

Spontaneous Dissociation of ^{85}Rb Feshbach Molecules

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The spontaneous dissociation of ^{85}Rb dimers in the highest lying vibrational level has been observed in the vicinity of the Feshbach resonance that was used to produce them. The molecular lifetime shows a strong dependence on magnetic field, varying by 3 orders of magnitude between 155.5 G and 162.2 G. Our measurements are in good agreement with theoretical predictions in which molecular dissociation is driven by inelastic spin relaxation. Molecule lifetimes of tens of milliseconds can be achieved within approximately a 1 G wide region directly above the Feshbach resonance.

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Magnetic Feshbach resonances were first used to dramatically alter the strength and sign of interatomic interactions in ultracold atoms [1–6]. Several years ago it was predicted that they could also be used to produce molecules [7–11]. Today Feshbach resonances have become very useful tools for creating ultracold gases of diatomic molecules. In our initial experiments we saw molecules formed from a ^{85}Rb Bose-Einstein condensate (BEC) by nonadiabatic mixing of atomic and molecular states when the magnetic field was rapidly pulsed close to the Feshbach resonance [12]. Subsequently, it has been shown that both fermionic [13–15] and bosonic [16–18] atoms can be converted into molecules by adiabatically sweeping the magnetic field through a Feshbach resonance. Molecules formed using these techniques are very weakly bound and very highly vibrationally excited and are of considerable experimental and theoretical [19–21] interest. The lifetime of these molecules has varied widely under different conditions and their decay processes have not been fully established.

Several experiments have shown that such molecules can undergo rapid vibrational quenching in which they collide with atoms or other molecules and relax to lower vibrational states [22,23]. For the case of molecules created from a Fermi gas, it has been observed that near resonance the molecular lifetime increases by several orders of magnitude [23]. It is speculated that collisional relaxation is greatly suppressed close to the Feshbach resonance due to the Fermi statistics of the atoms [21]. A systematic study of the lifetime of molecules composed of bosons near a Feshbach resonance has not yet been published. However, it is believed that the observed low atom-molecule conversion efficiencies for bosonic atoms [18,22] are actually the result of very high vibrational quenching rates near the Feshbach resonance. In general, all of these experiments have started with an atom cloud with an initial peak density of $n_0 \sim 10^{13}$ to 10^{14} cm^{-3} . This collisional quenching mechanism will become much less significant at lower densities, such as the conditions we have used in

studying the conversion of ^{85}Rb atoms to molecules ($n_0 \sim 10^{11}$ cm^{-3}).

There were several experiments that raised questions as to the possible decay of these molecules even at densities where collisional quenching was implausible. These experiments emphasized the need for a better understanding of the molecular lifetime. First, there were some unanswered questions regarding the results of our initial experiments in which we observed coherent atom-molecule oscillations [12]. Comparisons with theoretical calculations [24,25] showed superb agreement in all respects except for the amount of atom loss that was observed. If the molecules were decaying in such a way that they were being ejected from the trap, this would resolve the disagreement. In addition, the damping of this atom-molecule coherence has yet to be explained in part because it was unknown whether molecular decay contributed to this damping, and if so, to what degree. Further questions about the molecular lifetime were raised by our subsequent unsuccessful attempts to observe the spatial separation of atomic and molecular clouds juxtaposed with successful experiments by Grimm *et al.* [16] and later Rempe *et al.* [18] demonstrating just such a separation. A possible explanation for this difference was that our molecules were for some reason decaying more quickly than theirs, quicker than the time required to spatially separate them from the remaining atoms. For all of these reasons, in addition to the intrinsic interest of better understanding the properties of these remarkable Feshbach resonance molecules, there has been considerable incentive to determine the fundamental (i.e., noncollisional) decay processes of these molecules and to learn how it might depend on magnetic field, and, assuming there was an intrinsic decay mechanism, what range of lifetimes could be achieved.

There is a rather practical use to a relatively short well understood and characterized molecular lifetime. This is to definitively settle the substantial disagreement [24,25] over the fraction of the sample that has been converted to molecules in our initial experiments [12]. If the molecular lifetime is known, one can simply observe what fraction of

the sample disappears with this characteristic time and this issue is easily resolved. All of the above issues inspired the theoretical work of Köhler *et al.* discussed below and the parallel experimental program presented here.

We have systematically investigated the molecular lifetime of ^{85}Rb dimers in the highest vibrational state as a function of magnetic field in a 7 G wide region directly above the Feshbach resonance. By starting with an ultracold but uncondensed gas of bosonic ^{85}Rb atoms in a magnetic trap we have been able to study the molecular lifetime at an initial atom density which is 2 to 3 orders of magnitude smaller than in other experiments and thus distinguish collisional destruction of molecules from the intrinsic lifetime of the molecular state. Köhler *et al.* have predicted that a novel decay mechanism should dominate under these conditions [26]. In short, they expect inelastic spin relaxation to lead to the spontaneous decay of these molecules. One of the atoms in the molecule experiences a spin flip that is similar to an inelastic spin relaxation collision between two atoms. This causes the molecule to dissociate and releases sufficient kinetic energy for both atoms to be lost from the trap. A high dependence of this dissociation rate on magnetic field is anticipated. Close to resonance, the size of the molecule increases and spin relaxation is suppressed. Our work directly tests this theoretical prediction and determines the range of experimentally accessible molecular lifetimes.

To carry out these lifetime measurements we start with what has become a rather standard technique for molecule production, namely, ramping the magnetic field adiabatically through a Feshbach resonance [13–18]. We have used the 10.7 G wide resonance at 155.0 G for this purpose and have observed a 30% atom-molecule conversion efficiency. We found the lifetime of the molecules by holding them for various lengths of time, then converting all remaining molecules back into atoms and measuring the number of atoms remaining versus the duration of the hold. We have repeated this process holding the molecules at several different magnetic fields.

The apparatus used in this study has been described in detail elsewhere [2]. We first prepared an ultracold (30 nK) thermal cloud of 100 000 ^{85}Rb atoms in the $F = 2$, $m_F = -2$ state in a magnetic trap at a bias field of 162.2 G. The standard deviation of the atom number from shot to shot was $\sim 3\%$. The spatial distribution of the atoms was Gaussian with a peak density of $n_0 = 6.6 \times 10^{11} \text{ cm}^{-3}$ and the trap frequencies were $(17.5 \times 17.2 \times 6.8) \text{ Hz}$. We then used the trapping coils to apply a magnetic field time sequence as shown in Fig. 1 to produce molecules and subsequently measure their lifetime.

Having performed evaporative cooling at 162.2 G where the scattering length is positive, we first ramped the magnetic field to 147.2 G as rapidly as experimentally convenient (an inverse ramp rate of $46 \mu\text{s/G}$) simply to get to the correct side of the resonance to begin molecule pro-

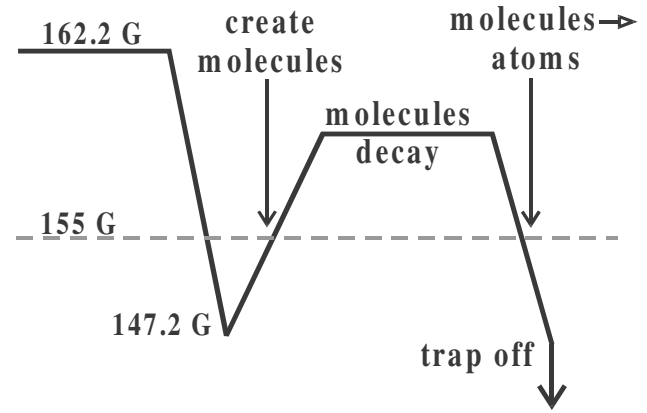


FIG. 1. Magnetic field ramp sequence for producing molecules and measuring their decay rate. The Feshbach resonance is indicated by the dashed line. The field is first swept as quickly as possible from the evaporation field to the opposite side of the resonance. A second ramp back across the resonance converts some atoms to molecules. The molecules are then held at a constant field above the resonance for a variable amount of time. A third ramp across the resonance then converts any remaining molecules back into atoms and the magnetic trap is turned off.

duction[27]. A second ramp ($57 \mu\text{s/G}$) back across the resonance then adiabatically converted 30% of the atoms into molecules. We did not observe any atom or molecule loss during this ramp; if we quickly shut the trap off after the ramp (converting any molecules back into atoms), all of the atoms in the original sample were still present. This field ramp continued to the chosen field B_{hold} above the resonance. The field was then held constant at B_{hold} for a variable amount of time t_{hold} , during which time a fraction of the molecules could decay. A third ramp across the resonance ($65 \mu\text{s/G}$) then converted any remaining molecules back into atoms. The trap was then turned off and the atom cloud was allowed to expand for 22 ms before destructive absorption imaging was used to determine the number of atoms in the cloud. By measuring the decrease in the number of atoms as a function of t_{hold} we were effectively measuring the decay of the molecules. This method, of course, relies on the assumption that the decaying molecules leave the magnetic trap so we do not see them in our absorption images. The observed exponential loss indicates that this must be true for at least a large fraction of them. On theoretical grounds it is likely all leave since it has been predicted that the decay energies associated with the various available decay channels are all on the order of several mK [26] and our trap depth is only $\sim 1 \text{ mK}$. Also, we have looked at absorption images at a large range of expansion times and have not seen any evidence for modestly energetic atoms arising from less energetic decay channels. By measuring the atom number as a function of t_{hold} and by fitting this to an exponential decay we were able to extract the molecular lifetime at B_{hold} .

Data from such a measurement are shown in Fig. 2(a) for $B_{\text{hold}} = 156.6$ G. We have investigated a range of B_{hold} from 155.5 G to 162.2 G. The decay we observe fits very nicely to an exponential. We found that the time constant for the decay depends very strongly on field; it changes by 3 orders of magnitude over this 7 G wide region. There is a minor complication in the data analysis for fields less than 156.5 G. At fields this close to the Feshbach resonance the atoms leave the trap via three-body collisions at a rate that is slower but not entirely negligible relative to the molecule loss rate, as we have previously studied [28]. This causes the apparent sloping baseline in Fig. 2(b). In principle we could fit the loss versus time curve with the correct func-

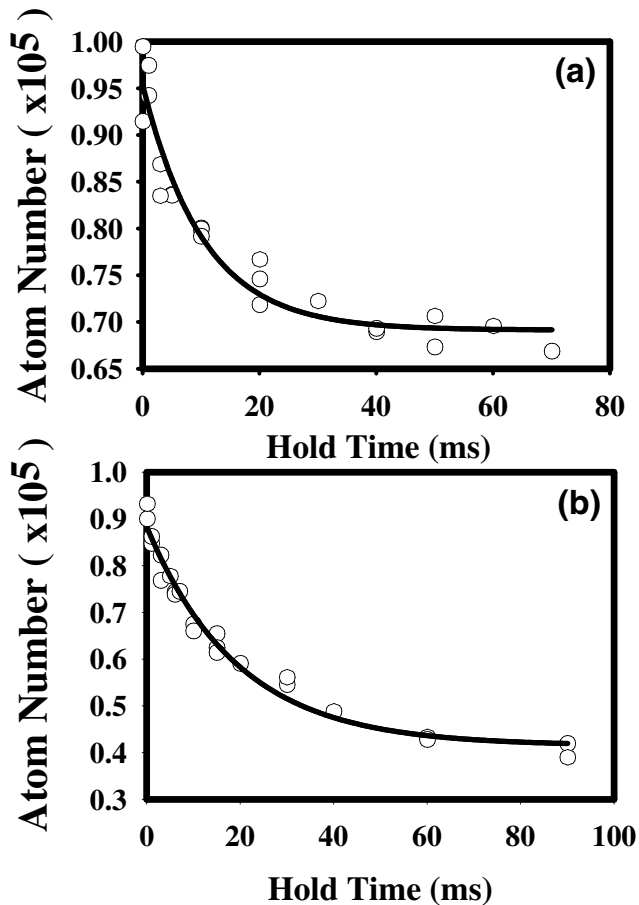


FIG. 2. Measurements of molecular lifetime. (a) Number of atoms remaining after holding the system at 156.6 G as a function of the hold time. The decay fits nicely to an exponential and from this we get a lifetime of 10.4(1.7) ms. The error represents the statistical error from the exponential fit to the data. The baseline indicates we have converted 30% of the atoms into molecules. (b) Number of atoms remaining after holding at 155.5 G as a function of the hold time. In this case atoms are being lost from the trap during the hold time due to three-body collisions and we never observe a horizontal baseline. Analyzing the data taking into account this atom loss we get a molecule lifetime of 24.7(6.4) ms.

tional form including molecular decay and three-body loss, but in practice the uncertainties are less if we simply measure the three-body loss versus time in a sample of pure atoms (no molecules) at 155 G and 156 G and subtract off this loss from the raw molecular decay data before fitting it to a single exponential. A similar technique was used by Regal *et al.* [23] in the measurements of the lifetime of ^{40}K molecules produced from a Fermi gas near a Feshbach resonance. This three-body atomic loss is strongly field dependent [28] and was negligible for data above 156.5 G. A summary of our molecule lifetime measurements is shown in Fig. 3. For B fields between 155.5 G and the peak of the Feshbach resonance at 155.0 G, the quantity $n_0 a^3$ (where a is the s -wave scattering length) is equal to or greater than one. In this regime the atomic and molecular states are not well defined and hence it becomes impossible to ascribe the observed loss to a “molecular decay.”

We have confirmed that the molecular lifetime is independent of density which illustrates the one-body nature of the decay. For an initial atom density $n_0 = 6.6 \times 10^{11} \text{ cm}^{-3}$ we measure a lifetime of 2.1(4) ms at 159 G. Repeating this measurement with $n_0 = 2.6 \times 10^{11} \text{ cm}^{-3}$ yielded a lifetime of 2.5(6) ms which agrees within the error bars.

The solid curve in Fig. 3 is the result of a coupled channels calculation done by Köhler *et al.* in Ref. [26] in which inelastic spin relaxation leads to the spontaneous

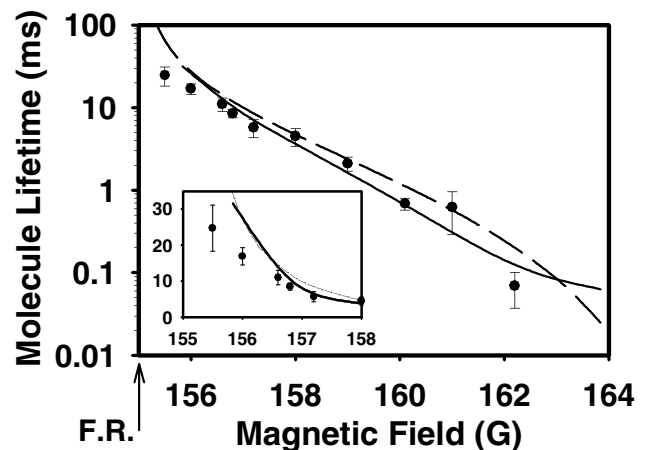


FIG. 3. Molecule lifetime as a function of magnetic field. The experimental data are represented by the closed points. The two lines are the results of theoretical calculations with *no free parameters* by Köhler *et al.* (Ref. [19]) in which molecules spontaneously decay due to inelastic spin relaxation. The solid line arises from an exact coupled channels scattering calculation. The dashed line results from a simpler calculation in which the detailed nature of the interatomic potentials is ignored, resulting in an analytic solution for the molecular lifetime. The inset shows the discrepancy between experiment and theory close to the 155.04 G Feshbach resonance.

decay of these molecules. There is good agreement between experiment and theory for fields greater than ~ 157 G covering a factor of 100 in lifetime. The discrepancy within ~ 1 G of the Feshbach resonance is most likely due to many-body effects becoming more important as $n_0 a^3$ becomes large and, as mentioned above, the loss processes cannot be simply described in terms of distinct molecular decay and three-body atomic loss. The essentially binary approach of the theoretical model is expected to fail in this regime. The dashed curve is the result a universal calculation which does not depend on the detailed nature of interatomic interactions, also by Köhler *et al.* in Ref. [26]. It predicts that the molecular lifetime as a function of magnetic field is given by $4\pi a^3(B)/K_2(B)$, where $a(B)$ is the s -wave scattering length and $K_2(B)$ is loss rate constant for inelastic spin relaxation collisions. This simple formula also does a good job of predicting the molecular lifetime over the magnetic field range we have investigated and in addition provides good physical insight into the decay mechanism. It has been theoretically shown that the spatial extent of the wave functions of these Feshbach molecules is of the order of the scattering length [19]. Thus, as $a(B)$ becomes large near resonance so does the volume containing the atom pair, and the spontaneous decay of the molecule is suppressed. As pointed out in Ref. [26], if $K_2(B)$ is known, such measurements of the molecular lifetime can be used as a direct probe of the size of the molecule.

In summary, we have measured the lifetime of ^{85}Rb dimers in the highest lying vibrational level in the vicinity of the Feshbach resonance. We have observed a very strong dependence of this lifetime on magnetic field which is in good agreement with theoretical predictions where molecules decay due to dissociation driven by inelastic spin relaxation. These results show that it is possible to create ^{85}Rb dimers with lifetimes of tens of milliseconds. Therefore, one should be able to carry out further experiments with these molecules if operating within a few gauss of the Feshbach resonance. These results also explain the unexplained atom-molecule loss observed in our previous experiments [7,12,24,25,29] creating coherent superpositions of atomic and molecular BECs of ^{85}Rb .

Because of our thorough understanding of this novel decay mechanism, we can use it as a tool for further investigation of the molecular production process which has received little experimental attention to date. For example, it is unclear what factors determine the atom-molecule conversion efficiency when the magnetic field is swept across the Feshbach resonance. By making molecules under a controlled set of conditions and then subsequently ensuring that they all undergo *one-body* decay before turning the magnetic trap off and imaging, we can

get an accurate measure of how many molecules were produced. This is the subject of current work.

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