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Deceleration of a continuous-wave (CW) molecular beam with a single quasi-CW semi-Gaussian laser beam*

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We propose a promising scheme to decelerate a CW molecular beam by using a red-detuned quasi-cw semi-Gaussian laser beam (SGB). We study the dynamical process of the deceleration for a CW deuterated ammonia (ND_3) molecular beam by Monte-Carlo simulation method. Our study shows that we can obtain a ND_3 molecular beam with a relative average kinetic energy loss of about 10% and a relative output molecular number of more than 90% by using a single quasi-cw SGB with a power of 1.5kW and a maximum optical well depth of 7.33mK.

Keywords: optical Stark deceleration, semi-Gaussian beam
PACC: 3380P, 4260H

1. Introduction

Much attention has been attracted to manipulate and control cold molecules in a gas phase for the past decade. Slow, cold molecules can be used for cold molecular spectroscopy and precision measurements,[1−3] studies of cold molecular collisions[4−6] and basic physics questions, quantum computing and information processing,[7] cold chemistry,[8] and so on. Until now, there have been two main approaches to generate slow and cold molecules. The first one is to prepare cold molecules from ultracold atoms via the photoassociation,[9−11] or Feshbach Resonances.[12−14] In this case, not all molecules are applicable as there are only a few species of laser-cooled atoms. The second method is to let fast and hot molecules be transformed to slow and cold molecules via the buffer gas cooling,[15] rotating the molecular jet,[16] decelerating polar (paramagnetic) supersonic molecular beam using the time-varying nonuniform electrostatic field[17,18] (time-varying nonuniform magnetostatic field,[19]) filtering out cold polar molecules from a thermal molecular ensemble by the bent quadrupole electrostatic field.[20,21] The inhomogeneous electrostatic field (magnetostatic field) can only be used to manipulate and control polar molecules (paramagnetic molecules) that have a permanent electric dipole moment (magnetic dipole moment). Recently, the methods of the cavity-assisted laser cooling of molecules have been proposed.[22−25] All molecules, no matter whether polar or paramagnetic, in a light field have a second-order Stark shift and their polarizability can interact with the light field, then they will experience a dipole interaction, which will exert a mechanical force on the molecule. A pulsed benzene (C_6H_6) supersonic beam has been slowed down using a single pulsed, focused Gaussian beam, and a relative reduction of ∼7.8% in the most probable velocity can be obtained.[26] A single laser pulse in a moving (or static) optical lattice can reduce the maximum kinetic energy of up to 50% for a pulsed NO (including C_6H_6 and H_2) supersonic beam.[27,28] These two schemes can only decelerate the pulsed molecular beam, and need a complex time-sequence control and synchronization system between the pulsed light field and the pulsed molecular beam. As far as we know, there has not been a promising scheme to decelerate a CW molecular beam until now. So it would be interesting and worthwhile to find a promising scheme to realize the Stark deceleration of

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a CW molecular beam.

In this paper, we propose a promising scheme to decelerate a CW molecular beam with a quasi-cw semi-Gaussian laser beam (SGB), and study the dynamical process of the deceleration of a CW deuterated ammonia (ND_3) molecular beam by Monte-Carlo simulations. In Section 2, we introduce the scheme to decelerate a CW molecular beam with a SGB. The phase space plot describing the motion of the molecules in the SGB and the deceleration results by Monte-Carlo simulations are given in Section 3. Some main results and conclusions are summarized in Section 4.

2. Deceleration scheme

A promising experimental setup that implements the deceleration of a cw molecular beam with a SGB is shown in Fig.1. The setup consists of two main parts: a quasi-cw SGB and a CW molecular beam. In the left part of Fig.1(c) is the experimental setup to generate a quasi-cw SGB that is used to decelerate the molecules. In the right part of Fig.1(c) is the incident molecular beam which is a preslowed, CW deuterated ammonia molecular beam, produced by the bent guiding of a buffer-gas-cooled effusive molecular beam in a quadrupole electrostatic guide.[29,30]

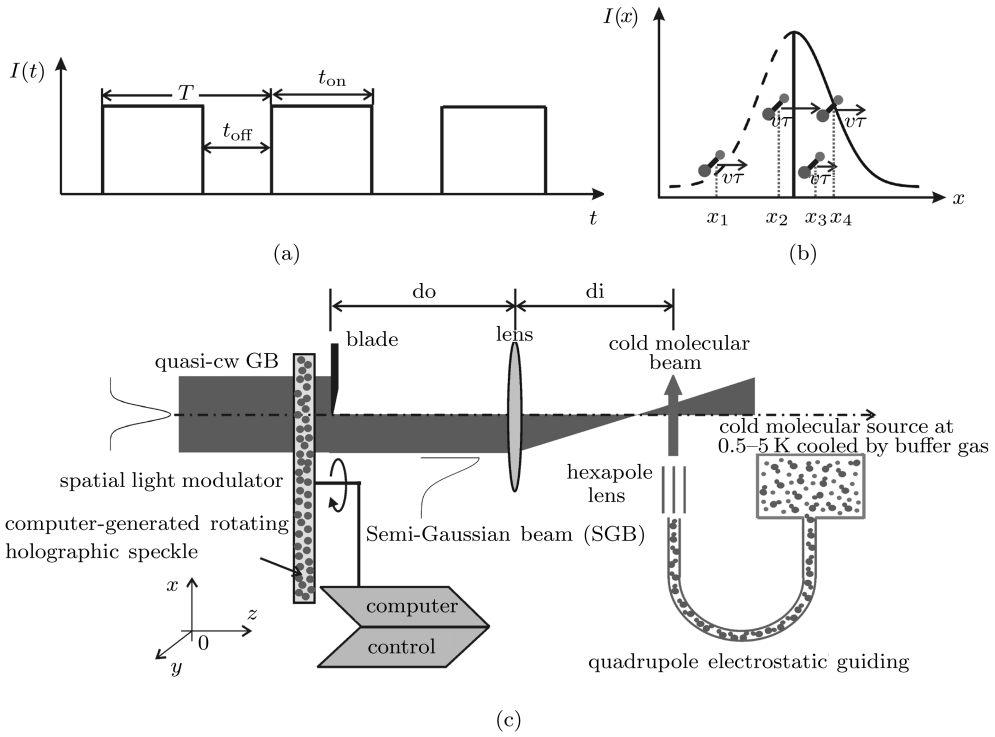


Fig.1. (a) The intensity distributions $I(t)$ of the incident Gaussian laser beam in the time domain. (b) Distributions of a quasi-cw semi-Gaussian beam in the spatial domains, which will be used to describe the basic deceleration principle of our SGB deceleration scheme. (c) Schematic diagram to generate a reduced SGB and form an optical Stark decelerator for a CW molecular beam.

A TEM_{00} CW Gaussian laser beam with a waist of 1.0 cm is propagating along the z-axis. An acousto-optic modulator (AOM)[31] is employed to modulate the CW laser to a quasi-cw one, and the modulated signal in the time domain is shown in Fig.1(a). We define its period as T, the light pulse duration (i.e. the switched-on time) as t_{on} and the switched-off time as t_{off}, and $T = t_{on} + t_{off}$. Then the above quasi-cw Gaussian beam with a waist of 10 mm is turned into a well-collimated quasi-cw Gaussian beam with a small waist of 1.0 mm by a telescope system. After that, the bottom straight edge of a thin and sharp blade is vertically inserted to the centre of the collimated Gaussian beam at the position $z = 0$, and the half of the Gaussian laser beam is blocked and a SGB with a horizontal and upward surface as well as some diffraction fringes is generated after the blade.[32] To obtain a neat and reduced SGB with a smaller waist of several 10-μm, a spatial light modulator (SLM), which is composed of the computer-generated rotating holo-

graphic speckle with a rotating frequency of several MHz and a piece of liquid crystal display (LCD), is put before the blade, which can decrease the coherence of the Gaussian laser.[33] Also, an imaging lens is placed at the propagation distance $z = d_{\rm o}$ and used to image for the straight edge of the blade, and finally a neat and reduced SGB will be produced near the image plane of the lens at $z = d_{\rm o} + d_{\rm i}$, here $d_{\rm o}$ and $d_{\rm i}$ are the object and image distances of the lens, respectively. In this case, the maximum well depth will be enhanced by M^2 times, where $M = d_{\rm o}/d_{\rm i}$.

Now let us introduce the basic deceleration principle of our SGB for a CW molecular beam. For the sake of simplicity, we choose four typical positions (or planes) $x = x_1$, x_2, x_3 and x_4 to analyse the molecular motion in the SGB light field, as shown in Fig.1(b). From ac Stark effect, the potential energy of a molecule moving in a red-detuned light field can be written as[26]

$$U(x, y) = -\alpha I(x, y)/(2\varepsilon_0 c), \tag{1}$$

where $I(x, y)$ is the SGB intensity profile, α is the average polarizability of the molecules, ε_0 is the dielectric constant in the vacuum, and c is the speed of light in vacuum. From $\boldsymbol{F}(x, y) = -\nabla U(x, y)$, Eq.(1) and Newtonian equations of motion, we can obtain the following motion equations of the molecules in the SGB field:

$$\begin{aligned} \frac{{\rm d}v_x}{{\rm d}t} &= -\frac{2\alpha I(x, y)x}{m\varepsilon_0 c w_{\rm SGB}^2}, \quad (x > 0); \\ \frac{{\rm d}v_y}{{\rm d}t} &= -\frac{2\alpha I(x, y)y}{m\varepsilon_0 c w_{\rm SGB}^2}. \end{aligned} \tag{2}$$

According to Eq.(2), as shown in Fig.1(b), when a red-detuned SGB pulse illuminates a CW incident molecular beam along the x direction, those molecules at the right of $x = 0$ (such as those in the x_3 or x_4 plane, etc.) will be decelerated because they experience a dipole force antiparallel to their moving direction. The molecules at the left of $x = 0$ and far away from the $x = 0$ plane (such as those in the x_1 plane, etc.) will not be disturbed by the SGB since they are not in the SGB field at $t = 0$ and cannot move into the SGB during $t_{\rm on}$. However, only less molecules at the left of $x = 0$ and near the $x = 0$ plane (such as those in the x_2 plane, etc.) will be accelerated. In this case, if a molecule moves from x_2 to x_2' during $t_{\rm on}$, its kinetic energy will be first increased by the maximum SGB well depth $-U(0)$, and then reduced by an optical Stark potential $U(x_2') - U(0)$, here $U(x)$ are given by Eq.(1). As a result, the molecule will obtain a net kinetic-energy increase of $-U(x_2')$. While those molecules at the right of $x = 0$ (such as those in the x_3 or x_4 plane, etc.) will be decelerated and will obtain a kinetic energy decrease of $U(x_3) - U(x_3')$ or $U(x_4) - U(x_4')$. Since the molecular-beam flux is a constant in the molecular propagation direction and the size of the acceleration region in the molecular-beam direction is far smaller than one of the deceleration region, the molecular number in the acceleration region is much less than one in the deceleration region, that is to say, the decelerated molecular number will be far greater than the accelerated one during a modulation period T when the semi-Gaussian beam comes. So the CW molecular beam, to sum up, will obtain a net deceleration. In addition, due to the focusing action of the transverse dipole attractive force from the red-detuned SGB, as a molecular lens,[34,35] then the output molecular beam will be focused and become a slow and thin molecular beam.

3. The phase space plot and Monte-Carlo simulation results

3.1. The phase space plot describing the motion of the molecules in the SGB

In this scheme, a 1.5 kW CW Ytterbium fibre laser with a wavelength of 1060–1080 nm and a beam quality of $M^2 < 1.1$,[36] which is far below the frequencies of all electronic transitions of ND_3 molecule, is used to generate a quasi-ideal reduced SGB with a waist radius of $w_{\rm SGB} = 20\,\mu$m, and the corresponding maximum well depth of SGB in the image plane of the lens is 7.33 mK. From Eq.(2), the velocity of the ND_3 molecules in the centre of the SGB light field (i.e. $y = 0$) can be described as

$$v_x = \sqrt{v_{x0}^2 + \alpha I(\exp(-2x^2/w_{\rm SGB}^2) - 1)/(m\varepsilon_0 c)}, \tag{3}$$

where v_{x0} is the initial velocity of the ND_3 molecule in the x direction and it fits to the functional form $(2v_x/v_{x\alpha}^2)\exp[-v_x^2/v_{x\alpha}^2]$, here $v_{x\alpha} = \sqrt{2k_{\rm B}T_l^i/m}$, m is the mass of a ND_3 molecule and $k_{\rm B}$ the Boltzmann constant, the longitudinal temperature of the precooled molecular beam $T_l^i = 10$ mK.[30]

The motion of the ND_3 molecules along the SGB axis in the x direction can be well described by the

phase space plot, which is calculated by using Eq.(3), as shown in Fig.2, showing energy contours of molecular motion in the SGB and are the trajectories of molecules in the phase space. The arrows indicate the direction of molecular motion along these energy contours. The thickest line shows the trajectories of the molecules with velocities of ± 2.47 m/s in the SGB, and as a boundary, it divides the phase space into two regions. Outside the enclosed region, the molecules are not trapped because their kinetic energies are higher than the maximum well depth of the SGB, but their velocities are significantly perturbed by the optical potential. Above the thickest line, most of the molecules (in the molecular beam) with higher kinetic energies will be decelerated by the red-detuned SGB and output from the SGB, which will form an output slow and cold molecular beam. Below the thickest line, the molecules with higher kinetic energies will be accelerated, but this case will not occur as the direction of the incident molecular beam is in the x direction in our scheme. Inside the enclosed region, some molecules with lower velocities are trapped in the optical well of the red-detuned SGB since their kinetic energy is smaller than the maximum well depth. It is interesting that the SGB optical well can only catches a small fraction of cold molecules from the molecular beam by choosing suitable parameters, and these cold molecules in the SGB can be collected and reused by using a suitable molecular guiding technique.

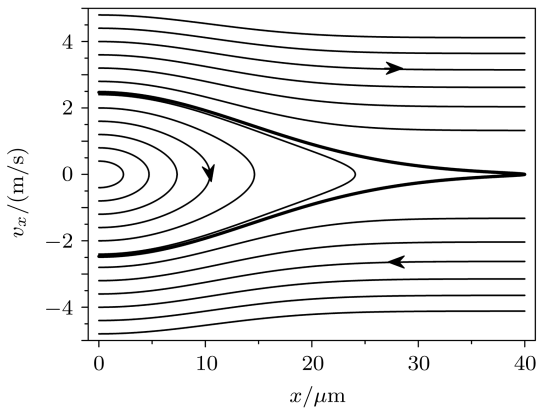


Fig.2. A phase space plot displaying the motion of ND_3 molecules within the SGB field. The thickest semi-closed line is the boundary that encloses the trajectories of molecules trapped in the SGB field. The arrows show the direction of the molecular motion along these lines.

3.2. Monte-Carlo simulation results

To demonstrate our optical Stark deceleration, we study the dynamic process of the slow CW ND_3 beam in the quasi-cw ideal SGB by using a classical Monte-Carlo simulation. In our simulation, we assume that neutral molecules in the different planes vertical to the x direction have the same velocity distributions, and the initial position of the molecular beam is at $x = 0$, and the molecules in the region of $0 < x < v_{\mathrm{m}}t_{\mathrm{on}}$ are in the SGB field as the SGB comes, where v_{m} is the mean velocity of the molecular beam. While the SGB is turned on at time t_{on}, part of the molecules in the region $-v_{\mathrm{m}}t_{\mathrm{on}} < x < 0$ can move into the SGB field and will be accelerated by the SGB during t_{on}. So during an AOM's modulation period T, the molecules in the region of $-v_{\mathrm{m}}t_{\mathrm{on}} < x < v_{\mathrm{m}}t_{\mathrm{on}}$ are disturbed. In the later periods, the above process will be repeated. In our simulation, we studied the motion of the molecules between the $x = -v_{\mathrm{m}}t_{\mathrm{on}}$ and $x = v_{\mathrm{m}}t_{\mathrm{on}}$ planes, and we divided this simulated range into forty planes.

By making the statistics of the moving kinetic energy of the Monte-Carlo simulated molecules, we find that the average kinetic energy of the decelerated molecules is decreased by 9.33% compared to that of the initial molecules, i.e. we have obtained a relative kinetic energy loss of $-\Delta\langle E_{\mathrm{k}}\rangle/\langle E_{\mathrm{k}}\rangle_i = 9.33\%$. In addition, our simulation result shows that about 95.15% of the molecules in the incident molecular beam has been decelerated and output from the SGB. The main molecular losses result from less transversely escaped molecules and those opposite-directional motional molecules, because their kinetic energies are lower than the well depth of the SGB, will be first decelerated to zero and then reflected back along the $-x$ direction by the optical dipole force. These results show that our scheme can be used to efficiently decelerate a CW molecular beam.

In order to find out the best deceleration effect, we study the influence of the t_{on} and T on the deceleration. We first make the switch-on time t_{on} unchanged and study the relation between the deceleration and the modulated period T. The results are shown in Fig.3 in the cases of $t_{\mathrm{on}} = \tau$ and $t_{\mathrm{on}} = 2\tau$, where $\tau = w_{\mathrm{SGB}}/v_{\mathrm{m}} = 7.82\,\mu$s is defined as the average time in which the molecular beam moves freely through the SGB region. The solid circles are the Monte-Carlo simulated results and the solid lines are the fitted curves. It is clear from Fig.3 that the relative average kinetic-energy loss first increases with increasing modulated period T and reaches the maximum, and then decreases. The fitting line shapes in the two cases are nearly the same, but their maxima and the values of T are different. For $t_{\mathrm{on}} = \tau$, the maximum relative av-

erage kinetic-energy loss is $-\Delta\langle E_{\rm k}\rangle/\langle E_{\rm k}\rangle_i = 9.33\%$ at $T = 2\tau$ (i.e. $t_{\rm off} = t_{\rm on}$), while for $t_{\rm on} = 2\tau$ the maximum relative average kinetic energy loss is $-\Delta\langle E_{\rm k}\rangle/\langle E_{\rm k}\rangle_i = 7.24\%$ at $T = 3.2\tau$.

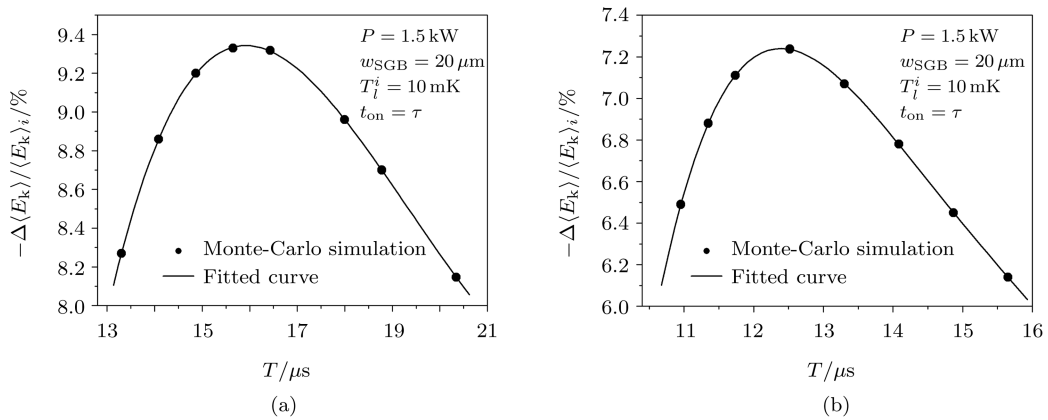


Fig.3. The dependence of the relative average kinetic-energy loss of the molecular beam for the modulated period T in the cases of (a) $t_{\rm on} = \tau$ and (b) $t_{\rm on} = 2\tau$. The solid circles are the Monte-Carlo simulated results, while the solid lines are the fitted curves.

Then we choose the optimal-modulated period T when we change the laser switch-on time $t_{\rm on}$, and study the relation between the deceleration and the switch-on time $t_{\rm on}$. The results are shown in Fig.4, where the solid circles, which represent the maximum relative loss of average molecular kinetic energy in the case of different $t_{\rm on}$, are the Monte-Carlo simulated results; while the solid line is the fitted curve. Obviously, with the increase of the laser switch-on time $t_{\rm on}$, the relative loss of the average kinetic energy of the molecular beam first increases and then decreases.

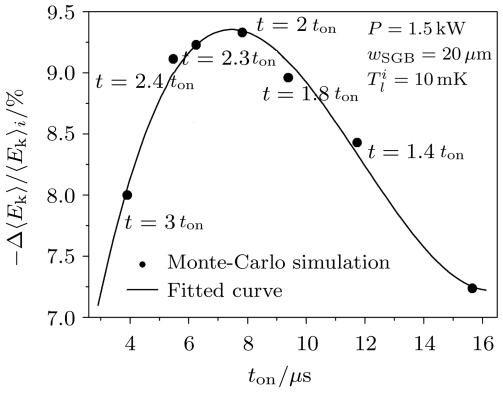


Fig.4. The dependence of the relative average kinetic-energy loss of the molecular beam on the pulse duration $t_{\rm on}$ for the case of different optimal modulated periods T. The solid circles are the Monte-Carlo simulated results, while the solid line is the fitted curve.

This shows that there is an optimal laser switch-on time $t_{\rm on}$ where the loss of the molecular kinetic energy arrives at the maximum value. The maximum kinetic-energy loss is about 9.33% for $t_{\rm on} = \tau$ and period $T = 2t_{\rm on}$. In this case, the modulated signal of AOM is a standard equal-periodic square wave, which is very easy to implement in the experiment. This result can be explained as follows: during the optimal laser switch-on time $t_{\rm on}$, most of the investigated molecules can pass through the whole SGB, so we can obtain the largest accumulation of the optical potential and the largest kinetic energy loss.

4. Summary

We have proposed a promising scheme to decelerate a CW molecular beam by using a single quasi-cw red-detuned SGB. We have used the Monte-Carlo method to simulate the dynamic process of a CW ND_3 molecular beam in the SGB field, and also studied the dependence of the deceleration on the modulated period T and the pulse duration $t_{\rm on}$. Our study shows that the proposed deceleration scheme can efficiently decelerate a CW ND_3 molecular beam and obtain a slower and slimmer ND_3 molecular beam with a relative average kinetic energy loss of about 10% and a relative output molecular number of more than 90% by using a single quasi-cw SGB with a power of 1.5 kW when the modulated signal is a standard equal-period square wave, i.e. the modulated period $T = 2t_{\rm on}$, and the switch-on time $t_{\rm on} = \tau$. Our optical Stark decelerator can be used not only to slow a CW polar, as well as non-polar or paramagnetic, molecular beam, but also to decelerate a cw atomic and cluster beam, even to slow an arbitrary pulsed molecular beam. Also, our SGB can be used not only to form various atom and

molecule optical elements, such as atomic or molecular lens, atomic or molecular mirror,[37] atomic or molecular grating and so on, but also to decelerate (or accelerate) some basic particles, such as electrons, plasma, etc. All these applications will be included in our future research.

In comparison with a pulsed electrostatic or pulsed optical Stark decelerator, our scheme has some new and unique advantages as follows: (1) Under the same flux of molecular beam, the obtainable number of cold molecules per second from a CW molecular beam is far higher than that from a pulsed molecular one. (2) Our decelerator does not need a complex time-sequence control and synchronization system in the experiment, and more than 90% of the decelerated molecules can be output within a small divergent angle due to the transverse self focusing-effect of the red-detuned SGB. (3) Our scheme, in principle, does not have any decelerating limitation. This shows that our Stark deceleration scheme might be able to decelerate a CW molecular beam from a few 10-mK (even $\sim 50\,\mathrm{K}$ in a high fineness cavity) to an ultracold region by using a cavity-enhanced technique and an optical or electrostatic storage ring.[38] In addition, our scheme is the simplest, more convenient and efficient. So it has some important applications in the fields of cold molecular physics, molecular optics, cold molecular spectroscopy[3,39] and cold chemistry,[8] even the realization of all-optical, chemically-stable molecular Bose–Einstein condensation in an optical dipole trap by using optical-potential evaporative cooling,[40] and so on.

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