Creating cold stationary molecular gases by optical Stark deceleration

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Abstract. The deceleration of molecules from cold molecular beams using electric and magnetic fields has become an important means for producing stationary, cold dipolar or paramagnetic molecular gases that can be trapped and potentially cooled to ultra-cold temperatures in the μK range. We report on a general scheme for the creation of cold molecules of essentially any type by deceleration of a cold molecular beam using intense optical fields. Deceleration of benzene molecules to zero velocity is achieved by utilizing a single half oscillation of the center-of-mass motion of the molecules within a moving optical lattice. The lattice traveling at half the speed of the molecular beam is created by the interference of two near-counter-propagating fields at different frequencies. We show that by rapidly switching on the optical lattice for approximately the time required for a half oscillation, a bunch of approximately $10^5$ benzene molecules can be rapidly decelerated and brought to rest in the laboratory frame.

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The creation of cold stationary molecular gases, and the control of their center-of-mass motion by external fields, has become an important new field in molecular and ultra-cold physics and in chemistry. The long interaction times of trapped cold molecular gases, coupled with their reduced momentum spread, allow increasingly higher resolution spectroscopy and therefore the study of molecular interactions that are normally masked by thermal averaging at temperatures above 1 K \([1, 2]\). At the same time, the ability to create a cold molecular beam in a single quantum state that is continuously tuneable over a wide kinetic energy range with a well-defined energy and spread has allowed access to a new regime in molecular collision physics that was previously unexplored \([3]\). A particularly important application of cold molecules is in precision spectroscopic measurements of cold heavy dipolar molecules to test physics beyond the standard model \([4]\). In a similar fashion, precision spectroscopy of cold chiral molecules offers a means to search for parity violation at the molecular level \([5]\). The non-spherical symmetry of molecules offers a set of new interactions that are not available in cold atomic physics. An important example is cold trapped dipolar gases \([6]\), which offer a route to the study of many-body condensed matter physics, exotic quantum phases \([7]\) and quantum information processing \([8]\) in a well-controlled system that can be tailored using external fields. When the de Broglie wavelength of ultra-cold molecular species is comparable to the molecular scale, access is gained to a largely unexplored regime in chemistry where reactions and collisions are dominated by tunneling and resonance phenomena \([9]\).

The creation of cold molecules has been challenging because laser cooling cannot in general be used to cool molecular gases because of the absence of a single or few cycling transitions. Ultra-cold diatomic molecular species can be produced by association of laser-cooled atomic species \([10]\), but the range of species that can be created is limited to the relatively small subset of atomic species that can be laser cooled. In addition, it is often difficult to get these systems into their absolute ro-vibrational ground state. The development of techniques to manipulate the center-of-mass motion of cold dipolar and paramagnetic molecular species has led to other methods for producing slow, cold and chemically stable molecules. These techniques typically start with a stable molecular species in a cold molecular beam or a buffer gas and then a conservative electrostatic or magnetic potential is used to trap and slow a well-defined quantum state. One well-established technique is electrostatic Stark deceleration \([11]\), where dipolar molecules in a well-defined quantum state and narrow velocity spread can be decelerated from a molecular beam using a traveling electrostatic Stark potential. More recently, Zeeman deceleration has been demonstrated \([12, 13]\). Molecular deceleration using a conservative optical potential has also been used to slow a range of molecules. In this scheme,
strong optical fields are used to induce a dipole moment and an optical potential results when the induced dipole moment interacts with the field that induced it creating potentials in excess of \( U/k_b = 100 \text{ K} \) using conventional pulsed lasers, where \( k_b \) is Boltzmann’s constant \([14–19] \). This method typically uses a rapidly switched optical field to briefly trap and decelerate molecules to rest \([17–19] \). Since the well depths created by pulsed lasers can be much larger than in electrostatic Stark and Zeeman deceleration, we utilize the concept of a half phase space rotation, or half oscillation, in a deep optical potential to decelerate cold molecules \([14, 18] \). Optical Stark deceleration is an important variant of these methods because in principle any molecular or atomic species could be manipulated and can be slowed. Importantly, the force that can be applied to a molecule is proportional to the polarizability-to-mass ratio, and this does not change by more than an order of magnitude for most atomic and molecular species in their ground state. In previous experiments on the deceleration of NO using this scheme \([18] \), the well depth was insufficient to decelerate molecules to zero velocity, while in the deceleration of benzene \([19] \), the pulse duration of the lattice was too long or could not be sufficiently controlled to achieve a half or integer half phase space rotation. In this paper, we describe the application of a well-tailored temporal lattice pulse that has sufficient well depth and duration to decelerate benzene molecules to zero velocity for the first time.

2. Optical Stark deceleration in a pulsed lattice

A pulsed optical lattice for molecular deceleration is formed by the interference of two near-counter-propagating optical fields \( E_1 \) and \( E_2 \), as shown in figure 1. An optical lattice potential,
$U(x,t)$, is formed along the $x$-direction by the interaction between the induced dipole of a molecule and the fields that induced it. The force on molecules within the field in the $x$-direction is given by $F(x,t) = -\nabla U(x,t)$. An interference pattern is utilized to maximize the gradient and therefore the force that can be applied to molecules. Molecules in the potential are high field seeking when the optical field is far detuned below any electronic resonances and this force leads to an oscillatory motion within each interference anti-node of the lattice. The induced molecular center-of-mass dynamics have the same form as the motion of a pendulum and the equation of motion of a molecule in the lattice is given by \[ \frac{d^2x}{dt^2} = -a(t)q \sin[q(x-v_l t)], \] (1)

where $a(t) = \alpha q E_1 E_2 / 2m$ is the maximum force per unit mass applied by the lattice to a molecule, $\alpha$ is its polarizability, $q \approx 4\pi/\lambda$ the lattice wave vector and $\lambda$ is the wavelength of the optical fields. Each molecule has mass, $m$, and $E_1$ and $E_2$ are the amplitudes of the electric fields of the counter-propagating optical fields that form the lattice. The lattice travels at a velocity, $v_l$, that is co-moving with the molecular beam along the axis of interference pattern $x$, as shown in figure 1. The lattice velocity is determined by the small angular frequency difference, $\Delta \omega$, between the two beams and is given by $v = \Delta \omega / q$. When the lattice velocity is half that of the molecular beam and the optical potential is deep enough to trap molecules, a significant fraction of molecules can be decelerated to zero velocity in the laboratory frame. This occurs when the lattice is switched on for the duration of one half of an oscillation of the molecules in the periodic potential. This half oscillation within the lattice potential is known as a half phase space rotation. For these molecules the mean velocity is shifted from supersonic velocities to a mean velocity of zero. The period for a single oscillation within the lattice is given by \[ \tau = \frac{8\pi}{q} \sqrt{\frac{M}{\alpha E_1 E_2}}. \] (2)

For most molecular species this oscillation period corresponds to the timescale of several nanoseconds for fields with an intensity in the range of $10^{16}$ W m$^{-2}$ or an amplitude of $10^9$ V m$^{-1}$. The ability to slow molecules to rest depends on both the lattice well depth and pulse duration. The well depth must be sufficient to trap molecules moving with the molecular beam, while the pulse duration must be sufficient to allow only a half or integer half oscillation. The dynamics of the molecules in the lattice depend on the intensity of the lattice. Higher laser intensities lead to deeper optical lattices (greater well depth), which are able to trap molecules of higher velocity. The molecules oscillation frequency in the wells is also proportional to the intensity, and hence in deeper wells the fields have to be switched more rapidly to decelerate molecules to a well-defined velocity. Figure 2 is a time series ($\frac{1}{4} \tau$ steps) illustrating the deceleration of molecules within the lattice. The left panel at each time step is a plot of the velocity and position of molecules that were initially ($t < 0$) traveling at a well-defined velocity and spread uniformly in phase across the lattice. The center panel at each time step is the spatial variation in intensity of the interference pattern that creates the lattice. When the field is turned on ($t = 0\tau$), molecules are attracted towards the anti-node with some accelerating and others decelerating (figure 1(a)). At $t = \frac{1}{2} \tau$ (figure 2(b)) many molecules reach the anti-node and are bunched in phase. This fraction then begins to slow as they are pulled back towards the anti-node. The field starts to turn off at $t = 0.5\tau$ (figure 2(c)) when many of the molecules have reached an average velocity of zero in the laboratory frame. Note that not
Figure 2. A time series in $\frac{1}{4}\tau$ steps illustrating the dynamics of molecules within a pulsed optical lattice. At each time step shown, the velocity and position of the molecules are plotted in the left panel and the moving lattice field in the center panel. (a) The lattice is turned on and molecules are attracted towards the high-intensity regions of the field. (b) Many molecules reach the anti-node and are bunched in phase. (c) This trapped fraction begins to slow down and the lattice is turned off at $t = 0.5\tau$ with many molecules reaching near zero velocity in the laboratory frame. In this timeframe, molecules undergo a single half oscillation within the lattice. Movie 1 (available from stacks.iop.org/NJP/12/073028/mmedia) is an animation of this process.

all the trapped molecules are decelerated to near zero velocity because the potential is not harmonic and the oscillation period is both position and velocity dependent. Movie 1 (available from stacks.iop.org/NJP/12/073028/mmedia) is a more detailed animation of this molecular deceleration process. We have up to this point ignored molecular motion that is orthogonal
to the lattice direction. For a typical beam focus with an $e^{-2}$ spot size of 100 $\mu$m, the optical gradient is approximately 200 times larger than in the axial (lattice) direction [14]. This means that the dipole force is approximately $1/200$ of that along the axial direction and thus the induced motion over the timescale of the lattice pulse is negligible in comparison. Molecules do, however, receive a velocity kick during this pulse which acts to focus the molecules further downstream as has been observed in previous experiments using pulse high-intensity lasers [15, 16]. This motion may be used to increase the density of the decelerated molecules.

3. Experiment

To prepare cold benzene molecules for deceleration, they are produced in a molecular beam containing a mixture of xenon and benzene. This mixture is supplied to a pulsed valve at a pressure of 1.2 bar that is expanded into a vacuum chamber, creating a supersonic expansion. The central part of the expansion is skimmed and passes into the main vacuum chamber (1 $\times$ $10^{-6}$ Torr base pressure) containing the optical lattice. The translational temperature of the benzene molecules is $(1.6 \pm 0.2)$ K and the mean velocity of the jet is $(420 \pm 10)$ m s$^{-1}$. The number density of benzene molecules is estimated to be $10^{11}$ cm$^{-3}$.

The optical lattice is formed by the intersection of two laser beams at an angle of $167.5^\circ$. Each beam is formed from a single longitudinal mode, injection-locked, Q-switched Nd:YAG pulsed laser operating at 1064 nm. One laser supplies a 300 mJ pulse over an approximately Gaussian temporal profile of 7 ns, while the other laser provides a 350 mJ pulse energy over a 15 ns pulse. Benzene molecules undergo a half rotation in phase space in a time that is considerably shorter than the laser pulse length in either of the two lasers used in this experiment. To create a lattice of shorter duration, the output pulse of one laser was chopped optically using a fast Pockels cell producing a duration of approximately 4.1 ns [20]. The lattice beams are subsequently focused into the chamber using 300 mm focal length lenses and the $1/e^2$ width of the beams at the focus is approximately 60 $\mu$m (FWHM) producing a peak intensity of $1.4 \times 10^{12}$ W cm$^{-2}$. The optical lattice has a period of 535 nm in the direction of the molecular beam and its velocity can be controlled by adjusting the frequency of each laser with respect to the other as measured by their beat frequency on a photodiode. A frequency difference of 700 MHz gives rise to a lattice velocity of approximately 375 m s$^{-1}$. Each laser is injection locked to a low-power, narrow linewidth, Nd:YAG laser called a seeder. However, while the pulsed Nd:YAG lasers should in principle be locked to the frequencies of their seeders, which would not change during the high-intensity laser pulse, a frequency chirp, created by refractive index changes in the laser medium, is induced during the laser pulse. This results in a time-dependent frequency change (frequency chirp) in each beam, as well as in the lattice produced by the two beams. This effect was measured in the pulsed output of each beam by mixing the output of each laser with a fixed frequency, narrow linewidth laser. The measured chirp in the lattice, derived from each of these beams, is primarily a linear chirp that is less than 10% of the frequency difference between the two laser beams during the deceleration period. The measured chirp originates primarily in one of the laser systems (Power-Lite, Continuum lasers) and could in principle be eliminated by using a single laser beam split into two components with one component shifted in frequency using an electro-optic modulator.

Benzene molecules transported by the lattice are ionized 60 ns after the peak in the lattice intensity by an ultraviolet laser beam at a wavelength of 254 nm. This beam is focused to a $5 \times 150 \, \mu m$ sheet oriented parallel to the direction of the molecular beam. The laser sheet
intersects with molecules distributed over many periods of the optical lattice. The wavelength
of the laser is chosen to ionize benzene via a 1 + 1 resonantly enhanced multi-photon ionization
(REMPI) scheme [19]. The polarization is chosen to be perpendicular to the motion of the
molecules to limit the recoil associated with photon absorption and electron emission. The
ionization of benzene molecules enables highly efficient detection of single molecules using
a microchannel plate (MCP). The ionization occurs well after the lattice pulses have decayed
(60 ns) to reduce the influence of the second-order Stark shift on the energy levels of benzene.

The lattice is created within a Wiley–MacLaren-type time-of-flight mass spectrometer
(TOFMS) and ions are subsequently detected on an MCP. A slit spaced 50 µm in the vertical
direction limits detection of only molecules that are in the lattice and the resulting velocity
distribution is acquired on an oscilloscope and up to 6000 pulses are averaged. The design
of the spectrometer ensures that spatially distributed molecules of the same velocity arrive
simultaneously at the MCP. The performance of the TOFMS and its dispersion were determined
using the standard ion optics software SIMION and are in accordance with experiment to within
3%. The calibration of the spectrometer for zero velocity was verified by filling the chamber to
a pressure of approximately 1 × 10⁻⁴ Torr with the xenon/benzene mix and observing the center
of the velocity profile, which corresponds to 0 m s⁻¹ for gas nominally at rest.

4. Results

Figure 3(a) is a plot of the velocity distribution of benzene molecules in the molecular beam
before the lattice is turned on. The distribution function is calculated from the recorded time-
of-flight spectrum and shows that the molecular beam has a velocity of 420 m s⁻¹ and a
velocity spread of 31 m s⁻¹ consistent with a temperature of 1.6 K. The peak at 310 m s⁻¹ is
due to the heavier 6% of benzene molecules, which contain a single ¹³C atom, that are also
traveling at 420 m s⁻¹, but are delayed in the time-of-flight spectrum. Figure 3(b) is the velocity
distribution measured 60 ns after the lattice has been turned off showing that a slowed bunch
(7.8% of the molecules that overlapped the lattice) reaches an average velocity of zero at the
end of the deceleration period. For the 1 mm long lattice, this corresponds to approximately
10⁸ molecules in the slowed bunch, assuming a density of 10¹¹ cm⁻³. This has a velocity
spread of approximately 80 m s⁻¹, corresponding to a one-dimensional (1D) energy spread of
approximately 10 K. Two other decelerated peaks occur at 160 and 300 m s⁻¹, in addition to
the broadened central peak of untrapped molecules which are much less strongly perturbed by
the lattice. The lattice was traveling at a velocity of 187 m s⁻¹ (Δ = 350 MHz). Figure 3(c) is
a simulation of the slowing process in the lattice, which takes into account the measured laser
intensity profiles, the frequency chirp in each laser, as well as the isotope observed at 310 m s⁻¹
in figure 3(a). The simulation reproduces the overall structure of the velocity distribution
function observed in the experiment (figure 3(b)) and four major peaks are observed in both
figures 3(b) and (c). These simulations indicate that during the lattice pulse duration many
molecules in the lattice undergo a half rotation in phase space over approximately 4.0 ns and are
decelerated to 80 m s⁻¹ during this time. After this time, they then interact with the low-intensity
tail of the lattice pulse which produces the two slowed velocity groups at 0 and 160 m s⁻¹ in the
final distribution function. A more detailed comparison between the experimental results and
the simulations shows that there are differences in the position of the slowed peaks and their
widths. We have determined that the variation in peak position can be attributed to uncertainty
in the laser intensity as a function of time (20%) and the measurement of the chirp in the
Figure 3. The velocity distribution function of (a) the molecular beam in the absence of the lattice and (b) 60 ns after the lattice has been turned off showing the deceleration of molecules over the lattice duration. (c) A simulation of the velocity distribution 60 ns after the lattice fields have been turned off. In each case, the velocity axis is converted from the time-of-flight, assuming using a mass of 78 amu. This velocity scale is not valid for the heavier but lower abundance isotope shown at 310 m s\(^{-1}\) in panel (a).

laser (20%). This was verified by changes to these parameters to the simulations within the measurement uncertainty. The larger width of the slowed peaks cannot be completely attributed to the uncertainties in our experiment or as due to collisions, even when the molecular density is briefly increased by a factor of three during the deceleration period. It is anticipated, however, that the effects of laser-induced molecular alignment on different rotational states [16], which
is not included in the simulation, will produce different dipole forces and thus oscillation frequencies for different rotational states within the lattice, which may account for the spread in the decelerated peaks. This effect on the deceleration of molecules in a lattice will be the subject of our future work.

5. Conclusion

We have demonstrated the deceleration of benzene molecules in a molecular beam to zero velocity in the laboratory frame using a near half phase space rotation. The optical trap depth, or the ability to trap molecules, depends on the polarizability for a fixed intensity. As this scales roughly with the mass of a complex particle, it can be used to compensate, to some extent, for their greater kinetic energy seeded into a molecular beam of the same velocity. The force for deceleration, however, depends on their polarizability-to-mass ratio, and since this value is constant within an order of magnitude for most atomic, molecular and even larger nanoscale particles and clusters, the same optical fields could be used to decelerate particles over a wide range providing they are not ionized or dissociated. The probability of ionization, and therefore the generality of this technique in this intensity regime for a large range of species, is, however, difficult to ascertain and requires further study. It does, however, appear feasible that the intensity used in this work can be applied to a wide range of molecular and atomic species before ionization would significantly reduce the efficiency [21]. This work demonstrates that optical Stark deceleration is now a viable method for producing cold stationary molecules suitable for trapping and further cooling to below mK temperatures and leads the way to trapping a large range of cold stationary non-polar and non-paramagnetic molecules. Unlike other schemes that filter out a narrow velocity distribution, optical Stark deceleration using deep potentials produces slowed bunches with a molecular beam energy spread that is greater than or equal to the initial molecular beam energy spread. However, because a significant fraction of these molecules are produced at zero velocity, a narrower energy spread can be created by trapping them in an optical, magnetic or electrostatic trap after deceleration. Such a scheme may prove useful for sympathetic [22] and cavity cooling [23], and for low-energy collision and cold chemistry studies. A detailed discussion of the trapping and sympathetic cooling of optical Stark decelerated molecules (H$_2$ and C$_6$H$_6$) has been given elsewhere [22]. Here, optical well depths of 150 mK for C$_6$H$_6$ and 12 mK for H$_2$ appear feasible in a high-finesse ($F = 20\,000$) buildup cavity. Assuming that the density of benzene can be increased up to $10^{13}$ cm$^{-3}$, which is feasible in a pulsed molecular beam, approximately $10^7$ trapped molecules could be decelerated based on our experiments. For a well depth of 150 mK, at most 16% of the slowed fraction at 10 K could be available for trapping. If we further estimate that only 10% of this fraction could be loaded into a trap, this corresponds to $10^5$ molecules trapped at a density approaching $10^{11}$ cm$^{-3}$. Although larger numbers could conceivably be loaded into deeper magnetic and electrostatic traps with larger volumes, the trapped density would be much lower. All of these types of trap could be loaded by first decelerating and then switching on the trap, or by deceleration directly inside the trap as the molecules pass through them.

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