

Optical manipulation of molecular ensembles in the gas phase

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Abstract. A review of developments in the manipulation of gases using strong, off-resonant, optical fields is given. Important applications, which manipulate the velocity distribution function of gases are discussed, including non-intrusive high spatial and temporal resolution gas diagnostics (Coherent Rayleigh and Rayleigh-Brillouin scattering). I also discuss more recent work, which has utilized the much stronger laser-gas interactions in high intensity fields to transport molecules gases on nanosecond timescales.

Keywords: optical lattices, non-resonant forces, manipulation of molecules

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INTRODUCTION

In the 1600's Johannes Kepler was one of the first people to suggest that light could have an important effect on the mechanical motion of particles. He realized that comet tails tend to point away from the sun. But it was not until 1891 that James Clerk Maxwell used his electromagnetic theory of light to quantitatively determine that these light forces have a very small effect on macroscopic objects. Light forces can, however, have large mechanical effects on the center-of-mass motion of microscale particles and particularly on atoms and molecules in gases. Over the last thirty years these light forces have been utilized to manipulate, trap and cool atomic gases. In this paper I will briefly describe the established uses of light forces before describing in detail the use of larger optical forces to significantly modify the velocity distribution function of gases at room temperature and above.

A well-established application of light forces on microscale objects is optical tweezers. Here light scattering and gradient forces are used to manipulate both dielectric and absorbing particles that are usually immersed in a fluid. Applications include sorting and manipulation of biological objects and measurements of small forces including rheometry [1]. Optical forces have also been used extensively for laser cooling of atomic gases. Here, scattering by near resonant excitation and the subsequent spontaneous emission forms a dissipative force, which allows cooling of these gases to micro-Kelvin temperatures [2]. These cold gases can themselves be trapped by conservative forces, which result from the stimulating scattering of photons. The ability to manipulate atoms in this way has revolutionized atomic physics and allowed the production and study of long sought after effects and processes such as Bose Einstein condensation [3].

Until recently these forces have not been of much use for the many gases at room temperature or higher that are of interest for engineering applications. In this paper I discuss a new direction in the application of strong optical forces to molecular and atomic gases, which allows whole distribution function manipulation of gases above room temperature. This introduces a new tool for both engineering and physics.

STRONG FIELD MANIPULATION

The requirement to manipulate gases at room temperature and above implies the use of strong optical fields in the 10^{14} - 10^{16} W/cm² range. Due to current technology this limits the interaction times to durations of 1-100 ns. The optical field must be far from resonance, which for the typical infrared lasers used allows, in principle, the manipulation of any atomic or molecular gas. The interaction of an optical field with the polarizability of a molecule or atom leads to the creation of an optical potential of a single focused laser beam given by [4]

$$U = -\frac{1}{4} \alpha E^2 \quad (1)$$

where α is the effective polarizability (usually the static polarizability) and E is the amplitude of the rapidly oscillating electric field of the light that illuminates the particles. For molecules, which have an anisotropic polarizability, the force is dependent on the orientation of the molecule with the field.

Optical well depths can be produced in the 1000 K range without ionization and dissociation over the timescale of the pulse. This type of potential typically induces molecular motion on timescales of nanoseconds. This method of manipulation can be seen as a type of high-powered optical tweezers for atoms and molecules. The force from a single beam is simply the negative gradient of equation (1), which acts to push molecules towards the highest field intensity.

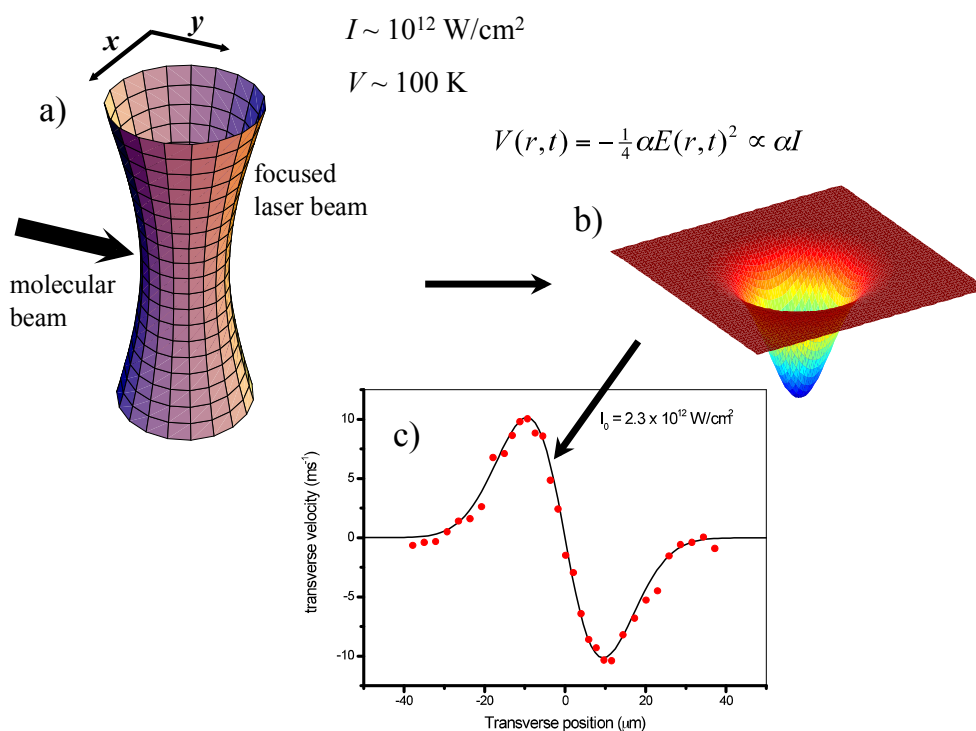


FIGURE 1. Illustration of optical forces induced in a jet by a single pulse focused laser beam including a measurement of the velocity imparted to a xenon jet.

A simple application of such forces is the focusing of an atomic or molecular jet/beam. Figure 1(a) shows a single focused laser beam in which particles from a molecular jet/beam pass through it. As the particles pass into the beam they are pushed towards the center of the light beam. This process can be seen to be due to the Gaussian spatial profile of the beam, which creates a force, attracting particles towards the central high intensity region of the beam. This results at a later time in a higher density of atoms/molecules that are focused downstream. Figure 1(c) is a typical measurement of the velocity imparted to a molecular beam of xenon atoms that are focused by a short pulse (15 ns) Nd:YAG laser producing optical potential of modest well depth of 100 K [5].

MANIPULATING GASES IN OPTICAL LATTICES: WEAK PERTURBATIONS

For any given intensity of light, the largest force that can be created is given by the largest gradient in the optical field. Such a large optical gradient can be created when two counter-propagating optical fields form an interference pattern that has an optical period of half the wavelength of light.

The optical potential created in this case is often termed an optical lattice and is shown schematically in Fig. 2.

Here, two near-counter-propagating optical fields, E_1 and E_2 , interfere. As described previously, the optical potential is formed by the interaction between the induced dipole of a molecule, whose magnitude is determined by the polarizability, and the fields that induced it. In the absence of collisions the induced molecular center-of-mass dynamics of the gas particles in the lattice is the same form as the oscillatory motion of a pendulum. The force on a particle in the lattice is given by [6]

$$F = -\frac{1}{2} \alpha \nabla E^2 \propto \alpha q \cos(qx - \Omega t) \quad (2)$$

where $q=4\pi/l$ is the grating wave vector, Ω is the angular frequency difference between the two counter propagating beams and l is the wavelength of light. The perturbation to the distribution function of the gas can be determined by introducing this force into the 1-D Boltzmann equation given by

$$\frac{\partial f}{\partial t} + v \frac{\partial f}{\partial x} + F \frac{\partial f}{\partial v} = I_c \quad (3)$$

where I_c is the collision integral. If the intensity is not too high ($<10^{10} \text{ Wcm}^{-2}$), a weak perturbation is induced in the velocity distribution function and an analytical solution to the distribution function can be found. This is given by Ref. [7]

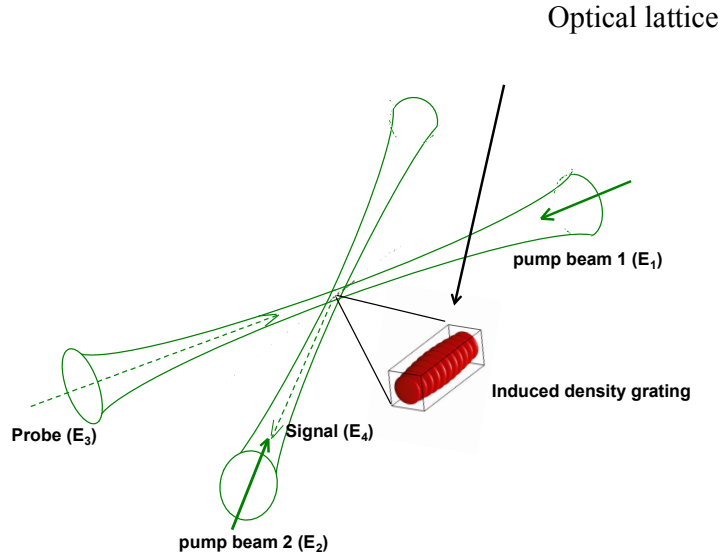


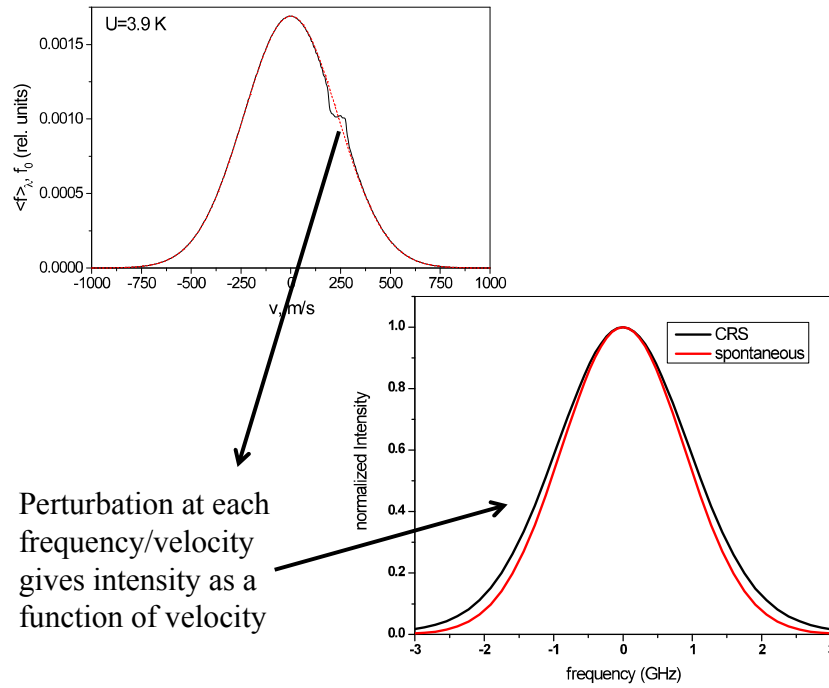
FIGURE 2. The creation of an optical lattice by the interference between two counter-propagating fields. The optical lattice leads to the creation of an induced density grating by optical dipole force.

$$f = f_0(v) \left\{ 1 + \frac{\alpha E_1 E_2}{k_b T} \frac{qv [\cos(qx - \Omega t) - \cos(qx - qvt)]}{qv - \Omega} \right\}. \quad (4)$$

When averaged over an optical period, the distribution function forms a weak plateau in the distribution function centered at a velocity $v = \Omega/q$, determined by the frequency difference between the two beams, Ω . Figure 3(a) shows a plateau created in the velocity distribution function of CO_2 gas at a velocity of 250 m/s by the optical force.

Equation (4) also shows that the induced perturbation is spatially periodic with a period of $\lambda/2$. This periodicity allows the Bragg diffraction of a separate light beam from the induced density perturbation. This process, called coherent Rayleigh (CRS) or coherent Rayleigh-Brillouin scattering (CRBS) can be used to probe the induced structure and obtain information on the thermodynamic properties of the gas. The diffracted signal is proportional to the square of the density perturbation, $\delta\rho$, at a particular frequency difference, Ω .

When the frequency difference between the two beams is scanned, a variation in the signal strength versus frequency is obtained. This scattering profile is shown in Fig. 3(b) for a low-pressure gas of carbon dioxide when the mean free path of the gas is much larger than the period of the lattice. The spectral profile of the diffracted signal as a function of frequency difference is approximately a Gaussian shape, with a width that is 10% wider than that expected from spontaneous Rayleigh scattering from a single laser beam.



Perturbation at each frequency/velocity gives intensity as a function of velocity

FIGURE 3. The creation of a plateau in the 1-D velocity distribution function by an optical lattice in a gas of pure CO₂. The plot on the upper left is the perturbation averaged over a lattice period. The plot on the lower right is the coherent Rayleigh scattering (CRS) profile of Bragg diffracted light from the induced perturbation as a function of the frequency difference between the two lattice beams. The well depth (U/k_b) of the lattice is 3.9 K [8].

This type of Bragg scattering has been used as a gas diagnostic in both single and multi-species gases. Figure 4 contains a plot of a CRS spectrum taken in a butane-air flame. The temperature of the flame (1740 K) was determined with an uncertainty of a few percent, based on a measurement of the CRS spectral profile [9]. Only three species needed to be included in the modeling to accurately model the flame temperature. In principle a single shot measurement of the spectral profile is feasible but has yet to be demonstrated.

At higher pressures, where bulk motion of the gas is induced, two peaks occur in the spectrum and the Bragg scattering is more generally termed coherent Rayleigh Brillouin scattering (CRBS) [10]. These peaks correspond to the efficient excitation of sound waves in the medium. It has been shown by Pan et al. that these CRBS scattering profiles are particularly useful for studying bulk viscosity in gases and temperature through detailed fitting of the spectral profiles to well-developed models of this process.

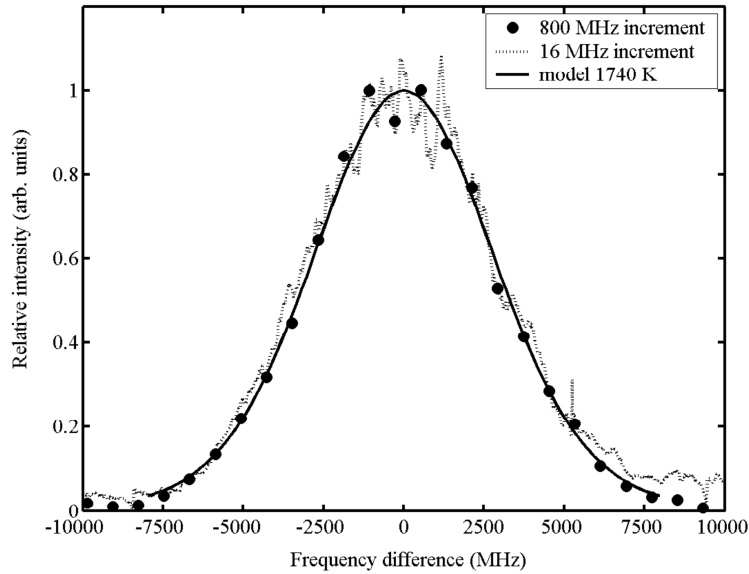


FIGURE 4. The CRS spectral profile of a butane-air flame. The bold line is a fit to the measurements (thin line and round circles).

LARGE PERTURBATIONS IN AN OPTICAL LATTICE

When higher intensities ($> 10^{10} \text{ W/cm}^2$) are applied to the gas, the induced optical potential can be comparable to the average energy of the gas particles. In this regime the distribution function can be completely modified by much larger forces. One of the first observations of this effect was the spectral narrowing of the coherent Rayleigh scattering line shape in a room temperature CO_2 gas with fields in the 10^{15} W m^{-2} range [8]. The line shape was observed to saturate to a width of approximately half that observed at low pump intensities and indicates a transition from scattering primarily from untrapped molecules to molecules that for the duration of the pulse were trapped and untrapped localized by the deep (60 K) optical potentials. At higher densities ($5 \times 10^{24} \text{ m}^{-3}$), where collisions between the trapped and untrapped molecules become important the spectral profile broadened to that observed in conventional CRS. This narrowing process is shown in Fig. 5 at two lattice beam intensities. At an intensity of $3.97 \times 10^{15} \text{ Wm}^{-2}$ the spectral profile narrows to approximately half the width of the spectrum taken at $2.48 \times 10^{14} \text{ Wm}^{-2}$ where a conventional CRS profile is observed. These measurements show a narrowing of the spectral profile, which indicates that much of the distribution function can be trapped and strongly perturbed by the optical potential. Not only is this observation important to show that this effect is possible with room temperature gases, but also that this strong modification to the line shape must be taken into account when using high intensity lasers to boost CRS signals.

Perhaps the clearest example of strong perturbation to the velocity distribution function of a gas induced by high intensity lattices is the deceleration of a molecular beam. In these experiments a moving optical lattice was used for a controlled manipulation of molecular velocity using optical fields in the $10^{11} \text{ W cm}^{-2}$ range. By using the oscillatory motion of nitric oxide molecules in these potentials, evident from equation (2), a rapid deceleration or acceleration in a molecular beam to well-defined velocities was demonstrated. A simulation of the oscillatory motion of the whole distribution function is illustrated in Fig. 6 for nitric oxide molecules in a molecular beam. Here the velocity distribution function undergoes oscillations in velocity with time as the whole distribution function is trapped by the optical potential. The optical lattice in this case is travelling at 325 m/s with the induced oscillations centered around it. By turning the optical field off rapidly, the force on the molecules can be switched off and the molecules can be left at approximately the same velocity at the time the field was turned off. The intensities of the two lattice beams determine the well depth and therefore the amplitude of the velocity oscillation. This process can

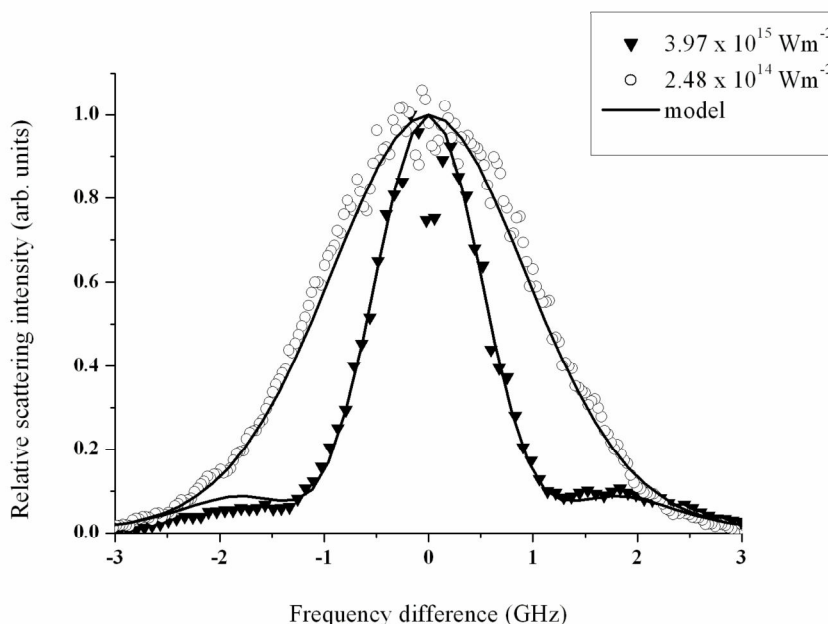


FIGURE 5. CRS spectral profile for two laser beam intensities showing a transition for the weak perturbation regime to the strong perturbation regime and the concomitant narrowing of the spectrum.

be used to create molecular beams over a wide range of velocities with little broadening [6]. Using this scheme we have decelerated and accelerated a range of molecules and atoms over length scales of less than 0.54 microns. By utilizing deceleration, we have been able to reduce the kinetic energy of the molecules in a cold molecular beam by up to 100% in a single laser pulse over a few nanoseconds, thus demonstrating the precise control of molecular-beam velocity that is possible with optical forces [11].

The results of this process are shown in Fig. 7, where we have also compared the experiment with a simulation of the process. The initial distribution function is calculated from the recorded time-of-flight spectrum obtained in a time-of-flight mass spectrometer following ionization of molecules by a UV laser. The molecular beam has an initial velocity of 420 m/s and a temperature of 1.6 K. A peak in the velocity distribution function at 310 m/s before deceleration is due to an isotope of benzene. The velocity distribution is measured 60 ns after the lattice, which was travelling at 187 m/s, has been turned off, showing a fraction (8%) of the molecules that were within the lattice at 0 m/s in the laboratory frame. The simulation reproduces the overall structure of the decelerated velocity distribution function observed in the experiment, and indicates that molecules in the lattice undergo the half oscillation required for deceleration to approximately 0 m/s. The deceleration occurred over 4 ns and indicates a deceleration of greater than 10^{11} g .

CONCLUSIONS

I have provided a review of key experiments that have been carried out to explore the ability to control the motion of gases at room temperature and above. In addition, I have demonstrated how these forces can be used to manipulate the motion of cold molecular beams. Such controlled light-matter interactions are now providing useful means for measurement of gas flow properties. The application of strong optical forces to gases is relatively new and there are still many potential applications that, although have been explored theoretically, are yet to be realized in the laboratory. These include the acceleration of gases to hyperthermal velocities [12] and the creation of jets in static gases for gas heating and mixing [13]. We have only just begun to use these techniques to create slow cold molecules that can be trapped for long time periods by optical fields. Trapping and storing molecules within a light

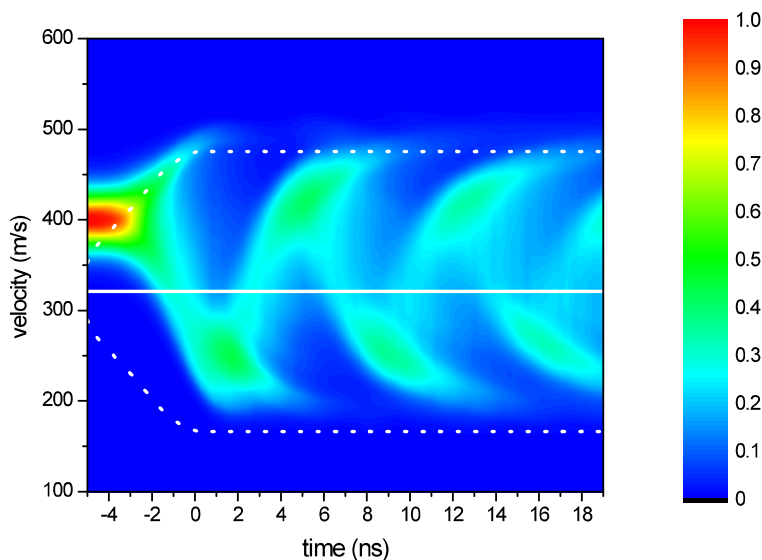


FIGURE 6. A simulation illustrating the oscillation of the distribution function of molecules in a cold jet (1 K) in an optical lattice. The distribution function oscillates around the optical lattice velocity, shown as a solid white line. The color bar shows the relative density of molecules at each velocity.

field will allow us to study detailed collisional interactions of a wide range of gases at an unprecedented resolution [14].

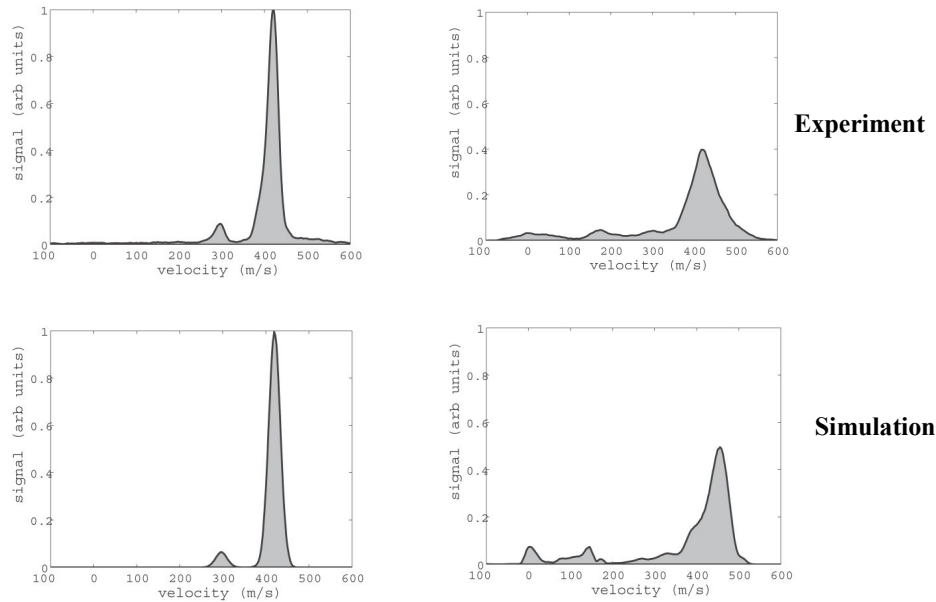


FIGURE 7. Measurements and a simulation of the velocity distribution function of the molecular beam before (on the left) and after (on the right) deceleration of benzene. The deceleration of the molecular beam originally travelling at 420 m/s occurred over 4 ns and produced a significant fraction of the molecules (8%) at 0 m/s.

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