

Laser cooling: Beyond optical molasses and beyond closed transitions

Vladan Vuletić, Andrew J. Kerman, Cheng Chin, and Steven Chu

Department of Physics, Stanford University, Stanford, CA 94305-4060

Abstract. We present a simple and general optical cooling method based on 3D degenerate Raman sideband cooling with adiabatic release that goes significantly beyond the density and temperature limitations of optical molasses. In 10 ms we cool a sample of 3×10^8 cesium atoms to a temperature of 330 nK at a density of $1.1 \times 10^{11} \text{ cm}^{-3}$, which corresponds to a phase-space density $n\lambda_{dB}^3 = 1/500$.

We further propose to cool atoms or molecules inside an optical cavity that enhances the scattering of blue-detuned photons and show that the dissipative mechanism can be viewed as a cavity-induced generalized Doppler cooling. Since the cooling depends on the atom's internal level structure only through the photon scattering rate, cavity Doppler cooling is applicable to particles that do not possess a closed optical transition, which may allow one to extend laser cooling to a greater class of atoms or molecules. Large samples are cooled at the same rate as single atoms if the effect of one atom on the cavity resonance frequency is small. We also show how to achieve 3D cooling with a single optical cavity.

BEYOND OPTICAL MOLASSES: 3D DEGENERATE RAMAN SIDEBAND COOLING

Optical molasses has long been the best simple optical cooling method available for atoms in free space and it remains the most widely used technique to prepare large samples of cold atoms for various experiments. Some applications, such as atomic fountain clocks, mainly require a simple and fast method that produces the lowest possible temperatures with the largest number of atoms, while other experiments, among them those where the atoms are loaded into magnetic or optical traps, require moderately low temperatures at high atomic densities. A simple and fast method that can cool large atomic samples to high phase-space density is particularly useful for evaporative cooling to Bose-Einstein condensation, since it allows one to reduce the atom loss during evaporation, and to produce significantly larger condensates in a shorter time.

Here we present a simple and general optical cooling method that requires only one weak laser in addition to the lasers used to create a magneto-optical trap (MOT), while it increases the phase-space density in 10 ms by almost three orders

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of magnitude over the value attainable with optical molasses for all 3×10^8 atoms prepared in our cesium MOT. In particular, this technique may allow an even stronger relative improvement for atoms where the excited-state hyperfine structure is not resolved and therefore sub-Doppler cooling is not effective in optical molasses. Important atoms belonging to this class are lithium and potassium, that have both bosonic and fermionic isotopes.

The new method is based on Raman sideband cooling [1–3] inside a 3D optical lattice in combination with adiabatic release [4,5]. The 3D lattice is formed by two counterpropagating beams along the x-axis and two running waves along the y and z-axes. This four-beam setup does not require interferometric stabilization of the relative time phases of different beams, since variations of the time phases correspond merely to a translation of the lattice in space. As these variations occur at a rate that is much lower than the typical lattice vibration frequency of $\omega/2\pi = 30$ kHz, the atoms that are trapped at the lattice sites follow the motion of the lattice adiabatically and consequently the cooling performance is not affected. We choose e^{-2} beam waists of 1 mm at the position of the atoms, such that the lattice is only slightly larger than the trapped cloud in the MOT containing up to 5×10^8 atoms, which reduces the power requirement for the lattice laser to a total of 20 mW for all four beams. The polarizations of the four beams are linear and both the polarization directions and the relative beam intensities are independently adjusted to optimize the cooling. A typical detuning for the lattice laser is ± 10 GHz relative to the $6S_{1/2}, F = 3 \rightarrow 6P_{3/2}, F'$ transitions. As the lattice geometry differs significantly for red and for blue detuning of the lattice laser, different beam intensity ratios and polarization angles are found to yield the lowest temperatures in these two cases.

As discussed in Refs. [3,5], degenerate Raman sideband cooling in the lower hyperfine manifold starts with atoms optically pumped into the lowest-energy magnetic sublevel $F = 3, m_F = 3$ in a trap where the vibration frequency exceeds the recoil energy (Lamb-Dicke regime). A small magnetic field on the order of 50 mG is applied that produces a Zeeman splitting between the neighboring magnetic sublevels $m_F = 3$ and $m_F = 2$ that equals the vibration energy. The atoms are transferred from $m_F = 3$ to $m_F = 2$ by means of an energy selective Raman transition with two photons of equal frequency but different polarizations, such that the vibrational quantum number is lowered by one unit. Optical pumping back to the sublevel $m_F = 3$ preferentially maintains the vibrational state, and the atoms are cooled by one vibration energy in the cycle. The cooling continues until the atoms accumulate in the vibrational ground state of the $m_F = 3$ sublevel that is dark to both the optical pumping light and to the Raman transition. Since the $P_{3/2}$ state of cesium has a sufficiently large hyperfine splitting, our optical pumping is performed on the $F = 3 \rightarrow F = 2$ transition of the D_2 line. For atoms where the hyperfine structure of the excited P state is not resolved optical pumping with circularly polarized light on the D_1 line that always provides a dark state should be used for Raman sideband cooling.

To attain fast cooling in our 3D lattice where the trapping potential at each

site is neither isotropic nor harmonic, we adjust the polarizations of the lattice beams to achieve a large Raman coupling that is comparable to the trap vibration frequency. Consequently, and in contrast to the best cooling in the 1D case where the red cooling sideband and its second harmonic were clearly resolved [3], here our cooling spectrum exhibits only a broad minimum for the final temperature as a function of magnetic field, as shown in Figure 1.

Although the large Raman coupling leads to a non-negligible off-resonant rate out of the nominally 'dark' vibrational ground state of the $m_F = 3$ sublevel, we still achieve a very good cooling performance. Figure 2 shows the phase-space density $n\lambda_{dB}^3$ as a function of density for all 3×10^8 atoms prepared in our MOT [5]. The dotted and the dashed line indicate the results for standard red-detuned polarization gradient cooling (bright molasses) and for blue-detuned polarization gradient cooling ('grey' molasses) for cesium from Ref. [6], respectively. The phase-space density obtained with our cooling scheme exceeds that of optical molasses by almost three orders of magnitude. This improvement results from a combination of three factors: First, the low-density temperature limit of our scheme of 290 nK is lower than both for bright optical molasses ($3\mu\text{K}$) and for dark optical molasses ($1.1\mu\text{K}$). More important is the fact that for our method the final temperature increases very slowly with density. We measure a linear density dependence of only $8\text{nK}/10^{10}\text{cm}^{-3}$, which is 75 times lower than the coefficient of $600\text{nK}/10^{10}\text{cm}^{-3}$ measured for dark optical molasses [6], while standard red-detuned polarization gradient cooling performs even worse. The observed low value of density dependent heating is probably partly due to the fact that in the lattice the atoms quickly bind to the lattice sites and are then isolated from each other, which leads to a suppression of radiative collisions, and partly due to the fact that in the Lamb-Dicke regime the relevant recoil heating (expressed in terms of change of quantum state per scattering event) is smaller than in free space. The weak dependence of the heating on density allows us to significantly increase the density before the

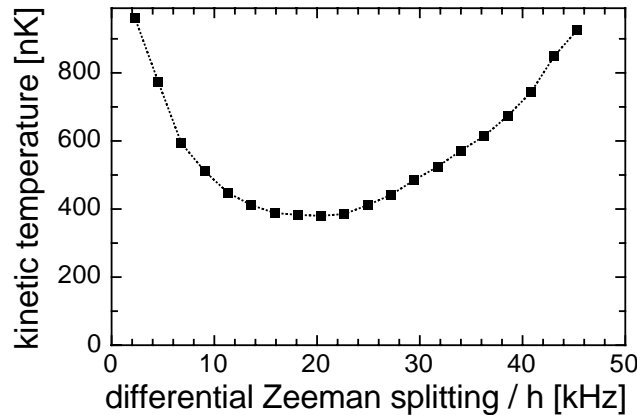


FIGURE 1. The final temperature after adiabatic release as a function of Zeeman energy splitting between two neighboring magnetic sublevels for 3D cooling with large Raman coupling.

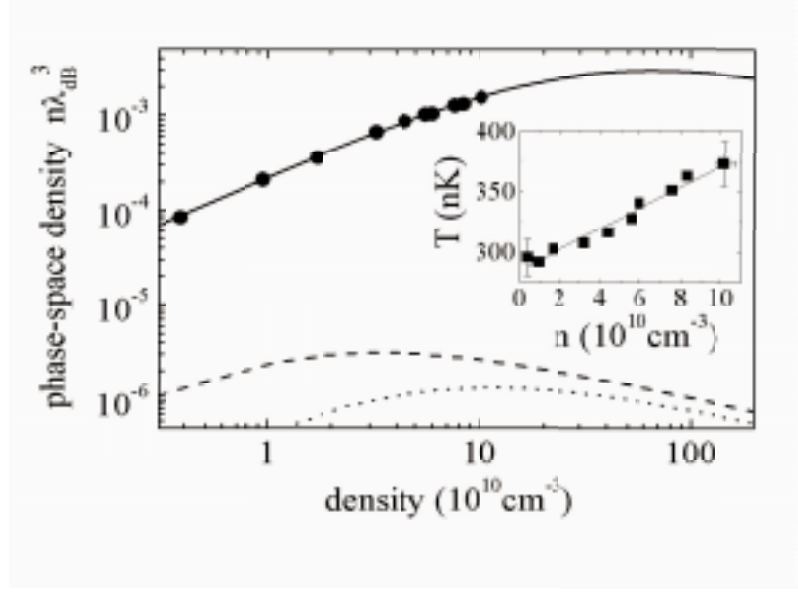


FIGURE 2. The phase space density as a function of atomic density for red detuned polarization gradient cooling (dotted line), blue detuned polarization gradient cooling (dashed line) and for 3D Raman sideband cooling with adiabatic release (solid line).

temperature increase eventually overcomes the effect of the density increase and limits the attainable phase-space density. Finally, our method simultaneously spin-polarizes the sample into one magnetic sublevel, while optical molasses leaves the atoms largely unpolarized with respect to an external quantization axis.

The highest phase-space density achieved with all 3×10^8 atoms prepared in our MOT is $n\lambda_{dB}^3 = 1/500$ at a density of $1.1 \times 10^{11} \text{ cm}^{-3}$ and a temperature of 330 nK. In this experiment the cooling was optimized with the constraint that it should not induce a loss of atoms and the measured transfer from the MOT to the cold sample was better than 95%. Higher values for the phase-space density are possible if one allows for atom loss during the cooling or if one uses smaller samples. Under such conditions a further improvement of the phase-space density by more than one order of magnitude has been recently demonstrated in Ref. [7], and we also observe higher phase-space density in combination with significant loss due to radiative collisions at densities above 10^{12} cm^{-3} . The critical value for Bose-Einstein condensation of $n\lambda_{dB}^3 = 2.6$ after release into free space would be obtained if one could prepare one atom at each lattice site and cool each atom to the local 3D vibrational ground state. However, this will be difficult to achieve since two or more atoms at one lattice site are lost by a radiative collision [8,4] long before they can be optically pumped, while a Poisson distribution of occupation numbers permits only a maximum single-atom occupancy of the fraction e^{-1} of sites. Even if it should not be possible to go beyond unity phase-space density using Raman sideband cooling alone, a simple cooling technique that provides significant improvement is important for various applications and precision experiments with cold atoms, while the phase-

space density attained for a given atom number in a given cooling time represents a good measure for the brightness of the cold atom source.

BEYOND CLOSED TRANSITIONS: CAVITY DOPPLER COOLING

When an atom of mass m illuminated by monochromatic light of wavevector \vec{k}_i scatters a photon into a mode with wavevector \vec{k}_s (Figure 3), the atom's momentum changes from $\vec{p} = m\vec{v}$ to $\vec{p}' = \vec{p} + \hbar\vec{k}_i - \hbar\vec{k}_s$, and its kinetic energy from $W = \vec{p}^2/2m$ to

$$W' = W + \hbar(\vec{k}_i - \vec{k}_s) \cdot \vec{v} + \frac{\hbar^2(\vec{k}_i - \vec{k}_s)^2}{2m}. \quad (1)$$

Energy conservation then requires that the angular frequency of the scattered photon differs from that of the incident photon by an amount

$$\Delta = ck_s - ck_i = \frac{W - W'}{\hbar} = -(\vec{k}_i - \vec{k}_s) \cdot \vec{v} - \frac{\hbar(\vec{k}_i - \vec{k}_s)^2}{2m}. \quad (2)$$

This formula shows that the frequency of the scattered light is determined by the Doppler effect along the two-photon wavevector $\vec{k}_i - \vec{k}_s$, while the last term describes recoil heating that is independent of the atomic velocity. If $\Delta > 0$, i.e. if the scattered photon is blue-detuned relative to the incident photon, the atom's kinetic energy will be reduced in the scattering event. An atomic gas is cooled if on average $\Delta > 0$, i.e. if on average the two-photon Doppler effect $\langle (\vec{k}_i - \vec{k}_s) \cdot \vec{v} \rangle$ is negative and exceeds the two-photon recoil heating $\langle \hbar(\vec{k}_i - \vec{k}_s)^2/2m \rangle$ in magnitude.

In conventional Doppler cooling, the direction of the scattered photon is random and therefore $\langle \vec{k}_s \cdot \vec{v} \rangle = 0$, while the desired negative Doppler effect for the incident beam $\langle \vec{k}_i \cdot \vec{v} \rangle < 0$ is achieved by tuning the laser frequency below the atomic

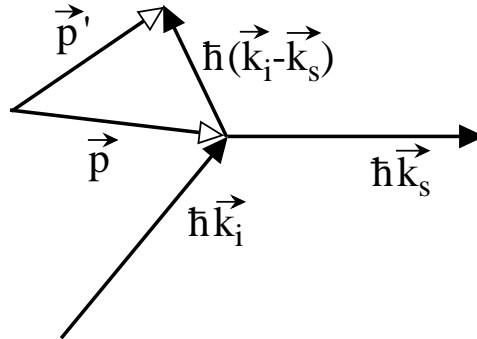


FIGURE 3. The atom scatters a photon from a mode with wavevector \mathbf{k}_i into a mode with wavevector \mathbf{k}_s and changes its momentum from \mathbf{p} to $\mathbf{p}' = \mathbf{p} + \hbar(\mathbf{k}_i - \mathbf{k}_s)$.

resonance, leading to a preferential absorption of photons from a beam opposing the atomic velocity. Conventional Doppler cooling requires a closed two-level system and a small detuning relative to the atomic resonance that is comparable to the Doppler effect of the moving atom. If the two-level system is not closed, the atom will be optically pumped to a different internal state where the detuning is different and typically large, and the cooling will stop.

To design a cooling scheme that works at arbitrary detuning from atomic transitions and that therefore can be applied to multilevel atoms, one can use an optical cavity to enhance scattering events where the two-photon Doppler effect $\langle (\vec{k}_i - \vec{k}_s) \cdot \vec{v} \rangle$, rather than the incident-photon Doppler effect $\langle \vec{k}_i \cdot \vec{v} \rangle$, is negative. In a quantum mechanical description the cavity changes the density of electromagnetic modes in a frequency-dependent manner [9,10] which according to Fermi's Golden Rule leads to a scattering rate that depends strongly on the frequency $ck_s = ck_i + \Delta$ of the scattered photon. If the cavity is blue-detuned relative to the incident light, it will enhance scattering events with $\Delta > 0$, i.e. those events where the scattered photon carries away a larger energy than that of the incident photon. Energy conservation then requires that the atom's energy is reduced by $\hbar\Delta$ in such a process.

In a classical description, the cavity acts to direct the scattered field back onto the atom with a phase that depends on the frequency of the scattered light. The field that has been reflected by the cavity mirrors interferes with the field scattered by the atom at a later time, "stimulating" further scattering through the addition of field amplitudes. This frequency-dependent feedback provided by the cavity leads to a scattering rate into the cavity mode that is proportional to the classical intensity enhancement function L of the cavity, in agreement with the quantum mechanical result. This cavity response function $L(\delta)$ not too far from resonance is given by the Lorentzian form [11,12]

$$L(\delta) = \frac{2E}{1 + \delta^2/\gamma_c^2}. \quad (3)$$

Here E is the cavity intensity enhancement factor that is related to the cavity finesse F by $E = F/\pi$, while δ is the detuning of the scattered light from cavity resonance and γ_c is the cavity decay rate constant for the field amplitude.

With respect to atomic properties, cavity-induced Doppler cooling relies on the fact that at low saturation of the atomic transition and at a detuning that exceeds the atomic linewidth, essentially all the scattered power is concentrated in the coherent Rayleigh scattering peak [13]. The phase and frequency of the scattered light are then completely determined by the incident light in combination with the atomic position and velocity. (For an atom fixed in space, the frequency spectrum of this classically scattered light is a delta function at the frequency of the incident light, while the amplitude depends on the real part of the atomic polarizability [13].) Since at low saturation the frequency of the scattered light is independent of the atomic level structure, temperatures below the atomic Doppler limit can be reached

with cavity Doppler cooling. Furthermore in contrast to conventional Doppler cooling the atomic properties and in particular the detuning between the incident light and the atomic transitions enter only through the free-space photon scattering rate. It follows that cavity Doppler cooling is possible at arbitrary detuning from atomic resonances, provided sufficient photon scattering rates can be achieved with an intense laser beam. This should enable one to cool simultaneously different atoms or isotopes in the same light field, and to cool atoms with a complicated level structure or even molecules that possess a sufficiently large polarizability in the optical frequency domain.

To calculate the cooling force, let us consider an atom scattering photons at a rate Γ_1 , where the index indicates that the atom is illuminated by a single linearly polarized running plane wave. The scattering rate Γ_w along one direction \vec{k}_s of a Gaussian mode with a polarization that is parallel to that of the incident beam and that has a waist w centered on the atom (Figure 4) is then given by $\Gamma_w = (3/k_s^2 w^2) \Gamma_1$. (This formula can be obtained by considering the dipole force that an atom experiences if an external light field is applied in the same mode. As the dipole force arises from the interference of the field that is already present in a particular mode with the scattered field, momentum conservation requires that the force on the atom equals the rate at which net momentum is carried away by the total (incident and scattered) electromagnetic field. The known expression for the dipole force [14] can then be used to calculate the scattered field.) The factor 3 takes into account that such a mode with parallel polarization is coupled three times stronger to the incident light field than the spatial average of the dipole pattern $\langle \cos^2 \theta \rangle = 1/3$, while the denominator $k_s^2 w^2$ defines the solid angle subtended by the scattering mode.

If a linear cavity is placed around the scattering mode, the modified scattering rate is obtained by multiplying the free-space scattering rate Γ_w into mode \vec{k}_s with the frequency-dependent cavity response function $L(\delta)$. For an atom moving at velocity \vec{v} , the scattered light will be shifted in frequency by the Doppler and

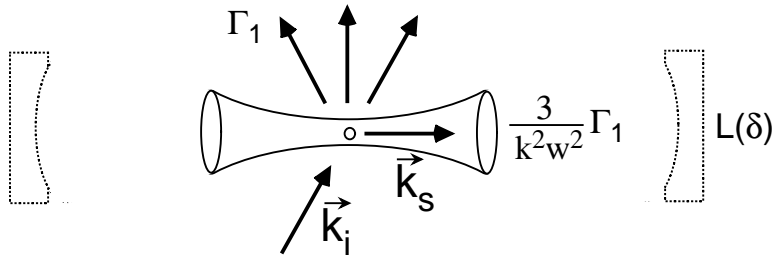


FIGURE 4. The atom with a total scattering rate Γ_1 scatters a fraction $3/k_s^2 w^2$ of the light along one direction of a mode with waist w . The polarization of the scattering mode is parallel to the polarization of the incident light.

recoil effects as described by Eq. (2). It follows that the photons scattered along the two directions of the cavity $\pm \vec{k}_s$ will be detuned by $\delta_{\pm} = \delta_l - (\vec{k}_i \mp \vec{k}_s) \cdot \vec{v} - \hbar(\vec{k}_i \mp \vec{k}_s)^2/2m$ relative to the cavity resonance. Here δ_l denotes the detuning of the incident light relative to the cavity resonance. The cooling force \vec{f} is simply given by the sum of the two possible scattering rates into the cavity mode $\Gamma_w L(\delta_{\pm})$, multiplied by the corresponding two-photon momenta $\hbar(\vec{k}_i \mp \vec{k}_s)$:

$$\vec{f} = \hbar(\vec{k}_i - \vec{k}_s)\Gamma_w L(\delta_+) + \hbar(\vec{k}_i + \vec{k}_s)\Gamma_w L(\delta_-). \quad (4)$$

Note the close resemblance of this formula with the expression for conventional Doppler cooling, where the absorption momentum $\pm \vec{k}_i$ has been replaced by the two-photon momentum $\vec{k}_i \mp \vec{k}_s$, the total free-space scattering rate Γ_1 has been replaced by the free-space scattering rate Γ_w into the cavity mode, and the atomic lineshape has been replaced by the cavity line $L(\delta)$ in the Lorentzian approximation. Eq. (4) clearly shows that cooling by means of an optical cavity at low saturation of the atomic transition can be viewed as a generalized Doppler cooling technique.

In the special case that the incident light is a standing wave that circulates inside the cavity or encloses a very small angle with the cavity mode, one finds the following result for the 1D cooling force [15]:

$$f = 2\hbar k \Gamma_{sc} \frac{12E}{k^2 w^2} \frac{2xy}{[1 + (2x + y)^2][1 + (2x - y)^2]} \quad (5)$$

Here $\Gamma_{sc} = 2\Gamma_1$ is the average free-space scattering rate in the standing wave, while $x = kv/\gamma_c$ and $y = (\delta_l - 4(\hbar k)^2/2m)/\gamma_c$ are the Doppler shift and recoil-shifted incident-light detuning relative to the cavity resonance, respectively, both normalized to the cavity linewidth. The cooling force is proportional to the free-space scattering rate in the incident beam of a given intensity, but otherwise independent of atomic properties. The force is cooling ($f \cdot v < 0$) if $y < 0$, i.e. if the incident light is red detuned *relative to the cavity resonance*, independent of the detuning relative to the atomic resonance. The last factor in Eq. (5) shows that the velocity dependence of the force, and therefore the cooling limit, are determined by the cavity parameters alone. The minimum attainable temperature is given by the usual Doppler limit $kT = \hbar\gamma_c$ for the Lorentzian cavity line in the case that the enhanced scattering into the cavity at a rate $(6E/k^2 w^2)\Gamma_{sc}$ exceeds scattering into free space that proceeds at the rate Γ_{sc} [15].

We point out that in the limiting case of low saturation of the atomic transition, the sole purpose of the cavity is to enhance the scattering into electromagnetic modes with a frequency which exceeds that of the incident light by the two-photon Doppler effect $-(\vec{k}_i - \vec{k}_s) \cdot \vec{v}$. Therefore it is by no means necessary that the incident light itself circulate inside the cavity, a feature which is less obvious in the time domain treatment [16,15] of the problem.

In the time domain description the atom acts as a position-dependent refractive index, tuning the cavity in and out of resonance as the atom moves along the standing wave, while the intracavity field changes with a time delay that is determined

by the resonator linewidth. The delayed circulating light field then acts back on the atom via the light shift, which under appropriate conditions gives rise to a Sisyphus-like cooling force [16]. This dissipative force is proportional to the square of the real part of the atomic polarizability α , since both the detuning of the cavity by the atom and the ac light shift exerted by the intracavity field on the atom are proportional to $\text{Re}(\alpha)$. It follows that in the classical limit the cooling force is proportional to the photon scattering rate as given by the imaginary part of the atomic polarizability [15].

To understand in the time domain picture why it is not necessary that the incident field circulate in the cavity, we note that the refractive index has its origin in the interference between the incident field and the field radiated in quadrature by the atom, which in lowest order leads to a phase shift of the total field. Even if the incident field encloses a nonzero spatial angle with the scattered field circulating inside the cavity, the interference between incident and scattered field still produces a delayed time-dependent light shift that can give rise to a cooling force.

The fact that the incident field need not circulate inside the cavity can be used to achieve cooling in all three spatial dimensions with a single optical resonator. Since the cooling mechanism depends on the two-photon Doppler shift $(\vec{k}_i - \vec{k}_s) \cdot \vec{v}$, for 3D cooling it is sufficient to illuminate the atom with four running waves along $\pm x$ perpendicular to the cavity axis that is oriented along z (Figure 5). Each incident running wave \vec{k}_i in combination with scattering along one direction $\pm \vec{k}_s$ of the cavity leads to a dissipative force along one diagonal direction $\vec{k}_i \mp \vec{k}_s$. (It is also possible to use a symmetric arrangement of three incident beams in the xy -plane.) The force is cooling if the cavity is blue detuned relative to the incident field. This

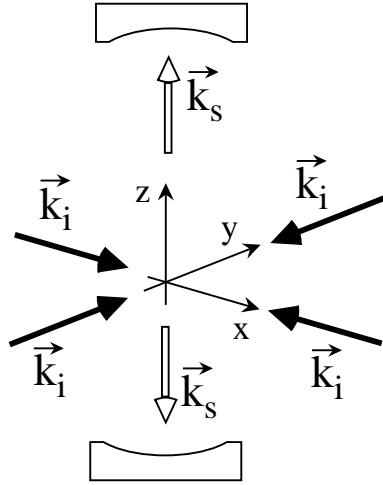


FIGURE 5. Setup for three-dimensional cooling using only one optical cavity. Since the cooling force is directed along the two-photon wavevector $\mathbf{k}_i - \mathbf{k}_s$, three-dimensional cooling can e.g. be achieved with the cavity oriented along the z axis and four incident beams along the $\pm x$ and $\pm y$ axes.

simple setup that requires only one high-finesse cavity should greatly facilitate 3D cavity Doppler cooling of an atomic or molecular gas.

When the effect of a single atom on the cavity resonance is small, i.e. when the atom inside the cavity changes the cavity resonance frequency by much less than a cavity linewidth, then a sample containing more than one atom will be cooled at the same rate as a single atom: If a sample of N uncorrelated atoms is placed inside the cavity, the effects of most of the atoms on the cavity will cancel each other. However, fluctuations from equilibrium will lead to an effective coupling parameter β_N between sample and cavity mode that is given by $\beta_N = \sqrt{N}\beta$, where $\beta = (k_s/\pi w^2)Re(\alpha)/\epsilon_0$ is the coupling between a single atom and the cavity [15]. As long as $\beta_N \ll 1$, i.e. as long as the coupling of the sample to the cavity is below saturation, the scattering rate into the cavity mode will be proportional to β_N^2 , and therefore to the atom number N . Therefore the sample will act as if it consisted of N independent scatterers, and it follows that the cooling time will be comparable to that of a single atom. (Note that the same line of argument applies to an optically thin sample of N independent scatterers in free space, where the total scattered power is simply N times the power scattered by a single atom, even when the scattering of the incident monochromatic radiation is coherent and the correct treatment is to add up the electric fields from all scatterers.) Therefore cavity Doppler cooling of a sample for $\beta_N \ll 1$ resembles Doppler cooling of an optically thin sample in free space, where the atoms act effectively as independent scatterers. This makes cavity Doppler cooling of large samples significantly more favorable than stochastic cooling [17], where the cooling slows down as the sample size increases. The scaling will be different and less favorable in the case of critical coupling $\beta \approx 1$ between the atom and the cavity [18], that although leading to a larger cooling force, is difficult to achieve with atoms at large detuning from atomic transitions and out of reach with molecules.

When the scattering rate of photons into the cavity by a single atom or by an atomic sample exceeds the cavity linewidth, the number of photons in steady state inside the resonator will exceed one, and the coherent coupling between atoms and scattered light becomes important. Although this situation will give rise to complicated dynamics of the particle motion and of the intracavity field, energy conservation requires that the atom's motion is cooled at the rate at which the blue-detuned photons leaking out of the cavity carry away the excess energy $\hbar\Delta = \hbar(ck_s - ck_i)$. Therefore even if the cavity contains more than one photon, the cooling force will be given by the simple formula Eq. (4) or (5) derived above.

In conclusion, cavity Doppler cooling relies on the classical scattering of light by particles and therefore may allow one to extend laser cooling to atomic or even molecular species with a complicated internal level structure. Simultaneous cooling of different species by the same light field is possible. The application of cavity Doppler cooling at large detuning from atomic transitions may open a new way to cool high-density samples. The free-space cooling considered here can be easily generalized to trapped particles (cavity sideband cooling) and to internal degrees of freedom, such as rovibrational states of molecules, or possibly even to transitions

in liquids or solids. The basic idea is always that the cavity is used to enhance the scattering of higher-energy and suppress the scattering of lower energy photons. The irreversibility of the coherent scattering process is ensured by the coupling of the intracavity field to the vacuum outside the cavity.

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