Creation of coherent superpositions between metastable atomic states in Doppler-broadened media

N. Sandor, J.S. Bakos, Zs. Sörlei and G.P. Djotyan
Research Institute for Particle & Nuclear Physics, Budapest, POBox 49, Hungary
sandor.nora@rmki.kfki.hu

Abstract. We propose and analyze a scheme for creation of coherent superposition of metastable states in a tripod-structured atom using frequency-chirped laser pulses with negligible excitation of the atoms. The underlying physics of the scheme is explained using the formalism of adiabatic states. We show that the proposed scheme may be equally efficient in homogeneously and Doppler-broadened media. By numerically solving master equation for the density matrix operator, we analyze the influence of relaxation processes on the efficiency of the creation of superposition states. We show that the proposed scheme is robust against small-to-medium variations of the laser field’s parameters.

1. Introduction
Coherent control of the atomic quantum states is a useful tool in numerous fields of optical science and technology. The preparation of the atoms or molecules in coherent superposition states leads to quantum interference between possible quantum paths which may result in drastic variations in the optical properties (absorption and refraction) of the prepared media. As a consequence, significant changes may be experienced in the propagation characteristics of the laser pulses (Electromagnetically Induced Transparency [1]), the efficiency of nonlinear processes can be enhanced[2] and important applications, such as quantum computing[3] or optical information processing [4]-[5] become possible.

In this communication a novel scheme is presented which allows robust creation of large (in certain cases 0.5) coherence between two metastable states of an atom having tripod-like level structure (see Figure 1). It was an important aim to find a scheme that is applicable in presence of spontaneous emission (which means that laser pulses longer than the relaxation time can be used) and in gases of room-temperature. Adiabatic passage schemes based on frequency-modulated (chirped) laser pulses are generally not sensitive to the resonance conditions[4],[6] and allow transition of the populations between metastable states of a multilevel atom without considerable excitation of the atom (in contrary to the other two, widely used adiabatic following methods[7]: stimulated Raman
adiabatic passage (STIRAP)[8]-[9] is sensitive to the two-photon resonance condition between the interacting pulses, while Stark chirped rapid adiabatic passage scheme (SCRAP) is accompanied by temporary excitation of the atom[10].

The creation of the coherent superposition between two metastable states is reached by coupling the metastable states of the atom to the excited state by three equally chirped laser pulses, two in Raman-resonance, the third one out of it. Initially, all the population is set in the metastable state coupled by the pulse out of Raman-resonance. We show that, as a result of the interaction, the population is transferred into a superposition of the other two ground states with a well-defined relative phase, while the excitation of the atom is suppressed during the whole interaction time, that may minimize the effect of the spontaneous emission. As the scheme is not sensitive to the one-photon resonance condition, it may be applied in Doppler-broadened media.

The remainder of the paper is organized as follows. In section 2, we present the mathematical formalism describing the interaction of three frequency chirped (FC) lasers with the tripod-structured atom based on the master equations for the density matrix operator taking into account longitudinal and transverse relaxation processes. In section 3, the underlying physics of creation of the coherent superposition states is discussed using dressed states analysis and the results are compared with the result of the numerical analysis in case of no relaxation or broadening processes. The effect of Doppler-broadening of the transition lines in an atomic gas of the tripod-structured atoms is analyzed in section 4. In section 5, the influence of longitudinal and transverse relaxation processes is discussed on the efficiency of the coherence creation. The results are summarized in section 6.

2. Mathematical formalism
We consider the interaction of three FC laser pulses with a model atom having tripod-structured working levels (see Figure 1). Each laser field is acting on corresponding allowed electric-dipole transition in the atom (between a ground state \(|n\rangle\), \(n\in\{1,2,3\}\) and the excited state \(|0\rangle\) according to selection of the carrier frequencies or polarizations of the laser fields. Transitions between the ground states are forbidden in the electric-dipole approximation. The electric field strength of the laser field has the form \(E(t) = \sum_{i=1}^{3} E_{i}(t) \cos\left(\int_{-\infty}^{t} \omega_{i}(t') dt'\right)\).

A same linear variation in time (linear frequency chirp) of the carrier frequencies of all the interacting laser pulses is assumed in this paper: \(\omega_{i}(t) = \omega_{i}^{(0)} + \beta t\), where \(\omega_{i}^{(0)}\) is the central carrier frequency of the \(k^{th}\) field and \(\beta\) is the speed of the chirp. The central frequency detuning \(\delta_{k}\) from the resonance atomic frequency \(\omega_{k}\) is \(\delta_{k} = \omega_{k} - \omega_{k}^{(0)}, k\in\{1,2,3\}\). Since we assume Raman resonance between the fields \(E_{1}\) and \(E_{2}\), we have equal single-photon detuning for these waves: \(\delta_{1} = \delta_{2} = \Delta,\) see Figure 1.

As we wish to take the relaxation processes into account, the system can be described by the density matrix \(\hat{\rho}\), for which the time evolution is given by the master equation

\[
\dot{\hat{\rho}} = \left[\hat{H}, \hat{\rho}\right] + \hat{R},
\]

where

\[
\hat{H} = \begin{pmatrix}
0 & \Omega_{1} & \Omega_{2} & \Omega_{3} \\
\Omega_{1}^{*} & -\Delta & 0 & 0 \\
\Omega_{2}^{*} & 0 & -\Delta & 0 \\
\Omega_{3}^{*} & 0 & 0 & -\delta_{k}
\end{pmatrix}, \quad \hat{R} = i\hbar\begin{pmatrix}
-\gamma_{10} & -\gamma_{11} + \Gamma/2 & -\gamma_{12} + \Gamma/2 & -\gamma_{13} + \Gamma/2 \\
-\gamma_{12} + \Gamma/2 & 0 & -\gamma_{23} + \Gamma/2 & -\gamma_{24} + \Gamma/2 \\
-\gamma_{13} + \Gamma/2 & -\gamma_{23} + \Gamma/2 & 0 & -\gamma_{34} + \Gamma/2 \\
-\gamma_{14} + \Gamma/2 & -\gamma_{24} + \Gamma/2 & -\gamma_{34} + \Gamma/2 & 0
\end{pmatrix}
\]

are the Hamiltonian in interaction picture and relaxation operator, respectively. The time-dependent Rabi frequencies are \(\Omega_{j} = \sqrt{E_{j}(t)}d_{j}\exp(i\beta t^{2}) = W_{j}f(t)\exp(i\beta t^{2})\), with (in general, different) complex amplitudes \(W_{j}\), \(j = 1,2,3\). We assume that the three pulses have the same Gaussian envelope function \(f(t) = \exp\left(-\left(t/t_{e}\right)^{2}\right)\). \(d_{j}\) is the dipole moment matrix element for transition between the ground state \(|j\rangle\) and the excited state \(|0\rangle\). \(\Gamma = \Gamma_{01} + \Gamma_{02} + \Gamma_{03}\) is the spontaneous decay rate of the excited state \(|0\rangle\) with \(\Gamma_{0j}\) being spontaneous relaxation rate to the ground state \(|j\rangle\); \(\gamma_{kj}\) is the dephasing rate for optical coherence between the states \(|k\rangle\) and \(|j\rangle\), \((k,j = 0,1,2,3)\).
Before presenting and analyzing the results of the numerical simulation of Eq. (1), and to gain an insight into the underlying physics of the processes in the atomic system, let us discuss the behaviour of the system in the case when the relaxation processes may be neglected assuming duration of the pulses much shorter than the relaxations times scales.

For the analysis provided below, it is convenient to introduce new basis functions consisting of the following superpositions of the bare atomic states of the tripod-like atom:

\[
|db_k\rangle = \frac{W_1|1\rangle - W_2|2\rangle}{\sqrt{|W_1|^2 + |W_2|^2}}; \quad |db_2\rangle = \frac{W_1|1\rangle + W_2|2\rangle}{\sqrt{|W_1|^2 + |W_2|^2}}; \quad |db_3\rangle = \frac{W_3|3\rangle}{|W_3|}; \quad |db_4\rangle \equiv |0\rangle
\]

In the new basis, the Hamiltonian of Eq.(4) can be written as:

\[
\hat{H}_{db} = \hbar \begin{pmatrix} 0 & 0 & 0 & f(t) \langle W_1|^2 + |W_2|^2 \rangle \\ 0 & 0 & 0 & \delta_{13} f(t) \langle W_3| W_2 \rangle \\ 0 & f(t) \langle W_1|^2 + |W_2|^2 \rangle & 0 & 2\beta + \Delta \\ f(t) \langle W_3| W_2 \rangle & \delta_{13} f(t) \langle W_3| W_2 \rangle & 0 & 0 \end{pmatrix}.
\]

3. Dressed state analysis

The state vector of the atom in the dark-bright basis can be represented on the basis of the eigenfunctions of the interaction Hamiltonian \( \hat{H}_{db} \) as \( |\psi(t)\rangle = \sum_k n_k(t) |\tilde{b}_k\rangle \exp\left(\int_{-\infty}^{t} \lambda_k(t') dt'\right) \), with initial condition \( |\psi(-\infty)\rangle = \sum_n n_n(-\infty) |\tilde{b}_n\rangle \) where \( \lambda_k \) and \( \tilde{b}_k \) are the eigen-values and eigen-vectors of the Hamiltonian \( \hat{H}_{db} \).

According to the adiabatic theorem[11], in the adiabatic regime of interaction \( n_k(t) = n_k(-\infty) \), \( k \in \{1,2,3,4\} \), which means that if the atom is in one (or superposition) of its eigen-states initially (before the interaction process, at \( t \to -\infty \)), it remains in the same eigen-state (or the superposition of the eigen-states) during the interaction process.

The following eigenvectors can be obtained from the eigen-equation of the Hamiltonian (4):

\[
|\tilde{b}_k\rangle = \begin{pmatrix} f(t) \frac{\sqrt{|W_1|^2 + |W_2|^2}}{\sqrt{N}} \\ \lambda_k (\Delta + 2\beta t) \langle W_1|^2 + |W_2|^2 \rangle \\ f(t) \langle W_3| W_2 \rangle \frac{\lambda_k}{\sqrt{N}} \end{pmatrix}^{(T)}, k \in \{2,3,4\},
\]

and \( |\tilde{b}_1\rangle = (1 \quad 0 \quad 0)^{(T)} \), with the normalization factor

\[
N = \left( f(t)^2 \langle |W_1|^2 + |W_2|^2 \rangle + \frac{1}{\rho 2|W_3|^4} \lambda_k (\Delta + 2\beta t) - f(t) \langle |W_1|^2 + |W_2|^2 \rangle \right)^{1/2},
\]

where \( \lambda_k \) are the eigenvalues of the Hamiltonian, which are plotted in Figure 2. As it can be seen, there is a quasienergy of the system (\( \lambda_3 \)) that has zero value at the beginning and during all the interaction process: \( \lambda_3 = 0 \). This quasienergy corresponds to the dark superposition state \( |db_3\rangle \), (see Eq.(3)). In the same time, there is another quasienergy, \( \lambda_4 \) (see Figure 2), which value, in contrast to the other two quasienergies \( \lambda_2 \) and \( \lambda_3 \), does not depend strongly on the Rabi frequencies and is restricted by the value of the Raman detuning \( \delta_{3}\delta_{3} \):

\[
0 \leq |\lambda_4| \leq |\delta_{3}| \quad (6)
\]

Parameters are normalized to the pulse duration \( \tau_p \).

Figure 2: Time dependence of the eigen-values (quasi-energies) of Hamiltonian (4), \( |\delta_{3}| \tau_p = 250 \).
The energy of this eigenvalue coincides with the energy of the bare state \( |3\rangle \) in the beginning of the interaction \( (t \to \infty) \), and the corresponding eigenstate \( b_3^\lambda \) coincides with \( |3\rangle \). This means that if the atom is prepared in state \( |3\rangle \), its time evolution will follow the evolution of \( b_3^\lambda \), provided the conditions of adiabaticity are fulfilled. At the end of the interaction \( b_3^\lambda \) coincides with the bright superposition of bare states \( |1\rangle \) and \( |2\rangle \), hence a coherent superposition has been established as a result of the interaction.

It is important to note that if the Rabi frequencies are sufficiently large, the excitation of the atom can be suppressed, namely

\[
\left| \frac{b_3^{(t)}}{b_3^{(0)}} \right| = \left| \frac{\lambda_3}{\left( |W_1|^2 + |W_2|^2 \right)^{1/2}} \right| \leq \left| \frac{\delta_3}{\left( |W_1|^2 + |W_2|^2 \right)^{1/2}} \right| \quad \text{(c.f. Eqs. (5) and (6))}.
\]

The results of the dressed state analysis are confirmed by the numerical solution of the master equation (1), if we neglect the relaxation processes in the system. We will show in further parts in this communication that this is a good approximation if the characteristic interaction time is shorter than the time of any relaxation (longitudinal and transversal) process of the system. It can be seen in Figure 3, that as a result of the interaction, a maximum coherence of 0.5 has been induced between states \( |1\rangle \) and \( |2\rangle \) with the complex phase 0, which corresponds to bright state. It is also important to observe that the population of the excited state \( \rho_{00} \) remains negligible during the whole interaction time.

![Figure 3: Time evolution of the populations and coherences. The parameters applied are:](image)

\( W_1 \tau_p = 500, \quad W_2 \tau_p = 475, \quad W_3 \tau_p = 525, \quad \beta \tau_p = 2500, \quad |\delta_3|\tau_p = 250. \)

4. Creation and control of the coherent superposition states in Doppler-broadened media

A Doppler-broadened medium of a gas of tripod-structured atoms is modelled by averaging the created coherence over distribution of the resonance frequencies (velocities) of atoms in the gas at different values of temperature assuming all three FC laser pulses propagating in a same direction.

Considering a gas of \(^{87}\text{Rb}\) atoms at temperature \( T \) and assuming Maxwell-Boltzmann distribution for the velocities of the atoms, we have for the (normalized) probability distribution \( P(\tilde{\Delta}) \) for an atom to have single-photon detuning \( \tilde{\Delta} = \Delta \tau_p \):

\[
P(\tilde{\Delta}) = \left\{ mc^2 \left[ (2\pi)^3 kT \left( f_0 \tau_p \right)^2 \right] \right\}^{1/2} \exp \left\{ -mc^2 \left( \tilde{\Delta} \right)^2 \left[ 8\pi^2 kT \left( f_0 \tau_p \right)^2 \right] \right\},
\]

where \( k \) is the Boltzmann constant, \( m = 86.909 \, \mu \) is the mass of \(^{87}\text{Rb} \) (\( \mu \) being the atomic unit), \( f_0 = 384.230 \, \text{THz} \) is the frequency distance between the excited and the ground states (\( F = 1 \) and \( F' = 0 \) hyperfine states in the \( D_2 \) line of \(^{87}\text{Rb}\)).

The average values of the density matrix elements: \( \langle \rho_{kl} \rangle \), \( (k,l = 0,1,2,3) \) are calculated numerically. First the master equation (1) is solved numerically for the values of \( \tilde{\Delta} \) corresponding to nonzero probability values (see Eq.(7)) in order to obtain the density matrix elements \( \rho_{kl}^{\text{final}}(\tilde{\Delta}) \) at the end of the interaction \( (t \to \infty) \). One can find that there is a range of values of the Doppler shift \( \tilde{\Delta} \), where the final coherences (populations) are independent on \( \tilde{\Delta} \). This feature is due to the frequency-chirping of the laser pulses: as long as the Doppler shift is smaller than the frequency range...
[−4βτ_p, 4βτ_p] covered by the chirp span during the interaction time (approximately equal to 4τ_p), the velocity of motion of the atoms does not have an impact on the resulting population and coherence distribution.

The absolute value of the average induced coherence \( \langle \rho_{12} \rangle \) established after the interaction with the laser field is presented in Figure 4 as a function of the normalized frequency span \( \beta \tau_p^2 \) of the laser pulses for different values of the gas temperature. As it can be seen from this Figure, the average value of the induced coherence does not depend on the Doppler-broadening for sufficiently large frequency span of the pulses during the interaction time due, for example to a sufficiently high speed of the frequency chirp.

5. Effect of the relaxation processes

The dependence of the states final populations and of the phase of the created coherence established after the interaction with the laser pulses is shown in Figure 5 as a function of the longitudinal relaxation rate. One could anticipate a negligible influence of the spontaneous relaxation processes on the populations and coherences of the atom when no considerable excitation of the atom takes place. However, the results of the numerical simulations show, that even for the negligible excitation of the atom, the final populations and induced coherences depend on the longitudinal relaxation rate. The reason is linked to the optical coherences \( \rho_{kk} \), \( (k=1, 2, 3) \) that are not negligibly small and depend on the longitudinal relaxation rate \( \Gamma \), (see Eqs. (1) and (2)). As it can be seen from Fig.9, the larger the longitudinal relaxation rate (or longer the laser pulses), the more of the atomic poopulation is transferred (optically pumped) into states \|1\rangle \) and \|2\rangle \) connected by the laser pulses in Raman resonance. Insets in the Figures show behavior of the phase of the induced coherence, which imply that, for longer laser pulses, the atom ends up in the dark superposition in both cases of the positive and negative Raman detuning.

Since the quantum interference processes are the basis for the schemes of creation of coherent superposition states, the phase relations between the states probability amplitudes (the values and the phases of the corresponding coherencies) must play an important role in the considered processes. That is why a strong effect of the transverse relaxation (dephasing) processes may be anticipated on creation and control of the coherent superposition states as well as on the population transfer between the atomic states. Our numerical analysis shows that the effect of the transverse relaxation begins to be imperative at \( \tau_p \approx 1/(\gamma \cdot 10^{12}) \). If the interacting pulses are longer, the transverse relaxational
processes destroy the adiabatic transfers. At large transverse relaxation rates, all the states are equally populated and there is no coherent superposition created as a result of the interaction.

6. Conclusions
We have presented and investigated a scheme for creation of arbitrary coherent superposition of two ground states in a tripod-like atom without considerable excitation of the atom using FC laser pulses.

We have analyzed the applicability of the scheme in a Doppler-broadened medium of a gas composed of tripod-structured atoms by averaging the induced coherence over the velocity distribution of the atoms in the gas at different temperatures. The results show that the scheme is effective even for relatively large widths of the Doppler–broadened transition lines if the frequency span of the laser pulses due to the chirp exceeds the width of the Doppler-broadening.

We have analyzed the influence of the relaxation processes on the population transfer and creation of the superposition states by numerical simulation of the master equation for the density matrix elements. We have shown that the considered scheme allows minimizing the effect of the spontaneous decay of the excited states by suppression of the population of the excited state. However, even under the condition of negligible excitation of the atom, for longer laser pulses, the longitudinal relaxation may influence the induced coherences and the resulting population distribution among the ground states. For laser pulses longer that the decay time of the excited state, optical pumping of the atom by a pair of the pulses in Raman resonance results in accumulation of the atomic population in dark superposition of the ground states linked by the laser pulses in Raman resonance. It is worth noting at this point that the influence of the spontaneous decay may be minimized by increasing the speed of the population transfer.

As it may be anticipated, the transverse relaxation (dephasing) has a strong destructive effect in the considered scheme. The numerical simulation of the master equation has shown that already at duration of the laser pulses close to the dephasing constant of the medium the transverse relaxation processes destroy the adiabatic transfers. At larger transverse relaxation rates or longer laser pulses, all the states are equally populated as a result of interaction and there is no coherent superposition states created. While the detrimental effect of the transverse relaxation may be avoided by utilizing sufficiently short laser pulses, this effect may be also diminished by increasing the chirp speed of the FC pulses.

The presented adiabatic scheme of coherence creation may find applications in the fields of quantum and nonlinear optics, atomic interferometry and in mapping and long-time storage of optical information in populations and coherences of metastable atomic states.

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