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Laser cooling of atoms

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Various aspects of the laser cooling of atoms are investigated theoretically. More generally, the authors investigate a process through which the kinetic energy of a collection of resonant absorbers can be reduced by irradiating these absorbers with near-resonant electromagnetic radiation. The process is described here as anti-Stokes spontaneous Raman scattering. Cooling mechanisms, rates, and limits are discussed for both free and bound atoms.

I. INTRODUCTION

In the past few years, there has been increasing interest in the use of near-resonant photon scattering to cool a collection of atoms, ions, or molecules. This interest is motivated in part by the practical need to reduce first- and second-order Doppler shifts in ultra-high-resolution spectroscopy and in part by the esthetic appeal of controlling the positions and velocities of a collection of atomic particles to within the limits imposed by quantum fluctuations. Recent proposals and experiments using narrow-band tunable lasers suggest that such control may soon become a reality. It is not difficult to imagine that the concepts and techniques which are being developed may have application in a variety of areas not initially anticipated.

Current interest in the possibility of cooling began with independent proposals to reduce the temperature of a gas of neutral atoms[1] or ions which are bound in an electromagnetic "trap"[2] with near-resonant laser radiation. This method of cooling has subsequently been incorporated into the interesting schemes for trapping of particles using near-resonant optical fields.[3] The first demonstration[4] of cooling using the basic techniques described here was made for a slightly modified situation; specifically, the magnetron motion of an electron bound in a Penning trap was "cooled" by a technique called motional sideband excitation,[5] which is formally equivalent to the laser cooling of atoms. Cooling of ions bound in an electromagnetic trap was more recently demonstrated.[6,7] The cooling which is potentially achievable should permit spectroscopy of unprecedented resolution and accuracy.

As discussed below, the technique can variously be described in terms of radiation pressure, motional sideband excitation, optical pumping, or anti-Stokes spontaneous Raman scattering; this last concept is the one primarily used here because of its generality. It should be mentioned that cooling by Raman scattering is not a new idea; "lumino-refrigeration" was hypothesized as early as 1950 by Kastler.[8] We also note that other cooling processes are possible; for example, one could use optical pumping followed by collisional relaxation, as discussed in Sec. II, or cooling by collisionally aided fluorescence.[9] The process described in this paper is, however, more direct and does not rely on atom-atom collisions to alter the atom kinetic energy.

The paper is divided as follows. In Sec. II we describe the general aspects of the cooling process and treat the problem combining simple classical and quantum ideas. Section III introduces the concepts and notation of the quantum-mechanical treatment which is then applied to free atoms in Sec. IV and bound atoms in Sec. V. In order to make the problem somewhat more tractable, we limit the discussion to simple systems which exhibit the salient features of the process.

II. SIMPLE DESCRIPTION OF THE COOLING PROCESS

A. Analogy with optical pumping

The basic features of the cooling process have been outlined previously (Refs. 1, 2, 5, 6, 7). The attempt is made here to describe the qualitative aspects of the problem more completely; however, the general problem becomes quite complicated, and therefore several limiting cases will be treated.

First, recall that in optical pumping we have a way of drastically altering the temperature of a specific degree of freedom in an atom or molecule. Assume, for example, that we have an alkali-like atom which has ground-state "hyperfine" structure. This atom can also have many

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excited electronic states, but we assume that we have a narrow-band laser which can excite the atom from one of the ground states to only one excited electronic state. The relevant levels are shown in Fig. 1.

Without laser irradiation, the atoms eventually reach thermal equilibrium by collisions or interaction with the background blackbody radiation. Therefore, the ratio of the number of atoms in state 2 to those in state 1 is given by the Boltzmann law:

$$N_2/N_1 = \exp[-(E_2 - E_1)/k_B T], \quad (1)$$

where k_B = Boltzmann's constant, T = temperature, and E_1, E_2 and N_1, N_2 are the respective energies and numbers of atoms in the states. For simplicity we assume that the ground state has only two nondegenerate energy levels; hence statistical weight factors are absent in Eq. (1).

If we now apply the laser radiation to the atoms, optical pumping occurs. Atoms are excited to level 3, but can decay into either ground state. If we neglect the finite frequency widths of the laser and of the optical transition, this process continues until all of the atoms are in level 1. They remain there until another process (say, the collisions) depopulates this level. However, in the pumping process, $N_2/N_1 \to 0$, and, via Eq. (1), we may say that $T \to 0$ also. In this simple example, we see that we can cool an internal degree of freedom of the atom (the hyperfine structure) by optical pumping. In principle, we could continue this optical pumping process and, using collisions to transfer kinetic energy to the internal degree of freedom, could reduce the translational temperature of the gas if there were sufficient isolation from the rest of the environment. The process of laser cooling discussed below is very similar to the optical pumping case except that the translational degrees of freedom are optically pumped directly.

B. Laser cooling of free atoms

Assume that we have an unbound gas of atoms (or resonant absorbers in general) which possess a resonant electric dipole transition (frequency, ν_0) in some convenient spectral region with radiative linewidth $\gamma/2\pi$ (full width at half-intensity points). Now suppose that we irradiate these atoms with monochromatic, directed, low intensity radiation tuned near, but slightly lower than, the resonance frequency. We assume that the intensity is well below that which would cause saturation (the case of saturation is treated in Ref. 10), and that the thermalizing collision rate γ_c between atoms is much less than the natural linewidth γ, but is larger than the optical absorption rate ($\gamma \gg \gamma_c \gg$ absorption rate, see Sec. V F). Those atoms of a particular velocity class moving against the radiation are Doppler shifted toward the resonant frequency ν_0 and scatter the incoming light at a higher rate than those atoms moving with the radiation which are Doppler shifted away from resonance. For each scattering event, the atom receives a momentum impulse $\hbar\vec{k}$ ($\vec{k}$ is the photon wave vector) in the absorption process. For an atom which is moving against the radiation, this impulse retards its motion. This retardation can also be described in terms of radiation pressure.[1,10] The average momentum per scattering event transferred to the atom by the reemitted photons is zero, because of the randomness of the photons' directions (if we neglect terms of second order in $|\vec{v}|/c$, where $\vec{v}$ is the atom velocity and c is the speed of light). The average net effect then is that the atomic velocity is changed by an amount $\Delta\vec{v} \cong \hbar\vec{k}/M$ per scattering event, where M is the atomic mass. When $\vec{v}$ and $\vec{k}$ are antiparallel, this leads to a net cooling, provided $|\vec{v} + \Delta\vec{v}| < |\vec{v}|$.[1] (See Fig. 2.) In a practical cooling experiment it would be desirable to irradiate the atoms from all sides with radiation that covered the entire lower half of the Doppler profile.[1] Alternatively, narrow-band laser schemes might be employed where the laser frequency is swept from some very low value to a value approaching the rest frequency.[3] This requirement is substantially relaxed if the

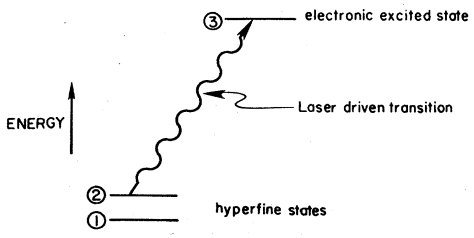

FIG. 1. Levels of interest in a hypothetical alkali-like atom. Optical pumping into state 1 occurs while driving the $2 \longleftrightarrow 3$ transition with a laser.

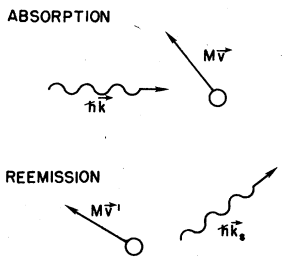

FIG. 2. Qualitative description of radiation-pressure cooling. In the absorption process, the atomic velocity is changed (reduced for $\vec{k}\cdot\vec{v} < 0$) by an amount $\Delta\vec{v} = \hbar\vec{k}/M$. In the reemission process, the average change in velocity is zero. Therefore in the overall scattering process, the kinetic energy can be reduced.

atoms are bound, as is described later. (See Sec. II C.)

The above argument gives the essence of the cooling process but neglects residual heating effects due to recoil. For example, in the reemission process, as explained below, the average momentum transfer is zero (neglecting terms of order $|\vec{v}|^2/c^2$); however, the atom undergoes a random walk in momentum space because of the finite momentum transfer in each emission. Thus, the limiting kinetic energy for a single atom in the cooling process must be at least the recoil energy $R=(\hbar k)^2/2M$, and, as we will see below, may be considerably larger than the recoil energy. To be more quantitative, we first write the *resonance* absorption and spontaneous emission frequencies, which are given by:

$$\omega_{abs}=\omega_0+\vec{k}_{abs}\cdot\vec{v}-\tfrac{1}{2}\omega_0\beta^2+R/\hbar\,, \tag{2a}$$

$$\omega_{em}=\omega_0+\vec{k}_{em}\cdot\vec{v}'-\tfrac{1}{2}\omega_0\beta^2-R/\hbar\,, \tag{2b}$$

where $\omega_0=2\pi\nu_0$, $|\vec{k}_{abs}|=\omega_{abs}/c$, $|\vec{k}_{em}|=\omega_{em}/c$, $\vec{v}$ = atom velocity in the ground state, $\vec{v}'$ = atom velocity in excited state, c = speed of light, $\beta=|\vec{v}|/c$, $\hbar\cdot 2\pi$ = Planck's constant. These equations, valid to order β^2 in the atom velocity, and to first order $R/\hbar\omega_0$ in the recoil energy, can be obtained by invoking conservation of energy and momentum in the absorption and emission processes. The second term on the right-hand side of these equations is the ordinary first-order Doppler shift, the third term is the second-order Doppler or time dilation shift, and the last term is usually called the recoil shift. We will use the nonrelativistic approximation and therefore neglect the third term in Eq. 2. Also, as explained in Sec. III, the net effect of averaging over all possible directions of reemission, is equivalent to setting the second term in Eq. 2(b) equal to zero (neglecting terms of order β^2 and higher). Hence, the average energy change of the photon per resonant spontaneous scattering event is given by

$$\Delta E(\text{photon})=\hbar(\omega_{em}-\omega_{abs})=-\hbar\vec{k}_{abs}\cdot\vec{v}-2R\,.$$

Here we note that this equation applies to both resonant and nonresonant scattering and can also be simply derived in general by considering conservation of energy and momentum in the overall scattering process. For this more general case, $\vec{k}_{abs}$ is replaced by $\vec{k}$. The change in photon energy is accounted for by a change in atom kinetic energy per scattering event

$$\Delta E_K(\text{atom})=\hbar\vec{k}\cdot\vec{v}+2R\,. \tag{3}$$

This leads to a net average cooling as long as $\hbar\vec{k}\cdot\vec{v}<-2R$, and net heating for the opposite condition. As an example, suppose we have an atom with $M=100$ amu, $\nu_0=5\times10^{14}$ Hz($\lambda=600$ nm) at thermal energies ($T=300$ K), then $v_{rms}=2.2\times10^4$ cm/s, $\hbar|\vec{k}|v_{rms}=2.5\times10^{-18}$ ergs, $2R=7.3\times10^{-23}$ ergs. Clearly, the recoil heating does not play a significant role until very low temperatures are achieved. Of course, at higher frequencies ν_0 it becomes more important. If we interpret the energy changes in terms of temperature changes by the relation $\Delta E_K=\frac{3}{2}k_B\Delta T$, then $\Delta T\cong0.012$ K per scattering event at $T=300$ K, and it therefore takes on the order of 10^4 scattering events to do substantial cooling at optical frequencies.

1. Cross section and cooling rates

The atomic cross section for absorption, neglecting saturation,[10] has the form

$$\sigma(\omega)=\sigma_0(\tfrac{1}{2}\gamma)^2/[(\omega-\omega_{abs})^2+(\tfrac{1}{2}\gamma)^2]\,, \tag{4}$$

where σ_0 is the resonance scattering cross section ($\sigma_0=2\pi\lambda^2$ for unpolarized atoms). Therefore, if the radiation is incident along the x direction, the rate of kinetic energy change is, using Eq. (3):

$$\frac{dE}{dt}=\frac{I}{\hbar\omega}\sigma(\omega)(\hbar kv_x+2R)\,, \tag{5}$$

where I is the energy flux of the laser beam (ergs/sec cm^2) and where σ is given by Eq. (4). (This expression will be more fully justified in Sec. III.) Equation (5) gives the cooling (heating) rate for a single atom. If we consider an ensemble of atoms we must average v_x over the velocity distribution. If, for example, the distribution is given by a Maxwell-Boltzmann distribution, Eq. (4) becomes a Voigt profile:

$$\sigma(\omega)=\sigma_0\int_{-\infty}^{\infty}\frac{\exp[-(v_x/u)^2]}{\sqrt{\pi}\,u}\times\frac{dv_x}{1+[(2/\gamma)(\omega-\omega_0'-kv_x)]^2}\,, \tag{6}$$

where $u=(2k_BT/M)^{1/2}$ and $\omega_0'=\omega_0+R/\hbar$. Similarly, Eq. (5) becomes

$$\frac{dE}{dt}=\frac{I\sigma_0}{\hbar\omega}\int_{-\infty}^{\infty}\frac{\hbar kv_x+2R}{1+[(2/\gamma)(\omega-\omega_0'-kv_x)]^2}\times\frac{\exp[-(v_x/u)^2]}{\sqrt{\pi}\,u}dv_x\,. \tag{7}$$

Equation (7) can also be written in the form

$$\frac{dE}{dt}=\frac{I\sigma_0\gamma^2}{4ku\omega}\left[\mathrm{Re}\,Z(F,Q)+\frac{2}{\gamma}\left(\omega_0'-\omega-\frac{2R}{\hbar}\right)\mathrm{Im}Z(F,Q)\right],$$

where $F\equiv(\omega_0'-\omega)/(ku)$, $Q\equiv\gamma/(2ku)$, and $Z(F,Q)$

is sometimes called the plasma dispersion function[11] defined as

$$Z(F,Q)=\pi^{-1/2}\int_{-\infty}^{\infty}\frac{dx\,e^{-x^2}}{x-F-iQ}$$

for $Q>0$. We can numerically solve Eq. (7) for dE/dt; however, when γ and $R/\hbar$ are much less than the Doppler width we can approximate the integral and the cooling rate by

$$\frac{dE}{dt}=\frac{I\sigma_0}{\hbar\omega}[\hbar(\omega-\omega_0)+R]\frac{\gamma\sqrt{\pi}}{2\omega_D}\exp\left[-\left(\frac{\omega-\omega_0'}{\omega_D}\right)^2\right], \quad (8)$$

where $\omega_D=\omega_0 v/c$.

For the case of unbound atoms, radiation incident along only one direction would retard the motion only in that direction and heat the motion in the other directions due to recoil. Therefore, in a practical situation we might irradiate the atoms with six narrow-band laser beams directed along the $\pm x$, $\pm y$, and $\pm z$ directions of a Cartesian coordinate system. In this case we must sum terms like Eq. (5) for each laser beam. If (i) the frequencies of the lasers are equal giving standing waves, or (ii) if the waves are not plane, we must consider the effect of the induced dipole forces responsible for optical trapping.[3] In order to disregard this effect we will assume that the incident (plane-wave) radiation has low enough power—that is, the limiting kinetic energy is larger than the potential energy "hills" created by the standing-wave-induced dipole force. In this limit, the induced forces average to zero. When the atoms are bound this complication need not arise, since one laser beam may be sufficient to cool all degrees of freedom.

2. *Cooling limit*

We can estimate the limit on cooling with the following argument whose general features were advanced by Purcell.[12] Assume that substantial cooling has already taken place so that the Doppler width of the atomic transition is smaller than the natural linewidth γ. We further assume that we irradiate the unpolarized atoms with six laser beams as described above, which are tuned near the lower half-power point of the optical resonance, $\omega-\omega_0\cong-\frac{1}{2}\gamma$. Considering the rate of energy change in the x direction, we can write

$$\frac{dE_x}{dt}=\frac{I}{\hbar\omega}[\sigma_+(\hbar k v_x+2R)+\sigma_-(-\hbar k v_x+2R)], \quad (9)$$

where I is the energy flux of each laser beam and where $\sigma_+(\sigma_-)$ is the scattering cross section for the laser light moving with (against) the atomic motion.

With the approximation that the Doppler width is much less than the natural width, we can show that the maximum cooling rate is achieved when $\omega=\omega_0'-\frac{1}{2}\gamma$. In this case Eq. (9) reduces to

$$\frac{dE_x}{dt}=\frac{2I\sigma_0}{\hbar\omega}\left(-\frac{\hbar k^2\langle v_x^2\rangle}{\gamma}+R\right). \quad (10)$$

We find the minimum kinetic energy (E_K) when $dE_x/dt=0$, in which case

$$\langle E_{Kx}\rangle_{\min}=\tfrac{1}{4}\hbar\gamma, \quad (11)$$

which is independent of intensity. In a real situation, of course, other causes of heating would be present, making high intensity desirable. (See, however, Sec. V F). Note that Eq. (11) holds for a single atom or an ensemble of weakly thermalized atoms. If the atoms have a Maxwell-Boltzmann distribution, then $E_{Kx}=\frac{1}{2}k_BT$. If $\gamma=2\pi\cdot 10$ MHz, we find $T_{\min}\cong 2.4\times10^{-4}$ K.

From Eq. (11) the rms velocity $v_{\rm rms}=(\langle v_x^2\rangle)^{1/2}$ can be used to arrive at an approximate Doppler broadening $\Delta\omega_D\cong\omega_0 v_{\rm rms}/c$. In deriving Eq. (11), we have assumed that this Doppler broadening is much less than the natural linewidth, which then implies that $R\ll\hbar\gamma$. In our example, $\hbar\gamma\cong 0.7\times10^{-19}$ ergs $\gg R\cong 4\times10^{-23}$ ergs, and our assumption is justified. However, for weakly allowed transitions in light atoms, this condition will be violated and the minimum kinetic energy will be limited as shown below to approximately the recoil energy R.

When the condition $R\ll\hbar\gamma$ is not satisfied we must accurately evaluate Eq. (7) to find the cooling limit. Assuming the thermalizing collision rate between atoms is large compared to the optical absorption rate, we can evaluate the plasma dispersion function to find the average energy

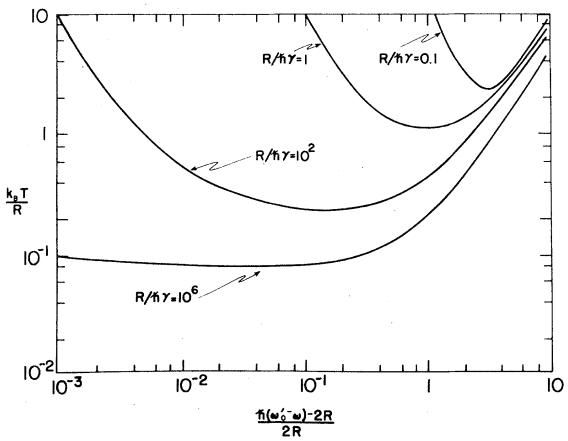

FIG. 3. Plots of $k_BT_{\min}$ versus $\hbar(\omega_0'-\omega)/R$ for four values of $R/\hbar\gamma$. We have assumed that the thermalizing collision rate (γ_c) is much greater than the optical absorption rate.

which makes Eq. (7) vanish for particular values of $\omega_0' - \omega$, R, and γ. (We note that cooling is possible only if $(\omega_0' - \omega) > 2R/\hbar$.) In Fig. 3 we show the minimum energy thus obtained for various values of $\hbar(\omega_0' - \omega)/R$ and $R/\hbar\gamma$. We note that it is possible to cool to an average energy less than the recoil energy; however, we must also note the difficulty in obtaining the proper conditions under which these results hold. In particular, free atoms cannot really be thermalized amongst themselves. Therefore some kind of confinement is required, in which case the required degree of thermal isolation from the environment would be difficult to achieve. (Ideally, we would like a transparent box whose walls have zero heat capacity.) This situation can be nearly realized for electromagnetically bound ions, in which case we might be able to obtain temperatures less than R/k_B, when $\gamma < R/\hbar$. For the atoms described in the previous example ($R \cong 4\times 10^{-23}$ ergs), this would imply $T < 2.7\times 10^{-7}$ K, when the natural linewidth of the transition $\gamma/2\pi$ was less than 6 kHz. We also note that practical limitations on laser power would occur for the case of free atoms since the diffusion of the atoms during the cooling process would require large volumes ($\gg 1$ cm^3).

C. Laser cooling of bound atoms

For the case of bound atoms we consider the special situation where they are harmonically bound in all dimensions. The assumption of harmonic binding is not essential, but it allows a simple interpretation and exhibits the most important features found in any bound system. For simplicity, we consider an atom constrained in the x, y, and z directions by springs of negligible mass as shown in Fig. 4. Of course, in a real situation, the springs might be replaced by electromagnetic restoring forces. We assume that the springs are attached to a massive block and that they act independently and may have different spring constants giving rise to different frequencies of oscillation Ω_x, Ω_y, and Ω_z in the three directions, where we assume $\{\Omega_i\} \ll \omega_0$, $i = x, y, z$.

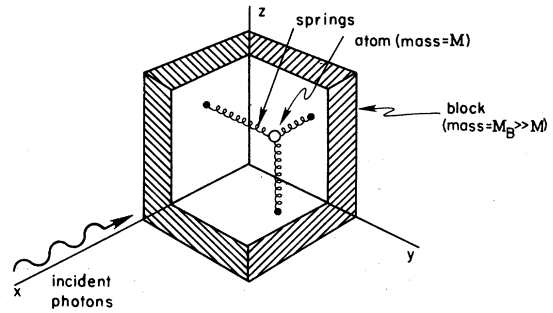

FIG. 4. Idealized representation of bound atom. In this picture the binding forces are due to (massless) springs; in an actual experiment these springs might be replaced by electromagnetic restoring forces.

1. Strong binding

When the binding is "strong", $(\gamma \ll \{\Omega_i\})$, the gross features of the absorption spectrum are significantly altered. We can see this by considering the optical electrical field seen by a particular atom. If the incident (plane-wave) radiation is directed along the x axis,

$$\vec{E}_{\text{atom}} = \vec{E}_0 \sin(kx - \omega t), \tag{12}$$

where x is the atomic x coordinate and k and ω are the wave vector and frequency of the incident radiation. Now

$$x = x_a \sin(\Omega_x t + \phi_x),$$

where x_a is the oscillation amplitude and ϕ_x is a phase factor. Choosing $\phi_x = 0$, we have

$$\vec{E}_{\text{atom}} = \vec{E}_0 \sin(kx_a \sin\Omega_x t - \omega t). \tag{13}$$

This expression is familiar as the signal derived from an oscillator of frequency ω which is frequency modulated at frequency Ω_x with modulation index kx_a. Equation (13) can be expanded in terms of a series of Bessel functions and gives rise to the spectrum shown in Fig. 5(a) when $kx_a = 1.5$. We note that the atom sees a spectrum comprised of a "carrier" at frequency ω with equally spaced first-order Doppler effect gen-

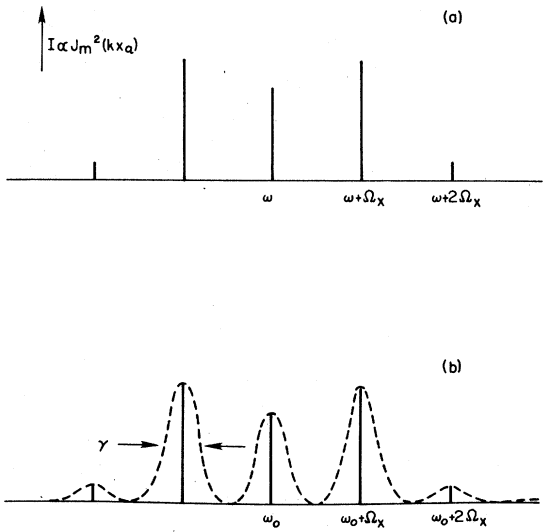

FIG. 5. Radiation spectrum when $\Omega_x \gg \gamma$. Part (a) illustrates the spectrum of an incident wave $\vec{E} = \vec{E}_0 \sin(kx - \omega t)$ as seen by an atom which oscillates according to $x = x_a \cos\Omega_x t$, where we have chosen $kx_a = 1.5$. Part (b) shows the emission spectrum as observed in the laboratory including the broadening due to the lifetime of the upper electronic state.

erated sidebands at the frequencies $\omega + m\Omega_x$ ($m = \pm 1, \pm 2, \pm 3, \ldots$) having intensities proportional to $J_m^2(kx_a)$. Similarly, the spectrum of the atom as seen by an observer in the laboratory will have the same character as in Fig. 5(a), but each component will be broadened by the natural linewidth γ as shown in Fig. 5(b).

To approximate the cooling rate, we note that when $\gamma \ll \Omega_x$ we can tune the incident radiation to one of the resolved lower sidebands. Thus, an atom can be made to absorb photons predominantly of energy $\hbar(\omega_0 + m\Omega_x)$ where m is a (negative) integer. Photons of average energy approximately equal to $\hbar\omega_0$ are reemitted, leading to a net cooling.[2] One can, therefore, approximate the cooling rate to be:

$$\frac{dE}{dt} = (\hbar m\Omega_x)\frac{I\sigma_0}{\hbar\omega} J_m^2(kx_a) . \tag{14}$$

In Sec. V we will see that this expression is strictly valid only when the recoil effect is negligible. For this approximation, as an example, suppose $\Omega_x/2\pi = 12.5$ MHz, $M = 100$ amu, $\lambda = 600$ nm, and $m = -28$. Let $\gamma/2\pi = 1.0$ MHz, and suppose that the intensity is such that $\sigma_0 I/\hbar\omega_0 = 5$ MHz. If the x motion initially has an energy corresponding to room temperature, so that $\frac{1}{2}M\Omega_x^2\langle x^2\rangle = \frac{1}{4}M\Omega_x^2 x_a^2 = \frac{1}{2}k_B T$ ($T = 300$ K) then $x_a \cong 2.8\times 10^{-4}$ cm, $kx_a \cong 30$, $J_{28}^2(kx_0) \cong 0.046$ and $dE/dt \cong 0.3$ eV/sec, $I = 2.7$ mW/cm^2. We therefore see that when the atom is bound, rather substantial cooling can be achieved with modest incident power tuned to a single frequency. (We have, however, chosen a strong sideband). Additionally, we can simultaneously cool all degrees of freedom with a single laser beam if the incident radiation is directed along, for example, the $\hat{i}+\hat{j}+\hat{k}$ direction when the vibration frequencies are related by rational numbers (but are not equal), or if, as in a cloud of ions, the long-range electrostatic forces tend to thermalize all degrees of freedom. This technique[2] has been called "cooling by motional sideband excitation."[5]

In a collection of bound atoms that are weakly thermalized (i.e., the collision rate $\gamma_c \ll \gamma$) a more realistic estimate of the cooling can be obtained if we average Eq. (14) over a thermal distribution. We have

$$\left\langle\frac{dE}{dt}\right\rangle = (\hbar m\Omega_x)\frac{I\sigma_0}{\hbar\omega} \times\left(\frac{1}{k_B T}\int \exp\left(\frac{-E}{k_B T}\right) J_m^2(kx_a)\, dE\right).$$

Since $E = \frac{1}{2}M\Omega_x^2 x_a^2$ the factor in parentheses can be integrated to give the result

$$\left\langle\frac{dE}{dt}\right\rangle = (\hbar m\Omega_x)\frac{I\sigma_0}{\hbar\omega}\exp(-k^2\langle x^2\rangle) \times I_m(k^2\langle x^2\rangle) , \tag{15}$$

where I_m is the modified Bessel function of order m and $k^2\langle x^2\rangle = 2Rk_B T/(\hbar\Omega_x)^2$. If we assume that we irradiate somewhere within the Doppler profile, then we can show [see Eq. (41)] that $|m| \lesssim k\langle x^2\rangle^{1/2}$. In the high-temperature or short-wavelength limit, $k(\langle x^2\rangle)^{1/2} \gg 1$. Therefore we can use the following asymptotic expansion[13] of I_m:

$$I_m(z) \to (2\pi z)^{-1/2}\exp(z - m^2/2z) \quad z \gg m \gg 1 \tag{16}$$

and we obtain

$$\left\langle\frac{dE}{dt}\right\rangle = (\hbar m\Omega_x)\frac{I\sigma_0}{\hbar\omega}\ \frac{\Omega_x}{\sqrt{\pi}\,\omega_D}\exp\left(\frac{-(\omega-\omega_0)^2}{\omega_D^2}\right), \tag{17}$$

where $\omega - \omega_0 = m\Omega_x$. The last factor shows the Doppler profile. For the above conditions, $\langle dE/dt\rangle \cong 0.057$ eV/sec.

The above description is not quite correct because we have not considered in detail momentum and energy conservation in the process of absorption and reemission. It happens that the absorption and emission spectra are also altered by recoil. Qualitatively we can say that in the emission spectrum, for example, components occur at frequencies ω_0 and $\omega_0 + m\Omega_x$, but the spectrum is now slightly skewed. For example, suppose we look at the emissions at $\omega_0 \pm \Omega_x$ when $kx_a < 1$. For photon emission at frequency $\omega_0 - \Omega_x$, conservation of energy tells us that the energy of oscillatory motion must increase; hence, during the emission, kx_a increases and so also does the amplitude of the spectral component at $\omega_0 - \Omega_x$. The opposite argument holds for the component at $\omega_0 + \Omega_x$ and, consequently, the average energy spectrum is shifted to a value slightly below $\hbar\omega_0$. The solution to this problem is straightforward when quantum mechanics is used and, therefore, we defer discussion to Sec. V. It has been treated by a semiclassical approach[7] when $kx_a \ll 1$.

A further complication arises when we ask for the cooling limit when $\gamma \ll \Omega_x$. We find that the minimum kinetic energy achieved from a classical treatment is less than the zero-point energy of the harmonic oscillator and therefore we defer this discussion to the quantum-mechanical treatment.

At this point we can see the similarity of this problem (when $\gamma \ll \Omega_x$) to the Mössbauer effect. Indeed, when $kx_a \lesssim 1$, the central peak or "carrier" becomes dominant; excitation at this central peak is equivalent to absorption on the Mössbauer line—the main difference being that at γ-ray energies the recoil plays a much larger role. As

we will see below, the quantum-mechanical treatment of laser cooling can use some of the same formalism as the treatment of the Mössbauer effect.

Finally, another picture which provides a connection with a familiar situation is given by considering the "molecule" formed by the block and the atom bound to it. Cooling of the vibrational motion is achieved by anti-Stokes Raman scattering. However, the ordinary selection rules for vibrational transitions do not apply in general since the "molecular" dimensions may be considerably larger than the optical wave length.

2. *Weak binding*

When $\gamma \gg \Omega_i$, the period of the atomic center-of-mass oscillation is much larger than the optical decay time. Therefore, during one oscillation period the atomic velocity gradually reaches the value where the incident radiation is shifted into resonance; scattering occurs, and for each scattering event Eq. (3) holds. In this limit we have treated the interaction of the radiation with the atom as occurring at an instant of time. This is reasonable since the atom loses optical phase memory during one oscillation period and therefore coherence during successive oscillations (which gives rise to the resolved sidebands) can be neglected. An important difference from the free-atom case is that the momentum of the photon is transferred to the center-of-mass motion of the block and atom. In the limit that the mass of the block becomes infinite, the change in velocity of the center of mass, per scattering event, goes to zero and the energy change must occur in the kinetic energy of the bound atom.

Therefore, in the weak binding limit we may apply Eq. (5) and find the average cooling rate by averaging v_x over one center-of-mass oscillation period. In an ensemble of weakly thermalized atoms Eqs. (7) and (8) therefore also apply. Neglecting recoil, we obtain Eqs. (2) and (3) of Ref. 6. However, if we assume only one laser beam is present to do the cooling we must assume that the transition rate is small enough to avoid heating by recoil in directions perpendicular to the laser beam. (See Sec. V F). If three mutually perpendicular laser beams are used to do the cooling this restriction is not necessary, but the power must be held low enough to avoid the effects of the induced dipole force. Note that the estimated cooling limit given in Ref. 6 is too low by a factor of 2. This is because only the recoil heating in the absorption process was considered and not the contribution from reemission.

D. Generalization of the cooling process

In this paper we address a rather specific problem, that is, the laser cooling of atoms. Abstracting the problem slightly, we have excited one "oscillator"—the atom's internal electronic resonance at frequency ω_0—with radiation at a lower frequency ω. In the frame of the harmonically bound atom, this excitation occurs due to one or more of the upper sidebands shown in Fig. 5(a). For the free atom these "sidebands" become a single line.

Instead of the internal atomic "oscillator," consider an electron (or in general a charged particle) which is constrained like the atom shown in Fig. 4. Further suppose that the resonance frequencies Ω_y, Ω_z are very high so that $\hbar\Omega_y, \hbar\Omega_z \gg \hbar\Omega_x$. If incident radiation is polarized along the z axis and is directed along the x axis at frequency $\Omega_z - \Omega_x$, then the z oscillation is excited much the same way that the electronic transition in the atom was excited by the frequency $\omega_0 - \Omega_x$. Thus, cooling of the x motion can be achieved at the expense of driving (heating) the z motion. This is the essence of "magnetron" cooling which was demonstrated for an electron bound in a Penning trap.[4] Treating this problem classically is reasonably straightforward,[5,14,15] but a general quantum-mechanical treatment is complicated by two things. (a) Recall that for the laser cooling of atoms, the electronic states can be assumed to comprise a two-level system which is not saturated; this is a good approximation for many practical situations. However, when the atomic transition is replaced by the harmonic oscillator, many levels are involved and saturation between levels can occur. (b) In addition, the condition $k_B T \ll \hbar\Omega_y, \hbar\Omega_z$ is not generally satisfied and one must contend with the effects of thermal background radiation. Nevertheless, certain cases of this more general problem can be treated.[16] Fortunately, these two problems usually do not occur for the laser cooling of atoms and many aspects of the quantum-mechanical treatment are quite straightforward.

We remark that the velocity selectivity provided by the narrow-band (laser) source might be provided by other means. For example, assume the atom is polarized in the z direction and is constrained to move along the x axis. If white light irradiates the atom at an angle with respect to the x axis, then, because the absorption rate is dependent on the angle the radiation is received (in the frame of the atom), we obtain a differential effect which is dependent on the atom velocity as is required above. This is analogous to the state selectivity provided by optical polarization in the

case of ordinary optical pumping.

We note that in terms of the above description we are discussing the case of a nearly ideal refrigerator: for each scattering event (i) we extract an amount of energy $\hbar\Omega_x$ from the "x reservoir" at temperature T_x, (ii) we supply an amount of energy $\hbar(\Omega_z-\Omega_x)$, and (iii) we transfer the amount of energy $\hbar(\Omega_z-\Omega_x)+\hbar\Omega_x=\hbar\Omega_z$ to the "z reservoir" at temperature T_z. The second law of thermodynamics then implies that the minimum obtainable temperature T_x is given by the relation[17]

$$T_x/T_z \geq \Omega_x/\Omega_z . \tag{18}$$

The competing heating mechanism which gives rise to this limit arises because the thermally excited z motion (frequency Ω_z) modulates the Lorentz force due to the incoming radiation (frequency $\Omega_z-\Omega_x$) and gives a component at frequency Ω_x which then excites the x motion.[18]

In the following we will assume that $T_z=0$; equivalently, we will assume that the atoms are normally in their ground electronic state and the background blackbody radiation is negligible. Then the minimum temperature achieved will be determined by recoil effects. We must, however, keep the thermodynamic limit in mind in certain cases.

We further remark that in terms of the above description, the cooling process is entirely analogous to the Overhauser effect where the flipping of an electron spin in a solid is accompanied by a flipping of the nuclear spin system.[17]

We also note that cooling by the technique of collisionally aided fluorescence[9] shares many similarities with cooling described here. When collisions are present, the atoms' absorption spectrum is broadened, and, if the collisions were very regular like the ion "collisions" with the electric restoring forces in an electromagnetic trap, then the spectrum might appear as in Fig. 5(b). Of course, for the general case of collisions the line broadening is not so regular; however, the cooling principle still applies, so that if we irradiated the atom at a frequency lower than the center of gravity of its emission spectrum, cooling would occur.

Finally, we observe that the cooling process can be described in terms of radiation pressure. The radiation pressure force on atoms has usually been developed in terms of scattering[1,10]; however, we note that an alternative approach sometimes used in elementary texts[19] describes the radiation pressure force on a conductor in terms of the Lorentz force interaction of the oscillating magnetic field with the induced current. For the atomic case, a dynamic polarization is established which is usually given in terms of the susceptibility. This atomic polarization interacts with the magnetic component of the radiation field via the Lorentz force leading to a force in the direction of the radiation:

$$\vec{F}=\frac{d\vec{p}}{dt}=\frac{I}{\hbar\omega}\sigma\hbar\vec{k} .$$

The effects of recoil must be added separately in this approach. The treatment of the problem as scattering has the advantage of completeness and is therefore used in the following. We finally remark that the interest in radiation pressure forces has had a long history. Some of the very early experimental and theoretical papers have been summarized by Nichols and Hull.[20] The first experiment observing radiation pressure on atoms was reported by Frisch.[21]

III. QUANTUM-MECHANICAL TREATMENT—GENERAL ASPECTS

For the quantum-mechanical treatment we will make some simplifying assumptions in order to illustrate the basic features of the problem. We can, however, assume that the atom (or molecule, or resonant absorber in general) moves in three dimensions. We assume that the motions in the different directions are independent, although it will sometimes be convenient to assume for a collection of atoms that they are thermalized with each other by some relatively weak long-range forces yielding a collision rate $\gamma_c \ll \gamma$. It is also assumed that the time required for the kinetic energy to thermalize with the outside environment is extremely long. This situation is closely approximated for a cloud of ions stored in an electromagnetic trap, for example. We further assume that the internal structure of the atom comprises a two-level system with ground-state energy E_g and excited-state energy E_e such that $E_e-E_g \gg k_BT$, where T is the ambient temperature. Within the limits imposed by thermodynamics (Sec. II D) this allows us to neglect the effects of background blackbody radiation; that is, in the absence of laser irradiation, we assume that the atoms are in the ground state. Finally, we assume that the incident radiation is highly monochromatic (spectral width $\ll\gamma$) and that the intensity is well below saturation.

In the following we will assume that the Hamiltonian for the system can be written

$$H=H_0+H' ,$$

where H_0 is the unperturbed Hamiltonian and H' describes the interaction of the radiation with the atom. We have

$$H_0=H_a+H_t+H_{\text{rad}}$$

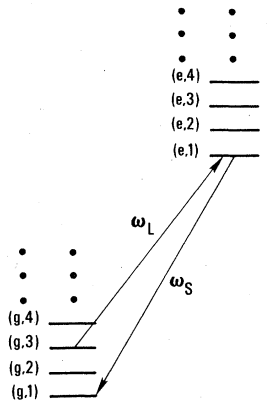

FIG. 6. Pictorial representation of electronic [ground (g) and excited (e)] energy states and translational energy states (denoted by integers) for an atom. When the atom is very weakly bound or unbound the spacing between translational energy levels goes to zero. The cooling can be described as anti-Stokes Raman scattering or optical pumping where the frequency (ω_s) of the scattered photon is greater than the frequency (ω_L) of the "laser" photon.

where H_a is the Hamiltonian for the two internal states of the atom, H_t is the Hamiltonian for the translational degrees of freedom, and $H_{\rm rad}$ is the Hamiltonian of the radiation field. Assuming that the various degrees of freedom are not coupled in the absence of H', we write the total wave function as a direct product:

$$|\psi\rangle = |\mathrm{int}\rangle|\mathrm{trans}\rangle|\mathrm{rad}\rangle\ .$$

In the absence of the radiation field, the internal and translational energy levels are depicted in Fig. 6. In the nonrelativistic limit, and assuming low intensity, we make the usual approximation

$$H' \simeq (e/mc)\vec{\mathrm{P}}_{\rm el}\cdot\vec{\mathrm{A}}(\vec{\mathrm{R}}_{\rm el})\ ,$$

where $\vec{\mathrm{P}}_{\rm el}$ and $\vec{\mathrm{R}}_{\rm el}$ are the momentum and position of the optically active electron, $\vec{\mathrm{A}}$ is the vector potential of the radiation field evaluated at the position of the electron, and m is the electron mass. We have neglected the $\vec{\mathrm{A}}^2$ term in the Hamiltonian.[22]

For the cooling problem, we are primarily interested in a scattering process that changes the translational and radiation states but leaves the internal atomic state unchanged. In particular, we can formulate the problem by asking for the cross section for scattering a photon of wave vector $\vec{\mathrm{k}}$ and polarization $\hat{\epsilon}$ where the translational energy has changed from $E_l(\mathrm{trans})$ to $E_f(\mathrm{trans})$ and where the scattered photon has wave vector $\vec{\mathrm{k}}_s$ and polarization $\hat{\epsilon}_s$. The differential cross section for this process can be obtained as a particular result from the Kramers-Heisenberg formula, which can be derived from time-dependent perturbation theory.[22] Specifically, the differential scattering cross section for scattering from initial state l to final state f can be written

$$\frac{d\sigma_{l\to f}}{d\omega_s d\Omega} = \sum_s \frac{\omega_s}{\omega}\left(\frac{r_e}{m\hbar}\right)^2 \left|\sum_j \frac{\langle f|\hat{\epsilon}_s\cdot\vec{\mathrm{P}}_{\rm el}\exp(-i\vec{\mathrm{k}}_s\cdot\vec{\mathrm{R}}_{\rm el})|j\rangle\langle j|\hat{\epsilon}\cdot\vec{\mathrm{P}}_{\rm el}\exp(i\vec{\mathrm{k}}\cdot\vec{\mathrm{R}}_{\rm el})|l\rangle}{\omega_j-\omega-\frac{1}{2}i\gamma}\right|^2$$

$$\times\,\delta(\omega-\omega_s-[E_f(\mathrm{trans})-E_l(\mathrm{trans})]/\hbar)\ , \qquad (19)$$

where $d\Omega$ is the differential element of solid angle into which the photons are scattered, ω is the incident photon frequency ($|\vec{\mathrm{k}}| = \omega/c$), $\hbar\omega_j = \hbar\omega_0 + E_j(\mathrm{trans}) - E_l(\mathrm{trans})$, r_e is the classical electron radius e^2/mc^2, subscript s denotes the two possible directions of the polarization of the scattered photon, j denotes the intermediate state, and where the wave functions now describe only the electronic and translational degrees of freedom

$$|j\rangle = |\mathrm{int}(j)\rangle|\mathrm{trans}(j)\rangle\ .$$

The δ function in Eq. (19) ensures conservation of energy for the scattering process; that is, $\hbar(\omega_s-\omega) = E_l(\mathrm{trans}) - E_f(\mathrm{trans})$. The operator in the matrix elements of Eq. (19) is proportional to H'. From this equation we see qualitatively that the process is described as the absorption of a photon which promotes the atom to a distribution of possible intermediate (virtual) states $\{j\}$ (where energy need not be conserved) followed by spontaneous reemission. The denominator indicates that a resonance is involved in the absorption. This is the main cause for the cooling, since a resonance can occur when the kinetic energy of the atom changes, that is, when $\omega = \omega_0 + [E_j(\mathrm{trans}) - E_l(\mathrm{trans})]/\hbar$. Equation (19) can, of course, also describe ordinary Raman and Rayleigh scattering.[22]

To evaluate the matrix elements in Eq. (19), it is useful to express the electron position and momentum as functions of the atomic center-of-mass position and momentum $\vec{\mathrm{x}}$ and $\vec{\mathrm{P}}$, the position of the electron relative to the atomic core $\vec{\mathrm{r}}$, and the velocity of the electron relative to the atomic core $\vec{\mathrm{p}}/m_r$ (m_r in this expression is the electron reduced mass given by $m(1+m/M_c)^{-1}$, where M_c is the mass of the core. In the approximation that $m \ll M_c$ we have:

$$\vec{R}_{el} = \vec{x} + \vec{r}\,, \tag{20a}$$

$$\vec{P}_{el} = \frac{m}{M}\vec{P} + \vec{p}\,, \tag{20b}$$

where $M = M_c + m$.

Before applying Eq. (19), we make the qualitative observation that the cooling process can be represented as in Fig. 6, where we have illustrated a process involving specific initial, intermediate, and final states. In general we must, of course, sum over all possible intermediate states, average over the initial-state distribution, and sum over all final states to get the net result. We see, however, that the cooling process can be described as spontaneous anti-Stokes Raman scattering or as a type of optical pumping as described in Sec. II.

IV. LASER COOLING OF FREE ATOMS

A. Matrix elements

For a free atom we have

$$H_t = \frac{\vec{P}^2}{2M}$$

and the spatial eigenfunctions are

$$|\text{trans}\rangle = C_N \exp(i\vec{K}\cdot\vec{x}) \equiv |\vec{K}\rangle\,,$$

where C_N is a normalization constant and

$$\langle\vec{K}|\vec{P}|\vec{K}\rangle = \hbar\vec{K}\,,\quad \langle\vec{K}|H_t|\vec{K}\rangle = (\hbar K)^2/2M\,,$$

$$\langle\vec{K}'|\vec{K}\rangle = \delta(\vec{K}',\vec{K})\,,$$

where δ denotes the Kronecker δ.

Using Eqs. (20), the matrix elements in Eq. (19) take the form:

$$\begin{aligned}\langle j|\hat{\epsilon}\cdot\vec{P}_{el}\exp(i\vec{k}\cdot\vec{R}_{el})|l\rangle &= \left\langle\vec{K}'\left|\hat{\epsilon}\cdot\frac{m}{M}\vec{P}\,\exp(i\vec{k}\cdot\vec{x})\right|\vec{K}\right\rangle\langle e|\exp(i\vec{k}\cdot\vec{r})|g\rangle \\ &+\langle\vec{K}'|\exp(i\vec{k}\cdot\vec{x})|\vec{K}\rangle\langle e|\hat{\epsilon}\cdot\vec{p}\exp(i\vec{k}\cdot\vec{r})|g\rangle\,,\end{aligned} \tag{21}$$

where e and g denote the internal atomic excited and ground states, respectively.

Qualitatively, the first term in Eq. (21) represents a transition of the center-of-mass motion which is caused by a field component resulting from the laser field "modulated" by the internal electronic oscillation. The second term represents an electronic transition which is caused by a field component of the laser field which has been Doppler shifted by the motion. For both terms, however, a simultaneous change in electronic and kinetic energy state has occurred.

We have

$$[\hat{\epsilon}\cdot\vec{P}, \exp(i\vec{k}\cdot\vec{x})] = 0 \text{ (Coulomb gauge)}\,.$$

Therefore,

$$\langle\vec{K}'|\hat{\epsilon}\cdot\frac{m}{M}\vec{P}\exp(i\vec{k}\cdot\vec{x})|\vec{K}\rangle = (m/M)\hbar\vec{K}\cdot\hat{\epsilon}\,\delta(\vec{K}+\vec{k},\vec{K}')\,, \tag{22a}$$

$$\langle\vec{K}'|\exp(i\vec{k}\cdot\vec{x})|\vec{K}\rangle = \delta(\vec{K}+\vec{k},\vec{K}')\,, \tag{22b}$$

$$\langle e|\exp(i\vec{k}\cdot\vec{r})|g\rangle \simeq i\vec{k}\cdot\langle\vec{r}\rangle\,, \tag{22c}$$

$$\langle e|\hat{\epsilon}\cdot\vec{p}\exp(i\vec{k}\cdot\vec{r})|g\rangle \simeq im\omega_0\hat{\epsilon}\cdot\langle\vec{r}\rangle\,, \tag{22d}$$

where we have made the usual dipole approximation ($1/|\vec{k}| \gg$ atomic dimensions) and have defined $\langle\vec{r}\rangle = \langle e|\vec{r}|g\rangle$. The ratio of the first term to the second term in Eq. (21) can be written:

$$\frac{\hbar(\vec{K}\cdot\hat{\epsilon})\vec{k}\cdot\langle\vec{r}\rangle}{M\omega_0\hat{\epsilon}\cdot\langle\vec{r}\rangle} \simeq \frac{v}{c}\,.$$

In the nonrelativistic approximation used here, the first term is therefore negligible and Eq. (19) becomes:

$$\frac{d\sigma_{l\to f}}{d\omega_s d\Omega} = C\sum_s \frac{\omega_s}{\omega}|\hat{\epsilon}_s\cdot\langle\vec{r}\rangle|^2|\hat{\epsilon}\cdot\langle\vec{r}\rangle|^2\left|\sum_j \frac{\langle\vec{K}_f|\exp(-i\vec{k}_s\cdot\vec{x})|\vec{K}_j\rangle\langle\vec{K}_j|\exp(i\vec{k}\cdot\vec{x})|\vec{K}_l\rangle}{\omega_0-\omega+(\hbar/2M)(K_j^2-K_l^2)-\frac{1}{2}i\gamma}\right|^2\delta(\omega-\omega_s-(\hbar/2M)(K_f^2-K_l^2))$$

or equivalently:

$$\frac{d\sigma_{l\to f}}{d\omega_s d\Omega} = C\sum_s \frac{\omega_s}{\omega}|\hat{\epsilon}_s\cdot\langle\vec{r}\rangle|^2|\hat{\epsilon}\cdot\langle\vec{r}\rangle|^2\frac{\delta(\vec{K}_f+\vec{k}_s,\vec{K}_l+\vec{k})\delta(\omega-\omega_s-(\hbar/2M)(K_f^2-K_l^2))}{[\omega_0-\omega+(\hbar/2M)(k^2+2\vec{k}\cdot\vec{K}_l)]^2+\frac{1}{4}\gamma^2}\,, \tag{23}$$

where $C = (r_e m\omega_0^2/\hbar)^2$. The δ functions in this equation express conservation of momentum and energy in the scattering process. We set $\vec{K}_f = \vec{K}_l + \vec{k} - \vec{k}_s$. After some manipulation, which is outlined in Ref. 23, Eq. (23) can be put in the form

$$\frac{d\sigma_{l\to f}}{d\omega_s d\Omega} = \frac{C\sum_s(\omega_s/\omega)|\hat{\epsilon}\cdot\langle\vec{r}\rangle|^2|\hat{\epsilon}\cdot\langle\vec{r}\rangle|^2\cdot N\delta(\omega_s-\omega_+)}{[\omega_0-\omega+(\hbar/2M)(k^2+2\vec{k}\cdot\vec{K}_l)]^2+\frac{1}{4}\gamma^2}\,, \tag{23a}$$

where N is an angular factor of order unity which depends on the angles between $\vec{k}$, $\vec{k}_s$, and $\vec{K}_i$. An exact expression for N is given in Ref. 23. Neglecting terms of order $R/\hbar\omega_0$, β^2 and higher, we have

$$N \simeq 1+\beta_i \cos\theta_{is}\,, \tag{23b}$$

where θ_{is} is the angle between $\vec{K}_i$ and $\vec{k}_s$ and $\beta_i = \hbar K_i/Mc$. ω_+ is the frequency of the scattered photon[23] and, neglecting the same order of terms as above, is given approximately by

$$\omega_+ \simeq \omega - (2R/\hbar)(1-\cos\theta_s) - \beta_i\omega(\cos\theta_i - \cos\theta_{is})\,, \tag{23c}$$

where θ_s is the angle between $\vec{k}$ and $\vec{k}_s$ and θ_i is the angle between $\vec{k}$ and $\vec{K}_i$.

B. Cross sections

The total cross section for scattering of the laser light by an atom in the translational state $|\vec{K}_i\rangle$, obtained by integrating the differential cross section obtained from Eq. (23a) over all angles of the scattered photon, is (neglecting terms of order $R/\hbar\omega_0$, β^2, and $(\omega-\omega_0)/\omega_0$)

$$\sigma_i = \int d\Omega\, d\omega_s \frac{d\sigma_{i\to f}}{d\omega_s d\Omega} = \frac{C\frac{8}{3}\pi|\hat{\epsilon}\cdot\langle\vec{r}\rangle|^2|\langle\vec{r}\rangle|^2}{(\omega_{\rm abs}-\omega)^2+\frac{1}{4}\gamma^2}\,,$$

where $\omega_{\rm abs}$ is given by Eq. (2a) (neglecting the β^2 term). Since γ is equal to $4e^2|\langle\vec{r}\rangle|^2\omega_0^3/(3\hbar c^3)$, then

$$\sigma_i = 6\pi\lambda^2|\hat{\epsilon}\cdot\langle\hat{r}\rangle|^2(\tfrac{1}{2}\gamma)^2/[(\omega_{\rm abs}-\omega)^2+(\tfrac{1}{2}\gamma)^2]\,, \tag{24}$$

where $\langle\hat{r}\rangle$ is the unit vector corresponding to $\langle\vec{r}\rangle$. If the orientation of the atom with respect to the polarization vector $\hat{\epsilon}$ is random, the average value of $|\hat{\epsilon}\cdot\langle\hat{r}\rangle|^2$ is $\frac{1}{3}$, and we arrive at Eq. (4). However, in a particular experimental situation, the orientation of $\langle\hat{r}\rangle$ and $\hat{\epsilon}$ may be fixed, in which case this averaging should not be performed.

We will assume as in Sec. II that we have six laser beams of equal intensity directed along the $\pm x, \pm y, \pm z$ directions. To avoid the effects of the induced dipole forces we must make the same approximations as in the classical treatment. In this case the initial wave function describing the radiation field becomes:

$$|n(\vec{k})\rangle = |n(\vec{k}_x), n(-\vec{k}_x), n(\vec{k}_y), n(-\vec{k}_y), n(\vec{k}_z), n(-\vec{k}_z)\rangle$$

where the occupation numbers are the same and $|\vec{k}_i| = k$. Equation (23) then becomes

$$\frac{d\sigma_{i\to f}}{d\omega_s d\Omega} = C\sum_s \frac{\omega_s}{\omega}|\hat{\epsilon}_s\cdot\langle\vec{r}\rangle|^2 \sum_m \frac{|\hat{\epsilon}_m\cdot\langle\vec{r}\rangle|^2\delta(\vec{K}_f+\vec{k}_s, \vec{K}_i+\vec{k}_m)\delta(\omega-\omega_s-(\hbar/2M)(K_f^2-K_i^2))}{[\omega_0-\omega+(\hbar/2M)(k^2+2\vec{k}_m\cdot\vec{K}_i)]^2+\frac{1}{4}\gamma^2}\,, \tag{25}$$

where the sum over m represents the sum over the directions of the six laser beams.

C. Cooling rates and limits

For each scattering event, the average change in the kinetic energy of an atom initially in the state $|\vec{K}_i\rangle$ is the negative of the average change in energy of the scattered photon and is equal to [with σ_i given by Eq. (24)]

$$\int d\Omega_s\, \hbar(\omega-\omega_s)\frac{d\sigma_{i\to f}}{d\Omega_s}\Big/\sigma_i = 2R + \hbar\vec{k}\cdot\vec{v}_i \tag{26}$$

in agreement with Eq. (3). Again, smaller terms of order $\beta_i^2\hbar\omega_0$, $\beta_i R$, and higher have been neglected. This same result was obtained in Sec. II by averaging over all possible directions of the scattered photon. The net cooling rate is given by

$$\left\langle\frac{d(E_f-E_i)}{dt}\right\rangle = \frac{I}{\hbar\omega}\int d\Omega \sum_{f,i} P_i(E_f-E_i)\frac{d\sigma_{i\to f}}{d\Omega}\,, \tag{27}$$

where I is the intensity of each laser beam, P_i is the probability for an atom to be in the initial state $|\vec{K}_i\rangle$, and $d\sigma_{i\to f}/d\Omega$ is given by Eq. (25). With the help of Eq. (26), we can write

$$\left\langle\frac{d(E_f-E_i)}{dt}\right\rangle = \frac{I\sigma_0}{2\hbar\omega}\sum_{m,i}|\hat{\epsilon}_m\cdot\langle\hat{r}\rangle|^2|G_{m,i}|^2 \times\left(2R+\frac{\hbar^2\vec{K}_i\cdot\vec{k}_m}{M}\right)P_i\,,$$

where

$$(1/\gamma')G_{m,i} \equiv [\omega_0+(\hbar/2M)(k_m^2+2\vec{k}_m\cdot\vec{K}_i)-\omega-\tfrac{1}{2}i\gamma]^{-1}$$

and $\gamma' = \frac{1}{2}(1-i)\gamma$. If we assume that the Doppler width is smaller than the natural width ($\hbar\vec{k}_m\cdot\vec{K}_i/M \ll \gamma$) and write $\omega = \omega_0 + R/\hbar - \frac{1}{2}\gamma + \Delta\omega$ we can write when $\Delta\omega \ll \gamma$

$$G_{m,i} \simeq 1 + \Delta\omega/\gamma' - (\hbar\vec{k}_m\cdot\vec{K}_i)/\gamma' M\,.$$

Assuming P_i is isotropic, the cooling rate is given by Eq. (10) (maximum cooling for $\Delta\omega = 0$) and the corresponding limit by Eq. (11). Similarly if the Doppler width is not negligible com-

pared to the natural width, we obtain Eq. (7), if P_i is given by a Maxwell-Boltzmann distribution. Consequently, the limits of Fig. 3 are also obtained. Thus the fully quantum-mechanical treatment is not necessary to describe the cooling for the case of free atoms. We shall see, however, that it is necessary for an accurate description of bound atoms.

V. LASER COOLING OF BOUND ATOMS

A. Matrix elements

To study the cooling process when the atoms are bound we will assume for simplicity that the atoms are harmonically bound in three dimensions as discussed in Sec. II. This case may not be realized in practical systems, but it may be a close approximation; moreover, it illustrates the important features of the problem. Thus we have

$$H_t = \frac{\vec{P}^2}{2M} + \tfrac{1}{2}M(\Omega_x^2 x^2 + \Omega_y^2 y^2 + \Omega_z^2 z^2) .$$

The solution for the eigenstates of a harmonic oscillator appears in many texts; using the operator formalism we have

$$H_t = \sum_{i=x,y,z} \hbar\Omega_i (N_i + \tfrac{1}{2}) , \quad N_i = a_i^\dagger a_i ,$$

where a_i and $a_i^\dagger$ are the lowering and raising operators for the states satisfying $[a_i, a_i^\dagger] = \delta_{ij}$. The position and momentum operators are given by

$$x = x_0(a_x + a_x^\dagger) , \quad P_x = ix_0 M\Omega_x (a_x^\dagger - a_x) .$$

$$x_0 \equiv (\hbar/2M\Omega_x)^{1/2} , \tag{28}$$

and similarly for y and z. We have $\Psi = \Psi_x\Psi_y\Psi_z$, where in the above representation

$$\Psi_i = |n_i\rangle \qquad \langle n_i|n_j\rangle = \delta_{n_i n_j}\delta_{ij}$$

$$a_i|n_i\rangle = (n_i)^{1/2}|n_i - 1\rangle \qquad \Psi = |n_x\rangle|n_y\rangle|n_z\rangle$$

$$a_i^\dagger|n_i\rangle = (n_i + 1)^{1/2}|n_i + 1\rangle \quad i,j = x,y,z$$

and in the Schrödinger representation[24]:

$$|n_x\rangle = (2^{n_x} n_x! \pi^{1/2})^{-1/2} \exp(-M\Omega_x x^2/2\hbar) \times H_{n_x}[(M\Omega_x/\hbar)^{1/2} x] , \tag{29}$$

where H_{n_x} is the Hermite polynomial of order n_x.

From Eqs. (19) and (20) we will be interested in matrix elements of the form

$$\langle n'|\hat{\epsilon}\cdot(m/M)\vec{P}\exp(i\vec{k}\cdot\vec{x})|n\rangle\langle e|\exp(i\vec{k}\cdot\vec{r})|g\rangle + \langle n'|\exp(i\vec{k}\cdot\vec{x})|n\rangle\langle e|\hat{\epsilon}\cdot\vec{p}\exp(i\vec{k}\cdot\vec{r})|g\rangle , \tag{30}$$

and therefore we are interested in harmonic oscillator matrix elements of the form

$$\langle n'|\exp(i\vec{k}\cdot\vec{x})|n\rangle = \langle n_x'|\exp(ik_x x)|n_x\rangle\langle n_y'|\exp(ik_y y|n_y\rangle \times \langle n_z'|\exp(ik_z z)|n_z\rangle .$$

Therefore, in general, we are interested in matrix elements of the type $\langle n'|\exp(ikx)|n\rangle$, where we have dropped the subscripts.

These matrix elements can be straightforwardly evaluated from the explicit form of the wave functions given in Eq. (29) or by the following operator method. Since

$$e^{A+B} = e^A e^B e^{-[A,B]/2}$$

when

$$[A,[A,B]] = [B,[A,B]] = 0 ,$$

we have

$$\exp(ikx) = \exp[ikx_0(a^\dagger + a)] = \exp[-\tfrac{1}{2}(kx_0)^2]\exp(ikx_0 a^\dagger) \times \exp(ikx_0 a) .$$

Since

$$a^m|n\rangle = \begin{cases} [n!/(n-m)!]^{1/2}|n-m\rangle & m \le n \\ 0 & m > n \end{cases}$$

then

$$\exp(ikx_0 a)|n\rangle = \sum_{m=0}^{\infty} \frac{(ikx_0)^m}{m!}\left(\frac{n!}{(n-m)!}\right)^{1/2}|n-m\rangle .$$

Thus the matrix elements can be written

$$\langle n'|\exp(ikx)|n\rangle = \exp[-\tfrac{1}{2}(kx_0)^2] \times \langle n'|\exp(ikx_0 a^\dagger)\exp(ikx_0 a)|n\rangle .$$

Operating to the right with $\exp(ikx_0 a)$ and to the left with $\exp(ikx_0 a^\dagger)$ and using orthogonality, we obtain

$$\langle n'|\exp(ikx)|n\rangle = \exp[-\tfrac{1}{2}(kx_0)^2](n!\,n'!)^{1/2}(ikx_0)^{\Delta n} \times \sum_{m=0}^{n_<} \frac{(-1)^m (kx_0)^{2m}}{m!\,(m+\Delta n)!\,(n_< - m)!} ,$$

where $\Delta n = |n' - n|$ and $n_<$ is the lesser of n and n'. From the explicit form of the generalized Laguerre polynomial

$$L_m^\alpha(X) = \sum_{m=0}^{n} (-)^m \binom{n+\alpha}{n-m}\frac{X^m}{m!}$$

we have

$$\langle n'|\exp(ikx)|n\rangle = \exp[-\tfrac{1}{2}(kx_0)^2][n_<!/(n_<+\Delta n)!]^{1/2} \times (ikx_0)^{\Delta n} L_{n_<}^{\Delta n}[(kx_0)^2] . \quad (31)$$

In the following we will see that the cooling problem for bound absorbers shares many similarities with the Mössbauer effect; therefore, some of the same mathematics[25,26] can be employed. In the study of the Mössbauer effect, one typically looks at the absorption and emission processes separately; for the cooling problem we recall that we are primarily interested in a scattering process. Nevertheless, it will be useful to look at the absorption and emission processes separately in the cooling problem as well. With the help of Eqs. (22c) and (22d), Eq. (30) reduces to

$$im\omega_0\langle n'|\exp(i\vec{k}\cdot\vec{x})(\hat{\epsilon}\cdot\langle\vec{r}\rangle - \frac{\vec{k}\cdot\langle\vec{r}\rangle}{M\omega_0}\hat{\epsilon}\cdot\vec{P})|n\rangle$$

and this expression can be substituted into Eq. (19) to obtain the cross section.

B. Spontaneous emission

With the approximation of low laser intensity used here, the scattered photon results from spontaneous emission. Therefore, if we look at the emission process separately, we are interested in amplitudes which are matrix elements of the operator:

$$A_{sp} \equiv \exp(-i\vec{k}_s\cdot\vec{x})[\hat{\epsilon}_s\cdot\langle\vec{r}\rangle + (\vec{k}_s\cdot\langle\vec{r}\rangle\hat{\epsilon}_s\cdot\vec{P})/(M\omega_0)] . \quad (32)$$

Strictly speaking, we must sum over all amplitude contributions of the intermediate states n_j as in Eq. (19); however, if only one intermediate state n_j is excited, which is a good approximation when $\gamma \ll \Omega_i$, then the probability for reemission is proportional to $|\langle n_f|A_{sp}|n_j\rangle|^2$.

It is instructive to first look at the average energy change of the translational energy states in the emission process. We have

$$\langle E_f - E_j\rangle = \frac{\int d\Omega\sum\sum_{sf}(E_f-E_j)|\langle n_f|A_{sp}|n_j\rangle|^2}{\int d\Omega\sum\sum_{sf}|\langle n_f|A_{sp}|n_j\rangle|^2} ,$$

where we can write

$$\sum_f (E_f-E_j)|\langle n_f|A_{sp}|n_j\rangle|^2 = \sum_f \langle n_j|A_{sp}^\dagger|n_f\rangle\langle n_f|[H_t,A_{sp}]|n_j\rangle = \langle n_j|A_{sp}^\dagger[H_t,A_{sp}]|n_j\rangle .$$

Using the expression

$$[H_t, \exp(-i\vec{k}_s\cdot\vec{x})] = [\exp(-i\vec{k}_s\cdot\vec{x})/2M](\hbar^2k_s^2 - 2\hbar\vec{k}_s\cdot\vec{P}) \quad (33)$$

we have

$$\langle n_j|A_{sp}^\dagger[H_t,A_{sp}]|n_j\rangle = R(\hat{\epsilon}_s\cdot\langle\vec{r}\rangle)^2 + R\left(\frac{\vec{k}_s\cdot\langle\vec{r}\rangle}{M\omega_0}\right)^2\langle(\hat{\epsilon}_s\cdot\vec{P})^2\rangle_j - \frac{2\hbar}{M^2\omega_0}\hat{\epsilon}_s\cdot\langle\vec{r}\rangle\vec{k}_s\cdot\langle\vec{r}\rangle\langle(\hat{\epsilon}_s\cdot\vec{P})(\vec{k}_s\cdot\vec{P})\rangle_j + \frac{i\hbar}{M\omega_0^2}(\vec{k}_s\cdot\langle\vec{r}\rangle)^2\langle\hat{\epsilon}_s\cdot\vec{P}\left(\sum_{m=x,y,z}\Omega_m^2 x_m\epsilon_{s_m}\right)\rangle_j , \quad (34)$$

where, for an operator J,

$$\langle J\rangle_j \equiv \langle n_j|J|n_j\rangle ,$$

and $|\vec{k}_s| \simeq |\vec{k}|$.

The third term in Eq. (34) is approximately equal to

$$|\langle\vec{r}\rangle|^2(RE_K/\hbar\omega_0) \simeq \hbar\omega_0\beta^2|\langle\vec{r}\rangle|^2 ,$$

and the fourth term is approximately equal to $|\langle\vec{r}\rangle|^2R(\Omega/\omega_0)^2$. The fourth term is clearly negligible with respect to the first, and the third term is negligible when $E_K \ll \hbar\omega_0$, which is reasonable and is assumed here.

With the above approximations and noting that

$$(\hat{\epsilon}_s\cdot\langle\vec{r}\rangle)^2 + \left(\frac{\vec{k}_s\cdot\langle\vec{r}\rangle}{M\omega_0}\right)^2\langle(\hat{\epsilon}_s\cdot\vec{P})^2\rangle_j = \sum_f |\langle n_f|A_{sp}|n_j\rangle|^2 , \quad (35)$$

we have

$$\langle E_f - E_j\rangle = R , \quad (36)$$

which is independent of the intermediate state. This is a familiar result from the studies of the Mössbauer effect and shows that a fundamental limit to the cooling process is caused by the recoil heating upon reemission. It also agrees with Eq. (2b) (averaged over emission angles) for the free atom case (neglecting β^2 terms). At this point we note that

$$[(\vec{k}_s\cdot\langle\vec{r}\rangle)/M\omega_0]^2\langle(\hat{\epsilon}_s\cdot\vec{P})^2\rangle_j \simeq |\langle\vec{r}\rangle|^2\beta^2 ,$$

and therefore we will neglect the second term in Eqs. (32) and (35) in what follows.

We can arrive at a better understanding of the emission spectrum by asking for the second moment of translational energy change in the emission process. This is interesting because it gives the spread of photon energies in the emission process. We have

$$\langle(\Delta E)^2\rangle = \langle(E_f - E_j - \langle E_f - E_j\rangle)^2\rangle$$
$$= \langle(E_f - E_j)^2\rangle - R^2,$$

where we have used Eq. (36). We have

$$\langle(E_f - E_j)^2\rangle = \int d\Omega \sum_{s,f} (E_f - E_j)^2 |\langle n_f|\exp(-i\vec{k}_s\cdot\vec{x})|n_j\rangle|^2 \Big/ \int d\Omega \sum_{s,f} |\langle n_f|\exp(-i\vec{k}_s\cdot\vec{x})|n_j\rangle|^2 .$$

Writing

$$(E_f - E_j)^2|\langle n_f|\exp(-i\vec{k}_s\cdot\vec{x})|n_j\rangle|^2$$
$$= |\langle n_f|[H_t, \exp(-i\vec{k}_s\cdot\vec{x})]|n_j\rangle|^2,$$

using Eqs. (28) and (33), and assuming the atoms to be unpolarized, we obtain

$$\langle(\Delta E)^2\rangle = \tfrac{2}{3}R\langle H_t\rangle . \quad (37)$$

If the motion is characterized by the Maxwell-Boltzmann distribution, this is just the expression for broadening due to the Doppler effect. Below we will show that in the classical limit the profile of the emission spectrum observed in a particular direction is a shifted Gaussian. In this case, for the recoil energy R approximately equal to the atom kinetic energy, the spectrum is shown in Fig. 7. We note that Eqs. (36) and (37) are also valid for the case of the free atom (neglecting terms of order β^2), since Eq. (33) holds for both cases; however, the spectrum is continuous and is shown by the dotted line in Fig. 7.

C. Absorption cross section

For the case of bound atoms, Eq. (19) becomes

$$\frac{d\sigma_{i\to f}}{d\omega_s\, d\Omega} = C\sum_s \frac{\omega_s}{\omega}|\hat{\epsilon}_s\cdot\langle\vec{r}\rangle|^2|\hat{\epsilon}\cdot\langle\vec{r}\rangle|^2 \left|\sum_j \frac{\langle n_f|\exp(-i\vec{k}_s\cdot\vec{x})|n_j\rangle\langle n_j|\exp(i\vec{k}\cdot\vec{x})|n_i\rangle}{\omega_0 - \omega + (E_j - E_i)/\hbar - \frac{1}{2}i\gamma}\right|^2 \delta\left(\omega - \omega_s - \frac{E_f - E_i}{\hbar}\right) . \quad (38)$$

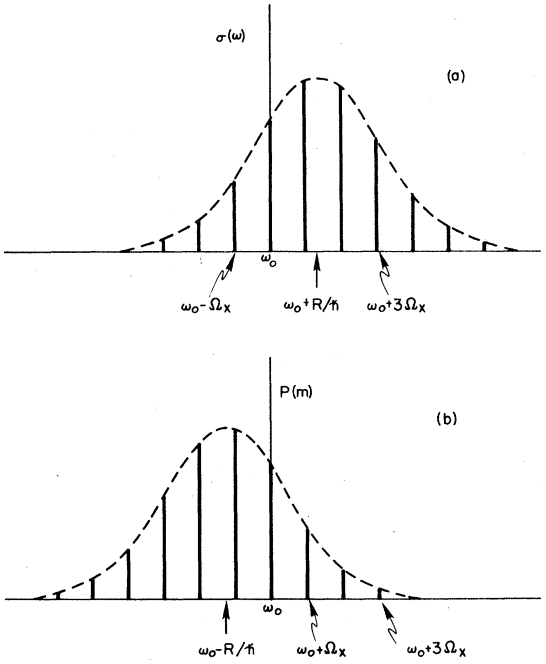

FIG. 7. Atomic spectra in classical limit ($\hbar\Omega_x \ll k_BT$) when $R \lesssim \hbar\omega_D$. Part (a) shows the absorption cross section for a laser directed along the x axis for the case when $\gamma \ll \Omega_x$ (giving the discrete lines) and when $\Omega_x \to 0$ (dashed curve) which is also the case for free atom. Part (b) shows the emission spectrum observed along the x direction for the same two cases.

We can obtain the absorption cross section by summing over the final states and integrating over possible scattered photon states. Following the procedure used to obtain Eq. (24), we have

$$\sigma_i = 6\pi\lambda^2|\hat{\epsilon}\cdot\langle\hat{r}\rangle|^2 \times \sum_j \left|\frac{\langle n_j|\exp(-i\vec{k}\cdot\vec{x})|n_i\rangle\frac{1}{2}\gamma}{\omega_0 - \omega + (E_j - E_i)/\hbar - \frac{1}{2}i\gamma}\right|^2 . \quad (39)$$

1. Strong binding

Equation (39) clearly shows the sideband structure in the absorption spectrum when $\Omega_i \gg \gamma$. For this case, essentially only one term contributes to the sum over j when the resonance condition $[\omega_0 - \omega + (E_j - E_i)/\hbar = 0]$ is met. For simplicity, assume that the atoms are unpolarized and that the laser radiation is incident along the x axis, so that $\omega = \omega_0 + m\Omega_x (m = 0, \pm1, \pm2, \ldots)$ that is, we are tuned to the mth sideband. Then Eq. (39) becomes

$$\sigma_i \simeq \sigma_0|\langle n_i + m|\exp(i\vec{k}\cdot\vec{x})|n_i\rangle|^2 , \quad (40)$$

where we have dropped the x subscripts in the wave functions.

It is useful to examine Eq. (40) when $R \ll k_BT$ and where n_i is approximately equal to the mean occupation number, i.e.,

$$n_l = n^* \simeq \langle n \rangle = \langle H_t \rangle / (\hbar\Omega_x) - \tfrac{1}{2} .$$

For the case of interest, the laser frequency must lie somewhere within the Doppler profile of the atomic resonance (i.e., $|m|\hbar\Omega_x \lesssim \beta\hbar\omega_0$). In this case, we can show that $|m| \ll n^*$. We have

$$|m|\hbar\Omega_x \lesssim \hbar\omega_0\Omega_x(\langle x^2\rangle)^{1/2}/c = \hbar\Omega_x k(\langle x^2\rangle)^{1/2} \tag{41}$$

and so

$$\frac{|m|}{n^*} \lesssim \frac{k\langle x^2\rangle^{1/2}}{n^*} \simeq \left(\frac{R}{n^*\hbar\Omega_x}\right)^{1/2} \simeq \left(\frac{R}{k_BT}\right)^{1/2} \ll 1 .$$

If n^* is large ($\hbar\Omega_x/E_x \ll 1$), we can approximate the Laguerre polynomial in the matrix element by its asymptotic form.[27] Dropping the * superscript, we have

$$\exp[-\tfrac{1}{2}(kx_0)^2](kx_0)^{|m|}L_{n-}^{|m|}{}_{|m|}((kx_0)^2) \simeq \frac{\Gamma(n+1)J_{|m|}[kx_0(4n-2|m|+2)^{1/2}]}{(n_l-\frac{1}{2}|m|+\frac{1}{2})^{|m|/2}(n-|m|)!} .$$

Since $n \gg |m|$, then

$$(n-|m|)! = n![n(n-1)(n-2)\ldots(n-|m|+1)]^{-1} \simeq n!n^{-|m|}$$

and

$$(n-\tfrac{1}{2}|m|+\tfrac{1}{2})^{|m|/2} \simeq n^{|m|/2}$$

so that

$$\exp[-\tfrac{1}{2}(kx_0)^2](kx_0)^{|m|}L_{n-}^{|m|}{}_{|m|}((kx_0)^2) \simeq n^{|m|/2}J_{|m|}(2n^{1/2}kx_0) .$$

Therefore

$$\langle n^* - |m||\exp(i\vec{k}\cdot\vec{x})|n^*\rangle \simeq (i)^{|m|}J_{|m|}(2n^{*1/2}(kx_0))$$

and since

$$n^*x_0^2 \simeq \tfrac{1}{2}\langle x^2\rangle = \tfrac{1}{4}x_a^2 ,$$

where x_a is the amplitude of the classical oscillatory motion, then Eq. (40) can be written

$$\sigma_l \simeq \sigma_0 J_{|m|}^2(kx_a),$$

which is equivalent to the cross section used in Eq. (14).

In general, we must average the absorption cross section over a distribution of initial state values. This averaged cross section is then given by:

$$\sigma(m) = \sum_l P_l\sigma_l , \tag{42}$$

where σ_l is given by Eq. (40) and where P_l is the probability of the atom initially being in the state l. In this more general case we can arrive at an expression for the cross section by assuming that P_l is given by a thermal distribution. That is,

$$P_l = [1-\exp(-\hbar\Omega_x/k_BT)]\exp(-n_l\hbar\Omega_x/k_BT) ,$$

so that when $m \leq 0$

$$\sigma(m) = \sigma_0\left[1-\exp\left(\frac{-\hbar\Omega_x}{k_BT}\right)\right]\sum_{n_l=-m}^{\infty}\exp\left(\frac{-n_l\hbar\Omega_x}{k_BT}\right)|\langle n_l+m|e^{ikx}|n_l\rangle|^2$$
$$= \sigma_0\left[1-\exp\left(\frac{-\hbar\Omega_x}{k_BT}\right)\right]\exp\left[-(kx_0)^2+\left(\frac{m\hbar\Omega_x}{k_BT}\right)\right](kx_0)^{-2m}\sum_{n=0}^{\infty}\exp\left(\frac{-n\hbar\Omega_x}{k_BT}\right)\frac{n!}{(n-m)!}[L_n^{-m}(k^2x_0^2)]^2 ,$$

where we have used Eq. (31). We obtain a similar result for $m \geq 0$. These sums can be evaluated with the generating function[28]

$$\sum_{n=0}^{\infty}\frac{n!}{\Gamma(n+\alpha+1)}L_n^{\alpha}(x)L_n^{\alpha}(y)z^n = (1-z)^{-1}\exp\left[-z\left(\frac{x+y}{1-z}\right)\right]\times(xyz)^{-\alpha/2}I_{\alpha}\left(\frac{2(xyz)^{1/2}}{1-z}\right) .$$

If we let

$$z = \exp(-\hbar\Omega_x/k_BT), \quad x = y = (kx_0)^2, \quad \alpha = |m| ,$$

and noting that $I_m = I_{-m}$, then

$$\sigma(m) = \sigma_0\exp\left[\frac{m}{2}\frac{\hbar\Omega_x}{k_BT} - 2(kx_0)^2 \times\left(\frac{1}{2}-\frac{1}{1-\exp(-\hbar\Omega_x/k_BT)}\right)\right] \times I_m\left(\frac{2(kx_0)^2\exp(-\frac{1}{2}\hbar\Omega_x/k_BT)}{1-\exp(-\hbar\Omega_x/k_BT)}\right) .$$

Noting that

$$\langle x^2\rangle = 2x_0^2(\langle n\rangle + \tfrac{1}{2}) ,$$

where $\langle n\rangle$ is the mean occupation number

$$\langle n\rangle = \sum_n nP_n = \left[\exp\left(\frac{\hbar\Omega_x}{k_BT}\right)-1\right]^{-1} , \tag{43}$$

then

$$\sigma(m)=\sigma_0\exp[\tfrac{1}{2}m(\hbar\Omega_x/k_BT)-k^2\langle x^2\rangle]$$
$$\times I_m[\exp(\hbar\Omega_x/2k_BT)2(kx_0)^2\langle n\rangle]\,. \qquad (44)$$

Examination of Eq. (44) shows the skewing of the cross section to high-energy values, i.e., $m>0$. This is plausible because, for example, in the low-temperature limit ($k_BT \lesssim R$) the atom more easily gains energy rather than losing it in the scattering process. (When $T=0$ it is impossible for it to lose energy.)

A result similar to Eq. (44) is obtained when one looks for the probability of emission at a certain sideband frequency. From Sec. V B we write a general expression for the probability of emission of a photon of frequency $\omega_0+m\Omega_x$, which we call $P(m)$. We have

$$P(m)=\sum_j |\langle n_j-m|\exp(-i\vec{k}_s\cdot\vec{x})|n_j\rangle|^2P_j\,,$$

where P_j is the probability of the atom being in the translational energy level j when in the excited state. $P(m)$ is normalized to unity as can be verified by summing over m. If we observe the emitted light in the x direction, then $\vec{k}_s\cdot\vec{x}\to k_sx$. If P_j is given by a Maxwell-Boltzmann distribution then we find

$$P(m)=\exp[-\tfrac{1}{2}m(\hbar\Omega_x/k_BT)-k^2\langle x^2\rangle]$$
$$\times I_m[\exp(\hbar\Omega_x/2k_BT)2(kx_0)^2\langle n\rangle]\,. \qquad (45)$$

This same result was derived by an alternative method and is given by Eq. (3.66a) of Ref. 26. Equation (45) clearly shows the skewing to low-energy values.

In a high-resolution experiment with lasers used to probe a bound atom, we might be particularly interested in the cross section for absorption on the unshifted carrier given by Eq. (44) for $m=0$. We have

$$\sigma(m=0)=\sigma_0\exp(-k^2\langle x^2\rangle)$$
$$\times I_0[\exp(\hbar\Omega_x/k_BT)2(kx_0)^2\langle n\rangle]\,.$$

A special case is when $\langle n\rangle\to 0$, yielding the cross section:

$$\sigma(0\to 0)=\sigma_0\exp(-k^2x_0^2)\,.$$

The exponential factor is known as the Debye-Waller factor and is familiar in studies of x-ray scattering and the Mössbauer effect. It shows the suppression of the cross section due to zero-point vibrations.

It is interesting to evaluate Eq. (44) in the classical limit, that is, when $\hbar\Omega_x \ll k_BT$. In this case, if we apply Eq. (16) we obtain

$$\sigma(m)=(\sigma_0\Omega_x/\sqrt{\pi}\,\omega_D)\exp\{-[(\omega-\omega_0')/\omega_D]^2\}\,, \qquad (46)$$

where $\omega-\omega_0'=m\Omega_x-R/\hbar$. This expression is analogous to the cross section used in Eq. (17), but now shows the dependence on recoil. We note that the envelope of the cross section for the sidebands has a normal Doppler shape; however, the center of gravity of this Doppler profile is shifted to the frequency $\omega_0'=\omega_0+R/\hbar$. Similarly, we can also find the expression for the emission spectrum which then has the same form as Eq. (46) except that $\omega_0'\to\omega_0''=\omega_0-R/\hbar$. Thus, we obtain a Doppler profile whose center of gravity is shifted to the frequency $\omega_0-R/\hbar$ as indicated in Fig. 7.

2. *Weak binding*

When the condition $\gamma \ll \Omega_x$ is not satisfied, we must return to the cross section given in Eq. (42), where σ_l is given by Eqs. (38) or (39). Assuming the atoms are thermalized and unpolarized, we have:

$$\sigma(\omega)=\sum_{m=-\infty}^{\infty}\frac{\sigma(m)}{1+[(2/\gamma)(\omega_0-\omega+m\Omega_x)]^2}\,, \qquad (47)$$

where $\sigma(m)$ is given by Eq. (44). In the classical limit ($\hbar\Omega_x \ll k_BT$), we can use Eq. (46) and convert the sum in Eq. (47) to an integral to obtain

$$\sigma(\omega)=\frac{\sigma_0}{\sqrt{\pi}\,\omega_D}\int_{-\infty}^{\infty}\frac{\exp[-(\Omega/\omega_D)^2]}{1+[(2/\gamma)(\omega_0'-\omega+\Omega)]^2}\,d\Omega\,,$$

which is identical to Eq. (6). Thus we see that the absorption cross section for a bound atom approaches that of the free atom in the limit of weak binding.

D. Cooling rate

The cooling rate can be derived from the differential cross section by means of Eq. (27), where $d\Omega_{l\to f}/d\Omega$ is given by Eq. (38). After some simplification, and assuming the atoms are unpolarized, we can write

$$\left\langle\frac{d(E_f-E_l)}{dt}\right\rangle=\frac{I\sigma_0}{\hbar\omega}\sum_{l,j}P_l\left((R+E_j-E_l)\frac{|\langle n_j|\exp(i\vec{k}\cdot\vec{x})|n_l\rangle|^2}{1+\{(2/\gamma)[\omega_0-\omega+(E_j-E_l)/\hbar]\}^2}\right). \qquad (48)$$

In this form we have separated out the effect due to reemission (R term) and the effect due to absorption ($E_j - E_l$ term).

1. Strong binding

In the limit $\gamma \ll \Omega_x$, and assuming we are tuned to resonance ($\omega = \omega_0 + m\Omega_x$), then only one term of the sum in Eq. (48) is important, and we obtain

$$\left\langle \frac{d(E_f - E_l)}{dt} \right\rangle_m = \frac{I\sigma(m)}{\hbar\omega} [R + m\hbar\Omega_x] , \tag{49}$$

where $\sigma(m)$ is given by Eq. (42) or by Eq. (44) when P_l is given by a Maxwell-Boltzmann distribution. In the classical limit $\sigma(m)$ is given by Eq. (46) and we obtain

$$\left\langle \frac{d(E_f - E_l)}{dt} \right\rangle_m = \frac{I\sigma_0\Omega_x}{\hbar\sqrt{\pi}\,\omega_0\omega_D} [R + m\hbar\Omega_x] \times \exp[-(\omega - \omega_0')^2/\omega_D^2] , \tag{50}$$

which is analogous to Eq. (17) but now includes the effect due to recoil.

2. Weak binding

When the condition $\gamma \ll \Omega_x$ is not satisfied, we must perform the sum in Eq. (48). In the classical limit $h\Omega_x \ll k_B T$ we obtain

$$\left\langle \frac{d(E_f - E_l)}{dt} \right\rangle = \frac{I}{\hbar\sqrt{\pi}\,\omega_0\omega_D} \times \int_{-\infty}^{\infty} \frac{(2R + \hbar\Omega)\exp[-(\Omega^2/\omega_D^2)]d\Omega}{1 + [(2/\gamma)(\omega_0' - \omega + \Omega)]^2} ,$$

which is identical to Eq. (7).

E. Cooling limits

It is also interesting to estimate the limits to the cooling processes; that is, what are the minimum temperatures which can be achieved by the technique? As explained in Sec. II, the limit is obtained when the heating rate due to recoil equals the cooling rate.

1. Strong binding

We first examine the case when $\Omega_x \gg \gamma$. We will assume that substantial cooling has already taken place, and therefore the spectrum of the atom consists of the "carrier" at frequency ω_0 and sidebands at $\omega_0 \pm \Omega_x$; that is, $k\langle x^2\rangle^{1/2} \ll 1$. Note also that for this model to work, we must require $R \ll \hbar\Omega_x$ or else the reemission spectrum does not satisfy the simple picture of a "carrier" and two adjacent sidebands; however, this seems to be a most interesting case in the optical domain. The case when $R \gg \hbar\Omega_x$ could be easily treated. For maximum cooling the incident radiation is tuned to $\omega = \omega_0 - \Omega_x$, i.e., the lower sideband. In Eq. (49) we have included the effect of only the resonant sideband in arriving at a cooling rate. However, to obtain the cooling limit, we will include the effect of both upper and lower sidebands and the carrier. From Eq. (48) we therefore find near the cooling limit:

$$\left\langle \frac{dE}{dt} \right\rangle = \frac{I\sigma_0}{\hbar\omega} R \left(\frac{5}{16} \frac{\gamma^2}{\Omega_x^2} - \langle n_x \rangle \right) , \tag{51}$$

which is independent of initial-state distribution but assumes that the y and z degrees of freedom are thermalized with the x degree of freedom and therefore have the same distribution. We might be tempted to apply Eq. (51) to the case of a single isolated atom, but we note that if the laser is directed along the x axis, then the y and z degrees of freedom are heated without bound by the recoil upon reemission. If the frequencies $\{\Omega_i\}$ are degenerate then this argument applies regardless of the direction of the laser since there are no unique axes for the bound atom. If the frequencies $\{\Omega_i\}$ are unequal, then Eq. (51) cannot hold for a single laser directed along, say, the $\hat{i} + \hat{j} + \hat{k}$ direction, because the laser frequency cannot simultaneously coincide with the first lower sidebands for the three independent oscillations. Each specific case is easily treated; for simplicity, however, we will assume as above that we have a collection of weakly thermalized atoms such that the collision rate between atoms is much larger than the optical transition rate. (See Sec. V F). To find the minimum energy possible, we set $\langle dE/dt \rangle = 0$ and obtain

$$\langle n_x \rangle = \tfrac{5}{16}(\gamma^2/\Omega_x^2) \ll 1 .$$

Note that this expression differs from that of Ref. 7. The difference appears to be due to the neglect of the heating effect due to recoil in the direction perpendicular to the laser beam in Ref. 7. That is, if the unshifted emission in the direction perpendicular to the laser beam is considered (the source of the "1+1" factor), then the heating due to recoil in this direction must also be considered.

The minimum energy is of course limited to the zero-point energy $\frac{1}{2}\hbar\Omega_x$; however, this can presumably be accurately determined if Ω_x is known, and therefore the uncertainty in energy will be on the order of $\langle n_x \rangle \hbar\Omega_x$. In this limit, if the atoms have a Maxwell-Boltzmann distribution, then the temperature of the atoms is given by Eq. (43). If $\Omega_x/2\pi = 12.5$ MHz and $\gamma/\Omega_x = 0.1$, then $T \cong 10^{-4}$ K; however, the fractional uncertainty in this minimum energy is about 3×10^{-3}. In this simple case where the absorption

spectrum consists of a strong carrier with weak adjacent sidebands ($k\langle x^2\rangle^{1/2} \ll 1$), we remark that if the incident radiation were tuned to ω_0 we would see in the scattered light a strong component at frequency ω_0, a weak Stokes line at $(\omega_0 - \Omega_x)$, and a weak anti-Stokes line at $(\omega_0 + \Omega_x)$. However, since the incident radiation is tuned to $\omega_0 - \Omega_x$, the scattered light will contain a component at this frequency, but a much stronger component at frequency ω_0. Because of this we might regard the cooling process as anti-Stokes or *inverse-Stokes* spontaneous resonance Raman scattering.

2. *Weak binding*

When the condition $\gamma \ll \Omega_x$ is not satisfied, there are two interesting cases to discuss. For both cases we will assume that γ_c is much larger than the transition rate or that we irradiate the atoms with three mutually perpendicular laser beams. (See Sec. V F.) In the first case we will assume that substantial cooling has already taken place so that the Doppler width is much less than the natural width (which as before requires $R \ll \hbar\gamma$). Maximum cooling will be obtained when $\omega - \omega_0 \simeq -\frac{1}{2}\gamma$ and we can expand the denominator in Eq. (48) to obtain

$$\frac{dE}{dt} = \left\langle \frac{d(E_f - E_i)}{dt} \right\rangle = \frac{I\sigma_0}{2\hbar\omega} \sum_i P_i \langle n_i | A(\vec{k})^\dagger (R\, A(\vec{k}) + [H_t, A(\vec{k})]) | n_i \rangle ,$$

where

$$A(\vec{k}) = \exp(i\vec{k}\cdot\vec{x}) + (1/\hbar\gamma')[\exp(i\vec{k}\cdot\vec{x}), H_t] .$$

After some simplification we obtain

$$\frac{dE}{dt} \simeq \frac{I\sigma_0}{\hbar\omega} R\left(1 - \frac{2\langle E_x\rangle}{\hbar\gamma}\right),$$

which does not depend on the distribution of initial states and therefore holds for an isolated atom if we use three mutually perpendicular laser beams. Minimum kinetic energy is obtained when $dE/dt = 0$, and we obtain Eq. (11) ($\langle E_{Kx}\rangle_{\min} = \frac{1}{4}\hbar\gamma$). It should be noted here that the total energy of a harmonic oscillator is, on the average, equally divided between the kinetic and potential energies.

The second case to discuss is when $R \gg \hbar\gamma$. In this limit we have shown that Eq. (7) applies when P_i is given by a Maxwell-Boltzmann distribution leading to the limits in Fig. 3.

F. Energy distribution for low thermalization rate

We are interested in the kinetic energy distribution when we use a single laser beam to cool a collection of bound atoms. For simplicity we discuss a particular example; other cases could be similarly treated.

Suppose we have a single laser beam incident along the x axis and consider the limit $\gamma \gg \{\Omega_i\}$. If we ask for the cooling limit, then we want $\omega - \omega_0 \simeq -\frac{1}{2}\gamma$ when $R \ll \hbar\gamma$. For this case the equation analogous to Eq. (10) is

$$\frac{dE_x}{dt} = \gamma_i\left(-\frac{4RE_x}{\hbar\gamma} + \tfrac{4}{3}R\right) + \gamma_c\left(\frac{E_y}{2} + \frac{E_z}{2} - E_x\right),$$

where $\gamma_i \equiv I\sigma_0/(2\hbar\omega)$ and where we have included the transfer of energy between degrees of freedom (via the "collision" rate γ_c) and have assumed for simplicity that the recoil upon reemission is distributed equally over the three degrees of freedom. Note, however, that this simple assumption is violated even for unpolarized atoms. For the y and z directions we have

$$\frac{dE_y}{dt} = \gamma_i \frac{R}{3} + \gamma_c\left(\frac{E_x}{2} + \frac{E_z}{2} - E_y\right),$$

$$\frac{dE_z}{dt} = \gamma_i \frac{R}{3} + \gamma_c\left(\frac{E_x}{2} + \frac{E_y}{2} - E_z\right).$$

In steady state, $dE_i/dt = 0$, and we have $E_x = \frac{1}{2}\hbar\gamma$, which is identical to Eq. (11), and $E_y = E_z = \frac{1}{2}\hbar\gamma + \frac{2}{3}R\gamma_i/\gamma_c$. Hence if $\gamma_c \ll \gamma_i$, the other degrees of freedom are substantially heated by recoil; for a single isolated atom they are heated without bound.

VI. SUMMARY

We have discussed laser cooling of atoms from the standpoint of anti-Stokes spontaneous Raman scattering. To isolate the essential features of the problem we have made several simplifying assumptions. We have assumed that the internal states of the atoms comprise a simple two-level structure and that radiative transitions between these two levels proceed via pure electric dipole transitions. We assume that the plane-wave radiation source (laser) has spectral width $\ll \gamma$ and power much less than that required for saturation. We also assume that the power is small enough to avoid the effects of the induced dipole force[3]; this is particularly important for the free-atom case where it is desirable to have several laser beams in order to cool in all directions (Sec. II B *1*). Finally, we assume that the laser power is low enough to avoid the effects of recoil heating in directions perpendicular to the laser beam when only one beam is used. The treatment has been nonrelativistic; hence we have neglected terms of order $\beta^2\hbar\omega_0$ and βR in the energy.

The cooling rates for free atoms are given in Eq. (5) (one laser beam in x direction), Eq. (7)

(one laser beam in x direction, atoms thermalized), and Eq. (8) [approximation of Eq. (7) when $\gamma, R/\hbar \ll \omega_D$]. These formulas are justified in Sec. IV. For bound atoms, the most general expression for the cooling rate (for one laser beam) is given in Eq. (48). Cooling rates for various approximations are given in Eq. (49) ($\gamma \ll \Omega_x$, one laser beam directed along the x axis and tuned to $\omega = \omega_0 + m\Omega_x$, where m is an integer), Eq. (50) [Eq. (49) in classical limit $\hbar\Omega_x \ll k_B T$], and Eq. (7) (classical limit when $\gamma \gg \Omega_x$, one laser beam directed along the x axis).

As an example, for $M = 100$ amu, $\nu_0 = 5\times 10^{14}$ Hz($\lambda = 600$ nm), $\gamma = 2\pi \times 10$ MHz, $I = 50$ mW/cm^2, $\sigma_0 = 5.7\times 10^{-10}$ cm^2, the cooling rate for "free" or weakly bound atoms ($\Omega_x \ll \gamma$) is given by Eq. (8) when the atoms are thermalized. This gives a rate $dE/dt \simeq -1.4$ eV/sec. We have assumed that the laser is tuned to $\omega - \omega_0 = -\omega_D/\sqrt{2}$; this is the condition for maximum cooling when $\gamma \ll \omega_D$. This result is independent of temperature as long as $\gamma \ll \omega_D$. (Note, however, that the laser must be swept closer to the line center as the atoms cool to satisfy the condition $\omega - \omega_0 = -\omega_D/\sqrt{2}$.) Equation (8) will also be invalid when $\omega_D \lesssim 10\gamma$ in this example due to saturation of the optical transition.

When the atom is tightly bound ($\Omega_x \gg \gamma$) we may apply Eq. (50) in the classical limit ($\hbar\Omega_x \ll k_B T$). We note that for the same approximate detuning (i.e., assuming the laser is tuned to the sideband which is closest to satisfying the condition $m\hbar\Omega_x = -\omega_D/\sqrt{2}$), the cross section is equal to the cross section in Eq. (8) times $2\Omega_x/(\pi\gamma)$, which results from the compression of the full Doppler profile into discrete sidebands. The cooling rate is increased by the same factor.

It is particularly interesting to examine the cooling limits for various cases. These are summarized in Table I. In each case the mechanism which limits the cooling is the recoil effect; however, this may not be apparent from the expressions. The limits given in Table I assume that there are no other heating mechanisms present; in particular, we have avoided the thermodynamic limits caused by blackbody radiation (Sec. II D) by assuming that blackbody radiation from the surroundings is absent.

For our example atom above, since $R/\hbar \ll \gamma$, the first limit for both free and bound atoms applies. (The assumption is that $\Omega \ll 2\pi \times 10$ MHz, which is usually the case for electromagnetically

TABLE I. Cooling limits for various limiting cases.

Case	Limit	Assumptions
Free atoms	$\langle E_{Kx}\rangle_{\min} = \frac{1}{4}\hbar\gamma$	$R/\hbar \ll \gamma$. Six laser beams in $\pm x$, $\pm y$, and $\pm z$ directions, tuned to $\omega = \omega_0 + R/\hbar - \gamma/2$. (Atoms need not be thermalized via collisions.) $\gamma/2\pi$ is the full width at half-maximum for the optical transition.
	$k_B T \lesssim R$ (Fig. 3)	$R/\hbar \sim \gamma$. Ensemble of atoms assumed to be thermalized with Maxwell-Boltzmann distribution.
Bound atoms	$\langle E_{Kx}\rangle_{\min} = \frac{1}{4}\hbar\gamma$	$R/\hbar, \Omega \ll \gamma$. Laser(s) tuned to $\omega = \omega_0 + R/\hbar - \gamma/2$. If atoms are thermalized only one laser beam need be present. If atoms are not thermalized we must have 3 mutually perpendicular laser beams or one laser beam along $\hat{i}+\hat{j}+\hat{k}$ direction when $\Omega_x \neq \Omega_y \neq \Omega_z \neq \Omega_x$.
	$k_B T \lesssim R$ (Fig. 3)	Same as for free atom. ($\gamma \gg \Omega_x$)
	$\langle n_x\rangle = 5\gamma^2/16\Omega_x^2$	$\gamma, R/\hbar \ll \Omega_x$. One laser beam directed along x axis and tuned to $\omega_0 - \Omega_x$, atoms thermalized. Same limit for noncolliding atoms if $\Omega_x = \Omega_y = \Omega_z$ and 3 mutually perpendicular beams are used.

confined ions, for example.) We have $\langle E_{Kx}\rangle_{\min} \simeq 10^{-8}$ eV or, if the atoms are thermalized, $T_{\min} \simeq 2.4\times 10^{-4}$ K. If the atomic transition was only very weakly allowed (given by the condition $\gamma \lesssim R/\hbar$), then the limits given in Fig. 3 apply. This would imply $T \lesssim 2.7\times 10^{-7}$ K for $M = 100$ amu, $\nu_0 = 5\times 10^{14}$ Hz. However, for this limit to apply we must have $\gamma/2\pi < 6$ kHz. This condition could be realized on an intercombination line, for example. Finally, if the atom was tightly bound, we could realize the last limit in Table I. In this case the minimum energy is given by the zero-point energy, but as discussed in Sec. V E, the uncertainty in this energy could be quite small.

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