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Comparison of trap-loss collision spectra for ^{85}Rb and ^{87}Rb

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We present measurements of excited-state trap-loss collisions of optically trapped ^{85}Rb and ^{87}Rb atoms as a function of the frequency of the light used to excite the colliding atom pairs. For detunings outside the excited-state hyperfine structure, the trap-loss rates are found to be the same for the two isotopes. For detunings inside the excited-state hyperfine structure, ^{87}Rb collisions occur at a substantially lower rate than those of ^{85}Rb. The spectra make it clear that the long-range dynamics of the collisions are strongly modified by the excited-state hyperfine structure, and that the dynamics are different for the two isotopes.

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Excited-state collisions between optically trapped atoms are of current interest due to their sensitivity to both long-range interatomic forces and spontaneous-emission processes. Experimentally, these collisions result in ejection of the colliding atoms from the trap [1] if the collision is inelastic. Such trap-loss collisions that involve one excited atom interacting with a ground-state atom can be the dominant loss process in a conventional spontaneous-force trap. In this paper we present measurements of the laser-frequency dependence of trap-loss collisions (trap-loss spectrum) of ^{87}Rb atoms and compare to our previous results for ^{85}Rb [2]. The comparison reveals that modification of the collision dynamics by the hyperfine interaction in the excited state is responsible for the previously observed [3] isotopic dependence of the collision rates.

The models currently used to describe trap-loss collisions [4,5] are extensions of the calculations of Gallagher and Pritchard [6] based on a simple picture of the collision dynamics and energy transfer process. In this picture, the trap-loss collisions are a result of absorption of a photon by a pair of ground-state atoms at large interatomic separation R (typically 500 Å) to an attractive excited-state potential curve, followed by acceleration of the atoms on the excited-state potential curves. If the atoms survive to small R (50 Å or so) without spontaneously radiating to the ground state, an energy-transfer process such as a change of fine-structure state or radiative redistribution can impart sufficient kinetic energy to the atoms so that they leave the trap. Thus the trap-loss rates can be considered to result from a product of two factors. The first factor is the rate at which atom pairs reach small R where energy transfer can occur. This rate depends on the dynamics of the collision at large R, including absorption and spontaneous-emission processes. The second factor is the energy-transfer probability once the atoms have reached small R. The physics of the energy-transfer process is not believed to be radically different for low as opposed to high temperatures, since the atom pairs that reach small R have been accelerated to relatively high energy by the excited-state molecular potential. Since the absorption rate and survival probability depend on R at the time of excitation, and hence on the frequency of the light used to cause the collisions, studies of the frequency dependence of the trap-loss collision rate, i.e., the trap-loss spectrum, reveal information about the collision dynamics. The expected shape of the trap-loss spectrum for Rb and Cs is a small rate coefficient at small detunings, rising to a peak near -100 MHz, and falling slowly for larger detunings. The small rate at small detunings arises from the atom pairs being excited at such large distances that the survival probability to small R is low. At large detunings the atom pairs are excited at small distances where the survival probability is high, but the number of atom pairs available to be excited decreases so the trap-loss rate is again small.

Two recent experiments on collisional loss from optically trapped clouds of Rb atoms [2,3] found several features in serious disagreement with the models [4–6] of excited-state collisions of trapped alkali atoms. In contrast, for small detunings and for detunings outside the excited-state hyperfine structure the models agree well with the only previous measurement of trap-loss collisions, for Cs [1,7]. The Rb experiments found a factor of 5 smaller trap-loss rate than expected [4] for Rb at small laser detunings [2,3]. Also, a much narrower peak was observed in the frequency dependence of the trap-loss rate for ^{85}Rb as compared to Cs [2], whereas a similar frequency dependence had been expected. For both ^{85}Rb and Cs the position of the peak was found to be near the frequency of the excited-state hyperfine structure, suggesting that the hyperfine structure is important. Finally, a large difference in the trap-loss rates was observed at about one natural linewidth detuning from the atomic resonance line used for trapping for the two stable Rb isotopes ^{85}Rb and ^{87}Rb [3], with ^{87}Rb being smaller by a factor of 3. Since the only substantial difference between the two isotopes is the hyperfine structure, this again suggests that the hyperfine interaction is very important for the trap-loss collisions.

It is reasonable that hyperfine structure should play an important role in trap-loss collisions, if the atom pairs are excited at separations that require the atoms to travel toward each other through interatomic radii where the

hyperfine interaction strongly affects the molecular potentials. Indeed, for ^{87}Rb and ^{85}Rb the characteristic interatomic radius R_τ for trap-loss collisions is larger than the distance at which the hyperfine interaction dominates. R_τ is the separation at which an atom pair must be excited by the light field in order that the atoms arrive at small R (where energy transfer can occur) in one radiative lifetime as a result of the resonant dipole-dipole interaction $-C_3/R^3$, and is given by

$$R_\tau = \left[\frac{3.59 C_3 \tau^2}{\mu}\right]^{1/5} = 530 \text{ Å} \quad (1)$$

for Rb. Here μ is the reduced mass of the atom pair, $C_3 = 70$ eV Å^3 for Rb, and we have chosen τ, the molecular lifetime, to be equal to the atomic lifetime [4]. A distance scale for which the hyperfine interaction is important can be found by setting the dipole-dipole interaction equal to the energy of the lowest excited-state hyperfine level for each isotope. For ^{87}Rb, the two are equal at 340 Å, while for ^{85}Rb the relevant distance is 450 Å. Clearly, trap-loss collisions may be induced at interatomic separations where effects of excited-state hyperfine interactions on the dynamics are important.

On the basis of the measurements described above, it is still not evident what the shape of the trap-loss spectrum for ^{87}Rb should be. One would expect from the ^{85}Rb-Cs comparison that the width of the spectrum should be larger for ^{87}Rb than for ^{85}Rb since the atomic excited-state hyperfine splittings are larger. An additional question is whether there is a significant isotope effect for detunings outside the excited-state hyperfine structure. This might arise if dynamics associated with the ground-state hyperfine structure were important, for example. If the excited-state hyperfine structure were the dominant effect, however, the trap-loss rates would be expected to be the same for the two isotopes at large detunings, as discussed below. We indeed find, as described in the rest of this paper, that the isotopic distinction at small detunings does not persist at large detunings, thereby isolating dynamics associated with the excited-state hyperfine structure as being primarily responsible for modifying the shapes and relative magnitudes of the spectra for ^{87}Rb and ^{85}Rb.

The experimental apparatus and techniques for obtaining the ^{87}Rb spectra are the same as what we presented briefly elsewhere for ^{85}Rb [2] and will present in full detail in a forthcoming article. Our trap is a standard Zeeman-tuned optical trap that continuously loads atoms directly from residual Rb vapor inside a stainless-steel UHV ($<10^{-9}$ Torr) chamber. Typical trapped-atom numbers and densities are 5×10^6 and 2×10^{10} cm^{-3}. The densities were measured using an absorption technique coupled with spatial distribution measurements from a CCD camera. The number of atoms was measured by detecting the fluorescence from the trapped atoms. The trap depth is sufficient to catch ground-state hyperfine-changing collisions, and so is at least 0.33 K. The three lasers used (two required for trapping, plus a "catalysis" laser whose role is explained below) were external-cavity grating-feedback diode lasers. The trapping laser was spatially filtered and frequency-stabilized to the atomic frequencies using saturation spectroscopy, and the catalysis laser frequency was set by reference to a 300-MHz free-spectral-range spectrum analyzer. The catalysis laser intensity used for the measurements was typically from 2 to 20 mW/cm^2.

The basic method for measuring the collision rate is as follows. The rate equation for the total number of trapped atoms is

$$\frac{dN}{dt} = L - \gamma N - \beta \int n^2(\mathbf{r},t) d^3r \ , \quad (2)$$

where N is the number of trapped atoms, L is the loading rate from the background vapor, γ is the loss rate due to collisions with room-temperature atoms (both Rb and vacuum contaminants), β is the rate coefficient for trap-loss collisions, and $n(\mathbf{r},t)$ is the trapped atom density at position $\mathbf{r}$ and time t. In steady state, the number of atoms is

$$N = \frac{L}{\gamma + \beta n_c f} \ , \quad (3)$$

where $f = \int n^2(\mathbf{r},t) d^3r / n_c N$ is a factor that accounts for the deviation of the density distribution of the cloud from a uniform density of value n_c.

To isolate the effects of excited-state trapped-atom collisions [β in Eq. (3)] we subject the trapped atoms to an additional "catalysis" laser whose frequency and intensity are chosen to cause collisions between the trapped atoms without affecting the operation of the trap. The total trap-loss rate coefficient due to trapped-atom–trapped-atom collisions is then $\beta = \beta_t + \beta_c(\Delta) I_c(\Delta)/I_{\text{ref}}$. Here β_t is the rate coefficient that arises from trap loss caused by the trapping laser, which is detuned 5–10 MHz to the red of the trapping transitions, $5S(F=2)-5P_{3/2}(F'=3)$ for ^{87}Rb and $5S(F=3)-5P_{3/2}(F'=4)$ for ^{85}Rb. The trap-loss coefficient that arises from the catalysis laser of detuning Δ and intensity I_{ref} is $\beta_c(\Delta)$. Detunings are measured from the trapping transition for the atom being studied. For consistency with our previous work we reference our trap-loss coefficients to the intensity $I_{\text{ref}} = 10$ mW/cm^2, assuming the trap-loss coefficient to be linear in intensity [1,2,7]. $I_c(\Delta)$ is the catalysis laser intensity used for measurements at detuning Δ. I_c is in general chosen to be different than I_{ref}, for reasons to be explained next. According to Eq. (3) the additional trap loss that arises from the addition of the catalysis laser will lower the number of trapped atoms. Changing the number of atoms changes the density distribution of the cloud of atoms, so the spatial distribution in principle must be measured for each value of β. Since the spatial distribution is the quantity most difficult to measure in this experiment, it is advantageous instead to determine the relative value of $\beta_c(\Delta)$ by changing the intensity $I_c(\Delta)$ when the detuning is changed in such a way as to hold the number of atoms, and hence the atomic density distribution, constant. Thus, since for each detuning $\beta_c(\Delta) I_c(\Delta) = \text{const}$, the trap-loss rate coefficient obeys the proportionality $\beta_c(\Delta) \propto 1/I_c(\Delta)$. Because everything in the experiment is held constant except the catalysis laser

intensity and detuning, we expect that the shape of the detuning dependence is insensitive to the trapping parameters. Experimentally we find this to be the case.

To determine absolute values of β_c, the trap loss at a single detuning (-550 MHz for ^{87}Rb) was measured by comparing the transient behavior of N governed by Eq. (2), with and without the catalysis laser affecting the trapped atoms. Absolute density measurements were deduced from absorption measurements and from density distribution measurements made with a CCD camera.

The measured values of $\beta_c(\Delta)$ for ^{87}Rb are shown in Fig. 1. These new results are displayed along with our previous measurements [2] for ^{85}Rb in Fig. 2. Note that there are gaps in the ^{87}Rb data (and ^{85}Rb data) in the proximity of the free-atom excited-state hyperfine structure. Data cannot be taken reliably in these detuning ranges because the spatial distribution of the trapped atoms is noticeably affected by the catalysis laser. At detunings close to the trapping transitions, the trap-loss rate coefficient is seen to be substantially smaller for ^{87}Rb than ^{85}Rb. This is consistent with the observations of Wallace *et al.* [3]. Two new features are evident in the detuning dependence. First, the trap-loss rate coefficients for the two isotopes become nearly the same at detunings outside the excited-state hyperfine structure. Second, there is a double-peaked structure around the other excited-state hyperfine transitions in ^{87}Rb (the positions of these transitions are shown in Fig. 1).

The observed near identity of the trap-loss rate coefficients for ^{87}Rb and ^{85}Rb for detunings outside the excited-state hyperfine structure, coupled with the strong distinction between the two isotopes for detunings within the hyperfine structure, shows that the origin of the isotopic difference is in the dynamical effects of the excited-state hyperfine interaction. At small detunings the atoms are excited at large distances, where the hyperfine interaction is larger than the dipole-dipole interaction.

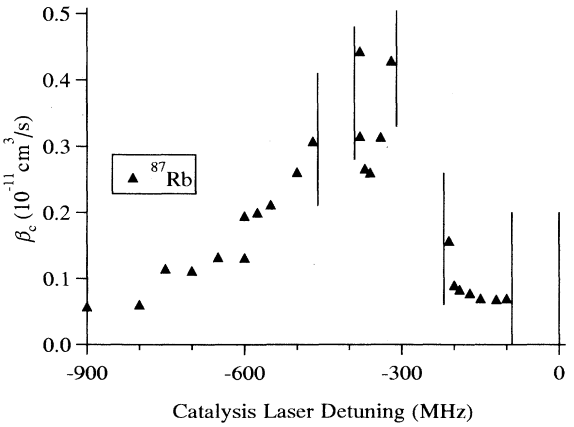

FIG. 1. Trap-loss rate coefficient β_c for ^{87}Rb as a function of detuning of the catalysis laser from resonance. The bars bound the frequency bands around the atomic hyperfine structure in which the catalysis laser produced a perturbing force on the trapped-atom cloud and/or caused a fast change in the atomic fluorescence.

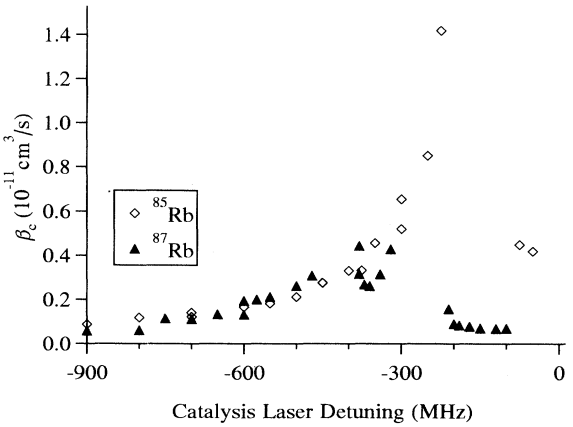

FIG. 2. Trap-loss spectra for ^{87}Rb and ^{85}Rb as a function of detuning from the atomic transition used for trapping (see text). Near zero detuning the trap-loss rate coefficients β_c are much smaller for ^{87}Rb than for ^{85}Rb. Outside the atomic hyperfine structure, however, the coefficients are the same, within error. This shows that the isotopic differences are due to excited-state hyperfine dynamics.

Clearly the absorption spectrum for the colliding atoms will be affected by the hyperfine structure. Also, the potential curves at these distances will include avoided crossings, so Landau-Zener transitions may be important in determining whether the atoms remain on attractive potential curves or not. For large detunings, the atoms are excited at distances where the dipole-dipole interaction has largely decoupled the excited-state hyperfine structure. In this region the absorption spectra and survival probabilities are similar for the two isotopes. As the atoms are accelerated to small R, they pass a second region where the dipole-dipole interaction is comparable to the ground-state hyperfine interaction, but the effects of this region are evidently similar for the two isotopes.

It is interesting to note that for K and Na with smaller hyperfine structure this picture is modified, since at R_τ the hyperfine structure will be largely decoupled. Thus excited-state hyperfine structure is expected to be much less important for these atoms. However, for ^{39}K with a ground-state hyperfine splitting of only 462 MHz and ^{41}K with 254 MHz, dynamics associated with the ground-state hyperfine structure should be important.

The second feature of the data of Figs. 1 and 2 that is new is a double-peaked structure in the ^{87}Rb spectrum. This feature is very reproducible. We believe that collisions are responsible for this feature and have considered and rejected the following other noncollisional mechanisms. One concern is that since the peaks are so near to the atomic hyperfine structure, modification of the atomic fluorescence by catalysis-laser-induced optical pumping might mimic a collisional change in the number of atoms. Evidence against this is that such a change would occur on a short (optical pumping) time scale when the catalysis laser is added or removed, as opposed to a collisional effect that takes many seconds to change the fluorescence. We measure no such rapid change in

the atomic fluorescence for the data of Fig. 2, and thus conclude that optical pumping is not affecting the data. However, when the catalysis laser was tuned near the atomic resonances in the gaps of Fig. 2 where data were rejected, submillisecond changes in the fluorescence signals were measured, rise-time limited by the bandwidth of our fluorescence detection. Under the same conditions rapid changes in the density distribution were usually observed on the video signal from the CCD camera, suggesting that the catalysis laser exerted a small force on the atoms. The second conceivable noncollisional source of the double-peaked structure might be a catalysis-laser-induced change in the loading rate L. This possibility was investigated by deducing L from loading transients. No systematic change in the loading rate was observed. From this evidence we believe that the structure in the ^{87}Rb spectrum arises from collisions.

A collisional source for the double-peaked structure may not, in fact, be too surprising since the characteristic width of the trap-loss spectrum for Rb (without hyperfine structure) should be approximately $\Delta_\tau = C_3/\hbar R_\tau^3 = 115$ MHz, which is smaller than the 157-MHz separation of the ^{87}Rb $F'=2$ and $F'=3$ atomic hyperfine levels. Thus a possible explanation for the observed shape is that the trap-loss spectrum actually consists of two contributions of the shape predicted by the Gallagher-Pritchard model for large detunings. For ^{85}Rb with excited-state hyperfine splittings of 120 MHz or less such structures would be unresolved. If this explanation is correct, an important question is why a similar structure was not observed in the Cs data [7]. The answer may lie in increased scatter in that experiment due to not holding the atomic spatial distribution constant.

In conclusion, we have measured the trap-loss spectrum for ^{87}Rb and not only find reduced collision rates at small detunings compared to ^{85}Rb, confirming the results of Ref. [3], but we also find no isotope effect for large detunings. This identifies excited-state hyperfine structure as the source of the different dynamics for the two isotopes, and illustrates the power of the catalysis laser technique for isolating and studying the dynamics of trapped-atom collisions.

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