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Light-induced ultracold spin-exchange collisions

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We study collisional loss rates due to spin-exchange collisions in very-low-intensity ^{87}Rb magneto-optical traps. The loss rate exhibits an unpredicted intensity dependence for low intensities, accounting for a large disparity between loss rates observed in light-force traps and magnetically confined Bose condensates. Possible collision mechanisms considered include light-altered collision phase shifts, flux enhancement, and excited-state hyperfine changing collisions.

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The development of magneto-optical trapping techniques has provided a unique environment for observing very-low-energy atomic collisions. The ability to reach submillikelvin temperatures means that collisions occur on time scales as long as microseconds and that long-range potentials play an important role in the collision process. Additionally, with the development of Bose-Einstein condensation techniques, a better understanding of these low-energy collisions is important. In particular, recent work has shown that it is possible to dramatically manipulate low-energy scattering processes with judicious application of external fields [1].

Collisional losses from light-force traps such as the magneto-optical trap (MOT) place important constraints on trap operation, in particular limiting the number of trapped atoms. For common vapor cell alkali-metal MOTs, which use light tuned within a few linewidths of an atomic resonance, excited-state collisions dominate trap loss when the trap laser intensity exceeds a few mW/cm^2. Extensive studies of excited-state collisions have produced a fairly deep and quantitative understanding of trap loss for light tuned substantially off resonance, but the near resonant case so important to the MOT remains poorly understood [2,3]. For low-intensity MOTs, studies [4–6] of Cs and Rb reported extremely large loss-rate coefficients (in excess of 10^{-11} cm^3/s) below an abrupt intensity threshold at 1–2 mW/cm^2. The threshold behavior is naturally explained as a turn-on of the ground-state spin-exchange collision channel:

$$(F=2)+(F=2)\rightarrow\begin{cases}(F=1)+(F=1)\\ \text{or}\\ (F=1)+(F=2)\end{cases}\tag{1}$$

for ^{87}Rb. Loss from this collision channel turns on when the trap depth becomes comparable to the ground-state hyperfine splitting. For such a purely ground-state process, the expectation is that once the trap depth becomes smaller than the hyperfine splitting the loss rate should be independent of intensity.

Inelastic spin-exchange collisions also constrain the performance of magnetic traps and the production of Bose-Einstein condensates (BECs) in mixed spin states, especially if the rate coefficients are comparable to those observed in MOTs. Thus one of the biggest recent surprises in the field of ultracold collisions was the discovery by the JILA BEC group of a ^{87}Rb Bose condensate consisting of atoms in two different spin states [7]. The double condensate was formed using evaporative cooling of $|F,m_F\rangle=|1,-1\rangle$ atoms, and sympathetic cooling of $|2,2\rangle$ atoms via elastic collisions with the $|1,-1\rangle$ atoms. The sympathetic cooling worked without disastrous spin-exchange induced trap loss only because the spin-exchange rate coefficient was not the theoretically expected 10^{-11} cm^3/s consistent with MOT data [8], but nearly three orders of magnitude smaller—$2.2(9)\times10^{-14}$ cm^3/s. The surprisingly low spin exchange rate was quickly explained theoretically as a serendipitous matching of the singlet and triplet scattering lengths for ^{87}Rb [9]. Under those conditions the phase shifts associated with the singlet and triplet potentials are nearly identical. The cross section for spin exchange is proportional to $\sin^2(\phi_s-\phi_t)$, and thus the coincidence in scattering lengths results in a destructive interference, suppressing the spin-exchange rate. This spin-exchange suppression appears to be unique to ^{87}Rb.

In this paper we demonstrate that the MOT spin-exchange rates are intensity dependent, indicating that light plays an essential role in spin-exchange collisions. This result again illustrates that low-energy-scattering processes are sensitive to externally applied fields [1].

It is not entirely surprising that the rate measured for the BEC is smaller than the rate measured for the traditional MOT. An obvious difference between the two experiments is the temperature. The temperature of the BEC was below the 500-nK transition temperature, while the MOT temperatures are typically on the order of 100 μK. Using the best potentials available, Williams [10] calculated the expected temperature dependence of spin-exchange collisions. At MOT temperatures, the maximum predicted rate is only on the order of 6×10^{-13} cm^3/s, a factor of 30 lower than the measured MOT rate. Further recent refinements increase this prediction by about a factor of 2 to 5, still substantially below the MOT measurements [12]. Of course, the second principal difference between the MOT and BEC experiments is the presence of the near-resonant trap laser used to produce the MOT. It is the trap laser effects on the spin-exchange rates that we report in this paper.

Our experiments were performed in a low-pressure, vapor-loaded ^{87}Rb MOT. The rubidium pressure was kept in the low 10^{-11} Torr range, so that losses due to collisions with hot rubidium atoms were small compared to the ultra-

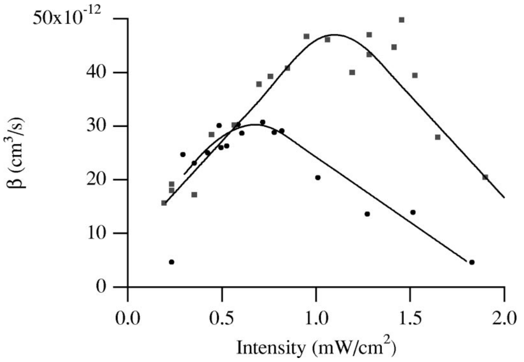

FIG. 1. Loss rates due to ultracold collisions as a function of total trap laser intensity with a detuning of $\Delta = -1\Gamma$. The circles and squares indicate data taken with a magnetic-field gradient of 10 and 18 G/cm, respectively. The solid lines have been included to guide the eye.

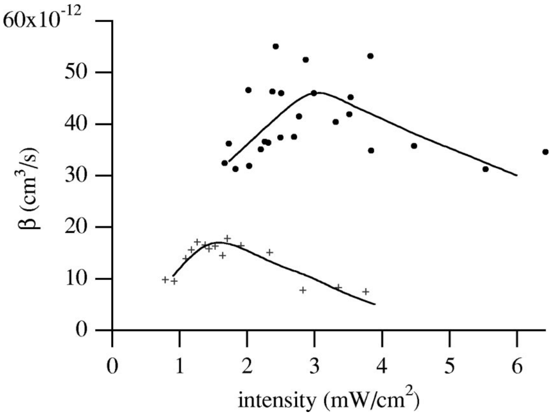

FIG. 2. Loss rates as a function of total trap laser intensity for larger laser detunings. The $+$'s and circles indicate data taken at $\Delta = -1.5\Gamma$ and -2Γ, respectively.

cold collisions. The vacuum chamber was carefully prepared prior to assembly in order to allow for base pressures as low as 10^{-11} Torr, with resulting trap lifetimes as long as 1000 s.

The basic experiment was performed by first loading the trap from the background at high intensity, using both the trap laser and an additional high-power laser. After loading, a shutter blocked the additional laser, and the trap laser intensity was switched, using a liquid-crystal retarder and polarizer to an intensity between 0.25 and 5 mW/cm^2. (The trap laser beams had a $1/e^2$ diameter of 1.0 cm.) The trap fluorescence was recorded as a function of time while the trap was allowed to decay for 10–15 min. A fluorescence image of the trapped atoms was taken with a liquid-nitrogen–cooled charge-coupled-device camera to determine the trap density. The density as a function of time was then fit to the analytic solution of the rate equation for trap loss:

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

where $n(\mathbf{r})$ is the position-dependent atom density, L is the loading rate, Γ is the loss rate due to collisions with hot background gas, and β is the loss rate due to cold collisions.

The measured ultracold collision loss rates as a function of intensity for a laser detuning of one linewidth and two different magnetic field gradients are shown in Fig. 1. Starting from high intensity we find the usual linear dependence on intensity (not shown) for intensities above 4 mW/cm^2, in agreement with previous experiments. As the intensity is decreased further, Fig. 1 shows the previously reported sharp increase in trap loss rate [5]. We then note, however, that at very low intensities the trap loss rate does not level off as expected, but instead decreases roughly linearly with decreasing intensity. As the temperature of the atoms does not vary strongly at the intensities and detunings of our MOT [13], we cannot attribute the observations to a temperature dependence.

The simplest interpretation of Fig. 1 is that the collision mechanism is intensity dependent, releasing a fixed quantity of energy per collision. At low intensity the trap depth is smaller than the energy release, causing the observed intensity dependence of the rate coefficient to reflect the inherent intensity dependence of the collision process. When the trap depth becomes comparable to the energy release, the rate coefficient decreases, as the atoms no longer leave the trap.

Figure 1 shows that the slope of the data at low intensity is the same for both magnetic-field gradients. As the intensity is increased, the data taken with the lower gradient turns over, while the higher gradient data continue to increase linearly. Thus the peak rate coefficient is larger for the higher gradient data. The trap depth is weaker at a given intensity for the higher-magnetic-field gradient that allows atoms to escape the trap at higher intensities, increasing the peak and shifting it to higher intensity. This interpretation is consistent with the predictions of trap depth models [6,11] for low-intensity traps with high field gradients. The intensity dependence for laser detunings of $\Delta = -1.5\Gamma$ and -2Γ is shown in Fig. 2. As in the previous data, there is a decrease in the loss rate at lower intensities. Clearly the collision process responsible for the low-intensity data shown in Figs. 1 and 2 is intensity dependent and releases a fixed amount of energy. A recent experiment has confirmed that spin-exchange rates decrease when near-resonant MOT light is removed [12].

An important figure of merit for these measurements is the quantity $2\beta n_o/\Gamma$ that compares the loss from ultracold collisions to background loss. To reliably distinguish the two loss mechanisms, it is important for this ratio to be larger than 1. In the $\Delta = -1\Gamma$ measurements at 10 G/cm, the trap could be operated at lower intensities than the data indicate; however, the figure of merit became prohibitively small (~ 1) at the lowest intensities, due to a rapid decrease in density. For the larger detuning data, the initial density was quite large. Unfortunately the density became large enough that radiation trapping effects were observed. To eliminate the radiation trapping, many fewer atoms were loaded into the trap. The reduced number of atoms caused a reduction in the trap fluorescence signal, which degraded our overall signal-to-noise ratio. In all cases, however, there is clearly an intensity dependence of the trap loss rates at low intensity. No intensity dependence of the background rate Γ was observed. At the lowest intensities, where the volume of the atom cloud is greatest, the limited depth of field of our cam-

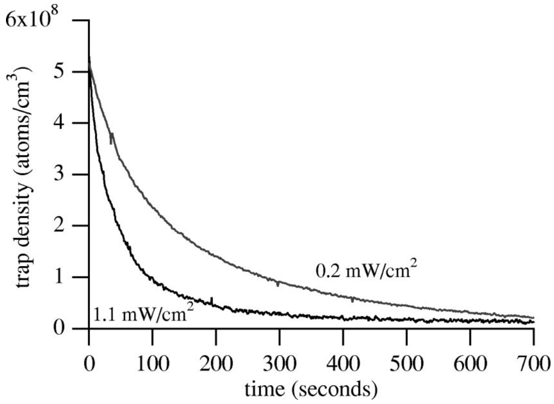

FIG. 3. Loss transients taken at intensities near the peak (1.1 mW/cm^2) and well below the peak (0.2 mW/cm^2). The more gradual decay at 0.2 mW/cm^2 clearly shows that the cold collision loss rate at the same density is greatly reduced as compared to the 1.1-mW/cm^2 rate.

era system produced a tendency to blur our trap images. This effect could produce systematically low measured trap densities, and hence overestimates of β. We took care to minimize this effect, but we note that any additional correction would tend to increase the slope of β at low intensity, further accentuating the strong observed intensity dependence. Loss transients taken near the peak and below the peak at nearly identical trap densities, shown in Fig. 3, show clearly the substantial reduction in the trap-loss rate at very low laser intensity.

The existence of a rapid onset strongly implies that the collision process releases a fixed quantity of energy. This fact makes it almost certain that the collision process is a hyperfine changing or spin-exchange mechanism.

The most straightforward explanation of the data is that the spin-exchange occurs in an excited-state hyperfine-changing process, namely,

$$5S(F=2)+5P_{3/2}\rightarrow 5S(F=1)+5P_{3/2},$$

with the release of 6.8/2 GHz of energy to each of the two outgoing atoms. This process is somewhat analogous to excited-state fine-structure changing collisions that occur at higher trapping intensities [2]. Such a process would begin with excitation of the colliding atoms at long range $\sim$1000 Å to radiatively stable attractive excited-state potentials. The atoms are accelerated to the region around 150 Å where the hyperfine interaction is decoupled by the dipole-dipole interaction. The decoupling would then result in transfer of the atoms to a potential curve that dissociates to the $5S(F=1)+5P_{3/2}$ asymptote.

Assuming excited-state hyperfine changing collisions are responsible for our observations, it is difficult to understand why the rate coefficient is $\sim 10\times$ larger than for the high-intensity excited-state loss mechanism. For the high-intensity loss mechanism, strong arguments [14] favor radiative escape as its source. In this case radiative escape begins with excitation at long range to long-lifetime $S+P_{3/2}$ states—analogous to the radiatively stable 2_u state when hyperfine interactions are ignored—followed by transfer to strongly radiating states in the hyperfine decoupling region. Because the transfer from radiatively stable to unstable states occurs in the hyperfine decoupling region, the collision dynamics of radiative escape and excited-state hyperfine changing collisions are nearly identical. The two loss processes differ only in the states to which the atoms are transferred. In the case of radiative escape the atoms are transferred from a radiatively stable state to an unstable state. On the other hand, excited-state hyperfine changing collisions involve transfer to states correlated with a specific ground-state hyperfine asymptote. If anything, this argument favors a smaller rate coefficient for excited-state hyperfine changing collisions as compared to radiative escape collisions.

Given that the excited-state hyperfine changing process is an unlikely explanation for the data, we turn to ground-state spin-exchange collisions that involve light. The key to the explanations for the low spin-exchange rates for magnetically trapped ^{87}Rb was the near identity of the singlet and triplet scattering phase shifts. Thus a possible explanation for the observed light intensity dependence is that the presence of the near-resonant light upsets the delicate balance between the singlet and triplet phase shifts that suppressed the spin exchange in the double BEC experiment. The presence of the trap laser couples the $1/R^3$ excited potentials to the ground-state $1/R^6$ potential. This coupling may alter the singlet/triplet phase balance in at least two ways. First, the excited-state coupling most certainly occurs at long range but may also be important at small interatomic separations where ground-state spin exchange occurs. We note, for example, that the Movre-Pichler fine-structure potentials [15] for Rb predict that the trap laser can resonantly excite the repulsive $0_g^+\ P_{1/2}$ potential at roughly 11–12 Å, precisely the region in which ground-state spin exchange occurs. Whether this nearly diabatic crossing or coupling to other states can disrupt the phase balance is an open question. The excited-state couplings can also affect spin exchange at long range. Even in the absence of significant ground-state singlet/triplet splitting, we note that at long range different hyperfine levels will couple with different strengths to the various singlet and triplet excited-state potentials. This difference in coupling produces an effective spin-exchange potential at long range that might be sufficient to disrupt the singlet-triplet phase balance.

Another possible explanation is flux enhancement due to the presence of the trap laser, which has been previously observed for excited-state collisions [16]. In this case, a colliding pair of atoms absorbs a photon and is accelerated on the excited-state potential. The atoms therefore return to the ground state via spontaneous emission with increased velocity. The effective collision energy is therefore increased, and in particular additional partial waves not normally present under MOT conditions can then overcome the centrifugal barrier and participate in spin exchange. In particular, we estimate that the *d*-wave contribution to the collision process may be significantly enhanced.

Spin-exchange collisions of $S_{1/2}$ atoms are important in a wide variety of physical phenomena, a partial list being the 21-cm line in radioastronomy [17], masers [18], spin-polarized H and D targets [19], spin-exchange optical pumping [20,21], atomic clocks [22], BECs [23], and spin-polarized hydrogen [24]. The spin-exchange collisions that occur in low-intensity MOTs are unique in that they are sensitive to the energy transfer properties of spin exchange.

In conclusion, we find a pronounced intensity dependence of ultracold spin-exchange collisions in low-intensity magneto-optical traps. It is clear that weak resonant light fields qualitatively and quantitatively change the behavior of low-temperature spin-exchange collisions. As the experimental signatures of the above possible explanations are all similar, further theoretical work is needed to distinguish them.

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