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Efficient stimulated slowing and cooling of the magnesium fluoride molecular beam

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Abstract

We theoretically investigate the possibility of stimulated light force deceleration and cooling of the diatomic magnesium fluoride molecular beam with near-cycling transitions in the bichromatic standing light wave of high intensity. The weighted degeneracy and force reduction factor are considered due to the behavior of the optical bichromatic force (BCF) in near-cycling transitions with internal degeneracies, and the two-level optical Bloch equations can estimate the actual behavior of the BCF. Our simulation shows that the stimulated force exceeding the spontaneous force by a factor of 2.8 can slow down the molecular beam to several m s^{-1} within centimeter-scale distance, and this slowing mechanism can eliminate the need of compensation of Doppler shift during the longitudinal deceleration of the molecular beam.

Keywords: laser slowing and cooling of molecule, magnesium fluoride, enhanced optical force, optical Bloch equations

(Some figures may appear in colour only in the online journal)

1. Introduction

Though the laser cooling techniques that have been tremendously successful in producing ultracold atoms are difficult to apply to molecules, in the past few years, substantial progress has been made in laser cooling and trapping of diatomic molecules [1–5]. The development of the molecular magneto-optical trap (MOT) should really mirror the huge historical success achieved by the atomic MOT [6], which has revolutionized the fields of atomic and quantum physics. Realizing such similar technique for producing a diverse set of dense, ultracold diatomic species will open the way for studies of precision measurements, ultracold chemical reactions, novel many-body quantum systems, quantum information processing [7–10]. Until now, the $X^2\Sigma^+ \rightarrow A^2\Pi_{1/2}$ electronic transition of diatomic molecules (SrF [3], YO [4], CaF [5]) have been demonstrated for radiative force from optical cycling [11], Doppler cooling [3] and the 2D [4], 3D [12] MOT. One of the complications behind the experiments of laser cooling and MOT of molecules is that it is hard to compensate the changes of the Doppler shift during the longitudinal slowing of the molecular beam. If adding an inhomogeneous dc magnetic field on the molecular beam is used to spatially vary the molecular resonance frequency, and keep the decelerating molecules in resonance with the fixed frequency laser, the existence of dark Zeeman sublevels X ($N = 1$) prevents the use of Zeeman slower, because they can't compensate the shifts of all the levels in the same direction as the magnetic field increasing. However, Barry *et al* used radiative forces produced by the cooling lasers with extra sidebands via modulating EOM to decelerate the SrF radical beam with initial velocity 140 m s^{-1} to less than 50 m s^{-1} for 6% initial flux [13]. Zhelyazkova *et al* also demonstrated slowing and longitudinal cooling of a supersonic beam of CaF radicals using resonant radiative force [5]. Without chirping, the molecules were slowed by up to 17 m s^{-1} with the central velocity 583 m s^{-1} and cooled to 330 mK. In principle, the velocity can be further reduced by chirping the frequency of the light to keep it on resonance as the molecules slow down. But they found that with chirping the deceleration became worse when the chirp rate increased to 30 MHz m^{-1} s^{-1}.

In order to increase the density and number of the decelerated molecules for loading into MOT, it is necessary to

explore the more efficient slowing method for the longitudinal velocity of molecular beam. The bichromatic force (BCF) uses stimulated emission to produce forces much larger than the usual spontaneous radiative force [14–23], and recently BCF was observed under without spontaneous emission, this maybe open the way to allow the extension of laser cooling to systems without closed cycling transitions [24]. Such forces can span a much larger range of atomic velocities than radiative forces, and decelerate the atomic beam from thermal velocities down to several m s^{-1} within a short distance. The other important property is cooling effect, force profile as a function of the atomic velocity shows the rather sharp lower velocity limit, and the decelerated atoms will pile up at the lower velocity limit. Except SrF [3], YO [4], CaF [5, 25] diatomic molecules, there are many more candidates as well suitable for laser cooling experiment such as the ongoing YbF [26], BH [27], CaH [28]. As a representative example, we select magnesium fluoride (MgF) as a prototype molecule to study the detailed dynamic process in optical bichromatic field, and it is both because it has an easily accessible near-cycling transition, and because the structure and parameters of MgF are suitable for laser cooling as a new candidate [29]. The $X^2\Sigma^+ \rightarrow A^2\Pi_{1/2}$ electronic transition of MgF is the main cooling cycling with wavelength of 359.3 nm, the vibrational branching of $A^2\Pi_{1/2}$ is suppressed by highly diagonal Franck–Condon factors, only two additional lasers at 368.6 and 368.1 nm are needed to repump the $v'' = 1,\ 2$ levels to limit the vibrational branching loss to $<10^{-6}$. In section 2 of this paper, we discuss the calculation of the BCF for a near-cycling transition, and In section 3 we present the numerical calculation and analysis of the BCF for MgF molecular beam. In section 4 we summarize and conclude.

2. Calculation of the BCF for a near-cycling transition

We employ the $X^2\Sigma^+_{1/2}(v = 0, N = 1) \rightarrow A^2\Pi_{1/2}(v' = 0, J' = 1/2^+)$ electronic transition frequency ω of MgF molecule with 359.3 nm wavelength for cycling and bichromatic slowing, and the first electronically excited $A^2\Pi_{1/2}$ state has lifetime 7.2 ns [30], the corresponding natural width $\Gamma = 2\pi \times 22$ MHz, and the bichromatic detuning is $\pm\delta$. We set the parameters $\delta = 25\ \gamma$ (~3.5 GHz), the adjacent level spacing of $N=0$ and $N=1$, $N=1$ and $N=2$ for the $X^2\Sigma_{1/2}$ $(v=0)$ state is 31 GHz (~8.6δ) and 62 GHz (~17.7δ), respectively. The laser frequency of the detuning $\pm\delta$ is far away from the adjacent rotational splitting levels of the $X^2\Sigma_{1/2}$ $(v=0)$ state, shown in figure 1. The BCF has a much wider decelerating velocity range than the radiative force, for $\delta = 25\gamma$, $\Delta v_{\mathrm{BCF}} = \delta/k = 198$ m s^{-1}, and $\Delta v_{\mathrm{radiative}} = \gamma/k = 8$ m s^{-1}. Due to the small splitting among the four spin-rotation hyperfine levels, we consider they can participate in the stimulated absorption and emission simultaneously. The repumping laser beams for $v=1$ (368.6 nm) and $v=2$ (368.1 nm) transitions of $X^2\Sigma_{1/2}$ are overlapped with the counter-propagating molecular beam under a small angle.

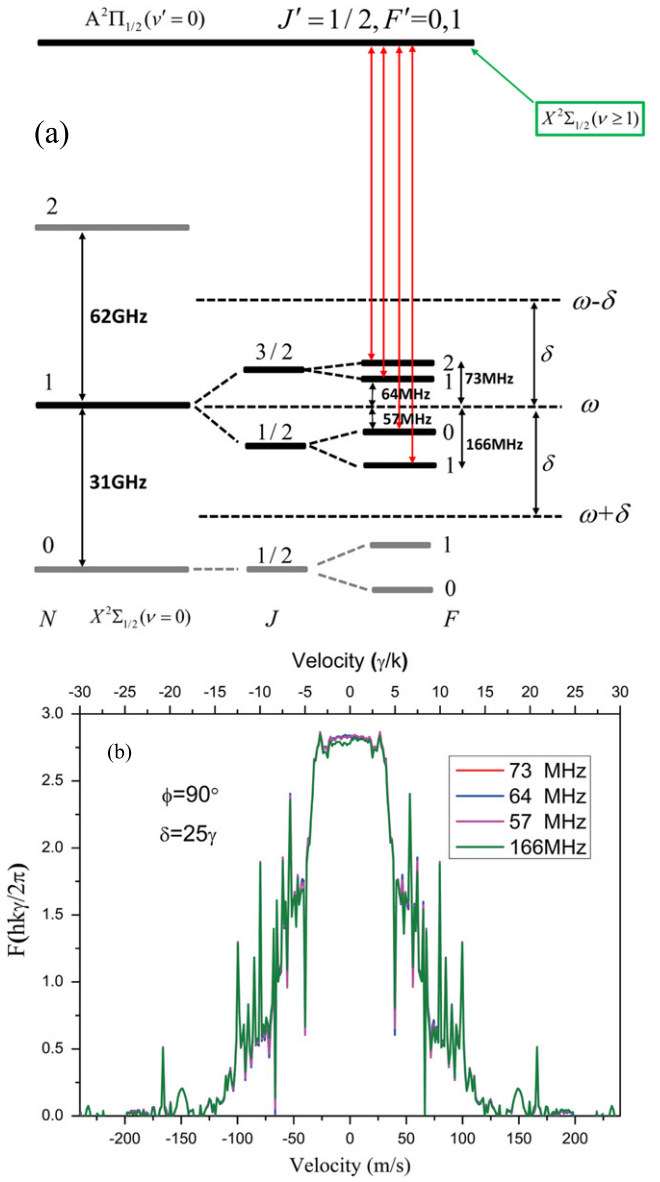

Figure 1. (a) Relevant rotational energy levels of MgF bichromatic cycling scheme. Two different frequencies ($\omega - \delta$, $\omega + \delta$) shifted from one laser beam (ω) are used to make the four HFS (hyperfine structure) levels participate in the stimulated absorption and emission simultaneously under the laser detuning $\delta = 25\ \gamma$; (b) bichromatic force as a function of the molecular velocity for four HFS levels when the relative beat phase ϕ is 90° and the optimal Rabi frequency Ω is set as $\sqrt{3/2}\,\delta$ [22].

The intensity requirement of the repumping lasers is only linearly increased from the saturation intensity I_s for each hyperfine transition by a factor of approximately δ/γ, on the scale of several hundred mW cm^{-2}, which is much less than the power of the main BCF beams.

The MgF molecular beam produced by laser ablation is cooled by a 4 K cryogenic cell and collimated by a pair of magnets, and then the molecular beam flies free for a distance of L_1 and is slowed by bichromatic fields for a distance of L_2 [25, 31–33]. In the end, a probe beam is used to detect the longitudinal velocity distribution of the slowed molecules.

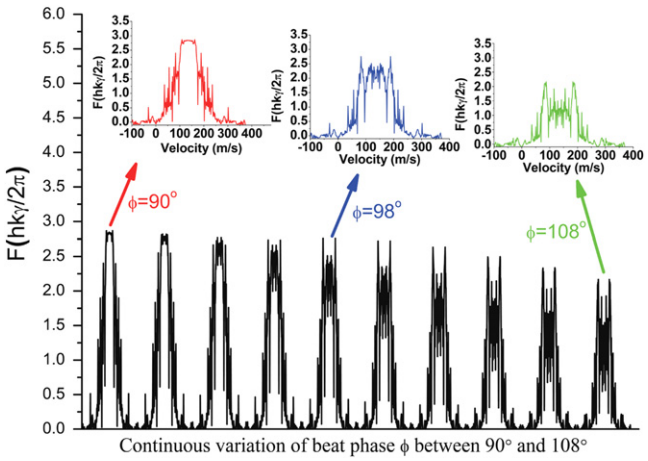

Figure 2. During the process of bichromatic force deceleration, the profiles of continuous beat phases and the relation between molecular beam velocity and the force exerted on the molecule when $\delta = 25$ and $\Omega = \sqrt{3/2}\,\delta$.

The basic physical image of the BCF on a molecule that interacts with two suites of beats traveling in opposite directions can be explained as follows. Standing on the molecule moving with a right-traveling velocity of υ, the left-traveling beat and the right-traveling one are both formed by two kinds of frequencies $\omega + \delta$ and $\omega - \delta$. If the beat has a pulse area of π and the relative phase of the two beats is $\phi = \pi/2$, a molecule will absorb a photon from the left-traveling beat and emit another photon to the right-traveling beat [19]. A momentum of two photons is transferred during a period of such a beat with a magnitude of π/δ, so we can estimate that the magnitude of the force is $2\hbar k\delta/\pi$ which is much larger than the radiative force $\hbar k\gamma/2$ exerted on a molecule.

The two-level optical Bloch equations (OBEs) can establish the tolerance of the BCF and estimate the actual behavior of the BCF when each molecule spends some of its time in the hyperfine levels and sees the same time-averaged force [20]. We get the BCF of the four different transitions via numerical solving of OBEs of two-level systems. In reality, the magnitude of the stimulated force is limited by the laser intensity and the efficiency of the photon transfer, which is given by the optical Rabi frequency. The power of the single bichromatic beam component scales quadratically with the optimal Rabi frequency, $I = 3I_s(\delta/\gamma)^2$, which causes power broadening that makes all of the hyperfine levels nearly resonant. The level spacing between these hyperfine levels is 9, 120 and 110 MHz, respectively, here we assume these four hyperfine splittings of the $X^2\Sigma_{1/2}(v=0)$ state have the same optimal Rabi frequency for the BCF. In such a case, the four sets of HFS levels will cycle independently in the BCF laser field, considering the Hönl–London factors which give the dependence of spectroscopic line intensities of Zeeman sublevels (m_F quantum states) of four hyperfine structure levels for decays from the $A^2\Pi_{1/2}(v' = 0, J' = 1/2^+)$ state to the $X^2\Sigma^+_{1/2}(v = 0, N = 1)$ state [34, 35], and each molecule sees the same time-averaged force by multiplying the probabilities decaying from the excited level to the four different HFS levels.

The force exerted on molecules against velocity in different relative beat-note phase can be attained using numerical calculations of OBEs. Using rotation wave appropriation and constant velocity appropriation we can get the OBEs:

$$\begin{cases} \dot{r}_1 = (\omega_{mi} - \omega)r_2 - \dfrac{\gamma}{2}r_1 - 4\Omega r_3 \sin(k\upsilon t)\sin\left(\dfrac{\phi}{4}\right)\sin(\delta t) \\ \dot{r}_2 = (\omega_{mi} - \omega)r_1 - \dfrac{\gamma}{2}r_2 + 4\Omega r_3 \cos(k\upsilon t)\cos\left(\dfrac{\phi}{4}\right)\cos(\delta t) \\ \dot{r}_3 = 4\Omega\left[r_1 \sin(k\upsilon t)\sin\left(\dfrac{\phi}{4}\right)\sin(\delta t) \right. \\ \qquad \left. - r_2 \cos(k\upsilon t)\cos\left(\dfrac{\phi}{4}\right)\cos(\delta t)\right] + \gamma(1 - r_3), \end{cases} \tag{1}$$

where the variables r_1, r_2, r_3 are called the Bloch variables, k is the wave vector of the incident laser, γ is the natural width of the excited state $A^2\Pi_{1/2}(v' = 0, J' = 1/2^+)$, υ is the velocity of the molecule, Ω is the Rabi frequency, ϕ is the beat-note phase, δ is the bichromatic detuning, ω_{mi} and ω are the frequency between one of the four hyperfine levels participating in the cycle and the excited level, the frequency of the incident laser, respectively. From numerical calculation of the OBEs we can get the magnitude of the three Bloch variables and consider the force reduction factor accounting for possible further reductions of BCF due to the behavior of the optical BCF in near-cycling transitions with internal degeneracies [20], which are then taken into the following equation of the BCF:

$$F_b = 2\left(\frac{\frac{g_e}{2} + g_a}{\frac{g_e}{2} + g_a + g_d}\right)\hbar k\Omega\left(-r_1\cos(\delta t)\cos\left(\frac{\phi}{4}\right)\sin \right. \\ \left. \times (k\upsilon t) + r_2 \sin(\delta t)\sin\left(\frac{\phi}{4}\right)\cos(k\upsilon t)\right). \tag{2}$$

Here g_e is the degeneracy of excited state, g_a is the degeneracy of the ground-state levels that are active in the BCF, and g_d is the degeneracy of 'dark' ground-state levels that are populated but do not participate in the BCF. With the force in bichromatic fields we can do some simulations to describe the dynamic process of molecule in such a slower. Figure 1(b) gives BCF exerted on an MgF molecule as a function of the molecular velocity for four hyperfine structure levels when the beat-note phase ϕ is 90° , the laser detuning δ is 25 γ and the optimal Rabi frequency Ω is set as $\sqrt{3/2}\,\delta$. The gap of the four cycling ω_{mi} is a bit different from each other, so the force is slightly different from each other. The cooling time can be estimated from the BCF profile, the initial velocity distribution should accumulate at the edge of the force profile after the cooling time.

3. Numerical calculation and analysis

First, we determine the range of molecular beam velocity during which the molecules can be slowed. The *X*–*A* electronic transition of MgF molecule has the saturation intensity I_s for each hyperfine transition of about 30 mW cm^{-2}. The power of the single bichromatic beam component scales quadratically with the optimal Rabi frequency, $I = 3I_s(\delta/\gamma)^2$. When the laser detuning δ is 25 γ and the optimal Rabi frequency Ω is set as $\Omega = \sqrt{3/2}\,\delta$, the cw laser beams are tightly focused to waists with top-hat diameter of 1mm to provide the required irradiance of about 56.25 W cm^{-2} for each of the four BCF beam components, the total power of four BCF beams is 1.77 W at wavelength 359.3 nm. Figure 2 shows how the BCF changes as the beat-note phase increases when the molecule situates different position. From the figure we can find that the magnitude of the force exerting on the molecule is decreasing as the beat phase increases, but the relation between the velocity and the force is different from each other. The variation of the beat-note phase ϕ should be taken into account and we set the initial value as $\pi/2$ with the maximum BCF.

Monte-Carlo simulations of molecule deceleration on the basis of the calculated force profiles have been performed. In order to take full advantage of the velocity capture range, we detune the main slowing laser by kv (=17 γ) (the frequencies of the components of the bichromatic beams are $\omega_m \pm \delta - kv$, $\omega_m \pm \delta + kv$), by this way the velocity center of the BCF profile can be located to a specific velocity (=17 γ /$k \approx$ 135 m s^{-1}). The molecular beam has a forward velocity of $v_{//} = 140$ m s^{-1} with a spread of $v_{//} = \pm 20$ m s^{-1}, the transverse velocity spread of the collimated beam is $\Delta v_\perp = \pm 1$ m s^{-1}. Figure 3(a) shows how the velocity of molecular beam changes under different laser detuning δ. We can find the molecules with lower velocity can be obtained under the lager detuning; however, the number of molecules is getting less. Because the larger detuning means the larger velocity capture range, we can get the slower molecular beam while the velocity center of the BCF profile is set as the same. From the velocity profile under different detuning, we can get the longitudinal velocity of the molecules as slow as about 6.5 m s^{-1}, which can be captured by MOT of molecules [4, 12]. Figure 3(b) also shows the deceleration efficiency of the cooled packet under different laser detunings. Filled squares are the overall efficiency obtained from comparing the total molecule-number slowed in each packet to the total number of molecules input into the slower as a function of different laser detuning. It can be seen from figure 3(b) that with the increase of the laser detuning, the deceleration efficiency first is increased to the maximum value about 10.8% at the position of $\delta = 15\,\gamma$ and then is decreased. The reason is that the larger detuning means larger velocity capture range, but the transverse diffusion of the molecular beam during the process of deceleration also becomes serious due to the larger magnitude of BCF. So when these two mechanisms are balanced, the largest efficiency is obtained. Though the deceleration efficiency can reach 10.8% for $\delta = 15\,\gamma$, the finial velocity of the molecular packet is about 58.5 m s^{-1}, which is larger than 6.5 m s^{-1} for $\delta = 15\,\gamma$ with efficiency of 2.85%.

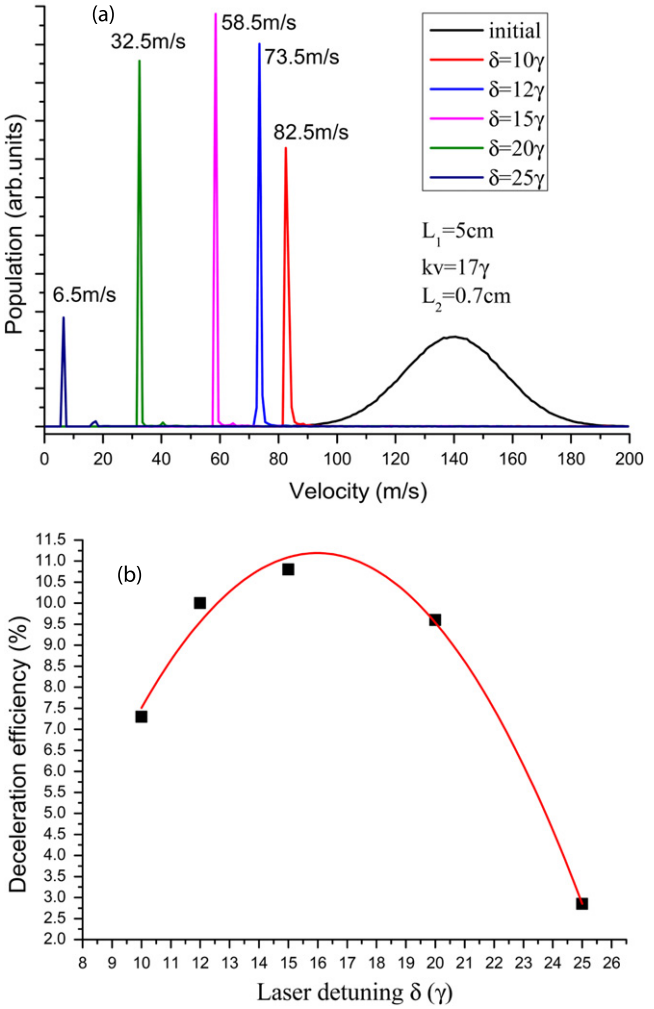

Figure 3. Deceleration results of molecular packets for varying laser detuning δ when the kv is set as 17 γ, and the Rabi frequency of the laser is set as $\Omega = \sqrt{3/2}\,\delta$, (a) the molecular velocity distributions for various laser detuning, (b) the dependence of the molecule-number of the decelerated molecular packets on the various laser detuning.

4. Summary

We have described an efficient approach to produce intense and slow molecular beams by the BCF. The frequency doubling Tisapphire laser can provide such power to produce larger BCF and the larger velocity capture ranges for the deceleration of MgF molecular beam. From the velocity profile under different detuning, we are able to theoretically decelerate the longitudinal velocity of the molecules as slow as about 6.5 m s^{-1}, which can be captured by molecular MOT. From above simulation and discussion, we can find the advantages of bichromatic slowing used in the system of molecules compared with the molecular beam slowing with the radiative force. We also find that the bichromatic slowing mechanism can eliminate the need of compensation of

Doppler shift while the velocity of the molecular beam is getting slower and slower. The compensation of Doppler effect with the way of Zeeman effect as used in the slowing of atoms has been found useless and the molecule deceleration via the frequency chirp of the cooling laser is not as good as that used in the system of atoms. The new proposals of the molecular beam deceleration and cooling with a stimulated light force, direct loading into a molecule MOT from a cryogentic two-stage buffer gas beam and Sisyphus process are proposed and demonstrated [36–40].

Acknowledgments

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