic samples and off-resonant light pulses allow us to store quantum information corresponding to a single harmonic oscillator (mode) in one multiatomic system. We discuss the possibility to involve several coherences of each atom so that the atomic sample can store information contained in several quantum modes. This is achieved by the coupling of different magnetic sublevels of the relevant hyperfine level by additional Raman pulses. This technique allows us to design not only the quantum non-demolition coupling, but also beam splitterlike and two-mode squeezerlike interactions between light and collective atomic spin.

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To enable quantum communication over long distances using quantum repeaters \cite{1}, one needs to transform a quantum signal from light pulses into material media and vice versa. Recently, this has been achieved by the quantum non-demolition (QND) interaction between alkali atom vapors and off-resonant optical pulses \cite{2,3} and also by the light-induced transitions between atomic ground states in a cloud of atoms \cite{4,5}. The principles of the off-resonant interaction between light and polarized atoms were given in \cite{6}, and recently their application for collective spin measurements and quantum noise suppression has been suggested \cite{7} and demonstrated \cite{8,9}. It turns out that the QND scheme can be applied to many valuable quantum-information protocols \cite{10}. However, for most of them either the QND interaction has to be applied several times in sequence (e.g., for swapping a quantum state between light and matter), or measurements with feedback must be used (e.g., for transferring state from light to matter that was prepared in a well-defined initial state). This makes many procedures rather cumbersome (one would have to store long optical pulses to use the multi-pass schemes) so that more straightforward protocols are highly desirable. Also, many questions remain as how to take full advantage of the atomic degrees of freedom.

In this Letter, we propose a method to increase the amount of quantum information stored and processed in the atomic sample. We exploit the structure of magnetic atomic levels and use additional Raman coupling lasers to store information about several optical modes into various atomic ground-state coherences. By properly choosing the frequency of the coupling laser, it is possible to design various effective light-atom interaction Hamiltonians such as QND coupling, two-mode squeezing and beam splitter-like coupling. Especially the last one would be very useful to enable us quantum state exchange between light pulses and atomic samples. The scheme could thus greatly enhance our ability to process quantum information at the light-matter interface.

The principle of the interaction is as follows. All atoms in the sample are initially pumped into a particular magnetic state $|m_F = -F\rangle_x$ of some hyperfine level $F$ of the electronic ground state, with $x$ being the quantization axis. The information is then encoded into the coherence between the state $|m_F = -F\rangle_x$ and a weakly populated neighboring state $|m_F = -F + 1\rangle_x$ by means of light pulses traveling in the $z$ direction (see Figs. 1 and 2). Each pulse has a strong component which is $x$-polarized and a weak quantum component which is $y$-polarized. With $z$ as the quantization axis, the influence of the field on the atoms can be described as the ac Stark shift induced by the imbalance between the right- and left-circularly polarized components (see Fig. 2) changing the phases between different $|m_F\rangle_z$ states. The influence of the atoms on the field can be understood as the Faraday effect with asymmetry in the $\sigma^{(2)}_{m,n}$ populations causing slight rotation of the linear polarization (here and in what follows $\sigma_{m,n}$ denotes the operator $|m\rangle\langle n|$ involving the atomic states $|m\rangle$ and $|n\rangle$ and the upper index denotes the quantization axis). With $x$ as the quantization axis, the interaction can be under-
stood as coherent Raman scattering of the field on the $\sigma_{F,F+1}^{(x)}$ coherences. In a typical experiment the atoms are placed in an $x$-oriented magnetic field which causes rotation of the atomic polarization. The effect of the atoms on the optical field is then observed on the sideband with the Larmor frequency $\Omega \propto B$, where $B$ is the magnetic induction. In the experiments so far, the information has been written into the $\sigma_{F,F+1}^{(x)}$ coherence and its conjugate only, whereas all the remaining coherences $\sigma_{F,F,m}^{(x)}$ with $m \neq -F + 1$ were negligible. A natural question arises: could one use also these coherences as a medium for storing quantum information? The coherence $\sigma_{F,F,m}^{(x)}$ would contribute to the oscillation of the atomic populations on the $z$-quantization magnetic states with frequency $(m + F)\Omega$. Photodetection on this sideband should reveal information about the corresponding coherence.

Let us first briefly discuss the traditional single-coherence QND scheme with a multilevel atom interacting with an off-resonant field (see Fig. 2). The atom-field interaction Hamiltonian is $H = \hbar \sum_m \sigma_{m,m}^{(z)}(g_{mR}\varepsilon_L^{(x)} + g_{mL}\varepsilon_R^{(x)})$, where $\sigma_{m,m}^{(z)}$ is the population in the magnetic state $|F,m\rangle$ with the quantization axis $z$, and $\varepsilon_R^{(x)}$ is the positive-frequency operator of the electric intensity of the right (left) circularly polarized field. The coupling constants are $g_{mR(L)} = 2/(\hbar^2\Delta)\sum_{F'}\mu_{F,F',m+1}^2$ where $\Delta$ is the detuning from a given fine-structure level (assumed to be much larger than the hyperfine splitting in that level, but much smaller than detuning from any other atomic level), $\mu_{F,F,m}$ is the dipole moment element between the hyperfine state $|F,m\rangle$ of the ground electronic state and the hyperfine state $|F',m'\rangle$ of the relevant excited electronic state, and the summation runs over all hyperfine levels $F'$ in the fine-structure level. In the cesium atoms are prepared in the hyperfine level $F = 4$ of the atomic ground state $6S_{1/2}$ and the field is coupled to the transition to states on the $6P_{3/2}$ level; the dipole moments sum up as $\sum_{F'}\mu_{F,F,m+1}^2 = \mu_0^2(8 \pm m)/48$, where $\mu_0^2 = e^2/(6S_{1/2}|r||6P_{3/2}|)^2$ and the reduced matrix element can be expressed in terms of the decay rate $\gamma$ of the $6S \leftrightarrow 6P$ transition, $(6S_{1/2}|r||6P_{3/2}|)^2 = 3\gamma^2/(\alpha\omega_0^3)$, where $\alpha$ is the fine structure constant [11]. Thus, the relevant part of the Hamiltonian is

$$H_{AF} = -\frac{\mu_0^2}{24\hbar\Delta}(\varepsilon_R^{(x)}\varepsilon_R^{(x)} - \varepsilon_L^{(x)}\varepsilon_L^{(x)}) \sum_m m\sigma_{m,m}^{(x)}.$$  (1)

We assume that the strong coherent $x$-polarized field has frequency $\omega_0$. The atoms then resonantly couple to two $y$-polarized field modes $a_{y\pm}$ oscillating on the Larmor sidebands $\omega_\pm \pm \Omega$ and we can write $\varepsilon_R(L) = E_0(a_{x \pm} \pm i(a_{y+} - a_{y-}))/\sqrt{2}$, where $E_0$ is the vacuum electric field, $E_0^2 = \hbar \omega_0/(2\epsilon_0 V)$, $V = A\ell T$ is the quantization volume, $A$ and $T$ are the transversal area and duration of the optical pulse, respectively, and $a_x$ is the annihilation operator of the strong $x$-polarized field. It is convenient to work with nonmonochromatic modes $\phi_{y\pm} = 2^{-1/2}(a_{y+} - a_{y-})$, whose field quadratures $X_j = 2^{-1/2}(a_j + a_j^\dagger)$, $P_j = -i2^{-1/2}(a_j - a_j^\dagger)$ are measured using homodyne detection as the sine and cosine signal components oscillating at frequency $\Omega$. Transforming from the $z$-quantization to the $x$-quantization, and averaging out fast oscillating terms, the interaction Hamiltonian becomes

$$\hat{H}_{AF}^{(1)} = i\hbar \sum_m a_x^\dagger G_m^{(x)}(a_{y+} + \sigma_{m+1,m}^{(x)} + a_{y-} - \sigma_{m,m+1}^{(x)}) + h.c.,$$  (2)

where $G_m^{(x)} = \mu_0^2 E_0^2/(48h^2\Delta)|20 - m(m + 1)|$. When the $a_x$ mode is in a strong coherent state $|\alpha_0\rangle$ with $\alpha_0$ real and the atoms are initially prepared in state $|F,m = -F\rangle$ so that only the coherences $\sigma_{F,F,m}^{(x)}$ and $\sigma_{F,F,-m}^{(x)}$ are non-negligible, after summing over all $N_A$ atoms, the interaction Hamiltonian $H_{AF}^{(1)} = \sum_{k=1}^{N_A} \hat{H}_{AF,k}$ becomes

$$H_{AF}^{(1)} = i\hbar \rho^{(x)}(X_C X_A + X_S P_A).$$  (3)

This is the QND-type Hamiltonian in which the atomic quadratures are $X_A = \frac{\sqrt{2N_A}}{\sqrt{2N_A}} \sum_{k=1}^{N_A} (\sigma_{F,F+1}^{(x)} + \sigma_{F,F-1}^{(x)} + \sigma_{F,F+1}^{(x)} - \sigma_{F,F-1}^{(x)})$, the coupling constant is $\kappa = -E_0^4\mu_0^2 \sqrt{V_L N_A}/(12h^2\Delta)$, with the photon number $N_L = |\alpha_0|^2$, and the index $k$ denotes individual atoms.

To couple the light field to higher coherences we propose applying additional strong fields co-propagating
with the signal fields. A single left-circularly polarized beam propagates in the $z$-direction, with frequency $\omega_c$, slightly off-resonance with respect to the Raman transition to the other hyperfine level $F_2$ of the ground state, $\omega_c - \omega_0 = \Delta_{HF} - \delta$ (see Fig. 3). In the $z$-quantization picture, such a field enhances interaction between the atom and the right-circularly polarized signal which is coupled to the moment $\sum_m m^2 \sigma_m(x)$, oscillating at frequency $2\Omega$. The Hamiltonian is most straightforwardly derived in the $x$-quantization. If the coupling field annihilation operator is $a_x$, we get after adiabatic elimination of the upper states and of the lower $F_2 = 3$ state the additional interaction Hamiltonian in the form

$$H_{AF}^{(2)} = i\hbar \sum_m G_m^2 a_x^\dagger a_x a_y^\dagger a_y + \sigma_m(z) + \hbar c.$$

Here the $y$-polarized fields $a_y^\dagger a_y$ have frequencies $\omega_y = \pm 2\Omega$ and the couplings are $G_m^{(2)} = E_0 E_{c0} M_m^{(4)}/(\hbar^4 \delta_{m+}^2 \Delta)$, where $M_m^{(4)}$ stands for the sum of all products of the dipole moment elements relevant in the transition from $|F, m\rangle$ to $|F, m+2\rangle$ via the intermediate states in the $P_{3/2}F'$, $S_{1/2}F_2$, and $P_{3/2}F'$ levels. For cesium with $F = 4$ we find $M_m^{(4)} = -\mu_0^4 \sqrt{(3-m)(4-m)(5+m)(6+m)}/48^2$. The Stark-shifted detunings $\delta_{m\pm}^\prime$ are

$$\delta_{m\pm}^\prime = \delta + \frac{\mu_0^2}{3\hbar^2 \Delta} (E_x^2 - E_c^2) + (2m + 2 \pm 1)\Omega,$$

where $E_x$ and $E_c$ are the electric intensities of the $x$-polarized field and of the coupling field. The second term and the hermitian conjugate of the first term of the Hamiltonian (4) with $m = -F$ are shown in Fig. 3a.

If $2\Omega \ll |\delta_{m\pm}^\prime|$, then the $G_{m\pm}^{(2)}$ couplings are almost equal, $G_{m+}^{(2)} \approx G_{m-}^{(2)}$, and Eq. (4) leads to a similar expression as Eq. (5), now coupling field quadratures at the $2\Omega$ sidebands to the atomic quadratures of the $\sigma_{m, m\pm2}$ coherences. This would enable us to apply the QND interaction similar to that in Fig. 2 in an additional channel. Even more interesting results are obtained if the detunings $\delta'$ are smaller than $\Omega$ and the couplings $G_{m\pm}^{(2)}$ for transitions $a_x a_x^\dagger a_y a_y^\dagger$ and $a_x^\dagger a_x a_y^\dagger a_y$ become very different. Let us assume that the atoms were initially prepared in state $|F, m = -F\rangle_x$ and let the coupling field be coherent with amplitude $\alpha F$ and the $a_x$ mode be coherent with real amplitude $\alpha_0$. If $|\delta_{m = -F, -}^\prime| \ll |\delta_{m = -F, +}^\prime|$, then the dominant term in Eq. (4) becomes

$$H_{AF}^{(2,SQ)} = i\hbar G_{SQ}^{(2)} a_x^\dagger a_x a_y a_y^\dagger \left( a_y^\dagger a_y - a_y a_y^\dagger \right),$$

where $G_{SQ}^{(2)} = G_{m = -F, -}^{(2)}$, and the atomic operators are $\sigma_{-} = \sigma_{-F, -F+2}$ and $\sigma_{+} = \sigma_{+F, -F+2}$. Operator $a_x^\dagger a_x$ acts as a two-mode squeezing operator, simultaneously creating or annihilating two quanta, one in the field $a_y^\dagger a_y$ and one in the atomic magnetic states. This would allow a direct preparation of entangled states between light and the atomic medium. If, on the other hand $|\delta_{m = -F, -}^\prime| \ll |\delta_{m = -F, +}^\prime|$, then the dominant term in Eq. (4) is

$$H_{AF}^{(2,BS)} = i\hbar G_{BS}^{(2)} a_x^\dagger a_x a_y a_y^\dagger \left( a_y^\dagger a_y + a_y a_y^\dagger \right),$$

where $G_{BS}^{(2)} = G_{m = -F, +}^{(2)}$. This Hamiltonian corresponds to a beam splitter which exchanges excitations between the field $a_y^\dagger a_y$ and the atomic magnetic states. This would be suitable for swapping quantum information between the field and the medium.

Let us define the annihilation operator of the effective atomic mode associated with the coherence between magnetic levels $m = -F$ and $m = -F + 2$, $a_{A2} = \sum_{x, k} \sigma_{A2}|x\rangle\langle x|$. The total Hamiltonian is obtained by summing the single-atom contributions over all $N_A$ atoms. For the squeezer-type Hamiltonian we obtain

$$H^{(2,SQ)}_{int} = i\hbar \kappa^{(2)} (a_{A2}^\dagger a_y^\dagger a_y - a_{A2} a_y),$$

and the beam splitter-type Hamiltonian is

$$H^{(2,BS)}_{int} = i\hbar \kappa^{(2)} (a_{A2}^\dagger a_y + a_{A2} a_y^\dagger).$$

The coupling constant can be expressed as $\kappa^{(2)} = E_0 E_{c0} \mu_0^2 \sqrt{7N_A N_c}/(576 \hbar^4 \delta^2 \Delta^2)$, and $N_c$ stands for...
the number of photons in the coupling field. Note that the Hamiltonians operating on the different sidebands approximately commute with each other (their commutators being $\sim N_A^{-1/2}$ times smaller than squares of the Hamiltonians themselves). This means that the processes in the different channels can run independently of each other so, e.g., an entangled state of two light modes can be stored in the medium and then be read out again.

The Hamiltonian $H_{\text{int}}^{(2,\text{BS})}$ enables us to straightforwardly store the quantum state of light into the atomic ensemble and to retrieve it later on. If $\kappa^{(2)} T = \pi/2$, where $T$ is the effective interaction time, then the quantum states of the light and atoms will be mutually exchanged. Note that the storage or retrieval would require only a single passage of the light beam through the atoms. Our method also does not involve any measurement followed by feedback and can achieve high fidelity without prior squeezing of atoms or light, in contrast to the protocol of Ref. [2].

We can see that the four-photon coupling increases with the intensity of the coupling field and with decreasing the detuning $\delta'$. It becomes comparable to the two-photon coupling if $\Omega^2 \approx 48\Delta' \sqrt{v T}$, where $\Omega = E_{2\alpha} \alpha \mu_0 / \hbar$ is the Rabi frequency of the coupling field. The magnitude of the detuning $\delta'$ is limited from below by Doppler broadening of the hyperfine frequency $\Delta_H$, $|\delta'| \gg \Delta_H v / c$, where $v$ is the RMS thermal speed of the atoms and $c$ is speed of light. Note that the much stronger Doppler shift of the single-photon transitions $\omega_0 v / c \sim 10^3 \text{s}^{-1}$ does not cause any problem here since the signal and coupling fields propagate in the same direction and their Doppler shifts subtract in the two-photon and four-photon transitions. This is the same trick which was used for achieving ultraslow group velocity in hot gases [13]. For cesium atoms used in the experiments [2, 3] the RMS thermal speed is $v \approx 100 \text{m/s}$ and the hyperfine splitting is $\Delta_H = 9.1 \text{GHz}$ so that $\Delta_H v / c \approx 3 \text{kHz}$. If we take $\delta \approx 30 \text{kHz}$ and the single-photon detuning as in [2, 3], $\Delta \approx 700 \text{MHz}$, we find $\Omega_\infty \approx 10^7 \text{s}^{-1}$. This value corresponds to the light intensity $\sim \text{mW/cm}^2$ which is of the same order of magnitude as the intensity of the $x$-polarized field used in the experiments. This suggests that the proposed method should work with present experimental setups.

In principle, by cascading multiphoton Raman transitions using additional intermediate levels, one should be able to realize Hamiltonians coupling higher sidebands with the corresponding atomic coherences. Another option could be going closer to resonance with one of the upper hyperfine levels $F'$. Such a scheme would be between the far off-resonant QND schemes and the resonant EIT schemes with the field coupled to multipole coherences [14], or the resonant scheme for selective addressing of higher polarization moments [15].

In conclusion, we have proposed a scheme for involving higher coherences of the atomic Zeeman sublevels by means of multiple Raman transitions to store quantum information carried by light. The scheme enables us to work with a broader class of Hamiltonians than those of the QND type and opens a way to involve higher amount of modes to be stored in parallel in the atomic media.

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