Deceleration of molecules by dipole force potential: a numerical simulation

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New Journal of Physics 11 (2009) 055023 (16pp)
Received 2 December 2008
Published 14 May 2009
Online at http://www.njp.org/
doi:10.1088/1367-2630/11/5/055023

Abstract. We propose a new method for making ultracold molecules below 300 µK by cooling via the dipole force of an intense infrared (IR) standing wave created in a high finesse cavity. Deceleration can be achieved by periodically switching an IR standing wave slightly red-detuned from the resonance frequency of a molecular vibration–rotation transition. The potential depth becomes as high as 50 mK with an incident laser power of 3 W. By properly switching the potential, one can remove the translational energy of molecules from a couple of hundred mK to 300 µK or below. Since all molecules except homonuclear diatomic molecules have one or more optical transitions in the IR range that can be used for a source of the dipole force, the present method is applicable to almost all molecules of chemical interest. Moreover, by choosing an appropriate rotation–vibration transition, one can decelerate and trap molecules in the completely ground state in any degrees of freedom. Numerical simulations showed sufficiently wide phase-space areas of decelerated molecules through use of this technique for practical applications. The effect of various parameters on the efficiency of the cavity decelerator has been examined by numerical simulations.

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1. Introduction

Recent interest in quantum phenomena observed in ultracold atoms has been expanding to ultracold molecules in the last few years [1]. It is now widely believed that ultracold molecules below 1 mK may offer completely new possibilities beyond those offered by cold atoms to study a wide range of fundamental and practical problems in both physics and chemistry. For example, non-uniform internal charge distribution in molecules may enhance the sensitivity of measurements of the electric dipole moment (EDM) of the electron [2, 3], the existence of permanent dipole moments makes molecular Bose–Einstein condensates (BECs) different from those of atoms [4], and the molecular matter wave may lead to new chemistry at low temperatures [5] or ‘superchemistry’ [6].

Despite their importance, it is still difficult to create ultracold molecules. Currently, two different approaches are being employed to create ultracold molecules. One of these approaches is the production from laser-cooled ultracold atoms and the other is the direct cooling of room temperature molecules.

Magnetic Feshbach resonance [7] and photoassociation [8] have been applied to various atomic systems to create ultracold diatomic molecules [9]–[11]. Molecules thus produced are in a highly vibrationally excited state, and a couple of techniques have been successfully applied to bring them to the ground state [12]–[14]. The limitation of these techniques, however, is that they are only applicable to diatomic (or triatomic etc) molecules of alkali and rare earth metal atoms.

In order to study the field of ultracold chemistry, direct deceleration of molecules from room temperature is indispensable. Various techniques have been proposed and tested for slowing room temperature molecules. These techniques include buffer gas cooling [15], a molecular beam from a rotating nozzle [16], a ‘billiard-like’ collision with another atomic beam [17], a velocity filter by Stark potential [18]–[20], electrostatic Stark deceleration [21, 22] and magnetic field Zeeman deceleration [23, 24]. In addition, a technique to decelerate molecules by optical forces has been examined [25, 26], in which a far-off resonant dipole
force created in a slowly traveling standing wave was used to decelerate molecules. All of these
methods are useful for making cold molecules of chemical interest, but the lowest temperature
is still limited to 50–500 mK. The development of a new method to create molecules below
1 mK is eagerly desired in order to study molecule characteristics at ultralow temperature.

For a precise control of molecular velocity to achieve temperatures below 1 mK, optical
manipulation must be employed. Unfortunately, laser cooling techniques cannot be applied
directly to molecules due to the existence of many vibrational and rotational energy levels in the
frequency range from microwave to ultraviolet (UV) [27]. Far-off resonant force traps (FORT)
have been widely used for the trapping of ultracold atoms [28], but they may not be effective for
polyatomic molecules because of the existence of numerous rotation and vibration states at any
wavelength from microwave to UV. Dipole forces of nearly detuned radiation would be better
to provide sufficiently large and well-controlled force on molecules.

Here, we discuss a new technique for making ultracold molecules below 300 µK using a
dipole force driven by the standing wave of an intense infrared (IR) field created in an optical
cavity. Infrared radiation slightly detuned from the resonance frequency of a vibration–rotation
transition of molecules provides a dipole force to molecules, which can be used for deceleration.
One of the advantages of the proposed method over other decelarators is that one can decelerate
molecules in the ground rotational state by choosing an appropriate rotation–vibration transition
for the dipole force. The method is a modification of a scheme we have proposed previously for
the deceleration of polar molecules by using a microwave standing wave in a cavity [29]. The
MW decelerator has the advantage of larger spatial confinement of molecules because of the
large diameter of beam waist. The advantage of the IR decelerator is that it is applicable to a
broad range of molecules because any type of molecule, with the exception of homonuclear
diatomic molecules, has one or more infrared transitions available for the deceleration. In this
paper, we examine the efficiency of the IR decelerator by changing various parameters of the
standing wave in a cavity.

2. Dipole force in a cavity

A dipole force originates from the interaction between an oscillating electric field and an
induced EDM. In an optical cavity, the intensity of an electric field changes periodically along
the cavity axis. The dipole force potential of a standing wave in a cavity is given by [30, 31]

\[ U_{\text{dip}} = \frac{1}{2} \hbar \delta \ln \left[ 1 + s \cos^2 \left( \frac{\phi + \frac{\pi}{2}}{2} \right) \right] \]
\[ = \frac{1}{2} \hbar \delta \ln \left[ 1 + \frac{1}{2} s (1 - \sin \phi) \right] \]

where \( \delta = \omega_L - \omega_0 \) is the detuning of laser frequency \( \omega_L \) from a resonance frequency \( \omega_0 \), and
\( s \) is the saturation parameter defined as \( s = 2 \Omega_R^2 / [\Gamma^2 + 4 \delta^2] \) with \( \Omega_R = \mu E / \hbar \) being the Rabi
frequency defined by the transition dipole moment \( \mu \) and the amplitude of the light field \( E \) at an
antinode of the standing wave, and \( \tau = 1 / \Gamma \) being the natural lifetime of the excited state. The
phase \( \phi \) corresponds to the position of a molecule along a standing wave. We define the phase
\( \phi \) as \( \phi = (4n + 1) \pi / 2 \) at nodes and \( \phi = (4n - 1) \pi / 2 \) at anti-nodes of the standing wave with \( n \)
being an integer as shown in figure 1. The dipole force potential in a cavity provides a periodic
potential with a period of \( \lambda / 2 \) where \( \lambda \) is the wavelength of the laser field. In the following, we
call the length \( \lambda / 2 \) one stage.
Figure 1. Principle of the cavity decelerator. (a) A standing wave inside a cavity produces a dipole force potential on molecules traveling along the cavity axis. The definitions of the phase $\phi$, and the position of a molecule $\Phi(t)$ at time $t$ are shown. The length of one stage of the potential is $\lambda/2$. (b) A modulation of the intensity $f(\Phi)$ in a cavity for the case of $N = 3$. The switching phase $\phi_0$ is shown. The period of the switching becomes $2N\pi$ ($= 6\pi$ for $N = 3$). (c) The effective potential for $N = 3$. (d) Associated dipole force for $N = 3$. (e) The change of the kinetic energy for $N = 3$. The effective work per period of the modulation is given as $W$.

The dipole force potential changes its sign with the detuning $\delta$. In the case of a near-resonant red-detuned field ($\delta < 0$), the antinodes of the standing wave become the minima of the potential with $U_{\text{dip}} = \frac{1}{2}\hbar\delta \ln [1 + s]$, while the nodes become the maxima of the potential with $U_{\text{dip}} = 0$ (see figure 1(a)). The dipole force potential becomes maximum at $\Omega_R = 2.8|\delta|$ when $\Gamma$ is significantly smaller than $\Omega_R$ and $\delta$, in which the saturation parameter is approximated as $s = \frac{1}{2}(\Omega_R/\delta)^2$.

The associated dipole force is obtained by taking a spatial derivative of the potential with respect to $\phi$ as

$$F_{\text{dip}} = -\frac{dU_{\text{dip}}}{d\phi} = \hbar\delta \frac{s \cos \phi}{4 + 2(s - \sin \phi)}.$$  (2)

For an order estimation, we have considered methane (CH$_4$) as an example. Methane has an infrared transition at 3.3 $\mu$m, which is the $\nu_3$ triply degenerate stretching mode. Frequency
Table 1. The resonance wavelength ($\lambda$), transition dipole moment ($\mu$), maximum potential depth ($A_{\text{max}}$), number of stages per period ($N$), and effective work ($W$) for various molecules. A standing wave with build-up power of 1 kW and a beam diameter of 1 mm was assumed.

| Species | $\lambda$ ($\mu$m) | $\mu$ (D) | $A_{\text{max}}$ (mK) | $N$ | $W$ (mK) |
|---------|------------------|----------|---------------------|-----|----------|
| OH      | 2.8              | 0.002$^c$| 0.8                 | 11  | 0.05     |
| NO      | 5.3              | 0.050$^c$| 20                  | 5   | 2.6      |
| CO      | 4.7              | 0.062$^c$| 25                  | 5   | 3.2      |
| NH$_3$  | 6.1              | 0.048$^c$| 20                  | 5   | 2.5      |
| CH$_4$  | 3.3              | 0.053$^{c,d}$ | 22                  | 9   | 1.6      |

$^a$The number of stages necessary for a molecule at 0.4 K, and a modulation frequency of 1 MHz.

$^b$ A linear modulation with $\phi_0 = -\pi$ was assumed.

$^c$ Rao [32].

$^{d}$ Fox [33].

stable IR radiation with a few watts of power is now commercially available in the wavelength range between 2 and 5 $\mu$m based on the optical parametric oscillation (OPO) of a periodically poled lithium niobate (PPLN) crystal in a ring-type resonator pumped by a fiber-amplified 1 $\mu$m fiber laser. High-finesse cavity mirrors of $R > 99.99\%$ in the wavelength range between 2 and 5 $\mu$m are also commercially available as cavity-ringdown mirrors. With a high finesse cavity of $R = 99.9\%$, for example, the IR power can be built up inside the cavity on the order of 1 kW creating an intense standing wave with a period of 1.65 $\mu$m. The electric field of such a standing wave is $E = 1.4 \times 10^6$ V m$^{-1}$ with a beam diameter of 1 mm. Since the transition dipole moment of the $v_3$ methane transition has been reported as $\mu = 0.053$ debye [32, 33], the Rabi frequency becomes $\Omega_R = 2.5 \times 10^9$ rad Hz, which is orders of magnitude faster than the lifetime of the $v_3$ excited state of $\tau = 40$ ms ($1/\tau = 25$ rad Hz). When we detune the infrared frequency by $-140$ MHz ($\delta = -9.0 \times 10^8$ rad Hz) to the resonant frequency, the standing wave in the cavity provides a periodic dipole force potential on the methane with a depth of 22 mK. In general, the depth of the dipole force potential becomes as high as 10–50 mK, which is the basis of the present method. The dipole force potential itself can be used as a linear optical trap for cold molecules, but we use it to decelerate molecules as well by actively switching the potential as described below.

Table 1 summarizes the dipole force potential and efficiency of the IR cavity decelerator for several molecules of interest.

3. Switching of potential

When a molecule travels from a local bottom of the dipole potential (an antinode for red-detuned radiation) to a neighboring top of the potential (a node), the molecule’s potential energy increases by $\frac{1}{2} \hbar \delta \ln [1 + s]$, which in turn causes the molecule’s kinetic energy to drop by $\frac{1}{2} \hbar \delta \ln [1 + s]$, resulting in the deceleration of the molecule. On the other hand, when a molecule travels from the top to the bottom of the potential, the molecule’s potential energy decreases by the same amount, which results in acceleration. Thus, there is no change of average velocity if molecules just travel along a continuous standing wave.
When proper switching of the potential is applied with respect to the motion of molecules, deceleration (or acceleration) of molecules can be realized. If the potential is switched on and off instantaneously when a molecule arrives at the bottom and at the top of the potential, respectively, the molecule loses its kinetic energy by $\frac{1}{2} \hbar \delta \ln [1 + s]$ along the standing wave at every stage. Therefore, when the depth of the dipole force potential is 50 mK, for example, and the length of the one stage is 1.5 $\mu$m, one can stop room temperature molecules (300 K) within only 1 cm! This is possible only if one can switch the potential at a frequency of 1 THz or faster. Such a fast switching of the laser field has not been realized up to now. Therefore, one has to take into account the effect of slow switching of the potential for a realistic decelerator.

Switching of laser radiation at a rate of 1 MHz is available by using an acoustooptic modulator (AOM). Due to the cavity lifetime, however, the intensity inside an optical high-finesse cavity may not follow the switching of external laser intensity. A confocal cavity with reflectivity $R$ and separation $\ell$ has a cavity lifetime of $-\ell/(c \ln R)$, where $c$ is the speed of light [34]. For example, the reflectivity of $R = 99.98\%$ and a separation of $\ell = 10$ cm gives a cavity lifetime of about 1.7 $\mu$s. In such a cavity, it takes 7.8 $\mu$s to build power up to 99% of the maximum power. Therefore, the power inside the cavity never reaches the maximum build-up power when the external laser is switched at 1 MHz.

When the external radiation is switched on and off at a rate of $f_s$ Hz with a duty cycle of 50%, the intensity inside a cavity with a cavity lifetime of $\tau_c$ increases as $I_{\text{cav}} - (I_{\text{cav}} - I_0) \exp[-(t - t_0)/\tau_c]$ for $t_0 \leq t < t_0 + 1/(2 f_s)$, and decreases as $I_0 \exp[-(t - t_0)/\tau_c]$ for $t_0 + 1/(2 f_s) \leq t < t_0 + 1/f_s$ in one period. Here, $I_{\text{cav}}$ is the maximum build-up intensity in the cavity, $I_0$ is the intensity of the standing wave when the electric field is switched on at $t = t_0$ and $I_0 = I_{\text{cav}} - (I_{\text{cav}} - I_0) \exp[-a_{cs}]$ with $a_{cs} = 1/(2 f_s \tau_c)$. When the radiation is switched on and off for a sufficient period of time, the modulation inside the cavity reaches equilibrium with $I_0 = I_{\text{cav}} \exp(-a_{cs})/[1 + \exp(-a_{cs})]$ and $I_0 = I_{\text{cav}}/[1 + \exp(-a_{cs})]$. When the switching rate $f_s$ is twice the value of the inverse of the cavity lifetime $1/\tau_c$, for example, the potential inside the cavity modulates between 0.44 $\times$ $I_{\text{cav}}$ and 0.56 $\times$ $I_{\text{cav}}$, and therefore the effective potential depth becomes only 10% of the maximum depth. An important point, however, is that even if the switching is faster than the cavity lifetime, there is a modulation of intensity inside the cavity that can be used for the deceleration of molecules.

Since molecules faster than 1.5 m s$^{-1}$ (corresponding to 4.5 mK for CH$_4$) travel more than one stage in 1 $\mu$s in a potential with $\lambda = 3$ $\mu$m, we have to consider the effect of multi-stage deceleration, in which a molecule travels more than one stage before the potential switches. The timing of the switching has to be controlled relative to the position of a molecule, $\Phi(t)$, at a time $t$ along the axis of the standing wave for efficient deceleration. In the following we limit our discussion to the case where the standing wave is switched every time a target molecule arrives at $\Phi(t) = \phi_0 + n N \pi$. Here the switching phase, $\phi_0$, has a constant value throughout the deceleration, $N$ is an integer corresponding to the number of stages, and $n$ is an integer with $n = \text{odd}$ being switched on and $n = \text{even}$ switched off. The case of $N = 3$ is depicted in figure 1(c). Hereafter, we call the length of $N\lambda/2 = 2N \pi$ one period. We call the target molecule a synchronous molecule.

The dipole force potential given in equation (1) may be approximated as $U_{\text{dip}} = (A_{\text{eff}}/2) (\sin \phi - 1)$, where $A_{\text{eff}}$ is the effective depth of the potential. When the potential varies as $f(\Phi)$ with respect to the position of the synchronous molecule $\Phi(t)$, the effective force provided by such a modulated potential to the synchronous molecule is given by

$$F(\Phi) = -(dU_{\text{dip}}/d\phi)|_{\phi=\Phi} \times f(\Phi) = -(A_{\text{eff}}/2) f(\Phi) \cos \Phi.$$ (3)

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Here, we discuss two simple cases. One is the modulation with a sinusoidal form, and the other is a simple triangle type linear modulation of the potential. When a modulation is given by

\[ f(\Phi) = \frac{1}{2} \cos((\Phi(t) - \phi_0)/N) + 1, \]

which is a simple sinusoidal modulation, the effective work per period is obtained as

\[ W = \int_{\phi_0}^{\phi_0 + 2N\pi} F(\Phi) d\Phi = 0 \]

for any \( N \) and any \( \phi_0 \). Therefore, the work becomes zero after one period for the sinusoidal modulation. Since the work \( W \) must be equal to the change of the kinetic energy of a molecule, there is no change of velocity due to this modulation.

When a modulation is given by

\[ f(\Phi) = (\Phi - \phi_0)/N\pi \]

for \( \phi_0 < \Phi \leq \phi_0 + N\pi \), and

\[ f(\Phi) = (\Phi_0 + 2N\pi - \Phi)/N\pi \]

for \( \phi_0 + N\pi < \Phi \leq \phi_0 + 2N\pi \), which is a simple triangle type linear modulation of the potential, the situation becomes different. The effective work per period for such a modulation is obtained as

\[ W = \int_{\phi_0}^{\phi_0 + 2N\pi} F(\Phi) d\Phi = \begin{cases} 0, & \text{for } N = \text{even}, \\ 2A_{\text{eff}} \frac{N}{N\pi} \cos(\phi_0), & \text{for } N = \text{odd}. \end{cases} \]

In this case, deceleration \( (W < 0) \) can be achieved when the number of stages \( N \) is odd with \( \cos(\phi_0) < 0 \). In addition, it is seen that the larger \( N \) gives less efficient deceleration because of the \( 1/N \) factor. The above examples clearly show that the deceleration of molecules can be achieved by a proper switching of the dipole force potential inside a cavity.

4. Simulation

The dipole force given in equation (2) contains the saturation parameter \( s \) in the denominator. Since the saturation parameter depends on the intensity of the electric field, the effective force is no longer a simple form as in equation (3) when the field intensity varies with time. Therefore, we have carried out numerical simulations to investigate the efficiency of the infrared decelerator. We have solved the equation of motion of molecules under a dipole force by the fourth-order Runge–Kutta method.

We chose methane (CH\(_4\)) as the sample molecule. In the simulation, an IR field was red-detuned to the resonance frequency of the \( R(0) \) transition of the \( v_3 \) triply degenerate stretching mode at 3028.763 cm\(^{-1}\). The lifetime of the \( v_3 \) excited state of methane is \( \tau = 40 \) ms, which can be calculated from the transition dipole moment of \( \mu = 0.053 \) debye [32, 33]. Since all simulations in the following showed that the deceleration of molecules in a cavity finishes within a time period of 10 ms or less, we ignored the effect of spontaneous emission and the associated heating effect. In general, the vibrational excited state of molecules has a lifetime of 100 ms or longer [32], these assumptions must hold for most molecules, which is an advantage of using a rotation–vibration transition of molecules for deceleration.

The cavity we used in the simulation consists of two spherical mirrors with a curvature of 1 m and a reflectivity of 99.986\%. With a mirror separation of 0.1 m, the beam waist diameter of the stable cavity mode is \( \sim 1 \) mm [34] with the difference of the IR beam diameter at the center and at the mirror positions being 3\%. Therefore, the depth of the periodic dipole potential was treated as constant everywhere in the cavity along the cavity axis. We consider the lowest mode of a cavity (TEM\(_{00}\)), which gives a monopolar standing wave along the axis.

There are six parameters we can adjust to optimize the deceleration efficiency. They are (i) the cavity length \( \ell \), (ii) the number of stages \( N \) in one period, (iii) switching phase \( \phi_0 \), (iv) switching frequency \( f_s \), (v) initial velocity \( v_0 \) and (vi) incident laser power \( P_0 \). In the following, we discuss the effects of each parameter on the efficiency of the decelerator in detail.
Table 2. Efficiency of the deceleration of a synchronous molecule in a cavity with various cavity lengths $\ell$. The beam waist radius $w_c$, cavity lifetime $\tau_c$, the maximum potential depth $A_{\text{max}}$, the effective potential depth $A_{\text{eff}}$, and the maximum initial velocity $v_{\text{max}}$ to be decelerated within the given cavity length $\ell$ are shown. The effective potential depth is calculated at the switching frequency $f_s$ of 1 MHz. See text for details of other conditions. The ratio $F$ between the travel distance and the cavity length for a molecule with the initial velocity of $v_0 = 14.0 \text{ m s}^{-1}$ is also shown.

| $\ell$ (m) | $w_c$ (mm) | $\tau_c$ ($\mu$s) | $A_{\text{max}}$ (mK) | $A_{\text{eff}}$ (mK) | $v_{\text{max}}$ (m s$^{-1}$) | $F$  |
|-----------|-----------|------------------|----------------------|----------------------|----------------------|-----|
| 0.01      | 0.27      | 0.24             | 112                  | 87                   | 21.5                 | 0.92|
| 0.05      | 0.40      | 1.19             | 82                   | 17                   | 18.2                 | 0.64|
| 0.10      | 0.48      | 2.38             | 70                   | 7.4                  | 18.2                 | 0.75|
| 0.20      | 0.56      | 4.77             | 59                   | 3.1                  | 18.2                 | 0.89|
| 0.50      | 0.67      | 11.91            | 48                   | 0.98                 | 14.9                 | 0.49|

Note: $w_c = \left(\frac{\lambda}{2\pi}\right)^{1/2}(2\ell R - \ell^2)^{1/4}$, $\tau_c = -\frac{\ell}{2ln R}$, $A_{\text{max}} = \hbar \Omega R \times 0.6$ when the detuning $\delta$ is adjusted to give the maximum potential depth [30] and $A_{\text{eff}} = \frac{1 - \exp(-a_{cs})}{1 + \exp(-a_{cs})} \times A_{\text{max}}$.

4.1. Cavity length $\ell$

The cavity lifetime depends linearly on the distance $\ell$ between the mirrors. The effective potential relative to the maximum build-up power is proportional to $[1 - \exp(-a_{cs})]/[1 + \exp(-a_{cs})]$ at the equilibrium, with the factor $a_{cs}$ being inversely proportional to the length $\ell$. By ignoring the difference of the beam waist radius for different $\ell$, we found that the ratio between the travel length of a synchronous molecule until stopping versus the cavity length $\ell$ becomes the smallest when $a_{cs} = 1.28$. A cavity with a smaller ratio can be considered as a more efficient one in terms of the deceleration. At the switching frequency of $f_s = 1$ MHz, the ratio becomes smallest with the cavity lifetime $\tau_c = 0.4 \mu s$. With a cavity reflectivity of $R = 0.99986$, the distance between mirrors of $\ell = 1.6$ cm gives the smallest ratio.

Since the beam waist radius is a function of the distance $\ell$, the best condition may be slightly different. Table 2 shows the beam waist radius $w_c$, cavity lifetime $\tau_c$, the maximum potential depth $A_{\text{max}}$, the effective potential depth $A_{\text{eff}}$, and the maximum initial velocity $v_{\text{max}}$ of a synchronous CH$_4$ molecule to be decelerated within the given cavity length $\ell$. An incident laser power of 3 W and a reflectivity of the cavity mirrors of $R = 0.99986$ were assumed. The initial switching frequency was set to as close as possible to 1 MHz to derive the maximum initial velocity. The effective potential depth is larger for shorter cavity length. Molecules with a velocity of 21.5 m s$^{-1}$ can be decelerated with a 1 cm cavity, while 18.2 m s$^{-1}$ is the maximum for a cavity of $\ell = 5$–20 cm. Speeds of 18.2 m s$^{-1}$ correspond to a translational temperature of 660 mK. This analysis shows that the precooling of molecules down to 500 mK is prerequisite for the cavity decelerator.

The value $F$ in table 2 shows the ratio between the travel length of a decelerated synchronous molecule versus the cavity length. Since molecules may distribute everywhere within a cavity before the deceleration, the smaller ratio must be better in order to have a larger number of decelerated molecules inside the cavity. The ratio also shows shorter cavity length.
Figure 2. The final velocity of synchronous molecules for different numbers of stages \(N\) and different values of the switching phase \(\phi_0\). The initial velocity of the synchronous molecules was set to \(v_0 = 14.0 \text{ m s}^{-1}\) for all \(N\), which is shown by the dotted line in the figure.

is better. A cavity length of 5–10 cm may be practically the best in terms of easier loadings of molecules into the cavity. In the following, we assume a cavity length of 10 cm and a reflectivity of \(R = 0.99986\).

4.2. The number of stages \(N\)

Figure 2 shows the final velocity of synchronous molecules as functions of the number of stages \(N\) in one period and the switching phase \(\phi_0\). The initial velocity of molecules was set to \(v_0 = 14.0 \text{ m s}^{-1}\) for all \(N\). The potential was switched every time when the synchronous molecule arrived at the position corresponding to the switching phase \(\phi_0\) after traveling \(N\) stages. Incident laser power of \(P_0 = 3 \text{ W}\) was assumed for the simulation.

It is seen that for \(N = \text{odd integers}\), synchronous molecules can be decelerated at a switching phase of \(-\pi/2 < \phi_0 < \pi/2\). The behavior is similar to that of the simple linear triangle modulation of the potential discussed in section 3. Figure 2 also shows that some deceleration may be achieved for even \(N\) around the switching phase of \(\phi_0 \sim \pi\), but the efficiency is much less than for the case of odd \(N\). Figure 2 clearly shows that the number of stages \(N\) has to be odd for an efficient deceleration.

4.3. Switching phase \(\phi_0\) and phase space stability

So far, we have discussed the motion of synchronous molecules in table 2 and figure 2. From this section, we discuss motion of other molecules in a cavity. When molecules are distributed continuously along the standing wave at the beginning, only molecules at a proper position relative to the dipole force potential can be decelerated. This problem has been discussed in detail as a ‘phase stability requirement’ [21, 35]. For the longitudinal motion along the cavity axis (\(z\)-axis), the ‘phase stability requirement’ limits the number of molecules to be decelerated in the \(z-v_z\) phase-space area. All adjustable parameters have to be optimized in order to obtain as a large phase-space area as possible. Here, we first discuss the phase space stability as a function of the switching phase \(\phi_0\).

Figure 3 shows the final velocity distribution for different values of the switching phase \(\phi_0\). The initial velocity was set around \(v_z = 14.0 \text{ m s}^{-1}\), corresponding to the temperature of
Figure 3. (a) The phase stability for various switching phases $\phi_0$. (b) Final velocity distribution of decelerated molecules for each switching phase $\phi_0$. All velocities are those with respect to the cavity, or the laboratory frame.

0.381 K. An incident laser power of $P_0 = 3$ W and a switching frequency of 1 MHz were assumed. It is seen that molecules of initial velocity $v_0 = 13-15$ m s$^{-1}$ and initial phase $-\pi < \phi < \pi/2$ are decelerated. There is no deceleration with $\phi_0 = 1.96\pi$, except for the synchronous molecule. The switching phase of $\phi_0 = 0.47\pi$ has the largest phase-space stability among the five switching phases shown in figure 3.

Panel (b) of figure 3 shows the final velocity distribution of molecules whose initial velocities were between 13.8 and 15.5 m s$^{-1}$, and whose initial phases were between $-\pi$ and
Table 3. Efficiency of the decelerator $D$, final mean velocity $v_{\text{fin}}$, final velocity distribution $\Delta v_{\text{fin}}$, and total travel length $\ell_{\text{tot}}$.

| Parameter $^a$ | $D$ (%) | $v_{\text{fin}}$ (m s$^{-1}$) | $\Delta v_{\text{fin}}$ (m s$^{-1}$) | $\ell_{\text{tot}}$ (mm) |
|----------------|---------|-------------------------------|--------------------------------|------------------|
| $N$ constant$^b$, $P_0 = 1$ W | 1.47 | $-0.03$ | 0.41 | 29 |
| $N$ adjusted$^b$, $P_0 = 1$ W$^c$ | 1.47 | 0.11 | 0.41 | 26 |
| $P_0 = 0.25$ W | 1.01 | $-0.14$ | 0.53 | 53 |
| $P_0 = 1$ W$^c$ | 1.47 | 0.11 | 0.41 | 26 |
| $P_0 = 3$ W$^d$ | 1.97 | $-0.03$ | 0.40 | 14 |
| $\phi_0 = 0.31\pi$ | 1.44 | 0.55 | 0.46 | 14 |
| $\phi_0 = 0.47\pi$ | 4.43 | $-0.24$ | 0.72 | 78 |
| $\phi_0 = 1.58\pi$ | 3.30 | $-0.19$ | 0.54 | 27 |
| $\phi_0 = 1.66\pi^d$ | 1.97 | $-0.03$ | 0.40 | 14 |
| $\phi_0 = 1.96\pi$ | 0.0 | – | – | – |
| $f_s = 0.5$ MHz | 1.43 | $-0.17$ | 0.49 | 25 |
| $f_s = 1$ MHz$^c$ | 1.97 | $-0.03$ | 0.40 | 14 |
| $v_0 = 7.8$ m s$^{-1}$ | 3.85 | $-1.30$ | 0.74 | 1 |
| $v_0 = 14.0$ m s$^{-1}d$ | 1.97 | $-0.03$ | 0.40 | 14 |

$^a$The standard condition used in the calculation was $P_0 = 3$ W, $f_s = 1$ MHz, $\phi_0 = 1.66\pi$, $v_0 = 14.0$ m s$^{-1}$ and ‘$N$ adjusted’. The parameters changed from the standard conditions are shown in the first column.

$^b$‘$N$ constant’ means the number of stages $N$ is fixed throughout the deceleration process. ‘$N$ adjusted’ means the number of stages $N$ was always adjusted so that the switching frequency became as close as possible to 1 MHz, but keeping $N$ as odd numbers.

$^c$These are exactly the same conditions.

$^d$These are exactly the same conditions.

We terminated the simulations when the velocity of the synchronous molecule became $v_z = 0.00$ m s$^{-1}$, that is, the synchronous molecule was completely decelerated and stopped with respect to the cavity. The velocity of other molecules were distributed around 0.0 m s$^{-1}$, and therefore some molecules have negative velocities with respect to the cavity as seen in figure 3(b). It is seen that the switching phase of $\phi_0 = 1.66\pi$ has the narrowest velocity distribution of the decelerated molecules, while the switching phase of $\phi_0 = 0.47\pi$ has the widest phase-space area. Since the temperature of cold molecules has to be defined by the velocity distribution $\Delta v$, but not the average velocity $v$, a narrower final velocity distribution is better. Wider phase-space area does not mean better deceleration in terms of temperature. The switching phase of $\phi_0 = 1.66\pi$ gave the coldest molecules whose final velocity distribution was 0.40 m s$^{-1}$ corresponding to the temperature of 320 µK.

Table 3 summarizes the efficiency of the decelerator $D$, final mean velocity $v_{\text{fin}}$, final velocity distribution $\Delta v_{\text{fin}}$, and total travel length $\ell_{\text{tot}}$ for various values of the switching phase $\phi_0$. The final mean velocity $v_{\text{fin}}$ is the average value of the final velocities of decelerated molecules. The efficiency $D$ was defined as a ratio between the number of molecules decelerated to 5 m s$^{-1}$ or below versus the total number of molecules whose initial velocity was within $\pm 1$ m s$^{-1}$ of a certain initial velocity. Under this definition, the efficiency for the switching phase of $\phi_0 = 1.66\pi$ becomes about 2%.
Figure 4. The phase-space stability of different switching frequency $f_s$ and the initial velocity $v_0$ of synchronous molecules. (a) $f_s = 0.5$ MHz, $v_0 = 14.0$ m s$^{-1}$, (b) $f_s = 1$ MHz, $v_0 = 14.0$ m s$^{-1}$ and (c) $f_s = 1$ MHz, $v_0 = 7.8$ m s$^{-1}$.

4.4. Switching frequency $f_s$

We have also examined the dependence on the switching frequency. The switching frequency of $f_s = 0.5$ MHz is compared with that of $f_s = 1$ MHz. The results are shown in table 3. It is seen that the switching at 1 MHz gives wider phase-space area and narrower final velocity distribution than the switching at $f_s = 0.5$ MHz. It is apparent that faster switching is better for efficient deceleration.

Figure 4(a) and (b) compare the phase-space stability for the cases of $f_s = 1$ MHz and $f_s = 0.5$ MHz. Initial velocities between 0 m s$^{-1}$ and 18 m s$^{-1}$ are shown in order to observe the global picture of the deceleration. In both cases, molecules whose initial velocity is not around 14 m s$^{-1}$ are also decelerated, which can be seen as a couple of stripes in figure 4. A similar effect has been discussed in the Stark decelerator as the higher-order resonance [36]. In general, the switching frequency $f_s = 1$ MHz is more efficient than $f_s = 0.5$ MHz (see table 3). All molecules whose initial velocity is slower than 5 m s$^{-1}$ are decelerated and trapped, except when the initial phase $\phi$ is around 0.5 $\pi$, because their initial kinetic energy is already below the depth of the dipole force potential.

When we fix the number of stages $N$ for all the time, the switching frequency becomes slower as the synchronous molecules are decelerated. We can keep the switching frequency close to 1 MHz by reducing the number of stages $N$. We have examined the effect of the adjustment of the number of stages $N$. The results are summarized in table 3 as ‘$N$ constant’ and ‘$N$ adjusted’. For the ‘$N$ constant’ simulation, we fixed the number of stages $N$ to be 9. For the ‘$N$ adjusted’ simulation, the initial number of stages was set to $N = 9$, and then reduced to 7, 5 and so on, when the velocity of the synchronous molecule became slow enough to keep the switching frequency at 1 MHz with the next lower $N$. The number of stages $N$ was always kept
Figure 5. Experimental setup for the IR cavity decelerator. The output from a frequency-stabilized IR laser is switched on and off by an AOM. The cavity is stabilized by a technique such as the PDH method.

4.5. Initial velocity $v_0$ of synchronous molecules

We have examined the dependence of the initial velocity $v_0$ of synchronous molecules on the phase-space stability. Figure 4(c) shows the phase-space stability when a molecule whose initial velocity was $v_0 = 7.8 \text{ m s}^{-1}$ was used as a synchronous molecule. The slower initial velocity gives a more clear area of decelerated molecules. Compared with the phase-space stability for the initial velocity of $v_0 = 14.0 \text{ m s}^{-1}$ shown in figure 4(a), the decelerated area for the case of $v_0 = 7.8 \text{ m s}^{-1}$ becomes almost twice as large, which is also given in table 3. The final velocity distribution with the slower synchronous molecules resulted in the average final velocity of $-1.3 \text{ m s}^{-1}$, whose absolute value is significantly larger than other cases. Some fine adjustments of the switching condition may be necessary to improve the final velocity and its distribution for molecules with slower initial velocity. It is apparent that slower molecules are more sensitive to the fine conditions of the decelerator.

4.6. Laser power

Table 3 also shows the laser power dependence on the phase-space stability. We have examined three different powers, that is, $P_0 = 0.25 \text{ W}$, $1.0 \text{ W}$ and $3.0 \text{ W}$, corresponding to the maximum potential depths of 20, 40 and 70 mK, respectively. It is clear that higher power gives wider phase-space area and narrower final velocity distribution. At present, we can obtain infrared laser power of up to 3 W using a commercially available laser. It is better to employ the maximum laser power for efficient deceleration.

5. Experimental setup

Figure 5 shows the experimental setup we propose. Frequency stabilized continuous wave (CW) IR radiation is switched on and off by using an AOM before being introduced into a cavity. Part of the radiation is separated before the AOM and used to stabilize and lock the cavity length using the Pound–Drever–Hall (PDH) method [37]. Circulating polarized radiation inside the
cavity is preferred to create the standing wave in order to employ the PDH locking as well as to minimize the possibility of multi-photon ionization by the intense IR electric field.

In order to keep the number of trapped molecules large enough to be detected, loading of molecules in the cavity must be well designed. There are three critical conditions. (i) Since the effective potential is roughly proportional to $1/N$ (see equation (4)), precooling of molecules down to 100–500 mK is a prerequisite in order to decelerate molecules effectively within a cavity length. (ii) Since the present apparatus is a one-dimensional decelerator, the transverse velocity of molecules has to be less than the depth of the dipole force potential, otherwise molecules would escape from the cavity. Therefore, a well collimated cold molecular beam has to be introduced into the cavity. (iii) As seen in figure 3, molecules with an initial velocity distribution of $\pm0.25 \text{ m s}^{-1}$ along the longitudinal direction will be mainly decelerated, so that smaller initial velocity distribution is better for efficient cooling.

The simplest loading scheme is to start with an ensemble of cold molecular gas obtained by buffer gas cooling, and load it into a cavity through a tiny aperture placed along the optical cavity. A cold molecular beam at 0.5 K with a density of $10^8 \text{ cm}^{-3}$ has been reported for NH molecules [38] by buffer gas cooling. Pulsing of such a cold molecular beam in order to combine with the present method is possible by combining a pulsed valve at cryogenic temperature [20, 39]. By introducing such a cold molecular flow through an aperture, 0.4% of molecules have a longitudinal velocity along the standing wave of $14.0 \pm 0.25 \text{ m s}^{-1}$, and transverse velocity of less than $v_t = 4.5 \text{ m s}^{-1}$ (400 mK), for example, by assuming an ideal effusive beam [40]. Therefore, the final density of molecules trapped in the cavity is expected to be $1 \times 10^4 \text{ cm}^{-3}$. An advantage of the buffer gas loading is that one can introduce any type of molecules into the cavity. On the other hand, a problem of this loading method may be that the transverse temperature is still high (40 mK), although they are trapped in the standing wave optical trap.

If we combine the present method with a Stark or Zeeman decelerator, more efficient slowing of molecules may be achieved, although applicable molecules will be limited by the Stark or Zeeman decelerator technique. A cold molecular beam created by a Stark decelerator has a small velocity distribution in both transverse and longitudinal directions, and therefore losses during the present decelerator would become minimal. For example, a cold molecular beam of NH$_3$ with a longitudinal velocity of $7 \text{ m s}^{-1}$ (50 mK) and a velocity distribution of $\pm0.5 \text{ m s}^{-1}$ has been obtained [21, 41]. In such a collimated molecular beam, at least 3% of molecules should be decelerated by our method. The advantage of using such a collimated beam is that the transverse velocity is already low so that the decelerated molecules have extremely slow velocities in all directions.

6. Conclusions

We examined the efficiency of an IR cavity decelerator for making ultracold molecules. It was predicted that the decelerator becomes more efficient with higher laser power and faster switching rate. The initial switching phase of $\phi_0 = 1.66\pi$ gave the best phase-space stability and the narrowest velocity distribution among the simulations we did in the present work. The proposed infrared decelerator can create ultracold molecules at 5 $\mu$K with a velocity distribution of less than 300 $\mu$K when precooled molecules of approximately 500 mK are introduced into a cavity. The simulation showed that the efficiency for the deceleration will be more than 2% when the velocity distribution of precooled molecules is $\pm1 \text{ m s}^{-1}$ along the axis of the cavity.
Narrower initial velocity distributions would result in better efficiency, so it is important that the precooling stage is well-designed.

In the present simulation, we only considered one-dimensional deceleration along the cavity axis. By taking into account the three-dimensional motion of molecules, one may get slightly different phase-space stability. In the case of the Stark decelerator, enhancement of the phase-space stability has been found in a three-dimensional calculation relative to a one-dimensional treatment [42]. A similar effect may be expected in the present IR decelerator.

Experimental realization of the proposed IR cavity decelerator is now underway at UBC. One of the advantages of the current method is that a commercially available IR laser based on the OPO process in a PPLN crystal emits a wide range of infrared frequency between 2 and 5 μm. This frequency range covers most of the vibrational transitions of molecules of chemical interest. For a practical application, one has to examine the parameter dependences more carefully to optimize the conditions for each target molecule. Optimization of parameters by an optimal control theory may be effective for such a purpose.

Acknowledgments

This study was supported by a National Sciences and Engineering Research Discovery Grant in Canada, and Japan Science and Technology Agency under the CREST project, Quantum Information Processing.

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