Multiphoton adiabatic passage for atom optics applications

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We study the force exerted on two-level atoms by short, counterpropagating laser pulses. When the counterpropagating pulses overlap each other partially, multiphoton adiabatic processes are possible in several configurations, which amplify the force exerted on the atoms. We investigate the practical usefulness of such multiphoton adiabatic transitions for the manipulation of the atoms' mechanical state. In particular, we compare the efficiency of a pair of constant frequency, oppositely detuned laser pulses and that of a pair of frequency-chirped pulses. We also consider the case of prolonged exposure to a sequence of laser pulses for a duration that is comparable to or much larger than the spontaneous lifetime of the atoms. We use numerical methods to calculate the reduction of the force and the heating of the atomic ensemble when spontaneous emission cannot be neglected during the interaction. In addition, we derive simple approximate formulas for the force and the heating, and compare them to the numerical results. © 2009 Optical Society of America

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1. INTRODUCTION

The forces that resonant laser radiation can exert on atoms have been used extensively in past decades for cooling, trapping, and manipulation of the atoms' mechanical state [1]. The simplest type, the so-called radiation force (or scattering force) is mediated by absorption-spontaneous emission cycles and was the first to gain widespread use in atomic physics. Another force, mediated by a sequence of consecutive, counterpropagating laser pulse pairs has also been investigated in detail. A pair of such pulses (under appropriate conditions) can first excite the atoms, then return them to the ground state in a controlled way, delivering $2\hbar k$ momentum during the process. Since the repetition frequency can be much greater than the inverse spontaneous lifetime Γ, this force does not saturate as the radiation force does. One way to realize this is with the usage of π pulses [2–4]. Another way is to use frequency-chirped laser pulses that invert the atomic state via adiabatic population transfer, also called adiabatic passage (AP) or sometimes adiabatic rapid passage (ARP). This procedure has been first proposed for isotope separation [5], and later suggested for the coherent acceleration of atoms in atom optical applications, as well as the amplification of the cooling effect for atoms or molecules with narrow line transitions [6–8]. The downside of the AP force (and the analogous π-pulse force) is that the excitation and deexcitation of the atoms must be achieved by the pulses propagating in the appropriate direction. Should spontaneous emission occur, the force will be reversed as the atom is accelerated in the wrong direction by the subsequent pulses. At first sight, the AP force is therefore useful only for times that are short compared to the spontaneous lifetime, for if a considerable fraction of the atoms is allowed to go through spontaneous transitions, the atomic ensemble is dispersed very quickly.

Experimental realizations of the AP force, however, show that present technology does not allow a great number of pulses to interact with the atoms within a single spontaneous lifetime. The typical atomic lifetime used in the experiments is of the order of 10–100 ns, but the use of picosecond pulses, whose length would allow numerous interaction cycles within the lifetime, presents bandwidth problems. The full bandwidth of the chirped pulses may easily span a range where other, unwanted, transitions can be excited. Thus, pulses of a few nanoseconds duration are usually used [9–12].

On the other hand, it has been shown that the force exerted on the atoms may continue to be large even if the detrimental effect of spontaneous emission is taken into account [13]. The requisite to this is that the timing between the forward and backward propagating pulses should be asymmetric—the time delay between the first and second pulses of a pulse pair should be considerably smaller than the time delay between subsequent pulse pairs. In this case, the average force on the atoms, as well as the heating due to random reversal of the force tend to a finite (nonzero) value. This bears a strong resemblance to the effect of bichromatic standing waves, where the force acting on the atoms can be attributed to a sequence of counterpropagating π pulses due to the beating of the light field. Here the proper choice of the beat phase can supply the asymmetry necessary for a prolonged duration of the force [14–17]. Using a sequence of separately generated pulses has the advantage that the asymmetry of the delay times can be much greater. Using frequency-chirped pulses (as opposed to π pulses) has the advantage

that Doppler broadening of the atomic ensemble or the transverse intensity distribution of the laser pulses are not much of an issue.

Recently it has also been shown that when two counterpropagating pulses overlap each other, multiphoton adiabatic processes are possible [18,19]. These transitions can be generated either with a pair of pulses that are detuned from the atomic resonance in opposite directions, or with a pair of frequency-chirped laser pulses, and they can be understood in terms of adiabatic Floquet theory [20–23]. The overall effect of such an interaction is very similar to that of two counterpropagating separated laser pulses in both cases. The atoms are returned to the ground state at the end of the cycle and the momentum of the whole ensemble is changed uniformly, i.e., all atoms are accelerated by an equal amount. The only difference is that the momentum the atoms gain is $2N\hbar k$ where $N=1$ is the usual separated pulse scheme, but depending on pulse amplitude, pulse delay, and the magnitude of the detuning or the chirp speed, higher values of the integer N are possible. This means that the force exerted on the atoms can be amplified several times over the usual counterpropagating-pulse scheme. Nevertheless, the statement in the previous paragraphs still remains true: it is difficult to realize experimentally a pulse sequence where a large number of pulse pairs interact with the atoms in a time shorter than the spontaneous lifetime of the atoms.

Multiphoton adiabatic passage (MAP) was also shown to be useful in numerous other applications: for coherent population transfer between Rydberg states of an atom [24,25], between vibrational states of diatomic molecules [26], or the population transfer between the metastable states of a Λ or tripod atom using a single chirped pulse [27,28].

In this paper, we examine the force exerted on the atoms by a sequence of counterpropagating, partially overlapping laser pulses. We consider both a pair of constant frequency, oppositely detuned laser pulses and a pair of frequency-chirped pulses. We focus on the regime when the overlapping laser pulses generate MAP. We show that much smaller pulse intensity is necessary to generate such transitions using chirped pulses. In addition, we examine the force when the laser pulses interact with the atoms for a time that is comparable to or much larger than the spontaneous lifetime. In particular, we consider the heating suffered by the atomic ensemble as a consequence of spontaneous emission induced force reversal. A simple model is also developed that can be used to estimate the long-term effect of the pulse sequence on the atoms.

2. AMPLIFICATION OF THE FORCE DUE TO MULTIPHOTON ADIABATIC PASSAGE

A. Basic Theory

To describe the interaction of a two-level atom and counterpropagating resonant laser fields, we use the Hamiltonian

$$\hat{H} = \frac{\hat{p}^2}{2m} + \hat{H}_a - \hat{\mathbf{d}}\mathbf{E}, \tag{1}$$

where the first term is the kinetic energy due to center of mass motion, the second term is the internal atomic energy $\hat{H}_a=\hbar\omega_g|g\rangle\langle g|+\hbar\omega_e|e\rangle\langle e|$ and the last one describes a dipole interaction with a classical light field. Without any loss of generality, we can write the classical electric field in Eq. (1) as

$$E(x,t) = E^+(t)\cos[kx - \omega_{ge}t - \phi^+(t)] + E^-(t)\cos[-kx - \omega_{ge}t - \phi^-(t)], \tag{2}$$

where $E^{\pm}(t)$ are the slowly varying amplitudes of the two counterpropagating components, $\phi^{\pm}(t)$ are the phases that may contain a detuning or frequency chirp, and $\omega_{ge}=\omega_e-\omega_g$ is the frequency of the atomic transition. The sign that indexes the amplitude and the phase shows the direction of propagation. To calculate the coherent interaction between the laser pulses and the atoms, it is most convenient to use the relevant Schrödinger equation for the momentum-space probability amplitudes $\alpha(p,t),\beta(p,t)$, which constitute the two components of a spinor wave function. The two probability amplitudes are related to the ket corresponding to the physical state of the system by

$$|\Psi\rangle = \int [\alpha(p',t)e^{-i\omega_g t}|g,p'\rangle + \beta(p',t)e^{-i\omega_e t}|e,p'\rangle]\mathrm{d}p', \tag{3}$$

where $|g,p'\rangle,|e,p'\rangle$ are the usual electrotranslational states, i.e., plane-wave momentum states with a well-defined electronic state. Inserting Eqs. (2) and (3), and the Hamiltonian (1) into the Schrödinger equation and projecting with $\langle g,p|,\langle e,p|$ we obtain equations of motion for the probability amplitudes $\alpha(p,t),\beta(p,t)$. In doing this, one must not forget that the spatial variable x in Eq. (2) must be considered an operator for this purpose, and the decomposition of the cosine into exponentials will yield the translational operators in momentum space necessary for momentum conservation. With the rotating wave approximation and defining the Rabi frequencies $\hbar\Omega^{\pm}(t)=d_{ge}E^{\pm}(t)$ the equations will read [19]

$$i\hbar\partial_t\alpha(p,t) = \frac{p^2}{2m}\alpha(p,t) - \frac{\hbar\Omega^+(t)}{2}e^{i\phi^+(t)}\beta(p+\hbar k,t) - \frac{\hbar\Omega^-(t)}{2}e^{i\phi^-(t)}\beta(p-\hbar k,t),$$

$$i\hbar\partial_t\beta(p,t) = \frac{p^2}{2m}\beta(p,t) - \frac{\hbar\Omega^+(t)}{2}e^{-i\phi^+(t)}\alpha(p-\hbar k,t) - \frac{\hbar\Omega^-(t)}{2}e^{-i\phi^-(t)}\alpha(p+\hbar k,t). \tag{4}$$

Here $d_{ge}=\langle g|\hat{d}|e\rangle$ is the dipole matrix element and $\Omega^{\pm}$ are taken to be real (again, without loss of generality). Equations (4) can be solved using a computer quite conveniently, and yield a valid description of the interaction for short times when spontaneous emission can be neglected

altogether. For prolonged interaction, however, we must use the relevant Liouville equation for the density operator, which can be augmented by phenomenological decay terms to describe relaxation [22],

$$i\hbar\partial_t\hat{\rho} = [\hat{H},\hat{\rho}] - i\hbar\hat{\Gamma}. \quad (5)$$

The operator $\hat{\Gamma}$ is taken to describe spontaneous emission processes from the excited to the ground state, but no other transitions (to different atomic levels) or phase interrupting processes (such as collisions) are considered. For one-dimensional motion the equations for the density matrix elements are still tractable, though considerably more time consuming to solve than Eqs. (4).

In this paper we will consider Gaussian pulses and two different cases for the phase evolution. First, we consider pulses with a constant detuning, which are tuned symmetrically above and below the resonance frequency—the so-called $\delta,-\delta$ configuration. Second, we consider pulses with a linear chirp, whose central frequency is exactly resonant with the atomic transition—the so-called β,β configuration. The forward and backward propagating pulses are assumed to be identical in this case. In both cases, the second pulse is assumed to arrive with a delay T_1, which is the same order of magnitude as the pulse length τ (see Fig. 1). Thus,

$$\Omega^+(t) = \Omega_0\sqrt{\frac{\pi}{2}}\exp\left[-\frac{t^2}{2\tau^2}\right], \quad \Omega^-(t) = \Omega^+(t-T_1), \quad (6)$$

$$\phi^+(t) = \delta t + \phi_0^+, \quad \phi^-(t) = -\delta(t-T_1) + \phi_0^-, \quad (7)$$

for the $\delta,-\delta$ configuration or

$$\phi^+(t) = \frac{\beta}{2}t^2 + \phi_0^+, \quad \phi^-(t) = \frac{\beta}{2}(t-T_1)^2 + \phi_0^-, \quad (8)$$

for the β,β case. Here Ω_0 is a constant amplitude parameter such that the area of a pulse (the time integral of the envelope function) is just $\Omega_0\tau\times\pi$, δ is a constant detuning, and β is the linear chirp speed. Note that the parameter τ called the pulse length in this paper is related to the usual pulse width in intensity FWHM by $\tau_{\text{intensity}} = \tau 2\sqrt{\ln 2} \approx 1.67\tau$.

B. Multiphoton Adiabatic Passage

The usual scenario for using π pulses or AP for the manipulation of the atoms' mechanical state is when the counterpropagating pulses arrive in succession, interacting with the atoms separately. Provided that the pulses and the delay are short enough for spontaneous emission to be neglected ($T_1,\tau \ll 1/\Gamma$), the effect of a single pair of pulses on the atomic momentum-space distribution functions $a(p,t)=\rho_{gg}(p,p,t)=\alpha(p,t)\alpha^*(p,t)$ and $b(p,t)=\rho_{ee}(p,p,t)=\beta(p,t)\beta^*(p,t)$ will simply become

$$a(p,t') = a(p-2\hbar k,t),$$

$$b(p,t') = b(p+2\hbar k,t). \quad (9)$$

The traditional AP force is mediated by a repetition of this basic cycle many times in rapid succession.

A similar process is also possible when the two counterpropagating pulses do not interact with the atoms quite separately, but overlap each other partially—the second one arrives with a distinct delay, the same order of magnitude as the pulse length $T_1 \sim \tau$. Multiphoton adiabatic processes are possible either when the two pulses are detuned symmetrically to either side of the transition [18] or when both pulses have a linear chirp [19]. When the pulse amplitudes, the delay, and the detunings or the chirp have appropriate values, the overall effect of the laser pulse pair on the atomic distributions can be similar to that of successive laser pulses in the AP case. The electronic state of the atoms is conserved and so are the shapes of the ground and excited state momentum-space distributions. However, the momentum transfer is increased:

$$a(p,t') = a(p-2N\hbar k,t),$$

$$b(p,t') = b(p+2N\hbar k,t), \quad (10)$$

where N is a small integer and $N=1$ is the traditional AP case. Figure 2 shows the average momentum transferred to the atom $\Delta\bar{p}=\bar{p}(t')-\bar{p}(t)$ (where $\bar{p}=\int p[a(p)+b(p)]dp$) by a pair of overlapping laser pulses as a function of the pulse amplitude $\Omega_0\tau$ (solid black curves). The figure also shows the population of the excited state after the interaction (multiplied by five to be visible). The data have been obtained by numerically integrating Eqs. (4) using parameters (a) $\delta=100/\tau$, $T_1=2\tau$ and (b) $\beta/\tau^2=20$, $T_1=1.5\tau$ (parameters for which the adiabatic condition is well fulfilled) and assuming that the atoms are in the ground state just before the interaction. The momentum transfer displays distinct plateaus of $2N\hbar k$ momentum transfer, where MAP is realized and the atoms are returned to the ground state at the end of the interaction. It is notable that the required pulse amplitude is much smaller in the β,β configuration (i.e., the chirped-pulse scheme)—the $\Delta\bar{p}=6\hbar k$ plateau occurs at around $\Omega_0\tau=600$ for the $\delta,-\delta$ case, and at $\Omega_0\tau=60$ for the β,β case, which means that in pulse intensity the difference is 2 orders of magnitude. It is also interesting to note that the $\delta,-\delta$ configuration yields a counterintuitive direction for the momentum transfer (i.e., the atoms are pushed opposite to the direction of propagation of the first pulse), and this is irrespective of whether δ is positive or negative. [This is why $-\Delta\bar{p}$ has been plotted in Fig. 2(a)]. In the β,β scheme, on the other hand, atoms are pushed in the direction of propagation of the first pulse, again, irrespec-

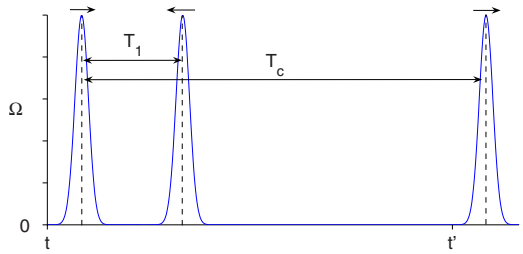

Fig. 1. (Color online) Timing of the laser pulse pairs. The second pulse of the pair, which counterpropagates the first one arrives with a delay T_1, which is the same order of magnitude as the pulse length τ. The cycle then repeats itself after $T_2=T_c-T_1 \gg T_1$. (Arrows above the pulses symbolize the propagation direction.)

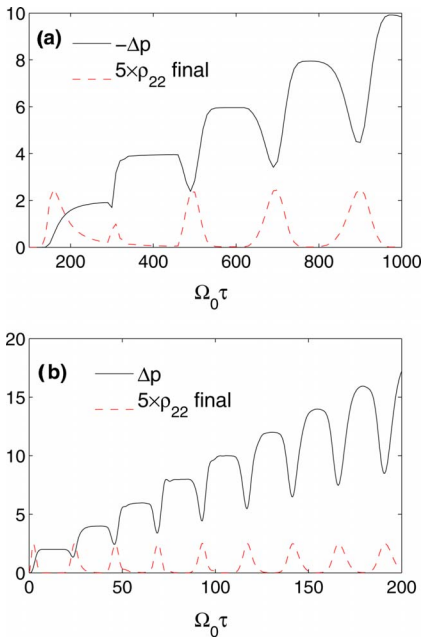

Fig. 2. (Color online) Solid black curves: average momentum obtained by atoms in the ground state $\Delta\bar{p}$ during the interaction with a pair of counterpropagating pulses as a function of the normalized pulse amplitude $\Omega_0\tau$. (a) δ, $-\delta$ configuration, with $\delta=100/\tau$ and $T_1=2\tau$. (b) β, β configuration with $\beta=20/\tau^2$ and $T_1=1.5\tau$. Dashed curves: population of the excited state after the interaction multiplied by 5 to be visible. (Note that $-\Delta\bar{p}$ has been plotted for the δ, $-\delta$ case; see the text for an explanation.)

tive of whether β is positive or negative. Thus creating pulse overlap in the usual AP force scheme can amplify the force acting on the atoms several times. Since the population transfer is adiabatic, the process is robust with respect to a slight change of the amplitude.

While the momentum transfer has been plotted here as a function of pulse amplitude, it is clear that, as in any adiabatic transition, these too are robust with respect to a slight change of all other parameters. This means that changing parameters like the time delay T_1, or the detuning or the chirp β one may observe similar curves with plateaus of various orders N. In particular, at a given amplitude and chirp, decreasing the delay (i.e., increasing the overlap) will yield higher order transitions—up to the point where the conditions of AP are violated.

C. Elements of a Floquet Analysis

Multiphoton adiabatic transitions can be analyzed conveniently with the use of adiabatic Floquet theory [20–23]. To present a concise summary of the elements of this theory that are relevant to the current problem, we start from a simplified form of Eqs. (4) that does not take into account the atoms' translational degree of freedom (this approximation will be justified later),

$$i\hbar\partial_t\alpha(t) = -\frac{\beta(t)}{2}[\hbar\Omega^+(t)e^{i\phi^+(t)} + \hbar\Omega^-(t)e^{i\phi^-(t)}],$$

$$i\hbar\partial_t\beta(t) = -\frac{\alpha(t)}{2}[\hbar\Omega^+(t)e^{-i\phi^+(t)} + \hbar\Omega^-(t)e^{-i\phi^-(t)}]. \quad (11)$$

The 2×2 Hamiltonian matrix of these equations depends on time through the amplitudes Ω^+,Ω^-, and the phases ϕ^+,ϕ^-. Since the phases appear in the exponent the time dependence of the matrix is not slow; the usual method of diagonalization to study adiabatic passage cannot be used. Now we assume that the frequency difference between the two fields is a constant, i.e., $\phi^+(t)-\phi^-(t)=\Delta t$, introduce the notation δ^+ for the instantaneous detuning of the Ω^+ wave from the resonance and writing $\beta'(t)=\beta(t)\exp(i\phi^+(t))$, we perform a Fourier expansion of the probability amplitudes $\alpha(t),\beta'(t)$,

$$\alpha(t) = \sum_m A_m(t)\exp(-im\Delta t),$$

$$\beta'(t) = \sum_m B_m(t)\exp(-im\Delta t). \quad (12)$$

From Eq. (11) we can then derive the equation of motion for the vector composed of the Fourier amplitudes $\Psi=(\ldots,B_{m-1},A_{m-1},B_m,A_m,B_{m+1},A_{m+1},\ldots)$ to be

$$i\partial_t\Psi = \mathbf{H}_{\mathrm{Floq}}\Psi, \quad (13)$$

where the matrix $\mathbf{H}_{\mathrm{Floq}}$ is an infinite tridiagonal matrix of the form

$$-\mathbf{H}_{\mathrm{Floq}} = \begin{pmatrix}
\ddots & \ddots & & & & & & & \\
\ddots & (m-1)\Delta+\delta^+ & \Omega^+/2 & & & & & & \\
 & \Omega^+/2 & (m-1)\Delta & \Omega^-/2 & & & & & \\
 & & \Omega^-/2 & m\Delta+\delta^+ & \Omega^+/2 & & & & \\
 & & & \Omega^+/2 & m\Delta & \Omega^-/2 & & & \\
 & & & & \Omega^-/2 & (m+1)\Delta+\delta^+ & \Omega^+/2 & & \\
 & & & & & \Omega^+/2 & (m+1)\Delta & \ddots & \\
 & & & & & & \ddots & \ddots &
\end{pmatrix}. \quad (14)$$

This form of the equations lends itself to the classical theory of the adiabatic following much better, as the fast time exponents have been removed, the matrix $\mathbf{H}_{\mathrm{Floq}}$ depends on time only through the slowly varying pulse amplitudes Ω^+,Ω^- and the detuning δ^+. Note that while we have assumed Δ to be time independent for convenience,

this is not essential; one can use this method with a slight twist even when Δ depends slowly on time. The coefficients A_m and B_m are merely Fourier components of two probability amplitudes in this formalism, but they can be shown to have a deeper meaning. The correspondence $A_m \leftrightarrow |m,g\rangle$ can be made, where the Floquet state $|m,g\rangle$ denotes a combined state of field and atom, with the latter being in the ground state and the former having m photons removed from the Ω^+ component and emitted into the Ω^- component, i.e., m photons exchanged between the two components compared to the initial state [20,23]. Likewise, the correspondence $B_m \leftrightarrow |m,e\rangle$ stands, where $|m,e\rangle$ denotes a Floquet state with the atom in the excited state, m photons removed from the Ω^+ component and $m-1$ emitted into the Ω^- component. Though we neglected the kinetic energy of the atom when writing Eqs. (11), we can clearly infer the momentum change of the atom during the process by noting the number of photons that have been exchanged—a $|0,g\rangle \rightarrow |N,g\rangle$ transition implies that a momentum of $2N\hbar k$ has been transfered to the atom.

Computing the eigenvalues and eigenvectors of the Floquet matrix (14) (i.e., calculating the dressed states of the atom in the field of the two pulses) as a function of time with various parameters (amplitude, delay, and detuning or chirp), one can analyze easily the possible outcomes of the interaction. Figure 3(a) shows a plot of Floquet eigenvalues for a pair of symmetrically detuned pulses in the $\delta,-\delta$ scheme. The thick black curve marks the energy of the dressed state that is connected with the $|0,g\rangle$ Floquet state before the interaction, i.e., a ground state atom and the fields in their initial states. This dressed state then transforms into the $|-3,g\rangle$ Floquet state, which corresponds to three photons removed from the Ω^- field and emitted into the Ω^+ field. Clearly, the atomic momentum must change by $-6\hbar k$ during this process. So if the changing of the fields is slow enough for adiabatic following to take place (with respect to the energy difference between neighboring levels), the atom will go through a six-photon adiabatic process.

Further insight is provided by plotting the Floquet eigenvalue of the $|0,g\rangle$ state when only the first pulse is present (thick black dashed curve), and the Floquet eigenvalue of the $|-3,g\rangle$ state when only the second pulse is present (thick black dashed-dotted curve). A single field couples these with only one other Floquet state (Ω^+ couples $|0,g\rangle$ to $|0,e\rangle$ and Ω^- couples $|-3,g\rangle$ to $|-2,e\rangle$), and causes a strong Stark shift at the pulse peak. With only one pulse present, the atom is returned to its initial state without the possibility of MAP (broken curves). But when both pulses are present, the two states that are Stark shifted close to each other by the leading edge of the Ω^+ field and the trailing edge of the Ω^- field connect during the overlap reagion and a six-photon transition is realized. For the parameters that were used to generate the figure, $\Omega^{\pm}\tau=420$, $\delta^{\pm}\tau=\pm 70$ and $T_1=2\tau$ the conditions of adiabaticity are already fulfilled quite well. Figure 3(b) shows a similar plot for a pair of counterpropagating chirped pulses. This time, the adiabatic dressed state that connects $|0,g\rangle$ and $|3,g\rangle$ is shown with a thick black solid curve, while the two broken (dashed and dashed-dotted) thick black curves show the evolution of the initial and final states's Floquet eigenvalue with only the first and the second pulses, respectively. It is visible on the plot that the frequency sweep in itself provides a considerable shift of the dressed state eigenvalues. Thus to move the inital and final states close to each other, we do not have to rely solely on a Stark shift, so the pulse amplitude required may be much smaller in the chirped-pulse scheme. Indeed, the parameters used to generate this plot are $\Omega^{\pm}\tau=60$, $\beta^{\pm}\tau^2=20$, and $T_1=1.5\tau$.

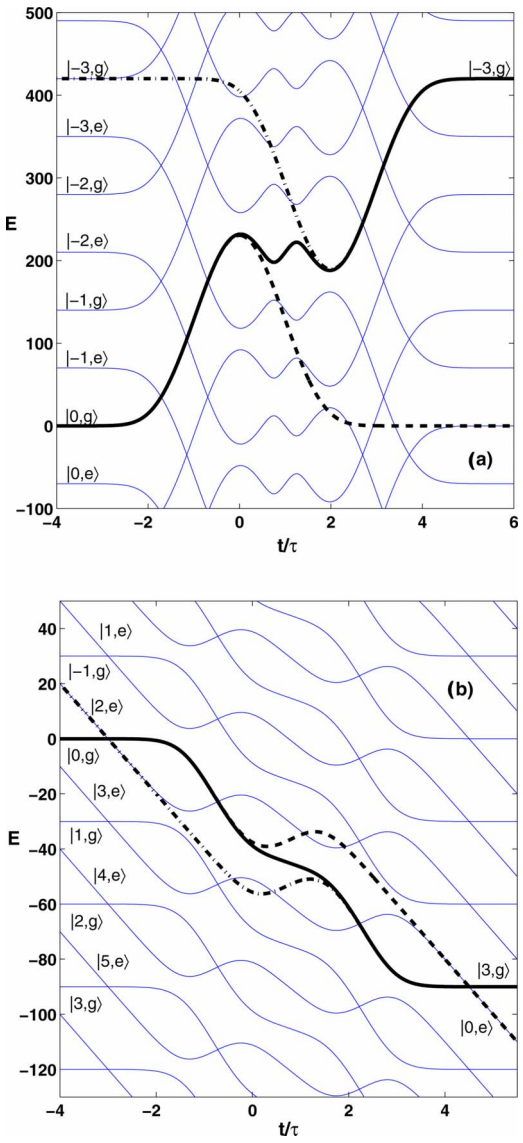

Fig. 3. (Color online) Floquet eigenvalues as a function of time during interaction with overlapping pulses. (a) δ, $-\delta$ configuration for $\Omega^{\pm}\tau=420$, $\delta^{\pm}\tau=\pm 70$ and $T_1=2\tau$ and (b) β, β configuration for $\Omega^{\pm}\tau=60$, $\beta^{\pm}\tau^2=20$ and $T_1=1.5\tau$. Some levels are labeled with the index of the corresponding Floquet state at the beginning and at the end of the interaction. In particular, the evolution of the Floquet state that is connected with the $|0,g\rangle$ state before the interaction is marked with a thick black curve.

Figure 3 helps us understand why neglecting the translational energy present in Eqs. (4) is justified. The relevant energy difference between neighboring states on the diagram is given by Δ. Provided we have pulses with $\tau\sim$ nanosecond duration (to beat spontaneous emission), the order of magnitude of $\Delta\sim$ gigahertz is much larger

than the slight correction due to the kinetic energy term <1 MHz. Clearly, the approach outlined in this subsection can be used only to obtain a qualitative understanding of the possible outcomes of the interaction. Whether the pulse parameters really do suffice to create MAP must be deduced from a numerical simulation of Eqs. (4), which take into full account of any nonadiabatic transitions as well.

3. SPONTANEOUS EMISSION DURING PROLONGED INTERACTION

As mentioned in the Introduction, present technology does not allow the AP force to be used in its pure form. While one can readily generate frequency-chirped laser pulses whose duration is much shorter than the spontaneous lifetime of the atoms, the interaction of a large number of pulses during a time short compared to the lifetime of the excited state is not possible. The main reason for this is that very short pulses require a great chirp speed and present bandwidth problems, opening the possibility of unwanted atomic transitions. Therefore, in practice, one has to be satisfied with a scenario where individual pulses are indeed much shorter than the excited state lifetime, but the whole interaction continues for much longer. This means that spontaneous emission cannot be left out of consideration. Even if only a small fraction of the atoms goes through such a process during a single cycle, the cumulative effect during many cycles can be considerable.

A. Numerical Calculations of the Density Matrix

As the most obvious step to include spontaneous emission in the description, we have solved Eqs. (5) using a computer. To facilitate the solution, the recoil due to spontaneous emission has been neglected. This is convenient because including a fractional change of the atomic momentum requires a much finer resolution in momentum space, with the corresponding increase of matrix elements. Figure 4 shows the result of a calculation in which the atoms interacted with 30 pairs of frequency-chirped laser pulses. The parameters of the simulation were chosen to correspond to the parameters of metastable He, which was used in several experiments to demonstrate the AP force [9,11], and the pulse parameters were similar to those used to produce Fig. 2(b), where several higher-order plateaus can be seen. Thus $\Gamma=1/97$ ns, $\tau=2$ ns (which clearly fulfils $\tau \ll 1/\Gamma$), $\Omega_0=100/\tau$, $\beta=20/\tau^2$. The delay was chosen to be $T_1=1.714\tau$, which at this amplitude corresponds to the $N=4$ plateau and the cycle length was chosen to be $T_c=80$ ns. Since $T_c \approx 1/\Gamma$, the whole process lasts much longer than the spontaneous lifetime, $T_{int}=2400$ ns in this case. Figure 4(a) displays the momentum-space distribution of the atoms before and after the interaction. Figure 4(b) shows the changing of the average momentum and that of the square of the momentum space width in each cycle, i.e., $\Delta\bar{p}$ and $\Delta\sigma_p^2 = \sigma_p^2(t') - \sigma_p^2(t)$, in recoil units. The most important property of the evolution is, that both $\Delta\bar{p}$ and $\Delta\sigma_p^2$ converge to stationary-state values. Therefore, in the long run there is a constant force acting on the atoms (which, however, is somewhat smaller than $8\hbar k/T_c$, the ideal case without spontaneous emission) and a diffusive spreading in momentum space—the width is proportional to $\sqrt{T_{int}}$. The behavior depicted is typical—the stationary-state force and diffusion constant depend a great deal on the parameters used (most importantly the T_1 and T_c) but the general behavior remains the same.

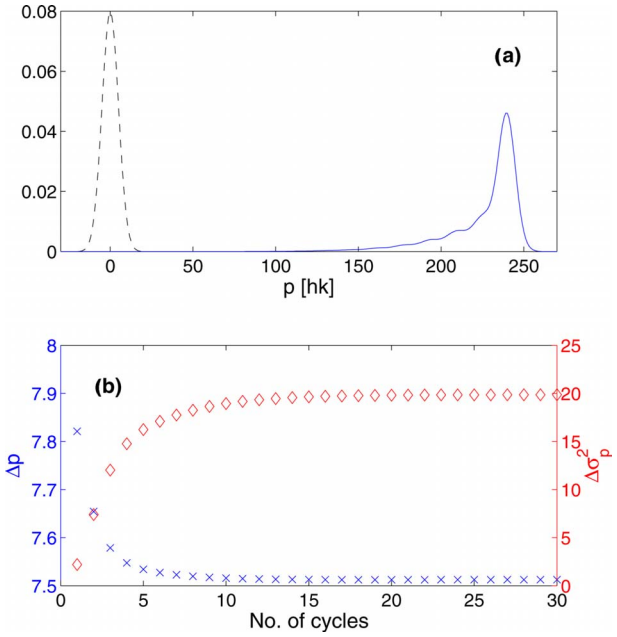

Fig. 4. (Color online) (a) Momentum-space distribution functions before (dashed curve) and after (solid curve) the interaction with 30 pairs of overlapping frequency-chirped laser pulses. The parameters used for the calculation are $\Gamma=1/97$ ns, $\tau=2$ ns, $\Omega_0=100/\tau$, $\beta=20/\tau^2$, $T_1=1.714\tau$ (which corresponds to the $N=4$ plateau), $T_c=80$ ns. (b) The value of $\Delta\bar{p}$ (crosses) and $\Delta\sigma_p^2$ (diamonds) in units of $\hbar k$ after each cycle.

B. Simple Model of the Amplified Adiabatic Passage Force

The behavior of the long-term effect of the overlapping laser pulses just seen is very similar to that of the usual AP force. For this latter case a very simple model can be used to calculate the detrimental effects of spontaneous emission during prolonged interaction [13]. In this subsection we generalize this simple model to the case of multiphoton adiabatic processes. Even though the assumptions used to derive the original model have limited validity in this case, the results turn out to be very useful.

To start, we need a set of equations that describe the evolution of the momentum-space distribution functions which also encompass the effect of spontaneous emission to replace Eqs. (10). Defining $q_1=1-\exp(-\Gamma T_1)$ to denote the probability of an atom emitting a photon spontaneously during T_1, and $q_2=1-\exp[-\Gamma(T_c-T_1)]$ to be the same for T_c-T_1 we now write,

$$a_{n+1}(p) = (1-q_1)a_n(p-2N\hbar k) + q_1q_2a_n(p) + q_2b_n(p + 2N\hbar k),$$

$$b_{n+1}(p) = (1-q_2)b_n(p+2N\hbar k) + q_1(1-q_2)a_n(p), \qquad (15)$$

as the basic iteration rule for the momentum-space distribution functions. In these formulas subscript n denotes

the distribution functions just before the $(n+1)$th cycle (at t, see Fig. 1) and subscript $n+1$ denotes distribution functions after the $(n+1)$th cycle, [just before the $(n+2)$th cycle at t']. The physical content of the single terms is clear: an atom in state $|g,p\rangle$ (ground state with momentum p) at the end of the cycle could have either (i) started from $|g,p-2N\hbar k\rangle$ and without emitting any photons spontaneously, received $2N\hbar k$ momentum; (ii) started from $|g,p\rangle$ and decayed twice obtaining no net momentum overall; or (iii) started from $|e,p+2N\hbar k\rangle$, received $-2N\hbar k$ momentum and decayed after the second pulse. In a similar manner, an atom in state $|e,p\rangle$ at the end of the cycle could have either (i) started from $|e,p+2N\hbar k\rangle$ and escaped any spontaneous transitions receiving $-2N\hbar k$ momentum, or (ii) started from $|g,p\rangle$ and decayed after the first pulse, but not after the second one, obtaining no net momentum overall.

Equations (15) can be used to calculate the the momentum distribution functions after each cycle. They are very convenient to use, because the evolution of the distribution functions can easily be studied for many cycles without actually solving Eqs. (4) and (5). They are basically the MAP counterpart of the original model of [13] with simply $2N\hbar k$ inserted for the momentum transfer. As such, they have several shortcomings: First of all, they have been written to separate the effects of the laser pulses and spontaneous transitions completely—however, with overlapping pulses such a separation can only be approximate, so our assumption has to be justified *a posteriori* by comparing the results derived from Eqs. (15) to a full simulation of Eqs. (5). Also (as with the original model) the recoil of spontaneously emitted photons has been neglected, so the only source of heating included in the model is that due to random force reversal at each spontaneous emission event. This approximation can also be justified *a posteriori* by observing that the latter is far greater than the heating that could be attributed to random single-photon recoils using the time averaged population of the excited state. Furthermore, these formulas can no longer be used for arbitrary delays—the delay has to correspond to one of the plateaus where there is full adiabatic population transfer and the atoms return to the ground state at the end of the interaction. The order N of the plateau that corresponds to the delay has to be inserted into the formulas. Finally, any nonadiabatic transitions are also neglected.

By integration and straightforward algebraic manipulation of Eqs. (15) we can also derive simple iteration rules for important quantities. The first of these are the level probabilities after n cycles defined as $P_{a,n}=\int a_n(p)\mathrm{d}p$ and $P_{b,n}=\int b_n(p)\mathrm{d}p$. An iteration rule for them can be obtained simply by integrating Eqs. (15) with respect to p to get

$$P_{i,n+1}=P_i^{st}+(P_{i,n}-P_i^{st})(1-q_c) \quad i\in\{a,b\}, \tag{16}$$

where $q_c=1-\exp(-\Gamma T_c)$, i.e., for prolonged interaction, the excited and ground-state probabilities converge to stationary values,

$$P_a^{st}=\frac{q_2}{q_c}, \quad P_b^{st}=\frac{q_1(1-q_2)}{q_c}. \tag{17}$$

By multiplying both of Eqs. (15) by p, adding the two, and again integrating with respect to p we get an iteration rule for the average momentum,

$$\Delta\bar{p}=\bar{p}_{n+1}-\bar{p}_n=2N\hbar k[(1-q_1)P_{a,n}-P_{b,n}]. \tag{18}$$

As the average momentum gain depends only on the level probabilities, for prolonged interaction $\Delta\bar{p}$ also converges to a stationary value

$$\Delta\bar{p}^{st}=2N\hbar k\frac{q_2-q_1}{q_c}. \tag{19}$$

Finally, we can also obtain an iteration rule for the momentum space width of the distribution σ_p^2 if we multiply Eqs. (15) by p^2, add them, integrate with respect to p, and use Eq. (18). With some straightforward algebraic manipulation it is not difficult to show that the asymptotic form for the iteration rule has the form

$$\Delta\sigma_p^2=\sigma_{p,n+1}^2-\sigma_{p,n}^2=K^{st}, \tag{20}$$

where K^{st} is a constant and can be calculated to be

$$K^{st}=8N^2\hbar^2k^2\frac{q_1q_2(1-q_1)(1-q_2)(2-q_1)(2-q_2)}{q_c^3}$$
$$+4N^2\hbar^2k^2\frac{q_1q_2(4-q_1-q_2-2q_c)}{q_c^2}. \tag{21}$$

So the model predicts the long-term effect of the counterpropagating, overlapping laser pulses that induce MAP to be a constant force acting on the atoms and a constant heating of the ensemble. This is in accordance with the findings of Subsection 3.A. The results are identical to the formulas derived in [13] for the usual AP force case except for the factor N and N^2 in the momentum gain and momentum diffusion formulas.

C. Discussion

The first question that arises is obviously the applicability of the model for the quantitative estimation of the MAP force. The basic assumption that could be used for separated laser pulses, that there is a full, "instantaneous" inversion of the atomic populations, is not valid for multiphoton adiabatic passage. Plotting the population histories for various delay values corresponding to different MAP plateaus shows that the population of the excited state never reaches unity during the interaction. On the other hand, the time interval, during which there is a considerable population in the excited state, can be somewhat longer than T_1 assumed by the simple model. Overall, the effect of the two laser pulses and spontaneous emission cannot be separated in time as for nonoverlapping pulses, calculating the probability of spontaneous emission as before is rather an order of magnitude estimate. Moreover, nonadiabatic transitions that can have considerable cumulative effect are neglected by the model altogether.

To evaluate the usefulness of the simple formulas for the steady-state MAP force Eqs. (19) and (21), we have

calculated the evolution of the atomic momentum-space distribution for numerous different values of T_c, T_1, and τ using Eqs. (5). From the solutions, we have extracted the relevant constants $\Delta\bar{p}^{st}$ and $\Delta\sigma_p^2$ and compared them with the predictions of the simple model. Figure 5 shows an example of the comparison for a pulse length of $\tau=2$ ns and two different cycle lengths $T_c=80$ and 40 ns (other parameters are the same as those used to produce Fig. 4). The figures show the values of $\Delta\bar{p}^{st}$ and $\Delta\sigma_p^2$ as a function of the plateau order N calculated from the simulation (squares) and the simple model (stars). Note that the delay T_1 is different for each N. The figures show that the value of the momentum transfer per cycle is predicted very well by the model—the exact result is just slightly above the value obtained from the model. The increase of the momentum distribution width shows a much larger discrepancy between model and simulation. However, one can see from the figures that the estimate of the model is conservative in all cases, i.e., the force is (slightly) underestimated, while the heating is overestimated. Therefore the model can be very useful for estimating the force and the heating in a given situation.

Equation (19) readily shows us that in case of prolonged interaction, the momentum transferred per cycle is less than the ideal $2N\hbar k$ value. In case of a symmetric time sequence of the pulses $T_1=T_c/2$ the steady-state force is zero [$q_1=q_2$ in Eq. (19)] and as the asymmetry (the ratio T_c/T_1) increases it gets closer and closer to the ideal value. The reason for this is intuitively clear: the decrease of the force is due to atoms that emit spontaneous photons and thus "get out of phase" with the rest of the ensemble, being accelerated in the opposite direction. The shorter T_1 is compared to T_c-T_1 the bigger the chance that an atom already "out of phase" with the majority will return via another spontaneous transition before the start of the next cycle. The use of MAP on higher-order plateaus is thus not only advantageous because the momentum transfered per cycle is proportional to N, but because $\Delta\bar{p}$ also increases as T_1 is decreased. Indeed, the avoidance of pulse overlap was a fundamental limit in the previous experimental realizations of the usual ARP force. The asymmetry realized in the studies of [9,11] was about $T_c/T_1=4$, which yields an average force of about $\hbar k/T_c$ instead of the ideal $2\hbar k/T_c$.

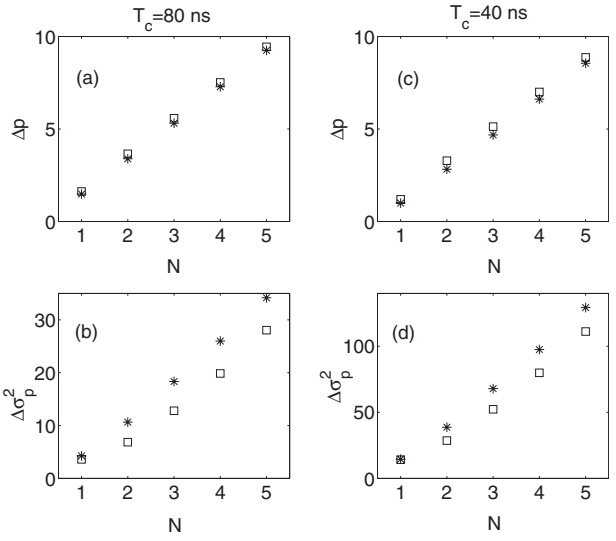

Fig. 5. (a) Comparison of the simulation results (squares) and the simple model (stars): (a) and (c) depict the steady state momentum transfer per cycle $\Delta\bar{p}^{st}$ for $T_c=80$ and 40 ns, respectively. (b) and (d) depict the heating per cycle $\Delta\sigma_p^2$ for $T_c=80$ and 40 ns, respectively. All quantities are plotted as a function of N, so the delay T_1 is different in each case. Other parameters are the same as for Fig. 4: $\Gamma=1/97$ ns, $\tau=2$ ns, $\Omega_0=100/\tau$, $\beta=20/\tau^2$.

The formula that describes the heating of the atomic ensemble Eq. (21) can also be investigated easily to show that the heating is largest in the case $T_1=T_c/2$ and decreases rapidly with increasing ratio of T_c/T_1. On the other hand, the stationary value of $\Delta\sigma_p^2$ is proportional to N^2, so using a higher-order plateau to increase the force has its price. It may be more favorable to take two $4\hbar k$ cycle steps instead of one $8\hbar k$ step if experimental possibilities and interaction times permit, because the overall heating will be smaller in the former case. Another disadvantage of higher-order peaks is that they are narrower in the space of parameters, so the interaction is less robust. This may have the consequence, for example that the Doppler shift the atoms acquire during acceleration will drive them out of the parameter range of the peak sooner.

From the behavior shown in Fig. 4 it can also be seen that if the atoms start from the ground state, the force converges to the stationary-state value from above, and the diffusion constant converges from below—which means that for practical purposes, the longer the transient behavior the better.

The simple formulas Eqs. (19) and (21) contain only the delay and cycle times T_1 and T_c as parameters. But the question of what values could be used for T_1 with any given physical pulse parameters τ, Ω_0 and β can only be calculated from numerical solutions of Eqs. (4). Thus the simple model can be thought of as a substitute for solving Eq. (5) for all parameters only in conjunction with Eqs. (4). Nevertheless, solving the former is much more time consuming than the latter—not only are the number of variables necessary to consider in the density-matrix description much larger, the equations also have to be solved for a time much longer than $1/\Gamma$ to find the steady-state evolution. Conversely, Eqs. (4) have to be solved only for one cycle with any given set of parameters to find out if they are suitable for realizing a full multiphoton adiabatic passage with some N, and then the simple model can be used to estimate the steady-state behavior for large interaction times. For the most promising parameter values, the full density-matrix simulation can also be performed to confirm the estimates, but the simple model makes it unnecessary to do the most time-consuming calculations for every set of parameters.

4. SUMMARY

In this paper, we have investigated the effect of short counterpropagating laser pulses on the mechanical state of two-level atoms. In particular, we have considered the situation when the laser pulses overlap each other partially and induce multiphoton adiabatic passage (MAP). We have investigated pairs of constant frequency, oppositely detuned pulses as well as pairs of frequency-chirped

pulses. MAP in these cases can be understood with the use of adiabatic Floquet theory, and this also shows why inducing MAP with chirped pulses requires much smaller intensities than with oppositely detuned pulses.

We have examined the behavior of the momentum-state distribution of the atoms when they interact with a sequence of pulse pairs for a prolonged time, such that spontaneous emission cannot be neglected. It was shown that the long-term effect of such a pulse sequence tends to be a constant force and a momentum-space diffusion (heating). The force acting on the atoms is identical in nature to the usual adiabatic passage force that is exerted by separated, counterpropagating chirped laser pulses, but its magnitude is amplified several times. There are two reasons for the amplification of the force: on the one hand, the atoms receive $2N\hbar k$ momentum from each pulse pair where N may be a small integer that is larger than one. On the other hand, allowing the two pulses of the cycle to overlap increases the asymmetry of timing between the two pulses of a cycle and the start of the next cycle, and this too increases the overall force acting on the atoms for long interaction times.

We have also derived a simple model that, in conjunction with the numerical solution of the momentum-space Schrödinger equation for a single pair of pulses, can be used to estimate the long-term effect of the sequence of laser pulses. The model correctly accounts for the dependence of the force on the timing parameters (the delay T_1 between pulses in the pulse pairs and the cycle time T_c). Using the model, one can avoid time consuming numerical solutions for the density-matrix equations for a large number of parameter sets.

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REFERENCES

1. H. Metcalf and P. van der Straten, *Laser Cooling and Trapping* (Springer, 1999).
2. B. Nölle, H. Nölle, J. Schmand, and H. J. Andrä, "Atomic-beam deflection by double-Pi-pulse laser technique," Europhys. Lett. **33**, 261–266 (1996).
3. T. G. M. Freegarde, J. Walz, and T. W. Hänsch, "Confinement and manipulation of atoms using short laser pulses," Opt. Commun. **117**, 262–267 (1995).
4. A. Goepfert, I. Bloch, D. Haubrich, F. Lison, R. Schütze, R. Wynands, and D. Meschede, "Stimulated focusing and deflection of an atomic beam using picosecond laser pulses," Phys. Rev. A **56**, R3354–R3357 (1997).
5. I. Nebenzahl and A. Szöke, "Deflection of atomic beams by resonance radiation using stimulated emission," Appl. Phys. Lett. **25**, 327–329 (1974).
6. J. S. Bakos, G. P. Djotyan, G. Demeter, and Zs. Sörlei, "Transient laser cooling of two-level quantum systems with narrow natural linewidths," Phys. Rev. A **53**, 2885–2888 (1996).
7. G. P. Djotyan, J. S. Bakos, G. Demeter, and Zs. Sörlei, "Manipulation of two-level quantum systems with narrow transition lines by short linearly polarized frequency-chirped laser pulses," J. Opt. Soc. Am. B **13**, 1697–1705 (1996).
8. T. Freegarde, G. Daniell, and D. Segal, "Coherent amplification in laser cooling and trapping," Phys. Rev. A **73**, 033409 (2006).
9. M. Cashen, O. Rivoire, L. Yatsenko, and H. Metcalf, "Coherent exchange of momentum between atoms and light," J. Opt. B: Quantum Semiclassical Opt. **4**, 75–79 (2002).
10. J. S. Bakos, G. P. Djotyan, P. N. Ignacz, M. A. Kedves, M. Serenyi, Z. Sorlei, J. Szigeti, and Z. Toth, "Interaction of frequency modulated light pulses with rubidium atoms in a magneto-optical trap," Eur. Phys. J. D **39**, 59–66 (2006).
11. X. Miao, E. Wertz, M. G. Cohen, and H. Metcalf, "Strong optical forces from adiabatic rapid passage," Phys. Rev. A **75**, 011402(R) (2007).
12. J. S. Bakos, G. P. Djotyan, P. Ignacz, M. A. Kedves, M. Serenyi, Z. Sorlei, J. Szigeti, and Z. Toth, "Acceleration of cold Rb atoms by frequency modulated light pulses," Eur. Phys. J. D **44**, 141–149 (2007).
13. G. Demeter, G. P. Djotyan, and J. S. Bakos, "Deflection and splitting of atomic beams using counter-propagating, short, chirped laser pulses," J. Opt. Soc. Am. B **15**, 16–24 (1998).
14. V. S. Voitsekhovich, M. V. Danileiko, A. M. Negrijko, V. I. Romanenko, and L. P. Yatsenko, "Observation of a stimulated radiation pressure of amplitude-modulated light on atoms," JETP Lett. **49**, 161–164 (1989).
15. J. Söding, R. Grimm, Y. B. Ovchinnikov, P. Bouyer, and C. Salomon, "Short-distance atomic beam deceleration with a stimulated light force," Phys. Rev. Lett. **78**, 1420–1423 (1997).
16. M. R. Williams, F. Chi, M. T. Cashen, and H. Metcalf, "Measurement of the bichromatic optical force on Rb atoms," Phys. Rev. A **60**, R1763–R1766 (1999).
17. M. R. Williams, F. Chi, M. T. Cashen, and H. Metcalf, "Bichromatic force measurements using atomic beam deflections," Phys. Rev. A **61**, 023408 (2000).
18. V. I. Romanenko and L. P. Yatsenko, "Scattering of atoms in a bichromatic field of oppositely propagating light pulses," JETP **90**, 407–414 (2000).
19. G. Demeter, G. P. Djotyan, Zs. Sörlei, and J. S. Bakos, "Mechanical effect of retroreflected frequency-chirped laser pulses on two-level atoms," Phys. Rev. A **74**, 013401 (2006).
20. J. F. Shirley, "Solution of the Schrödinger equation with a Hamiltonian periodic in time," Phys. Rev. **138**, B979–B987 (1965).
21. S. Guérin, L. P. Yatsenko, and H. R. Jauslin, "Dynamical resonances and the topology of the multiphoton adiabatic passage," Phys. Rev. A **63**, 031403(R) (2001).
22. B. W. Shore, *The Theory of Coherent Atomic Excitation* (Wiley, 1990).
23. S. Guérin, F. Monti, J.-M. Dupont, and H. R. Jauslin, "On the relation between cavity-dressed states, Floquet states, RWA and semiclassical models," J. Phys. A **30**, 7193–7215 (1997).
24. C. W. S. Conover, M. C. Doogue, and F. J. Struwe, "Chirped-pulse multiphoton transition between Rydberg states," Phys. Rev. A **65**, 033414 (2002).
25. H. Maeda, J. H. Gurian, D. V. L. Norum, and T. F. Gallagher, "Coherent population transfer in an atom by multiphoton adiabatic rapid passage," Phys. Rev. Lett. **96**, 073002 (2006).
26. G. N. Gibson, "Adiabatic passage on high-order multiphoton transitions," Phys. Rev. A **72**, 041404(R) (2005).
27. G. P. Djotyan, J. S. Bakos, G. Demeter, and Zs. Sörlei, "Population transfer in three-level Λ-atoms with Doppler-broadened transition lines by a single frequency-chirped short laser pulse," J. Opt. Soc. Am. B **17**, 107–113 (2000).
28. G. P. Djotyan, J. S. Bakos, G. Demeter, Zs. Sörlei, J. Szigeti, and D. Dzsotjan, "Creation of a coherent superposition of quantum states by a single frequency-chirped short laser pulse," J. Opt. Soc. Am. B **25**, 166–174 (2008).