Limits of Doppler Cooling in Pulsed Laser Fields

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As a result of transient atomic behavior, the achievements of laser cooling in pulsed fields are very different from those in continuous fields. Semiclassical and quantal treatments are used to identify the conditions for optimal cooling. For light atoms and narrow lines, energies below the one-photon recoil limit are obtained.

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Cooling of atoms and ions by near-resonant laser radiation is a powerful tool to improve conditions for a variety of atomic-physics experiments.[1] If the field interacts with the atoms for a long time, the stochastic nature of spontaneous emission implies, in classical terms, diffusion in addition to a friction force. In the case of a short interaction time, spontaneous emission has less effect, and the interaction causes deflection and diffraction of the atomic motion.[2] Pulsed cooling of antihydrogen has been considered, because intense Lyman-α radiation has so far been generated by pulsed lasers only,[3] and the use of π pulses has been described as a means for increasing the light pressure force in intense fields.[4] Further interest relates to the application of laser cooling in heavy-ion storage rings, where the interaction between the ions and the laser field is effectively being chopped as the ions circulate in the ring.[5,6] This paper presents a theoretical analysis of laser cooling in chopped fields, with results deviating significantly from the results obtained in continuous (cw) fields. The limiting atomic kinetic energies in cw laser cooling are proportional to or larger than the one-photon recoil energy,[7] which corresponds typically to microkelvin temperatures. Since a major source of imprecision in atomic frequency standards is the kinetic motion of the atoms, it is an important result of the present study that, in pulsed fields, atoms with very long-lived excited states can be cooled below this limit into the nanokelvin regime. The corresponding narrow lines may be good candidates for new frequency standards.

To distinguish different regimes, we introduce the dimensionless quantity

$$m = M\Gamma/\hbar k^2 , \tag{1}$$

where M is the atomic mass, Γ is the natural width, and k is the wave number of the atomic transition. If m is much larger than unity, the Doppler effect is insensitive to the velocity change $\hbar k/M$ caused by absorption or emission of a single photon, and a semiclassical analysis of the atomic motion is valid. This gives for cooling in a weak standing wave the mean kinetic energy of the atoms[1,7]

$$\bar{E}_K = \tfrac{7}{80}\,(2|\delta|/\Gamma + \Gamma/2|\delta|)\hbar\Gamma . \tag{2}$$

Here, δ is the (negative) detuning of the laser with respect to the atomic transition, and the lowest temperature $k_B T_{\min} = 2\bar{E}_{K,\min} = \frac{7}{20}\hbar\Gamma$ obtains for $\delta = -\Gamma/2$. If m is small, a quantum treatment of the atomic motion is required. For atoms continuously absorbing and emitting light, the finite photon momentum implies a cooling limit proportional to the recoil energy,[7]

$$\bar{E}_{K,\min} \simeq 0.53(\hbar k)^2/2M , \tag{3}$$

where $\delta' = \delta - \hbar k^2/2M \simeq -2.2\hbar k^2/M$ is the corresponding recoil-shifted frequency detuning. In a periodic field of short pulses, the mean kinetic energy will oscillate as a function of detuning around the cw result (2) for $m \gtrsim 1$, but there will be no improvement of the minimum value. For $m \ll 1$, however, the use of a pulsed field will lead to kinetic energies below the recoil energy.

We first consider a weak monochromatic traveling-wave field that is periodically turned on and off in time intervals τ and θ [Fig. 1(a)]. For simplicity we assume that the atoms decay completely between pulses. Within a semiclassical analysis the solution of the optical Bloch equations for the atomic density matrix is known. We shall need the excited-state population only,[8]

$$\Pi_e(t,v) = \begin{cases} \dfrac{\kappa^2/4}{(\delta - kv)^2 + \Gamma^2/4}\left[1 + e^{-\Gamma t} - 2e^{-\Gamma t/2}\cos(\delta - kv)t\right], & 0 < t \le \tau, \\ e^{-\Gamma(t-\tau)}\Pi_e(\tau,v), & \tau < t \le \tau + \theta, \end{cases} \tag{4}$$

where κ denotes the Rabi frequency of the atom-field interaction, $\kappa \ll \Gamma$, and where v is the atomic velocity along the wave vector of the laser field. The v dependence is due to the Doppler effect. In Fig. 1(a) are shown examples of the population of the excited state for two different velocities. With each spontaneous emission, a laser photon with momentum $\hbar\mathbf{k}$ is converted into a fluorescence photon emitted with equal probability in opposite directions. The average atomic momentum change per light pulse therefore equals $\hbar k$ times the spontaneous emission probability, and since we assume that atoms excited at $t=\tau$ decay before the next pulse, this probability, which we consider to be much smaller than unity, reads

$$P(v)=\int_0^\tau \Gamma\Pi_e(t,v)dt+\Pi_e(\tau,v)$$
$$=\frac{\kappa^2/4}{(\delta-kv)^2+\Gamma^2/4}\left\{\Gamma\tau+2-2e^{-\Gamma\tau/2}\cos(\delta-kv)\tau - \frac{2\Gamma}{(\delta-kv)^2+\Gamma^2/4}\left[\frac{\Gamma}{2}-e^{-\Gamma\tau/2}\left(\frac{\Gamma}{2}\cos(\delta-kv)\tau-(\delta-kv)\sin(\delta-kv)\tau\right)\right]\right\}. \quad (5)$$

$P(v)$ is shown in Fig. 1(b) for two values of τ and δ. Generalization to different pulse shapes amounts in the weak-field limit to a modification of the function $P(v)$, and is deferred to a later publication. With the optical Bloch equations it is easily shown that $\hbar kP(v)$ equals the integral of the light-induced mean force on the atoms. Since $P(v)\ll 1$, both the average value and the variance of the number of photons emitted after N pulses equal $NP(v)$ (Poisson process). The angular distribution of spontaneous emission contributes $\frac{2}{5}(\hbar k)^2$ per photon to the atomic momentum variance along the axis of interest, and the mean increase is $(1+\frac{2}{5})(\hbar k)^2P(v)$ per laser pulse. In many applications of weak standing waves we can simply add the contributions from each of the traveling-wave components to obtain the average force and diffusion coefficient,

$$\bar F(v)=\overline{\frac{d}{dt}\langle p\rangle}=\hbar k\frac{P(v)-P(-v)}{\tau+\theta},$$
$$\bar D(v)=\overline{\frac{1}{2}\frac{d}{dt}\mathrm{Var}(p)}=\frac{7}{10}(\hbar k)^2\frac{P(v)+P(-v)}{\tau+\theta}. \quad (6)$$

In the limit of very heavy atoms, the (quasi) stationary velocity distributions are Gaussians with $\bar E_K=\frac{1}{2}\bar D/(-d\bar F/dv)|_{v=0}$. If τ is large, the first term in the curly brackets in Eq. (5) dominates, and we recover the usual cw Doppler cooling result. But for $\Gamma\tau$ of the order of 1, the friction and diffusion coefficients oscillate as functions of detuning. In Fig. 2, the resulting energy $\bar E_k$ is shown for an infinitely heavy atom in the cw case, and for $\tau=4\Gamma^{-1}$. As a function of detuning the energy oscillates around the cw result, but it exceeds the minimum

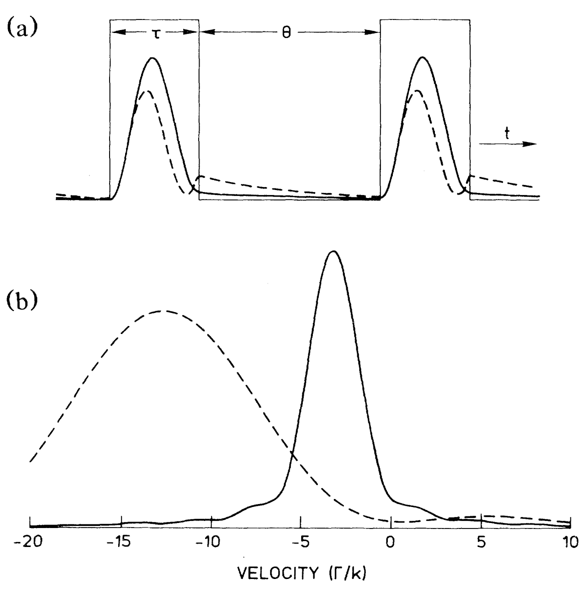

FIG. 1. (a) The time-dependent intensity in a chopped field. With the pulse duration $\tau=\Gamma^{-1}$, $\theta=2\Gamma^{-1}$, the excited-state population is indicated for atoms with velocity $v=0$ (solid curve) and $v=\Gamma/k$ (dashed curve); $\delta=-2\pi\Gamma$. (b) $P(v)$ [Eq. (5)] for two choices of the pulse duration and detuning: $\tau=2\Gamma^{-1}$ and $\delta=-\pi\Gamma$ (solid curve); $\tau=0.5\Gamma^{-1}$ and $\delta=-4\pi\Gamma$ (dashed curve).

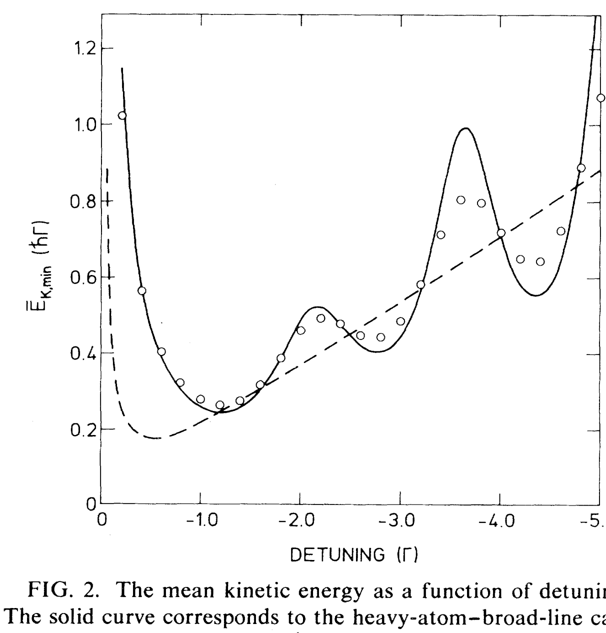

FIG. 2. The mean kinetic energy as a function of detuning. The solid curve corresponds to the heavy-atom–broad-line case for a pulse duration $\tau=4\Gamma^{-1}$; the dashed curve shows the cw results. The open circles are for $\tau=4\Gamma^{-1}$, but for atoms with $m=20$.

of Eq. (2). For any value of τ the minimum energy can be found numerically, and for pulse durations between $0.01\Gamma^{-1}$ and $4\Gamma^{-1}$ it is approximated within a few percent by the expression $\bar{E}_{K,\min} \simeq 0.45(\Gamma\tau)^{-1/2}\hbar\Gamma$. As $\tau \to 0$, $\tau\delta_{\text{opt}} \to -2\pi$: The atoms make a complete (Rabi) oscillation back to the ground state during the light pulse. For some values of δ, $P(v)$ has positive slope around $v=0$ and the force leads to heating. This point was also made in Ref. 3, supported by Monte Carlo simulations with both square and Gaussian laser pulses. When the mass is finite, the velocity distribution explores the functions $\bar{F}(v)$ and $\bar{D}(v)$ over a wider range, and with $m=20$ the stationary solution to the Fokker-Planck equation for the momentum distribution gives the mean kinetic energy, indicated by open circles in Fig. 2. Since both $\bar{F}(v)$ and $\bar{D}(v)$ are proportional to κ^2, the stationary velocity distributions are independent of the intensity in weak fields.

We now turn to a quantum description, valid also when $m<1$. Here we consider counterpropagating laser fields of orthogonal polarizations and a $j=0$ to $j=1$ atomic transition. The laser photons of momentum $\pm\hbar\mathbf{k}$ couple the ground state $|g\rangle$ to the excited states $|e\pm\rangle$, and coherences exist only within the closed families of atomic internal and momentum eigenstates $\{|g,p\rangle, |e+,p+\hbar k\rangle, |e-,p-\hbar k\rangle\}$. This simplifies the quantum treatment, and the generalized optical Bloch equations,[7] which are not restricted to large values of $\Gamma\theta$ or small values of κ/Γ, are solved numerically with a chopped field. The time evolution shown in Fig. 3 is obtained with parameters realistic for fine-structure transitions in heavy ions. With $M=200$ amu, $\Gamma=10^4\ \text{s}^{-1}$, and $2\pi/k=3500$ Å, the quantity m equals 0.1. With an energy of 100 keV in the Aarhus storage ring ASTRID,[6] the ions spend $\tau=25\ \mu\text{s}=0.25\Gamma^{-1}$ in the 8-m cooling section and $\theta=100\ \mu\text{s}=\Gamma^{-1}$ in the dark parts of the ring. A Rabi frequency $|\kappa|=2\Gamma$ per wave is used together with a detuning of $\delta'=-25\Gamma$ for ions with the average velocity in the beam. A possible redistribution among excited states between pulses is neglected, but the varying position of the atoms upon entrance to the cooling section is accounted for by a random phase angle.

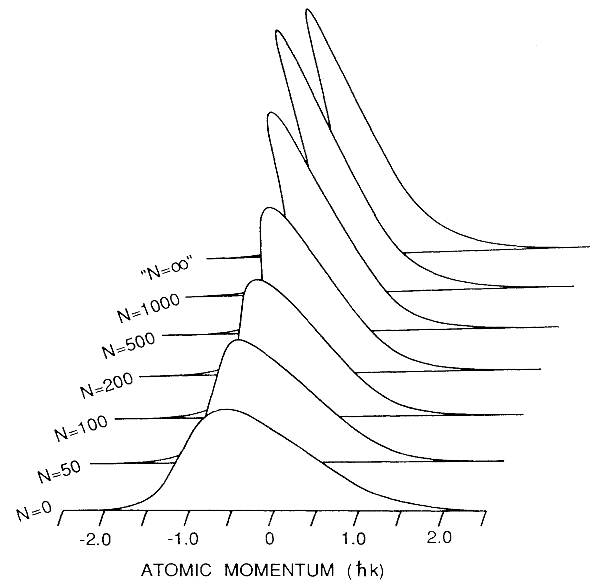

FIG. 3. Pulsed cooling of atoms with $m=0.1$; N denotes the number of pulses. The initial distribution is the coldest one obtainable in cw fields; the "$N=\infty$" curve is the steady-state solution of Eq. (7). The calculations are performed within the interval $-10\hbar k \le p \le 10\hbar k$ with a discretization of fifteen points per $\hbar k$.

The lifetime of the beam in a storage ring may be of the order of seconds,[5,6] and it is therefore realistic to let the distribution evolve over a thousand round trips, or light pulses. The initial distribution in the calculation is the coldest one obtainable with cw light, and it is notable that further cooling below the recoil energy occurs.

In weak fields we can give a simpler description based on ground-state populations only. Changes in the momentum distribution $w(p)$ occur when atoms, excited from the state $|g,p\rangle$, decay to the ground states of neighboring momentum families, $|g,p\pm\hbar k+u\hbar k\rangle$, $-1\le u\le 1$, with the distribution function of fluorescence photon momentum, $N(u)=\frac{3}{8}(1+u^2)$. Equation (4) yields the ratio between the populations of $|e\pm,p\pm\hbar k\rangle$ and $|g,p\rangle$ in weak fields and the change in the distribution per pulse is

$$\Delta w(p) = -[P(p/M)+P(-p/M)]w(p) + \int_{-1}^{1} du\, N(u)\left[P\left(\frac{p-\hbar k+u\hbar k}{M}\right)w(p-\hbar k+u\hbar k)+P\left(-\frac{p+\hbar k+u\hbar k}{M}\right)w(p+\hbar k+u\hbar k)\right]. \quad (7)$$

This applies for all values of the quantity m, provided δ' replaces δ in Eqs. (4) and (5). In the heavy-atom-broad-line limit, variations in $w(p)$ are small on the photon momentum scale, and from a second-order expansion of the integrand in Eq. (7) around the value p, and the substitution $\Delta w(p)=(\tau+\theta)dw(p)/dt$, we obtain a Fokker-Planck equation with the force and the diffusion coefficient of Eq. (6). In the light-atom-narrow-line limit the steady state can be found as the solution to Eq. (7), written as a band matrix equation with $\Delta w=0$. The "$N=\infty$" curve in Fig. 3 was determined in this way, and it is in good agreement with the results based on the generalized optical Bloch equations. When τ is small, and $\delta'\simeq -2\pi/\tau$, the probability of changing momentum family shows a minimum around $p=0$ [Fig. 1(b)]. This im-

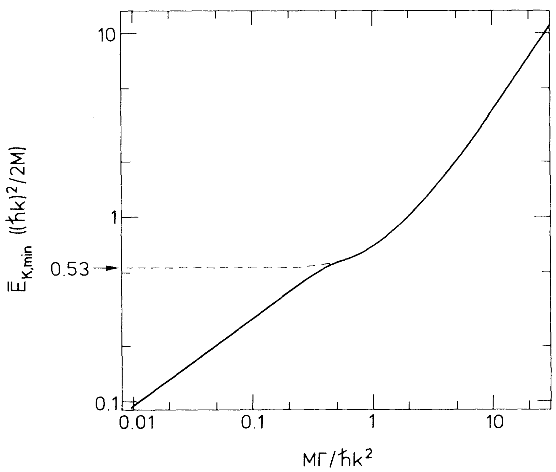

FIG. 4. The minimum kinetic energy as a function of $m = M\Gamma/\hbar k^2$. The dashed curve shows the cw result.

plies that atoms spend more time at $p=0$ than at higher momenta. The Rabi oscillation mimics a "dark resonance" at $v=0$,[9] and cooling below the recoil energy can be understood if the width of the minimum in the probability function $P(v)$ is smaller than $\hbar k/M$. Around $v=0$, $P(v)$ depends on velocity through the quantity $kv\tau$, and the pulse duration should not be too small on the scale $M/\hbar k^2$. On the other hand, the minimum in $P(v)$ disappears when τ is too large, cf. Fig. 1(b), and with Eq. (7), the pulse duration and detuning leading to minimum energy have been found for different values of m. For $m \ll 1$ we find an optimum pulse duration independent of the excited-state lifetime, $\tau_{opt} \simeq 2.5M/\hbar k^2$, and a detuning $\delta'_{opt} \simeq -2\pi/\tau_{opt} \simeq -2.5\hbar k^2/M$, which is not far from the one pertaining to cw conditions. But the minimum energy, shown in Fig. 4, is well approximated by the expression $\bar{E}_{K,min} \simeq 0.65[\hbar\Gamma(\hbar k)^2 2M]^{1/2}$, and this is very different from the cw result.

A single pulse of the monochromatic field corresponds to a frequency distribution centered around the laser frequency ω_L, $I(\omega) \propto \sin^2[\frac{1}{2}\tau(\omega-\omega_L)]/(\omega-\omega_L)^2$. Were the moving atoms to interact independently with each frequency component, the velocity dependence of the total photon scattering rate would be very similar to the function $P(v)/(\tau+\theta)$, cf. Fig. 1(b). This provides a qualitative interpretation of our results in terms of frequencies; a broadband laser with zero intensity at the resonance of atoms with velocities smaller than $\hbar k/M$ was recently suggested for cooling below the recoil limit in the light-atom-narrow-line case.[10]

We have seen that the transient atomic behavior leads to fundamentally new results for pulsed Doppler cooling of two-state atoms. Since not only the amplitude of the applied laser field but, for instance, also its polarization can be varied during experiments, a number of new observable effects might be anticipated in laser cooling with explicitly time varying fields.

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[1] See, for example, the special issues of J. Opt. Soc. Am. B **2**, 1706–1860 (1985), edited by P. Meystre and S. Stenholm; **6**, 2019–2278 (1989), edited by S. Chu and C. Wieman.

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