Optical Catalysis in a Sodium Vapor Cell MOT

L.G. Marcassa, S.R. Muniz and J. Flemming

Departamento de Física e Ciência dos Materiais, Instituto de Física de São Carlos - USP

Cx. Postal 369 - São Carlos - 13560-970 - SP - Brazil

Received January 1, 1997

The frequency dependence of the collisional trap loss rate for ultracold sodium atoms held in a magneto-optical trap was measured using the optical catalysis technique. The experiment was carried out near the $3S_{1/2} \Longrightarrow 3P_{3/2}$ and $3S_{1/2} \Longrightarrow 3P_{1/2}$ transitions. The experimental spectra are compared with a simple semiclassical model, and a good qualitative agreement is observed. The results also indicate that radiative escape is an important process for trap loss in sodium.

Introduction

Over the past decade advances in laser trapping and cooling have made it possible to produce atomic samples that are cooled to temperatures of 100 μK and below. In these samples a series of experiments involving ultracold collisions have been done over the past five years[1-4]. These collisions are of interest because of the long collision time for trapped atoms, that result from the combination of low velocities and sensibility to long range interactions. In such conditions the spontaneous radiative decay dominates, the system must be considered "open" and dissipatively coupled to the bath of empty modes of radiation field.

In an earlier work our group studied the photoassociative ionization process[5], but here we will turn our attention to exoergic processes leading to escape from the trap. These processes are radiative escape (RE) and fine structure change (FS). Such processes limit the density and confinement time of a 3-D optical trap and the ultimate "brightness" of a 2-D confined atomic beam. The radiative escape process begins when two ultracold ground-state atoms collide in the presence of a radiation field. This system absorbs a photon (ν_1) and is promoted at long range to an attractive $-C_3/R^3$ potential, which is asymptotically the $S+P_{3/2}$ level. In this level the atoms are accelerated against each other. During this acceleration the atomic pair may decay and emit a photon to the red of the atomic transition ($\nu_2, \nu_2 < \nu_1$), and the difference in energy goes to kinetic energy. If this kinetic energy is larger than the depth of the trap ($\sim$ 1K) the pair will escape and this becomes a loss mechanism. The fine structure change starts very similarly to the radiative escape, but spontaneous decay does not happen during the acceleration step. So the pair reaches short internuclear separation and there may change to the $S+P_{1/2}$ level. Both processes are shown in Fig. 1. The difference in energy goes again to kinetic energy. For all alkali the difference in energy between the $P_{3/2}$ and $P_{1/2}$ states is much bigger than the trap depth, with exception of Li, so such process should be important for loss also. The contribution of each process to the total loss is not clear until now, and there are several open questions in theory and experiment[6]. The understanding of these ultracold collisions is crucial to the design of cold dense atomic sources.

Earlier investigations[7-9] explored the importance of collisional loss to ultimate trap density, the absolute rate constants for collisional loss as functions of light intensity, and manipulation of trap loss rate with an "optical catalysis laser". In these optical catalysis experiments it was possible to obtain the frequency dependence of the collisional processes. However these experiments were carried out with rubidium[9] and cesium[10], both alkali have several hundred MHz hyperfine excited state splitting, which difficultates very much the comparison with theory.

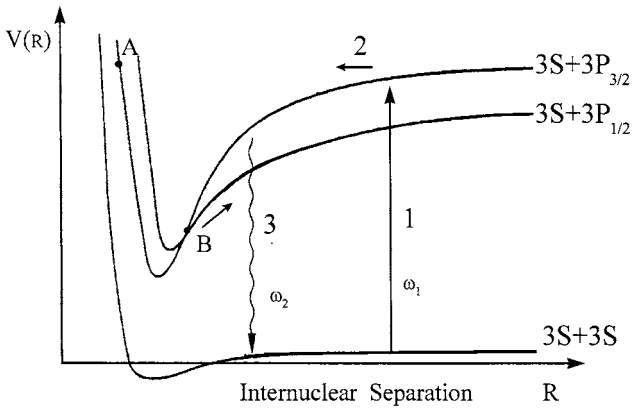

Figure 1. Schematic diagram of the trap loss processes: (i) Radiative escape. The atomic pair starts the collision in the $3S_{1/2}$+$3S_{1/2}$ potential, by photon absorption (1) it is excited to $3S_{1/2}$+$3P_{3/2}$. The atoms accelerate against each other (2), during this process spontaneous decay may happen and a redder photon is emitted (3). Depending on the frequency of the photon the pair may escape from the trap. (ii) Fine structure change. The atomic pair starts the collision in the $3S_{1/2}$+$3S_{1/2}$ potential, by photon absorption (1) it is excited to $3S_{1/2}$+$3P_{3/2}$. The atoms accelerate against each other (2). Point A is the return, and B is the point where the pair may have a transition to the $3S_{1/2}$+$3P_{1/2}$ potential. The difference in energy goes to kinetic energy.

Here we report a study of optical "catalysis" in a magneto optical trap for sodium atoms performed close to the D1 and D2 lines. The frequency dependence is presented and also the absolute loss rates introduced by the "catalysis" laser. From these rates we can estimate, for the first time, the contribution of each process for the total loss, and compare the results with a simple Gallagher-Pritchard model[11].

Optical catalysis technique

The rate equation that governs the number of atoms in the trap N in the regime of radiation trapping[10], is given by:

$$\frac{dN}{dt} = L - (\gamma + \beta n_c)N \qquad (1)$$

where L is the loading rate which depends on the trap parameters (Na vapor pressure, diameter of laser beams, magnetic field gradient, laser frequency, etc); γN is the rate of collisions between trapped atoms and hot atoms from the background gas; and $\beta n_c N$ is the rate of cold collisions. We should point out that the experiment was carried out in the regime of radiation trapping, and the time behavior of our trap can be fitted by this simple exponential law.

The linear dependence of excited state collisions with the laser intensity is a unique signature of this kind of collision, because it involves only one atom in the excited state. Therefore the addition of the catalysis laser introduces more losses in the trap due to excited state collisions. In this condition we can rewrite the rate coefficient as $\beta = \beta_t + \beta_c$, where β_t represents the losses only with the trapping beams present and β_c is the contribution from the catalysis laser. The trap loss spectrum (frequency dependence) can be obtained from the steady state behavior of equation (1). In the steady state equation (1) becomes:

$$N = \frac{L}{\gamma + \beta n_c} \qquad (2)$$

The catalysis laser, with an intensity $I_{\rm ref}$ and detuned by Δ_c, affects both β and the atomic density (n_c). As we vary Δ_c we have to adjust its intensity $I_c(\Delta_c)$ to hold the number of atoms constant, therefore the density will remain constant also. If we consider that β_c presents a linear intensity dependence we can rewrite

$$\beta_c = \beta_c(\Delta_c)\frac{I_c(\Delta_c)}{I_{\rm ref}} \qquad (3)$$

which is independent of Δ_c, because $\beta_c \propto 1/I_c(\Delta_c)$. The advantage in this technique is that it is not necessary to measure number of atoms and density, which simplifies very much the experiment, once the main source of error are the measurements of these parameters. Corrections to account for the fact that the density is not uniform, for small number of atoms, were made by Hoffmann and co-workers[12]. However these corrections are minor and we will not consider them in this work.

Gallagher - Pritchard Model

This semiclassical model has been the most used model for trap loss to date. The trap loss rate can be separated into two factors. The first factor takes into account the rate at which atoms reach short internuclear separation where energy transfer can occur (either RE or FS); this represents the dynamics of the process. This rate depends on the laser excitation rates, the acceleration of the atoms in the excited state potential curves, and the spontaneous emission probability.

The second factor is the probability that energy transfer occurs once the atoms arrive at short internuclear separations.

However, the first factor is the most important because it involves radiative effects, in particular spontaneous emission during the collision, which is not present in high temperature experiments. In the original G-P model the multiple excited state potential curves are replaced by a single effective potential curve, and the trap loss rate is given as:

$$\beta \propto \int 4\pi R^2 dR P(R,\Delta_L) \exp(-t(R)/\tau) \qquad (4)$$

where $4\pi R^2 dR$ is the number of atom pairs at a distance R, $P(R,\Delta_L)$ is the excitation rate of these pairs to the excited state potential by a laser detuned from the atomic resonance by Δ_L, $t(R)$ is the time for the atoms to reach small separations where energy transfer can occur, and τ is the excited state spontaneous emission time. The probability that the excited atom pair reaches small internuclear separation without decay is given by $\exp(-t(R)/\tau)$. The shape of the excited potential is important in two different parts of equation (4): (i) It determines at which distance the excitation will take place; (ii) And it determines also the time $t(R)$for the atoms to reach the region where trap loss happens.

To simplify this expression we can consider the situation when the laser is tuned to the red of the atomic transition by an amount that is large compared with the natural linewidth of the transition, the laser excites only pairs that obey the resonance condition $h\Delta = -C_3/R^3$. We consider that the atoms are at rest when the excitation happens. In such situation, the acceleration arises from a single excited state potential of the form $-C_3/R^3$, and the trap loss rate coefficient β becomes:

$$\beta \propto \left(\frac{\Delta_\tau}{\Delta_l}\right)^2 \exp\left[-\left(-\frac{\Delta_\tau}{\Delta_l}\right)^{5/6}\right] \qquad (5)$$

where Δ_L is a characteristic frequency scale, and for sodium is 240 MHz[11]. If we excite the atoms to the $S_{1/2}$+$P_{1/2}$ potential, we have to take into account only radiative escape as the loss mechanism, and in this condition β simplifies to:

$$\beta \propto (\Delta_L^2 \sinh[(-\Delta_\tau/\Delta_L)^{5/6}])^{-1} \qquad (6)$$

which presents a maximum at $\Delta_L = -0.36\Delta_\tau$. This expression allows us to test the frequency dependence in a straightforward way.

Experimental Setup

Our measurement technique is similar to the one of ref. [13]. The MOT operates in a closed vapor cell, and the trap is loaded with atoms from the low-velocity tail present in the vapor. Three mutually orthogonal retroreflected laser beams, tuned $\approx$ 10 MHz to the red of an atomic transition frequency, intersect at the center of the quadrupolar magnetic field generated by a pair of anti-Helmholtz coils. At the center, the magnetic field is zero and grows linearly in all directions. The Zeeman splitting of the atomic levels and the use of appropriate laser polarizations produce a spatially dependent light force with a net effect of restoring the atomic position to the trap center where the magnetic field is null. In addition to the trapping force, the red detuning produces a viscous, damping force, cooling the trapped atoms to about 300 μK. The cell is a stainless-steel chamber containing a partial pressure of sodium vapor at 350 K, with a background base pressure maintained below 10^{-9} torr. The magnetic field coils are located externally to the chamber and produce an axial field gradient of about 20 Gauss/cm.

Light from a dye laser (laser I) passes through an electro-optic modulator, introducing red and blue sidebands, which are separated from the carrier by 1712 MHz; the blue one works as a repumper (Fig. 2). After the modulator the beam is divided into three beams of equal intensity along the orthogonal trapping axes. Laser I is locked in an atomic transition using a saturation absorption cell, in such way that the carrier frequency is close to the $3S_{1/2}$(F=2) $\Longrightarrow$ $3P_{3/2}$(F'=3) and the blue sideband near the $3S_{1/2}$(F=1) $\Longrightarrow$ $3P_{3/2}$(F'=2). A second dye laser (laser II) is used as an independent "catalysis laser", which can be varied in the intensity range of 50-810 mW/cm^2 and tuned in the frequency range of -1000 MHz $< \Delta <$ -50 MHz from the two possible transitions: $3S_{1/2}$(F=2) $\Longrightarrow$ $3P_{3/2}$(F'=3) (D2 line) and $3S_{1/2}$(F=2) $\Longrightarrow$ $3P_{1/2}$(F'=1) (D1 line).

We determine the number of atoms by imaging the fluorescent volume onto a calibrated photomultiplier

tube (PMT). The collisional reduction of N caused by the catalysis laser is on the order of 10%. We have performed tests, similar to ref. [13], to ensure that the catalysis is acting only on the collisions and not on the trap performance. These tests were carried out to guarantee that: (i) there is not any optical pumping during the presence of the catalysis laser; (ii) the catalysis laser do not affect the loading rate L; (iii) and it does not provide a force on the atomic cloud. This procedure restricted us in the region of $\Delta_c < -50$ MHz and $\Delta_c < -100$ MHz for D1 and D2 lines respectively. We also verified that β_c presents a linear dependence with intensity.

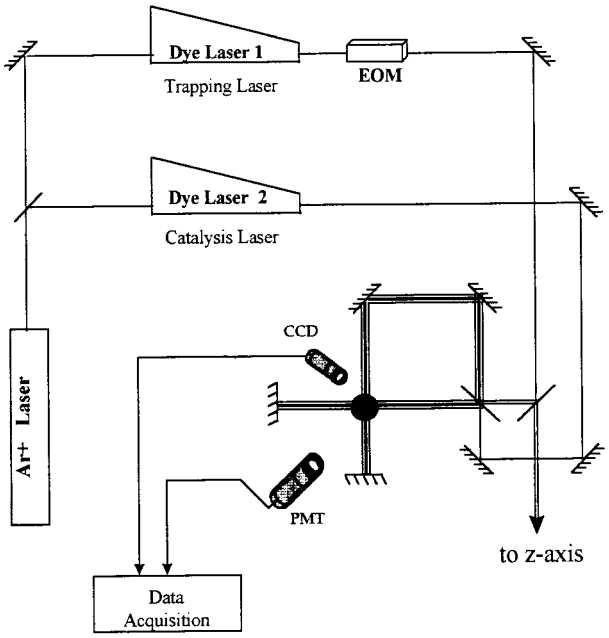

Figure 2. Diagram of the experimental setup. Dye laser I is used for trapping and laser II is the catalysis laser.

Results and discussion

In Fig. 3 we show the frequency spectra for both lines (D1 and D2). As we can observe, the spectra do not depend strongly on the hyperfine splitting of the excited state. It is also shown the predicted spectrum, obtained from equations (5) and (6) for D2 and D1 line respectively, using $\Delta_{\tau D2} = 300$ MHz and $\Delta_{\tau D1} = 210$ MHz.

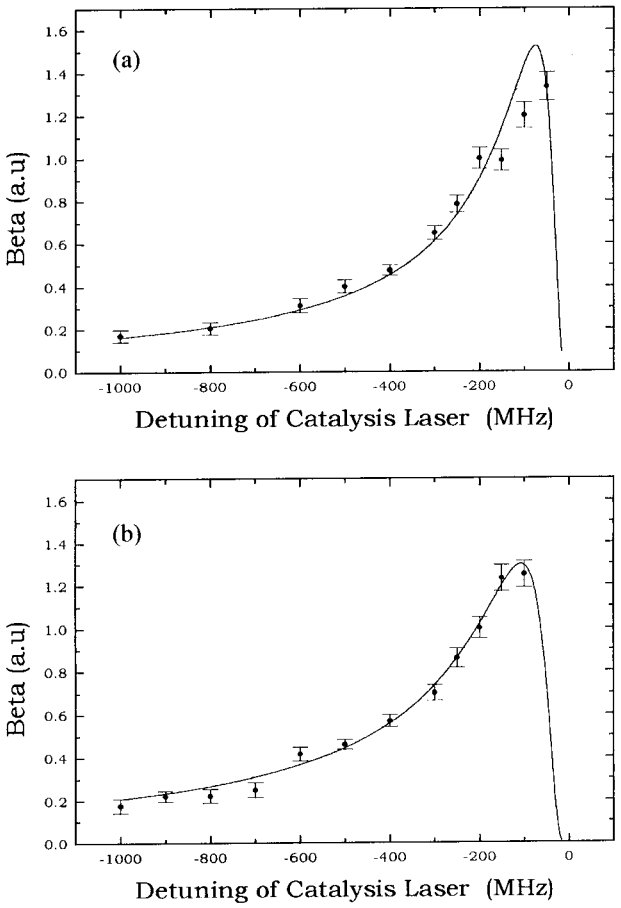

Figure 3. Frequency dependence of trap loss processes obtained by the catalysis laser technique. (a) Catalysis laser is operating close to the $3S_{1/2}(F=2) \Longrightarrow 3P_{1/2}(F'=1)$ transition (D1 line); (b) and close to the $3S_{1/2}(F=2) \Longrightarrow 3P_{3/2}(F'=3)$ transition (D2 line). The solid lines are the fittings using equations (5) and (6) and the parameters $\Delta_{\tau D2}=$ 300 MHz and $\Delta_{\tau D1}=$ 210 MHz.

Three comments should be made concerning the interpretation of Fig. 3. First, this weak dependency with the hyperfine splitting of the excited state was expected, because for sodium Δ_τ (240 MHz) is larger than the excited state splitting. In Rb and Cs we have the opposite situation. Second, the difference between $\Delta_{\tau D2}$ and $\Delta_{\tau D1}$ is an evidence of the influence of the hyperfine structure of the excited state. In the D2 line there are more potential curves involved during the collision than the D1 line, and each curve has a different C_3. Therefore there are several Δ_τ ($\Delta_\tau \propto C_3^{2/5}$), which contribute for an "effective Δ_τ". Third, we should address the issue of the maximum. For the sodium case, the maximum would be in the range of $|\Delta_\tau| < 100$ MHz. However with the present technique we can not access this region because it disturbs the trap performance. A possible way to reach this region is to carry

out the experiment in a cold dense atomic beam.

One more experiment performed was to measure the absolute value of the rate coeffficient for the catalysis laser, detuned -300 MHz from the transitions $3S_{1/2}(F=2) \Longrightarrow 3P_{3/2}(F'=3)$ (D2 line) and $3S_{1/2}(F=2) \Longrightarrow 3P_{1/2}(F'=1)$ at the intensity of 800 mW/cm^2. We measured $\beta_c = (1.0 \pm 0.2)10^{-11}$ cm^3/s and $\beta_c = (0.8 + 0.2)10^{-11}$ cm^3/s for D2 line and D1 line respectively. In the D1 line there is only radiative escape, therefore from these results we can say that fine structure chance contribution of the total loss process for sodium is in the range of 0-50%.

This is only an estimatlon, for more reliable results it is necessary to measure the population in $P_{1/2}$ directly, for example by photoionization. Such experiments are being prepared. However this result indicates that theory has to be reviewed to take into account radiative escape as an important loss process for sodium.

Conclusions

We have carried out the first optical catalysis experiment in sodium atoms close to D1 and D2 line. The results show a weak dependence of the frequency spectrum with the hyperfine excited state splitting. The overall shape of the spectrum agrees well with the semiclassical G-P model. And for the first time, experimental results indicate that fine structure change is not the dominant process for trap loss in sodium.

This work was supported by FAPESP (Fundação de Amparo à Pesquisa do Estado de São Paulo). The authors specially acknowledge V.S. Bagnato and S.C. Zilio for helpful discussion. We also thanks R. Howoricz.

References

1. For a general review of ultracold collisions: P.S. Julienne, A.M. Smith, and K. Burnett, Adv. At. Mol. Opt. Phys. **30**, 141 (1993), T. Walker and P. Feng, *ibid* **34**, 125 (1994), J. Weiner *ibid* **35**, 45 (1995).
2. V.S. Bagnato, L.G. Marcassa, C. Tsao, Y. Wang, and J. Weiner, Phys. Rev. Lett. **70**, 3225 (1993).
3. L.G. Marcassa, S.R. Muniz, E. de Queiroz, S.C. Zilio, V.S. Bagnato, J. Weiner, P.S. Julienne, and K.-A. Suominen, Phys. Rev. Lett. **73**, 1911 (1994).
4. L.P. Ratliff, M.E. Wagshul, P. Lett, S. Rolston, and W.D. Phillips, J. Phys. Chem. **101**, 2638 (1994).
5. V.S. Bagnato, L.G. Marcassa, and S.C. Zilio - Braz. J. Phys **22**, 389 (1995).
6. P.S. Julienne - private communication.
7. M. Prentiss et al., Opt. Lett. **13**, 452 (1988).
8. L.G. Marcassa, V.S. Bagnato, Y. Wang, C. Tsao, J. Weiner, O. Dulieu, Y.B. Band, P.S. Julienne, Phys. Rev. A **47**, R4563 (1993).
9. D. Hoffmann, P. Feng, R.S. Williamson III, and T. Walker, Phys. Rev. Lett. **69**, 753 (1992).
10. T. Walker, D. Sesko and C. Wieman, Phys. Rev. Lett. **64**, 408 (1990).
11. A. Gallagher and D.E. Pritchard, Phys. Rev. Lett. **63**, 957 (1989).
12. D. Hoffmann, P. Feng, and T. Walker, J. Opt. Soc. Am. B **11**, 712 (1994).
13. M.G. Peters, D. Hoffmann, J.D. Tobiason, and T. Walker, Phys. Rev. A **50**, R906 (1994).