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Collisions of Spin-polarized YO Molecules for Single Partial Waves

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Efficient sub-Doppler laser cooling and optical trapping of YO molecules offer new opportunities to study collisional dynamics in the quantum regime. Confined in a crossed optical dipole trap, we achieve the highest phase-space density of 2.5×10^{-5} for a bulk laser-cooled molecular sample. This sets the stage to study YO–YO collisions in the microkelvin temperature regime, and reveal state-dependent, single-partial-wave two-body collisional loss rates. We determine the partial-wave contributions to specific rotational states (first excited $N = 1$ and ground $N = 0$) following two strategies. First, we measure the change of the collision rate in a spin mixture of $N = 1$ by tuning the kinetic energy with respect to the p- and d-wave centrifugal barriers. Second, we compare loss rates between a spin mixture and a spin-polarized state in $N = 0$. Using quantum defect theory with a partially absorbing boundary condition at short range, we show that the dependence on temperature for $N = 1$ can be reproduced in the presence of a d-wave or f-wave resonance, and the dependence on a spin mixture for $N = 0$ with a p-wave resonance.

There has been immense progress recently in the field of dipolar quantum gases of molecules, culminating in the achievement of both degenerate Fermi gases [1–4] and Bose-Einstein condensate [5]. The strong electric dipole moments of these dipolar gases have allowed the study of dipole-mediated spin many-body dynamics [6] with a further promise to study strongly correlated phases [7, 8] and exotic states of matter [9–11]. However, these degenerate gases have thus far been limited to associated alkali molecules. An alternative route, the direct laser cooling of molecules [12–16] allows a complementary set of neutral molecules to be cooled to the ultracold regime [17–20] for a wide range of applications including the search for the electron’s electric dipole moment [21, 22] and ultracold chemistry [23–25].

The achievement of quantum degeneracy through evaporative cooling is facilitated by two ingredients: an initially high phase-space density, and a good ratio of elastic to inelastic collisions [26, 27]. Thus far, no directly laser-cooled molecules have been brought to quantum degeneracy, largely due to the initial phase-space density of the molecular gas being insufficiently high. A main limiting factor was low transfer efficiencies from a magneto-optical trap (MOT) into conservative traps [28], but this has since been resolved with the demonstration of the first sub-Doppler blue detuned molecular MOT in YO molecules [29]. Subsequently, blue detuned MOTs have also been demonstrated for SrF [30], CaF [31], and CaOH [32] molecules, with bulk-gas collisions reported in an optical dipole trap (ODT) of SrF [30].

Shielding of detrimental inelastic collisions has been proposed and demonstrated via DC electric [2, 33–36] and microwave fields [37–39], resulting in efficient evap-

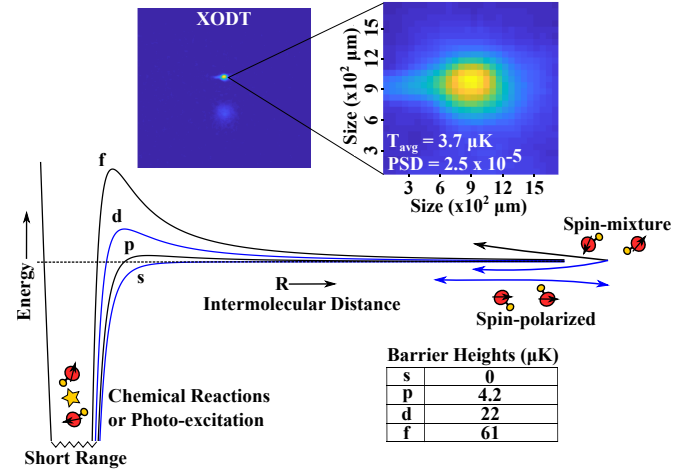


FIG. 1: Top: Images of YO molecules loaded into a crossed optical dipole trap (XODT). The unloaded molecules are seen falling, and a record phase-space density of 2.5×10^{-5} is achieved inside the XODT. Bottom: Molecules interact through a long-range van der Waals potential. At sufficiently low collision energies, distinguishable molecules enter through both even (s,d) and odd (p,f) partial-wave channels, where partial waves beyond s-wave feature a centrifugal barrier. The corresponding centrifugal barrier heights are shown in the table. Since YO is a boson, indistinguishable molecules collide only via even partial waves. Upon reaching short range, molecules undergo inelastic collisional loss.

orative cooling of alkali dipolar molecules [2–5, 40]. With a large dipole moment of 4.5 Debye in the molecular frame, we expect YO to be amenable to both methods

of shielding, ensuring prospects for evaporation.

In this Letter, we report the loading of laser-cooled YO molecules into a crossed ODT (XODT), with a record phase-space density of 2.5×10^{-5} . This achievement is based on robust cooling of YO in both free space and within optical traps, due to its favorable ground-state level structure. YO features a large Fermi-contact interaction between the electron and nuclear spin, resulting in a separation of 760 MHz between the $G = 0$ and $G = 1$ hyperfine manifolds [17]. This structure supports efficient gray molasses cooling (GMC) and we routinely achieve a temperature of $1.5 \mu\text{K}$ in free space, and a few μK in an XODT (top of Fig. 1). This provides favorable conditions to explore YO collision dynamics by tuning the thermal energy to control different partial-wave contributions to the inelastic loss rates. We model the rate coefficients using quantum defect theory (QDT), and show that the temperature dependence can be reproduced in the presence of a d-wave or f-wave scattering resonance. Further, we spin-polarize YO molecules in the absolute ground state $N = 0, G = 1, F = 1$ with the use of microwave sweeps between $N = 1$ and $N = 0$. We compare the collision rates for spin-polarized molecules and an incoherent spin mixture of hyperfine sublevels. The ratio of the rate coefficients for polarized and unpolarized samples can be reproduced in the presence of a p-wave resonance.

In the absence of external static or oscillating electric fields that polarize the heteronuclear molecules in the lab frame, the intermolecular interaction is well described by the long-range van der Waals potential

$$V(R) = -\frac{C_6}{R^6} + \frac{\hbar^2 L(L+1)}{2\mu R^2}, \quad (1)$$

where μ is the reduced mass. The low-energy collision dynamics are dominated by the resulting centrifugal barriers for different partial waves L . As depicted in the bottom of Fig. 1, the symmetrization requirement of the wave-function for a pair of bosonic YO molecules dictates that they interact through even partial waves when they occupy the same internal state. Distinguishable molecules can enter through both even and odd partial waves. For a partial wave that features a centrifugal barrier, the colliding molecules can reach short range only if they can tunnel through the barrier or have sufficient kinetic energy to overcome it. This results in a strong energy dependence on the scale of the barrier heights.

To illustrate the effect of these collision channels on total loss rate, we begin with the case of identical bosons subject only to two-body loss according to the form

$$\dot{n} = -(2k_e)n^2, \quad (2)$$

where $\dot{n}$ represents the density loss rate, and k_e is the even partial wave loss rate coefficient. The factor of two in Eq. 2 arises because each inelastic collision removes two molecules from the trap [41].

In contrast, molecular loss in a spin mixture is modeled by a set of coupled rate equations. For example, the total density for a mixture of three different spin states is

$$n = n_1 + n_2 + n_3. \quad (3)$$

Collisions for both even and odd partial waves occur between different spin states while only even partial waves occur within individual spin states. If there are no purely spin-changing collisions, the two-body loss rate equations for each spin state are

$$\begin{aligned} \dot{n}_1 &= -(2k_e)n_1^2 - (k_e + k_o)n_1n_2 - (k_e + k_o)n_1n_3, \\ \dot{n}_2 &= -(2k_e)n_2^2 - (k_e + k_o)n_2n_1 - (k_e + k_o)n_2n_3, \\ \dot{n}_3 &= -(2k_e)n_3^2 - (k_e + k_o)n_3n_2 - (k_e + k_o)n_3n_1, \end{aligned} \quad (4)$$

where we have introduced the loss rate coefficient k_o for odd partial waves, and assumed both k_e and k_o are independent of spin. This reveals a clear distinction for distinguishable collisions, as each collision gives rise to a loss of a single molecule from each respective spin state. Summing the terms in Eq. 4 while assuming equal population for each spin state, $n_1 = n_2 = n_3 = \frac{n}{3}$, results in a two-body loss rate

$$\dot{n} = -\left(\frac{4}{3}k_e + \frac{2}{3}k_o\right)n^2. \quad (5)$$

Following the example above, the two-body loss rate for collisions between q equally populated spin states scales according to the simple analytical form,

$$\dot{n} = -\left(\frac{q+1}{q}k_e + \frac{q-1}{q}k_o\right)n^2. \quad (6)$$

As a consequence of Eq. 6, the rate coefficient among increasingly large distributions of spin states will simply reduce to the sum of k_e and k_o . Therefore, we can use experiments with different numbers of populated states and temperatures to disentangle the contributions to the total loss rate. In particular, we experimentally access five different conditions: unpolarised $N = 1$ (12 states) at three temperatures; unpolarised $N = 0, G = 1$ (3 states); and polarised $N = 0$ (1 state).

Several physical mechanisms can cause loss. YO is strongly bound, by 7.3 eV [42, 43], so the bond-exchange reaction, $\text{YO} + \text{YO} \rightarrow \text{Y}_2 + \text{O}_2$, is energetically forbidden [44, 45]. It is not known whether the trimer-formation channels $\text{Y}_2\text{O} + \text{O}$ and $\text{YO}_2 + \text{Y}$ are energetically accessible, but loss could also be caused by photo-excitation of the collision complex or three-body collisions. Any of these processes is likely to be enhanced by long-lived “sticky” collisions [46–49]. Molecules prepared in excited states can also be lost by rotational and/or hyperfine relaxation.

To capture the multiple potential causes of loss, we use a QDT approach. The loss is modeled by a short-range

absorbing boundary condition, regardless of the physical mechanism. This is controlled by the loss parameter y , ranging from no loss at $y = 0$ to complete “universal” short-range loss at $y = 1$, and a short-range phase shift δ^s [50–52]. This approach has been successful in modeling loss in similar systems [53, 54]. We calculate cross sections and thermally averaged two-body loss rate coefficients [51] using analytic QDT functions [55, 56], including all partial waves needed for convergence.

The total density loss rate $\dot{n}_{\text{mol}}$ is defined as

$$\dot{n}_{\text{mol}} = -\alpha n_{\text{mol}} - (2k_2)n_{\text{mol}}^2, \quad (7)$$

where α and k_2 are the one-body and two-body loss rate coefficients. The density is determined using the molecule number and V_{eff} which is the effective volume defined as $V_{\text{eff}} = 8\sqrt{\pi^3}\sigma_x\sigma_y\sigma_z$ where σ_i is the rms width of the thermal distribution along each direction in the XODT with $i = \{x, y, z\}$. The rms widths are determined by the equipartition theorem $m\omega_i^2\sigma_i^2 = k_B T_i$, where k_B is Boltzmann’s constant, T_i is the temperature, and m is the mass of YO. The temperature of the molecules is measured by time-of-flight expansion in the radial $\{x, y\}$ direction and the axial $\{z\}$ direction. The trapping frequencies ω_i are determined by parametrically heating the molecules out of the XODT, which is formed by two independent 1064 nm laser beams intersecting at 45 degrees.

To optimally load our XODT, we apply 40 ms of GMC while ramping on the 1064 nm beams. Collisional studies of an incoherent mixture of molecules in different internal states of the $N = 1$ rotational manifold follow naturally after GMC, as the molecules are spread over 12 spin states during the overlap with the XODT beams. The number of molecules left in the XODT as a function of hold time is then measured by applying a 1.5 ms readout pulse. The total loss rate for three different temperatures is shown in Fig. 2. The first trace (blue-filled squares) and the second trace (green-filled circles) is taken at a trap depth of 85 μK . The average temperatures of the molecules, defined as $T_{\text{avg}} = T_{\text{radial}}^{2/3} \times T_{\text{axial}}^{1/3}$, are 3.7(5) μK and 8.3(1.1) μK respectively, which are below and above the p-wave barrier height of 4.2 μK . The third trace (red-filled circles) has a higher trap depth of 150 μK . The average temperature of the molecules is 14.5(1.7) μK , which is well above the p-wave but below the d-wave barrier height of 22 μK . The number of trapped molecules is ~ 5000 in all cases with a 40% capture efficiency from the blue detuned MOT. The one-body lifetime of 590(22) ms is measured at long hold times (Fig. 2 inset).

Using Eq. 7, a weighted fit for the curves in Fig. 2 is performed while fixing α . We do not measure an increase in temperature vs. time within error, hence a fixed trapping volume V_{eff} . The experimental results are $k_2(3.7 \mu\text{K}) = 2.2(0.6) \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$, $k_2(8.3 \mu\text{K}) = 5.6(1.8) \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ and $k_2(14.5 \mu\text{K}) = 4.9(1.4) \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$. The results of our universal-loss QDT

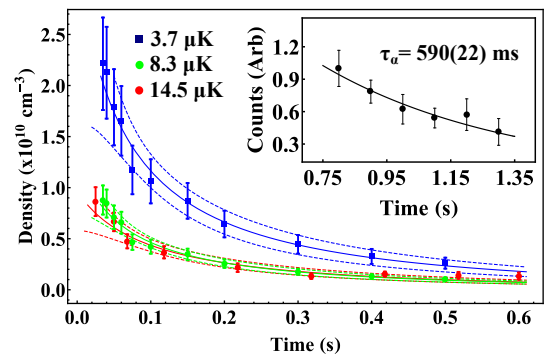


FIG. 2: Inelastic loss in an incoherent spin mixture of YO molecules in the $N = 1$ rotational manifold. The collisional loss rate is measured at three different average temperatures of 3.7(5) μK (blue-filled squares) and 8.3(1.1), 14.5(1.7) μK (green-filled and red-filled circles) which are below and above the p -wave centrifugal barrier height, respectively. The solid lines represent the fit of the data points to the density loss rate according to Eq. 7, and the dashed lines represent the 95% confidence interval bands from the fits. By tuning the temperature below and above the p -wave barrier height we determine two-body loss rates of $k_2(3.7 \mu\text{K}) = 2.2(0.6) \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$, $k_2(8.3 \mu\text{K}) = 5.6(1.8) \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ and $k_2(14.5 \mu\text{K}) = 4.9(1.4) \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$. To quantify the fitting results we find χ^2/ndf of 0.2, 0.3 and 1.4 for the respective fits. The inset shows the one-body lifetime $\tau_\alpha = 590(22)$ ms.

calculations are $k_2(3.7 \mu\text{K}) = 1.3 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ and $k_2(14.5 \mu\text{K}) = 1.5 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$. At the lower temperature, there is a fair agreement between theory and experiment. Both show an increased loss at the higher temperature, although more pronounced in the experiment. Such a feature could arise if there is a resonant enhancement for higher partial waves.

To study collisions in a spin-polarized case, the molecules are next prepared in the absolute rovibrational ground state, the $N = 0, G = 1, F = 1$ manifold, both in an incoherent mixture of the m_F sublevels and spin-polarized in $m_F = -1$. The transfer of the molecules into the absolute ground state begins by optically pumping into the single quantum state $N = 1, G = 1, F = 0$ by applying two laser tones blue detuned from $N = 1, G = 0, F = 1$ and $N = 1, G = 1, F = 2$. This process is shown diagrammatically in Fig. 3(a). A magnetic field of ~ 16 G is applied along z to split the degeneracy of the $N = 0, G = 1, F = 1$ hyperfine sublevels; applying a microwave sweep transfers the molecules into a particular sublevel of the absolute ground state. To produce an incoherent mixture, no magnetic field is applied during the sweep. The resonant frequency that addresses the transition $N = 1, G = 1, F = 0 \rightarrow N = 0, G = 1, F = 1$ transition is 23.282405 GHz [57].

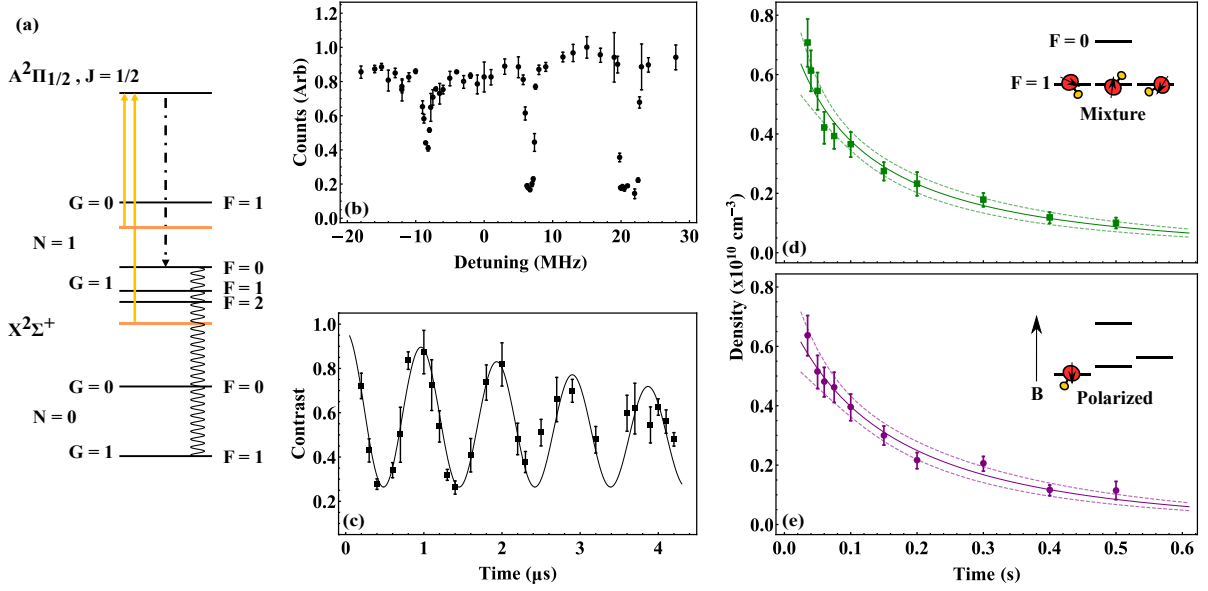


FIG. 3: (a) The optical pumping sequence to transfer molecules in $N = 1$ into a single hyperfine level $G = 1, F = 0$. This is achieved by applying two beams blue-detuned from $N = 1, G = 0, F = 1$ and $N = 1, G = 1, F = 1, 2$. Microwave fields are then swept across the transition $N = 1, G = 1, F = 0 \rightarrow N = 0, G = 1, F = 1$ to transfer the molecules into the absolute ground state. (b) Microwave spectroscopy of the transition with an applied magnetic field, showing splitting of the hyperfine sublevels in $N = 0, G = 1, F = 1$. (c) Rabi oscillations between $N = 1$ and $N = 0$. (d) Inelastic loss of YO molecules prepared in an equal mixture of spin states in $N = 0, G = 1, F = 1$ (green-filled squares). (e) Inelastic loss in single spin-polarized state $N = 0, G = 1, F = 1, m_F = -1$ (purple-filled circles). The solid lines represent the fitting of the data points to the rate equation given in Eq. 7, and the dashed lines represent the 95% confidence interval bands from the fits. To quantify the fitting results we find χ^2/ndf of 0.8 and 0.7 for the respective fits. We determine rate coefficients of $k_{\text{mix}} = 5.4(1.5) \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ and $k_{\text{pol}} = 3.2(1.0) \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ at 5.8 μK in the $N = 0$ ground state.

Microwave spectroscopy is shown in Fig. 3(b) with an applied magnetic field. A clear separation of the three sublevels is observed. As the microwave frequency is red-detuned at zero field, no mixing of the population between $N = 1, G = 1, F = 1, 2$ and $N = 0, G = 1, F = 1$ is observed, suggesting that the optical pumping to $N = 1, G = 1, F = 0$ has high fidelity ($> 95\%$). Figure 3(c) shows Rabi oscillations between the $N = 1$ and $N = 0$ rotational levels in free space. Rabi rates of $\sim 1 \text{ MHz}$ are achieved. Fast dephasing of the oscillations is measured with a coherence time of 8.8 μs . We attribute this to an inhomogeneous microwave field inside our metal vacuum chamber. This limits our coherent-transfer fidelity to the desired sublevel(s) in $N = 0$ to 70%. After removing all unwanted population, the transferred $N = 0$ population is measured as a function of time, recovering the associated collision loss rate.

The collisional loss rates for both an incoherent mixture (purple-filled circles) and spin-polarized YO molecules (green-filled squares) are shown in Fig. 3(d) and (e), all under an XODT trap depth of 85 μK . Due to optical pumping, the molecules are heated to an average temperature of 5.8(5) μK , just above the p-wave barrier height. Fitting to Eq. 7 for the total loss

rates, we determine $k_{\text{mix}} = 5.4(1.5) \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ and $k_{\text{pol}} = 3.2(1.0) \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$. The corresponding calculated values are $k_{\text{mix}} = 1.3 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ and $k_{\text{pol}} = 1.3 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$. The experimental k_{mix} is larger than the theory; this again could be a sign of slight resonant enhancement, most likely in p-wave scattering.

The discrepancy in the data compared to the theory may arise from the density determination, which is derived from the molecule number, temperature, and trap frequencies. However, our results for both rotational states could suggest the presence of resonances, particularly in $N = 1$. We therefore focus on ratios of rates, between high and low temperature for $N = 1$ and between spin mixtures and polarized samples for $N = 0$. Resonances cannot exist for universal loss ($y = 1$) since there is no reflection to produce interference. However, if $y < 1$, resonances occur when the short-range phase shift δ^s produces constructive interference. Our QDT model represents only shape resonances explicitly, but broad Feshbach resonances will give the same observable effects [51, 58]. Figure 4 shows the ratios from the QDT model and compares them with experiment. For $N = 1$, the experimental ratio can occur in two regions with y below 0.25 and δ^s just below $5\pi/8$ and $7\pi/8$; these correspond

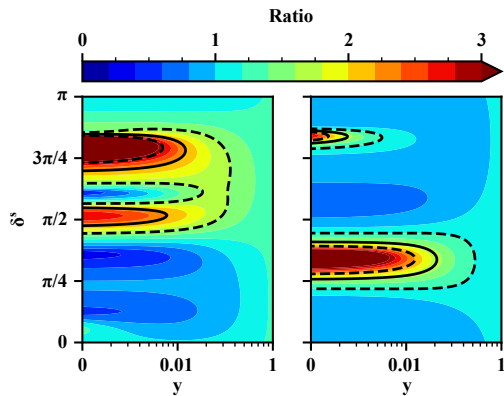


FIG. 4: Contour plots of the calculated ratios $k_2(14.5\,\mu\text{K})/k_2(3.7\,\mu\text{K})$ for $N = 1$ (left), and $k_{\text{mix}}/k_{\text{pol}}$ for $N = 0$ (right). Experimentally measured ratios are shown as solid black lines, with uncertainties indicated by dashed lines. The vertical and horizontal axes represent the short-range phase shift δ^s and the loss parameter y respectively.

to d- and f-wave shape resonances, respectively [51, 59]. Notably, a p-wave shape resonance is incapable of reproducing the experimental ratio. For $N = 0$ the region of agreement is again at low y , but now near $\delta^s = 3\pi/8$, corresponding to a p-wave shape resonance. This provides valuable insight into the dynamics of this new ultracold system.

In conclusion, we report the first direct measurement of loss rates in the lowest partial waves for laser-cooled molecules. Our results suggest that there is incomplete loss at short range, and that resonances in higher partial waves play an important role. With an understanding of the inelastic loss rates and with the large permanent dipole moment in YO molecules, collisional shielding can be achieved by the application of either DC or microwave fields to significantly enhance the ratio of elastic to inelastic collision rates. Evaporative cooling will then be applied with the aim of achieving a quantum degenerate gas of laser-cooled molecules.

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