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Subrecoil cooling of Rydberg atoms

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Two mechanisms for production of an ensemble of atoms in a highly excited state with very narrow distribution of the atomic center-of-mass momentum (cold Rydberg atoms) are proposed. The mechanisms are based on velocity-selective π pulses and coherent population trapping in three-level cascade systems with the uppermost state being the Rydberg one. Both methods permit one to reach subrecoil temperatures of the Rydberg atoms.

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I. INTRODUCTION

Atoms in highly excited states (Rydberg atoms) have been under active study since the beginning of the century. Their unique behavior in different physical situations enables one to study a wide variety of physical phenomena as well as to work out new technical applications [1–3]. In the past few years some interest has been shown in the mechanical action of light on Rydberg atoms. It was realized, for example, that a sufficiently slow beam of atoms may be trapped or reflected by a microwave field in the micromaser configuration [4,5]. Another example is scattering of Rydberg atoms by standing light [6] or microwave [7] waves. Manipulation of the center-of-mass motion of Rydberg atoms in a well controlled manner offers interesting perspectives for applications. Laser cooled Rydberg atoms, i.e., those ones with a narrow distribution of the momentum of the center of mass, seem to be attractive in such fields of investigation as laser spectroscopy of the highly excited states, physics of the Rydberg atom collisions, cavity quantum electrodynamics including quantum nondemolition measurements and single-atom maser operation, and investigations of a quantum manifestation of quasiclassical chaos. Once atoms are cooled to very low temperatures, their motion can be significantly altered by the relatively feeble forces. Thus, cold Rydberg atoms show a very high sensitivity of both internal and translational motion to different external fields. This suggests using them for a detection of very weak interactions (for example, slow Rydberg atoms were proposed to be used for measurement of vacuum cavity forces [8]). It was recently shown that cold Rydberg atoms can be considered also as realizable analogs of Chern-Simons theory [9]. Narrow distribution of the atomic center-of-mass momentum implies a large spatial extent of the atomic wave packet due to the uncertainty principle. Therefore an ensemble of low-temperature Rydberg atoms is an interesting object for observing the quantum statistical effects, e.g., phase transitions.

Moreover, cold Rydberg atoms are of fundamental interest. They combine a quasiclassical dynamics of the motion of the excited electron with full quantum-mechanical dynamics of the atomic center-of-mass motion. Thus, we have a very strange composite quantum object with macroscopic size: the wave packet of the electron is well localized in space, while the coherence length of the whole atomic wave packet extends over a large volume in space.

We propose two schemes permitting us to obtain Rydberg atoms with very low translational temperature corresponding to a velocity distribution spread less than 1 cm/sec, hence to a micrometer-sized spatial wave packet width. The methods are based on the cw laser excitation of Rydberg states via a three-level cascade (Fig. 1).

A three-level quantum system is the simplest one where effects of interference between transition channels manifest themselves. The effects influence dramatically the mechanical action of laser light on atoms and make possible the development of very effective methods for the manipulation of the atoms. The velocity selection technique (VST) [10] and laser cooling of atoms by velocity-selective coherent population trapping (VSCPT)

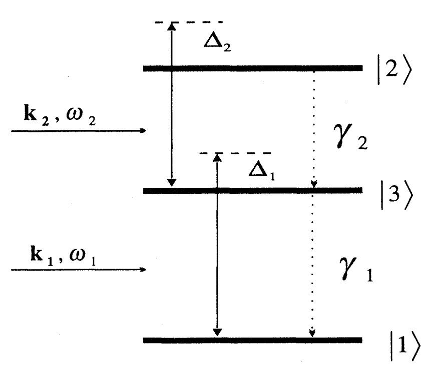

FIG. 1. Three-level cascade scheme of interaction of an atom with two light waves with the wave vectors $\vec{k}_m$ and frequencies ω_m. Detuning and spontaneous relaxation rate for the $|m\rangle - |3\rangle$ transitions are denoted by Δ_m and γ_m, respectively (m = 1, 2).

[11] are among the methods. They allow one to obtain atomic ensembles with translational temperatures in the nanoKelvin range. Usually these two techniques are applied to laser-atom interaction schemes where all excited states are used as the intermediate ones and cold atoms are accumulated in one of the substates of the ground atomic state. The simplest and most popular example of such a scheme is a three-level Λ system. However, the above methods can be exploited by the use of the cascade systems as well [12]. This fact permits one to produce very cold Rydberg atoms.

II. VELOCITY SELECTION TECHNIQUE

The first method, the velocity selection technique, is performed in coherent scattering of atoms by traveling light waves when one can neglect spontaneous relaxation. In this regime, Hamiltonian evolution of atomic population reveals oscillations between the atomic states (Rabi oscillations). Under specific conditions the population can be completely inverted between the states by means of a π pulse [13]. For cascade systems, the atoms are initially in the state $|1\rangle$ (Fig. 1). Having interacted with two laser fields with parameters corresponding to a π pulse between the states $|1\rangle$ and $|2\rangle$ via an intermediate state $|3\rangle$, the atoms get into the state $|2\rangle$ with stimulated transitions. For the moving atoms the process of laser excitation is selective in velocity due to the Doppler effect. Therefore, only atoms with velocities close to a resonance velocity reach the state $|2\rangle$. The degree of the selectivity is determined by the laser intensities and by the duration of the laser-atom interaction [14]. Thus, by a proper choice of laser parameters one can obtain an ensemble of atoms in the desired quantum state with desired velocity distribution. In the case of three-level cascade systems this velocity-selective coherent excitation technique can be achieved when spontaneous relaxation from both the intermediate $|3\rangle$ and the uppermost $|2\rangle$ states is negligible during the process. The requirement is satisfied naturally for the $|2\rangle$ state being the highly excited Rydberg state. In order to suppress spontaneous emission from the $|3\rangle$ state, the detunings Δ_1 and Δ_2 must be large. With the conditions

$$|\Delta_1|, |\Delta_2| \gg |g_1|, |g_2|, \gamma_1, \gamma_2,$$

where g_m is the Rabi frequency for the $|m\rangle - |3\rangle$ transition, the population n_2 of the Rydberg state evolves in time as [15], [14]

$$n_2(v,t) = n_1(v, t=0)\frac{4|g_1|^2|g_2|^2}{\Omega_1^2\varpi^2}\sin^2\left(\frac{\varpi t}{2}\right). \qquad (1)$$

Here we denote

$$\varpi^2 = \left(\frac{|g_1|^2}{\Omega_1} + \frac{|g_2|^2}{\Omega_2} - \delta\right)^2 + 4\delta\frac{|g_1|^2}{\Omega_1}$$

and

$$\Omega_1 = \Delta_1 + \tfrac{\hbar\vec{k}_1\vec{k}_2}{2M} - \vec{k}_1\vec{v},$$
$$\Omega_2 = -\Delta_2 + \tfrac{\hbar\vec{k}_1\vec{k}_2}{2M} + \vec{k}_2\vec{v},$$
$$\delta = \Omega_1 - \Omega_2.$$

It is seen from Eq. (1) that the Rydberg $|2\rangle$ state is excited most effectively for velocity $\vec{v}_0$ satisfying the condition of the Doppler-shifted two-photon resonance in the cascade scheme:

$$\Delta_1 - \vec{k}_1\cdot\vec{v}_0 = -\Delta_2 + \vec{k}_2\cdot\vec{v}_0. \qquad (2)$$

Note that for $|\vec{k}_1| \simeq |\vec{k}_2|$ the resonance condition is velocity selective for copropagating light waves, in contrast to the Λ system where atoms are selected by means of a stimulated Raman transition with counterpropagating waves [10]. Maximum population in the Rydberg state is achieved at a time τ_n (time of the $n\pi$ pulse)

$$\tau_n \cong \pi\,(2n+1)\,\frac{\Delta}{g^2}, \qquad (3)$$

where $n = 0, \pm1, \pm2, ..., g^2 = |g_1|^2 + |g_2|^2$ and we set $\Delta_1 = -\Delta_2 (\equiv \Delta)$ for simplicity. Thus, a narrow peak in the velocity distribution of the Rydberg atoms is formed (Fig. 2). The width δv of the peak (the half-width on the half maximum) is determined by the interaction time: $(k_1 + k_2)\,\delta v \simeq 4/t$, and at time of the π pulse $t = \tau_1$ the velocity width is

$$\delta v \simeq 1.21\frac{g^2}{\Delta\,(k_1 + k_2)}. \qquad (4)$$

In order to obtain the width less than the recoil velocity $v_R = \frac{\hbar(k_1+k_2)}{M}$, one has to choose atom-laser interaction parameters so that $t > \omega_R^{-1}$ and $g^2/\Delta < \omega_R$ $[\omega_R = \hbar\,(k_1 + k_2)^2/2M$ is the recoil frequency]. When this takes place, it is necessary to avoid the influence of

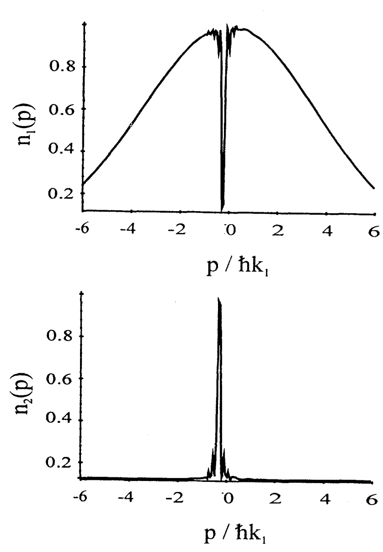

FIG. 2. Velocity selection of Rydberg atoms. The interaction parameters correspond to the first π pulse: $g_1 = g_2 (\equiv g)$, $\Delta_1 = \Delta_2 (\equiv \Delta)$, $g^2/\Delta = 0.2\omega_R$, $t = \pi\Delta/g^2$.

spontaneous relaxation. This implies that the interaction time t should be smaller than the lifetime γ_2^{-1} of the Rydberg state and laser detunings should be much larger than the spontaneous relaxation rate of the intermediate state $\Delta \gg \gamma_1$. For an example of ^{23}Na atoms excited to the $40\,^2S_{1/2}$ state via the $3\,^2S_{1/2} - 3\,^2P_{1/2(3/2)} - 40\,^2S_{1/2}$ cascade, the width 1 cm/sec of the velocity distribution of selected Rydberg atoms is achieved in 10^{-5} sec $(\approx 10^4 \gamma_1^{-1})$ for the Rabi frequencies $g \cong 2\gamma_1$ and detunings $\Delta \cong 10^3 \gamma_1$, while the lifetime of the excited states are $\tau_1 = 16 \times 10^{-9}$ sec and $\tau_2 \simeq 6 \times 10^{-5}$ sec.

III. LASER COOLING BY VSCPT

The technique discussed above is just a selection in the velocity space. It does not increase the number of "cold" atoms as the laser cooling methods do. Therefore, the narrower the velocity distribution width, the smaller the number of the selected "cold" Rydberg atoms. This can be favorable in some applications, for instance, in experiments with single atoms. But it is more often necessary to cool as much atoms from the initial distribution as possible.

We propose here a method based on the velocity-selective coherent population trapping (VSCPT) phenomenon [11], allowing cooling of atoms to translational energies far below the recoil energy $E_R = \hbar\omega_R$. In this method, the cooling is due to optical pumping of atoms in a superposition of the atomic states which is not excited by the laser radiation to the rest of the quantum system. Accumulation of atoms in this "trap" state, if it is velocity selective, leads to a strong narrowing of the atomic momentum distribution. Recently, we have found that atoms can be cooled by VSCPT in the three-level cascade systems [12]. The relevant trap state is the following superposition:

$$|NC\rangle = \frac{g_2}{\sqrt{|g_1|^2 + |g_2|^2}} \left|1, \vec{p}_0 - \hbar\vec{k}_1\right\rangle - \frac{g_1}{\sqrt{|g_1|^2 + |g_2|^2}} \left|2, \vec{p}_0 + \hbar\vec{k}_2\right\rangle \quad (5)$$

with $\vec{p}_0 = M\vec{v}_0$, where $\vec{v}_0$ satisfies the resonance condition Eq. (2).

The peculiarity of the VSCPT in cascade systems is that the $|NC\rangle$ is the superposition of the ground state $|1\rangle$ and the highest excited state $|2\rangle$ which decays spontaneously. Therefore, the $|NC\rangle$ state is not absolutely stable. Nevertheless, coherent population trapping (and subrecoil cooling by VSCPT) is realized if the rate at which the trap state fills is much higher than its relaxation rate. This requirement results in specific conditions on the laser field parameters and relaxation constants [12]. In particular, the relaxation rate of the $|2\rangle$ state should be less than that of the $|3\rangle$ state:

$$\gamma_2 \ll \gamma_1. \quad (6)$$

This condition is well satisfied for the $|2\rangle$ state being the highly excited Rydberg state.

In order to obtain the main fraction of the cooled atoms in the Rydberg state, one has to adjust the laser intensities so that $g_1 \gg g_2$. In this case for atoms trapped in $|NC\rangle$, the probability for the Rydberg state is $(g_1/g_2)^2$ ($\gg 1$) times larger than the probability for the ground state [see Eq. (5)].

We have calculated the VSCPT cooling process for the example of the $3\,^2S_{1/2} - 3\,^2P_{1/2(3/2)} - 40\,^2S_{1/2}$ cascade in ^{23}Na (Figs. 3 and 4). The formation of two narrow peaks in the momentum distribution corresponding to the accumulation of atoms in the trap state Eq. (5) is clearly seen in Fig. 3(a). The peak centered at $p = -\hbar k_1$ belongs to atoms in the ground $|1\rangle$ state and the one at $p = +\hbar k_2$ belongs to the Rydberg state atoms. The formation of the peak in the Rydberg state is shown in Fig. 3(b). Time dependence of the parameters of the Rydberg atoms distribution is shown in Fig. 4. We note two important features of the cooling process. The first one is the decrease of the peak width with time. Simple arguments analogous to those presented in [16] show that the width follows the dependence $\delta p \sim t^{-1/2}$. Our calculations confirm this dependence for the initial stage of the evolution quite well [see Fig. 4(a)]. One can produce in such a way an ensemble of very cold Rydberg atoms with temperatures below the recoil limit $T_R = E_R/k_B$. However, there is a limiting temperature attainable at VSCPT cooling in cascade systems, in contrast to VSCPT schemes with absolutely stable trap state. Spontaneous relaxation of the Rydberg state induces an instability of the trap state. Therefore atoms have a possibility to escape from the trap state. This process equalizes the pumping into the trap state for long interaction times. This leads, in the asymptotical limit, to the width independent of time. Exact analytical solution to the density matrix equations gives the following asymptotical value for δp [12]:

$$\delta p = \hbar\,(k_1 + k_2)\,\frac{2g_1^2}{g_2\omega_R}\sqrt{\frac{\gamma_2}{\gamma_1}} \quad (7)$$

[note that Eq. (7) is calculated for the case of our present interest: $g_1^2 \gg g_2^2 \gg \gamma_2\gamma_1$].

Unfortunately, efficiency of such a cooling method is rather poor [see Figs. 4(b) and 4(c)]. This is due to the light pressure force on the cascade atoms excited by traveling light waves. One can easily calculate a quantum-mechanical analog of the force $\vec{F}(\vec{p},t)$ determined as follows:

$$\vec{F}(\vec{p},t) = \left\langle \frac{\partial\vec{p}}{\partial t} \right\rangle \equiv Tr\left(\vec{p}\frac{\partial\hat{\rho}(\vec{p},t)}{\partial t}\right) \quad (8)$$

with the help of the "closed family approach" [16,17]:

$$\vec{F}(\vec{p},t) = \hbar\vec{k}_1\gamma_1\rho_{33}(\vec{p},t) + \hbar\vec{k}_2\gamma_2\rho_{22}(\vec{p},t). \quad (9)$$

In Eqs. (8) and (9) the operator $\hat{\rho}(\vec{p},t)$ denotes an atomic density matrix and $\rho_{mm}(\vec{p},t)$ is the population of the $|m\rangle$ state. The force is always of constant sign for any velocity of atoms. Therefore, the same force which decelerates atoms with negative velocities accelerates atoms with positive ones. This process happening on the initial stage of the evolution is clearly seen in Fig. 3(a) as a shift of a large part of the distribution towards high

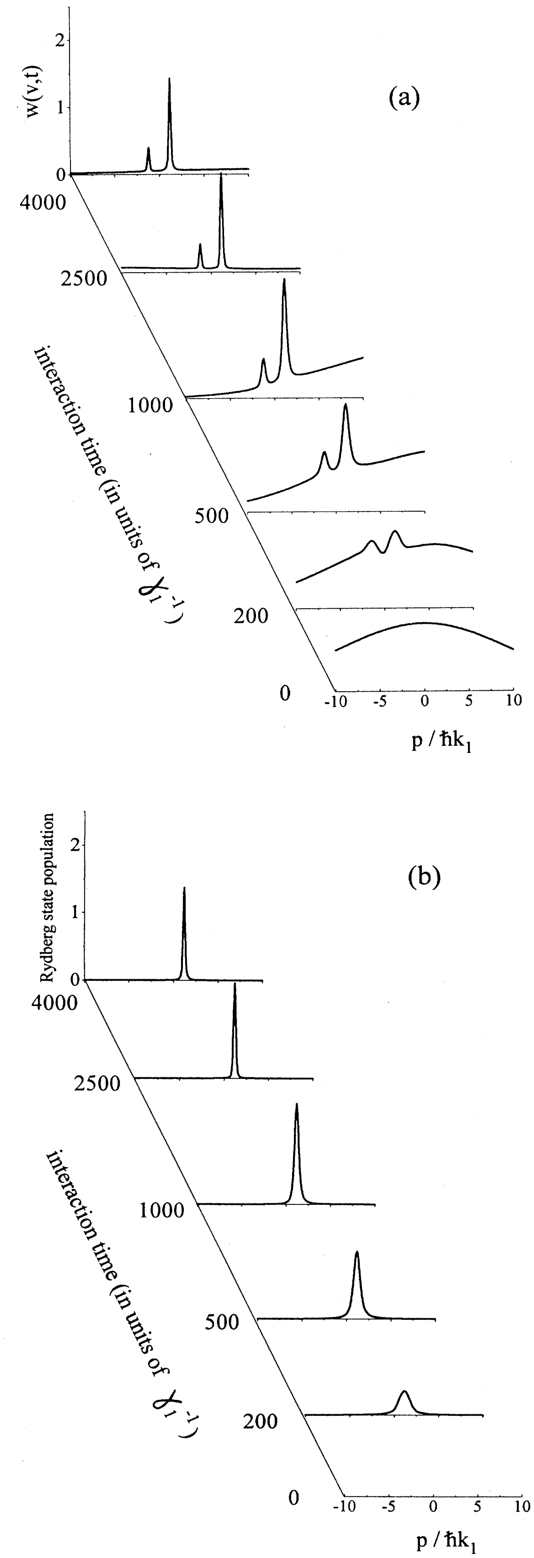

FIG. 3. VSCPT cooling in the $3\,^2S_{1/2} - 3\,^2P_{1/2(3/2)} - 40\,^2S_{1/2}$ cascade in ^{23}Na. (a) Temporal evolution of the whole atomic momentum distribution, (b) evolution of the population of the Rydberg state. Rabi frequencies: $g_1 = 0.1\gamma_1$, $g_2 = 0.05\gamma_1$; detunings: $\Delta_1 = -0.1\gamma_1$, $\Delta_2 = 0.1\gamma_1$.

positive velocities. For longer interaction times the force is small in the range of velocities close to the resonance value Eq. (2). Nevertheless, the instability of the trap state mentioned above induces slow but continuous decrease of the fraction of cooled atoms. Probably, one can choose parameters of the interaction (Rabi frequencies and detunings of the laser fields, interaction time) which improve cooling efficiency. We do not believe, however, that the efficiency can be improved substantially.

An evident solution to this problem is to find a laser configuration where light forces confine atoms in the momentum space. Such a force can be provided by standing light waves rather than traveling ones. In this case the VSCPT cooling coexists with a Sisyphus sub-Doppler cooling [18]. The role of Sisyphus cooling is twofold. First, it precools the initial momentum distribution to a few recoil momenta, and second, it confines the atoms in this momentum range. It can be shown that the trap

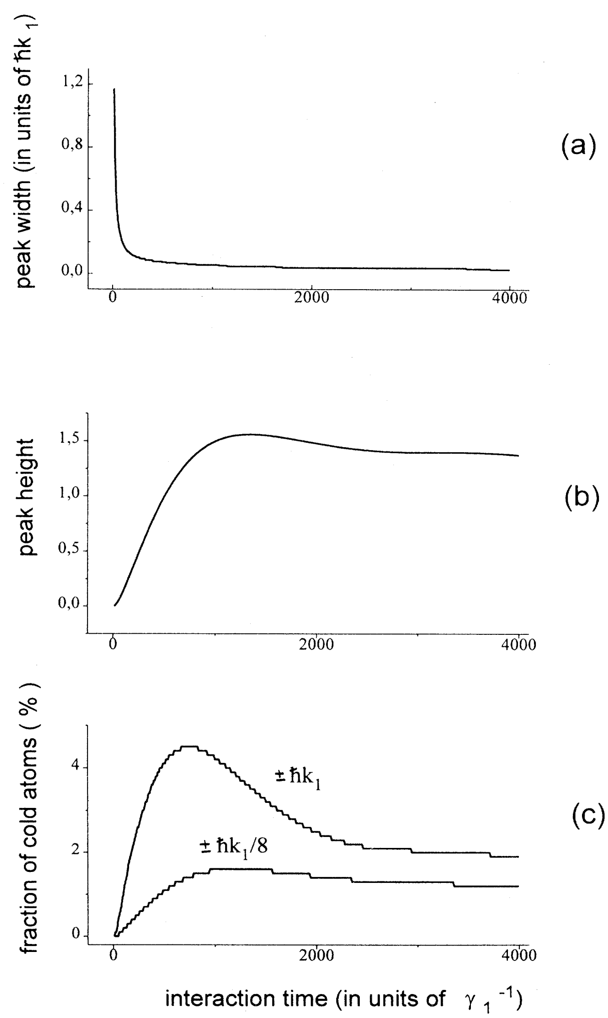

FIG. 4. Parameters of the cold Rydberg atoms peak in Fig. 3 as a function of time. (a) Peak width, (b) peak height (in arbitrary units), (c) fraction of cold Rydberg atoms with respect to the initial number of atoms; 1: in the momentum interval $\pm\hbar k_1$, 2: in the momentum interval $\pm\hbar k_1/8$.

state in the case of standing waves has the following form:

$$|NC\rangle = \frac{g_2}{\sqrt{|g_1|^2+|g_2|^2}}\left(e^{i\varphi}|1,-\hbar k_2\rangle + e^{-i\varphi}|1,+\hbar k_2\rangle\right) - \frac{g_1}{\sqrt{|g_1|^2+|g_2|^2}}\left(|2,-\hbar k_1\rangle + |2,+\hbar k_1\rangle\right) \quad (10)$$

with the condition for laser detunings:

$$\Delta_1 - \frac{\hbar k_1^2}{2M} + \frac{\hbar k_2^2}{2M} = -\left(\Delta_2 - \frac{\hbar k_1^2}{2M} + \frac{\hbar k_2^2}{2M}\right).$$

Here φ is the spatial phase shift between the standing waves.

Calculations of the cooling dynamics are in progress. Qualitative considerations show no principal differences compared to the case of Sisyphus-assisted VSCPT cooling in Λ systems [19]. The most important distinction concerns the asymptotical behavior of atomic momentum distribution. We expect a limit for the temperature due to the trap state instability discussed above.

IV. DISCUSSION

In this paper we have shown the possibility of producing an ensemble of atoms in a highly excited state with very narrow distibution of the atomic center-of-mass momentum (cold Rydberg atoms). The two proposed mechanisms are based on velocity-selective π pulses and on VSCPT cooling in three-level cascade systems with the uppermost state being the Rydberg one. Both mechanisms permit us to reach subrecoil temperatures of the Rydberg atoms. The lowest temperature attainable is limited by the radiative relaxation rate γ_2 of the Rydberg state for both methods. Therefore, the higher the target Rydberg state is, the colder the atoms that could be obtained. On the other hand, the frequency distance between the neighboring Rydberg states decreases as n^{-3} with the principal quantum number n. This fact imposes the upper limit on n for the methods presented, since they require the exact two-photon resonance condition. Therefore, the distance should be larger than both the laser bandwidth and the Doppler width of the initial atomic ensemble. One more source for limitation of the temperature is due to any process destroying the laser-induced coherence between the $|1\rangle$ and $|2\rangle$ states [12]. Besides the spontaneous relaxation contribution, these are basically laser fluctuations. The latter factor, however, can be substantially reduced by the use of correlated laser sources.

It is worthwhile to note that Zeeman degeneracy of the participating states does not considerably influence the cooling methods presented here, if atomic states and laser polarizations are properly chosen. One can use atomic states with angular momenta $J_1 = J$, $J_3 = J+1$, $J_2 = J$, as the states $|1\rangle$, $|3\rangle$, $|2\rangle$, respectively, excited by two (with frequencies ω_1 and ω_2) linear or circular polarized laser waves. In this case some similar three-level cascade systems with $m_J = 0, \pm 1, ..., \pm |J|$ are formed. The systems are closed with respect to the stimulated transitions and are coupled to each other by spontaneous decay transitions only. For weak enough external magnetic field, the two-photon resonance condition [Eq. (2)] is satisfied for the same velocity v_0 in each cascade. Therefore, the m_J Rydberg states population transferred by the velocity-selective π pulse in VST or pumped into $|NC\rangle$ in VSCPT cooling is summing up at v_0 for each system. For the example of sodium atoms, the states $|1\rangle$, $|3\rangle$, $|2\rangle$ are the hyperfine sublevels $3\,^2S_{1/2}$, $F = 1$, $3\,^2P_{1/2}$, $F = 2$, and $40\,^2S_{1/2}$, $F = 1$, respectively. The problem of the optical pumping into the $3\,^2S_{1/2}$, $F = 2$ ground state can be solved by applying an additional repumping laser field to the $3\,^2S_{1/2}$, $F = 2$ to $3\,^2P_{3/2}$, $F = 3$ transition.

We have considered here the methods for production of Rydberg atoms with narrow momentum distribution only in one dimension. At the same time, both π pulses and coherent population trapping are realized not only in three-level atomic systems but in N-level ($N > 3$) ones as well [20,13]. Therefore it should be possible to find atom-laser cascade configurations where these processes are velocity selective in two and three dimensions.

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