Photonic quantum memory using intra-atomic frequency comb

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Photonic quantum memory, such as atomic frequency comb (AFC), is essential to make photonic quantum computation and long distance quantum communication scalable and feasible. In standard AFC the frequency of different atoms must be stable relative to each other which presents difficulties in realizing the quantum memory. Here we propose a quantum memory using intra-atomic frequency comb which does not require frequency stabilization. We show that the transitions between two degenerate energy levels of a single atom can be used to construct the frequency comb. The spacing between the teeth of the comb is controlled by applying external magnetic field. Since the frequency comb is constructed from individual atoms, these atoms can be used alone or in ensembles to realize the quantum memory. Furthermore, the ensemble based quantum memory with intra-AFC is robust against Doppler broadening which makes it useful for high-temperature quantum memory. As an example, we numerically show the intra-AFC in the Cesium atoms and demonstrate a photon-echo which is essential for quantum memory.

Single photons are essential for long-distance quantum communication and linear optical quantum computation [11-10]. The probabilistic nature inherent in conventional single-photon sources hampers the scalable implementation of such protocols [11-16]. Quantum memory, a device which can store and reemit single photons on demand, can help overcome this problem and allows efficient photonic quantum technologies [17-20].

The basic idea behind a quantum memory is the light-matter interaction which allows the controlled reversible transfer of the quantum information between the photonic and the matter systems. Several protocols have been used to store single photons in atomic, condensed matter and superconducting systems [21-27]. Electromagnetically induced transparency [4, 28, 29], controlled reversible inhomogeneous broadening [23, 30-32] and the atomic frequency comb (AFC) [33-39] are the most used quantum memories in the atomic ensembles. In all these quantum memories the incoming photon interacts with a carefully designed spectrum of the atomic ensemble. By controlling the shape and the characteristics of the spectrum one can store and retrieve photons from the ensemble.

Implementing AFC in an ensemble (in the usual way) relies on the frequencies of different atoms being stable relative to each other. This can be achieved in solids to some reasonable approximation at sufficiently low temperature. However, in gases Doppler broadening is a major limiting factor. To overcome this problem, in this manuscript we propose intra-atomic frequency comb (I-AFC). We show that the frequency comb (FC) can be constructed by putting together multiple transitions between the hyperfine levels of a single atom. The spacing between the different transition frequencies is controlled by external magnetic field. In order to show that such systems can be used for photonic quantum memories first we show the photon-echo from an ensemble of such atoms. To achieve long storage time one can transfer the excitation from the excited state space to spin state space by applying a $\pi$ pulse. As an example, we numerically show the FC in the Cesium (Cs) atoms. We show that an ensemble of Cs atoms with this FC results in a prominent photon-echo which is robust against the Doppler broadening.

The I-AFC can be used alone or in ensemble to realize efficient quantum memories. Since the FC is constructed from different transitions of individual atoms, the I-AFC in ensemble is also robust against environmental effects such as Doppler broadening, which makes it useful for high-temperature quantum memory. Another important advantage of the proposed scheme is that the spacing between different teeth of the FC is controlled by the external magnetic field. Therefore, the finesse of the comb can be controlled by varying the magnetic field which can influence the efficiency and the photon-echo time of the quantum memory.

We start by introducing the conventional AFC. The AFCs are typically consist of rare-earth ions doped crystals that have optical transition between the ground state $|g\rangle$ and the excited state $|e\rangle$ [33-39]. This transition has a narrow homogeneous bandwidth $\gamma$ and a large inhomogeneous bandwidth $\Gamma_{in}$ ($\Gamma_{in} \gg \gamma$). The transition $|g\rangle-|e\rangle$ is spectrally shaped such that the atomic density function consists of a series of equispaced narrow peaks (teeth), with spacing $\Delta$, resulting in a comb like structure in frequency modes, Fig. 1(a).

A single photon with spectral width $\tau^{-1} = \tilde{\omega} \gg \Delta$ is absorbed in the AFC system at time $t = 0$, which is stored as a collective excitation delocalized over all the teeth in the system. Formally, the state of the AFC, after
absorbing a single photon, can be written as

$$|\Psi_{\text{AFC}}\rangle = \sum_{j=1}^{M} \left( c_j e^{i \delta_j t} |\{e_j\}\rangle \prod_{k \neq j} |\{g_k\}\rangle \right) .$$  (1)

Here $|\{g_j\}\rangle \equiv |g_1 g_2 \cdots g_{N_j}\rangle$ and $|\{e_j\}\rangle \equiv \sum_n \alpha_n |g_1 \cdots e_n \cdots g_{N_j}\rangle$ represent the ground and collective single-excitation state of all the atoms with detuning $\delta_j$, respectively, and the $c_j$'s represent the absorption coefficient of each tooth in the comb. The coefficients $\alpha_n$ characterize the absorption by individual atoms.

The photon emission probability $P(t)$ from this setup is proportional to

$$P(t) \propto |\langle G | S_- |\Psi_{\text{AFC}}\rangle|^2 ,$$  (2)

where $S_- = \sum_j |\{g_j\}\rangle \langle \{e_j\}|$ is the step down operator and $|G\rangle = \prod_k |\{g_k\}\rangle$ is the collective ground state of the ensemble. Here the sum is over all the teeth. It can be seen from the state (1) and the expression for the photon emission probability $P(t)$ (2) that the probability is maximum at times $t$ which are integer multiples of $2\pi/\Delta$ [Fig. 1(a)]. The unabsorbed light comes out at $t = 0$ whereas at time $2n\pi/\Delta$ we get the $n$-th photon-echo.

The efficiency $\eta$ of the quantum memory in this protocol is defined as the ratio of the total amount of light coming out in the first echo (at time $t = 2\pi/\Delta$) and the total input intensity, i.e.,

$$\eta = \frac{\int_{2\pi/\Delta}^{3\pi/\Delta} |E_{\text{out}}(t)|^2 dt}{\int |E_{\text{in}}(t)|^2 dt} .$$  (3)

Theoretically, the maximum efficiency that can be achieved in the forward propagation is 54% which can be 100% in the backward propagation.

As mentioned earlier, the usual ways of implementing AFC require the frequencies of different atoms in the ensemble to be stable relative to each other which is often difficult to achieve in gaseous ensemble. In the following, we present a model where the FC is constructed from individual atoms which does not have the above said problems.

Consider an atom with a single ground state and $N$ number of degenerate excited states (Fig. 1(b)). We lift the degeneracy in the excited state space by applying external magnetic field (Zeeman effect). The excited states are chosen such that the transition is possible only between the ground state and the excited states. The mean transition frequency is $\omega_L$, and the spacing between different transitions is $\Delta$; hence, the transition spectrum for this atom will resemble an AFC. The question is whether a photon-echo can be observed in this atom in order to be eligible for quantum memory.

Consider the Hamiltonian of this atom in the presence of electromagnetic field (see Appendix for details)

$$H = \sum_{n=1}^{N} \hbar \delta_n |n\rangle\langle n| - \frac{\mathcal{E}(z,t)}{2} \sum_{n=1}^{N} (|n\rangle\langle 0| + |0\rangle\langle n|)$$  (4)

where $\delta_n$ is the detuning between the mean frequency $\omega_L$ of light and the frequency of the $n$-th excited state $|n\rangle$, $|0\rangle$ is the ground state and $d$ is the magnitude of the transition dipole operator which we have taken to be same for all the transitions. $\mathcal{E}(z,t)$ represents the amplitude of the electric field.
If the state of the atom at time $t$ is given as the superposition of all the states, i.e., $|\psi(t)\rangle = \sum_{n=0}^{N} c_n(t) |n\rangle$, then the dynamical equation in terms of the coefficients $c_n(t)$ reads

$$\dot{c}_n = \frac{i\hbar}{2} \mathcal{E}(z,t) c_0 - i\delta_n c_n, \quad \dot{c}_0 = \frac{i\hbar}{2} \mathcal{E}(z,t) \sum_{n=1}^{N} c_n(t). \quad (5)$$

Often the interaction between the photon and the atom is weak. Hence, the probability of finding the atom in the ground state is almost unity, i.e., $|c_0|^2 \simeq 1$. Therefore,

$$c_n(t) = \frac{i\hbar}{2} e^{-i\delta_n t} \int_{-\infty}^{t} e^{i\delta_n t'} \mathcal{E}(z,t') \, dt'. \quad (6)$$

For a Gaussian input, i.e., electric field of the form $\mathcal{E}(z,t) = \mathcal{E}_0 \exp(-t^2/2\tau^2)$ with the temporal width $\tau \ll t$ Eq. (6) yields

$$c_n(t) = \frac{i\Omega}{2} \sqrt{\frac{\pi}{\omega}} \exp\left[-(\delta_n \tau)^2/2 - i\delta_n t\right],$$

where $\Omega = \frac{d\mathcal{E}_0}{\hbar}$. Since we have assumed equispaced excited states we can write $\delta_n = \delta_0 + n\Delta$ for some constant $\delta_0$. The photon emission probability $P(t)$ (for forward propagation) for this atomic system can be redefined as

$$P(t) = |\langle 0 \mid s_- | \psi(t) \rangle|^2 = \left| \frac{i\Omega}{2} \sqrt{\frac{\pi}{\omega}} \sum_{n=1}^{N} e^{-i\eta t} \right|^2, \quad (7)$$

where $s_- = \sum_n |0\rangle \langle n|$ and the sum is over all the states in the excited level. Here we have assumed $\omega = t^{-1} \gg \Delta$ which implies that $\exp[-(n\Delta)^2/2\omega^2] \rightarrow 1$. Clearly, Eq. (7) results in a photon-echo at $t = 2\pi/\Delta$ when the probability of the photon emission is maximum.

This calculation confirms that an ideal 1-AFC is capable of demonstrating photon-echo, which can ultimately be used as a quantum memory. However, in a real atom the situation can be slightly different. Generally, atoms have a number of degenerate states in both the ground and the excited levels [40,41]. The transitions are allowed only between certain combinations of ground and excited states with non-uniform transition amplitudes (Fig. 1[c]). Fortunately, by choosing external magnetic field strength appropriately the transition frequencies can be restricted around some mean frequency $\omega_L$. If the input photon has mean frequency close to $\omega_L$ and spectral width large enough to cover all these transition, then the photon can be absorbed in the atom as a delocalized excitation over the excited level.

For example, consider the Cs atom. Here 16-fold degenerate $5p^66s$ energy level is chosen as the ground level and 32-fold degenerate $5p^68p$ level constitute the excited state. The mean transition frequency between the two levels is 773.21THz [42,43]. The natural line-width of all the allowed transitions is taken to be equal (5MHz) [42]. The degeneracy in the ground-levels and the excited-levels is lifted by applying an external magnetic field of 0.1T.

In Figs. 2(a) and 2(b) we calculate the most prominent absorption amplitudes and the optical depths between the chosen energy levels for $\Delta m = \pm 1$ transitions for an ensemble of Cs atoms in the vapour form with density $10^{18}$m$^{-3}$ and the vapour cloud cell of length 5cm (see appendix). The spectrum in these figures resemble the AFC but with non-uniform spacing between the teeth and with unequal heights.

In order to observe a photon-echo, initially all the Cs atoms are prepared in an equal superposition of all the ground states. Upon absorption of a single photon the atom will acquire the state which is a superposition of different excited states. In Figs. 2(c) and 2(d) we show numerically the photon emission probability $P(t)$ for the forward propagation. The first peak at $t = 2$ns in both the plots is the emission probability of the unabsorbed photon. The second peak is the photon echo which occurs 3ns after the input photon.

In Fig. 3 we plot the photon-echo efficiency $\eta$ as a function of the cell length ($L$). We see that the efficiency increases as we increase the cell length, reaching
A maximum value at a critical length $L_\text{c}$ and then starts to fall. Also, the $L_\text{c}$ for $\Delta m = \pm 1$ is different and the maximum efficiency we get is $\eta \sim 35\%$ under the given circumstances. Note that the maximum efficiency in this case is limited only by the non-ideal form of the FC. One may achieve optimal efficiency by choosing the initial atomic state appropriately (in order to compensate for the non-uniform FC) and by choosing appropriate values of the magnetic field.

Note that both the ground states and the excited states have FC like structure which might create complications; however, we find that the FC in the ground state does not affect the photon-echo (see appendix). Furthermore, the degenerate energy level $5p^65d$ for Cs atom can serve as the long-lived spin state where the photon can be stored (in principle) indefinitely.

So far we have shown that an I-AFC can result in photon-echo. Our results are supported by numerical calculations for Cs atoms. Next we show that the I-AFC is robust against environmental effect, particularly, Doppler broadening.

In an ensemble of Cs atoms in a thermal equilibrium at temperature $T$ the state of each of the atoms is $|v\rangle \otimes |\psi_v\rangle$ where $|v\rangle$ is the kinetic state of the atom and $|\psi_v\rangle$ is its electronic state labeled by the velocity $v$. The total state of the atomic ensemble can be written as

$$\rho = \int dv \, p(v) \, |v\rangle \langle v| \otimes |\psi_v\rangle \langle \psi_v|,$$

where $p(v) = \exp(-mv^2/2k_B T)$ is the thermal distribution with $m$ being the mass of the atom and $k_B$ being the Boltzman constant. The motion of the atom makes it perceive the frequency of the incoming photon shifted from $\omega_L$ to $\omega_L + v \cdot k$ where $k$ is the wave vector of the photon. Or, it can be perceived as a shift of $-v \cdot k$ frequency in the FC, which would yield an additional phase $\exp(i v \cdot k z/c)$ in the state, i.e., $|\psi_v(t)\rangle \rightarrow e^{i v \cdot k z/c} |\psi_v(t)\rangle$. The photon-emission probability $P(t)$ can be written as (see appendix)

$$P(t) = \int v \, dv \, p(v) \, |G\rangle \, s - |\psi_v(t)\rangle |^2,$$

which is independent of the phase $\exp(i v \cdot k z/c)$. Here $|G\rangle$ is the collective ground state. Therefore, the photon-echo is the thermal average of the echos from all the atoms having different velocities. Since the velocity of the atom causes the whole spectrum to move by a factor $v \cdot k$ the echo appears at the same time for all the velocities. Therefore, the thermal average also results in the photon-echo to appear at exactly the same time. Hence, Doppler broadening has no effect on the I-AFC. In Fig. 4 we plot the output from the Cs vapours at 100K. We can see a clear photon-echo at exactly 3ns after the input.

To conclude, we have proposed an I-AFC which is constructed using different transitions of a single atom. This AFC is important for trapped-ions and on-chip quantum computation, single-atom quantum memory and microwave-to-optical transducers. Since the I-AFC is constructed from individual atoms, this FC (in atomic ensembles) is immune against Doppler broadening; hence can be used for high-temperature photonic quantum memory. The spacing between the neighboring teeth in the comb which characterize the time of the photon-echo is controlled by external magnetic field. Therefore, the time of the photon-echo can be controlled by tuning the magnetic field. We considered the example of Cs atom and showed that I-AFC can be realized in Cs atoms.

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Multi-level atom: single ground state and multiple excited states

Here we solve for the dynamics of an atom containing a single ground state and $N$ number of excited states interacting with oscillating electric field $E$. The Hamiltonian for such an atom can be written as \[ H = \sum_{n=1}^{N} \hbar \omega_n |n\rangle\langle n| - \frac{\mathcal{E}(z,t)}{2} \sum_{n=1}^{N} (d_{n0} |n\rangle \langle 0| + d_{n0}^* |0\rangle \langle n|) e^{-i \omega_L t}, \] (10)

where $|n\rangle$ represents the $n$-th excited state corresponding to energy $\hbar \omega_n$, $|0\rangle$ being the ground state with zero energy, and $d_{n0}$ is the transition dipole moment between $|0\rangle$ and $|n\rangle$. Here we are assuming that the transition between the different excited states is forbidden. Therefore, $d_{nn'} = 0$ for $n, n' \neq 0$. We can write the oscillating electric field as

\[ E(z,t) = \mathcal{E}(z,t) \cos(\omega_L t - kz), \] (11)

where $\mathcal{E}(z,t)$ gives the temporal shape of the electric field and $\omega_L$ is the mean frequency of light.

The Hamiltonian $H$ in the interaction picture can be calculated as $H_I = e^{i H_0 t} H e^{-i H_0 t} - H_0$ using $H_0 = \sum_{n=1}^{N} \hbar \omega_n |n\rangle\langle n|$. The Hamiltonian $H_I$ reads

\[ H_I = \sum_{n=1}^{N} \hbar \delta_n |n\rangle\langle n| - \frac{\mathcal{E}(z,t)}{2} \sum_{n=1}^{N} (d_{n0} |n\rangle \langle 0| + d_{n0}^* |0\rangle \langle n|), \] (12)

where $\delta_n = \omega_n - \omega_L$ is the detuning between the $n$-th energy level and the mean frequency of the laser light.

The state of the atomic system is represented by the density operator $\rho$ where $\rho_{nn}$ is the population of the $n$-th energy level and $\rho_{nn'}$ represent the coherence. In our system we consider that the number of photons are much smaller than the number of atoms in the ensemble. Therefore, $\rho_{00} \simeq 1$ which would imply that $\rho_{nn} \simeq \rho_{nn'} \simeq 0$ but $\rho_{00} \neq 0$. With these approximations we can write the dynamical equation for the coherence terms as

\[ \frac{\partial \rho_{n0}(z,t)}{\partial t} + i \left( \frac{\delta_n}{2} + \gamma \right) \rho_{n0}(z,t) = \frac{i \hbar d_{n0}}{2} \mathcal{E}(z,t), \] (13)

where the term with $\gamma$ is added phenomenologically to incorporate the environmental effects.

The Eq. (13) characterize the dynamics of the atomic state when it interacts with external electromagnetic field. In order to solve for the (forward) propagation of light through the ensemble of atoms we need to consider the effect of the atomic polarization on the electromagnetic field as well which can be written as

\[ \left( \frac{\partial}{\partial z} + \frac{1}{c} \frac{\partial}{\partial t} \right) \mathcal{E}(z,t) = \frac{i \omega_L}{2 \epsilon_0 c} \mathcal{P}(z,t). \] (14)

Here $\mathcal{P}(z,t)$ is the amplitudes of the atomic polarization which is defined as the expectation value of the aggregate transition electric dipole moment operator $\mathbf{d}$ of the entire ensemble,

\[ P(z,t) = \mathcal{N} \text{Tr} [\rho \mathbf{d}] = \mathcal{N} \left( \sum_{n=1}^{N} d_{n0}^* \rho_{n0} e^{-i(\omega_L t - kz)} + \sum_{n=1}^{N} d_{n0} \rho_{n0}^* e^{i(\omega_L t - kz)} \right), \] (15)

where $\mathcal{N}$ is the density of atoms. The atomic polarization is driven by the external electric field. Therefore, the time dependence of the polarization and the electric field is the same. Hence, we can write

\[ P(z,t) = \frac{1}{2} \mathcal{P}(z,t) e^{-i(\omega_L t - kz)} + cc. \] (16)

Comparing Eq. (16) with Eq. (15) results in

\[ \mathcal{P}(z,t) = 2 \mathcal{N} \sum_{n=1}^{N} d_{n0}^* \rho_{n0}. \] (17)
Eqs. (13) and (14) together describe the propagation of light through an atomic ensemble [32, 45]. We are interested in finding the state of light at time \( t \) and position \( z \) which requires solving these two equations simultaneously. We can obtain the solution by taking the Fourier transform of \( \rho \), \( P \) and \( E \) in the time domain. This would yield
\[
\hat{\rho}_{n0}(z, \omega) = \frac{i d_{n0}}{2\hbar} \frac{\hat{E}(z, \omega)}{i(\omega + \delta_n) + (\gamma/2)}, \\
\hat{P}(z, \omega) = 2N \sum_n \frac{|d_{nm}|^2}{2\hbar} \frac{\hat{E}(z, \omega)}{\gamma/2 + i(\delta_n + \omega)},
\]
and propagation equation for \( \hat{E}(z, \omega) \)
\[
\left( \frac{\partial}{\partial z} + \frac{i\omega}{c} \right) \hat{E}(z, \omega) = \frac{i\omega_L}{2\epsilon_0 c} \hat{P}(z, \omega).
\]
From here it is straightforward to solve for \( \hat{E}(z, \omega) \) which reads
\[
\hat{E}(z, \omega) = \hat{E}(0, \omega)e^{-Dz}
\]
where \( D \) and \( g \) are defined as
\[
D = \sum_n \frac{g}{\gamma/2 + i(\delta_n + \omega)} + \frac{i\omega}{c}, \quad g = N^2 |d_{nm}|^2 \omega_L/2\epsilon_0 c.
\]
Clearly the first term in \( D \) is a comb where the Lorentzian distributions in \( \omega \) are placed at \( \delta_n \). The second term results in a position dependent phase where \( \omega/c = k \). Inverse Fourier transform of \( \hat{E}(z, \omega) \) gives the output in the time domain.

Multi-level atom: multiple ground and excited states

Here we solve for the dynamics of an atom containing \( N_g \) number of ground state and \( N_e \) number of excited states interacting with oscillating electric field \( E \). The Hamiltonian for such an atom reads
\[
H = \sum_{n=1}^{N_g} \hbar \omega_n^e |e_n\rangle\langle e_n| + \sum_{m=1}^{N_e} \hbar \omega_m^g |g_m\rangle\langle g_m| - \frac{\mathcal{E}(z,t)}{2} \left( \sum_{n,m} d_{nm} |e_n\rangle\langle g_m| e^{-i\omega_L t} + H.C \right)
\]
where \( |e_n\rangle \) (\( |g_m\rangle \)) represents the \( n \) (\( m \))-th excited (ground) state corresponding to energy \( \hbar \omega_n^e \) (\( \hbar \omega_m^g \)), \( |g_i\rangle \) being the ground state with zero energy, and \( d_{nm} \) is the transition dipole moment between \( |e_n\rangle \) and \( |g_m\rangle \). Here we are assuming that the transition among the ground states and among the excited states is forbidden. Therefore, \( d_{nn'} = d_{mm'} = 0 \).

The Hamiltonian for this atom in the interaction picture can be calculated as \( H_I = e^{itH_0}He^{-itH_0} - H_0 \) using \( H_0 = \sum_{n=1}^{N_g} \hbar \omega_n^e |e_n\rangle\langle e_n| \). The Hamiltonian \( H_I \) reads
\[
H_I = \sum_{n=1}^{N_g} \hbar(\omega_n^e - \omega_L) |e_n\rangle\langle e_n| + \sum_{n=1}^{N_e} \hbar \omega_m^g |g_n\rangle\langle g_n| - \frac{\mathcal{E}(z,t)}{2} \left( \sum_{n,m} d_{nm} |e_n\rangle\langle g_m| + H.C \right)
\]
Similar to the previous case of single ground state and multiple excited states, we can assume that the population in the ground state is much smaller than the excited states, i.e., \( \sum_m \rho_{nm} \simeq 1 \) and \( \rho_{nn} \simeq \rho_{nm'} \simeq \rho_{mm'} \simeq 0 \) but \( \rho_{nm} \neq 0 \). This yields the dynamical equation for the coherence
\[
\frac{\partial \rho_{nm}(z,t)}{\partial t} + \left( i\delta_{nm} + \frac{\gamma}{2} \right) \rho_{nm}(z,t) = \frac{id_{nm}}{2\hbar} \rho_{mm} \mathcal{E}(z,t).
\]
where \( \delta_{nm} = [\omega_n^e - \omega_m^g] - \omega_L \) is the detuning between the transition \( |e_n\rangle \leftrightarrow |g_m\rangle \) and the mean frequency of the laser light.
Using the definition for the atomic polarization
\[ P(z, t) = 2N \sum_{n,m} d^*_{nm} \rho_{nm}, \quad (26) \]
we can arrive at the equations
\[ \rho_{nm}(z, \omega) = i \frac{d_{nm} \rho_{mm}}{2\hbar} \frac{\tilde{E}(z, \omega)}{\gamma/2 + i(\delta_{nm} + \omega)}, \quad (27) \]
\[ P(z, \omega) = 2N \sum_{n,m} i |d_{nm}|^2 \rho_{mm} \frac{\tilde{E}(z, \omega)}{2\hbar} \frac{\gamma/2 + i(\delta_{nm} + \omega)}, \quad (28) \]
and propagation equation for \( \tilde{E}(z, \omega) \)
\[ \tilde{E}(z, \omega) = \tilde{E}(0, \omega) e^{-Dz}, \quad (29) \]
where \( D \) and \( g \) are defined as
\[ D = \sum_{n,m} g_{nm} \frac{\gamma}{\gamma + 1(\delta_{nm} + \omega)} + \frac{i\omega}{c}, \quad g_{nm} = N |d_{nm}|^2 \rho_{mm} \omega_L, \quad (30) \]
Clearly, \( D \) in this case is identical to the one we obtained for the case of single ground state. Mathematically both the case are the same hence we get an echo in these generalized atoms.

**Effect of Doppler broadening on the output electric field**

In this section we consider the effect of Doppler broadening on the output light. An atomic ensemble at temperature \( T \) contains \( p_v \) fraction of atoms moving with velocity \( v \) where \( p_v \) has the form of thermal distributions, i.e., \( p_v = \exp(-mv^2/2k_BT) \). Here \( m \) is the mass of the atom and \( k_B \) is the Boltzman constant. An atom with velocity \( v \) and wave vector \( k \) experience the modified frequency of the incoming light from \( \omega_L \) to \( \omega_L + k \cdot v \). This would modify the detuning \( \delta \) to \( \delta + k \cdot v + \omega \) which shows in \( D \) in Eq. (22) as
\[ D_v = \sum_n \frac{g_n}{\gamma + i(\delta_n + k \cdot v + \omega)} + \frac{i\omega}{c}. \quad (30) \]
From this modified \( D_v \) we can calculate the output electric field
\[ \tilde{E}(z, \omega, v) = \tilde{E}(0, \omega) e^{-D_v z}. \quad (31) \]
Hence the light emitted from an atom moving with velocity \( v \) is \( \tilde{E}(z, \omega, v) \) which is different from \( \tilde{E}(z, \omega, v = 0) \) by only an over all phase which can be calculated by replacing \( \omega \rightarrow \omega' = k \cdot v + \omega \). Thus the additional phase the \( \tilde{E}(z, \omega, v) \) gets is \( \exp\{i k \cdot vz/c \} \). Since, the velocity distribution is incoherent over the atomic ensemble, we need to add the intensity of light coming from each atom which is independent of the velocity. Thus, the intensity of light in the photon-echo is unchanged due to the Doppler effect.

**Transitions in Cs atom**

In this section we calculate the transitions between the states \( 5p^6 6s \) (degenerate ground state) and \( 5p^6 8p \) (degenerate excited state). The Hamiltonian of the atom in the presence of an external magnetic field can be written as
\[ H = H_0 + H_{\text{hfs}} + H_{\mu} \quad (32) \]
where \( H_0 \) is the unperturbed spin-less Hamiltonian plus the fine-structure Hamiltonian of the atom and \( H' = H_{\text{hfs}} + H_{\mu} \) is the perturbation. The Hamiltonian \( H_{\text{hfs}} = A(I \cdot J) \) is the hyperfine correction and \( H_{\mu} = -(\hat{\mu}_j + \hat{\mu}_l) \cdot B = (g_j \mu_B J_z - g_l \mu_N I_z) B \) is the interaction of the atom with the magnetic field. Here \( A \) is the hyperfine structure
constant, $I$ and $J$ are nuclear spin operator and total (spin and orbital angular momentum) operator, and $\mu_B$ and $\mu_N$ are the Bohr and nuclear magneton. $g_J$ and $g_I$ are given as

$$g_J = 1 + \frac{J(J+1) + S(S+1) - L(L+1)}{2J(J+1)} ,$$

$$g_I = 0.7369$$

for a Cesium atom.

We are interested in only the states $5p^66s$ and $5p^68p$ which are the eigenstates of the Hamiltonian $H_0$. The energy difference between these states is $773.20996\text{THz}$ which lies in the optical frequency range $[42, 43]$. The $5p^66s$ states which we call ground states have 16-fold degeneracy and the $5p^68p$ energy level (excited states) has 32-fold degeneracy.

The perturbation $H'$ helps lift the degeneracy in these states which we exploit for AFC purpose.

To construct the AFC in the Cs atom we need to find the absorption spectrum of the transitions between $5p^66s$ and $5p^68p$ in the presence of the perturbative corrections $H'$. For that purpose we write the Hamiltonian $H = H_0 + H'$ in the basis $| F, M_F, I, J \rangle$ which is the simultaneous eigenbasis of $I, J, F$ and $F_z$. Here $F = I + J$. We numerically solve the Hamiltonian $H$ to find the eigenvectors and the eigenvalues.

The absorption amplitudes are calculated from the imaginary part of electric susceptibility $\chi_e$ where the atomic polarization $P = \varepsilon_0 \chi_e E$ \cite{19}. Therefore the absorption coefficients reads

$$\text{Im}\chi_e = \frac{N}{\hbar \varepsilon_0} \sum_n \frac{|d_{n_e n_0}|^2 \gamma}{\gamma^2 + (\delta_n + \omega)^2} .$$

Optical depth $\alpha$ is defined as

$$\alpha = \frac{N|d|^2 \omega L}{2\hbar \varepsilon_0 c \gamma} .$$

Here $L$ is the length of the cell which we have taken to be 5cm, $\gamma = 5\text{MHz}$ is the decay rate of the Cs transition at 100k temperature \cite{42}. $N$ is the number density of the Cs atoms in the ensemble which is taken to be $10^{18} \text{m}^{-3}$. The magnetic field strength is taken to be 0.1T. The dipole matrix elements $d_{n_e n_0} = \langle n_e | d | n_0 \rangle$, where $|n_e\rangle$ is one of the excited states and $|n_0\rangle$ is one of the ground states which can easily be calculated numerically. In Figs. 5(a) and 5(b) we show the absorption coefficients and the optical depths for the $\Delta m = \pm 1$ transitions.