High Resolution Feshbach Spectroscopy of Cesium

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(Received 27 April 2000)

We measure high-resolution Feshbach resonance spectra for ultracold cesium atoms colliding in different hyperfine and magnetic sublevels. More than 25 resonances are observed for magnetic fields up to 230 G and their positions are measured with an accuracy down to 0.03 G. From these spectra several ground-state molecular interaction parameters can be extracted with sufficient accuracy to permit for the first time an unambiguous and accurate determination of cesium's ultracold collision properties [P. J. Leo, C. J. Williams, and P. S. Julienne, following Letter, Phys. Rev. Lett. **85**, 2721 (2000)].

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

Cross sections observed in collisions between ultracold ground-state atoms can exceed the atoms' geometric size by many orders of magnitude when the de Broglie wavelength is larger than the range of interatomic forces [1–4]. In this limit, the collision properties are determined by the asymptotic behavior of the molecular potential, and can be resonantly altered by states near the dissociation threshold.

While resonantly enhanced atom-atom interactions constitute a major source of systematic error for precision experiments using cold atoms, such as atomic clocks [5] or T-violation experiments [6], they also facilitate evaporative cooling to Bose-Einstein condensation (BEC) [7]. In this context the cold-collision properties of cesium, which defines the primary time and frequency standard, have been the subject of a considerable number of theoretical [8–10] and experimental studies. The experimental data include density-dependent frequency shifts in atomic clocks [5], measurements of giant elastic cross sections [1,3,4] and inelastic rate coefficients [2,3] in magnetic traps, the direct detection of s- and p-wave collisions [11], and the observation of Feshbach resonances [4]. However, because the van der Waals coefficient C_6 [12] and the magnitude of the indirect spin-spin coupling [13,14] are not accurately known, these data are insufficient even to determine whether the effective interactions are repulsive or attractive [14]. This has prevented the accurate modeling of density-dependent effects in current and proposed precision experiments [5,6], which is particularly important in view of the large cross sections that have been observed [1–4].

Atomic Feshbach resonances [8], first observed in a sodium Bose condensate [15], establish a unique and direct link between atom-atom scattering and the underlying rovibrational spectrum of the associated diatomic molecule. They occur when a molecular bound state that is supported by a potential belonging to a different internal quantum number is nearly degenerate with the scattering state. In the presence of a coupling between these states, the asymptotic phase shift of the scattering wave function, and therefore the collision cross section [16], will be resonantly affected. If these two states have different magnetic moments, an external magnetic field can be used to adjust the energy splitting between them, giving rise to magnetically tunable atomic collision properties [8]. The observation of Feshbach resonances as a function of magnetic field can be regarded as a spectroscopic technique where the energies of weakly bound molecular states are measured via the known Zeeman energy of the scattering state. Since Feshbach spectroscopy involves only electronic ground states, it has an inherently high resolution that is limited solely by the temperature of the sample.

In this Letter, we report on precision Feshbach spectroscopy of weakly bound Cs_2 states with an energy resolution $\Delta E/h$ below 30 kHz. Collisions of atoms prepared in various hyperfine and magnetic sublevels probe different parts of the molecular potential, permitting for the first time a precise and exhaustive determination of the long-range molecular interaction parameters and hence the cold-collision properties of ^{133}Cs. In the accompanying Letter [17] it is shown that our observed spectra, which contain more than 25 Feshbach resonances, can be fit to the results of coupled-channel calculations to obtain singlet and triplet scattering lengths of $a_S = (280 \pm 10)a_0$ and $a_T = (2400 \pm 100)a_0$, a van der Waals coefficient of $C_6 = (6890 \pm 35)$ a.u., as well as the magnitude of the second order spin-orbit interaction. These scattering lengths differ significantly in both sign and magnitude from those obtained in previous calculations [8,9]. The high resolution of our spectroscopic technique permits the energies of weakly bound states to be determined to a few parts in 10^{10} of the Cs_2 molecular potential depth, and an extension of the analysis should allow one to obtain an even larger set of accurate molecular parameters.

The Feshbach spectra are measured using an experimental setup similar to that described in Ref. [4]. The 3×10^8 atoms are collected in 500 ms in a magneto-optical trap (MOT) from a Cs vapor. A vertically oriented, far-detuned 1D optical lattice trap formed by a retroreflected single-frequency laser beam is superimposed on the MOT. This beam is produced by a Nd:YAG laser operating at 1064 nm, has an intensity of up to 8 W, and has an e^{-2} intensity waist of 250 μm at the position of the atoms, corresponding to a trap depth of 76 μK. Up to 10^8 atoms are loaded into the 1D lattice trap using two 10 ms long periods of 3D degenerate Raman sideband cooling [18]

separated by 20 ms. The atom number is determined from the fluorescence induced by light resonant with the $6S_{1/2}$, $F = 4$ to $6P_{3/2}$, $F = 5$ transition, and a time-of-flight method is used to measure the axial and radial temperatures which have typical values between 1 and 6 μK. Scattering of photons from the trapping beams at a rate of 0.2 s^{-1} leads to negligible heating, and the 2.5 s lifetime of the trapped sample is limited by the Cs background vapor pressure. The axial and radial vibration frequencies with typical values of $f_{ax} = 50$ kHz and $f_{rad} = 80$ Hz are measured by observing a parametric heating of the sample when the trapping laser intensity is modulated at $2f_{ax}$ or $2f_{rad}$. These values are used, along with the measured atom number and temperature, to calculate the density of the trapped atoms which reaches peak values near 10^{13} cm^{-3}.

For state preparation and detection, we use a combination of optical pumping and microwave excitation. Since Raman sideband cooling almost completely spin polarizes the sample into the lowest-energy sublevel $|F = 3, m_F = 3\rangle$ [18], only a short additional optical pumping for 5 ms is necessary to optimally polarize the sample in this state. For this purpose, we use elliptically polarized light on the $6S_{1/2}$, $F = 3 \rightarrow 6P_{3/2}$, $F = 2$ transition that contains no σ^- component along a bias field of 0.5 G oriented at an angle of 30° relative to the beam. A depumping laser on the $6S_{1/2}$, $F = 4 \rightarrow 6P_{3/2}$, $F = 4$ transition is used to empty the upper $F = 4$ ground state. To study resonances in the low field-seeking state $|3, -3\rangle$, we quickly reverse the direction of the bias field after sideband cooling, and then apply the same optical pumping. To prepare the sample in the maximally spin-polarized states $|4, \pm 4\rangle$, we apply optical pumping (after a fast field reversal for $m_F = -4$) with circularly polarized light on the $F = 4 \rightarrow F = 4$ transition in combination with repumping light on the $F = 3 \rightarrow F = 4$ transition. To measure the populations of different magnetic sublevels in the lower (upper) hyperfine manifold, we use energy-selective π microwave transitions $|F, m_F\rangle \rightarrow |F', m_F\rangle$ in a bias field of 120 mG and subsequently detect the transferred population via the fluorescence induced by a laser on the closed $F = 4 \rightarrow F = 5$ ($F = 3 \rightarrow F = 2$) transition. Typically, we prepare 95% of the atoms in the desired state, less than 5% in the neighboring magnetic sublevel, and less than 1% in each of the other states. State selective microwave transitions are also used to prepare mixtures of sublevels. For example, to study collisions between atoms in the states $|3, 3\rangle$ and $|4, 2\rangle$, we transfer a fraction of a sample prepared in $|3, 3\rangle$ to $|4, 2\rangle$ with a microwave transition.

A variable homogeneous magnetic field in the horizontal direction is created by reversing the direction of the current in one of the coils that create the MOT quadrupole field. These coils, whose inner radius is 1.1 cm, have 20 water-cooled windings each and are spaced by 2.2 cm, producing a field of 230 G at a maximum current of 70 A. This field is calibrated to 1 mG by measuring the frequency $\nu = \nu_0(1 + \varepsilon B^2)^{1/2}$ of the $|3, 0\rangle$ to $|4, 0\rangle$ clock transition, where $\nu_0 = 9.192\,631\,77$ GHz and $\varepsilon = 9.299\,905 \times 10^{-8}$ Hz/G^2 [19]. The field variation across the trapped cloud is inferred from the observed width of the microwave lines to be less than 10 mG at the maximum field of 230 G, for samples whose typical vertical and horizontal Gaussian waists are 600 and 36 μm. At the highest fields, the accuracy of the directly measured resonance positions is limited to 10 mG by a small drift of the coil current in the time between the observation of the collisional resonance and the field calibration.

Since the lowest-energy magnetic sublevel $|3, 3\rangle$ is stable against exothermic two-body collisions, Feshbach resonances affect only its elastic cross section σ_{el}, which in the low-temperature limit is related to the s-wave scattering length a_0 by $\sigma_{el} = 8\pi a_0^2$. We detect the corresponding variations sensitively by allowing atoms in the high-energy tail of the thermal distribution to leave a shallow trap. Elastic collisions replenish these states, leading to a continued evaporation loss that is determined by the collision rate. Although this loss is a nonlinear function of the elastic cross section, it can still be used to find minima in σ_{el} with high precision. The corresponding magnetic field values can then be compared with a theoretical analysis [17]. The maxima of σ_{el} do not reveal Feshbach resonance positions with high sensitivity, since when a_0 exceeds the de Broglie wavelength λ_{dB}, the cross section is unitarity limited and independent of a_0 [4,16].

Figure 1 shows the atom number after evaporation for 300 ms in a shallow trap as a function of magnetic field for a sample prepared in the state $|3, 3\rangle$ at a density of approximately 10^{12} cm^{-3}, and at a final temperature of 1.1 μK. Negligible trap loss occurs for a 5 times larger trap depth, showing that the loss is not due to inelastic collisions. The trap depth is adjusted by means of the angle between the linear polarizations of the counterpropagating

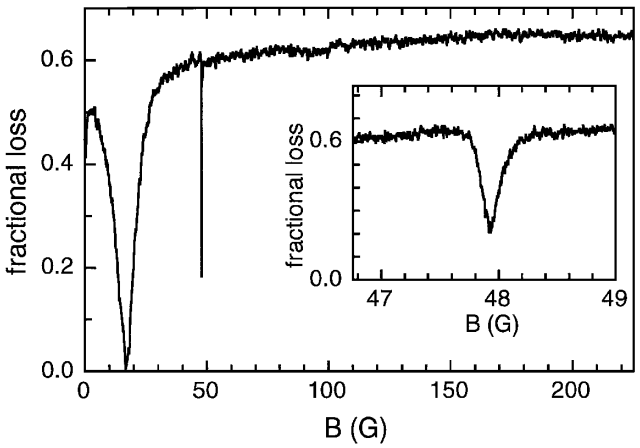

FIG. 1. Evaporative loss for atoms in the $|F = 3, m_F = 3\rangle$ state in 300 ms with a final temperature of 1.1 μK. The evaporative loss is smallest when the elastic cross section has a minimum. The inset shows an expanded view of the resonance near 48 G.

beams that form the 1D lattice, which does not substantially weaken the radial confinement. The broad feature, previously reported in Ref. [4], has a minimum at (17.064 ± 0.030 ± 0.026) G. The first uncertainty is the estimated systematic error (1σ), and the second is the statistical error (1σ) in the position of the extremum from a polynomial fit. In addition, we observe a very narrow resonance with a minimum at (48.017 ± 0.030 ± 0.002) G. The first feature arises from coupling to a molecular state with orbital angular momentum $\ell = 0$, while the narrow Feshbach resonance is due to a molecular state with $\ell = 2$ [17]. The spacing between these two minima is a sensitive measure of the second order spin-orbit coupling [17].

Feshbach resonances involving atomic states other than $|3, 3\rangle$ can be detected via the trap loss induced by inelastic collisions. Figure 2 shows the binary loss coefficient observed in this way for the state $|4, -4\rangle$, while Fig. 2 of the accompanying Letter [17] shows that for $|3, -3\rangle$, both at a temperature of 5.3 μK. These two spectra provide complementary information, since resonances involving solely atoms in the upper hyperfine state $F = 4$ can arise only from states whose binding energy is smaller than the Zeeman energy, while those observed in the lower hyperfine manifold [4,17] result from states bound by the much larger hyperfine energy. For this reason, the two exhibit different dependences on molecular parameters, and the simultaneous fitting of both resonance types allows the van der Waals coefficient C_6 to be extracted with high accuracy [17]. The two resonances in Fig. 2 occur at fields of (105.90 ± 0.03 ± 0.01) G and (205.03 ± 0.03 ± 0.01) G, and another resonance is observed at (206.6 ± 0.1 ± 0.01) G when atoms in the state $|4, -3\rangle$ are added to the sample, and is due to collisions between atoms in $|4, -4\rangle$ and $|4, -3\rangle$. These resonances, in combination with those shown in Fig. 2 of Ref. [17], would allow C_6 to be determined to 2 a.u. (0.03%), if the short range potential were known more accurately. We estimate an uncertainty of a factor of 3 for the overall scale of the loss coefficient, arising mainly from uncertainties in the density calibration and the fact that when the inelastic rate exceeds the thermalization rate, the radial density distributions deviate from the assumed Gaussian shape, since the loss occurs preferentially in the dense central trap regions. We have also verified that the highest-energy state $|4, 4\rangle$ exhibits no resonance structures, and at a temperature of 5 μK has a loss coefficient of 2×10^{-12} cm^3 s^{-1} in agreement with Ref. [2].

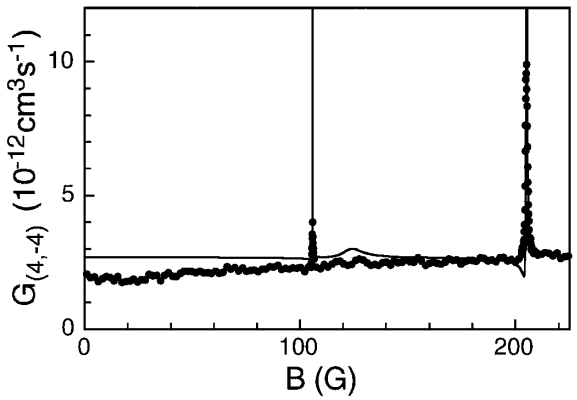

FIG. 2. Binary loss coefficient for atoms in the upper hyperfine state $|F = 4, m_F = -4\rangle$ at a temperature of 5.3 μK with calculation (smooth line) [21]. The two resonances arise from states bound only by the Zeeman energy.

Collisions between atoms in different internal states involve both even and odd partial waves. Figures 3(a) and 3(b) show the fractional loss of atoms from the states $|4, 3\rangle$ and $|4, 2\rangle$ in a time of 400 ms for a sample population of more than 80% in $|3, 3\rangle$ and 15% in $|4, 3\rangle$ or $|4, 2\rangle$. The initial density is 1.5×10^{12} cm^{-3} at a temperature of 5.3 μK. The accompanying analysis [17] shows that the resonances near 175 G for $m_F = 3$ and near 70 and 185 G for $m_F = 2$ are due to p-wave collisions. The two small peaks near 130 and 160 G in Fig. 3(b) which are not reproduced by the calculation are due to a small impurity of atoms in the $|4, 3\rangle$ state. We observe no resonances between 0 and 220 G for collisions between $|3, 3\rangle$ and $|4, 4\rangle$ state atoms.

We have investigated the possibility that our optical trapping potential could shift the observed Feshbach resonance positions. One such effect occurs if the polarizabilities of the bound and free states differ at the frequency of the trapping light. This effect is expected to be small in our far-detuned trap, since the dominant contribution to the light shift of weakly bound states should arise from coupling near the classical outer turning point. Here the excited state energy shift is much smaller than the laser detuning, which results in almost the same light shift for bound and free atoms. Possible effects due to the strong axial confinement [20] are also expected to be small for our temperatures, which typically exceed the axial vibration frequency by a

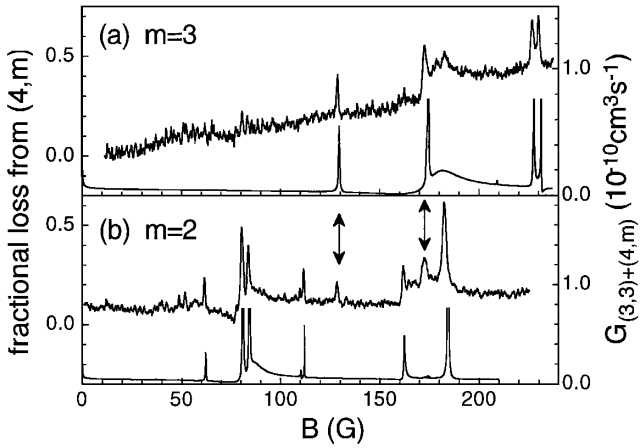

FIG. 3. Fractional loss of atoms from the state $|F = 4, m_F\rangle$ in 400 ms for a mixed sample $|3, 3\rangle + |4, m_F\rangle$, where $m_F = 3$ (a), and $m_F = 2$ (b). The populations of $|3, 3\rangle$ and $|4, m_F\rangle$ are 80% and 15%, respectively. Also shown are calculated event rate coefficients (smooth lines that refer to the right ordinates) for $|3, 3\rangle + |4, m_F\rangle$ s- and p-wave collisions [21].

factor of 3. Indeed, the measured shift of the resonance positions as the trapping light intensity is decreased is less than 30 mG. Within our statistical error of typically 10 mG we find a linear intensity dependence. Since the observed resonance shift may contain light shifts, as well as temperature and dimensionality effects, we include it conservatively in our systematic uncertainty. This error is small compared to the uncertainty of the calculated resonance positions of approximately 500 mG [17].

The full characterization of cesium's cold collision properties now allows us to reexamine possible routes to BEC. At the low bias fields typical of magnetic trapping, neither the $|4, 4\rangle$ nor the $|3, -3\rangle$ state is promising; the former has a prohibitively large dipolar loss coefficient [2], while the latter has a large negative a_0 [17]. However, according to Fig. 3 of Ref. [17], the $|3, -3\rangle$ scattering length is positive in field regions near 70, 110, and 130 G, while the inelastic rate remains relatively small. It should be noted that a_0 is so large in these regions that in typical traps the collision rate will far exceed the trap vibration frequencies. In this hydrodynamic regime the thermalization rate saturates at approximately the vibration frequency, while the inelastic loss rate continues to increase with density [4], such that the ratio $\mathcal{R}$ of thermalization rate to inelastic loss rate decreases as evaporation proceeds. For the above parameters, the hydrodynamic regime cannot be avoided completely without reducing the trap vibration frequencies to below 1 Hz; however, evaporation at large $\mathcal{R}$ in the hydrodynamic regime may still be possible, and requires further study. In this case a large a_0 may even be advantageous since the strong repulsion reduces the density and hence the two-body losses in a BEC. Finally, a promising alternative is the lowest-energy state $|3, 3\rangle$ which experiences no binary loss but must be optically trapped [4]. Its scattering length can be tuned to arbitrary values with a magnetic field near 17 G.

In conclusion, we have observed more than 25 Feshbach resonances for different hyperfine and magnetic sublevels and have measured their positions as a function of magnetic field with high accuracy. From the measured spectra all relevant interaction parameters governing cold cesium collisions can be extracted [17]. This will have important implications for predicting and modeling collision-induced systematic effects in atomic clocks and other precision experiments.

This work was supported in part by grants from the AFOSR and the NSF. V. V. acknowledges support from the Humboldt Foundation.

Note added.—After completion of this manuscript, results on photoassociative spectroscopy of a Cs_2 0_g^- excited electronic state were published [22]. Although ultimately a negative triplet scattering length and a different C_6 are inferred in that work, their observations can be explained with the parameters obtained by Leo, Williams, and Julienne [17].

[1] M. Arndt, M. Ben Dahan, D. Guéry-Odelin, M. W. Reynolds, and J. Dalibard, Phys. Rev. Lett. **79**, 625 (1997).
[2] J. Söding, D. Guéry-Odelin, P. Desbiolles, G. Ferrari, and J. Dalibard, Phys. Rev. Lett. **80**, 1869 (1998).
[3] C. R. Monroe, E. A. Cornell, C. A. Sackett, C. J. Myatt, and C. E. Wieman, Phys. Rev. Lett. **70**, 414 (1993); D. Guéry-Odelin, J. Söding, P. Desbiolles, and J. Dalibard, Opt. Express **2**, 323 (1998); D. Guéry-Odelin, J. Söding, P. Desbiolles, and J. Dalibard, Europhys. Lett. **44**, 25 (1998); S. A. Hopkins, S. Webster, J. Arlt, P. Bance, S. Cornish, O. Maragó, and C. J. Foot, Phys. Rev. A **61**, 032707 (2000).
[4] V. Vuletic, A. J. Kerman, C. Chin, and S. Chu, Phys. Rev. Lett. **82**, 1406 (1999).
[5] K. Gibble and S. Chu, Phys. Rev. Lett. **70**, 1771 (1993).
[6] M. Bijlsma, B. J. Verhaar, and D. J. Heinzen, Phys. Rev. A **49**, R4285 (1994).
[7] M. H. Anderson, J. R. Ensher, M. R. Matthews, C. E. Wieman, and E. A. Cornell, Science **269**, 198 (1995); C. C. Bradley, C. A. Sackett, J. J. Tollett, and R. G. Hulet, Phys. Rev. Lett. **75**, 1687 (1995); K. B. Davis, M.-O. Mewes, M. R. Andrews, N. J. van Druten, D. S. Durfee, D. M. Kurn, and W. Ketterle, Phys. Rev. Lett. **75**, 3969 (1995).
[8] E. Tiesinga, A. J. Moerdijk, B. J. Verhaar, and H. T. C. Stoof, Phys. Rev. A **46**, R1167 (1992); E. Tiesinga, B. J. Verhaar, and H. T. C. Stoof, Phys. Rev. A **47**, 4114 (1993).
[9] S. J. J. M. F. Kokkelmans, B. J. Verhaar, and K. Gibble, Phys. Rev. Lett. **81**, 951 (1998).
[10] P. J. Leo, E. Tiesinga, P. S. Julienne, D. K. Walter, S. Kadlecek, and T. G. Walker, Phys. Rev. Lett. **81**, 1389 (1998).
[11] K. Gibble, S. Chang, and R. Legere, Phys. Rev. Lett. **75**, 2666 (1995); R. Legere and K. Gibble, Phys. Rev. Lett. **81**, 5780 (1998).
[12] A. Derevianko, W. R. Johnson, M. S. Safronova, and J. F. Babb, Phys. Rev. Lett. **82**, 3589 (1999), and references therein.
[13] F. H. Mies, C. J. Williams, P. S. Julienne, and M. Krauss, J. Res. Natl. Inst. Stand. Technol. **101**, 521 (1996).
[14] S. J. J. M. F. Kokkelmans and B. J. Verhaar (private communication); P. Leo (private communication).
[15] S. Inouye, M. R. Andrews, J. Stenger, H.-J. Miesner, D. M. Stamper-Kurn, and W. Ketterle, Nature (London) **392**, 151 (1998).
[16] N. F. Mott and H. S. W. Massey, *The Theory of Atomic Collisions* (Clarendon Press, Oxford, 1965).
[17] P. J. Leo, C. J. Williams, and P. S. Julienne, following Letter, Phys. Rev. Lett. **85**, 2721 (2000).
[18] A. J. Kerman, V. Vuletic, C. Chin, and S. Chu, Phys. Rev. Lett. **84**, 439 (2000).
[19] J. Vanier and C. Audoin, *The Quantum Physics of Atomic Frequency Standards* (Adam Hilger, Bristol and Philadelphia, 1989), Vol. 1.
[20] D. S. Petrov, M. Holzmann, and G. V. Shlyapnikov, Phys. Rev. Lett. **84**, 2551 (2000).
[21] Calculated by P. Leo *et al.,* using the parameters described in Ref. [17].
[22] C. Drag, B. Laburthe Tolra, B. T'Jampens, D. Comparat, M. Allegrini, A. Crubellier, and P. Pillet, Phys. Rev. Lett. **85**, 1408 (2000).