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# A Multistage Optical Stark Decelerator for use with a Pulsed Molecular Beam with an Electrostatic Storage Ring *

XIA Yong(夏勇), YIN Ya-Ling(尹亚玲), JI Xiang(吉翔), YIN Jian-Ping(印建平)**

*State Key Laboratory of Precision Spectroscopy, Department of Physics, East China Normal University, Shanghai 200062*

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*We propose a promising scheme to repeatedly decelerate a pulsed molecular beam using a red-detuned quasi-cw semi-Gaussian laser beam (SGB) and an electrostatic storage ring. Using the Monte-Carlo simulation method, we demonstrate that this promising optical Stark decelerator can be used to efficiently slow a pulsed $ND_3$ molecular beam extracted from a Stark decelerator or a cryogenic reservoir by using a single SGB. The deceleration effect of this scheme on the intensity of the SGB is discussed in detail.*

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It is well known that cold molecular systems play an important role in the fields of cold molecular physics, cold chemistry,[1,2] etc. Until now, two main approaches have been taken to generate slow and cold molecules. One is to prepare cold molecules from ultracold atoms via photoassociation[3] or Feshbach resonance.[4] In this case, molecules are limited to only a few species of laser-cooled atoms and their short lifetime. In the second approach, fast and hot molecules are transformed to slow and cold molecules by using methods such as buffer gas cooling,[5] using a rotating jet,[6] deceleration with a time-varying non-uniform electrostatic field[7] or magnetostatic field,[8] applying an even optical field,[9,10] and so on. Among these methods, Stark decelerators or Zeeman slowers can make it possible to decelerate bunches of about $10^6$ molecules at densities of $10^8\,\mathrm{cm}^{-3}$ and velocity down to tens of m/s. In Ref. [11], we have proposed a scheme to decelerate an $ND_3$ molecular beam efficiently by using a single semi-Gaussian beam with a well depth of 7.3 mK, and obtain a relative average kinetic-energy loss of 9.33%. In order to obtain the accumulated deceleration effect of a single SGB, and bring the molecular beam into a much colder region, we use an electrostatic storage ring[12] to guide the $ND_3$ molecular beam extracted from a Stark decelerator and slow it down by using a single quasi-cw SGB, which will form a multistage optical Stark decelerator. In the storage ring, the guided $ND_3$ molecules will be decelerated many times (that is, this SGB deceleration effect can be accumulated), and finally trapped in the red-detuned SGB.

Our proposed multistage Stark deceleration scheme is depicted in Fig. 1(b). A well-collimated pulsed cold molecular beam generated by the electrostatic Stark decelerator is coupled into an electrostatic storage ring along the $z$-axis. An SGB is incident into the storage ring vertically. An acousto-optic modulator (AOM) is employed to modulate the cw SGB to a quasi-cw one with a period $T$ in the time domain. We define the light pulse duration (i.e. the switched-on time) as $t_{\mathrm{on}}$ and the switched-off time as $t_{\mathrm{off}}$, and $T{=}t_{\mathrm{on}}{+}t_{\mathrm{off}}$. The SGB has a waist radius of $w_{\mathrm{SGB}} = 20\,\mu\mathrm{m}$ and a sharp border width of $w_B = 1.2\,\mu\mathrm{m}$.[13,14] The intensity profile of the SGB in the $z$ direction is a semi-Gaussian one. The deceleration principle will be introduced in detail in the following.

The radius of the electrostatic storage ring is $R_{\mathrm{ring}} = 20\,\mathrm{mm}$, the hexapole structure is taken with a radius of the rod electrode equal to $R = 2\,\mathrm{mm}$ and with an inner radius $2R$. The voltages of the adjacent electrodes are taken to be $U = \pm 5\,\mathrm{kV}$. The electric field distribution inside the hexapole can be numerically calculated by using finite-element simulation software. We assume that the storage ring is located in the horizontal plane, for an $ND_3$ molecule in the low-field-seeking state, the centrifugal force on the molecule captured in the storage ring is balanced by the radial component of the hexapole force, $F = mv_\phi^2/(R_{\mathrm{ring}}+r')$, where $v_\phi$ is the longitudinal velocity or tangential velocity of the molecule in the ring, $r'$ is the radial coordinate. The equilibrium radius for the trapped molecules (the position offset from the center of hexapole electric field) $r_0' = mv_\phi^2/(R_{\mathrm{ring}}k)$, where $k = 3\mu_e U|MK|/[R^3\,J(J+1)]$, the total angular momentum $J$, its projection $M$ on the space fixed axis defined by the electric field direction, and

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**Corresponding authors. Email: jpyin@phy.ecnu.edu.cn

its projection $K$ on the molecular symmetry axis, $\mu_e$ is the permanent dipole moment of the molecule. Thus the round-trip time of a molecule is given by $t_r = 2\pi(R_{\rm ring} + r_0')/v_\phi$.

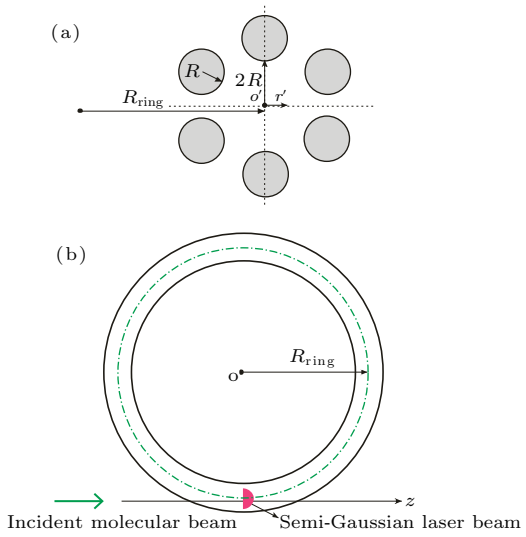


**Fig. 1.** (a) Cross-sectional view of the hexapole storage ring; (b) a principle scheme to form a multistage optical Stark decelerator for a pulsed molecular beam.

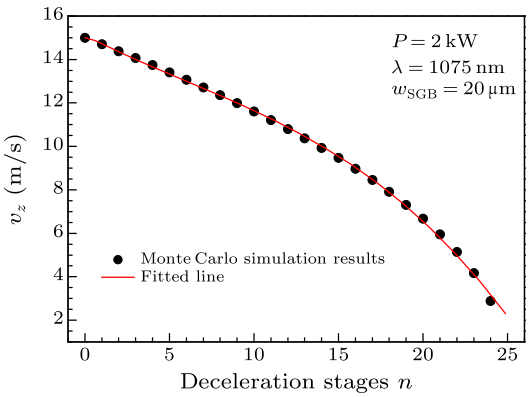


**Fig. 2.** Dependence of the velocity of the slowed molecular beam on the number of deceleration stages.

For the ac Stark effect, the potential energy of a molecule moving in a red-detuned SGB field in the $z$ direction is

$$U(z) = -\frac{\alpha}{2\varepsilon_0 c} e^{-\frac{2z^2}{w_{\rm SGB}^2}}, \quad z \geq 0,$$

where $w_{\rm SGB}$ is the waist radius of the SGB, $\alpha$ is the molecular average polarizability, $\varepsilon_0$ is the dielectric constant in vacuum, and $c$ is the speed of light in vacuum.[9] Based on the analysis in Ref. [11] we know that the influence of our SGB's sharp border width on the deceleration effect is very small, since the border width $w_B$ (=1.2 μm) of the reduced SGB is far smaller than its waist $w_{\rm SGB}$ (=20 μm). For simplicity, in this scheme, we only consider that the SGB is an ideal one. From $\boldsymbol{F} = -\nabla U(z)$, we can obtain the optical dipole force that molecules feel in the SGB light field, and analyze the motion of neutral molecules in the red-detuned SGB. The deceleration principle of a single SGB is the same as that in Ref. [11]. When the SGB is switched on at time $t_{\rm on}$, the incident molecular beam is decelerated and becomes colder. When the SGB is switched off at time $t_{\rm off}$, the decelerated molecules moving along the electrostatic storage ring form a circle. As soon as the molecules return to the SGB region, the SGB is turned on, and the molecules are again decelerated. In the later periods, the above process will be repeated. Thus the deceleration effect of a single SGB will be accumulated by using a synchronous time-sequence and control system so as to lower the incident molecular velocity to a final value that is lower than the well depth of the SGB. In this scheme, molecules with different velocities in the $z$ direction have their own moving orbits in the electrostatic storage ring, and the corresponding moving time for a circle is different. Thus a certain AOM modulation period $T$ can only work for molecules with a certain velocity in the $z$ direction. This deceleration principle is similar to that of a multistage electrostatic Stark decelerator used for molecules.[7] From the phase stability theory, we can only use a synchronous time-sequence and control system to slow a finite portion of molecules in a beam where velocities have a $\pm\Delta v_z$ deviation of the synchronized velocity $v_z$.[7]

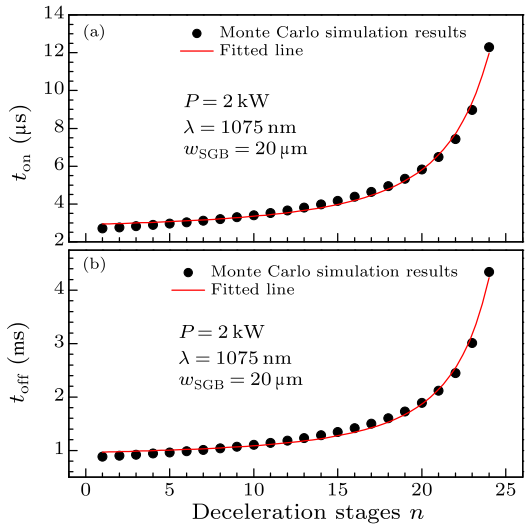


**Fig. 3.** Dependence of the switched-on time (a) and switched-off time (b) on the number of the deceleration stages.

To demonstrate our multistage optical stark deceleration, we study the dynamic process of a pulsed $ND_3$ beam in an electrostatic storage ring with a quasi-cw ideal SGB by using a classical Monte-Carlo simulation. A 2 kW cw ytterbium fiber laser with a wavelength of 1075 nm and a beam quality of $M^2 < 1.2$[15] is used to generate a reduced SGB with the waist radius of $w_{\rm SGB} = 20\,\mu$m, and the corresponding maximum well depth is 10.9 mK for $ND_3$, and the corresponding max-

imum trapped velocity is 3 m/s. The incident molecular beam is generated by the electrostatic Stark decelerator and has a central velocity of $v_z = 15$ m/s and a longitudinal temperature of 18 mK.[16]

Some typical simulated results are shown in Figs. 2–4, where the solid circles are the Monte-Carlo simulated data and the solid lines are the fitting curves. Figure 2 shows the dependence of the decelerated molecular velocity $v_z$ on the deceleration stages $n$. We can see from Fig. 2 that with the increase of the number of the deceleration stages $n$, the flow velocity of the molecular beam $v_z$ will be gradually reduced. In particular, after 24 deceleration stages, the mean flow velocity of the $ND_3$ beam will be slowed from 15 m/s to 2.9 m/s, and the decelerated cold molecules will be trapped in the SGB. This shows that our proposed multistage optical Stark decelerator can be used to efficiently slow a pulsed molecular beam.

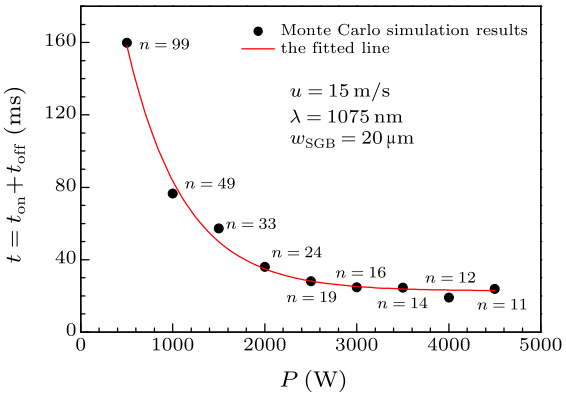


**Fig. 4.** Dependence of the deceleration time and the deceleration stage number on the SGB laser power.

Based on the analysis above, we know that an appropriate AOM time sequence is related to the synchronous molecular velocity in different deceleration stages. We have studied the appropriate switched-on time $t_{on}$ and switched-off time $t_{off}$ using the Monte-Carlo method and the results are shown in Fig. 3. We find that both the switched-on time $t_{on}$ and the switched-off time $t_{off}$ of the AOM increase nonlinearly with the increase of the deceleration stage numbers $n$, so the modulation frequency of the AOM is decreased nonlinearly with increasing the deceleration stage numbers, which is the same as that found in electrostatic Stark decelerators. We also find that the whole deceleration time ($t_{on}$+$t_{off}$) for 24 stages is 36.1 ms.

If we increase the laser power of the SGB, the deceleration ability will greatly increase and the number of required deceleration stages will decrease. In order to demonstrate this, we study the dependence of deceleration time and deceleration stages on SGB laser power and the simulated results are shown in Fig. 4. We can see that with increasing SGB laser power, both the whole deceleration time and the number of deceleration stages gradually decrease. For example, when the SGB laser power is 500 W, we need $n = 99$ deceleration stages to finish the deceleration process. When the SGB laser power increases to 4500 W, the deceleration stage number decreases to $n = 11$. All these results show that our multistage deceleration scheme can be used to realize an efficient optical Stark deceleration for a pulsed molecular beam.

In summary, we have proposed a promising scheme to realize multistage deceleration for a pulsed molecular beam in an electrostatic storage ring with a red-detuned quasi-cw SGB, simulated the dynamic process of our optical Stark decelerator for a pulsed $ND_3$ molecular beam, and studied the dependence of the deceleration effect on SGB laser power by using the Monte-Carlo method. Our study shows that the proposed scheme can be used to efficiently slow a pulsed $ND_3$ molecular beam from 15 m/s to 2.9 m/s by using a 24-stage quasi-cw SGB with a power of 2 kW. Our proposed optical Stark decelerator has some important applications in the experimental research of cold molecular physics and molecule optics, cold molecular spectroscopy and precision measurement, cold chemistry, quantum computing and information processing, and so on.

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