Photoionization of Neutral Barium

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Abstract

A photo-ionization scheme was developed for loading ions into an ion trap. The photo-ionization required 791 nm and 450 nm laser light suitable for addressing the relevant energy levels in neutral Barium. Using these lasers, spectroscopy of the 450nm transition was carried out and the isotope shift of ¹³⁵Ba and ¹³⁷Ba was measured. The spectroscopy allowed us to be able to achieve ¹³⁷Ba⁺ isotope selectivity in an ion trap with geometry. In this thesis, we discuss the experimental setup and characterization of the laser system used in the photo-ionisation scheme.

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Chapter 1

Introduction

1.1 Motivation for this work

The development of the ion trap system for quantum information applications has resulted in a number of impressive demonstrations of the fundamental building blocks necessary for larger scale devices. More recently efforts have focussed on scaling up to a larger number of ions. The use of shuttling techniques in which ions are moved from one location to another [1] have shown promise [2, 3]. For larger networks however it would be desirable to use photons as a means to mediate information transfer between distant locations. Experiments towards this goal have utilized the entanglement between an atom and a photon which occurs when atom decays from an excited state [4, 5]. Although very impressive, these experiments are inherently limited due the very low inefficiency associated with the collection of spontaneous emission. One possibility to overcome this inefficiency is to utilize the techniques developed for cavity quantum electrodynamics (CQED).

In CQED experiments, strong coupling of between an atom and a photon is mediated by a high finesse cavity. Very crudely, the high finesse cavity enhances the atom photon interaction by allowing the photon to make many round trip passes by the atom before its eventual decay. This enhanced interaction can be used to provide a highly efficient exchange of information between the atom and the cavity photon [6]. Combining the ion trap system with cavity QED would then provide the means of efficiently exchanging information between ions and photons and ultimately providing an optical link between remotely located ions.

Implementing the principles of CQED requires very high quality cavities and this requires mirrors with exceptionally high reflectivity and low loss. Since the mirror performance tends to degrade as the wavelength of interest decreases, it is desirable to use an ion with transitions in the visible or infrared part of the spectrum. The wavelengths relevant to Barium are the longest of the ionic qubit candidates with the transitions lying in the visible part of the spectrum. These transitions are conveniently accessed by diode lasers and high finesse, robust mirror coatings and optical fibers are readily available at the associated wavelengths. This makes it an ideal candidate for remote entanglement and CQED studies.

Barium has five stable isotopes available (¹³⁸Ba, ¹³⁷Ba, ¹³⁶Ba, ¹³⁵Ba, ¹³⁴Ba) with natural abundances 71.7%, 11.3%, 7.8%, 6.6%, 2.4% respectively. For qubits, it is desirable to use an isotope with a hyperfine structure. This provides readily available qubit states that allow state sensitive detection and the possibility of magnetically insensitive qubit transitions. Isotopes with the desired hyperfine structure are provided by the odd isotopes ¹³⁵Ba⁺ and ¹³⁷Ba⁺. We use ¹³⁷Ba⁺ as it has the highest natural abundance. In principle, isotope enriched sources are available but these are very expensive and not overly pure. Thus it is important to provide the means to load ions in an isotope selective way.

Loading ions into a trap typically involves ionizing neutral atoms within the trapping region. There are essentially two ionizing techniques used: electron bombardment and photo-ionization. Electron bombardment basically creates thermal electrons using a hot filament and they are accelerated towards the trap using a high voltage of a few tens of volts or more. The resulting collision with a neutral liberates a bound electron creating an ion. Photo-ionization, on the other hand, uses light induced excitation of the bound electron into the continuum. This excitation can be by a single photon absorption or by the absorption of many photons. The ionization scheme used prior to this work made use of single excitation photo-ionization. A xenon flash lamp which emits wavelengths ranging from 200nm to 2000nm illuminated the trap with absorption of photons in the lower 200nm range resulting in ionization. This technique has inherent disadvantages similar to electron bombardment. Firstly the wavelengths produced the Xenon flash lamp readily produce photo-electrons from all irradiated metal surfaces. This results in the accumulation of charges on insulating surfaces which destabilize the trap in a time dependent and unpredictable way. Secondly, there is no isotope selectivity as the xenon flash lamp ionizes all isotopes due to the wavelength insensitive nature of the process.

In this work we develop a photoionization scheme which excites a narrow intercombination transition at 791 nm followed by a transition at 450 nm. The 791 nm transition is very narrow and provides the ability to selectively excite desired isotopes by tuning the laser wavelength appropriately. All excitation steps are done via lasers. Since these can be tightly focussed, the generation of photon electrons is highly suppressed which minimises charging effects. The technique is also very efficient enabling fairly reliable loading within a few tens of seconds.

1.2 Organization of this thesis

In this thesis, we describe the details of the photo-ionization scheme used to load ions in an isotope selective way and the laser system needed for its implementation. In Chapter 2, we give a detailed description of our photoionization scheme. We also give an account of the spectroscopy principles used to establish frequency references for the photo-ionization lasers. In Chapter 3, we describe the experimental setup of the lasers needed to perform spectroscopy of the transitions involved in the photo-ionization. We also describe the implementation of a heat pipe vapour cell needed for the spectroscopy. In Chapter 4, we present the results of the spectroscopy of the transitions involved. This includes measurements of the hyperfine splitting of the upper $6p^2$ ³P₁ level which we believe is the first such measurements for this level. We also discuss the trapping of ions via this scheme and the procedures used to establish operating parameters. Finally in Chapter 5, we summarize our findings and outline future directions.

Chapter 2

Background

In this chapter, we will be looking at the process of photo-ionization in barium as well as the interaction of atoms with radiation. We start with a detailed description of our photo-ionization scheme, the transitions involved and motivations behind our chosen photo-ionization path. We then give an account of the spectroscopy techniques used in this work. This account provides us with sufficient detail to estimate beam intensities, detuning, etc needed for the implementation of both our spectroscopy set up and the photo-ionization.

2.1 Photo-ionization

Photo-ionization is the process of ionizing a neutral atom through the absorption of photons. When an atom absorbs a photon an electron is excited to a higher energy state. If the photon has sufficient energy the electron can be promoted to the continuum and an ion is formed. Single photon photo-ionization typically requires UV light ($\approx 240 \text{ nm}$ for Barium) and is not isotope selective. Here we use a multi-photon excitation scheme in which the atom is first promoted to an excited state and then subsequently photo-ionized by further absorption. The first excitation(s) provide isotope selectivity due to the narrow resonances involved.

Energy levels Ba and Ba⁺ relevant to this work are shown in Fig 2.1 and Fig 2.2. Following Chapman's work, previous barium photo-ionization schemes have utilized the narrow intercombination line at 791 nm followed by an excitation above the ionization threshold by a nitrogen pulsed laser at 337 nm [7]. However, in our photo-ionization scheme, we have chosen to use a 450 nm laser after the excitation at 791 nm to excite it to the $6p^2$ ³P₁ level as can be seen from Fig 2.1. The $6p^2$ ³P₁ level is very close to the ionization threshold so that any laser currently used in the Barium ion experiment can then complete the excitation. We use these lasers because all these lasers are readily available. The 791 nm laser is easily obtainable from the same lasers used to excite the rubidium D1 and D2 lines in the rubidium lab, while the 450 nm laser is already being explored for use at 455 nm to address the P_{3/2} level in the ¹³⁷Ba⁺ ion. A laser at 450 nm or 457 nm (either would do) could be used also for Raman transitions in the ion due to there proximity to the S_{1/2} to P_{3/2} transition in the ion.



Figure 2.1: Energy levels of Barium.



Figure 2.2: Barium ion energy levels.

2.2 Spectroscopy

The frequency reference for our photo-ionization lasers was provided by an atomic vapour cell. Here we give a theoretical description of the spectroscopy techniques we have used. We start with a very basic description of absorption through a gas of atoms at rest. We then illustrate how this treatment can be generalized to account for Doppler broadening. Using these discussions, we outline the principles of saturated absorption spectroscopy and how this is used to find a narrow atomic transition within a Doppler broadened absorption profile. We also discuss the two step excitation process and how it differs from the usual single excitation spectrum. This discussion form the basis of our measurements of the hyperfine splittings of the $6p^2$ ³P₁ level transition. We restrict ourselves to a two level atom as this is all that is needed for this work. Our discussion follows that given in [8].

2.2.1 Optical Absorption cross section

We consider a laser beam travelling through a sample of atoms as shown in Fig.2.3.

The attenuation of the beam can be determined by considering the power lost due to the photons scattered. For the area element A we have

$$dP = AdI = -\hbar\omega\gamma_s nAdz \tag{2.1}$$



Figure 2.3: Atoms with number density, N, distributed in a slab of thickness dz, and surface area, A. We assume that the motion of the target atoms can be ignored.

where, γ_s scattering rate, I is the intensity of the light incident on the sample of atoms, n is the density of atoms. For a two level atom the scattering rate is given by

$$\gamma_s = \frac{\Gamma}{2} \frac{s_0}{1 + s_0 + 4\Delta^2 / \Gamma^2}$$
(2.2)

where Γ is the linewidth of the transition, Δ is the laser detuning from resonance, and s_0 is the saturation parameter. In terms of the laser intensity we have $s_0 = I/I_{sat}$ where

$$I_{sat} = \frac{\pi \Gamma hc}{3\lambda^3}.$$
(2.3)

We then have

$$\frac{dI}{dz} = -n\frac{\hbar\omega\Gamma}{2}\frac{s_0}{1+s_0+4\Delta^2/\Gamma^2}
= -n\frac{\hbar\omega\Gamma}{2I_{sat}}\frac{1}{1+4\Delta^2/\Gamma^2}\left(\frac{1+4\Delta^2/\Gamma^2}{1+s_0+4\Delta^2/\Gamma^2}\right)I
= -n\frac{\sigma_{abs}}{1+4\Delta^2/\Gamma^2}\left(1-\frac{s_0}{1+s_0+4\Delta^2/\Gamma^2}\right)I$$
(2.4)

where we have introduced the absorption cross section $\sigma_{abs} = \frac{\hbar\omega\Gamma}{2I_{sat}} = \frac{3\lambda^2}{2\pi}$. Since the term in parentheses is just the difference in populations we can write this expression in the simple form

$$\frac{dI}{dz} = -(n_g - n_e)\sigma(\Delta)I, \quad \sigma(\Delta) = \frac{\sigma_{abs}}{1 + 4\Delta^2/\Gamma^2}$$
(2.5)

where n_g (n_e) is the population density of the ground (excited) state. This treatment is only valid for a two level atom. When more levels are involved we must replace the absorption cross section by

$$\sigma_{abs} = \frac{3\lambda^2}{2\pi} \frac{2J'+1}{2J+1} \frac{\gamma_p}{\gamma_{total}}$$
(2.6)

where γ_{total} is then the total rate of decay from the upper state, γ_p is the decay rate back to the lower state, J(J') is the orbital angular momentum of the lower (upper) state.

At 791nm, the transition is from the ${}^{1}S_{0}$ to the ${}^{3}P_{1}$ level. The partial rate of decay, γ_{p} , of the ${}^{3}P_{1}$ level back to the ${}^{1}S_{0}$ level was found to be 56.5kHz. For the ${}^{3}P_{1}$ state, γ_{total} is 132 kHz [9]. The absorption cross section is calculated to be 1.3×10^{-13} m² and the saturation intensity is $0.35 \ \mu W/cm^{2}$.

Similiarly, we can do the same calculations for the 450nm transition. The total rate of decay, γ_p of the 6p² ³P₁ level was found to be 61.9MHz and the partial rate of decay back to the the state of interest (6s6p ³P₁) is 16.6MHz

[9]. The absorption cross section, σ , for this transition is calculated using Eq 2.6 to be 2.55×10^{-14} m². The saturation intensity is calculated to be 0.88 mW/cm^2 .

2.2.2 Doppler Broadening



Figure 2.4: Doppler effect on the observed frequency of radiation

In the previous section, we have assumed that the atoms are stationary, however, in reality, the velocity distribution of a gas follows the Maxwell speed distribution. In a gas, the fraction of atoms with velocity in the range v to v+dv is

$$f(v)dv = \sqrt{\frac{M}{\pi 2k_BT}} \exp\left(-\frac{Mv^2}{2k_BT}\right)dv$$
(2.7)

and this significantly modifies the absorption spectrum.

For an atom moving with velocity, \mathbf{v} , the relationship between the angular frequency ω of radiation in the laboratory frame of reference and the angular frequency seen in a frame of reference moving at velocity, \mathbf{v} , is

$$\omega' = \omega - \mathbf{k.v} \tag{2.8}$$

and when the angular frequency of radiation is in the opposite direction as the atom moving at \mathbf{v} ,

$$\omega'' = \omega + \mathbf{k.v} \tag{2.9}$$

where **k** is the wavevector of the radiation and has magnitude of $\mathbf{k} = 2\pi/\lambda$. If the atom is at rest, the atom absorbs radiation at frequency ω_0 , that is $\omega'' = \omega' = \omega_0$. It is the component of the velocity along **k** that leads to the doppler effect.

To account for this shift the absorption must be averaged over the velocity distribution. This can be done in a similar manner to the calculation presented in Appendix C. The effect is to replace the lorentzian absorption profile to the Gaussian profile determined by the Maxwell-Boltzmann distribution with an appropriate reduction in the resonant absorption cross section σ_{abs} . From [10], Eq 2.6 is replaced with,

$$\sigma = \frac{\sqrt{\pi}}{2} \frac{\gamma}{\Gamma_D} \sigma_{abs} \tag{2.10}$$

Where Γ_D , the Doppler's width is given by,

$$\Gamma_D = \frac{2\pi}{\lambda} \sqrt{\frac{2k_B T}{M}} \tag{2.11}$$

At 900k, the Doppler's width is found to be 418MHz with an absorption cross section of 1.44×10^{-15} m². The Doppler's width and the altered absorption cross section due to Doppler broadening calculated using Eq 2.11 and 2.10

are 734MHz and $1.02{\times}10^{-16}{\rm m}^2$ respectively.

2.2.3 Saturated Absorption Spectroscopy

A common method to overcome the effects of Doppler broadening is to make use of saturation effects in absorption spectroscopy. From Eq 2.4, the absorption for a laser on resonance with the atoms is given by

$$\frac{d\bar{I}}{dz} = -\alpha \frac{\bar{I}}{1+\bar{I}} \tag{2.12}$$

where α is given by $\frac{-n\hbar\omega\gamma_s}{2I_s}$ and it is on atomic resonant frequency.

In the limit that $\bar{I} \ll 1$ we have,

$$\frac{d\bar{I}}{dz} = -\alpha\bar{I} \tag{2.13}$$

Solving for \overline{I} , we obtain the following solution for when \overline{I} is less than 1,

$$\bar{I}(z) = \bar{I}(0) \exp{-\alpha z} \tag{2.14}$$

Thus for weak intensities the beam has a fixed percentage of absorption for a given cell length and the transmission is given by

$$T = \frac{\bar{I}(0)}{\bar{I}(0)} = \exp \alpha z \tag{2.15}$$

On the other hand, when $\bar{I} \gg 1$ we have

$$\frac{d\bar{I}}{dz} = -\alpha \tag{2.16}$$

which has the following solution,

$$\bar{I}(z) = \bar{I}(0) - \alpha z \tag{2.17}$$

When \bar{I} is less than 1, the percentage transmission given by

$$T = \frac{\bar{I}(0)}{\bar{I}(0)} = \exp{-\alpha z} \Rightarrow 0$$
(2.18)

The percentage transmission given by

$$T = \frac{\bar{I}(0) - \alpha z}{\bar{I}(0)} = 1 - \frac{\alpha z}{\bar{I}(0)} \Rightarrow 1$$
(2.19)

which increases to unity as the intensity increases.



Figure 2.5: A typical saturated absorption spectroscopy setup where the pump beam and the probe beam are overlapped.

We exploit this saturation of absorption in saturated absorption spectroscopy. Fig 2.5 below shows a typical absorption setup. A strong pump beam is sent into the sample and an attenuated probe beam in the opposite direction is sent back into the sample. The pump beam and the probe beam are overlapped. The equations to describe the pump beam and the probe beam is shown previously in Eq 2.8 and Eq 2.9 respectively. Again, as discussed earlier, at rest, the the atom absorbs radiation at frequency ω_0 , that is $\omega'' = \omega' = \omega_0$. A typical absorption signal has the shape of a Gaussian function as discussed earlier.



Figure 2.6: A typical saturated absorption signal. The peaks are that of the ¹³⁵Ba and ¹³⁷Ba in a gaussian background.

When the probe beam is added, the atoms which have zero velocity will absorb both the pump and the probe beam at the atomic resonance frequency, ω_0 . In addition, we approach the case of when \bar{I} is greater than 1 as seen in Eq. 2.19 causing the transmission of the beam to decrease, resulting in a peak in the absorption signal at the atomic resonance frequency as seen in Fig.2.6.

2.2.4 Two-step excitation spectroscopy

In a single photon absorption, we obtain a saturated absorption signal as described in the previous section. However, in our photoionization scheme, two photons of different frequencies are being absorbed by the atom. This tells us that for the second excitation to take place, the atom must first be in the first excited state. Fig 2.7 shows us the absorption signal when two photon absorption takes place. The 791nm transition (first excited state) is that of a typical saturated absorption signal peak in a gaussian doppler broadened absorption signal while the 450nm transition (second excited state) directly gives an absorption signal which is already lorentzian and not doppler broadened. In this section, we will use the isotopes of 135 Ba and 137 Ba of F state 5/2 in our calculations shown below to illustrate.



Figure 2.7: Figure (a) shows the transmission signal of the 791nm laser while Figure(b) shows the transmission signal of the 450nm laser.

Consider Fig 2.8, for an atom moving at velocity, \mathbf{v} , from Eq 2.8 and with the two lasers of different frequencies co-propagating, we have



Figure 2.8: An atom moving at velocity, \mathbf{v} , with two different lasers of different frequencies, which we have labelled as ω_R and ω_B . ω_R' is the returning probe beam from ω_R in a typical saturated absorption spectroscopy setup.

$$\omega_0 = \omega_R - k_R \cdot \mathbf{v} \tag{2.20}$$

for the first laser ω_R and,

$$\omega_0' = \omega_B - k_B \cdot \mathbf{v} \tag{2.21}$$

for the second laser ω_B . k_R and k_B refers to the wavevector of the 791nm laser and the 450nm laser repectively. ω_0 refers to the atomic resonant frequency of the first transition from the ground state to the first excited state and ω_0' refers to the atomic resonant frequency of the second transition from the first excited state to the second excited state.

The velocity class of atoms, \mathbf{v} , in both equations Eq 2.20 and Eq 2.21 are the same when we consider the case of both laser beams overlapping, because for the second transition to occur, the same atom must be first excited from the ground state to the first excited state. Hence, the second transition is not doppler broadened because the atoms that undergo the



Figure 2.9: Energy levels of $^{135}\mathrm{Ba}$ and $^{137}\mathrm{Ba}$ with their different hyperfine splittings.

second excitation are from the same velocity class. (Refer to Section 2.2.2) In our calculations, we can then eliminate velocity, \mathbf{v} from the equations,

$$\omega_0' = \omega_B - \frac{k_B}{k_R} (\omega_R - \omega_0) \tag{2.22}$$

Consider Fig 2.9 which shows the energy levels of 135 Ba and 137 Ba.If we apply the Eq 2.22 to both isotopes, we have

$$\omega_1' = \omega_{B1} - \frac{k_B}{k_R} (\omega_{R1} - \omega_1) \tag{2.23}$$

$$\omega_2' = \omega_{B2} - \frac{k_B}{k_R} (\omega_{R2} - \omega_2) \tag{2.24}$$

where ω_{R1} , ω_{R2} , ω_{B1} , ω_{B2} are the frequencies of the lasers of the 791nm laser and the 450nm laser respectively. Looking at Eq. 2.23 and Eq 2.24, we can combine these two equations to obtain a general expression,

$$\omega_1' - \omega_2' = \omega_{B1} - \omega_{B2} - \left[\frac{k_B}{k_R}(\omega_{R1} - \omega_1) - \frac{k_B}{k_R}(\omega_{R2} - \omega_2)\right]$$
(2.25)

When the red laser is scanning and the blue laser locked, i.e. $\omega_{B1} = \omega_{B2}$, we can further simplify this equation to

$$\omega_1' - \omega_2' = \frac{k_B}{k_R} [(\omega_{R2} - \omega_{R1}) - (\omega_2 - \omega_1)]$$
(2.26)

It can be seen from the equation above that by knowing the seperations between the two isotopes in the first transition, we can calculate the seperations of the the two isotopes in the second transition. We will make use of this equation in Chapter 4 to deduce the isotope seperations between 135 Ba and 137 Ba.

For the measurement of the hyperfine splittings between the F-states of each isotope, the same equation can be used as Eq B.1, but with a slight modification to the expression because the red laser is now at the same frequency. We obtain the equation below to measure the hyperfine splittings.

$$\omega_1' - \omega_2' = \frac{k_B}{k_R} [-(\omega_2 - \omega_1)]$$
(2.27)

We can also do a similar calculation if the red laser is locked and the blue laser is left to scan, with slight changes in the equations.

2.2.5 Frequency Modulation spectroscopy

The laser passes through an EOM of resonant frequency 22.75MHz. The EOM is a nonlinear crystal that allows its refractive index in certain directions to be changed by the application of an electric field. The laser is then phase modulated by the EOM and sidebands of ± 22.75 MHz are formed.



Figure 2.10: Error signal above the saturated absorption peaks of $^{138}\mathrm{Ba},$ $^{134}\mathrm{Ba}$ and $^{136}\mathrm{Ba}.$

If the laser frequency, v is being modulated at the frequency, Ω , with amplitude, m, then the transmitted intensity, I_T through the cell will be

$$I_T = I_T(\upsilon + m\sin(\Omega t)) \tag{2.28}$$

Here, if we consider the case where Ω is lesser than Γ the linewdith of absorption, we can expand I_T as a taylor series.

$$I_T(v + m\sin(\Omega t)) = [I_T(v) + \frac{m^2}{4} \frac{d^2 I_T}{d^2 v} (m\sin(\Omega t)) \frac{dI_T}{dv} + ...] + \sin\Omega t [m \frac{dI_T}{v} + \frac{m^3}{8} \frac{d^3 I_T}{d^3 v} + ...] + ...$$
(2.29)

If phase sensitive detection is performed at Ω using a lock in amplifier, the coefficient of the $\sin(\Omega t)$ term can be extracted. In particular since we have assumed that m is small, the coefficient of the $\sin(\Omega t)$ term is just $m \frac{dI_T}{v}$ which is just the first derivative of the transmitted intensity of the beam [11].

When we lock the laser to a saturated absorption peak, it can be seen from Fig 2.10 that the error signal generated is an antisymmetric function corresponding to the first derivative of the saturated absorption peak. The atomic transition corresponds to a zero level in the error signal. The board then controls how much voltage would be applied to the PZT driver and the actuator, which changes the round trip phase of the laser so that the laser stays on resonance with the atomic transition that we have locked the laser to.

Note that a more general treatment is given in [12] which applies to case in which modulating frequency is larger than the linewidth of absorption.

Chapter 3

Experimental Setup

Implementation of our photo-ionization scheme requires our laser frequencies to be tuned to the appropriate atomic resonances to maximize the rate of photo-ionization ¹. To achieve this we have developed a frequency reference based on spectroscopy of neutral Barium. Our set up provides both saturated absorption spectroscopy of the 791 nm transition and two photon spectroscopy of the 450 nm transition. In this chapter, we will discuss the workings of our vapor cell used for the spectroscopy and the optical set up of the photo-ionization lasers.

 $^{^1{\}rm additional}$ offsets in the laser frequency are needed to compensate any residual Doppler shifts dependent on oven geometry

3.1 Neutral Barium Vapour Cell

The set up and operation of a vapour cell for neutral barium is complicated as it needs to be heated to above 800 K in order achieve sufficient absorption, particularly for the isotopes with low natural abundance. These temperatures are well above the allowable operating temperatures of standard vacuum windows and so the windows need to be kept at a much lower temperature. This results in a migration of barium onto the windows and ultimately blocking the transmission of the lasers. To overcome this difficulty we have employed a heat pipe oven similar to that discussed in [13].

The workings of a heat pipe oven is similiar to that of a heat pipe. While a heat pipe has one end hot and another end cold [14], a heat pipe oven is a tube with the middle section heated and the ends cooled. As the material in the centre of the heat pipe oven is heated, it vaporises within the centre section. When it reaches the cooled ends of the oven, it condenses and falls onto a wick which draws the liquid back to the centre region of the oven by capillary effect where it will be heated up again. An inert buffer gas is introduced into the heat pipe oven. As the vapor pressure of the heated material at the centre of the heat pipe oven approaches that of the buffer gas, equilibrium between the buffer gas and the vapor will cause the vapor and the gas to seperate with the buffer gas remaining in the cooled section next to the window. This prevents the vapor from condensing and depositing on the windows of the cell because the vapor cannot propagate through the cooler section of the chamber as it will collide with the buffer gas. This will cool and condense the material before it reaches the windows.

For the design of our vapor cell, a long stainless steel chamber cell of 0.8m was made with two glass windows at each end. A piece of barium solid of about 2 grams was placed on a crucible and placed in the middle of the chamber. A thin wire mesh of 30 microns was also placed in the middle of the chamber which acts as the wick. In order to perform a bake-out of the cell, the cell was then heated up to a temperature of 600K using a heating tape wound around the middle section of the cell. Heating coils were also placed around the windows. Ceramic wool and aluminum foil was used to wrap around the heating elements in order to achieve insulation against the surroundings. The cell was attached to a vaccum pump system at the same time. The cell was then allowed to cool back down slowly and we achieved a pressure of an order of magnitude 10^{-5} torr inside the cell.



Figure 3.1: This is a schematic diagram of the vapor cell. Not to scale.

Argon gas as the inert buffer gas was then pumped into the cell to create

a pressure of 10^{-2} torr in the cell. At this pressure, P, the mean free path of argon, l, is calculated using the equation below,

$$l = \frac{k_B T}{\pi d^2 \rho \sqrt{2}} \tag{3.1}$$

where k_B is the Boltzmann constant, T is the temperature of the buffer gas, ρ , the density of argon and d, the collision diameter. For argon at room temperature and atmospheric pressure, the collision diameter, d is 362pm [15]. The argon gas is estimated to be at 400K, the temperature of the cooler ends of the cell. The mean free part of argon, l, was calculated to be 680 microns. The length of 10cm from the end of the heated region to the window is sufficient for the range of argon to collide with any barium atoms attempting to move towards the windows. It is noted that if barium ends up on the windows, however, heating the windows up to 600K for several hours is sufficient to remove the barium.

To operate the vapor cell, the heating tape remained in the middle section of the cell. The windows were kept cool, with cooling rings added at each end of the heating tape as a precaution against barium reaching the windows. For the absorption of the most abundant isotope, ¹³⁸Ba, to be seen, the temperature of the cell must be at least around 800K.

There are however problems associated with the barium vapor cell that we have constructed. The barium still tended to migrate towards the windows. This causes the absorption signal to decrease as time passes and leads to difficulties in locking the lasers for photo-ionization. This is a common problem for heat pipe ovens constructed for heavier metals because the melting point of these metals tend to be very high, and in order for the wick to draw the liquid material back to the middle section of the cell, the cell would need to be operating at a very high temperature where collision broadening is on the order of Doppler broadening which obscures the atomic resonance lines. Hence, the cell's operating temperature is lower than expected of a true heat pipe oven.

3.2 Spectroscopy set up

3.2.1 The Extended Cavity Diode Lasers

Both the 791 nm and 450 nm lasers were provided by commercial laser diodes using a standard extended cavity diode laser (ECDL) design. In this design, feedback from a diffraction grating in the Littrow configuration provides the means to tune laser frequency simply by shifting the grating position via a piezo electric transducer (PZT). The optics setup is shown in Fig 3.2 and is essentially the same for both lasers. The 791nm laser and the 450nm laser beams overlapped each other in order for us to achieve the two step excitation mentioned in the previous chapter.

The 791nm laser exits the housing is slightly elliptical and has vertical polarization. It first passes through an anamorphic prism to obtain a circular



Figure 3.2: This is a schematic diagram of the experimental setup. On top, we have the 791nm laser system, below the 450nm laser system. Light is split out using a polarizing beam splitter and coupled to a fiber which goes to the ion trap system as shown. Not to scale.

beam. It is then passed through a half-waveplate and then passed through an optical isolator, which rotates the polarisation of the beam by 45° , obtaining an output of horizontal polarisation. The isolator also prevents feedback from the optical circuit. The laser beam then passes through a half waveplate and then into an electric optic modulator (EOM) which use will be discussed later. The laser beam is passed through a half-waveplate again. This half-waveplate can be rotated to control the intensity of the laser used in the ion trap or through the vapor cell. Right after this half-waveplate, there is a polarising beam splitter (PBS) that directs vertically polarised light towards the vapor cell and transmits the horizontally polarised light to a fibre coupler. The horizontally polarized beam is used in the ion trap experiment, while the vertically polarized beam is used to lock the frequency of the laser to the desired wavelength through a lock in detection method which was discussed in the Section 3.2.2. The vertically polarized beam is also coupled into a fiber before it is shone into the vapor cell in order to obtain a single mode in the beam that is passing through the vapor The beam is also blown up by a telescope so that we can reduce cell. the intensity of the 791nm laser without reducing the power of the laser. For the transmitted beam, another half-waveplate is put after the polariser beam splitter and before the fiber coupler. This half-waveplate rotates the horizontally polarised beam to a desired angle. This half-waveplate controls the intensity of the beam that goes into the ion trap.

The 450nm laser system has a similar configuration as the 791nm laser system. However, one difference in the setup of the 791nm laser and the 450nm laser is that the beam of the 450nm laser does not go through a fiber before it is shone into the vapor cell. This is because the 450nm laser only passes through the vapor cell once so we do not have to deal with any spatial problems associated with a beam with different modes as compared to the 791nm laser beam which requires the pump beam and the probe beam to overlap with each other when we do saturated absorption spectroscopy on the 791nm transition. The 450nm laser passes through an EOM of resonant frequency 40.42MHz and we use a similiar lock in technique for the locking of the 450nm laser.

3.2.2 Spectroscopy Cell

We do not want to saturate the transitions in the spectroscopy cell, hence it is important to find out the saturation parameters of the transition so that we are able to shine laser beams of suitable intensities as well as operate the vapor cell at a lower temperature so as to preserve the lifetime of the vapor cell. Although we have given estimates of the saturation parameters in Chapter 2, there are several other broadening mechanisms which have an effect on the absorption profile in the spectroscopy cell. Some of these include the laser's linewidth and pressure broadening.

The change in the saturation parameter of the transition due to such

broadening effects can be found in detail in [16] for the interested reader. However, very simply, the saturation parameter changes as such,

$$s_0 \to \frac{S_0}{1 + \frac{\gamma}{\Gamma} + \frac{2\alpha}{\Gamma}}$$
 (3.2)

where s_0 is the saturation parameter of the transition, α is the collisional width of the absorption profile, Γ is the linewidth of the transition and γ is the linewidth of the laser. From this equation, the combined effects of the pressure broadening and the laser linewidth decreases the saturation parameter which in turns increases the saturation intensity.

For pressure broadening, in a simple treatment, the width of a transition whose natural width is Γ is broadened to

$$\Gamma_{coll} = \Gamma + \Gamma_{br} \tag{3.3}$$

where Γ_{coll} is the collisional width, Γ_{br} is the collisional broadening rate, and is given by

$$\Gamma_{br} = Nk_{br} \tag{3.4}$$

where N is the number density of argon and k_{br} is the collisional broadening coefficient [17]. The value of k_{br} given in [17] is 2.35×10^{-9} cm³s⁻¹ measured at 810K for the 791nm transition, which is close to the temperature at which we are operating the vapor cell. At this temperature, the number density of argon, N, is 1.19×10^{20} m³. Under these conditions, the collisional width is calculated to be about 411kHz. The laser linewidth is estimated to be about 300kHz. The s₀ in our calculations decreases by a factor of 10 times under these conditions.

To find out the actual saturation intensity in the spectroscopy cell, we start off by first measuring the level of absorption at a very low intensity for 791nm, we then slowly increase the intensity of the 791nm laser until the absorption level observed earlier decreases by half. The intensity at this point was measured to be 4.5 mW/cm^2 . This is about 10 times the calculated saturation intensity calculated in Chapter 2, which was 0.35 mW/cm^2 , as expected.

Chapter 4

Measurements

In this chapter, we present the results of the spectroscopy of the transitions involved. This includes measurements of the hyperfine splitting of the upper $6p^2 \ ^3P_1$ level which we believe to be the first such measurements for this level. We also give a brief account of how we load the ions into the ion trap.

4.1 Spectroscopy of 450nm transition

The isotope shifts of the ${}^{1}S_{0} - {}^{3}P_{1/2}$ transition at 791nm in barium of different F states due to odd isotopes having a non-zero nuclear spin has been reported in [18] and the hyperfine splittings in [19]. The isotope shift of 135 Ba and 137 Ba has been reported to be 143.3 MHz(1.4). In Eq B.1 and referring to Fig 2.9, this means that ω_{2} - ω_{1} = 143.3 MHz. Substituting values



Figure 4.1: A plot of ¹³⁵Ba and ¹³⁷Ba saturated absorption peaks taken from a digital oscilloscope for the 791nm transition. The gaussian background due to doppler broadening has been taken care of and the peaks are fitted to a lorentzian fit as expected of such transitions due to radiative broadening.



Figure 4.2: A plot of 135 Ba and 137 Ba peaks taken from a digital oscilloscope for the 450nm transition, as the 791nm laser is scanning. The peaks are fitted to a lorentzian fit as expected of such transitions due to radiative broadening.

of the known frequencies and from the Lorentzian fits of Fig 4.1 and Fig 4.2, we are able to calculate the isotope shifts.

We believe the scan of the PZT to be very linear because we are scanning over a small range when the measurement were taken. We estimate the error of our results to be on the order of 1 MHz characterized by collisional broadening of 411MHz and the laser linewidth as mentioned in Chapter 3 and an estimated collisional shift of 100kHz [17].

The relevant isotope separation and the hyperfine splittings can be summarized in the tables below.

F state	Isotope pair	$\triangle v (\mathrm{MHz})$
5/2	137-135	200.6
3/2	137-135	210.7

Table 4.1: Isotope separation between 135 Ba and 137 Ba.

Isotope	F-state	$\triangle v (\mathrm{MHz})$
135	5/2 and $3/2$	11.0
137	5/2 and $3/2$	7.6

Table 4.2: Hyperfine splittings between 135 Ba and 137 Ba for F=5/2 and 3/2.

We have also attempted to measure the isotopic shifts between the rest of the isotopes, however, the peaks on the right, that of 136 Ba and 134 Ba, are not resolvable, as seen in the Fig 4.3.



Figure 4.3: Scan of the 138 Ba and 136 Ba and 134 Ba in the 450nm transition.



Figure 4.4: Schematic of the ion trap and the angle at which the laser beams and the atomic beam of barium is at. The one on the left shows that the laser beams and the atomic beam is at a 45° angle, while the one on the right shows them perpendicular to each other.

4.2 Trapping ions

4.2.1 Doppler shift in the ion trap

When implementing photo-ionization we must take into account residual Doppler shifts associated with direction of the beams in relation to the direction vector of the atomic beam. This Doppler shift is given by $\mathbf{k} \cdot \mathbf{v}$ where \mathbf{k} is the wave vector associated with the photo-ionization laser(s) and \mathbf{v} is the velocity vector associated with the atoms from the barium oven. Consider Fig 4.4 which illustrates the beam and oven orientations for the two ion traps currently in use in the ion trap lab. For the trap on the right, where the atomic beam and the laser beams are perpendicular to one another. In that case, $\mathbf{k} \cdot \mathbf{v}$ has a mean of zero and there is only a small residual broadening due the angular spread of the atomic beam [8]. For the trap on the left, $\mathbf{k} \cdot \mathbf{v}$ will have a significant mean Doppler shift associated with the non-zero mean thermal velocities of the beam as well as significant broadening due to the thermal spread of velocities along the beam direction. In this case it will be important to have an estimate of these effects.

As shown in Appendix C the fraction of atoms in the excited state for a given transition is given by

$$R_e = \frac{\pi s_0 \Gamma}{8} \frac{\Gamma}{\Gamma_D} \left(\frac{\Delta}{\Gamma_D}\right)^3 e^{-\frac{1}{2} \left(\frac{\Delta}{\Gamma_D}\right)^2} \tag{4.1}$$

provided $\Gamma' \ll \Gamma_D, \Delta$ where s_0 is the saturation parameter, Γ is the natural

linewidth of the transition, $\Gamma' = \Gamma \sqrt{1 + s_0}$ is the power broadened linewidth, Δ is the detuning of the laser from the transition of interest, and $\Gamma_D = k_{eff} \bar{v}$ is a frequency scale that is characteristic of both the Doppler shift and the Doppler broadening. In particular the distribution given by Eq 4.1 has a maximum at $\sqrt{3}\Gamma_D$ and a full width half maximum (FWHM) of $\approx 1.64\Gamma_D$.

It is difficult to determine our oven temperature, but assuming a value of 600 K as reported in [20], we find $\Gamma_D \approx 2\pi \times 170$ MHz for the 791 nm transition and $\Gamma_D \approx 2\pi \times 300$ MHz for the 450 nm transition. Based on Eq 4.1 it would seem reasonable to tune the lasers to maximize R_e for each transition. However the isotope shifts for both transition are comparable to the associated values of Γ_D . Thus if we tune the lasers to maximize the excitation of the the ¹³⁷Ba isotope we will also have a significant excitation of the ¹³⁵Ba isotope and thus lose the desired isotope selectivity of the photoionization process. A better strategy is to lock both lasers to the ¹³⁵Ba isotope transitions. Excitation of this isotope will only occur for near zero velocity atoms and is thus suppressed since the velocity distribution scales as $(v/\bar{v})^3$ in this region. In Fig 4.5 and 4.6 we plot the distributions $R_e(\Delta)$ for both transitions for this particular choice of laser settings.

4.2.2 Ion Trapping

As mentioned in the previous section, both the 791nm laser are locked to the 135 Ba isotope. In addition to the 791nm and the 450nm lasers, doppler



Figure 4.5: 791nm transition: Velocity distribution of the atomic beam of barium in the ion trap at 600K. The seperation between 135 Ba and 137 Ba is 143.3 MHz. The detuning in this graph is from 135 Ba and the solid vertical line is the detuning of 137 Ba from 135 Ba.[18]



Figure 4.6: 450nm transition: Velocity distribution of the atomic beam of barium in the ion trap at 600K. The seperation between 135 Ba and 137 Ba is 200MHz. The detuning in this graph is from 135 Ba and the solid vertical line is the detuning of 137 Ba from 135 Ba.

cooling lasers (493nm and 650nm) was used in the trapping of the ions. The oven current was initially ran at 4.3 amperes. The power of the doppler cooling lasers were kept roughly constant. The 493nm laser's power was 40μ W, while the 650nm laser's power was 23.5μ W in total. The 493nm laser had a frequency of 245.2MHz on Cesium reference and the wavelength of the 650nm laser was kept at 649.86957nm on the wavemeter. The time taken to load ions is about 30 seconds each time.

We now want to minimize the ion loading parameters for the 791nm laser and the 450nm laser as well as the oven's current for the loading of ¹³⁷Ba ions. Minimizing the oven's current increases the oven lifetime and reduces the effects of Barium deposition on the trap electrodes which is thought to cause heating problems. Minimizing the power in the 791nm laser reduces unwanted excitations of the ¹³⁵Ba isotope and improves the isotope selectivity of the loading process. Minimizing the power in the 450nm laser reduces unwanted excitation of the $S_{1/2}$ to $P_{3/2}$ transition in the ion. In our initial experiments the power of the 450nm laser was very high at 200 μ W. Being of high intensity, this caused the barium that were being loaded to be excited into the $P_{3/2}$ state after it had been photo-ionized. From the $P_{3/2}$ state, it decays down into the metastable $5D_{5/2}$ state which has a lifetime of 32s. Because we had no available 615nm laser to pump it out from the $5D_{5/2}$ state, we were not unable to tell if there were any ions loaded into the trap until we blocked the 450nm laser beam and waited for it to decay back down to the ground state of the ion.

To minimise the oven's operating current, we first lock the 791nm laser and the 450nm laser to ¹³⁸Ba and reduced the oven current to a point where only a few ¹³⁸Ba ions were loaded. This is because ¹³⁸Ba is the most abundant isotope of Barium. Doing so allows us to know the threshold at which the oven in the ion trap is no longer able to produce sufficient barium for photo-ionization.

While we decreased the intensity of the 450nm laser, the photo-ionization cross section was also compromised which required us to turn up the oven current if we wanted to continue loading ions. Instead, we realised that a 650nm raman laser could be used in the process of photo-ionization and used as a third laser to photo-ionize the atoms as mentioned in Chapter 2, without changing the oven current. Because the $6p^2 \ ^3P_1$ state is very close to the continuum, any laser invovled in other ion trap experiments with sufficient power can be used to complete the photo-ionization process. The power of the 450nm laser was eventually reduced to 23μ W where it did not bring any ions into the dark state wile the 650nm raman laser was used in the photo-ionization with a power of 20 mW and we kept these 2 parameters constant for the rest of our experiment. We then varied the 791nm laser's power and recorded the number of ¹³⁷Ba ions loaded. Between each load, we waited for an average of 5-10 minutes in order to ensure that the oven could cool down sufficiently. If the oven was still warm during a new loading, we found that we would load a lot more number of ions than expected. The raw data is kept on file and is available with the micro-traps group.

The minimum loading parameters i found was when the oven was operating at a current of 3.50 amperes, with a 791nm power of 650 nW. At these parameters, the number of ions loaded each time, on average, was 1.8 ions and the maximum number of ions loaded was 3 ions. Any oven current value below 3.50 amperes loaded almost no ions, although where were instances where ions were loaded because the oven had not cooled down sufficiently. 650nW power of 791nm laser was sufficient to photo-ionize the barium atoms. A power higher than 650nW resulted in inconsistent results, probably because the intensity of the laser was still too high which would result in power broadening and lead to the loading of ¹³⁵Ba ions. The ¹³⁵Ba ions could then probably lead to the heating of the ion trap and instability in any ¹³⁷Ba ions loaded and suppress the appearance of any ¹³⁷Ba ions. Furthurmore, in all instances, only ¹³⁷Ba ions were loaded.

Chapter 5

Discussion and Conclusion

In this thesis we have discussed the development and successful implementation of an efficient method of loading ions into an ion trap based on a multi-step photo-ionization process. This required the implementation of a frequency reference based on multi photon spectroscopy using a barium vapor cell constructed on the principle of a heat pipe oven. There was a residual doppler shift in the ion trap. Through the spectroscopy of the transitions involved in the photo-ionization, we were able to lock the lasers to the appropriate atomic transitions so as to compensate the Doppler shifts and reliably load the desired ¹³⁷Ba isotope. This isotope selective loading can be readily modified for other isotopes and other trap geometries. Thus it will be a convenient and accessible technique for all future ion trap experiments within the micro-traps group.

For the spectroscopy of the 450nm transition, there is no known work in

literature that we can compare our experimental values to. Thus this work represents the first steps towards accurate measurements of this particular transition. Our spectroscopy was limited to the isotopes and hyperfine states of interest to this work but the system could be easily modified to include all isotopes and hyperfine states of the $6p^2$ ³P₁ level.

The barium vapor cell that we have constructed provides reliable operation over a period of a few months. However, there is a still a slow migration of barium to the cooler sections of the cell. This results in a need to run the cell at higher temperatures until ultimately there is a need to open the vacuum and reload more barium. This is only a minor inconvenience as this only needs to be done every few months. However the use of a quartz cell would allow the entire vapour cell to be heated including the windows. This may provide a long term solution to the problem.

Appendix A

Experimental Setup



Figure A.1: Barium vapor cell. The middle is wrapped with layers of ceramic wool and aluminum foil.



Figure A.2: Setup overview

Appendix B

Isotope shifts and Hyperfine

splittings

F state	Isotope pair	$\triangle v (\text{MHz})$
5/2	137-135	-143.4 (1.4)
3/2	137-135	-144.3 (1.4)
1/2	137-135	-360.4(1.4)

Table B.1: Isotope shifts of the ${}^{1}S_{0}$ to the ${}^{3}P_{1}$ transition at 791nm in barium of different F states due to odd isotopes having a non-zero nuclear spin [19].

From Eq. 2.25, when we leave the red laser to scan and the blue laser locked, we have

$$\omega_1' - \omega_2' = \frac{k_B}{k_R} [(\omega_R^{137} - \omega_R^{135}) - (\omega_2 - \omega_1)]$$
(B.1)

From Fig 4.1, the fits shows that the peak to peak distance between 135 Ba and 137 Ba is 0.299955, and from Fig 4.2, the peak to peak distance between

¹³⁵Ba and ¹³⁷Ba (F state=5/2) is 0.539 .We susptitute the values into the Eq B.1 above and the the values of k_B and k_R ,

$$\omega_1' - \omega_2' = \frac{\frac{2\pi}{450 \times 10^{-9}}}{\frac{2\pi}{791 \times 10^{-9}}} \left[\frac{143.3 \times (0.539)}{(0.299955)} - 143\right] = 200.6 MHz \tag{B.2}$$

Similarly, for 135 Ba and 137 Ba (F state=3/2), the peak to peak distance from Fig 4.2 is 0.5525, and we octain the following expression,

$$\omega_1' - \omega_2' = \frac{\frac{2\pi}{450\times10^{-9}}}{\frac{2\pi}{791\times10^{-9}}} [\frac{143.3\times(0.5525)}{(0.299955)} - 143] = 210.7MHz$$
(B.3)

For the hyperfine splittings, the equation to use is Eq 2.27,the peak to peak distance between 135 Ba (F state=5/2 and 3/2) is 0.0435 and 137 Ba (F state=5/2 and 3/2) is 0.03 in the 450nm transition lorentzian fits. We have for 135 Ba,

$$\omega_1' - \omega_2' = \frac{\frac{2\pi}{450 \times 10^{-9}}}{\frac{2\pi}{791 \times 10^{-9}}} [-(\omega_2 - \omega_1)] = 11.0MHz$$
(B.4)

and for ^{137}Ba ,

$$\omega_1' - \omega_2' = \frac{\frac{2\pi}{450 \times 10^{-9}}}{\frac{2\pi}{791 \times 10^{-9}}} [-(\omega_2 - \omega_1)] = 7.6MHz$$
(B.5)

If the blue laser is scanning and the red laser is unlocked, we obtain the

following equations from Eq 2.25, for the measurement of isotope shifts,

$$\omega_1' - \omega_2' = \omega_B^{135} - \omega_B^{137} - \left[\frac{k_B}{k_R}(-\omega_1 - \omega_2)\right]$$
(B.6)

For the measurement of hyperfine splittings, the equation is the same as Eq 2.27.

Appendix C

Doppler shift calculations

In this appendix we derive the effects of Doppler broadening for an oven geometry in which the atomic beam is at a 45° angle to the photon-ionization beams. Since the Doppler shifts will be dominated by the thermal spread of velocities along the beam axis we do not need to account for the angular spread of atomic velocities. The distribution of velocities along the axis of the atomic beam is given by [21]

$$f(v) = \frac{1}{2} \frac{v^3}{\bar{v}^4} \exp\left(-\frac{v^2}{2\,\bar{v}^2}\right)$$
(C.1)

where \bar{v} is a characteristic thermal velocity given by

$$\bar{v} = \sqrt{\frac{k_B T}{M}} \tag{C.2}$$

For given detuning, Δ the excitation rate to the excited state for a stationery atom is given by

$$R_e = \frac{\Gamma}{2} \frac{s_0}{1 + 4\Delta^2 / \Gamma^2} \tag{C.3}$$

where s_0 is the saturation parameter for the transition and Γ is the net linewidth [10]. For the atomic beam we must add up the contributions from all velocity classes giving

$$R_e = \frac{\Gamma s_0}{2} \int_0^\infty \frac{f(v)}{1 + 4(\Delta - \mathbf{k} \cdot \mathbf{v})^2 / \Gamma^2} dv$$
$$= \frac{\Gamma s_0}{2} \int_0^\infty \frac{f(v)}{1 + 4(\Delta - k_{eff}v)^2 / \Gamma^2} dv$$
(C.4)

where $k_{eff} = k \cos \theta = k/\sqrt{2}$ for the 45° geometry considered here. Assuming the linewidth, Γ , is much less than both the Doppler broadening and the detuning Δ we can replace f(v) with its value at the maximum of the Lorentzian to get

$$R_{e} = \frac{\Gamma s_{0} f(\Delta/k_{eff})}{2} \int_{0}^{\infty} \frac{1}{1 + 4(\Delta - k_{eff}v)^{2}/\Gamma^{2}} dv$$

$$= \frac{\Gamma s_{0} f(\Delta/k_{eff})}{2} \frac{\Gamma}{2k_{eff}} \int_{-\infty}^{2\Delta/\Gamma} \frac{1}{1 + x^{2}} dx$$

$$\approx \frac{\Gamma s_{0} f(\Delta/k_{eff})}{4} \frac{\Gamma}{k_{eff}} \int_{-\infty}^{\infty} \frac{1}{1 + x^{2}} dx$$

$$= \frac{\Gamma \pi s_{0} f(\Delta/k_{eff})}{4} \frac{\Gamma}{k_{eff}}$$

$$= \frac{\pi \Gamma s_{0}}{8} \left(\frac{\Gamma}{\Gamma_{D}}\right) \left(\frac{\Delta}{\Gamma_{D}}\right)^{3} e^{-\frac{1}{2}\left(\frac{\Delta}{\Gamma_{D}}\right)^{2}}$$
(C.5)

where we have introduced $\Gamma_D = k_{eff} \bar{v}$ which is a frequency scale associated with the Doppler broadening. In particular the distribution given by Eq C.5 has a maximum at $\sqrt{3}\Gamma_D$ and a full width half maximum (FWHM) of \approx 1.64 Γ_D . In terms of Γ_D , our assumptions used to derive Eq C.5 may be expressed as $\Gamma \ll \Gamma_D, \Delta$.

For the special case in which $\Delta = 0$ we must carry out the integral in Eq C.4 explicitly. With the aid of *Mathematica* we find, to second order in Γ/Γ_D ,

$$R_e(\Delta = 0) = \frac{\Gamma}{16} s_0 \left(\frac{\Gamma}{\Gamma_D}\right)^2 \tag{C.6}$$

We note that these are the excitation rates from ground state to excited state and neglects stimulated emission processes. This is valid provided the upper state is not significantly populations. This can still be the case for large values of s_0 provided the excitation rate for the next step in the photo-ionization rate is sufficiently fast.

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