Quantum control of population inversion in the presence of spontaneous emission

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Abstract: The detrimental effect of spontaneous emission on the performance of control schemes designed to achieve population inversions between the ground state and a highly excited atomic state is studied using computer simulations.

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We consider a \( N \)-level quantum system with energy levels \( E_n \) and corresponding energy eigenstates \( |n\rangle \) for \( n = 1, 2, \ldots, N \). Assuming that the system is initially in the ground state \( |1\rangle \), our objective is to maximize the population difference between the ground state and the excited state \( |N\rangle \) by applying a sequence of simple control pulses. For simplicity, we shall restrict our attention here to control schemes involving only transitions between adjacent energy levels in the model and control pulses that are resonant with one of the allowed transition frequencies. It will be assumed that the allowed transition frequencies \( \mu_n = E_{n+1} - E_n \) for \( n = 1, 2, \ldots, N - 1 \) are distinct and sufficiently separated so that off-resonant effects can be neglected.

If we neglect the finite lifetimes of the excited states and assume that each control pulse interacts only with its resonant transition, then it can easily be shown, e.g., using Lie group decompositions [1], that a complete population transfer from the ground state to the highest excited state \( |N\rangle \) can be achieved by applying a sequence of \( N - 1 \) control pulses such that the \( n \)th pulse is resonant with the \( n \)th transition frequency \( \mu_n \) and has a total pulse area of \( \pi/(2d_n) \), where \( d_n \) is the absorption oscillator strength of the transition \( |n\rangle - |n + 1\rangle \). The length and shape of the pulses do not influence the outcome of the control process as long as the pulse areas remain fixed. For a real system, however, the decay of the excited states by spontaneous emission reduces the populations of the excited states and ultimately leads to the repopulation of the ground state, which is detrimental to achieving population inversion. Our aim is to study the effect of spontaneous emission on the outcome of the control process using computer simulations. Due to space constraints, we only consider a four-level model of Rubidium with energy levels and transitions as indicated in Fig. 1 as a concrete example. If dissipative effects are neglected then it can easily be seen that a complete population transfer from the ground state \( |1\rangle = |5S_{1/2}\rangle \) to the excited state \( |4\rangle = |6P_{3/2}\rangle \) can be achieved by applying a sequence of three control pulses of frequencies \( \mu_1, \mu_2 \) and \( \mu_3 \) and pulse areas \( \pi/(2d_1), \pi/(2d_2) \) and \( \pi/(2d_3) \), respectively, and that the result of the control process is independent of the shape and length of the control pulses. If dissipative effects such as the finite lifetimes of the excited states are taken into account, however, then a complete population transfer can usually not be achieved, and the performance of the control process depends significantly on the shape and length of the control pulses employed.

Fig. 2 shows the difference between the populations of states \( |1\rangle \) and \( |4\rangle \), \( \rho_{44} - \rho_{11} \), at the conclusion of the control process as a function of the total control time \( T_f \) for various ratios of the pulse lengths, as obtained from a computer simulation of the control process. Note that for very short control times the final yield is around 90% of the theoretical maximum value of one. As the total control time increases, the final yield \( \rho_{44} - \rho_{11} \) decreases since dissipative effects accumulate. However, the computer simulation suggests that the final yield also depends considerably on the ratio of the pulse lengths, not just the total control time \( T_f \). For our four-level Rubidium system, the naive choice of equal pulse lengths, i.e., pulse ratios \( 1:1:1 \), consistently results in the worst yield, independent of the total control time \( T_f \). Another obvious choice of the pulse lengths according to the ratio of the lifetimes of the excited states results only in a marginal improvement. However, the final yield and the threshold value of \( T_f \) for population inversion increase considerably as the ratio of the first two pulses versus the last pulse increases. These at first quite startling results can be partly explained by the vastly different lifetimes of the excited states of Rubidium. The lifetime of state \( |2\rangle = |5P_{3/2}\rangle \) is less than a third of that of \( |3\rangle = |4D_{3/2}\rangle \) and almost one fourth of that of \( |4\rangle = |6P_{3/2}\rangle \). Hence, state \( |2\rangle \) is the weakest link in the chain and it is therefore imperative to minimize the time the system spends in state \( |2\rangle \) in order to reduce dissipative losses. However, choosing the pulse lengths according to the ratio of the
lifetimes of the excited states is not sufficient since the amount of time the system spends in state $|n\rangle$ in the control scheme under consideration is roughly proportional to $(\Delta t_n - 1 + \Delta t_n)/2$, not simply $\Delta t_n$. We see that it is advantageous to choose the pulse lengths $\Delta t_n$, $n = 1, 2, 3$, such that $\Delta t_2 + \Delta t_3 > 3(\Delta t_1 + \Delta t_2)$, which explains why increasing the ratio of the pulse length of the third pulse compared to that of the first two pulses tends to reduce the repopulation of the ground state due to spontaneous emission and thus improves the performance of the control scheme.

Fig. 3 shows the (envelopes of the) control pulses and the corresponding evolution of the energy-level populations if dissipative effects due to spontaneous emission are neglected (center) and if realistic lifetimes of the excited states are assumed (right). The total pulse length in the example is 30 ns, where the first and second pulse are both 6 ns long while the third pulse is 18 ns. The cumulative effect of spontaneous emission is clearly noticeable by comparing the graphs.

The example studied in this paper suggests that it is possible to achieve substantial population inversions between the ground state and a highly excited state of an atom (or possibly ion or molecule) by applying a sequence of short control pulses, even if the finite lifetimes of the intermediate excited states are taken into account. In spite of inevitable population losses due to spontaneous emission, pulse lengths and ratios can be adjusted to minimize dissipative effects and achieve substantial yields. We believe such a scheme could be used to calculate the pulse configurations required to build short wavelength pulse lasers in the UV or even X-ray regime by applying a sequence of short optical-frequency laser pulses to a cloud of trapped atoms or ions to achieve population inversion between the ground state and a highly excited state.

References