Laser Cooling of Atoms, Ions, or Molecules by Coherent Scattering

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We point out a laser cooling method for atoms, molecules, or ions at low saturation and large detuning from the particles' resonances. The moving particle modifies the field inside a cavity with a time delay characteristic of the cavity linewidth, while the field acts on the particle via the light shift. The dissipative mechanism can be interpreted as Doppler cooling based on preferential scattering rather than preferential absorption. It depends on particle properties only through the coherent scattering rate, opening new possibilities for optically cooling molecules or interacting atoms.

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Laser cooling [1], a tremendously successful technique for creating high-brightness atomic sources for various applications, has been limited to the alkalis, a small number of other atomic species, and several trapped ions [2]. Doppler cooling, which represents the dominant mechanism at all but the lowest velocities, is based on the preferential scattering of photons from a laser beam opposing the atomic motion into a random direction. Since many photons are required to significantly change the atom's momentum, cooling has been demonstrated only with species that can be optically cycled many times. In particular, molecules have a large number of vibrational and rotational states that cannot be addressed by the same frequency, while each of the many transitions carries only a small transition strength. Once the molecule is optically pumped into a different internal state, it no longer interacts with the light and the cooling ceases.

The application of many different laser frequencies to address molecules in all ground states simultaneously represents a possible solution that has been proposed for alkali dimers [3]. The alternative of cooling with an intense single-frequency beam coupling to all transitions simultaneously is prevented by the requirement for Doppler cooling that the laser detuning match the Doppler shift of the moving particle. A dissipative mechanism that allows large detuning is provided by the dipole force [4] in a blue-detuned standing wave [5]. This force, however, cools only on a saturated transition, which for large detuning requires exceedingly high laser power and is accompanied by strong heating due to force fluctuations [5].

In this Letter, we point out a general optical cooling scheme for the translational motion of polarizable particles at low saturation and large detuning from the particles' resonances. The cooling is achieved by coupling the particles to a far-detuned light field inside an optical cavity. The essence of this method and other ideas for atom cooling by means of a resonator [6–9] is that the cavity-induced frequency dependence of the electromagnetic mode density [6] can be used to tailor the energy transfer from laser field to atom and vacuum, and for strongly coupled single atoms heating has been observed recently [10]. In contrast to previous proposals that concentrate on two-level atoms [6,9], the present work is motivated by the application to molecules or dense samples of interacting multilevel atoms, and considers cooling by coherent scattering, which is dominant at large detuning and low saturation [11], rather than by incoherent spontaneous emission. When saturation is negligible, a classical analysis of the particle-cavity coupling yields a general and simple expression for the velocity-dependent force that resembles the Doppler cooling result. Incident light *red-detuned relative to the cavity* gives rise to a cooling force that is proportional to the coherent scattering rate, but otherwise *independent of the particle's internal level structure.* The temperature limit is estimated using quantum mechanical results for the recoil heating and the dipole force heating [5] in a two-level system. At not-too-high scattering rate, temperatures at or below the recoil limit can be reached. Possible applications of this technique include the cooling of molecules, of dense samples with interaction-induced level shifts, of species where lasers matching the transition frequencies are not readily available, and the simultaneous cooling of different isotopes.

When interpreted in the time domain, the cooling arises from the time-delayed adjustment of the optical potential to the atomic motion [9]. Consider an incident light field tuned to the red of the *cavity* resonance. An atom in the cavity alters the local index of refraction in proportion to the local intensity, and near an antinode will tune the cavity closer to (away from) resonance for red (blue) detuning δ_{at} relative to the *atomic* resonance. It follows that in both cases the intracavity intensity will be higher when the atom is at a minimum of the optical potential. A moving atom experiences a time-delayed potential due to the finite cavity response time, which slightly increases the decelerating force when the atom is moving away from the potential minimum, and slightly reduces the acceleration when the atom is moving towards the minimum. At large detuning δ_{at} this results in a net cooling force independent of the sign of the light shift.

In the frequency domain, the cooling originates from the cavity-induced asymmetry of the coherent scattering [11] peak. Because of the Doppler effect, backward scattered photons from the beam opposing the atomic motion are

blueshifted relative to the incident light, while backscattered photons from the copropagating beam are redshifted. (Photons scattered in the forward direction experience no frequency shift.) A cavity blue-detuned relative to the incident light will enhance the scattering of high-energy photons and suppress the scattering of low-energy photons, which transfers energy from the atom's motion into the light field that then leaks out of the cavity. Consequently the mechanism can be interpreted as Doppler cooling by coherent scattering, with a rate proportional to the cavity finesse and to the solid angle subtended by the cavity mode. While conventional Doppler cooling arises from a velocity-dependent absorption cross section [1], here the symmetry is broken by the scattered rather than the absorbed photon.

In the following we consider the one-dimensional motion of an atom, ion, or molecule inside a cavity. At low saturation we can model the atom as a classical oscillator with complex polarizability α [12,13] driven by a light field of frequency $\omega_L = ck$. Let $2E$ denote the field amplitude at an antinode inside the standing-wave cavity of length L (Fig. 1), $\tau = 2L/c$ the round-trip time, $\Delta = \omega_L - \omega_c$ the detuning of the incident light relative to the cavity resonance ω_c, and $\omega = kv_0$ the Doppler shift of the atom moving at velocity v_0. We consider a cavity with a damping dominated by the cavity linewidth γ_c, defined as the decay rate of the field amplitude. We further assume γ_c, Δ, $\omega \ll \tau^{-1}$, such that the field changes slowly compared to the round-trip time and retardation effects due to the atomic motion can be neglected. We introduce a dimensionless parameter $\beta = (k/\pi w^2)\,\mathrm{Re}(\alpha)/\varepsilon_0$, characterizing the coupling between the atom and a single TEM_{00} cavity mode with e^{-2} intensity waist w. An oscillating dipole radiates a field $\pi/2$ out of phase [12,13], which far from atomic resonances ($|\beta| \ll 1$) for an incident traveling Gaussian beam results in amplitude reflection and transmission coefficients $i\beta$ and $1 + i\beta$, respectively. (These coefficients, correct to first order in β, can be established by noting that the dipole force in a standing wave [4,5] arises from an interference of transmitted and reflected waves. Depending on the atom's position, the two resulting waves traveling to the left and to the right have in general different intensities, and carry away momentum at a rate that equals the dipole force.) A simple equation governing the time evolution of the amplitude E is then obtained by retaining the reflected and transmitted waves to first order in β:

$$\tau\dot{E}(t) = qE_i + [g_0 - 1 + 4i\beta \sin^2 kx(t)]E(t). \quad (1)$$

E_i is the time-independent incident field amplitude, each mirror has an amplitude reflection coefficient $-r$ and transmission coefficient q (Fig. 1), where r and q are real with $r^2 + q^2 = 1$, $g_0 = r^2 e^{2ikL}$ is the round-trip gain for the empty cavity, and $x(t)$ is the atom's distance from the second mirror at time t. According to Eq. (1), the atomic dipole, radiating a field in quadrature, acts as a refractive index that depends on the atom's position [9]. The atom has no effect at a node, where the field vanishes.

If the kinetic energy of the atom far exceeds the light shift, the atom's unperturbed motion $x(t) = x_0 + v_0 t$ can be inserted into Eq. (1). The solution to first order in β is given by $E = E_{\mathrm{st}}[1 + \beta(h_+ + h_-)]$, where $E_{\mathrm{st}} = qE_i/(1 - g_0 - 2i\beta)$ is the stationary field amplitude, and

$$h_\pm(t) = \pm e^{\mp 2ikx_0}\,\frac{e^{\mp 2i\omega t} - e^{i\Omega t}}{(2\omega \pm \Omega)\tau}. \quad (2)$$

Here the Doppler shift $\omega = kv_0$ is real, while the complex frequency $\Omega = i(1 - g_0)/\tau \approx i\gamma_c + \Delta$ characterizes the cavity. The term $e^{i\Omega t}$ associated with the transient cavity response decays away on a time scale γ_c^{-1} and is ignored in the following. Equation (2) shows that the moving atom modulates the intracavity field and creates Doppler sidebands with different amplitudes $h_\pm$ at frequencies $\omega_L \pm 2\omega$. Such sidebands can be used to extract the particle motion inside the cavity [14].

The atom-induced fractional change of the intracavity power to first order in β is given by $2\beta\,\mathrm{Re}(h_+ + h_-)$. The light field exerts a position-dependent dipole force $f = f_{\mathrm{st}} + f_\eta$ on the atom, where f_{st} is due to the steady-state field E_{st}, while $f_\eta = f_{\mathrm{st}} 2\beta\,\mathrm{Re}(h_+ + h_-)$ originates from the small atomic motion-induced intensity variation. The atomic velocity v_0 is perturbed by these forces to $v = v_0 + v_{\mathrm{st}} + v_\eta$ with $f_{\mathrm{st}} = m\dot{v}_{\mathrm{st}}$ and $f_\eta = m\dot{v}_\eta$, where m is the atom's mass. The dissipative work ΔE done on the atom to lowest order is then given by $\Delta E = v_0 \int f_\eta\, dt + \int (f_\eta v_{\mathrm{st}} + f_{\mathrm{st}} v_\eta)\, dt$. The last integral is simply $m v_{\mathrm{st}} v_\eta$ and negligible compared to the first term $m v_0 v_\eta$ if the atom's kinetic energy far exceeds the optical potential. Dissipative work is done on the atom at a rate $P = v_0 f_\eta$ that is averaged over one wavelength with the result

$$\overline{P} = \frac{8k}{\pi w^2}\,\frac{d^2}{\varepsilon_0}\,\frac{\omega^2}{\tau}\,\frac{\mathrm{Re}(\Omega)\,\mathrm{Im}(\Omega)}{|4\omega^2 - \Omega^2|^2}. \quad (3)$$

The square of the induced dipole $d = \mathrm{Re}(\alpha)E_{\mathrm{st}}$ establishes a relation to the Larmor formula for the power

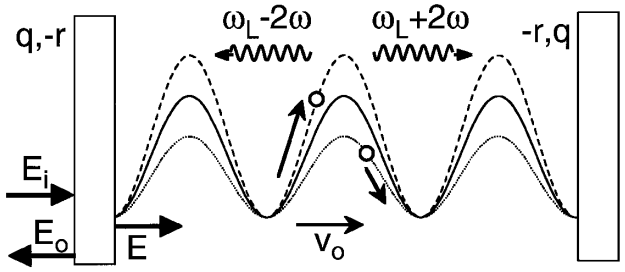

FIG. 1. Cooling of an atom. The dashed and the dotted lines indicate the optical potential when the atom (open circle) with velocity v_0 is traveling uphill and downhill in the optical potential, respectively. The coherently backscattered photons parallel and antiparallel to v_0 are frequency shifted by $2\omega = 2kv_0$ and -2ω, respectively.

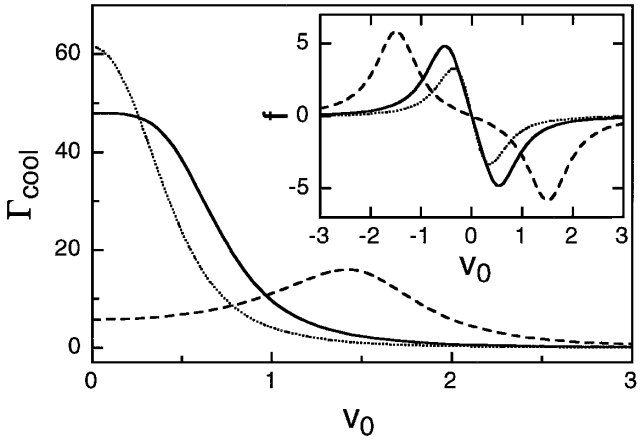

FIG. 2. The cooling rate constant $\Gamma_{\rm cool} = 2\overline{P}/mv_0^2$ [in units of $\Gamma_{\rm sc}(E_{\rm rec}/\hbar\gamma_c)Q/k^2w^2$] as a function of atomic velocity v_0 (in units of γ_c/k) for detunings of $\Delta = -\gamma_c/2$ (dotted line), $\Delta = -\gamma_c$ (solid line), and $\Delta = -3\gamma_c$ (dashed line) relative to the cavity. The inset shows the cooling force in units of $\hbar k\Gamma_{\rm sc}Q/k^2w^2$.

radiated by an oscillating dipole [12]. The corresponding averaged free-space scattering rate is $\Gamma_{\rm sc} = k^3d^2/6\pi\varepsilon_0\hbar$. Using $\Omega \approx i\gamma_c + \Delta$, the power transferred to the atom can finally be written as

$$\overline{P} = \hbar\gamma_c\Gamma_{\rm sc}\frac{12Q}{k^2w^2}\frac{(2x)^2y}{[1 + (2x + y)^2][1 + (2x - y)^2]}. \tag{4}$$

Here $Q = q^{-2} = (\gamma_c\tau)^{-1}$ is the cavity intensity enhancement factor [12], while $x = \omega/\gamma_c$ and $y = \Delta/\gamma_c$ are the normalized Doppler shift and detuning of the incident light relative to the cavity, respectively. Equation (4) has been derived without the rotating-wave or two-level approximations. It has the same form as the result for conventional Doppler cooling [1,4], if the width of the atomic transition and the detuning from atomic resonance $\delta_{\rm at}$ are replaced by the cavity linewidth γ_c and the detuning from cavity resonance Δ, and the free-space scattering rate $\Gamma_{\rm sc}$ is replaced by the enhanced scattering rate into the cavity mode with solid angle $(kw)^{-2}$, given by $\Gamma_{\rm sc}12Q/(kw)^2$. For small waist size w this rate can significantly exceed the free-space rate, since mirrors allowing $Q \geq 10^6$ have been demonstrated [10]. (Note, however, the trade-off between cooling rate that scales as w^{-2}, and cooling volume, given by $2z_Rw^2 = 2\pi w^4/\lambda$ for $z_R < L$ and πw^2L for $z_R > L$, where z_R is the Rayleigh range.) Cooling arises if the incident light is red-detuned relative to the cavity ($\Delta = \gamma_c y < 0$), independent of the sign of the light shift. In contrast to other methods, cooling is therefore possible at large detuning $\delta_{\rm at}$, provided that the intensity is sufficient to achieve the desired scattering rate. Conventional Doppler cooling or heating is suppressed by a factor $kv_0/\delta_{\rm at}$.

Figure 2 shows the cooling rate constant, $\Gamma_{\rm cool} = \overline{P}/W$, defined as ratio of cooling power and atomic kinetic energy $W = mv_0^2/2$, as a function of velocity. If $\Delta = -\gamma_c$ is chosen, the cooling power for small velocities $|2kv_0| < \gamma_c$ is given by

$$\overline{P}_0 = -E_{\rm rec}\Gamma_{\rm sc}\frac{W}{\hbar\gamma_c}\frac{48Q}{k^2w^2}, \tag{5}$$

where $E_{\rm rec} = (\hbar k)^2/2m$ the recoil energy.

The dissipative mechanism can be interpreted as Doppler cooling by cavity-induced frequency-dependent scattering. The resonator enhances scattering into modes with higher frequency than the incident light, which according to Eq. (2) produces asymmetric sidebands $\beta h_\pm$ at frequencies $\omega_L \pm 2\omega$ in the light E_0 that leaks out of the cavity (Fig. 1). The power gain relative to the incident light must equal the cooling power on the atom [14], which can be used to derive Eq. (3). Optimum cooling occurs when the denominator $(|2\omega| + \Delta) - i\gamma_c$ in the term $\beta h_\pm$ is minimized, i.e., when the cavity detuning $-\Delta$ equals twice the Doppler shift ω (compare Fig. 2). While in conventional Doppler cooling the direction of the scattered photon is random, here the cooling proceeds with backscattered photons, which accounts for the factor 2. In fact, since the time-varying refractive index associated with the moving atom leads to a modulation of the cavity resonance at frequency 2ω, cooling occurs for light tuned to the red sideband $\omega_c - |2\omega|$ of the system "cavity + atom." (Compare Ref. [7] for trapped ions.) Note that the recoil shift increases the entropy of the coherently scattered light over that of the incident light, and the dissipative mechanism presented here does not violate the general result that time-dependent conservative potentials cannot give rise to cooling [15].

As long as the cavity mode subtends a solid angle $(kw)^{-2} \ll 1$, scattering into free space will heat the atom at a rate $2E_{\rm rec}\Gamma_{\rm sc}$, while cavity-enhanced scattering, as calculated from the modified electromagnetic mode density [6], heats at a rate $2E_{\rm rec}\Gamma_{\rm sc}(6Q/k^2w^2)$ for $\Delta = -\gamma_c$ and $|2kv_0| \ll \gamma_c$. The calculated temperature limit of $k_BT = (h\gamma_c/2)(1 + k^2w^2/6Q)$ can be smaller than $E_{\rm rec}$. In analogy to Doppler cooling on a narrow transition, one expects for $\hbar\gamma_c/2 < E_{\rm rec}$ the temperature to be limited to the recoil energy [16], although even lower values of order $\hbar\gamma_c$ may be attainable with broadband incident light [17].

In a cavity, heating due to dipole force fluctuations [5] for $|kv_0| \ll \gamma_c$ can be well approximated by the free-space value in a standing wave at an intensity determined by the atom's position [18]. Since the fractional change of intracavity intensity is of order βQ, the heating rate for $|\beta|Q \ll 1$ is the same as in free space and for a two-level system with excited state lifetime Γ^{-1} given by [5]

$$P_{\rm dip} = 20E_{\rm rec}\Gamma_{\rm sc}\left(\frac{\Gamma_{\rm sc}}{\Gamma}\right)^3\left(\frac{\delta}{\Gamma}\right)^2. \tag{6}$$

For fixed scattering rate the heating increases with detuning or intensity, but its strong dependence on $\Gamma_{\rm sc}$ allows one to reach significantly lower temperatures at a reduced cooling rate. Dipole heating is negligible if $\Gamma_{\rm sc}/\Gamma \ll (1 + 6Q/k^2w^2)^{1/3}(\Gamma/\delta)^{2/3}$.

Regarding the cooling of a large sample, Eq. (4) contains only the scattering rate and implies a cooling rate that is independent of atom number. On the other hand, if N atoms were to move phase coherently relative to the lattice, the coherent interaction would lead to a force on each atom that is N times larger than for a single particle. Since on average $\sqrt{N}$ more or less atoms will be found at a node than at an antinode, the corresponding $\sqrt{N}$ times larger friction force acting on a fraction $1/\sqrt{N}$ of the sample will lead to a cooling time comparable to that of single atom. Similarly, the effective coupling β_N between sample and cavity will be approximately $\sqrt{N}$ times larger than the single-atom parameter β. Collective motions of the atoms mediated by the cavity that could enhance the force fluctuation heating are negligible for $2|\beta_N|Q \ll 1$ [19].

The above classical analysis is valid as long as the photon number p in the scattering mode $\omega_{\rm sc} = \omega_L + 2k\upsilon$ is smaller than one. For M participating atoms, the balance of scattering into $\omega_{\rm sc}$ at a rate $M\Gamma_{\rm sc}(3Q/k^2w^2)$ (only one out of four different forward and backscattering events contributes) and damping at a rate $2\gamma_c$ leads to an average photon number given by $\overline{p} = (M\Gamma_{\rm sc}/2\gamma_c)(3Q/k^2w^2)$. For $p \geq 1$ the p-dependent Rabi frequency $\Omega_p = (p+1)^{1/2}\Omega_0$, where Ω_0 is the vacuum Rabi frequency, will govern the complicated photon exchange between laser and scattering mode. However, since independent of the exact evolution the photons in the scattering mode $\omega_{\rm sc}$ leave the cavity through the end mirrors and are blue-detuned relative to the incident light ω_L, energy conservation implies a cooling power per atom of $\overline{p}2\gamma_c(\hbar\omega_{\rm sc} - \hbar\omega_L)/M = \hbar\Gamma_{\rm sc}k\upsilon(6Q/k^2w^2)$. This shows that even for $\overline{p} \geq 1$ the cooling proceeds at the classical rate given by Eq. (4).

As an atomic example, let us consider a sample of fermionic ^{6}Li atoms in a far-detuned CO_2 dipole trap [20]. We assume that a cavity mode with $Q = 10^3$, linewidth $\gamma_c/2\pi = 80$ kHz, and waist size $w = 12\lambda$ is superimposed. The detuning $\delta_{\rm at}$ from the D line and the scattering rate are chosen $\delta_{\rm at} = 10^4\Gamma$ and $\Gamma_{\rm sc} = 10^2{\rm s}^{-1}$, respectively, yielding negligible dipole heating. For 10^6 trapped atoms the coupling and the collective parameter are $\beta_N \approx 5 \times 10^{-5}$ and $2\beta_N Q \approx 0.1$. The calculated cooling limit of 3.7 μK exceeds only slightly the recoil energy of 3.5 μK. Note that the method is well suited for cooling dense samples, possibly to quantum degeneracy, since it is unaffected by interaction-induced level shifts.

As a molecular example, we consider cooling of Cs_2 on the B-X band. We estimate for $\lambda \approx 766$ nm the polarizability for the lower vibrational levels of the electronic ground state to be ${\rm Re}(\alpha)/\varepsilon_0 \approx 2 \times 10^{-26}$ m^3 [3]. Choosing $w = 35$ μm, $L = {\rm kw}^2 = 1$ cm, $\Gamma_{\rm sc} = 4 \times 10^4$ s^{-1}, and $Q = 2 \times 10^4$, we find a maximum deceleration of 10^2 m/s^2, available for a velocity class of 0.1 m/s width. A larger velocity range can be addressed by readjusting the detuning as the atoms are slowed ("chirped cooling" [2,4]).

In summary, we have proposed a laser cooling method that can be used at large detuning and low saturation to cool particles inside an optical cavity. Light tuned below the cavity resonance then gives rise to Doppler cooling by coherent scattering. The method depends on molecular parameters only through the scattering rate and should be applicable to atoms, molecules, or ions.

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