

## Super-Gaussian mirror for high-field-seeking molecules

Guangjiong Dong,<sup>1,\*</sup> Sverker Edvardsson,<sup>1</sup> Weiping Lu,<sup>2</sup> and P. F. Barker<sup>2</sup>

<sup>1</sup>Electronic Division, ITM, Mid Sweden University, 851 70, Sundsvall, Sweden

<sup>2</sup>Physics Department, School of Engineering and Physical Science, Heriot-Watt University, Edinburgh, Scotland

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A matter wave mirror using a single, pulsed, super-Gaussian (SG) optical beam for specular reflection of neutral ground-state molecules is studied. The mirror has a high reflectivity close to 100% and nearly perfect specular reflection over a large incident angle. This mirror avoids the usual problems due to surface roughness and the van der Waals interactions that occur in conventional atomic mirrors. Further, it is capable of reflectance and transmittance with applications to velocity filtering and deceleration of cold molecules.

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Recent progress in producing ultracold molecules [1–6] has led to a new field of molecular optics [7–12], opening up a new route to nanoscale material design [10,12,13] and ultrahigh resolution spectroscopy [14]. A molecular mirror is an important optical element in molecular optics. So far, design of such a mirror by extending techniques in atom optics has not been reported. Conventional atomic mirrors utilize a repulsive potential produced by an evanescent optical field [15] or by a periodically magnetized surface [16], reflecting atoms with velocities up to 10's of cm/s with a high reflectivity ( $\sim 100\%$ ). However, the tangential component of the dipole force to the interface used to produce an evanescent field limits the incidence angle to a few milliradians [17]. Reflection may be adversely affected by surface roughness and the van der Waals interaction. Moreover, an optical evanescent reflector cannot reflect high-field-seeking ground-state molecules, although it may reflect molecules in Rydberg states since they are low-field seekers [18]. So far, two alternative methods have been investigated. Reflecting neutral molecules and atoms by an optical standing wave has been studied [19,20]. This scheme has only a low reflectivity ( $< 20\%$ ), and the momentum distribution of the reflected beam seriously deviates from the initial distribution due to anharmonic motion within the potential [20]. More recently, reflection of a state-selected polar molecular beam by a microstructured switchable electrostatic mirror, consisting of a planar array of equidistant gold electrodes with a voltage difference between adjacent electrodes, was demonstrated [21]. This mirror can only reflect low-field-seeking molecules and strongly deflects molecules approaching the surface within several micrometers.

In this paper, we present a method for specular reflection of high-field-seeking molecules by using a super-Gaussian (SG) optical field [22,23]. We show that an ultracold molecular beam with velocities in the tens of centimeters per second range can be efficiently reflected by a single pulsed SG optical beam with intensities of 10's of MW/cm<sup>2</sup>. The reflection of molecules is realized by the dipole force, which is proportional to the gradient of the optical intensity. Specular

reflection is achieved by applying the SG field, which has a large intensity gradient at its edge due to its flat top profile. Unlike those demonstrated in atom optics [15,16], this mirror is an almost perfect specular reflector over relatively large incident angles. It is free from an interface, and thus is capable of transmittance, leading to applications as a velocity filter for molecules and as a molecular decelerator.

A far-off-resonant SG optical beam [22] has a near-flat-top optical intensity profile in the transverse direction, given by  $I(x, y, t) = I_0 g(t) \exp[-(x/w_x)^{2n} - (y/w_y)^{2m}]$ , where  $I_0$  is the maximum optical intensity;  $g(t)$  is a temporal profile,  $x$  and  $y$  are transverse coordinates,  $w_x$  and  $w_y$  are the  $e^{-1}$  widths of the light beam in the two transverse directions, and  $n$  and  $m$  are the orders of the SG field and are treated as equal in this paper. The SG optical field can be readily produced with current techniques in diffractive optics [22,23]. We assume that the optical field is rapidly turned on and off [19,20,24], i.e.,  $g(t) = 1$ , for time  $t$  less than the pulse duration  $t_d$ , and  $g(t) = 0$  otherwise. The pulse can be produced by pulse shaping techniques [25]. The interaction between a far-off-resonant laser and molecules is conservative and produces a dipole potential  $U(x, y, t)$ , given by  $U(x, y, t) = -\alpha Z_0 I(x, y, t) / 4$  [26], where  $\alpha$  is the polarizability and  $Z_0$  the impedance of the vacuum. Using a far-off-resonant optical field at the price of a high power, the spontaneous-emission events can be greatly reduced to produce a specular reflection [27].

We consider an ultracold ( $\ll 1$  mK) molecular beam that is within the SG field and travels in the  $x$  direction with velocity  $v_B$ . The transverse width of the molecular beam and transverse distance that the molecules can move during a typical reflection process is much less than the SG beam width  $w_x$ . We can therefore treat the dipole potential of normal incident molecules as a quasi-one-dimensional (1D) potential, i.e.,  $U(x, t) = -\alpha Z_0 I(x, 0, t) / 4$ . To understand the dynamics of molecules in 1D potential wells for  $t < t_d$ , we plot in Figs. 1(a) and 1(b) the potential wells (solid line) produced by SG optical fields with the orders  $n=1$  (Gaussian) and  $n=12$  (super-Gaussian), and the corresponding dipole forces (dot-dashed line). In contrast with a Gaussian potential, the SG potential well has a flat bottom resembling a square well, and the dipole force is significant only near a boundary of the potential well. Molecules have different dynamics in the two cases, as shown in the position and veloc-

\*Present address: Department of Physics, Hong Kong Baptist University, Kowloon Tong, Hong Kong. Email address: [dong.guangjiong@gmail.com](mailto:dong.guangjiong@gmail.com)

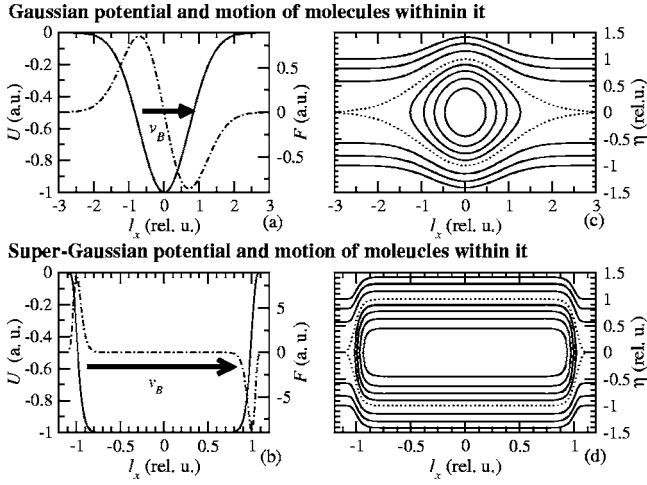


FIG. 1. The dipole potential (solid line),  $U$ , and corresponding dipole force (dashed line),  $F$ , as functions of the position  $l_x$  are shown in (a) and (b) for Gaussian and super-Gaussian intensity profiles. An incident molecular beam with velocity  $v_B$  is within the potential. Position and velocity ( $\eta_x$ ) phase-space plots (c) and (d) indicate the trajectory of molecules within the two potentials. The dotted lines are separatrix within which trapped molecules are reflected.

ity phase space plots in Figs. 1(c) and 1(d), in which the scaled coordinate  $l_x \equiv x/w_x$  and the scaled velocity  $\eta_x \equiv v_x/v_c$ , where  $v_c \equiv \sqrt{M\alpha Z_0 I_0}/8$  is the maximum velocity of molecules that can be reflected. In both figures, the dotted curves are separatrix within which molecules are trapped and reflected. The length of the pulsed molecular beam that is reflected can be no longer than  $2w_x$ . The trajectories of molecules within the Gaussian potential [Fig. 1(c)] are similar to that in the potential of a pulsed standing wave [20], in which the velocity distribution after reflection has been shown to be distorted. By contrast, molecules trapped and reflected within the SG potential well [Fig. 1(d)] have stadium tracklike trajectories. Trapped molecules move almost freely until they are in the curved region where a significant dipole force is exerted. Velocities of reflected molecules are nearly the same as their initial velocities, so that a higher-quality specular reflection could be achieved with a higher-order SG optical field by turning off the optical field when all trapped molecules are reflected.

We use the Wigner function [28] to determine the distribution of the center-of-mass motion of molecules in phase space. When the induced momentum of molecules is much greater than the recoil momentum [19] and the collisions between ultracold molecules are rare (i.e.,  $v_B t_d \ll l_c$  with  $l_c$  the average collision length), the equation of motion for the Wigner function is simplified to the Liouville equation [19,20],

$$\frac{\partial p(l_x, \eta_x, \tau)}{\partial \tau} + v \frac{\partial p(l_x, \eta_x, \tau)}{\partial l_x} + F(l_x, \tau) \frac{\partial p(l_x, \eta_x, \tau)}{\partial \eta_x} = 0, \quad (1)$$

where  $p(l_x, \eta_x, \tau)$  is the Wigner function at the scaled time  $\tau \equiv t/(w_x/v_c)$ , and  $F(l_x, \tau)$  is the scaled dipole force per mass, given by  $F(l_x, \tau) = -g(\tau)n l_x^{2n-1} \exp(-l_x^{2n})$ . Equation (1) is nu-

merically solved with the method of characteristics [19,20] with the initial distribution function  $p(l_x, \eta_x, 0) = \exp[-(\eta_x - \eta_B)^2/\sigma^2]/\sqrt{(l_1 - l_0)\pi\sigma}$  for  $l_0 \leq l_x \leq l_1$ , where  $l_0$  and  $l_1$  are, respectively, the end and the front positions of the molecular beam and the scaled beam velocity  $\eta_B \equiv v_B/v_c$ .

The velocity of the molecules can change after reflection due to the difference between the initial and final potentials, consequently the velocity distribution in the reflected beam deviates from the initial Gaussian distribution. For applications to interferometry and lithography, the deviation is a loss of coherence [29], thus reducing fringe visibility or focusing sharpness. On reflection from the SG mirror, there will be less distortion of the incident velocity distribution function, depending on its velocity spread. For an ultracold molecular beam, the initial velocity spread is so narrow that little distortion of the velocity distribution function occurs after reflection by an SG optical field with a high order ( $n, m \gg 1$ ). By setting the pulse duration equal to the time that a molecule initially at the end (front) of the molecular beam moves to the front (end) after reflection, the distortion can be minimized. In our simulations, we set the pulse duration for reflection to be the time required for slow molecules, initially with velocity  $\eta_x = \eta_B - 2\sigma$ , to travel from the end ( $l_x = l_0$ ) to the front position ( $l_x = l_1$ ) of the incident beam following a reflection. This scaled pulse duration is given by

$$\tau_d = \int_{l_0}^{l_r} \frac{dl_x}{\sqrt{\epsilon + \exp(-l_x^{2n})}} + \int_{l_1}^{l_r} \frac{dl_x}{\sqrt{\epsilon + \exp(-l_x^{2n})}}, \quad (2)$$

with the total scaled energy of the molecule  $\epsilon = (\eta_B - 2\sigma)^2 - \exp(-l_0^{2n})$  and the turning point  $l_r = \sqrt[2n]{-\log(-\epsilon)}$  at which the velocity of the molecule is zero.

Using Eq. (1), we have simulated the reflection of a molecular beam whose initial distribution is uniform in position space from  $l_0 = -0.45$  to  $l_1 = 0.55$  and has a Gaussian distribution in velocity with a normalized mean molecular beam velocity of  $\eta_B = 5.851 \times 10^{-1}$  and an  $e^{-1}$  width of  $\sigma = 9.25 \times 10^{-3}$ . These parameters correspond to a rubidium molecular beam ( $\alpha = \alpha_{||} + \Delta\alpha/3$  [26],  $\alpha_{||} = 135 \text{ \AA}^3$ ,  $\Delta\alpha = 70 \text{ \AA}^3$  [30]) with a mean velocity 44.27 cm/s at a translational temperature 1  $\mu\text{K}$  in an SG field with an intensity 2.4 MW/cm<sup>2</sup>. The simulation is performed with both Gaussian and super-Gaussian beams with the same width and potential well depth. Figure 2 plots the final velocity distributions  $\rho(\eta_x)$  of the reflected beams, respectively, by the SG potential well (solid curve) and the Gaussian potential well (dashed curve). A Gaussian velocity distribution (diamond curve) with the same  $e^{-1}$  width as the initial, but an average velocity  $-\eta_B$  corresponding to a specular reflection, is plotted in the same figure. Overlapping of the solid curve and the diamond curve demonstrates the specular reflection by the super-Gaussian mirror. In contrast, the velocity distribution of the molecular beam reflected by the Gaussian mirror deviates from the Gaussian profile with an extended velocity distribution.

For an obliquely incident molecular beam, deflection may occur during the reflection process due to the change of the transverse velocity resulting from optical dipole force in this direction. For  $|l_x| < 1$ , the transverse force is  $F_y \approx -m\alpha Z_0 I_0 / (4w_y) (l_y)^{2m-1} \exp(-l_y^{2m})$ , where  $l_y \equiv y/w_y$ . Its

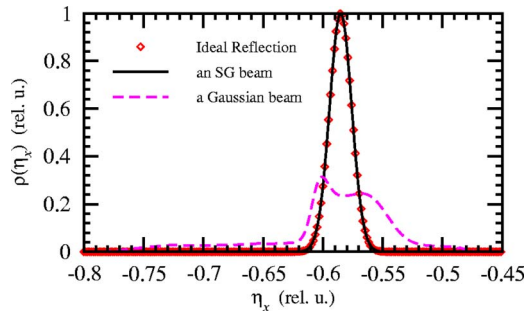


FIG. 2. (Color online) Comparison of the velocity distribution functions,  $\rho(\eta_x)$ , of reflected molecules by a super-Gaussian mirror (solid) and a Gaussian mirror (dashed), with a Gaussian velocity distribution (diamond) corresponding to an idea reflection.

functional dependence on the position  $y$  is the same as that of  $F_x$  on  $x$ , shown in Fig. 1(b). The transverse force vanishes in the central region of the SG optical beam, i.e.,  $F_y$  by an SG beam with the order  $m=12$  is less than one-thousandth of the maximum force for  $|l_y| < 0.7$  [31], but increases rapidly beyond this limit. Therefore, the boundary for specular reflection can be considered as  $|l_y| \leq 0.7$  [31] for a 12th-order SG beam. This leads to a limit to the maximum incident angle at which specular reflection can be achieved. The maximum incident angle is roughly estimated as  $\arctan\{(0.7\omega_y - y_0)/[2(\omega_x - x_0)]\}$  on the assumption that the end of the molecular beam is initially at the position  $(x_0, y_0)$  and reflects to  $(x_0, 0.7\omega_y)$ . We illustrate a case study in Fig. 3 for molecules at the same initial position ( $x_0 = -0.5w_x$ ,  $y_0 = -0.2w_y$ ) and the same initial velocity  $\eta_0 = 0.5$  but with four different incident angles  $\theta_i = 5^\circ, 10^\circ, 15^\circ$ , and  $20^\circ$  within a square SG field ( $\omega_x = \omega_y$ ,  $m = n = 12$ ). Figure 3(a) plots their spatial trajectories, and the corresponding transverse velocities  $\eta_y$  as a function of  $l_y$  are shown in Fig. 3(b).  $\eta_y$  remains unchanged from its initial value for the first three incident angles as the molecules are kept within the region of  $|l_y| < 0.7$ . Once they ap-

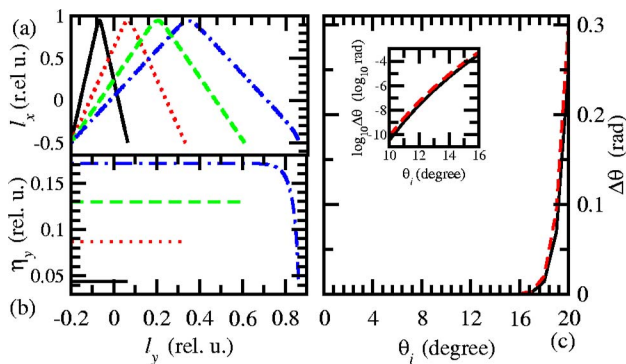


FIG. 3. (Color online) (a) Trajectories of molecules with the same velocity but different incident angles  $0^\circ$  (solid line),  $5^\circ$  (dotted line),  $10^\circ$  (dashed line), and  $20^\circ$  (dot-dashed line) are shown. (b) The corresponding scaled transverse velocity  $\eta_y$  as a function of the position  $l_y$  is shown. (c) The deflected angle  $\Delta\theta$  (radian) for two initial velocities  $\eta_0 = 0.5$  (solid line) and  $\eta_0 = 0.8$  (dashed line) is plotted as a function of the incident angle  $\theta_i$  (degree). Inset shows the common logarithm of the deflected angle for incident angles less than  $16^\circ$ .

proach the right-hand boundary, as shown in the last case  $\theta_i = 20^\circ$ ,  $\eta_y$  is dramatically reduced and consequently deflection occurs. Deflection  $\Delta\theta$  as a function of the incident angle  $\theta_i$  is plotted in Fig. 3(c) for this case study. Here  $\Delta\theta$  is defined as the difference between the angle  $\theta_r = \arctan(\eta_y/\eta_x)$  at the final position and the incident angle. As shown, for incident angles  $\leq 16^\circ$  the deflection angles are much smaller than the typical angular divergence of a molecular beam ( $\sim 1$  mrad) [10,11]. Figure 3(c) shows the deflected angle  $\Delta\theta$  as a function of the incident angle of molecules for two different initial velocities  $\eta_0 = 0.5$  (solid curve) and  $\eta_0 = 0.8$  (dashed curve). The almost overlapping curves show that the deflection is insensitive to the initial velocity. As a result, an obliquely incident ultracold molecular beam can maintain its initial velocity distribution after reflection.

We further study the application of the SG potential as a velocity filter and a decelerator of molecules. The high reflectivity is achieved when the SG potential is sufficiently high to trap all molecules. When the beam velocity is set to be equal to or larger than the critical velocity ( $\eta_B \geq 1$ ), molecules with energy larger than the potential well are untrapped and will transmit through it. This process is a unique feature of this surface free reflector, because using evanescent mirrors, the untrapped molecules [21] or atoms [15] adhere to or are inelastically scattered from the surface. The SG potential can therefore reflect and transmit molecules, which is controlled by beam intensity, and may function as a velocity-selective device by separating molecules of different velocities. The SG potential can also be used to slow molecules because untrapped molecules initially within the potential well cannot recover their kinetic energy in climbing up the potential well. As the potential is switched on when the molecules are inside the SG beam, the dipole potentials for the transmitted molecules are much smaller than their initial value when its final position  $l_x > 1$ . For these molecules with the initial position  $l_{x0}$  and velocity  $\eta_{x0}$ , the final velocity is approximated by

$$\eta_{x,tran} \approx \sqrt{\eta_{x0}^2 - \exp(-l_{x0}^{2n})} \approx \sqrt{\eta_{x0}^2 - 1}. \quad (3)$$

Equation (3) predicts that the velocities of transmitted molecules with initial velocities close to 1 can be greatly decreased when initially they are at the bottom of the potential well [ $\exp(-l_{x0}^{2n}) \approx 1$ ], i.e., the velocity of a transmitted molecule with the initial velocity  $\eta_{x0} = 1 + \sigma$  at the position  $l_{x0} = l_0$  is about 0.19 ( $l_0$  and  $\sigma$  are the same as in Fig. 2). When the beam velocity is set to be equal to the critical velocity, about 50% of the incident molecules after transmittance will have final velocities less than 0.2. This would be an efficient way to decelerate molecules in a molecular beam.

In summary, we have shown that an SG optical field can function as a mirror for reflecting neutral molecules, and achieve a true specular reflection. An SG mirror is applicable to ground-state high-field-seeking polarizable molecules, and needs no state-selection processes. Further, the SG mirror allows incident angles much larger than those in other mirrors [17,21]. Being free from an interface, the mirror allows transmission which could be useful for velocity filtering, and decelerating molecules. Finally, we note that experimental

implementation of the SG mirror is feasible as an SG beam of the required order has been realized in diffractive optics [22,23], while the required intensity can be readily produced by available pulsed laser systems.

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