Dissociation and Decay of Ultra-cold Sodium Molecules

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The dissociation of ultracold molecules is studied by ramping an external magnetic field through a Feshbach resonance. The observed dissociation energy shows non-linear dependence on the ramp speed and directly yields the strength of the atom-molecule coupling. In addition, inelastic molecule-molecule and molecule-atom collisions are characterized.

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Recently, it has become possible to create ultracold molecular gases from precooled atomic samples \(1, 2, 4, 5, 6, 7, 8, 9, 10, 11\). Extending the ultralow temperature regime from atoms to molecules is an important step towards controlling the motion of more complicated objects. The complex structure of molecules may lead to new scientific opportunities, including the search for a permanent electric dipole moment, with sensitivity much higher than for heavy atoms \(12\), and the realization of quantum fluids of bosons and fermions with anisotropic interactions \(13\). Furthermore, stable mixtures of atomic and molecular condensates are predicted to show coherent stimulation of atom-molecule or molecule-atom conversion, constituting quantum-coherent chemistry \(14\).

To date, all realizations of ultracold molecules have bypassed the need for direct cooling of the molecules, which is difficult due to the complicated ro-vibrational structure. Rather, molecules were formed from ultracold atoms using Feshbach resonances \(2, 4, 5, 6, 7, 8, 9, 10, 11\), where a highly-vibrational excited molecular state is magnetically tuned into resonance with a pair of colliding atoms.

In this paper, we study the dissociation and decay of such highly excited molecules. Figure 11 shows the relevant energy levels. For magnetic fields above the Feshbach resonance, the molecular state crosses the free atomic states, shown here as discrete states in a finite quantization volume. The interaction between atoms and molecules turns these crossing into anti-crossings. When the magnetic field is swept very slowly through the resonance, the molecules will follow the adiabatic curve and end up in the lowest energy state of the atoms. For faster ramps, the molecular populations will partially cross some of the low-lying states, and the dissociation products will populate several atomic states.

The stronger the coupling between the molecular state and the atomic states, the faster the molecules dissociate and the smaller the energy release in the dissociation. Observing the atom-molecule coupling in one-body decay (dissociation) is a new method to experimentally determine the strength of a Feshbach resonance. Previous measurements used two- or three-body processes to characterize the Feshbach resonance and therefore required accurate knowledge of the atomic density distribution.

Collisional properties of the molecules were also studied. Inelastic collisions limit both the production of molecules and their lifetime. We observed loss of molecules by collisions both with atoms and other molecules. These two processes were studied separately because we could produce atom-molecule mixtures, as well as pure molecular samples, by separating atoms and molecules with short pulses of laser light \(6\).

To generate molecules, sodium condensates in the \(|F, m_F\rangle = |1, -1\rangle\) state were prepared in an optical dipole trap. The trap frequencies of 290 Hz in the radial direction and 2.2 Hz in the axial direction yielded typical densities of \(1.7 \times 10^{14}\) cm\(^{-3}\) for 5 million atoms. Atoms were then spin-flipped using an adiabatic radio frequency sweep to the \(|1, 1\rangle\) state, where a 1 G wide Feshbach resonance exists at 907 G \(12\).

The magnetic field sequence used to create and detect Na\(_2\) molecules was identical to our previous work \(9\). Briefly, the axial magnetic field was ramped to 903 G in 100 ms using two pairs of bias coils. In order to prepare the condensate on the negative scattering length side of the resonance, the field was stepped up to 913 G as quickly as possible (\(\sim 1\) \(\mu\)s) to jump through the resonance with minimal atom loss. The field was then ramped back down to 903 G in 50 \(\mu\)s to form molecules. In order to remove non-paired atoms from the trap, the sample was irradiated with a 20 \(\mu\)s pulse of resonant light. Because 903 G is far from the Feshbach resonance, the mixing between atomic and molecular states was small, and therefore molecules were transparent to this “blast” pulse. By ramping the field back to 913 G molecules were converted back to atoms. Absorption images were taken at high fields (either at 903 G or 913 G) after 10 to 17 ms ballistic expansion, with the imaging light incident along the axial direction of the condensate.

To study the momentum distribution of the back-converted atoms, the magnetic field was ramped up immediately after turning off the optical trap, or for a reference, at the end of the ballistic expansion. The difference between the energies of ballistic expansion is the released dissociation energy. Energies were obtained from the rms width of the cloud \(\langle x^2 \rangle\) as \(E = 3m\langle x^2 \rangle/2t^2\), where \(t\) is the ballistic expansion time, and \(m\) is the atomic mass. Figure 5 shows that faster field ramps created hotter atoms.

An earlier theoretical treatment assumes a constant predissociation lifetime of the molecules and predicts a
molecular state is lying continuum states due to the coupling with a bound
mean field energy 4
where ∆
π
mean field energy
magnetic moments,
B
where
V
two atoms in a volume
nance with second-order perturbation theory. Assuming
by comparing the energy shift near a Feshbach reso-
dence. Linear behavior would be expected if the lifetime
served. Furthermore, our data shows a non-linear depen-
ence (and therefore smaller dissociation energy) than was ob-
rate [16]. This theory predicts a much faster dissociation
linear relation between dissociation energy and field ramp
rate [14]. This theory predicts a much faster dissociation
and therefore smaller dissociation energy) than was ob-
erved. Furthermore, our data shows a non-linear depend-
ence. Linear behavior would be expected if the lifetime
of the molecules was independent of the energy ϵ from the
dissociation threshold. The fact that the slope be-
comes smaller for increasing ramp rate indicates that the
lifetime of molecules decreases with the ramp rate. As
we will show, this can be explained by an increase of the
density of atomic states, leading to a √τ dependence of
the molecular decay rate (Wigner threshold law [13]).

The decay rate Γ(ϵ) follows from Fermi’s golden rule as
hΓ(ϵ) = 2π|Vma(ϵ)|2D(ϵ) [18], where Vma is the matrix
element between atomic and molecular states, which to
leading order is independent of ϵ. The density of states
D(ϵ) is given by

D(ϵ) = \frac{V}{(2\pi)^2} \left( \frac{m}{\hbar^2} \right)^{3/2} ϵ^{1/2},

(1)

where V is the quantization volume for free atomic states.

An expression for the matrix element Vma is obtained by
comparing the energy shift near a Feshbach resonance
with second-order perturbation theory. Assuming two
atoms in a volume V, the energy shift of the low-
lying continuum states due to the coupling with a bound
molecular state is

δ(ϵ) = \frac{|Vma|^2}{ε} = \frac{|Vma|^2}{∂μ(B − B_0)},

(2)

where Δμ is the difference between atomic and molecular
magnetic moments, B the applied magnetic field, and B_0
the position of the Feshbach resonance.

The energy shift can also be expressed in terms of the
mean field energy 4πℏ^2a/mV, where a = a_bgΔB/(B −
B_0) is the scattering length near the Feshbach resonance
(a_bg is the background scattering length and ΔB is the
resonance width [19]):

δ(ϵ) = \frac{4πℏ^2 a_bg ΔB}{mV B − B_0}.

(3)

Comparing eq. [26] and eq. [3] yields

|Vma|^2 = \frac{4πℏ^2 a_bg Δμ ΔB}{mV}.

(4)

If the entire population is initially in the molecular
state, the fraction of molecules, m(ϵ), at energy ϵ follows
the rate equation,

n \frac{dm(ϵ)}{dϵ} = \frac{dm(ϵ)}{dt} \left( \frac{de}{dt} \right)^{-1} = Γ(ϵ)m(ϵ) \left( \frac{de}{dt} \right)^{-1}

(5)

Using Eqs. [11] and [4], we solve the differential equation for m(ϵ)

m(ϵ) = e^{-\frac{2}{C} C^3/2},

(7)

In the lab frame, the atoms have kinetic energy ϵ/2 and
therefore the average energy of an atom after dissociation
is

δE = \int_0^∞ \frac{ϵ}{2} (-dm(ϵ)) = 0.591 \left( \frac{ℏ}{ma^2_{bg}} \frac{ℏB}{2ΔB} \right)^{2/3}.

(8)

FIG. 1: Schematic diagram of energy levels for molecules and
atoms. The diabatic energy levels are shown as dashed lines.
The adiabatic curves (solid lines) include the atom-molecule
coupling. When the magnetic field is swept from positive to
negative scattering length, dissociated molecules end up in
one or several atomic states, depending on the ramp rate of
the magnetic field. The spheres represent the distribution of
the population before and after the ramp.

FIG. 2: Dissociation energy of molecules as a function of mag-
netic field ramp rate. The dashed line represents the linear
relation described in ref. [16] the solid line shows the result of
our theory with no free parameters (using a theoretical valu-
e for ΔB = 0.98 G), and the dotted line shows a curve with
ΔB as a fitting parameter.
FIG. 3: Temperature of the molecular cloud. After 15 ms, the temperature saturates at $\sim 250$ nK. Error bars represent the statistical error (standard deviation). The inset shows finer resolution data for holding times up to 6 ms. The solid line is a guide to the eye.

Using theoretical values $a_{bg} = 3.4$ nm, $\Delta \mu/h = 3.65 \times 1.4$ MHz/G, and $\Delta B = 0.98$ G \(\square\square\), our parameter-free prediction (solid line in Fig. 2) shows good agreement with the experimental data. Alternatively, we may regard the width of the Feshbach resonance as a free parameter to obtain a fitted value of $\Delta B = 1.07 \pm 0.02$ G (dotted line in Fig. 2). Compared to previous mean-field measurements \(\square\square\), our determination of the resonance width is more accurate and free from systematic errors associated with the determination of atomic densities.

Further experiments with ultracold sodium molecules will critically depend on their collision properties. Therefore we also studied heating and inelastic collision processes. As shown in Fig. 3, we observed monotonic heating of the pure molecular sample over $\sim 30$ ms. In addition, we observed short timescale oscillations (6 ms) in the fitted temperature (inset of Fig. 3). Such breathing oscillations were excited because the molecules were formed over the volume of the atomic condensate, which was larger than the equilibrium volume for the molecules. The absence of damping implies a collision time of at least 6 ms, or a molecular scattering length smaller than 17 nm (obtained using the expression for the collision rate $8\pi a^2 v_{th} n_m$ where $v_{th}$ is the thermal velocity). It is unclear whether the oscillation disappeared due to collisions or limited signal-to-noise ratio.

The temperature of the molecular cloud saturated at $\sim 250$ nK after 15 ms. A possible explanation is the balance between heating due to inelastic molecular decay and the evaporative cooling caused by the finite trap depth (1.7 $\mu$K). This would imply a collision time of 15 ms. However, we have no clear evidence that thermalization has occurred. Clearly, further studies of elastic collisions between ultracold molecules are necessary.

Molecules formed via Feshbach resonances are created in high vibrational states. Therefore, one expects vibrational relaxation to be a strong, inelastic decay mechanism. Vibrational energy spacings are much larger than the trap depth, leading to loss of molecules from the trap.

Figure 4(a) shows the decay of a pure molecular sample. The decay was analyzed with the rate equation

$$\frac{N_m}{N_m} = -K_{mm} n_m.$$  \(9\)

Here $n_m$ is the density of the molecules, and $K_{mm}$ is the molecule-molecule collision rate coefficient. Because of the changing size and temperature of the molecular cloud during the first $\sim 15$ ms (Fig. 3), we only fit data points at later times, assuming a thermal equilibrium volume for the molecules. The decay at earlier times is slower, consistent with a larger molecular cloud. The fit yields a molecule-molecule collision coefficient of $K_{mm} \sim 5.1 \times 10^{-11}$ cm$^3$/s, about 2 orders of magnitude larger than the typical values reported for fermions \(\square\).

FIG. 4: Decay of ultracold molecules trapped alone (a) or together with atoms (b). The solid lines in (a) and (b) are fits of eq (9) and (10) to data, which assume vibrational relaxation in the collision of molecules (a) or collisions between molecules and atoms (b). The insets illustrate the experimental sequences.
Inelastic collisions between molecules and atoms were also observed by keeping atoms in the trap (Fig. 1b)). The decay was analyzed assuming that the loss of molecules occurred mainly due to collisions with atoms, resulting in an exponential decay:

$$\frac{N_m}{N_m} = -K_{am}n_a. \quad (10)$$

Here $N_m$ is the number of the molecules, $n_a$ is the density of atoms, and $K_{am}$ is the atom-molecule collision rate coefficient. From the fit, we extract a lifetime of $106 \mu s$ and a rate coefficient $K_{am} \sim 5.5 \times 10^{-11} \text{ cm}^3/\text{s}$, which agrees well with theoretical predictions [20, 21].

The inelastic losses determine the maximum conversion efficiency from atoms to molecules. For an adiabatic ramp, one expects close to 100% conversion efficiency. Indeed, in experiments with fermionic atoms, efficiencies up to 85% have been observed [7]. Figure 5 shows the results for magnetic field ramps of different durations. The two sets of images show that applying the blast pulse dramatically improved the molecular number and temperature. Without it, a slower ramp time (4 ms) appeared to be more favorable for molecule formation (open circles in Fig. 5b)). No molecules were observed for a 50 $\mu s$ ramp time. However, with the blast pulse, nearly the same number of molecules was obtained for all ramp times between 50 $\mu s$ to 4 ms (closed circles in Fig. 5c)).

We interpret our data as the interplay of two competing processes. The adiabatic condition requires a relatively slow field ramp for efficient conversion. However, this means that the atoms and molecules spend more time near or at the Feshbach resonance, where inelastic collision rates are enhanced. In contrast to Fig. 5b), the absence of molecular signal in Fig. 5a) for 50 $\mu s$ ramp time reflects that the atomic density reduction due to the mean-field expansion is too slow for the molecules to survive the inelastic collisions with the atoms.

In conclusion, we observed a Wigner threshold behavior in the dissociation of ultracold molecules. We were able to characterize a Feshbach resonance using a one-body decay (dissociation) process. The rapid decay of the molecules due to collisions with atoms and other molecules imposes a severe limit to further evaporative cooling for bosons. This also explains the low conversion efficiency ($\sim 4\%$), in contrast to recent experiments with fermions.

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[1] R. Wynar et al., Science 287, 1016 (2000).
[2] E. A. Donley et al., Nature (London) 417, 529 (2002).
[3] C. A. Regal et al., Nature (London) 424, 47 (2003).
[4] S. Jochim et al., cond-mat/0308095 (2003).
[5] J. Cubizolles et al., cond-mat/0308018 (2003).
[6] K. E. Strecker et al., Phys. Rev. Lett. 91, 080406 (2003).
[7] S. Dürr et al., cond-mat/0307440 (2003).
[8] C. Chin et al., Phys. Rev. Lett. 90, 033201 (2003).
[9] K. Xu et al., Phys. Rev. Lett. 91 210402 (2003).
[10] M. Greiner et al., cond-mat/0311172 (2003).
[11] S. Jochim, et al., Science Express Reports, 1093280 (2003).
[12] J. J. Hudson et al., Phys. Rev. Lett. 89, 23003 (2002).
[13] M. Baranov et al., cond-mat/0201100 (2002).
[14] D. J. Heinzen et al., Phys. Rev. Lett. 84, 5029 (2000).
[15] S. Inouye et al., Nature (London) 392, 151 (1998).
[16] F. A. van Abeelen, and B. J. Verhaar, Phys. Rev. Lett. 83, 1550 (1999).
[17] E. P. Wigner, Phys. Rev. 73, 1002 (1948).
[18] F. H. Mies et al., Phys. Rev. A 61, 022721 (2000).
[19] E. Timmermans et al., Phys. Rep., 315, 199 (1999).
[20] V. A. Yurovsky and A. Ben-Reuven, Phys. Rev. A 67, 043611 (2003).
[21] V. A. Yurovsky, A. Ben-Reuven et al., Phys. Rev. A 62, 043605 (2000).
[22] J. Stenger et al., Phys. Rev. Lett. 82, 2422 (1999).