Coherent interactions in nonlinear multilevel media.

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Abstract. We propose in this work an alternative method of easier calculation of necessary conditions for lossless propagation of short laser pulses in multilevel atomic media. Method is based on the quasienergy approach and illustrated with example of a five-level system. We also demonstrated effective population transfer in this system.

1. Introduction

Coherent interaction of light signals with quantum systems attract considerable interests for their importance in both fundamental science and practical applications. The most observable experimentally and applicable are adiabatic coherent interactions which rely upon sufficiently slow evolution of a quantum system [1-3]. Constructing desired coherent superposition states of atoms interacting with laser pulses in macroscopic media is one of key problems in quantum informatics and related topics.

Microscopic theory of adiabatic interaction between laser pulses and individual multilevel atoms is sufficiently complete [4, 5, 6] but the realization of adiabatic interaction in macroscopic volumes is not completely clear and sufficiently studied as of today. In adiabatic approximation the populations of atomic levels follow the instantaneous values of field envelopes. So, the adiabatic approximation corresponds to a dispersion-free medium. Even in linear optics the approximation of dispersion-free medium is restricted by propagation lengths where the group delay (caused by the group velocity which is different from c) of the pulse in the medium may be neglected. In nonlinear media the situation is more complicated because of the dependence of the group velocity on the intensities of available laser fields. In the simplest, two-level, model more intense portions of the pulse are moving faster, resulting in steepening of the leading edge and formation of a shock wave (see, e.g., [7, 8, 9]). In addition to this mechanism, nonlinearity of medium leads to parametric broadening of spectrum (self-phase modulation [10]) which again shortens the pulse duration during its propagation and also furthers the violation of adiabaticity. So adiabatic processes in medium are quite robust if and only if they are implemented at sufficiently short optical lengths, where adiabaticity of interaction is preserved. However, it was shown as early as in seventies that at interaction of a laser pulse with a V-system the length of adiabaticity of interaction increases essentially, as compared with the two-level system, due to compensation for the medium dispersion [11]. After that the possibility of soliton regime of pulse propagation in such media was studied in detail [12]. This mechanism of compensation for the nonlinear dispersion of medium is based on destructive interference of dipole moments induced by radiation at the adjacent transitions of medium atoms.
Another striking example of bleaching of resonant medium via adiabatic interaction is electromagnetically induced transparency (EIT) [13, 14] which is based on the so-called dark state formed in a three-level system [15]. The dark state does not involve the intermediate state because of quantum interference; due to this fact the field-induced dipole moments vanish leading to medium bleaching.

The problem of medium transparency at certain frequencies which enables distortion-less propagation of laser pulses in media, is of considerable importance because it is concerned directly with such applications as optical information storage in media with its subsequent retrieval and optical communication lines [14], adiabatic population transfer [15], creation of superposition states, control of chemical processes [16], implementation of logic gates and many others.

In this work we propose a relatively simple calculation technique to find the necessary (but not always sufficient) condition for similar bleaching of a medium consisting of atomic systems with more complicated level diagrams. The method relies on the system quasienegries, which allows obtaining analytical expressions describing controllable population transfer and pulse propagation without distortion in media. We also obtain the necessary conditions for the propagation length where the adiabaticity of interaction does not break in the medium. For visualization, we demonstrate these effects in media of M-type and W-type atomic system.

2. Formalism

We consider the multilevel schemes (see, Fig.1) interacting with incident multipulse field 

\[ E(z, t) = \sum_i E_i \exp(i\omega_i z/c - i\omega_i t) + \text{c.c.} \]

Frequencies \( \omega_i \) of pulses may be in resonance with one or several atomic transitions, i.e. \( |\omega_i - \omega_{ij}| << \omega_{ij} \), where \( \omega_{ij} \) are the frequencies of respective atomic transitions. Conditions imposed on intensities of pulses, their on/off switching sequences, ranges of overlapping, choice of polarizations (if magnetic sublevels are employed) etc. are determined while solving the problem (see Section 3) depending on specific configurations. We assume that considered pulses have durations \( T_i \) much shorter than all times of relaxation and at the same time much longer than the inverse frequency distance between the closest quasienergies i.e. \( |\lambda_i - \lambda_j|/T_i >> 1 \) (\( \lambda_{ij} \) are eigenvalues of the interaction Hamiltonian), in order to ensure the adiabaticity of interaction. In approximation of non-overlapping atomic wave functions, the polarization of medium may be represented in the form \( P = N\langle d \rangle \), with \( N \) being the number density of atoms and \( \langle d \rangle \) being the atomic dipole moment induced by an external field, \( \langle d \rangle = \langle \psi | d | \psi \rangle \), where \( |\psi\rangle \) is the state vector of the atom in the external field. Within approximations listed above we have

\[ \langle \psi | d | \psi \rangle = \sum b_i^* b_{i+1} \langle i | d | i+1 \rangle e^{-i\omega_i t} + \text{c.c.} \] (1)

Here \( |i\rangle \) is the bare state of atom corresponding to the energy \( E_i \), \( \langle i | d | j \rangle = d_{ij} \) is the matrix element of the dipole moment of atomic transition, \( b_i \) is the coefficient of bare state \(|i\rangle\) in superposition state \(|\psi\rangle\) and \( b_i^* b_{i+1} \langle i | d | i+1 \rangle e^{-i\omega_i t} = \langle d_i \rangle e^{-i\omega_i t} \) is the dipole moment at frequency \( \omega_i \).

It is known that in the approximation of slowly varying amplitudes, propagation of pulses in a dielectric homogeneous isotropic medium can be described by the system of truncated equations in running coordinate system \( x = z, \tau = t - z/c \) (see, e.g., [1]):

\[ \frac{\partial E_i}{\partial x} = -\frac{2\pi \omega_i}{c} P_i \]  (2)

where \( P_i \) is the complex amplitude of medium polarization at frequency \( \omega_i \), \( P_i = N\langle d_i \rangle \). As shown in works [14,15], in the resonant approximation and adiabatic interaction with medium,
the system of truncated equations [22,23] may be represented in the form

$$\frac{\partial E_j}{\partial x} = -i \frac{2\pi N_j \omega_j \hbar}{c} \frac{\partial \lambda_i}{\partial E_j^2}$$

(3)

Here $\lambda_i$ is the eigenvalue of the interaction Hamiltonian corresponding to eigenstate realized in adiabatic interaction. The last equation is correct only at such lengths of propagation where the approximation of dispersion-free medium is valid. Limitations of applicability of this equation are considered in Section 4. If the system has a quasienergy which remains constant during the overall time of interaction, i.e., $\partial \lambda / \partial E_j^* = 0$, then according to [1], the induced dipole moment in such a system is zero and the pulse propagates in such medium without change in shape at the group velocity equal to $c$. It is known [1] that the quasienergies (eigenvalues) of the system are the roots of characteristic equation

$$\det(H - \lambda I) = 0$$

(4)

where $I$ is the unit matrix and $H$ the interaction Hamiltonian (in frequency units) which in the dipole approximation and the resonance approximation may be represented in the form

$$H = \sum \sigma_{i,j} \delta_{i-1} - (\sum \sigma_{i,i+1} \Omega_i + h.c.)$$

(5)

with $\sigma_{i,j}$ being the projection matrices, $\Omega_i$ amplitude of Rabi frequencies of the pulse interacting with the transition $i \rightarrow i + 1$: $\Omega_i = \Omega e^{i\varphi_i} = -E_i d_{i,i+1} / \hbar$, and $\delta_{i-1}$ the $(i-1)$-photon detuning ($\delta_0=0$). The phases of complex Rabi frequencies $\Omega_i$, which can vary when traveling in the medium, are included into one-photon detunings $\Delta_i$. So the quantities $\Omega_i$ are real and positive. (The one photon detunings are defined as $\Delta_i = \omega_{i+1,i} - \omega_i + \varphi_i$, if $\omega_{i+1,i} > 0$ and $\Delta_i = \omega_{i+1,i} - \omega_i + \varphi_i$, if $\omega_{i+1,i} < 0$, where $\varphi_i$ are the phases of corresponding Rabi frequencies). Definition of multiphoton detunings depends on the specific scheme of interaction. By differentiating the characteristic equation with respect to $E_j^*$ and setting to zero the derivatives of quasienergy, we obtain a system of algebraic equations, from which the needed values of interaction parameters may be determined. The problem can, however, be simplified. Really, at turning off the fields the interaction Hamiltonian is diagonalized and the roots of [2] go to the following constant values:

$$\lambda_i \rightarrow \delta_{i-1}$$

(6)

The problem of determination of necessary (but not always sufficient) conditions for medium transparency is thus reduced to the search of such parameters of adiabatic interaction for which the system quasienergy remains equal to one of multi-photon detunings always during interaction. By substituting these values successively into equation (2) we obtain the needed conditions for the interaction parameters.

3. Criterion of adiabaticity of interaction

Procedure considered above allows determining only one eigenvalue of the interaction Hamiltonian, but determination of the adiabaticity criterion for a single atom requires knowledge of all other eigenvalues, i.e., finding the algebraic roots of fourth-order equation. This can be done in general only numerically. Since we are interested in the propagation problem, we must determine the adiabaticity criterion for the medium, rather than for a single atom; it may be found directly from the propagation equations. As follows the dipole moment $< \mathbf{d} >$ of an atom interacting with electromagnetic field and residing the state $|\psi\rangle$, can be expressed in terms of the coefficients $b_i$ of bare state $|i\rangle$ in superposition state $|\psi\rangle$ and matrix elements of the dipole moment of atomic transition $d_{ij}$. The coefficients $b_i$ are determined by non-stationary Schroedinger equation with Hamiltonian (3). 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 Schrödinger equation, in general case we obtain 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 of medium consisting of W-type atoms (Fig. 1c) we obtain:

\[
\begin{align*}
\frac{\partial \Omega_1}{\partial x} &= -q_1 \frac{\partial |b_1|^2}{\partial \tau} \\
\frac{\partial \Omega_2}{\partial x} &= q_2 \left( \frac{\partial (|b_1|^2 + |b_2|^2)}{\partial \tau} \right) \\
\frac{\partial \Omega_3}{\partial x} &= q_3 \left( \frac{\partial (|b_1|^2 + |b_2|^2 + |b_3|^2)}{\partial \tau} \right) \\
\frac{\partial \Delta_i}{\partial x} &= q_i \frac{\partial \text{Re}(b_i^* b_{i+1})}{\partial \tau} \\
\end{align*}
\]  

(7)

where \( q_i = 2\pi N \omega_i |d_{i,i+1}|^2 / \hbar c \) For other atomic level schemes equations for intensities differ in only the signs of derivatives with respect to populations. As it follows from equations, during
propagation in the medium not only the shapes of pulses may vary essentially, but also the conditions for detuning of resonances may be violated.

Since it is the time derivatives that stand in right hand side of these equations, by use for atomic populations the expressions following from the wave functions obtained above, we take into account the first nonadiabatic correction which lead to the change in spectral and temporal characteristics of pulses. Correspondingly, the conditions of smallness of these changes are criteria of adiabaticity in the medium.

4. Conclusion
We have developed and analyzed, for the example of a five-level system, a relatively simple method for finding necessary conditions under which adiabatic short light pulses can travel in the medium without distortion of the shape and phase. It was shown that the necessary conditions for such bleaching of medium (i.e., vanishing of dipole moments induced in the medium at the frequencies of all interacting fields) is the equality of system quasienergies to one of the values of multi-photon detunings of resonances. In this case the medium bleaching may be caused by both interference of quantum states and interference of dipole moments of different transitions if pumping is degenerate. The existence of such regimes means that at the propagation lengths where the interaction adiabaticity does not break, stable superposition states are produced all over the medium and these states may easily be controlled by adjusting the parameters of pulses. Finding of the sufficient conditions requires determination of roots of algebraic equations which may be done only numerically for the order of equation higher than three. However, the sufficiency of found necessary conditions may easily be checked directly by calculation of the induced dipole moments. In addition, we demonstrated for the examples of W-systems the possibility to find a generalized criterion of adiabaticity of interaction in the medium without determination of all eigenvalues of interaction Hamiltonian; we analyzed population transfer in this system and its peculiarities during propagation in the medium.

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