Influence of radiative interatomic collisions on dark-state cooling

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We discuss the role of light absorption by pairs of atoms (radiative collisions) in the context of a model for velocity selective coherent population trapping with emphasis on its application to metastable triplet helium. Our main interest is the application of such dark-state cooling schemes to the regime where individual quantum states get multiply occupied, a regime which is sometimes referred to with the term atom laser. We show that, because of radiative collisions, for positive detuning of the driving light fields from an atomic resonance the conditions for proliferation of the occupation numbers of the lowest lying states can only be marginally met. Multiple occupation of these states only occurs if a very efficient sub-Doppler precooling mechanism is operative. In the case of negative frequency detuning the requirements on this sub-Doppler mechanism are less restricting, provided one avoids molecular resonances. [S1050-2947(96)07211-3]

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I. INTRODUCTION

The investigation of macroscopic quantum phenomena is one of the prime motivations for the study of ultracold atomic gases. Recent successful experiments on Bose-Einstein condensation (BEC) in trapped rubidium [1], lithium [2], and sodium [3] gave a tremendous boost to this field of research. The breakthrough leading to the achievement of BEC was the implementation of evaporative cooling schemes, where cooling and thermal quasiequilibrium are provided by interatomic elastic collisions. It is important that interaction between atoms also governs the formation kinetics of a Bose condensate [4].

A principal question with regard to quantum statistical effects concerns the possibility to reach large or even macroscopic occupation numbers for a single-particle state in a collisionless Bose gas, where cooling and phase-space compression proceed through interaction of atoms with light rather than through interatomic collisions. The central idea is to exploit the bosonic nature of the particles: once the occupation number of a single-particle state becomes larger than unity, this enhances the rate at which the state is filled and strongly influences the population dynamics [5,6]. This is equivalent to the gain mechanism in lasers. Accordingly, the term ''atom laser'' [7–10] has been coined to describe a gas far from thermal equilibrium, with atoms accumulating in a single quantum state or at least in a very small region of phase space. In this paper we will use the term atom laser in this sense, namely, to indicate the fact that low-lying atomic states acquire potentially large occupation numbers through radiative processes rather than elastic interatomic collisions. We will refer to such a gas as being in the collisionless regime, meaning that ordinary elastic collisions are essentially absent and the states are not necessarily occupied according to a thermal distribution.

Optical cooling of a gas in the collisionless regime requires the condition

$$n\lambda\!\!\!^{-3} \ll 1, \tag{1}$$

where n is the gas density and $2\pi\lambda\!\!\!^{-}$ the wavelength of light. In the opposite limiting case the evolution of excited atomic states is mainly governed by interatomic collisions induced by resonance dipole interaction [11]. These collisions proceed at a rate much larger than the rate of spontaneous emission and destroy cooling. In view of Eq. (1), to achieve occupation numbers of the order of unity or larger the atoms should gather in a momentum range smaller than the single photon recoil. Cooling schemes based on dark states, such as velocity selective coherent population trapping (VSCPT) [12–14] or schemes involving Raman pumping between different hyperfine states [15] can be used for this purpose. Whatever the scheme chosen, it will necessarily involve laser fields which drive the pumping, and hence spontaneous emission which, under certain conditions, can lead to compression of atoms in momentum space. Reabsorption of spontaneously emitted photons, destroying the momentum-space compression, can be, hopefully, circumvented by selecting a high-frequency detuning of driving light [7] or by choosing at least one of the sample dimensions smaller than the mean free path of a photon. Such geometrical means of reducing the effect of multiple reabsorption were successfully exploited in the optical cooling of atomic hydrogen [16] and have been discussed in the context of atom lasers [17].

Exchange of longitudinal virtual photons between excited and ground-state atoms leads to resonance dipole interaction. The influence of the mean field of this interaction on VSCPT cooling was found small under the condition Eq. (1) [18]. We will consider the case where the frequency detuning δ of the laser fields from resonance with an excited state is large compared to the natural line width Γ of this state. In this case resonance dipole interaction manifests itself in interatomic pair collisions (see, e.g., [19]). At interparticle distances where resonance dipole interaction compensates the frequency detuning a colliding pair is resonant with the light. This is the origin of the well known process of light absorption in pair collisions, which we will refer to as radiative collisions. Of particular interest is the limit of ultracold collisions, reviewed in, e.g., Ref. [20].

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At ultralow temperatures corresponding to the s-wave scattering limit, under the condition of Eq. (1) the rate of radiative collisions is normally much smaller than the rate of absorption of light by single atoms. The situation changes drastically for atoms in so-called dark states which are decoupled from the driving light field. In this case the coupling of the dark-state atoms with the driving light is induced by their radiative collisions with the atoms in coupled states. This process, unlike reabsorption of spontaneously emitted photons, can not be eliminated by arranging the sample geometry. In this paper we argue that for $|\delta| \gg \Gamma$ at any gas density the phase-space compression to occupation numbers larger than unity can be frustrated by this type of radiative collisions. The physical reason that these collisions can be important even in a very dilute gas is the following: If we consider a small sphere of radius $\tilde{p}$ around $p=0$ in momentum space, the filling rate of this momentum region from the gas cloud of spatial density n is roughly proportional to $[1+n(\tilde{p})]\tilde{p}^3 n$, where $n(\tilde{p})$ is the characteristic occupation number of momentum states in the small sphere. The collisional loss of atoms from this sphere is proportional to $n(\tilde{p})\tilde{p}^3 n$. The ratio of filling to loss rate, being large for small occupation numbers, decreases with increasing $n(\tilde{p})$ and tends to a constant for $n(\tilde{p})>1$. The key point is that both the filling rate and the rate of radiative collisions depend on the degree of excitation of the atoms in a similar way. However, the phase space available for the filling is ultimately set by the momentum change in the last spontaneous emission event, i.e., the photon momentum, whereas the phase space available for collisions can be much larger. In view of this the question whether radiative collisions can prevent the occupation numbers from reaching unity is principal.

We will analyze a simple general model for an atom laser which includes radiative collisions and apply the results to VSCPT cooling on a $J=1$ to $J=1$ transition, successfully used to cool helium atoms in the metastable $2\,^3S_1$ state to below the recoil energy [12–14]. For this scheme we discuss both the case of large positive and large negative detuning and show that the latter case is more promising for realizing an atom laser.

II. GENERAL SCHEME

We consider a general scheme for an atom laser, such as presented in Ref. [7]. This scheme is depicted in Fig. 1. All atoms are confined to a volume much larger than the optical wavelength and such that the eigenstates of the translational motion of the atoms are (approximate) momentum eigenstates. We have two sets of atomic states called the system (labeled s) and bath (labeled b). The system s comprises just the atoms in the compressed momentum space described above. The laser fields are tuned close to a resonance involving an excited atomic state e, which optically pumps the atoms from the bath into the system. In Fig. 1 the pumping scheme is schematically represented as a single transition connecting the bath states to the excited state followed by spontaneous emission into the system states, although in practice the pumping scheme can be more complicated. Spontaneous emission makes the atom end up in a system state with momentum $\mathbf{p}$, chosen so to say ''by chance'' as a result of the random direction of the emitted photon. As the focus of this paper is the role of radiative collisions with bath atoms, we omit all other potential loss mechanisms (in contrast to, e.g., Ref. [7]) such as decay of the system states due to absorption of spontaneously emitted photons. Then the rate equation for the occupation numbers $n_s(\mathbf{p})$ of the system states with momentum $\mathbf{p}$ takes the form:

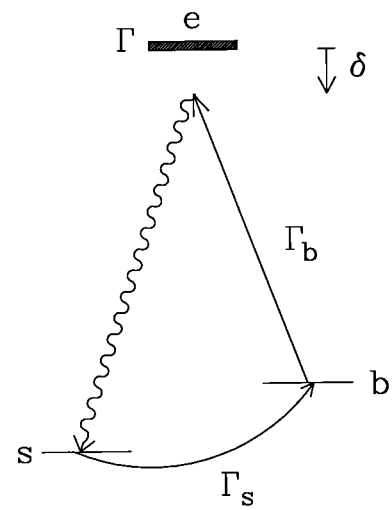

FIG. 1. Schematic depiction of the atom laser model. The bath and the system states are denoted b and s, respectively. The pumping rate from the bath states is Γ_b and the repumping from the system back into the bath is denoted by Γ_s. We assume that Γ_s vanishes for zero momentum. The wiggly line represents a spontaneously emitted photon. The excited state involved in the process is denoted as e and its inverse lifetime is Γ.

$$\dot{n}_s(\mathbf{p}) = -\Gamma_s(\mathbf{p})n_s(\mathbf{p}) + [1+n_s(\mathbf{p})] \times \int \frac{d\mathbf{q}}{8\pi^3} n_b(\mathbf{q})\Gamma_b(\mathbf{q})P(\mathbf{p},\mathbf{q}) - n_s(\mathbf{p})\int \frac{d\mathbf{q}}{8\pi^3} G(\mathbf{p},\mathbf{q})n_b(\mathbf{q}). \tag{2}$$

The first term denotes the loss rate of isolated atoms in the system due to the presence of the light fields. It involves optical pumping from the system states back into the bath. Clearly it is advantageous if the lifetime of systems states increases with decreasing momentum. We will assume that this repumping rate vanishes quadratically with momentum for small p: $\Gamma_s(\mathbf{p}) = \tilde{\Gamma}(p/p_*)^2$. Such a quadratic dependence is naturally encountered in schemes based on VSCPT or on velocity selective Raman pumping. The region $p<p_*$ can be called a trap in momentum space. The second term in Eq. (2) is the pumping rate from bath states into the system. Here $\Gamma_b(\mathbf{q})$ is the probability per unit time that an atom leaves the bath state having momentum $\mathbf{q}$, $n_b(\mathbf{q})$ is the occupation number of the corresponding bath state, and $P(\mathbf{p},\mathbf{q})$ is the probability density for a bath atom with momentum $\mathbf{q}$ to end up in a system state with momentum $\mathbf{p}$ after a spontaneous emission event. The prefactor $1+n_s(\mathbf{p})$ in the second term is the Bose enhancement factor which is responsible for the ''lasing'' process. The third term in Eq. (2) is the focus of this paper. It describes the absorption and subsequent reemis-

sion of a photon from the laser fields in a collision between the system atom and the bath atom. As the photon absorption strongly changes the relative motion of colliding atoms, such radiative collisions will be a loss mechanism for system atoms trapped in the space of low momenta $p<p_*$. For large frequency detuning δ the rate constant of radiative collisions $G(\mathbf{p},\mathbf{q})$ becomes momentum independent and can be written as $G=\beta\tilde{\Gamma}\lambda\!\!\!^{-3}$. The cubic dependence on $\lambda\!\!\!^{-}$, and the proportionality to $\tilde{\Gamma}$ are a consequence of the resonant dipole interaction. The coefficient β depends on δ, the Rabi frequency and details of the level structure.

In view of Eq. (1) it is natural to assume that the momentum trap size $p_*<k$, where k is the photon momentum. In order to simplify the picture we describe the bath by introducing a sphere p_{max} in momentum space, with the bath occupation numbers independent of $\mathbf{p}$ for $p<p_{max}$ and zero otherwise. Also Γ_b is momentum independent. Accordingly, $n_b(\mathbf{q})\equiv n_b\Lambda_b^3$, where n_b is the real space density of bath atoms, and their De Broglie wavelength $\Lambda_b=(6\pi^2)^{1/3}/p_{max}$. In a nonthermal gas, the kinetic energy of bath atoms is maintained by a dissipative optical cooling mechanism and thus higher than the recoil energy, i.e., $p_{max}>k$. Since the momentum change during spontaneous emission is of order $\mathbf{k}$ and $p_*<k$, the integral in the second term of Eq. (2) can be written as $\alpha\Gamma_b n_b\Lambda_b^3$, where α is a numerical coefficient which depends on the cooling scheme and the level structure of the atoms. With $\Gamma_b\approx\tilde{\Gamma}$, the rate equation (2) now reduces to:

$$\dot{n}_s(\mathbf{p})=\tilde{\Gamma}\left[-\left(\frac{p}{p_*}\right)^2 n_s(\mathbf{p})+\alpha n_b\Lambda_b^3[1+n_s(\mathbf{p})]-\beta n_s(\mathbf{p})n_b\lambda\!\!\!^{-3}\right]. \tag{3}$$

We introduce the parameter $\eta\equiv(\alpha\Lambda_b^3)/(\beta\lambda\!\!\!^{-3})$. Apart from the ratio α/β, the parameter η essentially denotes the ratio of the recoil energy $E_r=k^2/2m$ ($\hbar$ is set equal to unity throughout) to the bath "temperature" $T_b\sim 2\pi/m\Lambda_b^2$. As we necessarily have $T>E_r$ ($\Lambda_b<\lambda\!\!\!^{-}$) we may expect η to be less than unity. For $\eta<1$ we find a steady state solution for the occupation numbers in the trap:

$$n_s(\mathbf{p})=\eta/[(p/p_*)^2/\beta n_b\lambda\!\!\!^{-3}+(1-\eta)]. \tag{4}$$

The maximum occupation number is achieved for $p\to 0$. It is smaller than unity unless η is very close to 1. The fraction of particles accumulated in the momentum-space trap is of the order of $(p_*/p_{max})^3\ll 1$, hence for $\eta<1$ the atoms predominantly remain in bath states.

In the case $\eta>1$ there is no steady state solution. The occupation numbers of states with $(p/p_*)^2<(\eta-1)\beta n_b\lambda\!\!\!^{-3}$ grow exponentially, with a characteristic inverse growth time $\tilde{\Gamma}(\eta-1)\beta n_b\lambda\!\!\!^{-3}-\tilde{\Gamma}(p/p_*)^2$. We have an atom laser. There is no threshold due to the fact that we omitted all loss mechanisms except radiative collisions. Ultimately the bath will be depleted and the above approximations break down. Clearly, two criteria have to be met in order to make the atom laser work: in order to have $\eta>1$ the prefactor β which governs radiative collisions should be much smaller than α and the bath temperature should be kept as close to E_r as possible.

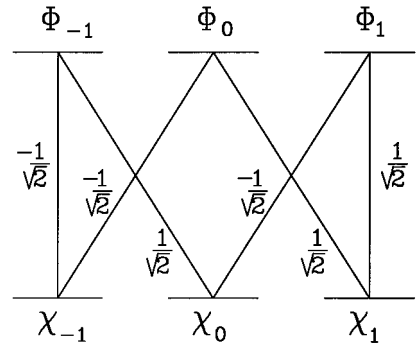

FIG. 2. Level scheme involved in VSCPT cooling of He*. The lower manifold is $2\,^3S_1$, the upper is $2\,^3P_1$. The numbers are the nonzero Clebsch-Gordan coefficients.

III. APPLICATION TO He*

We will apply the general scheme for the atom laser described above to the case of VSCPT cooling of He in the metastable 2^3S state (He*). In Fig. 2 we show the relevant levels involved in VSCPT cooling of He*. For simplicity we will first consider a one-dimensional picture and later generalize it to 3D. The model in the previous section relates only to the 3D case. The 1D calculation presented here is not meant as a 1D variant of this model but serves only to obtain numerical results which we will show to be independent of the dimension and which we will subsequently use in the generalization to 3D.

In the 1D VSCPT case the sample is irradiated with a σ_+ and a σ_- polarized beam propagating in the positive and negative z direction, respectively. The Hamiltonian of interaction of an isolated atom with the light field has six eigenstates. Optical pumping ensures that after a comparatively short time only three of the states in Fig. 2 remain populated [12]. In the absence of light the wave functions of two of these states can be written in the form:

$$\chi_{c,u}(\mathbf{p})=\frac{1}{\sqrt{2}}[\chi_1\exp\{i(\mathbf{p}+\mathbf{k})\cdot\mathbf{R}\}\pm\chi_{-1}\exp\{i(\mathbf{p}-\mathbf{k})\cdot\mathbf{R}\}]. \tag{5}$$

Here the labels c and u stand for coupled and uncoupled states, the plus sign relating to $\chi_c(\mathbf{p})$ (using the phase convention of Ref. [12]). The atom coordinate and momentum are $\mathbf{R}$ and $\mathbf{p}$, and χ_M is the wave function of the $2\ ^3S$ atomic state with spin projection M on the direction of light propagation, which we select as quantization axis. The state χ_0 in Fig. 2 is depopulated by optical pumping. The states χ_1 and χ_{-1} are coupled by the σ_- and σ_+ beams, respectively, to the excited $2\,^3P_1$ state ϕ_0, with zero projection of the total electron angular momentum. The wave function of this state can be written as $\phi_0(\mathbf{p})=\phi_0\exp(i\mathbf{p}\cdot\mathbf{R})$.

The state $\chi_u(\mathbf{p})$ is called uncoupled because in the limit of $p\to 0$ it is completely decoupled from the driving light fields. The rates Γ_c and Γ_u at which $\chi_c(\mathbf{p})$ and $\chi_u(\mathbf{p})$ scatter photons are given by [12]:

$$\Gamma_c = (\Omega^2/2)\frac{\Gamma}{\delta^2+\Gamma^2/4}, \tag{6}$$

$$\Gamma_u(p) = \begin{cases} (kp/m)^2 \dfrac{\Gamma}{2\Omega^2}; & p \lesssim p_*, \\ \Gamma_c; & p \gtrsim p_*, \end{cases} \tag{7}$$

where $p_* = \Omega^2 m/k\delta$. The Rabi frequency is defined as $\Omega = dE$, where d is the dipole moment of the $2\,^3S - 2\,^3P_1$ atomic transition and E the electric field amplitude for each beam. The scattering rate for atoms in the state $\chi_u(\mathbf{p})$ is proportional to p^2 for small momenta. Hence we can identify the states $\chi_u(\mathbf{p})$ for $p < p_*$ as our system states. The states $\chi_c(\mathbf{p})$, as well as the states $\chi_u(\mathbf{p})$ with $p > p_*$, can be considered as comprising the bath. To complete the correspondence with Eqs. (2) and (3) we note that the excited state decays into χ_1 and χ_{-1} (and hence into χ_c and χ_u) with equal probability. Accordingly, the coefficient $\alpha = 1/2$ (see also Ref. [12]).

Light absorption in pair interatomic collisions requires at least one of the colliding atoms to be in a coupled state, since for a pair of atoms both in uncoupled states the resonance dipole interaction is practically absent. Therefore, the collisional loss term for the system atoms in Eq. (2) will be proportional to the occupation number $n_c(k)$ of the coupled states with momenta around k. We defined our bath as containing both coupled and uncoupled states, but only the coupled part contributes to the radiative collisions. Except for very small momenta, the time scale on which the populations change is long compared to the optical pumping time, therefore detailed balance ensures that the ratio of the occupation numbers of coupled and uncoupled states satisfies the condition

$$n_c(p)\Gamma_c(p) = n_u(p)\Gamma_u(p). \tag{8}$$

Hence, as $\Gamma_c = \Gamma_u = \tilde{\Gamma}$ for $p > p_*$, we can express the decay of $n_s(p)$ in terms of the bath occupation numbers by substituting $n_c(p) = n_b(p)/2$.

The effect of radiative collisions can not be reduced by relaxing the above assumption $p_* < k$. Let us demonstrate this for the extreme case, where $p_* \gtrsim p_{max}$. We still assume $\Omega \ll \delta$. In this case it is more natural to define the system states as uncoupled states in a small momentum range $p < \tilde{p}$ near zero ($\tilde{p} < k$). Then, for bath states we have $\Gamma_u(q) \approx \Gamma_c(q/p_*)^2$ and, hence, most of the atoms will be pumped into the uncoupled state [see Eq. (8)]. As only the population of the coupled states contributes to the collisional loss of system atoms, the rate of radiative collisions involving coupled-state atoms with momentum q is reduced by a factor $\sim(q/p_*)^2$ compared to the case $p_* < k$. On the other hand, the optical pumping rate is reduced by the same factor. This is again clear from Eq. (8) which shows that the optical pumping rate from coupled states should be exactly half the total pumping rate from coupled and uncoupled states, just as we found above for $p_* < k$.

IV. RADIATIVE COLLISIONS: POSITIVE DETUNING

Let us now consider light absorption in pair collisions of atoms in the uncoupled state $\chi_u(\mathbf{p})$ with atoms in the coupled state $\chi_c(\mathbf{p}')$. We will first discuss the case of positive frequency detuning δ, where the light is at resonance with continuum states of the excited quasimolecule. The Hamiltonian of resonance dipole interaction for a pair of atoms labeled by (1) and (2) is given by

$$\hat{V} = \frac{(\hat{\mathbf{d}}^{(1)}\hat{\mathbf{d}}^{(2)})r^2 - 3(\hat{\mathbf{d}}^{(1)}\mathbf{r})(\hat{\mathbf{d}}^{(2)}\mathbf{r})}{r^5}, \tag{9}$$

where $\hat{\mathbf{d}}^{(1)}$ and $\hat{\mathbf{d}}^{(2)}$ are the dipole moment operators of the colliding atoms, and $\mathbf{r}$ the vector of interparticle separation. Under the condition $\delta \gg \Gamma = 4d^2/3\lambda\!\!\!^{-3}$ radiative transitions predominantly occur at interparticle distances $r \ll \lambda\!\!\!^{-}$ where the resonance dipole interaction $V \propto d^2/r^3$ compensates the frequency detuning. At such distances we can omit the factors $\exp[i(\mathbf{p}+\mathbf{k})\cdot\mathbf{R}]$ and $\exp[i(\mathbf{p}-\mathbf{k})\cdot\mathbf{R}]$ in the expressions for $\chi_{c,u}(\mathbf{p})$, and the initial-state wave function of a colliding pair takes the form

$$\Psi_i = \hat{P}_g \chi_u^{(1)} \chi_c^{(2)}, \tag{10}$$

where $\hat{P}_g$ is the symmetrization operator with respect to interchange of electrons and their inversion. The index g shows that the initial electronic state of the quasimolecule is *gerade*. The two atoms forming the pair are labeled by the superscripts (1) and (2).

Excited quasimolecular states to which radiative transitions occur are *ungerade*. For $\delta > 0$ the quasimolecule formed in the light absorption process corresponds to repulsive potential of interaction. Diagonalizing the Hamiltonian of resonance dipole interaction Eq. (9) we find five such states:

$$\tilde{\Phi}_{21} = \hat{P}_u \frac{1}{\sqrt{2}}(\tilde{\chi}_1^{(1)}\tilde{\phi}_0^{(2)} + \tilde{\chi}_0^{(1)}\tilde{\phi}_1^{(2)}),$$

$$\tilde{\Phi}_{2-1} = \hat{P}_u \frac{1}{\sqrt{2}}(\tilde{\chi}_{-1}^{(1)}\tilde{\phi}_0^{(2)} + \tilde{\chi}_0^{(1)}\tilde{\phi}_{-1}^{(2)}),$$

$$\tilde{\Phi}_{11} = \hat{P}_u \frac{1}{\sqrt{2}}(\tilde{\chi}_1^{(1)}\tilde{\phi}_0^{(2)} - \tilde{\chi}_0^{(1)}\tilde{\phi}_1^{(2)}),$$

$$\tilde{\Phi}_{1-1} = \hat{P}_u \frac{1}{\sqrt{2}}(\tilde{\chi}_{-1}^{(1)}\tilde{\phi}_0^{(2)} - \tilde{\chi}_0^{(1)}\tilde{\phi}_{-1}^{(2)}),$$

$$\tilde{\Phi} = \frac{1}{\sqrt{6-2\sqrt{3}}}[\sqrt{2}\tilde{\Phi}_{20} + (\sqrt{3}-1)\tilde{\Phi}_{00}]$$

$$= \hat{P}_u \frac{1}{\sqrt{6-2\sqrt{3}}}[\tilde{\chi}_1^{(1)}\tilde{\phi}_{-1}^{(2)} + (\sqrt{3}-1)\tilde{\chi}_0^{(1)}\tilde{\phi}_0^{(2)} + \tilde{\chi}_{-1}^{(1)}\tilde{\phi}_1^{(2)}], \tag{11}$$

where $\tilde{\Phi}_{JM}$ is the electron wave function of the excited $(2\,^3P_1-2\,^3S_1)$ quasimolecular state, with total electron angular momentum J and projection M, and $\tilde{\phi}_m$ is the wave function of the $2\,^3P_1$ atom, with projection m of the total angular momentum. The tilde is used to denote that the quantization axis is here the internuclear axis. The first four states are characterized by the potential

$$V(r)=\frac{d^2}{2r^3}, \tag{12}$$

and the fifth one by

$$V_*(r)=\left(\frac{\sqrt{3}+1}{2}\right)\frac{d^2}{r^3}. \tag{13}$$

One can transform the states of Eq. (11) on the original quantization axis (direction of light propagation) by using the transformation

$$\tilde{\Phi}_{JM'}=\sum_M\ (-1)^{M'-M}\Phi_{JM}D^J_{-M',-M}(\theta,\varphi,0), \tag{14}$$

where $D^J_{M'M}(\theta,\varphi,\phi)$ is a finite rotation matrix. The angles θ,φ determine the orientation of the internuclear axis with respect to the axis of quantization.

Radiative transitions couple the initial state with the states $\Phi_{11},\Phi_{1-1},\Phi_{21},\Phi_{2-1}$. The dipole moment of the corresponding transitions is equal to $d/\sqrt{2}$. Accordingly, the dipole moment d_{JM} of transitions from the initial state (10) to the first four states (11) is

$$d_{JM}=\frac{d}{\sqrt{2}}(D^J_{-M,-1}+D^J_{-M,1}). \tag{15}$$

The dipole moment of the transition to the state $\tilde{\Phi}$ is

$$d_*=-\frac{d}{\sqrt{6-2\sqrt{3}}}(D^2_{0,-1}+D^2_{01}). \tag{16}$$

In our limit of large detuning the light absorption is dominated by distances in a narrow vicinity of the resonance separation r_δ determined by the condition $V(r)=\delta$ [or $V_*(r)=\delta$], and the number of absorption events per unit time and unit volume is given by (see, e.g., [19]):

$$\nu=2\pi\Omega^2 n_c n_s\int d^3r\left[\left|\frac{d_*(\theta,\varphi)}{\sqrt{2}d}\right|^2\delta(V_*(r)-\delta)+\sum_{JM}\left|\frac{d_{JM}(\theta,\varphi)}{\sqrt{2}d}\right|^2\delta(V(r)-\delta)\right], \tag{17}$$

where n_s is the density of atoms in system states (uncoupled states with $p<p_*$), n_c is the density of coupled states, and the summation should be performed over the first four states (11). Using Eqs. (15), (16), (12), and (13) we obtain from Eq. (17):

$$\nu=7.4\left(\frac{\Omega}{\delta}\right)^2\Gamma n_s(n_c\lambda\!\!\!^{-3}). \tag{18}$$

If we replace n_s in Eq. (18) by $n_s(\mathbf{p})$ we obtain the decay rate of the occupation number of system atoms with momentum $\mathbf{p}$ due to pair radiative collisions with bath atoms in uncoupled states. As those represent only a part of the bath, the above defined effective rate constant of radiative collisions G (and parameter β) are proportional to the ratio n_c/n_b. As we already mentioned for $p_*<k$ this ratio is equal to 1/2. Then, comparing Eqs. (18), (6), and (7) with Eq. (3) we find $\beta=7.4$ and (with $\alpha=1/2$) obtain

$$\eta=0.068(\Lambda_b^3/\lambda\!\!\!^{-3})=4.0k^3/p^3_{max}\approx 3(E_r/T_b)^{3/2}. \tag{19}$$

Unless p_{max} is within a factor 1.5 of k, the parameter η is less than unity. In other words the atom laser can only be realized in the case of positive δ if the bath is essentially cooled down to the recoil energy. As noted above Eq. (19) remains unchanged for $p_*>k$ since in this case the ratio n_c/n_b becomes smaller than 1/2, but the filling rate reduces by the same factor.

The above results are easily generalized to 3D and perhaps surprisingly the result does not change. The key point is that the final states of the colliding pair given in Eq. (11) remain unchanged and we need only to reconsider the initial state. In a 3D VSCPT cooling scheme different configurations of laser fields are possible. It has been shown [21] (see also Ref. [13]) that for a $J=1$ to $J=1$ transition there always exists an uncoupled state with $\mathbf{p}=\mathbf{0}$. This uncoupled state is a vector $\boldsymbol{\psi}(\mathbf{R})$ which satisfies the condition that the local spin vector is everywhere proportional and parallel to the polarization vector of the applied light field:

$$\boldsymbol{\psi}(\mathbf{R})=c\mathbf{E}(\mathbf{R}), \tag{20}$$

where c is a normalization coefficient, and $\mathbf{E}(\mathbf{R})$ is the laser electric field at position $\mathbf{R}$. Commonly the geometry of the light fields is selected such that it consists of three, mutually orthogonal, pairs of light fields each consisting of counterpropagating σ_+ and σ_- beams, just as in the 1D case. The resulting field $\mathbf{E}(\mathbf{R})$ is rather complicated, giving rise to a light field potential and a pumping rate which are modulated in real space. As in the 1D case, one can generalize Eq. (20) and obtain the expression for the ''uncoupled state'' with $\mathbf{p}\neq 0$. Then the loss rate from such ''uncoupled'' states is again proportional to p^2 [21,13]. When considering radiative collisions we can again omit all momentum labels, since for $|\delta|\gg\Gamma$ the dominant contribution to the rate of light absorption comes from interatomic distances $r\ll\lambda$, while the optical potential (and the function $\boldsymbol{\psi}$) vary on a length scale of order λ. At each point $\mathbf{R}$ we can define a local quantization axis perpendicular to the vector $\boldsymbol{\psi}(\mathbf{R})$ and find two orthogonal coupled states χ_{c1} and χ_{c2} which form the complement of $\boldsymbol{\psi}(\mathbf{R})$. The uncoupled state plays the role of the state χ_u, introduced above for the 1D case [see Eq. (5)], and the two coupled states correspond to superpositions of χ_c and χ_0.

One difference from the true 1D case is that due to the nonlocal nature of the pumping process neither of the states χ_{c1} and χ_{c2} is depopulated. However, as χ_{c1} and χ_{c2} are

related by a simple unitary transformation, one immediately finds that the probability that the atom is optically pumped into the state $\psi(\mathbf{R})$ from either of the coupled states is exactly 1/2, just as in the 1D case described above. Similarly, the result of the calculation presented above for the rate of radiative collisions carries over without change: We should only replace n_c in Eq. (18) by the total density of atoms in the states χ_{c1} and χ_{c2}.

The results of this section rely on perturbation theory and do not take into account the influence of light on the wave function of the relative motion of atoms in the initial state. In fact this wave function was implicitly put equal to unity at r close to the resonance separation r_δ, which assumes that the ratio Ω/δ is sufficiently small. The situation is different if $(\Omega/\delta)kr_\delta \gg 1$, where $k=\sqrt{m\delta}$ is the momentum of the relative motion acquired by a colliding pair in the light absorption process. Then the light will provide a repulsion between the potential curve $V(r)$ [or $V_*(r)$] and the potential curve of the ground electronic state (shifted by the photon energy). This decreases the probability for two atoms to approach each other to distances $r \sim r_\delta$ where the light absorption is most efficient. Hence, in principle, there is a possibility to reduce the rate of radiative collisions by increasing Ω/δ. In the case of He* for realistic frequency detuning this requires the Rabi frequency to be significantly larger than δ, as the quantity kr_δ will not be much greater than unity.

V. NEGATIVE DETUNING

For negative values of δ the situation is completely different from the case described above, as the light can only be at resonance with discrete vibrational levels (having orbital angular momentum equal to 1 and high vibrational quantum number) of the electronically excited molecule. This means that radiative collisions will be nothing else than photoassociation, a process well investigated in ultra-cold alkali atom gases (for a review see [22,23]). In analogy to Eq. (11), diagonalizing the Hamiltonian of resonance dipole interaction Eq. (9) we find four attractive excited electronic states:

$$\tilde{\Phi}_{22}=\hat{P}_u\tilde{\chi}_1^{(1)}\tilde{\phi}_1^{(2)},$$

$$\tilde{\Phi}_{2-2}=\hat{P}_u\tilde{\chi}_{-1}^{(1)}\tilde{\phi}_{-1}^{(2)},$$

$$\tilde{\Phi}_{10}=\hat{P}_u\frac{1}{\sqrt{2}}(\tilde{\chi}_1^{(1)}\tilde{\phi}_{-1}^{(2)}-\tilde{\chi}_{-1}^{(1)}\tilde{\phi}_1^{(2)}),$$

$$\tilde{\Phi}_-=\frac{1}{\sqrt{6+2\sqrt{3}}}[(\sqrt{3}+1)\tilde{\Phi}_{00}-\sqrt{2}\tilde{\Phi}_{20}]$$

$$=\hat{P}_u\frac{1}{\sqrt{6+2\sqrt{3}}}[\tilde{\chi}_1^{(1)}\tilde{\phi}_{-1}^{(2)}-(\sqrt{3}+1)\tilde{\chi}_0^{(1)}\tilde{\phi}_0^{(2)}+\tilde{\chi}_{-1}^{(1)}\tilde{\phi}_1^{(2)}]. \tag{21}$$

The first three states are characterized by the interaction potential $V_-(r)=-d^2/r^3$ and the fourth one by $V_{*-}(r)=(1-\sqrt{3})d^2/2r^3$. The transition dipole moments d_{JM} to the first three states of Eq. (21) are again given by Eq. (15) and the dipole moment of the transition to the state $\tilde{\Phi}_-$ is

$$d_{*-}=\frac{d}{\sqrt{6+2\sqrt{3}}}(D_{0,-1}^2+D_{01}^2). \tag{22}$$

The exact location of discrete vibrational levels in these potentials can only be found if one knows the short-range form of the interaction potentials. Nevertheless, the spacing $\Delta\varepsilon_\nu$ between adjacent levels with binding energies ε_ν and $\varepsilon_{\nu+1}$ is determined by the above given resonance dipole potentials V_- and V_{*-}: $\Delta\varepsilon_\nu \sim \varepsilon_\nu(r_t/r_0)^{1/2} \sim (\varepsilon_\nu/\Gamma)^{5/6}(\lambda\!\!\!^{-}/r_0)^{1/2}$, where r_t is the outer turning point for the relative motion of atoms in the bound state with vibrational quantum number ν, and $r_0=md^2 \gg r_t$. Hence, we can find the photoassociation rate as a function of the frequency detuning from the nearest vibrational resonance. If the light is nearly resonant with the vibrational level ν, the rate of photoassociation will be

$$\nu_{pa}=4\pi a_J\Omega^2\frac{\Gamma}{(\delta-\varepsilon_\nu)^2+\Gamma^2}\left|\int_0^\infty \psi_\nu(r)\psi_g(r)r^2dr\right|^2 n_c n_s. \tag{23}$$

Here $\psi_\nu(r)$ and $\psi_g(r)$ are the radial wave functions of the relative motion of atoms in the initial and final electronic states of the quasimolecule. The coefficient $a_J=1/(2J+1)$ for the states $\tilde{\Phi}_{JM}$ given in Eq. (21) and $a_J=0.042$ for the state $\tilde{\Phi}_-$. The main contribution to the integral in Eq. (24) comes from the vicinity of the tuning point r_t. Unless δ and ε_ν are very large, the wave function ψ_g can be put equal to unity at distances $r \sim r_t$. Calculating the integral by using a linear approximation for the potentials $V_-(r)$ and $V_{*-}(r)$ in the vicinity of r_t we find

$$\nu_{pa}=\pi b_J\left(\frac{\Omega}{\delta}\right)^2\frac{\Gamma^2}{(\delta-\varepsilon_\nu)^2+\Gamma^2}\Delta\varepsilon_\nu\lambda\!\!\!^{-3}n_c n_s. \tag{24}$$

For transitions to the state $\tilde{\Phi}_{JM}$ we have $b_J=a_J$, and for transitions to the state $\tilde{\Phi}_-$ the coefficient $b_J=0.015$.

At resonance the rate of photoassociation Eq. (24) is larger by a factor $\sim\Delta\varepsilon_\nu/\Gamma$ than the rate of radiative collisions given by Eq. (18) for similar but positive detuning. However, for large detuning the level spacing becomes very much larger than Γ and for most values of δ one will miss the vibrational resonances. The photoassociation rate is the smallest when the frequency detuning is just in between two resonances, i.e., is of order $\Delta\varepsilon_\nu$. In this case the two nearest resonances will give the dominant contribution and, assuming $\delta-\varepsilon_\nu \sim \Delta\varepsilon_\nu$, we have

$$\nu_{pa} \sim (\Omega/\delta)^2(\Gamma^2/\Delta\varepsilon_\nu)n_c n_s\lambda\!\!\!^{-3}, \tag{25}$$

which is smaller by factor of order $\Gamma/\Delta\varepsilon_\nu$ than the rate of radiative collisions for similar but positive detuning. Accordingly, we have $\eta \sim (\Lambda_b/\lambda\!\!\!^{-})^3(\Delta\varepsilon_\nu/\Gamma)$. Hence, for the negative δ case η can in principle be increased to a value above unity for higher bath ''temperatures'' than in the case of positive δ.

In practice, however, negative detuning does not lead to sub-Doppler cooling in a VSCPT scheme on a $J=1$ to $J=1$ transition. Therefore if we only rely on Doppler cooling to cool the bath, the increase in η resulting from the reduced rate of radiative collisions is counteracted by the decrease of Λ_b. It is not clear whether this problem can be easily dealt with.

VI. CONCLUSIONS

We have shown that the operating conditions for an atom laser based on VSCPT of He* are strongly limited by the loss mechanism associated with radiative collisions. For positive detuning it is necessary to precool the gas very close to the recoil energy. For negative detuning the situation is more favorable but the lack of sub-Doppler cooling in VSCPT schemes for negative δ may offset this advantage. We have shown that it is not fundamentally impossible to realize the operating conditions for an atom laser using VSCPT but in practice it may be rather difficult. Clearly all other loss mechanisms should be carefully eliminated.

Although we did not analyze in detail other atom laser schemes, we believe that in general it is crucial to take the effect of radiative collisions into account when considering these models.

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