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Effects of resonant dipole interaction between atoms on laser cooling: The case of two identical atoms

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We study the effects of the resonant dipole interaction between identical atoms undergoing polarization-gradient laser cooling. A generalized master equation, which includes both the atom-field interaction and the dipole-dipole interaction between atoms, is derived. We then apply this equation to the particular case of two identical atoms cooled simultaneously by a one-dimensional cooling field. We show that, as the average center-of-mass separation between the two atoms is reduced to be less than an optical wavelength $\lambda/2\pi$, the population of the ground motional state of the atoms begins to decrease sharply, indicating a breakdown of the cooling mechanism due to the enhanced atom-atom interaction.

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In the past few years, one has witnessed remarkable progress in the field of laser cooling and trapping of neutral atoms. In most of the existing theories on sub-Doppler laser cooling [1–3], one neglects the atom-atom interactions, and calculates the interaction between individual atoms and the cooling fields only. Such single-atom theories have their obvious limitations. With the development of new cooling and trapping techniques, the achievable densities of cold atomic vapors at the present stage are such that the average separations between vapor atoms are on the order of a few wavelengths of the cooling field. For higher vapor densities, it remains interesting to see how the interactions between identical atoms in the field affect the outcome of laser cooling.

There are some previous studies [4,5] on the effects of the long-range resonant interactions between atoms undergoing laser cooling or trapping. They are carried out in the so-called semiclassical regime, in which the center-of-mass (c.m.) motion of atoms is treated classically. A recent calculation [6] does include a quantal treatment of the atomic c.m. motion, but the spatial localization of atoms in the cooling field is absent from the model. The experimental observations of discretized motional energy spectra of atoms in one-dimensional optical molasses [7] provide evidence of the quantization of the atomic motion, as well as the spatial localization of atoms in the cooling field (atom crystals). It seems necessary to extend the earlier semiclassical theories to include the quantized c.m. motion of atoms and the spatial localization effects of atoms. In this Rapid Communication, we derive a generalized master equation describing the interaction between N atoms undergoing laser cooling. Then, based on this equation, we calculate the steady-state density matrix of two identical atoms simultaneously cooled by a pair of counterpropagating, linearly polarized fields with orthogonal polarizations (optical molasses).

The Hamiltonian describing a system of N atoms interacting with one-dimensional incident fields, which propagate along the z direction, and the spontaneous modes of the vacuum can be written as

$$H=H_a+H_f+H_{int} \ , \tag{1}$$

where H_a and H_f are the free Hamiltonians of the atoms and the vacuum field, given by

$$H_a=\sum_{l=1}^{N}\left(\frac{\mathbf{P}_l^2}{2m}+\hbar\omega_0\mathbf{P}_e^l\right) \ , \quad H_f=\sum_{\mathbf{k},\lambda}\hbar\omega_k a_{\mathbf{k},\lambda}^{\dagger}a_{\mathbf{k},\lambda} \ , \tag{2}$$

respectively, where $\mathbf{P}_l$ is the c.m. momentum of the lth atom, ω_0 is the unperturbed atomic transition frequency, $\mathbf{P}_e^l$ is the excited-state projection operator of the lth atom, and $a_{\mathbf{k},\lambda}$ ($a_{\mathbf{k},\lambda}^{\dagger}$) is the annihilation (creation) operator for the field mode $(\mathbf{k},\lambda)$. Notice that H_a includes the atomic translational degree of freedom as well through the momentum operator $\mathbf{P}_l$. H_{int} describes the interaction between the atoms at c.m. positions $\mathbf{R}_l$'s and both the incident fields of frequency ω and the vacuum modes. Under the rotating-wave approximation (RWA), it is given by

$$\begin{aligned}H_{int}=&\sum_{l=1}^{N}\frac{\hbar\Omega}{2}[\boldsymbol{\sigma}_{eg}^{l}\cdot\boldsymbol{\epsilon}(z_l)e^{-i\omega t}+\boldsymbol{\sigma}_{ge}^{l}\cdot\boldsymbol{\epsilon}^{*}(z_l)e^{i\omega t}]\\&+i\sum_{l=1}^{N}\sum_{\mathbf{k},\lambda}\hbar g_k[(\boldsymbol{\sigma}_{eg}^{l}\cdot\boldsymbol{\epsilon}_{\mathbf{k},\lambda})a_{\mathbf{k},\lambda}e^{i\mathbf{k}\cdot\mathbf{R}_l}\\&-a_{\mathbf{k},\lambda}^{\dagger}e^{-i\mathbf{k}\cdot\mathbf{R}_l}(\boldsymbol{\sigma}_{ge}^{l}\cdot\boldsymbol{\epsilon}_{\mathbf{k},\lambda}^{*})] \ .\end{aligned} \tag{3}$$

In the above equation, the atomic raising operator $\boldsymbol{\sigma}_{eg}^{l}$ is given by

$$\boldsymbol{\sigma}_{eg}^{l}=\sum_{q=-1}^{1}(-1)^q\sigma_{eg}^{l}(q)\boldsymbol{\epsilon}_{-q} \ , \tag{4}$$

where, using appropriate Clebsh-Gordan coefficients for coupling, the components $\sigma_{eg}^{l}(q)$ are given by

$$\sigma_{eg}^{l}(q)=\sum_{m_g,m_e}\langle J_g m_g,1q|J_e m_e\rangle|J_e m_e\rangle_l\langle J_g m_g| \ , \tag{5}$$

and J_g,J_e are the ground- and excited-state angular momenta.

$$\Omega=\frac{\mathcal{E}d_{eg}}{\hbar\sqrt{2J_e+1}} \ , \quad g_k=\frac{d_{eg}}{\sqrt{2J_e+1}}\sqrt{\frac{\omega_k}{2\hbar\epsilon_0 V}} \ , \tag{6}$$

are the Rabi frequencies associated with the incident and vacuum fields, where $\mathcal{E}$ is the cooling field amplitude, d_{eg} is

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the dipole matrix element between the ground and the excited state, and V is the quantization volume. Finally, $\boldsymbol{\epsilon}_{\mathbf{k},\lambda}$ and $\boldsymbol{\epsilon}(z_l)$ are the polarization vectors of the vacuum mode $(\mathbf{k},\lambda)$ and the cooling field, respectively.

Following the standard procedure of treating the vacuum as a reservoir and eliminating the vacuum field modes [8], one obtains the equation for the atomic density matrix as

$$\frac{d\rho(t)}{dt}=\frac{1}{i\hbar}[H_{eff}\rho(t)-\rho(t)H^{\dagger}_{eff}]+\Gamma\sum_{l}\int dp'\sum_{q=-1}^{1}N_q(p')$$
$$\times\sigma^{l}_{ge}(q)e^{-ip'z_l}\rho(t)e^{ip'z_l}\sigma^{l}_{eg}(q)$$
$$+\Gamma\sum_{l\neq l'}\boldsymbol{\sigma}^{l}_{ge}\cdot\rho(t)\alpha(\mathbf{R}_{ll'})\cdot\boldsymbol{\sigma}^{l'}_{eg}\ , \tag{7}$$

where Γ is the natural linewidth of the excited state. The angular distribution functions $N_q(p')$ for the emission of a spontaneous photon with a polarization component q and a momentum $\hbar p'$ along the z direction for an isolated atom are given by [9]

$$N_{\pm 1}=\frac{3}{8k}\left(1+\frac{p'^2}{k^2}\right)\ ,\quad N_0=\frac{3}{4k}\left(1-\frac{p'^2}{k^2}\right)\ . \tag{8}$$

The non-Hermitian effective Hamiltonian H_{eff} in Eq. (7) is given by

$$H_{eff}=\sum_{l}\left[\frac{\mathbf{P}_l^2}{2m}+\hbar\left(-\Delta-i\,\frac{\Gamma}{2}\right)\mathbf{P}^{l}_{e}+\frac{\hbar\Omega}{2}[\boldsymbol{\sigma}^{l}_{eg}\cdot\boldsymbol{\epsilon}(z_l)\right.$$
$$\left.+\mathrm{H.c.}]\right]-\frac{\hbar\Gamma}{2}\sum_{l\neq l'}\boldsymbol{\sigma}^{l}_{eg}\cdot[i\boldsymbol{\alpha}(\mathbf{R}_{ll'})+\boldsymbol{\beta}(\mathbf{R}_{ll'})]\cdot\boldsymbol{\sigma}^{l'}_{ge}\ , \tag{9}$$

where $\Delta=\omega-\omega_0$ is the cooling field detuning, and $\mathbf{R}_{ll'}=\mathbf{R}_l-\mathbf{R}_{l'}$. The two tensor operators $\boldsymbol{\alpha}$ and $\boldsymbol{\beta}$ are given by

$$\boldsymbol{\alpha}(\mathbf{R})=\frac{3}{2}\,(\mathbf{I}-\hat{\mathbf{R}}\hat{\mathbf{R}})\frac{\sin kR}{kR}+\frac{3}{2}\,(\mathbf{I}-3\hat{\mathbf{R}}\hat{\mathbf{R}})\left(\frac{\cos kR}{k^2R^2}-\frac{\sin kR}{k^3R^3}\right)\ , \tag{10}$$

$$\boldsymbol{\beta}(\mathbf{R})=\frac{3}{2}\,(\mathbf{I}-\hat{\mathbf{R}}\hat{\mathbf{R}})\frac{\cos kR}{kR}-\frac{3}{2}\,(\mathbf{I}-3\hat{\mathbf{R}}\hat{\mathbf{R}})\left(\frac{\sin kR}{k^2R^2}+\frac{\cos kR}{k^3R^3}\right)\ , \tag{11}$$

respectively, where $\hat{\mathbf{R}}$ is the unit vector in the direction of $\mathbf{R}$. They describe the dependence of the strength of the resonant interaction between two atoms on their mutual separation. In particular, $\boldsymbol{\alpha}$ is related to the modification of the atomic relaxation rate due to the resonant interaction, and $\boldsymbol{\beta}$ to the shift of the energy levels of one atom in the presence of another [10].

It is known for polarization-gradient cooling [11] that optimal cooling effects can be achieved in the limit of weak fields and large detuning (the secular limit), i.e.,

$$|\Delta|\gg\Gamma,\Omega\ . \tag{12}$$

At present, we assume that the excited-state energy shifts of the atoms due to their mutual interaction, which are of order $\Gamma|\boldsymbol{\beta}(\mathbf{R}_{ll'})|$, remain small compared to $\hbar|\Delta|$. As a result, adiabatic elimination of the electronic excited state is possible. This assumption imposes a lower limit on the closest average distance b (impact parameter) that two atoms can approach each other in our model. However, under the secular limit, the distance b can be much smaller than the laser wavelength λ without invalidating this assumption.

To eliminate the electronic excited state, one makes the substitutions

$$\boldsymbol{\sigma}^{l}_{ge}\rightarrow\boldsymbol{\xi}^{l}=-\frac{i\Omega/2}{\Gamma/2-i\Delta}\boldsymbol{\sigma}^{l}_{ge}[\boldsymbol{\sigma}^{l}_{eg}\cdot\boldsymbol{\epsilon}(z_l)]+\sum_{l'\neq l}\frac{\Gamma\Omega/4}{(\Gamma/2-i\Delta)^2}$$
$$\times\boldsymbol{\sigma}^{l}_{ge}\{\boldsymbol{\sigma}^{l}_{eg}\cdot[i\{\boldsymbol{\alpha}(\mathbf{R}_{ll'})+\boldsymbol{\beta}(\mathbf{R}_{ll'})]\cdot\boldsymbol{\sigma}^{l'}_{ge}\}$$
$$\times[\boldsymbol{\sigma}^{l'}_{eg}\cdot\boldsymbol{\epsilon}(z_{l'})]\ ,$$
$$\mathbf{P}^{l}_{e}\rightarrow(\boldsymbol{\xi}^{l})^{\dagger}\boldsymbol{\xi}^{l}\ , \tag{13}$$

in Eqs. (7) and (9) [12]. To lowest order in $(\Gamma/|\Delta|)|\boldsymbol{\beta}(\mathbf{R}_{ll'})|$ and $\Gamma/|\Delta|$, the resulting density-matrix equation for the atomic ground states is then given by

$$d\rho(t)/dt=[H^{0}_{eff}+H'_{eff},\rho(t)]/i\hbar+[\dot{\rho}]_{relax}\ . \tag{14}$$

The zeroth-order Hamiltonian H^{0}_{eff} is the sum of the atomic kinetic energy and the light-shift potentials associated with the atomic ground-state sublevels, given by

$$H^{0}_{eff}=\sum_{l}\left\{\frac{\mathbf{P}_l^2}{2m}+U_0[\boldsymbol{\sigma}^{l}_{ge}\cdot\boldsymbol{\epsilon}^{*}(z_l)][\boldsymbol{\sigma}^{l}_{eg}\cdot\boldsymbol{\epsilon}(z_l)]\right\}\ , \tag{15}$$

where the potential depth U_0 is

$$U_0=\hbar\Delta(\Omega/2)^2/(\Gamma^2/2^2+\Delta^2). \tag{16}$$

The modification to the ground-state Hamiltonian due to the resonant dipole interaction between different atoms, H'_{eff}, is given by

$$H'_{eff}=-\hbar\Gamma'\sum_{l\neq l'}[\boldsymbol{\sigma}^{l}_{ge}\cdot\boldsymbol{\epsilon}^{*}(z_l)]$$
$$\times\boldsymbol{\sigma}^{l}_{eg}\cdot\boldsymbol{\beta}(\mathbf{R}_{ll'})\cdot\boldsymbol{\sigma}^{l'}_{ge}[\boldsymbol{\sigma}^{l'}_{eg}\cdot\boldsymbol{\epsilon}(z_{l'})]\ , \tag{17}$$

where

$$\Gamma'=\Gamma(\Omega/2)^2/(\Gamma^2/2^2+\Delta^2). \tag{18}$$

The relaxation term $[\dot{\rho}]_{relax}$ in Eq. (14) can be written as

$$[\dot{\rho}]_{relax}=-\frac{1}{2}\sum_{l}[A^{l}\rho(t)+\rho(t)A^{l}]+\Gamma'\sum_{l}\int dp'\sum_{q}N_q(p')\sigma^{l}_{ge}(q)[\boldsymbol{\sigma}^{l}_{eg}\cdot\boldsymbol{\epsilon}(z_l)]e^{-ip'z_l}\rho(t)e^{ip'z_l}[\boldsymbol{\sigma}^{l}_{ge}\cdot\boldsymbol{\epsilon}^{*}(z_l)]\sigma^{l}_{eg}(q)$$
$$+\Gamma'\sum_{l\neq l'}\boldsymbol{\sigma}^{l'}_{ge}[\boldsymbol{\sigma}^{l'}_{eg}\cdot\boldsymbol{\epsilon}(z_{l'})]\cdot\rho(t)\boldsymbol{\alpha}(\mathbf{R}_{ll'})\cdot[\boldsymbol{\sigma}^{l}_{ge}\cdot\boldsymbol{\epsilon}^{*}(z_l)]\boldsymbol{\sigma}^{l}_{eg}\ , \tag{19}$$

where

$$A^l=\Gamma'[\boldsymbol{\sigma}^l_{ge}\cdot\boldsymbol{\epsilon}^*(z_l)][\boldsymbol{\sigma}^l_{eg}\cdot\boldsymbol{\epsilon}(z_l)]+\Gamma'\sum_{l'\neq l}[\boldsymbol{\sigma}^l_{ge}\cdot\boldsymbol{\epsilon}^*(z_l)]\boldsymbol{\sigma}^l_{eg}\cdot\boldsymbol{\alpha}(\mathbf{R}_{ll'})\cdot\boldsymbol{\sigma}^{l'}_{ge}[\boldsymbol{\sigma}^{l'}_{eg}\cdot\boldsymbol{\epsilon}(z_{l'})]\ . \tag{20}$$

From Eqs. (15)–(18), the ratio between the magnitudes of the dipole interaction energy H'_{eff} and the single-atom light shift H^0_{eff} is given approximately by the factor $(\Gamma/|\Delta|)|\boldsymbol{\beta}(\mathbf{R}_{ll'})|$. In the secular limit (12), H'_{eff} can be small compared to H^0_{eff}.

In the following, we apply Eq. (14) derived above to the specific case of two identical atoms undergoing one-dimensional polarization-gradient cooling. The cooling fields are two counterpropagating, cross-polarized fields. The polarization vector of the total field $\boldsymbol{\epsilon}$ is given by

$$\boldsymbol{\epsilon}(z)=(-i)\sin(kz)\boldsymbol{\epsilon}_{+1}+\cos(kz)\boldsymbol{\epsilon}_{-1}\ . \tag{21}$$

We calculate the steady-state atomic density matrix with the inclusion of the dipole-dipole interaction between these atoms. In this one-dimensional case, the atomic motion along the field propagation direction needs to be quantized, while the atomic motion transverse to the laser axis remains arbitrary. As a model, we fix the transverse separation between the two atoms at a distance b, while allowing the quantized motion of atoms along the laser direction. We are primarily interested in the dependence of the two-atom density matrix on the separation b. For the internal level scheme of the atoms, we choose a simple $J_g=1/2\rightarrow J_e=3/2$ transition. Transitions between states with higher angular momentum J could also be studied starting from Eq. (14), but we expect the main features to be similar.

Since we restrict ourselves to the case where H'_{eff} is small compared to the single-atom Hamiltonian H^0_{eff}, we can expand the atomic density matrix in the eigenstate basis of H^0_{eff}, i.e., the product space of the Bloch states of the two atoms $|\nu_1,q_1,\epsilon_1\rangle_1|\nu_2,q_2,\epsilon_2\rangle_2$[2], where $\nu_i=0,1,\ldots,$ is the band index, q_i is the Bloch index, and $\epsilon_i=\pm$ denotes the two internal ground-state sublevels $|m_g=\pm1/2\rangle$ of the atoms. In general, the density-matrix elements can be written as

$$\rho(\nu_1q_1\epsilon_1\nu_1'q_1'\epsilon_1';\nu_1q_1\epsilon_1\nu_1'q_1'\epsilon_1')$$
$$={}_1\langle\nu_1,q_1,\epsilon_1|_2\langle\nu_2,q_2,\epsilon_2|\rho|\nu_1',q_1',\epsilon_1'\rangle_1|\nu_2',q_2',\epsilon_2'\rangle_2\ . \tag{22}$$

Certain terms of the density matrix can be neglected under given conditions. In the secular limit (12), the energy separations between Bloch states with different band indices ν (which are on the order of $|U_0|$) are much greater than the relaxation rates of these states due to optical pumping (of order Γ'). As a result, one is able to neglect the fast oscillating, off-diagonal terms of the density matrix and keep only those matrix elements with $\nu_1+\nu_2=\nu_1'+\nu_2'$ [13]. Even with this simplification, the number of remaining density matrix elements can still be enormous. To minimize the numerical computation, we choose to include the lowest Bloch states with $\nu\leqslant8$, which proves to be sufficient in describing the localization of atom in the light-shift potential wells.

The steady-state solution of the atomic density matrix ρ can be obtained by integrating Eq. (14) starting, for example, from the initial density matrix of two isolated atoms. Figure 1 shows the dependence of the population of the ground motional state of each atom,

$$\pi_0=\sum_{q,\epsilon}\ \sum_{\nu',q',\epsilon'}\rho(0q\epsilon0q\epsilon;\nu'q'\epsilon'\nu'q'\epsilon')\ , \tag{23}$$

on their mean separation b, for a potential depth $U_0=-100E_k$ and a detuning $\Delta/\Gamma=-10$, where $E_k=\hbar^2k^2/2m$ is the photon recoil energy. For $b\geqslant\lambda/2\pi$, the value of π_0 is virtually indistinguishable from that of an isolated atom. In this distance range, the dipole-dipole interaction between the two atoms is dominated by the long-range radiative part, which decreases with the atom separation R_{12} as $1/(kR_{12})$. The fact that the modifications to π_ν's are small in this distance range indicates that the long-range radiative interaction between the two atoms has negligible effects on the outcome of Sisyphus cooling for this particular system. As b is further reduced below the wavelength $\lambda/2\pi$, the static dipole-dipole interaction between two atoms, which varies as $1/(kR_{12})^3$, becomes dominant. One can see from Fig. 1 that the population of the ground motional state starts to decrease sharply below this distance range, indicating a breakdown of the conventional polarization-gradient cooling scheme.

In Fig. 2, we show the cooperative populations of the two atoms, defined as

$$\pi(\pm,\pm)=\sum_{\nu,\nu',q,q'}\rho(\nu q\pm\nu q\pm;\nu'q'\pm\nu'q'\pm)\ ,$$
$$\pi(\pm,\mp)=\sum_{\nu,\nu',q,q'}\rho(\nu q\pm\nu q\pm;\nu'q'\mp\nu'q'\mp)\ , \tag{24}$$

as a function of b. As b is reduced below $\lambda/2\pi$ and the population of the ground motional state starts to decrease, the probability of both atoms occupying the same internal ground sublevel (either $m_g=1/2$ or $-1/2$) becomes less than the probability of their occupying the opposite ones (one atom in $m_g=1/2$ and the other in $m_g=-1/2$), i.e., $\pi(\pm,\pm)<\pi(\pm,\mp)$. Since the internal state of an atom localized in a potential well is determined by the polarization of the field at the position of the well (either σ^+ or σ^-), Fig. 2 also shows that atoms tend to be localized in light-shift potential wells where the field polarizations are opposite as their mutual interaction becomes enhanced.

The above results can be qualitatively understood as follows. First, the radiative part of the dipole-dipole interaction, established through exchange of transverse photons between both atoms, has negligible effects on the outcome of cooling. The photon-exchange processes can create coherences between atomic motional states with the same band indices ν

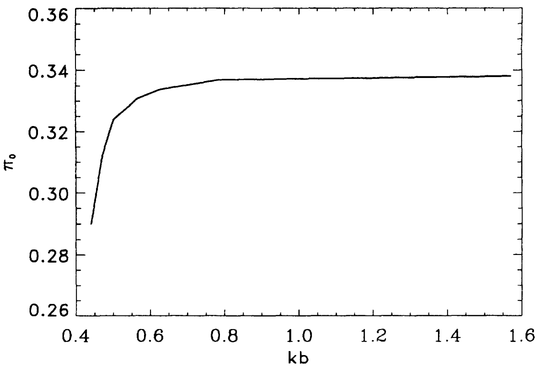

FIG. 1. Population of the atomic ground motional state π_0. The potential depth $U_0/E_k = -100$, and detuning $\Delta/\Gamma = -10$.

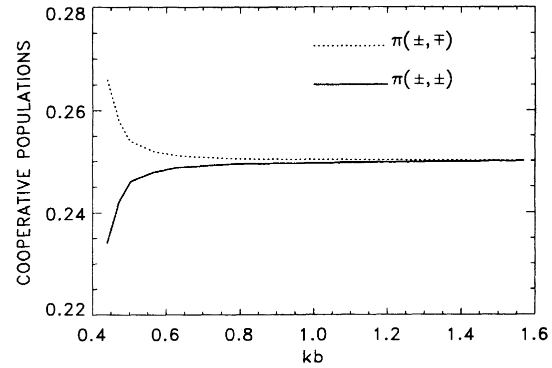

FIG. 2. Cooperative populations of two interacting atoms $\pi(\pm,\pm)$ and $\pi(\pm,\mp)$. The parameters are the same as in Fig. 1.

but different Bloch indices q (Rayleigh coherences), as well as coherences between states with different band indices ν (Raman coherences). The magnitudes of both the Rayleigh and Raman coherences are proportional to the factor $|\chi_{fi}|^2|p_{fi}|$, where χ_{fi} is the transition matrix element between the initial and final states, and p_{fi} denotes the population difference between the initial and final states. For the Rayleigh coherence, although the transition probability $|\chi_{fi}|^2$ can have appreciable magnitude (given approximately by the optical pumping rate Γ'), the population differences between Bloch states with the same ν and different q's are small for the lowest motional bound states[14]. For the Raman coherences, the opposite is true: the population differences between Bloch states of different indices ν's can be of order unity; however, due to the small degree of overlap of the wave functions between these different vibrational states, the transition probability $|\chi_{fi}|^2$, or the Franck-Condon factor, is small[14]. As a result, both the Rayleigh and the Raman coherences established through exchange of transverse photons have negligible values, and the modifications to the atomic density matrix induced by the radiative dipole-dipole interaction are small.

Second, as the transverse separation b decreases below $\lambda/2\pi$, the strength of the static dipole-dipole interaction between both atoms increases rapidly. The dipole interaction leads to extra couplings between different motional states of each atom, which redistribute populations among these states. As a result, the population of the ground motional state π_0 is reduced. One can also understand the results shown in Fig. 2 based on the above argument. The two atoms interact most strongly when they are separated only by the distance b in the transverse direction, and localized in the light-shift potential wells associated with the same internal atomic state. The strong dipole-dipole interaction leads to an increase in the populations of the excited motional states. An atom in an excited motional state can be transferred quickly to a different internal state due to enhanced optical pumping, which then becomes localized in the light-shift potentials associated with this other internal state. As a result, atoms are less likely to occupy the same internal states as the opposite ones.

Although we consider a one-dimensional cooling configuration in this calculation, the results here should help us to understand the situations in two- or three-dimensional molasses. For atomic vapor densities sufficiently low that, on the average, there are much less than one atom per potential well, one expects the effects related to the dipole-dipole interaction to be small, since the separation between adjacent potential wells is at least $\lambda/2$, which is beyond the distance where the static dipole interaction between atoms starts to become important. However, as the density of cold atoms increases to a point where there can be more than one atom in a single potential well, cooling will be significantly limited due to the strong resonant dipole interaction between atoms.

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