Subrecoil laser cooling by velocity-selective coherent population trapping in cascade systems

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A theoretical analysis of laser cooling of atoms by velocity-selective coherent population trapping in a three-level cascade system is presented. We show that the temperature of the cooled atoms can be well below the value determined by the recoil energy. Conditions for coherent population trapping in the cascade scheme to be established are derived and different implementations of cascade schemes are discussed. The analysis of laser cooling dynamics is performed with both full quantum and quasiclassical (for translational motion of atoms) approaches. We study the temporal evolution of the atomic momentum distribution and the efficiency of the cooling method considered. The analytical solution for the atomic density matrix is obtained in the limit of large interaction time and small kinetic momentum of the atom. © 1995 American Institute of Physics.

1. INTRODUCTION

Laser cooling by means of velocity-selective coherent population trapping (VSCPT) has attracted a lot of attention in last few years. There are some important reasons to study this cooling method in detail. First of all, the mechanism permits the cooling of atoms below not only the Doppler limit temperature $T_{\rm D} = \hbar \gamma / 2k_{\rm B}$, but also below the photon recoil temperature $T_R = R/k_B$, where k_B is the Boltzmann constant, γ is the relaxation rate of the atomic transition, $R = \hbar^2 k^2 / 2M$ is the one-photon recoil energy, M is the mass of the atom, and k is the wave number of the photon.^{1,2} Another circumstance is that VSCPT, being a general quantum mechanical phenomenon, can be realized in a wide variety of atom-field interaction schemes. Therefore VSCPT can be applied to get very low temperatures for ensembles of quite different quantum objects including atoms, ions and molecules, and to achieve two-3-6 and three-dimensional⁵ cooling by proper choice of laser and atomic configurations. VSCPT provides an interesting example of a physical mechanism performing local (in momentum and, possibly, in coordinate space and in time) freezing of the degrees of freedom of quantum objects by means of diffusion redistribution.

The phenomenon of coherent population trapping (CPT) is based on destructive quantum interference between the amplitudes of transitions in a laser-irradiated multilevel atom. This interference occurs under certain conditions of interaction, in particular, for certain values of the velocities of the atom. As a result, superpositional 'dark' states $|\Psi_{NC}\rangle$ appear, which are not coupled to the rest of the system by laser radiation but can be filled by means of spontaneous emission. Thus, after some fluorescence cycles most of the atomic population is optically pumped into $|\Psi_{NC}\rangle$ and trapped there.

Up to now all the experimental realizations and perspective proposals of VSCPT have exploited the configurations of laser-atom interaction, where superpositions $|\Psi_{NC}\rangle$ are generated by radiative-stable lower states (such as three-level Λ -system and various multilevel systems of the magnetical sublevels of two states).¹⁻¹⁰ The use of such configurations allows one to avoid completely the decay of the population of $|\Psi_{NC}\rangle$ through spontaneous relaxation, but it restricts, in some respects, the possible applications of laser cooling by VSCPT. At the same time, the phenomenon of CPT occurs in cascade system of excited states (Fig. 1)¹¹⁻¹⁴ as well. The present paper is designed to show the feasibility of laser cooling with VSCPT in a three-level cascade scheme for the interaction of an atom with laser radiation. This scheme allows the number of quantum objects which can be cooled down to sub-recoil temperatures to be extended considerably.

In general, the states $|\Psi_{
m NC}
angle$ are never 'perfect traps' for the population even if they are superpositions of stable ground states. There always exist processes (laser fluctuations, collisions, etc.) violating the phase correlation between transition amplitudes and thereby destroying the interference. These processes cause the atomic coherence to relax with the rate Γ . Therefore the trap states $|\Psi_{\rm NC}\rangle$ are always decaying ones, that leads to incomplete population trapping. Another reason for instability is that the $|\Psi_{
m NC}
angle$ are not exact eigenstates of the Hamiltonian of the unperturbed atom for some interaction schemes.^{7–9} Nevertheless, if the loss rate from the trap state is low enough, the population trapping is coherent in the sense that a large fraction of the atomic population is trapped in $|\Psi_{\rm NC}\rangle$ for rather a long time. In particular, atoms can be laser-cooled to temperatures lower than the recoil limit T_R .^{1,7,10}

One can estimate the temperature limit for cooling by VSCPT with the following simple qualitative considerations. For moving atoms the state $|\Psi_{\rm NC}(p)\rangle$ is real trap for population only at certain value p_0 of atomic momentum. The loss rate $\Gamma_{\rm NC}(p)$ of $|\Psi_{\rm NC}(p)\rangle$ is close to zero (but not equal to zero) for $p = p_0$ and increases with momentum as $(p - p_0)^2$



FIG. 1. Three-level (or Ξ -) scheme of interaction of an atom with the two travelling electromagnetic waves with the wave vectors \mathbf{k}_m and frequencies ω_m . Detuning and spontaneous relaxation rate for the $|m\rangle - |3\rangle$ transitions are denoted by Ω_m and $2\gamma_m$ respectively (m=1,2).

(for the range of p close to p_0 the rate is $\Gamma_{\rm NC}(p) \cong 2[k(p-p_0)/M]^2/(g^2/\gamma)$, with g being the Rabi frequency of the optical transition; see, e.g., Eq. (3.16) in Ref. 3). The minimum value of $\Gamma_{NC}(p)$ is equal to Γ , the decay rate of the coherence between the states generating the trap state superposition.^{10,15} During the interaction with light the atoms accumulate in $|\Psi_{\rm NC}(p=p_0)\rangle$ so that the width δp of the atomic momentum distribution asymptotically tends to the value which determines the minimum loss rate, $\Gamma_{\rm NC}(p) \cong 2(k \, \delta p/M)^2/(g^2/\gamma) \cong \Gamma$. Therefore the minimum width attainable VSCPT in cooling is δp $\approx \hbar k (g \sqrt{\Gamma/2 \gamma/2 \omega_R})$, where $\omega_R = R/\hbar$ is the recoil frequency, with a corresponding effective temperature $T = (\delta p)^2 / 2M k_{\rm B} \approx (g^2 \Gamma / 8 \omega_R^2 \gamma) T_R$. The CPT establishment process has a threshold: the saturation parameter $g^2/2\gamma^2$ has to be larger than the small quantity $\Gamma/2\gamma$ to allow CPT to occur.¹⁴ Therefore the lowest temperature to which atoms can be cooled by the VSCPT method is $T_{\min} = (\Gamma^2 / 8\omega_R^2)T_R$. A rigorous derivation of these formulas on the basis of the quantum kinetic equations for atomic density matrix is presented in Ref. 10. Thus, the atoms are cooled below the recoil limit when the parameter Γ/ω_R is less then unity.

In case of a cascade atomic system (Fig. 1) the 'dark' state is a superposition of the ground state $|1\rangle$ and the highest excited state $|2\rangle$ which decays spontaneously, so that the loss rate Γ is increased by the contribution of the spontaneous emission. However, under certain conditions CPT in cascade scheme is possible¹¹⁻¹⁴ as well as in schemes where $|\Psi_{\rm NC}\rangle$ is composed only of ground stable states.¹⁻¹⁰ In this paper we give explicit expressions for these conditions and extend the study of coherent population trapping in cascade schemes to VSCPT and to laser cooling by VSCPT. Apart from the possibility of subrecoil cooling of a new class of atoms, VSCPT in cascade schemes deserves attention since the 'usual' mechanism of VSCPT cooling by diffusion redistribution in momentum space 1,3,15 is mediated by the radiation force, as in asymmetrical schemes of interaction.^{7,16} The force has a special dependence on atomic velocity that causes strong deceleration of the cooled atoms. The presence of such a force considerably improves the cooling efficiency, allowing us to consider this method as a powerful tool to manipulate atomic motion.

The paper is organized as follows. In Sec. 2 we derive the explicit form of the trap state $|\Psi_{NC}\rangle$. The state $|\Psi_{NC}\rangle$ is

really non-coupled only for atoms with a center-of-mass velocity satisfying the condition of Doppler-shifted two-photon resonance in the cascade scheme. The conditions for the atomic parameters and the laser radiation to establish CPT are presented and discussed in Sec. 3. Dynamics of laser cooling by VSCPT in cascade systems is examined in this paper by use of both full quantum and quasiclassical (for the translational motion of atoms) approaches. It allows us to study the process of cooling in different regimes for a wide range of interaction parameters, as well as to find explicit expressions for the temperature limit of this cooling method. In Sec. 4 a quasiclassical approach is used to analyze the action of radiation force and momentum diffusion and the influence of different parameters on the efficiency of the cooling mechanism and to find the time scales of the process under study. We further propose two regimes of interaction of atoms with laser beams dramatically improving the efficiency, so that an atomic ensemble can be cooled almost without losses. Finally, we present in Sec. 5 an analytical solution for the atomic density matrix, treating both internal and translational degrees of freedom of the atoms quantum mechanically. The solution is obtained in the asymptotic limit of large interaction times and small atomic momenta. As a result we get the expressions for the asymptotic dependence of populations on time and momentum as well as for the width of the momentum distribution (temperature) and for the number of cold atoms. Appendix A contains the general expressions for the force and for the diffusion coefficient in the quasiclassical approach, while the analytical calculations presented in the last Section are developed in Appendix Β.

2. STATE OF VELOCITY-SELECTIVE TRAP FOR THREE-LEVEL CASCADE SYSTEM

In the present paper we consider an atom interacting with two travelling electromagnetic waves as a three-level cascade (Ξ) quantum system (Fig. 1). The Hamiltonian of the system under consideration has the form

$$\hat{H} = \frac{\hat{\mathbf{p}}^2}{2M} + \sum_{\mu=1}^{3} \varepsilon_{\mu} |\mu\rangle \langle \mu| + \sum_{\mu=1}^{2} \varepsilon_{\mu} |3\rangle$$
$$\times \langle 3|\hat{V}|\mu\rangle \langle \mu| + \text{H.c.}, \qquad (1)$$

where the first two terms in the right-hand side constitute the Hamiltonian \hat{H}_0 of an unperturbed atom with the energies ε_{μ} of the time-independent states $|\mu\rangle$ and with the kinetic energy operator $\hat{\mathbf{p}}^2/2M$. The last two terms describe the interaction of the atom with the applied fields. We assume that the interaction is an electric-dipole one for the $|1\rangle - |3\rangle$ transition and that it can be electric- or magnetic-dipole one for the $|2\rangle - |3\rangle$ transition (Fig. 1).

The aim of this Section is to find explicit expression for the trap state $|\Psi_{NC}(\mathbf{p})\rangle$ uncoupled to the other states by the laser-atom interaction. The state $|\Psi_{NC}\rangle$ responsible for the CPT is a solution of the Schrödinger equation (in the interaction representation):

$$i\hbar \frac{\partial |\Psi_{\rm NC}\rangle}{\partial t} = \hat{U}(\mathbf{r}, t) |\Psi_{\rm NC}\rangle$$
 (2)

with

$$\hat{U}(\mathbf{r},t) = \exp(i\hbar^{-1}\hat{H}_0 t)(\mathbf{r},t)$$

$$\times \exp(-i\hbar^{-1}\hat{H}_0 t), \qquad (3)$$

which satisfies the condition

$$\hat{U}(\mathbf{r},t)|\Psi_{\rm NC}\rangle = 0. \tag{4}$$

We will solve for $|\Psi_{NC}\rangle$ as a superposition of the eigenfunctions of the unperturbed Hamiltonian and the kinetic energy operator, requiring this state to be strictly stationary with respect to free motion of the atom:

$$|\Psi_{\rm NC}(\mathbf{r})\rangle = \sum_{\mu=1}^{3} a_{\mu}(\mathbf{r})|\mu\rangle.$$
(5)

Here $a_{\mu}(\mathbf{r})$ are the eigenfunctions of the kinetic-energy operator and $\hbar^2 \mathbf{b}_{\mu}^2 / 2M$ are the respective eigenvalues:

$$\frac{\hat{\mathbf{p}}^2}{2M} a_\mu(\mathbf{r}) = -\frac{\hbar^2}{2M} \hat{\Delta} a_\mu(\mathbf{r}) = \frac{\hbar^2 \mathbf{b}_\mu^2}{2M} a_\mu(\mathbf{r}), \qquad (6)$$

where $\hat{\Delta}$ is the Laplace operator. The solutions of Eq. (6) are the functions

$$a_{\mu}(\mathbf{r}) = A_{\mu} \exp(i\mathbf{b}_{\mu}\mathbf{r}), \tag{7}$$

where the amplitudes A_{μ} do not depend on **r** and *t*. The interaction operator $\hat{U}(\mathbf{r},t)$ in the resonance approximation has the form

$$\hat{U}(\mathbf{r},t) = -\hbar \left[\sum_{\mu=1}^{2} g_{\mu} \exp[(-1)^{\mu} i (\Omega_{\mu} t - \mathbf{k}_{\mu} \mathbf{r}) + i \omega_{R\mu} t] |3\rangle \langle \mu| + \text{H.c.} \right] \exp\left(i \frac{\hbar t}{2M} \hat{\Delta} \right).$$
(8)

In the expression (8) we denote the Rabi frequencies by $g_{\mu} = \langle 3 | \hat{V} | \mu \rangle / 2\hbar$, the detunings by $\Omega_1 = \omega_1 - (\varepsilon_3 - \varepsilon_1)/\hbar$, $\Omega_2 = \omega_2 - (\varepsilon_2 - \varepsilon_3)/\hbar$ and the recoil frequencies of the atom by $\omega_{R\mu} = \hbar k_{\mu}^2 / 2M$ ($\mu = 1,2$). Substitution of (7) and (8) into (4) gives

$$\sum_{\mu=1}^{2} A_{\mu}g_{\mu} \exp[(-1)^{\mu}i(\Omega_{\mu}t - \mathbf{k}_{\mu}\mathbf{r}) + i(\omega_{R\mu} - \hbar \mathbf{b}_{\mu}^{2}/2M)t + i\mathbf{b}_{\mu}\mathbf{r}] = 0, \quad A_{3} = 0.$$
(9)

Equations (9) for the amplitudes A_{μ} have a solution only when the oscillation frequencies of the summands in (9) are equal:

$$\Omega_1 - \omega_{R1} + \hbar \mathbf{b}_1^2 / 2M = -\Omega_2 - \omega_{R2} + \hbar \mathbf{b}_2^2 / 2M, \qquad (10)$$

$$\mathbf{k}_1 + \mathbf{b}_1 = -\mathbf{k}_2 + \mathbf{b}_2. \tag{11}$$

Conditions (10), (11) are satisfied for

$$\mathbf{b}_1 = \mathbf{b}_0 - \mathbf{k}_1,\tag{12a}$$

$$\mathbf{b}_2 = \mathbf{b}_0 + \mathbf{k}_2,\tag{12b}$$

$$\boldsymbol{\Omega}_1 - \mathbf{k}_1 \mathbf{v}_0 = -\,\boldsymbol{\Omega}_2 + \mathbf{k}_2 \mathbf{v}_0,\tag{13}$$

where the vector $\mathbf{v}_0 = \hbar \mathbf{b}_0 / \mathbf{M}$ is the velocity of the center of mass of the atom. Normalizing $|\Psi_{NC}\rangle$ by $\langle \Psi_{NC} ||\Psi_{NC}\rangle = 1$ we obtain $A_1 = g_2/g_0$, $A_2 = -g_1/g_0$, where $g_0^2 = |g_1|^2 + |g_2|^2$. Thus,

$$|\Psi_{\rm NC}(\mathbf{r})\rangle = \frac{g_2}{g_0} \exp[i(M\mathbf{v}_0/\hbar - \mathbf{k}_1)\mathbf{r}]|1\rangle$$
$$-\frac{g_1}{g_0} \exp[i(M\mathbf{v}_0/\hbar + \mathbf{k}_2)\mathbf{r}]|2\rangle. \tag{14}$$

In the momentum representation the wave function of the trap state is a superposition of the lowest ground state $|1\rangle$ and the highest excited one $|2\rangle$, each multiplied by a δ -function:

$$|\Psi_{\rm NC}(\mathbf{p})\rangle = \frac{g_2}{g_0} \,\delta(\mathbf{p} - (M\mathbf{v}_0 - \hbar\mathbf{k}_1))|1\rangle$$
$$-\frac{g_1}{g_0} \,\delta(\mathbf{p} - (M\mathbf{v}_0 + \hbar\mathbf{k}_2))|2\rangle. \tag{15}$$

Velocity selectivity of the trap state is provided by the condition (13) for Doppler-shifted two-photon resonance. Note that CPT in cascade scheme is velocity-selective only for co-propagating light waves in the case of $|\mathbf{k}_1| \cong |\mathbf{k}_2|$, in contrast to the Λ -scheme where laser cooling by VSCPT is usually realized by counterpropagating waves. In particular, the velocity-selective trap state $|\Psi_{NC}\rangle$ can arise when the Ξ -atom is excited by only one travelling laser wave. The problem is that such a trap state can not be populated by optical pumping cycles, so that actually VSCPT is not realized for the case of one exciting wave. This conclusion follows from the conditions under which CPT in the cascade scheme is established. We present and discuss these conditions in the next Section.

3. CONDITIONS FOR CPT IN CASCADE SYSTEMS

In order to describe the dynamics of the Ξ -atoms we use as a basis the formalism of the density matrix in the Wigner representation. This permits us to present both the full quantum and the quasiclassical (for translational motion of atoms) approaches to the problem of laser cooling by VSCPT. The relevant set of equations for the density matrix elements in the basis of the bare states $|1\rangle$, $|2\rangle$, $|3\rangle$ (Fig. 1) is given (in the rotating-wave approximation) by

$$\frac{d}{dt} \,\tilde{\rho}_{11}(\mathbf{r},\mathbf{p},t) = 2 \operatorname{Re}\left[ig_1\tilde{\rho}_{13}\left(\mathbf{r},\mathbf{p}+\frac{1}{2}\,\hbar\mathbf{k}_1,t\right)\right] \\ + 2\gamma_1 \int d\mathbf{n}\Phi_{31}(\mathbf{n})\rho_{33}\left(\mathbf{r},\mathbf{p}\right) \\ + \mathbf{n}\,\frac{\hbar\,\omega_{31}}{c},t \right], \\ \frac{d}{dt}\,\tilde{\rho}_{22}(\mathbf{r},\mathbf{p},t) = 2 \operatorname{Re}\left[ig_2\tilde{\rho}_{23}\left(\mathbf{r},\mathbf{p}-\frac{1}{2}\,\hbar\mathbf{k}_2,t\right)\right] \\ - 2\gamma_2\rho_{22}(\mathbf{r},\mathbf{p},t),$$

$$\begin{aligned} \frac{d}{dt} \,\tilde{\rho}_{33}(\mathbf{r},\mathbf{p},t) &= -2 \,\operatorname{Re} \bigg[ig_1 \tilde{\rho}_{13} \bigg(\mathbf{r},\mathbf{p} - \frac{1}{2} \,\hbar \mathbf{k}_1, t \bigg) \bigg] \\ &- 2 \,\operatorname{Re} \bigg[ig_2 \tilde{\rho}_{23} \bigg(\mathbf{r},\mathbf{p} + \frac{1}{2} \,\hbar \mathbf{k}_2, t \bigg) \bigg] \\ &- 2 \,\gamma_1 \rho_{33}(\mathbf{r},\mathbf{p},t) + 2 \,\gamma_2 \int d\mathbf{n} \Phi_{23}(\mathbf{n}) \rho_{22} \\ &\times \bigg(\mathbf{r},\mathbf{p} + \mathbf{n} \,\frac{\hbar \omega_{23}}{c}, t \bigg), \\ \frac{d}{dt} \,\tilde{\rho}_{13}(\mathbf{r},\mathbf{p},t) &= - (\gamma_1 + i\Delta_1) \tilde{\rho}_{13}(\mathbf{r},\mathbf{p},t) + ig_1^* \bigg[\tilde{\rho}_{11} \bigg(\mathbf{r},\mathbf{p} \\ &- \frac{1}{2} \,\hbar \mathbf{k}_1, t \bigg) - \tilde{\rho}_{33} \bigg(\mathbf{r},\mathbf{p} + \frac{1}{2} \,\hbar \mathbf{k}_1, t \bigg) \bigg] \\ &+ ig_2^* \tilde{\rho}_{12} \bigg(\mathbf{r},\mathbf{p} + \frac{1}{2} \,\hbar \mathbf{k}_2, t \bigg), \end{aligned}$$

$$\begin{aligned} \frac{d}{dt} \,\tilde{\rho}_{23}(\mathbf{r},\mathbf{p},t) &= - (\gamma_1 + \gamma_2 + i\Delta_2) \tilde{\rho}_{23}(\mathbf{r},\mathbf{p},t) \\ &+ ig_2^* \bigg[\tilde{\rho}_{22} \bigg(\mathbf{r},\mathbf{p} + \frac{1}{2} \,\hbar \mathbf{k}_2, t \bigg) - \tilde{\rho}_{33} \bigg(\mathbf{r},\mathbf{p} \\ &- \frac{1}{2} \,\hbar \mathbf{k}_2, t \bigg) \bigg] + ig_1^* \tilde{\rho}_{21} \bigg(\mathbf{r},\mathbf{p} - \frac{1}{2} \,\hbar \mathbf{k}_1, t \bigg), \end{aligned}$$

$$\begin{aligned} \frac{d}{dt} \,\tilde{\rho}_{12}(\mathbf{r},\mathbf{p},t) &= - [\Gamma + i(\Delta_1 - \Delta_2)] \tilde{\rho}_{12}(\mathbf{r},\mathbf{p},t) \\ &+ ig_2 \tilde{\rho}_{13} \bigg(\mathbf{r},\mathbf{p} - \frac{1}{2} \,\hbar \mathbf{k}_2, t \bigg) - ig_1^* \tilde{\rho}_{32} \bigg(\mathbf{r},\mathbf{p} \\ &+ \frac{1}{2} \,\hbar \mathbf{k}_1, t \bigg). \end{aligned}$$

Here we have used the notation

The functions $\Phi_{31}(\mathbf{n})$ and $\Phi_{23}(\mathbf{n})$ determine the relative probability of spontaneous photon emission in the direction of the unit vector \mathbf{n} for spontaneous atomic decay through the $|3\rangle$ $-|1\rangle$ and $|2\rangle-|3\rangle$ channels. However, for the analysis of one-dimensional VSCPT the spherical symmetry approximation is sufficient for these functions.^{3,17,18}

$$\Phi_{31}(\mathbf{n}) = \Phi_{23}(\mathbf{n}) \cong 1/4 \,\pi. \tag{17}$$

We denote by Γ the cumulative decay rate of the coherence ρ_{12} :

$$\Gamma = \gamma_2 + \Gamma_0, \tag{18}$$

where the value of Γ_0 includes collisional (Γ_{coll}), laser fluctuations (Γ_{fl}), and transit-time (Γ_{tr}) broadening and other factors destroying the laser-induced coherence.

If the trap state is absolutely stable against any relaxation process, then once the atomic population falls into the $|\Psi_{NC}\rangle$ state it remains in it forever. Thus, after a few fluorescent cycles a large fraction of atomic population will be trapped in $|\Psi_{NC}\rangle$.

The peculiarity of CPT in cascade scheme is that $|\Psi_{NC}\rangle$ is a superposition of the stable lower state $|1\rangle$ and the uppermost excited one $|2\rangle$, which decays to the intermediate state $|3\rangle$ with the rate $2\gamma_2$. Nevertheless, establishment of CPT in cascade scheme is possible, if the rate at which $|\Psi_{NC}\rangle$ fills is much higher than its relaxation rate. This requirement results in specific conditions on the laser field parameters and relaxation constants.

The conditions are analyzed by means of the density matrix equations in the basis of the states $|\Psi_{NC}\rangle$, $|\Psi_{C}\rangle$, $|3\rangle$ $(|\Psi_{C}\rangle$ is the 'coupled' state orthogonal to $|\Psi_{NC}\rangle$ and $|3\rangle$), so that the matrix element $\langle \Psi_{NC} | \tilde{\rho} | \Psi_{NC} \rangle$ is equal to the population of the trap state. CPT occurs in a quantum system when most of the population is in $|\Psi_{NC}\rangle$ in the steady state. Thus, solving the steady-state equations for the atoms with velocity satisfying Eq. (13) (e.g., $v = v_0 = 0$ for $\Omega_1 = -\Omega_2 \equiv \Omega$) we obtain $\langle \Psi_{NC} | \tilde{\rho} | \Psi_{NC} \rangle \approx 1$ under the following conditions:

$$\Gamma \ll \gamma_1, \tag{19a}$$

$$g_1^2 + g_2^2 \gg (\Gamma / \gamma_1) (\gamma_1^2 + \Omega^2),$$
 (19b)

$$g_2^2 \gamma_1 \gg g_1^2 \gamma_2. \tag{19c}$$

These conditions are in good agreement with the qualitative picture of filling the weakly-decaying state $|\Psi_{NC}\rangle$ by optical pumping through the intermediate state $|3\rangle$. Hence, the atomic population is largely trapped in $|\Psi_{NC}\rangle$ when the relaxation rate (which is proportional to Γ) of $|\Psi_{NC}\rangle$ is much lower than its pumping rate (which is proportional to γ_1 for strong saturation of optical transition and to $g_0^2 \gamma_1 / (\gamma_1^2 + \Omega^2)$ for weak saturation), resulting in the conditions (19a), (19b). The inequality (19c) reflects the asymmetry between the $|3\rangle$ $-|1\rangle$ and $|2\rangle - |3\rangle$ channels with respect to the filling of the $|\Psi_{NC}\rangle$ state, taking place because of the nonzero natural width $2\gamma_2$ of the state $|2\rangle$.

The conditions (19) impose specific limitations on the parameters of both the atomic systems and the applied radiation, allowing CPT to occur in the cascade scheme. The value of Γ_0 can be substantially reduced by the use of correlated laser fields.^{12,19,20} Note that for the cascade scheme $\Gamma_{\rm fl} = \bar{\Delta}_1 + \bar{\Delta}_2 + 2\bar{\Delta}_{12}$ (where $\bar{\Delta}_m$ is the bandwidth of a laser with frequency ω_m and $\bar{\Delta}_{12}$ is the cross-spectral bandwidth) is reduced with negative critical cross-correlation $\bar{\Delta}_1 = \bar{\Delta}_2 = -\bar{\Delta}_{12}$,¹² in contrast to the Λ -system where positive cross-correlation should be used.¹⁹ The value of $\Gamma_{\rm tr}$ can be reduced by preliminary deceleration of the atomic beam. However, the total magnitude of Γ (18) can not be as small as desirable because of the presence of the transverse damping rate γ_2 . Thus, it follows from (19a) and (18) that the natural width of the state $|2\rangle$ should be well below than that of the state $|3\rangle$:

$$\gamma_2 \ll \gamma_1. \tag{20}$$

There are two ways to satisfy the inequality (20). The first, "natural," way is to use atomic schemes where the $|3\rangle - |2\rangle$ transition is weak because it is an intercombinational or electric quadrupole, or magnetic dipole transition. In this paper we consider the example of alkaline-earth metals Mg, Ca, Zn, Cd, Hg, where the intercombinational optical transition ${}^{1}S_{0} - {}^{3}P_{1}$ corresponds to the $|1\rangle - |3\rangle$ transition, whereas the magnetic dipole microwave (infrared) transition ${}^{3}P_{1} - {}^{3}P_{2}$ corresponds to the $|3\rangle - |2\rangle$ one. In this scheme $\gamma_{1} = 10^{3} - 10^{7} \sec^{-1}$ and $\gamma_{2} = 10^{-3} - 10^{-1} \sec^{-1}$. The second, "artificial," way is to use the so-called combinational transition as the upper $|3\rangle - |2\rangle$ one, which becomes electric-dipole allowed only by applying an external dc electric field.²¹ The probability of such a transition is proportional to the squared electrical field strength, allowing the conditions (19), (20) to be satisfied by a proper choice of the field strength.

We note finally that the condition (19c) cannot be satisfied if both transitions are driven by one laser wave. One can reduce (19c) to the following form: $E_2^2/k_2^3 \ge E_1^2/k_1^3$ (where E_m and k_m are the amplitude and the wave number of the laser wave applied to the transition $|m\rangle - |3\rangle$), so that for one laser wave $(E_1 = E_2, k_1 = k_2)$ above condition is not satisfied.

4. QUASICLASSICAL APPROACH TO LASER COOLING BY VSCPT

VSCPT provides a mechanism for laser cooling which works rather effectively in both the sub-Doppler and subrecoil temperature ranges.^{1,2,16,22} The physical content of this mechanism can be described by the action of the light pressure force and momentum diffusion,^{4,7,8,16} exactly as for other cooling mechanisms. This fact is in accord with Bogolubov's well known conclusion on the simplification of statistical systems description as time increases (see, e.g. Ref. 17). In the case of laser cooling phenomena this is possible due to the large number of induced and spontaneous photons reemitted by atoms during the interaction. The most natural (and usual) way to study the dynamics of laser cooling is to use the quasiclassical approach, assuming the atomic translational motion to be the classical motion of Brownian particles. Unfortunately, such a classical treatment of atomic translational motion leads to some essential restrictions to the theory. First of all, the quasiclassical theory can not describe variations of momentum distribution function on a scale smaller than the photon momentum $\hbar k$, which is the measure of quantum fluctuations of atomic momentum. Therefore, in particular, the temperature limit of the approach is the recoil temperature T_R . However, quasiclassical analysis has the advantage of clarity of the physical picture of the cooling process. Besides, it permits one to study different regimes of the dynamics in a wide range of interaction parameters.

The characteristic feature of VSCPT is a sharp narrow dip, centered at the velocity v_0 (13), in the velocity dependence of the fluorescence rate of the trap state.^{16,22,23} This dip

is a consequence of destructive interference between the amplitudes of the $|1\rangle - |3\rangle$ and $|2\rangle - |3\rangle$ transitions which prevents the incident light being absorbed by atoms with velocity close to v_0 . Therefore the atoms are affected by radiation and change their translational and internal state as their velocities become weaker or closer to the value v_0 (or, in other words, the closer the atoms are to the 'bottom' of the velocity dip). At the same time, the atoms at the edges of the dip are affected most by light. For these reasons, one should consider the width δv_C of the dip as a "first velocity scale" or as the velocity capture range of laser cooling by VSCPT. It is therefore worthwhile to consider quasiclassically the evolution of the atomic velocity distribution, when both δv_C and the width δv_0 of the initial distribution are much larger than the recoil velocity $v_R = \hbar k/M$.

Quasiclassical analysis usually starts with the equations (16) for the Wigner density matrix $\tilde{\rho}(\mathbf{r},\mathbf{p},t)$. For times $t \ge \tau'$ (τ' is the time scale on which the atomic internal degrees of freedom change for Ξ -atoms it is $\tau' = \gamma_1^{-1}$ for large laser intensities $(g_0^2 \ge \gamma_1^2)$ and $\tau' = \gamma_1/g_0^2$ for $\gamma_1^2 \ge g_0^2 \ge \gamma_1 \Gamma^{14})$ one can reduce the density matrix equations to a single Fokker-Planck equation (FPE) for the atomic distribution function $w(\mathbf{r},\mathbf{p},t) = \rho_{11}(\mathbf{r},\mathbf{p},t) + \rho_{22}(\mathbf{r},\mathbf{p},t) + \rho_{33}(\mathbf{r},\mathbf{p},t)$, where \mathbf{r} and \mathbf{p} are the position and momentum of the atomic center of mass.^{16,17} Considering the one-dimensional problem of atomic motion along the z-axis and supposing that the distribution function w is spatially uniform, $w(\mathbf{r},\mathbf{p},t) = w(p_z,t)$, we write the FPE in the following form:

$$\frac{\partial w}{\partial t} = -\frac{\partial}{\partial p_z} \left(F_z w \right) + \frac{\partial^2}{\partial p_z^2} \left(D_{zz} w \right), \tag{21}$$

where p_z is the z-projection of the atomic center-of-mass momentum, F_z is the z component of the radiation force affecting the atoms and D_{zz} is the diagonal z component of the momentum diffusion tensor.

To derive the FPE we expand the Wigner density matrix elements in a power series in the small parameters $\hbar \mathbf{k}_{\mu}/\Delta \mathbf{p}$, where $\Delta \mathbf{p}$ is the characteristic momentum scale of the variation of the density matrix elements. In the presence of CPT, $\Delta \mathbf{p}$ is determined by the width $k_1 \delta \nu_c = \Gamma/2 + g_0^2/\gamma_1^{24,25}$ of the coherent trapping resonance: $|\Delta \mathbf{p}| = M \delta \nu_c \cong M(\Gamma/2 + g_0^2/\gamma_1)/k_1$. Therefore the condition for the applicability of expansion $(\hbar \mathbf{k}_{\mu}/\Delta \mathbf{p} \ll 1)$ limits the intensities for which the quasiclassical approach is valid:

$$g_0^2 \gg \omega_{R\mu} \gamma_{\mu},$$

where the condition (19b) is assumed to be fulfilled.

The light pressure force and the elements of the momentum diffusion tensor are determined by the steady-state populations ρ_{22}^0 and ρ_{33}^0 of the excited states in the Ξ -atom:

$$F_{z}(\nu) = \hbar k_{1} \gamma_{1} \rho_{33}^{0}(\nu) + \hbar k_{2} \gamma_{2} \rho_{22}^{0}(\nu), \qquad (22)$$

$$D_{zz}(\nu) = \hbar^2 k_1^2 \gamma_1 \rho_{33}^0(\nu) + \hbar^2 k_2^2 \gamma_2 \rho_{22}^0(\nu),$$

$$v = p_z / M.$$
 (23)

For simplicity we neglect in (23) the small nonadiabatic corrections related to the statistics of reemitted photons and suppose the photon emission to be isotropic.



FIG. 2. Time evolution of atomic velocity distribution w(v,t). Time is measured in units of ω_{R1}^{-1} ($\tau=t\omega_{R1}^{-1}$). Inset shows the velocity dependence of the radiation force F_z acting on Ξ -atom. Recoil frequency $\omega_{R1}=0.011\gamma_1$ (²⁰¹Hg), Rabi frequencies $g_1=g_2=0.3\gamma_1$, detuning $\Omega_1=\Omega_2=0$, transverse relaxation rate $\Gamma=0.001\gamma_1$, initial distribution width $k_1\delta v_0=0.5\gamma_1$, corresponding to the Doppler precooled atomic beam.

Explicit expressions for the velocity dependence of the force F_z and the diffusion coefficient D_{zz} are given in Appendix A. It is essential to note that both functions have the same shape, with a narrow sharp dip centered at the velocity v_0 (see formula (A1) and inset in Fig. 2). Force and diffusion coefficients of this shape are known^{16,22} to cause rapid formation of a narrow atomic velocity distribution peak near the velocity v_0 , i.e., atomic cooling. When this takes place, the diffusion plays a constructive role in the cooling process^{15,16} at the initial stage of atomic evolution in contrast to other cooling mechanisms (such as Doppler²⁶ or polarizationgradient²⁷ cooling) where diffusion broadens the atomic velocity distribution at any time. It is characteristic of the Ξ -scheme that the pure diffusion regime of cooling can not be recognized, as it can for Λ -scheme of interaction, since the Ξ -scheme is inherently asymmetric. Therefore the force affecting the Ξ -atoms under CPT conditions gives the main contribution to the cooling. 16,22 Computer calculations of the evolution of the velocity distribution of Hg atoms are presented in Fig. 2. It is seen that the atomic ensemble is effectively cooled in times of order $(1-10)\omega_{R1}^{-1}$, that is, about $(10^{-5}-10^{-4})$ sec for ²⁰¹Hg atoms cooled by CPT in the cascade scheme $6^{1}S_{0}-6^{3}P_{1}-6^{3}P_{2}$. This time corresponds to the time scale $\tau_{\rm fr} = \beta^{-1}$ for the action of the frictional force determined by the value of friction coefficient β : $M\beta = -(\partial F_z/\partial v)_{v=\langle v \rangle}$ for mean atomic velocity $\langle v \rangle$ close to zero.²²

However, the peak of the cold atoms can not be increased and narrowed indefinitely because of the incomplete trapping of population in the $|\Psi_{NC}\rangle$ state. The degree of the incompleteness of trapping is determined in general by the ratio Γ/γ (Section 3 of this paper; see also Ref. 16). With respect to the quasiclassical description of cooling by VSCPT, this fact is reflected by nonzero values of the radia-

tion force and diffusion coefficient at the velocity corresponding to the bottom of the dip in the velocity dependence (e.g. at $v = v_0 = 0$ for $\Omega_1 = \Omega_2 = 0$). Hence the peak of the cold atoms is shifted by a constant force $F_z^0(F_z^0 = F_z(v=0))$ for $\Omega_1 = \Omega_2 = 0$) and smeared out by the diffusion $D_{zz}^0(D_{zz}^0 = D_{zz}(v=0))$ for $\Omega_1 = \Omega_2 = 0$). One can estimate the time required for the atoms to be removed from the dip by the force F_z^0 as $\tau_F = (\Delta p) \cdot (F_z^0)^{-1}$ and by the diffusion D_{zz}^0 as $\tau_D = (\Delta p)^2 (D_{zz}^0)^{-1}$, where Δp is determined by the width of the dip: $\Delta p \cong Mg_0^2/(\gamma_1k_1)$. For the case when $|3\rangle - |2\rangle$ is a magnetic-dipole transition $(k_1 \ll k_1)$ these times are:

$$\tau_{F} = \frac{2g^{2}(g^{2} + (\gamma_{2}/4\gamma_{1})\Omega^{2})}{\omega_{R1}\gamma_{1}(g^{2}(2\Gamma + \gamma_{2}) + 2\Gamma(\gamma_{2}/\gamma_{1})\Omega^{2})},$$

$$\tau_{D} = \frac{4g^{4}(g^{2} + (\gamma_{2}/4\gamma_{1})\Omega^{2})}{\omega_{R1}^{2}\gamma_{1}^{2}(g^{2}(2\Gamma + \gamma_{2}) + \Gamma(\gamma_{2}/\gamma_{1})\Omega^{2})}.$$
 (24)

In the case of ²⁰¹Hg atoms the values of the time scales are $\tau_F \cong \tau_D \cong 10^{-3}$ sec for the parameters of Fig. 2. Thus, over the interaction times $\tau_{\rm fr} \le t \le \tau_D$, τ_F the atomic ensemble is cooled down to very low temperatures.

One of the most important questions of any cooling method is the cooling efficiency, which is generally specified: i) by the limiting value of the width of the velocity distribution of the cold atoms or by the 'temperature limit' reachable in a given method, ii) by the fraction of cold atoms with respect to initial number of atoms, and iii) by the density of cold atoms in velocity space. In order to improve the efficiency of laser cooling by VSCPT some methods of so-called 'pre-cooling' have been proposed^{4,28-30} The main principle is to supplement the VSCPT mechanism by a force accumulating atoms in the range of velocities near v_0 and to provide in this way a constant rate of pumping into the trap state. Quasiclassical analysis allows us to consider this problem in the simplest way and to propose new facilities that will considerably improve the cooling efficiency.

The above qualitative analysis and computer calculations (Fig. 2) clearly demonstrate that the narrow peak of 'cold' atoms is formed in the range of velocities close to v_0 , i.e., inside the CPT-dip. The number of cold atoms (calculated from the area of the peak) is determined by the relation between the initial distribution width δv_0 and the value of the dip width $\delta v_{\rm c}$. Under CPT conditions (19a) the width $\delta v_{\rm c}$ is determined only by the intensity of the applied fields. Increasing the laser intensity thus leads to an increase of the fraction of cooled atoms, accompanied by an increase in the width of the peak (Fig. 3). Hence it would be possible to accumulate most of the atoms in the narrow peak, if the laser intensity (which is proportional to g_0^2) is adiabatically decreased from the value $g_0^2 \cong \gamma_1 k_1 \delta v_0$ (so that initial CPT-dip width is of order of initial distribution width) to the value $g_0^2 \cong \gamma_1 \Gamma$ (the threshold value for CPT to be established).

Another effective method for intense one-dimensional cooling of the whole atomic ensemble (in most experimental realizations the one-dimensional cooling is the transverse cooling of an atomic beam) is proposed here on the basis of the following considerations. One of the characteristic features of cooling with VSCPT is that the sign of the light pressure force F_z is the same in the whole range of velocity



FIG. 3. Parameters of the cold atoms peak as a function of time $\tau = t \cdot \omega_{R1}^{-1}$ for different values of the Rabi frequencies $g = g_1 = g_2$. (a) The height of the peak (normalized to the height of initial distribution); (b) The width of the peak; (c) The fraction of atoms in the peak (normalized to the number of atoms in initial distribution). Recoil frequency $\omega_{R1} = 0.011 \gamma_1$ (²⁰¹Hg), detuning $\Omega_1 = \Omega_2 = 0$; transverse relaxation rate $\Gamma = 0.001 \gamma_1$.



FIG. 4. Two-zone scheme for the production of intensive well-collimated atomic beam.

variation. Therefore, decelerating the atoms with $v_z < 0$, the same force accelerates atoms with positive velocities, leading to a considerable loss in the final intensity of the atomic beam.

We consider here a technique which yields a collimated atomic beam almost without intensity loss. The production of such a beam is possible using the two-zone interaction scheme shown in Fig. 4. In zone A the frequency detuning of the exciting fields is shifted so that the narrow CPT resonance is in the region of positive velocities at $v_A = (\Omega_1 + \Omega_2)/(k_1 + k_2)$. As a result, narrow peak in the velocity distribution develops for $v \cong v_A$ (Fig. 5a). The peak accumulates the atoms with initial velocity $v_2 < v_A$. In order to concentrate the whole Maxwellian distribution in this peak, values $v_A \cong (2-3) \delta v_0$ are necessary. On the other hand, the atomic beam is now deflected after the interaction in the zone A. The narrow peak of atoms returns to the region of small transverse velocities in zone B. Here the atomic beam interacts with light fields, the wave vector directions of which are opposite to those of zone A, and the detunings are equal $(\Omega'_1 = -\Omega'_2 = \Omega)$. The light-pressure force F_z in zone B is negative and the CPT resonance is in the zero-velocity region (see inset of Fig. 5b). The effect of F_z shifts the velocity distribution peak formed in zone A to the zero-velocity region and further narrows it. As can be seen from Fig. 5b, almost all the atoms of the beam are concentrated in a narrow velocity region with the width $\Delta v \cong 0.01(\gamma_1/k_1)$ (corresponding to ≈ 2 cm/s for Hg) near v = 0.

Note also that the momentum diffusion slightly affects the implementation of this technique, since the transit time of atoms through zones A and B is small, so that the diffusive broadening of the transverse velocity distribution is slight.

Concluding this Section we point out that in the quasiclassical description of laser cooling the final temperature of the cooled atoms can be evaluated by the Einstein formula for Brownian motion. We recall that the quasiclassical approach is valid in the temperature range $T \gg T_R$, while the mechanism of laser cooling by VSCPT provides temperatures well below T_R . In order to obtain an explicit expression for the temperature of VSCPT-cooled atoms one should solve the full quantum equations (16) rather then the Fokker–Planck equation (21). This is what we do in the next Section.



FIG. 5. Time evolution of the atomic velocity distribution w(v,t) for the two-zone laser-atom interaction scheme. (a) Interaction in zone A: wave vectors $\mathbf{k}_m = k_m \mathbf{e}_z$, $k_m > 0$; detunings $\Omega_1 = \Omega_2 = 0.5 \gamma_1$; (b) Interaction in zone B: wave vectors $\mathbf{k}_m = -k_m \mathbf{e}_z$, $k_m > 0$; detunings $\Omega_1 = \Omega_2 = 0$. Insets show the velocity dependence of the radiation force F_z acting on the Ξ atom (upper right corner) and the time dependence of the number of cold atoms (normalized to the number of atoms in initial distribution). Recoil frequency $\omega_{R1} = 0.011 \gamma_1$ (²⁰¹Hg), Rabi frequencies $g_1 = g_2 = 0.3 \gamma_1$, transverse relaxation rate $\Gamma = 0.001 \gamma_1$, and initial distribution width $k_1 \delta v_0 = 0.5 \gamma_1$, corresponding to the Doppler precooled atomic beam.

5. ASYMPTOTIC BEHAVIOR AND THE LIMIT OF SUBRECOIL COOLING BY VSCPT

As we pointed out before, there is an evident shortcoming of the quasiclassical theory: it is impossible to describe structures in the atomic momentum distribution narrower than the photon momentum $\hbar k$. However, VSCPT is known to permit the atoms to be cooled below the recoil limit¹ in such a way that the width of the distribution is decreased to very low magnitudes when the laser-atom interaction time is increased. Therefore the asymptotic behavior of the momentum distribution, which is of great importance in the theory of cooling by VSCPT, should be studied without making the approximation that the atomic translational motion is classical. The aim of this Section is to study the asymptotic evolution of the atomic density matrix. In order to obtain the full quantum solution to the problem under consideration, we introduce new definitions for the density matrix elements:

$$\sigma_{11}(\mathbf{p},t) = \rho_{11} \left(\mathbf{r}, \mathbf{p} - \frac{1}{2} \hbar(\mathbf{k}_{1} + \mathbf{k}_{2}), t \right),$$

$$\sigma_{13}(\mathbf{p},t) = \rho_{13} \left(\mathbf{r}, \mathbf{p} - \frac{1}{2} \hbar \mathbf{k}_{2}, t \right),$$

$$\sigma_{22}(\mathbf{p},t) = \rho_{22} \left(\mathbf{r}, \mathbf{p} + \frac{1}{2} \hbar(\mathbf{k}_{1} + \mathbf{k}_{2}), t \right),$$

$$\sigma_{23}(\mathbf{p},t) = \rho_{23} \left(\mathbf{r}, \mathbf{p} + \frac{1}{2} \mathbf{k}_{1}, t \right),$$

$$\sigma_{33}(\mathbf{p},t) = \rho_{33} \left(\mathbf{r}, \mathbf{p} + \frac{1}{2} \hbar(\mathbf{k}_{1} - \mathbf{k}_{2}), t \right),$$

$$\sigma_{12}(\mathbf{p},t) = \rho_{12}(\mathbf{r}, \mathbf{p}, t).$$
(25)

After this substitution, which corresponds to a transition to the "closed family" basis,³ one gets from (16) the following set of equations for $\sigma_{ij}(\mathbf{p},t)$:

$$\frac{d}{dt} \sigma_{11}(\mathbf{p},t) = 2 \operatorname{Re}[ig_{1}\sigma_{13}(\mathbf{p},t)] + 2\gamma_{1} \int d\mathbf{n} \Phi_{31}(\mathbf{n}) \sigma_{33} \Big(\mathbf{p} - \hbar \mathbf{k}_{1} + \mathbf{n} \frac{\hbar \omega_{31}}{c}, t \Big), \frac{d}{dt} \sigma_{22}(\mathbf{p},t) = 2 \operatorname{Re}[ig_{2}\sigma_{23}(\mathbf{p},t)] - 2\gamma_{2}\sigma_{22}(\mathbf{p},t), \frac{d}{dt} \sigma_{33}(\mathbf{p},t) = -2 \operatorname{Re}[ig_{1}\sigma_{13}(\mathbf{p},t)] - 2 \operatorname{Re}[ig_{2}\sigma_{23}(\mathbf{p},t)] - 2\gamma_{1}\sigma_{33}(\mathbf{p},t) + 2\gamma_{2} \int d\mathbf{n} \Phi_{23}(\mathbf{n}) \sigma_{22} \Big(\mathbf{p} - \hbar \mathbf{k}_{2} + \mathbf{n} \frac{\hbar \omega_{23}}{c}, t \Big), \frac{d}{dt} \sigma_{13}(\mathbf{p},t) = -[\gamma_{1} + i(\Delta_{1} + \tilde{\omega}_{R})]\sigma_{13}(\mathbf{p},t) + ig_{1}^{*}[\sigma_{11}(\mathbf{p},t) - \sigma_{33}(\mathbf{p},t)] + ig_{2}^{*}\sigma_{12}(\mathbf{p},t),$$

$$\frac{d}{dt} \sigma_{23}(\mathbf{p},t) = -[\gamma_1 + \gamma_2 + i(\Delta_2 + \tilde{\omega}_R)]\sigma_{23}(\mathbf{p},t)$$
$$+ ig_2^*[\sigma_{22}(\mathbf{p},t) - \sigma_{33}(\mathbf{p},t)]$$
$$+ ig_1^*\sigma_{21}(\mathbf{p},t),$$

$$\frac{d}{dt} \sigma_{12}(\mathbf{p},t) = -[\Gamma + i(\Delta_1 - \Delta_2)]\sigma_{12}(\mathbf{p},t)$$
$$+ ig_2\sigma_{13}(\mathbf{p},t) - ig_1^*\sigma_{32}(\mathbf{p},t), \qquad (26)$$

where $\tilde{\omega}_R = \hbar(\mathbf{k}_1 \mathbf{k}_2 / 2M)$.

Because of the substitution (25) the distribution function $w(\mathbf{r},\mathbf{p},t)$ of atoms is expressed in the "closed family" basis as (see also Ref. 3):

$$w(\mathbf{r},\mathbf{p},t) = \sigma_{11} \left(\mathbf{r},\mathbf{p} + \frac{\hbar}{2} (\mathbf{k}_1 + \mathbf{k}_2), t \right) + \sigma_{22} \left(\mathbf{r},\mathbf{p} - \frac{\hbar}{2} (\mathbf{k}_1 + \mathbf{k}_2), t \right) + \sigma_{33} \left(\mathbf{r},\mathbf{p} - \frac{\hbar}{2} (\mathbf{k}_1 - \mathbf{k}_2), t \right).$$
(27)

Therefore it is necessary to calculate the time and momentum dependence of the diagonal elements $\sigma_{ii}(p,t)$ in order to investigate the dynamics of atomic cooling. We recall that due to (15) the atoms being cooled by VSCPT, are accumulated in the superposition of the states $|1\rangle$ and $|2\rangle$ localized in momentum space in the two planes $\mathbf{p}=M\mathbf{v}_0-\hbar\mathbf{k}_1$ and $\mathbf{p}=M\mathbf{v}_0+\hbar\mathbf{k}_2$. This is the reason why we, anticipating the cooling by VSCPT, are interested mostly in the behavior of the diagonal elements $\sigma_{11}(p,t)$ and $\sigma_{22}(p,t)$ in the range of momenta close to $\mathbf{p}=M\mathbf{v}_0$ [in particular, close to p=0 for $\Omega_1=-\Omega_2$; see (13)].

In the last few years some analytical^{10,18,23} and numerical³¹ techniques for solving the problem of cooling by VSCPT in the limit of large interaction time have been developed. Here we use an approach based on that of Refs. 10 and 18. We were forced, however, to modify this method substantially because of the strong asymmetry of the cascade scheme, so that the solution algorithm was totally changed. In the present form this technique can be used for a number of different laser-atom interaction schemes.

An exact algorithm for the asymptotic solution of Eqs. (26) is given in Appendix B. Thus, we obtain the following formula describing populations for small values of atomic momentum and for large interaction time:

$$\sigma_{11}(p,t) = \sigma_{22}(p,t) = \frac{1}{\pi} \frac{(\delta p)}{p^2 + (\delta p)^2} \frac{(k_1 + k_2)}{k_1} \frac{g}{\omega_{R12}} \\ \times \sqrt{\frac{3(\Gamma + \gamma_2)}{16\gamma_1}} \exp\left(-\frac{1}{2}(\Gamma + \gamma_2)t\right) \\ \times I_0\left(\frac{1}{2}(\Gamma - \gamma_2)t\right), \qquad (28)$$
$$\delta p = \hbar(k_1 + k_2) \frac{g}{\omega_{R12}} \sqrt{\frac{\Gamma + \gamma_2}{2\gamma_1}}, \\ \omega_{R12} = \hbar(k_1 + k_2)^2/2M, \qquad (29)$$

for the case of equal frequency detuning $\Omega_1 = -\Omega_2 \equiv -\tilde{\omega}_R$ and Rabi frequencies $g_1 = g_2 \equiv g$. In deriving (28) we assume the conditions (19) of CPT in Ξ -system to be fulfilled.

Equation (28) shows that the momentum distribution peaks at time $t \ge \Gamma^{-1}$ have Lorentzian shape. The time dependence of the distribution is described by an exponential de-

crease with the rate $(\Gamma + \gamma_2)/2$, combined with slow increase defined by the modified Bessel function $I_0((\Gamma - \gamma_2)t/2)$.³²

The half-width δp (29) of the peak is constant in time. This is in contrast to the conclusions of Refs. 3,18,23, where the relaxation of the coherence σ_{12} is not taken into account, thus resulting in a $t^{-1/2}$ dependence of the width. The relative number of atoms N involved in the cooling process is given by $N = \int \sigma_{11}(p) dp$, where the integral is taken over the peak of the trapped atoms only. From (28) it is easy to find that

$$N = \frac{(k_1 + k_2)g}{k_1 \omega_{R12}} \sqrt{\frac{3(\Gamma + \gamma_2)}{16\gamma_1}} \times \exp\left[-\frac{1}{2}(\Gamma + \gamma_2)t\right] I_0\left(\frac{1}{2}(\Gamma - \gamma_2)t\right).$$
(30)

It is interesting that neither the half-width of the peaks nor the number of cold atoms depends on the initial atomic distribution width in agreement with the conclusions of other studies of the asymptotic behavior of the process.^{10,18,23,31} The reason is that the atomic distribution is divided into two parts during cooling by VSCPT: an ensemble of 'cold' atoms and another one of 'hot' atoms, with substantially different temporal evolution (this is clearly seen from Fig. 2; see also discussions in Refs. 9, 15, and 18). In the initial stage the narrow peaks of cold atoms are formed and the remaining (which we call 'hot') part of the distribution is a reservoir for the process of accumulation of atoms in $|\Psi_{NC}\rangle$. At the same time, atoms from the 'hot' ensemble diffuse in momentum space to the range of large momenta as well. Therefore, beginning at a certain time (estimates of its value are presented in^{15}) the two parts of the atomic distribution are more and more weakly coupled to each other as the interaction time increases, so that for large (asymptotic) times their mutual influence is negligible.

Note that the atomic momentum distribution given by Eq. (28) does not permit us to calculate the mean value of p^2 , because the integral $\int_{-\infty}^{\infty} p^2 \sigma_{11}(p) dp$ diverges. This difficulty arises because of the asymptotic meaning of (28), where p is assumed to be small. The "wings" of the atomic momentum distribution, of course, decrease more rapidly than p^{-2} at large p. But the half-width (29) of the distribution can be associated with the effective temperature of the Ξ atoms cooled by VSCPT:

$$T = \frac{(\Gamma + \gamma_2)g^2}{2\gamma_1 \omega_{R12}^2} T_R, \quad T_R = \frac{\hbar \omega_{R12}}{k_{\rm B}}.$$
 (31)

Thus, the temperature can be much less than the recoil limit for a wide range of laser intensities. The process of CPT establishment has a threshold (19b): the saturation parameter g^2/γ_1^2 has to be larger than the small quantity Γ/γ_1 to allow CPT to occur. Therefore the lowest temperature of cold atoms that can be achieved by VSCPT method is

$$T_{\min} = \frac{(\Gamma + \gamma_2)\Gamma}{2\omega_{R12}^2} T_R, \qquad (32)$$

and the minimum achievable half-width of the momentum distribution is, accordingly,

$$\delta p_{\min} = \hbar (k_1 + k_2) \sqrt{\frac{(\Gamma + \gamma_2)\Gamma}{2\omega_{R12}^2}}.$$
(33)

Note that the formulas (29), (31)–(33) agree very well with those derived in the Introduction from qualitative considerations. In the case of alkaline-earth atoms cooled by CPT in the ${}^{1}S_{0}-{}^{3}P_{1}-{}^{3}P_{2}$ cascade scheme one can estimate the limiting values of temperature as 10^{-15} K, corresponding to an atomic distribution with velocity spread (in one dimension) of 10^{-5} cm/sec and a minimum wave-packet width of 0.05 cm for 201 Hg, 0.1 cm for 40 Ca, and 0.3 cm for 24 Mg at $\Gamma=10$ sec ${}^{-1}$.

On the whole, the asymptotic behavior of the momentum distribution of the cooled Ξ -atoms is similar to that of a symmetric Λ -system when relaxation of the coherence σ_{12} with the rate Γ is introduced.¹⁰ The narrow peaks of the distribution, formed at the early stage of evolution, is smeared out so that width of a narrow peak is constant as the number of cooled atoms decreases with time. The values of δp and N depend on the value of Γ in such a way that the smaller the value of Γ the more efficient the cooling process. Note that the values of both δp and N are proportional to the Rabi frequency. It is interesting also that the related frequency parameter for the cooling of Ξ -atoms is the 'sum' recoil frequency $\omega_{R12} = \hbar (k_1 + k_2)^2 / 2M$ rather than the individual ones ω_{R1} and ω_{R2} .

6. CONCLUSIONS

We have investigated the feasibility of one-dimensional laser cooling of atoms by the use of VSCPT in the three-level $(\Xi$ -) cascade interaction scheme. We have demonstrated that the temperature of cooled Ξ -atoms can reach values much lower than the recoil limit obtained for interaction schemes with the trap state superposition composed only of stable ground states. However, to implement VSCPT in a cascade system, one has to satisfy specific conditions (19) imposed on both the atomic level scheme and the laser radiation parameters. The conditions are due to restrictions induced by the presence of spontaneous decay of the upper state $|2\rangle$ involved to the trap state $|\Psi_{\rm NC}\rangle$ as well as by relaxation of coherence between states $|1\rangle$ and $|2\rangle$. These processes destroy the superposition $|\Psi_{\rm NC}\rangle$ and modify the dynamics of cooling as compared with cooling by VSCPT in the case of an absolutely stable state $|\Psi_{NC}\rangle$.³ The evolution of the atomic momentum distribution consists of the rapid formation of narrow peaks of cold atoms, followed by a slow decrease in their height and the number of cold atoms. In the asymptotic limit of large interaction times the width of the peaks reaches, a constant value. Both the time scales of the evolution and the limiting value of the width are determined by the relaxation rates of the $|2\rangle$ state and of coherence between the $|1\rangle$ and $|2\rangle$ states. It is important that under the conditions (19) of CPT the atoms are cooled down to subrecoil temperatures for rather a long time. We have also demonstrated, using the quasiclassical approach, some schemes of interaction permitting a substantial improvement in the effectiveness of the cooling.

Thus, we have shown that effective subrecoil cooling in one dimension can be achieved by the use of VSCPT in cascade systems where one atomic transition is optical while the second one can be in the optical or the microwave (infrared) band. As an example, we have considered in this paper the subrecoil cooling of the alkaline-earth atoms Ca, Mg, Zn, Cd, Hg, which have attracted attention in recent years because of their application to high-quality infrared frequency standards (see, e.g., Ref. 33 and references therein). We believe that the cascade scheme of laser-atom interaction can be preferable for subrecoil cooling of some other types of atoms and possibly molecules.

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APPENDIX A

In this Appendix we present exact expressions for the force F_z and diffusion coefficient D_{zz} in the quasiclassical approach:

$$\begin{split} F_z &= \hbar k_1 \gamma_1 a B_F L^{-1}, \\ D_{zz} &= \hbar^2 k_1^2 \gamma_1 a B_D L^{-1}, \\ a &= \alpha^2 + (4\Gamma/\gamma_1) g^2, \\ B_{F,D} &= 2g^2 \, \frac{2\Gamma + \gamma_2 \theta_{F,D}}{\gamma_1} + \frac{2\Gamma \gamma_2}{\gamma_1^2} \, \Omega^2 \theta_{F,D} - \frac{2\gamma_2}{\gamma_1} \, \Omega \alpha \\ &+ \left(1 + \frac{\gamma_2}{\gamma_1} \, \frac{\Omega^2}{g^2}\right) \alpha^2 + \frac{2\gamma_2}{\gamma_1} \, \frac{\Omega}{g^2} \, q_2 \alpha^3 + \frac{\gamma_2}{\gamma_1} \, \frac{q_2^2}{g^2} \, \alpha^4, \\ \theta_F &= 1 + k_2/k_1, \quad \theta_D &= 1 + k_2^2/k_1^2, \\ L &= \frac{16\Gamma}{\gamma_1} \, g^2 \left(g^2 + \frac{\gamma_2}{4\gamma_1} \, \Omega^2\right) + \frac{8\Gamma}{\gamma_1} \, g^2 \Omega \, \alpha + 4 \left(g^2 + \frac{2(\Gamma + \gamma_2)}{\gamma_1} \, \Omega^2 + \frac{\Gamma \gamma_2}{2\gamma_1^2} \, \frac{\Omega^4}{g^2}\right) \alpha^2 + 2\Omega \left(1 - \frac{2\Gamma \gamma_2}{\gamma_1^2} \, \frac{\Omega^2}{g^2} \, (q_1 - q_2)\right) \alpha^3 + 2 \left(q_2 + \frac{\gamma_1^2 + 4\Omega^2}{8g^2} + \frac{\gamma_2}{2\gamma_1} \, \frac{\Omega^4}{g^4}\right) \alpha^4 - \frac{2\Omega}{g^2} \left(q_1 + \frac{\gamma_2}{\gamma_1} \, \frac{\Omega^2}{g^2} \, (q_1 - q_2)\right) \alpha^5 \\ &+ \frac{1}{g^2} \left(1 + \frac{\gamma_2}{\gamma_1} \, \frac{\Omega^2}{g^2} \, (1 - 8q_1q_2)\right) \alpha^6 \\ &+ \frac{\gamma_2}{\gamma_1} \, \frac{2\Omega}{g^4} \, q_1 q_2 (q_1 - q_2) \alpha^7 + \frac{\gamma_2}{\gamma_1} \, \frac{q_1^2 q_2^2}{g^4} \, \alpha^8, \\ q_j &= k_j / (k_1 + k_2), \quad (j = 1, 2), \end{split}$$

$$\alpha = (k_1 + k_2)v. \tag{A1}$$

Here $v = p_z/M$ is the z-projection of the atomic velocity. For simplicity the detuning and the Rabi frequency are taken the same for both transitions in the Ξ -atom: $\Omega_1 = -\Omega_2 \equiv \Omega$ and $g \equiv g_1 = g_2$. In deriving (A1) we also assume the conditions (19) for coherent population trapping in the Ξ -system to be fulfilled.

APPENDIX B

Here we present the asymptotic solution of the set of equations (26). First, the Laplace transformation is taken:

$$\sigma_{\mu\nu}(p,s) = \int_0^\infty dt \exp(-st) \sigma_{\mu\nu}(p,t)$$

with the initial conditions

$$\sigma_{11}(\mathbf{p},t=0) = W_1(\mathbf{p}), \quad \sigma_{22}(\mathbf{p},t=0) = W_2(\mathbf{p}), \quad (B1)$$

while the other initial density matrix elements are equal to zero.

In the one-dimensional approximation the equations for the Laplace transforms read as follows:

$$s\sigma_{11}(p,s) - W_{1}(p) = 2 \operatorname{Re}[ig_{1}\sigma_{13}(p,s)] + \frac{\gamma_{1}}{\hbar k_{1}}$$

$$\times \int_{-\hbar k_{1}}^{+\hbar k_{1}} du\sigma_{33}(p - \hbar k_{1} + u, s),$$

$$s\sigma_{22}(p,s) - W_{2}(p) = 2 \operatorname{Re}[ig_{2}\sigma_{23}(p,s)]$$

$$-2\gamma_{2}\sigma_{22}(p,s),$$

$$s\sigma_{33}(p,s) = -2 \operatorname{Re}[ig_{1}\sigma_{13}(p,s)] - 2 \operatorname{Re}[ig_{2}\sigma_{23}(p,s)]$$

$$-2\gamma_{1}\sigma_{33}(p,s) + \frac{\gamma_{2}}{\hbar k_{2}} \int_{-\hbar k_{2}}^{+\hbar k_{2}} du\sigma_{22}(p)$$

$$-\hbar k_{2} + u, s),$$

$$s\sigma_{13}(p,s) = -[\gamma_{1} + i(\Delta_{1} + \tilde{\omega}_{R})]\sigma_{13}(p,s)$$

$$+ ig_{1}^{*}[\sigma_{11}(p,s) - \sigma_{33}(p,s)]$$

$$+ ig_{2}^{*}\sigma_{12}(p,t),$$

$$s\sigma_{23}(p,s) = -[\gamma_{1} + \gamma_{2} + i(\Delta_{2} + \tilde{\omega}_{R})]\sigma_{23}(p,s)$$

$$+ ig_{1}^{*}\sigma_{21}(p,s),$$

$$s\sigma_{12}(p,s) = -[\Gamma + i(\Delta_{1} - \Delta_{2})]\sigma_{12}(p,s) + ig_{2}\sigma_{13}(p,s)$$

$$- ig_{1}^{*}\sigma_{32}(p,s),$$
(B2)

where the integration over the direction **n** of spontaneous decay in the spontaneous feeding terms for $\sigma_{11}(p,s)$ and $\sigma_{22}(p,s)$ is changed by the integration over the spontaneous photon momentum *u* along the *z*-axis and the approximation (17) is used. Here *p* is the *z*-projection of the momentum **p**, and we consider the case when both e.m. waves propagate

along the positive z-axis: $\mathbf{k}_1 = k_1 \mathbf{e}_z$, $\mathbf{k}_2 = k_2 \mathbf{e}_z$, $k_{\mu} > 0$. From the fourth, fifth and sixth equations of (B2) one can write

2 Re
$$(ig_1\sigma_{13}) = a_1\sigma_{11} + a_2\sigma_{22} + a_3\sigma_{33},$$

2 Re $(ig_2\sigma_{23}) = b_1\sigma_{11} + b_2\sigma_{22} + b_3\sigma_{33}.$ (B3)

Throughout this article we consider the case of laser intensities, weakly saturating atomic transitions: $g_{\mu}^2/(\gamma_{\mu}^2 + \Omega_{\mu}^2) \ll 1$, so that at any time $\sigma_{33} \ll \sigma_{11}$, σ_{22} holds and one can write $\sigma_{11} - \sigma_{33} \cong \sigma_{11}$, $\sigma_{22} - \sigma_{33} \cong \sigma_{22}$. Therefore the coefficients a_{μ} , b_{μ} for interaction times $t \gg \gamma_1^{-1}$ are given by:

$$a_{1} = \operatorname{Re}[-2g_{1}^{2}(\varepsilon_{2}\Gamma_{p} + g_{1}^{2})D^{-1}],$$

$$a_{2} = b_{1} = \operatorname{Re}(2g_{1}^{2}g_{2}^{2}D^{-1}),$$

$$b_{2} = \operatorname{Re}[-2g_{2}^{2}(\varepsilon_{1}\Gamma_{p} + g_{2}^{2})D^{-1}],$$

$$a_{3} = b_{3} = 0,$$

$$\varepsilon_{1} = \gamma_{1} + i(\Delta_{1} + \tilde{\omega}_{R}),$$

$$\varepsilon_{2} = \gamma_{1} + \gamma_{2} - i(\Delta_{2} + \tilde{\omega}_{R}),$$

$$\Gamma_{p} = s + \Gamma + i(\Delta_{1} - \Delta_{2}),$$

$$D = \varepsilon_{1}g_{1}^{2} + \varepsilon_{2}g_{2}^{2} + \varepsilon_{1}\varepsilon_{2}\Gamma_{p}.$$
(B4)

After inserting (B3) into the first three equations of (B2), for the function $\sigma_2(p,s)$ defined as

$$\sigma_{2}(p,s) \equiv \sigma_{22}(p,s)\beta(p,s),$$

$$\beta(p,s) \equiv \frac{s-a_{1}}{b_{1}} (s+2\gamma_{2}-b_{2})-a_{2},$$
 (B5)

we get the integral equation:

$$\sigma_{2}(p,s) - W_{1}(p) = \frac{\gamma_{1}}{\hbar k_{1}(s+2\gamma_{1})} \int_{-\hbar k_{1}}^{+\hbar k_{1}} du_{1}$$

$$\times \left\{ \frac{\gamma_{2}}{\hbar k_{2}} \int_{-\hbar k_{2}}^{+\hbar k_{2}} du_{2} \right\}$$

$$\times \left[\frac{\sigma_{2}(p-\hbar k_{1}+u_{1}-\hbar k_{2}+u_{2},s)}{\beta(p-\hbar k_{1}+u_{1}-\hbar k_{2}+u_{2},s)} \right]$$

$$- \frac{\sigma_{2}(p-\hbar k_{1}+u_{1},s)}{\beta(p-\hbar k_{1}+u_{1},s)} \left[\frac{a_{1}+b_{1}}{b_{1}} (s+2\gamma_{2}-b_{2}) + a_{2}+b_{2} \right]_{p=p-\hbar k_{1}+u_{1}}, \quad (B6)$$

while the Laplace transform $\sigma_{11}(p,s)$ of the population of lower state $|1\rangle$ is expressed in terms of $\sigma_{22}(p,s)$ as

$$\sigma_{11}(p,s) = \frac{s+2\gamma_2 - b_2}{b_1} \,\sigma_{22}(p,s),\tag{B7}$$

where we assume for the sake of simplicity that there is no initial population in the uppermost state $|2\rangle$: $W_2(p)=0$.

The next step in obtaining the asymptotic solution to the set of equations (26) is the Fourier transformation of (B6):

$$f(x) \equiv \tilde{f}(p) \equiv \int_{-\infty}^{+\infty} e^{ipx/\hbar} f(p) dp.$$

Introducing the operators $\hat{S}_1(x)$, $\hat{S}_2(x)$, $\hat{K}_1(x,s)$, $\hat{K}_2(x,s)$, which act on arbitrary function $\eta(x)$ so that

$$\hat{S}_{1}(x) \eta(x) = \int_{-\infty}^{+\infty} e^{ipx/\hbar} \left(\int_{-\hbar k_{1}}^{\hbar k_{1}} du \right) \times \eta(p - \hbar k_{1} + u) dp,$$

$$\hat{S}_{2}(x) \eta(x) = \int_{-\infty}^{+\infty} e^{ipx/\hbar} \left(\int_{-\hbar k_{2}}^{\hbar k_{2}} du \right) \times \eta(p - \hbar k_{2} + u) dp,$$

$$\hat{K}_{1}(x,s) \eta(x) = \int_{-\infty}^{+\infty} e^{ipx/\hbar} \frac{\eta(p)}{\beta(p,s)} dp,$$

$$\hat{K}_{2}(x,s) \eta(x) = \int_{-\infty}^{+\infty} e^{ipx/\hbar} \eta(p) \left[\frac{a_{1} + b_{1}}{b_{1}} (s + 2\gamma_{2} - b_{2}) \right]$$

$$+a_2+b_2 dp, \tag{B8}$$

we obtain from (B6) an equation for the Fourier transform $\sigma_2(x,s)$:

$$\sigma_{2}(x,s) - W_{1}(x) = \frac{\gamma_{1}}{\hbar k_{1}(s+2\gamma_{1})} \hat{S}_{1}(x) \left[\frac{\gamma_{2}}{\hbar k_{2}} \\ \times \hat{S}_{2}(x) \hat{K}_{1}(x,s) \sigma_{2}(x,s) \\ - \hat{K}_{1}(x,s) \hat{K}_{2}(x,s) (\sigma_{2}(x,s)) \right].$$
(B9)

The solution to (B9) can be written as the operator relation

$$\sigma_2(x,s) = \hat{L}(x,s)W_1(x) \tag{B10}$$

with the operator $\hat{L}(x,s)$ defined by

$$\hat{L}(x,s) = \left[1 - \frac{\gamma_1}{\hbar k_1 (s+2\gamma_1)} \,\hat{S}_1 \left[\frac{\gamma_2}{\hbar k_2} \,\hat{S}_2 \hat{K}_1 - \hat{K}_1 \hat{K}_2\right]\right]^{-1}.$$
(B11)

Thus, the solution to (B2) is expressed as

$$\sigma_{22}(p,s) = J(p,s)\phi_2(p,s),$$

$$\sigma_{11}(p,s) = J(p,s)\phi_1(p,s),$$
(B12)

where

$$J(p,s) = \frac{1}{2\pi\hbar} \int_{-\infty}^{+\infty} e^{-ipx/\hbar} \hat{L}(x,s) W_1(x) dx$$
(B13)

and functions $\phi_1(p,s), \phi_2(p,s)$ are

It is easy to obtain explicit forms of the operators $\hat{S}_1(x)$ and $\hat{S}_2(x)$, just by changing the order of integration in (B8):

$$\hat{S}_{1}(x) = 2\hbar k_{1} e^{ik_{1}x} \frac{\sin(k_{1}x)}{k_{1}x},$$
$$\hat{S}_{2}(x) = 2\hbar k_{2} e^{ik_{2}e} \frac{\sin(k_{2}x)}{k_{2}x}.$$
(B15)

As for the operators $\hat{K}_1(x,s)$, $\hat{K}_2(x,s)$, one can find only their asymptotic representation.

We investigate now the dynamics of atomic populations in the asymptotic limit of long interaction times $t \to \infty$ (corresponding to $s \to 0$ [10]) and small atomic momenta $p \to 0$ (corresponding to $x \to \infty$). Thus, we see that $\hat{S}_1(x)|_{\substack{x\to\infty\\(p\leqslant\hbar k_1)}} \to 0$ and $\hat{S}_2(x)|_{\substack{x\to\infty\\(p\leqslant\hbar k_2)}} \to 0$ for the case when both k_1 and k_2 belongs to the optical frequency range. For the case when k_2 belongs to the microwave range, we have $(\gamma_2/\hbar k_2)\hat{S}_2\hat{K}_1|_{x\to\infty} \to 0$, since the value of γ_2 is very small. The latter expression corresponds just to neglect of the 'feeding-by-spontaneous-relaxation' term for $\sigma_{33}(p,s)$ in Eq. (B.2). Therefore in both cases one can write the asymptotic (for small values of atom momenta) expression for the operator $\hat{L}(x,s)$:

$$\hat{L}(x,s)|_{x\to\infty} \cong \left[1 + \frac{\gamma_1}{\hbar k_1(s+2\gamma_1)} \hat{S}_1 \hat{K}_{10} \hat{K}_{20}\right]^{-1}, \quad (B16)$$

where \hat{K}_{10} , \hat{K}_{20} denote the asymptotic forms of the operators $\hat{K}_1(x,s)$, $\hat{K}_2(x,s)$. From (B8) one can easily find

$$\hat{K}_{10} = \hat{K}_1 \Big|_{\substack{x \to \infty \\ (p \to 0)}} = \beta^{-1}(p,s) \Big|_{p=0},$$

$$\hat{K}_{20} = \hat{K}_2 \Big|_{\substack{x \to \infty \\ (p \to 0)}} = \left[\frac{a_1 + b_1}{b_1} \left(s + 2\gamma_2 - b_2 \right) + a_2 + b_2 \right]_{p=0}.$$
(B17)

Thus the asymptotic form of the operator $\hat{L}(x,s)$ is

$$\hat{L}(x,s)|_{x \to \infty} \cong \left\{ 1 + \frac{2\gamma_1}{(s+2\gamma_1)} e^{ik_1 x} \times \frac{\sin(k_1 x)}{k_1 x} \beta^{-1}(p,s) \Big|_{p=0} \left[\frac{a_1 + b_1}{b_1} \times (s + 2\gamma_2 - b_2) + a_2 + b_2 \right] \Big|_{p=0} \right\}^{-1},$$
(B18)

so that $\hat{L}(x,s)|_{x\to\infty}$ is equivalent to the multiplication operator.

For small values of momenta one can approximate the integral J(p,s) in (B13) by its asymptotic function J(p = 0,s). At the same time, the functions $\phi_1(p,s), \phi_2(p,s)$ tend to nonzero functions of p for $s \rightarrow 0$. Thus, retaining only

the leading functions, one can separate the variables p and s in expressions (B12) for $\sigma_{11}(p,s)$ and $\sigma_{22}(p,s)$, factoring them in the asymptotic limit:

$$\sigma_{22}(p,s) \Big|_{\substack{p \to 0 \\ s \to 0}} = J(p=0,s) \phi_2(p,s=0),$$

$$\sigma_{11}(p,s) \Big|_{s \to 0}^{p \to 0} = J(p=0,s) \phi_1(p,s=0).$$
 (B19)

Setting

$$\Theta(s) \equiv \beta^{-1}(p,s)|_{p=0} \left[\frac{a_1 + b_1}{b_1} \left(s + 2\gamma_2 - b_2 \right) + a_2 + b_2 \right]|_{p=0},$$
(B20)

and supposing the initial distribution to be Maxwellian, $W(p) = \exp(-p^2/2 \, \delta p^2)$ so that $W(x) = \exp(-Cx^2)$, C > 0, we obtain for $J(p=0,s)|_{s\to 0}$ from (B13)

$$J(p=0,s)|_{s\to 0} = \frac{1}{2\pi\hbar} \int_{-\infty}^{\infty} \frac{\exp(-Cx^2)dx}{1+\exp(ik_1x)\Theta(x)[\sin(k_1x)/k_1x]} \approx \frac{1}{2\pi\hbar[1+\Theta(s)]} \int_{-\infty}^{\infty} \frac{dx}{1-(k_1x)^2\Theta(s)/6[1+\Theta(s)]} = \frac{\sqrt{3/2}}{\hbar k_1\sqrt{1+\Theta(s)}\sqrt{-\Theta(s)}}.$$
 (B21)

In deriving (B21) we have taken into account that the func-

tion $W(x) = \exp(-Cx^2)$ is negligible except for x close to zero, thus acting as a filter in x space. This enables us to consider the integrand only for $x \rightarrow 0$.

Now we find, assuming $\Omega_1 = -\Omega_2 = -\tilde{\omega}_R$, that under the conditions of CPT (19) and of weak saturation, $g_{\mu}^2/(\gamma_{\mu}^2 + \Omega_{\mu}^2) \ll 1$, the functions for the asymptotic solution are

$$\phi_{1}(p,s=0) = g_{0}^{6} \gamma_{1}^{3} (\gamma_{1} \gamma_{2} g_{0}^{2} + g_{2}^{4}) Q^{-1},$$

$$\phi_{2}(p,s=0) = g_{1}^{2} g_{2}^{2} g_{0}^{6} \gamma_{1}^{3} Q^{-1},$$

$$Q = 2g_{1}^{2} g_{0}^{6} \gamma_{1}^{3} (\gamma_{2} g_{1}^{2} + \Gamma g_{2}^{2}) + 2g_{1}^{2} g_{0}^{2} \gamma_{1}^{4} (\gamma_{1} \gamma_{2} (g_{0}^{2} + g_{1}^{2}) + g_{2}^{2} g_{0}^{2}) \alpha^{2} + 2g_{1}^{2} g_{2}^{2} \gamma_{1}^{6} \alpha^{4},$$
(B22)

$$\alpha = (k_1 + k_2) p / M,$$

$$\Theta(s) = \frac{g_1^2 \{ [g_2^2 - g_1^2 - \gamma_1(s + \Gamma)](s + 2\gamma_2) - 2g_2^2(s + \Gamma) \}}{2g_1^2 g_2^2(s + \Gamma) + g_1^4(s + 2\gamma_2)g_2^4 s},$$
(B23)

and the integral $J(p=0,s)|_{s\to 0}$ takes the form

$$J(p=0,s)|_{s\to 0} = \sqrt{\frac{3}{2}} \frac{1}{\hbar k_1 \sqrt{(1+\Theta(s))} \sqrt{-\Theta(s)}}$$

= $\sqrt{\frac{3}{2}} \frac{1}{\hbar k_1} \frac{2g_1^2 g_2^2 (s+\Gamma) + g_1^4 (s+2\gamma_2) + g_2^4 s}{g_1 \sqrt{[-g_2^2+g_1^2+\gamma_1(s+\Gamma)]} (s+2\gamma_2) + 2g_2^2 (s+\Gamma)} \sqrt{g_2^4 s + g_1^2 (s+2\gamma_2) [g_2^2-\gamma_1(s+\Gamma)]}}.$ (B24)

Thus, inserting (B24) and (B22) into the asymptotic representation (B19) of $\sigma_{ii}(p,s)$, after the corresponding inverse Laplace transformation one obtains formulas for the populations of the Ξ -atoms. In order to simplify the analysis it is convenient to use the expressions for $J(p=0,s)|_{s\to 0}$ and $\phi_i(p,s=0)$ for different limiting cases. For example, in the case of equal Rabi frequencies $g_1 = g_2 \equiv g$

$$J(p=0,s)|_{s\to 0} \approx \sqrt{\frac{3}{2}} \frac{\Gamma + \gamma_2}{\hbar k_1 \sqrt{(s+\gamma_2)(s+\Gamma)}}$$
(B25)

and, taking reciprocal Laplace transformation, one obtains

$$\sigma_{11}(p,t) = \sigma_{22}(p,t) = \frac{1}{\pi} \frac{(\delta p)}{p^2 + (\delta p)^2} \frac{(k_1 + k_2)}{k_1} \frac{g}{\omega_{R12}} \\ \times \sqrt{\frac{3(\Gamma + \gamma_2)}{16\gamma_1}} \exp\left(-\frac{1}{2} (\Gamma + \gamma_2)t\right) \\ \times I_0\left(\frac{1}{2} (\Gamma - \gamma_2)t\right), \qquad (B26)$$

where $I_0(\zeta)$ is the modified Bessel function of order zero and

$$\delta p = \hbar (k_1 + k_2) \frac{g}{\omega_{R12}} \sqrt{\frac{\Gamma + \gamma_2}{2\gamma_1}},$$

$$\omega_{R12} = \hbar (k_1 + k_2)^2 / 2M.$$
(B27)

The case of laser fields with substantially different intensities gives for $g_2^2 \gg g_1^2$

 $J(p=0,s)\big|_{s\to 0}$

$$\approx \sqrt{\frac{3}{2}} \frac{2\Gamma(g_1/g_2)}{\hbar k_1 \sqrt{(s+2\gamma_2(g_1/g_2)^2](s+2\Gamma-2\gamma_2)]}}$$
(B28)

and

$$\sigma_{11}(p,t) = \frac{1}{\pi} \frac{(\delta p)^2}{p^2 + (\delta p)^2} \frac{g_2}{g_1} \frac{\sqrt{3}/2}{\hbar k_1} \exp\left[-\left(\Gamma - \gamma_2 + \frac{g_1^2}{g_2^2} \gamma_2\right)t\right] I_0 \left[\left(\Gamma - \gamma_2 - \frac{g_1^2}{g_2^2} \gamma_2\right)t\right],$$

$$\sigma_{22}(p,t) = \frac{1}{\pi} \frac{(\delta p)^2}{p^2 + (\delta p)^2} \frac{g_1}{g_2} \frac{\sqrt{3}/2}{\hbar k_1} \exp\left(-\left(\Gamma - \gamma_2 + \frac{g_1^2}{g_2^2} \gamma_2\right)t\right),$$

$$+ \frac{g_1^2}{g_2^2} \gamma_2 t\right) I_0 \left(\left(\Gamma - \gamma_2 - \frac{g_1^2}{g_2^2} \gamma_2\right)t\right),$$

$$\delta p = \hbar (k_1 + k_2) \frac{g_2}{\omega_{B12}} \sqrt{\frac{\Gamma}{4\gamma_1}}.$$
 (B29)

In the opposite case of $g_1^2 \gg g_2^2 \gg \Gamma \gamma_1$ we have

$$J(p=0,s)|_{s\to 0} \approx \sqrt{\frac{3}{2}} \frac{(s+2\gamma_2)(g_1/g_2) + 2\Gamma(g_2/g_1)}{\hbar k_1(s+2\gamma_2)}$$
(B30)

and

$$\sigma_{11}(p,t) = \frac{(\delta p)^2}{p^2 + (\delta p)^2} \frac{\Gamma \sqrt{3/2}}{\hbar k_1} \frac{g_2(g_2^4 + \gamma_1 \gamma_2 g_1^2)}{g_1^3(\Gamma g_2^2 + \gamma_2 g_1^2)}$$

$$\times \exp(-2\gamma_2 t),$$

$$\sigma_{22}(p,t) = \frac{(\delta p)^2}{p^2 + (\delta p)^2} \frac{\Gamma \sqrt{3/2}}{\hbar k_1} \frac{g_2^3}{g_1(\Gamma g_2^2 + \gamma_2 g_1^2)}$$

$$\times \exp(-2\gamma_2 t),$$

$$\delta p = \hbar (k_1 + k_2) \frac{g_1}{g_2 \omega_{R12}} \sqrt{\frac{\Gamma g_2^2 + \gamma_2 g_1^2}{4\gamma_1}}.$$
(B31)

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