Doppler cooling with coherent trains of laser pulses and a tunable velocity comb

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We explore the possibility of decelerating and Doppler cooling an ensemble of two-level atoms by a coherent train of short, nonoverlapping laser pulses. We derive analytical expressions for mechanical force exerted by the train. In frequency space the force pattern reflects the underlying frequency comb structure. The pattern depends strongly on the ratio of the atomic lifetime to the repetition time between the pulses and pulse area. For example, in the limit of short lifetimes, the frequency-space peaks of the optical force wash out. We propose to tune the carrier-envelope offset frequency to follow the Doppler-shifted detuning as atoms decelerate; this leads to compression of atomic velocity distribution about comb teeth and results in a "velocity comb"—a series of narrow equidistant peaks in the velocity space.

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I. INTRODUCTION

Laser cooling is one of the key techniques of modern atomic physics [1–3]. Radiative force originates from momentum transfer to atoms from a laser field and subsequent spontaneous emission in random directions. The Doppler effect makes the force velocity dependent.

Here we develop a systematic theory of Doppler cooling by a coherent train of short laser pulses (see Fig. 1). A qualitatively new effect comes into play: atomic quantum-mechanical amplitudes induced by subsequent pulses *interfere*, resulting in a periodically varying radiative force as a function of frequency. This structure of the force reflects the comblike pattern of the Fourier image of the pulse train, the so-called frequency comb (FC) [4]. Here we derive the force and show that, for sufficiently weak pulses and long atomic lifetimes, each tooth acts as if it were an independent cw laser. In the opposite limit of short lifetimes (short compared to the repetition time between pulses), we recover the force due to an isolated laser pulse. Earlier works on the mechanical effects of FCs include a proposal involving two-photon transitions [5]. Following the proposal in [6], pulse trains from mode-locked lasers were also used in cooling experiments [7,8]. An analytical analysis of the FC's radiative force is presented here.

Notice that over the past few years the power and spectral coverage of FCs have grown considerably. A fiber-laser-based FC with 10 W average power was demonstrated [9] and the authors argue that the technology is scalable above 10 kW average power. The spectral coverage was expanded from optical frequencies to ultraviolet and to IR spectral regions [10]. These advances pave the road for new applications of FCs, such as laser cooling.

As an application, we consider mapping the frequency comb to a "velocity comb." We demonstrate that, during pulse-train cooling, continuous velocity distributions gravitate toward a series of sharp peaks (of a typical Doppler width of m/s for strong lines and mm/s for weak lines such as intercombination transition in Sr) in the velocity space. Velocity combs could be used for studying velocity-dependent (e.g., shape) resonances where traditional beam techniques with their broad velocity distributions would fail [11]. Moreover, since groups of atoms with different velocities would arrive at the target at different times, the experiment may be carried out "in parallel" for many velocities (cf. molecular fingerprinting [12].) Notice that the moniker "velocity comb" was used in a work [13] on optical pumping with FCs; we retain this label here as a natural visual for the resulting velocity distribution.

II. SETUP

In a typical FC setup, a train of phase-coherent pulses is produced by multiple reflections of a single pulse injected into an optical cavity. A short pulse is outcoupled every round trip of the wave packet inside the cavity, determining a repetition time T between subsequent pulses. At a fixed spatial coordinate, the electric field of the train may be parametrized as

$$\mathbf{E}(t) = \hat{\varepsilon} E_p \sum_m \cos(\omega_c t - \phi_m) g(t - mT), \tag{1}$$

where $\hat{\varepsilon}$ is the polarization vector, E_p is the field amplitude, and ϕ_m is the phase shift. The frequency ω_c is the carrier frequency and $g(t)$ is the shape of the pulses. We normalize $g(t)$ so that $\max g(t) \equiv 1$; then E_p has the meaning of the peak amplitude. While typically pulses have identical shapes and $\phi_m = m\phi$, one may want to install an active optical element at the output of the cavity as in Fig. 1 that could vary the phase and the shape of the pulses. Also repetition time and intensity of pulses could be controlled by varying the reflectivity of the cavity mirror.

We focus on two-level systems as these are amendable to analytic treatment and much insight may be gained from analyzing the derived expressions. Technically, we solve the optical Bloch equations (OBEs) for density matrix elements (excited- and ground-state populations are ρ_{ee} and ρ_{gg} and coherences ρ_{eg} and ρ_{ge})

$$\dot{\rho}_{ee} = -\gamma \rho_{ee} + \frac{i}{2}(\rho_{ge}\Omega_{eg}(z,t) - \rho_{eg}\Omega_{ge}(z,t)), \tag{2}$$

$$\dot{\rho}_{eg} = -\left(\frac{\gamma}{2} - i\delta_{\text{eff}}\right)\rho_{eg} + \frac{i}{2}\Omega_{eg}(z,t)(\rho_{gg} - \rho_{ee}), \tag{3}$$

where $\delta_{\text{eff}} = \delta + \mathbf{k}_c \cdot \mathbf{v}$ is the Doppler-shifted detuning ($\delta = \omega_c - \omega_{eg}$, $k_c = 2\pi/\omega_c$, and v is the atomic velocity). The time- and space-dependent Rabi frequency is $\Omega_{ge}(z,t) = \Omega_p \sum_{m=0}^{N-1} g(t + z/c - mT) e^{i\phi_m}$, with the peak Rabi frequency $\Omega_p = \frac{E_p}{\hbar}\langle e|\mathbf{D}\cdot\hat{\varepsilon}|g\rangle$ expressed in terms of the dipole

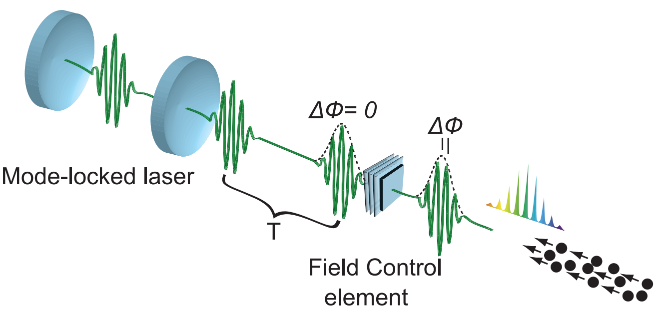

FIG. 1. (Color online) Schematic of a typical experimental setup. An atomic beam is slowed and cooled by a train of laser pulses. The phase and shape of pulses may be varied in time to attain optimal cooling.

matrix element. Once the OBEs are solved, radiative force may be determined in terms of the coherence

$$F_z = -p_r \,\mathrm{Im}[\rho_{eg}\Omega^*_{eg}], \tag{4}$$

where $p_r = \hbar k_c$ is the photon recoil momentum.

We start by observing that, as long as the duration of the pulse is much shorter than the repetition time, the atomic system behaves as if it were subject to a perturbation by a series of δ-function-like pulses. In this limit, the only relevant parameter affecting the quantum-mechanical time evolution is the effective area of the pulse, $\theta = \Omega_p \int_{-\infty}^{\infty} g(t)dt$. As an illustration we use a Gaussian-shaped pulse, $g(t) = e^{-t^2/2\tau_p^2}$; we assume that $\tau_p \ll T$.

We distinguish between prepulse (left) and postpulse (right) elements of the density matrix; e.g., $(\rho^m_{eg})_l$ and $(\rho^m_{eg})_r$ are the values of coherences just before and just after the mth pulse. Between the pulses the dynamics is determined by the spontaneous decay

$$\begin{aligned}\rho_{eg}(t) &= \left(\rho^m_{eg}\right)_r \exp\left[-\left(\frac{\gamma}{2} - i\delta_{\text{eff}}\right)(t - mT)\right],\\ \rho_{ee}(t) &= \left(\rho^m_{ee}\right)_r \exp[-\gamma(t - mT)].\end{aligned} \tag{5}$$

We neglect the spontaneous decay *during* the pulse, since for femtosecond pulses $\tau_p\gamma \ll 1$. Then

$$(\rho^m)_r = e^{i\theta/2\sigma_m}(\rho^m)_l e^{-i\theta/2\sigma_m}, \tag{6}$$

with $\sigma_m = \cos\phi\,\sigma_x - \sin\phi\,\sigma_y$, where $\sigma_{x,y}$ are the Pauli matrices. Analogs of Eqs. (5) and (6) were derived earlier [14]. By stacking single-pulse (6) and free-evolution (5) propagators, one may evolve a given initial ρ over the duration of the entire train. In Fig. 2 we show results of such a calculation for the excited-state population (atom remains at rest).

III. RADIATIVE FORCE

Now we focus on the evaluation of the radiative force. The laser field is present only during the pulse, so we deal with a sum over instantaneous forces. The change in the atomic momentum due to a single pulse is

$$\frac{-\Delta\mathbf{p}_m}{p_r} = \left[\left(\rho^m_{ee}\right)_r - \left(\rho^m_{ee}\right)_l\right]\hat{\mathbf{k}}_c; \tag{7}$$

i.e., a laser pulse imparts a fractional momentum kick equal to the difference of populations before and after the pulse. Since

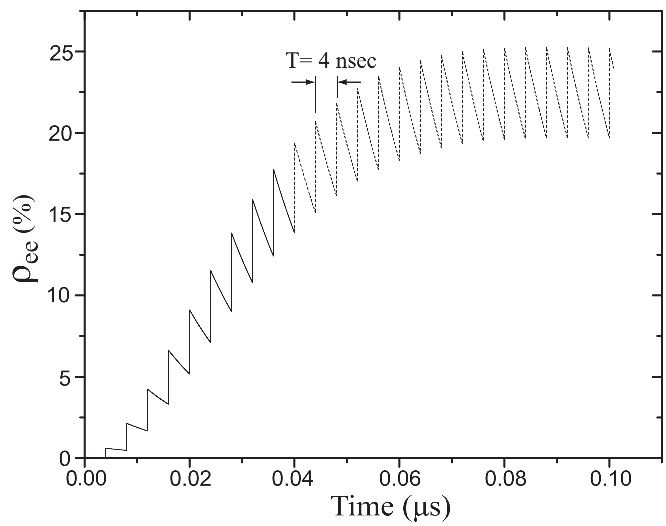

FIG. 2. Evolution of the excited-state population due to interaction with a train of laser pulses. The atom is initially in the ground state, and it is driven by a train of pulses separated by $T = 4$ ns and of pulse area $\theta = \pi/10$. Radiative lifetime is 16 ns.

$0 \leqslant \rho_{ee} \leqslant 1$, the maximum momentum kick per pulse is equal to the recoil momentum.

By combining Eqs. (5)–(7) we find the radiative force. The time evolution of the population, Fig. 2, separates into two regimes: an initial transient phase and the quasi-steady-state (QSS) regime when the radiative-decay-induced drop in the population following a given pulse is fully restored by the subsequent pulse. Doppler cooling requires many scattering cycles and we focus on the QSS (or the "coherent accumulation" [14]) regime.

In the QSS regime, $\rho_{ee}(t) = \rho_{ee}(t + nT)$ and pre- and postpulse values $(\rho^m_{ee})_{l,r}$ do not depend on the pulse number m; we simply denote these values as $(\rho^s_{ee})_{l,r}$. Then Eq. (7) becomes $-\Delta p_s/p_r = (\rho^s_{ee})_r(1 - e^{-\gamma T})$. We find (ρ^s_{ee}) using nonperturbative propagators Eqs. (5) and (6) and arrive at the fractional momentum kick per pulse,

$$\frac{-\Delta\mathbf{p}_s}{p_r} = \frac{\sin^2(\theta/2)\sinh(\gamma T/2)}{\cosh(\gamma T/2) - \cos^2(\theta/2)\cos\eta}\hat{\mathbf{k}}_c. \tag{8}$$

Here the Doppler-shifted phase η is

$$\eta = (\delta + \mathbf{k}_c\cdot\mathbf{v})T - \phi. \tag{9}$$

Finally, the radiative force is $F_{\text{train}} = \Delta p_s/T$.

As a function of phase η (or frequency or velocity), the force spikes at the positions of the Doppler-shifted frequency comb teeth, $\eta_n = 2\pi n$, with n being integer numbers (see Fig. 3). In Fig. 3 we also investigate the dependence on the values of the parameter γT. Let us focus on one of the teeth (e.g., $\eta = 0$). As γT is increased the momentum kick grows, reaches a maximum, and then declines; apparently for a given θ there is an optimal value of γT. By analyzing Eq. (8), we find this optimal value to be $(\gamma T)^F_{\text{opt}} = 2\cosh^{-1}[1/\cos^2(\theta/2)]$. For example, for $\theta = \pi/10$, the optimal value is $(\gamma T)^F_{\text{opt}} \approx 0.447$; i.e., the radiative lifetime is roughly twice the repetition period.

Equation (8) is nonperturbative. It remains valid even for strong laser pulses, as long as the pulses do not overlap. For

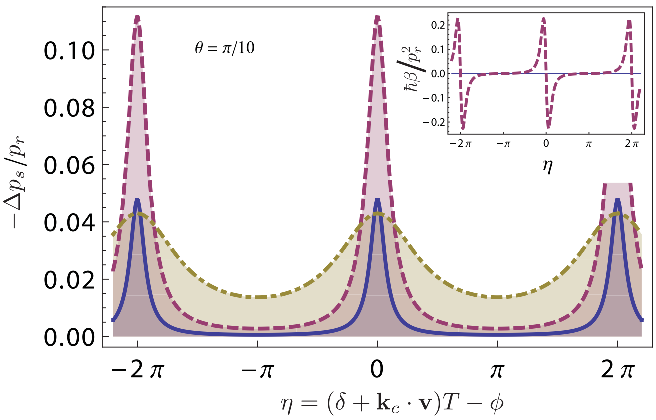

FIG. 3. (Color online) Frequency dependence of the fractional momentum for a pulse area $\theta = \pi/10$. The three curves differ by the values of the parameter γT (solid curve, $\gamma T = 0.1$; dashed, $\gamma T = 0.447$; and dot-dashed, $\gamma T = 2.5$). The spikes reflect the underlying frequency-comb teeth structure of the pulse train in the frequency domain. Inset: friction coefficient β as a function of phase for the dashed curve of the main graph.

$\theta = 2\pi$ the force vanishes since the 2π pulse does not redistribute the population. For π pulses, the right-hand side of Eq. (8) reduces to a frequency-*independent* value, $\tanh(\gamma T/2)$.

In the limiting case of weak pulses ($\theta \ll 1$) and fast repetition rates ($\gamma T \ll 1$), Eq. (8) reduces to

$$F_{\mathrm{train}}|_{\gamma T \ll 1, \theta \ll 1} \approx p_r \Omega_p^2 \left(\frac{\sqrt{\pi}\tau_p}{T} \right)^2 \times \sum_n \frac{\gamma/2}{(\gamma/2)^2 + [\delta + \mathbf{k}_c \cdot \mathbf{v}_0 - (\phi + 2\pi n)/T]^2}. \quad (10)$$

By comparing Eq. (10) with relevant cw expressions (see, e.g., Ref. [3]), we arrive at the qualitative picture where each tooth acts as an independent cw laser, the intensity of which has been reduced by a factor of $\sqrt{\pi}\tau_p/T$. This factor is roughly equal to the number of teeth fitting inside the overall frequency envelope of the FC.

Does a single tooth have enough power for cooling? The relevant parameter is the saturation intensity, I_s. Its typical value (sodium atom) is 6.4 mW/cm^2. Typical FC parameters ($\tau_p = 100$ fs, $T = 1$ ns, and average power 1 W) translate into the power per tooth of 0.1 mW; thereby I_s can be attained by focusing the laser output to a spot of 1.4 mm diameter. Notice that a fiber-laser-based FC with 10 W average power has been demonstrated [9] and the authors argue that the demonstrated technology is scalable to 10 kW average power. With this new generation of combs the cross section of the interaction region may be increased dramatically.

IV. COOLING DYNAMICS AND VELOCITY COMB

Now we turn to the dynamics of slowing down and cooling an entire atomic ensemble, characterized by some velocity distribution $f(v,t)$ (time dependence is caused by radiative force). To be specific, consider a typical use of radiative force for slowing down an atomic beam. The laser pulses would impinge on the atoms (see Fig. 1), countering their motion. The radiative force (8) depends on the atomic velocity via Doppler shift. As velocity is varied across the ensemble, the maxima of the force occur at discrete values of velocities (n are integers)

$$v_n = (2\pi\, n - T\,\delta + \phi)/(k_c T). \quad (11)$$

The force peaks are separated by $v_{n+1} - v_n = \lambda_c/T$ in the velocity space. The comb may have multiple teeth effectively interacting with the ensemble.

Cooling can be characterized by introducing the friction coefficient β, where $F_{\mathrm{train}}(v + \Delta v) \approx F_{\mathrm{train}}(v) - \beta(v)\Delta v$. If $\beta > 0$, there is a compression of velocity distribution around v. In the limiting case of $\gamma T \gg 1$ or $\theta = \pi$ the force does not depend on velocity; thereby $\beta = 0$ and, while the ensemble slows down, there is no cooling. The friction coefficient may be derived analytically from the force (8). We plot the dependence of β on η in Fig. 3. It acquires the maximum value at $\eta = \bar{\eta}$,

$$\cos \bar{\eta} = \tfrac{1}{2} \sec^2(\theta/2) \times \left(\sqrt{8\cos^4(\theta/2) + \cosh^2(\gamma T/2)} - \cosh(\gamma T/2)\right). \quad (12)$$

In the cw limit, this expression leads to detuning of $\gamma/2$ below the atomic resonance as expected. One could optimize β by varying θ or γT.

As the atoms slow down, they come in and out of resonance with the FC teeth, leading to periodic variation in the sign of β; no cooling results due to this variation. By analyzing Eq. (11) we see that this problem may be solved [1–3] by modulating the atomic resonance frequency (e.g., by spatially varying B fields as in the Zeeman slowers) or by employing chirped pulses (which would modulate the carrier frequency). Both approaches affect the detuning $\delta = \omega_0 - \omega_c$. The velocities at the resonance, Eq. (11), depend also on the carrier-envelope phase offset ϕ. Based on this observation, here we alternatively propose to vary ϕ by a laser field control element (see Fig. 1). As the atoms slow down, one would decrease the phase, keeping the Doppler-shifted atomic frequency in resonance with one of the FC teeth.

If for a given velocity group initially centered at $v_{mp}(t = 0)$, the phase detuning is kept at $\bar{\eta}$, and there is a compression of the velocity distribution around $v_{mp}(t)$. We may satisfy this requirement by tuning the phase according to $\phi(t) = [\delta + k_c v_{mp}(t)]T - \bar{\eta}$. As $v_{mp}(t)$ becomes smaller due to radiative force, the offset phase needs to be reduced. Using Eq. (8) we find the required pulse-to-pulse decrement of the phase

$$\Delta\phi_T = \frac{p_r^2 T}{\hbar M_a} \frac{\sin^2(\theta/2)\sinh(\gamma T/2)}{\cos\bar{\eta}\cos^2(\theta/2) - \cosh(\gamma T/2)}. \quad (13)$$

When the phase offset is driven according to Eq. (13), the entire frequency-comb structure shifts toward lower frequencies. As the teeth sweep through the velocity space, atomic $v(t)$ trajectories are "snow-plowed" by teeth, ultimately leading to narrow velocity spikes collected on the teeth. Formally, we may separate initial velocities into groups $v_{mp}(t = 0) + (\bar{\eta} + 2\pi n)/k_c T < v(t = 0) < v_{mp}(t = 0) + [\bar{\eta} + 2\pi(n + 1)]/k_c T, n = 0, \pm 1, \ldots$. The width of each velocity group is equal to the distance between neighboring

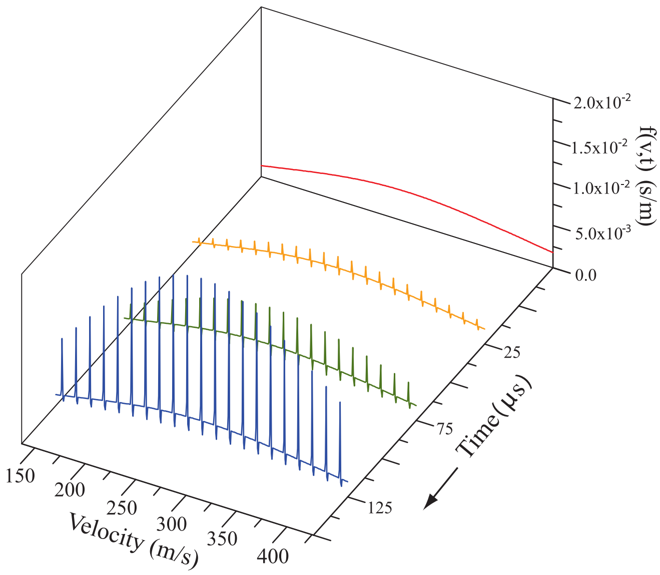

FIG. 4. (Color online) Time evolution of the velocity distribution for a thermal beam subjected to a coherent train of laser pulses. The pulse-to-pulse phase offset of the train is varied linearly in time as prescribed by Eq. (13). Atomic and pulse train parameters are $\gamma T = 0.0026, \theta = 0.0019$. The optimal phase detuning is $\bar{\eta} = 0.001$. The characteristic initial temperature of the ensemble is 293 K.

teeth in velocity space, λ_c/T. As a result of snow-plowing, the nth group will be piled up at $v_n(t) = v_{mp}(t) + n\lambda_c/T$. The final velocity spread of individual velocity groups will be limited by the Doppler temperature, $T_D = \hbar\gamma/2k_B$.

The formation of a velocity comb is illustrated in Fig. 4, where we consider cooling and slowing a one-dimensional thermal beam of ^{88}Sr by a pulse train. The initial velocity distribution is characterized by $f(v,t=0) = \frac{9}{2}\frac{v^3}{v_{mp}^4}\exp(-\frac{3v^2}{2v_{mp}^2})$, where v_{mp} is the most probable velocity at $t = 0$. In this example we use the weak $5s^2\,{}^1S_0 \rightarrow 5s5p\,{}^3P_1$ transition with $\gamma = 5.3 \times 10^4$ 1/s. Parameters of the train are $T = 50$ ns and $\theta = 0.0019$. The simulation was carried out by solving the OBEs numerically using the standard Runge-Kutta techniques for finite-duration pulses and also using analytical propagators for δ-function-like pulses; results from both techniques were in close agreement. During the time evolution, atoms followed classical trajectories and photon recoil effects were neglected. The phase offset was driven according to Eq. (13). At the end of the process we ended up with a velocity comb separated by 13.8 m/s and of Doppler-limited width of 7.6 mm/s (this is comparable to the recoil limit). About 14% of the total number of atoms are snow-plowed into the teeth within 125 μs. Notice that by shining two counterpropagating pulse trains on the atoms, one could control the positions of the velocity teeth at will, as shifting the phase of one train with respect to the other would change the balance of two counteracting radiative forces exerted by the trains.

V. SUMMARY

We demonstrated that radiative force exerted by laser pulse trains has unique features and expands the toolbox of laser cooling techniques. For example, one may engineer velocity combs that may be used for studies of narrow collision resonances and thresholds [11,15]. In some cases, the frequency comb may be already a part of experimental setup, e.g., in optical atomic clocks [16]. Using it for cooling would reduce the number of lasers. Also the setup does not require Zeeman slowers, whose fields may be detrimental for precision measurements [17].

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