Efficient creation of molecules
from a cesium Bose-Einstein condensate

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Abstract. – We report a new scheme to create weakly bound Cs_2 molecules from an atomic
Bose-Einstein condensate. The method is based on switching the magnetic field to a narrow
Feshbach resonance and yields a high atom-molecule conversion efficiency of more than 30%, a
factor of three higher than obtained with conventional magnetic-field ramps. The Cs_2 molecules
are created in a single g-wave rotational quantum state. The observed dependence of the
conversion efficiency on the magnetic field and atom density shows scattering processes beyond
two-body coupling to occur in the vicinity of the Feshbach resonance.

The newly emerging field of quantum-degenerate molecules provides intriguing possibilities for controlled studies of multicomponent matter-wave systems. Chemical reactions are expected to show effects of coherence, matter-wave interference, quantum tunneling, and bosonic stimulation. Recently, coherent atom-molecule couplings [1] have been observed in a Bose-Einstein condensate (BEC), and molecular quantum gases [2,3] and molecular BECs [4] have been realized. The key ingredient in these experiments has been the presence of magnetically induced Feshbach resonances [5]. These resonances provide the variable coupling between atoms and molecules as a function of an external magnetic field and allow the conversion of atoms to molecules and vice versa.

In the previous experiments on the creation of ultracold Cs_2, Na_2 and Rb_2 molecules from the corresponding atomic BECs [2, 3, 6] the molecules are formed by ramping the magnetic field through a Feshbach resonance; see illustration in fig. 1. It is expected that during the ramping process the ground-state atom population in the trap is adiabatically and efficiently converted into molecules in a weakly bound state [7]. However, the reported efficiencies using this method are relatively low: Typically 5%–10% are observed, whereas up to 50% to 70% of the atoms are lost during the ramping process. The missing fraction, the lost atoms which are not converted into weakly bound molecules, is generally believed to result from the creation
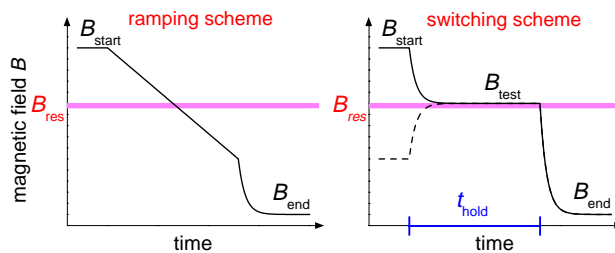
Fig. 1 – Schemes for molecule creation near a Feshbach resonance, located at B_{res}. In the ramping scheme, we linearly ramp the magnetic field from B_{start}, well above the resonance, to some value well below the resonance, and then quickly to B_{end}. In the switching scheme, we first switch from B_{start} to B_{test}. After a hold time t_{hold}, we switch the field to B_{end}. The switching scheme also works for B_{start} below the resonance (dashed line). The finite response time of the magnetic field in our experiment is due to eddy currents in the stainless-steel vacuum chamber.

of molecules in states which cannot be detected by the conventional imaging method, or to consist of "hot" atoms which quickly leave the trap [2, 8].

In this paper, we report a high atom-molecule conversion efficiency in excess of 30% from an atomic BEC based on a novel switching scheme. This scheme is illustrated in fig. 1. The magnetic field is quickly switched from an off-resonant value B_{start} to a field B_{test}, near the resonance position B_{res}. After a variable hold time t_{hold}, the magnetic field is quickly lowered well below the resonance B_{end}, where atoms and molecules decouple and can be independently measured. Our new scheme works for initial magnetic fields B_{start} both well above or well below the resonance. In the latter case, the creation of molecules cannot be explained in terms of the two-body adiabatic conversion picture [7]. An investigation on the atom loss and molecule creation efficiencies suggests that different scattering processes are involved near the narrow Feshbach resonance.

The cesium molecules we create are of special interest since they have a large orbital angular momentum ($l = 4$). Coupling from ultracold atoms in an *s*-wave scattering state to the *g*-wave molecular states is observed only for cesium atoms due to the large indirect spin-spin coupling [9]. Many narrow Feshbach resonances of this kind were observed at low magnetic fields for cesium atoms polarized in the lowest internal state $|F = 3, m_F = 3\rangle$ [10], where F is the total angular momentum and m_F is the magnetic quantum number. Based on these narrow resonances, the formation of thermal molecules was investigated [11] and a pure molecular quantum gas was created from an atomic BEC [2].

Our experiments start with a pure BEC of cesium with up to 2.2×10^5 atoms in the ground state $|F = 3, m_F = 3\rangle$ [12, 13]. The magnetic field is set to $21\,\text{G}$, corresponding to an atomic scattering length of $210 a_0$, where a_0 is the Bohr radius. The magnetic field gradient is set to $31.3\,\text{G/cm}$ for levitation of the atoms [12]. The condensate is confined in a crossed dipole trap formed by two horizontally intersecting laser beams, which are derived from a broad-band Yb fiber laser at $1064\,\text{nm}$. One tightly focused beam with a waist of $35\,\mu\text{m}$ and a power of $0.5\,\text{mW}$ essentially provides the radial confinement; the other beam with a waist of $300\,\mu\text{m}$ and power of $350\,\text{mW}$ essentially provides the axial confinement. The radial and axial trap frequencies are $\omega_r/2\pi = 17.5\,\text{Hz}$ and $\omega_z/2\pi = 4.7\,\text{Hz}$, respectively. The chemical potential is $k_B \times 11\,\text{nK}$, where k_B is Boltzmann's constant.

Before we start the molecule creation, we first compress the condensate by adiabatically increasing the power of the tightly focused laser in $0.7\,\text{s}$ to $40\,\text{mW}$. The higher laser power provides a stronger trapping force and allows us to turn off the levitation field. The absence
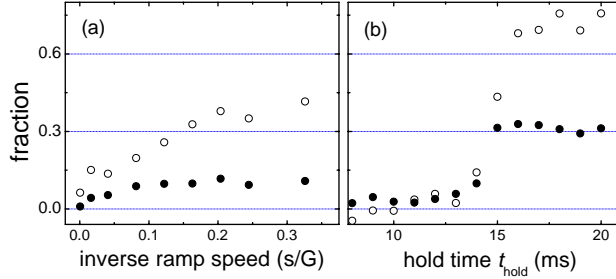
Fig. 2 – Comparison of the two schemes of molecule creation. Molecule fraction (solid circles) and atom loss fraction (open circles) are shown for (a) the ramping scheme, where the fractions are measured for different ramp speeds, and for (b) the switching scheme for different hold times t_{hold}. In (b), B_{test} is set right on resonance.

of the magnetic field gradient is crucial to ensure that all atoms experience the same magnetic field and can simultaneously participate in the molecule formation process. In the compressed trap, the trap frequencies are $\omega_r/2\pi = 170\,\text{Hz}$ and $\omega_z/2\pi = 6.5\,\text{Hz}$, the chemical potential is $k_{\text{B}} \times 86\,\text{nK}$ and the peak density is $1.7 \times 10^{14}\,\text{cm}^{-3}$. We then slowly change the magnetic field in 200 ms to a starting value of B_{start}, typically 0.5 G above the Feshbach resonance B_{res}. Note that this 0.5 G offset is much larger than the resonance width of a few mG. The condensate at B_{start} is not influenced by the resonance. We then switch off the dipole trap and release the atoms into free space and, at the same time, tune the magnetic field toward the Feshbach resonance to create molecules. At the end of the molecule formation phase, we quickly lower the magnetic field down to $B_{\text{end}} \approx 18\,\text{G}$ to decouple the molecules and atoms.

The resulting molecule and atom numbers can be determined independently by absorption imaging [2]. The atoms are directly imaged at 18 G. We verify that the molecules are insensitive to the imaging beam at this magnetic field. To detect the molecules, we first blast away the atoms at 18 G with a resonant beam [3], and then ramp the magnetic field back above the resonance to 21 G. The weakly bound molecular state is then above the continuum and the molecules quickly dissociate into free atoms [14]. By imaging the cloud of the resulting atoms, we can determine the molecule number. We define the corresponding molecule fraction as the number of atoms detected after dissociation normalized to the initial atom number. This is then, by definition, equal to the atom-molecule conversion efficiency.

We employ both the ramping scheme and the switching scheme for molecule production (see fig. 1) and compare their performances. In the ramping scheme, we tune the magnetic field across the resonance with a constant ramping speed. Based on the resulting molecule number and the loss in atomic number, we calculate the conversion fractions, shown in fig. 2(a). A maximum molecule fraction of 10% is observed when the ramps are slower than 10 G/s. The atom loss for these ramps is about 40%, which indicates a missing fraction of about 30%. This result is similar to all previous experiments using the same method [2,3,6]. For the switching scheme, we quickly tune the magnetic field onto the Feshbach resonance $B_{\text{test}} \approx B_{\text{res}}$, wait for various hold times t_{hold}, and quickly lower the magnetic field to $B_{\text{end}} = 18\,\text{G}$. Due to the finite response time of the magnetic field, the field approaches the Feshbach resonance after about 12 ms [15]. At this time, the peak density of the expanding condensate reduces to $1.1 \times 10^{12}\,\text{cm}^{-3}$ [16]. For hold times $t_{\text{hold}} > 15\,\text{ms}$, molecule fractions of $30 \sim 35\%$ and atom loss fractions of $\sim 70\%$ are reached as shown in fig. 2(b). The conversion efficiency is by more than a factor of three higher than obtained from the ramping scheme. Note that in order
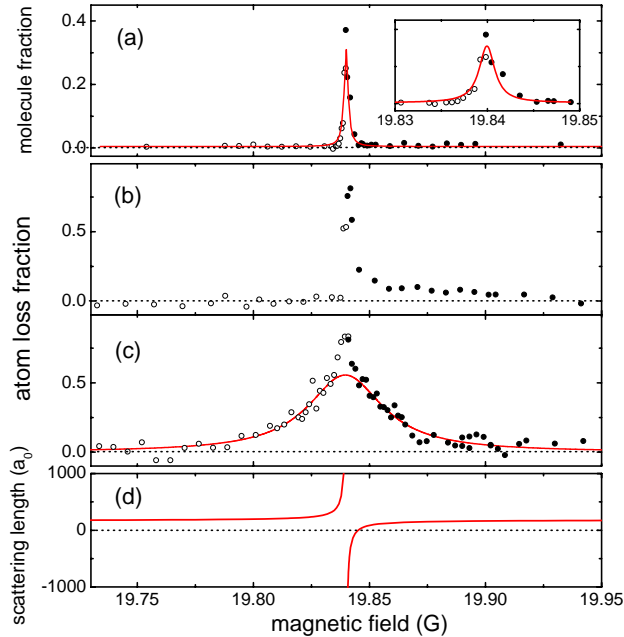
Fig. 3 – Molecule creation and atom loss near the Feshbach resonance. Based on the switching scheme, molecule increase in $t_{\text{hold}} = 18\,\text{ms}$ (a) and atom loss in $18\,\text{ms}$ (b) are measured for various test fields B_{test}. The inset shows the expanded view of the molecule signal. For comparison, the atom trap loss in the compressed trap is shown in (c). The scattering length is shown in (d) for reference. Solid circles (open circles) show the measurements above (below) the resonance. In (a), a Lorentzian fit (solid line) yields a width of $2.1\,\text{mG}$ and the resonance position of $B_{\text{res}} = 19.840\,\text{G}$, subject to a calibration uncertainty of $4\,\text{mG}$. Fitting both wings in (c) gives a Lorentzian width of $40(2)\,\text{mG}$.

to precisely set the magnetic field right on the narrow Feshbach resonance, we synchronize the experiment with the $50\,\text{Hz}$ line voltage to reduce the effects of the ambient magnetic-field ripple, for which we measure an amplitude of $4\,\text{mG}$. This suppresses uncontrolled magnetic-field variations to about $1\,\text{mG}$.

To understand the different performance of the two schemes, we study the atom loss and molecule increase at different magnetic fields B_{test} based on the switching scheme, see fig. 3(a) and (b). For comparison, we also show the atom loss in the compressed trap in fig. 3(c), where the initial peak density is $1.9 \times 10^{14}\,\text{cm}^{-3}$ [17]. The calculated scattering length is shown in fig. 3(d) [18]. For all measurements with B_{test} above the resonance, we prepare the condensate as previously described at $B_{\text{start}} = B_{\text{res}} + 0.5\,\text{G}$. For B_{test} below the resonance, we prepare the condensate at a magnetic field below the resonance by quickly switching the magnetic field from the initial value to $B_{\text{res}} - 0.5\,\text{G}$. No appreciable atom loss, molecule formation or condensate excitation is observed in this process. We then follow the same experimental procedure, but approach the resonance from below. These two different preparation procedures for magnetic fields above and below the resonance are necessary to avoid a slow field-sweep across the resonance, which can lead to systematic atom loss or molecule increase.

In the molecule creation spectrum (fig. 3(a)) we observe a very narrow linewidth of $2.1\,\text{mG}$, which is consistent with the predicted resonance width. Notably, our molecule creation scheme also works for B_{start} below the resonance, which suggests that coupling beyond the adiabatic
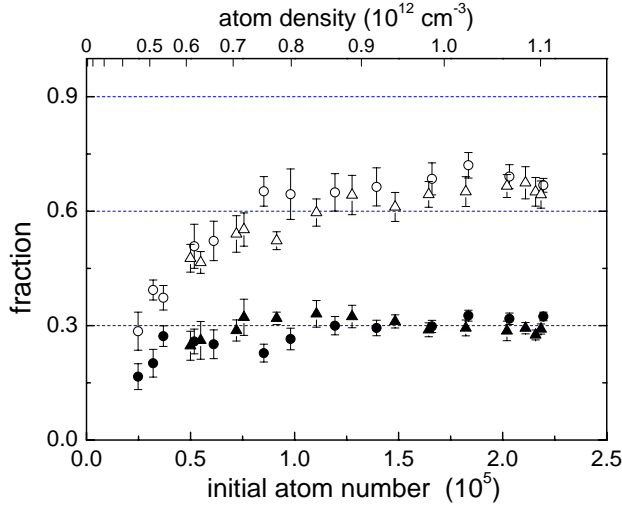
Fig. 4 – Density dependence of the atom loss fraction (open symbols) and molecule fraction (solid symbols). The atom number in the BEC is varied by either adjusting optical cooling efficiencies (solid and open circles) or the loading efficiencies into the crossed dipole trap (solid and open triangles). The corresponding atomic density of the sample right before the molecule formation (after 12 ms expansion in free space) is given on the top axis.

conversion model plays an important role in the creation process. In the adiabatic passage picture, molecules cannot be created when the creation field is below the resonance. The atom loss, shown in fig. 3(b), is asymmetric and seems to include two components, a narrow peak on resonance and a much broader and weaker loss feature for magnetic fields above the resonance. The narrow peak has a similar width as in the molecule production spectrum in fig. 3(a), and is clearly related to the observed molecule formation. The broad and weak feature on the high magnetic-field side has a width of 80(20) mG as determined from a one-sided Lorentzian fit. To obtain further information about the atom loss process, we measure the atom loss in the compressed trap, where the atom density is higher by a factor of ~ 170 than in fig. 3(a) and (b). The result shown in fig. 3(c) displays a wide and symmetric loss feature. By fitting the two wings to a Lorentzian profile, we find a width of 40(2) mG.

The different lineshapes suggest that different scattering processes are involved near the Feshbach resonance. The molecule formation width is close to the predicted width of the Feshbach resonance and can be interpreted in terms of the two-body Feshbach coupling. The asymmetric loss feature in fig. 3(b) and the trap loss may be due to three-body recombination or many-body effects. These broad atom loss features are puzzling, since they are a factor of 20 or more wider than the Feshbach resonance width of 2 mG. The physical origin of the associated loss mechanisms requires further investigation.

The large width of the atom loss feature, however, does provide a qualitative explanation why the switching scheme is more efficient than the ramping scheme. In a linear ramp, atoms sample all magnetic fields near the resonance which, for a large fraction of time, leads to atom loss without molecule increase. With the switching scheme, the atoms spend more time in the magnetic-field range where the molecules can be created.

To further investigate and differentiate the physical mechanisms that are responsible for the missing fraction and for the molecule increase, we measure the dependence of the atom loss

fraction and molecule fraction on the atom number in the condensate, as shown in fig. 4. Atom numbers ranging from 2.5×10^4 to 2.2×10^5 correspond to peak densities of $7.9 \times 10^{13}\,\mathrm{cm}^{-3}$ to $1.9 \times 10^{14}\,\mathrm{cm}^{-3}$ in the compressed trap and to $4.6 \times 10^{11}\,\mathrm{cm}^{-3}$ to $1.1 \times 10^{12}\,\mathrm{cm}^{-3}$ in free space at the moment the molecules are created. For the calculation of the densities in free space, we take into account the 12 ms expansion of the condensate in the Thomas-Fermi regime after the dipole trap is turned off.

Several interesting features show up in the density dependence. The molecule fraction grows and saturates to $\sim 30\%$ at densities higher than $5 \times 10^{11}\,\mathrm{cm}^{-3}$. The saturation of the molecule fraction resembles observations in a thermal gas [11,19], where a thermal equilibrium is reached with a constant molecule fraction in the sample [20]. The missing fraction is very small at low densities and continues to grow up to a density of $8 \times 10^{12}\,\mathrm{cm}^{-3}$. The stronger density dependence of the missing fraction suggests that scattering processes involved in the atom loss are of higher order than for the molecule increase. Similar enhancement of the collision loss near the Feshbach resonance was also observed in a ^{85}Rb condensate [21] and in a thermal Cs gas [22]. A further analysis on the scattering dynamics and the possible thermal equilibrium condition is necessary.

In conclusion, we show that an atom-molecule conversion fraction of more than 30% can be reached based on a magnetic-field switching scheme. The performance of this scheme is superior to the conventional linear magnetic-field ramping scheme since the molecules are created only within the narrow Feshbach resonance width of 2 mG, while the atoms are lost over a much large range of ~ 40 mG. The density dependence of both the missing fraction and the molecule fraction suggests that in our scheme the molecules are created via Feshbach coupling, while the missing fraction comes from higher-order scattering processes. Based on the new creation scheme, we are now able to obtain samples with up to 40000 ultracold molecules. This provides a good starting point to investigate the trapping, the interactions, and the matter-wave nature of ultracold molecules.

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[15] The response time of $t_{1/e} = 1.54$ ms for the magnetic field is limited by the eddy currents from the vacuum chamber. Starting from 500 mG above the resonance, we estimate the magnetic field will take $\ln(500\,\mathrm{mG}/2\,\mathrm{mG})t_{1/e} \approx 9$ ms to settle within the resonance width. This result is close to the measured delay time of 12 ms.

[16] We observe that the expansion rate of the molecules is very close to that of the expanding condensate.

[17] This value is slightly higher than the value given before because of the reduced scattering length.

[18] The calculated resonance position is 20.1 G. To reflect the correct resonance position, we shift the numerical data by centering the resonance at our observed value of 19.84 G.

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