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Robust population transfer in atomic beams induced by Doppler shifts

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Abstract The influence of photon momentum recoil on adiabatic population transfer in an atomic three-level lambda system is studied. It is shown that the Doppler frequency shifts, due to atomic motion, can play an important role in adiabatic population transfer processes of atomic internal states by a pair of laser fields. For the limiting case of slow atoms (Doppler shift much smaller than the photon recoil energy), the atoms occupy the same target state regardless of the order of switching of laser fields, while for the case of fast atoms interacting with the intuitive sequence of pulses, the target state is the intermediate atomic state. Furthermore, it is shown that this novel technique for adiabatic population transfer is related to a level crossing in the bright–intermediate state basis (rather than in the original atomic basis). It is shown that these processes are robust with respect to parameter fluctuations, such as the laser pulse area and the relative spatial offset (delay) of the laser beams. The obtained results can be used for the control of temporal evolution of atomic populations in cold atomic beams by externally adjustable Doppler shifts.

1 Introduction

The transitions between atomic internal states induced by resonant laser field are always accompanied by momentum transfer from photons to atoms. Many elements of atom optics, e.g., deflectors and splitters, are based on this effect [1]. These elements separate the atomic wave function into several components with different momenta [2–7]. A common approach for implementing such elements is to apply laser pulses with suitable temporal pulse areas (e.g., the so-called π or $\pi/2$ pulses). This technique, however, is not robust because it requires a carefully controlled duration and power of the pulse in order to assure the desired area. An important paper by Marte et al. [5] showed that an atomic beam deflection by the stimulated Raman adiabatic passage (STIRAP) technique [6] can be implemented via stimulated Raman transitions induced by counterpropagating laser beams. Later on, this idea was demonstrated independently by Lawall et al. [7] and Goldner et al. [8]. Those publications stimulated the recognition of STIRAP, which was initially developed for efficient excitation of molecules. Later, Theuer et al. [9] developed a laser-controlled variable beam splitter based on the tripod-STIRAP scheme [10–12], by varying the spatial overlap of lasers.

For more details concerning current theoretical, experimental developments and applications of STIRAP, the interested readers are referred to the review articles [13–17]. For completeness, however, we give a brief description of STIRAP. The underlying physical mechanism of STIRAP is the existence of an adiabatically decoupled, or dark state [18], which at early times coincides with the initial state and at late times is aligned with the target state. When the pump and Stokes frequencies together maintain two-photon resonance, then the only conditions to be fulfilled for successful transfer are those of adiabaticity [13–17, 19], i.e., large pulse area, and counterintuitive application of the pulses, i.e., Stokes before pump.

The schemes suggested in Refs. [5] and [9] operate in the regime where Doppler shifts of atomic transition frequencies due to the photon recoil and the recoil energy are small compared to the Rabi frequencies and the transit-time

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broadening. In this limit, the combined system of atom plus fields is equivalent to a lambda or tripod atomic system in which internal states can be labeled by different photon recoil momenta. The idea of the beam splitters is then very clear: A population transfer between these internal states via STIRAP [6] or via tripod-STIRAP [10–12] necessarily means photon momentum transfer to the particles in the atomic beam. Thus in this regime, because of the darkness of the state, the photon recoil momentum does not play a role in the evolution of internal atomic states. It should be noted that standing wave laser beams (see, e.g., [20–22] and references therein) offer more efficient deflection of atoms than those with traveling wave beams as in Refs. [5–9]. Our goal, however, will not be to find an efficient way to deflect atoms; rather, we ask a different question, namely whether is it possible to control the dynamics of atomic internal states in a *robust* way via the velocity-induced Doppler shift of the atomic transition frequency. The problem, as far as the author knows, with such formulation within the context of robustness has never been discussed before (see the recent review preprint [23]). The novelty of the present study is focused on the fact that due to Doppler shifts, the Landau–Zener transition [24–26] may occur between the bright and intermediate atomic states (rather than in the bar atomic basis). In the original atomic basis, for the case when laser beams are ordered intuitively, i.e., the bright state is populated, this transition corresponds to a robust population transfer from the initial to intermediate atomic state, while for the counterintuitive sequence, STIRAP remains valid for both cases of slow and fast atoms as long as the effective Rabi frequency is sufficiently large.

2 Model

Let us consider a monoenergetic atomic beam, crossing two laser beams but not necessarily at a right angle. In particular, we consider an atomic Λ system with three levels coupled by two counterpropagating optical fields with the same frequency ω and wave vectors k whose parallel propagation axes are spatially shifted. The geometry of the interaction between atoms and lasers and the atomic level scheme are shown in Fig. 1.

The sequence of interaction of atoms with laser beams is controlled by the spatial displacement of their axes. The initial atom velocity in the direction of the laser beams can be varied by changing the intersection angle α between atomic velocity $\mathbf{v}$ and laser beams $\mathbf{k}$. The motion of atoms causes a Doppler shift $\sim \mathbf{v} \cdot \mathbf{k}$. The Doppler shift is large for small crossing angles $\alpha \approx 0$, and for large crossing angles α close to $\pi/2$ it tends to zero. As we will see later on, the possibility of controlling the Doppler shift could

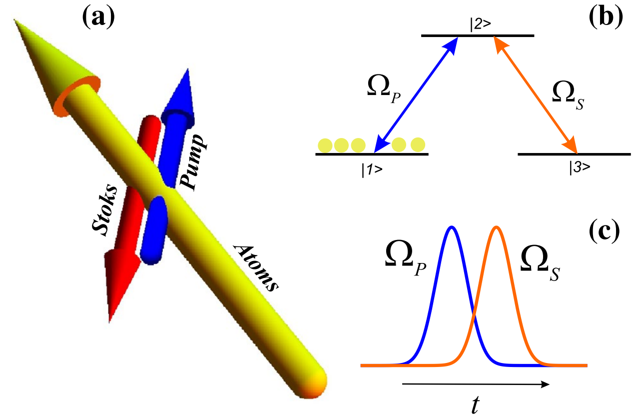

Fig. 1 **a** Sketch of the geometry of the interaction between atoms with two counterpropagating laser fields. **b** Three-level system with initial population in state $|1\rangle$. **c** Pulses in the intuitive sequence, the pump pulse precedes the Stokes pulse

provide a new way for controlling adiabatic evolution processes of internal states of atoms.

The laser fields are detuned from the atomic transition by the single-photon detuning $\Delta = \omega_{21} - \omega = \omega_{23} - \omega$. In what follows, we will consider only the case of exact one-photon resonance, $\Delta = 0$. The generalization to nonzero detuning is straightforward and requires one only to replace the recoil energy

$$E_{\mathrm{r}} = \frac{\hbar^2 k^2}{2M} \tag{1}$$

with $E_{\mathrm{r}} + \Delta$. We assume that the atom–laser interaction time is much shorter than the spontaneous lifetime of state $|2\rangle$. This condition can be fulfilled, for example, for fast atomic beams.

The atoms are described by atomic flip operators $\sigma_{nm} = |n\rangle\langle m|, n, m = 1, 2, 3$. The Hamiltonian for the atom-field coupling along the z axis (z is directed along the propagation direction of the one of the lasers) in rotating wave approximation has the following form [5]

$$\begin{aligned} H = & -\frac{\hbar^2}{2M}\frac{\partial^2}{\partial z^2} \\ & + \frac{\hbar\Omega_{\mathrm{P}}(t)}{2} e^{-ikz}\sigma_{12} + \frac{\hbar\Omega_{\mathrm{S}}(t)}{2} e^{+ikz}\sigma_{32} + \mathrm{H.c.}, \end{aligned} \tag{2}$$

where M is the mass of the atom. $\Omega_{\mathrm{P}}(t)$ and $\Omega_{\mathrm{S}}(t)$ are space-independent, time-delayed Rabi frequencies of the pump and Stocks fields, respectively. We have omitted the transverse part of the kinetic energy since it is a conserved quantity. Using the gauge transformation

$$\begin{aligned} G = & \exp\left[ikz(\sigma_{11} - \sigma_{33})\right] \\ = & \sigma_{22} + \sigma_{11}\exp(ikz) + \sigma_{33}\exp(-ikz) \end{aligned} \tag{3}$$

the Hamiltonian (2) can be transformed to a more convenient form

$$H \to G \cdot H \cdot G^{-1} \to \tag{4}$$

$$= \frac{\hbar k}{M} \cdot p(\sigma_{11} - \sigma_{33}) - \frac{\hbar^2 k^2}{2M}\sigma_{22} + \frac{\hbar \Omega_{\mathrm{P}}(t)}{2}\sigma_{12} + \frac{\hbar \Omega_{\mathrm{S}}(t)}{2}\sigma_{31} + \mathrm{H.c}$$

Here the terms containing the constants of motion have been omitted. Note that the atomic momentum commutes with the transformed Hamiltonian. The momentum operator then is no longer a dynamical variable in this frame, and it has been replaced by a real parameter p. We see that the states $|1\rangle$ and $|3\rangle$ are shifted from the two-photon resonance condition by the Doppler detunings $\frac{\hbar k p}{M}$. The energy of the state $|2\rangle$ is shifted by the recoil energy $\frac{\hbar^2 k^2}{2M}$.

The state vector $|\Psi\rangle$ transforms under the gauge transformation (3) into $G|\Psi\rangle$. Throughout the paper, we assume that the initial state of the system is $|\Psi\rangle = \exp\left(-\frac{i}{\hbar} p_0 \cdot z\right)|1\rangle$, where p_0 is the atom's momentum projection onto the direction of the laser beams. We are interested in the evolution of the system with the Hamiltonian (4) when the Rabi frequencies $\Omega_{\mathrm{P}}(t)$ and $\Omega_{\mathrm{S}}(t)$ are varied adiabatically in time.

2.1 Adiabatic evolution

To analyze adiabatic evolution of the system, it is convenient to introduce dark, bright and atomic states by the following orthogonal transformation

$$U(t) = \begin{pmatrix} \cos\theta(t) & 0 & -\sin\theta(t) \\ 0 & 1 & 0 \\ \sin\theta(t) & 0 & \cos\theta(t) \end{pmatrix}. \tag{5}$$

The mixing angle $\theta(t)$ is defined by

$$\tan\theta(t) = \frac{\Omega_{\mathrm{P}}(t)}{\Omega_{\mathrm{S}}(t)}. \tag{6}$$

In the adiabatic limit of slowly changing mixing angle,

$$\frac{\mathrm{d}\theta(t)}{\mathrm{d}t} << \Omega_{\mathrm{eff.}}(t), \tag{7}$$

$$\Omega_{\mathrm{eff.}}(t) = \sqrt{\Omega_{\mathrm{P}}^2(t) + \Omega_{\mathrm{S}}^2(t)}, \tag{8}$$

the corresponding Hamiltonian transforms into (for sake of simplicity, we omit below the argument t).

$$H = H_{DD} + H_{D\overline{D}} + H_{\overline{D}D} + H_{\overline{D}\overline{D}}, \tag{9}$$

where

$$H_{DD} = \frac{\hbar k p}{M}|D\rangle\langle D| \cos 2\theta, \tag{10}$$

$$H_{D\overline{D}} = H_{\overline{D}D}^{\dagger} = \frac{\hbar k p}{M}|D\rangle\langle B| \sin 2\theta, \tag{11}$$

$$H_{\overline{D}\overline{D}} = -E_{\mathrm{r}}\sigma_{22} - \frac{\hbar k p}{M}|B\rangle\langle B| \cos 2\theta + \frac{\hbar\Omega_{\mathrm{eff.}}}{2}|B\rangle\langle 2| + \mathrm{H.c} \tag{12}$$

and the new orthogonal basis vectors are the dark $|D\rangle$ and bright $|B\rangle$ states defined in the following way

$$|D\rangle = |1\rangle\cos\theta - |3\rangle\sin\theta, \tag{13}$$

and

$$|B\rangle = |1\rangle\sin\theta + |3\rangle\cos\theta. \tag{14}$$

The structure of the Hamiltonian (9) tells us that the terms $H_{\overline{D}D}$ and $H_{D\overline{D}}$ induce transitions from the dark state to its complement subspace (bright and atomic states) and vice versa. A sufficient condition to neglect them is to ignore the term $\frac{\hbar k p}{M}\sin 2\theta$ compared to

$$\delta = \min\left[\left|\frac{\hbar k p}{M}\cos 2\theta - \mu_1\right|, \left|\frac{\hbar k p}{M}\cos 2\theta - \mu_2\right|\right] \tag{15}$$

i.e.,

$$\frac{\hbar k p}{M}\frac{\sin 2\theta}{\delta} << 1, \tag{16}$$

where $\mu_{1,2}$ are the eigenvalues of $H_{\overline{D}\overline{D}}$. One can verify that for relatively large Rabi frequencies

$$\Omega_{\mathrm{eff.}} >> \frac{\hbar k p}{M}\sin 2\theta, \tag{17}$$

(along with the assumptions $kp > 0$ and $\min[\mu_1, \mu_2] < \frac{\hbar k p}{M}\cos 2\theta < \max[\mu_1, \mu_2]$) condition (16) is fulfilled and the dark state decouples from the dynamics. By combining this condition with the condition of the adiabaticity (7), we arrive at the sufficient condition

$$\Omega_{\mathrm{eff.}} >> \max\left[1/T, \frac{\hbar k p}{M}\sin 2\theta\right], \tag{18}$$

where T is a time characterizing the pulse duration. This condition ensures that the system remains either in the initial dark state or in its complement subspace. In particular, if a counterintuitive pulse sequence is applied, i.e., if Ω_{S} is switched on and off before Ω_{P}, i.e., the mixing angle starts at zero and then increases to $\pi/2$, thus the system is initially in the dark state $|D\rangle$ and will remain in this state, while according to Eq. (13) the initial state $|1\rangle$ will transform, up to an irrelevant phase (proportional to the Doppler shift), into $|3\rangle$. This dynamics has been described in reviews [13–17]. The inverse gauge transformation (3) will yield atomic beam deflection due to the momentum exchange $2\hbar k$ between the atoms and laser photons. As

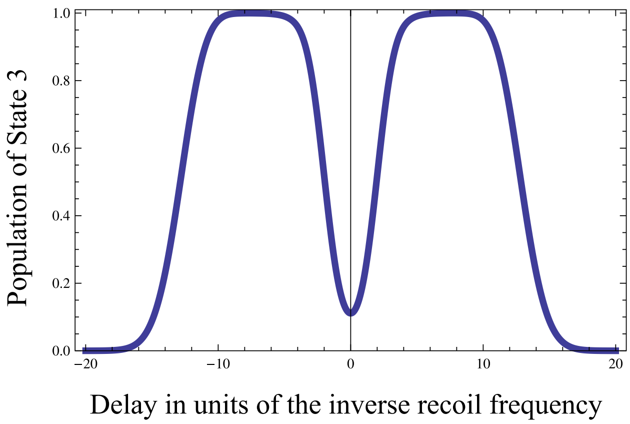

Fig. 2 Population of the state 3 as a function of the time delay between pulses. The pulses are as in Eq. (19) with equal pulse durations $E_r T_P = E_r T_S = 10\hbar$ (time is measured in units of the inverse recoil frequency), and equal areas $\hbar\Omega_0 = 5E_r$. Positive values of the delay correspond to STIRAP. The initial state of the atom is $|1\rangle$ and $p_0 = 0.1\hbar k$

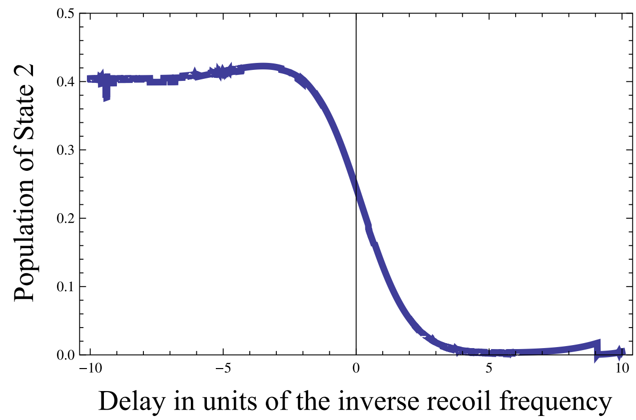

Fig. 3 Maximum population of state $|2\rangle$ during the evolution as a function of the time delay between the pulses. Other parameters and units are the same as in Fig. 2

was mentioned in the introduction, in this case, the Doppler shift does not play a role in dynamics of the system.

3 Population dynamics

3.1 Slow atomic beams

In this subsection, we consider the case of the initial momentum of the atoms p_0 in the direction of the laser beam being much smaller than the recoil momentum $\hbar k$. This corresponds to the case when the atomic beam is almost perpendicular to the laser beams.

Figure 2 shows the final atomic population of the state $|3\rangle$ as a function of the delay between the coupling pulses. This figure was obtained by numerically integrating the Schrödinger equation with the full Hamiltonian (9).

The pulses have Gaussian shape

$$\Omega_P(t) = \Omega_0 \exp\left[-\left(\frac{t-\tau}{T}\right)^2\right],$$
$$\Omega_S(t) = \Omega_0 \exp\left[-\left(\frac{t+\tau}{T}\right)^2\right] \quad (19)$$

with equal pulse durations $E_r T_P = E_r T_S = 10\hbar$ (in units of the inverse recoil frequency), and equal areas $\hbar\Omega_0 = 5E_r$. The initial momentum of the atoms is $p_0 = 0.1\hbar k$. The atoms are prepared initially in the state $|1\rangle$. Positive values of the delay correspond to STIRAP, and negative delays to an intuitive pulse sequence; i.e., the pulse Ω_P precedes Ω_S. For both positive and negative delays, we observe an efficient population transfer from the initial state $|1\rangle$ to the state $|3\rangle$. The transfer efficiency approaches unity for a relative broad range of delays. The case of STIRAP was discussed in [5]. In addition to that, Fig. 3 shows that for negative delays in contrast to ordinary STIRAP, the state $|2\rangle$ is substantially populated during the time evolution of the system. This indicates that the transfer does not occur via adiabatic rotation of the dark state from $|1\rangle$ to $|3\rangle$.

The state vector for the initial problem can be found by the inverse gauge transformation (3) which leads to the change of the atomic momentum from p_0 to $p_0 + 2\hbar k$. Hence, if the timescale of the interaction is smaller than the radiative lifetime of state $|2\rangle$, and the initial momentum of the atoms p_0 is much smaller than the recoil momentum, an intuitive pulse sequence also transfers completely the population of the initial state $|1\rangle$ to the state $|3\rangle$. Hence, an atomic beam deflection can be implemented by spatially shifted laser beams with large pulse areas.

The mechanism of the population transfer for the negative delays (intuitive sequence of pulses) can be understood qualitatively as follows: If an intuitive pulse sequence is applied, the system starts its evolution from the bright state $|B\rangle$, see Eq. (14). Due to the large recoil energy ($\hbar k >> p\cos 2\theta$), which plays the role of a detuning, the bright state undergoes an adiabatic return process under the condition

$$E_r T >> \hbar. \quad (20)$$

Because of the chosen intuitive pulse order, the bright state transforms to the bare atomic state $|3\rangle$. This mechanism is similar to the so-called b-STIRAP process [6, 27, 28]. The condition (20), however, might be questionable for atomic beams, because of the constraint on the radiative lifetime. Indeed, it is important to recall that the duration of the interaction of atoms with lasers should be shorter than the upper state lifetime. Therefore, the condition $E_r\tau_{sp} >> \hbar$

should be satisfied, where τ_{sp} is the lifetime of state $|2\rangle$. In fact, the opposite situation occurs in many experiments, e.g., with noble atoms [32].

3.2 Fast atomic beams

Consider now the case where the initial momentum of the atom is larger than the photon momentum. Figure 4 shows the results of numerical simulation for the final populations of states $|2\rangle$ and $|3\rangle$ for fast atoms ($p_0 = 4\hbar k$) depending on the time delay of pulses. For positive large delays, the system ends its evolution in state $|3\rangle$, i.e., the process is STIRAP-like. The atom thus receives a momentum kick of $2\hbar k$. On the other hand, for negative delays (around $-0.8T$) the population transfer from $|1\rangle$ to $|2\rangle$ occurs and, therefore, the momentum of the atom in the direction of laser beams changes by $\hbar k$. The transfer probability from $|1\rangle$ to $|2\rangle$ is robust for a wide range negative delays. Figure 5 shows the dependence of the final population in state $|2\rangle$ for $\tau = -0.75T$ and $\tau = -0.5T$ as a function of the pulse area $\Omega_0 T$. We thus see that the adiabatic transfer to state $|2\rangle$ is more efficient for the case of largely delayed pulses. Figure 5 also shows that there is a pronounced threshold area starting at $\Omega_0 T \gtrsim 30$ for an efficient population transfer into state $|2\rangle$. These observations are in full agreement with condition (17).

In the following, we examine the dynamics of atomic populations in the case of substantially delayed laser pulses corresponding to small values of $\sin 2\theta$. In this case, condition (17) is easy to satisfy for small Rabi frequencies. Hence, the coupling between the dark and bright components is negligible.

For a counterintuitive pulse sequence, the system is initially in a dark state and the dynamics of the system is STIRAP-like. We, therefore, discuss only the case when the Stokes and pump pulses are ordered intuitively, i.e., the system starts its evolution from the bright state (14). By neglecting the term (11) in Hamiltonian (9), we arrive at the finite Landau–Zener Hamiltonian [24–26]

$$H = \Delta(\theta)|B\rangle\langle B| + \frac{\hbar\Omega_{\mathtt{eff}}}{2}(|B\rangle\langle 2| + |2\rangle\langle B|). \quad (21)$$

Here $\Delta(\theta) = E_r - \frac{\hbar k p}{M}\cos 2\theta$ is the effective detuning between $|B\rangle$ and $|2\rangle$ states, whose asymptotics is given by $E_r + \frac{\hbar k p}{M}$ early times and by $E_r - \frac{\hbar k p}{M}$ at late times. Depending on the ratio $E_r / \frac{\hbar k p}{M}$ level crossings (resonance) between the bright and atomic states may or may not occur. It is easy to verify that the condition $p > \frac{\hbar k}{2}$ is essential for the occurrence of a Landau–Zener-like transition at the crossing point $\cos 2\theta(t_0) = \frac{\hbar k}{2p}$. For Gaussian pulses shapes (19), the slope of the effective detuning $\frac{\mathrm{d}\Delta(\theta)}{\mathrm{d}t}$ at the crossing point is given by

$$\beta = \left.\frac{\mathrm{d}\Delta(\theta)}{\mathrm{d}t}\right|_{t=t_0} = 4E_r \frac{p}{\hbar k}\left(1 - \frac{\hbar k}{2p}\right)\frac{\tau}{T^2}. \quad (22)$$

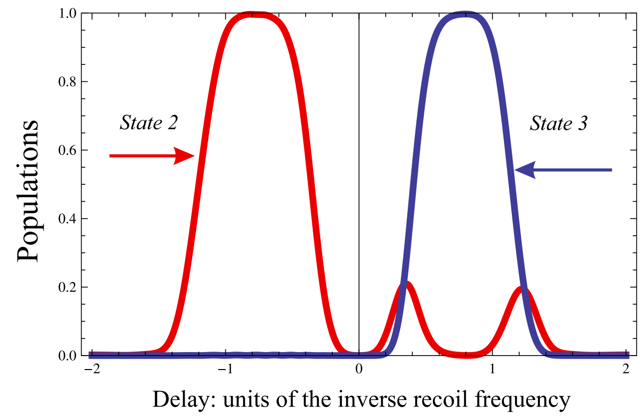

Fig. 4 Final populations of states $|2\rangle$ (red) and $|3\rangle$ (blue) as a function of the time delay between pulses. The initial momentum of atoms is $p_0 = 4\hbar k$. Other parameters are $E_r T = \hbar, \hbar\Omega_0 = 50E_r$

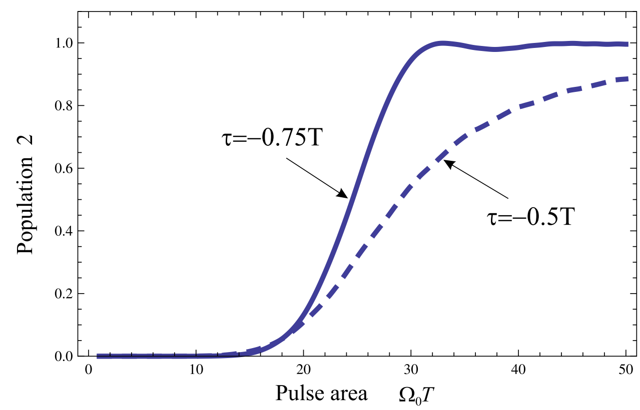

Fig. 5 Final population of state $|2\rangle$ under conditions of Fig. 4 as a function of $\Omega_0 T$, for different delay times of pulses

The condition $p > \frac{\hbar k}{2}$ with the adiabatic condition $\Omega_{\text{eff.}} T >> 1$ guarantee a robust population transfer from the bright to the atomic state. In this scheme, the atoms receive a momentum kick only from the pump photons. The role of the Stokes laser is to assist the Landau–Zener transition between the bright $|B\rangle$ and $|2\rangle$ states.

It is of interest to estimate a scaling relationship for the sensitivity of the Landau–Zener transition to the Doppler shift. For an atom initially in the bright state, the model (21) suggests the following expression [24–26]

$$P_2 = 1 - \exp\left(-\frac{\pi\Omega^2_{\mathtt{eff.}}}{2\beta}\right) \quad (23)$$

for the population of the middle state. Here β is the slope of the effective detuning Eq. (22). It is easily seen from this expression that the condition,

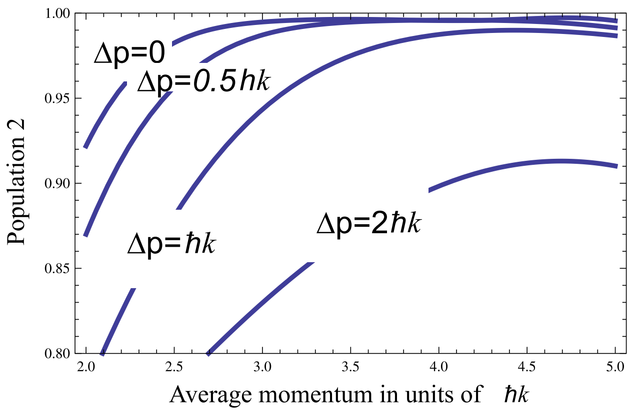

Fig. 6 Average final population of state $|2\rangle$ over the momentum under conditions of Fig. 4 as a function of average momentum $\bar{p}$, for different values of Δp

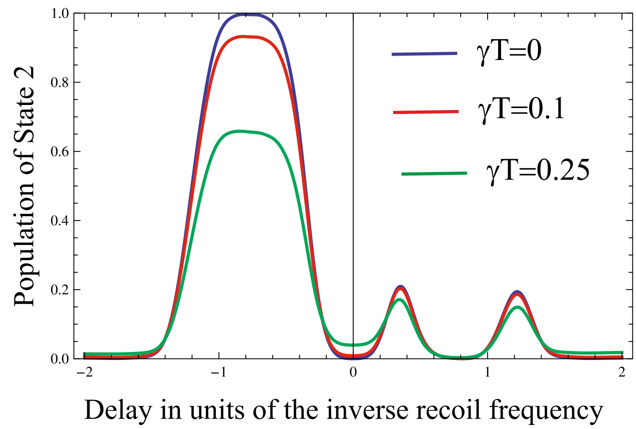

Fig. 7 Final population of the state $|2\rangle$ versus the time delay for different values ($\gamma T = 0, 0.1$ and 0.25) of the pulse duration relative to the lifetime of the middle state. Other parameters and units are the same as in Fig. 4

$$\hbar k \lesssim |p| \lesssim \frac{\pi}{8}\hbar k \frac{(\Omega_{\mathtt{eff}}.T)^2}{\frac{E_r\tau}{\hbar}\ln\frac{1}{1-P_0}} \tag{24}$$

is required in order to achieve transfer efficiency above the level P_0. Analogously, expressions for the efficiency of STIRAP and b-STIRAP processes with the two-photon detuning have been derived in previous studies [29, 30] (see also references in the review preprint [23]).

Hence, as Eq. (24) shows in order to have a higher transfer efficiency to the middle state the speed of atoms must be within certain interval. In real atomic beam experiments, the atomic beam is never perfectly monoenergetic nor do all the atoms move with the same speed. The spread in speed results in different Doppler shifts. Hence, for some fraction of slow atoms the condition (24) can be violated and some population will go to the state 3. Therefore, one has to perform an averaging over the velocity distribution of the transition probability. We expect that the transfer efficiency to the middle state would be high even for atom beams with a poor quality as long as the spread of velocities distribution is much smaller than $\hbar k$. Indeed, the simulation results (see Fig. 6) confirm this prediction. Figure 6 shows how the transfer efficiency eventually decreases as the width Δp of the Maxwell distribution $\frac{1}{\sqrt{2\pi}\,\Delta p}\exp\left(-\frac{(p-\bar{p})^2}{2\Delta p^2}\right)$ increases. The transfer efficiency is seen to be essentially independent of Δp if $\Delta p < \hbar k$. However, the beam parameters $\Delta p > 2\hbar k$ can have an appreciable effect on the transfer efficiency.

Before concluding this subsection, we briefly discuss V and cascade atomic schemes. We note that, in limiting cases (slow atoms and absence of the spontaneous decay) the distinction between lambda, V and ladder linkage patterns is not essential. For an atomic ensemble and a ladder linkage, the two-photon resonance condition is independent of velocity. Therefore, regardless of the ratio $p/\hbar k$ and order of switching of laser fields, the ladder-atoms will end up in the same target state due to STIRAP or b-STIRAP processes. In the absence of decoherence, all possible adiabatic processes for Λ and V schemes are the same.

3.3 Decoherence processes

In an atomic beam experiment, collisions have no influence, but the possibility of spontaneous emission is always present. Only this mechanism can produce changes not predicted within the coherent dynamics of the Schrödinger equation. As described here, the successful population transfer to the atomic state 2 takes place by means of adiabatic time evolution. We expect that when pulses are shorter than the spontaneous emission lifetime, meaning $\gamma T << 1$, then description of the dynamics can be provided by the time-dependent Schrödinger equation. But as the pulses become longer, a situation requires treatment using a density matrix. To verify the validity of our approximation, we have carried out simulations using more elaborate density matrix equations of motion. We took these to be of Lindblad form [31]

$$\frac{\mathrm{d}\rho}{\mathrm{d}t} = \frac{1}{i\hbar}[H,\rho] + \frac{\gamma}{2}\sum_{n=1}^{2}\left(\left[L_n\rho, L_n^\dagger\right] + \left[L_n, \rho L_n^\dagger\right]\right), \tag{25}$$

where $L_1 = \sigma_{12}$ and $L_2 = \sigma_{32}$ are the Lindblad operators modeling the longitudinal relaxation of the upper state to lower states 1 and 3, with equal rates γ. We have neglected here the irreversible decay of the upper state 2. It can be satisfactorily treated by simply including a loss (imaginary energy) from the upper state. Figure 7 shows the transfer process to the state 2 versus the time delay for different values of the pulse duration relative to the lifetime of the middle state. As can be seen, considerable population is lost from the state 2 when one has pulses as long as $\gamma T > 0.25$. From the results shown in Fig. 7, it is clear that the results predicted here by the Schrödinger equation are generally valid for $\gamma T < 0.1$.

4 Conclusion and discussion

It is shown that the Doppler shift can play an important role in the population transfer in three-level atoms driven by two counterpropagating spatially shifted laser fields. In particular, when the atoms interact with intuitively ordered laser pulses, depending on the ratio of the initial atomic and photonic momenta, the final atomic states are different. Namely, for $p_0 << \hbar k$ and large recoil energies $E_r T >> 1$ the atoms occupy state $|3\rangle$ and receive $2\hbar k$ momentum regardless of the order of switching the laser fields, while for the case of fast atoms and an intuitive sequence of pulses, the target state is the intermediate state $|2\rangle$ and the corresponding momentum kick is $\hbar k$. We showed that in the case of slow atoms and intuitive sequence of laser pulses, the bright state, which is a linear combination of the initial $|1\rangle$ and target $|3\rangle$ states, undergoes an adiabatic return process (due to the large recoil energy). In the bare atomic basis, this process corresponds to the $|1\rangle \to |3\rangle$ transition, whereas in the second case (fast atoms), the Landau–Zener transition occurs between the bright and atomic state $|2\rangle$ and the system transforms from the initial state $|1\rangle$ into state $|2\rangle$.

For an experimental observation of these effects, the interaction time between atoms and laser fields should be short enough $T << \tau_{sp.}$. To fulfill this condition, a fast atom beam is required. For implementing such a situation, a metastable cold helium beam with $^3S_1$ and $^3P_1$ transitions could be a possible candidate. Simple calculations show that to avoid the spontaneous emission from the excited state $^3P_1$, the helium atoms must enter the interaction region with velocity in the range of 10–100 m s^{-1}. This is achievable for metastable cold helium beams as was reported by Oberst et al. [33]. Another possible system which would be immune to the effects of the spontaneous decay can be an ensemble of atoms with two lower stable levels both coupled to a Rydberg state by laser fields [34]. Atoms in highly excited Rydberg states are remarkably stable against spontaneous emission. The interaction between Rydberg atoms, however, could play an important role in the evolution of atomic populations [35]. An interesting extension of this work will be to study the influence of atom–atom interactions on the effects considered above.

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