# Collisionless Boltzmann equation with an external periodic traveling force: Analytical solution and application to molecular optics 

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#### Abstract

We present an analytical solution to the collisionless Boltzmann equation for describing the distribution function of molecular ensembles subject to an external periodic traveling force of pulsed optical fields. We apply our solution to study a pulsed standing wave mirror for neutral molecules, recently proposed [P. Ryytty et al., Phys. Rev. Lett. 84, 5074 (2000)]. Using our analytical solution we study the effects of the anharmonicity of optical potential on the reflectivity of the molecular mirror and the corresponding optimal pulse duration. We demonstrate that the reflectivity of the molecular mirror can be significantly improved by optimizing the pulse duration of the external optical fields when taking into account the anharmonicity of molecular motion.


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## I. INTRODUCTION

The Boltzmann equation describes the evolution of the position and velocity distribution of an ensemble of particles. It has extensive and important applications in the fields such as rarefied gas dynamics [1], statistical physics [2], and solid state physics [3], but can rarely be solved analytically [4-6]. The difficulty of solving the Boltzmann equation arises from the nonlinear nature of the collision term. However, if the mean-free path for an ensemble of particles is much larger than the typical dimension of the system, or the duration of the external force exerted on the system is much shorter than the mean collision time, the collision term can be neglected as a first approximation $[2,4,5]$. A collisionless environment is often realized in high temperature plasmas [7], molecular beams [8,9], atomic and molecular optics [10], and astrophysics [11]. More than 50 years ago, Landau derived the dispersion relation for electron plasma oscillations (Lagmuir oscillation) using the collisionless Boltzmann equation and the Poisson equation [7]. In most recent dipole force experiments using short pulsed lasers, solutions to the collisionless Boltzmann equation agree well with experimental results [12-14]. The collisionless Boltzmann equation without external force can be solved readily $[15,16]$. However, it is not a trivial task to find an analytical solution to this equation when an external force is present [11-14,17-19]. If the external force is weak, a perturbative technique can be used to obtain an approximate analytical solution [11-13]. For arbitrary external forces, numerical techniques such as the second-order McCormack method [20] can be applied to solve the equation [17-19].

In this paper, we study the one-dimensional (1D) collisionless Boltzmann equation with a periodic traveling force, which arises from the current research on manipulation of molecules with a one-dimensional far-off resonant optical lattice. An optical lattice is a periodic dipole potential created by two counterpropagating optical fields [12-14,21,22]. In previous experiments on coherent Rayleigh scattering [12] and its application to a noninvasive measurement of temperature of a gas [13], this equation was used to describe the
longitudinal position and velocity distribution of molecules in an optical lattice and was solved with a perturbation technique for weak optical dipole forces. In a study of a pulsed standing wave mirror, this equation was solved numerically [21,22] to model the reflection of a molecular beam by a pulsed optical standing wave. Here, we present an analytical solution to the equation for any initial distribution by using the method of characteristics [15]. The method of characteristics reduces the problem of the distribution function of an ensemble of particles to that of a probability density function of a single particle; the distribution function of the system equals the initial probability density of the single particle. Two steps were required to obtain the solution. The first step obtained a general solution to the (Newtonian) equation of motion for the particle, referred as the characteristics of the collisionless Boltzmann equation with external force. The second step was to recover the initial state (backward characteristics) of the particle using the characteristics. Using this procedure we obtain the analytical expression for the distribution function of the collisionless Boltzmann equation in terms of the initial condition and the parameters of the optical fields and molecules, such as optical intensity and the polarizability of molecules.

In the second part of this paper, we apply our analytical solution to investigate a pulsed standing wave mirror for neutral molecules, which Ryytty and Kaivola studied numerically [21]. Such a mirror may be important for designing matter wave cavities [23,24], atomic wave guides [25,26], and interferometers [27,28]. Mirrors for reflecting atoms by an evanescent optical wave have been realized previously [29,30], but this approach fails for molecules and atoms with complex energy structure [21,22]. Ryytty and Kaivola recently proposed a new method for a molecular mirror based on the properties of the motion of molecules trapped in the optical potential wells formed by two counterpropagating optical fields $[21,22]$. They suggested that a molecular mirror could be realized when the pulse duration of the standing wave was chosen to be half a period of the simple harmonic motion of trapped molecules [21,22]. In this paper, we use our analytical model to extend their approach considering the more general case of the anharmonic motion of molecules in


FIG. 1. An optical lattice is created by two optical fields at angle $\beta \approx 180^{\circ}$. A molecular beam with longitudinal velocity $v_{D}$ is injected into the interference pattern of the two crossed optical fields.
a periodic potential. We study the effects of the anharmonicity on optimal pulse duration and the reflectivity of the molecular mirror.

## II. THE ONE-DIMENSIONAL COLLISIONLESS BOLTZMANN EQUATION WITH AN EXTERNAL OPTICAL PERIODIC TRAVELING FORCE

We study the interaction of a molecule in a collimated molecular beam with a quasi-one-dimensional optical lattice, as shown in Fig. 1. A pulsed optical lattice is created by two counterpropagating optical fields $\varepsilon_{1}(\mathbf{r}, t)=E_{1}(t) \sin \left(\mathbf{k}_{1} \cdot \mathbf{r}\right.$ $\left.-\omega_{1} t\right)$ and $\varepsilon_{2}(\mathbf{r}, t)=E_{2}(t) \sin \left(\mathbf{k}_{2} \cdot \mathbf{r}-\omega_{2} t\right)$, where $E_{1}$ and $E_{2}$ are the amplitudes of the two optical fields, $\mathbf{r}$ is a coordinate vector, $\mathbf{k}_{1}$ and $\mathbf{k}_{2}$ are the wave vectors, and $\omega_{1}$ and $\omega_{2}$ are frequencies of optical waves chosen to be far-off resonant to form quasielectrostatic potential [31]. We consider optical pulses that have a square temporal profile, where $E_{1}(t)=E_{2}(t)=E_{0}$ for $0<t \leqslant t_{d}$, with a pulse duration $t_{d}$, which is typically in the nanosecond range $[12,13,18,21,22,32]$. The $x$ axis is set to be parallel to the axis of the collimated molecular beam. The dipole potential of a molecule in the nonresonant field is given by $U(\mathbf{r}, t)=-\alpha E_{1} E_{2} / 2 \cos \left[\left(\mathbf{k}_{1}-\mathbf{k}_{2}\right) \cdot \mathbf{r}-\left(\omega_{2}-\omega_{1}\right) t\right]$, where $\alpha$ is polarizability, and the dipole force $\mathbf{G}(\mathbf{r}, t)$ exerted on a molecule is given by $\mathbf{G}=-\nabla U(\mathbf{r}, t)$ $=-\alpha E_{1} E_{2} / 2 \sin \left[\left(\mathbf{k}_{1}-\mathbf{k}_{2}\right) \cdot \mathbf{r}-\left(\omega_{2}-\omega_{1}\right) t\right]\left(\mathbf{k}_{1}-\mathbf{k}_{2}\right)$. In many experimental schemes [12,13,18,21,22,32], where the two optical fields are nearly counterpropagating, the angle between the optical fields and the supersonic beam is very small $\left(<2.5^{\circ}\right)$, so that the transverse wave vectors $\mathbf{k}_{1 \perp}$ and $\mathbf{k}_{2 \perp}$ are small. In these experiments, there is a small difference in the frequencies of the two beams and, therefore, $\mathbf{k}_{1 \perp} \approx \mathbf{k}_{2 \perp}$. With these approximations, $\quad \sin \left[\left(\mathbf{k}_{1}-\mathbf{k}_{2}\right) \cdot \mathbf{r}-\left(\omega_{2}-\omega_{1}\right) t\right]=\sin \left[\left(k_{1 x}\right.\right.$ $\left.\left.+k_{2 x}\right) x+\left(\mathbf{k}_{1 \perp}-\mathbf{k}_{2 \perp}\right) \cdot \mathbf{r}_{\perp}-\left(\omega_{2}-\omega_{1}\right) t\right] \approx \sin \left[\left(k_{1 x}+k_{2 x}\right) x-\left(\omega_{2}\right.\right.$ $\left.\left.-\omega_{1}\right) t\right]=\sin \left[q\left(x-v_{L} t\right)\right]$, where $k_{1 x}$ and $k_{2 x}$ are the components of wave vectors $\mathbf{k}_{1}$ and $\mathbf{k}_{2}$ in the direction of $x$ axis, and $q=k_{1 x}+k_{2 x}$, and $v_{L}=\left(\omega_{2}-\omega_{1}\right) / q$. Thus, the optical force along the $x$ axis, $G_{\|}=-\alpha E_{1} E_{2} / 2 \sin \left[\left(\mathbf{k}_{1}-\mathbf{k}_{2}\right) \cdot \mathbf{r}\right.$ $\left.-\left(\omega_{2}-\omega_{1}\right) t\right]\left(k_{1 x}+k_{2 x}\right)$, is approximated by

$$
\begin{equation*}
G_{\|} \approx-G_{0} \sin \left[q\left(x-v_{L} t\right)\right], \tag{1}
\end{equation*}
$$

where $G_{0}=\alpha q E_{1} E_{2} / 2$ is the maximum force in the longitudinal direction. In the transverse direction, the force $\mathbf{G}_{\perp}$ $=-\alpha E_{1} E_{2} / 2 \sin \left[\left(\mathbf{k}_{1}-\mathbf{k}_{2}\right) \cdot \mathbf{r}-\left(\omega_{2}-\omega_{1}\right) t\right]\left(\mathbf{k}_{1 \perp}-\mathbf{k}_{2 \perp}\right)$ is approximated by

$$
\begin{equation*}
\mathbf{G}_{\perp} \approx-G_{0} \sin \left[q\left(x-v_{L} t\right)\right]\left(\mathbf{k}_{1 \perp}-\mathbf{k}_{2 \perp}\right) / q \tag{2}
\end{equation*}
$$

We note that the ratio $\left|\left(\mathbf{k}_{1 \perp}-\mathbf{k}_{2 \perp}\right) / q\right|$ is much less than $1 \%$ for nearly counterpropagating beams $\left(\beta>175^{\circ}\right.$ in Fig. 1) and, therefore, the dipole force $\mathbf{G}_{\perp}$ in the transverse direction is much less than the longitudinal force $G_{\|}$. In order to demonstrate that we can neglect the transverse force in further calculations we estimate its effect on the molecular beam for the duration of the optical field $t_{d}$. The velocity change induced by the force is given by $\left|\Delta \mathbf{v}_{\perp}\right|<\mid G_{0}\left(\mathbf{k}_{1 \perp}\right.$ $\left.-\mathbf{k}_{2 \perp}\right) /(m q) \mid t_{d}$, and the displacement is $|\Delta \mathbf{s}|<\left|\mathbf{v}_{\perp 0} t_{d}\right|$ $+\left|G_{0}\left(\mathbf{k}_{1 \perp}-\mathbf{k}_{2 \perp}\right) /(q m)\right| t_{d}^{2} / 2$, where $m$ is the mass of the molecule and $\mathbf{v}_{\perp 0}$ is the initial transverse velocity. For a $\mathrm{Rb}_{2}$ (mass $m=2.82 \times 10^{-25} \mathrm{~kg}, \alpha=135 \AA^{3}$ ) molecular beam at a temperature of 1 mK (the most probable velocity of $v_{m}$ $\approx 0.63 \mathrm{~m} / \mathrm{s}$, we estimate a transverse velocity change of $\left|\Delta \mathbf{v}_{\perp}\right|<8.5 \mu \mathrm{~m} / \mathrm{s}$, and a transverse displacement $|\Delta \mathbf{s}|$ $<12.6 \mathrm{~nm}$. These calculations are carried out using an optical field with an intensity $10 \mathrm{GW} / \mathrm{cm}^{2}$, a wave length 1064 nm , a pulse duration $t_{d}=10 \mathrm{~ns}$, and the ratio $\left|\left(\mathbf{k}_{1 \perp}-\mathbf{k}_{2 \perp}\right) / q\right|=1 \%$. The initial transverse velocity $\mathbf{v}_{\perp 0}$ we used in this estimation is twice the most probable velocity determined by the temperature. As in this example, where the duration of typical experimental schemes is in the nanosecond range, the transverse velocity change $\left|\Delta \mathbf{v}_{\perp}\right|$ induced by the potential is much less than the most probable velocity $v_{m}$, and the transverse displacement $|\Delta \mathbf{s}|$ is much less than the typical width of the optical lattice $(100 \mu \mathrm{~m})$. Therefore, the weak transverse force $\mathbf{G}_{\perp}$ does not significantly change the transverse position or velocity of molecules within the pulse duration we consider. We conclude that only a very small fraction of the molecules will enter or escape from the potential during these time periods.

We consider the motion of the molecules in a high intensity optical lattice where the momentum transferred to the molecules is several orders of magnitude larger than the recoil momentum. Under these conditions we can treat the molecules as classical particles [21,22]. Almost collisonless conditions can be created within a pulsed supersonic beam [8,9], when the interaction time between the optical fields and molecules is shorter than the collision time. For this situation the position and velocity distribution function $f(\mathbf{r}, \mathbf{v}, t)$ can be described by the collisionless Boltzmann equation $[2,4,5]$,

$$
\begin{equation*}
\frac{\partial f(\mathbf{r}, \mathbf{v}, t)}{\partial t}+\mathbf{v} \cdot \frac{\partial f(\mathbf{r}, \mathbf{v}, t)}{\partial \mathbf{r}}+\frac{\mathbf{G}(\mathbf{r}, t)}{m} \cdot \frac{\partial f(\mathbf{r}, \mathbf{v}, t)}{\partial \mathbf{v}}=0 . \tag{3}
\end{equation*}
$$

During the short interaction time, the transverse motion of molecules cannot be significantly affected by the weak transverse force $\mathbf{G}_{\perp}$, we thus treat $\mathbf{G}_{\perp}$ as a perturbation, and neglect $\mathbf{G}_{\perp}$ in Eq. (3) in the zero-order approximation of the position and velocity distribution function $f(\mathbf{r}, \mathbf{v}, t)$. With this approximation, we can decouple the transverse motion to the longitudinal motion and make the approximation that $f(\mathbf{r}, \mathbf{v}, t)=f_{\|}\left(x, v_{x}, t\right) f_{\perp}\left(\mathbf{v}_{\perp}, \mathbf{v}_{\perp}, t\right)$, where the transverse molecular beam distribution function $f_{\perp}\left(\mathbf{v}_{\perp}, \mathbf{v}_{\perp}, t\right)$ and the longitudinal molecular beam distribution function $f_{\|}\left(x, v_{x}, t\right)$ are independent. As the effect of the external fields on the
transverse molecular beam distribution can be neglected, the parallel molecular beam distribution function $f_{\|}\left(x, v_{x}, t\right)$ can be determined by the 1D Boltzmann equation

$$
\begin{equation*}
\frac{\partial f_{\|}\left(x, v_{x}, t\right)}{\partial t}+v \frac{\partial f_{\|}\left(x, v_{x}, t\right)}{\partial x}+\frac{G_{\|}(x, t)}{m} \frac{\partial f_{\|}\left(x, v_{x}, t\right)}{\partial v}=0 . \tag{4}
\end{equation*}
$$

The use of Eq. (4) is borne out by recent experimental work in coherent Rayleigh scattering [12] and its applications [13] where the distribution function was measured using light scattering techniques. The experimental results can be well described by Eq. (4). This equation has also been used in theoretical treatments to describe a pulsed standing wave mirror $[21,22]$ for molecules, and the deceleration of cold molecules in a molecular beam using a one-dimensional far-off resonant optical lattice [18]. This formalism could also be used to model recent experiments demonstrating the decelerating, trapping, and bunching of molecules by a Stark deceleration [33-35] and measurement of the polarizability of molecules with the optical dipole force [32].

In the following, we study how to solve Eq. (4) analytically. For convenience, we drop the subscript || from the distribution function $f_{\|}\left(x, v_{x}, t\right)$.

## III. ANALYTICAL SOLUTION TO THE ONE-DIMENSIONAL COLLISIONLESS BOLTZMANN EQUATION WITH AN EXTERNAL PERIODIC TRAVELING FORCE

The one-dimensional collisionless Boltzmann equation with an external periodic traveling force, given by Eq. (4), has been used to model the position and velocity distribution of molecules in 1D optical lattices. When the optical dipole force is weak, 1D perturbation solutions can be used to accurately model experiments [12-14]. However, no analytical solution to this equation for arbitrarily strong fields has been presented. In this section, using the method of the characteristic, we find an analytical solution, for arbitrarily strong fields, which are of interest for a number of applications such as a pulsed standing wave mirror $[21,22]$ and for the creation of cold molecules by deceleration [18,33-35].

Before we solve Eq. (4), we introduce a normalization transformation. We denote the phase of the traveling force by $\theta$,

$$
\begin{equation*}
\theta=q\left(x-v_{L} t\right), \tag{5}
\end{equation*}
$$

and introduce the normalized time $\tau$ and normalized velocity $\eta$ by

$$
\begin{gather*}
\tau=t \omega_{0},  \tag{6}\\
\eta=\left(v_{x}-v_{L}\right) / v_{n}, \tag{7}
\end{gather*}
$$

where $\omega_{0}=\sqrt{G_{0} q / m}$ and $v_{n}=\sqrt{G_{0} /(m q)}$. We further introduce the normalized distribution function $f(\theta, \eta, \tau)$ in the new normalized phase space $(\theta, \eta)$ by

$$
\begin{equation*}
f(\theta, \eta, \tau)=v_{n} f(x, v, t) / q \tag{8}
\end{equation*}
$$

Inserting Eqs. (5)-(8) into Eq. (4), we obtain the normalized collisionless Boltzmann equation,

$$
\begin{equation*}
\frac{\partial f(\theta, \eta, \tau)}{\partial \tau}+\eta \frac{\partial f(\theta, \eta, \tau)}{\partial \theta}-\sin (\theta) \frac{f(\theta, \eta, \tau)}{\partial \eta}=0 \tag{9}
\end{equation*}
$$

which we solve by using the method of characteristics [15] by rewriting this equation into the following three equations:

$$
\begin{gather*}
\frac{d f(\theta(\tau), \eta(\tau), \tau)}{d \tau}=0  \tag{10}\\
\frac{d \theta(\tau)}{d \tau}=\eta(\tau)  \tag{11}\\
\frac{d \eta(\tau)}{d \tau}=-\sin [\theta(\tau)] \tag{12}
\end{gather*}
$$

The function $f(\theta(\tau), \eta(\tau), \tau)$ in Eq. (10) can be interpreted as the probability density function of a molecule [4], the motion of which is described by Eqs. (11) and (12), which are the equations of motion for a pendulum driven by a force of sine form. From Eq. (10), we obtain

$$
\begin{equation*}
f(\theta(\tau), \eta(\tau), \tau)=f(\theta(0), \eta(0), 0)=f_{0}(\theta(0), \eta(0)) \tag{13}
\end{equation*}
$$

which shows that the probability density in the phase space is unchanged in time, even with the presence of the external force. This is due to the collisionless environment of the molecules. This result is consistent with the recent experiments with a Stark decelerator $[33,34]$. Using Eq. (13) and the normalization relation Eq. (8), the distribution function is given by

$$
\begin{equation*}
f(x, v, t)=q f_{0}(\theta(0), \eta(0)) / v_{n} \tag{14}
\end{equation*}
$$

indicating that the distribution function of an ensemble of molecules is proportional to the probability density function of a single molecule in the collisionless environment. However, the initial position $(\theta(0), \eta(0))$ of the molecule is unknown, therefore, to obtain the analytical relation (characteristics) between $(\theta(\tau), \eta(\tau))$ and $(\theta(0), \eta(0))$, we need to solve Eqs. (11) and (12) for $\theta(\tau)$ and $\eta(\tau)$, and then carry out an inverse procedure to determine $(\theta(0), \eta(0)$ ) (backward characteristics) from $(\theta(\tau), \eta(\tau))$.

Equations (11) and (12) are the equations of motion for a nonlinear pendulum, which has been extensively studied in mechanics [36] and engineering fields [37,38]. In previous works, the periodic behavior of the nonlinear pendulum was studied [36-39] and some solutions were presented for special initial conditions [39]. However, in this paper we study the motion of an ensemble of molecules, which requires general solutions to Eqs. (11) and (12) so that the inverse procedure can be done.

To solve the equations of motion (11) and (12), we first present the corresponding normalized Hamiltonian of a molecule $H(\theta(\tau), \eta(\tau))$ by

$$
\begin{equation*}
H(\theta(\tau), \eta(\tau))=\eta(\tau)^{2} / 2-\cos [\theta(\tau)] \tag{15}
\end{equation*}
$$

From Eq. (15), we obtain the normalized velocity

$$
\begin{equation*}
\eta(\tau)= \pm \sqrt{N^{2}-\sin ^{2}[\theta(\tau) / 2]} \tag{16}
\end{equation*}
$$

where the parameter $N$ is defined as

$$
\begin{equation*}
N=\sqrt{[H(\theta(\tau), \eta(\tau))+1] / 2}=\sqrt{\eta(\tau)^{2} / 4+\sin ^{2}[\theta(\tau) / 2]} . \tag{17}
\end{equation*}
$$

Since the system is conservative, the parameter $N$ is time independent. The dynamical behavior of the molecules is classified by the parameter $N$. When $N \leqslant 1$ molecules are trapped by the potential well [the second term in the right side of Eq. (15)] and for $N>1$ molecules are untrapped [39].

If a molecule is untrapped by the potential wells, it travels from one potential well to another, whereas if trapped, it oscillates within one of the potential wells [39]. We discuss these two motion states separately. In this section we present analytical results for trapped molecules. The analytical results for untrapped molecules are given in the Appendix A. The mathematical procedures for the two cases are similar.

For the trapped case $N \leqslant 1$, we define a function $\xi(\tau)$ by

$$
\begin{equation*}
N \sin [\xi(\tau)]=\sin [\theta(\tau) / 2] \tag{18}
\end{equation*}
$$

Differentiating both sides of Eq. (18) with respect to the normalized time $\tau$, and using Eq. (16) we obtain

$$
\begin{equation*}
\frac{d \xi(\tau)}{d \tau}= \pm \sqrt{1-\{N \sin [\xi(\tau)]\}^{2}} \tag{19}
\end{equation*}
$$

which has the solution

$$
\begin{equation*}
\xi(\tau)=\operatorname{am}(\tau+\phi, N), \tag{20}
\end{equation*}
$$

where $\operatorname{am}(\tau+\phi, N)$ is a Jacobian elliptical amplitude function $[40,41]$ and $\phi=\int_{0}^{\xi(0)}\left\{1-[N \sin (\beta)]^{2}\right\}^{-1 / 2} d \beta$ is a constant. Using Eqs. (18) and (20), the normalized velocity [Eq. (16)] becomes

$$
\begin{equation*}
\eta(\tau)=2 N \operatorname{cn}(\tau+\phi, N) \tag{21}
\end{equation*}
$$

where $\operatorname{cn}(\tau+\phi, N)$ is a Jacobian elliptical cosine function [40,41]. The velocity $\eta(\tau)$ is a periodic function of $\tau$, and its period $P_{\text {trap }}(N)$ is given by

$$
\begin{equation*}
P_{\text {trap }}(N)=4 \int_{0}^{\pi / 2}\left\{1-[N \sin (\beta)]^{2}\right\}^{-1 / 2} d \beta=4 K(N), \tag{22}
\end{equation*}
$$

where $K(N)$ is the complete Jacobian elliptic integral of the first kind $[40,41]$. Equation (22) shows that the period of a trapped molecule depends only on the parameter $N$.

We integrate Eq. (21) with respect to the normalized time $\tau$ and obtain

$$
\begin{align*}
\theta(\tau)= & \theta(0)+2 \operatorname{sgn}[\operatorname{sn}(t+\phi, N)] \arccos [\operatorname{dn}(t+\phi, N)] \\
& -2 \operatorname{sgn}[\operatorname{sn}(\phi, N)] \arccos [\operatorname{dn}(\phi, N)] \tag{23}
\end{align*}
$$

where $\operatorname{sgn}(x)$ is a $\operatorname{sign}$ function, $\operatorname{sgn}(x)=1$ for $x>0$, and -1 otherwise. $\theta(\tau)$ and $\eta(\tau)$ given by Eqs. (23) and (21) are a general solution (characteristics) of Eqs. (11) and (12).

Now let us study the inverse process to obtain the initial phase and velocity $(\theta(0), \eta(0))$ from $(\theta(\tau), \eta(\tau))$. The initial velocity $\eta(0)$ is given by Eq. (21) for $\tau=0$. Using $\phi$ $=\tau+\phi-\tau$ and the addition theory of Jacobian elliptic functions [40,41], we have

$$
\begin{equation*}
\eta(0)=2 N \frac{\operatorname{cn}(\tau+\phi, N) \operatorname{cn}(\tau, N)+\operatorname{sn}(\tau+\phi, N) \operatorname{sn}(\tau, N) \operatorname{dn}(\tau+\phi, N) \operatorname{dn}(\tau, N)}{1-N^{2}[\operatorname{sn}(\tau+\phi, N) \operatorname{sn}(\tau, N)]^{2}} . \tag{24}
\end{equation*}
$$

To find the relations of all $\phi$-dependent terms in Eq. (24) to the state $(\theta(\tau), \eta(\tau))$, we differentiate both sides of Eq. (21) with respect to the normalized time $\tau$, and compare this expression for $d \eta(\tau) / d \tau$ with Eq. (12) to obtain

$$
\begin{equation*}
\sin [\theta(\tau)]=2 N \operatorname{sn}(\tau+\phi, N) \operatorname{dn}(\tau+\phi, N) \tag{25}
\end{equation*}
$$

Using Eqs. (17), (21), (25), and the relations between the Jacobian elliptical functions, from Eq. (24), we finally get an analytical expression for the initial velocity $\eta(0)$ by $\theta(\tau)$ and $\eta(\tau)$,

$$
\begin{equation*}
\eta(0)=\frac{\eta(\tau) \operatorname{cn}(\tau, N)+\sin [\theta(\tau)] \operatorname{sn}(\tau, N) \operatorname{dn}(\tau, N)}{1-\left[\sin \left(\frac{\theta(\tau)}{2}\right) \operatorname{sn}(\tau, N)\right]^{2}} \tag{26}
\end{equation*}
$$

Next, we determine the expression for initial phase $\theta(0)$ from Eq. (23). Using $\phi=t+\phi-t$ and the addition theory of elliptical functions [40,41], $\operatorname{sn}(\phi, N)$ and $\operatorname{dn}(\phi, N)$ in Eq. (23) are given as

$$
\begin{gather*}
\operatorname{sn}(\phi, N)=\frac{\operatorname{sn}(t+\phi, N) \operatorname{cn}(t, N) \operatorname{dn}(t, N)-\operatorname{cn}(t+\phi, N) \operatorname{dn}(t+\phi, N) \operatorname{sn}(t, N)}{1-[N \operatorname{sn}(t+\phi, N) \operatorname{sn}(t, N)]^{2}}  \tag{27}\\
\operatorname{dn}(\phi, N)=\sqrt{1-[N \operatorname{sn}(\phi, N)]^{2}} \tag{28}
\end{gather*}
$$

From Eq. (21) and the relations between the Jacobian elliptical functions [40,41], $\operatorname{dn}(t+\phi, N)$ is determined by

$$
\begin{equation*}
\operatorname{dn}(t+\phi, N)=\sqrt{1-N^{2}+[N \operatorname{cn}(t+\phi, N)]^{2}}=\sqrt{1-N^{2}+\eta(\tau)^{2} / 4} \tag{29}
\end{equation*}
$$

Further using Eq. (25), we obtain

$$
\begin{equation*}
\operatorname{sn}(t+\phi, N)=\sin [\theta(\tau)] /\left[2 N \sqrt{1-N^{2}+\eta(\tau)^{2} / 4}\right] \tag{30}
\end{equation*}
$$

and by inserting Eqs. (21), (29), and (30) into Eq. (27), we arrive at

$$
\begin{equation*}
\operatorname{sn}(\phi, N)=\frac{\sin [\theta(\tau)] \operatorname{cn}(t, N) \operatorname{dn}(t, N)-\eta(\tau)\left[1-N^{2}+\eta(\tau)^{2} / 4\right] \operatorname{sn}(t, N)}{2 N\left\{1-\left[N^{2}-\eta(\tau)^{2} / 4\right] \operatorname{sn}(t, N)^{2}\right\} \sqrt{1-N^{2}+\eta(\tau)^{2} / 4}} \tag{31}
\end{equation*}
$$

Substituting Eqs. (28)-(31) into Eq. (23), we can determine $\theta(0)$ with $\theta(\tau)$ and $\eta(\tau)$.

In summary, the procedure for calculating the distribution function of the collisionless Boltzmann equation (4) is the following.
(1) Calculate $\tau, \eta(\tau), \theta(\tau)$ by the normalization transformation, Eqs. (5)-(7), from $t, v, x$.
(2) Calculate $N$ by Eq. (17).
(3) If $N<1$, first determine $(\theta(0), \eta(0))$ from $(\theta(\tau), \eta(\tau))$ using Eqs. (26) and (23), and then obtain the distribution function $f(x, v, t)$ by inserting $(\theta(0), \eta(0))$ into Eq. (14).

## IV. APPLICATION TO A PULSED STANDING WAVE MIRROR FOR NEUTRAL MOLECULES

In this section, we apply our analytical results to the study of a pulsed standing wave mirror, recently proposed as an optical element for reflecting a molecular beam $[21,22]$. A schematic diagram of the two optical fields interacting with a supersonic molecular beam to produce a pulsed standing wave is shown in Fig. 1. The two optical fields have the same frequency, and the force in Eq. (1) is time independent $\left(v_{L}\right.$ $=0$ ).

To understand the dynamics of molecules in pulsed standing wave mirror (stationary optical lattice), we present a phase space $(\theta, \eta)$ plot in Fig. 2. Each line is an equal energy line corresponding to a different value of the parameter $N$ defined in Eq. (17). Dotted lines are separatrix, which define two dynamical regions: molecules enclosed by them are trapped, whereas molecules outside are untrapped. Arrows show the direction of the motion of the molecules. As shown in Fig. 2, untrapped molecules move from one potential well to another and keep their initial motion direction unchanged, whereas trapped molecules move along close trajectories and the directions of their motions change periodically. Therefore, only trapped molecules can be reflected by the dipole potential well. The cross points of the solid, dashed, and dot-dashed orbits of trapped molecules with the lines $\eta$ $=\eta_{D}$ and $\eta=-\eta_{D}$ are denoted by $A, B, C, \ldots, G, H$ as shown in this figure. It can be seen that there are three dynamical processes contributing to reflection of molecules from $\eta_{D}$ to $-\eta_{D}$. The first of these processes involves molecules whose initial phases are between 0 and $\pi$. This cor-
responds to those traveling from $A$ to $B$ in Fig. 2. For this situation, the time required for reflection is shorter than a half-oscillation period. The duration is given by $\tau_{r}$ $=2\left[K(N)-\mathrm{cn}^{-1}\left(\eta_{D} / N, N\right)\right]$, where $\mathrm{cn}^{-1}$ is the inverse function of the Jacobian cosine function cn. The time $\tau_{r}$ for this process, and the following two processes, are obtained with the results presented in Sec. II and are given in Appendix B. The second process involves two groups of molecules whose initial phases are symmetrical with $\theta=0$. This corresponds to the groups at $F$ and $C$ traveling to $D$ and $E$, respectively, as shown in Fig. 2. The two groups reverse their initial velocity after traveling half their orbit and, therefore, with a half of the period, given by $\tau_{r}=2 K(N)$. A third process involves molecules whose initial phases are between $-\pi$ to 0 , which are reflected after traveling more than a half


FIG. 2. (a) Each line in the $(\theta, \eta)$ space is an equal energy line corresponding to a value of the parameter $N$ defined in Eq. (17). The dotted lines are separatrix and the region enclosed by them is a trapping region. The arrows show the direction of motion, and the points denoted by $A, B, C, \ldots, G, H$ are the crossing points of the orbits of the trapped molecules with the lines $\eta=\eta_{D}$ and $\eta$ $=-\eta_{D}$. Here $\eta_{D}$ is the initial velocity. The three dynamical reflection processes are revealed: the first is from $A$ to $B$, the second is from $C$ to $E$ and from $F$ to $D$, and the third is from $G$ to $H$. (b) The time $\tau_{r}$ required for reflecting molecules with the initial velocity $\eta_{D}=0.264$ varies with the parameter $N$.
of the period. These are molecules that travel from $G$ to $H$, with time $\tau_{r}=2\left[K(N)+\mathrm{cn}^{-1}\left(\eta_{D} / N, N\right)\right]$. The time $\tau_{r}$ required for each of the three processes, corresponding to different equal energy lines, varies in a wide range as shown in Fig. 2(b), which implies that the trapped molecules cannot be reflected at the same time. Consequently, there is a question of what is the optimal pulse duration to realize maximum reflectance. In the following we apply the analytical results obtained in Sec. III to study the velocity distribution of the molecular beams and determine the optimal pulse duration.

The initial longitudinal velocity distribution of a free molecular beam is described by a Maxwellian model, $f_{0}\left(x, v_{x}\right)$ $=\exp \left[-\left(v_{x}-v_{D}\right)^{2} / v_{m}^{2}\right] /\left(2 \pi \sqrt{\pi} v_{m}\right) \quad$ with $\quad v_{m}=\sqrt{2 K_{B} T / m}$, where $K_{B}$ is the Boltzmann constant and $v_{D}$ is the molecular beam velocity. As the dipole force is of the form of Eq. (1), the distribution of a molecular beam after the optical fields are turned on is $f\left(x, v_{x}, t\right)=q f(\theta, \eta, \tau) / v_{n}$, as given by Eq. (14), where the normalized distribution $f(\theta, \eta, \tau)$ is read as

$$
\begin{equation*}
f(\theta, \eta, \tau)=\eta_{m} \exp \left\{-\eta_{m}^{-2}\left[\eta_{0}(\theta, \eta, \tau)-\eta_{D}\right]^{-2}\right\} /(2 \pi \sqrt{\pi}) \tag{32}
\end{equation*}
$$

where $\eta_{m}=v_{m} / v_{n}$ is the normalized $1 / e$ width of the initial distribution, $\eta_{D}=v_{D} / v_{n}$ is the normalized beam velocity, and $\eta_{0}(\theta, \eta, \tau)$ is the initial velocity of a molecule with phase $\theta$ and normalized velocity $\eta$ at normalized time $\tau$. $\eta_{0}(\theta, \eta, \tau)$ depends on the parameter $N$ and is calculated by Eq. (26) for $N \leqslant 1$, otherwise by Eq. (A10). Due to the periodic motion of the molecule, the velocity $\eta_{0}(\theta, \eta, \tau)$ is a periodic function with a period given by Eq. (22) or Eq. (A5). Consequently, the normalized distribution $f(\theta, \eta, \tau)$ is also a periodic function.

Based on Eq. (32), we now study the reflection of the molecular beam by the pulsed standing wave. As an example, we consider the beam with a normalized velocity $\eta_{D}=2.64 \times 10^{-1}$ and normalized width $\eta_{m}=6.90 \times 10^{-3}$, which corresponds to $\mathrm{Rb}_{2}$ molecules with the beam velocity $v_{D}=10 \mathrm{~m} / \mathrm{s}$ and temperature $T=700 \mu \mathrm{~K}$ in an optical lattice formed by optical field of intensity $7.25 \mathrm{GW} / \mathrm{cm}^{2}$. For this parameter set, the motion of the molecules in the region around the antinodes is quasiharmonic and, therefore, is similar to the case discussed in references [21,22].

The velocity distributions of reflected molecular beam are shown in Fig. 3(a) for several pulse durations. The normalized velocity distribution $p\left(\eta, \tau_{d}\right)$ is calculated by $p\left(\eta, \tau_{d}\right)$ $=\int_{-\pi}^{\pi} \exp \left\{-\eta_{m}^{-2}\left[\eta_{0}\left(\theta, \eta, \tau_{d}\right)-\eta_{D}\right]^{-2}\right\} /\left(2 \pi \sqrt{\pi} \eta_{m}\right) d \theta$. The dotted and solid lines, respectively, correspond to $\tau_{d}=\pi$, which is half a period of the simple harmonic motion, and $\tau_{d}=3.18$. Both the distributions are similar to Gaussian centered at $\eta=-\eta_{D}$ and have almost the same width as that of the initial distribution. On the contrary, the dashed line for $\tau_{d}=0.53$ is asymmetric with $\eta$, whereas the dot-dashed line for $\tau_{d}=3.22$ is double peaked. The peak for $\tau_{d}=3.18$ is the highest. Figure 3(a) reveals that the velocity distributions sensitively depend on the pulse duration and the optimal pulse duration occurs at $\tau_{d}>\pi$. With our analytical results we can quickly calculate the velocity distribution $p\left(-\eta_{D}, \tau_{d}\right)$ for different pulse durations to determine an optimal duration $\tau_{d}$. Figure 3(b) gives the dependence of


FIG. 3. The reflection of a molecular beam with velocity $\eta_{D}$ $=2.64 \times 10^{-1}$ and an initial $1 / e$ velocity width of $\eta_{m}$ $=6.90 \times 10^{-3}$ by a pulsed standing wave. (a) The velocity distribution for ensemble of reflected molecules corresponding to several pulse durations. (b) The dependence of the distribution function $p\left(-\eta_{D}, \tau_{d}\right)$ on the pulse duration $\tau_{d}$.
$p\left(-\eta_{D}, \tau_{d}\right)$ for different values of $\tau_{d}$. On increasing the duration, the first peak appears at $\tau_{d}=0.53$ corresponding to the first dynamical process of transferring molecules from $\eta_{D}$ to $-\eta_{D}$ (reflection) as revealed in Fig. 2, and the second occurs at $\tau_{d}=3.18$ corresponding predominantly to the latter two processes discussed in Fig. 2. The following peaks occur because of the periodic nature of the molecular motion in the potential wells. However, the molecules that contribute to the second peak have different periods, therefore at a later time not all of them arrive at the same velocity $-\eta_{D}$ at the same time. Consequently, the peak height decreases with time. The second peak gives the highest concentration of reflected molecules at $\eta=-\eta_{D}$ and therefore $\tau_{d}=3.18$ is the optimal pulse duration.

Figure 4 shows a more general relation of the optimal duration $\tau_{d, \text { opt }}$ and their corresponding peak heights $p\left(-\eta_{D}, \tau_{d, \text { opt }}\right)$ to beam velocity $\eta_{D}$. On increasing the beam velocity $\eta_{D}$, it shows that the optimal duration $\tau_{d \text {,opt }}$ increases from half a period of simple harmonic period $\pi$, but the peak height $p\left(-\eta_{D}, \tau_{d \text {,opt }}\right)$ decreases. These results


FIG. 4. The dependence of the optimal duration $\tau_{d, \text { opt }}$ (solid line) and the velocity distribution $p\left(-\eta_{D}, \tau_{d, \text { opt }}\right)$ (dashed line) on the beam velocity $\eta_{D}$. Due to anharmonicity, the optimal pulse duration $\tau_{d, \text { opt }}$ increases with increasing the beam velocity $\eta_{D}$, while the velocity density at the velocity $-\eta_{D}, p\left(-\eta_{D}, \tau_{d, \text { opt }}\right)$, decreases.
are consistent with the previous finding [21] that anharmonicity is more profound for higher velocity such that the reflectance is decreased. We note, however, even when the motions of molecules are predominantly anharmonic at higher beam velocity, reflection can still be achieved, though with significantly reduced peak height. This result agrees with the dynamical analysis of reflecting molecules shown in Fig. 2.

## V. SUMMARY

We have obtained a general analytical solution to the collisionless Boltzmann equation describing the distribution of an ensemble of molecules in an external periodic traveling field for arbitrary initial conditions. The analytical solution enables us to investigate the relation of the final distribution function to the initial velocity distribution, its bulk velocity and its initial velocity width, as well as the magnitude of the external force. Our results can be used to aid in the design of pulsed standing wave mirrors [21,22], coherent Rayleigh scattering using arbitrary fields [12-14], deceleration of molecular beams with optical lattices [18], and the measurement of molecular polarizability by an optical dipole force [32]. As a case study, we have applied our analytical method to study the pulsed standing wave mirror for $\mathrm{Rb}_{2}$ molecules and show that the velocity distribution of the reflected molecules is sensitively dependent on the pulse durations of the optical fields. We have further studied how the optimal pulse duration is found as a function of the molecular beam velocity and show that it is, in general, larger than half a period of the simple harmonic motion. Moreover, we show that the reflectance of the pulsed standing wave mirror can be significantly improved by optimizing the pulse duration when compared to the simple harmonic model previously considered.

## APPENDIX A: DISTRIBUTION FUNCTION FOR UNTRAPPED MOTION $(N>1)$

Like the analysis of the trapped motion presented above, we introduce a function $\zeta(\tau)$ by

$$
\begin{equation*}
\sin [\zeta(\tau)]=\sin [\theta(\tau) / 2] \tag{A1}
\end{equation*}
$$

Differentiating both sides of Eq. (A1) and by using Eq. (16) we obtain

$$
\begin{equation*}
\frac{d \zeta(\tau)}{d \tau}= \pm N \sqrt{1-N^{-2} \sin ^{2}[\zeta(\tau)]} \tag{A2}
\end{equation*}
$$

which has the solution

$$
\begin{equation*}
\zeta(\tau)= \pm \operatorname{am}\left(N \tau+\phi, N^{-1}\right), \tag{A3}
\end{equation*}
$$

where $\phi$ is a constant. Inserting Eq. (A3) into Eq. (16), we obtain the velocity

$$
\begin{equation*}
\eta(\tau)= \pm 2 N \operatorname{dn}\left(N \tau+\phi, N^{-1}\right) \tag{A4}
\end{equation*}
$$

where $\operatorname{dn}\left(N \tau+\phi, N^{-1}\right)$ is a Jacobian elliptic tangent function $[40,41],+$ is for initial $\eta(0)>0$, and - for initial $\eta(0)<0$. Equation (A4) shows that an untrapped particle performs periodic motion whose period is given by

$$
\begin{equation*}
\widetilde{P}_{\text {untrap }}=\frac{2}{N} K\left(N^{-1}\right) \tag{A5}
\end{equation*}
$$

where $K\left(N^{-1}\right)$ is a complete Jacobian elliptic integral of the first kind $[40,41]$. Integrating Eq. (A4), the difference between the phase $\theta(\tau)$ and the initial phase $\theta(0)$ is determined by

$$
\begin{align*}
\theta(\tau)-\theta(0)= & \pm i \ln \left(\frac{\operatorname{cn}\left(N \tau+\phi, \frac{1}{N}\right)-i \operatorname{sn}\left(N \tau+\phi, \frac{1}{N}\right)}{\operatorname{cn}\left(\varphi, \frac{1}{N}\right)-i \operatorname{sn}\left(\varphi, \frac{1}{N}\right)}\right) \\
& \pm 2 m \pi \tag{A6}
\end{align*}
$$

with $i^{2}=-1, m$ is equal to the integer part of $\tau / \widetilde{P}_{\text {untrap }}$, and $\varphi=2 m K\left(N^{-1}\right)+\phi$. Equations (A4) and (A6) are solutions of Eqs. (11) and (12).

Now we begin to perform an inverse procedure to recover the initial states $\theta(0)$ and $\eta(0)$ from $\theta(\tau)$ and $\eta(\tau)$. From Eq. (A4), we obtain the initial velocity

$$
\begin{equation*}
\eta(0)= \pm 2 N \operatorname{dn}\left(\phi, N^{-1}\right) . \tag{A7}
\end{equation*}
$$

Using $\phi=N \tau+\phi-N \tau$ and the addition theory of elliptic functions, from Eq. (A7), we obtain the initial velocity

$$
\begin{equation*}
\eta(0)=2 N \frac{\operatorname{dn}\left(N \tau+\phi, \frac{1}{N}\right) \operatorname{dn}\left(N \tau, \frac{1}{N}\right)+\frac{1}{N} \operatorname{sn}\left(N \tau+\phi, \frac{1}{N}\right) \operatorname{sn}\left(N \tau, \frac{1}{N}\right) \operatorname{cn}\left(N \tau+\phi, \frac{1}{N}\right) \operatorname{cn}\left(N \tau, \frac{1}{N}\right)}{1-\left(\frac{\operatorname{sn}\left(N \tau+\phi, N^{-1}\right) \operatorname{sn}\left(N \tau, N^{-1}\right)}{N}\right)^{2}} \tag{A8}
\end{equation*}
$$

We next determine the relation of the $\phi$-dependent terms in Eq. (A8) with $\theta(\tau)$ and $\eta(\tau)$. By differentiating both sides of Eq. (A4) and using Eq. (12), we obtain

$$
\begin{equation*}
\sin [\theta(\tau)]= \pm 2 \operatorname{sn}\left(N \tau+\phi, N^{-1}\right) \operatorname{cn}\left(N \tau+\phi, N^{-1}\right) \tag{A9}
\end{equation*}
$$

Using Eqs. (A4) and (A9), from Eq. (A8) we obtain the initial velocity

$$
\begin{equation*}
\eta(0)=\frac{\eta(\tau) \operatorname{dn}\left(N \tau, \frac{1}{N}\right)+N^{-1} \sin [\theta(\tau)] \operatorname{sn}\left(N \tau, N^{-1}\right) \operatorname{cn}\left(N \tau, N^{-1}\right)}{\left[\operatorname{cn}\left(N \tau, N^{-1}\right)\right]^{2}+\left(\frac{\eta(\tau) \operatorname{sn}\left(N \tau, N^{-1}\right)}{2 N}\right)^{2}} . \tag{A10}
\end{equation*}
$$

Next we investigate $\theta(0)$ using Eq. (A6). Using the addition theory of Jacobian elliptical functions, $\mathrm{cn}\left(\varphi, N^{-1}\right)$ and $\operatorname{sn}\left(\varphi, N^{-1}\right)$ are determined by

$$
\begin{equation*}
\operatorname{sn}\left(\varphi, \frac{1}{N}\right)=\frac{\operatorname{sn}\left(N \tau+\phi, \frac{1}{N}\right) \operatorname{cn}\left(N \tau^{\prime}, \frac{1}{N}\right) \operatorname{dn}\left(N \tau^{\prime}, \frac{1}{N}\right)-\operatorname{cn}\left(N \tau+\phi, \frac{1}{N}\right) \operatorname{dn}\left(N \tau+\phi, \frac{1}{N}\right) \operatorname{sn}\left(N \tau^{\prime}, \frac{1}{N}\right)}{1-\left[N^{-1} \operatorname{sn}\left(N \tau+\phi, N^{-1}\right) \operatorname{sn}\left(N \tau^{\prime}, N^{-1}\right)\right]^{2}} \tag{A11}
\end{equation*}
$$

and

$$
\begin{equation*}
\operatorname{cn}\left(\varphi, \frac{1}{N}\right)=\frac{\operatorname{cn}\left(N \tau+\phi, \frac{1}{N}\right) \operatorname{cn}\left(N \tau^{\prime}, \frac{1}{N}\right)+\operatorname{sn}\left(N \tau+\phi, \frac{1}{N}\right) \operatorname{dn}\left(N \tau+\phi, \frac{1}{N}\right) \operatorname{sn}\left(N \tau^{\prime}, \frac{1}{N}\right) \operatorname{dn}\left(N \tau^{\prime}, \frac{1}{N}\right)}{1-\left[N^{-1} \operatorname{sn}\left(N \tau+\phi, N^{-1}\right) \operatorname{sn}\left(N \tau^{\prime}, N^{-1}\right)\right]^{2}}, \tag{A12}
\end{equation*}
$$

where $\tau^{\prime}=\tau-m \widetilde{P}_{\text {untrap }}$. From Eq. (A4), we have

$$
\begin{equation*}
\operatorname{dn}\left(N \tau+\phi, N^{-1}\right)= \pm \eta(\tau) /(2 N) \tag{A13}
\end{equation*}
$$

Using the relation

$$
\operatorname{sn}\left(N \tau+\phi, N^{-1}\right)=N \sqrt{1-\operatorname{dn}\left(N \tau+\phi, N^{-1}\right)^{2}}
$$

we have

$$
\begin{equation*}
\operatorname{sn}\left(N \tau+\phi, N^{-1}\right)=\sqrt{4 N^{2}-\eta(\tau)^{2}} / 2 . \tag{A14}
\end{equation*}
$$

By further inserting Eq. (A14) into Eq. (A9), we obtain

$$
\begin{equation*}
\operatorname{cn}\left(N \tau+\phi, N^{-1}\right)= \pm \sin [\theta(\tau)] /\left[2 N \operatorname{sn}\left(N \tau+\phi, N^{-1}\right)\right] . \tag{A15}
\end{equation*}
$$

By inserting Eqs. (A11)-(A15) into (A6), we can get $\theta(0)$, and inserting Eq. (A10) and Eq. (A6) into Eq. (14), we obtain $f(\theta(\tau), \eta(\tau), \tau)$.

## APPENDIX B: TIME REQUIRED FOR EACH DYNAMICAL REFLECTION PROCESS

The time required for the second process is half a period of the oscillation, i.e., $P_{\text {trap }}(N) / 2=2 K(N)$, where $P_{\text {trap }}(N)$ is defined in Eq. (22).

In Fig. 2, the time needed for molecules to travel from $F$ to $C$ is two times that from $F$ to $(0,2 N)$, which is the middle point of the trajectory from $F$ to $C$. By setting $\theta(\tau)=0$, $\eta(\tau)=2 N$, and $\eta(0)=\eta_{D}$ in Eq. (26) we obtain that the time for molecules to travel from $F$ to $(0,2 N)$ is $\mathrm{cn}^{-1}\left(\eta_{D} /(2 N), N\right)$. The time required for molecules to move from $C$ to $D$, corresponding to the first process, is the time for moving from $F$ to $D$ minus that from $F$ to $C$, given by $2\left[K(N)-\mathrm{cn}^{-1}\left(\eta_{D} /(2 N), N\right)\right]$. The time for the third process can be obtained in a similar manner.
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