Precise determination of $^6$Li cold collision parameters by radio-frequency spectroscopy on weakly bound molecules

M. Bartenstein$^1$, A. Altmeyer$^1$, S. Riedl$^1$, R. Geursen$^1$, S. Jochim$^1$, C. Chin$^1$, J. Hecker Denschlag$^1$, and R. Grimm$^{1,2}$

$^1$Institut für Experimentalphysik, Universität Innsbruck, Technikerstraße 25, 6020 Innsbruck, Austria
$^2$Institut für Quantenoptik und Quanteninformation, Österreichische Akademie der Wissenschaften, 6020 Innsbruck, Austria

A. Simoni, E. Tiesinga, C. J. Williams, and P. S. Julienne

Atomic Physics Division, National Institute of Standards and Technology
100 Bureau Drive Stop 8423, Gaithersburg, Maryland 20899

(Dated: March 23, 2022)

We employ radio-frequency spectroscopy on weakly bound $^6$Li$_2$ molecules to precisely determine the molecular binding energies and the energy splittings between molecular states for different magnetic fields. These measurements allow us to extract the interaction parameters of ultracold $^6$Li atoms based on a multi-channel quantum scattering model. We determine the singlet and triplet scattering lengths to be $a_s = 45.167(8)a_0$ and $a_t = -2140(18)a_0$ ($a_0 = 0.0529177$ nm), and the positions of the broad Feshbach resonances in the energetically lowest three $s$–wave scattering channels to be $83.41(15)$ mT, $69.04(5)$ mT, and $81.12(10)$ mT.

PACS numbers: 34.50.-s, 05.30.Jp, 32.80.Pj, 67.40.Hf

Molecular level structure near a collision threshold uniquely determines the scattering properties of ultracold atoms. When a molecular state is tuned near the scattering threshold, the atomic scattering amplitude can be resonantly altered. Magnetically tuned Feshbach resonances in ultracold fermionic gases have recently led to ground-breaking observations, including the condensation of molecules and the study of the crossover physics from a molecular Bose-Einstein condensate to atomic Cooper pairs in the Bardeen-Cooper-Schrieffer state (BEC-BCS crossover). These studies are of general importance in physics as the ultracold Fermi gas provides a unique model system for other strongly interacting fermionic systems.

In spin mixtures of $^6$Li atoms, a broad Feshbach resonance in the energetically lowest $s$-wave channel allows for precise interaction tuning. This, together with the extraordinary stability of the system against inelastic decay, makes $^6$Li the prime candidate for BEC-BCS crossover studies. Precise knowledge of the magnetic-field dependent scattering properties is crucial for a quantitative comparison of the experimental results with crossover theories. Of particular importance is the precise value of the magnetic field where the $s$–wave scattering diverges. At this unique point, the strongly interacting fermionic quantum gas is expected to exhibit universal properties. Previous experiments explored the $^6$Li resonance by measuring inelastic decay, elastic collisions, and the interaction energy, but could only locate the exact resonance point to within a range between 80 mT and 85 mT.

An ultracold gas of weakly bound molecules is an excellent starting point to explore the molecular energy structure near threshold. Improved knowledge on the exact $^6$Li resonance position was recently obtained in an experiment that observed the controlled dissociation of weakly bound $^6$Li$_2$ molecules induced by magnetic field ramps. These measurements provided a lower bound of 82.2 mT for the resonance position. Studies of systematic effects suggested an upper bound of 83.4 mT. Within this range, however, we observe the physical behavior of the ultracold gas still exhibits a substantial dependence on the magnetic field. In this Letter, we apply radio-frequency (rf) spectroscopy on weakly bound molecules to precisely determine the interaction parameters of cold $^6$Li atoms. Together with a multi-channel quantum scattering model, we obtain a full characterization of the two-body scattering properties, essential for BEC-BCS crossover physics.

The relevant atomic states are the lowest three sublevels in the $^6$Li ground state manifold, denoted by $|1\rangle$, $|2\rangle$ and $|3\rangle$. Within the magnetic field range investigated in this experiment, these levels form a triplet of states, essentially differing by the orientation of the nuclear spin ($m_I = 1, 0, -1$). Figure 1 shows the energy level structure of the two scattering channels $|1\rangle + |2\rangle$ and $|1\rangle + |3\rangle$, denoted by $|1,2\rangle$ and $|1,3\rangle$, respectively. The broad Feshbach resonance occurs in the $|1,2\rangle$ channel near 83 mT. When the magnetic field is tuned below the resonance, atoms in the $|1,2\rangle$ channel can form weakly bound molecules. For the $|1,3\rangle$ channel, a similar Feshbach resonance occurs near 69 mT.

Starting with molecules formed in the $|1,2\rangle$ channel, we drive the rf transition to the $|1,3\rangle$ channel at various magnetic field values. The rf excitation can dissociate a molecule into two free atoms (bound-free transition; see Fig. 1) or, for $B < 69$ mT, it can also drive the transition between the molecular states in the $|1,2\rangle$ and $|1,3\rangle$ channels (bound-bound transition). In both processes, the rf excitation results in loss of molecules in the $|1,2\rangle$ channel. This loss constitutes our experimental signal. We perform measurements at different magnetic
fields for both the bound-free and the bound-bound transitions.

Our experimental procedure is similar to Ref. 5. We start with a pure condensate of typically $2 \times 10^9$ molecules at a magnetic field of 76.4 mT 6. The condensate is confined in a weak optical trap, where the peak molecular density is near $10^{11}$ cm$^{-3}$. We then linearly ramp the magnetic field to a specific value between 66 mT and 72 mT in typically 200 ms. After the ramp, we apply a single rf pulse for 200 ms with its frequency tuned close to the atomic transition $|2\rangle$ to $|3\rangle$. Following the rf pulse, we apply state-selective absorption imaging, which is sensitive to free atoms in state $|2\rangle$ and molecules in the $(1, 2)$ channel.

To precisely determine the magnetic field, we employ rf spectroscopy on thermal atoms with temperature $T = 6T_F$, where $T_F$ is the Fermi temperature. Here, the rf transition energy corresponds to the internal energy difference between the states $|2\rangle$ and $|3\rangle$, $hf_0$, where $h$ is Planck’s constant. This energy is magnetic field dependent and the transition frequency is about 83 MHz in the magnetic field range we study. The measured transition has a narrow linewidth of less than 1 kHz, and the center position can be determined to within a few 100 Hz. This high resolution allows us to calibrate our magnetic field to an accuracy of a few $\mu$T based on the Breit-Rabi formula and the $^6$Li parameters given in 21. Within our statistical uncertainty, we do not observe any density-dependent frequency shifts 19.

For bound-free transitions, the molecules in the $(1, 2)$ channel make a transition to the $(1, 3)$ scattering continuum. The excitation rate from a stationary molecule to an atomic scattering state with kinetic energy $2E_k$ is determined by the Franck-Condon factor between the bound and free wavefunctions 22.

The rf dissociation spectra at 72.0 mT and 69.5 mT are shown in Fig. 2. An important feature of the spectra is the sharp rising edge on the low frequency side. This threshold corresponds to the dissociation of a molecule into two atoms with zero relative momentum. Therefore, the position of the edge relative to the atomic transition directly indicates the molecular binding energy.

We determine the dissociation threshold and thus the molecular binding energy by fitting the full lineshape. The lineshape function 22 depends on both the $(1, 2)$ molecular binding energy $E_b$ and the scattering length $a_{13}$ in the $(1, 3)$ channel. In the range of magnetic fields we investigate, $a_{13}$ is much larger than the interaction range of the van der Waals potential of $\sim 30a_0$. The lineshape function $P(E)$ is then well approximated by

$$P(E) \propto E^{-2}(E - E_b)^{1/2}(E - E_b + E')^{-1},$$

where $E = hf - hf_0$ and $E' = h^2/ma_{13}^2$. From the fits to the experimental data 23, we determine the threshold positions, given in Table I. Together with the atomic transition frequencies, we conclude that the molecular binding energies are $E_b = h \times 134(2)$ kHz at 72.013(4) mT and $E_b = h \times 277(2)$ kHz at 69.483(4) mT.

For magnetic field $B < 69$ mT, we can drive the rf transition between the $(1, 2)$ and $(1, 3)$ molecular states. Here, the resonance frequency is given by the energy

Fig. 1: Energy level structure near the $^6$Li$_2$ dissociation threshold as a function of magnetic field $B$. The threshold energy of the $(1, 3)$ scattering channel (upper dotted line) is plotted relative to the $(1, 2)$ threshold (lower dotted line). In the $(1, 2)$ channel, a molecular state (lower solid line) exists below the Feshbach resonance at $\sim 82$ mT. In the $(1, 3)$ channel, another molecular state (upper solid line) exists below the resonance at $\sim 69$ mT. The bound-free and bound-bound transitions of molecules in the $(1, 2)$ channel are illustrated by the arrows.

Fig. 2: Bound-free rf spectra at 72.013(4) mT (a) and 69.483(4) mT (b). Fractional loss in state $|2\rangle$ is measured as a function of the radio frequency. The solid lines are the fit based on Eq. (1). The atomic transition frequencies, which are measured independently, are indicated by the vertical dashed lines.
difference of the two molecular states. To avoid possible systematic mean-field shifts at these lower magnetic fields 19, we prepare a thermal mixture of atoms and molecules with temperature $T \approx T_F$ by a controlled heating method 8. Rf spectroscopy is performed at 67.6 mT and 66.1 mT. The bound-bound transition signal at 66.1 mT is shown in Fig. 3. By fitting the narrow transition line with a Lorentzian profile, we determine the resonance frequency, see Table I. Notably, below the resonance in the $(1, 3)$ channel at $\sim 69$ mT, the bound-free transition is much weaker due to a Fano-type interference effect 22.

Because of the high precision of the measured transition frequencies, a careful analysis of systematic effects is necessary. Possible systematic shifts include differential light shifts of the two molecular states and density-dependent many-body shifts. In order to characterize these possible systematic errors, we experimentally investigate these shifts by varying the trap depth of the optical potential. In a deeper trap, both the differential light shifts and mean-field shifts are expected to increase. We repeat the bound-free and bound-bound rf spectroscopy in traps with different laser powers $P$ between 3.8 mW and 310 mW. We do not see systematic shifts within our statistical uncertainty. The measurements show that these systematic shifts do not exceed the uncertainties given in Table I.

Given the measured data summarized in Table I, it is possible to predict the location of the scattering resonances in the $(1, 2)$, $(1, 3)$ and $(2, 3)$ channels if we have an accurate theoretical model of the collision. We use a standard multi-channel model for the interaction of two $^2$S atoms with nuclear spin 22 to calculate the scattering lengths and bound state energies for these channels. It is only necessary to include $s$-waves in the basis set, since we find that there is a negligible change within the experimental uncertainties if we also include higher partial waves in the basis set. The interaction potential model is the same as described in Ref. 14. It uses a combination of Rydberg-Klein-Rees and $ab$ initio potentials for the singlet $^{1}\Sigma_g^+$ and triplet $^{3}\Sigma_u^+$ states at short range, and joins them smoothly onto long range potentials based on the exchange 23 and van der Waals dispersion energy 24, the lead term of which is $C_6 = 1393.39(16)$ au $(1 \text{ au} = 9.57344 \times 10^{-6} \text{ J nm}^6)$. As in Ref. 14, the singlet $^{1}\Sigma_g^+$ and triplet $^{3}\Sigma_u^+$ scattering lengths, $a_s$ and $a_t$, respectively, are varied by making small variations to the inner wall of the potential. Once $a_s$ and $a_t$ are specified, all other scattering and bound state properties for all channels of two $^6$Li atoms are uniquely determined, including the positions of the resonances. Consequently, varying $a_s$ and $a_t$ to fit the binding energies and energy differences from rf spectroscopy determines the values of these two free parameters.

Fitting the data of the present experiment determines $a_s = 45.167(8)a_0$ and $a_t = -2140(18)a_0$. The uncertainty includes both the uncertainty in the measured value of the magnetic field and the uncertainty in the rf measurements. Our scattering lengths agree within the uncertainties with previous determinations: $a_s = 45.1591(16)a_0$ 18 and $a_t = -2160(250)a_0$ 27. Table I shows a comparison of the measured and best fit calculated energies. The calculated positions of the broad $s$-wave resonances for the $(1, 2)$, $(1, 3)$, and $(2, 3)$ channels are 83.41(15) mT, 69.04(5) mT, and 81.12(10) mT respectively.

Figure 4 shows the scattering lengths calculated for several different channels in the magnetic field range of interest to BEC-BCS crossover experiments. We find that the formula $\alpha = a_0[1 + \Delta(B - B_0)^{-1}][1 + \alpha(B - B_0)]$ fits the calculated scattering lengths to better than 99% over the range of 60 to 120 mT. This expression includes the standard Feshbach resonance term 28 with the background scattering length $a_0$, resonance position $B_0$ and resonance width $\Delta$, and a leading-order correction parameterized by $\alpha$. The respective values for $a_0$, $B_0$, $\Delta$, and $\alpha$ are $-1405a_0$, 83.4149 mT, 30.0 mT, and 0.0040 mT$^{-1}$ for channel $(1, 2)$, $-1727a_0$, 69.043 mT, 12.23 mT, 0.0070 mT$^{-1}$ for channel $(1, 3)$, and $-1903a_0$, 69.043 mT, 12.23 mT, 0.0090 mT$^{-1}$ for channel $(2, 3)$.

![FIG. 3: Bound-bound rf spectrum at 66.144(2) mT. The fractional population loss in state $|2\rangle$ shows a narrow resonance.](image-url)
and 0.0020 mT$^{-1}$ for channel (1, 3), and $-1490 a_0$, 81.122 mT, 22.23 mT, and 0.00395 mT$^{-1}$ for channel (2, 3).

The (1, 3) channel molecular bound state can decay to the (1, 2) channel by a very weak spin-dipolar coupling. We have used the methods of Ref. [29] to calculate the two-body lifetime of the (1, 3) bound state due to pre-dissociation to the (1, 2) channel, and find that it is very long, greater than 10 s at 60.0 mT, increasing to 1000 s at 68.5 mT very close to resonance. However, (1, 3) molecules might be quenched by collisions with [2] atoms or (1, 2) channel molecules, since with three different spin states involved in the collision, there would be no fermionic suppression of collision rates according to the mechanism of Ref. [11].

In conclusion, radio-frequency spectroscopy on ultracold, weakly bound molecules allowed us to precisely determine the molecular binding energies and the energy splittings between two molecular states for different magnetic fields. Based on the measured data and a multi-channel quantum scattering model, we determine the scattering lengths as a function of magnetic field and the Feshbach resonance positions in the lowest three channels with unprecedented precision. With this data, we can fully characterize the interaction strength between particles in the BEC-BCS crossover regime for future experiments based on $^6$Li atoms.

We acknowledge support by the Austrian Science Fund (FWF) within SFB 15 (project part 15) and by the European Union in the framework of the Cold Molecules TMR Network under Contract No. HPRN-CT-2002-00290. P. J. thanks the Office of Naval Research for partial support. C. C. is a Lise-Meitner research fellow of the FWF.

[1] E. Tiesinga, B.J. Verhaar, and H.T.C. Stoof, Phys. Rev. A 47, 4114 (1993); S. Inouye et al., Nature 392, 151 (1998).
[2] S. Jochim et al., Science 302, 2101 (2003); published online 13 November 2003 (10.1126/science.1093280).
[3] M. Greiner, C. A. Regal, D. S. Jin, Nature 426, 537 (2003).
[4] M. Zwierlein et al., Phys. Rev. Lett. 91, 250401 (2003).
[5] T. Bourdel et al., Phys. Rev. Lett. 93, 050401 (2004).
[6] R.G. Hulet, KITP Conference on Quantum Gases, Santa Barbara, CA, 10 - 14 May 2004.
[7] M. Bartenstein et al., Phys. Rev. Lett. 92, 120401 (2004); C. A. Regal, M. Greiner, D. S. Jin, Phys. Rev. Lett. 92, 040403 (2004); M. W. Zwierlein et al., Phys. Rev. Lett. 92, 120403 (2004); J. Kinast et al., Phys. Rev. Lett. 92, 150402 (2004); M. Bartenstein et al., Phys. Rev. Lett. 92, 203201 (2004).
[8] C. Chin et al., Science 305 1128 (2004); published online 22 July 2004 (10.1126/science.1100818).
[9] D. M. Eagles, Phys. Rev. 186, 456 (1969); A. J. Leggett, in Modern Trends in the Theory of Condensed Matter, edited by A. Pekalski and R. Przystawa (Springer-Verlag, Berlin), 1989; P. Nozières, S. Schmitt-Rink, J. Low Temp. Phys. 59, 195 (1985); Q. Chen, J. Stajic, S. Tan and K. Levin, cond-mat/0404274.
[10] M. Houmiers, H.T.C. Stoof, W.I. McAlexander, and R.G. Hulet, Phys. Rev. A 57, R1497 (1998).
[11] D. S. Petrov, C. Salomon, and G. V. Shlyapnikov, cond-mat/0309010.
[12] H. Heiselberg, Phys. Rev. A 63, 043606 (2001); T.-L. Ho, Phys. Rev. Lett. 92, 090402 (2004).
[13] K. Dieckmann et al., Phys. Rev. Lett. 89, 203201 (2002).
[14] K.M. O’Hara et al., Phys. Rev. A 66, 041401(R) (2002).
[15] S. Jochim et al., Phys. Rev. Lett. 89, 273202 (2002).
[16] T. Bourdel et al., Phys. Rev. Lett. 91, 020402 (2003).
[17] C. A. Regal, C. Ticknor, J. L. Bohn, and D. S. Jin, Nature 424 47 (2003).
[18] C. H. Schunck et al., cond-mat/0407377.
[19] S. Gupta et al., Science 300, 1723 (2003); published online May 8, 2003 (10.1126/science.1085335).
[20] J. Cubizolles et al., Phys. Rev. Lett. 91, 240401 (2003); S. Jochim et al., Phys. Rev. Lett. 91, 240402 (2003).
[21] J. Vanier and C. Audoin, The Quantum Physics of Atomic Frequency Standards, (Adam Hilger Ltd, Bristol), 1989.
[22] C. Chin and P. Julienne, cond-mat/0408254.
[23] We assume an exponential decay of the particle number and fit $1 - e^{-\lambda P(E) t}$ to the loss signal, where $\lambda$ depends on the rf power. In principle, we can also determine interaction parameters from $E'$. However, our fits yield large uncertainties for $E'$.
[24] H. T. C. Stoof, J. M. V. A. Koelman and B. J. Verhaar, Phys. Rev. B 38, 4688 (1988).
[25] R. Côté, A. Dalgarno, and M.J. Jamieson, Phys. Rev. A 50, 399 (1994)
[26] Z.-C. Yan, J.F. Babb, A. Dalgarno and G.W.F. Drake, Phys. Rev. A 54, 2824 (1996)
[27] E.R.I. Abraham, W.I. McAlexander, J.M. Gerton, R.G. Hulet, R. Côté and A. Dalgarno, Phys. Rev. A 55, R3299 (1997).
[28] A.J. Moerdijk, B.J. Verhaar, and A. Axelsson Phys. Rev. A 51, 4852-4861 (1995)

[29] T. Köhler, E. Tiesinga, and P. S. Julienne