Pulse propagation, population transfer and light storage in five-level media

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We consider adiabatic interaction of five-level atomic systems and their media with four short laser pulses under the condition of all two-photon detunings being zero. We derive analytical expressions for eigenvalues of the system’s Hamiltonian and determine conditions of adiabaticity for both the atom and the medium. We analyse, in detail, the system’s behaviour when the eigenvalue with non-vanishing energy is realized. As distinct from the usual dark state of a five-level system (corresponding to zero eigenvalue), which is a superposition of three states, in our case the superposition of four states does work. This seemingly unfavourable case is nevertheless demonstrated to imitate completely a three-level system not only for a single atom but also in the medium, since the propagation equations are also split into those for three- and two-level media separately. We show that, under certain conditions, all the coherent effects observed in three-level media, such as population transfer, light slowing, light storage, and so on, may efficiently be realized in five-level media. This has an important advantage that the light storage can be performed twice in the same medium, i.e., the second pulse can be stored without retrieving the first one, and then the two pulses can be retrieved in any desired sequence.

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I. INTRODUCTION

Coherent interaction of light signals with quantum systems attracted considerable interest for their importance in both fundamental science and practical applications. A prominent example of coherent interactions is electromagnetically induced transparency (EIT)1–3 which can be used to eliminate the resonant absorption of a laser beam incident upon a coherently driven medium with appropriate energy levels. EIT technique allows controlled manipulations of the optical properties of atomic or atom-like media via coupling them with signal and control fields. In particular, it is possible to greatly slow down the optical (signal) pulse1–3 and even stop it to attain reversible storage and retrieval of information7–9.

Despite a huge number of publications, light storage remains in the focus of attention of researchers, since it is one of key components in optical (quantum) information processing1–14. Another application of coherent interactions is controllable population transfer between the atomic levels and constructing desired coherent superpositions of different states12–17. These effects are also employed widely in such fields of research as laser cooling of atoms, lasing without inversion, new precision techniques of magnetometry, coherent control of chemical reactions, and so on.

All the above-listed phenomena are comprehensively studied, both theoretically and experimentally, for various three level systems and their media18–24. Multilevel atomic and atom-like systems, although do not provide new physical principles in addition to quantum interference and principle of superposition, they widen essentially the possibilities of experimental realizations and practical applications. The idea of a double-EIT(DEIT) regime is introduced in27 and modified in28. The laser cooling scheme for trapped atoms or ions which is based on DEIT is discussed in27. DEIT in a medium, consisting of four level atoms in the inverted-Y configuration is discussed in28. DEIT in a ring cavity is studied in29. Enhanced cross-phase modulation based on DEIT is reported in30. Work31 examines dark state polariton formation in a four-level system. Quantum memory for light via stimulated off-resonant Raman process is considered in32 beyond the three-level approximation. Work33 proposes, through numerical calculations, to use multilevel systems involving hyperfine structure in problems of localization of excitations via dark state formation in the EIT processes. Work34 investigates five-level atoms and media driven by four light pulses in nonadiabatic regime. Two of four pulses are assumed weak and treated as perturbation in the first order. Work35 observed experimentally off-resonance EIT-based group delay in multilevel D2 transition in rubidium. Enhancement of EIT in a double-lambda system in cesium atomic vapor by specific choice of atomic velocity distribution is observed in36. A scheme based on two sequential STIRAP processes with four laser fields is proposed in37 for measurement of a qubit of two magnetic sublevels of the ground state of alkaline-earth metal ions. Another topic where multilevel systems were used was generalization of the notion of dark-state polariton38 and discussion of possibility to apply multilevel EIT to quantum informa-
tion processing.

In the present paper we study both analytically and numerically a five-level atomic system interacting adiabatically with four co-propagating laser pulse of different durations and different sequences of turning on and off. We require that each laser pulses interacts (be resonant) with only one of adjacent transition and assume all the two-photon detunings to be zero. Two examples of such level diagrams are shown in Fig. 1. Another example is the ladder-system which may turn out to be rather useful for problems of excitation of Rydberg states.

As distinct from all above-sited works, we concentrate on those eigenstates of interaction Hamiltonian (see below) whose eigenvalues are different from zero. We will show that these eigenstates are similar to the well-known dark and bright states in a three-level system. As distinct from the state in the M-system considered in [4], the levels 2 and 4 are at interaction with laser fields populated, but the population of level 3 remains zero. We will demonstrate for our case that an efficient and more flexibly controllable population transfer and light storage becomes possible. We also study the advantages of this technique. Specifically, we show the possibility of successive storage of two pulses with their subsequent retrieval.

The first pulse is stored into the coherence $\rho_{31}$, which exactly reproduces, after turning off the interaction, the shape of the pulse $\Omega_2$. Since the coherence $\rho_{31}$ remains zero during all time of interaction, the same medium can be used again for storage of the pulse $\Omega_1$. For this purpose, the pulses $\Omega_1$ and $\Omega_2$ should be divided into two beams before the first storage attempt and one part of each pulse should be sent to a delay line, to be used for the second storage. Note that at the second storage we can write the pulse $\Omega_2$, instead of $\Omega_1$; in this case we must change the succession of turning on the pulses (and exchange their polarizations, if the magnetic sublevels are involved).

The paper is organized as follows: In section II we derive eigenfunctions and eigenvalues of systems under consideration and discuss the relevant cases. In section III we study adiabatic population transfer in five-level systems. Section IV derives the equations of propagation and presents their analytical solution. In the same section the regime of adiabaton is demonstrated. In section V we show the possibility to store optical information in considered media. We conclude with a final discussion in section V.

II. EIGENFUNCTIONS AND EIGENVALUES OF INTERACTION HAMILTONIAN.

Consider a five-level atomic system as shown in Fig. 1. Four (in general) laser pulses are close to resonance with respective transitions (Fig. 1). Hamiltonian of interaction in rotating-wave approximation, and under the assumptions that the carrier frequencies of laser pulses are tuned near resonance with one of the adjacent atomic transitions, and pulse durations are much shorter compared to relaxation times in the system, has the following form:

$$H = \sum_i \sigma_{ij} \delta_{i,j} - \sum_i \sigma_{i,i+1} \Delta_i + \hbar \cdot c$$  \hspace{1cm} (1)$$

with the projection matrices $\sigma_{ij}$, the Rabi frequencies $\Omega_i$ at transitions ($i \rightarrow i+1$), and $\delta_{i,j}$ representing $(i-1)$-photon detunings (with $\delta_0 = 0$). The Rabi frequencies are assumed to be real and positive. Phases, which can vary during propagation, are included in the single-photon detunings ($\Delta_i = \omega_{i+1,1} - \omega_i - \phi_i$, if $\omega_{i+1,1} > 0$ and $\Delta_i = \omega_{i+1,1} - \omega_i + \phi_i$ if $\omega_{i+1,1} < 0$). Definition of multi-photon detunings depends on the specific scheme of interaction. For an M-system (see Fig. 1(b)) the multi-photon detunings are $\delta_2 = \Delta_1 - \Delta_2$, $\delta_3 = \Delta_3 + \Delta_4 - \Delta_3$. For an extended $\Lambda$-system (see Fig. 1(a)), the multi-photon detunings are $\delta_2 = \Delta_1 + \Delta_2$, $\delta_3 = -\Delta_3 + \Delta_2$, $\delta_4 = -\Delta_4 + \delta_3$. Similarly to the $\Lambda$ system, we will assume now all the three two-photon detunings to be zero (exact two photon resonances), i.e. $\delta_2 = \delta_4 = 0$ and $\delta_1 = \delta_3 = \Delta$. For an M-system this condition means equal single-photon detunings, while for the extended $\Lambda$-scheme the single-photon detunings have equal absolute value, but differ in sign (see Fig. 1(a)). In this case, one of five eigenvalues of the Hamiltonian is $\lambda = 0$. We can also easily calculate the other four eigenvalues (see Appendix A). Consider now a special case,
when the pulses $\Omega_1$ and $\Omega_4$ coincide by their temporal profiles (but the frequencies and phases of pulses may be different). In this case, the eigenvalues of the Hamiltonian (1) are

$$\lambda_{1,3} = \frac{1}{2} \left( \Delta \mp \sqrt{\Delta^2 + 4\Omega_1^2} \right),$$

$$\lambda_{2,4} = \frac{1}{2} \left( \Delta \mp \sqrt{\Delta^2 + 4(\Omega_1^2 + \Omega_2^2 + \Omega_3^2)} \right)$$

We note, that when the fields are turned off, we get $\lambda_{1,2} \to 0$ and $\lambda_{3,4} \to \Delta$. It should be emphasized that the eigenvalues $\lambda_{1,3}$ depend upon only the field $\Omega_1$ and coincide with the eigenvalues of a two-level system, driven by field $\Omega_1$. Similarly, the eigenvalues $\lambda_{2,4}$ are equal to the eigenvalues of a two-level system, driven by an effective field $(\Omega_1^2 + \Omega_2^2 + \Omega_3^2)^{1/2}$. Adiabatic evolution requires the following conditions to be met (see Appendix A for details):

$$\Delta T \gg 1,$$

$$\frac{(\Omega_3^2 + \Omega_3^2)T}{\Delta} \gg 1,$$

$$\frac{\Omega_1 T}{\Delta} \gg 1$$

with the duration $T$ of the shortest pulse. The first condition mirrors the adiabaticity condition for a two-level system. The second condition corresponds to the adiabaticity condition for a three-level system. The third condition is only relevant in the time interval, when all pulses overlap (i.e., when $\Omega_3^2 + \Omega_3^2 \neq 0$).

To write the eigenvectors corresponding to the eigenvalues $\lambda_1$ and $\lambda_2$ we introduce the following notations:

$\Omega^2 = \Omega_2^2 + \Omega_3^2, \tan \theta = \frac{\Omega_2}{\Omega_3}$,

$$\tan \Phi_1 = -\frac{\lambda_1}{\Omega_1}, \tan \Phi_2 = -\frac{\lambda_2}{\Omega_1},$$

$$\tan \Phi = -\frac{\Omega}{\Omega_1} \cos \Phi_2$$

Then, eigenvector corresponding to the eigenvalue $\lambda_1$ is

$$|\lambda_1\rangle = |\psi_1\rangle \cos \theta - |\psi_2\rangle \sin \theta$$

where $|\psi_1\rangle$ and $|\psi_2\rangle$ are superposition states of two-level systems $1 \to 2$ and $5 \to 4$:

$$|\psi_1\rangle = \cos \phi_1|1\rangle - \sin \phi_1|2\rangle$$

$$|\psi_2\rangle = \cos \phi_1|5\rangle - \sin \phi_1|4\rangle$$

It is apparent that the eigenvector corresponding to $\lambda_1$ does not involve state $|3\rangle$ and is equal to the dark state of a three-level Lambda-system, if we replace the lower states by superposition states $|\psi_1\rangle$ and $|\psi_2\rangle$.

Similarly, the eigenvector corresponding to the eigenvalue $\lambda_2$ yields

$$|\lambda_2\rangle = |\psi_1\rangle \cos \Phi \sin \theta - \sin \phi|3\rangle + |\psi_2\rangle \cos \Phi \cos \theta,$$

where

$$|\psi_1\rangle = \cos \Phi_2|1\rangle - \sin \Phi_2|2\rangle$$

$$|\psi_2\rangle = \cos \Phi_2|5\rangle - \sin \Phi_2|4\rangle$$

As in the previous case, the eigenvector $|\lambda_2\rangle$ is equal to that of the bright state of a three-level Lambda system, if we replace the lower states by superposition states $|\psi_1\rangle$ and $|\psi_2\rangle$. The time behavior of eigenvalues $\lambda_1$ in the special case above for different pulse sequences is demonstrated in Fig. 2 and Fig. 3.

### III. POPULATION TRANSFER

In case of large one-photon detunings, the angle $\Phi_1$ defined in (4) is proportional to $\Omega_1/\Delta$, so we obtain a pure dark state to the terms of the order of $\Omega_1/\Delta$, i.e., the five-level system imitates, in this case, the three-level lambda system. Thus, we can use the state $|\lambda_1\rangle$ to transfer the system from state $|1\rangle$ to state $|5\rangle$ by a STIRAP-like process, driven by the pulse sequence introduced above (see Fig. 1). In contrast to a simple three-level Lambda system, during the interaction some transient population shows up in the intermediate levels $|2\rangle$ and $|4\rangle$ of the five-level Lambda system.
system. However, these transient populations become smaller if the one-photon detuning is sufficiently large, but still satisfies the adiabaticity condition (3). It should be noted that the condition of large one-photon detuning is not very crucial for the population transfer. In this case, the intermediate states |2⟩ and |4⟩ mediate a coupling between states |1⟩, |3⟩, and |5⟩, but are only weakly populated during the process. The dynamics of populations in the described case is shown in Fig. 4.

Similarly, the state |λ2⟩ is analogous to the bright state of a lambda-system and we can use these states for adiabatic transfer from state |1⟩ to state |5⟩ by a b-STIRAP-like process [39, 40] driven by the pulse sequence in Fig. 3, according to definition of angle Φ. We emphasize that the STIRAP-technique is applicable for both schemes of pulse sequence in Figs. 2 and 3. The dynamics of populations in the b-STIRAP case is demonstrated in Fig. 5. Note that the two eigenstates |λ1⟩ and |λ2⟩ render the five-level system, driven by a considered pulse sequence, fully reversible. Thus, we can transfer atomic population from state |1⟩ to state |5⟩ by a STIRAP-like process and from state |5⟩ to state |1⟩ by a b-STIRAP-like process with the same sequence of pulses.

IV. MEDIUM OF ATOMS

Now we move from a single-atom case to that of a medium consisting of the described atoms. We start from the well-known truncated Maxwell equation in running coordinates z = x, τ = t − x/c:

$$\frac{\partial E_i}{\partial x} = i \frac{2\pi\omega_i}{c} N d_i$$

(9)

FIG. 3. Same as in Fig. 2 but for another sequence of pulses.

Here $E_i$ are the complex amplitudes of electric fields of the pulses, $N$ is the number density of medium atoms, and $d_i$ are the amplitudes of induced dipole moments of each individual atom at a frequency $\omega_i$ ($\langle \psi | d_i | \psi \rangle = d_i \exp(-i\omega_i t)$). These amplitudes can be expressed in terms of the amplitudes of atomic populations $b_i$ of bare states and the matrix elements of the dipole moment $\langle i | d | i+1 \rangle$: $d_i = b_i^* b_{i+1} (i | d | i+1) + b_{i+1}^* (i | d | i+1)$ if $\omega_{i+1,i} > 0$ and $d_i = b_i b_{i+1}^* (i | d | i+1)$ if $\omega_{i+1,i} < 0$. The coefficients $b_i$ are determined by the non-stationary Schroedinger equation with Hamiltonian $H$.

Separating real and imaginary parts in the truncated equation of propagation, differentiating the equation for the phase with respect to time, and combining the obtained equations with the Schroedinger equation, we obtain in general case a self-consistent system of equations describing variation of frequencies (one-photon detunings) and intensities (Rabi frequencies) of pulses during propagation in medium. For example, in the case...
optical length of the medium is sufficiently short, the variations of detunings and intensities can be negligibly small.

Since it is only the time derivatives that enter the right-hand sides of equations (10), we can use expressions (9) and (7) for the atomic amplitudes in these equations and this will be equivalent to allowance for the first non-adiabatic corrections. Correspondingly, the conditions of smallness of rhs of (10) serve as a criteria of insignificance of changes in spatial and temporal characteristics of pulses and thus a criteria of adiabaticity of interaction in the medium. For simplicity we will restrict ourselves to the case of equal oscillator strengths in all transitions (in case of different oscillator strengths, we can proceed to the case of equal oscillator strengths in all transitions in usual lambda systems with the three-level 2-3-4 systems supported by two-level 1-2 and 4-5 systems in two-level-atom medium [12]. This peculiarity is important because both problems are studied in sufficient detail in literature and have analytical solutions. In particular, we can realize all phenomena taking place in usual lambda systems with the three-level 2-3-4 system which is supported by two-level 1-2 and 4-5 systems pumping level 2 and depleting level 4, respectively. As an example, we obtain, in the considered five-level system, propagation of the adiabaton [15] in the five-level system. Fig. 6 visualizes this phenomenon (details in figure caption).

In case of state \(|\lambda_2\rangle\) the system of equations (11) is interesting and important peculiarity. Propagation of fields \(\Omega_2\) and \(\Omega_3\) occurs independent of \(\Omega_1\) and \(\Omega_4\) and is described by propagation equations for three-level-atom medium in conditions of dark-state formation [1]. The fields \(\Omega_1\) and \(\Omega_4\) are described by propagation equations for two-level-atom medium [12]. This peculiarity is important because both problems are studied in sufficient detail in literature and have analytical solutions. In particular, we can realize all phenomena taking place in usual lambda systems with the three-level 2-3-4 system which is supported by two-level 1-2 and 4-5 systems pumping level 2 and depleting level 4, respectively. As an example, we obtain, in the considered five-level system, propagation of the adiabaton [15] in the five-level system. Fig. 6 visualizes this phenomenon (details in figure caption).

In case of state \(|\lambda_2\rangle\) the system of equations is no longer split.

Equations (11) are valid if state \(|\lambda_1\rangle\) is formed on entire length of medium. This requires fulfillment of two conditions: i) the detunings \(|\Delta_i| = \Delta\) for all \(i\) and Rabi frequencies \(\Omega_1 = \Omega_4\) and ii) the adiabaticity of interaction in all of the medium. Let us examine when these conditions are met. Equations (11) show that detunings \(\Delta_2\) and \(\Delta_3\) are preserved during propagation (as should be in a three-level system), whereas \(\Delta_1\) and \(\Delta_4\) can vary with propagation length because of self-phase modulation (as in two-level system), but, as shown in [12], these
On the same length we can take $\Omega_1 = \Omega_4$ (adiabatic approximation for two-level system). As follows from the results of cited works the adiabaticity of interaction in two-level system breaks at the lengths when $(q_1 x/\Delta^2 T) \sim 1$, whereas the interaction adiabaticity in three-level medium does not break at all. Another condition imposed on the length requires non-depletion of pump pulse in three-level medium for an effective population transfer.

$$\frac{q_1 L}{\Delta} \frac{1}{\Delta T} \ll 1,$$

(12)

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$$\frac{q_2,3 L}{\Delta} \frac{\Delta}{\Omega^2 T} \sim 1$$

(13)

It follows from (12) and (13) that the influence of medium is determined by the factor $q_{i} x/\Delta$, times the adiabaticity conditions for a single atom. This means that it is sufficient to require the medium parameter $q_{i} x/\Delta$ to not exceed unity by much. If we express this parameter in terms of the linear coefficient of absorption of medium $\alpha_0$, we obtain restriction for the optical length in the form:

$$\frac{q L}{\Delta} = \alpha_0 x \frac{\Gamma}{\Delta} \sim 1$$

(14)

with $\Gamma$ being the maximum of the relevant widths. So, in the case of large one-photon detuning the length of adiabaticity of interaction can exceed the length of linear absorption in medium several times. On this length equations (11) can be solved analytically and the solution has the form:

$$\Omega_1 = \Omega_4 = \Omega_{10}(\tau),$$

$$\Omega_2 = \Omega_0(\tau) \sin \theta_0(\xi(x, t)),$$

$$\Omega_3 = \Omega_0(\tau) \cos \theta_0(\xi(x, t))$$

(15)

where $\Omega_{10}$, $\Omega_0$, $\theta_0$ are the boundary conditions given at the entrance of the medium and $\xi(x, t)$ is an implicit function defined by the following expression

$$\int_\xi^\tau \Omega_0^2 dt = qx$$

(16)

FIG. 6. Distortion-free propagation of the signal pulse $\Omega_2$ at the subluminal group velocity. Shapes of pulses are chosen to be $\Omega_1 T = \Omega_4 T = \Omega_3 T = 25e^{-0.2x^2}$, $\Omega_2 T = 0.1e^{-5x^2}$ (linear case). The single-photon detuning is $\Delta = 50/T$. The scaled length $L = \Omega_2^2/(t = 0)T/q$ and the time delay in medium is $\Delta t/T = x/L$.

V. LIGHT STORAGE

It follows from the solution (15) that, after turning off all pulses, the coherence $\rho_{15}$ induced by these pulses remains in the medium (like in three level system):

$$\rho_{15} = -\sin \theta(\xi) \cos \theta(\xi)$$

(17)

where function $\xi(x)$ is defined by the following expression:

$$\int_\xi^\infty \Omega_0^2 dt = qx$$

(18)

Fig. 7 shows $x$-dependence of the $\xi$-function and coherence $\rho_{15}$ after all pulses are turned off, together with the input shape of probe pulse. Figure demonstrates that the distribution of coherence along $x$ mirrors the arbitrary $t$-shape of the probe pulse at medium entrance. It is also apparent that the $\xi$-function has two asymptotes, $x = 0$ and $x = x_{max}$. Existence of the maximal length of medium, i.e., the length where probe pulse disappears, is the essence of light storage phenomenon. For realization of storage (and mapping of $t$-dependence onto $x$-distribution) the medium must be not shorter than $x_{max}$. It follows from (13) that the maximal length is representable in the form $x_{max} = N/N_{ph}$, where $N$ is the number density of resonant atoms in medium and $N_{ph}$ is the overall photon fluxes in control and probe pulses at the medium input:

$$N_{ph} = \int_{-\infty}^{\infty} (cE_{p0}^2/h\omega_p + cE_{s0}^2/h\omega_s) dt$$

(19)
FIG. 7. The $x$-dependence of $\xi(x)$-function and the spatial distribution of coherence $\rho_{51}$ after the interaction is switched off (top) and the temporal profile of the pulse $\Omega_2$ at the medium input (bottom).

Thus, in order to write completely a light pulse into a medium, it is necessary that the number of atoms interacting with radiation be comparable with the total number of relevant photons. Note that $x_{\text{max}}$ does not depend on $\Omega_1$ and $\Omega_4$ (the latter enters only the adiabaticity condition). Note that in linear approximation in $\Omega_2$ we can construct a dark-state polariton similar to that in lambda system [7]. Fig. 8 shows, by means of numerical solution of corresponding equations with the use of Lax-Wendroff method [46, 47], writing of a light pulse into a medium. We emphasize that, as distinct from the generalized lambda-system where the described process of storage is more visualisable, but has little advantages as compared to the usual lambda system, the M-system is much more interesting because it enables double storage, i.e., we can write two different pulses, one after another with possibility of subsequent retrieval in any desired succession. Indeed, during the whole interaction time (and also after the first writing), the coherence $\rho_{31}$ remains zero. Thus, the population of the level 1 is close to unity in linear approximation in $\Omega_2$. This means that the same medium is now ready for the usual lambda-storage of the second pulse. The detailed study of the process of retrieval of optical information in a five-level system, of the efficiency of writing with allowance for influence of losses from the intermediate level, as well as of influence of the ratio of oscillator strengths of adjacent transitions, is performed in our preceding work.

FIG. 8. Propagation of the signal pulse $\Omega_2$ (light storage). Shapes of pulses are chosen to be $\Omega_1T = \Omega_4T = 25e^{(-0.2t^2)}$, $\Omega_3T = 25e^{(-t^2)}$, $\Omega_2T = 0.1e^{(-5t^2)}$. The single-photon detuning is $\Delta = 50/T$. The group velocity $u = c/(1 + qc/\Omega_2^2) \to 0$ when $\Omega_2^2 \to 0$

VI. CONCLUSION

We considered the behavior of a five-level atomic system and a medium of such systems driven by four laser pulses of different amplitudes and frequencies. We showed for such a system the possibility of analytical determination of system eigenfunctions and eigenvalues in case where all two-photon detunings are zero. We have obtained that, in addition to the traditional zero eigenvalue, there exists a non-zero one, for which the propagation equations in the medium are split into the equations for two- and three-level system media, i.e., two of four laser pulses travel independently of two other. This splitting is caused by the fact that in this case the five-level system reduces to a certain effective "lambda"-system whose ground states are superpositions of two states. We derive the dressed states and dressed energies of the system, as well as conditions for adiabatic evolution, and show that the length of medium where adiabaticity is preserved exceeds several times the linear absorption length.
We show that adiabatic passage permits reversible transfer of atomic population from an initial to a target state, and back again. The obtained mechanism of the population transfer may be employed for excitation of Rydberg states in atoms. We analysed the traveling of pulses in the medium and obtained, in particular, adiabaton (distortion-free) propagation at the group velocity lower than c. Also the process of information storage in five-level medium was examined. We propose a possibility of double storage of light pulses in the same medium with subsequent retrieval of the two stored pulses in desired sequence. We note that the relaxation processes have not been taken into account throughout the work. Allowance for these processes requires separate investigation. Finally we note that the considered five-level systems can experimentally be realized in a number of media, such as hyperfine structures of D-lines of alkali-metal atoms, in optical transitions of rare-earth-ion impurities in crystal matrices, in rovibrational levels of different electronic states in molecules, in problems of population transfer in entangled three two-level atoms and so on.

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APPENDIX A

Equation for eigenvalues of the interaction Hamiltonian, i.e., the equation $\det(H - \lambda I) = 0$, has, under conditions $\delta_2 = \delta_4 = 0$ and $\delta_1 = \delta_3 = \Delta$, the following form:

$$\lambda^2(\lambda - \Delta)(\lambda(\lambda - \Delta) + \Omega_1^2) + V^4\lambda = 0 \quad (20)$$

where $\Omega_1^2 = \Omega_1^2 + \Omega_2^2 + \Omega_3^2 + \Omega_4^2$ and $V^4 = \Omega_1^2\Omega_2^2 + \Omega_1^2\Omega_3^2 + \Omega_1^2\Omega_4^2$. With the notation $x = \lambda(\lambda - \Delta)$ the equation above becomes $\lambda[x^2 - \Omega_1^2x + V^4] = 0$ and the eigenvalues are obtained directly:

$$\lambda_0 = 0, \quad \lambda_{3,1} = \frac{1}{2}[\Delta \pm (\Delta^2 + 4x_1)^{1/2}], \quad \lambda_{4,2} = \frac{1}{2}[\Delta \pm (\Delta^2 + 4x_2)^{1/2}]$$

where $x_{2,1} = (1/2)[\Omega_1^2 \pm (\Omega_1^4 - 4V^4)^{1/2}]$. We note that the condition $\Omega_1^4 \geq 4V^4$ is always met.

Conditions of interaction adiabaticity for a single atom, $|\lambda_i - \lambda_j|T \gg 1$ for any $i \neq j$ with $T$ being the time of interaction, leads to following requirement imposed on the parameters of pulses.

$$\frac{(x_2 - x_1)T}{(\Delta^2 + 4x_{1,2}^2)^{1/2}} \gg 1,$$
$$\frac{(\Delta^2 + 4x_{1,2}^2)^{1/2}T \gg 1,}{x_{1,2}T} \gg 1$$.  

Note that the last condition can be fulfilled only for $V^4 \neq 0$, i.e., in the range of overlapping of pulses.

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