Suppression of Atomic Radiative Collisions by Tuning the Ground State Scattering Length

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We investigate the radiative collision rate in an ultracold, optically trapped cesium gas near a magnetically tunable Feshbach resonance. The radiative trap loss is suppressed by a factor of up to 15 when the node of the ground state pair correlation function is tuned to the Condon point. This constitutes a direct measurement of both the sign and the magnitude of the *s*-wave scattering length. In addition, we observe several narrow, superimposed loss resonances. Their widths are determined by the gas temperature, and their positions are independent of external parameters to within 5 mG.

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Collisions between ultracold atoms, in addition to exhibiting a rich variety of physical phenomena interesting in their own right [1], are also of crucial importance in the fields of Bose-Einstein condensation (BEC) [2], laser cooling and trapping [3], and precision measurements of atomic and molecular properties and fundamental quantities [4]. Ultracold collisional effects can, depending on the situation, be either a fundamental obstacle or an extremely useful tool. Inelastic collisions are a major limitation in the production and manipulation of high-density ultracold samples and condensates [5], and elastic collisions set important limits for many precision experiments [4]. On the other hand, ultracold collisions can serve as an important probe of atomic and molecular parameters [1,6] and quantum statistical properties of degenerate atomic gases [7]. In addition, elastic collisions may be usable as a means of coherent quantum-state control [8], and are required for the production of BECs by evaporative cooling [2].

Collisions in the presence of a light field are especially important since in many cases light is used to probe or manipulate the atomic sample. If one of the atoms is transferred to the excited state during such a collision, the strong dipole-dipole interaction accelerates the atoms to high relative kinetic energies before they return to the ground state via spontaneous emission [9,10]. For a cold trapped atomic gas, these so-called radiative collisions lead to heating and trap loss. Radiative collisions represent an important limitation in all attempts to reach the quantum degenerate regime using optical techniques alone, such as sub-Doppler cooling using dark states [11], Raman sideband cooling in Lamb-Dicke traps [12,13], or polarization gradient cooling in far-detuned optical lattices [14]. At typical condensate densities of 10^{14} cm⁻³, the radiative collision rate will limit the condensate lifetime to values smaller than the single-atom photon scattering time [10].

For a light field with a given detuning from the atomic resonance, excitation occurs at the Condon point, defined as the interatomic distance at which the laser frequency equals the energy spacing between the two-atom ground state and an excited quasimolecular state (Fig. 1). In a very good approximation, the excitation probability amplitude is proportional to the ground state wave function governing the relative motion of the two atoms, evaluated at the Condon point [10]. By monitoring the loss rate as this point is tuned, it is possible to map out the ground state wave function, and even to observe quantum statistical effects [15]. Conversely, the radiative loss is minimized when a node of the two-atom ground state wave function is tuned to the Condon point.

In the low-energy limit, the location of the wave function's first node outside the range of the interatomic potential is given, independent of energy, by the *s*-wave scattering length a_s , if a_s is positive. For $a_s < 0$ no such energy-independent node exists. Therefore, in a cold thermal ensemble the radiative loss will be strongly reduced when a positive scattering length coincides with the Condon point, whereas for $a_s < 0$ no such suppression will be observed (Fig. 1). This effect can be used to determine unambiguously the sign of a_s , and to obtain the magnitude of a positive a_s , using only a simple loss measurement. In contrast, previous studies have either relied on



FIG. 1. Ground state and repulsive excited state potentials for the Cs_2 molecule. The blue detuned light is resonant with the potential difference at the Condon point R_c . Also shown is a ground state wave function in the low-energy limit for positive scattering length a_s . The radiative loss is minimized when the node of the scattering wave function is tuned to the Condon point.

the analysis of photoassociation spectra [1], on the large mean-field energy in a condensate [16], or else on thermalization measurements which are sensitive only to the cross section $\sigma(a_s^2)$ and therefore do not directly reveal the sign of a_s [17,18].

In this Letter, we report a suppression of radiative losses in a nondegenerate, high-density gas by a factor of up to 15 when we adjust the scattering length to a positive value equal to the Condon point. This is accomplished by means of a recently discovered magnetically tunable Feshbach resonance in cesium [18]. The dependence of the radiative collision rate on a_s is described by a simple theoretical model introduced in Ref. [10], that we have extended to include the cesium fine structure and a thermal average of collision energies. In addition, we observe several very narrow resonances with greatly enhanced light-assisted loss in the vicinity of the Feshbach resonance. Their line shapes are asymmetric with widths significantly smaller than the natural linewidth of the atomic transition. Their positions in magnetic field are independent of all external parameters within our experimental uncertainty, including the detuning and polarization of the light that induces the loss and determines the Condon point.

Since the radiative loss comes predominantly from binary events, the rate is proportional to the particle density. A perturbative calculation in the light intensity yields the following result for the binary loss rate coefficient: $K_{\text{loss}} = (8\pi^3 \hbar^3 / \mu k) b_C^2 \Omega_A^2 f_C$ [10]. Here μ is the reduced atomic mass, $\hbar k = (2\mu E)^{1/2}$ is the asymptotic relative momentum, b_C is a unitless structure factor for the coupling to a particular molecular level, and Ω_A is the atomic Rabi frequency. The Franck-Condon factor f_C is proportional to the square of the ground state wave function Ψ_{ρ} at the Condon point R_C , and for blue detuning is given by $f_C = (R_C^4/3C_3) |\Psi_g(R_C, E)|^2$ [10]. For a detuning Δ_A from the atomic resonance that is small compared to the fine-structure splitting, the Condon point R_C is given by $\hbar\Delta_A \approx C_3/R_C^3$, where C_3 is the dipole-dipole coefficient. Since the loss rate is proportional to the probability density of the scattering state evaluated at R_C , the radiative loss induced by a tunable laser constitutes a spatially selective probe of the ground state atomic scattering wave function.

At finite gas temperature *T*, the loss coefficient must be averaged over a thermal distribution of collision energies yielding $K_{\text{loss}} = 8\pi^2 b_C^2 f_3 \lambda_A^3 h_C \Gamma_{\text{atom}}$, with $h_C = [1 - R_B^4/R_C^4][1 - \exp(-2k_{\text{th}}^2\rho_C^2/3)]/k_{\text{th}}^2R_C^2$, which in the lowenergy limit reduces to the expression derived in Ref. [10]. Here $R_B = (\mu C_6/10\hbar^2)^{1/4}$ is the characteristic distance of the van der Waals interaction ($R_B = 49.7$ Å for Cs, with $C_6 = 6331$ a.u. from Ref. [19]), which has been included to lowest order in R_B/R_C [10]; $\rho_C = \rho(R_C)$ with $\rho(R) = R - a_s - 2R_B^4/3R^3$, where $\rho(R) = 0$ defines the position of the node of the ground state wave function. Γ_{atom} is the photon scattering rate for a single atom, and f_3 is the unitless dipole-dipole coefficient defined by $C_3 = f_3 \hbar \gamma_A \chi_A^3$ [10]. Here $\gamma_A = 2\pi \times 5.3$ MHz and $\lambda_A = (852/2\pi)$ nm are the natural atomic decay rate and reduced wavelength for the Cs D_2 line, respectively. For $R_B < a_s < k_{\text{th}}^{-1}$, where $\hbar k_{\text{th}} = (3\mu k_B T)^{1/2}$ is the thermal average of the asymptotic relative momentum, the wave function has a node near a_s independent of collision energy, while for $a_s \leq R_B$ the node position depends only weakly on a_s and is determined primarily by the strength of the van der Waals interaction.

Our atomic sample is a gas of ultracold cesium atoms trapped in a far-detuned 1D optical lattice. The trap is produced by a single-mode Nd:YAG laser operating at 1064 nm, with an output power of up to 17 W. The loading procedure starting from a magneto-optical trap (MOT) is described elsewhere [13]. We trap a total of 10^7 atoms in 4000 pancake-shaped traps with typical axial and radial vibration frequencies of 60 kHz and 60 Hz, respectively. Degenerate Raman sideband cooling [13] is used to lower the temperature and increase the density. The density is calculated from the measured atom number, trap vibration frequencies, and temperature, with typical values of 3×10^{12} cm⁻³ at 4 μ K. The lifetime of the trapped gas is limited by the Cs background pressure to 2 s.

After loading, more than 90% of the atoms are optically pumped in 5 ms to the lowest energy ground state $F = m_F = 3$. Its s-wave scattering length can be changed by applying a magnetic field near the Feshbach resonance [18]. Radiative collisions are induced by an independent, continuously tunable laser (loss laser) near the D_2 line with a power of up to 100 mW, whose intensity is stabilized to 2%. The elliptical beam with e^{-2} beam waists at the trap of 2.5 and 0.6 mm uniformly illuminates the atomic sample, whose vertical and horizontal extensions are 2 mm and 60 μ m, respectively. The detuning of the loss laser is varied between $2\pi \times 10$ GHz and $2\pi \times 1$ THz to the blue of the atomic resonance, and is measured with a resolution of $2\pi \times 1$ GHz using a commercial wave meter. We observe a light-induced change in atom number whose time evolution is well described by a binary process for losses smaller than 30%. Larger losses result in additional heating of the sample since atoms are expelled preferentially from the central region of the trap. Even in this case fits to the time evolution strongly favor a two-body over a three-body process. To measure the loss coefficient as a function of detuning or magnetic field, we record the initial decay rate over a typical time of 100 ms for a total loss near 20%. Figure 2 shows the loss rate versus magnetic field for a detuning of $2\pi \times 32$ GHz, at an atomic peak density of 3×10^{12} cm⁻³ and a temperature of 5 μ K. The broad feature exhibiting suppressed loss is the expected resonance when the scattering length is tuned to the Condon point. Also shown is a fit using the position and width of the Feshbach resonance obtained from our previous thermalization measurements [18], with the overall size of the loss and the residual minimum loss as the only fitting parameters. Here we have included only the two strongest



FIG. 2. Radiative loss rate for a detuning of $2\pi \times 32$ GHz and an average intensity of 26 mW/cm². The gas temperature and peak density are 5.0 μ K and 3×10^{12} cm⁻³, respectively. The broad feature corresponds to the suppression of light-assisted collisions when a_s is positive and close to the Condon point R_c . Superimposed are very narrow resonances with high radiative loss.

couplings to the excited-state molecular levels 2_g and 1_g , with averaged oscillator strengths of $\frac{1}{2}$ and $\frac{5}{12}$ [9], and unitless dipole-dipole coefficients $f_3 = 0.75$ and 0.16, respectively. The absolute measured loss rate agrees with our simple model to within a factor of 2, and we observe a reduction of the loss by up to a factor of 15 relative to the loss near zero magnetic field. The residual loss rate may be due to the fact that different Condon points are associated with other weakly coupled excited-state molecular potentials, or to the residual atomic population in the F = 3, $m_F = 2$ state, which has a different a_s .

To demonstrate that the radiative loss can be used to determine the magnitude of a positive scattering length, we have measured the magnetic field value where minimum loss occurs for different detunings, i.e., for different Condon points. Since the loss is smallest when the node of the wave function occurs at the Condon point, the corresponding scattering length can be calculated from the condition $\rho_C(a_s) = 0$. As the inset of Fig. 2 shows, the scattering lengths thus obtained as a function of magnetic field are in good agreement with our previous measurements of the elastic cross section [18], as indicated by the solid line. This method is applicable only in the region $R_B < a_s < k_{\text{th}}^{-1}$, where the node position nearly coincides with a_s . An extension of this technique would be to map out the entire ground-state pair correlation function for fixed a_s of either sign by measuring the light-induced loss as a function of detuning. A fit to a thermally averaged numerical solution of the Schrödinger equation can then be used to extract both the van der Waals coefficient and the asymptotic phase defined by a_s . We estimate that a determination of a_s to 10% and of C_6 to 1% should be possible with a careful analysis that includes all excited states.

The most striking features in spectra like Fig. 2 are the strong and narrow resonances of increased loss at certain magnetic fields. We observe three strong peaks at 11.07(1), 14.44(1), and 20.00(2) G, and four weaker resonances for a larger detuning than shown in Fig. 2 at 15.17(1), 20.73(4), 23.57(3), and 35.58(5) G. Within our resolution of 5 mG, these values are independent of the detuning and the polarization of the loss laser, the atomic density, and the gas temperature. By extinguishing the trapping laser before the loss laser is turned on, we have also found that the loss is independent of the trapping laser within 50 mG. Finally, we observe that the lost particles cannot be recaptured by the MOT when it is turned on only a few ms after the loss is induced. We conclude that the lost atoms either have an energy E larger than the MOT depth, typically E/h > 10 GHz, or else that they leave the trap in a form that is not recaptured by the MOT, i.e., as molecules. The time evolution of the loss on the peaks favors a three-body over a two-body process.

The width of the peaks depends on the gas temperature. Figure 3 shows an enlarged view of the peak near 14.44 G for temperatures of 3.3 and 8 μ K. The asymmetric shape is well fitted to a Boltzmann distribution $(B - B_0)^{1/2} e^{-(B - B_0)/\Delta B}$. The fitted width ΔB is proportional to the measured temperature in frequency units with coefficients between 1 and 2 mG/kHz that differ for different resonances. These facts suggest that the resonance is associated with a long-lived two- or three-body bound state whose energy is scanned across the thermal distribution of collision energies as the magnetic field is changed. The peak heights depend on the loss laser detuning. For example, the peak near 14.4 G increases slightly with detuning up to $\Delta_A = 2\pi \times 100$ GHz, and falls off beyond that value faster than Δ_A^{-1} . The loss rate increases linearly with intensity for small intensities and saturates at high intensity to a value determined by the density. These data support the conclusion that the loss results from a onephoton transition coupling a bound state to the repulsive excited-state continuum, with a rate proportional to the



FIG. 3. Expanded view of the narrow loss peak near 14.48 G for 3.3 μ K (solid squares) and 8 μ K (open circles) with fitted Boltzmann distributions. The sharp temperature-independent left edge indicates a linewidth of $2\pi \times 5$ kHz.

bound state probability density at the Condon point, and to the single atom photon-scattering rate. The detuning dependence would then reflect the spatial variation of the corresponding molecular wave function.

A possible explanation for the coupling of colliding atoms into these long-lived molecular states is a weak mixing with the bound state responsible for the Feshbach resonance, perhaps provided by the second-order spin-orbit interaction, which is known to be large in cesium [20]. The appearance of several resonances within a field interval of only 10 G indicates very closely spaced molecular levels, possibly rotational states of a weakly bound molecule with large mean interatomic distance. The sharp low-energy edge of the resonances indicates an intrinsic linewidth smaller than $2\pi \times 5$ kHz. The width suggests that the atoms spend more than 30 μ s in these bound states, and thus are likely to be found at close range near the Condon point where they can be excited by the light. This could account for the strongly increased loss which we observe. Extrapolation to a BEC density of 10¹⁴ cm⁻³ yields a radiative loss rate that exceeds the single-atom photon scattering rate by a factor of 3000 for a three-body process or a factor of 100 for a two-body process.

In summary, we have demonstrated that radiative collisions can be suppressed by tuning a zero of the ground state atomic scattering wave function onto the Condon point. This allows a significant reduction of radiative losses when optically manipulating atoms with detuned light in high density samples or condensates. We have shown how the sign and the magnitude of a_s can be directly determined from simple measurements of the radiative loss. In addition, we have observed strong and extremely narrow lightinduced loss resonances as a function of magnetic field. These resonances may arise from coupling to molecular states near the Feshbach resonance. If these molecular states can be identified, the sharp resonances should allow the determination of cesium ground state interaction parameters with unprecedented accuracy.

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