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Magneto-optical trap and its application to ultra-cold atom collision studies

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Rice University, 1994
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Magneto-Optical Trap and Its Application to Ultra-Cold Atom Collision Studies

by

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A Thesis Submitted in Partial Fulfillment of the Requirements for the Degree Master of Arts

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Abstract

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We have successfully trapped $^7$Li in a vapor cell magneto-optical trap (MOT). The trap is studied in detail and the parameters are measured from the experiment. The trapping mechanism has been understood in terms of the radiation force and a simple one dimensional model is used in interpreting and predicting the experimental results. The cold-cold atom collision rate coefficient is measured in a beam MOT. The two collision mechanisms, fine structure changing and radiative escape, are discussed in a molecular picture. One of the channels, the fine structure changing channel can be turned on and off by the experimental conditions, namely the laser intensity in this experiment. The rate coefficient for each mechanism has been determined from the experiment.
Acknowledgements

Not until the last page of this thesis did I realize how enjoyable the two-year work has been. I have to thank my advisor, Randy Hulet, for letting me finish this project and reading of the thesis. I want to thank my colleagues Jeff Tollett, Curtis Bradley, Eric Abraham, Cass Sackett, Ian McAlexander for making the life in the lab joyful and memorable. I want to thank my committee members Dr. King Walters and Dr. Ian Duck for their interest in my work and their approval of my Ph.D candidacy, despite my leaving of this department. I also have to thank Nick Ritchie for teaching me a good lesson on humanity.

This work is for my father and my mother, who led me the way to the nature and the science. I want to thank my sister for her cheeriness and good heart. Last, but never the least, I want to thank Ke for his expert help on experiment and computer.
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Introduction

The laser cooling technique, that is, the technique using the interaction between the atoms and a near atomic resonance laser field to slow the atomic motion or trap the atoms, have advanced rapidly since its initial studies in late 70's by Ashkin [1978], Wineland [1979] and Gordon [1980]. The work that has been done in the 80's by Phillips [1982], Chu [1985], Ertmer [1985], Prodan [1985] and Lett [1988] realized the slowing in experiments. Since then, many new cooling schemes have been developed and optical molasses is used to trap atoms optically [Chu, 1986], [Pritchard, 1986]. The latest trapping technique, the magneto-optical trap was developed in 1987 by Raab et al [1987]. This technique can be applied to both atomic diffusive vapor cell or a properly slowed atomic beam from an effusive atomic oven. Both cases have been realized in our experiment. The cell MOT has the advantage of small size and simple operation. The beam MOT, on the other hand, involves rather complicated laser cooling techniques, which in turn relies on other techniques, such as quantum electronic devices, but has a larger number of trapped atoms without contaminating the vacuum by the background atoms.

The large number of these ultra-cold atoms in the trap provides a unique ultra-cold atom source for new applications as well as to study new physics. Much physics can be re-studied in the ultra-cold regime, with collisions being one of the most important. Collisions are important for atom trapping for the reason that it is one of the limiting factors in achieving large number and density of
cold atoms. It is also important to collision physics because the new features arising from the fact of slow atom collisions are a new test and stimulus to present collision theory, especially to quantum collision theory since quantum properties are more prominent in this ultra-cold system. Both theory and experiments are under development in this new regime. We measured the cold-cold Li collision rate coefficient in the previously mentioned beam MOT and I will discuss the experiment and theory in Chap 3 after the discussion of the realization of the MOT in Chap 1 and MOT parameters in Chap 2.
Chapter 1

Trap Li Atoms in a Magneto-Optical Trap

1.1 Lithium magneto-optical trap realization

The first magneto-optical trap (MOT) was demonstrated in 1987 [Raab, 1987]. Because of its compactness and simple operation, it is becoming an attractive technique for obtaining cold neutral atoms.

A MOT utilizes the interaction between the atomic magnetic moment, the external inhomogeneous magnetic field and the laser light field. The atoms are slowed down by the Doppler force, and are trapped in a magnetic field minimum. In a simplified two level system of a $J=0$ ground state and a $J = 1$ excited state, as shown in Fig. 1.1, the excited state energy levels are Zeeman-shifted by the magnetic field. A pair of anti-Helmholtz coils provide a magnetic field that has its minimum zero field at the center and a constant field gradient in the small region around the center. A slightly red detuned, circularly polarized laser beam is retro-reflected back with opposite polarization. Because of the different Zeeman energy shifts of the $m_J$ levels, the atoms interact more strongly with one polarization than with another. For the atoms in $z > 0$ region, $\sigma^-$ exerts a larger force than $\sigma^+$, because $\sigma^-$ transition is closer to resonance, the atoms are pushed towards the origin. Similarly for the atoms in $z < 0$ region.

To trap the atoms three dimensionally, six laser beams from the $x$, $y$, $z$ directions are crossed in the center of the trap as shown in Fig. 1.2. The polarization is with respect to propagation direction of each beam. The laser beams naturally set up a
Figure 1.1 Magneto-optical trap in a two level system: $J = 0$ ground state and $J = 1$ excited state. Two coils in the anti-Helmholtz configuration provide the "spherical quadrupole" magnetic field; trapping laser beams are circularly polarized in opposite directions.

Cartesian coordinate system where $z$ is along the vertical direction, $x$ and $y$ are in the horizontal plane. The magnetic field in $x$ and $y$ axes are also linear but are half of that in the longitudinal $z$ direction in the anti-Helmholtz configuration. For the carefully chosen magnetic field direction and laser polarization as in Fig. 1.2, the trapping along each of the three axes can be satisfied independently.

Our experiment was first carried out in a vapor cell magneto-optical trap similar to the Cs trap of Monroe et al [1990]. The cell is a ten-way-cross vacuum chamber, pumped by a turbo pump. The vacuum pressure is read by a hot cathode ionization gauge. Because of the low lithium vapor pressure at room temperature, the cell is heated up by several heating tapes. A finger-size oven containing lithium metal was built under one of the arms. The cell is normally run at 200°C and the oven at 300°C.
Figure 1.2 3D Magneto-optical trap. Six laser beams propagating along three perpendicular axes, together with chosen field direction form the trap.

The magnetic field coils are thick copper wire, capable of carrying 40 A current. They were placed around the top and bottom arms in the anti-Helmholtz configuration.

The experimental setup is shown in Fig. 1.3. A Spectra-Physics CW ring dye laser is pumped by an argon ion laser to produce an output of $\sim 400$ mW at 671 nm for $^7$Li $2S_{1/2}$ to $2P_{3/2}$ transition (excited state lifetime 27.29 ns, transition linewidth $\Gamma = 2\pi \times 5.8$ MHz). The frequency is measured by a home-made Michelson interferometer with a resolution of 0.5 GHz. The laser frequency is stabilized by locking the laser to a lithium heatpipe, using Doppler-free saturation spectroscopy [Demtroder, 1981]. The pump laser frequency is shifted to the blue relative to the probe frequency by double passing the acousto-optical modulator (AOM) in the saturation absorption locking system. The laser is locked to the crossover-resonance point of $F = 1$ and
Figure 1.3 Vapor cell MOT schematic (top view). Six circularly polarized laser beams (top beam not shown) and the cell. The source of the hot atoms is a finger-size oven under one of the arms of the chamber. This is the type of setup studied in this and the next chapter.

\[ F = 2 \text{ of } 2S_{1/2} \text{ to } 2P_{3/2} \text{ transitions.} \]  
As a result, the laser frequency was accurately stabilized to within the linewidth of the transition frequency. Other than occasional large frequency jumps, the laser can stay locked within a workday.

The laser excited transitions and \( ^7 \text{Li} \) energy levels are shown in Fig. 1.4. Because of hyperfine structure, \( ^7 \text{Li} \) (actually, same for all the other alkali species) is not a simple two level system. Two laser frequencies coupling both \( F = 1 \) and \( F = 2 \) of \( 2S_{1/2} \text{ to } 2P_{3/2} \) are required, otherwise the atoms will eventually be stranded in the
unpumped ground state and be lost from the trap. (The $2P_{3/2}$ is referred sometime without specifying the hyperfine state, because the hyperfine structure splitting is only three transition linewidths and is not well resolved such as in the absorption measurement in the next chapter.)

![Diagram of energy levels](image)

**Figure 1.4** Laser excited transitions and $^7$Li energy levels (not drawn in scale). The EOM first order sidebands provide both trapping transitions $F = 1$ and $F = 2$ of $2S_{1/2}$ to $2P_{3/2}$. The inset shows the laser central frequency and first-order sideband after the EOM.

We used the first-order sidebands from an electro-optical frequency modulator (EOM) to conveniently provide both frequencies ($\omega^+$ and $\omega^-$ in Fig. 1.4). The EOM uses a LiTaO$_3$ crystal in a split-ring resonator originally designed for use in magnetic resonance at 200-2000 MHz [Hardy, 1981], [Kelly, 1987]. The essentially lumped LC resonator has a Q factor of about 100 and requires only $\sim 300$ mW rf power at a 406 MHz modulation frequency to achieve the maximum first-order sideband power (34%
of the incident power in each sideband). The design principle and schematic are in Appendix A.

The frequency-locked output from the laser is focused down to less than 500 μm and passes through the EOM. The AOM in the main beam shifts the frequency (including the sidebands) 40 MHz to the blue. The AOM also deflects the laser beam so that it can be used as a shutter to turn the laser beam on and off in the experiment. This deflection AOM combined with the tunable AOM in the saturation absorption locking system result in a laser frequency several linewidths detuned from both atomic hyperfine transitions.

The laser intensity is carefully controlled by using two polarizing beamsplitters and two λ/2 plates to balance the intensity in the three directions. Highly reflecting dielectric mirrors are used to reduce the intensity loss of the retro-reflected beams. Because the number of atoms in the trap increases approximately to the 4th power with the trapping beam diameter for constant laser intensity (see Chap 1.3), the laser beam is collimated and expanded to a beamwaist of about 6 mm, where the beamwaist w is defined by the Gaussian profile of the radial intensity distribution:

$$I(r) = \frac{2P}{\pi w^2} \exp \left( -\frac{2r^2}{w^2} \right)$$

(1.1)

and P is the total power of the laser beam.

The atoms can be trapped for a large range of trapping parameters. The detuning can be varied from one to more than ten linewidths; the longitudinal field gradient from 10 to 40 Gauss/cm limited by the power supply and the amount of heat required to dissipate from the field coils. We typically run at two to four linewidths detuning and 20 Gauss/cm field gradient. The variation of trap with these parameters are discussed in the next chapter. The laser intensity is limited by the laser output power and to maintain a saturation absorption. The beamwaist is 6 mm, and the
intensity is typically 30 mW/cm² in each beam in one first-order sideband. The saturation intensity is 7 mW/cm² for both \( F = 1 \) and \( F = 2 \) to \( 2P_{3/2} \) transitions.

Because of the strong fluorescence at 671 nm of Li, the trapped atoms form red dense glowing cloud and can be seen by the naked eye or monitored by a CCD camera.

### 1.2 Radiation force and trap mechanism

Atoms that are interacting with the light field are subject to a radiation force. The trap mechanism can be understood by this radiation force. Consider an ensemble of moving atoms and a slightly red detuned near-resonance laser beam of frequency \( \omega_f \) as in Fig. 1.5. \( \omega_{\text{laser}} - \omega_{\text{resonance}} = \Delta \) is the detuning. Atoms that are moving towards the laser beam can be Doppler shifted into resonance with a red-detuned (\( \Delta < 0 \)) laser beam, if \( \mathbf{k} \cdot \mathbf{v} = \Delta \). These atoms can absorb photons and be excited to the excited states. Each absorption receives a momentum kick of \( \hbar \mathbf{k} \) from the photon which provides a force in the direction of \( \mathbf{k} \). The re-emission of a photon through spontaneous emission is isotropic: the average momentum change is zero.

![Atomic ensemble](image)

**Figure 1.5** Atoms that are moving towards the near-resonance laser beams are slowed down by repeatedly absorbing and re-emitting photon.
Thus, after many absorption and re-emission processes, some of the atoms are slowed down [Wineland, 1979].

For a particular atomic transition of resonant frequency $\omega_0$ and excited state lifetime $\tau$, the radiation force (Doppler force) in terms of momentum transfer in one dimension is:

$$F_{Dop} = \hbar k R = \hbar k \frac{I\sigma}{\hbar \omega}$$  \hspace{1cm} (1.2)

where $k$, $\omega$ and $I$ are the laser wave-vector, frequency and intensity, respectively. The excitation rate $R$ is related to the atomic absorption cross section $\sigma$ as shown. Including the power broadening, the excitation rate has a Lorentzian profile of

$$R = \frac{\Omega^2 \Gamma}{2 \Omega^2 + 4 \Delta^2 + \Gamma^2}$$  \hspace{1cm} (1.3)

where $\Gamma = 1/\tau$, is the transition linewidth in radian/s. The Rabi frequency $\Omega$ is defined as $\Omega = \sqrt{I/I_s \Gamma}$, and $I_s$ is the saturation intensity of this particular transition. $I_s = \hbar \omega \Gamma / \sigma_0$, where $\sigma_0$ is the on-resonance absorption cross section.

In the MOT, taking into account the thermal velocity $v$ and the Zeeman shift in the linear magnetic field gradient of $b$, the effective detuning $\Delta = \omega - \omega_0 \pm kv \pm \mu_B b z$, and the Doppler force in the trap in one dimension can be written as:

$$F_{Dop} = F_{\sigma^+} - F_{\sigma^-} = \hbar k \left[ \frac{\Omega^2 \Gamma}{2 \Omega^2 + 4 (\Delta - kv - \mu_B b z)^2 + \Gamma^2} - \frac{\Omega^2 \Gamma}{2 \Omega^2 + 4 (\Delta + kv + \mu_B b z)^2 + \Gamma^2} \right]$$  \hspace{1cm} (1.4)

where $\mu_B / 2\pi = 1.40$ MHz/Gauss is the Bohr magneton, and $\Omega$ is the 1D Rabi frequency for $F = 1$ or $F = 2$ to $2P_{3/2}$ transition.

The force as a function of the position is shown in Fig. 1.6. The force is localized around the origin for slow atoms. For fast moving atoms, the force is merely displaced opposite to its velocity. As atoms are slowed down, the force changes with the velocity. The potential well is similar to that of harmonic oscillator at trap center and thus the
Figure 1.6  Top: Doppler force as a function of position for a set of atomic velocities. The parameters are $b = 20$ Gauss/cm, $\Omega = 0.4 \Gamma$, $\Delta = -2.0 \Gamma$.
Bottom: Potential well for the same parameters from the integration of $F_{Dop}$ over $z$ (in unit of $\hbar k \Gamma$ cm.)

motion of the trapped atoms can be approximated as harmonic oscillation. Solving for the motion equation under this force, we found there is an initial maximum thermal velocity (capture velocity) for a certain trap configuration of which atoms can be trapped. The calculated capture velocity ranges from 30 to 100 m/s in our trap depending on the actual trapping parameters, i.e., the laser intensity, detuning and
the magnetic field gradient. Fig. 1.7 is the calculated capture velocity from this 1D model vs detuning for various parameters.

![Graph](image-url)

**Figure 1.7** Calculated capture velocity vs detuning for various trapping conditions. The solid line is a spline interpolation fit.

In the above 1D model, I neglected the hyperfine splitting in both the ground and the excited states. The Zeeman shift of a hyperfine state is $g_F m_F \mu_B b z$. The multihyperfine states are simplified if we assume a strong optical pumping in the trap. The trapping states are the $m_F = \pm 2$ of the ground $F = 2$ state and the $m_{F'} = \pm 3$ of the excited $2P_{3/2}, F' = 3$ state. The Landé $g$ factors are $g_F = 1/2$ for the $F = 2$ and $g_{F'} = 2/3$ for the $F' = 3$. The energy difference between the ground and the excited states Zeeman shifts are the same as in the simplified two level system as in Fig. 1.1, i.e. $\Delta g \equiv g_{F'} m_{F'} - g_F m_F = \pm 1$. There is also a significant number of atoms in the ground $F = 1$ hyperfine state. The trapping states are $m_F = \pm 1$ of the ground $F = 1$ state and the $m_{F'} = \pm 2$ of the excited state. The $g$ factors are -1/2 and 2/3, respectively. Thus $\Delta g = \pm 11/6$. Because of the optical pumping effect of the laser transition from $F = 1$ to $2P_{3/2}$ ($\omega^+$ in Fig. 1.4), there are typically two
times atoms in the $F = 2$ state than that in the $F = 1$ state at two to five linewidths laser detunings. In the model, we used $\Delta_\mu = \pm 1$ for approximation.

1.3 Trap dynamics

In this experiment, we desired to obtain a large number of atoms as well as a large density. The trap dynamics are examined more carefully to achieve this. When the trap is first turned on, the number of the atoms in the trap can be described by the simple rate equation:

$$\frac{dN(t)}{dt} = L - \gamma_6 N(t)$$

(1.5)

where $L$ is the trap loading rate, $\gamma_6$ is the collision rate of one cold atom with hot background gas. (Ignore cold-cold atom collision at this moment.) Solving the equation for $N(t = 0) = 0$, gives

$$N(t) = N_s (1 - e^{-\gamma_6 t})$$

(1.6)

The steady state number of atoms $N_s = L/\gamma_6$.

The background collision rate $\gamma_6$ is determined by the cell pressure $P$ (i.e., cell temperature $T_c$). If we ignore the vapor pressure other than Li.

$$\gamma_6(T_c) = n_c(T_c) \sigma_c \sqrt{\frac{8k_BT_c}{\pi m}}$$

(1.7)

where $n_c$ is Li density in the cell, $\sigma_c$ is the collision cross section.

The Li pressure $P$ is approximated by a logarithm of $T_c$ at low pressure [O'Haulon, 1980].

$$P(\text{torr}) = 10^{-11}10^{(T_c-425)/20.3}$$

(1.8)

where $T_c$ is in Kelvin. Therefore, $\gamma_6$ increases approximately linearly with cell pressure. $\sigma_c$ is obtained from the experiment as I will describe in the next chapter. Low pressure is required to observe the cold-cold collisional process as the cold-cold collision will be swamped by the large background collision if the vacuum is not high.
enough. I will discuss this more in detail in the context of the cold-cold collision rate measurement.

The other quantity, the loading rate \( L \), is determined by the trap region dimension, capture velocity and the Li background gas density in the cell. Assume a thermal equilibrium in the vacuum chamber, the loading rate is determined from the thermal flux into the trapping area with thermal velocity less than capture velocity. In the experiment, the trap region is defined by the trapping laser diameter which is apertured down to \( d =1.5 \) cm and is approximated as a cubic region.

The loading rate is the number of atoms that enters this cubic region per unit time.

\[
L = 6d^2 \int_0^{v_z} dv_y \int_0^{v_z} dv_z \int_0^{v_z} v_z f(v_x, v_y, v_z) dv_x
\]
\[
= \frac{1}{\pi^{3/2}} d^2 n_c a^3 v_c^3 2v_c (2v_c + v_c') \approx \frac{3}{\pi^{3/2}} d^2 n_c v_c^4 a^3
\]  
(1.9)

where \( f(v_x, v_y, v_z) \) is the Maxwell-Boltzmann distribution:

\[
f(v_x, v_y, v_z) = n \left( \frac{a^3}{\pi^{3/2}} \right) \exp \left[ -a^2 (v_x^2 + v_y^2 + v_z^2) \right]
\]  
(1.10)

and

\[
a^2 = \frac{m}{2k_B T}.
\]  
(1.11)

In Eq. 1.9, I used the capture velocity from the one dimensional model. \( x \) and \( y \) axes are symmetric if we assume equal intensity in all axes, while \( z \) axis is a little different resulting from the larger field gradient. In the experiment, because the laser intensity has a Gaussian distribution, the off-axis Rabi frequency is smaller than the on-axis one, while the off-axis atoms are under the influence of laser beams from two or three directions. These factors may be cancelled with each other as we observe a rather uniform spherical (or elliptical, depending on the intensity in the \( z \) axis relative to \( x \) and \( y \)) distribution in the experiment. The measurement, such as the loading rate,
is also close to the 1D model. However, some features in later collision experiment may suggest that the system is dynamically non-uniform (such as the trap depth), although the average property is close to the 1D model.

The steady state number of atoms in the trap, combining Eq. 1.7 and 1.9, is thereby

\[ N_s = \frac{L}{\gamma_b} = \frac{0.12}{k_B} \cdot d^2v_c^2 \cdot \frac{m^2}{\sigma_c T^2}. \]  
(1.12)

In the first order approximation, we can take \( v_c^2 = 2ad \), where \( a \) is the Doppler cooling acceleration, \( a \propto I/m \) at low laser intensity \( I \) (i.e. \( \Omega \simeq 1 \)) and a few linewidths detunings. Therefore,

\[ N_s \propto \frac{d^4I^2}{\sigma_c T^2}. \]  
(1.13)

At low intensities, since \( I = 2P/\pi w^2 \simeq P/d^2 \) (\( w \) is the laser beamwaist), the number of atoms in the trap will not be affected by \( d \) for the same laser power. We once doubled the laser beamwaist for the same laser power of total \( \sim 150 \text{ mW} \), we only see a factor of two improvement in the number of trapped atoms. But if the intensity is kept as constant, \( N_s \) will increase dramatically with \( d \), as has been seen in Gibble [1992] and Lindquist [1992]. The index to the \( d \) is measured to be 3.65 in the Cs MOT in Gibble instead of 4 from Eq. 1.13.
Chapter 2

Lithium Vapor Cell MOT Studies

The results presented in this chapter are based on the vapor cell magneto-optical trap. The trap is studied to obtain large number of trapped atoms and to measure the parameters for later collision experiments.

2.1 Cloud shape

The size and shape of the trapped atoms are determined by grabbing the image from the CCD camera. The size is defined by the complex combination of the trapping parameters: laser detuning, intensity and magnetic field gradient. Once the atoms are trapped, the atoms are slowed to a small velocity (mean velocity at 1 mK is 1.7 m/s) and are confined within a small range around the trap center. Notice that in Eq. 1.4, $ke, \mu b z \ll \Delta$ when the atoms are trapped. The force is then approximated as:

$$F_{rpp} = \frac{16F_o \cdot \Omega^2}{(2\Omega^2 + 4\Delta^2 + \Gamma^2)^2} \cdot (-kz - \mu b z)$$

$$= -\alpha r - Kz$$ (2.1)

where $F_o = \hbar k \Gamma$. The atomic motion in the trap is therefore approximated as damped harmonic oscillation with the spring constant $K$ and damping coefficient $\alpha$. The photon scattering causes random fluctuation in the atomic motion. This random nature of photon scattering can be modeled in analogy with the Brownian motion [Stenholm, 1984]. From the Langevin's equation or the Fokker-Planck equation of a harmonic oscillator in a damping medium, we obtain a Gaussian density distribution...
Figure 2.1  Grabbed image from CCD camera (top) and the Gaussian fit to the intensity (bottom).

[Chandrasekhar, 1954]. The steady-state distribution as $t \to \infty$ is:

$$n \propto \exp \left( -\frac{z^2}{2 < z^2 >} \right)$$  \hspace{1cm} (2.2)

where $< z^2 >$ is the mean square of the displacement and it satisfies the equipartition principle,

$$k_B T = K < z^2 > .$$  \hspace{1cm} (2.3)
$T$ is the trap temperature (not the cell temperature $T_c$). The radius $w_z$ given by the $1/e^2$ radius is

$$w_z = \sqrt{2 < z^2 >}.$$  \hspace{1cm} (2.4)

The three-dimensional motion can be broken down to three one-dimensional cases, since the Gaussian distribution in each axis is uncoupled from any other axes. Fig. 2.1 shows the CCD image of the trapped atoms and the Gaussian fit. The typical radius is $200 \sim 500 \mu m$. The variation with detuning is shown in Fig. 2.2. This data set was taken at $x$ axis with Rabi frequency $\Omega = 2.0 \Gamma$, field gradient $b_x = 13$ Gauss/cm. The data was fitted to Eq. 2.3, where $K$ is defined as in Eq. 2.1. The fitting parameter $T$ is found to be 2.5 mK. ($T$ is actually varied with detuning as discussed in the next section, but the variation is quite slow at high intensity and several linewidths detunings. See Eq. 2.8.)

![Graph showing variation of cloud radius vs detuning](image)

**Figure 2.2** Variation of the cloud radius vs detuning. The solid line is the fit from Eq. 2.1 and 2.3.
In the experiment, we observed that the larger the laser detuning, the bigger the radius; while the larger the field gradient, the smaller the size. These observations are in agreement with the Doppler cooling theory, but are very different from that of sub-Doppler cooling, which predicts a constant radius for different detunings. The trap temperature from the fit is 2.5 mK, which is consistent with the time-of-flight measurement discussed below.

2.2 Trap temperature

The time-of-flight technique was incorporated to measure the trap temperature in the early stage of the experiment. After we trapped the atoms, the trap was turned off instantly. The fluorescence signal from the time-of-flight probe placed at a position about 1 cm below the trap was taken vs time. One of the data sets taken is shown in Fig. 2.3. The peak was positioned at 4.0 ms, the initial most probable velocity is then 2.0 m/s, which corresponds to a trap temperature of 1.7 mK, i.e.,

\[ T = \frac{1}{2} m v_p^2 / k_B = \frac{1}{2} m \left( \frac{D_z}{t_p} \right)^2 / k_B. \]  

(2.5)

\( v_p \) is the most probable velocity at temperature \( T \). \( D_z = 0.8 \text{ cm} \) is the distance between the detector and trap center and \( t_p \) is the peak position in the TOF spectrum. Gravity is a small effect for the time scale of \( 10^{-3} \) seconds. It changes the result by less than 3%. The result is mainly limited by the time resolution of the measurement, which is 0.5 ms. Therefore the major uncertainty is

\[ \frac{\Delta T}{T} = 2 \frac{\Delta t}{t_p} \simeq 25\% \]  

(2.6)

i.e. \( T = 1.7 \pm 0.4 \text{ mK} \).

From the thermal velocity distribution, we can calculate the expected theoretical curves for various temperatures. The TOF signal is proportional to the number of
Figure 2.3 A typical time-of-flight spectrum shown in dots. The peak position is the signature of the trap temperature. Detector was 0.8 cm underneath the trap. Lines show results (normalized to 1) from Eq. 2.7 for temperatures of 0.1, 1.7 and 10 mK.

Atoms that falls into the detection region after the trap being turned off, i.e.

\[ S \propto \int_{-\frac{h_i}{2}}^{\frac{h_i}{2}} dv_x \int_{-\frac{h_y}{2}}^{\frac{h_y}{2}} dv_y \int_{-\frac{h_z}{2}}^{\frac{h_z}{2}} dv_z f(v_x, v_y, v_z) dv_y. \]  

(2.7)

In the experiment, \( 2h_i, i=x,y,z \) is the detection region dimensions in each axis and is 0.4, 0.85, 0.1 cm, respectively. The decay factor of \( e^{-\gamma_i t} \) (\( \gamma_i \) is about 3 s\(^{-1}\)) is negligible for the time scale of \( 10^{-3} \) sec. The results are shown in Fig. 2.3 for temperatures of 0.1, 1.7 and 10 mK.

The trap temperature is consistent with the Doppier cooling theory. The Doppler cooling limit \( T_{Dop} \) when intensity goes to zero and detuning at half linewidth [Wineland, 1979] is 140 \( \mu \)K for Li. Lett et al [1989] gave the temperature dependence on laser intensity and detuning for 3D optical molasses in a two level system:

\[ k_B T = D_{\nu}/\mu \]
\[
\frac{\hbar \Gamma}{\Delta} \left( \frac{\omega^2 + 4 \Delta^2 + \Gamma^2}{2 \Delta \Gamma} \right) = k_B T_D \frac{\omega^2 + 4 \Delta^2 + \Gamma^2}{4 \Delta \Gamma}
\] (2.8)

where $\omega$ is the Rabi frequency of one beam in one first-order sideband, $D_p$ is the momentum diffusion constant, and $\alpha$ is the damping coefficient.

Figure 2.4 Time-of-flight spectrum for various field gradients (top) and detunings (bottom). The temperature indicated by the peak position is unchanged.

This is valid in the MOT because at the trap center, the magnetic field is approaching zero. Most importantly, the equilibrium temperature is determined by the equilibrium of the heating and the dissipative processes, that is, the momentum diffusion caused by the random photon scattering and the damping part of the Doppler
force, whereas the magnetic field provides the conservative restoring force. The result from Eq. 2.8 is $9T_{Dop}$ at $\Omega = 2.0\Gamma$, $\Delta = -1.7\Gamma$, agrees with the $12T_{Dop}$ from the experiment.

From the above argument, the temperature should not be affected by the field gradient. Also from Eq. 2.8, for $\Omega = 2.0\Gamma$, the temperature varies from $9T_{Dop}$ to $7.5T_{Dop}$ for detunings between -1.7 to -2.5 linewidths, which is within the experimental precision. Fig. 2.4 shows the result. The larger width in the top figure of Fig. 2.4 is because the detector is further away from the trap center (0.95 mm).

2.3 Number of atoms in the trap

Now, to measure the number of trapped atoms, one method is to take a fluorescence measurement, which determines the excited state number of atoms. The fluorescence is detected by a calibrated low noise, photodiode detector. (Hamamatsu S2387-06R photodiode, sensitivity 0.43 A/W. Detector amplifier bandwidth 20 Hz, detector sensitivity $2.5 \times 10^{-7}$ V/W). Amplifier circuit is included in Appendix C. Fig. 2.5 is the steady state fluorescence signal and background level. The time of the scan is 18 seconds, and the signal-to-noise level is over 10:1. The calibration factor is $2.1 \times 10^{6}$ excited state number of atoms per volt. A typical number is $10^{6}$ atoms in the excited state and $10^{7}$ total in the trap, depending on the detuning. The thermal noise (Johnson noise) and quantum shot noise in the detector amplifier are negligible as indicated by the background level. The major noise source has been the instability of the laser intensity, mostly fluctuations in long time scale (1 second), which causes fluctuations in the fluorescence signal. The room light was blocked out by a bandpass filter (centered at 671nm, filter ratio 1000:1, attenuation coefficient 0.5) as well as by wrapping and sealing the detector.
Figure 2.5  Fluorescence signal and background level vs time.

Because of the hyperfine splitting in the ground and excited states as in Fig. 1.4, lithium is a multilevel system. The excited state fraction $\rho_{ee}$ cannot be determined accurately due to the different detunings to the excited hyperfine states, and thus the total number of the atoms in the trap is uncertain.

To accurately determine the total number, we used absorption spectroscopy. We took an absorption probe and passed it through a spatial filter to obtain good spatial mode quality. It is then focused down to a 120\(\mu\)m beamwaist and passed through the center of the trap. The probe power is set at about 30 nW (Rabi frequency $\sim 0.1\Gamma$) to avoid any mechanical disturbance on the trapped atoms. Good alignment was ensured by tuning the probe to resonance and pushing the atoms out of the trap. The probe was detected by a photomultiplier tube (Thorn EMI 9828B, gain $1.0 \times 10^4$). Simultaneously, a second reference beam, derived from the same laser was sent directly to a photodiode, so that the intensity fluctuation of the probe laser can be divided out. Because of the relatively large number of atoms in the trap, the fluorescence
from the trapped atoms significantly increased the noise level, as did the scattering of the trapping beam from the chamber internal walls. In some cases the absorption was only 2-3% and a direct absorption measurement while the trapping laser was on was almost completely masked by the noise and thus no accurate measurement could be achieved. Therefore, we electronically integrated the signal and turned off the trapping laser simultaneously at the start of the integration period to enhance the signal-to-noise level.

![Graphs showing absorption signal for transitions](image)

**Figure 2.6** Absorption signal for $2S_{1/2}$ to $2P_{3/2}$ transition. Top: $F=1$ to $2P_{3/2}$. Bottom: $F=2$ to $2P_{3/2}$. 
The timing of the measurement is as follows: The trapping beam was chopped on and off rapidly by turning on and off the deflection AOM. The trap lifetime is on the order of seconds, so the momentarily switching off (50 μs off time, 500 μs on time) had little effect on the trap dynamics but trapped about 90% as many of atoms as without turning it off. After the short period of turning off (~5μs, because of the delay in AOM), the integrators were turned on by a CMOS switch, so the absorption and reference probes were integrated (during 1 gate-on time) and accumulated over the preset number of gates, and then recorded by the computer. The computer then reset the integrator and incremented the probe laser frequency and the same processes were repeated. At the time the absorption measurement was taken, the trapping beam was turned off, so the scattering from the trapping beam and the fluorescence from the atoms were essentially eliminated, and we attained a high signal-to-noise level. The circuit diagram and timing schematics are included in Appendix C.

Fig. 2.6 shows a typical absorption signal for the probe laser tuned closely to ground state $F = 1$ and $F = 2$ to $2P_{3/2}$ transitions, respectively. The FWHM on both spectrum is 20 MHz and the kink structure on $F = 2$ spectrum is 13 MHz to the blue of the peak position. The width and the small kink structure are due to the hyperfine structure of the $2P_{3/2}$ excited state.

The partial cross section for each of the dipole transitions between ground and excited hyperfine states is: [Sobelman, 1991].

$$
\sigma(F \rightarrow F') = \frac{\lambda^2}{2\pi} \frac{1}{(2F' + 1)(2J_u + 1)} \frac{(\Gamma/2)^2}{(\omega_l - \omega_u)^2 + (\Gamma/2)^2} \left\{ \begin{array}{ccc} J & F & I \\ F' & J' & 1 \end{array} \right\}^2
$$

where $J_u$ is the angular momentum of upper state, $I = 3/2$ is the nuclear spin and $F, F'$ is the total angular momentum for lower and upper states, respectively. The calculated partial cross sections are shown in Fig. 2.7. The total cross section for the $F=1$ and $F=2$ transitions is the summation of all the possible hyperfine transitions.
Figure 2.7  Partial cross sections (in $\lambda^2/4\pi$) for indicated dipole transition. Left: $2S_{1/2}$ to $2P_{3/2}$ transition. Right: $2S_{1/2}$ to $2P_{1/2}$

The width (FWHM) from the calculated total absorption cross section is 15 MHz and the kink structure is 11 MHz to the blue of the peak absorption from Fig. 2.8. The Doppler broadening at trap temperature of 1.7 mK is

$$\delta_D = 2\sqrt{\ln 2} \frac{k}{2\pi} \sqrt{\frac{2k_BT}{m}} = 5.0 \text{ MHz}$$  \hspace{1cm} (2.10)

where $k$ is the wave vector. Clearly, the Doppler broadening in the trap is negligible and the transition is predominated by the hyperfine structure. From Fig. 2.8, we see $F = 2$ transition has a well resolved absorption peak, which corresponds to $F = 2$ to $F' = 3$ on-resonance transition. It is possible to precisely lock the laser to this $F' = 3$ hyperfine state although it's more difficult than Cs which has a well resolved hyperfine structure.
Figure 2.8  Calculated total cross section vs frequency for $2S_{1/2}$ to $2P_{3/2}$ transition: Top: $F=1$; Bottom: $F=2$.

Besides enhancing the signal-to-noise level by turning off the trap, another advantage is that this is a direct measurement of the total number of atoms in the trap, since for the off period of 50\(\mu\)s, the excited state atoms (lifetime 27.29 ns) have all decayed back to the ground states. The total number of atoms is obtained as following. At the peak absorption frequency, the absorption coefficient $\alpha$ is (probe along $y$
direction):

\[ \sigma(N, x, z) = \int_{-\infty}^{\infty} \sigma_m n(x, y, z) dy \]
\[ = \frac{\sigma_m N}{\pi w_x w_z} \exp \left[ - \left( \frac{x^2}{w_x^2} + \frac{z^2}{w_z^2} \right) \right]. \quad (2.11) \]

\( w_x, w_z \) are the cloud Gaussian radii in the \( x \) and \( z \) directions, \( \sigma_m = 2.34\lambda^2/\pi \) for \( F = 1 \) and \( 2.86\lambda^2/\pi \) for \( F = 2 \) transition is the peak absorption cross section. The transmitted intensity is

\[ I_f(x, z) = I_0(x, z)e^{-\alpha}. \quad (2.12) \]

The transmitted power is the integration over the probe Gaussian intensity distribution:

\[ P_f = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} I_f(x, z) \exp \left[ - \frac{2(x^2 + z^2)}{w_p^2} \right] dx dz. \quad (2.13) \]

In the linear region, the integration of \( I_f(x, z) \) over the cloud is expressed as:

\[ \frac{\Delta P}{P_o} = \frac{P_f - P_o}{P_o} = \frac{N \sigma_m}{\pi w_x w_z} \quad (2.14) \]

for a probe beam waist much smaller than cloud radii.

The total number of trapped atoms is around \( 10^7 \) and the highest density observed was \( 2 \times 10^{10} \) atoms/cm\(^3\) in the cell MOT. We once measured the number of trapped atoms and the density for various trapping conditions. We found that the density is quite insensitive to the detunings from -2 to -6 linewidths (within three times and is not monotonic), since \( N_e \) increases with detuning at this range but cloud radii decrease. However, at pressure \( \leq 8 \times 10^{-9} \) torr, \( N_e \) increases monotonically with cell temperature (i.e. pressure), and approaches to a constant. This is because, at low pressure, the background collision is due to both non-Li species and hot Li, whereas at higher pressure, \( \gamma_b \) is mostly due to hot Li background gas. Since \( L \) and \( \gamma_b \) are both linear with \( n_e \) (Li density in the cell), \( N_e \) is independent of \( n_e \). For a certain detuning,
there is an optimal field gradient where we can trap most atoms. For this detuning range (-2 to -6 linewidths), it increases with detuning but is around 20 Gauss/cm.

We also did \(2S_{1/2}\) to \(2P_{1/2}\) absorption in the beam MOT described in the next chapter. The hyperfine splitting for \(2P_{1/2}\) is simpler and larger (91.8 MHz) and is well within the experiment frequency resolution. The partial cross section for the transition is also shown in Fig. 2.7. Fig. 2.9 is the absorption signal.

Figure 2.9  Absorption signal for \(2S_{1/2}\) to \(2P_{1/2}\) transition. Top: \(F=1\) to \(2P_{1/2}\). Bottom: \(F=2\) to \(2P_{1/2}\).
The uncertainty of the absorption measurement comes from several sources. One is the uncertainty of the alignment of the probe, which is reduced by improving the mode quality and focusing the beam down to a small beamwaist and by pushing the atoms further away from the trap. We repeatedly aligned the probe several times, and the variation of the absorption signals is within 20%. Within each absorption scan, the precision is limited by the frequency resolution. The resolution of 2 MHz is a compromise between the length of the scan period (each data point is about 0.5 sec, so one scan is about 1 min) and reasonable resolution. In the worst case, where the peak absorption lies halfway between two adjacent data points, the error is 15% in the $F = 2$ to $2P_{3/2}$ transition. The other type of error comes from the parameters in modeling absorption over a Gaussian density distributed cloud. The important parameters are the Gaussian radii of the cloud. The uncertainty in measuring the cloud radii is ±1 pixel in the CCD camera giving a relative error of 5%. Therefore, from Eq. 2.14 the relative error in the number of atoms is

$$\frac{\Delta N}{N} = \frac{\Delta S}{S} + \frac{\Delta w_z}{w_x} + \frac{\Delta w_z}{w_z} = 25\%$$  \hspace{1cm} (2.15)

where $S = \Delta P/P_0$.

### 2.4 Density of trapped atoms

As discussed in the earlier section, the density is a Gaussian distribution. The peak density is

$$n_o = \frac{N_s}{\pi^{3/2} w_x w_y w_z}.$$  \hspace{1cm} (2.16)

From the one-dimensional model

$$n_o \propto \frac{d^2 v_x^4 m^2}{\sigma_T c^2} \cdot \left( \frac{K}{2k_B T} \right)^{3/2}$$  \hspace{1cm} (2.17)

where $T_c$ is the cell temperature. The density is an important parameter in collision study. At low density, it is limited by the background collision rate for certain trapping
conditions. However, at high density it will be limited by the cold-cold atom collision and the photon pressure of the re-scattered photons between trapped atoms. I will discuss this in more detail in the next chapter.

2.5 Dynamical parameters

![Graph](image)

**Figure 2.10** Trap loading rate and time constant measurement. Dotted line is the data and solid line is the fitted curve.

The trap dynamical parameters are the trap loading rate $L$, which is determined by the capture velocity and trap size as discussed in Chap 1, and the trap lifetime $\tau = 1/\gamma_b$, where $\gamma_b$ is the background collision rate. The fluorescence signal was monitored when the trapping beams were instantly chopped on. Fig. 2.10 is the data taken at a trap detuning -5.0 $\Gamma$ and cell pressure $3.0 \times 10^{-9}$ torr. The number of atoms in the trap is $N_s = 1.7 \times 10^7$, and the density is $2.2 \times 10^{10}$ cm$^{-3}$. The data was fit to the solution of the rate equation (Eq. 1.6).

From the fit, the time constant $\tau$ is 1.05 s, the background collision rate $\gamma_b = 1/\tau = 0.95$ s$^{-1}$. The loading rate $L = N_s \gamma_b \sim 1.6 \times 10^7$ s$^{-1}$. The good exponential fit
indicates the cold-cold collision rate is small compared with the background collision. From Eq. 1.7 and 1.8,

$$
\gamma_b(P) = \frac{11.7P\sigma_c}{\sqrt{\log P + 31.9}} \times 10^{21}
$$

(2.18)

where $P$ is in torr, $\sigma_c$ in cm$^2$, $\gamma_b$ in second. The measured $\gamma_b$ vs $P$ is fit to above equation as shown in Fig. 2.11, and the total collision cross section $\sigma_c$ (including collisions with non-Li species and hot Li background) is found to be $9 \times 10^{-14}$ cm$^2$.

Here, I assume at pressure $\geq 10^{-9}$ torr, the pressure is entirely due to Li vapor. This result is of the same order of magnitude of Cs, which is measured to be $2 \times 10^{-13}$ cm$^2$ [Monroe, 1990].

The average capture velocity from Eq. 1.9 is 73 m/s. The calculated 1D capture velocity from the equation of motion (Eq. 1.4) is 63 m/s. The trap depth, however is not defined by the capture velocity. Instead, we define the escape velocity $v_e$, which is the maximum velocity required to escape from the trap center. The trap depth is
Figure 2.12 Trap depth as functions of trapping conditions from the 1D model. Top: trap depth vs laser detuning for various magnetic field gradients, $\Omega = 1.0\Gamma$. The solid lines are the spline interpolation fit to the calculated points. Bottom: trap depth vs Rabi frequency for various detunings. $b=20$ Gauss/cm. The dotted line corresponds to 0.24 K, the trap depth where fine structure collision channel is turned on. See next chapter.

$$T_D = \frac{1}{2}mv_z^2/k_B. \quad (2.19)$$

The trap depth depends on the trap conditions similar to the capture velocity. The variation of trap depth with the detuning and magnetic field is calculated again by solving the equation of motion for the initial conditions $z = 0$ and $\dot{z} = 0$. The results are shown in Fig. 2.12.
The trap depth and its dependency on trapping conditions are essential in later collision experiments. The collision rate and the dominant collision mechanism varies significantly with trap depth. We also define the escape energy to be two times the trap depth for later reference: \( E_{\text{es}} = 2k_B T_D \).
Chapter 3

Ultra-Cold Atom Collision Rate Coefficient Measurement of Lithium

There are several distinct collisional processes occurring in the trap which result in atoms being lost from the trap. The first is the collision of cold atoms with hot background gas, which is the primary trap loss mechanism presently in our trap. However, at higher density, cold-cold atom collisions are expected to be the dominant processes and will be a limiting factor for very high density atom traps. Together with the interesting, challenging properties of cold-cold atom collisions, such as long de Broglie wavelengths at low temperatures and the strong effect of spontaneous radiation because the collision time is comparable to the excited state lifetime, the subject has aroused intense study in both theoretical and experimental work. The work that has been done in Na [Prentiss, 1988] (beam MOT), $^{85}$Rb [Hoffmann, 1992] and $^{87}$Rb [Feng, 1992] (cell MOT), $^{85}$Rb and $^{87}$Rb [Wallace, 1992] (beam MOT), Cs [Sesko, 1989] (beam MOT) are in agreement to some extent with the theoretical model by Gallagher and Pritchard [1989], and as refined by Julienne and Vigué [1991], though there is still a lack of enough experimental data to satisfactorily compare with theory. There are also other proposed limiting factors for achieving high density such as photon pressure resulting from the scattering of photons from one atom to another in the trap as observed by Walker et al [1990].

Our motivation in studying Li collisions is based on the theoretical study that predicted at least a 10 times smaller collisional loss rate than that of the other alkali
species [Julienne, 1991]. If this holds true, Li is potentially the most suitable candidate for a high density source for other studies, such as Bose-Einstein condensation, alkali metallic clusters or technically, diatomic excimer lasers in the blue-green wavelength region. Secondly, the hyperfine splitting of Li is 800 MHz in the $2S_{1/2}$ ground state, and only 20 MHz and 90 MHz in the $2P_{3/2}$ and $2P_{1/2}$ excited states, respectively. Neglecting the hyperfine structure in most theories because of its complexity is most likely valid in Li. In other words, Li provides a suitable system to compare with presently available theory. Thirdly, in $^7$Li MOT, we found that we are able to separate the two proposed kinds of mechanisms (fine structure changing and radiative escape) and demonstrate to measure them separately. This is the first time in experiment to separate these two mechanisms and directly compare with the theory.

3.1 Collisional processes in the MOT

To begin with, I would like to examine closely what are the collisional processes in an alkali atom trap. Besides collisions with hot background atoms, there are two kinds of cold-cold collisional processes that contribute to trap loss: fine structure (FS) changing collision and radiative escape (RE) process. The collisions are two body molecular processes involving one ground $S$ state atom and one excited $P$ state atom. The long-range potential for the $S + P$ excited state is the $R^{-3}$ dipole-dipole interaction, whereas it is a $R^{-6}$ van der Waals interaction for the $S + S$ ground state. Obviously the ground state potential is negligible in the long range region compared with the excited states.

For all the alkali species, the FS changing process arises from the significant fine structure splitting in the $P$ excited state. As illustrated in Fig. 3.1 for a generic alkali element, if the excited atomic pair of $S + P$ are drawn close enough by the $R^{-3}$ interaction to reach the small-$R J$-crossing region and undergo a non-adiabatic curve
crossing, the atomic pair will gain a kinetic energy equal to the energy difference of $P_{3/2}$ and $P_{\frac{1}{2}}$, and can escape from the trap if the energy is greater than the escape energy (defined at the end of Chap 2). This is the fine structure changing collision (also known as a $J$-changing collision). The process can be expressed as:

$$A(S_{\frac{1}{2}}) + A(S_{\frac{1}{2}}) + \hbar \omega_{\frac{1}{2}} \rightarrow A(S_{\frac{1}{2}}) + A(P_{\frac{1}{2}}) = A^2(S_{\frac{1}{2}} + P_{\frac{1}{2}})$$

$$\rightarrow A(S_{\frac{1}{2}}) + A(P_{\frac{1}{2}}) + \Delta_{FS}$$ (3.1)

The other process is the radiative escape (RE), similar to the FS changing in the molecular motion but due to a different nature.

$$A(S_{\frac{1}{2}}) + A(S_{\frac{1}{2}}) + \hbar \omega_{\frac{1}{2}} \rightarrow A(S_{\frac{1}{2}}) + A(P_{\frac{1}{2}}) = A^2(S_{\frac{1}{2}} + P_{\frac{1}{2}})$$

$$\rightarrow A(S_{\frac{1}{2}}) + A(S_{\frac{1}{2}}) + \hbar \omega$$ (3.2)

The re-emission by the $A_2$ molecule of a photon $\hbar \omega$ with $\hbar \omega < \hbar \omega_{\frac{1}{2}}$ for an attractive potential can lead to escape if $\hbar (\omega - \omega)$ is greater than the escape energy. The RE process clearly depends on the trap depth. It is also illustrated in Fig. 3.1.
The dominant feature of these cold-cold atomic collisional processes is two-fold: 1) Both $J$-changing and RE processes occur in the small-$R$ region while the formation of the collision pair is at large atomic separation (for a small red-detuned trapping laser). Because of the slow thermal velocity at the trap temperature, the duration of collisional processes can be comparable to the excited state lifetime. The atomic pair needs to survive before a spontaneous decay in order to reach the collision region. The collisional processes are therefore greatly modified and complicated by spontaneous emission because of this survival factor. It is possible that this survival factor requires a quantum treatment since the results from a fully quantum mechanical calculation [Boeselten, 1993] showed some different features from that of the classical motion along the trajectory.

2) The Li de Broglie wavelength in a 1 mK trap is $\lambda_D \simeq 400 \text{Å}$, comparable to the effective scattering length. The consequence is that a quantum treatment of the collisional processes is essential. This is especially true for Li because of the small mass and therefore, large $\lambda_D$. The number of the partial waves can be estimated by setting the collision kinetic energy $3k_BT/2$ equal to the height of the centrifugal barrier for $C_3/R^3$ potential.

$$l_{\text{max}} = \left( \frac{81\mu^2 C_3^2 k_BT}{\hbar^6} \right)^{1/6}$$

(3.3)

where $\mu$ is the reduced mass and $T$ is the trap temperature. $l_{\text{max}}$ is 13 for Li, whereas it is 26 for Na and 76 for Cs for the same collision channel $(0^+_1)$. The relative small number of partial waves requires a quantum collision theory for Li for a better understanding of the process.

From now on, I shall concentrate on the collisional processes in Li only. The analysis is similar for all the alkali species, though the dominant process and the collision cross section can vary significantly among them.
3.2 Molecular physics of lithium dimer

As mentioned before, both FS changing and RE process are small-$R$ region processes, where it is imperative to understand the molecular physics in these processes. I summarize some terminology and what we know about the Li dimer in the rest of this section.

First since I will frequently refer to the coupling Hund’s cases, a brief outline of these cases is as follows: Various couplings between electronic spin and orbital motion with the rotating molecular axis are categorized by Hund’s cases [Herzberg, 1950]. Case (a) is where the rotational motion of the molecular axis is small and the coupling between the spin and the axis is dominant, as most often found in stable molecules. Case (b) is the opposite to case (a), where the rotational motion is important as in light molecules (very likely in Li$_2$) because of the large rotational energy splitting for small moment of inertia. Case (c) is dominant in long range molecules, where the coupling to the molecular axis is so weak that the electronic spin and orbital coupling is predominant as in most alkali species (strong spin-orbital coupling).

The electronic structure and spectra of Li$_2$ has long been closely studied. Various methods have been developed and employed to numerically compute the long range electronic states as well as the molecular states potential curves for low-lying electronic states [Konовалow, 1979], [Konovalow, 1983], [Bussery, 1985]. The molecular potential curves from Schmidt-Mink et al [1985] are shown in Fig. 3.2. Two molecular states arise from $2S + 2S$ asymptote: $^1\Sigma_g^+(X)$ and $^3\Sigma_u^+(a)$. The long range van der Waals coefficient for $2S + 2S$ is $1.385 \times 10^3$ a.u.. The energy shift due to this interaction term is less than 1 MHz for $R > 200a_o$ ($a_o$ is the Bohr radius). I shall neglect this term hereafter.
Figure 3.2 Molecular potential curves for states correlated to $2S + 2S$ and $2S + 2P$ asymptotes. The ungerade states are shown in dotted line. [Schmidt-Mink, 1985]

The $2S + 2P$ asymptote on the other hand give rise to 8 molecular states: $^1\Sigma_u^+(A), ^3\Sigma_d^+, ^1\Pi_g, ^3\Pi_u(b), ^1\Pi_u(B), ^3\Pi_u, ^1\Sigma_g^+(A), ^3\Sigma_u^+$. The dipole-dipole $C_3$ coefficients is 5.51 a.u. [Schmidt-Mink, 1985]. The long-range potential (determined from $C_3, C_6, C_8$ are shown in Fig. 3.3 [Konowalow, 1983]. The pair with configuration of $^1\Lambda_i$ and $^3\Lambda_j$, where $i \neq j \in (g,u)$ has similar long range dependence. The interaction in the same pair is different at small-$R$ due to the exchange interaction. The long range potentials are symmetric with respect to the $E = 0$ line. Therefore 4 states out of the 8 are attractive and 4 are repulsive at long to intermediate range.

By diagonalizing the Hamiltonian matrix which includes the long-range non-relativistic potentials and the relativistic spin-orbit coupling terms, the long range potentials are correlated with the specific $2P_j + 2S_{1/2}$ asymptote [Dashevskaya, 1969]. The enlarged correlation is illustrated in Fig. 3.4.
Figure 3.3  Molecular potential curves for states correlated to $2S + 2S$ (top) and $2S + 2P$ (bottom) asymptotes. Notice $2S + 2S$ is much flatter in this region compared with $2S + 2P$. The small fine structure splitting (0.34 cm$^{-1}$) is not shown.

In Hund's case (c) $\Omega_{g/u}^{\lambda}$ representation, there are six molecular states: $0^+_g$, $0^-_g$, $0^+_u$, $0^-_u$, $1_g$, $1_u$ disassociating to $2S + 2P_{1/2}$ and ten molecular states: $0^+_g$, $0^-_g$, $0^+_u$, $0^-_u$, $2 \times 1_g$, $2 \times 1_u$, $2_g$, $2_u$ dissociating to $2S + 2P_{3/2}$. Crossings occur at large separation due to $0^+_g$ of $2S + 2P_{1/2}$ with $0^+_u$, $1_g$, $2_u$ of $2S + 2P_{3/2}$ and at a smaller region due to $1_u$ of $2S + 2P_{1/2}$ with $0^+_u$, $2_u$, $1_g$ of $2S + 2P_{3/2}$. These crossings are the possible FS transition channels.
3.3 Collision mechanism and rate coefficient

3.3.1 RE process

Among the two processes, the RE mechanism is fairly straightforward. In Fig. 3.2, six molecular states are attractive in the small-$R$ region. However, the attractive $^1\Pi_u(B)$ and $^2\Sigma^+_g$ have slight repulsive walls at intermediate- and large-$R$ region. The barrier for the shallower $^1\Pi_u(B)$ is 0.002 a.u. = 600 K, much greater than the atomic kinetic energy in the trap. Therefore they cannot be collision channels for RE process. The other four molecular states in the small-$R$ region correlate to eight asymptotic states in the long range region, where five of them are correlated to the...
$2S + 2P_{3/2}$ asymptote but $2_u$ is forbidden dipole transition to ground state. The possible collision channels with bounding energy greater than the escape energy ($\sim 1$ K in our setup) so that both atoms can escape from the trap are $0_u^+$ and $1_g$.

The collision rate coefficient calculation has been carried out very nicely in Julienne and Vigué [1991]. I hereby outline the physical picture and the results so as to apply to Li in our experiment and compare with the experimental results later in this chapter.

1) The optical transition to the excited state is treated semi-classically. The excitation rate depends on the separation $R_o$ of the atomic pair.

$$\varepsilon(R_o, \Delta) = \frac{\Gamma_M^2(\beta)}{4 \left( \Delta + \frac{\mathcal{C}_3}{R_o^3} \right)^2 + \Gamma_M^2(\beta)} \times \left( \frac{\Gamma_{\sigma o}}{\hbar \omega_1} \right)$$ (3.4)

where $\Delta, \omega_1, I$ is the trapping laser detuning, frequency and intensity. $\beta = 0_u^+$ or $1_g$ is the collision entrance channel, $\sigma_o$ is the on-resonance atomic absorption cross section. The factor 8 comes from the 2 entrance channels out of 16 possible ones.

2) The motion of the excited atomic pair under the $-\mathcal{C}_3/R^3$ interaction, is treated classically. The excited atomic pair is drawn closer by the attractive interaction. The probability of reaching the $R \leq R_E$ without any spontaneous decay back to the ground state is given as a survival factor:

$$S(R_o, \beta) = \exp \left( - \int_{R_o}^{R_E} \Gamma_M(\beta) dt \right)$$ (3.5)

where $\Gamma_M(\beta)$ is the molecular spontaneous decay rate for channel $\beta$ and

$$R_E = \left( \frac{\mathcal{C}_3}{2k_BT_D} \right)^{1/3} \approx 60 \text{Å}$$ (3.6)

for Li at 0.3 K trap depth. The survival factor is determined by the time it takes to travel from the excitation position $R_o$ to the inner zone $R \leq R_E$ where spontaneous emission within this zone transfers enough energy to the atoms to escape from the trap, and is therefore dependent on $R_E$, which in turn depends on trap depth.
3) Once the excited atomic pair reaches the inner zone, the probability of undergoing a spontaneous emission is calculated by the time it spends in the inner zone over the molecular excited state lifetime:

\[ P(\beta) = c(\beta) \frac{\Gamma_M(\beta)C_3^{1/3}\mu^{1/2}}{(2k_BT_D)^{17/8}} \]  \hspace{1cm} (3.7)

where \( \mu \) is the reduced mass, \( c(\beta) \) is an integration constant. The probability is strongly related to the trap depth due to both the survival factor and the molecular excited state lifetime. Li has a stronger trap depth dependency than other alkali elements because of the additional lifetime dependency on the interatomic distance. For a deep trap, the effective spontaneous emission is further into the smaller-\( R \) region, where the decay rate is smaller for these two collision channels.

4) Finally the number of ground state pairs per unit volume between \( R_o \) and \( R_o + dR_o \) which approach each other with thermal energy between \( E \) and \( E + dE \) and with angular momentum \( l \leq l_{\text{max}} \) is \( dN(R_o, E, l) \). The collision rate coefficient \( K_{RE} \) is the integration of \( S(R_o, \beta)P(\beta)c(\Delta, R_o)dN(R_o, E, l) \) averaged over the Maxwell-Boltzmann distribution:

\[ K_{RE} = \text{const} \cdot \frac{\Gamma_A C_3}{(2k_BT_D)^{17/6}(k_BT)^{1/6}}. \]  \hspace{1cm} (3.8)

The calculated Li RE trap loss for 1 K trap depth and 1 mK trap temperature is \( 4.5 \times 10^{-14} \text{ cm}^3/\text{s} \) at \( \Delta = -1.0 \Gamma_A \) and \( I = 10 \text{ mW/cm}^2 \).

The rate coefficient from above equation again shows a strong dependence on the trap depth. Because the trap depth is dependent on the trapping conditions (laser intensity, detuning and magnetic field gradient), the rate coefficient will also be varied with these parameters. The change of the intensity will change the excitation rate as well as the trap depth. The changes are independent in the theory. This gives us the way to scale the theoretical prediction to the experimental trapping condition to compare with the experimental results as will be done in the next section. The magnetic
field gradient will have an effect on the trap depth only. The detuning dependence is more complicated because of different Condon points for different detunings and excitation rate as well as the trap depth. This dependence has not yet been fully addressed in this model.

3.3.2 FS process

Historically, the fine structure changing mechanism was examined closely by a group of Russian scientists 20 years ago in connection with the experimental studies of alkali atom collisions by a Canadian group [Dashevskaya, 1969], [Dashevskaya, 1979], [Nikitin, 1975](theory); and [Czajkowski, 1965], [Rae, 1965] (experiment). The work being done is at room temperature or higher, nonetheless provides basic insights and guidelines to the problem.

The FS collisional process is introduced as a trap loss by Gallagher and Pritchard [1989]. The fine structure splitting of alkali atoms except Li is hundreds times the trap depth. It is believed to be one of the most important trap loss mechanisms in an alkali-metal trap. For Na and K, the calculation shows a 20 times larger than the RE process. In Li, however, the fine structure splitting (10 GHz = 0.48 K) is comparable to the escape energy. Therefore the FS changing channel can be purposely turned on or off as we change the escape energy by varying the trapping conditions: laser intensity, detuning and magnetic field gradient. In this way, we can directly measure the pure RE (assume other collision channels are negligible) at large trap depth or FS plus RE in a shallow trap. The RE rate should be deductible from the previous information so that the rate for FS may be measured independently.

The fine structure changing mechanism, as I mentioned before, can be understood from the long range correlation and curve crossings. In Fig. 3.5, I outline the correlation between the long range asymptotes and the intermediate range $\Omega_{g/u}$
representation and the short range $\Lambda_{g/u}$ representation from previous figures. The 8 molecular states are grouped to 4 pairs with each pair having the same intermediate dependency. The $\Lambda_{g/u}$ representation is related to the $\Omega_{g/u}$ representation only with the same $g/u$ inversion symmetry.

There are 6 possible crossings in the intermediate range as indicated by the dots in the graph. The selection rules for the $J$-changing is:

$$g \leftrightarrow g, \ U \leftrightarrow u, 0^+ \leftrightarrow 0^+, 0^- \leftrightarrow 0^-; \Omega = 0, \pm 1. \quad (3.9)$$

since the transition (not an optical transition) should not change the symmetry of the states. So the possible transition channels are the $0^+_g$ of the $^3\Pi_u$ with $1_g$ of the $^1\Pi_u; 1_u$ with $2_u$ or with $0^+_u$ all of the $^3\Pi_u$.

Dashevskaya [1969] demonstrated that the first is negligible because of the vanishing transition matrix element $\langle 0^+_g | J_z | 1_g \rangle$ at both long range and short range.
The other crossings $1_u$ with $0_u^+$ and $1_u$ with $2_u$ are states that corresponding to the same state $^3\Pi_u$. The matrix elements are calculated to be considerable because of the Coriolis interaction between the vibrational and rotational motion within that state [Landau, 1969]. These two crossings are responsible for transitions in the intermediate range.

There are 8 more crossings which occur in the inner molecular zone ( $R_o < 15a_o$ for Li in Fig. 3.2). After eliminating the crossings that are from different symmetry (i.e., crossings of g state and u state) as well as those are not reachable for the trap thermal energy, we identify the crossing of $0_u^+$ of $^3\Pi_u(b)$ and $^1\Sigma_u^+(A)$ as a possible collision channel.

In summary there are three crossings that contribute to the FS changing process: 1) $0_u^+$ and $1_u$, 2) $2_u$ and $1_u$ and 3) $0_u^+$ of $^3\Pi_u(b)$ and $^1\Sigma_u^+(A)$. The first two occur at $20 \sim 60a_o$ and $20 \sim 40a_o$ for alkali species, respectively. There is no solid calculation for Li because of the small fine structure splitting. In all the theoretical calculation these crossings are approximated to occur at $R_o = 0$ because of the fast motion in the inner molecular zone.

The details of the computation involved is similar to the RE process. The transition probability is evaluated by using the interaction matrix element at the crossing point of the non-relativistic potentials and either the Landau-Zener formula for curve crossing transition or quantum mechanically partial wave integration.

The results in Julienne and Vigué show that the contribution from the two channels $0_u^+$ and $2_u$ vary significantly among the alkali elements. The lighter elements Na and K have 99% of the FS rate from the $2_u$ entrance channel since spontaneous decay rate from this state is small as it has dipole forbidden transition to the ground states. The smaller decay rate results in a larger survival factor, giving it a large fraction of collision rate coefficient. In the heavy species Rb and Cs, $0_u^+$ channel is also important
(more important than $2_u$ in Cs) because of the large spin-orbital interaction. Because of the relatively shorter lifetime of this state, Rb and Cs have smaller FS changing coefficient than Na and K.

Similar calculations have not yet been done on Li. Lithium RE process is estimated much smaller than the other alkali elements because of the small decay rate of $0_u^+$ and $1_g$ for the $R$ range of interest ($R_F \approx 60$ Å). However, for the very same reason, the FS of Li can be considerably large because of the small spontaneous decay rate for both $0_u^+$ and $2_u$ channels at the small $R$ range. In the experiment, we did see a comparable FS collision rate in comparison with other elements. In analogy with Na and K, the major contribution will be from $2_u$ channel. It will be very interesting to carry out the calculations in above picture.

A simplified calculation suggested in Gallagher and Pritchard was carried out for Li at 10 mW/cm$^2$ total intensity into the trap. The details are included in the Appendix B. Fig. 3.6 shows the results. The transition probability for a round-trip curve crossing is 0.02 from Julienne and Vigué. It is smaller than the probability at the room temperature because of the slow atom collisions. The coefficient is on the order of $3 \times 10^{-11}$ cm$^3$/s at the peak, much larger than the RE process at the same laser intensity ($4.5 \times 10^{-14}$ cm$^3$/s from previous subsection).

The FS coefficient is expected to be a linear function of the laser intensity at small Rabi frequency (negligible power broadening). The coefficient should be relatively constant with respect to various magnetic field gradients (when the channel is open), assuming the transition mechanism is not affected by the magnetic field. The detuning dependence is determined by both the absorption cross section and the survival factor. At large detuning, closer atomic pairs are excited because of the energy shift of $C_3/R^3$. The time is less for this atomic pair to reach the small-$R$ collision region, so the survival factor is larger. However, the number of collision partners and the excitation
rate are smaller. The combination results in a peak coefficient at some intermediate detuning. The peak for Li is anticipated to be at -20 MHz from this model.

3.4 Experimental results

3.4.1 RE rate coefficient

We proceeded in measuring the cold-cold collision coefficient in a beam MOT. The cold-cold collisional processes are incorporated in the trap as a trap loss mechanism. The rate equation taking into account these two body processes is:

\[
\frac{dN(t)}{dt} = L - \gamma_b N(t) - \beta^* \int n_p(r,t)n_e(r,t)d^3r = L - \gamma_b N(t) - \beta \int n^2(r,t)d^3r
\]  

(3.10)

where \(n(r,t)\) is the trapped atom density, \(\rho\) is the excited state fraction, \(\beta = \beta^*\rho(1-\rho)\) is the total collision rate coefficient. The \(N(t)\) from this equation will deviate from an exponential because of the non-linear term \(n^2(r,t)\).
The experiment was eventually realized in a beam MOT trap. The previous cell MOT setup has the drawback that the cell needs to be running at high temperature to achieve the necessary density, which increased the cell pressure and thus the background collision rate. The non-exponential cold collision term was not large enough compared to the background collision to observe the effect. In the beam MOT, on the other hand, the trap region is separated from the hot oven. The experimental setup is shown in Fig. 3.7. We utilized the available laser cooling techniques in this lab, namely relay chirp cooling [Bradley, 1992] to laser cool the fast atoms from the Li effusive source oven. This technique, uses a travelling wave EOM to sweep the laser frequency to compensate the changing Doppler shift as atoms are slowed down, so the atoms are always in close resonance with the laser. The MOT is placed \( \sim 0.65 \)
m away from the oven. The unslowed atomic flux at a 600°C oven temperature with 0.5 × 1 mm² oven aperture is:

\[ \Gamma = \frac{1}{4} n \bar{v} A \Omega = 3.7 \times 10^{12} \text{ atoms/s.} \quad (3.11) \]

\( \Omega \) is the solid angle of the trap region (0.65 m away from the oven and trapping beam diameter 1.5 cm). The loading rate calculated from the number of trapped atoms and the background collision rate is \( L = 1.6 \times 10^7 \text{ s}^{-1} \) (Chap 2). The loading efficiency including the chirp cooling is roughly 5 parts per million. The total number of trapped atoms is no longer defined by the cell pressure. As we optimize the loading (chirp cooling power, chirp frequency offset and rf power), we can obtain \( 10^8 \) atoms maximum.

The loss coefficient is obtained from measuring the decay of the trap. After filling the trap, the atomic beam is blocked using a mechanical beam block. Simultaneously the chirp cooling beam is turned off by the AOM chopper in the cooling path. The decay fluorescence signal was monitored by the photodiode. Fig. 3.8 is the decay signal for \(-2.4\Gamma\) detuning and 85 mW/cm² laser intensity. The data is fit to the solution of the rate equation at \( L = 0 \). The integration of \( \alpha^2(r,t) \) is carried out by assuming a Gaussian density distribution with constant radius. The rate equation is therefore:

\[ \frac{dN(t)}{dt} = -\gamma_0 N(t) - \frac{\beta}{V_c} N^2(t) \quad (3.12) \]

where \( V_c = (2\pi)^{3/2} w_r^2 w_z^2 \) is a constant, and \( w_r, w_z \) are the Gaussian radii defined in Chap 2. The solution is

\[ N(t) = \frac{N_0 \exp(-\gamma_0 t)}{\frac{2\gamma_0}{\gamma_0 + \frac{\beta}{V_c}} [1 - \exp(-\gamma_0 t)] + 1}. \quad (3.13) \]

\( \beta \) is found to be \((3.4 \pm 0.9) \times 10^{-12} \text{ cm}^3/\text{s} \) averaged over ten scans of the same parameters as that shown in Fig. 3.8. For trap temperature of 1.7 mK, the mean velocity is 1
m/s. We estimate the cold-cold collision cross section to be \((3.4 \pm 0.9) \times 10^{-14} \text{ cm}^2\). This is about 0.4 time the background collision cross section \(\sigma_c\), which is \(9 \times 10^{-14} \text{ cm}^2\).

Notice that the FS channel, which transfers an energy of 0.48 K can only be turned on at a trap depth \(\leq 0.24\) K. As discussed in Chap 2, the trap depth can be varied by the trapping conditions, such as laser intensity. The trap depth as a function of laser intensity (Rabi frequency) predicted by the one dimensional Doppler cooling model is illustrated in Fig. 2.12. In the experiment, \(I = 85 \text{ mW/cm}^2\) is the total laser intensity in the six beams in one first-order sideband. The Rabi frequency in the 1D model is defined as \(\sqrt{I_1/I_\pi \Gamma}\), where \(I_1\) is the laser intensity in one beam. Therefore, \(I_1 = 14 \text{ mW/cm}^2\), and \(\Omega = 1.37\Gamma\). The trap depth for this trapping condition (\(\Omega = 1.37\Gamma, \Delta = -2.4\Gamma, \text{ and } b = 20 \text{ Gauss/cm}\)) is 0.37 K, much greater than the FS turning-on trap depth 0.24 K. Therefore, the collision is from the RE channel only. For intensity of 85 mW/cm², trap depth 0.37 K and trap temperature 1.7 mK, the
theory in previous section predicts a $5.9 \times 10^{-12}$ cm$^3$/s for $-1.0\Gamma$ detuning, 1.8 times the measured $\beta_{RE}$ at $-2.4\Gamma$ detuning. There is not yet a theoretical calculation on the detuning dependence of $\beta$. Without making a comparison, I show in Fig. 3.9 the measured detuning dependence. The intensity is 85, 90, 68, 87 mW/cm$^2$ for $-2.4$ to

![Graph showing the relationship between $\beta$ and Detuning.](image)

**Figure 3.9** Measured collision rate coefficient vs detuning.

-5.2 linewidths detunings, respectively and $b = 20$ Gauss/cm. From the 1D model, the collisional process is RE only.

The uncertainty in the experiment comes from several sources. As discussed in the absorption measurement, the trapping parameters contains significant error. From Eq. 3.13, the relative error of $\beta$ is approximately

$$\frac{\Delta \beta}{\beta} = \sqrt{\left(\frac{\Delta N}{N}\right)^2 + \left(\frac{\Delta V_c}{V_c}\right)^2 + \left(\frac{\Delta \gamma_b}{\gamma_b}\right)^2}. \quad (3.14)$$

$\Delta N/N = 25\%$ from Chap 3, and $\Delta V_c/V_c \simeq 3\Delta w_e/w_e = 15\%$. $\Delta \gamma_b/\gamma_b$ from the fit program scatters less than 10%. Therefore, I estimate the uncertainty from the
experimental parameters to be 40%. Another source of error is from the modeling of the decay. In the modeling, we assumed constant cloud radii, whereas in the experiment we found they decrease with decay signal. The uncertainty from this source has not yet be carried out. The fit routine itself, critically depends on the background level. Once including the fit to the background level, the fit program works well. However, the estimated variance on the fitted parameter $\beta$ is still about 100%. So the uncertainty from one scan is 140%. The $\beta$ from the average of the ten scans is 30%. This is consistent with the data scattering. One of the fitted curve is included in Fig. 3.8. The non-linearity in Fig. 3.8 is clearly determined by $f \equiv \beta n_o / V_c \gamma_b = \beta n_e / \gamma_b$, where $n_e = 2^{-3/2} n_o$, $n_o$ is the peak density. To observe the non-linear effect and improve the measurement, we need to have large density and small background collision rate to enhance the non-linear fraction $f$. In Fig. 3.8, $n_o = 2.8 \times 10^{10}$ cm$^{-3}$, $\gamma_b = 0.22$ s$^{-1}$. Therefore $f = 0.15$.

### 3.4.2 FS rate coefficient

As I pointed out several times in the previous sections, we can turn on the FS channel by lowering the intensity so as to lowering the trap depth (Fig. 2.12). So, to measure the FS coefficient, we measured the intensity dependence of $\beta$. The results are shown in Fig. 3.10. The results show a slight increase at large intensity, which is the intensity dependence of the RE process. (It is slower than a linear dependence, because the intensity changes both the excitation rate and the trap depth.) At some small detunings, there is a rather sudden jump of $\beta$ as we expected because the FS channel is turned on. From the one-dimensional Doppler cooling model, the FS channel is expected to turn on at $\Omega = 1.0 \Gamma$ at $b = 20$ Gauss/cm, i.e. intensity of 45 mW/cm$^2$ for $\Delta = -2.4 \Gamma$ and $\Omega = 0.45 \Gamma$ (intensity of 9 mW/cm$^2$) for $\Delta = -4.2 \Gamma$. The slope of turning-on in the experiment may be caused by the non-uniformity of the
trap depth. Also the turning-on for the −2.4Γ detuning is not well resolved because of lack of experimental data points between 20 and 60 mW/cm².

The FS coefficient can be obtained by deducting β_{RE} from measured β. β_{RE} after the turning-on are calculated from β_{RE} at high intensity as a reference and the scaling discussed in Chap. 3.3.1. The trap depth are calculated from the 1D model. The results of two such data points (intensity of 16 and 8 mW/cm² in Fig. 3.10) are tabulated in Fig. 3.11, where intensities are in mW/cm². T_D in Kelvin and β in 10⁻¹² cm³/s.

<table>
<thead>
<tr>
<th>Δ</th>
<th>β^{rc}_{RE} @ I, T_D</th>
<th>I_{FS-on}</th>
<th>T_D</th>
<th>β_{RE} @ I_{FS-on}, T_D</th>
<th>β</th>
<th>β_{FS}</th>
<th>β^{calc.}_{FS}</th>
</tr>
</thead>
<tbody>
<tr>
<td>-2.4Γ</td>
<td>3.4 @ 85, 0.37</td>
<td>16</td>
<td>0.17</td>
<td>5.8</td>
<td>18</td>
<td>12</td>
<td>29</td>
</tr>
<tr>
<td>-4.2Γ</td>
<td>3.0 @ 136, 0.58</td>
<td>8</td>
<td>0.24</td>
<td>2.2</td>
<td>35</td>
<td>33</td>
<td>21</td>
</tr>
<tr>
<td>Source</td>
<td>Exp., Exp., 1D</td>
<td>Exp. 1D</td>
<td>J&amp;V + β^{rc}_{RE}</td>
<td>Exp. β, β_{RE}</td>
<td>G&amp;P</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**Figure 3.10** Collision rate coefficient measured for laser detuning -2.4 and -4.2 linewidths.

**Figure 3.11** Results of β vs intensity measurement.
These results agree qualitatively with other alkali atoms and are consistent with the prediction of both Gallagher, Pritchard’s model and Julienne, Vigué’s model that $\beta_{FS}$ is larger than the $\beta_{RE}$ for light alkali atoms. The calculated FS rate coefficient from Gallagher and Pritchard model shown in Fig. 3.6 is $29 \times 10^{-12}$ cm$^3$/s for -2.4 $\Gamma$ (-14 MHz) at intensity 16 mW/cm$^2$ and $21 \times 10^{-12}$ cm$^3$/s for -4.2 $\Gamma$ (-25 MHz) at intensity 8 mW/cm$^2$.

The simplified Gallagher and Pritchard model gives a very clear physical picture but may not be adequate. A refined model as in Julienne and Vigué as well as more experimental data may provide a better comparison.

In comparing the experimental measurement with theory, the major uncertainty comes from the inhomogeneity of the trap. In our experiment, although the intensities in the three axes were balanced to within 20%, the field gradient in the $z$ axis is two times that in the $x$ and $y$ axes. Moreover, the laser beams have Gaussian distribution, which means the intensity is not uniform over the trap. $\beta_{RE}$ is very sensitive to trap depth, so the non-uniform intensity which results in non-uniform trap depth may significantly vary the results from that of a uniform trap. We have not yet investigated these questions.

### 3.5 Discussion

1) The collision study related to laser cooled and trapped atoms has been an intensely studied topic recently. There are other techniques of measuring the collision coefficient. Sesko et al [1989] has utilized a catalysis laser, a technique using a far red detuned laser to probe the trapped atoms, because the redder detuned laser enhances the collision without exerting much effect on the trap. This technique has been used to measure the coefficient of Cs [Sesko] and Rb [Hoffmann, 1992]. For Li, the peak position is expected to be at about three linewidth compared to eight linewidths for
Cs and more than ten linewidths for Rb. The small detunings disturb the trap also causing additional fluorescences. The coefficient measurement is not possible at the small detunings, while it will be useful in extending our measurement to the redder region where trapping in that region is either weak or not possible.

2) Wallace et al [1992] has reported some interesting features in the Rb collision coefficient measurement. Their results show $\beta$ increase with intensity, while at low intensity it decreases with intensity, similar to our observation, which they ascribe to the turning-on of ground state hyperfine changing collisions as a loss mechanism. The apparent advantage of Li is that its negligible hyperfine structure simplifies the problem and we can separate the FS and RE processes because of the small fine structure splitting.

3) In our experiment, we measured RE and FS coefficient independently. This is the first time in experiment to isolate these two mechanisms and measured each of them separately. This provides a better comparison with theory.

4) In isotope collision studies, Rb isotope have hyperfine splitting comparable to the trap depth, while it is 18 MHz and 5 MHz for $^7$Li and $^6$Li, both much smaller than the trap depth and therefore negligible. However, their relative mass difference is significant, which means they are quantum mechanically different. It will be very useful to measure the coefficient of $^6$Li and compared with that of $^7$Li in understanding the quantum effect in these collisional processes. In the near future, we are intended to measure isotope $^6$Li and perform molecular fluorescence spectroscopy of the trapped atoms.
Conclusion

We have successfully trapped $^7$Li in both vapor cell and beam magneto-optical trap. We studied in detail on the trap mechanism and measured the trap parameters. We expect this technique to become very useful in applications as well as physics studies, such as collision study. In our collision experiment, we are able to isolate the two cold-cold atom collision mechanisms and measured them separately. The comparison of the experimental results with present available theory indicated that quantum theory may be required in treating Li cold-cold collisions. We further expect to study these collisional processes in $^6$Li and compare with $^7$Li in the hope that the large quantum difference between these two isotopes will bring us new information about these cold-cold atom collisions.
Appendix A

Electro-Optical Modulator

One of the EOMs we used in the MOT experiment is a 400 MHz split-ring resonator. It utilizes a LiTaO$_3$ crystal as the modulating medium. The large dielectric constant of the crystal together with the cavity essentially form a lumped capacitor. The design followed that in Hardy [1981] and Kelly [1987].

The crystal is placed between the gap of a well polished OHFC resonant cavity as in Fig. A.1. The linearly polarized incident laser field (frequency $\omega_0$, polarization along $z$ axis) is transversely phase modulated by the resonant field in the cavity (modulation frequency $\omega_m$). The output at the end of the crystal of length $L$ is:

$$E(t) = E_0 \sin(\omega_0 t + \eta \sin \omega_m t)$$

$$= E_0 \sum_{n=-\infty}^{\infty} J_n(\eta) \sin(\omega_0 + n \omega_m) t$$  \hspace{1cm} (A.1)
where

\[ \eta_{ij} = \frac{\pi L n_i^2 r_{ij} E_j}{\lambda_o} \]  \hspace{1cm} (A.2)

is the phase modulation index. \( n_i \) is the index of refraction for the \( i \)th direction of light polarization, \( E_j \) is the applied electric field strength of modulation, \( r_{ij} \) is the linear electro-optical coefficient. The maximum fraction of power in the first-order sideband is \( J_1^2(\eta = 1.84) = 0.34 \). The modulation frequency \( f_m \) is the resonant frequency of the lumped \( LC \) circuit.

\[
\begin{align*}
    f_m & = \frac{1}{2\pi} (LC)^{-1/2} \\
    & = \frac{c}{2\pi v_o} \left( \frac{d}{\pi w K G} \right)^{1/2} \left( 1 + \frac{A_1}{A_2 - A_1} \right)^{1/2}.
\end{align*}
\]  \hspace{1cm} (A.3)

The frequency is determined by the ratio of the depth \( d \) and the width \( w \) of the crystal as well as the cavity radius \( r_o \). In current design, the crystal is \( d = 1.3 \) mm, \( w = 1.0 \) mm, \( L = 25 \) mm, \( r_o = 1.5 \) cm. \( K \) is the relative dielectric constant and is 43 for \( \text{LiTaO}_3 \). \( G = 0.65 \) is the correction factor for finite solenoid. \( A_1 \) and \( A_2 \) is the cross section of the cavity and the shielding box (4" \( \times \) 2.75").

The rf power is coupled to the cavity using a single loop of RG-58 coax cable terminated to its sheath. The frequency is adjustable for \( \pm 10 \) MHz by a tuning ring placed at one end of the cavity. The cavity is held by a low dielectric loss delrin clamp. The assembly is covered by the aluminum shielding box.

The power loss is mainly from the skin effect in the cavity and the dielectric loss in the crystal. The quality factor is about 100 and the power required for the full modulation at 406 MHz is about 300 mW.

We are currently working on a new modulator that will have first-order sideband at \( \pm 114 \) MHz to be used on trapping \( ^6\text{Li} \) which has a hyperfine structure splitting 228 MHz in the ground state.
Appendix B

Fine Structure Changing Collision Coefficient

This calculation is based on the Gallagher and Pritchard model.

1) As pointed out in Gallagher's paper, the excitation should be considered from a molecular viewpoint. There are four excited states arising from the $2S+2P$ electronic configuration that are attractive in the intermediate region. The excitation cross section is half of the atomic one. Therefore, the Rabi frequency of the excitation is:

$$\Omega^2 = \frac{I \sigma_o}{\hbar \omega} \cdot \Gamma_M$$  \hspace{1cm} (B.1)

where $I$, $\omega$ is the laser intensity and frequency. $\sigma_o = \lambda^2 / \pi$ is the on-resonance atomic absorption cross section. $\Gamma_M$ is the molecular decay rate. The semi-classical excitation rate depends on the separation $R_o$ of the atomic pair.

$$E(R_o, \Delta) = \frac{\Omega^2 \cdot \Gamma_M}{2\Omega^2 + 4(\Delta + \frac{\sigma_o}{\hbar R_o})^2 + \Gamma_M^2}.$$  \hspace{1cm} (B.2)

$\Delta = \omega_l - \omega_o$ is the laser detuning relative to the atomic transition. Fig. B.1 shows the excitation rate for different detunings.

2) The excited atomic pair is then drawn closer by the $C_3/R^3$ interaction. To undergo the $J$-changing process, the atomic pair needs to reach the inner $J$-changing region without spontaneous emission back to the ground state. Because of the relative slow velocity, the process is greatly affected by the spontaneous emission. If we neglect the initial thermal velocity, the time needed to reach the $J$-changing region is almost the same as to reach the origin because of the large velocity at small-$R$ region.

$$t(R_o) = \left(\frac{\mu}{2}\right)^{1/2} \int_0^{R_o} \frac{dR}{\sqrt{\frac{C_3}{R^3} - \frac{C_3}{R_o^3}}}$$
**Figure B.1** Excitation rate for different detunings. The larger the detuning, the smaller R the excitation occurs, and narrower the profile; vice versa.

\[
\phi(R_o) = \exp(-\Gamma_M t(R_o))
\]  

(B.3)

where \( \mu \) is the reduced mass. It is clear that the redder the detuning, the smaller is \( R_o \) where the excitation occurs, the longer is the time that needs to reach the \( J \)-changing region, therefore the larger the possibility to reach the \( J \)-changing region.

The probability of reach this small-\( R \) region without spontaneous emission is

\[
\phi(R_o) = \exp(-\Gamma_M t(R_o))
\]  

(B.4)

The total probability of the \( J \)-changing is the sum due to multiple curve crossings:

\[
P_J(R_o) = \eta_J \phi + \eta_J \phi \cdot (1 - \eta_J) \phi \cdot \phi + ... \\
= \frac{\eta_J \phi}{1 - (1 - \eta_J) \phi^2}
\]  

(B.5)

where \( \eta_J \) is the probability of undergoing the \( J \)-changing at each round trip curve crossing. \( \phi(R_o) \) and \( P_J(R_o) \) are shown in Fig. B.2.
Figure B.2 $\phi(R_o)$ and $P_j(R_o)$. $\eta_J$ is taken to be 0.02.

Taking into account the probability of finding an atom in the shell with distance $R_o$ in a uniform distributed density, the total rate per unit volume is therefore the product of all the probability integrated over the separation:

$$R_J(\Delta) = \frac{\eta^2}{2} \int_0^\infty E(R_o, \Delta) \cdot P_j(R_o) \cdot D(R_o) dR_o$$  \hspace{1cm} (B.6)

where

$$D(R_o) = 4\pi R_o^2 \cdot \exp \left( -\frac{4\pi R_o^3}{3} \cdot n \right).$$  \hspace{1cm} (B.7)

The total collision rate coefficient is the collision rate per atom with the entire ensemble.

$$K_{FS}(R_o) = \frac{2R_J(\Delta)}{n^2} = \frac{2}{n^2} \int_0^\infty E(R_o, \Delta) \cdot P_j(R_o) \cdot D(R_o) dR_o$$  \hspace{1cm} (B.8)

where the factor of 2 comes from the fact that both atoms escape from the trap. The results are shown in Fig. 3.6. The parameters used in the calculation are: $I = 10$ mW/cm$^2$, $n = 1 \times 10^{16}$ atoms/cm$^3$, $\Gamma_M = 7.2 \times 10^7$ s$^{-1}$, $\eta_J = 0.02$, $C_3 = 5.51$ a.u.
Appendix C

Circuit Schematics and Timing Diagram

Figure C.1 Fluorescence photodiode schematic.

Figure C.2 Timing diagram for absorption measurement.
Figure C.3  Gating circuit schematic.

Figure C.4  Integrator circuit schematic.
References


