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Slowing of supersonically cooled atoms and molecules by time-varying nonresonant induced dipole forces

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We describe a versatile method for slowing molecules (or atoms) which relies on high-field-seeking states created by the polarizability interaction with a nonresonant laser field. A pulsed supersonic beam expansion is employed to precool the molecules internally and to narrow their velocity spread. The molecules are scooped at right angles by a nonresonant laser beam steered by a scanner and decelerated on a circular path by gradually reducing the beam's angular speed.

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The scope of magnetic trapping has been recently expanded to include most paramagnetic atoms and molecules. This feat was made possible by the development of the buffer-gas cooling technique [1,2] which relies on elastic collisions with a cryogenically cooled He gas and, therefore, unlike laser cooling [3], is not limited to species with particular energy level patterns. Buffer-gas loading combines naturally with magnetic trapping as both make use of cryogenics. While many atoms are paramagnetic (about two-thirds of the ground-state atoms in the periodic system [1]), few ground-state molecules have the requisite magnetic dipole moment of 1 Bohr magneton or greater [2]. Moreover, magnetic trapping relies on low-field-seeking states that can relax to high-field-seeking states of lower energy [4]. Once this occurs, the high-field seekers are ejected from the magnetic trap and thus are lost. Here we describe a method for slowing molecules (and atoms) based on the nonresonant interaction of a laser field with molecular (or atomic) polarizability, which offers the means to overcome both limitations. The method makes use of a time-varying electric field produced by a laser beam steered by a scanner with a variable angular speed. A pulsed supersonic beam expansion precools the molecules internally and narrows their velocity spread. The molecules are scooped at right angles by the nonresonant laser beam and brought to a halt by gradually reducing the beam's angular speed. The arrested molecules can then be evaporatively cooled [4] and accumulated in a nonresonant light trap [5–8] or another suitable trap [3].

The interaction of a nonresonant laser field with molecular polarizability occurs for any molecule (no matter whether polar or paramagnetic) as all molecules are polarizable, and increasingly so as their size increases [9]. Moreover, for molecules of any other than spherical symmetry the nonresonant interaction creates an induced dipole moment that is coupled to one of the molecular axes [8]. This has been shown [8,10–14] to give rise to directional states (termed *pendular*) in which the molecular axis librates about the electric field vector. The directionality of pendular states arises from hybridization (linear superposition) of the field-free rotor states. The hybridization enhances the induced electric dipole moment over that of a spherical species (i.e., an atom or a spherical top molecule) of the same average polarizability. Since the nonresonant polarizability interaction is purely attractive, the energies of the corresponding eigenstates decrease with increasing field strength (laser intensity) and, therefore, the states are all high-field seeking. At the same time, focused radiation in free space gives rise to a maximum of field strength and thus produces a trap for the high-field seekers. In short, trapping based on the nonresonant polarizability interaction is versatile, lacks relaxation losses, and lends directionality to nonspherical species.

In previous work with atoms, the nonresonant light trap (far-off-resonance trap) was loaded using optical molasses [5] or by transfer of a trapped atomic ensemble from a magnetic or magneto-optic trap [6,15]. Magnetic or magneto-optic traps were also used as stages for producing molecules, via either photoassociation [16,17] or ternary collisions [18] of the ultracold atoms. The resulting translationally cold molecules could then be trapped in a nonresonant light trap [18].

A possible experimental setup which implements the slowing scheme based on the time-varying nonresonant polarizability interaction is shown in Fig. 1. The setup consists of three main parts: a pulsed supersonic beam, a steerable

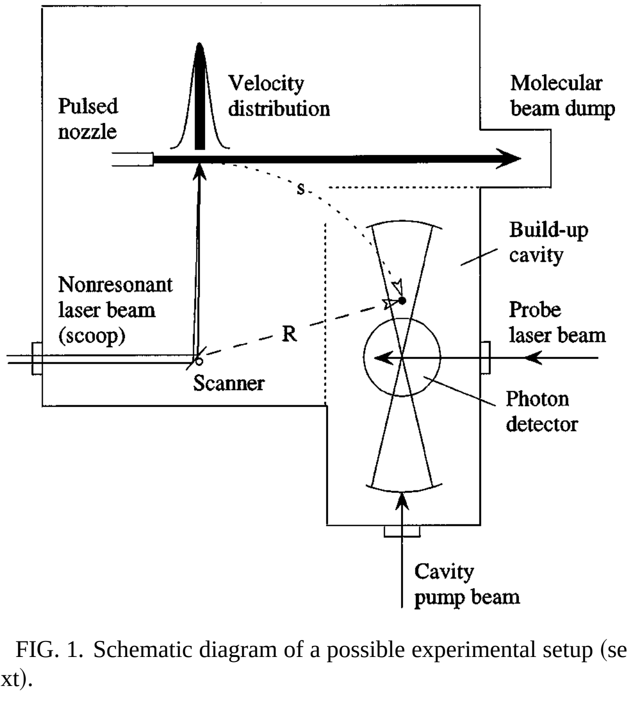

FIG. 1. Schematic diagram of a possible experimental setup (see text).

nonresonant laser beam (referred to as the scoop), and a trap. A pulsed supersonic beam expansion is employed to internally precool the molecules to be loaded and to narrow their velocity distribution. The scoop, initially at right angles to the molecular beam, is swept at the most probable velocity of the molecules, picking up those whose kinetic energy spread falls within the depth of the trap created by the interaction of the scoop with the molecules (the corresponding speed group is indicated by the shaded fraction of the velocity distribution). The angular sweep of the scoop is then gradually brought to a halt on a path of about 1 m while the time-varying dipolar force exerted by the scoop on the molecules counteracts the centrifugal and deceleration forces. The arrested molecules are deposited in the resonator of a build-up cavity (or another type of trap) where they can evaporatively cool and accumulate. We describe a model indicating that the initial number of molecules in a single scoop, given by the terminal temperature (≈ 1 K) and density ($\approx 10^{13}$–10^{14} molecules/cm^3) of the molecular beam expansion, and the depth (≈ 10 mK) and volume of the scoop (10^{-8} cm^3), may range between 10^3 and 10^6.

We limit our consideration to a continuous-wave laser field, $\varepsilon(t)$, interacting adiabatically [14] with a $^1\Sigma$ molecule, treated as a rigid rotor with polarizability components $\alpha_\parallel$ and $\alpha_\perp$ parallel and perpendicular to the axis. The oscillation frequency of the field is far removed from any molecular resonance and much higher than any rotational periods. The field-molecule Hamiltonian thus is averaged over these rapid oscillations, which quenches interaction of the permanent dipole (if any) with $\varepsilon(t)$ and reduces the time dependent factor in the polarizability interaction to $\langle \varepsilon^2(t)\rangle = I$, with I the laser intensity. The strength of the nonresonant interaction is characterized by the parameter $\Delta\omega \equiv \omega_\parallel - \omega_\perp$, with $\omega_{\parallel,\perp} \equiv \frac{1}{2}\alpha_{\parallel,\perp} I/B$ and B the rotational constant of the molecule.[1] In the low-field limit (i.e., for $\Delta\omega \leqslant 2$ [8]) pertinent here (see below), the eigenenergies are available in analytic form and were summarized in Table I of Ref. [8]. For pendular states that correlate with field-free rotor states up to $J \leqslant 10$, the eigenenergies are given, within a factor of unity, by $E \approx -\frac{1}{2}\alpha I$, with $\alpha \equiv \frac{1}{3}(\alpha_\parallel + 2\alpha_\perp)$. Since at rotational temperatures $\leqslant 5$ K, attainable in supersonic expansions [20], the most probable rotational states of typical linear molecules (see below) have $J \leqslant 10$, E is a good approximation to the trap depth for such molecules.

As demonstrated by the far-off-resonance technique for atom trapping [5,6,15], the Gaussian intensity profile of a TEM$_{00}$ mode provides an inhomogeneous field suitable for trapping high-field-seeking states. At any point, the trap depth produced by the nonresonant polarizability interaction is proportional to laser intensity. The TEM$_{00}$ profile,

$$I = (2P/\pi w^2)\exp[-2(r/w)^2],$$

with

$$w \equiv w_0[1 + (\lambda z/\pi w_0^2)^2]^{1/2},$$

P the laser power, r and z the distances from and along the laser beam axis, λ the laser wavelength, and w_0 the beam waist (where $z=0$), has a maximum $I_0 \equiv I(r=z=0) = (2P/\pi w_0^2)$ and the intensity is greater than half the maximum intensity, $I \geqslant \frac{1}{2}I_0$, over a volume $V = 2\pi w_0^4/\lambda$ (the effective volume of the trap).

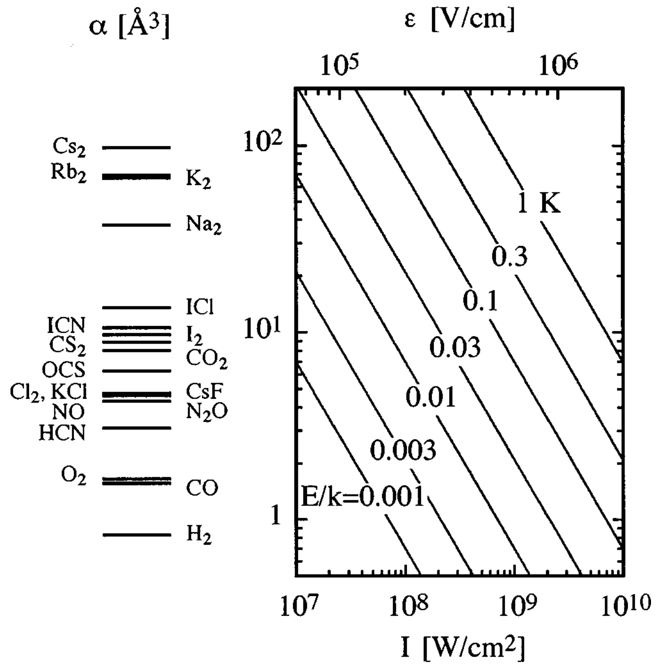

FIG. 2. Nomogram for the trap depth E calculated from the polarizability and laser intensity (lower abscissa) or electric field strength (upper abscissa) for a number of typical linear molecules.

For a laser power $P \approx 100$–300 W and waist $w_0 \approx 5$–50 μm, the maximum laser intensity ranges between 10^7 and 10^9 W/cm^2. Figure 2 provides a nomogram that facilitates estimates of the trap depth E from the polarizability and laser intensity (or electric field strength) for a number of typical linear molecules. One can see that a trap depth of 10 mK should be attainable for all but H$_2$ with laser intensities between 10^7 and 10^9 W/cm^2. In this intensity range the interaction of most of the molecules included in the nomogram is well described by the assumed low-field limit. Notable exceptions are the Rb$_2$ and Cs$_2$ molecules, whose interaction with the laser field reaches into the strong-field limit and so the corresponding trap depths are even greater.

The ability of the scoop to decelerate molecules depends on the attainable balance between the deceleration and centrifugal forces on the one hand, and the counteracting tangential and radial forces exerted by the scoop on the other. The latter, proportional, respectively, to $\alpha\partial I(r,z)/\partial z$ and $\alpha\partial I(r,z)/\partial r$, are about equal to one another over the volume of the scoop and can be well approximated by $E/w_0 \approx F_{scoop}$. The centrifugal and deceleration forces are, respectively, given by $F_{cent} = mv^2/R$ and $F_{dec} = ma$, with m and v the molecular mass and velocity, a the deceleration, and R the perpendicular distance between the scanner and the

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[1] With quantities expressed in customary practical units, $\omega_{\parallel,\perp} = 1.438 \times 10^{-11} \alpha_{\parallel,\perp}$ [Å^3] I [W/cm^2]/B[K].

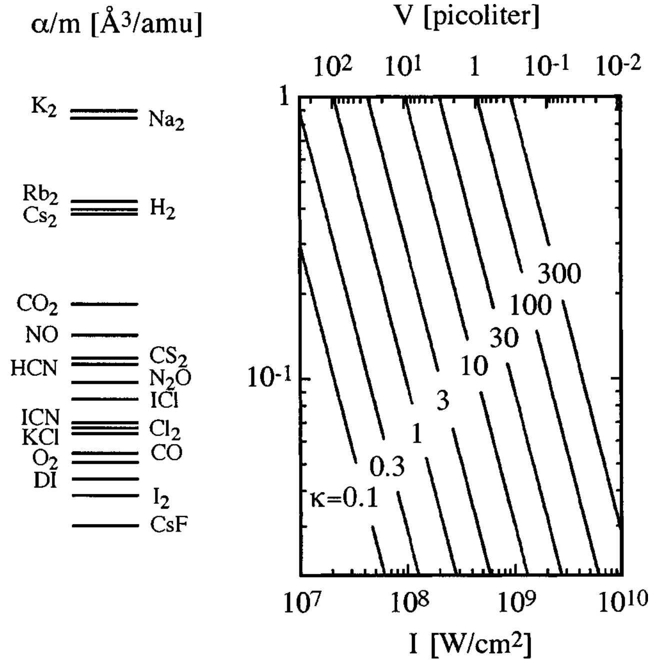

FIG. 3. Nomogram for the ratio $\kappa \equiv |F_{scoop}/F_{dec}|$ calculated for $R=s=1$ m and $v=0.5$ km/s as a function of laser intensity and the ratio of the polarizability to molecular mass for the same molecules as in Fig. 2. The upper abscissa shows the trap volume V obtained by assuming a laser power of 200 W and a diffraction-limited waist of 10 μm.

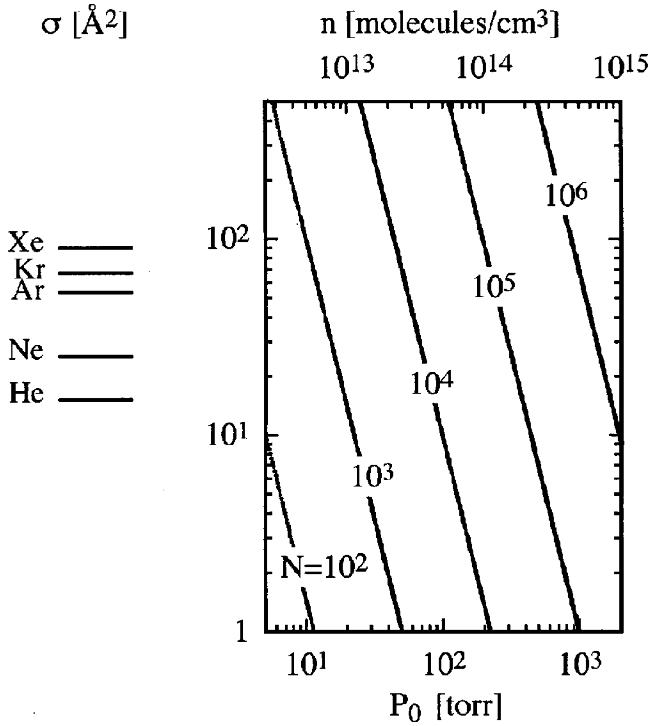

FIG. 4. Nomogram for the number of molecules N seized by a scoop of depth $E/k=10$ mK and volume $V=10$ pl from a supersonic molecular beam seeded in a monatomic carrier gas as a function of source pressure P_0 (lower abscissa) or molecular number density n (upper abscissa); calculation based on a source temperature of 300 K, nozzle diameter of 0.05 cm, 10% concentration of the seeded molecules, and a pick-up distance of 5 cm from the nozzle. The ordinate shows the corresponding gas-kinetic cross sections needed to attain the requisite terminal temperatures.

molecular beam (or the laser beam's waist, see Fig. 1). For $R=1$ m, $v=0.5$ km/s (see below), and a deceleration path s of 1 m, $F_{cent} \leqslant F_{dec}$, and so the balance of the forces involved can be characterized by a ratio $\kappa \equiv |F_{scoop}/F_{dec}|$. Note that the corresponding deceleration time is ≈ 5 ms and the deceleration $a \approx 10^7$ cm/s^2; the scanner that steers the scoop has to rotate initially at an angular speed of ≈ 250 rad/s. Figure 3 shows a nomogram that surveys the attainable values of κ as a function of laser intensity and the ratio of the polarizability to molecular mass for the same molecules as in Fig. 2. The upper abscissa shows the volume of a scoop produced by a 200 W laser beam focused to a diffraction-limited waist of 10 μm that provides the intensity shown on the lower abscissa. One can see that for most of the listed molecules, the balance is favorable at laser intensities ranging, again, between 10^7 and 10^9 W/cm^2.

The total number, N, of molecules that can be seized by a single scoop of a given depth E and volume V is given by $N = fnV$, with n the number density of molecules at the pick-up point and f the fraction of molecules whose velocities range between $v-(2E/m)^{1/2}$ and $v+(2E/m)^{1/2}$ about the most probable velocity, v. Both f and n depend on the parameters of the pulsed supersonic beam expansion. For a seeded supersonic expansion [19], the velocity distribution of the molecules peaks at $v \approx [\gamma k T_0/(\gamma-1)m_c]^{1/2}$, with T_0 the source temperature, γ the Poisson coefficient (γ=5/3 for a monatomic gas), k the Boltzmann constant, and m_c the mass of the carrier gas; the width of the distribution is characterized by the terminal translational temperature, T. The fraction f is independent of mass and T_0 and is found to be given by $f \approx 0.64(3E/kT)^{1/2}$. The terminal temperature can, in turn, be expressed in terms of the source pressure, P_0, nozzle diameter, d, and the gas-kinetic collision cross section, σ, of the carrier gas [19]; for a monatomic carrier $T = 1.72 T_0 (P_0 d\sigma)^{-1.09}$ with P_0 in torr, d in cm, and σ in Å^2. The molecular number density n at the pick-up point (≈ 5 cm downstream from the nozzle) can be approximated by the product of the carrier-gas number density and the concentration of the seed gas, yielding $n \approx 7.6n \times 10^{13} P_0/T_0$ molecules/cm^3 for a 10% concentration and $d = 0.05$ cm. Put all together, the number of molecules seized by a scoop moving at the most probable velocity v is approximately $N \approx 6.41 \times 10^{13} V (E/kT_0^3)^{1/2} (d\sigma)^{0.545} P_0^{1.545}$. Figure 4 provides a nomogram that facilitates estimates of the number of molecules seized by a scoop with $E/k=10$ mK and $V=10^{-8}$ cm^3 from a seeded supersonic molecular beam as a function of the source pressure P_0 (lower abscissa) or molecular number density n (upper abscissa) at a source temperature $T_0 = 300$ K. The ordinate shows the corresponding gas-kinetic cross sections needed to obtain the requisite cooling. Also shown are values of the cross sections for rare gases — the favorite carriers. The above beam parameters are routinely attained in pulsed supersonic expansions, with pulse durations of 50 μs or less and repetition rates of 10 Hz or higher [19]; at the same time, the background pressure for such expansions can be maintained at less than 10^{-5} torr, affording long-enough mean free paths, of about 10 m or more.

The scooped molecules are separated from the molecular beam in less than 0.1 ms, a time significantly shorter than the collision time of over 1 ms within the beam at the pick-up point; this should minimize heating of the scooped molecules. The velocities within the scoop are then distributed according to an apodized Maxwell-Boltzmann distribution pertaining to the terminal temperature T. The apodization is due to the velocities $\pm(2E/m)^{1/2}$ that set the limits on the seized speed group. Since the deceleration time (≈5 ms) is considerably shorter than the (elastic) collision time within the scoop (≈100 ms, assuming a cross section of 100 Å^2), collisional equilibration among the scooped molecules will be suppressed. This should prevent the development of a high-speed tail of the velocity distribution and hence curb evaporation over the edge of the scoop. Only after releasing the decelerated molecules into a trap will there be sufficient time for equilibration. The molecules can be deposited into a static trap if they are polar or paramagnetic, or, better, released into a versatile nonresonant light trap realized by the resonator of a build-up cavity.

A cavity equipped with supermirrors (preferably in the near infrared where there are few molecular resonances) and locked to the TEM_{00} mode of a continuous-wave pump laser is expected to build up powers that are 10^3- to 10^4-fold the pump power [20]. With pump powers between 1 and 10 W, such a build-up cavity would allow creating a trap whose peak intensity is about the same as that of the scoop but whose volume exceeds the volume of the scoop by a factor between 10^3 and 10^4. Molecules deposited by the scoop into such a trap could be evaporatively cooled and accumulated. The intensity of the trapping field could be increased if a higher degree of alignment of the trapped molecules were desirable.

Apart from decelerating atoms or molecules and loading them into traps, the scoop could also be used to accelerate trapped atoms or molecules, including a Bose-Einstein condensate (BEC). In this slingshot mode, the scoop could create bursts of virtually monoenergetic (or, in the case of a BEC, coherent) atoms or molecules with up to hyperthermal velocities determined by the tangential speed of the scoop.

The advent of molecular trapping brought to prominence such areas as the study of ultracold molecular collisions and of collective effects in weakly interacting molecular ensembles. Loading and trapping based on the nonresonant polarizability interaction holds the promise of extending the scope of such studies to most molecules.

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[20] Supermirrors available from, e.g., Newport or Boulder Electro-Optics have reflectivities of over 0.9999 and withstand intensities exceeding 10^6 W/cm^2.