# Ultracold Molecule Production via a Resonant Oscillating Magnetic Field

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A novel atom-molecule conversion technique has been investigated. Ultracold $^{85}$Rb atoms sitting in a dc magnetic field near the 155 G Feshbach resonance are associated by applying a small sinusoidal oscillation to the magnetic field. There is resonant atom to molecule conversion when the modulation frequency closely matches the molecular binding energy. We observe that the atom to molecule conversion efficiency depends strongly on the frequency, amplitude, and duration of the applied modulation and on the phase space density of the sample. This technique offers high conversion efficiencies without the necessity of crossing or closely approaching the Feshbach resonance and allows precise spectroscopic measurements. Efficiencies of 55% have been observed for pure Bose-Einstein condensates.

The conversion of ultracold atoms to ultracold molecules by time varying magnetic fields in the vicinity of a Feshbach resonance is currently a topic of much experimental and theoretical interest. This particular conversion process lends itself well to the formation of molecular BECs [1–4] and atom-molecule superpositions [5]. These Feshbach molecules and their creation process are also important for understanding ultracold fermionic systems in the BCS-BEC crossover regime because they are closely related to the pairing mechanism in a fermionic superfluid that occurs near a Feshbach resonance [6–9]. Finally, Feshbach molecules are interesting themselves because they are very weakly bound and very large in spatial extent—comparable to the spacing between atoms in the sample from which they were created [10].

To date three Feshbach molecule creation techniques have been demonstrated. Atom to Feshbach molecule conversion was first directly observed by applying very rapid (tens of $\mu$s) time dependent magnetic fields to a $^{85}$Rb Bose-Einstein condensate (BEC) in a Ramsey type manner [5]. The magnetic field was pulsed very close to the Feshbach resonance which created a superposition of free atoms and Feshbach molecules. This technique was plagued by low conversion efficiencies (on the order of a few percent) that were difficult to control. It also lead to heating and loss of atoms from the atomic sample.

The most popular atom-molecule conversion scheme to date involves slowly sweeping the magnetic field through a Feshbach resonance. This has been demonstrated for both fermionic [11,12] and bosonic [13–17] atoms. Although high conversion efficiencies have been observed in degenerate Fermi systems, high vibrational quenching rates near the Feshbach resonance have lead to low conversion efficiencies for BECs of bosonic atoms [15,18]. There are also problems caused by density dependent heating processes. As the resonance is crossed, we observe significant heating. $^{85}$Rb BECs with peak densities of $n_0 = 1 \times 10^{13}$ cm$^{-3}$ were heated by a factor of roughly 250 (a few $\mu$K) which prevented any molecule formation in the

field sweep. This heating was much less, but still significant for thermal clouds that were 2 orders of magnitude less dense. For example, a 30 nK cloud with peak density $n_0 = 4 \times 10^{11}$ cm$^{-3}$ was heated by a factor of 0.5. This heating is likely due to three body recombination collisions. For this reason, we were only able to observe molecule production in thermal clouds. For adiabatic magnetic field sweeps (adiabatic with respect to the two body physics governing molecule formation, not in a thermodynamical sense) we have shown that the atom to molecule conversion efficiency is solely determined by the phase space density of the atomic sample after this heating has been taken into account [17]. Therefore, by limiting the achievable phase space density, this heating is also limiting the conversion efficiency.

A third atom-molecule conversion technique has been demonstrated in two experiments with fermionic atoms [19,20]. This technique utilizes the enhanced three body recombination collision rates near a Feshbach resonance to efficiently associate atoms into molecules. Molecules were formed simply by holding a degenerate Fermi cloud of atoms for several seconds on the positive scattering length side of a Feshbach resonance where a weakly bound molecular state exists. Conversion efficiencies as high as 85% have been reported [19]. This technique would not be useful in systems such as ours where the molecular lifetime is comparatively short and cannot exceed tens of ms [21].

Low bosonic atom-molecule conversion efficiencies and the heating observed when using both of these time dependent magnetic field techniques has prompted us to investigate alternative conversion methods. In this Letter we report on a novel atom-molecule conversion method in which atoms are resonantly associated to form molecules by applying a sinusoidally oscillating magnetic field modulation. Note that this molecular formation process is different from typical single photon photoassociation processes where a colliding pair of ground state atoms absorbs a photon to form an excited state molecule [22–24]. The photon *adds* energy to the system, allowing a molecule to

form. In our case, photons from our oscillating magnetic field have the opposite effect—they cause the system to *lose* energy by stimulating an atom pair to emit a very low frequency photon and thereby decay to a lower energy bound molecular state. These experiments were motivated in part by work done by Regal *et al.* where radio frequency photons were used to *dissociate* molecules formed from a degenerate two component Fermi gas [11].

This technique circumvents problems of heating and enhanced collision rates because it does not require crossing or closely approaching the Feshbach resonance. The maximum achievable conversion efficiency for a given phase space density is the same as was observed in the slow field sweep experiments [17]. However, since this new technique greatly reduces the heating the cloud experiences in the conversion process, we can achieve higher ultimate phase space densities and hence higher conversion efficiencies. It enables us to convert more than half of the atoms in a BEC to molecules. This new scheme also allows spectroscopy that is comparable or superior to the Ramsey technique discussed in Ref. [5].

A detailed description of the $^{85}$Rb experimental apparatus can be found in Ref. [25]. We use evaporative cooling in a purely magnetic trap at a bias field of 162 G to produce either degenerate or nondegenerate atomic samples. Bose-Einstein condensates used for these experiments contain 5000–20 000 atoms and range from having a 50% condensate fraction to nearly a 100% condensate fraction, and the nondegenerate clouds used typically have 40 000–200 000 atoms at temperatures ranging from 20–80 nK (BEC transition temperature is approximately 14 nK). The majority of these experiments were done with uncondensed samples. After producing an ultracold atomic sample, we utilize the 11 G wide Feshbach resonance at 155.0 G to convert a fraction of the sample to molecules. The molecular state exists above the resonance. We first ramp the magnetic field from 162 G to a selected value between 156 and 157 G in 5 ms. We then use the trapping coils to apply a sinusoidal modulation to the magnetic field for 0–50 ms whose peak-to-peak amplitude ranges from 130–280 mG and whose frequency is close to the molecular binding energy as measured in Ref. [26]. We then slowly ramp the field (in 5 ms) back to 162 G where the molecular lifetime is 700 $\mu$s [21]. The field is held here for about 10 ms to ensure that any molecules we have made decay, ejecting their constituent atoms from the trap and our field of view in the process [21]. The trap is then rapidly turned off and we measure the number of atoms remaining using absorption imaging. The loss of free atoms is negligibly small, so any loss of atoms in this process must be due to molecule production. We observe significant molecular formation that is very dependent on modulation frequency, time, and amplitude and on the initial phase space density of the sample.

As further confirmation that the observed loss is due to molecular formation and decay, we have done a slightly modified version of this experiment. After applying the sinusoidal modulation to create molecules, the trap is immediately turned off instead of ramping the magnetic field back to 162 G where all of the molecules would quickly dissociate. In turning the trap off, we are sweeping the magnetic field through the Feshbach resonance, converting any remaining molecules back into atoms. In this case, most of the original atoms are still present in the absorption image and the slight loss we observe is consistent with the molecular lifetime at the field at which the molecules were created.

Figure 1 shows the observed atom loss as a function of modulation frequency for three different modulation durations (coupling times). There is a clear resonant frequency. As the coupling time increases, more atoms are converted into molecules. Over the range of modulation amplitudes and coupling times we have investigated, the width of the loss feature increases linearly with peak loss to within our measurement uncertainty. For 37(2)% conversion at a field of 156.45 G the width is 1.88(16) kHz while for 17(2)% conversion it is 0.82(14) kHz. By fitting the linewidth versus conversion data to a straight line we find the width in the zero percent conversion limit to be 0.2(2) kHz. We would expect the width due to the spontaneous lifetime of the molecules to be $1/2\pi\tau$ which, with $\tau = 11$ ms [21,27], is 0.01 kHz.

We have investigated the dependence of the resonant frequency on the value of the dc magnetic field. One would expect this to provide information about the field dependent molecular binding energy. The results of all of our measurements are shown in Fig. 2. The solid line is the molecular binding energy that resulted from fitting our Ramsey measurements described in Ref. [26] with an exact coupled-channel scattering calculation and the dashed
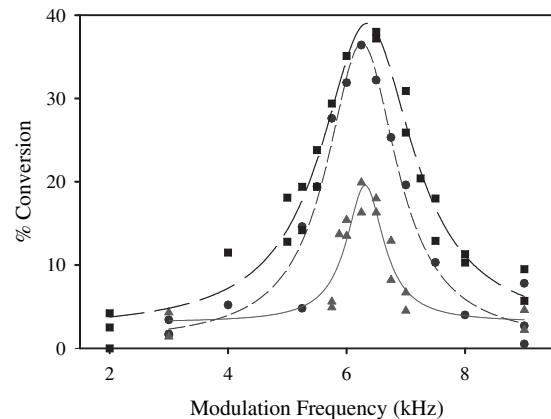


FIG. 1. Percentage of atoms converted to molecules at 156.45 G as a function of modulation frequency for three different coupling times. The modulation amplitude was 130 mG. The triangles are the data and the solid line a Lorentzian fit for a coupling time of 6 ms. The circles and short dashed line are for a 25 ms coupling time and the squares and long dashed line are for a 38 ms coupling time. The widths are 0.82(14), 1.44(18), and 1.88(16) kHz, respectively. The peak positions for all three curves agree within the 0.04 kHz fitting uncertainty.
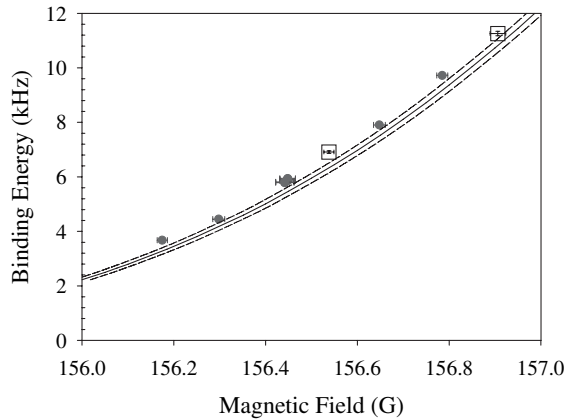
FIG. 2. Resonant frequency as a function of magnetic field. The solid line is from Ref. [26] and is the result of fitting our Ramsey measurements with a coupled-channels scattering calculation. The dashed line indicates the uncertainty in that fit. Measurements were made with both condensed and uncondensed ($T = 20$–$80$ nK) $^{85}$Rb clouds. The open squares indicate BEC measurements and the closed circles represent thermal cloud measurements. The data has been corrected for the temperature dependent shifts discussed in the text. The uncertainty in the measured frequency is represented by the width of the points.
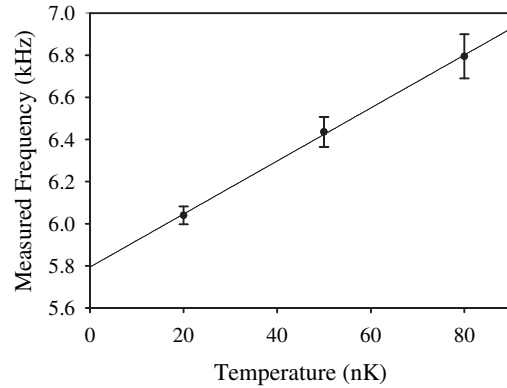


FIG. 3. Measured resonant frequency at 156.44 G as a function of the temperature of the atomic sample. Fitting the data to a straight line gives a frequency shift equal to $0.60(2) k_B T$ where $k_B$ is Boltzmann's constant and $T$ is the temperature.

lines represent the uncertainty in that fit. The solid circles are measurements made with uncondensed thermal clouds that have been corrected for the temperature shift discussed below. The open squares are measurements made with much denser, partially condensed clouds.

One would expect that the exact transition frequency would depend on the relative energy of the free atoms, and we see this as a temperature dependent shift in the measured resonant frequency as shown in Fig. 3. In order to form molecules, hotter, more energetic atoms require higher frequency photons to carry away their excess energy. We have measured the resonant frequency at 155.44 G for clouds with temperatures of 20, 50, and 80 nK. Over this range the shift in the resonant frequency is linear with a slope of 0.0126(4) kHz/nK which corresponds to $0.60(2) k_B T$ where $k_B$ is Boltzmann's constant and $T$ is the temperature of the sample. From the results of Ref. [17] we expect this to be lower than the $1.5 k_B T$ relative energy, but the precise factor requires a theory of the formation process which does not yet exist. In Ref. [17] we showed that the pairing is selective and favors atom pairs with a lower relative energy than the sample average. The thermal cloud data points shown in Fig. 2 have been corrected to show the zero temperature resonant frequency. Most of the error bars for these measurements overlap with the error bars on the binding energy curve but it is interesting that they are all consistently shifted to slightly higher frequency. This is consistent with the peak of the Feshbach resonance being about 40 mG lower than the value found in Ref. [26].

The two BEC measurements were carried out with clouds which had 50% condensate fractions. We found

the results of BEC measurements to show larger statistical fluctuations which may be due to shot-to-shot fluctuations in the phase space density of the partially condensed samples. The sample was not significantly heated during the molecule production process and the condensate fraction remained the same to within our level of uncertainty (approximately 15%). Small corrections (0.03–0.06 kHz) due to the 5–10 nK thermal cloud fractions have been made to the measurements shown in Fig. 2. We assumed that the thermal fraction and condensate fraction have slightly different resonant frequencies so that the frequency we measure is an average of the two. The measurement at 156.54 G lies above the best fit line for measurements made with lower density thermal clouds and hence may show evidence for a mean field shift to the binding energy. A shift consistent with this one was also observed using our Ramsey technique as reported in Ref. [26].

We have thoroughly investigated the resonant conversion efficiency at 156.45 G as a function of coupling time. Figure 4 shows that the conversion efficiency increases with coupling time until a saturated value is reached. Interestingly, for a given phase space density, this value is the same as we would expect if we had instead ramped the magnetic field adiabatically across the Feshbach resonance [17]. In both cases the maximum possible conversion is determined just by the phase space density. However, this new technique allows much higher phase space densities (and hence higher conversion efficiencies) because the cloud is heated much less. The relevant phase space densities to compare are the ones after the conversion process. We have confirmed that this same phase space limit on conversion efficiency applies at all magnetic field values we have investigated and at this time it is not understood why the efficiencies of these two conversion schemes have the same dependence on phase space density.

Figure 4(b) shows the converted fraction in the short coupling time regime. We observe Rabi-like oscillations in
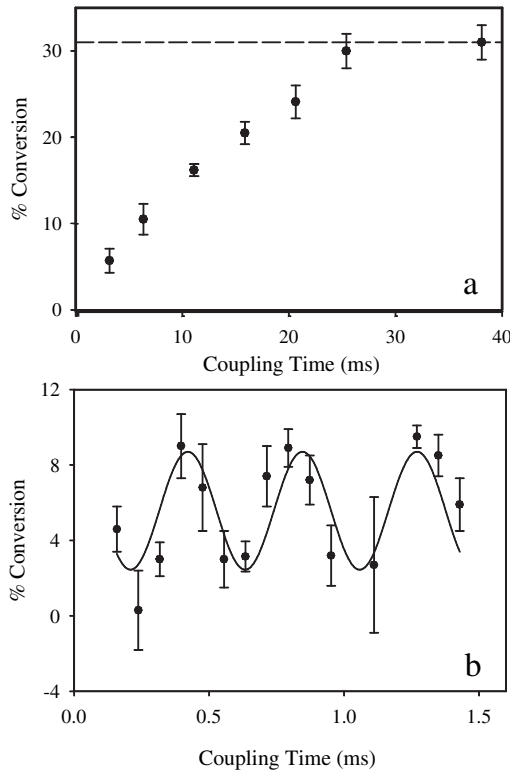
FIG. 4. (a) Percentage of atoms converted to molecules at 156.45 G (using 6.5 kHz resonant modulation frequency) as a function of coupling time. The number of molecules increases with coupling time until the conversion becomes saturated. (b) Oscillations in the converted fraction as a function of coupling time. The oscillation frequency is approximately 2.5 kHz and the damping time is approximately 4 ms.

the atomic population at a frequency of about 2.5 kHz. Note that the initial peak-to-peak amplitude of these oscillations is only 6% of the total atom number, making this measurement rather difficult. We have mapped out the oscillations for slightly longer coupling times and observe that, as one would expect, they damp out with a time constant of 4 ms which is of the same order as the molecular lifetime. One question that remains is why the initial oscillation amplitude (6%) is much less than the maximum converted fraction of 30%. In a simple two level system one would expect a Rabi oscillation amplitude of 30% because the maximum conversion would occur during the first Rabi cycle. Here, we observe a slow build up of molecules over a time scale much slower than the Rabi frequency. It is unclear what determines the coupling strength relevant to both these Rabi-like oscillations and the time scale associated with reaching the saturated conversion regime.

In summary, we have devised a new and very useful technique for atom-molecule coupling in the vicinity of a Feshbach resonance that offers superb control of the conversion process. It allows conversion efficiencies that are

higher than those achieved by ramping the magnetic field through the Feshbach resonance because it greatly reduces problems associated with working close to resonance such as heating and rapid collisional loss. For example, by applying a resonant modulation with an amplitude of 0.5 G to a pure BEC for 1.6 ms we observe 55% conversion. We have observed Rabi-like oscillations between atomic and molecular populations that damp out on the time scale of the molecular lifetime. This new technique also allows precise spectroscopic measurements that are comparable to the results of our Ramsey measurements in Ref. [26].

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