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Simultaneous deceleration of atoms and molecules in a supersonic beam

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Abstract
A unique property of Zeeman effect based manipulation of paramagnetic particle’s motion is the ability to control velocities of both atoms and molecules. In particular the moving magnetic trap decelerator is capable of slowing and eventually trapping mixtures of both cold atoms and cold molecules generated in a supersonic expansion. Here we report the deceleration of molecular oxygen together with metastable argon atoms. The cold mixture with temperature below 1 K is slowed from an initial velocity of 430 m s\(^{-1}\) down to 100 m s\(^{-1}\). Our decelerator spans 2.4 m and consists of 480 quadrupole traps. Our results pave the way for the study of sympathetic cooling of molecules by laser cooled atoms.

The production of cold and ultracold molecules is a growing field fueled by many possible applications in different disciplines such as cold chemistry, precision spectroscopy and degenerate quantum gas research. The internal molecular degrees of freedom such as rotation or vibration, which are absent in atoms, offer many opportunities. However, they also introduce complexity, presenting new experimental challenges, such that novel methods need to be developed to gain access and control over the additional degrees of freedom.

Despite the growing effort, production of ultracold molecular ensembles is still a formidable experimental challenge with no proven and general methods for cooling and trapping. Laser-cooling, while largely successful and straightforward for particular atoms, can only be applied to a small set of molecules with favorable vibrational transitions [1–3]. Several alternative approaches for producing cold molecules are currently being pursued in various labs around the world. The indirect strategy uses laser-cooled alkali-metal atoms, which are assembled into ultra-cold molecules via photo-association [4] and magneto-association[5, 6]. Utilizing the well established techniques from the field of cold atoms, few bi-alkali molecules have been produced at ultra cold temperatures and even close to the quantum degeneracy limit [7]. Direct approaches include buffer gas cooling, where molecules are cooled by collisions with helium atoms in cryogenic environment [8]. The obtained temperatures are typically in the range of 0.5–4 K where even lower temperatures can be reached by applying velocity filtering [9]. Using these techniques a large variety of atomic and molecular species have been cooled and few paramagnetic molecular species have been trapped in deep magnetic traps generated by superconducting coils [10]. Finally the efficient cooling of both internal and translational degrees of freedom taking place during adiabatic expansion of supersonic beams can be combined with a variety of methods that allow for the manipulation of the beam mean velocity.

The benefits of supersonic beams as a source of cold atoms or molecules have been already realized half a century ago. Here, the high flux of atoms and small molecules can be produced with all the degrees of freedom cooled to about 1 K but translationally accelerated in the forward direction to velocities of several hundreds to thousands of meters per second. Supersonic beams are produced by the adiabatic expansion of a high pressure gas through a small aperture into vacuum. In addition to the carrier gas, which is typically a noble gas such as He, Ne, Ar or Kr, many other atoms and molecules can be entrained or seeded into the beam by means of laser ablation or passing the beam through a vapor cell. For some applications, such as merged beams cold-collision experiments [11], the high beam velocity in the lab frame of reference does not impose a fundamental limit. However, in order to approach quantum degeneracy it is necessary to apply additional phase space compression
schemes which require decelerating and trapping of the molecules, while ideally conserving the initial phase space density (PSD). During the last two decades, experiments began to explore the possibility of reducing the mean velocity of supersonic beams by dynamically controlling inhomogeneous electromagnetic fields. Polar molecules [12, 13] and Rydberg atoms and molecules [14] can be decelerated using inhomogeneous electric fields, whereas paramagnetic atoms’ and molecules’ velocity can be manipulated using interaction with high magnetic fields [15, 16]. High intensity optical fields have been used to decelerate both atoms and molecules [17]. The most promising method to bring a supersonic beam to stop involves deceleration in a true three dimensional trap. In an ‘ideal’ adiabatic decelerator, particles can be brought to rest without losses and heating, thereby conserving the initial PSD [18].While molecules with different mass to dipole moment have been co-decelerated [19], the moving magnetic trap decelerator offers the unique advantage of allowing both atoms and molecules, with different mass to magnetic moment ratio, to be co-decelerated and trapped in a magnetic trap. In addition, our decelerator’s acceptance has a phase space volume that is two orders of magnitude larger than in other deceleration methods [20]. This opens a path towards application of either direct evaporative or sympathetic cooling of the molecules using high densities of co-trapped atoms as a thermal bath. According to DSMC gas flow simulation, very high densities of light atoms, up to $10^{13}$ atoms cm$^{-3}$ [21], amenable to laser cooling can be entrained into the supersonic expansion. A possible example is atomic lithium co-decelerated with NH molecule. Initial theoretical studies show that sympathetic cooling is favorable in this system [22].

In our approach a moving magnetic trap is created by a series of spatially overlapping quadrupole traps that are activated by a temporally overlapping pulse sequence. Each current pulse follows a half-sine shape. The timing sequence is illustrated in figure 1(B), where every consecutive trap it activated at the peak current of the preceding trap. As the velocity of the trapping region decreases, the current pulse duration has to increase accordingly. For constant deceleration, $a$, the amount of kinetic energy, $\Delta E_k$, that is removed by each trap is constant and given by $\Delta E_k = M \Delta x$, where $M$ is the mass of the particle and $\Delta x$ is the distance between adjacent traps. The maximal attainable deceleration $a_{\text{m}}$ is approximated by $a_{\text{m}} \approx a \mu B_{\text{m}} / M \Delta x$ where $\mu$ is the magnetic moment of the particle and $B_{\text{m}}$ is the maximal magnetic field that is generated by the front coil of the trap. The necessary current pulse waveform is conveniently generated by discharging a capacitor with capacitance $C$ through an inductor with inductance $L$, which results in a period of $2\pi \sqrt{LC}$. In addition, a thyristor is added in series to the LC circuit. The thyristor here plays two roles: first, it allows to set the discharge time of the capacitor by applying an electric pulse to its gate electrode. Second, it acts as a diode that opens the circuit after half an oscillation period when the current direction changes sign. Unlike the case of a multistage Zeeman decelerator,
the current is switched off at zero value. This property significantly relaxes the requirements from the electronic components.

In our earlier work we demonstrated deceleration of metastable neon from 430 m s\(^{-1}\) to 50 m s\(^{-1}\) using 213 overlapping traps extending over 1.14 m [23]. While this first generation decelerator has been successful as a proof-of-principle experiment, it had a few limitations that impeded its use in trapping experiments. First, its short length imposed a limit on the initial velocity or mass to magnetic moment ratio of the decelerated particles. The second shortcoming was the lack of flexibility in generating variable duration current pulses. In the proof-of-principle experiment, variation in pulse duration was achieved by adding an additional variable inductance in series with the traps. Only limited tunability can be achieved using this method, requiring a different set of fixed capacitors per each trap circuit for different deceleration sequences, along with high voltages reaching 900 V. Both of these issues have been solved in our new moving magnetic trap decelerator.

The new decelerator consists of 480 traps and spans 2.4 m long. Although we have scaled up the system by more than a factor of two, we have reduced its overall complexity by developing a new design of the high current driving electronics. The main improvement in the new design is moving from a dedicated capacitor per trap to ten configurable LC modules (CLCM), which drive all the traps. A single CLCM consists of three parts: a variable binary inductor (Lbox), a variable binary capacitor (Cbox) and a boost converter circuit (BCC). The schematic diagram of the electronic circuit is depicted in figure 1(A). The variable binary capacitor is made of seven capacitors with capacitances of 1, 2, 4, ..., 64 \(\mu F\) that can be connected in parallel electronically (using IGBTs) to form an effective capacitor with any capacitance between 1 and 127 \(\mu F\) with a resolution of 1 \(\mu F\). Similarly, the variable binary inductor consists of four coils with inductances of 2, 4, 8 and 16 \(\mu H\), which can be interconnected in series to form an effective inductor having any inductance between 0 and 30 \(\mu H\) with a resolution of 2 \(\mu H\). The boost converter power supply has a capacitor bank of 40 mF, which is charged to 150 V prior to the deceleration sequence and stores all the required energy. In addition, a BCC charges the Cbox to 600 V after each discharge cycle during the deceleration sequence. Since each CLCM drives 48 traps along the decelerator, multiplexing is achieved by having a thyristor in series with each trap. Two home built field programmable gate array boards are used to control the CLCMs and activate the traps by triggering the thyristors.

The mechanical dimensions of the traps are similar to the first generation design. Each trap is made of two coils where the front (back) coil consists of 4 \(\times\) 4 (2 \(\times\) 4) turns of 26 AWG magnetic wire. The coils’ bore diameter is 10.2 mm and the center to center distance between two neighboring traps is 5 mm. Every ten traps are mounted in an aluminum housing and encapsulated by thermally conductive epoxy. The aluminum housing provides structural integrity as well as heat sinking. These modules are mounted over the vacuum tube, which is made of Inconel with an outer diameter of 10 mm and wall thickness of 0.25 mm. The 2.5 m long tube is pumped only from the two sides, which we found to be sufficient when working at repetition rates below 1 Hz. The trap front coil produces a magnetic field of 0.8 Tesla at the peak current of 500 A. In the first half of the decelerator, where the velocity is high and the pulse duration is short (20–30 \(\mu s\)), the Lbox is bypassed (reducing parasitic resistance) and pulse duration tuning is achieved with the Cbox alone. This allows us to increase the peak current in this deceleration section to 600 A.

In order to demonstrate the capability of the moving trap decelerator to simultaneously slow atoms and molecules, we chose a mixture of molecular oxygen together with metastable argon (Ar*). While many radicals are paramagnetic in their ground-state, they need to be produced and seeded or entrained into the supersonic beam, whereas molecular oxygen, which is paramagnetic as well, can be directly used in a supersonic expansion. Therefore, a very large number of \(O_2\) molecules can be obtained and decelerated [25–27]. The electronic ground state \(\Sigma_g^+\) of \(O_2\) has two unpaired electrons, which in the ‘fully stretched’ state corresponds to magnetic moment of \(\approx 2 \mu_B\) (Bohr magneton) and a mass to magnetic moment ratio of 16 amu/\(\mu_B\). Due to the bosonic nature of \(^{16}O_2\) its rotational states can take only odd numbers. In addition, due to spin–rotation interaction \((S = 1, N = 1)\) the ground state is split to \(J = 0, 1\) and 2 levels. The energies of the Zeeman sub-levels as a function of the magnetic field are presented in figure 2(A). Out of the nine states, four are low field seekers. However, only the \(J = 2, \mathit{m}_J = 2\) has a linear Zeeman shift with a magnetic moment that is large enough at the relevant magnetic fields (below 1 Tesla) to undergo deceleration down to the lowest velocity. The metastable \(^3\)P\(_2\) state of argon exhibits a linear Zeeman effect, where the two low-field seeking sub-levels have a mass to magnetic moment ratio of 13 amu/\(\mu_B\) \((\mathit{m}_J = 2)\) and 26 amu/\(\mu_B\) \((\mathit{m}_J = 1)\).

Our experimental apparatus is presented in figure 1(C). A supersonic beam with a mixture of 99% Ar and 1% \(O_2\) is produced using an Even–Lavie pulsed valve [28]. A small fraction of the neutral Ar beam is excited to the 4\(S\) \(^3\)P\(_2\) metastable state by electron impact using a dielectric barrier discharge [29], which is mounted near the orifice of the nozzle. We cool the valve to 125 K at a stagnation pressure of 3.5 bar in order to reduce the initial mean velocity of the supersonic beam to 450 m s\(^{-1}\) with FWHM of 50 m s\(^{-1}\). The beam passes a 4 mm diameter skimmer, mounted 15 cm from the valve, before it reaches the first quadrupole trap that is located 37 cm from the valve. At the output side of the decelerator a micro-channel plate (MCP) detector is positioned 9 cm from the center of the last trap. A five fold increase in the number of Ar* atoms in fully stretched \(\mathit{m}_J = 2\) state is achieved by
means of optical pumping on the $^3\text{P}_4\text{s}(3/2) \leftrightarrow 4\text{p}(5/2)$ transition with a circularly polarized laser light at 811 nm. While Ar* atoms can be detected directly using the MCP, the O$_2$ has to be first ionized by resonance enhanced multiphoton ionization (REMPI) process, where only the product ions are then detected by the MCP. The 2+1 REMPI process consists of the spectroscopically resolved two-photon transition $C_3\sigma^+\Pi_g (\nu' = 2) \rightarrow X^\Sigma^-(\nu' = 0)$ at 287 nm, followed by a third photon that ionizes the molecule in a bound to continuum transition. We use 15 mJ nanosecond laser pulses generated by a pulsed dye laser (Sirah Cobra-Stretch pumped by Spectra Physics Quanta-Ray Pro) and focused by 200 mm lens onto the beam about 1 cm from the MCP. Figure 2(B) shows a measured REMPI spectrum of O$_2$ that was guided through the decelerator. This transition only addresses the population in the $J = 2$ manifold.

The experimental results are presented in figure 3 and consist of seven TOF traces including the free flight (decelerator is off) and decelerated bunches at final velocities in the range of 350–100 m s$^{-1}$ for both O$_2$ (blue line) and Ar* (red and inverted). In the case of Ar* the MCP amplified signal was acquired with 20 averages. To obtain time trace for O$_2$, the delay of the REMPI laser pulse was scanned with respect to deceleration sequence and the resulting ion signal was integrated for each delay time. The plotted traces of O$_2$ are the result of averaging over 4–16 measurements. The results for the Ar* and O$_2$ were acquired separately since they are measured with...
the same MCP, but require different gain settings for the current amplifier due to the two distinct detection schemes. Otherwise, the experiment sequences were identical. Each of the two sets were normalized according to the peak signal of the final velocity of 350 m s$^{-1}$ and the free TOF traces were multiplied by 100 for clarity. The lowest final velocity that we report here is 100 m s$^{-1}$, limited only by the expansion that occurs during the free propagation from the last decelerator coil to the detector. One can observe a small delay in the detection of Ar* compared to O$_2$ which increases with higher deceleration values. This is attributed to a small difference in the TOF distance, which for oxygen molecules is shorter since they are ionized about 1 cm in front of the MCP detector.

In figure 4(A) we plot the relative total number of Ar* atoms and O$_2$ molecules that were measured as a function of the final velocity. Both curves were separately normalized to the final velocity of 300 m s$^{-1}$. In both species there is a slight increase in the total number at a final velocity of 300 m s$^{-1}$ as compared with 350 m s$^{-1}$, due to the asymmetric trapping potential which at low deceleration values is limited by the height of the back effective barrier.

In the case of O$_2$, we see a drop in particle number at a final velocity of 250 m s$^{-1}$, which occurs due to the loss of the population in $|J_m=2, J=|0\rangle$ sub-level, which can no longer be decelerated. The same effect is absent in the case of Ar* due to the optical pumping to the $m_J=2$ state. The drop in particle numbers at the lowest final velocity can be partially explained by the reduced detection efficiency due to the slow bunch expansion during the free flight. The effective trapping potentials in the decelerated frame of reference with a final velocity of 100 m s$^{-1}$ are plotted in figure 4(B). At this deceleration value the trap depth of Ar* is more than twice as deep as that of O$_2$ due to the preferable mass to magnetic moment ratio.

In summary, we demonstrate for the first time the capability of the moving magnetic trap decelerator to simultaneously decelerate molecules and atoms. We have slowed molecular oxygen together with Ar* in a supersonic beam from an initial velocity of 430 m s$^{-1}$ to a final velocity of 100 m s$^{-1}$, removing 95% of the kinetic energy. One of the promising schemes to produce ultra-cold molecules is sympathetic cooling via collisions with laser cooled atoms. Therefore, decelerating a cold (300 mK) atom–molecule mixture into a permanent magnetic trap brings us within reach of testing such a cooling scheme. Although Ar* has been used in magneto optical trap (MOT) and laser cooled to a temperature below 1 mK [30, 31], it has a few disadvantages. First, various quenching processes limit the maximal number of atoms that we can create within the supersonic expansion to below $10^8$. More importantly, metastable argon collisions with neutral molecules may lead to Penning ionization losses that are not suppressed by spin polarization as in the case of metastable helium. Lithium on the other hand, is a more promising candidate for sympathetic cooling. With a mass to magnetic moment of 7 amu/μ$_B$, $^7$Li can easily be decelerated after being entrained into a supersonic beam [32]. Quantum degenerate gas of $^7$Li is routinely produce in many labs where the bosonic $^7$Li is used to sympathetically cool the fermionic $^6$Li. In addition, lithium was laser-cooled in Ioffe–Pritchard magnetic trap using a single laser beam [33]. Another important advantage of using a mixture of atoms and molecules may arise when considering the application of evaporative cooling [34]. Here, density and the elastic collision cross-sections are the most important parameters. However, most paramagnetic molecules are not easily produced in large numbers and the evaporation process itself trades the number of particles for lower temperatures. This problem can be
circumvented by having a large number of coolant atoms together with the desired molecule. In this case, one can apply forced evaporation only to the atoms, without sacrificing the number of molecules.

An alternative approach to obtain a mixture of molecules and atoms is to decelerate the molecules and merge them with a cloud of ultracold atoms in a MOT. Although MOT can reach atoms number as large as $10^{10}$ at density of $10^{12}$, the contradicting requirements of large optical access on one hand and a small magnetic trap on the other, impose a significant technical challenge. Whereas our approach has the potential of obtaining higher densities, good spatial overlap between atoms and molecules and with an overall reduced complexity.

Currently our group is focused on building a permanent magnetic trap, where mixtures of atoms and molecules can be stored for longer times, permitting studies of collisional properties at low trapping temperatures. We are also exploring efficient lithium entrainment methods.

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References

[1] Hummon M T, Yeo M, Stuhl B K, Collopy A L, Xia Y and Ye J 2013 Phys. Rev. Lett. 110 143001
[2] Barry J F, McCarron D J, Norrgard E B, Steinecker M H and DeMille D 2014 Nature 512 286–9
[3] Zhelevaova V, Cournois A, Wall T, Matsushima A, Hudson J J, Hinds E, Tarbutt M R and Sauer B E 2014 Phys. Rev. A 89 053416
[4] Sage J M, Sains S, Bergeman T and DeMille D 2005 Phys. Rev. Lett. 94 203001
[5] Danzl J G, Haller E, Gustavsson M, Mark M J, Hart R, Bouloufa N, Dulieu O, Ritsch H and Nagerl H C 2008 Science 321 1062–6
[6] Ospelkaus S, Pe’er A, Nï K, Zirbel J, Neyenhuis B, Kotochigova S, Julienne P, Ye J and Jin D 2008 Nat. Phys. 4 622–6
[7] Ospelkaus S, Ni K K, de Miranda M H G, Neyenhuis B, Wang D, Kotochigova S, Julienne P S, Jin D and Ye J 2009 Faraday Discuss. 142 351–9
[8] Hutzler N R, Lu H I and Doyle J M 2012 Chem. Rev. 112 4803–27
[9] Chevrenkov S, Wu X, Bayerl J, Rohlfs A, Gantzler T, Zeppenfeld M and Rempe G 2014 Phys. Rev. Lett. 112 013001
[10] Weinstein J D et al 1998 Nature 395 148–50
[11] Henson A B, Gersten S, Shagam Y, Narevicius J and Narevicius E 2012 Science 338 234–8
[12] Bethlem H L, Berden G and Meijer G 1999 Phys. Rev. Lett. 83 1558
[13] van de Meerrakker S Y, Smeets P H, Vanhaecke N, Jongma R T and Meijer G 2005 Phys. Rev. Lett. 94 023004
[14] Yamakita Y, Procter S R, Goodgame A L, Soffity T P and Merkt F 2004 J. Chem. Phys. 121 1419–31
[15] Narevicius E, Libson A, Parthey C G, Chavez J, Narevicius J, Even U and Raizen M G 2008 Phys. Rev. Lett. 100 093003
[16] Vanhaecke N, Meier U, Andrist M, Meier B H and Merkt F 2007 Phys. Rev. A 75 031402
[17] Fulton R, Bishop A L, Shielde M N and Barker P F 2006 Nat. Phys. 2 465–8
[18] Osterwalder A, Meek S A, Hammer G, Haak H and Meijer G 2010 Phys. Rev. A 81 051401
[19] Fitch N J, Estes D A, Fabrikannt M I, Bries T C, Shyur Y, Parazzoli L and Lewandowski H 2012 J. Mol. Spectrosc. 278 1–6
[20] Lavert-O, Ofir E, David L, Henson A B, Gersten S, Narevicius J and Narevicius E 2011 Phys. Chem. Chem. Phys. 13 18948–53
[21] Even U 2015 Private communication
[22] Wallis E O G, Longdon E J, Zwichtovic P S and Hutson J M 2011 Eur. Phys. J. D: At. Mol. Opt. Plasma Phys. 65 151–60
[23] Lavert-O, Gersten S, Henson A B, Shani I, David L, Narevicius J and Narevicius E 2011 New J. Phys. 13 103030
[24] Brown J M and Carrington A 2003 Rotational Spectroscopy of Diatomic Molecules (Cambridge: Cambridge University Press)
[25] Narevicius E, Libson A, Parthey C G, Chavez J, Narevicius J, Even U and Raizen M G 2008 Phys. Rev. A 77 0531401
[26] Wiederkehr A W, Schmutz J, Motjes M and Merkt F 2012 Mol. Phys. 110 1807–14
[27] Liu Y, Zhou S, Zhong W, Djuricin J and Pomose T 2015 Phys. Rev. A 91 021403
[28] Even U 2014 Adv. Chem. 2014 636042
[29] Luria K, Lave E and Even U 2009 Rev. Sci. Instrum. 80 104102
[30] Gauthier K, Hartl M, Schnelle D, Schnitzehl H, Pfau T and Mlynek J 1998 Phys. Rev. Lett. 81 5298
[31] Busch H C, Shaffer M K, Ahmed E M and Suenkel C I 2006 Phys. Rev. A 73 023406
[32] Borysov M 2012 Phd Thesis The University of Texas at Austin
[33] Schreck F, Ferrari G, Cosin K L, Cubizolles J, Khaykovich L, Mewes M O and Salomon C 2001 Phys. Rev. A 64 011402
[34] Stuhl B K, Hummon M T, Yeo M, Quéméner G, Bohn J L and Ye J 2012 Nature 492 396–400