

Deceleration of continuous molecular beams

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A method for decelerating a continuous beam of neutral polar molecules is theoretically demonstrated. This method utilizes nonuniform static electric fields and regions of adiabatic population transfer to generate a mechanical force that opposes the molecular beam's velocity. By coupling this technique with dissipative trap-loading molecular densities $\geq 10^{11}$ cm $^{-3}$ are possible.

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The electric dipole-dipole interaction between polar molecules is fundamentally different from most interactions between ultracold atoms. While atomic interactions are typically isotropic and comparatively short ranged, the dipolar interaction is strong, long range, tunable, and anisotropic. This interaction can lead to many novel and exciting phenomena, such as quantum chemistry [1,2], field-linked states [3], the possibility for quantum computation [4,5], and long-range topological order [6]. Furthermore, the closely spaced opposite-parity internal levels of molecules, e.g., Ω -doublet, rotational, and vibrational levels, present new possibilities for precision measurement of fundamental physics [7–11].

For these reasons, there has been much effort toward developing techniques to produce cold polar molecules. Current molecular cooling techniques can be characterized as either association of ultracold atoms or direct cooling of molecules. While the association of ultracold atoms, either via a Feshbach [12] or optical resonance [13], is capable of producing molecules near quantum degeneracy [14], these methods restrict experiments to a limited class of molecules, namely, those composed of laser-cooled atoms. Conversely, direct cooling techniques, such as buffer gas cooling [15] and Stark deceleration [16] are capable of producing cold samples from a wide range of polar molecular species. However, despite much work, the molecular density currently attainable via these methods is limited to $\lesssim 10^{7-8}$ cm $^{-3}$. In the case of buffer gas cooling, the molecular density is limited by the presence of the helium buffer gas, which prevents further cooling; for Stark deceleration the poor efficiency ($\sim 0.01\%$) of decelerating molecules from a supersonic beam is the limiting factor [17]. Furthermore, the “single-shot” nature of these techniques makes it inefficient to accumulate cold molecules over time.

Here we propose a technique for producing continuous beams of cold polar molecules. When coupled with irreversible trap loading [18,19] this continuous beam deceleration technique circumvents the poor efficiency associated with traditional beam deceleration by allowing the accumulation of molecules over many seconds, similar to the loading of a magneto-optical trap (MOT) [20]. Moreover, this method appears to allow the accumulation of densities appropriate for the study of the interesting phenomena mentioned above.

Because continuous beams are extended in space they are

not amenable to traditional Stark decelerators, which rely on well-timed electric field pulses and a spatially compact pulsed molecular beam [16]. Figure 1 shows a method of deceleration that instead uses static electric fields, coupled with appropriately placed transition regions, to control the molecular internal state. A molecule in a weak-field seeking state (dipole moment anti-aligned with the electric field) exiting the beam source will be decelerated as it moves into the high electric field region between the electrodes of the deceleration stage. If at the center of this deceleration stage the molecular internal state is changed to a less polarized weak-field seeking state, the molecule will exit the deceleration stage with less kinetic energy than when it entered. After exiting the stage, the molecule internal state can be returned to its original state and

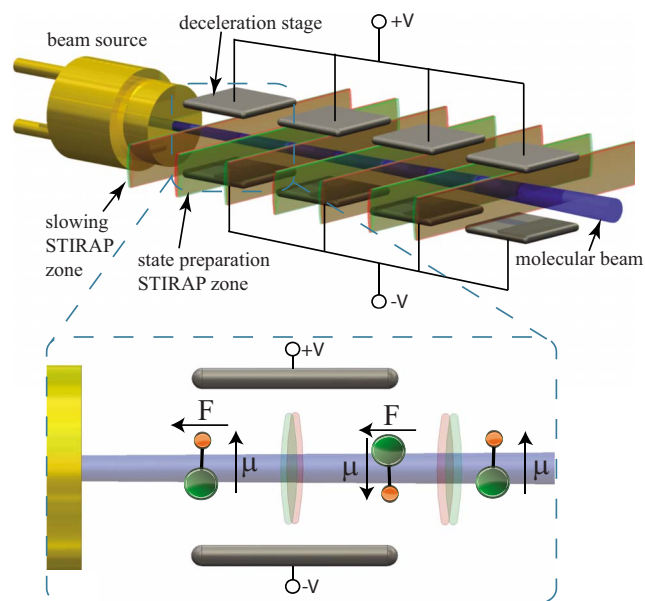


FIG. 1. (Color online) Schematic of continuous beam decelerator. Weak-field seeking polar molecules exiting the beam source are decelerated as they move into the high electric field region between the electrodes of the deceleration stage. At the stage center, STIRAP is used to transfer the molecules to either a less polarized weak-field seeking state or a strong-field seeking state (inset), resulting in net deceleration of the molecular beam. After the deceleration stage, the molecules are returned to their initial internal state via the inverse STIRAP and the process repeated. For clarity, we have omitted the low-voltage electrodes between the deceleration stages, which are used to maintain molecular orientation.

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the deceleration process repeated. These transitions can be driven with near unit efficiency by the technique of stimulated Raman adiabatic passage (STIRAP) [21]. A similar idea for the Rydberg atom acceleration, relying on spontaneous emission rather than adiabatic transfer, has been put forward by Breeden and Metcalf [22].

This approach has several advantages over traditional Stark deceleration. First, this technique allows for continuous guidance of the transverse molecular motion during the deceleration process, which been shown to be the limiting factor in traditional decelerator efficiency [23]. Second, as shown in the inset of Fig. 1, if the molecule's internal state is instead changed to a strong-field seeking state (dipole moment aligned with the electric field) at the electrodes center, the molecule will be decelerated as it moves out of the high electric field region. Since strong-field seeking states typically experience the largest Stark shift, this technique makes it possible to decelerate a beam of molecules to rest with substantially fewer deceleration stages than required in a traditional decelerator. Third, because molecules can be decelerated in strong-field seeking states and transversely refocused in weak-field seeking states, this technique is an alternative to alternate-gradient deceleration [24]. Fourth, this technique is amenable to the use of a buffer gas beam sources [25], which provide higher molecular flux at much lower initial beam speed than room-temperature beams; this is advantageous since the required number of deceleration stages scales with the square of the molecular-beam velocity. Finally, because this technique produces a continuous beam of decelerated molecules it is well suited for dissipative trap loading, either into a static trap [18] or the recently proposed molecular MOT [19].

For definiteness, consider a molecule with $^1\Sigma^+$ ground and excited states, e.g., any compound containing one alkaline-earth atom and one chalcogen atom. The Hamiltonian for the lowest vibrational level of each electronic state in the presence of an electric field is

$$H_o = T_{i,v} + B_{i,v}J^2 + \vec{\mu} \cdot \vec{E}_{dc}, \quad (1)$$

where $T_{i,v}$ is the vibronic term energy operator with eigenfunctions $|\psi_{i,v}\rangle$ where $i=X,A,\dots$, $v=0,1,2,\dots$, and $T_{X,0} \equiv 0$; $B_{i,v}$ is the rigid-rotor rotational constant for the i th electronic level with vibrational excitation v , J is the total angular momentum of the molecule, $\vec{\mu}$ is the electric dipole moment operator, and \vec{E}_{dc} is the externally applied electric field. The eigenvectors of this Hamiltonian are $|\Psi_{i,v,k}\rangle = \sum_J a_J^{i,k} |J, M\rangle |\psi_{i,v}\rangle$ with energies as shown in Fig. 2 as a function of electric field. Here the eigenvector label k is equal to the value of J that describes the zero-field wave function.

For the deceleration scheme considered here, the $|\Psi_{A,1,1}\rangle \leftarrow |\Psi_{X,0,2}\rangle$ and $|\Psi_{A,1,1}\rangle \rightarrow |\Psi_{X,0,0}\rangle$ transitions are used to adiabatically transfer population during the deceleration process. If deceleration by exclusively weak-field seeking molecules is desired (for transverse guiding purposes), the $|\Psi_{X,0,0}\rangle$ state can be replaced by the $|\Psi_{X,0,1}\rangle$ state. Note that a two-photon transition between states with $|\Delta J|=1$ is possible here because the static electric field mixes states of

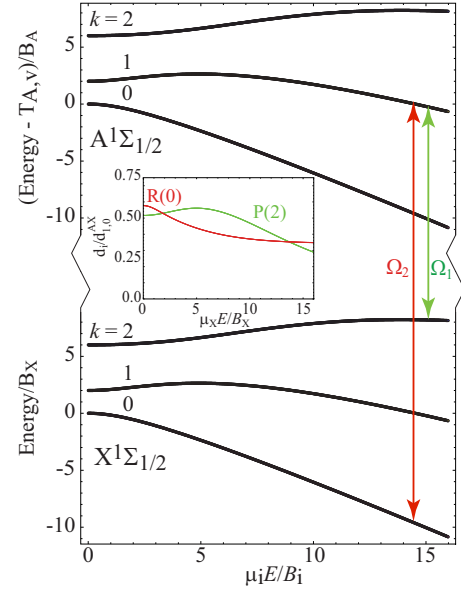


FIG. 2. (Color online) Stark shift of $^1\Sigma^+$ states and STIRAP transfer scheme. Molecules enter the deceleration stage in the $k=2$ state and exit in $k=0$. If the STIRAP transfer takes place at $\mu_x E/B_i=15$ then $\sim 3.1 \text{ cm}^{-1}$ is removed per deceleration stage. The figure inset shows the relevant Hönl-London factors for SrO as a function of electric field.

opposite parity. In the rotating frame, the Schrödinger equation for the evolution of the population of the relevant molecular states is given as [21]

$$i \frac{d}{dt} \begin{bmatrix} c_{0,2}^X \\ c_{1,1}^A \\ c_{0,0}^X \end{bmatrix} = \begin{bmatrix} 0 & -\frac{\Omega_1(t)}{2} & 0 \\ -\frac{\Omega_1(t)}{2} & -i\frac{\Gamma}{2} & -\frac{\Omega_2(t)}{2} \\ 0 & -\frac{\Omega_2(t)}{2} & \Delta \end{bmatrix} \begin{bmatrix} c_{0,2}^X \\ c_{1,1}^A \\ c_{0,0}^X \end{bmatrix}, \quad (2)$$

where Γ is the natural linewidth of the A state in radians per second, $\Omega_n(t) = \frac{d_n E_n(t)}{\hbar}$ with $E_n(t)$ as the envelope of the laser's electric field amplitude, and $|c_{v,k}^i|^2$ is the probability of the molecule being found in the state $|\Psi_{i,v,k}\rangle$. The transition dipole moments are given in terms of the dipole operator, d , as

$$d_n = \langle \Psi_{A,1,1} | d | \Psi_{X,0,2(2-n)} \rangle, \quad (3)$$

which can be calculated from the relevant transition moments:

$$\begin{aligned} \langle \Psi_{A,v} | \langle J+1, M | d | JM \rangle | \Psi_{X,v} \rangle \\ = d_{v',v}^{AX} \sqrt{(J+M+1)(J-M+1)/[(2J+1)(2J+3)]}, \end{aligned}$$

and

$$\langle \Psi_{A,v} | \langle J-1, M | d | JM \rangle | \Psi_{X,v} \rangle = d_{v',v}^{AX} \sqrt{J^2 - M^2/[(2J+1)(2J-1)]}$$

where $d_{v',v}^{AX}$ contains both the electronic and vibrational contributions to the transition dipole moment.

As a specific example, Eqs. (1) and (2) have been solved for the strontium monoxide molecule, SrO,

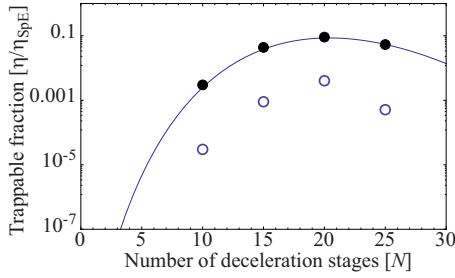


FIG. 3. (Color online) The fraction of total molecules that can be coupled into the trap is displayed versus the number of deceleration stages used. Open (closed) circles are for a traditional (laser-cooled) buffer gas beam of SrO molecules with $T_{\perp}=4$ K ($T_{\perp}=10$ μ K). The line is the maximum trappable fraction as predicted by Eq. (5).

($B_X=0.33798$ cm⁻¹, $\mu_X=8.9$ D, $B_A=0.3047$ cm⁻¹ [26], $\mu_A \approx 2.5$ D [27], $\Gamma=2\pi \times 3.7$ MHz [28], and $d_{1,0}^{AX}=0.43$ D [29]) in a 30 kV/cm electric field ($\mu_X E/B_X \approx 15$). For a geometry similar to Ref. [30], with laser beams of axial and transverse e^{-2} intensity waist $w_A \times w_{\perp}$ of 1×10 mm² and total power of 1 W, the transfer efficiency has been calculated to be $\geq 90\%$ throughout the deceleration process—because the pulse area scales as v^{-1} , the transfer efficiency increases with deceleration stage number, N , as $N^{1/2}$. At this field and with transfer from the $|2,0\rangle$ to $|0,0\rangle$ state at the stage center, ~ 3.1 cm⁻¹ is removed from the molecular kinetic energy at each deceleration stage, allowing a 100 m/s beam of SrO to be decelerated to rest with only 20 deceleration stages.

If the decelerated molecules are loaded into the microwave trap of Ref. [31] via a spontaneous emission loading scheme similar to that of Ref. [18], then molecules exiting the decelerator with a final velocity less than the maximum trappable velocity, v_{trap} , can be accumulated in the trap. Since, all molecules lose the same amount of energy ΔE per deceleration stage these trappable molecules enter the decelerator with a velocity in the range $(\sqrt{\frac{2N\Delta E}{m}}, \sqrt{v_{\text{trap}}^2 + \frac{2N\Delta E}{m}})$. Assuming the buffer gas beam source can be described by the longitudinal velocity distribution,

$$f(v) = \frac{1}{\Delta v} \sqrt{\frac{\ln(2)}{\pi}} \exp\left[-\left(\frac{v-v_0}{\Delta v/\sqrt{\ln 2}}\right)^2\right], \quad (4)$$

the fraction of trapped molecules is then given as

$$\eta = \frac{\eta_{\text{SE}}\eta_{\perp}}{2} \left(\text{erf}\left[\frac{\sqrt{v_{\text{trap}}^2 + \frac{2N\Delta E}{m}} - v_0}{\Delta v/\sqrt{\ln 2}}\right] - \text{erf}\left[\frac{\sqrt{\frac{2N\Delta E}{m}} - v_0}{\Delta v/\sqrt{\ln 2}}\right] \right), \quad (5)$$

where η_{SE} is the efficiency of the spontaneous emission loading [18] and η_{\perp} is the fraction of molecules that remain within the transverse bounds of the decelerator during the deceleration process [17]. For loading of the microwave trap by excitation of the $|\Psi_{A,1,0}\rangle \leftarrow |\Psi_{X,0,2}\rangle$ transition, followed by spontaneous emission on the $|\Psi_{A,1,0}\rangle \rightarrow |\Psi_{X,0,0}\rangle$ transition [32], and assuming excitation saturation, Eq. (3) yields $\eta_{\text{SE}} \approx 0.4$. For reference, Eq. (5) is plotted in Fig. 3 for SrO

($v_0=100$ m/s and $\Delta v=25$ m/s) and the microwave trap parameters of Ref. [31] ($v_{\text{trap}}=2$ cm⁻¹).

The transverse efficiency η_{\perp} depends sensitively on the details of the decelerator design and molecular beam and can only be estimated from detailed Monte Carlo trajectory simulations. Since most of the deceleration takes place in a strong-field seeking state and the strongest fields are near the electrode surface, molecules are defocused transversely as they are decelerated. Thus, a mechanism for transverse refocusing of the molecular beam must be added to the decelerator to prevent complete loss of the beam due to transverse defocusing. For definiteness, complete three-dimensional (3D) Monte Carlo simulations were performed for a decelerator geometry with a hexapole focusing element [33] ($r_0=1$ cm and $V_0=35$ kV) added after every fifth deceleration stage. As the techniques of transversely focusing molecular beams are the subject of much on-going work [23,34], it must be noted that this may not be the optimal beamline. The results of these simulations are shown in Fig. 3 as open circles for a beam of SrO molecules with $v_0=100$ m/s, $\Delta v=25$ m/s, and transverse beam temperature $T_{\perp}=4$ K. At twenty stages of deceleration, η reaches a maximum of ~ 0.002 . Thus, with an initial molecular-beam flux of 10^{13} molecules/s [25], trap volume of $V \approx \frac{4}{3}\pi$ cm³ [31], and background gas limited trap lifetime of $\tau \approx 100$ s, an average trap density of $\rho = K\tau \approx 5 \times 10^{11}$ cm⁻³ is expected.

A recent proposal has shown how certain molecules may be laser-cooled [19] and collected in a MOT. While most molecules are not amenable to the molecular MOT cooling scheme, many molecules possess relatively diagonal Franck-Condon factors allowing the possibility of scattering many photons before optical pumping to undesired states occurs. Thus, cooling only the transverse degree of freedom of a molecular beam, which requires the scattering of a maximum of a few thousand photons, may be possible for many molecular species. As a further check of the model, simulations were also performed for an idealized SrO molecular beam that has undergone transverse laser cooling to $T_{\perp}=10$ μ K. As shown in Fig. 3, as closed circles, the reduced transverse beam temperature makes it possible to recover the maximum trapped fraction as predicted by Eq. (5). Of course, it may be possible to achieve the maximum efficiency, even for $T_{\perp}=4$ K, with more elaborate transverse guiding schemes (e.g., addition of more focusing elements, different order focusing elements, and voltage scaling [23]) and this is the subject of on-going work.

For polar molecules, the collision rate is expected to be accurately given by semiclassical approximations as $\gamma = \frac{2p\mu^2}{3\hbar\epsilon_0}$ and is valid for temperatures larger than $T_{\text{min}} \approx (4\pi\epsilon_0)^2 \frac{\hbar^6}{m^3\mu^4}$, where m is the molecular mass [35]. Since for SrO $\gamma\tau \approx 10^7$ and $T_{\text{min}} \approx 1$ pK, the densities and lifetimes provided by this method are more than sufficient to allow for the study of interesting collisional effects mentioned in the introduction. Furthermore, though more sophisticated treatment is required, it may be possible that these densities also represent a sufficient starting point for evaporative cooling [36].

The main technical challenge in constructing the continuous beam decelerator is the required voltage stability. Since

the two-photon detuning, Δ , depends on the electric field, any change in the field during the deceleration process diminishes the STIRAP efficiency. At 30 kV/cm the energy dependence of the $|20\rangle|\psi_{x,v}\rangle$ state on electric field is weak compared to the field dependence of the $|00\rangle|\psi_{x,v}\rangle$ state. Therefore, Δ depends on electric field as $\Delta \approx \langle \mu_0 \rangle \delta|\vec{E}|$, where $\langle \mu_0 \rangle$ is the ground rotational state laboratory-frame dipole moment (6.9 D for SrO [31]) and $\delta|\vec{E}|$ is any drift of the electric field during the deceleration process. Maintaining maximum STIRAP efficiency requires $\Delta \lesssim 2\pi v/w_A$. Since the STIRAP efficiency is $\propto \sqrt{w_A}/v$, the beam waist may be decreased with the square of the velocity to maintain maximum transfer efficiency. Thus, the tightest constraint on field stability comes from the first deceleration stage which leads to $\Delta \lesssim 2\pi \times 100$ kHz or a required electric field stability of $\delta|\vec{E}|/|\vec{E}| \sim 10^{-6}$. For instance, a 1 cm electrode spacing requires a voltage stability of 0.1 V out of 30 kV; this level of stability can be obtained with commercial power supplies (Matsusada AE/AF Series) with long-term drift compensation. Any inhomogeneity between deceleration stages may be offset by local voltage adjustments. Moreover, it may be possible to *utilize* a field inhomogeneity to produce an adiabatic

rapid passage transfer [37], greatly reducing the constraints on field stability.

In summary, a general technique for producing cold polar molecules has been described. This method promises large improvement over other direct cooling methods by allowing deceleration of a continuous molecular beam as opposed to the single-shot nature of current techniques. By coupling the decelerated molecular beam into a trap, densities $\geq 10^{11}$ cm $^{-3}$ seem attainable, which represent an improvement of up to 4 orders of magnitude over current technology. At these densities it appears that study of the interesting effects predicted for polar molecules is possible. Finally, in this work STIRAP has been used to accomplish the necessary state transfer, however, in the presence of an electric field, parity is mixed and a single photon can transfer population between the relevant levels. Thus, it may be possible to perform direct transfer by THz radiation, further simplifying the design.

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