# Laser cooling of Rydberg atoms in bichromatic standing waves

J. Guo and E. Arimondo*
*JILA, University of Colorado and National Institute of Standards and Technology, Boulder, Colorado 80309-0440*
(Received 13 November 1995)

We present a calculation of cooling of Rydberg atoms in bichromatic standing light waves. The atoms in our model are assumed to have a cascade three-level transition scheme, with the second excited level being the Rydberg state. The cooling fields, which also excite the atoms to the Rydberg state, consist of two standing waves whose frequencies are tuned near the two transition frequencies of the cascade atomic system. It is shown that the mechanisms of Sisyphus cooling and velocity-selective coherent population trapping can coexist in such a configuration, and under certain conditions the majority of the laser-cooled atoms are in the Rydberg state.

PACS number(s): 32.80.Pj, 42.65.−k

Laser cooling of atoms has been an active field of research in the last few years. Among most of the existing laser cooling schemes, the goal has been to prepare a sample of cold atoms in their ground electronic states. In certain situations, it may be desirable to have a laser-cooled sample of atoms in a selected excited electronic state. In this Rapid Communication we explore the possibility of preparing cold Rydberg atoms with a combination of Sisyphus cooling [1] and velocity-selective coherent population trapping (VSCPT) [2], two cooling mechanisms that have been studied extensively in the past.

There exist many reasons for preparing laser-cooled atoms in a highly excited electronic state. Rydberg atom physics has been an active field of research in atomic physics for several decades [3]. The preparation of cold Rydberg atoms should be of interest to subject fields such as Rydberg atom collisions, cavity quantum electrodynamics, precision spectroscopy of Rydberg states, and nonlinear dynamics and chaos. Moreover, laser-cooled Rydberg atoms are unique objects in that although the motion of their outer electrons is well localized, their center-of-mass (c.m.) motion is quantum mechanical with a coherence length on the order of an optical wavelength. One expects that new experiments can be performed with cold Rydberg atoms, which may reveal interesting physics and lead to potential applications in the future.

The atoms in our model are assumed to have a nondegenerate cascade three-level structure as shown in Fig. 1, with the ground state $|g\rangle$, the first excited state $|e\rangle$, and the Rydberg state $|r\rangle$. This level scheme may correspond to, for example, the Na $3\,{}^2S_{1/2}\rightarrow 3\,{}^2P_{3/2}\rightarrow 45\,{}^2D_{5/2}$ transition. The Bohr frequencies of the two cascade transitions are denoted as $\omega_{10}$ and $\omega_{20}$. The spontaneous decay rates of the excited and the Rydberg states are given by $\gamma_1$ and $\gamma_2$, respectively. We assume that $\gamma_1 \gg \gamma_2$, which is the case for most Rydberg systems. The cooling fields are a pair of standing waves, with their frequencies $\omega_1$ and $\omega_2$ tuned near $\omega_{10}$ and $\omega_{20}$, respectively. In the case where $\omega_{10}$ and $\omega_{20}$ are nearly identical, a rectified dipole force may occur for such a system [4]. Here we will be concerned with the cases when $|\omega_{10}-\omega_{20}|$ have the same order of magnitude as $\omega_{10}$ or $\omega_{20}$.

VSCPT cooling of atoms with the same cascade three-level structure in bichromatic traveling waves has been considered recently in Ref. [5], where it was shown that in order to obtain a velocity-selective dark state, the two laser fields must propagate in the same direction. In this case the cooling effects are severely restricted by the presence of an unidirectional light pressure force due to the copropagating traveling waves, and the atomic population captured in the dark state is quite small [5]. The advantages of the present standing-wave configuration over the traveling-wave scheme are twofold: First, the replacement of the copropagating traveling waves with standing waves eliminates the unidirectional light pressure force, and cooling can occur over a much longer time scale; second, in addition to VSCPT cooling, there also exists Sisyphus cooling in the standing-wave case, and the combination of both mechanisms makes this configuration more efficient and useful for cooling of Rydberg atoms. The coexistence of VSCPT and Sisyphus cooling in this cascade scheme is similar to the case of a three-level $\Lambda$ transition previously investigated [6,7].

The total Hamiltonian of the system, including the c.m. motion of the atoms, can be written in a frame rotating at the laser frequencies as

$$H = H_0 + V, \tag{1}$$

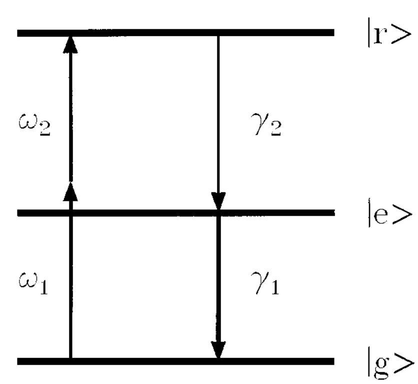


FIG. 1. The cascade level scheme and the incident fields.

*Permanent address: Dipartimento di Fisica, Università di Pisa, I-56126 Pisa, Italy.

 

where the atomic Hamiltonian $H_0$ is given by

$$H_0=\frac{p^2}{2M}(|g\rangle\langle g|+|e\rangle\langle e|+|r\rangle\langle r|)-\hbar\Delta_1|e\rangle\langle e|-\hbar(\Delta_1+\Delta_2)|r\rangle\langle r| \quad (2)$$

and the interaction Hamiltonian $V$ is written as

$$V=\hbar\left[\frac{\Omega_1}{2}\cos(k_1z)|e\rangle\langle g|+\frac{\Omega_2}{2}\cos(k_2z+\phi)|r\rangle\langle e|+\text{H.c.}\right], \quad (3)$$

where $\phi$ is a relative phase difference between the two standing waves. The field detunings and Rabi frequencies in Eqs. (2) and (3) are defined as

$$\Delta_1=\omega_1-\omega_{10},\quad \Delta_2=\omega_2-\omega_{20},$$
$$\Omega_1=\frac{d_{eg}E_1}{\hbar},\quad \Omega_2=\frac{d_{re}E_2}{\hbar}. \quad (4)$$

The noncoupled state $|\text{NC}\rangle$, defined as $V|\text{NC}\rangle=0$, is given by

$$|\text{NC}\rangle=\frac{1}{\sqrt{\Omega_1^2+\Omega_2^2}}[\Omega_2\cos(k_2z+\phi)|g\rangle-\Omega_1\cos(k_1z)|r\rangle]. \quad (5)$$

For stable VSCPT cooling, $|\text{NC}\rangle$ must also be a dark state which is not coupled to any other atomic states by the atomic c.m. motion. This requirement can be satisfied if $|\text{NC}\rangle$ is an eigenstate of the total Hamiltonian $H$ given in Eq. (2), which leads to the following requirement on the field detuning $\Delta_1$ and $\Delta_2$:

$$\Delta_1-\omega_{k_1}=-\Delta_2-\omega_{k_2}\equiv\Delta, \quad (6)$$

where $\omega_{k_i}=\hbar k_i^2/2M$ $(i=1,2)$. In what follows we assume that this requirement on the detunings $\Delta_1$ and $\Delta_2$ is always satisfied. We next assume a weak-field limit defined by

$$\Omega_1,\Omega_2\ll\gamma_1,|\Delta|, \quad (7)$$

which is the optimal limit for Sisyphus cooling that, as shown below, can exist in this standing-wave configuration. In this weak-field limit one can adiabatically eliminate the intermediate excited state $|e\rangle$ since its population is negligible. The resulting effective Hamiltonian $H_{\text{eff}}$ for the atomic ground and Rydberg states can be written as

$$H_{\text{eff}}=\frac{p^2}{2M}(|g\rangle\langle g|+|r\rangle\langle r|)-\hbar(\Delta_1+\Delta_2)|r\rangle\langle r|+U(z), \quad (8)$$

where the light-induced potential $U(z)$ is given by [9]

$$U(z)=U_0\left[\cos^2(k_1z)|g\rangle\langle g|+\left(\frac{\Omega_2}{\Omega_1}\right)^2\cos^2(k_2z+\phi)|r\rangle\langle r|+\frac{\Omega_2}{\Omega_1}\cos(k_1z)\cos(k_2z+\phi)(|r\rangle\langle g|+|g\rangle\langle r|)\right], \quad (9)$$

with the potential depth $U_0$ given by

$$U_0=\frac{\hbar\Delta\Omega_1^2/2}{(\gamma_1/2)^2+\Delta^2}. \quad (10)$$

In deriving the effective Hamiltonian $H_{\text{eff}}$ we have neglected terms proportional to $\gamma_2/\gamma_1$ or $\omega_{k_i}/|\Delta|$ $(i=1,2)$ which are small. The density matrix equation for the atomic ground state and the Rydberg state are given by

$$\dot{\rho}=\frac{1}{i\hbar}[H_{\text{eff}},\rho]-\frac{1}{2}[A\rho+\rho A]+\gamma_1\int_{-k_1}^{k_1}N_1(p')e^{-ip'z}B^\dagger\rho Be^{ip'z}dp'+\gamma_2\int_{-(k_1+k_2)}^{k_1+k_2}N_2(p')e^{-ip'z}|g\rangle\langle r|\rho|r\rangle\langle g|e^{ip'z}dp', \quad (11)$$

where the operators $A$ and $B$ are given by

$$A=\frac{\gamma_1(\Omega_1/2)^2\cos^2(k_1z)}{(\gamma_1/2)^2+\Delta^2}|g\rangle\langle g|+\frac{\gamma_1(\Omega_2/2)^2\cos^2(k_2z+\phi)}{(\gamma_1/2)^2+\Delta^2}|r\rangle\langle r|+\frac{\gamma_1\Omega_1\Omega_2/4\cos(k_1z)\cos(k_2z+\phi)}{(\gamma_1/2)^2+\Delta^2}\times(|g\rangle\langle r|+|r\rangle\langle g|)+\gamma_2|r\rangle\langle r|,$$

$$B=[i\Omega_1/2\cos(k_1z)|g\rangle\langle g|+i\Omega_2/2\cos(k_2z+\phi)|r\rangle\langle g|]/(\gamma_1/2+i\Delta), \quad (12)$$

and the functions $N_1(p')$ and $N_2(p')$ denote the spontaneous emission kernels associated with the decay from $|e\rangle$ to $|g\rangle$ directly and from $|r\rangle$ to $|g\rangle$ indirectly via $|e\rangle$, respectively.

One can diagonalize the optical potential $U(z)$, and the resulting eigenvalues, $U_0(z)$ and $U_1(z)$, are usually called the adiabatic potentials. They are given by

$$U_0(z)=0,$$
$$U_1(z)=U_0\left(\cos^2(k_1z)+\frac{\Omega_2^2}{\Omega_1^2}\cos^2(k_2z+\phi)\right). \quad (13)$$

In the absence of the atomic c.m. motion, the eigenstate associated with $U_0(z)$ is the noncoupled state $|\text{NC}\rangle$, while that associated with $U_1(z)$ is given by

$$|C\rangle=\frac{1}{\sqrt{\Omega_1^2+\Omega_2^2}}[\Omega_1\cos(k_1z)|g\rangle+\Omega_2\cos(k_2z)|r\rangle]. \quad (14)$$

Similar to the three-level $\Lambda$ configuration [6,7], Sisyphus cooling of the atoms is provided through the transitions from the adiabatic potential $U_1(z)$ to $U_0(z)$ and from $U_0(z)$ back to $U_1(z)$, which are induced by optical pumping and nonadiabatic coupling, respectively. The potential $U_1(z)$ must be positive, or $\Delta > 0$, such that the atoms can lose their kinetic energy when they climb up the potential $U_1(z)$ before being optically pumped into the potential $U_0(z)$. In what follows we assume that $\phi = 0$ without loss of generality.

The light-induced potential $U(z)$ given in Eq. (9) is, in general, nonperiodic in space for an arbitrary ratio of $k_2/k_1$. To facilitate an analysis of the cooling process based on the Bloch state description of the atomic state [1,7,8], we choose laser wave vectors such that the ratio $k_2/k_1$ is a rational number 1.5, which is close to the actual value of $\omega_{20}/\omega_{10}$ for the above sodium cascade transition [10]. As a result, $U(z)$ becomes periodic in space with a period determined by the difference wave vector $k = k_2 - k_1$ (i.e., $k_1 = 2k, k_2 = 3k$). The Bloch states are denoted as $|n,q\rangle$, where $n$ is the energy band index and $q \in [-0.5k, 0.5k)$ is the Bloch index. The Bloch states can be expanded in terms of the free-particle states $|m\hbar k + \hbar q\rangle$ and the atomic internal states $|\epsilon\rangle$ ($\epsilon = g, r$) as

$$|n,q\rangle = \sum_{\epsilon=g,r} \sum_{m=-N}^{N} C_{n,q}(m,\epsilon)|m\hbar k + \hbar q\rangle|\epsilon\rangle, \quad (15)$$

where $N$ is a cutoff value for the momentum expansion. For the results presented below we have chosen $N = 30$, which proves to be sufficient, and the Bloch index $q \in [-0.5k, 0.5k)$ is discretized on an interval of $0.1k$.

Under a secular limit defined by

$$\Delta \gg \gamma_1, \quad (16)$$

the energy separations between the Bloch states belonging to different energy bands are much greater than the relaxation rates of these states, and one can therefore neglect the coherences between these different Bloch states [1]. The density-matrix equation, Eq. (11), then becomes a set of rate equations for the populations of various Bloch states $\pi_{n,q}$. Starting from an arbitrary initial distribution of the atomic population among various Bloch states, the density matrix reaches a quasiequilibrium that corresponds to the results of Sisyphus cooling after a time scale of order $1/\Gamma'$, where $\Gamma' = \gamma_1 \Omega_1^2 / 4(\gamma_1^2/4 + \Delta^2)$ is the optical pumping rate of the system. The atoms then undergo a much slower process of VSCPT cooling. In the case of a decaying Rydberg state, i.e., $\gamma_2 \neq 0$, the noncoupled state $|\mathrm{NC}\rangle$ also decays through spontaneous emission at a rate given by

$$\Gamma_{\mathrm{NC}} = \frac{1}{1 + (\Omega_2/\Omega_1)^2} \gamma_2. \quad (17)$$

As a result, the density matrix can reach a steady state on a time scale given by $1/\Gamma_{\mathrm{NC}}$.

The time evolution of the c.m. momentum distribution function for all the atoms, $\rho(p)$, and that for atoms in the Rydberg state, $\rho_r(p)$, are presented in Figs. 2(a) and (b), respectively, for a potential depth $U_0 = 200E_k$, where $E_k = \hbar^2 k^2 / 2M = E_{k_1}/4$. For realistic atomic systems such as the Na $3\,^2S_{1/2} \rightarrow 3\,^2P_{1/2} \rightarrow 45\,^2D_{5/2}$ transition, the ratio $\gamma_2/\gamma_1$ is on the order of $10^{-4}$. For typical laser intensities and detunings for Sisyphus cooling, the ratio $\gamma_2/\Gamma'$ is on the order of 0.001. As one can see from Fig. 2, after an initial period of Sisyphus cooling on the order of $\Gamma' t = 10$, the atomic momentum distribution width is reduced to a few photon recoil momenta. The effect of VSCPT then leads to the appearance of subrecoil peaks in the distribution at $p = \pm \hbar k_1$ and $p = \pm \hbar k_2$, which is a manifestation of the accumulation of atoms in the noncoupled state $|\mathrm{NC}\rangle$. The peaks at $\pm \hbar k_1$ correspond to atoms in the Rydberg state, as shown in Fig. 2(b). For this particular choice of parameters, more than 65% of the atomic population is found to be in the Rydberg state at equilibrium. Notice that this result is independent of the initial condition for the atomic density matrix, provided that the initial atomic momentum distribution is sufficiently narrow to be within the velocity capture range of Sisyphus cooling. One may thus regard the current standing-wave configuration as both an excitation and a cooling scheme for the Rydberg atoms.

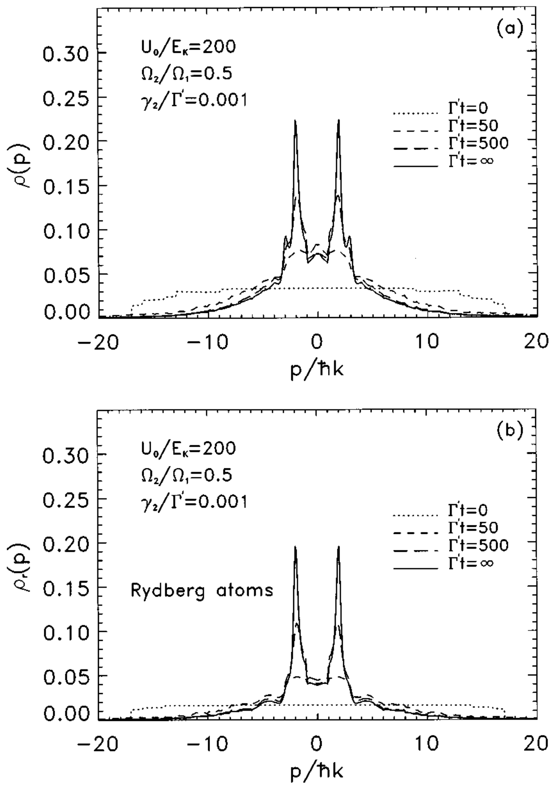


FIG. 2. The c.m. momentum distribution of (a) all the atoms and (b) the atoms in the Rydberg state $|r\rangle$, after various cooling times $t$. The potential depth $U_0 = 200E_k$, the ratio $\Omega_2/\Omega_1 = 0.5$, and the Rydberg state decay rate $\gamma_2 = 0.001\Gamma'$.

The steady-state population in the noncoupled state $|\mathrm{NC}\rangle$ is expected to be sensitive to the ratio between the Rydberg-state decay rate $\gamma_2$ and the optical pumping rate $\Gamma'$, since it is the balance between the feeding of $|\mathrm{NC}\rangle$ at a rate of order $\Gamma'$ and the leaking at a rate $\Gamma_{\mathrm{NC}}$ that determines the equilibrium population of $|\mathrm{NC}\rangle$ and the effects of VSCPT. The result of Sisyphus cooling, on the other hand, should remain insensitive to the ratio $\gamma_2/\Gamma'$ as long as $\gamma_2 \ll \Gamma'$. The above analysis has been confirmed by further calculations. For example, if one chooses a greater Rydberg-state decay rate $\gamma_2/\Gamma' = 0.005$, the magnitudes of the subrecoil peaks at $p = \pm \hbar k_1$ in the equilibrium momentum distributions decrease by nearly 50% as compared to the result in Fig. 2, while the wing of the distribution, which corresponds to the result of Sisyphus cooling, as well as the total population in the Rydberg state, is not significantly affected by the increase of $\gamma_2$.

The optimal value for the choice of the ratio between the two Rabi frequencies, $\Omega_2/\Omega_1$, can be determined based on two conflicting factors. First, since the percentage of the Rydberg atoms in the population of the noncoupled state $|\mathrm{NC}\rangle$ is given by $1/(1+\Omega_2^2/\Omega_1^2)$ [see Eq. (5)], it seems that one should choose a small value of $\Omega_2/\Omega_1$ in order to have most of the cooled atoms in the Rydberg state. On the other hand, as analyzed above, the process of Sisyphus cooling depends on the transition of atoms from $U_1(z)$ to $U_0(z)$ through optical pumping involving spontaneous emissions of the Rydberg state. The rate of this transition depends on the weight of the Rydberg state in the motional states associated with $U_1(z)$, which is proportional to $(\Omega_2/\Omega_1)^2/(1+\Omega_2^2/\Omega_1^2)$, as can be seen from Eq. (14). Therefore, if the ratio $\Omega_2/\Omega_1$ is too small, the effect of Sisyphus cooling will be greatly reduced due to the lack of optical pumping processes that are responsible for the dissipation of the atomic kinetic energy. In the asymptotic case when $\Omega_2/\Omega_1 \rightarrow 0$, the cascade system reduces to a two-level system, and Sisyphus cooling is lost. We illustrate these considerations in Fig. 3, where we plot the average kinetic energy $E_r$ and the total population $P_r$ of the Rydberg atoms versus $\Omega_2/\Omega_1$, respectively. As can be seen from Fig. 3, both $E_r$ and $P_r$ decrease with $\Omega_2/\Omega_1$, in agreement with our previous analysis. Therefore, the maximization of the Rydberg-state population with the decrease of $\Omega_2/\Omega_1$ will be accompanied by an increase of the average kinetic energy $E_r$, and vice versa.

Our analysis has not included some additional features of the real Rydberg systems, which may modify the fraction of cold atoms prepared in the Rydberg state. The Rydberg state may have a branching ratio toward lower states other than the first excited level considered in our analysis. However, as long as these intermediate states decay much faster than the Rydberg state, the effects of their existence mainly affect the feeding of the noncoupled state and should remain small. Furthermore, the lifetimes of Rydberg states are generally sensitive to the blackbody radiation, which can induce transitions to neighboring states or to the continuum [3]. The finite amount of blackbody radiation will pose some limitation on the efficiency of the VSCPT cooling of Rydberg atoms, and experiments based on this scheme should be performed in a cryogenic environment to reduce the effects of blackbody radiation.

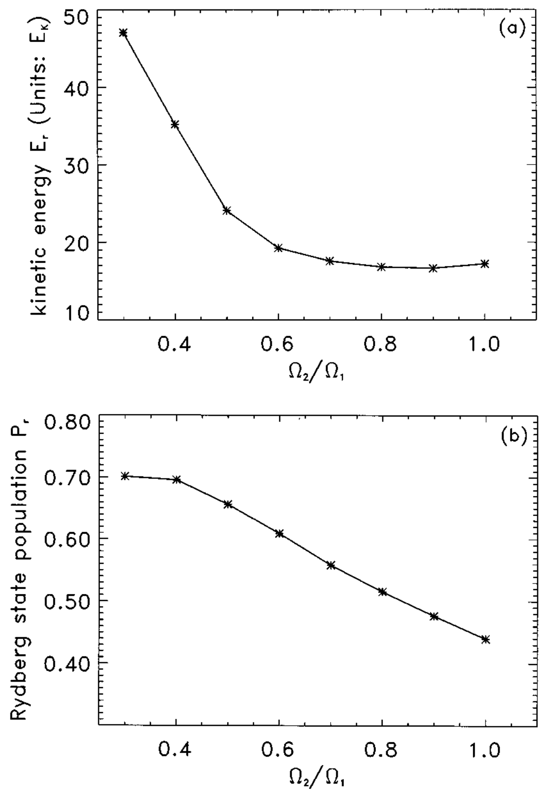


FIG. 3. (a) The average kinetic energy $E_r$ of the Rydberg atoms and (b) the total population of the Rydberg state $P_r$, as a function of the Rabi frequency ratio $\Omega_2/\Omega_1$. The potential depth $U_0=200E_k$, and the decay rate $\gamma_2=0.001\Gamma'$.

In conclusion, we have presented in this paper a model of laser cooling of Rydberg atoms in bichromatic standing-wave fields. At equilibrium, the majority of the atoms can be prepared in the Rydberg state with a narrow c.m. momentum distribution width resulting from the combination of VSCPT cooling and Sisyphus cooling. Although this present calculation is performed in a one-dimensional configuration, our model can be generalized to higher-dimensional cases by applying a pair of standing light waves in each spatial dimension. Research along this direction is currently under way.

We thank E. Korsunsky for helpful discussions. J.G. is supported by the E. U. Condon Fellowship at JILA.

[1] Y. Castin and J. Dalibard, Europhys. Lett. **14**, 761 (1991).
[2] A. Aspect *et al.*, J. Opt. Soc. Am. B **6**, 2112 (1989).
[3] T. F. Gallagher, *Rydberg Atoms* (Cambridge University Press, Cambridge, England, 1994).
[4] R. Gupta *et al.*, Phys. Rev. Lett. **71**, 3087 (1993); T. T. Grove *et al.*, Phys. Rev. A **51**, R4325 (1995).
[5] E. Korsunsky and Yu. Rozhdestvensky, Phys. Rev. A **52**, 3027 (1995).
[6] M. S. Shahriar *et al.*, Phys. Rev. A **48**, R4035 (1993).
[7] P. Marte *et al.*, Phys. Rev. A **49**, 4826 (1994).
[8] J. Guo and J. Cooper, Phys. Rev. A **52**, R1819 (1995).
[9] J. Guo, E. Korsunsky, and E. Arimondo (unpublished).
[10] The choice of 1.5 for $k_2/k_1$ is solely for the convenience of our calculation. With a rational number other than 1.5 for $k_2/k_1$, the results of cooling should remain essentially the same.