

## External-feedback laser cooling of molecular gases

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We analyze the laser cooling of a gas of polarizable particles by continuous dispersive position detection and active feedback. Microkelvin temperatures can be attained inside an optical resonator, while in free space cooling requires wavelength-size beams. The maximum cooling flux is set by the thermal Doppler width, with typical values between  $10^5$  and  $10^9$  molecules per second.

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A dissipative force on a particle can arise from an otherwise conservative potential in combination with a time delay. For instance, in polarization gradient cooling of atoms [1], delayed optical pumping between internal states results in a friction force. In stochastic cooling in particle accelerators [2], an electronic delay between a position measurement and a correction force is introduced, and in cavity cooling [3,4] an optical resonator provides the delay. Electronic feedback cooling has been applied to classical [5] and quantum systems [6–10], and theoretical concepts have been extended into the quantum domain [10–13]. For accelerators, an optical version of stochastic cooling has also been proposed [14].

The feedback cooling of an atom strongly coupled to an optical resonator has been analyzed [15], and in cavity experiments, single atoms have been trapped, but not cooled, by applying feedback [16,17]. In free space, various cooling schemes based on optical measurement and feedback [18–20] have been proposed, and in an experiment, a sample's center-of-mass oscillation has been damped [21] by feedback. A method using an optical resonator similar to the one analyzed below [22], has been numerically studied by Lu and Barker [23]. However, with the exception of Refs. [10,15,22], most treatments do not properly include the photon recoil heating that necessarily accompanies the optical measurement and feedback. As discussed below, this can lead to erroneous conclusions regarding the feasibility of the proposed method for a given detection geometry.

Here we analyze the laser cooling of a gas of polarizable molecules by continuous dispersive position measurement and optical feedback [22,23]. We show that the dissipative force is proportional to the radiation pressure associated with Rayleigh scattering into the detector, while its velocity dependence is determined by the frequency dependence of the loop gain. The maximum *collective* cooling rate for a sample, limited by collective heating, is given by the thermal Doppler effect. This results in a typical cooling flux of  $10^8$  molecules/s at room temperature, and  $10^5$  molecules/s at a sample temperature of  $100 \mu\text{K}$ . Using a resonator for

signal enhancement, the method can be applied to any gas of particles that scatter light at a sufficiently large rate.

The dissipative force arises from the application of a time-dependent conservative optical potential in response to an optical measurement of the particle's motion [22,23]. Consider a single molecule or atom of mass  $m$  moving as  $x_m = vt$  at constant velocity  $v$  in a weak periodic potential  $V(x, t) = U(t) \cos(2kx)$  whose depth  $U(t) \ll \frac{1}{2}mv^2$  depends on the molecule's motion. Since the time variation of the force on the molecule  $f(t) = 2kU(t) \sin(2kvt)$  arises both from the spatial variation  $\cos(2kx)$  of the potential, and from the dependence of the depth  $U(t)$  on the molecule's trajectory, a frequency component of  $U(t)$  in phase with the molecular-motion-induced force variation  $\sin(2kvt)$  produces a nonzero average force that can heat or cool the molecule. A standing light wave can be used to both monitor the position  $x_m(t)$  of the polarizable particle by means of its index of refraction [3,4,17], and to generate a periodic potential of adjustable depth  $U(t)$  via the light shift.

For a sample instead of a single particle, and linear feedback, the self-generated dissipative force of each molecule remains unchanged, while the fluctuations in  $U(t)$  due to other molecules constitute a source of heating. However, the heating is quadratic in loop gain, whereas the cooling varies linearly with gain [2]. Therefore the collective heating limits only the cooling rate, but not the final temperature attainable at low loop gain.

The optical signal arising from the particle's motion, and consequently the cooling force, can be increased by means of an optical resonator (Fig. 1). Light of angular frequency  $ck$  incident onto a standing-wave resonator of finesse  $F$ , waist size  $w$ , and field decay rate constant  $\gamma_c$ , produces an electric field of amplitude  $2E_c$  at an antinode ( $x=0$ ) inside the resonator. A molecule with complex polarizability  $\alpha$  moving on the resonator axis coherently scatters photons into free space at an average rate  $\Gamma_{sc} = k^3 |\text{Re}(\alpha)E_c|^2 / (6\pi\epsilon_0\hbar)$ , and experiences an optical potential  $U_0 \cos(2kx)$  of depth  $U_0$

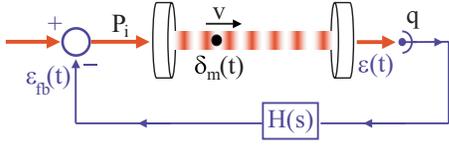


FIG. 1. (Color online) Schematic of continuous-feedback cooling. The moving molecule periodically detunes the standing-wave resonator by  $\delta_m(t)$ . The observed fractional change in cavity transmission  $\varepsilon(t)$  acts as an error signal that is applied to change the input power  $P_i$  by a fraction  $-\varepsilon_{fb}(t)$ . The feedback open-loop gain  $H(s)$  is set by an external electronic circuit.

$=-|E_c|^2 \text{Re}(\alpha)/2$ . Conversely, the forward-scattered field, or equivalently, the molecule's index of refraction, results in a molecular-position-dependent detuning  $\delta_m = \zeta \gamma_c \cos(2kx_m)$  of the resonator from its average resonance frequency  $\omega_c$  [3,4]. Here  $\zeta = \hbar \eta \Gamma_{sc} / U_0$  is a dimensionless parameter characterizing the molecule-cavity coupling, and the single-atom cooperativity  $2\eta = 12F / (\pi k^2 w^2)$  can be interpreted as the fraction of photons scattered into the resonant cavity.

For incident light detuned by an amount  $\delta_i = ck - \omega_c$  relative to the resonator, the time-varying detuning  $\delta_m(t)$  of the resonator by the moving molecule changes the resonator transmission by a fraction  $\varepsilon(t)$ . The transmitted power is measured and used as an error signal in a feedback loop to adjust the incident power by a fraction  $-\varepsilon_{fb}(t)$  (Fig. 1). In order to ignore cavity-induced forces arising from the finite response time of the resonator [3,4], we take its linewidth  $2\gamma_c$  to be much larger than both the feedback bandwidth and the Doppler frequency  $2kv$ , such that the intracavity power  $P_c(t)$  adjusts instantaneously to a value determined by the total light-resonator detuning  $\delta_i(t) = \delta_i - \delta_m(t)$ . If both the molecule and the feedback loop produce only small changes ( $|\delta_m|/\gamma_c, |\varepsilon_{fb}| \ll 1$ ), then the fractional deviation  $\varepsilon(t) = P_c(t)/P_0 - 1$  of the intracavity power  $P_c(t)$  from its unperturbed value  $P_0$  (for  $\delta_m = \varepsilon_{fb} = 0$ ) is

$$\varepsilon(t) = r \gamma_c^{-1} \delta_m(t) - \varepsilon_{fb}(t). \quad (1)$$

The moving molecule modulates the intracavity power by an amount  $r \delta_m / \gamma_c$  proportional to the normalized resonator slope  $r = 2 \delta_i \gamma_c / (\gamma_c^2 + \delta_i^2)$ , while the feedback loop adjusts the incident power by a fraction  $-\varepsilon_{fb}(t)$ . If the molecule's kinetic energy far exceeds the light shift  $U_0$ , then to lowest order  $\delta_m(t)$  is determined by the molecule's unperturbed motion  $x_m = vt$ . We introduce the open-loop feedback gain  $H(s)$  for the Laplace transformed quantities  $\tilde{\varepsilon}_{fb}(s)$ ,  $\tilde{\varepsilon}(s)$  via  $\tilde{\varepsilon}_{fb} = H \tilde{\varepsilon}$ . Then  $H(i\omega) = H_1(\omega) + iH_2(\omega)$ , with real and imaginary parts  $H_1$  and  $H_2$ , respectively, is the complex gain in the frequency domain. If the loop is stable, the steady-state solution of Eq. (1) in the time domain is given by

$$\varepsilon(t) = r \zeta \frac{[1 + H_1(2kv)] \cos(2kvt) + H_2(2kv) \sin(2kvt)}{|1 + H(2ikv)|^2}. \quad (2)$$

$H_1(2kv)$  and  $H_2(2kv)$  are the open-loop gain in phase and in quadrature with the molecular-motion-induced intensity modulation  $\delta_m(t) = \zeta \gamma_c \cos(2kvt)$ , respectively. They deter-

mine the closed-loop signal  $\varepsilon(t)$ , and thereby the time variation of the optical-potential depth  $U(t) = U_0[1 + \varepsilon(t)]$ . In the limit  $U_0 \ll \frac{1}{2}mv^2$ , work is done on the molecule at a rate  $\dot{W} = \varepsilon(t) f_u(t) v$  to lowest order, where  $f_u = 2kU_0 \sin(2kvt)$  is the unperturbed force. The component  $\sin(2kvt)$  of  $\varepsilon(t)$  in phase with  $f_u$  produces a dissipative force with time average  $f = \langle \dot{W} \rangle / v$  given by

$$f(v) = \hbar k \eta \Gamma_{sc} \frac{r H_2(2kv)}{|1 + H(2ikv)|^2}. \quad (3)$$

This expression, valid for arbitrary laser detuning from molecular resonances below saturation, shows that the friction force  $f$  depends on molecular parameters exclusively through the Rayleigh scattering rate  $\Gamma_{sc}$ .  $f$  is proportional to the rate of momentum transfer  $2\hbar k \eta \Gamma_{sc}$  due to backward scattering into the resonator at rate  $\eta \Gamma_{sc}$ , multiplied by a dimensionless function of the molecule velocity. In particular, the sign and velocity dependence of  $f$  are determined by the frequency-dependent loop gain  $H(i\omega)$ . The dissipative force  $f$  is maximum for a resonator-light detuning  $\delta_i = \pm \gamma_c$  that gives the largest slope  $r = \pm 1$ . In the following we assume  $\delta_i = -\gamma_c$  ( $r = -1$ ), such that  $H(s) > 0$  corresponds to negative feedback.

The cooling force  $f$  is proportional to the quadrature component  $\sin(2kvt)$  of the intracavity light modulation in closed loop, given by  $H_2/|1+H|^2$ , evaluated at the Doppler frequency  $2kv$ . In particular, for very small or very large open-loop gain ( $|H|^2 \ll 1$  or  $|H|^2 \gg 1$ ), this relevant quadrature of the intensity variation, and hence  $f$ , will be small. The velocity-dependent term takes on its maximum value  $1/(2 + 2H_1)$  when the quadrature gain  $H_2$  and the in-phase gain  $H_1$  are related by  $|H_2| = |1 + H_1|$ . When  $|1 + H_1| \ll 1$ , the feedback loop regeneratively amplifies the intensity variation caused by the moving molecule. However, the heating of the molecule due to noise amplification is then also increased.

The simplest stable cooling loop is a differentiator [17]  $H_d(i\omega) = i\omega / (2ku)$ , with unity gain frequency  $2ku$ . (The gain can be rolled off outside the velocity range of interest to the cooling.) We assume for simplicity that the laser is far detuned from molecular resonances compared to the Doppler shift  $2kv$ , such that  $\Gamma_{sc}$  is independent of  $v$ . The differentiator-induced cooling force  $f_d$  is then given by

$$f_d(v) = -\hbar k \eta \Gamma_{sc} \frac{uv}{u^2 + v^2}. \quad (4)$$

The maximum force  $\hbar k \eta \Gamma_{sc} / 2$ , attained for the unity-gain velocity  $v = u$ , is the same as in conventional Doppler cooling at the photon scattering rate  $\eta \Gamma_{sc} / 2$ , but the differentiator force, falling off as  $1/v$  rather than  $1/v^3$ , provides a significantly larger velocity capture range. Figure 2 shows that higher-order loops can substantially extend the velocity capture range while maintaining the low-velocity friction coefficient  $\alpha_0 = \partial f / \partial v$  that determines the final temperature.

Technical or quantum noise  $\hat{\varepsilon}(t)$  in the light-induced potential  $U(t) = U_0[1 + \hat{\varepsilon}(t)]$  will heat a particle moving at velocity  $v$  in proportion to the spectral noise density at the modulation frequency  $2kv$  of the unperturbed force. We define the single-sided fractional spectral noise density  $S_c(\omega)$

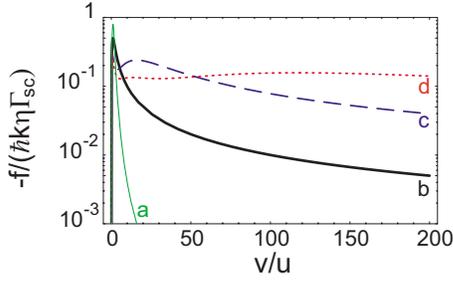


FIG. 2. (Color online) Cooling force versus molecular velocity  $v$  for conventional Doppler cooling (a), and for feedback cooling with loop gain  $H(s)=s$  (differentiator, b),  $H(s)=s(1+s/10)/(1+8s/10)$  (c), and  $H(s)=s(1+s/10)(1+s/100)/[(1+8s/10)(1+s/20)]$  (d). Here  $s=iv/u$ , where  $u$  sets the unity gain frequency for the differentiator loop.

$= (2/\pi) \int_0^\infty d\omega \langle \hat{\epsilon}(0)\hat{\epsilon}(t) \rangle \cos(\omega t)$ , normalized such that  $\int_0^\infty d\omega S_c(\omega) = \langle \hat{\epsilon}^2(0) \rangle$  is the mean-square fractional intensity noise, and calculate the heating power due to the force fluctuations [24],

$$\dot{W}_{\text{fluct}} = \frac{\pi k^2}{m} U_0^2 S_c(2kv). \quad (5)$$

This general formula describes the heating of a molecule moving at velocity  $v$  due to classical intracavity intensity noise with spectral noise density  $S_c(\omega)$ . For quantum noise, Eq. (5) needs to be doubled to take into account atomic-dipole fluctuations [24]. Neglecting technical noise, the photon shot noise of the incident light with power  $P_i$  corresponds to a fractional power spectral density given by  $S_i^{\text{psn}} = \hbar ck / (P_i \pi)$ . In closed loop, the intracavity noise density is  $S_c^{\text{psn}}(\omega) = S_i^{\text{psn}} / |1 + H(i\omega)|^2$  for noise frequencies  $\omega \ll \gamma_c$ . In addition, uncorrelated shot noise in the detector photocurrent at limited quantum efficiency  $q < 1$  arising from the undetected photons will cause the feedback loop to generate (classical) intracavity intensity noise. This results in a fractional noise density  $S_c^{\text{sn}} = S_i^{\text{psn}}(q^{-1} - 1) |H|^2 / |1 + H|^2$  in closed loop. The total shot noise  $S_c^{\text{sn}} = S_c^{\text{psn}} + S_c^{\text{sn}}$ , expressed in terms of the intracavity power  $P_c = P_i F / (2\pi)$  for the detuned cavity with  $|\delta_i| = \gamma_c$ , and inserted into Eq. (5) then yields a closed-loop heating  $\dot{W}_{\text{sn}}$  due to shot noise

$$\dot{W}_{\text{sn}} = E_r \eta \Gamma_{\text{sc}} \frac{2 + |H(2ikv)|^2 (q^{-1} - 1)}{|1 + H(2ikv)|^2}, \quad (6)$$

where  $E_r = \hbar^2 k^2 / (2m)$  is the recoil energy. We see that  $\dot{W}_{\text{sn}}$  can be expressed as the recoil heating rate  $2E_r \eta \Gamma_{\text{sc}}$ , multiplied by a dimensionless function of the loop gain  $H$  and detector quantum efficiency  $q$ . In the absence of feedback ( $H=0$ ), Eq. (6) reproduces the well-known recoil heating [24] due to scattering into the resonator, where each back-scattering event, occurring at a rate  $(\eta/2)\Gamma_{\text{sc}}$  for the detuned cavity, heats the molecule by an amount  $4E_r$ . For unity quantum efficiency  $q=1$ , the feedback loop reduces (increases) this heating by suppressing (enhancing) the intracavity fluctuations for  $|1+H| > 1$  ( $|1+H| < 1$ ). For  $q < 1$ , the feedback loop also generates intensity noise originating from random

photoelectron detection. In addition to the cavity heating  $\dot{W}_{\text{sn}}$ , scattering into free space heats the molecule at a rate  $\dot{W}_{\text{fs}} = 2E_r \Gamma_{\text{sc}}$  for isotropic scattering [24].

For the differentiator loop  $H_d(i\omega) = i\omega / (2ku)$ , the choice  $u = (q^{-1} - 1 + 2\eta^{-1}) \hbar k / m$  for the unity-gain velocity  $u$  minimizes the equilibrium temperature  $T_d$ , yielding

$$k_B T_d = 4E_r (\eta^{-1} + 1) (2\eta^{-1} + q^{-1} - 1), \quad (7)$$

where  $k_B$  is Boltzmann's constant. The temperature scales with the recoil energy  $E_r$ , but strongly depends on the cooperativity  $\eta$ , i.e., the molecule-cavity coupling. In the bad-cavity limit  $\eta \ll q < 1$  the final temperature is independent of detector quantum efficiency  $q$ , and given by  $T_d = 8\eta^{-2} E_r / k_B$ . For a typical bad-cavity setup with  $\eta = 0.05$  for Cs atoms [25], this corresponds to a temperature of  $320 \mu\text{K}$ , while the good-cavity limit  $\eta \gg 1$  [16,17] with  $q = 0.2$  would correspond to  $T_d = 1.6 \mu\text{K}$ .

For a thermal sample consisting of  $N+1$  molecules at temperature  $T = mv_{\text{th}}^2 / k_B$ , any chosen probe molecule at velocity  $v$  will be heated by the intracavity intensity noise induced by the other  $N$  randomly moving molecules. Substituting the corresponding closed-loop spectral noise density into Eq. (5) yields the collective heating rate  $\dot{W}_N$  as follows:

$$\dot{W}_N(v) = \frac{E_r \eta \Gamma_{\text{sc}}}{|1 + H(2ikv)|^2} \frac{\sqrt{2\pi} N \eta \Gamma_{\text{sc}}}{2kv_{\text{th}}} \exp\left(-\frac{v^2}{2v_{\text{th}}^2}\right). \quad (8)$$

As is typical of stochastic cooling, the heating overwhelms the cooling at too large sample size  $N$  or too large photon scattering rate  $\Gamma_{\text{sc}}$ . The collective heating rate is quadratic rather than linear in  $\Gamma_{\text{sc}}$ , and can be reduced below the single-particle heating at the expense of cooling speed. The cooling rate, however, is limited by the collective heating. For a differentiator loop, the net cooling power averaged over the thermal sample,  $\langle \dot{W}_d \rangle = \langle f_d v + \dot{W}_N \rangle$  is maximized for  $u \approx v_{\text{th}}$ . The cooling rate constant  $\gamma_d = -2\langle \dot{W}_d \rangle / (k_B T)$  can then be written in the form

$$\gamma_d \approx \frac{kv_{\text{th}}}{6N} (2\bar{\Gamma}_{\text{cav}} - \bar{\Gamma}_{\text{cav}}^2), \quad (9)$$

where  $\bar{\Gamma}_{\text{cav}} = 2N \eta \Gamma_{\text{sc}} \hbar / (k_B T)$  is the total scattering rate into the cavity normalized to the sample temperature. The cooling rate is maximized for  $\bar{\Gamma}_{\text{cav}} = 1$ , yielding a rate constant  $\gamma_d = kv_{\text{th}} / (6N)$ . We see that the thermal Doppler broadening  $kv_{\text{th}}$  takes the role of the stochastic-cooling bandwidth [2], and that the cooling of smaller subsamples can proceed faster [2,18]. For a sample of  $N = 10^8$  magnetically trapped CaH molecules at  $0.4 \text{ K}$  [26] cooled with light of wavelength  $760 \text{ nm}$ , the optimum cooling rate is  $\gamma_d = 0.1 \text{ s}^{-1}$ , attained at a photon scattering rate  $\eta \Gamma_{\text{sc}} = 3 \times 10^2 \text{ s}^{-1}$ . (Here we are assuming that the different degrees of freedom are mixed [2], e.g., by the trapping potential [18].) An ensemble of  $10^6$  CaH molecules at room temperature can be cooled at a rate  $\gamma_d = 340 \text{ s}^{-1}$  for a photon scattering rate of  $\eta \Gamma_{\text{sc}} = 3 \times 10^7 \text{ s}^{-1}$ . Figure 3 shows the differentiator cooling flux  $\gamma_d N$  for different sample sizes, ranging between  $10^6$  and  $10^9$  molecules/s for temperatures between  $100 \text{ mK}$  and

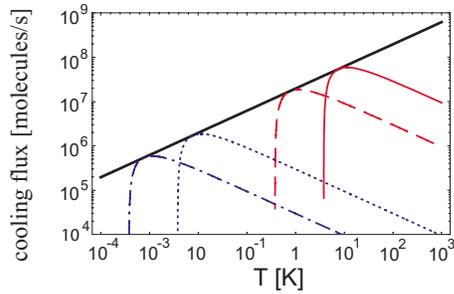


FIG. 3. (Color online) Differentiator cooling flux (cooling power divided by  $k_B T$ ) versus temperature  $T$  for various free-space scattering rates  $\Gamma_{sc}$  and sample sizes  $N$ , calculated for CaH with  $\lambda = 760$  nm inside a bad cavity ( $\eta = 0.05$ ). The parameters are (a)  $N = 10^6$ ,  $\Gamma_{sc} = 10^7$  s $^{-1}$  (thin solid line), (b)  $N = 10^5$ ,  $\Gamma_{sc} = 10^7$  s $^{-1}$  (dashed), (c)  $N = 10^8$ ,  $\Gamma_{sc} = 10^2$  s $^{-1}$  (dotted), and (d)  $N = 10^8$ ,  $\Gamma_{sc} = 10$  s $^{-1}$  (dash-dotted). The thick solid line is the optimum rate with  $\Gamma_{sc}$  adjusted to maintain  $\bar{\Gamma}_{cav} = 1$ .

1000 K. This is somewhat lower than, but comparable to, the atomic fluxes attained with standard laser cooling methods [27].

The prediction of heating for too large cooling speed  $\bar{\Gamma}_{cav} > 2$  by Eq. (9) is not necessarily correct. The reason is that self-organization of the atom-light system [25] may oc-

cur. The corresponding change from single-atom Rayleigh scattering to much stronger Bragg scattering by the sample could then lead to new collective effects not captured by the above model.

The role of the resonator is to enhance the photon scattering rate  $\eta\Gamma_{sc}$  into the detector, i.e., the signal-to-noise ratio [19]. In the absence of the resonator,  $\eta$  is replaced by the detection solid angle  $\Delta\Omega = 3/(k^2 w^2)$ , preventing cooling altogether except for the smallest beam size. For instance, the cooling limit for Rb atoms using a beam as small as  $w = 10$   $\mu\text{m}$ , according to Eq. (7) corresponds to  $T_d = 370$  K, i.e., no observable cooling. This explains why stochastic cooling has not been observed in free-space experiments using macroscopic beam size  $w \gg k^{-1}$  for detection [18,21].

In conclusion, we have derived simple analytic expressions for the cooling and heating of a gas of linearly polarizable particles interacting with a laser beam whose intensity is adjusted in response to the particles' motion. The velocity capture range can be tailored via the frequency-dependent external-loop gain, while the thermal Doppler width takes the role of the stochastic-cooling bandwidth, determining the maximum cold-molecule flux.

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