Pure Gas of Optically Trapped Molecules Created from Fermionic Atoms

S. Jochim, M. Bartenstein, A. Altmeyer, G. Hendl, C. Chin, J. Hecker Denschlag, and R. Grimm *Institut für Experimentalphysik, Universität Innsbruck, Technikerstraße 25, 6020 Innsbruck, Austria* (Received 5 August 2003; published 8 December 2003)

We report on the production of a pure sample of up to 3×10^5 optically trapped molecules from a Fermi gas of 6 Li atoms. The dimers are formed by three-body recombination near a Feshbach resonance. For purification, a Stern-Gerlach selection technique is used that efficiently removes all trapped atoms from the atom-molecule mixture. The behavior of the purified molecular sample shows a striking dependence on the applied magnetic field. For very weakly bound molecules near the Feshbach resonance, the gas exhibits a remarkable stability with respect to collisional decay.

DOI: 10.1103/PhysRevLett.91.240402

The formation of composite bosons by pairing of fermions is the key to many intriguing phenomena in physics, with superfluidity and superconductivity being prominent examples. In ultracold atomic gases, pairs of fermionic atoms can be combined to form bosonic molecules [1,2] or possibly Cooper pairs [3]. The pairing changes the properties of the gas, highlighted by the prospect of a molecular Bose-Einstein condensate or a Cooper-paired superfluid. The interatomic interactions play a crucial role for the nature of the pairing process. The ability to control the interaction via magnetically tuned Feshbach resonances [4–6] opens up exciting possibilities for experiments on ultracold fermionic gases, e.g., exploring superfluidity in different pairing regimes [7–10].

The formation of molecules near Feshbach resonances in ultracold gases has been reported for bosons [11–14] and fermions [1,2]. In the experiments [1,2,11,12], the molecules coexist with the atoms in a strongly interacting mixture. A generic feature of a Feshbach resonance is the existence of a bound molecular state with a magnetic moment that differs from that of the unbound atom pair. The binding energy thus depends on the magnetic field, and a properly chosen field can resonantly couple colliding atoms into the molecular state. The inherent difference in magnetic moments facilitates a Stern-Gerlach selection of molecules and atoms. Two recent experiments [13,14] demonstrate the separation of the molecular from the atomic cloud in free space.

In this Letter, we report the creation of a pure sample of up to 3×10^5 optically trapped molecules from a fermionic gas of ^6Li atoms. After the production of an atom-molecule mixture via three-body collisions, a Stern-Gerlach purification scheme efficiently removes all trapped atoms, while leaving all molecules trapped. This allows us to investigate the intriguing behavior of the pure molecular sample, which strongly depends on the magnetic field.

The lithium isotope ⁶Li is one of the two prime candidates in current experiments exploring the physics of fermionic quantum gases [15–19], the other one being

⁴⁰K [1,20]. A spin mixture composed of the lowest two sublevels in the hyperfine manifold of the electronic ground state is stable against two-body decay and exhibits wide magnetic tunability of s-wave interactions via a broad Feshbach resonance at about 850 G [21]. A calculation of the corresponding scattering length a as a function of the magnetic field [22] is shown in Fig. 1(a) [23]. The large cross section for elastic scattering near the resonance can be used for efficient evaporative cooling, in particular, above the resonance at negative scattering length where inelastic loss is negligible [16]. In the region of positive scattering length below the resonance, loss features have been observed [24]. At large positive a, a weakly bound molecular level exists with a binding energy approximately given by $\hbar/(ma^2)$, where \hbar is Planck's constant and m denotes the atomic mass. For the region of interest, Fig. 1(b) shows this binding energy as calculated

PACS numbers: 03.75.Ss, 05.30.Fk, 32.80.Pj, 33.80.Ps

The starting point of our experiments is a sample of 2.5×10^6 ⁶Li atoms in a standing-wave optical dipole trap realized with a Nd:YAG laser at a wavelength of 1064 nm [19,26]. The 50-50 spin mixture in the lowest two spin states is spread over ~ 1500 individual lattice sites of the standing-wave trap. In the central region of the trap, a single site contains typically 1800 atoms. The

from the scattering length data [25].

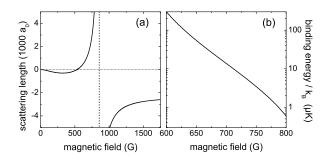


FIG. 1. (a) Magnetic-field dependence of the *s*-wave scattering length a in the 6 Li spin mixture. An additional, narrow Feshbach resonance near 550 G [22] is omitted in the plot. (b) Binding energy of the weakly bound molecular level in the region of large positive a.

axial and radial trap frequencies are 260 kHz and 390 Hz, respectively. The trap depth is $k_{\rm B} \times 27~\mu{\rm K}$ with $k_{\rm B}$ denoting Boltzmann's constant. At a temperature of 2.5 $\mu{\rm K}$, peak values for the number density and phase-space density are $3\times 10^{12}~{\rm cm}^{-3}$ and 0.04 [27,28], respectively. The ultracold gas is prepared by forced evaporative cooling after loading the optical trap at an initial depth of $\sim 1~{\rm mK}$ with 8×10^6 atoms from a magneto-optical trap (MOT). The evaporation is performed by ramping down the light intensity in 1 s at a magnetic field of 1200 G. The evaporation initially proceeds with very high efficiency similarly to [16,29], but finally loses its efficiency when the tightly confining lattice potential does not support more than one or two quantum states.

We form molecules in the weakly bound level at a field of 690 G, where we find optimum production rates at a large positive scattering length of $a=+1300a_0$. Here a_0 denotes Bohr's radius. To reach the production field of 690 G, we quickly ramp from the evaporation field of 1200 G down to this value with a speed of -7.5 G/ms. In contrast to other experiments with fermionic atoms [1,2], the molecule formation during this ramp is negligible and the molecules are predominantly formed after the ramp at the fixed production field.

The molecules are detected by dissociating them into atoms [1,2,13,14] and measuring their fluorescence. For this purpose, we apply a ramp across the Feshbach resonance to fields of typically 1200 G (speed +6 G/ms). This brings the weakly bound level above the scattering continuum and the molecules quickly dissociate. The dissociation turns out to be insensitive to variations of the ramp speed and the final field. After the dissociation ramp, we immediately ramp down to zero magnetic field. The ramp speed of -12 G/ms is fast enough to avoid molecule formation when crossing the region of positive scattering length. After reaching zero magnetic field, we recapture all atoms into the MOT. Their number is then determined by measuring the emitted fluorescence intensity using a calibrated photodiode [27]. This measurement provides the total atom number $2N_{\text{mol}} + N_{\text{at}}$, where N_{mol} and $N_{\rm at}$ denote the number of molecules and atoms after the production phase, respectively. To determine $N_{\rm at}$, we repeat the same measurement without the Feshbach dissociation ramp by immediately ramping down to zero from the production field. The ramp down to zero magnetic field increases the binding energy to a large value of about $k_{\rm B} \times 80~{\rm mK}$ and the molecules are lost without leading to any fluorescence light in the MOT. The number of molecules N_{mol} is then obtained by taking the difference in atom numbers measured in two subsequent runs with and without the dissociating Feshbach ramp.

The creation of molecules from the atomic gas is demonstrated in Fig. 2 for the optimum production field of 690 G. The time evolution of the measured numbers $2N_{\rm mol} + N_{\rm at}$ and $N_{\rm at}$ is shown together with the corresponding number of molecules $2N_{\rm mol}$. We attribute the molecule formation to three-body recombination into the

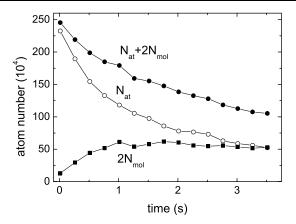


FIG. 2. Formation of molecules at a fixed magnetic field of 690 G. The measured numbers $N_{\rm at} + 2N_{\rm mol}$ and $N_{\rm at}$ are plotted as a function of time together with the resulting number of molecules $2N_{\rm mol}$.

weakly bound state [30,31]. Two-body processes cannot lead to bound dimers as a third particle is required for energy and momentum conservation. The three-body molecule formation process can be modeled with the differential equation $\dot{N}_{\rm mol}/N_{\rm at}=M_3\langle n_{\rm at}^2\rangle$, where $\langle n_{\rm at}^2\rangle$ denotes the mean quadratic density of the atoms. From the initial molecule formation rate of $\dot{N}_{\rm mol}=3.5\times10^5~{\rm s}^{-1}$, we thus derive a three-body formation coefficient of $M_3=1\times10^{-25}~{\rm cm}^6/{\rm s}^{-1}$ [27]. The maximum number of 3×10^5 molecules is reached after about 1 s. For longer times, the fraction of atoms forming molecules approaches a value of $\sim50\%$.

At the optimum production field of 690 G, the molecular binding energy amounts to $\sim k_{\rm B} \times 18~\mu{\rm K}$, which is in between the thermal energy of $k_{\rm B} \times 2.5~\mu{\rm K}$ and the trap depth of $k_{\rm B} \times 27~\mu{\rm K}$ for the atoms. For the molecules, the trap depth is a factor of 2 higher because of the 2 times larger polarizability. We have verified this fact by measuring the trap frequencies for atoms and molecules to be equal within the experimental uncertainty of a few percent. After a three-body recombination event both the atom and the molecule remain trapped. We believe that the recombination heat is cooled away by a evaporation of atoms out of the trap. Evaporative loss of molecules is strongly suppressed because of the higher trap depth.

To purify the created molecules we use a Stern-Gerlach selection technique. We apply a magnetic field gradient perpendicular to the standing-wave axis. This pulls particles out of the trap for which the magnetic force is larger than the trapping force. In order to be able to apply large enough field gradients, we lower the trap depth to $k_{\rm B} \times 19~\mu{\rm K}$ while applying the gradient for about 10 ms. Figure 3 demonstrates such a purification at 568 G. While all the atoms are lost above $B'_{\rm at} = 17~{\rm G/cm}$, the molecules start getting spilled at 20 G/cm, and are lost completely above $B'_{\rm mol} = 32.5~{\rm G/cm}$. This means that, under suitable conditions, we can remove all the atoms while keeping the molecule number constant.

240402-2 240402-2

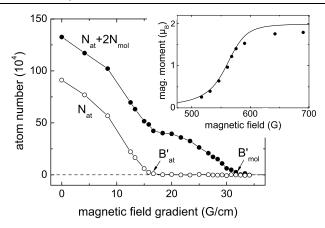


FIG. 3. Stern-Gerlach selection by applying a magnetic field gradient to the trapped atom-molecule mixture at 568 G and a trap depth of $k_{\rm B} \times 19~\mu{\rm K}$. Marked are the two gradients where all the atoms and all the molecules are lost. The inset shows the magnetic moment of the molecules estimated from the Stern-Gerlach selection at different magnetic fields together with the theoretical calculation.

The magnetic moment of the molecules μ_{mol} can be estimated to be $\mu_{mol} = 2\mu_{at}B'_{mol}/B'_{at}$, where μ_{at} is the magnetic moment of one free atom. At high magnetic field, μ_{at} equals Bohr's magneton μ_{B} . The inset of Fig. 3 shows the magnetic moments of the molecules determined at various magnetic fields. The data agree well with the magnetic field dependence calculated from theory (solid curve). We attribute the systematic deviation to slightly different trap parameters for atoms and molecules.

Starting with a pure molecular sample, we study its stability against inelastic molecule-molecule collisions. Corresponding decay curves are displayed in Fig. 4 for two different magnetic fields. At 546 G a rapid nonexponential decay is observed as a clear signature of inelastic molecule-molecule collisions. From the initial decay rate we derive a two-body loss coefficient of 5×10^{-11} cm³/s [27]. At 690 G, the observed behavior is strikingly different. The molecular sample shows a nearly exponential

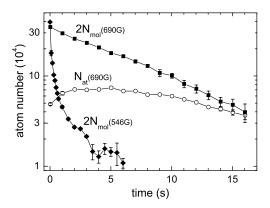


FIG. 4. Time evolution of an initially pure sample of molecules at 546 G (\spadesuit) and at 690 G (\blacksquare). At 690 G, atoms are observed to reappear (\bigcirc).

decay with a time constant as long as ~ 10 s. As similar lifetimes are observed for trapped atom samples under conditions where trapped molecules cannot be created, the observed molecular lifetime can be fully attributed to one-body effects such as heating in the optical trap. For a loss rate coefficient at 690 G our data provide an upper limit of 3×10^{-13} cm³/s [27], which is surprisingly low for inelastic collisions in a molecular system with many open exit channels.

The data at 690 G show another interesting collisional effect. Atoms reappear after purification of the molecular cloud, see (○) in Fig. 4. For long storage times (~15 s), an atom-molecule mixture is reestablished with a similar fraction of molecules as observed in the initial formation process at the same magnetic field (see Fig. 2). Collisions producing atoms from molecules are endoergic in nature as kinetic energy is required to provide the dissociation energy. The increasing atom fraction does not lead to any increased loss. This shows that the gas is remarkably stable both against molecule-molecule and atommolecule collisions.

The dependence of the molecular decay on the magnetic field is shown in Fig. 5. Here we store the initially pure gas of 1.8×10^5 molecules at a variable magnetic field for a fixed holding time of 1 s before we measure the number of remaining molecules and atoms. A sharp transition occurs around 650 G. For fields below $\sim\!600$ G, where the binding energy is relatively large ($>\!k_{\rm B}\!\times 100~\mu{\rm K}$), the observed decay is very fast and no atoms are found to reappear. Here inelastic collisions apparently lead to a rapid vibrational quenching. Furthermore, the kinetic energy of the molecules cannot provide the necessary energy for collisional dissociation. Consequently, we do not observe any atoms reappearing.

For fields above \sim 680 G, a completely different behavior is observed. In this regime, no significant loss occurs in the total number $2N_{\rm mol} + N_{\rm at}$. However, an increasing atom fraction is observed as a result of collisional dissociation of the molecules. Here the binding energy

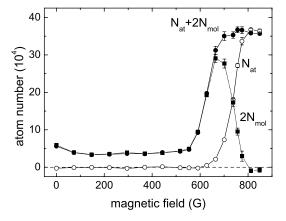


FIG. 5. Remaining number of atoms $N_{\rm at}$, $N_{\rm at} + 2N_{\rm mol}$ and $2N_{\rm mol}$ after a 1-s hold time at variable magnetic field starting with a pure molecular sample.

240402-3 240402-3

approaches the thermal energy and the sample tends towards a thermal atom-molecule equilibrium. Close to the Feshbach resonance, where the binding energy becomes comparable to thermal energy, the atomic fraction dominates in the atom-molecule mixture.

In conclusion we have produced an ultracold, pure molecular gas of ⁶Li dimers in an optical dipole trap. Close to the Feshbach resonance, where the molecular binding energy is small, there is a strong coupling of the atomic gas and the molecules. Three-body collisions between atoms form molecules and collisions break up molecules to produce atoms. Our observations show that this exchange between atomic and molecular fraction can be nearly lossless. The long molecular lifetime along with a large elastic collision rate between the particles opens up great perspectives for further evaporative cooling of the molecular gas to Bose-Einstein condensation. Given the maximum molecule number of 3×10^5 and a temperature of about 2.5 μ K, we reach a phase-space density of 0.01, only a factor of 4 lower than our initial atomic phase-space density. The molecular sample may be further cooled to condensation by efficient evaporation. Out of a mixture of atoms and molecules, mainly atoms will evaporate because they are more weakly trapped than the molecules. The gas is cooled further when molecules break up into atoms since this is an endoergic process. Once quantum degeneracy is accomplished it will be very interesting to cross the Feshbach resonance in order to observe the transition to a strongly interacting superfluid Fermi gas [7–10].

We thank G. Shlyapnikov for very stimulating discussions and V. Venturi for providing us with theoretical data on the scattering length and binding energy. We gratefully acknowledge support by the Austrian Science Fund (FWF) within SFB 15 (project part 15) and by the European Union in the frame of the Cold Molecules TMR Network under Contract No. HPRN-CT-2002-00290.

Note added.—After submission of the present Letter, molecule formation in ⁶Li using the narrow Feshbach resonance at 543 G was reported by Hulet's group [32].

- [1] C. A. Regal, C. Ticknor, J. L. Bohn, and D. S. Jin, Nature (London) **424**, 47 (2003).
- [2] J. Cubizolles, T. Bourdel, S. J. J. M. F. Kokkelmans, G. V. Shlyapnikov, and C. Salomon, preceding Letter, Phys. Rev. Lett. 91, 240401 (2003).
- [3] H. T.C. Stoof, M. Houbiers, C. Sackett, and R. Hulet, Phys. Rev. Lett. 76, 10 (1996).
- [4] H. Feshbach, Ann. Phys. (Leipzig) 19, 287 (1962).
- [5] S. Inouye, M. Andrews, J. Stenger, H.-J. Miesner, D. Stamper-Kurn, and W. Ketterle, Nature (London) 392, 151 (1998).
- [6] T. Loftus, C. Regal, C. Ticknor, J. Bohn, and D. Jin, Phys. Rev. Lett. 88, 173201 (2002).
- [7] M. Holland, S. JJ. M. F. Kokkelmans, M. Chiofalo, and R. Walser, Phys. Rev. Lett. 87, 120406 (2001).

- [8] E. Timmermans, K. Furuya, P.W. Milonni, and A. K. Kerman, Phys. Lett. A 285, 228 (2001).
- [9] M. L. Chiofalo, S. J. J. M. F. Kokkelmans, J. L. Milstein, and M. J. Holland, Phys. Rev. Lett. 88, 090402 (2002).
- [10] Y. Ohashi and A. Griffin, Phys. Rev. Lett. 89, 130402 (2002).
- [11] E. A. Donley, N. R. Claussen, S. T. Thompson, and C. E. Wieman, Nature (London) 417, 529 (2002).
- [12] C. Chin, A. J. Kerman, V. Vuletić, and S. Chu, Phys. Rev. Lett. 90, 033201 (2003).
- [13] J. Herbig, T. Kraemer, M. Mark, T. Weber, C. Chin, H.-C. Nägerl, and R. Grimm, Science **301**, 1510 (2003).
- [14] S. Dürr, T. Volz, A. Marte, and G. Rempe, cond-mat/ 0307440.
- [15] A. G. Truscott, K. E. Strecker, W. I. McAlexander, G. B. Partridge, and R. G. Hulet, Science 291, 2570 (2001).
- [16] K. O'Hara, S. Hemmer, M. Gehm, S. Granade, and J. Thomas, Science 298, 2179 (2002).
- [17] T. Bourdel, J. Cubizolles, L. Khaykovich, K. M. F. Magalhães, S. J. J. M. F. Kokkelmans, G. Shlyapnikov, and C. Salomon, Phys. Rev. Lett. **91**, 020402 (2003).
- [18] S. Gupta, Z. Hadzibabic, M. Zwierlein, C. Stan, K. Dieckmann, C. H. Schunck, E. G. M. van Kempten, B. J. Verhaar, and W. Ketterle, Science 300, 1723 (2003).
- [19] S. Jochim, M. Bartenstein, G. Hendl, J. Hecker Denschlag, R. Grimm, A. Mosk, and M. Weidemüller, Phys. Rev. Lett. **89**, 273202 (2002).
- [20] G. Modugno, G. Roati, F. Riboli, F. Ferlaino, R. J. Brecha, and M. Inguscio, Science 297, 2240 (2002).
- [21] M. Houbiers, H.T.C. Stoof, W. McAlexander, and R. Hulet, Phys. Rev. A 57, R1497 (1998).
- [22] K. O'Hara, S. Hemmer, S. Granade, M. Gehm, J. Thomas, V. Venturi, E. Tiesinga, and C. Williams, Phys. Rev. A 66, 041401(R) (2002).
- [23] The exact resonance position is uncertain within a few 10 G [17], whereas the location of the zero-crossing has been determined to 529 G within 3 G [19,22].
- [24] K. Dieckmann, C. Stan, S. Gupta, Z. Hadzibabic, C. Schunck, and W. Ketterle, Phys. Rev. Lett. 89, 203201 (2002).
- [25] From the scattering length data [22], we calculate the binding energy $\hbar^2/[m(a-\bar{a})^2]$ including a correction with $\bar{a}=29.8a_0$ for lithium. See G. Gribakin and V. Flambaum, Phys. Rev. A **48**, 546 (1993).
- [26] A. Mosk, S. Jochim, H. Moritz, T. Elsässer, M. Weidemüller, and R. Grimm, Opt. Lett. 26, 1837 (2001).
- [27] With fluorescence detection, we can determine the absolute atom number only within a factor of 2. All the derived quantities are subject to this systematic error.
- [28] With a ground-state population of 99% in the tightly confined direction, each lattice site contains a quasi-2D gas. This fact is taken into account in our calculations of number and phase-space densities.
- [29] S. R. Granade, M. E. Gehm, K. M. O'Hara, and J. E. Thomas, Phys. Rev. Lett. **88**, 120405 (2002).
- [30] H. Suno, B. D. Esry, and C. H. Greene, Phys. Rev. Lett. 90, 053202 (2003).
- [31] D. Petrov, Phys. Rev. A 67, 010703(R) (2003).
- [32] K. E. Strecker, G. B. Partridge, and R. G. Hulet, Phys. Rev. Lett. 91, 080406 (2003).

240402-4 240402-4