Laser cooling during velocity-selective adiabatic population transfer

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Velocity-selective adiabatic population transfer by time-delayed laser pulses is considered. In this process only a narrow part of an atomic momentum distribution follows the ''dark'' state adiabatically. The remaining atoms are excited and partially pumped into the dark state. We show that this effect can be used for cooling atoms below the photon recoil energy using pulsed laser radiation. [S1050-2947(96)51209-6]

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Stimulated Raman adiabatic passage (STIRAP) has become nowadays an established technique in atomic and molecular physics for efficient transfer of population between the states of an atomic system [1]. The idea of STIRAP is based on the concept of a ''dark'' state, which is a superposition of only ground atomic states and which is not coupled to an excited state. An atom in such a dark state can adiabatically follow a changing light field, transferring population from one ground state of Raman configuration to another. The process is accomplished without the intermediate excited state being significantly populated at any time, the state that would introduce population loss by spontaneous emission. Therefore the ideal STIRAP is a coherent process. Its high efficiency and selectivity, not very sensitive to shape, intensity, power, and frequency stability of the used laser pulses, have recently attracted much attention in relation to atom optics and interferometry [2–5]. The fact that the dark state is velocity-selective in traveling counterpropagating light waves is used, and a distinct momentum is associated with each atomic state. Therefore the adiabatic transfer of population proceeds not only in a space of internal atomic states, but also in a momentum space. It is important for coherent manipulation of atomic motion to avoid the excitation of intermediate states for all atoms from an ensemble. In this Rapid Communication we investigate the situation where one part of the ensemble is transferred coherently, while the other part does experience excitation and subsequent spontaneous relaxation. In such a case spontaneous relaxation can transfer atoms to the velocity-selective dark state. This leads to compression of atomic momentum distribution and may be used for efficient deep cooling of atoms.

The simplest quantum system where population transfer by STIRAP can be realized is a three-level Λ atom [Fig. 1(a)] driven by two laser pulses of frequencies ω_1 and ω_2 and wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ on transitions $|1\rangle-|3\rangle$ and $|2\rangle-|3\rangle$, respectively. Let us consider for simplicity the case of laser pulses counterpropagating along the z axis, having the same wave numbers $\mathbf{k}_1=-\mathbf{k}_2=k\mathbf{e}_z$. This scheme can be created in a $J=1\leftrightarrow J'=1$ transition excited by two σ^+ and σ^- laser waves. The Hamiltonian for the system in our one-dimensional problem is

$$\hat{H}=\frac{\hat{p}^2}{2M}+\sum_{m=1,2,3}\varepsilon_m|m\rangle\langle m|-\frac{i\hbar}{2}\gamma|3\rangle\langle 3| -\hbar[g_1(t)e^{i\omega_1 t-ik\hat{z}}|1\rangle\langle 3| +g_2(t)e^{i\omega_2 t+ik\hat{z}}|2\rangle\langle 3|+\text{H.c.}], \tag{1}$$

where ε_m are the energies of the eigenstates $|m\rangle$ of the unperturbed atom, and $g_m(t)$ are the Rabi frequencies for transitions $|m\rangle-|3\rangle$ $(m=1,2)$. The Hamiltonian includes kinetic-energy operator $\hat{p}^2/2M$ and a non-Hermitian term $-i\hbar\gamma/2$ accounting for spontaneous relaxation of the intermediate state $|3\rangle$ [2]. In the interaction picture and with rotating-wave approximation the Hamiltonian is transformed to the following form:

$$\widetilde{H}=\hbar\int dp\Bigg\{-(\Delta+\omega_R+i\gamma/2)|3,p\rangle\langle 3,p| +\sum_{m=1,2}[\pm\delta|m,p\mp\hbar k\rangle\langle m,p\mp\hbar k| -g_m(t)(|m,p\mp\hbar k\rangle\langle 3,p|+\text{H.c.})]\Bigg\}, \tag{2}$$

where

$$\delta=\frac{(\Delta_1-\Delta_2)}{2}-\frac{kp}{M} \tag{3}$$

is the two-photon or Raman detuning. In Eqs. (2) and (3) the upper (lower) sign corresponds to $m=1(2)$; $\Delta_m=\omega_m-(\varepsilon_3-\varepsilon_m)/\hbar$ are the one-photon detunings, $\Delta=(\Delta_1+\Delta_2)/2$ is the common detuning, and $\omega_R=\hbar k^2/2M$ is the recoil frequency. When deriving Eq. (2), we used the equality $\exp(\pm ik\hat{z})=\int_{-\infty}^{+\infty}dp|p\rangle\langle p\mp\hbar k|$, where p is the atomic momentum projection along the z axis. The Hamiltonian $\widetilde{H}$ couples only states within the family $|1,p-\hbar k\rangle$, $|2,p+\hbar k\rangle,|3,p\rangle$, and is therefore block-diagonal in the momentum basis. Under the two-photon resonance condition $\delta=0$ the atomic Hamiltonian has an adiabatic dressed-state

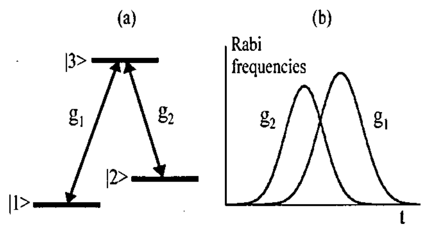

FIG. 1. (a) Three-level Λ system and (b) pulse sequence required for adiabatic population transfer between two ground states.

eigenvalue $E_0=0$. This means that the associated eigenvector is the so-called noncoupled or ''dark'' state $|\mathrm{NC}\rangle$. This state is given by

$$|\mathrm{NC}\rangle_0=(g_2/g)|1,p-\hbar k\rangle-(g_1/g)|2,p+\hbar k\rangle, \tag{4}$$

where $g=\sqrt{g_1^2+g_2^2}$. If the laser waves are arranged in such a way that $[g_1(t)/g_2(t)]_{t\to-\infty}\to 0$ and $[g_2(t)/g_1(t)]_{t\to+\infty}\to 0$, i.e., the $g_2(t)$ pulse precedes the $g_1(t)$ pulse [as shown in Fig.1(b)], then the atomic population, being initially (at $t=-\infty$) in the state $|1\rangle$, will adiabatically follow into $|2\rangle$ while remaining in the dark state. The adiabaticity condition is derived from the requirement that the coupling of the $|\mathrm{NC}\rangle_0$ state to the other two dressed states of the system should be small throughout the process of transfer. This results in the following conditions:

$$g_0T \gg 1 \quad \text{for } g_0 \gg \gamma, \tag{5}$$

$$g_0^2T/\gamma \gg 1 \quad \text{for } g_0 \ll \gamma, \tag{6}$$

where T is the characteristic time of the process and g_0 is the peak Rabi frequency of the pulses [6,7]. It is remarkable that the adiabatic transfer does not involve excited state $|3\rangle$ if the inequalities (5) or (6) are well satisfied.

Condition $\delta=(\Delta_1-\Delta_2)/2-kp/M=0$ implies that adiabatic population transfer (APT) is velocity selective. There is no zero eigenvalue of the Hamiltonian for atoms with velocities shifted from the two-photon resonance. However, for sufficiently small Raman detunings $\delta \ll g_0$, one finds the eigenvalue (for $\Delta+\omega_R=0$)

$$E_\delta \approx -\delta\frac{(g_1^2-g_2^2)}{g^2}-i\delta^2\frac{4\gamma g_1^2g_2^2}{g^6}, \tag{7}$$

which becomes exactly zero at $\delta=0$ and corresponds to the nearly dark state $|\mathrm{NC}\rangle_\delta$:

$$|\mathrm{NC}\rangle_\delta=\frac{g_2}{g}|1,p-\hbar k\rangle-\frac{g_1}{g}\left(1-i\frac{4\gamma\delta}{g^2}\right)|2,p+\hbar k\rangle$$
$$-\delta\frac{4g_1g_2}{g^3}\left(1-i\frac{\delta(g_1^2-g_2^2)}{\gamma g^2}\right)|3,p\rangle. \tag{8}$$

Solution of the Schrödinger equation gives that both the admixture of the excited state in the nearly dark state $|\mathrm{NC}\rangle_\delta$ and the nonadiabatic coupling of $|\mathrm{NC}\rangle_\delta$ to the other two dressed states are small at

$$\delta \ll g_0 \text{ and } g_0T \gg 1 \quad \text{for } g_0 \gg \gamma, \tag{9}$$

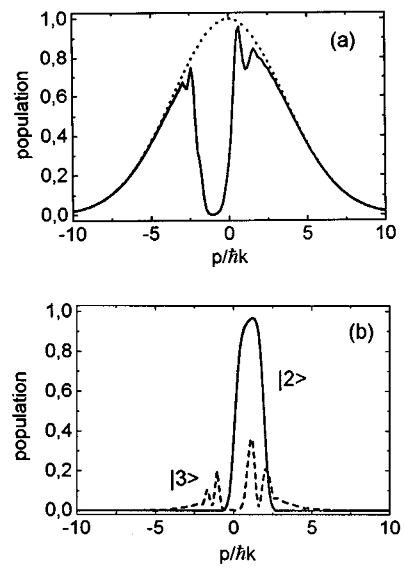

FIG. 2. Coherent population transfer of Λ atoms with pulse sequence from Fig. 1(b). Both light pulses have the same Gaussian shape with width $T=20$, amplitude $g_0=1.0$; time delay between the pulses $\tau=T=20$ and recoil frequency $\omega_R=0.134$ (in arbitrary units). Spontaneous emission is absent: $\gamma=0$. (a) Population of $|1\rangle$. The dotted line shows initial distribution; solid line, distribution after the transfer. (b) Momentum distribution of populations after the transfer. Dashed line, population of $|3\rangle$; solid line, population of $|2\rangle$.

$$\delta \ll g_0^2/\gamma \text{ and } g_0^2T/\gamma \gg 1 \quad \text{for } g_0 \ll \gamma, \tag{10}$$

Therefore, only atoms with z projection of momentum satisfying the conditions (9) or (10) follow adiabatically from $|1,p-\hbar k\rangle$ to $|2,p+\hbar k\rangle$. If the width of an initial momentum distribution of atoms is within the range of these conditions, then the whole atomic ensemble is transferred without the significant excitation of the intermediate state $|3\rangle$, which introduces incoherence in the process via spontaneous relaxation. Therefore, in atom optics and interferometry applications of APT [2–5] one should use precooled atomic ensembles in order to fulfill the conditions (9) or (10) where δ is the width of initial atomic distribution. Otherwise, for broad initial distribution, only a part of the atomic momentum distribution is transferred coherently. Figure 2 shows the process of velocity-selective APT in the latter case. These model calculations were performed for the Λ system, with spontaneous relaxation being switched off. As can be seen, the narrow part of the initial distribution centered at $p_0/M=(\Delta_1-\Delta_2)/2k$ is cut and transferred to $|2\rangle$ for small g_0. Atoms with momenta out of this narrow range undergo Rabi oscillations between the atomic states. In this situation, the intermediate state $|3\rangle$ is substantially populated for ''bad'' velocities throughout the interaction [Fig. 2(b)]. In a real process (with a relaxation of $|3\rangle$) this population decays to the ground states. In particular, there is a probability for atoms to be pumped into the nearly dark state $|\mathrm{NC}\rangle_\delta$. The decay rate of $|\mathrm{NC}\rangle_\delta$ changes during the process, but it is always very small for small momentum, and it vanishes at $p_0/M=(\Delta_1-\Delta_2)/2k$ [the decay rate is determined by the imaginary part of the eigenenergy, see Eq. (7)]. Hence, the

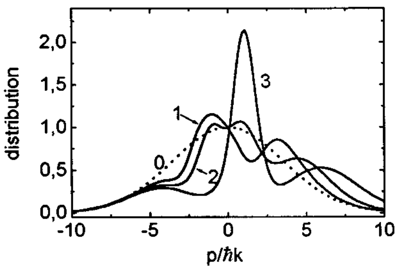

FIG. 3. Velocity-selective adiabatic population transfer accompanied by an accumulation of atoms in the dark state. Parameters of atomic transition correspond to Lyman-α $F=1$ to $F'=1$ transition in hydrogen. Parameters of the light pulses are the same as in Fig. 2 (in units of relaxation rate γ). Numbers at curves correspond to: 0, initial distribution; 1, distribution at time of maximum of $g_2(t)$ pulse; 2, at time when $g_1(t)=g_2(t)$; 3, after the transfer process.

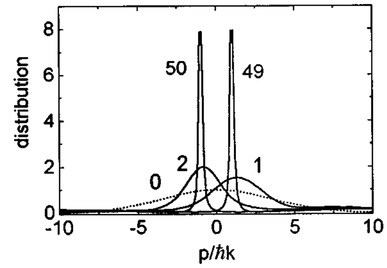

FIG. 4. Laser cooling by multiple repetition of velocity-selective APT's (for details see text). Parameters are the same as in Fig. 3. Rabi frequency g_0 is decreased during the sequence from $g_0=4.0$ to $g_0=2.0$. Momentum distribution is shown at times after the number of transfers indicated at the curves.

atoms are accumulated in the velocity-selective dark state during the transfer (see Fig. 3). This kind of atomic momentum distribution narrowing was recently observed with Rb atoms [8]. Note that the process takes place even if both states $|1\rangle$ and $|2\rangle$ are initially populated. In this case, all the atoms in $|2\rangle$ are not in the dark state at the initial stage of transfer. Therefore, they are rapidly pumped by the $g_2(t)$ pulse into the $|1\rangle$ state, and then velocity-selective APT proceeds as described above.

Thus, the presented phenomenon is a counterpart of velocity-selective coherent population trapping (VSCPT) [9]. It has all the features necessary for laser cooling below the photon recoil energy: (i) vanishing absorption of light for atoms in the narrow $\Delta p \ll \hbar k$ momentum domain; and (ii) momentum diffusion of atoms, bringing them into this domain from other regions of momenta. One may therefore consider the velocity-selective APT as a basis for subrecoil laser cooling with pulsed radiation. Multiple repetition of such pairs of time-delayed pulses will lead to continuous accumulation of atoms in the velocity-selective state $|\mathrm{NC}\rangle_\delta$, i.e., to the cooling of an atomic ensemble. In order to maintain the distribution in the zero-velocity region, one should alternate the sequence of laser pulses $g_1(t)$ and $g_2(t)$ from pair to pair (in contrast to the scheme in [5] where the directions of both laser beams are reversed after each transfer, giving a large momentum transfer). A high efficiency of cooling is achieved by applying the pulses, the intensity of which decreases successively from pair to pair, so that each laser pair transfers coherently the largest part of the atomic momentum distribution and only the wings are pumped into $|\mathrm{NC}\rangle$. However, one cannot decrease the laser intensity to arbitrarily low values. The parameter $g_0^2 T/\gamma$ should always be kept at $\gg 1$ for nonadiabatic transitions from $|\mathrm{NC}\rangle$ to be negligible [see (6)]. Figure 4 shows an example of such a cooling of hydrogen atoms on the Lyman-α $F=1-F'=1$ transition [10]. We take an initial distribution with the width $\delta p = 5\hbar k$ corresponding to a temperature of 16 mK, which can be achieved by the Doppler cooling [11]. One can start with broader distribution as well, but then larger peak Rabi frequencies are necessary to achieve a reasonable efficiency [cf. (9) and (10)]. We have found that H atoms are cooled to the halfwidth of the momentum distribution $\Delta p \simeq 0.2\hbar k$ with approximately 50% of the atoms from the initial distribution accumulated in a peak after the application of 50 laser pulse pairs. The extension of this cooling scheme to two and three dimensions should be possible either by adding additional laser beams along the x and y axes [12] or by alternating the direction of the one-dimensional process. One should note the limitation on velocity-selective APT and on the minimum width of momentum distribution attainable in this cooling method, which is imposed by laser fluctuations [6]. This factor, however, can be substantially reduced by the use of correlated laser sources.

So far our discussion has concentrated on APT only in the Λ system in the $J=1 \leftrightarrow J'=1$ atomic transition. However, it was both theoretically and experimentally shown that APT is accomplished in $J=J \leftrightarrow J'=J$ or $J-1$, $J>1$ transitions as well [2,4,5,13,14]. The dark states in such transitions are intrinsically leaky, since the ground-state magnetic sublevels comprising the dark state have different kinetic energies, due to their different momenta. Nevertheless, the difference in kinetic energies can be compensated for by the a.c. Stark shift (light shift) from a far-off-resonance laser, which suppresses the leakage [14]. We note that velocity-selective APT can also be realized in the three-level cascade system with a long-lived uppermost excited state. This may be especially useful for subrecoil cooling of Rydberg atoms [15] and atoms with a nondegenerate ground state like alkaline-earth-metal elements.

The mechanism proposed here has to be compared to other schemes of subrecoil cooling with pulsed laser radiation. One can consider, for example, a scheme where the Λ system is excited not by time-delayed but by coincident (or matched) laser pulses, which is essentially the usual VSCPT method [9]. In that method, each laser pulse pair generates a velocity-selective dark state that is a superposition of $|1,p_0-\hbar k\rangle$ and $|2,p_0+\hbar k\rangle$ with a definite phase relation between the probability amplitudes. In general, the proper phase relation cannot be maintained between two independent pulse pairs. Therefore, the next pulse pair destroys the dark state created by the previous one. Atoms even with zero velocity are excited to $|3\rangle$ and have a probability of leaving the zero-velocity range. We have checked by numerical calculations that this scheme leads to a very poor efficiency in cooling. In contrast, in velocity-selective APT the dark state always ends up in one of the states $|1,p_0-\hbar k\rangle$ or $|2,p_0+\hbar k\rangle$ after the interaction with each laser pulse pair, and the exact phase relation is not important for the next pair. Another technique using pulsed laser radiation that allows one to reach subrecoil temperatures is Raman cooling [16]. This method uses sequences of stimulated Raman π pulses and optical pumping pulses. It is therefore very sensitive to the exact value of the pulse shape and area. Since the process of velocity-selective APT is adiabatic, it is, within the validity of the adiabatic condition [Eqs. (9) and (10)], strongly resistant to changes of laser parameters, interaction time, etc. On the other hand, while velocity-selective APT requires a specific type of atomic transition, Raman cooling can be applied to any three-level system with two long-lived states. Finally note that the possibility of subrecoil cooling with laser pulses was pointed out in Ref. [17] for narrow atomic transitions.

In conclusion, we have shown that the adiabatic population transfer by delayed counterpropagating laser pulses in multilevel systems is a velocity-selective process. Under certain conditions only a part of atoms is transferred coherently while following the dark state. The remaining atoms are driven to the excited state and have the possibility of being pumped into this velocity-selective dark state. The phenomenon is considered as a basis for subrecoil cooling with laser pulses. This may be attractive for the cooling of atoms having optical transitions from the ground state in such spectral ranges where the sources of cw laser radiation have not been available until now (e.g., hydrogen).

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