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Laser-induced ultracold Rb($5S_{1/2}$)+Rb($5P_{1/2}$) collisions

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Measurements of collisional loss of optically trapped Rb atoms are presented for excitation of the colliding atoms by light tuned near the $5S_{1/2}\rightarrow 5P_{1/2}$ transitions of ^{85}Rb and ^{87}Rb. Using the $P_{1/2}$ state allows the effects of spontaneous emission on excited-state collisions to be studied with minimal complications arising from hyperfine structure. The shapes of the collision spectra are nearly identical for the two isotopes and are consistent with a simple model for the role of spontaneous emission during the collision.

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The durations of excited-state collisions of optically trapped atoms are comparable to radiative lifetimes. As a result, spontaneous radiative decay during the course of the collisions plays an integral role in the collision dynamics. While various theoretical models treat the effects of spontaneous emission [1,2], the experiments done to date [3–11] are difficult to interpret and thus have not tested the predicted role(s) of radiative decay in any significant way. The ambiguities arise from complications of hyperfine interactions in the alkali-metal $P_{3/2}$ states and substantial theoretical uncertainties in calculating energy-transfer probabilities for collisions that result in the ejection of the atoms from the trap.

We propose that theory can be meaningfully compared to experiments that measure the laser-frequency dependences of trap-loss collision rates ("trap-loss spectra") using light tuned in the vicinity of the $S_{1/2}\rightarrow P_{1/2}$ transitions of the heavy alkali-metal atoms. Frequency-dependent tests require no precise calculations of the energy-transfer probabilities, and the large hyperfine splitting of the $P_{1/2}$ state actually reduces the complications from hyperfine interactions. Additionally, energy transfer occurs only through the radiative escape mechanism since fine-structure-changing collisions are energetically forbidden.

Here we present measurements of $P_{1/2}$ trap-loss spectra for ^{85}Rb and ^{87}Rb that show no isotopic effect, indicating the insensitivity of the collision dynamics to hyperfine interactions. Our experimental results are quantitatively consistent with the Gallagher-Pritchard (GP) model for the role of spontaneous emission [1], modified in a simple way to account for hyperfine interactions. The data support multiple traversals of the potential wells by the colliding atoms.

Experimental progress on isolating the effects of spontaneous emission has been hindered in two important ways. First, absolute measurements of the trap-loss rates [3–7] are uninformative since the models must include calculations of the energy-transfer probabilities. Dulieu *et al.* recently showed that these calculations are extremely sensitive to the poorly known repulsive parts of the potential curves at small interatomic separations [12]. Second, measurements of the trap-loss spectra (the shapes of which should not depend on the energy-transfer rates) near the $S_{1/2}\rightarrow P_{3/2}$ transitions [3,11] show strong isotopic effects; it is therefore necessary for the models to include the effects of hyperfine interactions on the collision dynamics [5,11]. This is a difficult theoretical task, especially since the motion can be nonadiabatic [13]. It is therefore desirable to perform experiments where hyperfine interactions are absent or able to be accounted for in a straightforward manner, as for the experiment described here.

In order to explain how the hyperfine interactions can be accounted for in the current experiment, we review the essential features of the GP model, the simplest model of these collisions [1], depicted in Fig. 1. For this purpose we consider the case of $P_{1/2}$ excitation (for which there is no fine-structure-changing mechanism for trap loss), ignore hyperfine interactions, and assume that the laser is detuned many linewidths from the atomic resonance. The excited-state interaction potential for an atom pair at interatomic separation R is assumed to be $-C_3/R^3$ ($C_3=71$ eV Å^3 for Rb), and, because the radiative broadening is small compared to the detuning, the lasers can be considered to excite only those atom pairs that obey the resonance condition $h\Delta=-C_3/R_0^3$, where $\Delta=\nu_L-\nu_0$ is the detuning of the laser frequency ν_L from the atomic resonance frequency ν_0. Using a quasistatic theory and the resonance condition, atom pairs are assumed to be excited at a rate of

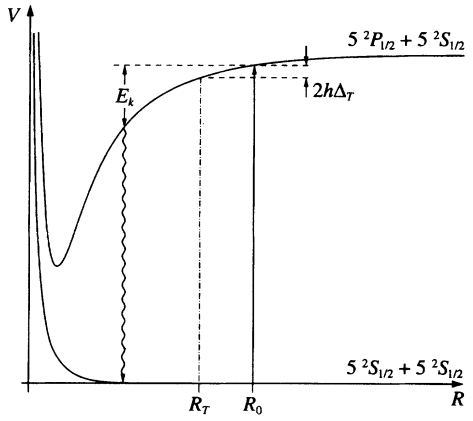

FIG. 1. Gallagher-Pritchard model of trap-loss collisions. The laser of detuning Δ excites atom pairs at interatomic separation R_0. The atoms are accelerated toward each other by the attractive excited-state potential curve. If the atoms radiate while separated by $R<R_T$, where $h\Delta-V(R_T)=2h\Delta_T$ and $h\Delta_T$ is the trap depth, the outgoing atoms have kinetic energy $E_k>h\Delta_T$ and escape the trap.

$$\mathscr{R} \propto R_0^2 |d\Delta/dR_0|^{-1} \propto \Delta^{-2}. \quad (1)$$

Once the atoms are excited at large interatomic separation R_0, they undergo acceleration by the attractive potential curve. The acceleration is rapid enough that neglect of the small initial thermal velocities is permissible. The motion is treated classically. If spontaneous emission occurs before the atoms reach $R=R_T$, where the acquired kinetic energy of the atoms is equal to twice the trap depth, the atoms will still be contained by the trap. However, if the atoms reach R_T without radiating and subsequently radiate in the region $0<R<R_T$, they will leave the trap, since their velocities will exceed the capture velocity of the trap. Defining t_0 as the classical time required to go from $R=R_0$ to $R=R_T$, t_1 as the time to go from $R=R_T$ to $R=0$, and Γ as the spontaneous emission rate, the trap-loss probability for a single orbit from $R=R_0$ to $R=0$ and back is $\exp(-\Gamma t_0) \times[1-\exp(-2\Gamma t_1)]$. By summing this probability over all possible repeated orbits, the total probability of trap loss becomes

$$P=\frac{\sinh(\Gamma t_1)}{\sinh[\Gamma(t_0+t_1)]}. \quad (2)$$

The half-orbit time is $t_0+t_1 \approx 0.746\sqrt{\mu R_0^5/2C_3} \equiv (-\Delta_\tau/\Delta)^{5/6}/\Gamma$, where μ is the reduced mass. We are interested in the case $R_0^3 \gg R_T^3$, implying that $t_0 \gg t_1$ and that t_1 is independent of detuning.

The detuning dependence of the total trap-loss rate is obtained from the product of the excitation rate (1) and the trap-loss probability (2):

$$\beta \propto \{\Delta^2 \sinh[(-\Delta_\tau/\Delta)^{5/6}]\}^{-1}, \quad (3)$$

which has a maximum at $\Delta=-0.36\Delta_\tau$. This prediction for the detuning dependence of β provides a straightforward way to test the validity of the assumptions of the model. Previous experiments using light tuned near the $P_{3/2}$ states of Cs [3] and Rb [4,11] found the maxima of β to correlate strongly with the frequencies of the excited-state hyperfine structure. Interpretation of the data for the $P_{3/2}$ states is difficult because $\Delta_\tau \approx 115$ MHz is very similar in magnitude to the splittings of the excited-state hyperfine levels. Thus, the mixing of the hyperfine states by the dipole-dipole interaction complicates the collision dynamics by making nonadiabatic motions important [13].

It is our goal to test the prediction of Eq. (3) with minimal complications of hyperfine interactions on the collision dynamics. This suggests that studying excited-state collisions of atoms without nuclear spin, such as Ca [14] or perhaps metastable ^{4}He [15] would be ideal. While this viewpoint seems reasonable, it is unnecessarily restrictive since the potential curves, including hyperfine structure, and resulting dynamics will still be simple if the hyperfine splittings are *large* compared to Δ_τ, as is the case for the heavy alkali-metal atoms, Rb and Cs. To illustrate, we show in Fig. 2 the calculated potential curves, including hyperfine interactions, that correlate to the states ^{87}Rb $^2P_{1/2}(F'=1,2)+{}^{87}$Rb $^2S_{1/2}(F=2)$. While the attractive $F'=2$ curves interact with the repulsive curves from $F'=1$, creating considerable complexity, the attractive

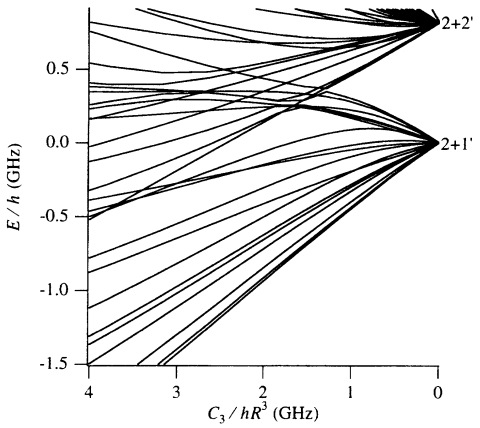

FIG. 2. Calculated potential curves for the relevant $P_{1/2}$ states of ^{87}Rb. The curves shown correlate at large separations to the atomic $S_{1/2}(F=2)+P_{1/2}(F=1',2')$ states. In the region on the low-energy side of the $(2+1')$ state the potential curves closely follow an R^{-3} dependence, facilitating a relatively straightforward comparison with simple models. In between the $(2+1')$ and $(2+2')$ states the curves and resulting hyperfine dynamics are more complicated.

curves from $F'=1$ obey an approximate R^{-3} power law and will be the dominant contribution to trap loss over the range of -1000–0 MHz detunings. There are approximately 15 such curves, but they do not strongly interact, and thus nonadiabatic effects should be unimportant. Even if the atoms make transitions between these curves, since they are all attractive the dynamics would be relatively unaffected.

Based on the above reasoning, we can extend the GP model to take into account the multiple curves by summing individual contributions of the form of Eq. (3), each potential curve having a different value of Δ_τ. From the curves of Fig. 2, we have estimated that the Δ_τ values for the different curves fall in a range of about 50–85 MHz, which means that the trap-loss spectrum should have a maximum at about -25 MHz. The relatively narrow variation in the values of Δ_τ arises from its dependence on $C_3^{2/5}$ and the clustering of the molecular lifetimes in the range of 0.8–1.2 atomic lifetimes. In addition, we find that the potentials for the two isotopes are similar, which should cause their spectra to also be nearly the same. To summarize, for detunings to the red of the ^{87}Rb $^2P_{1/2}(F'=1)$ state [or ^{85}Rb$(F'=2)$], the principal effect of including hyperfine structure is to change the value of Δ_τ from about 115 MHz to about 70 MHz. Thus meaningful comparisons to the models should be possible.

So far we have discussed the hyperfine effects strictly in terms of the GP model. We chose to do this because of its simplicity, ability to make predictions such as Eq. (3), and success in accounting for our observations, as discussed below. We believe that similar simplifications of hyperfine effects will be possible in other more sophisticated approaches [2,16] as well.

Our method for making the measurements of the $P_{1/2}$ trap-loss spectra is the same as described elsewhere [4,11,17]. Light from stabilized diode lasers traps and cools either ^{85}Rb(nuclear spin $I=\frac{5}{2}$) or ^{87}Rb$(I=\frac{3}{2})$ atoms in a vapor-loaded, magneto-optical trap [18] inside a vacuum chamber with partial pressures of Rb and non-Rb atoms of

$\sim 10^{-9}$ and $\sim 10^{-10}$ Torr, respectively. The laser that traps and cools the atoms is locked 7.5 MHz to the red of the $^2S_{1/2}(F=I+\frac{1}{2}) \rightarrow {}^2P_{3/2}(F'=I+\frac{3}{2})$ transition, while a second laser that maintains the atoms in the $F=I+\frac{1}{2}$ ground state is tuned to the $^2S_{1/2}(F=I-\frac{1}{2}) \rightarrow {}^2P_{1/2}(F'=I+\frac{1}{2})$ transition. The density of the trapped atoms is $\sim 2\times 10^{10}$ cm^{-3} at a temperature of ~ 100 μK.

We study excited-state collisions by illuminating the trapped atoms with an independent "catalysis laser" (intensity 1–30 mW/cm^2) that can be tuned in the frequency range -1200 MHz$<\Delta<1000$ MHz from the $^2S_{1/2}(F=I+\frac{1}{2}) \rightarrow {}^2P_{1/2}(F'=I-\frac{1}{2})$ transition of interest for this experiment. The catalysis laser causes additional collisional loss that reduces the number of trapped atoms. In the steady state, the number of trapped atoms is [11] $N=L/[\gamma_t+\beta(\Delta)n_c f]$, where L is the loading rate of atoms into the trap, γ_t is the collisional loss without the catalysis laser (background contribution as well as trapping laser-induced loss), $\beta(\Delta)$ is the collisional rate coefficient at the detuning Δ of the catalysis laser from the $^2S_{1/2}(F=I+\frac{1}{2}) \rightarrow {}^2P_{1/2}(F'=I-\frac{1}{2})$ transition, and f is a factor that accounts for the deviation of the density distribution of the trapped atoms from a uniform density of value n_c. The intensity dependence of the rate coefficient β is found to be linear, a signature of trap-loss collisions involving singly excited states. Since we are interested in finding only the detuning dependence of β, it is unnecessary to measure n_c and f, which have only a weak dependence on N. We therefore hold N (and hence n_c and f) constant by adjusting the intensity of the catalysis laser while scanning Δ. If $I_c(\Delta)$ is the intensity required to keep N fixed, then $\beta(\Delta) \propto 1/I_c(\Delta)$. The collisional reduction of N by the catalysis laser is typically 5–15 %.

Several tests are made to ensure the reliability of this procedure [11,17]. First, in order to guard against atomic optical pumping effects of the catalysis laser, we chop the laser and check for the absence of rapid (<1 msec) changes in the fluorescence. Second, we check that there is no difference in the loading rate L with the catalysis laser blocked or unblocked. Third, we ensure that the catalysis laser provides no force on the cloud, as observed by a video camera. We reject the data if any of the three tests fail, which restricts the detuning to be outside ± 50 MHz of the atomic transition. Finally, we obtain spectra for different values of N to check that their shapes remain the same.

The measured trap-loss spectra are shown for both isotopes in Fig. 3, where an arbitrary scale factor applied to the data enables the comparison of the shapes of the two curves. The shapes are identical within the scatter of the data, an important observation since the $P_{1/2}$ hyperfine splittings are more than twice as large for ^{87}Rb as for ^{85}Rb. We conclude that hyperfine interactions play a subordinate role for the data of Fig. 3. In contrast, the corresponding spectra for the $P_{3/2}$ state show a substantial difference between the two isotopes [11,17].

We also show in Fig. 3 the predicted spectrum, obtained from Eq. (3), using $\Delta_\tau = 70$ MHz. This function accounts for the overall shape of the data over the entire range, from -50 to -1000 MHz. Note that over most of this range Eq. (3) can be approximated by

$$\beta \approx \Delta^{-7/6}, \tag{4}$$

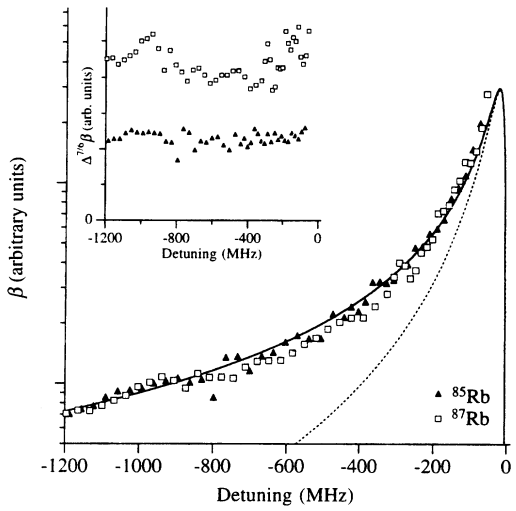

FIG. 3. Measurements of trap-loss spectra for ^{87}Rb and ^{85}Rb for detunings to the red of the lowest atomic $P_{1/2}$ hyperfine states. The lack of isotopic effect in the shapes of the trap-loss spectra indicates the relative unimportance of hyperfine dynamics for these data. The data are scaled to allow comparison of the shapes. Two model calculations are shown, the solid line including effects of multiple orbits of the atoms and the dashed line assuming a single orbit. The inset shows the data presented as $\Delta^{7/6}\beta$, with the scaling changed to allow separate comparison for the two isotopes.

so it is this predicted power law that is principally tested by our data and is not affected by the existence of a number of curves with different values of Δ_τ. For comparison, we also show the predicted Δ^{-2} dependence of a GP model that neglects multiple orbits [8]. For further confirmation, in the inset to Fig. 3 we show experimental values of $\Delta^{7/6}\beta$, which should be a constant according to Eq. (4). We again conclude that the data support the multiple orbit result.

Four comments should be made concerning the interpretation of Fig. 3 as supporting the simple model described above. First, despite the simplicity of the potentials in the detuning region -1000 MHz $<\Delta<0$ MHz, there are curve crossings and avoided crossings that will change the values of t_0 and t_1 at distances where the dipole-dipole interaction is comparable to the ground-state hyperfine splitting. Although this effect is important for t_1, it changes t_0 by only $\sim 10\%$ and therefore changes the absolute scale of the spectrum but not its predicted shape.

Second, one might suppose that, if multiple orbits of the colliding atoms are occurring, discrete resonances should be observed in the data. This reasoning is based on the vibrational splittings becoming larger than $\Gamma/2\pi$, signifying the breakdown of a semiclassical picture of the collision dynamics. However, since about 15 potential curves contribute to the trap loss, resolution of the vibrational structure should not arise until more negative detunings are used.

Third, it is important to address the issue of whether there is a maximum at $|\Delta|<50$ MHz. Distortion of the atom cloud and optical pumping affects the catalysis laser technique, which is unreliable in this range. A possible way of probing the physics of the $|\Delta|<50$ MHz region is to perform two-color $P_{1/2}$ experiments analogous to the $P_{3/2}$ photoassocia-

tive ionization experiment recently reported by Bagnato [10]. In this type of experiment, both lasers can be tuned far from atomic resonances, and the spontaneous emission effects show up in the collision rate as a function of the relative detuning of the two lasers. Since both lasers are off resonance with the isolated atoms, the distortion and optical pumping effects would not be a problem.

Finally, it is important to recognize which features of the model are really tested by our data. In particular, it is surprising that a quasistatic calculation of the excitation rate should be valid because of motion on both the ground- and excited-state potential curves. However, Julienne *et al.* [2] have shown that the same detuning dependence of the excitation rate results from a stationary-phase calculation where the atoms are moving but spontaneous decay is neglected. Thus two very different approaches to calculating the excitation rate give the same result. Based on this we suggest that the trap-loss probability [Eq. (2)] is what is principally being tested by our experiment.

So far we have concentrated on the spectra to the red of the ^{87}Rb($2\rightarrow1'$) and ^{85}Rb($3\rightarrow2'$) states. To the blue of these states, the trap-loss rate is smaller and increases for increasing blue detunings, as expected. However, in this region the distortions of the potential curves that are evident in Fig. 2 decrease the prospects for simple interpretation of the data.

To summarize, calculations of the hyperfine structure of colliding alkali-metal-atom pairs suggest that models of the collision dynamics can be meaningfully compared with experiments performed in a regime where the hyperfine splittings are larger than the characteristic frequency Δ_τ that governs the role of spontaneous emission during the collisions. Use of the lowest-energy hyperfine states reduces distortion of the attractive potential curves due to interactions with curves that originate from other hyperfine states. Accordingly, the $P_{1/2}$ states of ^{85}Rb and ^{87}Rb provide such a situation. This is supported by our measurements that show identically shaped spectra for the two isotopes, despite their different hyperfine splittings. In addition, the shapes of the spectra support the treatment of multiple orbits by the GP model [1].

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