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## Very-high-precision bound-state spectroscopy near a $^{85}\text{Rb}$ Feshbach resonance

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We precisely measured the binding energy ( $\epsilon_{\text{bind}}$ ) of a molecular state near the Feshbach resonance in a  $^{85}\text{Rb}$  Bose-Einstein condensate (BEC). Rapid magnetic-field pulses induced coherent atom-molecule oscillations in the BEC. We measured the oscillation frequency as a function of  $B$  field and fit the data to a coupled-channel model. Our analysis constrained the Feshbach resonance position [155.041(18) G], width [10.71(2) G], and background scattering length [ $-443(3)a_0$ ] and yielded new values for the Rb interaction parameters. These results improved our estimate for the stability condition of an attractive BEC. We also found evidence for a mean-field shift to  $\epsilon_{\text{bind}}$ .

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The phenomenon of a Feshbach resonance in ultracold collisions of alkali-metal atoms has received much theoretical and experimental interest in recent years, sparking interest in the subjects of resonant Bose-Einstein condensates (BECs) and superfluidity in degenerate Fermi gases. Here we focus on ultracold bosonic gases, in which magnetic-field Feshbach resonances [1,2] have been used both to control elastic and inelastic collisions [3–6] and to tune the self-interaction in BEC [7–11].

In a BEC, the magnetic field controls the self-interaction of the condensate by affecting the  $s$ -wave scattering length  $a$ . Close to resonance, the scattering length varies with  $B$  field according to

$$a = a_{\text{bg}} \left( 1 - \frac{\Delta}{B - B_{\text{peak}}} \right), \quad (1)$$

where  $B_{\text{peak}}$  is the resonance position and is defined to be the magnetic field where the magnitude of  $a$  becomes infinite,  $a_{\text{bg}}$  is the background scattering length,  $\Delta = B_{\text{zero}} - B_{\text{peak}}$  is the resonance width, and  $B_{\text{zero}}$  is the field where the scattering length crosses zero. The resonance is due a (quasi)-bound state that can be tuned in close proximity to the zero-energy threshold via a magnetic field. Measurements of Feshbach resonance positions and widths have been used in a variety of alkali-metal atoms to improve the determination of the interatomic potentials, which in turn can be used for calculating interaction properties relevant for cold atomic gases [12–15].

Recently, we applied rapid magnetic-field variations near a Feshbach resonance to create an atom-molecule superposition state in a  $^{85}\text{Rb}$  BEC [16], which has allowed us to precisely determine the Feshbach resonance position and width. Our technique for studying the Feshbach resonance relies on the presence of atom-molecule coherence [17–19]. By inducing periodic oscillations in the number of condensate atoms, we obtain a direct, high-precision measurement of the molecular bound-state energy  $\epsilon_{\text{bind}}$ . Exploiting the reso-

nance, we tune the molecular state very close to threshold ( $\epsilon_{\text{bind}} \sim 10^{-7} \text{ cm}^{-1}$ )—to our knowledge, this is the most weakly bound diatomic state ever observed in a spectroscopy experiment.

The present method for studying the Feshbach resonance through atom-molecule oscillations offers all of the many inherent advantages of a frequency measurement, including the possibility of high measurement precision, a lack of sensitivity to errors in the absolute atom number calibration, and a simple interpretation of the oscillation frequency in terms of the relative energy difference between the atomic and molecular states. When these advantages are combined with an improved method for magnetic-field calibration [20], the present technique for probing the Feshbach resonance is much more precise than previous experiments that examined such Feshbach resonance observables as variable rethermalization rates in a trapped cloud of atoms [4], enhancements of photoassociation rates [3] and inelastic loss rates [6] near the resonance, and variations of the mean-field expansion energy of a BEC [7].

To complete our precise characterization of the Feshbach resonance, we also made an improved measurement of  $B_{\text{zero}}$ , the magnetic field where the scattering length vanishes. This experiment is very similar to our previous work [14,21], where we determined the  $a=0$  field by measuring the critical number ( $N_{\text{crit}}$ ) for collapse of a BEC, and then we extrapolated to the magnetic field where  $N_{\text{crit}}$  would be infinite. We have improved the measurement precision by about a factor of 4 by improving our magnetic-field calibration and using a larger number of condensate atoms to measure the collapse. We find  $B_{\text{zero}} = 165.750(13)$  G.

The procedure used to generate atom-molecule oscillations in  $^{85}\text{Rb}$  Bose-Einstein condensates has been described in previous work [16], so we merely outline the method here. After creating condensates with initial number of atoms  $N_0 \approx 16\,000$  at a magnetic field  $B \approx 162$  G, we apply two short  $B$ -field pulses ( $\sim 40$ - $\mu\text{s}$  duration) that approach and then recede from the Feshbach resonance at  $B_{\text{peak}} \approx 155$  G. The intermediate value of magnetic field between the pulses,  $B_{\text{evolve}}$ , and the time spacing between pulses,  $t_{\text{evolve}}$ , are variable quantities. The double pulse sequence is followed by a slow change in the  $B$  field to expand the BEC [9], then

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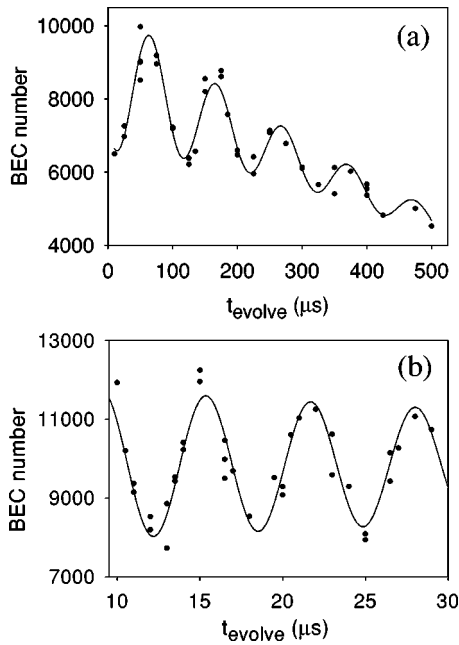


FIG. 1. BEC number versus pulse spacing,  $t_{\text{evolve}}$ . (a)  $B_{\text{evolve}} = 156.840(25)$  G. The oscillation frequency is very low [ $\nu_0 = 9.77(12)$  kHz] so that the damping and atom loss are significant—here  $\beta = 2\pi \times 0.58(12)$  kHz and  $\alpha = 7.9(4)$  atoms/ $\mu\text{s}$ . Nevertheless, the oscillations remain underdamped, with an effective frequency shift due to damping [see Eq. (2)] of only 0.2%, which is far smaller than the 1% statistical frequency error from the fit. (b)  $B_{\text{evolve}} = 159.527(19)$  G. Farther from resonance, the damping of the oscillations and atom loss are negligible in the relatively short time window used to determine  $\nu_0 = 157.8(17)$  kHz.

the trap is switched off ( $B \rightarrow 0$ ) and destructive absorption imaging is used to count the number of atoms remaining in the condensate.

As in Ref. [16], periodic oscillations in the BEC number were observed as a function of  $t_{\text{evolve}}$  (see Fig. 1). We fit the BEC number oscillation to a damped harmonic oscillator function with an additional linear loss term:

$$N(t) = N_{\text{avg}} - \alpha t + A \exp(-\beta t) \sin(\omega_e t + \phi), \quad (2)$$

where  $N_{\text{avg}}$  is the average number,  $A$  is the oscillation amplitude,  $\alpha$  and  $\beta$  are the number loss and damping rates, respectively, and  $\omega_e = 2\pi \sqrt{\nu_0^2 - [\beta/(2\pi)]^2}$ . The quantity of interest here is  $\nu_0$ , the natural oscillator frequency corresponding to the molecular binding energy,  $\nu_0 = \epsilon_{\text{bind}}/h$ . We measured the oscillation frequency for values of  $B_{\text{evolve}}$  from 156.1 G to 161.8 G. Over this range, the frequency varies by over two orders of magnitude (10–1000 kHz), but the linear loss rate of roughly  $-5$  atoms/ $\mu\text{s}$  changes very little [20]. The damping rate shows a significant  $B$ -field dependence, increasing from  $\beta \approx 2\pi \times 0.8$  kHz near 156 G to  $\beta \approx 2\pi \times 22$  kHz near 162 G. This dependence is most likely related to dephasing of the atom-molecule oscillations due to a finite  $B$ -field gradient.

To characterize the Feshbach resonance, it is necessary to know both the oscillation frequency and  $B_{\text{evolve}}$ . We pre-

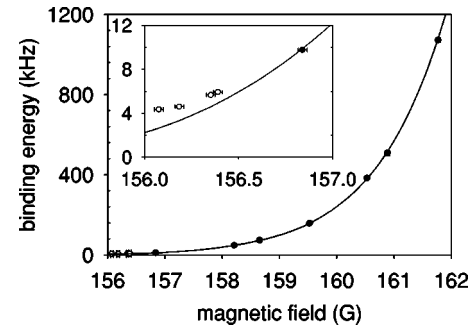


FIG. 2. Molecular binding energy versus magnetic-field  $B_{\text{evolve}}$ . The points are measured values of the atom-molecule oscillation frequency  $\nu_0$ , while the solid line fit is the molecular binding energy from a coupled-channel scattering theory. Only black points were included in the fit; white points were excluded because they experienced a statistically significant mean-field shift. To improve visibility, the points are larger than the error bars. The inset shows the deviation of the lowest frequency data from the fit to the rest of the data.

cisely measured  $B_{\text{evolve}}$  by transferring atoms to an untrapped spin state by driving  $\Delta m = +1$  spin-flip transitions with an applied pulse of rf radiation (pulse length = 5–25  $\mu\text{s}$ ). After measuring the rf transition frequency, we inverted the Breit-Rabi equation to obtain the corresponding  $B$  field. To ensure that the magnetic field was sufficiently constant during  $t_{\text{evolve}}$ , we mapped out  $B(t)$  using rf pulses with lengths short compared to  $t_{\text{evolve}}$ . Due to interference of the rf radiation with the magnetic-field control circuitry, there was a small systematic shift of the field as a function of the rf power used. The total uncertainty for each magnetic-field determination was  $\sim 25$  mG due to the quadrature sum of the uncertainty from the line shape measurements ( $\sim 15$  mG) and the uncertainty in the extrapolation to zero rf power ( $\sim 20$  mG).

The measured oscillation frequencies versus magnetic field are plotted in Fig. 2. We use these data and the zero-crossing field  $B_{\text{zero}}$  to completely characterize the scattering length and binding energy as a function of magnetic field near the Feshbach resonance. As a starting point, we use the coupled-channel analysis of van Kempen *et al.* [15], where several high-precision data for  $^{85}\text{Rb}$  and  $^{87}\text{Rb}$  were combined to perform an interisotope determination of the rubidium interactions with unprecedented accuracy. The predictive power of this analysis can be seen from Ref. [16], where the initial data on the atom-molecule coherence were already in good agreement with the predicted binding energy of the underlying Feshbach state. Another example of the accuracy of the analysis in Ref. [15] is its agreement with more than 40 Feshbach resonances recently discovered in  $^{87}\text{Rb}$  [22].

Van Kempen *et al.* used the best known values [14] for the resonant magnetic field  $B_{\text{peak}}$  and zero crossing  $B_{\text{zero}}$ . In this work, we ignore the relatively imprecise value of  $B_{\text{peak}}$  from Ref. [14], and instead use the measured dependence of binding energy on magnetic field along with the new  $B_{\text{zero}}$  measurement given above to determine the interaction parameters. We observe that the fitting procedure is mainly

TABLE I. Sensitivities of the determined interaction parameters  $v_{DS}$ ,  $v_{DT}$ , and  $C_6$  to fractional uncertainties in  $C_8$ ,  $C_{10}$ ,  $\phi_T^E$ , and  $J$ . For instance, the systematic error in  $C_6$  due to a 10% uncertainty in  $C_8$  is  $123 \times 0.10 = 12.3$  a.u.

	$\Delta C_8/C_8$	$\Delta C_{10}/C_{10}$	$\Delta \phi_T^E/\phi_T^E$	$\Delta J/J$
$\Delta v_{DS}$	$-1.53 \times 10^{-4}$	$-6.80 \times 10^{-5}$	$-2.59 \times 10^{-3}$	$1.72 \times 10^{-3}$
$\Delta v_{DT}$	$-4.14 \times 10^{-4}$	$-1.39 \times 10^{-4}$	$2.31 \times 10^{-3}$	$1.71 \times 10^{-3}$
$\Delta C_6$	123	33.4	-47.8	19.3

sensitive to only three parameters: the van der Waals dispersion coefficient  $C_6$  and the nonintegral vibrational quantum numbers at dissociation,  $v_{DS}$  and  $v_{DT}$ , which determine the position of the last bound state in the singlet and triplet potentials, respectively. Varying the additional parameters  $C_8$ ,  $C_{10}$ ,  $\phi_T^E$  (the first-order energy dependence of the phase of the oscillating triplet radial wave function), and  $J$ , the strength of the exchange interaction, does not improve the fitting because these changes can be absorbed in small shifts of  $v_{DS}$ ,  $v_{DT}$ , and  $C_6$ . Therefore, we take the mean values for these four parameters [23] from the most recent determination in Ref. [22].

The best fit to  $B_{\text{zero}}$  and the seven highest frequency data points yields a reduced  $\chi^2 = 0.30$  for 5 degrees of freedom. This value of  $\chi^2$  is improbably low due to the fact that the uncertainty in the data is dominated by the systematic uncertainty in magnetic field related to the magnitude of the rf power shift. Figure 2 shows the theoretical fit to the binding-energy data as a function of magnetic field. From the fit, we find substantially improved values for the Feshbach resonance position  $B_{\text{peak}} = 155.041(18)$  G, width  $\Delta = 10.71(2)$  G, and background scattering length  $a_{\text{bg}} = -443(3)a_0$ . These results may be compared to previously obtained results  $B_{\text{peak}} = 154.9(4)$  G and  $\Delta = 11.0(4)$  G [14], and  $a_{\text{bg}} = -450(3)a_0$  [24]. Our best interaction parameter values are  $C_6 = 4707(2)$  a.u.,  $v_{DS} = 0.00916(17)$ , and  $v_{DT} = 0.94660(29)$ . Here the error bars do not include systematic errors due to the uncertainties in other interaction parameters that are not constrained by our data. To compare our values with those of Ref. [15], we determined the sensitivity of our three interaction parameters to systematic shifts in the other parameters, as shown in Table I. Using the fractional uncertainties in  $C_8$ ,  $C_{10}$ ,  $\phi_T^E$ , and  $J$  from Ref. [15], we find  $C_6 = 4707(13)$  a.u.,  $v_{DS} = 0.0092(4)$ , and  $v_{DT} = 0.9466(5)$ . All of these values agree with those given in Ref. [15]:  $C_6 = 4703(9)$  a.u.,  $v_{DS} = 0.009(1)$ , and  $v_{DT} = 0.9471(2)$ . Our value for  $v_{DS}$  is more precise than that of Ref. [15], while  $v_{DT}$  and  $C_6$  are slightly less precise. If future experiments allow improvements to the other interaction parameters, then our results will also become more precise since the systematic errors are comparable to or larger than our statistical errors from the fit.

To understand the strong parameter constraints that we obtain with our bound state spectroscopy, one must consider the nonlinear dependence of the binding energy on magnetic field. The magnetic-field dependence of  $\epsilon_{\text{bind}}$  as it approaches the collision threshold depends sensitively on the exact shape of the long-range interatomic potentials, which are mainly

characterized by the van der Waals coefficient  $C_6$ . At magnetic fields far from resonance, the bound-state wave function is confined to a short internuclear distance and  $\epsilon_{\text{bind}}$  varies linearly with magnetic field. The linear dependence on  $B$  field gives relatively little information about  $C_6$ . As the  $B$  field approaches resonance, the detuning decreases until the bound-state lies just below threshold. Now the bound-state wave function penetrates much deeper into the classically forbidden region, which causes  $\epsilon_{\text{bind}}$  to curve toward threshold as a function of magnetic field. Because the energetically forbidden region stretches out as  $C_6/r^6$ , the observed curvature depends sensitively on the  $C_6$  coefficient. One can show [25] that an analytical Feshbach model that includes the correct potential range and background scattering processes [26] can reproduce the binding energy curve over the full range of magnetic field.

The coupled-channel theory used in this work applies to two-body scattering; therefore, this theory cannot account for many-body effects in the atom-molecule BEC system, such as a mean-field shift to the observed oscillation frequency [17,27]. Any such mean-field shift must be fractionally largest near the Feshbach resonance, where the binding energy approaches zero while the atom-atom scattering length increases to infinity. We searched for a mean-field shift to the oscillation frequency when  $B_{\text{evolve}}$  was decreased to  $\sim 156$  G. As shown in Fig. 2, the lowest magnetic-field data display a clear frequency shift with respect to the coupled-channel theory prediction. As  $B_{\text{evolve}}$  approaches resonance, the observed shift increases to 1.7 kHz, which significantly exceeds a simple estimate for the average atom-atom mean-field shift in the BEC:  $4\pi\hbar^2\langle n \rangle a/m \approx 0.5$  kHz at  $B_{\text{evolve}} = 156.1$  G. We are presently investigating new experimental techniques to further study the frequency shifts, including their density dependence.

We use a statistical test to exclude the low-frequency data from the (two-body) theory fit. We first fit the dataset including all frequency measurements with  $\nu_0 \geq 9$  kHz. Eliminating the lowest frequency point from this set causes the reduced  $\chi^2$  to decrease from 0.3 to 0.2, and there is no significant change in parameter values. In contrast, adding the next lower frequency point increases the reduced  $\chi^2$  to 1.9, causing a large systematic shift in the parameter values. The observed behavior seems sensible since we expect mean-field shifts to increase rapidly as one moves toward resonance. The fact that the two-body theory fits the oscillation data over a frequency range spanning two orders of magnitude, but fails when the  $B$  field approaches  $B_{\text{peak}}$ , strongly suggests the influence of many-body physics. We note that a recent theoretical treatment [27] of the atom-molecule oscillations that includes mean-field effects shows excellent agreement with our data over the entire range of magnetic field shown in Fig. 2.

As a result of our improved determination of the  $^{85}\text{Rb}$  Feshbach resonance parameters, we find that our new value for the off-resonant or background scattering length  $a_{\text{bg}} = -443(3)a_0$ , is inconsistent with the value given in Ref. [14], where  $a_{\text{bg}} = -380(21)a_0$ . The most plausible explanation we can find for disagreement is that the theoretical expression used to relate measured rethermalization rate to

cross section is insufficient for the requisite level of accuracy. However, the new value for  $a_{bg}$  allows us to revise our previous estimate for the stability condition of a BEC with negative scattering length [21]. The stability condition may be expressed in terms of a stability coefficient  $k_{collapse} = N_{crit}|a|/a_{ho}$ . Here  $k_{collapse}$  depends on the critical number of atoms,  $N_{crit}$ , the magnitude of the negative scattering length,  $|a|$ , and the harmonic oscillator length of the trapping potential,  $a_{ho}$ . To determine  $k_{collapse}$ , we first use Eq. (1) to obtain the linear slope of scattering length versus  $B$  field near  $B=B_{zero}$ . We then combine the value of  $\Delta a/\Delta B = -39.87(22)a_0/G$  with our previously measured slope of  $1/N_{crit}$  versus magnetic field [21] of  $0.00126(3)$  (atoms G) $^{-1}$ . We obtain the revised value  $k_{collapse} = 0.547(58)$ , where the error is dominated by a 10% systematic uncertainty in the determination of  $N_{crit}$ . The present

$k_{collapse}$  determination agrees with the most recent theoretical value of 0.55 [28].

In conclusion, we present a unique method for exploring a  $^{85}\text{Rb}$  Feshbach resonance. The observed atom-molecule coherence allows us to study the highly nonlinear dependence of the molecular binding energy on magnetic field. We find good agreement with an analysis of van Kempen *et al.* [15] and we also substantially improve the precision of the  $^{85}\text{Rb}$  Feshbach resonance parameters. In addition, we observe mean-field shifts to the molecular binding energy, offering the possibility for future studies of many-body effects in this exciting system.

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