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Off-resonant Raman quantum memory in impurity crystals: signal-to-noise ratio analysis

A.D. Berezhnoi, A.A. Kalachev

Abstract. **The possibility of implementing an off-resonant Raman scheme of optical quantum memory on the basis of an ensemble of three-level atoms is investigated under the condition of equal polarisations of resonant transitions forming the Λ-scheme. The developed model is used to analyse the signal-to-noise ratio at the output of an optical quantum memory device in $^{143}Nd^{3+}:Y^{7}LiF_4$. It is shown that this ratio can significantly exceed unity for single-photon input pulses. The required values of the parameters can be obtained by using an impurity crystal in the form of a whispering-gallery mode ring resonator.**

Keywords: *quantum memory, impurity crystal, resonator, signal-to-noise ratio.*

1. Introduction

The creation of optical quantum memory is one of the topical tasks of modern quantum optics and computer science [1–6]. Devices capable of storing and retrieving quantum states of light are necessary for the operation of optical quantum computers and for the implementation of long-distance optical quantum communication. In addition, quantum memory can be used to develop deterministic sources of Fock states of the electromagnetic field, i.e., states with a certain number of photons, and also to implement various quantum measurement protocols involving the transfer of nonclassical states of light into atoms.

At present, main attention is paid to quantum memory schemes utilising the interaction of weak light pulses with ensembles of atoms. These schemes are based on photon echoes [7–9], electromagnetically induced transparency [10, 11] and off-resonant Raman absorption and scattering [12], as well as make it possible to implement multimode quantum memory necessary for practical applications. As for quantum information carriers, dielectric crystals doped with rare-earth ions are among the most promising [13]. The coherence lifetime of impurity-ion ground-state hyperfine transitions, which determines the storage time of the recorded information, reaches several hours in such crystals [14]. Of particular interest are isotopically pure impurity crystals, which exhibit very narrow optical lines up to 10 MHz [15–19]. Such a small inhomogeneous broadening of optical transitions opens the possibility of implementing various schemes of off-resonant Raman quantum memory [20–24], which has so far been realised only in gas media [25–27] and in a diamond crystal [28].

The overwhelming majority of theoretical works devoted to the development of new optical quantum memory schemes is based on the use of an ideal three-level model of atoms with the Λ-scheme of transitions having orthogonal polarisations. Indeed, in an experiment it is sometimes possible to find close-to-ideal conditions. However, in some cases, in particular when analysing the signal-to-noise ratio at the output of a memory device, such a model is not complete. First of all, there is a need to take into account the actual structure of atomic levels that goes beyond the three-level model. For gases, the corresponding theory was developed in [29]. In the context of solid-state off-resonant Raman memory, one can note paper [30], in which, in addition to the analysis of noise, it was shown that it is possible to realise the Λ-scheme of orthogonal linearly polarised transitions in diamond NV centres due to the action of external electric and magnetic fields. Another factor that significantly affects the noise level is the nonorthogonal polarisation of the transitions forming the Λ-scheme, which is typical, for example, of optical ground- and excited-state hyperfine transitions in rare-earth ion-doped dielectric crystals. As an example, we can cite paper [31], where 10 Λ-like systems were experimentally identified in a $^{167}Er^{3+}:Y_2SiO_5$ crystal. Therefore, the theoretical analysis of the storage and retrieval of single photons in the regime of off-resonant Raman interaction of fields with impurity crystals doped by rare-earth ions, taking into account the real hyperfine structure of the working levels and the identical polarisation of the transitions forming a Λ-like scheme, is an important problem. This is the subject of this work.

2. Basic equations

The general scheme of quantum memory based on the Raman interaction of two fields with an ensemble of $N \gg 1$ atoms placed in a resonator is shown in Fig. 1. This scheme was analysed in detail in [32] under the assumption that polarisations of $|3\rangle - |1\rangle$ and $|3\rangle - |2\rangle$ transitions are orthogonal. This situation is realised quite easily in gases located in a magnetic field. In this paper, we consider the case of an arbitrary Λ-scheme, which is obtained, for example, by using hyperfine sublevels of the ground and excited states of impurity ions in dielectric crystals. In the case of nonorthogonal polarisation of the transitions forming the Λ-scheme, the control field with frequency ω acts on both transitions, producing a Raman com-

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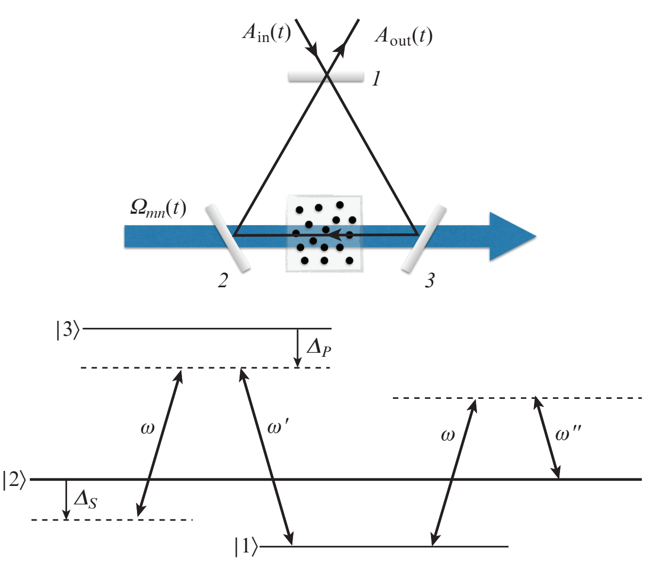

Figure 1. (Top) Schematic diagram of off-resonant Raman quantum memory and (bottom) scheme of working transitions: (*1–3*) mirrors that are transparent to the control field (frequency ω) and form a ring resonator with one input/output for a signal field (frequency ω') and an idler field (frequency ω''); $\Omega_{mn}(t)$ is the amplitude (Rabi frequency) of the control field corresponding to the $|m\rangle - |n\rangle$ transition; $A_{in}(t)$ and $A_{out}(t)$ are the amplitudes of single-photon pulses of the signal field at the resonator input and output, respectively.

bination with the signal (frequency ω') and idler (frequency ω'') fields. Then, the Hamiltonian of the system in the rotating wave approximation takes the form:

$$H = -\hbar[\Omega_{32}\sigma_{32}\exp(-\mathrm{i}\omega t) + \Omega_{31}\sigma_{31}\exp(-\mathrm{i}\omega t) + g_{31}\sigma_{31}a + g_{32}\sigma_{32}a + g_{31}\sigma_{31}b + g_{32}\sigma_{32}b] + \text{h.c.}, \quad (1)$$

where $a = A\exp(-\mathrm{i}\omega' t)$ and $b = B\exp(-\mathrm{i}\omega'' t)$ are the annihilation operators for the photons of the signal and idler fields, respectively, in the resonator modes; $\sigma_{31} = P^*\exp(\mathrm{i}\omega' t)$, $\sigma_{21} = S^*\exp[\mathrm{i}(\omega' - \omega)t]$, $\sigma_{32} = Q^*\exp(\mathrm{i}\omega t)$ are the atomic transition operators ($\sigma_{mn} = |m\rangle\langle n|$); and A, B and P, S, Q are slowly varying field and coherence amplitudes, respectively. For an ensemble of atoms, taking into account that in the process of interaction with a weak signal field almost all atoms are in the ground state ($\langle\sigma_{11}\rangle \approx N$, $\langle\sigma_{33}\rangle \approx \langle\sigma_{22}\rangle \approx 0$), from the Heisenberg–Langevin equations we obtain the equations

$$\dot{P} = -\mathrm{i}\Delta_P P - \gamma_P P + \mathrm{i}\Omega_{32}S + \mathrm{i}\Omega_{31}\sqrt{N}\exp(-\mathrm{i}\delta t) + \mathrm{i}g_{31}\sqrt{N}A + \mathrm{i}g_{32}AS\exp(\mathrm{i}\delta t) + \mathrm{i}g_{31}\sqrt{N}B\exp[\mathrm{i}(\delta' - \delta)t] + \mathrm{i}g_{32}BS\exp(\mathrm{i}\delta' t), \quad (2)$$

$$\dot{S} = -\mathrm{i}\Delta_S S - \gamma_S S + \mathrm{i}\Omega_{32}^* P - \mathrm{i}g_{31}Q^*A - \mathrm{i}\Omega_{31}Q^*\exp(-\mathrm{i}\delta t) + \mathrm{i}g_{32}^* PA^*\exp(-\mathrm{i}\delta t) - \mathrm{i}g_{31}Q^*B\exp[\mathrm{i}(\delta' - \delta)t] + \mathrm{i}g_{32}^* PB^*\exp(-\mathrm{i}\delta' t), \quad (3)$$

$$\dot{Q} = -\mathrm{i}(\Delta_P - \Delta_S)Q - \gamma_Q Q - \mathrm{i}g_{31}S^*A - \mathrm{i}\Omega_{31}S^*\exp(-\mathrm{i}\delta t) - \mathrm{i}g_{31}S^*B\exp[\mathrm{i}(\delta' - \delta)t], \quad (4)$$

$$\dot{A} = \mathrm{i}g_{31}^*\sqrt{N}P + \mathrm{i}g_{32}^*Q\sqrt{N}\exp(-\mathrm{i}\delta t) - kA + \sqrt{2k}A_{in}, \quad (5)$$

$$\dot{B} = \mathrm{i}g_{31}^*\sqrt{N}P\exp[-\mathrm{i}(\delta' - \delta)t] + \mathrm{i}g_{32}^*Q\sqrt{N}\exp(-\mathrm{i}\delta' t) - kB + \sqrt{2k}B_{in}, \quad (6)$$

where for all atomic coherences we used a normalisation of the form $P = \sum_{j=1}^{N} P_j/\sqrt{N}$ and introduced the notation $\delta = \omega - \omega'$; $\delta' = \omega - \omega''$; $g_{mn} = d_{mn}\sqrt{\omega/(2\hbar\varepsilon_0 V)}$ is the coupling constant of the field with atoms at the $|m\rangle - |n\rangle$ transition, which is characterised by the dipole moment d_{mn} (in this case we can set $\omega' = \omega'' = \omega$); $\gamma_P, \gamma_S, \gamma_Q$ are the decay rates of coherences; $2k$ is the decay rate of the field in the resonator; and V is the volume of the resonator field mode. The two-photon detuning Δ_S takes into account the frequency shift equal to $-|\Omega_{32}|^2/\Delta_P$ (Δ_P is the single-photon detuning), which occurs under the action of the control field. In the case of an exact two-phonon resonance, when $\Delta_S = 0$, we have $\delta = -\delta'$. Finally, the amplitude of the signal field at the output from the resonator, $A_{out}(t)$, is found from the boundary condition $A_{out}(t) = \sqrt{2k}A(t) - A_{in}(t)$. In the numerical solution of the system of equations (2)–(6), the operators are replaced by complex numbers corresponding to the amplitudes of the transition probability between the vacuum and single-photon states for the field and between the ground and excited states for atoms. In this case, the idler field B_{in} at the input is assumed to be zero. Since the signal field is considered to be weak, the contribution of atomic noise operators in the Heisenberg – Langevin equations can be neglected (see, e.g., [32]), and they are not included in this system of equations. In the case of orthogonal polarisation of the transitions forming the Λ-scheme, we must set $\Omega_{31} = 0$ and $g_{32} = 0$ if we consider the signal field acting on the $|1\rangle - |3\rangle$ transition and the control field acting on the $|2\rangle - |3\rangle$ transition.

3. Quantum memory in a $^{143}Nd^{3+}:Y^7LiF_4$ crystal

As an example, let us consider the possibility of storing and retrieving single-photon states in an isotopically pure $^{143}Nd^{3+}:Y^7LiF_4$ crystal, which has been recently used in experiments demonstrating quantum memory protocols based on atomic frequency combs [33, 34]. The presence of a hyperfine level structure in odd isotopes of impurity neodymium ions allows storing information about a signal photon in the form of coherence created on transitions between hyperfine sublevels, and the isotopic purity of the crystal provides an inhomogeneous optical transition broadening of 10–50 MHz [18, 33], which is much smaller than the hyperfine splitting of energy levels. The hyperfine structure and the relative probabilities of optical transitions between the sublevels of the ground state [$^4I_{9/2}(0)$] and the excited state [$^4F_{3/2}(0)$] (zero in parentheses is the lowest sublevel of the Stark structure) can be calculated using the effective spin Hamiltonians given in paper [18].

At certain values of the longitudinal magnetic field, clock transitions occur between hyperfine ground-state sublevels, which contributes to an increase in the phase relaxation time. In this case, there is one value of the magnetic field (~636 Gs), at which a symmetric and isolated Λ-scheme of transitions is obtained, when only one of the excited-state sublevels is related to only two ground-state sublevels and the probabilities of both transitions are identical. Moreover, in this case, the linear Zeeman effect is absent for all directions of the magnetic field, which corresponds to a full clock transition, or ZEFOZ-transition (zero first-order Zeeman transition) [35, 36]. According to calculations, the full clock transition occurring in a longitudinal magnetic field, equal to 636 Gs, has a frequency of 2087 MHz, and the polarisation of both transitions forming the Λ-scheme corresponds to linear polarisation perpendicular to the optical axis of the crystal.

The main problem in the numerical solution of the system of equations (2)–(6) was to determine the conditions under which a single-photon wave packet can be stored and retrieved in the regime of off-resonant Raman absorption and emission with a large signal-to-noise ratio. In this case, Gaussian single-phonon pulses with the amplitude $A_{\rm in}(t) = A_{\rm in}^0\exp[-R(t-T)^2]$ were considered, where the parameter R is related to the FWHM pulse duration τ by the relation $R = \tau^{-2}2\ln 2$, and the normalisation corresponding to a single-photon state of the signal field has the form $\int|A_{\rm in}(t)|^2{\rm d}t = 1$. The choice of Gaussian pulses for the analysis is due in particular to the fact that they are optimal for use in optical quantum schemes from the point of view of temporal synchronisation [37]. The change in the Rabi frequency of the control field $\Omega_{32}(t)$, which results in the emission of a Gaussian pulse in the off-resonant Raman scattering regime, can be described analytically (see the solution obtained in [38]):

$$\Omega_{32}^2(t) = \frac{\Omega_0^2}{C}\exp\{2[\gamma_P(t-T) - R(t-T)^2]\}$$
$$\times\left\{\frac{1}{C}\exp\left[\frac{\gamma_P^2}{2R}(1-C^2)\right] + \sqrt{\frac{\pi\gamma_P^2}{2R}}\exp\left(\frac{\gamma_P^2}{2R}\right)\right.$$
$$\left.\times\left[\operatorname{erf}\left(\frac{C\gamma_P}{\sqrt{2R}}\right) - \operatorname{erf}\left[(t-T)\sqrt{2R} - \frac{\gamma_P}{\sqrt{2R}}\right]\right]\right\}^{-1}, \qquad (7)$$

where $C = |g_{31}|^2 N(\gamma_P k)^{-1}(|\Omega_0|/\Delta_P)^2$ is the cooperative parameter, and Ω_0 is the maximum value of the Rabi frequency in the modulation process. To store an analogous Gaussian pulse in the off-resonance Raman absorption regime, it is necessary to use the time-reversed dependence (7). It is convenient to assume that the input pulse has a maximum amplitude at a time instant $-T$ ($T > 0$), so that the information storage time is $2T$. The noise that appears during the storage and retrieval of light pulses is defined as radiation at a signal field frequency that arises when the control field in the absence of an input pulse is applied to the atomic ensemble. In general, at the output of the device one observes the sum of the useful signal and the noise. For the Λ-scheme with the same polarisation of the transitions, we have $\Omega_{31} = \Omega_{32}$, and the noise level is determined primarily by the term $\Omega_{31}\sqrt{N}\exp(-{\rm i}\delta t)$ in Eqn (2). Thus, a large signal-to-noise ratio can be obtained by reducing the Rabi frequency of the control field and decreasing the total number of particles. In this case, the single-photon detuning Δ_P proves much smaller than the interval between the lower sublevels, which for an exact two-photon resonance is equal to $|\delta| = |\delta'|$. This fact imposes a restriction on the minimum duration of the signal field.

One of the results of the calculation is shown in Fig. 2. In calculations, the duration of single-photon pulses, τ, was set equal to 425 ns, and the information storage time $2T$ was 2 μs. The homogeneous width of all resonance transitions was considered equal to 10 kHz, which corresponds to a phase relaxation time of 100 μs. It is this order of magnitude of the relaxation time that we expect to obtain in the crystal under consideration in the case of a full clock transition between the hyperfine sublevels. The values of the other parameters are presented in the caption to Fig. 2. In this case, the total quantum memory efficiency η, defined as

$$\eta = \frac{\int|A_{\rm out}(t)|^2{\rm d}t}{\int|A_{\rm in}(t)|^2{\rm d}t},$$

is equal to 76%, and the contribution of noise to the signal field at the output is 1.4%. For the crystal in question, suitable values of the coupling constant g_{mn} are found by using a whispering-gallery mode ring resonator of about 1 mm in diameter with a Q-factor of 10^8, which at such diameters is not very large and corresponds to the resonator finesse of $\sim 10^4$. In addition, to obtain $N \approx 10^5$ particles requires a very low concentration of impurity ions. In fact, it can be a small segment of a ring resonator containing impurities.

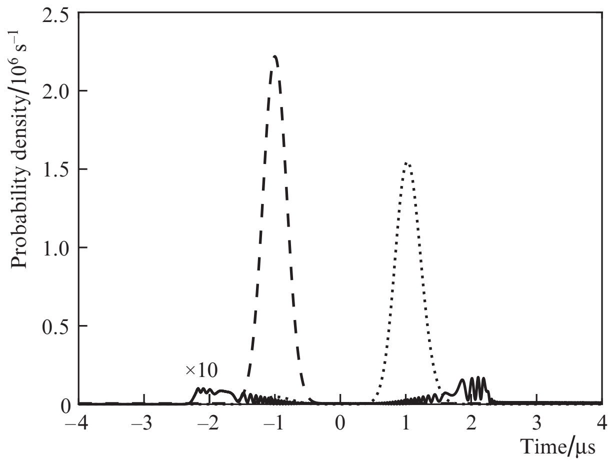

Figure 2. Input (dashed curve) and output (dotted curve) signal field pulses, as well as noise (solid curve) illustrating the operation of quantum memory in ^{143}Nd^{3+} : Y ^{7}LiF$_4$. The result of the numerical solution of equations (2)–(6) was obtained for $\Delta_P/2\pi = -25$ MHz, $\Omega_0/2\pi = 30.4$ MHz, $|\delta|/2\pi = |\delta'|/2\pi = 2087$ MHz, $g_{31}/2\pi = g_{32}/2\pi = 7.9$ kHz, $N = 1.9\times10^5$, $k/2\pi = 6.2$ MHz, $\gamma_P = \gamma_S = \gamma_Q = 2\pi\times 10$ kHz, $\tau = 425$ ns, $T = 1$ μs.

4. Conclusions

We have developed a theoretical model describing the storage and retrieval of weak light pulses in the regime of off-resonant Raman absorption and emission of photons in an ensemble of three-level atoms that have an Λ-scheme of nonorthogonally polarised transitions. Using this model, we have analysed the signal-to-noise ratio at the output of an optical quantum memory device based on an isotopically pure ^{143}Nd^{3+} : Y ^{7}LiF$_4$

crystal. For the given crystal, we have determined the conditions under which an ideal Λ-scheme of transitions is implemented, when only one of the hyperfine sublevels of the excited electronic state is related to only two hyperfine sublevels of the ground electronic state of impurity ions, and the probabilities of both transitions are equal. In this case, the lowest sensitivity of the Raman transition frequency to fluctuations of the magnetic field is attained, which corresponds to a full clock transition. The simulation results have shown that under these conditions the signal-to-noise ratio can significantly exceed unity for single-photon input pulses. The required values of the parameters can be obtained by using an impurity crystal in the form of a whispering-gallery mode ring resonator.

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