RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 51, NUMBER 2 FEBRUARY 1995

Trap-loss collisions of ultracold lithium atoms

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(Received 15 June 1994)

Accurate measurements are presented of the rate of trap-loss-producing collisions between ultracold magneto-optically trapped ^{7}Li atoms for a range of trap laser intensities and frequencies. Intensities from near the atomic saturation intensity to well above it are investigated. At low intensities, fine-structure-changing collisions cause trap loss with a rate constant of $\sim 10^{-10}$ cm^3/s. At sufficiently high intensity, the trap can be deep enough to effectively freeze out the dominant fine-structure-changing collisions as a loss mechanism, enabling an accurate comparison of the radiative escape loss rate with theory. At the lowest intensities of this radiative escape regime, the measured loss rates compare favorably with those calculated using an optical Bloch equation theory and a three-dimensional model of trap depth. However, the intensity dependence of the measured rates does not show the saturation predicted by the optical Bloch equation theory. It is shown that reliable knowledge of trap depth is necessary to accurately compare experiment with theory.

PACS number(s): 32.80.Pj, 34.50.Rk, 33.80.Ps

The magneto-optical trap (MOT) employs laser beams and an inhomogeneous magnetic field to both confine and cool atoms to the ultracold ($T \leq 1$ mK) regime [1]. In one of the first experiments using a MOT, the atomic density was observed to have a nonexponential decay, which was attributed to exoergic collisions between trapped atoms [2]. For alkali-metal atoms, two collisional loss mechanisms involving excited and ground-state ultracold atoms have been identified [3]. In both processes, two nearby (~ 1000 Å) ground-state atoms absorb a near-resonant photon from the trapping laser beams. The atoms can then accelerate toward each other via the attractive R^{-3} resonant dipole interaction. If the atom pair radiates at a smaller internuclear separation the atoms will have gained kinetic energy. The atom pair will be lost from the trap in a radiative escape (RE) event if the acquired kinetic energy of each atom is greater than the trap depth E_T, where $E_T = \frac{1}{2} m v_e^2$, m is the atomic mass, and v_e is the largest velocity an atom can acquire and remain trapped. The second mechanism, a fine-structure-changing collision (FS), occurs when the colliding atom pair is excited to a molecular potential correlating with the $S_{1/2}+P_{3/2}$ atomic states and exits on a molecular potential correlating with the $S_{1/2}+P_{1/2}$ atomic states. The difference in energy between the fine-structure levels $E_{\rm FS}$ is divided between the atom pair as kinetic energy. If $\frac{1}{2}E_{\rm FS}$ is greater than E_T, the pair is ejected from the trap.

Measurements of ultracold atom collisional loss rates have been done for most alkali-metal atom and some rare-gas atom species [4]. Although extensive theoretical investigations have also been performed [3], unanswered questions, including the role of hyperfine structure and isotopic differences [5–8], remain. Lithium is, in many respects, ideally suited for making accurate comparisons between experiment and theory. In general, collisional loss rates are believed to be extremely sensitive to fine details of the molecular potentials [9]. Theory is facilitated in the case of lithium by the relatively well-known interatomic potentials, which are well described by *ab initio* calculation [10]. In addition, the excited $2P_{3/2}$-state hyperfine structure of lithium is small (comparable to the radiative linewidth) and inverted, unlike for the heavier alkali-metal species, where excited-state hyperfine structure is thought to play an important role in trap-loss collisions [5,6]. Finally, since $E_{\rm FS}$ ($E_{\rm FS}/k_B = 0.48$ K for lithium) is comparable to E_T for a MOT, it is possible to suppress FS as a loss mechanism by ensuring that $E_T > \frac{1}{2}E_{\rm FS}$, as was observed recently in a Li MOT by Kawanaka *et al.* [11]. This ability to separate the contributions of RE and FS greatly simplifies the interpretation of the experiment. In this paper, we present measurements of the RE and FS rates in ^{7}Li for a range of trap laser intensities and detunings. The measured rates are compared with calculations that account for the variation of E_T with laser intensity and detuning.

The experimental apparatus and procedure are similar to other measurements of collisional loss in a MOT [2,4]. The MOT consists of six near-resonant laser beams along the three orthogonal axes that provide for dissipation of the atomic kinetic energy, and, when combined with an inhomogeneous magnetic field, also produce a restoring force [1]. The six beams are produced by a dye laser that is frequency locked relative to a saturated absorption feature of ^{7}Li in an absorption cell to provide long-term frequency stability and a relative frequency reference. The waist ($1/e^2$ intensity radius) of the laser beams is 0.64 cm. A pair of anti-Helmholtz configured coils generates an axial magnetic-field gradient of 3 mT/cm and a radial gradient of 1.5 mT/cm. To ensure a near-spherically-symmetric cloud of trapped atoms, the intensities of the four radial laser beams are equal, while the intensity of an axial beam is $\sim 60\%$ of the intensity of a radial beam. Transitions from both $F=1$ and 2 ground-state hyperfine levels to the excited states are driven with equal intensity using the upper and lower first-order sidebands produced by a standing-wave electro-optic modulator, modulated at 406.4 MHz. The lower first-order sideband is de-

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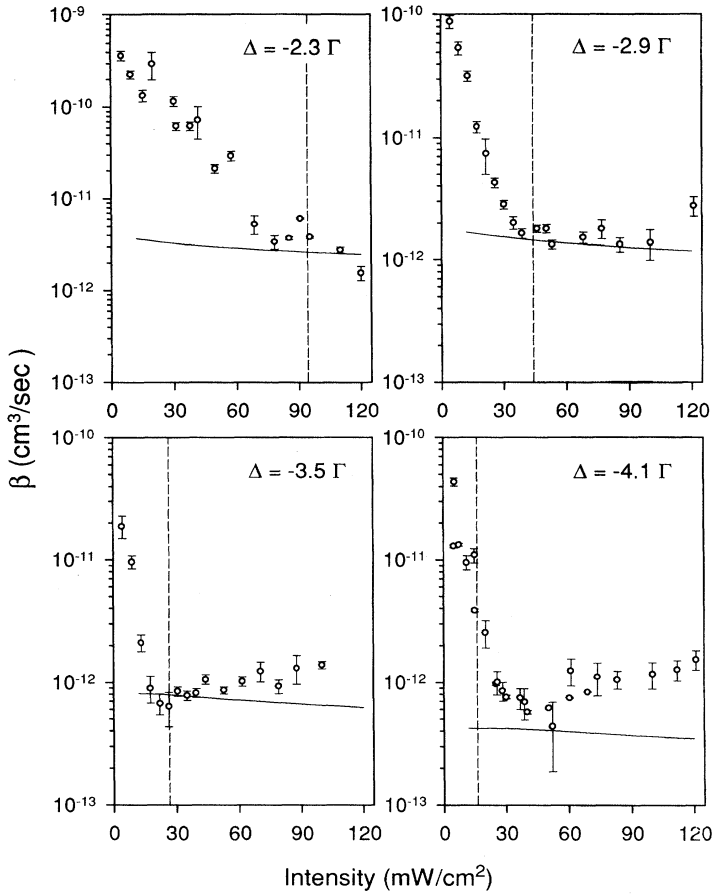

FIG. 1. Two-body trap-loss collision rates for ^{7}Li as a function of total trap laser intensity for trap laser detunings of $\Delta=-2.3\Gamma$, -2.9Γ, -3.5Γ, and -4.1Γ ($\Gamma=5.8$ MHz). The circles are the mean rates obtained from fitting the data to Eq. (1) for ten data runs. The error bars signify the one standard deviation of the mean range. The minimum in the observed loss rate corresponds to the intensity where the trap depth is sufficient to contain the products of fine-structure-changing collisions, and above this intensity only radiative escape can contribute to loss. The dotted vertical line in each plot is the calculated intensity for which this occurs. The solid line is the result of a semiclassical calculation of the radiative escape rate, using the calculated trap depth as input.

tuned relative to the $2S_{1/2},F=2\leftrightarrow 2P_{3/2},F=3$ resonance frequency, while the upper sideband is detuned by the same amount from the $2S_{1/2},F=1\leftrightarrow 2P_{3/2},F=2$ resonance frequency. The MOT is loaded from a laser-slowed atomic beam [12].

A loss rate measurement begins by chopping off the slowing laser beam, then measuring the decrease in the number of trapped atoms as a function of time. Losses from the trap are caused by collisions with hot background gas atoms and collisions with other trapped atoms. Therefore, the rate of change in the number of trapped atoms is given by

$$\frac{dN}{dt}=-\gamma N-\beta\int_{\text{all space}} n^2 dV, \tag{1}$$

where n is the density distribution of trapped atoms, γ is the collisional loss rate of a trapped atom with background gas atoms, and β is the loss rate coefficient for collisions between trapped atoms [2].

The density distribution is determined using a spatially calibrated charge-coupled-device (CCD) camera and imaging optics. The cloud is observed to be Gaussian in shape with a size that increases with the number of trapped atoms, presumably due to the increased optical thickness of the cloud and the reabsorption of fluorescence [13]. The observation that the density distribution remains Gaussian at the highest densities observed is unexpected and is discussed elsewhere [14]. By analyzing images recorded during the trap decay, the Gaussian parameters are found empirically to be

$$w_i(N)=a_i+b_i\frac{N(t)}{N_0}, \tag{2}$$

where i refers either to the axial (A) or to the radial (R) dimension, and N_0 is the initial number of atoms. The parameters a_i and b_i are measured for each combination of trap laser intensity I and detuning Δ. With this assumption, the integral in Eq. (1) simplifies to $N^2/[(2\pi)^{3/2}w_A w_R^2]$. $N(t)$ is determined by combining the measurement of the trap-laser-induced fluorescence using a calibrated silicon photodiode with a calculation of the steady-state fraction of excited-state atoms. The calculation includes all 24 relevant ground- and excited-state sublevels [14]. The excited-state fraction and density found this way agree with a direct absorption measurement using a weak probe laser beam. The ratio of the ground state $F=2$ to $F=1$ population is found by both methods to be approximately 3 to 1. For the experimental results presented here, the maximum density n_0 ranges between 2×10^9 cm^{-3} and 5×10^{10} cm^{-3}, and w_R and w_A are initially between 100 and 900 μm, depending on Δ and I. For most of the Δ,I combinations, $w_R\approx w_A$ to within 20% for all N. In most cases $b_i/a_i<0.5$, and in all cases $b_i/a_i<0.8$.

Figure 1 shows data for four values of Δ and a range of

intensities I. I is the total intensity of the six beams in each of the two frequencies. The circles represent the mean value of β obtained by fitting the data to Eq. (1) for ten different data runs, for each I,Δ pair. The error bars signify the one standard deviation of the mean range of the ten data runs. The estimated systematic uncertainties in β are $\sim 50\%$, due mostly to the uncertainty in the dimensions of the cloud. The estimated absolute uncertainty in Δ is ± 2 MHz, while the relative uncertainty is ± 1 MHz. The measured trap decay times γ^{-1} depend on the trap parameters and conditions, and range from 5 to 20 sec, although the majority of the data fall into the range of 10–15 sec.

For each of the four detunings, the measured loss rate is largest at small intensities and initially decreases with increasing I until reaching a distinct minimum, which occurs at a different value of I for each detuning. This loss rate minimum is attributed to the suppression of FS as a loss mechanism, which occurs when $E_T > \frac{1}{2}E_{\mathrm{FS}}$, caused by the increase of E_T with I. This effect is similar to the earlier observation of Kawanaka *et al.* where suppression of FS loss was caused by the increase of E_T with duty cycle of the trapping laser beams [11]. At higher values of I in our experiment, the loss is due entirely to RE. In this RE regime, the loss rate is significantly smaller than at lower intensities, where FS dominates.

Also shown in Fig. 1 as solid lines are the results of a calculation of the RE trap loss rates of Li [15]. The theory uses the multitrajectory, semiclassical optical Bloch equation (OBE) method of Ref. [16]. The individual contributions to RE trap loss from the Li_2 0_u^+ and 1_g molecular states are summed, using the excited-state adiabatic potentials and decay rates found by diagonalizing a molecular Hamiltonian that includes the fine-structure and resonant dipole-dipole interactions, but neglects the molecular hyperfine interaction. The rate coefficients were calculated as a function of I, Δ, T, and E_T. These quantitative calculations, as well as a qualitative discussion of possible modifications due to the hyperfine structure of the Li_2 molecule, are described in detail elsewhere [15]. The OBE method, which accounts for saturation of the optical transition, was used in order to treat the higher intensities of this experiment [15]. The calculated loss rates in the experimental range of E_T scale as $E_T^{-3.0}$ for fixed I, Δ, and T, which is close to the predicted $E_T^{-17/6}$ scaling for $E_T \gg E_{\mathrm{FS}}$ [17]. Because of the existence of this strong scaling with E_T, a comparison of theory and experiment *must* account for the experimental variation of E_T with I and Δ. The trap loss rate is calculated to be relatively insensitive to temperature in the range 0.3–3 mK, the rate at 0.3 mK being approximately 60% of the rate at 3 mK. The results shown in Fig. 1 assume that the temperature of the trapped atoms is 1 mK, based upon the result of a single measurement using the time-of-flight technique [18].

Since the theory predicts that the loss rate depends sensitively on E_T, a reliable estimate of E_T is essential. A model was developed to calculate the three-dimensional trajectory of an atom in a MOT for this purpose [4]. The force on the atom is calculated from a system of coupled rate equations solved in steady state for all relevant ground- and excited-state hyperfine levels. The atom is assumed to start at the center of the trap with an initial speed and direction (θ,ϕ). The trajectory is calculated to find whether the atom remains trapped. The trap depth is defined by $E_T(\theta,\phi) = \frac{1}{2}m[v_e(\theta,\phi)]^2$, where $v_e(\theta,\phi)$ is the maximum initial velocity that remains trapped. $E_T(\theta,\phi)$ is highly anisotropic, varying by as much as a factor of 4 between the shallowest and deepest directions for our experimental parameters. The calculated loss rates shown in Fig. 1 correspond to an average over all directions, $\beta = \beta_0 \int [E_T(\theta,\phi)/E_0]^{-3} d\Omega/4\pi$, where β_0 is calculated for a symmetric trap of depth E_0. We find, for E_0 equal to the trap depth in the shallowest direction, that $\beta/\beta_0 = 0.23 \pm 0.02$ and is relatively independent of I and Δ, in the ranges considered.

The calculated intensity I_c required to recapture an atom released in the shallowest direction with a kinetic energy $\frac{1}{2}E_{\mathrm{FS}}$ is shown in Fig. 1 as a dashed vertical line for each Δ. For $I > I_c$, only RE collisions should contribute to trap loss. The predicted value of I_c is in reasonably good agreement with the measurements for the three smallest detunings, while for $\Delta = -4.1\Gamma$ ($\Gamma = 5.8$ MHz, the radiative linewidth), the predicted I_c is somewhat smaller than the observed value. The actual disagreement in E_T is small, however, since E_T is a weak function of I for these parameters; E_T is calculated to be only 30% larger at $I = 45$ mW/cm^2 than at 17 mW/cm^2. We attribute the discrepancy in the predicted value of I_c to the breakdown of the validity of the approximation that all collisions occur at the center of the trap, since at larger detunings the trap spring constant is weaker [19,20], causing the trap cloud dimensions to increase. This deficiency can be eliminated by modifying the calculation to allow for a distribution of collision locations.

In the present experiment, β is measured to be relatively insensitive to I for $I > I_c$, while the calculated RE rates decrease with I. There are several factors determining the intensity dependence of the calculated rates. First, the excitation rate is predicted by the OBE theory to saturate at the highest intensities, thereby limiting the increase in RE with I. A recent experiment with Na in the high-intensity regime is unable to confirm or to rule out this prediction [21]. Second, E_T increases with I, significantly depressing the RE loss rate. For example, for $\Delta = -3.5\Gamma$, E_T/k_B in the shallowest direction increases from 0.24 K at $I = 22$ mW/cm^2 to 0.38 K at 120 mW/cm^2, so that the E_T^{-3} factor reduces the RE rate by a factor of 4.

The discrepancies between measured and calculated RE loss rates may be an indication of errors in the E_T calculation, or the neglect of effects of multichannel scattering due to hyperfine structure, or an incorrect theoretical treatment of the optical excitation process occurring during the collision. Although it is clear that the present implementation of the E_T calculation overestimates E_T somewhat for $\Delta = -4.1\Gamma$, depressing the theoretically calculated rates for this detuning, this effect is not large enough to account for the differing I dependence. Undoubtedly, hyperfine structure has a role to play and could affect the trends due to saturation and detuning [15]. Several groups have recently pointed out that the OBE method incorrectly describes the excitation process in the case of heavier atoms at very low temperatures [22–25]. The origin of this disagreement is still an open question. Interestingly, the differences in intensity dependence between theory and experiment shown in Fig. 1 are significantly reduced if the optical excitation rate is assumed to

increase linearly with I, rather than to saturate. Finally, although it has been shown that semiclassical methods are a good approximation for Li collisions at 1 mK, the detuning dependence can be affected by quantized bound-state motion that the semiclassical method does not include [23]. Any quantitative assessment of these various sources of error in the calculation awaits additional development of the theory. It is gratifying that a relatively simple theory obtains the basic magnitude of the low-intensity loss rate.

In the future, we will report a comparison between ^{6}Li and ^{7}Li, which should help to clarify the role of hyperfine effects and mass differences in the rate of collisional trap loss. Clearly, more theoretical and experimental work is needed to fully understand trap-loss collisions.

This work was supported by the National Science Foundation, the Texas Advanced Technology Program, and the Robert A. Welch Foundation. We gratefully acknowledge the valuable contributions to the imaging system made by Brian P. Anderson.

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