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Slowing molecules by optical microlinear deceleration

P. F. Barker
The Department of Physics, Heriot-Watt University, Edinburgh, EH14 4AS, Scotland, United Kingdom

M. N. Shneider
Department of Mechanical and Aerospace Engineering, Princeton University, Princeton, New Jersey, 08544

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We study the creation of stationary cold molecules by rapid deceleration of supersonically cooled molecules in a high-intensity pulsed optical lattice. Using the heavy molecule I_2 as an example we predict the evolution of the velocity distribution function of the ensemble during the deceleration period, and show that stationary cold molecules with temperatures below 1 K and densities of approximately 10^{12} cm^{-3} can be produced over length scales of hundreds of micrometers on submicrosecond time scales.

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The application of optical forces using tailored fields has allowed unprecedented control over the external degrees of freedom of atoms [1,2]. More recently, coherent manipulation of neutral molecules has demonstrated alignment [3,4], deflection [5,6], and rotation [7], utilizing the large conservative forces that can be applied using high-intensity fields far from resonance. The deflection of molecules by optical dipole forces has been used to measure molecular polarizability [8], and trapping and slowing in a focused laser beam has been suggested [9]. The periodic dipole forces produced within the interference pattern of a pulsed laser have been suggested for use as a molecular mirror [10], while accelerating lattices produced by frequency chirped cw fields have been used to study transport of ultracold atoms [11,12], and Bose-Einstein condensates [13,14]. Electrostatic techniques have also been used to successfully manipulate highly polarizable atoms [15] and dipolar molecules [16,17], by using time varying periodic fields. These methods have been used to create a cold source of stationary molecules in the milliKelvin range by slowing jet-cooled molecules moving at supersonic speeds [18]. Such a high density of cold molecules is of fundamental interest because it can be used for high-resolution molecular spectroscopy, studies of cold molecular collisions, or as a starting point for dissipative cooling schemes such as cavity Doppler cooling [19] or collisional cold buffer gas techniques [20].

In this Brief Report we study the creation of a cold ensemble of molecules by optical dipole or Stark deceleration in a decelerating optical lattice produced by nanosecond pulsed optical fields. This scheme is the optical analog of the successful electrostatic scheme which has been used to slow a range of dipolar molecules [16–18]. Much larger electrodeless electrical fields can be produced within a short pulse, far detuned, focused laser beam and these fields are strong enough to induce a dipole moment of sufficient strength to be able to decelerate most molecules. Our scheme relies on the application of a decelerating optical lattice created by a rapidly chirped, high-intensity, short pulse laser [21]. Although high-intensity fields are required, we operate far from resonance, while maintaining the intensity well below a value where multiphoton ionization, tunneling ionization, or dissociation would cause significant neutral molecule depletion. We previously explored the application of this general scheme for producing an accelerated ensemble of molecules or atoms. In this paper we show that this scheme could be used to slow heavy polarizable molecules and present results for I_2 as an example.

Our deceleration scheme uses nondissipative optical forces to transfer jet-cooled molecules in a supersonic expansion (10^{12} cm^{-3}) to zero velocity in the laboratory frame. To slow the molecules a decelerating optical lattice with an initial velocity equal to that of the supersonic gas is created. By chirping one of the two counterpropagating beams, the lattice velocity decelerates in time, reducing the velocity of the trapped molecules.

We assume that all fields are far detuned from resonance with a sufficiently slow chirp and field turn-on and turn-off times to ensure adiabatic evolution given by $dR/dt \ll \Delta^2$, where R is the effective Rabi frequency determined for each electronic state and $\Delta(\gg R)$ is the average detuning from the first single photon resonance. For infrared fields in the 1 μm range, the first single photon resonance of many molecules is in the UV and VUV region. Therefore, for an interference pattern traveling slowly with respect to the speed of light, the adiabatic condition can be met for many molecules, and the force is given by the quasielectrostatic approximation [22]

$$F(z,t) = \frac{1}{2}\alpha \nabla E(z,t)^2 \qquad (1)$$

where α is the static polarizability and $E(z,t)$ is the optical electric field. At high intensities (10^{12} $\mathrm{W/cm^2}$), pendular states can be created when the pulse duration is greater than the rotational period [3]. In this process, the molecule librates around the polarization direction of the optical field, effectively aligning the molecule with the field. This process increases the optical force since it leads to a higher effective polarizability when compared to the static polarizability. We consider the slowing of molecules traveling in a supersonic beam by a decelerating optical potential that also travels in the same direction as the beam and is created by the interference between a fixed frequency (ω_1) single mode field of wave vector k_1, and a second field of the same amplitude $E_0(t)$, with a time dependent wave vector $k_2(t)$ and fre-

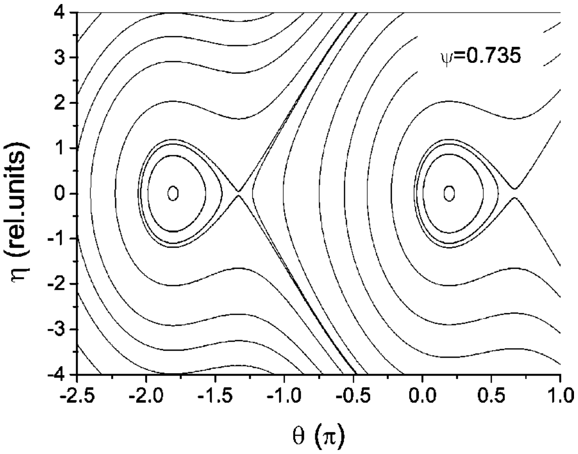

FIG. 1. Trajectories of both trapped and untrapped molecules in the phase space of Eqs. (3) for $\psi = 0.735$. The teardrop shaped closed trajectories define a stable region of phase space where molecules are trapped and slowed by the decelerating optical lattice.

quency $\omega_2(t)$. The initial frequency difference between the beams is given by $\Omega_0 = \omega_2(0) - \omega_1 = qv_0$, where $q = |k_1 + k_2(t)|$ is the wave number of the decelerating lattice and v is the initial velocity of the lattice and the supersonic beam. We limit our consideration to counterpropagating plane wave fields with square temporal intensity profiles. In the absence of collisions, and neglecting an arbitrary phase factor, the equation of motion for the molecule in the lattice is given by

$$\ddot{z} = -a(t)\sin(qz + \beta t^2 - qv_0 t), \tag{2}$$

where $a(t) = \frac{1}{2}\alpha q E_0(t)^2/m$ is the maximum force per unit mass supplied by the lattice, and $\beta(t) = d\omega_2(t)/dt$ is the frequency chirp. We consider the case where $\Omega_0 \ll \omega_1, \omega_2(t)$, and therefore where q is approximately constant over the chirped frequency range. The instantaneous lattice velocity and therefore the mean velocity of the trapped molecules is given by $v_0 - 2(\beta/q)t$.

We study the motion of the molecule in the decelerating reference frame with phase $\theta = qz + \beta t^2 - qv_0 t$ and the dimensionless phase velocity $\eta = (q\dot{z} + 2\beta t - qv_0)/\sqrt{\beta}$. To investigate the trajectory of particles in this phase space we find the critical points of the system of dimensionless equations derived from Eq. (2):

$$\frac{d\eta}{dT} = 2 - \frac{2}{\psi}\sin\theta,\ \frac{d\theta}{dT} = \eta, \tag{3}$$

where $T = \sqrt{\beta}t$ is defined as the dimensionless time, and $\psi = 2\beta/aq$. The critical points of this system of equations are given by $[\theta_1, \eta_1] = [2n\pi - \sin^{-1}\psi, 0]$, which are stable points where n is an integer, and $[\theta_2, \eta_2] = [(2n-1)\pi + \sin^{-1}\psi, 0]$, which are unstable equilibrium or saddle points. Figure 1 shows selected trajectories in the phase space of Eq. (2) for $\psi = 0.735$. The closed teardrop shaped regions separated by 2π define the boundaries between stable orbits in phase space, where particles that are initially trapped by the lattice stay trapped during the deceleration period and all other particles outside this region do not undergo stable deceleration. However, the velocity of untrapped particles can still be significantly perturbed by the lattice potential for short deceleration periods [21]. This phase space plot has the same form as that produced by periodic switching of electrodes as in Ref. [18] when represented in non-normalized coordinates.

The depth of the potential well in each case is determined by the difference in potential height between a saddle point and its closest equilibrium point. The potential well depth ΔU is given by [21]

$$\Delta U = \frac{ma}{q}[2\cos(\sin^{-1}\psi) - \psi(\pi - 2\sin^{-1}\psi)]. \tag{4}$$

It can be seen from Eq. (4) that no potential well exists for the case $\psi \geqslant 1$, because either the chirp is too high, or the force per unit mass supplied by the lattice is not sufficient to trap particles. The maximum well depth is given by $\Delta U_{\max} = 2ma/q$ when $\psi = 0$, which corresponds to the case with no chirp.

We are interested in slowing molecules that are initially uniformly distributed in space, but molecules cannot be introduced selectively into the periodic nanoscale stable region of phase space. Instead, a smaller fraction of the molecules that are initially at the correct phase and velocity are trapped and decelerated. The maximum initial velocity of the trapped species is phase dependent and is given by [21]

$$v_c(\theta) = V[\cos\theta - \cos(\pi + \sin^{-1}\psi) - \psi(\theta - \pi - \sin^{-1}\psi)]^{1/2} \tag{5}$$

where $V = 2\sqrt{\beta/\psi}/q$. Particles are trapped in stable orbits whenever $\psi < 1$. To estimate the fraction of the distribution that can be trapped by the decelerating field, we calculate the overlap between the region of stability and the initial velocity distribution function over a 2π region. For molecules that are initially in thermal equilibrium and homogeneously distributed in phase space, the trapped fraction ς_T is given by

$$\varsigma_T = \frac{1}{2\pi}\int_{\Theta_1}^{\Theta_2}\int_0^{v_c(\theta)} \sqrt{\frac{m}{2\pi kT}} e^{-mv^2/2kT} dv\, d\theta, \tag{6}$$

where Θ_1 and Θ_2 are the roots of Eq. (5). Figure 2 is a plot of the decelerated fraction of molecules as a function of ψ, as well as the minimum deceleration time.

We have chosen to study the deceleration of I_2 at 1 K, which can be produced in an isentropic supersonic expansion [6]. We study molecular iodine to demonstrate that a significant fraction of even very heavy nonpolar molecules can be decelerated using this scheme. We chose an initial I_2 velocity of 560 m/s at 1 K, contained within a supersonic jet of inert buffer gas such as helium and argon. In order to study the evolution of the velocity distribution function of both the trapped and untrapped molecules, we predict the evolution of the distribution functions for each of the mixture components under the influence of the decelerating lattice by numerical integration of the one-dimensional Boltzmann equation given by

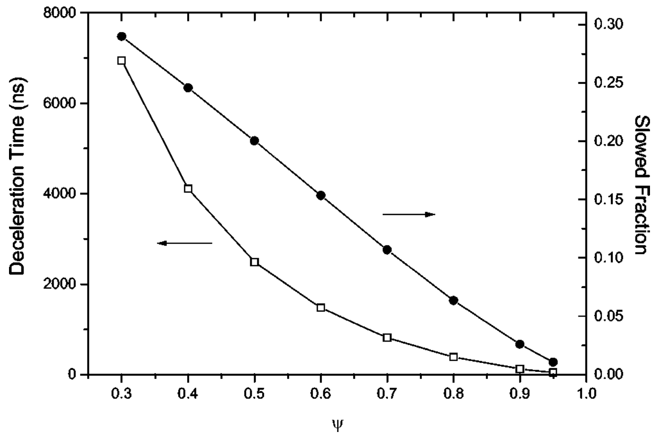

FIG. 2. A plot showing the minimum deceleration time and the maximum slowed fraction of I_2 molecules as a function of the dimensionless parameter ψ.

$$\frac{\partial f}{\partial t}+v\frac{\partial f}{\partial x}+\frac{F(x,t)}{m}\frac{\partial f}{\partial v}=\left(\frac{\partial f}{\partial t}\right)_c, \tag{7}$$

where $f=f(x,v,t)$ is the velocity distribution function, $(\partial f/\partial t)_c$ is the collision integral, and $F(x,t)$ is the optical force given by Eq. (1). The collisional term can be ignored during the deceleration period when the total deceleration time is less than the average time between collisions and therefore we can find solutions to the Boltzmann equation for the buffer gas and I_2 independently. The maximum pressure for collisionless deceleration can be estimated from $p<l_{m0}(T_0/300)/(v_0\tau+a\tau^2/2)$, where p is the pressure in torr, l_{m0} is the free-collision length at 1 torr and 300 K, and τ is the time of deceleration. We calculate the distribution function during the deceleration as detailed in our previous work on acceleration [21]. Figure 3 contains snapshots of the calculated velocity distribution function of I_2 as the lattice is decelerated over the 0.75 μs period. These conditions can be created in a supersonic pulsed valve by expanding I_2 diluted in a buffer gas into a vacuum through a small orifice. For I_2 we used the ratio $\psi_{I_2}=0.735$, which corresponds to a chirp of 5.82×10^{15} rad s^{-1} and $q=1.56\times10^{7}$ m^{-1} for two counterpropagating beams with an intensity of 12 GW/cm^2. At this intensity, essentially no alignment was previously observed [4], and therefore we use the static polarizability to determine the optical force on I_2. We also note that previous alignment experiments of I_2 using 1.064 μm radiation detected no significant ionization at intensities approximately two orders of magnitude higher than the value used here. The final accelerated fraction of 8% obtained at the end of the deceleration period agrees reasonably well with the value of 9.1% calculated analytically, with the small discrepancy due to numerical dissipation in our integration scheme. If we expand the I_2 contained in a buffer gas of argon, we predict, using the same arguments, that approximately 10^{14} cm^{-3} of argon will be trapped and decelerated simultaneously when expanded isentropically from an initial temperature of 300 K and pressure of 1 atm. The inset in Fig. 3 contains an expanded plot of the final distribution functions of I_2 and Ar after the 0.75 μs deceleration period. Although the velocity spread for each distribution of I_2 and Ar is the same, the optical potential and therefore the temperature of the decelerated fraction are lower for Ar (157 mK) than for I_2 (1 K). The final peak values for the decelerated distribution functions of I_2 and the buffer gas Ar are determined by the well depth, by the roots Θ_1 and Θ_2, and also by the initial velocity spread or temperature. The potential well depth and the roots are determined solely by the polarizability to mass ratio of each of the species in the same optical field. We note that the potential well depth for argon is smaller than that of I_2, so that if both were simultaneously trapped following deceleration the I_2 could be sympathetically cooled by the colder Ar.

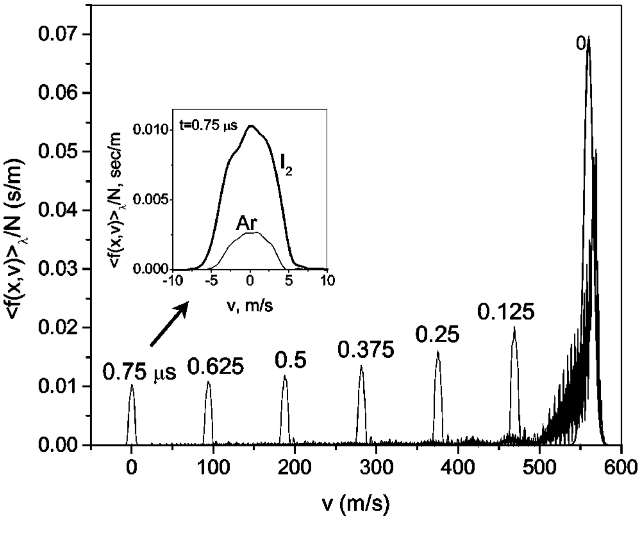

FIG. 3. The evolution of the I_2 distribution function, in 0.125 μs time increments, showing the creation of a stationary ensemble. The inset graph shows the distribution functions of both I_2 (1 K) and Ar (157 mK) following simultaneous slowing in the decelerating lattice.

To estimate an upper limit to the number of decelerated molecules we note that the momentum change $(N_m m v_0)$ imparted to N_m decelerated particles with initial velocity v_0 must be less than the final momentum in the two counterpropagating beams $(\pi r_b^2\beta\tau^2 I/2c\omega)$ over the duration τ. The maximum number of particles that can be trapped is then given by

$$N_m<\frac{1}{2}\frac{\pi r_b^2\beta\tau^2 I}{m\omega c v_0}, \tag{8}$$

where $\omega\approx\omega_1,\omega_2$, I is the intensity, r_b is the spot size of the focused laser beams, and m is the mass of the iodine molecules. Based on these criteria we estimate for an intensity of 12 GW/cm^2 that a maximum number of approximately 10^6 I_2 molecules could be decelerated. For a spot size of 40 μm and 1 cm overlap determined by the Rayleigh range of the focused beam, a density of 10^{12} cm^{-3} could be decelerated. We point out that the optical deceleration scheme that we propose does not cool the molecules, but instead maintains phase space density while transferring them from supersonic speeds to zero velocity in the laboratory frame.

Due to collisions with the buffer gas, or with nontrapped species, trapped particles can be lost from the decelerating potential. The effect of many of these collisions can be considered in the simplest form as a frictional force. The magnitude of this effective force is determined by the rate of the momentum change due to the collisions, and the equation of motion is modified by the effective force, which transforms the closed phase space trajectories for collisionless acceleration, shown in Fig. 1, into open spiral trajectories that are no longer locked to the center of the stable phase space. Instead, the center of the perturbed trajectories translates in time toward the unstable equilibrium point. When the trajectories eventually cross the boundary of the closed teardrop shaped stable region they are lost from the decelerating potential. Therefore a particle is decelerated when the deceleration time is less than its lifetime in the decelerating potential.

We have presented a general scheme for creating a high density of neutral molecules by optical deceleration in a lattice produced by a high-intensity pulsed laser. We predicted decelerated distribution function profiles, and the fractions, using molecular iodine as an example of a heavy, nonpolar molecular species, that can be slowed to create a stationary ensemble of cold molecules. This scheme appears feasible using current laser technology, and should be applicable to a large variety of both polar and nonpolar molecules. The volume and therefore total number density of the decelerated species are limited by the focused spot size of the laser beam, but this disadvantage is offset by the ability to decelerate a high density due to the rapid collisionless deceleration that can be achieved with short pulsed laser sources in the 100–1000 ns range. We point out that in order to use the quasielectrostatic approximation, and to ensure adiabaticity, careful selection of the laser wavelength is required. For lighter molecules, this condition should be easily met with infrared pulsed laser sources. However, for some of the heavy molecules with electronic transitions in the visible, far infrared sources such as a pulsed and chirped CO_2 laser may be required.

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