Spontaneous Dissociation of ⁸⁵Rb Feshbach Molecules

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The spontaneous dissociation of ⁸⁵Rb dimers in the highest lying vibrational level has been observed in the vicinity of the Feshbach resonance which was used to produce them. The molecular lifetime shows a strong dependence on magnetic field, varying by three 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 close to 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, 2, 3, 4, 5, 6]. Today they have become very useful tools for creating ultracold gases of diatomic molecules. In our initial experiments we saw molecules formed from a ⁸⁵Rb BEC by nonadiabatic mixing of atomic and molecular states when the magnetic field was rapidly pulsed close to the Feshbach resonance[7]. Subsequently it has been shown that both fermionic[8, 9, 10] and bosonic[11, 12, 13] 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 [14, 15, 16] interest. Near the Feshbach resonance the size of the molecules is comparable to the interatomic spacing. The lifetimes 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 [17, 18]. 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 [18]. It is speculated that collisional relaxation is greatly suppressed close to the Feshbach resonance due to the Fermi statistics of the atoms [16]. 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 [13, 17] 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 $10^{13} - 10^{14}$ cm⁻³. This collisional quenching mechanism will not be significant at lower densities, such as the conditions we have used in studying the conversion of ⁸⁵Rb atoms to molecules. However, in this Letter we show that the molecular lifetime can be quite short even in the low density regime.

Here we have systematically investigated the molecular lifetime of ⁸⁵Rb dimers in the highest vibrational state as a function of magnetic field near the Feshbach resonance. By starting with an ultracold but uncondensed gas of bosonic ⁸⁵Rb atoms in a magnetic trap we have been able to study the molecular lifetime at an initial atom density which is two to three 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 very different decay mechanism should dominate under these conditions [19]. 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, releasing 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[8, 9, 10, 11, 12, 13]. We have used the 11G wide resonance at 155G for this purpose and have observed a 30% atommolecule 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 ⁸⁵Rb 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 ~3%. The spatial distribution of



FIG. 1: Magnetic field ramp sequence for producing molecules and measuring their decay rate. The Feshbach resonance in 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 slower 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.

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 x 17.2 x 6.8) 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 μ s/G) simply to get to the correct side of the resonance to begin molecule production[21]. A second slower ramp (57 μ s/G) back across the resonance then adiabatically converted 30% of the atoms into molecules. 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 $(65 \,\mu s/G)$ 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 don't 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[19]and our trap depth is only ~ 1 mK. Also, we have looked at absorption images at a large range of expansion times and have not seen any evidence for modestly energetic



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 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. After correcting for this loss we get a molecule lifetime of 24.7(6.4) ms.

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 is shown in Fig. 2a for $B_{hold} = 156.6$ G. We have investigated a range of B_{hold} from 155 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 three orders of magnitude over this 7 G wide region.

We have observed that for values of B_{hold} within ~1 G of the Feshbach resonance the interpretation of the data is complicated by the fact that some atoms also leave the trap during t_{hold} mostly due to three body collisions. The exponential decay in Fig. 2b exhibits a decaying baseline due to this atom loss. To compensate for this atom loss we measured the loss of atoms directly (no molecules present) at the appropriate densities and magnetic fields and subtracted this loss from our raw molecular decay data. In this way the molecular lifetime

close to the Feshbach resonance was extracted. A similar technique needed to be employed by Regal *et al.* [18] in the measurements of the lifetime of their 40 K molecules produced from a Fermi gas. A summary of our molecule lifetime measurements is shown in Fig. 3. The three data points closest to the Feshbach resonance show the atom loss correction described above. For the point at 156.6 G the correction is negligible. Since we know that the three body loss rate decreases rapidly at higher fields[20] we can safely ignore this atom loss correction for all points above 156.6 G.

The solid curve in Fig. 3 is the result of a coupled channels calculation done by Köhler *et al.* in ref. [19] in which inelastic spin relaxation leads to the spontaneous 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 close to the Feshbach resonance is most likely due to other decay processes not included in the theory that may become significant close to resonance such as atom-molecule or molecule-molecule collisions. 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. [19]. 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 [14]. 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.[19], 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 ⁸⁵Rb 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 ⁸⁵Rb dimers with lifetimes of tens of milliseconds. These results also explain the unexplained atom/molecule loss observed in our previous experiments[7, 22, 23, 24] creating coherent superpositions of atomic and molecular BECs of ⁸⁵Rb.

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FIG. 3: Molecule lifetime as a function of magnetic field. The experimental data are represented by the closed points. The open points close to the Feshbach resonance are the raw data before the atom loss correction was applied. 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.

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