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Optical quantum memory with generalized time-reversible atom–light interaction

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Abstract. We examine a quantum memory scheme based on controlled de- and rephasing of atomic coherence of a nonresonant, inhomogeneously broadened Raman transition. We show that it generalizes the physical conditions for time-reversible interaction between light and atomic ensembles in the case of strong fields and nonlinear interactions. Furthermore, assuming weak input fields, we develop a unified framework for realizations exploiting either controlled reversible inhomogeneous broadening or atomic frequency combs, and discuss new aspects of the storage and manipulation of quantum states.

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1. Introduction

The study of time-reversible evolution in unitary dynamics was central to the development of thermodynamics and statistical mechanics [1] and CPT symmetry [2]. Furthermore, reversible interaction underpins reversible transfer of quantum states between light and atoms, i.e. quantum memory (QM) [3, 4], which is key for quantum repeaters [5, 6] and all-optical quantum computing [7].

Approaches to QM exploit atoms in cavities [8], nonresonant Raman transitions [9–11], electromagnetically induced transparency (EIT) [12–18] or photon-echo techniques [19–34]. The latter is of particular interest in the present context as it is the first in which time-reversible dynamics between light and atomic ensembles could be observed from the equations of motion alone [23]. Similar considerations in the limit of weak fields led later to an improved efficiency of EIT-based storage [16].

Here, we generalize the physical conditions allowing time-reversal symmetry in the mapping of quantum states between light and atomic ensembles to fields of arbitrary strength and nonlinear interactions beyond those considered in [19, 21, 23]. The scheme exploits reversible dephasing of the atomic coherence of a nonresonant, inhomogeneously broadened Raman transition (Raman echo quantum memory (REQM)). A simplified version of REQM has been proposed[4], further developed in [36, 37] and demonstrated in [32, 33]. We also compare the conditions for time-reversible dynamics for the storage of strong fields with those in the case of weak fields, which naturally leads to a unified framework for realizations of REQM based on controlled reversible inhomogeneous broadening (CRIB) or atomic frequency combs (AFC) [3, 25]. Our findings shed new light on time reversibility in the interaction between light and inhomogeneously broadened atomic ensembles, and also pave the way for the storage of macroscopic light fields in nano-sized atomic media. Finally, we discuss, with the example of frequency conversion, how REQM enables controlled manipulation of quantum light fields.

2. The scheme

Energy and temporal diagrams of the interaction scheme are depicted in figure 1. At time $t = 0$, the probe light field $\hat{E}_1(t, z) = \hat{A}_1(t, z)\exp\{ik_1 z\}$ with duration δt_1, carrier frequency ω_1, wave vector k_1 and spectral width $\delta\omega_1 = \delta t_1^{-1}$ enters the medium with three-level atoms (labeled by j) prepared in the long-lived level $|1\rangle = \prod_{j=1}^{N} |1\rangle_j$ along the $+z$-direction. The atoms are simultaneously exposed to an intense control (writing) field propagating along wave vector $\vec{K}_1$ with carrier frequency ω_1^c and resonant Rabi frequency $\tilde{\Omega}_1(t, \vec{r}) = \Omega_1(t)\exp\{-i(\omega_1^c t - \vec{K}_1\vec{r})\}$. It is reduced to zero after absorption of the probe field. The probe and writing fields are assumed to be in Raman resonance $\omega_1 - \omega_1^c \approx \omega_{21}$ with sufficiently large spectral detuning $\Delta_1 = \omega_{31} - \omega_1$ from the $1 \leftrightarrow 3$ transition to avoid populating level 3.

We take the $1 \leftrightarrow 2$, $1 \leftrightarrow 3$ and $2 \leftrightarrow 3$ transitions to feature inhomogeneous broadenings (IBs). Furthermore, we assume $|\Delta_1 + \Delta_{31}^j| \gg \delta\omega$ (with arbitrary Δ_1/Δ_{31}^j), $|\Delta_{21}^j| \ll |\Delta_{31}^j|, |\Delta_{32}^j|$ and $\frac{1}{\Delta_1+\Delta_{32}^j} \cong \frac{1}{\Delta_1+\Delta_{31}^j}$. The latter relates the detuning of each individual atom, j, from the center of the $1 \leftrightarrow 3$ transition (Δ_{31}^j) with its detuning from the center of the $2 \leftrightarrow 3$ transition (Δ_{32}^j). This allows us to express Δ_{32}^j in terms of Δ_{31}^j, which keeps formulae compact. The IB, together

[4] The idea of generalizing CRIB has been proposed independently in [35].

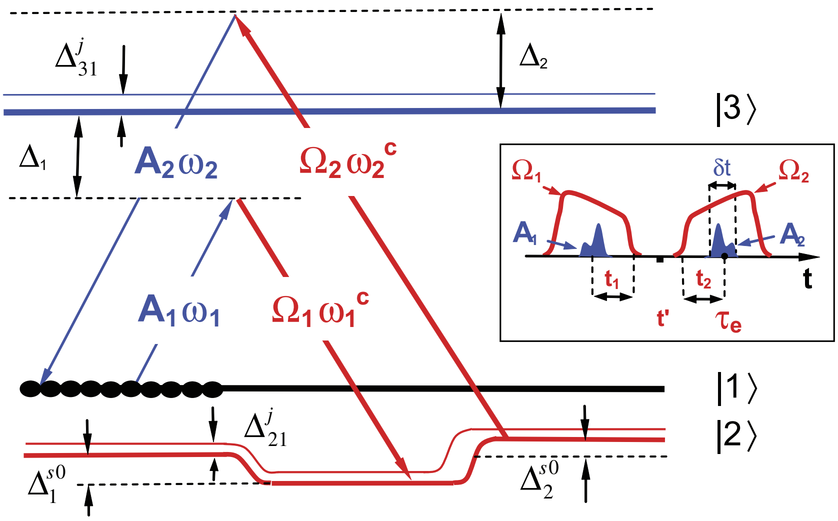

Figure 1. Energy level diagram showing atomic transitions, probe and echo fields (A_ν) with carrier frequencies ω_ν, and writing and reading fields with Rabi frequencies Ω_ν and carrier frequencies ω_ν^c. Also depicted is a temporal diagram of all light fields.

with the ac Stark shift $\Delta_1^s = \frac{\Omega_1^2}{\Delta_1+\Delta_{31}^j}$ introduced by the control field, results in a total two-photon detuning of each atom j from the Raman transition given by

$$\Delta_{R,\mathrm{tot}}^j(t) = \Delta_{21,\mathrm{tot}}^j - \Delta_1^s \cong \Delta_{21,\mathrm{tot}}^j + \Delta_{31,\mathrm{tot}}^j f_1(t) - \Delta_1^{s0}$$

with

$$\Delta_{k1,\mathrm{tot}}^j = \Delta_{k1,\mathrm{nat}}^j + \Delta_{k1,\mathrm{cont}}^j \equiv \Delta_{k1}^j.$$

Here, $k = 2, 3$ and Δ_1^{s0} is the Stark shift for atoms featuring zero detuning from the $1 \leftrightarrow 3$ transition ($\Delta_{31}^j = 0$): $\Delta_1^{s0} = \Omega_1^2/\Delta$. The indices refer to total and natural detuning, and detuning induced in a controlled way using e.g. external electric [24] or magnetic [32] fields. We denote the IBs $\Delta_{31,\mathrm{nat}}$, $\Delta_{31,\mathrm{cont}}$, $\Delta_{31,\mathrm{tot}} \equiv \Delta_{31}$, etc. We take the IB of the Raman transition to be large enough to absorb all spectral components of the probe, and assume the natural IB on the $1 \leftrightarrow 2$ transition to be negligible [16, 28]. $f_1(t)$ is related to the Rabi frequency of the write field, $f_1(t) = |\Omega_1(t)|^2/\Delta_1^2$, and to the Stark shift Δ_1^{s0}, $f_1(t) = \Delta_1^{s0}/\Delta_1$. It can in principle exceed unity.

The atom–light interaction leads to excitation of atomic (Raman) coherence, which rapidly decays due to IB. To retrieve the stored field, we apply a phase-matching operation, and launch at time $t > t'$ a second control (reading) pulse propagating roughly in the $-z$-direction and having wave vector, carrier and Rabi frequencies $\vec{K}_2$, ω_2^c and $\tilde{\Omega}_2(t,\vec{r}) = \Omega_2(t)\exp\{-\mathrm{i}(\omega_2^c t - \vec{K}_2\vec{r})\}$, respectively (see figure 1). In addition, we either actively invert the Raman broadening, as in CRIB [25] (now RECRIB), or, in the case of a generalization of AFC [26] (now REAFC), simply wait until the atomic coherence automatically rephases. The probe field, at frequency $\omega_2 \approx \omega_{21} + \omega_2^c$, will then be re-emitted at time τ_e as an echo in the backward $(-z)$-direction. We emphasize that IB on the optical transition is no impediment to time-reversible atom–light evolution. In the following, we limit our discussion to the case where the spectral width and duration of the echo are equal to those of the probe field: $\delta\omega_2 = \delta\omega_1 \equiv \delta\omega$, $\delta t_2 = \delta t_1 \equiv \delta t$.

3. Basic equations

We describe the interaction between N three-level atoms, probe and echo fields $\hat{A}_\nu(z,t)$, and control fields $\Omega_\nu(t)$ using the Hamiltonian

$$\hat{H} = \hat{H}_{\rm a} + \hat{H}_{\rm f} + \hat{V}_{\rm a-f} + \hat{V}_{\rm c}.$$

The indices a, f, a–f and c denote the Hamiltonian for the atoms, the quantum light fields and the interaction between the atoms and the quantum and classical light fields, respectively:

$$\hat{H}_{\rm a} = \sum_{j=1}^{N}\sum_{n=1}^{3} E^j_{nn}\hat{P}^j_{nn},$$

$$\hat{H}_{\rm f} = \hbar\sum_{\nu=1}^{2}\int {\rm d}z\, \hat{A}^+_\nu(z)[\omega_\nu + i(-1)^\nu \upsilon_\nu \frac{\partial}{\partial z}]\hat{A}_\nu(z),$$

$$\hat{V}_{\rm a-f} = -\hbar\sum_{j=1}^{N}\sum_{\nu=1}^{2}[g_\nu \hat{A}_\nu(z_j)\exp\{{\rm i}k_\nu z_j\}\hat{P}^j_{31} + {\rm h.c.}],$$

$$\hat{V}_{\rm c} = -\hbar\sum_{j=1}^{N}\sum_{\nu=1}^{2}[\Omega_\nu(t)\exp\{-{\rm i}(\omega^c_\nu t - \vec{K}_\nu\vec{r}_j)\}\hat{P}^j_{32} + {\rm h.c.}].$$

Here, $\hat{P}^j_{mn}$ denotes the atomic operator $|m\rangle_{jj}\langle n|$, E^j_{nn} is the energy of the nth level of atom j, and $\nu = 1, 2$ identifies storage and recall, respectively. Furthermore, $[\hat{A}_{\nu'}(z'), \hat{A}^+_\nu(z)] = \delta_{\nu',\nu}\delta(z'-z)$, $k_\nu = (-1)^{\nu+1}\,\frac{\omega_\nu n_\nu}{c}$, n_ν and $\upsilon_\nu = \partial\omega/\partial k|_{\omega=\omega_\nu}$ are the refractive indices and group velocities for the probe and echo fields in the absence of interaction with the three-level atoms, and g_ν is the photon–atom coupling constant. Note that $\hat{H}_{\rm f}$ takes into account that the propagating quantum light fields are described by localized temporal modes and hence feature broad spectra (see [40]).

In the following, we use the Heisenberg picture and derive the equations of motion for the slowly varying operators for the light fields $\hat{A}_{\nu,{\rm o}}$ and atomic coherences $\hat{R}^j_{mn,\nu}$ between states m and n, where

$$\hat{A}_\nu(z,t) = \hat{A}_{\nu,{\rm o}}(z,t)\exp\{-{\rm i}\omega_\nu t\}),$$

$$\hat{P}^j_{12}(t) = \hat{R}^j_{12,\nu}(t)\exp\{{\rm i}\varphi_\nu(\vec{r},z_j) - {\rm i}(\omega_\nu - \omega^c_\nu)(t + (-1)^\nu n_\nu z_j/c)\},$$

$$\hat{P}^j_{13}(t) = \hat{R}^j_{13,\nu}(t)\exp\{-{\rm i}\omega_\nu(t + (-1)^\nu n_\nu z_j/c)\},$$

$$\hat{P}^j_{32}(t) = \hat{R}^j_{32,\nu}(t)\exp\{{\rm i}\varphi_\nu(\vec{r},z_j) + {\rm i}\omega^c_\nu(t + (-1)^\nu n_\nu z_j/c)\},$$

$$\hat{P}^j_{mm}(t) = \hat{R}^j_{mm,\nu}(t),$$

$$\varphi_\nu(\vec{r},z_j) = -((-1)^\nu n_\nu\omega^c_\nu z_j/c + \vec{K}_\nu\vec{r}).$$

These definitions take into account changes in variables and operators associated with storage and recall. As for storage, we also assume $|\Delta_2 + \Delta^j_{31,2}| \gg \delta\omega$ (with arbitrary $\Delta_2/\Delta^j_{31,2}$), $|\Delta^j_{21,2}| \ll |\Delta^j_{31,2}|$, $|\Delta^j_{32,2}|$ and $\frac{1}{\Delta_2+\Delta^j_{32,2}} \cong \frac{1}{\Delta_2+\Delta^j_{31,2}}$. Hence, we can, now in full generality, express

$\Delta^j_{32,\nu}$ in terms of $\Delta^j_{31,\nu}$. Being necessary conditions for time-reversibility, we ignore all atomic decay as well as irreversible atomic dephasing while the probe state is mapped onto atomic coherence, and assume that the probe field is completely absorbed in the atomic media. To simplify the expressions, we use a coordinate system moving with the probe, or echo fields, respectively. For absorption ($t < t'$, $\nu = 1$), we use $\tau_1 = t - Z/v_1$, and for retrieval ($t > t'$, $\nu = 2$), we have $\tau_2 = t + Z/v_2 - \tau_e$, where $Z = z$. Finally, to simplify the equations for the light fields and atomic coherences, we introduce new variables for the fields:

$$\hat{\zeta}_\nu(\tau_\nu, Z) = -(-1)^\nu(\Omega^*_\nu(\tau_\nu)/\Delta_\nu)g_\nu\hat{A}_{\nu,\mathrm{o}}(\tau_\nu, Z),$$

and we assume $\omega_\nu - \omega^c_\nu = \omega_{21}$. All put together, taking into account that $\hat{R}^j_{33,\nu} \cong 0$, i.e. $\hat{R}^j_{11,\nu} + \hat{R}^j_{22,\nu} \cong 1$, and after adiabatic elimination of the excited atomic coherences

$$\hat{R}^j_{13,\nu}(t) \cong \frac{g_\nu\hat{A}_{\nu,\mathrm{o}}(t, z_j)\hat{R}^j_{11,\nu}(t) + \Omega_\nu(t)\hat{R}^j_{12,\nu}(t)}{\Delta_\nu + \Delta^j_{31,\nu}}$$

and

$$\hat{R}^j_{32,\nu}(t) \cong \frac{g_\nu\hat{A}^+_{\nu,\mathrm{o}}(t, z_j)\hat{R}^j_{12,\nu} + \Omega^*_\nu(t)\hat{R}^j_{22}(\tau)}{\Delta_\nu + \Delta^j_{31,\nu}},$$

we find that

$$\begin{aligned}
\frac{\partial}{\partial Z}\hat{\zeta}_\nu &= \frac{\mathrm{i}\beta_\nu}{2}\left(\frac{-(-1)^\nu}{\Delta_\nu}\hat{B}_{11,\nu}\hat{\zeta}_\nu + f_\nu\hat{B}_{12,\nu}\right),\\
\frac{\partial}{\partial \tau_\nu}\hat{R}^j_{12,\nu} &= -\mathrm{i}(-1)^\nu\hat{\zeta}_\nu\frac{\hat{R}^j_{11,\nu} - \hat{R}^j_{22,\nu}}{1 + \Delta^j_{31,\nu}/\Delta_\nu}\\
&\quad -\mathrm{i}\left(\Delta^j_{21,\nu} - \frac{\Delta_\nu}{1 + \Delta^j_{31,\nu}/\Delta_\nu}\left(f_\nu - \frac{\hat{\zeta}^+_\nu\hat{\zeta}_\nu}{|\Omega_\nu(\tau_\nu)|^2}\right)\right)\hat{R}^j_{12,\nu},\\
\frac{\partial}{\partial \tau_\nu}\hat{R}^j_{11,\nu} &= -\mathrm{i}(-1)^\nu\left(\frac{\hat{\zeta}^+_\nu\hat{R}^j_{12,\nu}}{1 + \Delta^j_{31,\nu}/\Delta_\nu} - \frac{\hat{R}^j_{21,\nu}\hat{\zeta}_\nu}{1 + \Delta^j_{31,\nu}/\Delta_\nu}\right),
\end{aligned} \tag{1}$$

where $\frac{\partial}{\partial t}\hat{R}^j_{22,\nu} = -\frac{\partial}{\partial t}\hat{R}^j_{11,\nu}$ and $\beta_\nu = 2\pi(n_\mathrm{o}S)|g_\nu|^2/v_\nu$ with atomic density n_o, and cross section of the probe (echo) field S. Furthermore, $\hat{B}_{mn,\nu} = \int \mathrm{d}\Delta^j_{21,\nu}\int \mathrm{d}\Delta^j_{31,\nu}G(\Delta^j_{21,\nu})$ $G(\Delta^j_{31,\nu})\hat{R}^j_{mn,\nu}(\tau, Z_j \approx Z)/(1 + \Delta^j_{31,\nu}/\Delta_\nu)$, with $G(\Delta^j_{mn,\nu})$ describing the IB of the $m \leftrightarrow n$ transition.

All variables in equations (1) depend on τ_ν and Z. We emphasize that these equations hold for arbitrary numbers of atoms N and photons N_p, provided $N > N_\mathrm{p}$. As compared to the usual Maxwell–Bloch equations [38, 39], they contain additional nonlinear terms. In the evolution of the atomic coherence $\hat{R}^j_{12,\nu}$, the predominant term is a Stark shift ($\sim \frac{\Delta_\nu}{1+\Delta^j_{31,\nu}/\Delta_\nu}f_\nu$) that is due to the interaction of the atoms with the off-resonant control fields. Another, generally much smaller, shift is caused by the probe and echo fields ($\sim \frac{\hat{\zeta}^+_\nu\hat{\zeta}_\nu}{|\Omega_\nu(\tau_\nu)|^2}\Delta_\nu$). Furthermore, a term that is proportional to the population of level 1 affects the refractive index, due to the large spectral detuning Δ_ν from the single-photon resonance, and causes dispersion in the evolution of $\hat{\zeta}_\nu$.

The analytic solution of equations (1) and their analysis for time-reversal symmetry have not been considered before.

4. General reversibility of quantum dynamics

4.1. Storage of strong probe fields

We now show that equations (1) allow for time-reversible evolution (i.e. storage and retrieval of the probe field) despite the nonlinear terms. This implies unit efficiency and fidelity without the necessity to solve any equations (a similar approach was first discussed in [23]). Indeed, the equations for retrieval coincide with the equations for absorption for time-reversed echo emission (i.e. $\tau_2 \rightarrow -\tau_2'$ and $\frac{\partial}{\partial \tau_2} \rightarrow -\frac{\partial}{\partial \tau_{2'}}$) if the following *strong field conditions of reversibility* are satisfied.

(i) $c(\vec{K}_1 - \vec{K}_2) = (n_1\omega_1 + n_2\omega_2)\vec{e}_z$, i.e. a phase matching operation that results in mapping the atomic coherence created by the forward propagating probe field onto coherence that can create a backwards propagating echo (i.e. ensures equal boundary conditions at the end of the forward evolution and the start of the backward evolution). It is found from $\hat{R}^j_{12,2}(t') = \exp\{-\mathrm{i}\alpha\}\hat{R}^j_{12,1}(t')\forall j$, with t' denoting a moment after complete probe absorption, and by expressing $\hat{R}^j_{12,1}$ and $\hat{R}^j_{12,2}$ through $\hat{P}^j_{12}$. The phase factor α contributes to the global phase of the echo. Note the absence of a similar equality for the light field operators, which is due to complete absorption of the probe field.

(ii) $\beta_1 = \beta_2$ and $f_1(\tau_1) = f_2(-\tau_2')$, i.e. equal coupling between the atomic coherence of the Raman transition and the probe and echo fields, respectively, and $|\Omega_1(\tau_1)| = |\Omega_2(-\tau_2')|$, i.e. temporal reversibility of the Rabi frequencies of the writing and reading fields (see figure 1).

(iii) $\Delta^j_{21,2} - \frac{f_2(-\tau_2')\Delta_2}{1+\Delta^j_{31,2}/\Delta_2} + \Delta^j_{21,1} - \frac{f_1(\tau_1)\Delta_1}{1+\Delta^j_{31,1}/\Delta_1} = \Delta^j_{21,2} + \Delta^j_{21,1} = 0$, i.e. rephasing of atomic coherence when reversing the IB, similar to CRIB[5]. The first equal sign requires meeting conditions (ii) and (iv).

(iv) $\Delta_2 = -\Delta_1$, and $\frac{\Delta^j_{31,2}}{\Delta_2} = \frac{\Delta^j_{31,1}}{\Delta_1}$, i.e. anti-correlated spectral detunings of the light fields. This condition completely determines ω_2 for a given ω_1 and Δ_1: $\omega_2 = \omega_{31} - \Delta_2 = \omega_1 + 2\Delta_1$, where we assumed that $\omega_{31} = \omega_\nu + \Delta_\nu$.

To support our claim, we consider the equation that describes the evolution of $\hat{R}^j_{11,\nu}$. As we replace storage ($\nu = 1$) with retrieval ($\nu = 2$), and taking into account condition (iv), we find that all terms on the right-hand side change sign. This sign change also applies to the left-hand side if we consider time-reversed evolution (i.e. $\partial/\partial\tau_2 = -\partial/\partial\tau_2'$). Hence, the equation describing the change in the population of level one during recall predicts a time-reversed evolution as compared to absorption. Taking into account conditions (ii)–(iv), the same is straightforward to show for all equations of motion. Given equal boundary conditions (as enforced by condition (i), this proves our claim.

The time reversibility hidden in equations (1) results in a temporally reversed replica of arbitrary probe fields in the echo, which is equivalent to the standard system of the Maxwell–Bloch equations [19, 23]. Thus any quantum state encoded into the input light field can be stored and recalled with unit efficiency and fidelity, despite the nonlinear interactions.

[5] It may be possible to generalize this expression, similar to the case of weak probe fields discussed below.

At the same time, the demonstrated reversibility enables new possibilities for the realization of optical QM, e.g. storage of multi-mode macroscopic light fields in nano-size memories where the usual weak-field approximation may not be satisfied [47][6].

4.2. Storage of weak probe fields

It is interesting to compare the previous conditions for time reversibility with the conditions in the case of weak probe fields, where $\langle \hat{R}_{11,\nu} \rangle \cong 1$ and $\langle \hat{R}_{22,\nu} \rangle \cong 0$. In the following, we also assume that the Stark shift due to the presence of the probe and echo fields is small compared to the spectral width of the stored light: $\frac{\Delta_\nu \langle \hat{\zeta}^+_\nu \hat{\zeta}_\nu \rangle}{|\Omega_\nu|^2} \ll \delta\omega$, where $\langle .. \rangle$ denotes the expectation value, $|\Delta^j_{31,\nu}| \ll \Delta_\nu$, and we use new variables

$$(\tilde{\hat{\zeta}}_\nu, \tilde{\hat{R}}_{12,\nu}) = (\mathrm{i}\hat{\zeta}_\nu, \hat{R}_{12,\nu}) \exp\left\{\mathrm{i}(-1)^\nu \frac{\beta_\nu}{2\Delta_\nu} Z - \mathrm{i}\psi_\nu\right\}$$

in equations (1), with $\psi_\nu = \int^{\tau_\nu} \Delta^{s0}_\nu(\tau_\nu)\mathrm{d}\tau_\nu$. These variable transformations result in a description of the atom–light evolution that is similar to the well-known Maxwell–Bloch equations:

$$\begin{aligned} \frac{\partial}{\partial Z}\tilde{\hat{\zeta}}_\nu &= -\frac{\beta_\nu f_\nu}{2}\tilde{\hat{B}}_{12,\nu}, \\ \frac{\partial}{\partial \tau_\nu}\tilde{\hat{R}}^j_{12,\nu} &= -\mathrm{i}\Delta^j_{R,\mathrm{tot},\nu}\tilde{\hat{R}}^j_{12,\nu} - (-1)^\nu \tilde{\hat{\zeta}}_\nu, \end{aligned} \tag{2}$$

where the centers of the Raman transitions are shifted by $\Delta^{s0}_\nu(\tau_\nu) = \Delta_\nu f_\nu(\tau_\nu)$ (see figure 1). Recall that $\Delta^j_{R,\mathrm{tot},\nu} = \Delta^j_{21,\nu} + \Delta^j_{31,\nu} f_\nu$. For a probe field with symmetric shape in time and a Raman broadening that is symmetric in frequency, these linearized equations allow again time-reversible evolution, but this time under the *weak field conditions of reversibility*[7]:

(i′) $c(\vec{K}_1 - \vec{K}_2) = \vec{e}_z[n_1\omega_1 + n_2\omega_2 + c(\beta_1/\Delta_1 + \beta_2/\Delta_2)]$, which is found from $\tilde{\hat{R}}^j_{12,2}(t') = \exp\{-\mathrm{i}\alpha'\}\tilde{\hat{R}}^j_{12,1}(t')$, as explained in (i). Note that this generalized mode matching condition coincides with (i) if conditions (ii) and (iv) are met. Yet, in the case of weak probe fields, these requirements are relaxed, e.g. due to the lack of the probe (echo)-induced Stark shift. In particular, this allows for continuous frequency conversion of the echo compared to the probe. Assuming resonance with the center of the IB Raman transition and constant Stark shifts, we find that $\omega_1 = \omega^s_1 + \omega_{21} - \Delta^{s0}_1$ and $\omega_2 = \omega_1 + \omega^c_2 - \omega^c_1 + \Delta^{s0}_1 - \Delta^{s0}_2$.

(ii′) $\beta_1 f_1(\tau_1) = \beta_2 f_2(-\tau_2)$, similar to (ii), but without the necessity to individually equalize each variable.

[6] We note that the diffraction limit can be overcome using plasmonic devices [41] and dielectric–metamaterial interfaces [42] and unity-absorption in media with small single-pass optical depth can be achieved through impedance-matched cavities [45, 46]. Furthermore, the analyzed Raman scheme exploits a direct transfer of the state of the probe field onto long-lived atomic coherence on the $1 \leftrightarrow 2$ transition. The relevant, time-dependent Rabi frequency for this direct transfer is given by the electric field of the control fields averaged over the spatial mode determined by the configuration of the atomic ensemble, regardless of its size [47]. This is similar to the cavity mode in a QED approach.

[7] The assumed symmetry leads to a symmetry in the excited $1 \leftrightarrow 2$ coherence under exchange of detuning $\Delta_1 \to \Delta_2$. Together with conditions (i′–iii′), this allows showing time-reversal symmetry. The imposed symmetry in the probe field shape is not required in the case of $k = 0$. It is included here for a unified presentation.

(iii′) $\Delta^j_{R,\mathrm{tot},2} t_2 + \Delta^j_{R,\mathrm{tot},1} t_1 = k n_j 2\pi + \mathrm{const.}$, where $k \in \mathbf{N}_0$, $n_j \in \mathbf{Z}$, and const. is an irrelevant constant. We emphasize that this generalized rephasing condition can be met by using additional experimental approaches compared to (iii). We can find those approaches, characterized by k, by identifying situations where all atoms accumulated the same phase (modulo 2π), i.e. where the initially excited collective coherence is recovered.

For $k = 0$, rephasing is achieved at time $t_1 = t_2$ by actively inverting the Raman detuning of each atom j, and we recover condition (iii). In analogy to CRIB, we refer to this protocol as RECRIB.

For $k = 1$, we analyze equations (2) for a symmetric comb structure, where each atom j is located in an absorption line detuned from the unbroadened transition by a multiple of δ_{comb} given by n_j: $\Delta^j_{31,\mathrm{tot},1} = \Delta^j_{31,\mathrm{tot},2} = n_j \delta_{\mathrm{comb}}$ (we assume for simplicity the broadening of the Raman transition to be due to the broadening of the $1 \leftrightarrow 3$ transition and f_ν). Using symmetry properties for absorption and retrieval to recover the coherence $\tilde{\hat{B}}_{12,\nu}$, we find an echo emitted at a time given by $f_1 t_1 + f_2 t_2 = 2\pi/\delta_{\mathrm{comb}}$, which can be controlled by varying δ_{comb}, f_1 or f_2. In analogy to the AFC QM, we refer to this approach as REAFC. Approaches with $k > 1$ are subclasses of $k \neq 0$. They also rely on AFC, but exploit that rephasing of atomic coherence is repetitive, i.e. can generate echoes at later times.

4.3. Discussion and conclusion

Obviously, experimental imperfection excludes perfect time-reversible evolution. Before we conclude this paper, let us briefly discuss how limited optical depth, e.g. due to large controlled broadening, and the remaining dephasing during the presence of the control fields impacts on the storage efficiency

$$\epsilon = \frac{\int \mathrm{d}t \langle \hat{A}_2^\dagger(t) \hat{A}_2(t) \rangle}{\int \mathrm{d}t \langle \hat{A}_1^\dagger(t) \hat{A}_1(t) \rangle}.$$

To evaluate ϵ for weak probe fields and Lorentzian line shapes, we solve a slightly modified version of equations (2):

$$\begin{aligned} \frac{\partial}{\partial Z} \tilde{\hat{\xi}}_\nu &= -\frac{\beta_\nu f_\nu}{2} \tilde{\hat{B}}_{12,\nu}, \\ \frac{\partial}{\partial \tau_\nu} \langle \tilde{\hat{R}}^j_{12,\nu} \rangle &= -(\Gamma_\nu + i \Delta^j_{\mathrm{cont},\nu}) \langle \tilde{\hat{R}}^j_{12,\nu} \rangle - (-1)^\nu \tilde{\hat{\xi}}_\nu, \end{aligned} \tag{3}$$

where $\langle \tilde{\hat{R}}^j_{12,\nu} \rangle = \int \mathrm{d}\Delta^j_{21,\mathrm{nat}} G(\Delta^j_{21,\mathrm{nat}}/\Delta_{21,\mathrm{nat}}) \int \mathrm{d}\Delta^j_{31,\mathrm{nat}} G(\Delta^j_{31,\mathrm{nat}}/\Delta_{31,\mathrm{nat}}) \tilde{\hat{R}}^j_{12,\nu}$ denotes the atomic coherence averaged over the natural broadenings on the $1 \leftrightarrow 2$ and $1 \leftrightarrow 3$ transitions, and $\Gamma_\nu = f_\nu \Delta_{31,\mathrm{nat}} + \Delta_{21,\mathrm{nat}}$ describes the irreversible dephasing.

Assuming no frequency conversion ($\beta_\nu \equiv \beta$, $\upsilon_\nu \equiv \upsilon$, $\Delta_\nu \equiv \Delta$, $f_\nu \equiv f$, $\Gamma_\nu \equiv \Gamma$) and following standard procedures to solve linearized Maxwell–Bloch equations [43, 44], we find

$$\epsilon = \exp\{-2\Gamma(t_1 + t_2)\} |1 - \mathrm{e}^{-\alpha_{\mathrm{eff}} L}|^2.$$

$\alpha_{\mathrm{eff}} L$ is the effective optical depth, which depends on the on-resonant absorption coefficient $\alpha_{\mathrm{o}} = \beta/\Delta_{31,\mathrm{tot}}$, and L is the length of the atomic medium. For RECRIB, assuming that the width of the initial, naturally broadened absorption line $\Delta_{31,\mathrm{nat}}$ (possibly reduced after spectral tailoring [24]) is small compared to the linewidth $\Delta_{31,\mathrm{cont}}$ after controlled broadening, we find that $\alpha_{\rm eff}^{\rm (RECRIB)} = \alpha_{\rm o} f \Delta_{31,\rm tot}/(f\Delta_{31,\rm cont} + \Delta_{21,\rm cont} + \Gamma)$ and $\Gamma^{\rm (RECRIB)} = f\Delta_{31,\rm nat} + \Delta_{21,\rm nat}$. For REAFC, taking the width γ of each individual comb line to be small compared to the width of the whole comb, and assuming many lines, we find that $\alpha_{\rm eff}^{\rm (REAFC)} = \alpha_{\rm o}\sqrt{2\pi}(\gamma/\delta_{\rm comb})$ and $\Gamma^{\rm (REAFC)} = f\gamma + \Delta_{21,\rm nat}$. The efficiency decreases with increased controlled broadening, increases with $\alpha_{\rm o}L$ and depends on Γ. The optimum value of Γ is determined by the trade-off between small irreversible dephasing, i.e. small unbroadened absorption lines ($\Delta_{31,\rm nat}$ and γ, respectively), and large absorption, i.e. large linewidth compared to $\Delta_{31,\rm cont}$ or $\delta_{\rm comb}$, respectively. Note that the values of $\alpha_{\rm o}L$ of 1800 and 32 have been reported for Cs vapor [11] and $Pr:Y_2SiO_5$ [29], respectively. Furthermore, the use of impedance-matched cavities promises unity absorption even in the event of small single-pass optical depth [45, 46].

Let us emphasize some interesting features of REQM. Firstly, the storage bandwidth, which depends on the IB of the Raman transition, not only relies on material properties but also can be changed by adjusting the control fields (i.e. f_1)[8]. Secondly, the direct Raman transfer allows using atomic materials with potentially short optical coherence times T_2, as determined by the duration of the control fields t_1, t_2 and Γ (which depends on f). Hence, for a given T_2, one can maximize the efficiency via f, although this also impacts on the storage bandwidth. These two properties result in a larger choice of materials compared to photon-echo QM without direct Raman transfer. Furthermore, as opposed to the storage of quantum states using a homogeneously broadened Raman transition, REQM does not require tailoring the temporal shape of the control field depending on that of the probe field [16], which makes it more universal and simpler to implement. Also, as in all photon-echo protocols, the multi-mode storage capacity scales better with respect to the optical depth [26]. Finally, we point out that dynamical frequency shifts induced by previous absorption of (possibly intense) pulses of light [49] are without consequences for our scheme as long as the absorption is not modified. As the storage bandwidth of our approach, determined by the total Raman broadening, is much larger than the frequency change due to repopulation of atomic levels, this requirement is well satisfied and cascades of transitions do not impact on the reversibility of the light–atom interaction, i.e. on the fidelity or efficiency.

In conclusion, we have shown that REQM generalizes time-reversible dynamics in photon-echo QM in the case of strong fields and nonlinear interactions. Furthermore, for weak input fields it unifies AFC and CRIB in an extension to off-resonant Raman transitions. It allows exploitation of additional degrees of freedom, e.g. wave vectors, and carrier and Rabi frequencies of the control fields, allowing us to influence Raman IB, i.e. storage bandwidth, and frequency conversion.

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[8] Note the lack of dependence of the bandwidth on the detuning Δ_1, in opposition to EIT and usual Raman QM. See [48].

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