

# Manipulation of dark states and control of coherent processes with spectrally broad light

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(Received 7 September 2008; published 25 November 2008)

The formation of dark states under an interaction of degenerate atomic states with incoherent broadband radiation (white light) is discussed. A simple coupling scheme in a three level  $\Lambda$  system, which allows both qualitative and quantitative analysis, is discussed. We found a stationary solution of the optical Bloch equations in a broad excitation line approximation that describes the dynamics of the atom-white light interaction and demonstrated its identity to a conventional dark state created with coherent laser fields. We then examine the efficiency of the population transfer induced by broadband radiation in a model  $\Lambda$  system and revealed that high efficiency (attaining 100%) of stimulated Raman adiabatic passagelike processes can be achieved with certain temporal control of light polarization. The corresponding criterion of adiabaticity was formulated and justified by means of numerical simulations.

DOI: [10.1103/PhysRevA.78.053415](https://doi.org/10.1103/PhysRevA.78.053415)

PACS number(s): 32.80.Qk, 32.60.+i

## I. INTRODUCTION

Coherent laser fields can be used to manipulate atomic and molecular quantum states in order to create coherent superpositions of quantum states, which are of interest in many important fields of research including laser cooling [1], ultracold matter [2], dark states [3], electromagnetically induced transparency [4], and laser driven states [5]. These techniques have many important applications at the forefront of technology and industry, including in such areas as lithography [6], quantum information [7], quantum chemistry [8], and others. Typically, control of quantum states is implemented within  $\Lambda$ -type and V-type systems driven by two (or more) laser fields. In the case of  $\Lambda$ -type excitation, dark (or population trapping) states ( $D$  states) can be formed, which become transparent for photons and thus cease interacting with light [3]. In contrast, another type of superposition of coherent states, bright states ( $B$  states), becomes more absorbing under the laser light action [9–11]. The dark states are useful in laser cooling processes [2,12], optical pumping [13–15], “lasing without inversion” [16,17], and others. One particular application of dark states is in the method of Stimulated Raman Adiabatic Passage (STIRAP), which can be used to couple an initial and final state to a common intermediate state and transfer atomic populations between initial and final state without loss [5]. This method has traditionally required highly stabilized laser fields that are strongly phase correlated. But are these requirements really so strict as traditionally thought? In this paper we discuss a scheme which allows one to achieve a high degree of control of quantum states by means of broadband (i.e., basically white) light from a single light source.

The basic equation that describes coherent processes in light-atom interaction under the density matrix formalism is the Liouville equation, which in atomic physics often is re-

ferred to by the term “optical Bloch equations.” When these equations describe a system illuminated by spectrally broad light, they formally allow a steady-state solution that has the structure of a pure quantum state similar to the solution for dark states in monochromatic laser fields. It turns out that spectrally broad light can form dark states thanks to a beneficial cancellation of photons of different frequencies. This result evidently opens new possibilities for controlling atomic states with incoherent light sources. As a specific case, we will discuss in detail the problem of transferring a population of atoms with spectrally broad light from an initial, stable discrete state to a desired target state, without loss of population.

The paper is organized as follows. In Sec. II we shall review the optimal scenario for population transfer; we discuss the nature of coherent dark states under monochromatic excitation and outline the main factors of the atom-coherent-light interaction that can destroy the  $D$  states by mixing them with bright states. In Sec. II B we shall consider cases that limit the control over coherence, such as two lasers that are only partially coherent. We will then be in a position to formulate the arguments for and against the possibility of using spectrally broad light to manipulate quantum states. The answer will be obtained in Sec. III. In Sec. III A we shall analyze the master equation for the density matrix that describes the dynamics of the atom-spectrally broad light interaction [18,19]. In Sec. III B we shall demonstrate the existence of  $D$  states under spectrally broad light excitation and coherent processes with broadband excitation. Then, in Sec. IV we shall present the results of numerical simulations that demonstrate that population transfer by spectrally broad light can achieve an efficiency of 100%. The effects of detuning from the two-photon resonance will be analyzed in Sec. IV B, and numerical simulations of the corresponding two photon line shapes will be presented. In Sec. V we will summarize the results and discuss the possibilities of coherent processes in atoms in the case of spectrally broad light excitation. Finally, in order to justify some statements related to  $D$  states and STIRAP in the case of spectrally broad light sources, an analogy between coherent and incoherent process for quantum state control is established in the Appendix.

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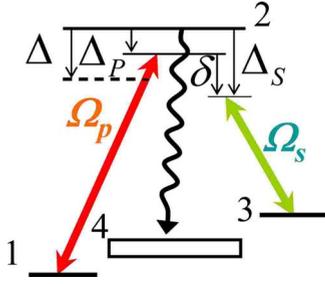


FIG. 1. (Color online) Schematic diagram of energy levels, laser Rabi frequencies, and detunings. The initial population is embedded on level 1.

## II. FORMULATION OF THE PROBLEM: ADIABATIC PASSAGE WITHIN BOUND STATES

We shall concentrate ourselves on the problem of how to transfer populations of atoms from an initial level 1 to some target level 3 (see Fig. 1) without loss of population. We first briefly recall the case of two coherent, *monochromatic* pulsed lasers, a pump laser and a Stokes laser, with fixed frequencies  $\omega_p, \omega_s$  and corresponding Rabi frequencies  $\Omega_p(t), \Omega_s(t)$  [5]. It is clear that, in any atom manipulation scheme, one should avoid involving the unstable upper-lying state 2, because from this state population could flow into other unwanted states, which are schematically depicted in Fig. 1 as a single level 4. Our first task is to find out how to create a wave function  $\Psi_D = C_1\Psi_1 + C_3\Psi_3$  as a linear combination of the two low-lying states 1 and 3, which is not coupled to the excited state 2. In other words, if the Hamiltonian is  $H = H_0 + V$ , where  $V$  describes the coupling of the system levels with light, we require that the matrix element  $\langle\Psi_2|V|\Psi_D\rangle$  is zero.

### A. Dark and bright states

In the rotating wave approximation, the total Hamiltonian  $H = H_0 + V$  of the system depicted in Fig. 1 has the well known form [20]

$$H = \hbar/2 \begin{bmatrix} 2\Delta_p & \Omega_p(t) & 0 \\ \Omega_p(t) & 0 & \Omega_s(t) \\ 0 & \Omega_s(t) & 2\Delta_s \end{bmatrix} \quad (1)$$

in the basis of the bare states  $\Psi_i (i=1, 2, 3)$ . The Hamiltonian  $H_0$  corresponds to a free atom and determines the bare state energies  $\varepsilon_i$ . We choose as the zero level of energy the value  $\varepsilon_2$  for the excited state 2. The quantities  $\Delta_{p,s}$  in the diagonal elements give the laser detunings ( $\Delta_p = \omega_p - \omega_{21}$ ;  $\Delta_s = \omega_s - \omega_{23}$ ) from the Bohr frequencies  $\omega_{21}, \omega_{23}$  of the corresponding optical transitions (see Fig. 1). The Rabi frequencies  $\Omega_p(t), \Omega_s(t)$  are determined from the coupling term  $V$  of the Hamiltonian. We neglect to mention here any relaxation terms and leave their proper discussion to Sec. III.

The required solution  $\Psi_D(t)$  of the equation  $\langle\Psi_2|V|\Psi_D\rangle = 0$  reads [5]

$$\Psi_D = \cos \Theta(t)\Psi_1 - \sin \Theta(t)\Psi_3,$$

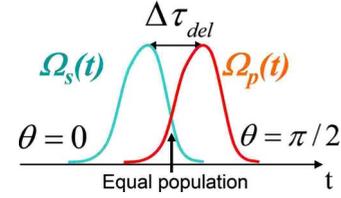


FIG. 2. (Color online) A sequence of laser pulses. The arrow indicates a moment when the populations of levels 1 and 3 are equal.

$$\sin \Theta = \Omega_p / \Omega_{eff}, \quad \Omega_{eff} = \sqrt{|\Omega_p|^2 + |\Omega_s|^2}, \quad (2)$$

and is known as a dark state. Since  $\Psi_D(t)$  does not share the population with the excited state, it does not radiate directly itself. Note that the mixing angle  $\Theta$  gives a convenient measure of population sharing between stable states: the value  $\Theta=0$  corresponds to a population that resides entirely in state 1, while  $\Theta=\pi/2$  describes a population that has been transferred entirely to the target state 3.

Since controlling the population means controlling the mixing angle, as can be seen from Eq. (2), an efficient transfer of population can be achieved by organizing a sequence of dark states with the mixing angle varying from  $\Theta=0$  (the initial state 1 populated) to  $\Theta=\pi/2$  (the target state 3 populated). In this sequence of states, the state vector  $\Psi(t)$  should be maintained as the current dark states  $\Psi_D(t)$  all the time. An experimental implementation of such a scenario is depicted in Fig. 2. The pulse sequence that provides the desired rotation of the mixing angle seems counterintuitive [21]: the pump laser pulse arrives after the Stokes laser pulse. What happens is that the Stokes laser prepares (dresses) the transition  $2 \rightarrow 3$  for accepting the population, which is delivered by the pump laser [5]. It is noteworthy that the states 1, 3 share the population equally at the moment when both lasers' Rabi frequencies are equal.

We now examine different unwanted factors that can restrict the efficiency of the desired population transfer. For this purpose, we consider another convenient concept, the so called bright state ( $B$  state)  $\Psi_B$  [5] as follows:

$$\Psi_B = \sin \Theta(t)\Psi_1 + \cos \Theta(t)\Psi_3,$$

$$\langle\Psi_2|V|\Psi_B\rangle / \hbar = \Omega_{eff}, \quad (3)$$

which is orthogonal to the dark state  $\Psi_D$ . Although the  $B$  state also does not contain the excited state 2, it is always mixed with the excited state by the light fields, as can be seen from the corresponding matrix element presented in Eq. (3). Having been coupled to the excited state, the  $B$  state thus is power broadened, and this coupling leads to radiation from state 2, which results in unwanted population losses to the marginal levels 4 due to spontaneous transitions (see Fig. 1).

As was mentioned above, although the  $D$  state is not coupled to state 2 and does not lead to radiation from that state, nevertheless it may be coupled with the bright state. The mixing frequency between the bright and dark states during the system's temporal evolution reads [20]

$$\langle \Psi_B | \frac{H}{\hbar} + i \frac{\partial}{\partial t} | \Psi_D \rangle \equiv i \frac{d\Theta}{dt} - \frac{\delta}{2} \sin 2\Theta, \quad (4)$$

where  $H$  is the Hamiltonian (1), and the temporal derivative corresponds to nonadiabatic linkage between states. Two important requirements follow from relation (4). First, to preserve the dark state, changes of the mixing angle [see Eq. (2)] should be slow enough, or adiabatically organized. The corresponding criterion is given via the inequality  $d\Theta/dt \ll \Omega_{eff}(t)$ , which yields, after integration over  $t$  [5],

$$\int_{-\infty}^{\infty} dt \Omega_{eff}(t) \gg \Delta\Theta = \pi/2. \quad (5)$$

It is seen that the applied laser pulses should be stronger than the  $\pi$  pulses. The second important requirement concerning the mixing of  $B$  and  $D$  states is that the difference  $\delta = \Delta_S - \Delta_P$  between laser detunings should be small. This difference  $\delta$  (see Fig. 1) is often called the double-photon detuning, and it opens a pathway for unwanted population flow, which may dramatically destroy the  $D$  state. A detailed study of the efficiency of STIRAP-like processes as a function of  $\delta$  (the so-called two-photon line shape) may be found in [22]. Note that the one photon detuning  $\Delta$ , determined as  $\Delta = 1/2(\Delta_S + \Delta_P)$  (see Fig. 1), does not enter itself into the mixing matrix element in Eq. (4), which explains the weak influence of  $\Delta$  on the population transfer [5,22].

### B. Phase-diffusion effects for partially coherent fields

Up to now we dealt with coherent radiation. However, in the real world, lasers typically are subject to vibrations and other environmental influences that cause phase diffusion and result in only partial coherence of the laser fields. Phase diffusion effects were studied in detail by [20] using the phase diffusion model, according to which the random walk of laser frequencies varies chaotically both the double-photon detuning  $\delta$  and the single-photon detuning  $\Delta$ . Drift of the one-photon detuning is not detrimental to STIRAP, but strong  $\delta$ -chaotic jumps dramatically decrease the STIRAP efficiency. However, the authors of paper [23] pointed out an important exception: if the radiation in both laser fields has the same source, a beneficial cancellation of the phase fluctuations may occur. Since laser phases are varying equally, the value of  $\delta$  remains equal to zero.

### III. DARK STATES IN SPECTRALLY BROAD LIGHT

With the above preliminaries, we are now ready to determine if quantum states can be controlled by means of spectrally broad light instead of coherent lasers. From Sec. II B it is clear that in case of spectrally broad light one has to use a single light source. Otherwise, if two distinct uncorrelated sources of broadband light were to be used, every dark state would be depopulated by mutual, multiple incoherences among the sources. In our analysis we deal with a fluctuating electric field  $\mathbf{E}(t)$  that has a well defined elliptical polarization

$$\mathbf{E}(t) = \text{Re } \mathbf{E}_0(t) \exp(-i\omega_0 t) \varepsilon(t),$$

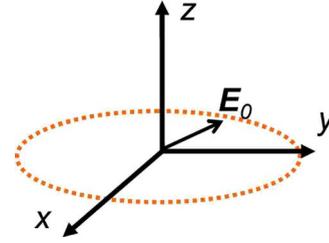


FIG. 3. (Color online) Light polarization.

$$\langle \varepsilon(t_1) \varepsilon^*(t_2) \rangle = A(t_1 - t_2), \quad (6)$$

where  $\omega_0$  is the carrier frequency of the light. We assume that the fluctuating part  $\varepsilon(t)$  of the light is a scalar, dimensionless, random, complex function of unit modulus  $|\varepsilon| = 1$  with the broadband correlation function  $A(t_1 - t_2)$ . As a result, the spectral distribution  $P(\omega)$  of the light [24]

$$P(\omega) = \frac{1}{\pi} \text{Re} \int_0^{\infty} dt \exp(-i\omega t) A(t) \quad (7)$$

is a smoothly varying function within the spectral interval  $\Delta\omega$  of interest. The application of adiabatic elimination procedure implies the following requirement  $\tau\Delta\omega \gg 1$  for the characteristic duration  $\tau$  of the matter-light interaction, which is directly related to the time of switching on and switching off the light beam [19,26]. When the interaction takes place during a very short time interval, i.e., when  $\tau$  becomes very small, the correlation function  $A(t_1 - t_2) \sim \delta(t_1 - t_2)$  should correspond to spectrally broad light (here  $\delta$  is the Dirac delta function). In the case of slow processes, i.e., in the case of adiabatic control of quantum states, the spectral interval  $\Delta\omega$  may be of finite size. It is important to note that the frequency  $\omega$  in Eq. (7) denotes a measure of the frequency shift from the center  $\omega_0$  of the light spectra.

In contrast, the envelope vector function  $\mathbf{E}_0(t)$  has regular behavior and determines the light polarization. If one chooses the direction of light propagation as the  $z$  axis (see Fig. 3), the polarization ellipse lies in the  $(x, y)$  plane. It is convenient to work with the polarization elements  $E^{(\pm 1)}$  represented by spherical components  $\mathbf{e}_{\pm}$  [25] of vector  $\mathbf{E}_0$ .

$$\mathbf{E}_0(t) = E^{+1}(t) \mathbf{e}_+ + E^{-1}(t) \mathbf{e}_-,$$

$$\mathbf{e}_{\pm} = \mp (\mathbf{e}_x \pm i\mathbf{e}_y) / \sqrt{2}. \quad (8)$$

When  $E^{(\pm 1)}$  are real, the polarization of light is such that the main semiaxes of the ellipse are oriented along the  $\mathbf{e}_x$  and  $\mathbf{e}_y$  axes. If  $E^{(\pm 1)}$  are complex numbers, the difference between their phases determines the double rotation angle of the ellipse in the  $(x, y)$  plane.

The quantum states may be controlled through an appropriately chosen, time-dependent variation of the light polarization. As an example we consider the simple interaction scheme presented in Fig. 4: a single broadband light beam excites a two-level system. The excited state ( $e$  state) has the angular momentum  $l=0$  and consists of one Zeeman component  $m'=0$ . The ground state ( $g$  state) possesses angular momentum  $l=1$  and therefore has three components, one of

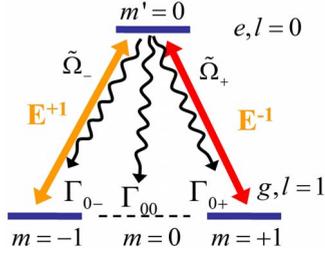


FIG. 4. (Color online) Energy level diagram.

which ( $m=0$ ) is not involved in the interaction because of the chosen light polarization plane. The light's central frequency  $\omega_0$  is assumed to be in resonance with the  $g \rightarrow e$  transition. The quantization axis is oriented along the  $z$  direction. As is apparent, the spherical component of light (8) couples independently the transitions  $m = \pm 1 \rightarrow m' = 0$ . In fact, the magnetic sublevels involved in this interaction effectively form a three-state  $\Lambda$  scheme.

In our model, we assume that at  $t = -\infty$  only the  $m = -1$  component is populated. Our aim is to analyze the efficiency of the STIRAP-like process that could transfer the population from  $m = -1$  to  $m = +1$ . This population transfer can be accomplished by applying a sequence of light pulses with Rabi frequencies  $\Omega_{\mp}$  (see Fig. 2) by varying (by changing light polarization) the relative strength of the polarization components  $E^{(\pm 1)}$ .

$$E^{(\pm 1)}(t) = E_S \exp[-(t - \Delta\tau_{\mp})^2/2\tau^2],$$

$$\Omega_{\mp}(t) = E^{(\pm 1)}(t) \langle m' = 0 | \mathbf{e}_{\pm} \cdot \mathbf{d} | m = \mp 1 \rangle / \hbar. \quad (9)$$

Here  $\mathbf{d}$  denotes the atomic dipole moment. The Rabi frequencies  $\Omega_{\pm}$  correspond to the strengths of the coupling interaction between the levels  $m = \pm 1$  and  $m' = 0$ , which is induced by the light's spherical components  $E^{\mp}$  (see Fig. 4). The parameter  $\tau$  in the arguments of the exponential factors determines the interaction time, while the parameters  $\Delta\tau_{\pm}$  give the temporal shifts of the applied impulses. It is worth emphasizing an important feature of the scheme presented here. Clearly, each frequency  $\omega$  of the light beam stimulates both transitions  $m = \pm 1 \rightarrow m' = 0$  with effective partial Rabi frequencies  $\tilde{\Omega}_{\pm}(\omega) = \Omega_{\pm} \sqrt{P(\omega)}$  and results in the appearance of dark states (2). Because the corresponding mixing angle  $\sin \Theta = \tilde{\Omega}_{+} / \sqrt{\tilde{\Omega}_{+}^2 + \tilde{\Omega}_{-}^2}$  turns out to be independent of  $\omega$ , the photons prepare a unique dark state, which therefore is not coupled to the upper excited state  $m' = 0$ .

Here, however, new additional unwanted factors in the formation of the dark state arise. Indeed, spectrally broad light actually consists of many uncorrelated photons with different frequencies  $\omega_0 + \omega$ . A photon with the fixed frequency  $\omega_0 + \omega$  that excites the transition  $m = +1 \rightarrow m' = 0$  [the Rabi frequency  $\tilde{\Omega}_{+}(\omega)$ ] combines with a variety of photons of frequencies  $\omega_0 + \tilde{\omega}$  that excite the transition  $m = -1 \rightarrow m' = 0$  [the Rabi frequency  $\tilde{\Omega}_{-}(\tilde{\omega})$ ]. We could expect the presence of many nonzero two-photon resonance detunings  $\delta = \omega - \tilde{\omega}$  to lead to strong mixing between the dark and bright states [see Eq. (4)], i.e., to a fast destruction of the dark state.

However, there is one favorable circumstance: it is possible to distribute  $\tilde{\omega}$  frequencies into pairs  $\tilde{\omega}_{1,2}$  with opposite two-photon detuning values  $\delta_1 = -\delta_2$ , so that the average value  $\langle \delta_{1,2} \rangle$  over the pair becomes zero. As a result, it may be possible to compensate the pair contribution in the mixing between dark and bright states. The total rate of unwanted population loss due to coupling to the  $B$  state could still be reduced to zero despite the presence of many light frequencies. To verify this hypothesis, we need a robust treatment of the dynamics of the system.

#### A. Basic equation for the density matrix under coupling with spectrally broad light

The evolution of the system should be studied within the framework of the density matrix  $\rho_{ij}$  formalism. At first glance, the problem seems to be intractably complicated because of the presence of multichromatic light. Fortunately, the broad spectrum of applied spectrally broad light results in an adiabatic elimination of the optical coherences [19,26] in the optical Bloch equations, i.e.,  $\rho_{eg}$  in our case. Briefly, under the action of spectrally broad light, the effective lifetime  $\tau_{ex} \sim 1/\Delta\omega$  of induced optical dipoles described by  $\rho_{eg}$  appears to be very short in comparison with the interaction time  $\tau$ . The optical dipoles adiabatically follow only the  $\rho_{ee}, \rho_{gg}$  elements (Zeeman coherences). The diagram techniques for solving the evolution of the density matrix developed by Konstantinov and Perel [27] and later by Keldish [28] justify the results that were first empirically obtained by Claude Cohen-Tannoudji [18] in the form of rate equations for the Zeeman coherences  $\rho_{ii}$  under the broad-line approximation. A detailed study of various problems under broad-line approximation may be found in [19]. In particular, the equations presented in [19] and adopted for our system (Fig. 4) may be easily reduced to the following system of equations, which describes the populations of the  $e$ -state ( $\rho'_{00}$ ), the  $g$ -state ( $\rho_{++}, \rho_{--}$ ), and the off-diagonal ( $\rho_{+-}, \rho_{-+}$ ) elements between  $g$ -state components  $m = \pm 1$  (Zeeman coherences):

$$\begin{aligned} \frac{d}{dt} \rho'_{00} = & -(|\tilde{\Omega}_{+}|^2 + |\tilde{\Omega}_{-}|^2 + \Gamma_0) \rho'_{00} + |\tilde{\Omega}_{+}|^2 \rho_{++} + |\tilde{\Omega}_{-}|^2 \rho_{--} \\ & + 2 \operatorname{Re} \tilde{\Omega}_{-} \tilde{\Omega}_{+}^* \rho_{-+}, \end{aligned} \quad (10)$$

$$\frac{d}{dt} \rho_{++} = -|\tilde{\Omega}_{+}|^2 \rho_{++} + (|\tilde{\Omega}_{+}|^2 + \Gamma_{0+}) \rho'_{00} - \operatorname{Re} \tilde{\Omega}_{-} \tilde{\Omega}_{+}^* \rho_{-+}, \quad (11)$$

$$\frac{d}{dt} \rho_{--} = -|\tilde{\Omega}_{-}|^2 \rho_{--} + (|\tilde{\Omega}_{-}|^2 + \Gamma_{0-}) \rho'_{00} - \operatorname{Re} \tilde{\Omega}_{-} \tilde{\Omega}_{+}^* \rho_{-+}, \quad (12)$$

$$\begin{aligned} \frac{d}{dr} \rho_{-+} = & \left[ 2i\omega_L - \frac{1}{2} (|\tilde{\Omega}_{+}|^2 + |\tilde{\Omega}_{-}|^2) \right] \rho_{-+} \\ & - \frac{1}{2} \tilde{\Omega}_{-}^* \tilde{\Omega}_{+} (\rho_{++} + \rho_{--} - 2\rho'_{00}). \end{aligned} \quad (13)$$

Note that  $\rho_{+-} = \rho_{-+}^*$ .

The structure of system (10)–(13) contains two types of terms. First, the majority of its terms contain the populations and, hence, represent a simple balance between population flow into different levels. Second, the contribution of coherence effects is represented by the Zeeman coherence element  $\rho_{+-}$ . Briefly, the spontaneous radiative constant  $\Gamma_0$  gives the total rate of the excited state  $m'=0$  decay into the low-lying levels. The values  $\Gamma_{0i}$  ( $i=-, 0, +$ ) yield the partial spontaneous transition rates from the  $e$  state to the individual  $i$  components of the  $g$  state; clearly  $\Gamma_0 = \Gamma_{0-} + \Gamma_{00} + \Gamma_{0+}$ . In our particular case (Fig. 4), one has  $\Gamma_{0i} = \Gamma_0/3$  [29]. The light polarization components  $E^{\pm 1}$  generate transitions, including stimulated transitions, between the excited and ground state. The efficiency of the coupling of states is determined by the effective “frequencies”  $\tilde{\Omega}_{\pm}$ , which turn out to be expressed via the above introduced Rabi frequencies  $\Omega_{\pm}(t)$  (9)

$$\tilde{\Omega}_{\pm}(t) = \hbar \sqrt{P(\omega=0)} \Omega_{\pm}(t) = \sqrt{P(\omega=0)} E^{\mp 1}(t) \|d\| / \sqrt{3}. \quad (14)$$

The parameter  $\|d\|$  corresponds to the reduced dipole matrix element [29], while the coefficient  $\sqrt{3}$  arises from 3- $j$  symbol related to  $m = \pm 1 \rightarrow m' = 0$  transitions according to the Wigner-Eckart theorem. Note that because of the factor  $\sqrt{P}$ , the dimension of  $\tilde{\Omega}_{\pm}$  is  $[s^{-1/2}]$ . The purely imaginary term in Eq. (13) appears in the presence of an external magnetic field that results in the Larmor energy shift  $m\hbar\omega_L$  of Zeeman components  $m$ , i.e., in energy splitting  $2\hbar\omega_L$  between  $m = -1$  and  $m = +1$  components.

It is convenient to measure the scale of the coupling in units of  $E_S$  (9), so that the pulse sequences (9) acquire the form

$$\begin{aligned} \tilde{\Omega}_{\pm}(t) &= \tilde{\Omega}_0 \exp[-(t - \Delta\tau_{\pm})^2 / 2\tau^2], \\ \hbar\tilde{\Omega}_0 &= \sqrt{P(\omega=0)} E_S \|d\| / \sqrt{3}, \end{aligned} \quad (15)$$

with  $\tilde{\Omega}_0$  again measured in  $s^{-1/2}$ .

Coherent processes, as was mentioned above, are described by the dynamics of the off-diagonal elements  $\rho_{+-} = \rho_{-+}^*$ . Because of these off-diagonal elements the balance equations result in some important specific features, such as, for instance, the existence of dark states. In particular:

(i) The system of Eqs. (10)–(13): describes an open system because of the presence of a spontaneous cascade into the uncoupled  $m'=0$  component of the  $g$  state. In accordance with Eqs. (10)–(13),

$$\frac{d}{dt}(\rho'_{00} + \rho_{++} + \rho_{--}) = -\Gamma_{00}\rho'_{00} \quad (16)$$

population flow into the uncoupled level depletes the population of the coupled system. Correspondingly, the population  $\rho_{00}$  of the ground  $m=0$  component may be found from the following equation:

$$\frac{d}{dt}\rho_{00} = \Gamma_{00}\rho'_{00}. \quad (17)$$

(ii) The initial conditions imply the absence of any coherence, i.e.,  $\rho_{+-}(t=-\infty) = 0$ . It is clearly seen from Eq. (13) that, if a magnetic field is absent ( $\omega_L = 0$ ) and  $\tilde{\Omega}_+ \tilde{\Omega}_-$  has a real value (i.e., the  $x, y$  axes are the main diagonals of the polarization ellipse), the imaginary part of the off-diagonal element  $\rho_{-+}$  remains zero.

(iii) The main feature of the case that occurs when  $\omega_L = 0$  is that the system (10)–(13) has a unique simple stationary solution which does not include the population of the excited state

$$\begin{aligned} \rho'_{00} &= 0, \quad \rho_{ii} = \frac{\tilde{\Omega}_- \tilde{\Omega}_-^*}{|\tilde{\Omega}_+|^2 + |\tilde{\Omega}_-|^2}, \\ \rho_{ij} &= -\frac{\tilde{\Omega}_i \tilde{\Omega}_j^*}{|\tilde{\Omega}_+|^2 + |\tilde{\Omega}_-|^2} \quad (i, j = +, -). \end{aligned} \quad (18)$$

### B. Dark states formed by spectrally broad light

The density matrix in Eq. (18) obviously corresponds to a pure quantum state and allows one to determine an angle for mixing the  $m = -1$  and  $m = +1$  sublevels of the ground state, as in the case of coherent laser fields. Since the excited state is not populated, the stationary solution (18) describes a dark state that has been formed with spectrally broad light. This result implies a beneficial cancellation of contributions from incoherent frequency components of spectrally broad light and consequently opens up new perspectives on the fruitful control of atomic states.

It is noteworthy that, under the absence of Zeeman separation ( $\omega_L = 0$ ), there is only a single stationary state of the density matrix that allows  $\rho'_{00} = 0$ . To prove this statement, let us set the left-hand side of the system (10)–(13) to be zero. Then Eqs. (11) and (12) for  $\rho'_{00} = 0$  yield

$$|\tilde{\Omega}_+|^2 \rho_{++} = -\text{Re} \tilde{\Omega}_- \tilde{\Omega}_+^* \rho_{-+} = |\tilde{\Omega}_-|^2 \rho_{--}. \quad (19)$$

Since the system (10)–(13) is a linear one, its solution may be normalized by one parameter; we may choose  $\rho_{++}$ , for instance, in the form  $\rho_{++} = |\tilde{\Omega}_-|^2 / (|\tilde{\Omega}_+|^2 + |\tilde{\Omega}_-|^2)$ . With such a choice, the relations in Eq. (19) acquire the form of Eq. (18), which therefore turns out to be unique. The normalization adopted in Eq. (18) follows from the requirement that  $\rho_{++} + \rho_{--} = 1$ , which reflects the population conservation. Significantly, the stationary case of Eqs. (10) and (13) (with zero the left-hand side) is satisfied automatically by the solution (18) provided that  $\omega_L = 0$ . The fact that the dark states formed by spectrally broad light are unique fits well with our previous qualitative considerations about  $D$  states [see the discussion after Eq. (9)]. In the Appendix we shall further explore why the density matrix in Eq. (18) is a solution of the master equations (10)–(13).

The density matrix in Eq. (18) corresponds to the pure quantum state with the  $\Psi$  function whose components have amplitudes  $c_i$ ,

$$c_+ = \tilde{\Omega}_- / \sqrt{|\tilde{\Omega}_+|^2 + |\tilde{\Omega}_-|^2},$$

$$c_- = -\tilde{\Omega}_+ / \sqrt{|\tilde{\Omega}_+|^2 + |\tilde{\Omega}_-|^2}, \quad c'_0 = 0. \quad (20)$$

In the case of a conventional  $\Lambda$  scheme with two coherent lasers of Rabi frequencies  $\tilde{\Omega}_i$  (which are complex in general), the coherent dark state in Eq. (2) has the same amplitude as determined in Eq. (20). Therefore, it is possible to analyze the situation with our spectrally broad light or “white light” dark states (*WD* states) using the rich information known about standard coherent *D* states. In particular, in the next section we will be able to express quantitatively to what extent the state control process needs to be adiabatic in order to ensure the survival of *WD* states under nonstationary conditions.

#### IV. SPECTRALLY BROAD LIGHT STIRAP

There are many interesting applications of the *WD* state. First of all, since the *WD* state (18) that arises in the scheme presented in Fig. 4 turns out to have the standard structure (20), it would be beneficial to consider the most recent developments in the applications of conventional dark states [3,5] when exploring applications of the *WD* state.

##### A. Population transfer

We start by examining now the efficiency of the STIRAP-like population transfer from the  $m=-1$  component to the  $m=+1$  component of the ground state when the analogue of the two-photon resonance detuning  $\delta=2\omega_L$  is zero [see the clarifying remarks in the Appendix after Eqs. (A3)–(A6)]. Initially, at ( $t=-\infty$ ) the population was found in the sublevel  $m=-1$  in a pure state that corresponds to

$$\begin{aligned} \rho_{--} &= 1, & \rho_{ii} &= 0 \quad (i \neq -), \\ \rho_{ij} &= 0 \quad (i \neq -j). \end{aligned} \quad (21)$$

We now consider how to drag most efficiently the population without loss from the  $m=-1$  sublevel to the  $m=+1$  sublevel by dynamically changing the properties of the light. Based on previous experience with STIRAP (Fig. 1), the mixing angle  $\Theta$  [see Eq. (2)] should be changed from 0 to  $\pi/2$ . Therefore, one has to modulate the polarization of the light in such a manner that the corresponding effective Rabi frequencies acquire pulse forms (15) in a necessary sequence: pulses are offset in time, and in the pulse sequence the  $\tilde{\Omega}_+(t)$  pulse should arrive first, i.e.,

$$\Delta\tau_{del} = \Delta\tau_- - \Delta\tau_+ > 0. \quad (22)$$

The pulses have duration  $\tau$ , and the temporal shift  $\Delta\tau_{del}$  between them has to be positive.

The efficient population transfer (without loss) should be performed adiabatically via *D* states. Any deviation from adiabaticity results in mixing between the state coupled to the excited state (the bright state) and the *WD* state that allows the population flow into an unwanted *g* level with  $m=0$ . The condition for implementing the adiabatic passage in the case of coherent lasers was discussed above and is expressed by relation (5). In the Appendix we derive a modi-

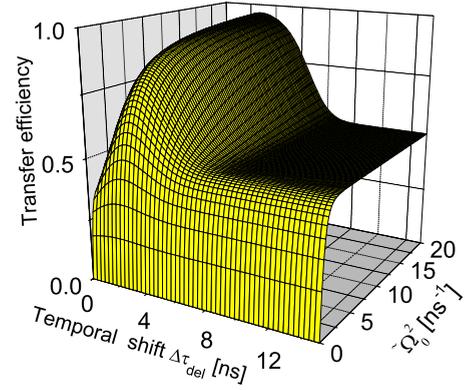


FIG. 5. (Color online) Simulation of STIRAP with white light. The population transfer efficiency was found from numerical solutions of system (10)–(13) and is plotted versus the square of the effective Rabi frequency  $\tilde{\Omega}_0^2$  ( $\text{ns}^{-1}$ ) and the temporal shift  $\Delta\tau_{del}$  (ns) between the pulses. The radiative decay constant is  $\Gamma_0=9 \text{ ns}^{-1}$ , while the duration of pulses is  $\tau=2 \text{ ns}$ .

fication of criterion (5) for spectrally broad light excitation

$$\tilde{\Omega}_0^2 \tau \gg 1, \quad (23)$$

where the effective frequency  $\tilde{\Omega}_0$  is defined in Eq. (15). This last requirement ensures the adiabaticity of STIRAP in the case of system (10)–(13).

In the Appendix we will examine as well the structure of the density matrix equation [see Eqs. (A3)–(A6)] for the case of two coherent laser fields with a small two-photon detuning  $\delta$  and a relatively large one-photon detuning  $\Delta$  (see Fig. 1). Such a one-photon detuning makes it possible to adiabatically eliminate the optical coherence elements, which reveals a close analogy between coherent [Eqs. (A3)–(A6)] and incoherent dynamics [Eqs. (10)–(13)]. With this analogy in mind, intuitively one may expect to attain high efficiency with spectrally broad light STIRAP. Figure 5 illustrates the above conclusions by showing the results of numerical simulations of population transfer. In this example we fix the duration of the pulses to  $\tau=2 \text{ ns}$ . The transfer efficiency is measured as the population  $\rho_{++}^{(f)} = \rho_{++}(t=\infty)$  of the target state after the pulse sequence concludes. Note that the surfaces of  $\rho_{++}^{(f)}$ , presented in Fig. 5 have properties identical to the case of coherent lasers, as is shown in the Appendix. The value  $\rho_{++}^{(f)}$  is optimal, for instance, when  $\Delta\tau_{del} \approx \tau$  [5]. The data exhibited in Fig. 5 illustrate as well criterion (23) of transfer adiabaticity: if we set a criterion for a successful population transfer at the level of  $\rho_{++}^{(f)}$  equal to 0.9, the saturation starts in the region  $\tilde{\Omega}_0^2 \tau > 10$ . If one wants to increase the efficiency of population transfer (increase the value of  $\rho_{++}^{(f)}$ ) further, one needs to increase  $\tilde{\Omega}_0^2 \tau$ . Note that the simulation shows that a variation of the decay constant  $\Gamma_0$  by up to an order of magnitude does not influence significantly the efficiency as a function of the pulse area  $\tilde{\Omega}_0^2 \tau$  and the delay  $\Delta\tau_{del}$  between the pulses.

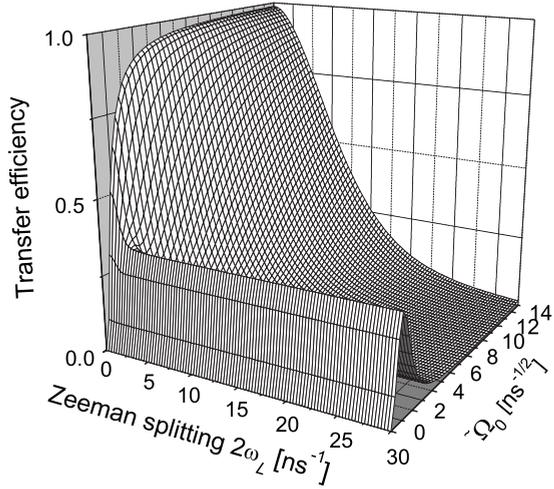


FIG. 6. Simulation of STIRAP with white light. Population transfer efficiency is plotted versus the effective Rabi frequency  $\tilde{\Omega}_0$  ( $\text{ns}^{-1/2}$ ) and Zeeman splitting  $2\omega_L$  ( $\text{ns}^{-1}$ ). The temporal shift  $\Delta\tau_{del}$  between the pulses is chosen to be 2 ns. The radiative decay constant is  $\Gamma_0=9 \text{ ns}^{-1}$  and the duration of pulses is  $\tau=2 \text{ ns}$ .

### B. Influence of Zeeman splitting

It is of particular interest for practical applications to examine what happens when a weak magnetic field is present. The presence of Zeeman splitting  $\omega_L$  in system (10)–(13) formally corresponds to two-photon detuning with the value  $\delta=2\omega_L$  (see the Appendix). The *WD* state (18) fails to be a stationary solution of system (10)–(13), as the terms corresponding to  $\omega_L$  mix the dark state with the bright state, and some fraction of the *D*-state population flows to unwanted states: *g* state  $m=0$  and the initial state  $m=-1$ . Figures 6 and 7 give some insight into how the unwanted processes change the desired transfer efficiency. We choose the situation with an optimal delay  $\Delta\tau_{del}=2 \text{ ns}$  in the pulse sequences and set

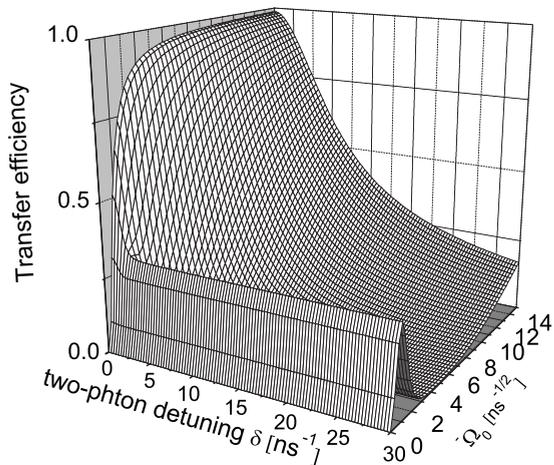


FIG. 7. Simulation of STIRAP with coherent lasers. Population transfer efficiency is plotted versus the effective Rabi frequency  $\tilde{\Omega}_0$  ( $\text{ns}^{-1/2}$ ) and two-photon detuning  $\delta=2\omega_L$  ( $\text{ns}^{-1}$ ) (see the Appendix). The temporal shift  $\Delta\tau_{del}$  between the pulses is chosen to be 2 ns. The radiative decay constant is  $\Gamma_0=9 \text{ ns}^{-1}$  and the duration of pulses is  $\tau=2 \text{ ns}$ .

the decay constant to be  $\Gamma_0=9 \text{ ns}^{-1}$ . Figure 6 corresponds to the case of state control with spectrally broad radiation, while Fig. 7 shows the results of solving system (A3)–(A6), that illustrates conventional monochromatic STIRAP. In the latter case the effective frequency  $\tilde{\Omega}_0$  is determined in Eq. (A8).

We point out for both Figs. 6 and 7 the somewhat curious behavior of  $\rho_{++}^{(f)}$  as a function of the light intensity in the region of large two-photon detuning values. Initially when effective Rabi frequencies  $\tilde{\Omega}_0$  or  $\tilde{\Omega}_0$  are small, the curves  $\rho_{++}^{(f)}(\omega_L=\text{const}, \tilde{\Omega}_0)$  rise linearly with increasing  $\tilde{\Omega}_0$  in the sequence of simple observation: the population transfer occurs because the levels couple to the light, where larger  $\tilde{\Omega}_0$  corresponds to stronger coupling and, consequently, to larger transfer efficiency. This observation is true as long as the interaction of an atom with photons is linear, i.e., light is unable to modify the bare states. When the light intensity starts to exceed the saturation value, the bare states are transformed into dressed states, each of which shares the population with the excited state (if  $\delta \neq 0$ ) that clearly stimulates the unwanted population flow. Moreover, the transformations are accomplished by energy shifts of the dressed states that lead to some effects with similarities to laser induced transparency. In the particular case of monochromatic STIRAP, because of the ac Stark shift of the transition  $m=-1 \rightarrow m'=0$ , the initial impulse  $\tilde{\Omega}_+(t)$  [see in the Appendix, Eq. (A7)] results in increasing detunings from the transition  $m=+1 \rightarrow m'=0$ . Both aforementioned factors dramatically decrease  $\rho_{++}^{(f)}$  in the region  $\tilde{\Omega}_0^2\tau > 18$  as follows from Figs. 6 and 7. The case of large  $\tilde{\Omega}_0$  values allows one to consider the problem under a perturbation approach in which the parameter  $\delta/\tilde{\Omega}_0^2$  becomes small. One of the dressed states has a structure close to that of a dark state [see Eq. (18)]. It shares only a small fraction  $\sim \delta/\tilde{\Omega}_0^2$  of the excited state and restores the population transfer efficiency in the region of very large  $\tilde{\Omega}_0$ .

### V. CONCLUSION

In this paper we have demonstrated that if the states between which we carry out population adiabatic transfer (STIRAP) are degenerate, this process can be implemented with broadband nonmonochromatic (white) light. The efficiency of the population transfer with broadband radiation is similar to the efficiency of the STIRAP process that can be achieved in the traditional way with monochromatic radiation and, in the case of sufficiently slow, adiabatic manipulation of the states, can approach 100%.

The existence of dark states in the manifold of magnetic sublevels of an atomic state created by the nonmonochromatic radiation was noticed earlier (see, for example, [30]) and was related to the well known phenomenon of optical pumping of atoms in the manifold of magnetic sublevels [13]. In the 1950s, a long time before the invention of lasers, the phenomenon of optical pumping was observed with a conventional spectrally broad light source by Brossel, Kastler, and Winter [31] and by Hawkins and Dicke [32]. With this in mind, it should be easy to understand the conclusions

of this paper, that with broadband radiation it is possible not only to create a dark state in the manifold of magnetic sublevels, but it is possible also to manipulate this state.

The demand that the states involved in the spectrally broad light STIRAP process are degenerate is essential. It ensures that the phase fluctuations of the radiation source are synchronous for the pump as well as the Stokes field and cancels in the atom-light interaction process. Even a weak magnetic field that splits magnetic sublevels on the order of the ground-level width destroys the dark state created by the broadband radiation.

It is obvious that the manipulation of coherent states created in the manifold of the magnetic sublevels by the broadband radiation is not limited to the STIRAP process only, but can be extended to other coherent processes, such as coherent control of atomic states with three light fields in a tripod configuration [33] or manipulation of many degenerate quantum states simultaneously [34].

If one uses a broadband radiation source with sufficient spectral density, population transfer through the continuum [35] can be foreseen as well. The advantage of white light is that it does not significantly perturb the continuum in contrast to the monochromatic lasers. The latter strongly modify continuum states and result in sharp Fano profiles of dipole matrix elements [36] that block the existence of stable dark states.

#### ACKNOWLEDGMENTS

This work was supported by the EU FP6 TOK Project LAMOL (Contract No. MTKD-CT-2004-014228), RFBR Grant No. 09-02-92428, by the Latvian Science Council, and the INTAS Projects No. 06-1000024-9075 and No. 06-1000017-9001. We thank Professor E. Arimondo, Professor K. Bergmann, Professor D. Budker, Dr. B. Shore, and Professor L. P. Yatsenko for their useful discussions.

#### APPENDIX: DENSITY MATRIX EQUATION WITH COHERENT LASERS UNDER ADIABATIC CONDITIONS

It is of interest to analyze the population transfer scheme presented in Fig. 4 for the case of two coherent lasers. We assume a large enough one-photon detuning value to ensure that the procedure of adiabatic elimination of the optical coherence elements [26] will be valid. The same procedure is the main approach used in the broad-line approximation to obtain system (10)–(13) for the dynamics of the density matrix driven with spectrally broad light. It is natural to expect some similarity between the coherent and incoherent cases and to take advantage of the well studied coherent case to predict important properties of the manipulation of states with broadband radiation.

We consider a three-level system, as depicted in Fig. 4, which is being excited by two independent coherent laser fields with Rabi frequencies  $\Omega_{\pm}(t)$ . In an experiment it would mean that one applies lasers with  $\pm$  circular polarization to a two-level atom with angular momentum  $l=0, 1$  for the upper and lower energy states, respectively. The ground state Zeeman sublevel  $m=0$  is not involved in the interaction

with the light. It collects the spontaneous population flow from the excited sublevel  $m'=0$  at the decay rate  $\Gamma_{00}$ . The lasers have identical frequencies  $\omega$ , i.e., they are detuned from the  $l'=0, m'=0 \leftrightarrow l=0, m=0$  Bohr transition frequency  $\omega_0$  at the same one-photon detuning  $\Delta = \omega - \omega_0$ . Because of the possible presence of an external magnetic field, the Zeeman sublevels  $m = \pm 1$  may have Zeeman energy shifts with a value  $m\tilde{\omega}_L$ . Clearly, the two-photon detuning  $\delta$  is then equal to the corresponding Zeeman splitting  $2\omega_L$ . If  $\Delta$  happens to be relatively large, for instance, if it effectively exceeds the inverse duration  $1/\tau$  of the laser pulses [we assume them to have Gaussian shapes (9)], the adiabatic approximation becomes valid for the density matrix elements  $\rho_{m,m'=0}$  (the indices  $m = \pm 1$  and  $m' = 0$  belong to the ground and the excited states, respectively) that make it possible to eliminate adiabatically  $\rho_{m,m'=0}$  [26]. In addition, a large spontaneous decay rate  $\Gamma_0\tau \gg 1$  ensures that adiabatic elimination is realized as well. Under the rotating-wave approximation [5] adiabatic elimination allows us to set  $d/dt\rho_{m,m'=0} = 0$  and, thus, to reduce  $\rho_{m,m'=0}$  to the form [19,26,37]

$$\begin{aligned} \left(i\Delta_m + \frac{1}{2}\Gamma_0\right)\rho_{m,m'=0}(t) = & -\frac{i}{2}\Omega_m^*\rho_{m'=0,m'=0}(t) + \frac{i}{2}\Omega_+^*\rho_{m,+}(t) \\ & + \frac{i}{2}\Omega_-^*\rho_{m,-}(t). \end{aligned} \quad (\text{A1})$$

The values  $\hbar\Delta_m = \hbar(\Delta + m\omega_L)$  give the energies of the  $m$ -bare states, provided that the energy of the  $m'=0$  bare state is chosen to be zero.

The coherent matrix elements  $\rho_{m,m'=0}$  describe optical oscillators in an atom with decay constant  $\Gamma_0/2$ . The oscillators are excited by the lasers' radiation field, which has detunings  $\Delta_m$ . It is well known [37,38] that the excitation time  $\tau^{(ex)}$  is determined by the relation  $\tau^{(ex)} \approx 1/\sqrt{\Delta_m^2 + \Gamma_0^2/4}$ . If  $\tau^{(ex)}$  is substantially smaller than the characteristic duration  $\tau$  of the laser pulses, i.e.,

$$\tau\sqrt{\Delta_m^2 + \Gamma_0^2/4} \gg 1, \quad (\text{A2})$$

then the evolution of the amplitude  $\rho_{m,m'=0}$  follows the excitation adiabatically, and Eq. (A1) comes to be valid. This fact allows the general equation of motion for the density matrix [19,37] to be reduced to the following system:

$$\begin{aligned} \frac{d}{dt}\rho'_{00} = & -(|\bar{\Omega}_+|^2 + |\bar{\Omega}_-|^2 + \Gamma_0)\rho'_{00} + |\bar{\Omega}_+|^2\rho'_{++} + |\bar{\Omega}_-|^2\rho'_{--} \\ & + 2\text{Re}\bar{\Omega}_-\bar{\Omega}_+^*\rho'_{+-}\left(1 + \frac{2i\omega_L}{\Gamma_0}\right), \end{aligned} \quad (\text{A3})$$

$$\begin{aligned} \frac{d}{dt}\rho'_{++} = & -|\bar{\Omega}_+|^2\rho'_{++} + (|\bar{\Omega}_+|^2 + \Gamma_0)\rho'_{00} \\ & - \text{Re}\bar{\Omega}_-\bar{\Omega}_+^*\rho'_{+-}\left(1 + \frac{2i\omega_L}{\Gamma_0}\right), \end{aligned} \quad (\text{A4})$$

$$\begin{aligned} \frac{d}{dt}\rho_{--} = & -|\bar{\Omega}_-|^2\rho_{--} + (|\bar{\Omega}_-|^2 + \Gamma_{0-})\rho'_{00} \\ & - \text{Re} \bar{\Omega}_- \bar{\Omega}_+^* \rho_{-+} \left(1 + \frac{2i\omega_L}{\Gamma_0}\right), \end{aligned} \quad (\text{A5})$$

$$\begin{aligned} \frac{d}{dr}\rho_{-+} = & \left[2i\omega_L - (|\bar{\Omega}_+|^2 + |\bar{\Omega}_-|^2)\left(\frac{1}{2} + \frac{i\omega_L}{\Gamma_0}\right)\right]\rho_{-+} \\ & - \frac{1}{2}\bar{\Omega}_-^* \bar{\Omega}_+ \left(1 + \frac{2i\omega_L}{\Gamma_0}\right)(\rho_{++} + \rho_{--} - 2\rho'_{00}), \end{aligned} \quad (\text{A6})$$

where we adopt the same notations for the density matrix elements as in the case of broadband radiation, Eqs. (10)–(13). In particular,  $\rho'_{00} \equiv \rho_{m'=0, m'=0}$ . The equation for  $\rho_{+-}$  is obtained from Eq. (A6) by complex conjugation. Two effective “frequencies”  $\bar{\Omega}_\pm$  are introduced as follows:

$$\begin{aligned} \bar{\Omega}_\pm(t) &= \frac{\sqrt{\Gamma_0}}{\sqrt{\Gamma_0^2 + 4\Delta_\pm^2}} \Omega_\pm(t), \\ \Omega_\pm(t) &= \Omega_0 \exp[-(t - \Delta_\pm)^2/2\tau^2], \end{aligned} \quad (\text{A7})$$

which have dimension ( $s^{-1/2}$ ) and Gaussian pulse shapes of the type (15), which have arrived, as before, in an appropriate sequence.

Systems (A3)–(A6) and (10)–(13) provide useful insight into how to apply knowledge of state control with two coherent fields to the case of state control with spectrally broad radiation. For instance, the reason for the existence of dark-states (18) becomes clear, namely, when Zeeman splitting is absent (i.e.,  $\omega_L=0$ ), systems (A3)–(A6) and (10)–(13) become identical (provided we identify the effective Rabi frequencies  $\bar{\Omega}_\pm$  and  $\bar{\Omega}_\pm$ ) and the coherent dark state (2) generates a spectrally broad light dark state in the form of Eq. (18). Note that this situation corresponds to two-photon resonance when  $\Delta_+ = \Delta_- = \Delta$ . It is clear as well that criterion (5) for STIRAP adiabaticity is reduced to the requirement  $\Omega_0^2 \tau^2 \gg 1$  for the pulses  $\Omega_\pm(t)$  [Eq. (A7)]. It is more instructive to express this requirement in terms of the effective value  $\bar{\Omega}_0$  for Rabi frequency as follows:

$$\bar{\Omega}_0 = \Omega_0 \sqrt{\Gamma_0} / \sqrt{\Gamma_0^2 + 4\Delta^2},$$

$$\bar{\Omega}_0^2 \tau (\tau \sqrt{\Gamma_0^2 + 4\Delta^2}) \sqrt{1 + 4\Delta^2/\Gamma_0^2} \gg 1. \quad (\text{A8})$$

Since system (10)–(13) was obtained under assumption (A2), relation (23) (with the clear substitution  $\bar{\Omega}_0 \rightarrow \bar{\Omega}_0$ ) turns out to be sufficient to satisfy inequality (A8). In other words, if we identify the effective Rabi frequency  $\bar{\Omega}_0$  with  $\bar{\Omega}_0$ , we obtain a new criterion (23) for efficient population transfer with spectrally broad light under the realization of two-photon resonance.

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