Experimental Investigation of Atomic Collisions in Time Scales Varying from Nanosecond to Microseconds

R.D. Glover, D.E. Laban, K.J. Matherson, W. Wallace and R.T. Sang

Centre for Quantum Dynamics, Griffith University, Nathan, Queensland 4111, Australia

R.Sang@griffith.edu.au

Abstract. We present the results from two experiments investigating collisions that differ in time scale by three orders of magnitude. The first experiment enables the determination of absolute total collision cross sections using a technique that measures a change in the loss rate of trapped atoms from a magneto optical trap (MOT). We also investigate light assisted collision processes between cold metastable neon atoms in the \(^3\)P\(_2\) metastable state within the MOT. A catalysis laser is scanned in frequency across the \(^3\)P\(_2\) – \(^3\)D\(_3\) cooling transition and the ionization rate was observed. Ionization spectra are obtained which demonstrate a dependence on the magnetic sublevels of the transition that the catalysis laser is exciting.

1. Introduction

Cold and ultra-cold atomic collision processes have been the subject of intense scientific investigations over the past decade as a result of the advent of laser cooling and trapping techniques [1]. These techniques enable the trapping of atoms and create a source of dense atomic gases at mK to \(\mu\)K temperatures and have served as the source of many detailed collision experiments. At low temperatures such as this, atomic collision times between trapped atoms are relatively long and of the order of microseconds [2], compared to room temperature collisions where collision partners have velocities in the order of \(10^3\) s yielding collision times of the order of \(10^{-12}\) s. Such long interaction times yield interesting long-range interactions that are negligible at short collision times. Long range interactions between trapped atoms can also be modified though the presence of light that is mediated by the dipole interaction, whereby ground and excited state atoms interact though a potential created by the light field and has a \(1/r^3\) dependence where \(r\) is the separation distance between the interacting atoms. At low temperatures, the induced potential can exceed the kinetic energy of the colliding species even at large separation distances of \(r > 100\) nm [2].

The experimental investigation of collisions of atoms that are in atomic metastable states has been limited. This is primarily a result of the difficulty in preparing experiments with large numbers of atoms in atomic beams with a significant fraction of the atoms in the metastable state of interest. The limited number of experiments is surprising given the number of technological devices such as fluorescent lights and lasers that rely on collision processes involving these states. The work presented in this paper describes a recently developed technique by us [3] to measure absolute total atom-atom collision cross sections between neon atoms in the \(^3\)P\(_2\) metastable state (designated as Ne*) with a
variety of collision partners. In these experiments collision cross sections are obtained for collisions between the cold trapped Ne* atoms and an introduced gas species at room temperature. Such collisions take place over times scales of the order of nanoseconds. The technique relies on the observation of the population dynamics of trapped metastable atoms in a magneto-optical trap (MOT). It has the significant advantage that the trap is a pure sample of atoms in this metastable state and does not require knowledge of the number density of the atoms in the trap. Accurate collision cross sections can be measured and the technique is capable of producing benchmark atom-atom collision cross sections [3].

In the last section of this paper we present the progress of the modification of cold collisions between trapped metastable atoms with the introduction of an additional laser. The additional laser can dramatically alter the ionization rate between colliding atoms in the MOT. This additional laser, called a catalysis laser, can produce an attractive or repulsive potential between colliding atoms that depends on the frequency tuning of the laser relative to an appropriate dipole allowed transition [1]. We present the progress on initial experiments that measure ionization rates as a function of catalysis laser frequency detuning and demonstrate a rich structure that is dependant on the magnetic sublevels of the optical transition being excited. This effect has not been observed in previous cold collision studies on rare-gas metastable atoms.

2. Atom-Atom Absolute Total Collision Cross sections Using a Magneto-Optical Trap

The rate of change of the number of trapped atoms is determined by the loading and loss rates from the trap and in the case of a low density MOT is given by [3]

\[
\frac{dN_t}{dt} = R_L - N_t \Gamma_B - \frac{\beta N_t^2}{V_{eff}}
\]  

(1)

where \(N_t\) is the number of trapped metastable neon atoms, \(R_L\) is the loading rate, \(\Gamma_B\) is the trap loss rate due to the collisions with the residual background gas atoms, \(\beta\) is the density-dependent binary collision loss rate between trapped atoms and \(V_{eff}\) the effective volume of the trap and is given by \(V_{eff} = 8\pi^{3/2} w\) where \(w\) is the full width at half maximum of the trap distribution. With the addition of a collision species introduced into the interaction region the trap population dynamics is modified to

\[
\frac{dN_t}{dt} = R_L - N_t (\Gamma_B + \Gamma_C) - \frac{\beta N_t^2}{V_{eff}}
\]  

(2)

where \(\Gamma_C\) is an additional trap loss rate due to collisions of the trapped atoms with the introduced collision species. The collisions cross-section between the trapped atoms and the collisions species is given by

\[
\sigma = \frac{\Gamma_c}{n \bar{v}}
\]  

(3)

where \(n\) is the density of the collision species and \(\bar{v}\) is the relative collision velocity which is approximately given by the mean velocity of the collision species viz:

\[
\bar{v} = \sqrt{\frac{8 k_B T}{\pi m}}
\]  

(4)

where \(T\) is the temperature and \(m\) the mass of the collision species. If the loading of the trap is suspended from steady state, the population in the trap can be found by solving (2) yielding:
\[ N_t(t) = \frac{N_t(0)e^{-\Gamma_C t}}{1 + \left( \frac{\beta N_t(0)}{V_{\text{eff}}}(\Gamma_B + \Gamma_C) \right) \left( 1 - e^{-\Gamma_B t} \right)} \] (5)

\( \Gamma_C \) can be determined by fitting eqn (5) to the measured trap decay. The cross section is then found through plotting \( \Gamma_C \) as a function of \( n \), the absolute density of the collision species. As seen in eqn (3) this is a linear relationship where the gradient is proportional to the cross section \( \sigma \).

The apparatus for the experiment is discussed in detail in the references given in [3], but we provide a brief description here. A beam of neon gas supersonically expands through a hollow cathode discharge source that is cooled via a liquid nitrogen cold finger. Neon atoms emerge from the source in a variety of internal quantum states at a mean velocity of 500 m/s. A small percentage of this atomic beam has Ne atoms excited to the \(^3\)P\(_2\) metastable state that has a lifetime of 14.7 s. Atoms in this metastable state are decelerated using a Zeeman slower in combination with a counter-propagating laser detuned to the red from the \(^3\)P\(_2\) – \(^3\)D\(_3\) transition at 640 nm. Atoms in the \(^3\)D\(_3\) state can only decay to the \(^3\)P\(_2\) state and hence there are no loss channels from the cooling transition. This optical transition is also used for the MOT trap beams. Following the Zeeman slower, the MOT is formed by the intersection of three pairs of counter propagating laser beams detuned to the red of the cooling transition in combination with a set of anti-Helmholtz coils which create the appropriate magnetic field gradient. A 12-bit CCD camera is used to monitor the trapped atom population via resonance fluorescence from the trapped atoms interacting with the MOT laser beams. All laser beams are produced by a Coherent 899 single-mode ring dye laser pumped by a Coherent Verdi V5 solid state pump laser.

The experiments involved the measurement of the change in the atom number (i.e. the measurement of a decay curve) with a collision gas added to the MOT vacuum chamber at different pressures. A calibrated residual gas analyzer was used for pressure measurements and was also used to verify that the partial pressure of the collision species in the vacuum system had come to equilibrium. Once this equilibrium was confirmed, the loading of the MOT was interrupted and the decay of the MOT was measured using the imaging system.

| Collision Species | \( \Upsilon \) (m/s) | \( E_{\text{rel}} \) (meV) | \( \sigma_M (\text{Å}^2) \) | \( \sigma_R (\text{Å}^2) \) | \( \sigma_{TI} (\text{Å}^2) \) |
|-------------------|-------------------|------------------------|-----------------|-----------------|-----------------|
| He                | 1249.0            | 27.0                   | 164.8 ±0.5 ±15.5 | 123             |                 |
| Ne                | 556.3             | 16.2                   | 499.1 ±4.1 ±47.1 | 143             |                 |
| Ar                | 395.4             | 10.9                   | 837.7 ±15.7 ±79.0 | 398             | 3.46 ±30.5     |
| H\(_2\)           | 1760.0            | 29.4                   | 230.1 ±2.8 ±21.7 | 2.6 ±23.4       |                 |
| O\(_2\)           | 441.8             | 12.5                   | 1026.8 ±12.7 ±96.9 | 25.5           |
| N\(_2\)           | 472.1             | 13.5                   | 1259 ±16.5 ±118.8 | 10.4           |

Table (a) the measurements for this work where \( E_{\text{rel}} \) is the relative collision energy. The errors quoted respectively are the statistical and systematic errors respectively. (b) Measurements are from reference [4] with \( E_{\text{rel}} \approx 115 \) meV and estimated uncertainties of the order of 25%. (c) The total ionization cross sections for collision with metastable Ne in a mixed composition of \(^3\)P\(_2\) and \(^3\)P\(_0\) states has been included for comparison purposes. In this case \( E_{\text{rel}} \) is between 33 and 47 meV [5].

The results in Table 1 show the measured total absolute cross sections for collisions between trapped Ne\(*\) atoms with six different collision species. The total cross section is a combination of elastic collisions plus, in some cases, ionization events. As a comparison to our measured cross sections, Table 1 also reports the measured cross sections of Rothe and Neynaber that were taken at higher relative energies using crossed atomic beam techniques [4]. Given that these experiments are at higher energy, it is expected that these cross sections are smaller than our cross sections. It should be
noted that the contribution to the total ionization cross section is relatively small as shown by the work of Yencha [5]. In the case of Ne*-He collisions, the cross section is purely elastic since the Ne* atoms do not have enough energy to ionize the He atom. At low velocities it is expected that elastic, van der Waals type interactions dominate and the cross section can be approximated as

\[ \sigma_{\text{tot}}(v_{\text{rel}}) \approx 8 C_6 \left( \frac{\hbar v_{\text{rel}}}{\eta} \right)^{2/5} \]

(6)

To estimate the total cross section over the velocity range of this experiment, the rate of collisions leading to trap loss is calculated at each velocity using equation (6) and integrated from the capture velocity of the MOT over the Maxwellian velocity distribution. Using this method and taking \( C_6 \) to be 340 kcal mol\(^{-1}\) Å\(^6\) [6], the total elastic cross section for Ne* in the \(^3\)P\(_2\) state with thermal ground state He is calculated to be 168.9 Å\(^2\). As can be seen from our data for this cross section our measured cross section is in good agreement with this value. Our data is unique due to the low energy of the collisions as well as the relatively low uncertainties. A full analysis of the contributions to errors are detailed in Matherson et. al. [3].

3. Cold Collision Experiments in the Presence of a Catalysis Laser

The collision processes discussed in the previous section investigated binary collisions that were dominated through long-range electrostatic dispersion forces. We have also commenced an experimental investigation on the effects of light assisted collision processes. In these experiments another retro-reflected laser beam is inserted into the MOT. Such a laser is termed a catalysis laser as it can alter the collisional interactions between atoms in the trap [7]. Consider two colliding Ne* separated by a distance \( r \) in the presence of such a catalysis laser. If the catalysis laser is frequency tuned to a dipole transition moment, then the two atoms will be influenced by the resonant dipole-dipole interaction potential given by [7]:

\[ V(r) = \pm a h \gamma \left( \frac{\lambda}{2\pi r} \right)^3 \]

(7)

where \( \lambda \) and \( \gamma \) are the wavelength and the natural linewidth of the atomic transition respectively and \( a \) is configuration constant of order 1. If the transition moment is large, such as in the case of a cooling transition, then the magnitude of this potential can be significantly greater than the kinetic energy of the trapped atoms even at relatively large separation distances. The effect of such a potential between an atom in the ground state and an atom in the excited state is to create an attractive potential between the approaching atoms, thereby enhancing the probability of collision if the laser is frequency detuned to the red of an appropriate transition. In the case of a blue detuned laser beam the induced interaction potential is repulsive and collision rates are reduced [1]. A MOT trapping rare-gas metastables can also undergo ionization in the binary collision process. There are two possible ionization channels that can occur; the first is Penning ionization whereby following a binary collision event, three collision products are produced, a neutral ground state atom, an ionized ground state atom and an electron. The second process is associative ionization in which an ionized neon molecule is produced as well as a free electron.

Ionization rates for atoms in metastable states can be greatly enhanced using a catalysis laser as shown by the work of Katori and Shimizu [7]. In this work Katori and Shimizu scanned the frequency of the catalysis laser over the cooling transition and observed the change in the ionization rate of trapped metastable Kr atoms. When the catalysis laser is frequency detuned to the red of the cooling transition, the ionization rate increased dramatically and peaked close to resonance. Past resonance, when the laser is frequency tuned to the blue side of the transition, the ionization rate decreased.
dramatically and was inhibited for a small frequency range due to optical shielding as a result of a repulsive potential induced by the catalysis laser [1].

The goal of this work was to investigate the enhancement of ionization processes in the Ne* MOT using such a catalysis laser. The apparatus described for the measurement of the absolute total collision cross sections was modified to facilitate the detection of charged products resulting from ionizing collisions in the MOT. Ionization processes from the trap were monitored by the use of a channel electron multiplier with a simple electrostatic discrimination system that assisted in the detection of Ne* ions. A catalysis laser beam was provided by an additional ring-dye laser (Spectra Physics 380D operating with krypton red laser dye pumped by a Verdi V5 laser) and was inserted into the MOT in a retro-reflecting configuration so as to minimize trap loss resulting from momentum transfer from the light beam due to photon absorption. The laser beam was circularly polarized in the same polarization configuration as the MOT trap beams and could be scanned over the cooling transition with approximately 1GHz frequency detuning range. The laser power was typically of the order 100 times the saturation intensity (~4mW/cm²) of the cooling transition.

The resulting ionization spectrum is shown in the left of figure 1 and displays a complex spectrum unlike that of the work of Katori and Shimizu. The spectrum shows multiple ionization enhancement peaks when the catalysis laser is frequency detuned over the cooling transition. At a first glance, one might deduce that this spectrum is displaying these features due to the formation of molecular ions through the associative ionization process with the peak structure a result of resolving molecular vibrational energy levels. However, this is unlikely due to the probability of Penning ionization having an order of magnitude greater probability [5]. A further point to note is the enhancement in the ionization peak at approximately +50 MHz when the catalysis laser is frequency detuned to the blue of the cooling transition. This is completely unexpected as optical shielding should occur in this region of the spectrum and therefore reduce Ne* ion production.

Further experimental investigation shows that the ionization spectrum is dependent on the magnetic field produced by the MOT coils. Increasing the current in the coils produced more peaks in the ionization spectrum as well as shifting the peaks as shown in the spectrum on the right of figure 1. The cooling transition that is used for the catalysis laser displays fine structure and the magnetic sublevels of the two participating atomic states in the MOT have their degeneracy lifted due to the Zeeman shift as a result of the magnetic field created by the MOT coils. In the case of the magnetic field gradient of 0.4 T/m the various transitions between magnetic sublevels that can be excited are given in table 2 at a distance of 1mm from the centre of the trap.

Figure 1: Ion count rate Vs catalysis laser frequency detuning relative to the 3P_2-3D_3 cooling transition; left figure MOT trap gradient ~ 0.25 T/m, right figure MOT trap gradient ~0.4 T/m.

Further experimental investigation shows that the ionization spectrum is dependent on the magnetic field produced by the MOT coils. Increasing the current in the coils produced more peaks in the ionization spectrum as well as shifting the peaks as shown in the spectrum on the right of figure 1. The cooling transition that is used for the catalysis laser displays fine structure and the magnetic sublevels of the two participating atomic states in the MOT have their degeneracy lifted due to the Zeeman shift as a result of the magnetic field created by the MOT coils. In the case of the magnetic field gradient of 0.4 T/m the various transitions between magnetic sublevels that can be excited are given in table 2 at a distance of 1mm from the centre of the trap.
Table 2: Zeeman frequency shifts for optical transitions between magnetic sublevels for the $^3P_2 - ^3D_3$ optical transition for $\sigma^-$ light ($\Delta m_J=-1$), $\pi$ light ($\Delta m_J=0$) and $\sigma^+$ light ($\Delta m_J=+1$) at a displacement of 1 mm from the trap centre with magnetic field gradient of 0.4 T/m. The left hand column displays the magnetic sublevel of the lower energy magnetic sublevel.

| $^3P_2$ | $\Delta m_J=-1$ (MHz) | $\Delta m_J=0$ (MHz) | $\Delta m_J=+1$ (MHz) |
|---------|------------------------|----------------------|------------------------|
| 2       | -58.9                  | -11.8                | 35.3                   |
| 1       | -53.0                  | -5.9                 | 41.2                   |
| 0       | -47.1                  | 0                    | 47.1                   |
| -1      | -41.2                  | 5.9                  | 53.0                   |
| -2      | -35.3                  | 11.8                 | 58.9                   |

The spectrum on the right of figure 1 shows that there are two peaks in the ionization spectrum at approximately ±50 MHz which are close to the two transitions for both, $\sigma^-$ from the $^3P_2$ ($m_J=+2$, $\Delta m_J=-1$), and $\sigma^+$ from the $^3P_2$ ($m_J=-2$, $\Delta m_J=+1$) transitions respectively. It is likely that the peaks in the ionization spectrum are from these transitions as well as contributions from the other transitions that are not frequency resolved. The general shape of each of the peaks is in good agreement with that of the work of Katori and Shimizu [7], in that each of the peaks in the spectrum show a gradual increase in the ionization rate on the red side of the transition with a fast drop off in the ionization rate when tuned to the blue of the transition. Our spectra also show that the background ionization rate is suppressed on the blue side of the transition compared to the background rate at far red detuning. This is consistent with optical shielding effects as demonstrated in previous experimental results [7]. We should point out that the work of Katori and Shimizu does not show a multi-peak structure like ours, however, the magnetic field gradient in that work is nearly an order of magnitude smaller than this work and it would be unlikely they could resolve the fine structure. This would also explain why their work shows a broader ionization peak due to the influence of all of the magnetic sublevel transitions.

To confirm our findings we could use a smaller magnetic field gradient but it is not currently possible with our current experimental configuration. Experiments are now underway that will perform these measurements in a pulsed mode so that the ionization rates are measured without the influence of any magnetic field to confirm our expectation.

4. Acknowledgements
This research was supported by the Australian Research Council and Griffith University.

References
[1] J. Weiner, V.S. Bagnato, S. Zilio, and P. S. Julienne, Rev. Mod. Phys. 71, 1 (1999).
[2] S.D. Gensemer and P.L. Gould, Phys. Rev. Lett 80, 936 (1998).
[3] K.J. Matherson, R.D. Glover, D.E. Laban, and R.T. Sang, Rev. Sci. Instrum. 78, 073102 (2007).
K.J. Matherson, R.D. Glover, D.E. Laban, and R.T. Sang, Phys. Rev. A 78, 042712 (2008).
[4] E.W. Rothe and R.H. Neynaber, J.Chem. Phys. 42, 3306 (1965).
[5] A.J. Yencha, Electron Spectroscopy: Theory and Technique, and Application, edited by C.R. Brundle and A.D. Baker (Academic Press, New York, 1984), Vol 5.
[6] T. Fukuyama and P. Siska, Chem. Phys. Lett. 39, 418 (1976).
[7] H. Katori and F. Shimizu, Phys. Rev. Lett. 73, 2555 (1994).