Bichromatic velocity-selective coherent population trapping

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We propose a scheme to achieve one-dimensional velocity-selective coherent population trapping in a five-level system under two-frequency laser excitation. It may be applied to either D line of alkali-metal atoms. The trapped state is a time-dependent superposition of three sublevels from two different hyperfine ground states. Stable trapping may be produced by correctly detuning the laser beams to compensate for different hyperfine and kinetic energies of these ground states. We present numerical results for a five-level model, as well as for the complete level systems appropriate for alkali-metal atoms.

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Thermodynamics demands dissipative irreversible processes in order to cool atomic samples. The primary such mechanism provided by atom-light interactions is spontaneous decay from excited states populated by the laser light. Such decays always produce discrete atomic momentum changes in random directions, and thereby limit the ultimate rms momentum of an atomic sample to a few times $P_R = MV_R = \hbar k$, the recoil momentum caused by exchange of one photon with the laser field ($k = 2\pi/\lambda$ is the optical wave number and M is the atomic mass). There are two demonstrated ways to overcome this limitation [1,2].

One of these is the phenomenon called velocity-selective coherent population trapping (VSCPT), introduced in [2], which produces peaks of subrecoil width in the atomic momentum distribution. In VSCPT, atoms interacting with laser light can fall into a "dark" (also called "noncoupled") state through spontaneous emission [2,3]. Such states consist of internal atomic ground states entangled with external momentum states, so that laser excitation is exactly canceled by interfering transition amplitudes. Dark states couple to other linear combinations of ground states (which are *not* dark) via the atomic and kinetic energy parts of the Hamiltonian. The *trapped* state is the dark state of some particular momentum for which this coupling vanishes, so that atoms accumulate in this state as long as the atom-laser interaction continues.

Most VSCPT schemes examined so far involve a three-level system of two ground states and one excited state ("Λ-VSCPT") [2–7]. The seminal work [2] used counter-propagating σ^+-σ^- laser beams on the $J_g = 1 \rightarrow J_e = 1$ transition in He*. Since VSCPT has only been observed in He [2,8,9], and since laser cooling is interesting for its application to alkali-metal atoms, e.g., for atomic clocks, it is useful to study VSCPT schemes appropriate for alkali-metal atoms. A straightforward extension of the He* scheme would be simply to use the $F_g = 1 \rightarrow F_e = 1$ transition on the $^2S_{\frac{1}{2}} \rightarrow {}^2P_{\frac{1}{2}}$ transition (D_1 line) or $^2S_{\frac{1}{2}} \rightarrow {}^2P_{\frac{3}{2}}$ transition (D_2 line) in the alkali-metal atoms. In this case a repumper must be added to recycle atoms that have decayed into the $F_g = 2$ hyperfine state.

In this paper we explore a different VSCPT scheme, where the trapped state is made up of magnetic sublevels of *both* hyperfine ground states. Our more general scheme achieves VSCPT in a five-level system. This system is closely analogous to the monochromatic inverted-W system of an $F_g = 2 \rightarrow F_e = 1$ transition studied in [10], which has been shown to produce transient VSCPT in theory. In that case, there is a dark state that is not completely trapped because the kinetic energies of the three ground states cannot be equal [11]. In our five-level system, a *bichromatic* inverted-W, there is also an energy difference between the ground states (see Fig. 1). However, it has the added freedom of a second frequency (hence the name "bichromatic VSCPT" or BVSCPT), that allows for *stable* trapping by choosing appropriate laser frequencies ω_1 and ω_2 to compensate for the energy difference.

Our five-level system is present in, and thus models, the situation of two separate hyperfine ground states of

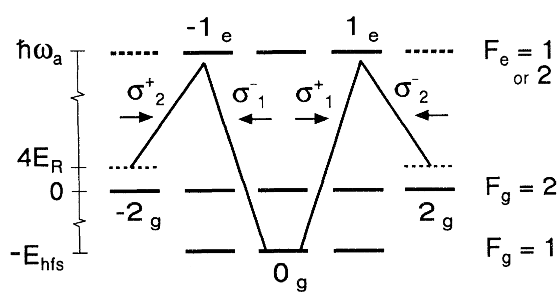

FIG. 1. Level scheme for BVSCPT. The relevant five levels that produce trapping are labeled. Energy differences between the five states are shown, including small kinetic energy differences (not to scale). $F_g = 2$ is the zero-energy state; $F_g = 1$ is at $-E_{\rm hfs}$, and F_e is at $+\hbar\omega_a$.

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many alkali-metal atoms, $F_g = 1$ and 2, that are connected by optical transitions to one hyperfine excited state, either $F_e = 1$ or $F_e = 2$, via two σ^+ laser beams counterpropagating with two σ^- laser beams (Fig. 1). One of the σ^+ beams is tuned near the $F_g = 1 \to F_e$ transition, the other near the $F_g = 2 \to F_e$ transition; the same holds for the σ^- beams. We assume it is possible to fix each of the four different σ^+ and σ^- beams so that each beam drives only *one* transition as shown in Fig. 1. Because the hyperfine ground states are separated by a large splitting $E_{\rm hfs}$, this is a good assumption. For Na, $E_{\rm hfs} = 1.7$ GHz, while our detunings ($\delta_1 = \omega_1 - \omega_a - E_{\rm hfs}/\hbar$ and $\delta_2 = \omega_2 - \omega_a$) may be much less than a natural linewidth $\Gamma = 2\pi \times 10$ MHz ($\hbar\omega_a$ is the internal energy of the excited state relative to the zero-energy $F_g = 2$ ground state, see Fig. 1). BVSCPT can be realized particularly easily on the D_1 line of the alkali-metals since it has only two excited hyperfine states, $F_e = 1, 2$ (separated by about 190 MHz in Na). The D_2 line contains extraneous transitions ($F_e = 0, 1, 2, 3$) that are less well separated.

We primarily restrict ourselves to a simple model to develop the fundamental aspects of BVSCPT. Both internal and external states must be quantized to study subrecoil phenomena accurately, so we use states $|p, m_F\rangle = |p\rangle \otimes |\, m_F\rangle$ with $|\, p\rangle$ a linear momentum eigenstate of eigenvalue p. Our model uses only the five magnetic sublevels labeled in Fig. 1. Central to the model are the families of states [3] $S_p = \{|\, p - 2\hbar k, -2_g\rangle, |\, p - \hbar k, -1_e\rangle, |\, p, 0_g\rangle, |\, p + \hbar k, 1_e\rangle, |\, p + 2\hbar k, 2_g\rangle\}$, which contain only those states relevant to BVSCPT. States within one family are connected to one another by absorption and stimulated emission in this five-level system, and different families are connected only by spontaneous emission [3]. In our model, the branching ratio from any excited state to any connected ground state is $\frac{1}{2}$ so all Clebsch-Gordan (CG) coefficients are set equal to 1. We have also studied the full system, including all magnetic sublevels of the ground and excited hyperfine states as indicated in Fig. 1, correct hyperfine transition rates ($6J$ symbols), and CG coefficients. We discuss these results later.

The five-level model Hamiltonian includes the atom's kinetic (external) and electronic (internal) energies, as well as the atom-field interaction: $H = H_{\rm ex} + H_{\rm in} + H_{\rm af}$. Here, $H_{\rm ex} = P^2/2M$ (P is the momentum operator along the direction of the laser beams, $\hat{\mathbf{z}}$). Inspecting Fig. 1, $H_{\rm in} = \hbar\omega_a \{|-1_e\rangle\langle -1_e| + |1_e\rangle\langle 1_e|\} - E_{\rm hfs}|0_g\rangle\langle 0_g| + 0\{|-2_g\rangle\langle -2_g| + |2_g\rangle\langle 2_g|\}$. In the electric-dipole approximation, the atom-field interaction is given by the product of the laser's electric field and the dipole moment operator, $H_{\rm af} = -\mathbf{E}\cdot\mathbf{d}$. The laser field is expressed as two σ^+ waves running in the $+\hat{\mathbf{z}}$ direction and two σ^- waves running in the $-\hat{\mathbf{z}}$ direction. For example, $\mathbf{E}_{2,-} = E_2\{\hat{\mathbf{x}}\cos(\omega_2 t + kz) - \hat{\mathbf{y}}\sin(\omega_2 t + kz)\}$ is the field of the σ^- laser beam that excites the $2_g \to 1_e$ transition. We set the wave numbers to be equal, $k_1 = k_2 = k$, justified when atoms travel only a distance Δz satisfying $\Delta z \mid k_1 - k_2 \mid \ll \frac{\pi}{2}$. For a typical slow atom with momentum on the order of P_R, this condition is fulfilled for interaction times $t \ll hc/V_R E_{\rm hfs}$ ($\approx 10^7\Gamma^{-1}$ for Na). Then

$$
\begin{aligned}
H_{\rm af} = \hbar\int_{-\infty}^{\infty} dp\{&\Omega_2 e^{-i\omega_2 t}\mid p - \hbar k, -1_e\rangle\langle p - 2\hbar k, -2_g \mid \\
&+\Omega_1 e^{-i\omega_1 t}(\mid p + \hbar k, 1_e\rangle + \mid p - \hbar k, -1_e\rangle)\langle p, 0_g \mid \\
&+\Omega_2 e^{-i\omega_2 t}\mid p + \hbar k, 1_e\rangle\langle p + 2\hbar k, 2_g \mid + \text{H.c.}\}\ ,
\end{aligned}
\tag{1}
$$

where we treat z as the position operator Z and use $e^{\pm ikZ} = \int_{-\infty}^{\infty} dp \mid p\rangle\langle p \mp \hbar k \mid$. The single-beam Rabi frequencies Ω_1 and Ω_2 are given by $\Omega_i = \Gamma\sqrt{s_i/2}$ with $s_i = 24\pi^2 I/hc\Gamma k^3$ the corresponding saturation parameter, and I is the laser intensity. $H_{\rm af}$ is thus expressed in the basis of the S_p family of states.

We now generate a ground-state basis that contains the trapped state, physically appropriate for exploring BVSCPT. This new basis is composed of linear combinations of the ground-state elements of S_p. Requiring one of these basis states to be dark means it is an eigenstate of $H_{\rm af}$ since the dark state $|\, N\,\rangle$ is defined by $H_{\rm af}\,|\, N\,\rangle = 0$. We construct the effective potential matrix $H_{\rm af}H^{\dagger}{}_{\rm af}$ that governs the evolution of the ground-state density matrix [12] and diagonalize it. Its eigenstates form a complete, orthonormal ground-state basis and the state with zero eigenvalue is the dark state $|\, N\,\rangle$.

$H_{\rm af}H_{\rm af}^{\dagger}$ is infinite and block diagonal in ground and excited states. The ground-state block, V_{gg}, is also block diagonal, each block being a 3×3 matrix in the ground-state basis of S_p. A representative block may be written in matrix form as

$$
V_{gg,p} \propto \hbar^2 \begin{pmatrix} \Omega_2^2 & \Omega_2\Omega_1 e^{i\omega_{21}t} & 0 \\ \Omega_1\Omega_2 e^{i\omega_{12}t} & 2\Omega_1^2 & \Omega_1\Omega_2 e^{i\omega_{12}t} \\ 0 & \Omega_2\Omega_1 e^{i\omega_{21}t} & \Omega_2^2 \end{pmatrix}\ , \tag{2}
$$

where $\omega_{21} = \omega_2 - \omega_1 = -\omega_{12}$. (The upper left element, $\langle p + 2\hbar k, 2_g \mid V \mid p + 2\hbar k, 2_g\rangle$, represents the light shift of state $\mid 2_g\rangle$.) The eigenvalues are $\lambda_S = \hbar^2(2\Omega_1^2 + \Omega_2^2), \lambda_W = \hbar^2\Omega_2^2$, and $\lambda_N = 0$. The associated eigenkets are

$$
\begin{aligned}
\mid N\rangle = A_0\{&\Omega_1 \mid p - 2\hbar k, -2_g\rangle - \Omega_2 e^{i\omega_{12}t}\mid p, 0_g\rangle \\
&+\Omega_1 \mid p + 2\hbar k, 2_g\rangle\}\ ,
\end{aligned}
\tag{3a}
$$

$$
\mid W\rangle = \frac{1}{\sqrt{2}}\{|p - 2\hbar k, -2_g\rangle - |p + 2\hbar k, 2_g\rangle\}\ , \tag{3b}
$$

$$
\begin{aligned}
\mid S\rangle = \frac{A_0}{\sqrt{2}}\{&\Omega_2|p - 2\hbar k, -2_g\rangle + 2\Omega_1 e^{i\omega_{12}t}|p, 0_g\rangle \\
&+\Omega_2 \mid p + 2\hbar k, 2_g\rangle\}\ ,
\end{aligned}
\tag{3c}
$$

where $A_0^{-1} = \sqrt{2\Omega_1^2 + \Omega_2^2}$. These strongly, weakly, and noncoupled states [10] form the "optical basis."

Notice that both $|\, N\,\rangle$ and $|\, S\,\rangle$ have nontrivial time dependence (i.e., not just an overall time-dependent phase). For one value of p, the noncoupled state is our trapped state. Thus the trapped state is not a *stationary* eigenstate of the full Hamiltonian. This is in contrast to Λ-VSCPT, where the time dependence of the trapped state is trivial. In the bichromatic case, at different times the dark state is a different combination of ground states. Thus the dark state must evolve synchronously with the atom-laser interaction for atoms to remain trapped.

We now develop the conditions under which a dark state produces trapping. We require the rate of escape *out* of the dark state, $[d\langle N|\sigma|N\rangle/dt]_{\text{out}} = \langle \dot{N}|\sigma|N\rangle + \langle N|\sigma|\dot{N}\rangle + \langle N|\dot{\sigma}|N\rangle$, to be zero for one value of p. The last term we obtain from the generalized optical Bloch equations for the noncoupled density matrix element, $i\hbar\dot{\sigma}_{NN} = [H,\sigma]_{NN}$ (not including the decay term, which only *feeds* the ground-state populations). The commutator is $[H,\sigma]_{NN} = \sum_I \{H_{NI}\sigma_{IN} - \text{c.c.}\}$, where the sum is over optical basis states $I = N, W, S, 1_e, -1_e$. Since the dark state is composed entirely of ground states and is not coupled to the laser field, $H_{N,1_e} = H_{N,-1_e} = 0$. The terms we need are

$$H_{NW} = \frac{8\Omega_1}{\sqrt{4\Omega_1^2 + 2\Omega_2^2}}\, p\, \frac{\hbar k}{2M}, \tag{4}$$

$$H_{NS} = \frac{\sqrt{2}\Omega_1\Omega_2}{2\Omega_1^2 + \Omega_2^2}(4E_R + E_{\text{hfs}}) \tag{5}$$

($E_R = P_R^2/2M$). The time dependence of the dark state itself results in

$$i\hbar(\langle \dot{N}|\sigma|N\rangle + \langle N|\sigma|\dot{N}\rangle) = \frac{\sqrt{2}\Omega_1\Omega_2}{2\Omega_1^2+\Omega_2^2}\hbar\omega_{21}\sigma_{SN} - \text{c.c.} \tag{6}$$

From Eqs. (4), (5), and (6), we obtain the total rate of loss of the dark-state population:

$$i\hbar\left[\frac{d\langle N \mid \sigma \mid N\rangle}{dt}\right]_{\text{out}} = \frac{8\Omega_1}{\sqrt{4\Omega_1^2 + 2\Omega_2^2}}\, p\, \frac{\hbar k}{2M}\sigma_{WN} + \frac{\sqrt{2}\Omega_1\Omega_2}{2\Omega_1^2 + \Omega_2^2}\{4E_R + E_{\text{hfs}} + \hbar\omega_{21}\}\sigma_{SN} - \text{c.c.} \tag{7}$$

If $p = 0$ and $4E_R + E_{\text{hfs}} + \hbar\omega_{21} = 0$, the derivative is zero. Using the difference detuning $\delta = \delta_2 - \delta_1$ and using $\hbar\omega_{21} + E_{\text{hfs}} = \hbar\delta$ gives

$$p = 0, \tag{8a}$$

$$4E_R/\hbar + \delta = 0 \tag{8b}$$

as the two conditions which must be fulfilled to achieve BVSCPT. Equations (8a) and (8b) reveal two routes out of the noncoupled state. One is the "motional coupling," proportional to p, which is also found in Λ-VSCPT [3]. The other is the "bichromatic coupling," proportional to $4E_R/\hbar + \delta$, due to the energy difference between the ground states that make up the trapped state. Since δ is an experimentally adjustable parameter, this bichromatic coupling can be made to vanish. BVSCPT causes atoms to collect in the dark state with family momentum $p = 0$. Then three peaks appear in the velocity distribution, at 0 and $\pm 2V_R$, the three ground-state velocities of this family.

In Figs. 2 and 3 we present numerical results demonstrating the trapping conditions for BVSCPT in the five-level system, for a model Li atom (for which the recoil parameter $\epsilon \equiv E_R/\hbar\Gamma = 1.1 \times 10^{-2}$). The calculations solve the equations of motion for the density matrix of the five-level system, as determined by the generalized optical Bloch equations (including spontaneous emission). We used a free-particle momentum basis spanning $[-8\hbar k, 8\hbar k]$ [13]. In Fig. 2(a) we show the time evolution of the velocity distribution up to $20\tau_R$ ($\tau_R = \hbar/E_R$ is the recoil time). The subrecoil-width peaks at velocity $V = 0, \pm 2V_R$ continue to grow and narrow for long interaction times. Figure 2(b) shows the corresponding population in the dark state as a function of p and t, with a subrecoil-width peak at the trapped-state family momentum $p = 0$. The population in the dark state was extracted from the numerical data used for Fig. 2(a) by projecting the density matrix onto the optical basis. These data illustrate stable VSCPT when Eqs. (8) are satisfied. Figure 3 shows how the population in the trapped state $\langle N(p=0) \mid \sigma \mid N(p=0)\rangle$ changes with δ, for fixed interaction time $20\tau_R$. Clearly, trapping is most efficient at $\delta = -4\epsilon\Gamma$.

In a Λ system, VSCPT promotes the growth of two

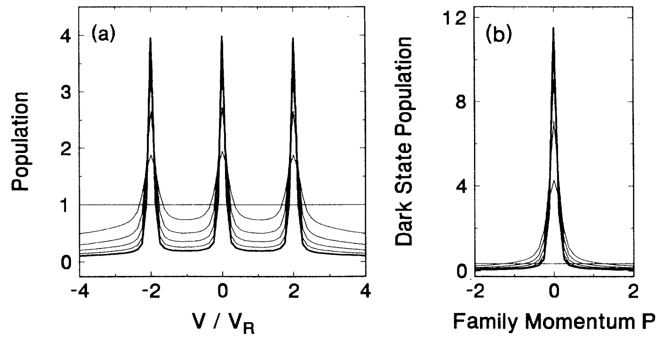

FIG. 2. Calculations for a five-level model Li atom ($\epsilon = 1.1 \times 10^{-2}$), when $\Omega_1^2 = \Omega_2^2 = 0.05\Gamma^2$. Detunings are $\delta_1 = -\delta_2 = 2\epsilon\Gamma$, so $\delta = -4\epsilon\Gamma$. Each profile represents the population at increments of $4\tau_R$, for total interaction time $T_{\text{tot}} = 20\tau_R$. (a) The velocity distribution exhibits three subrecoil peaks at $V = 0, \pm 2V_R$. (b) The dark state is most heavily populated at family momentum $p = 0$, where stable trapping occurs. Peak height increases with time in both plots.

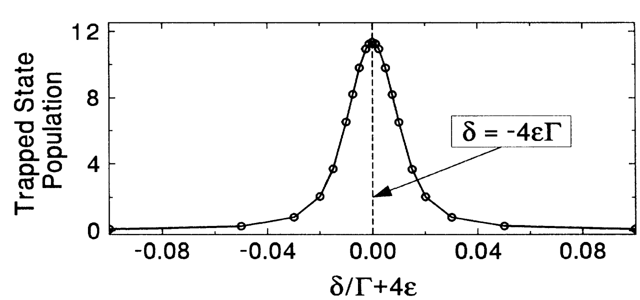

FIG. 3. Calculated population in the trapped state, $\langle N(p=0)|\sigma|N(p=0)\rangle$, at $T_{\text{tot}} = 20\tau_R$ with varying δ. Aside from δ, we used the same parameters as for Fig. 2, and kept $\delta_2 = -\delta_1$. Stable trapping occurs when $\delta = -4\epsilon\Gamma$.

peaks in the velocity distribution at $\pm V_R$. As discussed in [3], the relative peak heights of the two branches of the Λ system can be varied. An analogous phenomenon occurs in BVSCPT. If most of the population is in the noncoupled state, then the peak height in the velocity distribution at $V = \pm 2V_R$ is related to the peak height at $V = 0$ by $P_{\pm 2V_R}/P_0 = \Omega_1^2/\Omega_2^2$. Hence, we might trap virtually all the population at $V = 0$ in $| 0_g\rangle$ by taking $\Omega_2 \gg \Omega_1$.

So far we have discussed only the model system that describes the essence of BVSCPT. To model a real alkali-metal atom more closely, we have performed calculations that include all magnetic sublevels of the relevant hyperfine states, correct CG coefficients, and $6J$ symbols. Typical results are plotted in Fig. 4. In our model five-level system, the velocity distribution exhibits characteristic VSCPT peaks more quickly than in these full-system calculations. This is expected since trapping occurs when atoms make a random walk into the trapped state, and the full system contains many states irrelevant to the VSCPT process. However, for long enough interaction times, we again see strong peaks in the velocity distribution. We show calculated momentum distributions for $F_e = 1$ (2) in Figs. 4(a) [4(b)], with parameters for the sodium D_1 line. These plots indicate that BVSCPT is readily accessible experimentally. Interaction times needed to observe the corresponding trapping peaks for Li are within the range of beam experiments. For Na and heavier alkali-metal atoms, it might be more suitable to first confine the atoms with a magneto-optical trap, then turn off the trap and apply the BVSCPT fields, as in Ref. [9].

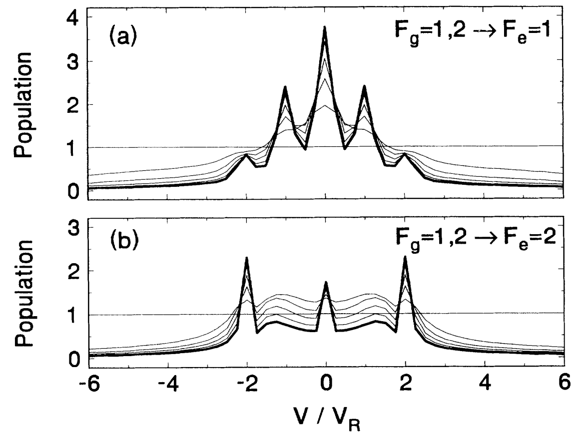

FIG. 4. (a) Full-system calculation of the velocity distribution for BVSCPT in Na ($\epsilon = 2.5 \times 10^{-3}$), with $F_e = 1$. The peaks at $V = \pm V_R$ are due to the presence of Λ systems. Peak heights differ because of different $6J$ symbols and CG coefficients. $T_{\rm tot} = 25\tau_R$, with increments of $5\tau_R$. Laser intensities are given by $\Omega_1^2 = \Omega_2^2 = 0.1\Gamma^2$. (b) Same as (a), but $F_e = 2$.

To conclude, we have used a model system with only five internal energy states to show the existence of VSCPT in a bichromatic laser field. We have shown that the velocity-dependent trapped state has nontrivial time dependence, and that the efficiency of the trapping process can be controlled by a difference in the detuning of the laser beams. This difference detuning can be used to compensate for the difference in internal and kinetic energies of the states that make up the coherent superposition of ground states. Our five-level system is present in the more complicated $F_g = 1, F_g = 2 \to F_e = 1$ and $F_g = 1, F_g = 2 \to F_e = 2$ configurations, both of which can be realized on the D_1 or D_2 lines of the alkali-metal atoms. We have demonstrated that the full configurations will produce BVSCPT in these atoms under experimentally accessible conditions. We conclude that BVSCPT may be employed as a useful tool in the production of subrecoil temperatures among the alkali-metal atoms.

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