Optimal control of population and coherence in three-level $\Lambda$ systems

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Abstract

Optimal control theory (OCT) implementations for an efficient population transfer and creation of maximum coherence in a three-level system are considered. We demonstrate that the half-stimulated Raman adiabatic passage scheme for creation of the maximum Raman coherence is the optimal solution according to the OCT. We also present a comparative study of several implementations of OCT applied to the complete population transfer and creation of the maximum coherence. Performance of the conjugate gradient method, the Zhu–Rabitz method and the Krotov method has been analysed.

1. Introduction

Efficient and selective transfer of population in quantum systems provides an essential tool for a variety of applications in physical chemistry, laser spectroscopy, quantum optics and quantum information processing [1–5]. Some of the population transfer techniques make use of the Rabi oscillations when the transfer efficiency is controlled by the pulse area of the external fields [6]. Sensitivity of the final population distribution in the system to the field parameters is sometimes considered as a drawback of these methods. Techniques utilizing adiabatic passage solution in the system dynamics are substantially more robust against moderate variations in the interaction parameters. Stimulated Raman adiabatic passage (STIRAP) is one example of such methods [7, 8].

In a three-level $\Lambda$ system, STIRAP provides a robust scheme for transferring population by using two strong pulses, called the pump and the Stokes [7, 8]. When the pump–Stokes pulse sequence is arranged in a counterintuitive order (the Stokes pulse precedes the pump pulse) the system dynamics takes place within a so-called dark state (a coherent superposition of the initial and the target states). The adiabatic change of the field amplitudes guaranties nearly 100% efficiency of population transfer to the target state while the intermediate state population is negligibly small during whole time evolution. There are a few modifications of the STIRAP scheme for more complex configurations of quantum systems [7–15].

The search for an efficient scheme of population transfer is also one of the major goals of optimal control theory (OCT), a powerful and sometimes mathematically very sophisticated technique [16–20]. The OCT algorithm has been applied successfully to a broad variety of physical and chemical systems [21–30]. It intrinsically accounts for the quantum-mechanical interference between many pathways connecting the initial and target state in a quantum system. There were some interesting arguments in trying to find a link between OCT and STIRAP. Despite initial negative prognosis [31], it was demonstrated recently that the STIRAP-type solution can emerge automatically from the global OCT method [32]. Besides, the STIRAP scheme has been obtained successfully using a local OCT [14].

In this work, we review the performance of the OCT algorithms [1, 2, 29, 33–36] applied for the population transfer and explore their potential to maximize the Raman coherence in the three-level $\Lambda$ system. Our motivation for maximizing coherence is related to the recent developments in coherent anti-Stokes Raman microscopy and remote detection using intense femto-second laser setups [37–42].

The paper is organized as follows. In section 2, basic equations of the OCT are developed using the method of variational calculus. Field equations are derived using the
penalty on the energy of the control field. A time-dependent penalty function is used which ensures an experimentally feasible profile of the laser pulses. A second penalty function is introduced to minimize the population of the excited intermediate state throughout the time evolution of the system. In section 3, we apply the OCT formalism to a three-level Λ system and analyse solutions of the OCT equations for population transfer and a maximum Raman coherence applying different optimization strategies: the conjugate gradient method [29, 33], the Zhu–Rabitz method [16] and the Krotov method [17–20]. Section 4 is the conclusion.

2. General equations of the OCT

The OCT is based on the definition of a cost functional $K$ which must have an optimal value when the desired transformation of the wavefunction is successfully achieved by the control laser field $\epsilon(t)$. An optimal solution requires that the system wavefunction at a final time, $|\psi(T)\rangle$, should be as close as possible to the target wavefunction, $|\phi(T)\rangle$. That is, the overlap $|\langle\psi(T)|\phi(T)\rangle|^2$ is maximal at final time $T$.

In order to derive the control equation for the field and obtain realistic field amplitude we minimize the energy fluency of the field. Another requirement is that the population of the excited intermediate states has to be minimal throughout the time evolution of the field. Another requirement is that the population of the intermediate states must have an optimal value when the desired transformation is successfully achieved by the control field $\epsilon(t)$.

These requirements lead to a complete cost functional of the form

$$K = |\langle\psi(T)|\phi(T)\rangle|^2 - \alpha(t) \int_0^T dt [\epsilon(t) - \epsilon_r(t)]^2$$

$$- \beta \int_0^T dt \langle\psi(t)|\hat{P}|\psi(t)\rangle$$

$$- 2 \text{Re} \left[ \int_0^T dt \langle\chi(t)|{\hat{\alpha}} + \frac{i}{\hbar} \hat{H}\psi(t)\rangle \right].$$

(1)

The factor $\alpha(t) = \alpha_0/s(t)$ is a time-dependent penalty function that determines the shape function $s(t)$, while $\alpha_0$ is a constant which should be determined due to the significance of the field energy value. The main purpose of $s(t)$ is to turn the field on and off smoothly to ensure the feasibility of experimental implementation of the optimal laser pulse. $\epsilon_r(t)$ denotes a reference field and $\beta$ is a penalty parameter for the total population of the intermediate state (or state manifold). The function $|\chi(t)\rangle$ can be regarded as a Lagrange multiplier introduced to ensure satisfaction of the Schrödinger equation.

Each of the terms in equation (1) depends explicitly or implicitly on the unknown driving field. The goal is to determine an optimal field $\epsilon(t)$ for which the cost functional has an extremum. Taking variations of the cost functional with respect to $|\chi(t)\rangle$, $|\psi(t)\rangle$ and $\epsilon(t)$ we find the following set of equations:

$$i\hbar \frac{\partial}{\partial t} |\psi(t)\rangle = \hat{H} |\psi(t)\rangle,$$

(2)

$$\frac{\partial}{\partial t} |\chi(t)\rangle = -\frac{i}{\hbar} \hat{H} |\chi(t)\rangle + \beta |\phi(t)\rangle,$$

(3)

$$\epsilon(t) = \epsilon_r(t) + \frac{1}{\alpha(t)\hbar} \text{Im} \left( \langle\chi(t)|{\hat{\alpha}} + \frac{i}{\hbar} \hat{H}\psi(t)\rangle \right).$$

(4)

Variation of the cost functional with respect to $|\psi(T)\rangle$ gives the initial condition for the Lagrange multiplier

$$|\chi(T)\rangle = |\langle\psi(T)|\phi(T)\rangle|\phi(T)\rangle.$$

(5)

Equations (2) and (3) determine the time evolution of the wavefunction and Lagrange multiplier which are used in equation (4) to find the optimal field, $\epsilon(t)$, maximizing the overlap $|\langle\psi(T)|\phi(T)\rangle|^2$ in equation (1).

3. Optimal control of three-level Λ system dynamics

3.1. General equations

To demonstrate an application of the general optimization formalism outlined in the previous section and to test the performance of various implementations of the OCT we consider two optimal control problems. First, we re-examine the population transfer from the initially occupied level $|1\rangle$ to the level $|3\rangle$. In the second problem, we utilize the OCT to create a maximum Raman coherence, in other words, the 50/50 coherent superposition of the states $|1\rangle$ and $|3\rangle$.

First, we consider the population transfer in a generic three-level Λ system excited by a pump–Stokes pulse sequence, see figure 1. We address the so-called nonimpulsive excitation when the pump pulse interacts with the $|1\rangle$–$|2\rangle$ transition while the Stokes pulse controls the transition $|2\rangle$–$|3\rangle$.

We assume that all relaxation times in the system are much longer than the pulse duration so that the dynamics of an arbitrary wavefunction

$$|\psi(t)\rangle = a_1(t)|1\rangle + a_2(t)|2\rangle + a_3(t)|3\rangle,$$

(6)

where $a_i(t)$ is the probability amplitude to be in the state $|i\rangle$, is governed by the Schrödinger equation with the Hamiltonian of the form

$$\hat{H} = \begin{pmatrix}
E_1 & -\mu_1 \epsilon_p(t) & 0 \\
-\mu_1 \epsilon_p(t) & E_2 & -\mu_2 \epsilon_s(t) \\
0 & -\mu_2 \epsilon_s(t) & E_3
\end{pmatrix}.$$

(7)
Here $E_i$ is the energy of the $i = 1, 2, 3$ state, $\mu_{12,23}$ are the dipole moments and $\epsilon_{P,S}(t)$ are the pump and Stokes fields.

Now, we are ready to solve the optimization problem by applying a general algorithm outlined in the previous section. Varying the cost functional with respect to the Lagrange multiplier vector $b_i(t)$, the probability amplitudes vector $a_i(t)$ and the external field $\epsilon_{P,S}(t)$, equations (2), (3) and (4) take the following form:

$$i\hbar \frac{\partial a_i(t)}{\partial t} = \hat{H}_{ij} a_j(t),$$

$$\frac{\partial b_i(t)}{\partial t} = -\frac{i}{\hbar} \hat{H}_{ij} b_j(t) + \beta a_2(t) \delta_{i2},$$

$$\epsilon_P(t) = \epsilon'_P(t) - \frac{1}{2\hbar \alpha(t)} \cdot \text{Im}[b_{i1}^*(t) \mu_{12} a_2(t) + b_{i2}^*(t) \mu_{23} a_1(t)],$$

$$\epsilon_S(t) = \epsilon'_S(t) - \frac{1}{2\hbar \alpha(t)} \cdot \text{Im}[b_{i2}^*(t) \mu_{23} a_3(t) + b_{i3}^*(t) \mu_{32} a_2(t)],$$

where $\epsilon'_P(t)$ and $\epsilon'_S(t)$ are the pump and Stokes reference fields.

The set of equations (8)–(11) provides optimal control fields which are based on the solution of the Schrödinger equation with the Hamiltonian in equation (7). In many cases, we consider the interaction of the three-level system with the external field as a resonant excitation and invoke the rotating wave approximation (RWA) which partially simplifies the solution of the problem and sometimes facilitates understanding of underlaying physical mechanisms.

In the interaction representation the Hamiltonian can be written in the form

$$\hat{H} = \frac{\hbar}{2} \begin{pmatrix} 0 & -W_P(t) e^{-i\Delta_{P,t}} & -W_S(t) e^{-i\Delta_{S,t}} \\ -W_P^*(t) e^{i\Delta_{P,t}} & 0 & 0 \\ -W_S^*(t) e^{i\Delta_{S,t}} & 0 & 0 \end{pmatrix},$$

where $W_{P,S}(t) = \Omega_{P,S}(t)(1 + e^{-2i\omega_{P,S} t})$, $\Omega_{P,S}(t) = \mu_{12,23} E_{P,S}(t)/\hbar$ are the pump and Stokes Rabi frequencies, $E_{P,S}(t)$ are the envelopes of the pump and Stokes pulses, $\Delta_{P,S} = \omega_{P,S} - \omega_{21,23}$ are the single-photon detunings of the pump and Stokes central frequency $\omega_{P,S}$ from the respective transition frequency $\omega_{21,23}$.

Figure 2. Population transfer in a Λ system using the conjugate gradient method with penalty on population of the state $|2\rangle$ (right panel) and without penalty $\beta = 0$ (left panel). (a), (d) The population of the state $|1\rangle$ (red line), $|2\rangle$ (blue line) and $|3\rangle$ (black line). (b), (c) The sequence of optimal pulses: Rabi frequency of the pump pulse (red line) and Stokes pulse (blue line). (c), (f) The convergence behaviour of the optimized transition probability (red line), penalty on the field energy (blue line), penalty on the intermediate state population (pink line) and final optimized cost functional (black line) versus the number of iteration steps.
Using the RWA we can replace $W_{P,S}(t)$ in equation (12) by the corresponding Rabi frequencies, that is we neglect the rapidly oscillating terms. As a result we rewrite the equations for the optimal fields, equations (10) and (11), in terms of the Rabi frequency envelopes of the pump and Stokes fields

$$\Omega_{P}(t) = \Omega'_{P}(t) - \frac{\mu_{P}^2}{2\hbar^2 \alpha(t)} \cdot \text{Im}[b_{11}^*(t)a_{2}(t) + b_{21}^*(t)a_{1}(t)],$$

(13)

$$\Omega_{S}(t) = \Omega'_{S}(t) - \frac{\mu_{S}^2}{2\hbar^2 \alpha(t)} \cdot \text{Im}[b_{22}^*(t)a_{3}(t) + b_{32}^*(t)a_{2}(t)],$$

(14)

where $\Omega'_{P}(t), \Omega'_{S}(t)$ are the reference Rabi frequencies of the pump and Stokes pulses. We assume $\mu_{12} = \mu_{21} = \mu_{P}$ and $\mu_{23} = \mu_{32} = \mu_{S}$.

### 3.2. Complete population transfer

To examine OCT implementation we consider three different optimization methods: the conjugate gradient method \[29, 33, 43\], the Zhu–Rabitz method \[16\] and the Krotov method \[17, 18\]. A detailed description of the numerical schemes is given in the appendix. We choose the Gaussian form for the pump and Stokes pulse envelopes as an initial guess

$$\Omega_{P,S}(t) = \Omega_0 \exp \left( -\frac{(t - t_c)^2}{2\tau_0^2} \right),$$

(15)

and our target time is equal to $T = 10$ in the units normalized by the time duration $\tau_0$. The reference envelope $\Omega'_{P,S}(t) = 0$ is used unless otherwise stated.

The goal of the first problem at hand is twofold: first, to design the shape and the sequence of the pump and Stokes pulses providing a complete population transfer to state $|3\rangle$; second to suppress population of the intermediate state during the excitation process by applying a penalty on the state population. By doing this we would also like to determine the efficiency of the methods mentioned above. For simplicity we restrict our consideration to the exact resonance conditions, $\Delta_{P} = \Delta_{S} = 0$.

The conjugate gradient method. The results obtained using the conjugate gradient method \[29, 33, 43\] are shown in figure 2. The set of plots (a)–(c) shows results obtained without the penalty on the intermediate state population, $\beta = 0$. Plots (d)–(f) show results obtained when the penalty is imposed on the population of state $|2\rangle$, $\beta \neq 0$. The population of states $|1\rangle, |2\rangle,$ and $|3\rangle$ as a function of time is shown in plots (a) and (d). Complete population transfer from the initial level $|1\rangle$ to the target level $|3\rangle$ is achieved at the
final time $T$ independently of the penalty parameter. The amount of population in the intermediate state is considerably reduced for a field obtained with the state-dependent constraint (figure 2(d)) in contrast to that resulting from unconstrained optimization (figure 2(a)). The optimized Rabi frequencies (obtained after 1000 iterations) are shown in figures 2(b) and (e). With a proper choice of the penalty parameters $\alpha_0$ and $\beta$, we remove almost all the intuitive solutions and obtain the STIRAP solution, a counterintuitive pulse sequence when the Stokes pulse precedes the pump. Plots (c) and (f) of figure 2 show the convergence behaviour of the optimized transition probability defined as $P = |\langle \psi(T)|\phi(T) \rangle|^2$, and the cost functional, $K$, versus the number of iteration steps. It is seen that the transition probability reaches nearly 100%. The Zhu–Rabitz method. Figure 3 shows the dynamics of population transfer in the three-level $\Lambda$ system optimized using the Zhu–Rabitz method [16]. The same order of illustrations and legends is kept in figure 3 as in the case of figure 2.

There is a complete transfer of the population from the initial state to the target state at final time $T$, figures 3(a) and (d). However, we observe two different mechanisms of population transfer. Optimization without a penalty on the intermediate state population (left panel of figure 3) produces an intuitive pulse sequence; first the pump and then the Stokes pulse is applied to the system (figure 3(a)). That pulse arrangement results in a sequential population transfer from state $|1\rangle$ to state $|2\rangle$ and then to the target state $|3\rangle$ as is demonstrated in figure 3(a), with almost 80% of population resided in state $|2\rangle$ at the central time.

Applying a penalty on the second state population changes the mechanism dramatically (see the right panel of figure 3). Now, we obtain almost hundred times more intense pulses, compare figures 3(b) and (e). More importantly, we automatically obtain a counterintuitive pulse sequence when the Stokes pulse precedes the pump pulse. Therefore, it is clear that the OCT algorithm finds a well-known STIRAP solution which has a highly pronounced signature of the suppressed intermediate state population, see figure 3(d). In that case population transfer takes place through the dark state consisting ideally only of the initial and the target state probability amplitudes and has no projection to state $|2\rangle$ [7].

The value of the optimized cost functional in figure 3(f) is a bit less than that of the cost functional value obtained by using the conjugate gradient method, figure 2(f). This is due to a slightly larger energy fluency of the fields as is clear from the field amplitudes in plot (e) in figure 3 compared to that in figure 2.

The Krotov method. The optimized results of population transfer using the Krotov method [17, 18] are shown in...
Figures 4 and 5. The same set of parameters is used in the optimization process as in the previous two cases. Figure 5 illustrates the results obtained using the Krotov method when $\Omega_{P,S}(t) \neq 0$. The population of the states, optimized Rabi frequencies, transition probability and cost functional are shown in plots (a)–(c) and (d)–(f) of figures 4 and 5.

As in the previous cases, we produce complete population transfer to the target state. However, using the Krotov method [17, 18] with $\Omega_{P,S}(t) = 0$ we were not able to find a set of penalty parameters which provides considerable suppression of the second state population, see figure 4(d). At most we are able to reduce the second state population to only about 40%. Optimized Rabi frequencies are arranged in the intuitive manner even for the reasonably large areas of the pulses, figures 4(b) and 4(e).

The situation changes considerably when we use the reference fields, $\Omega_{P,S}(t) \neq 0$, figure 5. In a manner similar to the previous two cases the STIRAP solution emerges from the optimization procedure when we impose a penalty on the intermediate state population, see the right panel in figure 5: the Stokes pulse precedes the pump, figure 5(e), and a detrimental population of the state $|2\rangle$ is suppressed almost to zero, figure 5(d).

Table 1 shows a comparison of the transition probability $P$, the optimized cost functional $K$ and the maximum population, $\rho_{22} = |a_2|^2$, which resides in the intermediate state at central times for all the methods. Note that the value of the cost functional obtained using the Krotov method for $\Omega_{P,S}(t) \neq 0$ is larger than the corresponding value in other methods. However, the computation cost of the conjugate gradient method is larger than that of the Zhu–Rabitz method and the Krotov method.

3.3. Optimal control to maximize coherence

In this section, we apply the OCT to create a maximum coherence $|\rho_{31}| = |a_3^* a_1|$ between the levels $|1\rangle$ and $|3\rangle$ at final time $T$. We use the same parameters and the initial guess function for the field envelopes as in the complete population transfer section. Figure 6 illustrates the results obtained using the conjugate gradient method (a)–(c), the Zhu–Rabitz method (d)–(f) and the Krotov method with $\Omega_{P,S}(t) \neq 0$ (g)–(i), respectively. This time we restrict our consideration to the case when a penalty on the intermediate state population is applied.

Figures 6(a), (d), (g) show the time evolution of the population in the three-level system excited by the optimized pulse sequence. The dynamics of the population presented in figure 6 is almost identical for all three versions of the OCT: at the target time we obtain a maximum coherence $|\rho_{31}(T)| \approx 1/2$, that is we create a 50/50 coherent superposition of the
states $|1\rangle$ and $|3\rangle$. The population of the intermediate state $|2\rangle$ is almost negligible during the excitation process.

The Rabi frequency of the pump and Stokes pulses obtained from the implementations of the OCT is shown in figures 6(b), (e) and (h), respectively, using the conjugate gradient method [29, 33, 43], the Zhu–Rabitz method [16] and the Krotov method with $\Omega_{P,S}(t) \neq 0$ [19]. All three methods give a similar solution for the optimal pulse sequence. The obtained pulse sequence and corresponding population dynamics allow us to conclude that the OCT finds the so-called half-STIRAP (also sometimes referred to as fractional STIRAP) scheme as the optimal pulse sequence to create maximum coherence in the three-level system. The same way as in the STIRAP scheme, the Stokes pulse is turned on first but both pulses are turned off simultaneously at the later time, see figures 6(b), (e) and (h). The mechanism of the solution can be explained using the dressed state basis as follows. It is easy to find the eigenvalues and eigenvectors (dressed states) of the system Hamiltonian, equation (12), in the RWA. The important dressed state with energy $\lambda_0 = 0$ has the form

$$|c_0(t)\rangle = \frac{\Omega_{P,S}(t)}{\sqrt{\Omega_{P}(t)^2 + \Omega_{S}(t)^2}} |0\rangle + \frac{\Omega_{P}(t)}{\sqrt{\Omega_{P}(t)^2 + \Omega_{S}(t)^2}} |1\rangle.$$  

(16)

That dressed state has zero projection on the pure state $|2\rangle$ while probability amplitudes to be in states $|1\rangle$ and $|3\rangle$ are controllable by the ratio of the pump and Stokes Rabi frequencies. Analysing equation (16) we can reproduce the population dynamics for the half-STIRAP scheme: at time $t = 0$ the pump Rabi frequency $\Omega_P(t = 0) = 0$. 

Figure 6. Maximizing coherence in a three-level system using the conjugate gradient method (a)–(c), the Zhu–Rabitz method (d)–(f) and the Krotov method (g)–(i). (a), (d), (g) The population of state $|1\rangle$ (red line), state $|2\rangle$ (blue line), state $|3\rangle$ (black line) and coherence $\varrho_{31}$ (pink line). (b), (e), (h) The sequence of optimal pulses: Rabi frequency of the pump pulse (red line) and the Stokes pulse (blue line). (c), (f), (i) The convergence behaviour of the optimized transition probability (red line), penalty on the field energy (blue line), penalty on the second state population (pink line) and final optimized cost functional(black line) versus the number of iterations.

Table 1. Comparison of results obtained using different implementations of the OCT for the population transfer dynamics in the three-level $\Lambda$ system.

<table>
<thead>
<tr>
<th>Method</th>
<th>$\alpha_0$</th>
<th>$\beta$</th>
<th>$P$</th>
<th>$K$</th>
<th>$\varrho_{22}$</th>
<th>Without state-dependent penalty</th>
<th>$\varrho_{22}$</th>
<th>With state-dependent penalty</th>
</tr>
</thead>
<tbody>
<tr>
<td>Conjugate gradient</td>
<td>0.01</td>
<td>0</td>
<td>0.999</td>
<td>0.836</td>
<td>0.48</td>
<td>0.00005</td>
<td>1.0</td>
<td>0.998</td>
</tr>
<tr>
<td>Zhu–Rabitz</td>
<td>0.01</td>
<td>0</td>
<td>0.999</td>
<td>0.882</td>
<td>0.74</td>
<td>0.0005</td>
<td>1.8</td>
<td>0.998</td>
</tr>
<tr>
<td>Krotov ($\Omega_{P,S}(t) = 0$)</td>
<td>0.01</td>
<td>0</td>
<td>0.999</td>
<td>0.883</td>
<td>0.74</td>
<td>0.005</td>
<td>0.2</td>
<td>0.998</td>
</tr>
<tr>
<td>Krotov ($\Omega_{P,S}(t) \neq 0$)</td>
<td>1.0</td>
<td>0</td>
<td>0.999</td>
<td>0.999</td>
<td>0.46</td>
<td>0.05</td>
<td>0.2</td>
<td>0.999</td>
</tr>
</tbody>
</table>
Table 2. Comparison of the results obtained using different implementation methods of the OCT to maximize coherence, $|\rho_{31}(T)|$, between the levels $|1\rangle$ and $|3\rangle$ in the three-level $A$ system; $\rho_{ii}(T) = |\langle i| \rho(T) |i\rangle|^2$ is the population of state $|i\rangle$, $i = 1, 2, 3$.

<table>
<thead>
<tr>
<th>Method</th>
<th>$\alpha_0$</th>
<th>$\beta$</th>
<th>$\rho_{11}$</th>
<th>$\rho_{22}$</th>
<th>$\rho_{33}$</th>
<th>$P$</th>
<th>$K$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Conjugate gradient</td>
<td>0.000 25</td>
<td>0.2</td>
<td>0.521</td>
<td>0.058</td>
<td>0.478</td>
<td>0.499</td>
<td>0.999</td>
</tr>
<tr>
<td>Zhu–Rabitz</td>
<td>0.000 5</td>
<td>1.8</td>
<td>0.538</td>
<td>0.030</td>
<td>0.461</td>
<td>0.498</td>
<td>0.998</td>
</tr>
<tr>
<td>Krotov ($\Omega_{p,s} \neq 0$)</td>
<td>0.1</td>
<td>0.2</td>
<td>0.503</td>
<td>0.011</td>
<td>0.496</td>
<td>0.499</td>
<td>0.999</td>
</tr>
</tbody>
</table>

and the Stokes Rabi frequency $\Omega_3(t) \neq 0$; therefore, the dressed state $|c_0(t = 0)\rangle$ correlates with the initial state $|1\rangle$. During the turn-off stage, the ratio between pump and Stokes Rabi frequencies is equal to 1, which results in creation of the state $|c_0(t = T)\rangle = 1/\sqrt{2}, 0, -1/\sqrt{2}$, the state of maximum coherence between the states $|1\rangle$ and $|3\rangle$. The whole dynamics of the system takes place in one dressed state and that is possible only in the limit of the adiabatic regime when nonadiabatic coupling to the other two dressed states is negligible. According to our observation the OCT finds the adiabatic mechanism as an optimal solution of the control problem.

To compare the performance of the different implementations the optimized transition probability $P$, the final optimized cost functional $K$, the penalty on the field energy and the penalty on the second state population as a function of the number of iterations are shown in figures 6(c), (f) and (i). The best results for the maximal coherence obtained using the discussed methods are also summarized in table 2.

4. Conclusion

In this paper, we have analysed a performance of several implementation methods of the OCT to design optimal pulse sequences for a complete population transfer and the creation of a maximum coherence in a three-level $A$ system. We have applied the conjugate gradient method [29, 33, 43], the Zhu–Rabitz method [16], and the Krotov method [17–20] to obtain the optimal solution.

Using the conjugate gradient method it was demonstrated earlier [32] that the STIRAP-type solution can emerge automatically from the global OCT method. Now, we have shown that the Zhu–Rabitz method [16] and the Krotov method [17–20] provide additional conformation that the counterintuitive pulse sequence is the optimal solution according to the OCT. It was demonstrated that the penalty on the population of the intermediate state in a three-level system is a crucial factor to obtain the optimal STIRAP scheme.

We have also demonstrated that the half-STIRAP scheme is the optimal solution according to the OCT for creation of maximum coherence. We have shown that the creation of a maximally coherent superposition is one more optimization problem where the OCT finds the adiabatic mechanism as an optimal solution.

Note that the related results using OCT to create predetermined coherent superposition have been published recently [29]. However, the target wavefunction used in [29] was different in comparison to the present studies: the population of the initially populated state at the later time was zero [29]. As a result, the generalized STIRAP pulse sequence was found as the optimal solution using the conjugate gradient method [29]. In our case, the optimization procedure reveals the half-STIRAP pulse sequence as the optimal solution since we require that half of the population should remain in the initially populated state. Comparing the results obtained using the above considered methods, it is clear that the methods can be applied to various problems on equal footing, since the value of the optimized transition probability $P$ is more than 99% for all the methods. However, the value of the optimized cost functional $K$ calculated using the conjugate gradient method is larger than the corresponding values calculated using the Zhu–Rabitz iterative method and the Krotov method for $\Omega_{p,s}(t) = 0$ and $\beta \neq 0$. Note that the computation cost of the conjugate gradient method is generally larger than that of the other two methods.

One might observe that the pulses in the optimal sequence obtained by optimizing the population transfer (figures 2–5) and the maximum coherence (figure 6) have a relatively similar structure in all considered implementation schemes. However, the Zhu–Rabitz method [16] provides shorter and more intense pulses in comparison to other methods. There is also a bit more pronounced asymmetry of the pump–pulse shape obtained by the Krotov method. We believe that these differences are due to the variations in the numerical implementation procedure and the values of the penalty parameters. The conjugate gradient method [29, 33, 43] incorporates one iteration to next iteration feedback from the control field while the Krotov method [17–20] incorporates one time step to next time step feedback of the control field, and the Zhu–Rabitz method [16] incorporates feedback from the control field in an entangled fashion [45], mixing up both feedback techniques.

The detailed description of the implementation procedure of these methods is given in the appendices.

Acknowledgments

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Appendix A. Conjugate gradient method

For given target time $T$ and the number of time steps $N$ ($t_i = i \times \Delta t$, where $i = 0, 1, 2, \ldots, N$ and $N \Delta t = T$), the conjugate gradient method [29, 33, 43] involves the following steps to obtain the optimal field.

1. **Step 1.** Choose an initial electric field $\epsilon_0(t)$.
2. **Step 2.** Set $k = 1$ and $e^k(t) = \epsilon_0(t)$.
3. **Step 3.** Propagate $|\psi^k(t = 0)\rangle$ forward in time using $e^k(t)$ according to equation (2) to obtain $|\psi(T)\rangle$. 

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Step 4. Evaluate the cost functional $K^k$ according to equation (1). The last term in this equation is zero as $|\chi(t)|$ is a solution of the time-dependent Schrödinger equation.

Step 5. If $k \geq 2$, compute $\Delta K^k = K^k - K^{k-1}$ and compare it with the convergence threshold, $\gamma$. If $\Delta K^k \leq \gamma$, then stop the iteration and declare that the optimal pulse has been obtained.

Step 6. Set $⟨\chi^k(T)⟩ = ⟨φ(T)|φ(T)⟩$. Propagate $⟨\chi^k(T)⟩$ and $|ψ^k(T)⟩$ backward in time using field $ε^k(t)$ to obtain $⟨\chi^k(0)|$ and $|ψ^k(0)⟩$.

Step 7. The gradient $g^k(t)$ of the cost functional $K^k$ defined in equation (1) with respect to the variation of $ε^k(t)$ at time $t$ is given by

$$
g^k(t) = \frac{∂K^k}{∂ε^k(t)} - 2\left(α(t)ε^k(t) - \text{Im}\langle χ^k(t)|\frac{∂H}{∂|ψ^k(t)|}⟩\right)Δt.
$$

(A.1)

Appendix B. Zhu–Rabitz method

The Zhu–Rabitz method [16] involves the following steps to find the optimal value of the control field.

Step 1. Choose an initial electric field $ε_i(t)$. Set $k = 1$ and $ε^k(t) = ε_0(t)$.

Step 2. Propagate $|ψ^k(0)⟩$ forward in time using field $ε^k(t)$ according to equation (2) to obtain $|ψ^k(T)⟩$.

Step 3. Evaluate the cost functional $K^k$ according to equation (1). If $k \geq 2$, compute $\Delta K^k = K^k - K^{k-1}$ and compare it with the convergence threshold, $\gamma$. If $\Delta K^k \leq \gamma$, then stop the iteration and declare that the optimal pulse has been obtained.

Step 4. Propagate the Lagrange multiplier $⟨χ^k(T)⟩ = ⟨φ(T)|φ(T)⟩|ψ^k(T)⟩$ backward in time using the new electric field $ε^{k+1}(t)$ defined by

$$
e^{k+1}(t) = \frac{1}{α(t)} \cdot \text{Im}\left\langle χ^k(t)|\frac{∂H}{∂|ψ^k(t)|}\right|ψ^{k+1}(t)⟩.
$$

according to equation (3) to obtain $⟨χ^k(0)|$.

Step 5. Propagate $|ψ^{k+1}(0)⟩$ forward in time using the new field $ε^{k+1}(t)$ given by

$$
e^{k+1}(t) = \frac{1}{α(t)} \cdot \text{Im}\left\langle χ^k(t)|\frac{∂H}{∂|ψ^k(t)|}\right|ψ^{k+1}(t)⟩.
$$

according to equation (2) to obtain $|ψ^{k+1}(T)⟩$.

Step 6. Go back to step 3 and repeat 3–8 until the required convergence has been achieved.

Appendix C. Krotov method

The Krotov method [17, 18] employs a slightly different iteration procedure.

Step 1. Choose an initial electric field $ε_i(t)$.

Step 2. Set $k = 1$ and $ε^k(t) = ε_i(t)$.

Step 3. Propagate $|ψ^k(t)⟩ = |ψ^k(0)⟩$ forward in time with the field $ε^k(t)$ according to equation (2) to obtain $|ψ^k(T)⟩$.

Step 4. Evaluate the cost functional $K^k$ according to equation (1). If $k \geq 2$, compute $\Delta K^k = K^k - K^{k-1}$ and compare it with the convergence threshold, $\gamma$. If $\Delta K^k \leq \gamma$, then stop the iteration and declare that the optimal pulse has been obtained.

Step 5. Set $⟨χ^k(T)⟩ = ⟨φ(T)|φ(T)⟩$. Propagate $⟨χ^k(T)⟩$ backward in time according to equation (1) using field $ε^k(t)$ to obtain $⟨χ^k(0)|$.

Step 6a. Set $|ψ^{k+1}(t)⟩ = |ψ^k(t)⟩$ and propagate forward in time according to the Schrödinger equation (2) with simultaneous evaluation of the electric field $ε^{k+1}(t)$ at each time step. To calculate $⟨χ(t)|$ use the old electric field, i.e. the electric field generated in the previous iteration step and calculate $|ψ^{k+1}(t)⟩$ with the new electric field.

Step 6b. Compute the new electric field $ε^{k+1}(t)$ from $⟨χ^k(T)⟩$ and $|ψ^{k+1}(t)⟩$ according to

$$
e^{k+1}(t) = \frac{1}{α(t)} \cdot \text{Im}\left\langle χ^k(t)|\frac{∂H}{∂|ψ^k(t)|}\right|ψ^{k+1}(t)⟩.
$$

Step 7. Go back to step 4 and repeat 4–6 until the required convergence has been achieved.

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