Metastable D-state spectroscopy and laser cooling of barium

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Document Version
Publisher's PDF, also known as Version of record

Publication date:
2006

Link to publication in University of Groningen/UMCG research database

Citation for published version (APA):

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Chapter 5

Laser Cooling of Heavy Alkaline Earth Atoms

Modern atomic physics techniques like laser cooling and trapping of neutral atoms has become a vital tool for a variety of experiments. Of them, to mention are Bose-Einstein condensation, high precision measurements, optical frequency standards and studies on fundamental symmetries. The advantage of neutral atom trapping is that atoms can be confined in a small volume in high densities, at low kinetic energies providing long observation and storage times with excellent isotope selectivity. Furthermore, optical pumping allows to prepare the trapped sample in well defined atomic states. Laser cooling and trapping has so far been demonstrated with alkali elements lithium (Li) [142], sodium (Na) [143], potassium (K) [144, 145], rubidium (Rb) [146, 147], cesium (Cs) [148, 149] and francium (Fr) [150], light alkaline earth elements magnesium (Mg) [151], calcium (Ca) [152] and strontium (Sr) [152], except beryllium (Be), noble gases helium (He) [153], neon (Ne) [154], argon (Ar) [155], krypton (Kr) [155, 156] and xenon (Xe) [157], except radon (Rn), transition elements chromium (Cr) [158, 159] and silver (Ag) [160], ytterbium (Yb) [161] and erbium (Er) [162]. Recently, radium has been added to the list [55]. The weak intercombination line $^1S_0 - ^3P_1$ is sufficient for trapping, but the trapping efficiency is small.

The light alkaline earth elements have a strong $^1S_0 - ^1P_1$ transition, which can be approximated by a two-level system. Magnesium has a closed 3s$^2$ $^1S_0$ - 3s3p $^1P_1$ transition with all the metastable $D$-states higher than this transition making it a two level system [151]. Strontium and calcium atoms have low lying singlet $D$-states connected to the first excited $^1P_1$ state with negligible leakage rate. For calcium [163] and strontium [164] longer storage times in a MOT have
been demonstrated when an appropriate repumping laser is added to the scheme. In calcium, atoms from the singlet $D$-state are repumped via the higher $4s5p^1P_1$ state rather than to the $4s4p^1P_1$ state used in the cooling transition.

In contrast to the above, the atomic level structure is different in heavy alkaline earth atoms barium and radium. They have the metastable $D$-states lying lower than the excited $^1P_1$ state. In barium the strong $6s^2\ 1S_0 - 6s6p\ 1P_1$ transition is the only option for cooling, while radium offers the possibility to use the weaker intercombination line $^1S_0 - ^3P_1$ for cooling in addition to the $7s^2\ 1S_0 - 7s7p\ 1P_1$ transition [55].

In barium, the leakage rate to the $D$-states is so large that laser cooling without repumping lasers is not possible. In addition, repumping has to go through the $6s6p\ 1P_1$ state, because excitation to higher levels would allow branching into many more states. Since the cooling transition and the repumping transition have a common excited state, we expect coherent Raman resonances. These will be discussed in detail in Section 5.1, together with their implication for laser cooling.

To illustrate the problem of repumping we will discuss briefly laser cooling of chromium. The level structure with its $\Lambda$-systems is shown in Fig. 5.1. Laser cooling and trapping has been demonstrated with and without repumping from the two metastable $D$-states and the cooling transitions use a common excited state [159]. Of all trapped atomic systems it has the largest similarity to laser

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**Figure 5.1:** Partial term scheme of chromium used for laser cooling and trapping experiment [159].
cooling of barium. We give a simple estimate to emphasize the issues for the case of barium.

In this estimate we compare the average velocity change of an atom without repumping. The average velocity change is given by

$$\Delta v = A_{\text{leak}}^{-1} \times v_r$$

(5.1)

where $A_{\text{leak}}^{-1} = 2500$ is the leak rate to the $D$-states in chromium and the recoil velocity $v_r = \hbar k/m = 1.8$ cm/s. Thus a velocity change of $\Delta v = 45$ m/s can be achieved, while still a fraction of $1/e$ of the atoms remain in the cooling cycle. Below 45 m/s there is a fraction $1 \times 10^{-5}$ of the atoms in thermal distribution from their atomic source. It was reported that less than 100 atoms could be trapped without repumping lasers. Adding the repumping of atoms using the additional diode lasers from the $D$-states helped to increase the number of atoms in the MOT by a factor of 45 and to achieve longer storage times. The repump lasers are focused on the trapped sample and high intensities are easily achieved. The repumping transitions are in the visible region that are conveniently accessible with diode lasers at 658.3 nm and 649.2 nm respectively.

Now we estimate what fraction of barium atoms we could stop without repump lasers. The branching ratio to the three $D$-states is given in Eq. 4.1. On average only $A_{\text{leak}}^{-1} = 330(30)$ photons are scattered before the atom is in one of the $D$-states. Also the recoil velocity $v_r = 0.52$ cm/s is smaller than for chromium. Using the relation 5.1, we arrive at a velocity change of only $\Delta v = 1.7$ m/s. At these low velocities there are no atoms in an atomic beam, because of collisions in the nozzle. This effect is known as the Zacharias effect [33].

To bring a barium atom from a velocity of 45 m/s to rest the atom has to scatter about $N_{45m/s} = 8650$ photons. This is $N_{45m/s}/A_{\text{leak}}^{-1} = 26$ times more than $A_{\text{leak}}$. A fraction of only

$$P_{cc} = \exp \left( -\frac{N_{45 m/s}}{A_{\text{leak}}^{-1}} \right) = 10^{-11}$$

(5.2)

remain in the cooling cycle. This makes trapping without repumping not feasible. To achieve a fraction of $1/e$ of the atoms remaining in the cooling cycle the atoms have to be optically pumped out of the metastable states about 26 times.

The time the atoms spend in the metastable $D$-states depends on the optical pumping rate, which is related to the Rabi frequency (Eq. 5.41). In addition, this drives two photon Raman transitions from the ground state to the metastable $D$-states. The consequence is that depending on the detuning of the repump lasers
and cooling laser the atoms can get transferred efficiently into the metastable states. On average this increases the population compared to a situation in which the optical pumping can be done via another level. In atoms with nuclear spin other states of the hyperfine manifold can be used for repumping.

Though Raman transitions are present for barium, they are not a strong constraint for laser cooling. At high enough intensities for the repump lasers we get an equal probability to find the atom in one of the states connected by the laser transitions. Depending on the number \( N \) of states involved, for barium, the ground state and three metastable \( D \)-states, we will have a fraction of \( 1/N \) of the atoms in each state.

We will describe the theoretical treatment and the experimental observation of coherent Raman resonances. Using these results we discuss the setup for slowing a barium atomic beam by light forces. We show the first successful slowing of barium and we give an analysis of slowing results using our knowledge of Raman resonances in a dynamic calculation.

### 5.1 \( \Lambda \)-System

#### 5.1.1 Theoretical Treatment of Coherent Raman Resonances in a \( \Lambda \)-System

Atoms having a common excited state with multiple ground states exhibit the phenomenon of coherent Raman resonances. As an example, consider a three-level system, having two ground states connected to a common excited state, a \( \Lambda \)-system. In the \( \Lambda \)-configuration the atoms interacting with near-resonant bichromatic light fields show additional resonances (Fig. 5.2).

When the detuning \( \Delta \) from the excited state of both lasers is the same, atoms are transferred into a superposition of state \( |1\rangle \) and \( |3\rangle \) being the two groundstates which is expressed by an additional two photon Rabi frequency \( \Omega_{13} \). Hence, resonant excitation and fluorescence are suppressed. To the observer this process appears as a *dark resonance* with a narrow line width, which is in principle determined by the excited state properties. Experimentally, in 1976, dark resonances were first observed in sodium [165]. A review on developments and status of the coherent dark state spectroscopy experiments can be found in for example [166, 167].

For the theoretical discussion we consider the \( \Lambda \)-system with the \( ^1S_0, ^1P_1 \) and one of the metastable states. In particular we use the \( ^1D_2 \) state as an example. This treatment can easily be extended to a four level system.
Interaction of a $\Lambda$-system with Coherent Light Fields

The energy level scheme of the barium atom used for the calculation is shown in Fig. 5.2. The $^1P_1$ excited state decays to the $^1S_0$ ground state and weakly to the $^1D_2$ metastable state. With the bichromatic excitation, the scattered light of the barium atom contains spectral components at 553.7 nm and negligible 1500.4 nm light.

A theoretical model describing the atom-laser interaction for a three-level $\Lambda$-system, interacting with two coherent light fields is described. The formalism adapted here is similar to the calculations for the barium ion [168, 169]. Here the atom is treated quantum mechanically where as the coherent laser fields are treated as classical electromagnetic waves $\vec{E}_g \sin(\omega_g t)$ and $\vec{E}_r \sin(\omega_r t)$ as they are of high photon numbers. They have linewidths $\Gamma_g$ and $\Gamma_r$ respectively. The spontaneous emission of photons is treated as a decay from $|2\rangle$ to $|1\rangle$ and $|3\rangle$ with the rates $\Gamma_{21}$ and $\Gamma_{23}$. The quantities $\Omega_{12}$ and $\Omega_{32}$ are Rabi frequencies and denote the strength of the coupling between the atom and the electric field.

The Hamiltonian of the atom, using the states 1, 2 and 3 as the basis, is
defined by

\[ \hat{H}_{\text{atom}} | a \rangle = \hbar \omega_a | a \rangle \quad \text{with} \quad a = 1, 2, 3 \]  \hfill (5.3)

with \( \omega_a \) being the atomic Bohr frequencies. The atom-laser interaction is restricted to the dipole interaction. Then the interaction Hamiltonian can be written as:

\[ \hat{H}_{\text{int}} = -\left\{ \bar{D}_{12} \bar{E}_g i |2\rangle \langle 1| e^{-i\omega_g t} - |1\rangle \langle 2| e^{i\omega_g t} \right\} + \bar{D}_{32} \bar{E}_r i |2\rangle \langle 3| e^{-i\omega_r t} - |3\rangle \langle 2| e^{i\omega_r t} \right\} \]  \hfill (5.4)

where \( \bar{D}_{12} \) and \( \bar{D}_{32} \) are the dipole matrix elements for the transition from \( |1\rangle \) to \( |2\rangle \) and \( |3\rangle \) to \( |2\rangle \), respectively. Here the terms which rotate at twice the optical frequency and the non-resonant terms e.g., \( \bar{D}_{12} \bar{E}_r \) are neglected (rotating wave approximation) [170].

The Hamiltonian can also be written in matrix formalism with

\[ \begin{pmatrix} c_1 \\ c_2 \\ c_3 \end{pmatrix} \equiv c_1 |1\rangle + c_2 |2\rangle + c_3 |3\rangle \]  \hfill (5.5)

\[ H_{\text{atom}} = \hbar \begin{pmatrix} \omega_{12} & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & \omega_{32} \end{pmatrix} \]  \hfill (5.6)

with \( \omega_{12} = \omega_1 - \omega_2 \) and \( \omega_{32} = \omega_3 - \omega_2 \). The energy zero has been set to \( |2\rangle \). The interaction Hamiltonian written as

\[ H_{\text{int}} = \hbar \begin{pmatrix} 0 & \frac{\Omega_{12}}{2} e^{-i\omega_g t} & 0 \\ \frac{\Omega_{12}}{2} e^{i\omega_g t} & 0 & \frac{\Omega_{32}}{2} e^{-i\omega_r t} \\ 0 & \frac{\Omega_{32}}{2} e^{i\omega_r t} & 0 \end{pmatrix} \]  \hfill (5.7)

where

\[ \hbar \Omega_{12} := \bar{D}_{12} \bar{E}_g \]  \hfill (5.8)

\[ \hbar \Omega_{32} := \bar{D}_{32} \bar{E}_r \]  \hfill (5.9)

The complete Hamiltonian can be written as
\[ H = H_{\text{int}} + H_{\text{atom}} = \hbar \left( \begin{array}{ccc} \omega_{12} & \frac{\Omega_{12}}{2} e^{-i\omega_0 t} & 0 \\ \frac{\Omega_{12}}{2} e^{i\omega_0 t} & 0 & \frac{\Omega_{32}}{2} e^{-i\omega_0 t} \\ 0 & \frac{\Omega_{32}}{2} e^{i\omega_0 t} & \omega_{32} \end{array} \right) \] (5.10)

The Hamiltonian does not describe the spontaneous decay of the \(|2\rangle\) level into the \(|1\rangle\) and \(|3\rangle\) levels. To understand our measurements it is not necessary to use a full quantum-mechanical treatment of the electromagnetic fields. It is sufficient to treat the spontaneous decay as a rate \(\Gamma_{21}\) (\(\Gamma_{23}\)) with which the state \(|2\rangle\) decays into \(|1\rangle\) (\(|3\rangle\)). The resulting state of the atom is no longer a pure state and one can use the density matrix formalism.

**Density Matrix Formalism**

The density operator \(\hat{\rho}\), written in the basis of atomic eigenstates \(|a\rangle\), is

\[ \hat{\rho} = \sum_{a,b=1,2,3} \rho_{ab} \langle a | b \rangle \] (5.11)

The elements \(\rho_{11} = \langle 1 | \rho | 1 \rangle\), \(\rho_{22}\) and \(\rho_{33}\) are the expectation values for finding the atom in the states \(|1\rangle\), \(|2\rangle\) and \(|3\rangle\) respectively, such that

\[ \text{Trace}(\hat{\rho}) = \rho_{11} + \rho_{22} + \rho_{33} = 1 \] (5.12)

The time evolution of the density operator is governed by the Liouville equation

\[ \frac{d\hat{\rho}}{dt} = -\frac{i}{\hbar} [H, \hat{\rho}] + \mathcal{L}_{\text{damp}}(\hat{\rho}) \] (5.13)

where the first part corresponds to the Schrödinger equation and \(\mathcal{L}_{\text{damp}}\) describes the spontaneous decay of the upper state. The operator \(\mathcal{L}_{\text{damp}}\) has the general form

\[ \mathcal{L}_{\text{damp}}(\hat{\rho}) = -\frac{1}{2} \sum_m [\hat{C}^\dagger_m \hat{C}_m \hat{\rho} + \hat{\rho} \hat{C}^\dagger_m \hat{C}_m - 2\hat{C}_m \hat{\rho} \hat{C}^\dagger_m] \] (5.14)

The operators \(\hat{C}^\dagger_m \hat{C}_m\) describe the different dissipative processes. The first two terms \(\hat{C}^\dagger_m \hat{C}_m \hat{\rho}\) and \(\hat{\rho} \hat{C}^\dagger_m \hat{C}_m\) describe the decay of the excited levels. The term \(2\hat{C}_m \hat{\rho} \hat{C}^\dagger_m\), which is called feeding term, describes the decay into the lower states. Here this is the decay from \(|2\rangle\rightarrow |1\rangle\) and \(|2\rangle\rightarrow |3\rangle\). Also, the decoherence through the finite bandwidth \(\Gamma_g\) and \(\Gamma_r\) of the driving laser fields can be included in \(\mathcal{L}_{\text{damp}}\).
In this case of three level atom we have the decay from the $P$ level to the $S$ level, described by

$$\hat{C}_{21} = \sqrt{1_2} | 1 \rangle \langle 2 |$$

(5.15)

and from the $P$ to the $D$ level

$$\hat{C}_{23} = \sqrt{1_3} | 3 \rangle \langle 2 |$$

(5.16)

The finite linewidth of the lasers can be introduced by the operators

$$\hat{C}_g = \sqrt{2\Gamma_g} | 1 \rangle \langle 1 |$$

(5.17)

$$\hat{C}_r = \sqrt{2\Gamma_r} | 3 \rangle \langle 3 |$$

(5.18)

By writing

$$\frac{d\hat{\rho}}{dt} = \mathcal{L} \hat{\rho}(t)$$

(5.19)

with

$$\mathcal{L} \hat{\rho} = -\frac{i}{\hbar} [H, \hat{\rho}] + \mathcal{L}_{damp}(\hat{\rho})$$

(5.20)

the density operator $\rho$ at the time $t$ can now be expressed in terms of $\rho(0)$ by

$$\hat{\rho}(t) = e^{\mathcal{L}t} \hat{\rho}(0)$$

(5.21)

Finally, we transform the system into the rotating frame of the laser light fields with

$$U = \begin{pmatrix} e^{-i\omega_0 t} & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & e^{-i\omega_0 t} \end{pmatrix}$$

(5.22)

and get the transformed density and Hamiltonian operators

$$\hat{\rho}' = U \hat{\rho} U^\dagger$$

(5.23)

$$H' = UHU^\dagger - i\hbar U \frac{dU^\dagger}{dt}$$

(5.24)

The resulting Hamiltonian $H'$ is
\[ H' = \hbar \begin{pmatrix} \Lambda_g/2 & \Omega_{12}/2 & 0 \\ \Omega_{12}/2 & 0 & \Omega_{23}/2 \\ 0 & \Omega_{23}/2 & \Delta_r \end{pmatrix} \] 

(5.25)

with the detunings

\[ \Delta_g = \omega_g - \omega_{12} \] 

(5.26)

\[ \Delta_r = \omega_r - \omega_{32} \] 

(5.27)

The damping terms remain unchanged.

**Optical Bloch Equations**

The Liouville equation 5.13 can be transformed into a system of linear equations by setting

\[ \vec{r} = (r_{11}, r_{12}, ..., r_{23}, r_{33}) \] 

(5.28)

\[ \frac{d\vec{r}}{dt} = \sum_j M_{ij} \vec{r}_j \] 

(5.29)

where \( \rho_{ab} = \langle a | \vec{r} | b \rangle \) and we have introduced the \( N^2 \times N^2 \) matrix \( M \). These equations are called optical Bloch equations. The time evolution \( \vec{r}(t) \) with a given \( \vec{r}(0) \) is

\[ \vec{r}(t) = \exp(Mt)\vec{r}(0). \] 

(5.30)

The matrix \( M \) is uniquely determined by \( \mathcal{L} \) in Eq. 5.13, i.e., it contains all information relevant for the evolution of the system, i.e., the laser intensities, detunings and decay constants. All the terms have been introduced in such a way that \( \rho \) remains normalized, i.e.,

\[ \sum_i \rho_{ii}(t) = 1 \] 

(5.31)

In order to obtain the steady state solution (\( \vec{r}(\infty) = \text{constant} \leftrightarrow \vec{r}=0 \)) is given by

\[ \sum_j M_{ij} \vec{r}_j = 0 \] 

(5.32)
To solve Eq. 5.32 with the correct normalization, one of the equations must be replaced by the normalization condition Eq. 5.31. The time evolution of $\tilde{\rho}(t)$ and the steady state solution $\tilde{\rho}(\infty)$ are calculated numerically with a computer program written in matlab [169,171].

The numerical approach allows us to calculate the Raman spectrum. In order to compare the numerical result to the experiment we have to include the finite time the atom spends in the laser beam. Since in the experiments the laser beams are perpendicular to the atomic beam in order to reduce the Doppler broadening, the time is given by the laser beam diameter and the velocity of the atoms. We use a Maxwell-Boltzmann velocity distribution for the atoms.

Depending on the detuning of the $^1D_2 - ^1P_1$ transition laser (1500.4 nm) with respect to the $^1S_0 - ^1P_1$ transition (553.7 nm) frequency the atoms are driven coherently between the $S$ and the $D$-states without passing through the $P$ state. If both detunings are equal the effective Rabi frequency can be approximated to

$$\Omega_{13} = \frac{\Omega_{12}\Omega_{32}}{2\Delta}$$

under the approximation that the detuning $\Delta$ is much larger than the linewidth $\Gamma$ of the transition. The dark resonance spectrum calculated with the parameters used in the experiments is shown in Fig. 5.5. Because we operate in the limit of $\Delta$ on the order of linewidth of the transitions we have to solve the full optical Bloch equations.

### 5.1.2 Coherent Raman Resonances: Experiment

The two photon Raman transition allowed us to determine the Rabi frequency of the repumping transitions. We will discuss the $^1D_2 - ^1P_1$ transition in detail. The experimental setup used to observe the dark resonances in a $\Lambda$-system is shown in Fig. 5.3. It consists of the 553.7 nm laser and the 1500.4 nm laser.

<table>
<thead>
<tr>
<th>Transition</th>
<th>Rabi frequency in rad/s @ $I = 1\text{mW/cm}^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^1S_0 - ^1P_1$</td>
<td>$22 \cdot 10^6$</td>
</tr>
<tr>
<td>$^1D_2 - ^1P_1$</td>
<td>$4.7 \cdot 10^6$</td>
</tr>
<tr>
<td>$^3D_2 - ^1P_1$</td>
<td>$2.0 \cdot 10^6$</td>
</tr>
<tr>
<td>$^3D_1 - ^1P_1$</td>
<td>$0.32 \cdot 10^6$</td>
</tr>
</tbody>
</table>

Table 5.1: Rabi frequencies for different transitions in barium. The numbers are calculated based on the wavelength and the decay rates to the different states.
Figure 5.3: Setup to observe the coherent Raman resonances. A natural barium atomic beam is used. 553.7 nm laser light and 1500.4 nm laser light are overlapped and passed perpendicular to the barium atomic beam to reduce the Doppler broadening. The fluorescence from the 6s6p $^1P_1$ state is detected with a PMT.

The two laser beams are passed perpendicular to the barium atomic beam and are counter propagating. They are collimated and are overlapped along the path of interaction with the atomic beam. The beam radius of the 1500.4 nm laser was 1.0(0.2) mm with Gaussian beam profile and the 553.7 nm laser beam was shaped by a rectangular aperture of size $3 \times 3$ mm$^2$. The fluorescence signal from the $^1P_1 - ^1S_0$ transition is detected by a photomultiplier. This probes the population in the excited state or $\rho_{22}$ of the density matrix. We compare the experimental spectra with numerical calculations taking the geometry of the setup into account. In particular, the laser beam at 553.7 nm was about a factor of two larger in diameter than the infrared laser and we have to take into account the time the atoms spend in the laser beams depending on their velocity.

The 1500.4 nm laser was red detuned by $\Delta = -13.5(5)$ MHz with respect to the $^1D_2 - ^1P_1$ transition of the $^{138}$Ba resonance and the 553.7 nm laser frequency was scanned across the resonance. The output power of the 1500.4 nm laser used was 10(1) mW corresponding to an intensity of 320(120) mW/cm$^2$. This corresponds to a Rabi frequency of $\Omega_{32} = 90(20) \cdot 10^6$ rad/s. For the 553.7 nm laser, a power
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Figure 5.4: Fluorescence spectrum of the $^1P_1$ state detected with a beam of natural barium. The upper figure (a) has the 1500.4 nm laser frequency detuned by -13.5 MHz with respect to the $^{138}$Ba resonance. The solid line is the result of the optical Bloch equations with parameters $\Omega_{12}=120 \cdot 10^6$ rad/s for the 553.7 nm laser and $\Omega_{32}=100 \cdot 10^6$ rad/s. The lower figure (b) is in the absence of the repump laser. The line is the result of the optical Bloch equation calculation with the parameters $\Omega_{12}=120 \cdot 10^6$ rad/s. For both calculations the laser linewidth for the 553.7 nm laser $\Gamma_s=1$ MHz and $\Gamma_r=50$ kHz for the 1500.4 nm laser. The spectra are normalized to the peak amplitude. The only parameter, which has been adjusted is the ratio of the coherent Raman transition fraction to the ordinary fluorescence spectrum. The latter is about 45% of the $^{138}$Ba fluorescence.
of 2.2(1) mW was used corresponding to an intensity of 24(1) mW/cm$^2$ or a Rabi frequency of $\Omega_{12} = 120(3) \cdot 10^6$ rad/s respectively. The laser linewidth for the two lasers is much less than the Rabi frequencies and the decay rate of the $^1P_1$ state.

In Fig. 5.4(a) we show the observed spectrum which contains different barium isotopes. The dip on the left side of the $^{138}$Ba resonance is the two photon Raman transition. The fluorescence is reduced because of the direct coupling of the $^1S_0$ ground state and the $^1D_2$ state. The depth of the observed dip contains information on the Rabi frequency $\Omega_{32}$. Further, the size of the dip depends on the overlapping of the two laser beams and their alignment with respect to the atomic beam. Shown in Fig. 5.4(b) is the spectrum in the absence of the 1500.4 nm
laser. The solid line in both figures is the numerical solution of the optical Bloch equations. The parameters are in good agreement with the calculated Rabi frequencies.

Dark resonances can also be observed in another experimental configuration. For this, the 553.7 nm laser was frequency locked to the peak of the $^{138}$Ba isotope resonance using the Doppler shift method described in Section 4.10 and the 1500.4 nm laser frequency was scanned over the resonance. The spectrum is shown in Fig. 5.5. As long as the 1500.4 nm laser detuning is different from the detuning at 553.7 nm we observe the unperturbed scattering from the $^{1}S_{0}^{1}P_{1}$ transition. When the 1500.4 nm laser and the 553.7 nm laser have the same detuning $\Delta$ the scattering rate decreases, because of coherent population transfer to the metastable $^{1}D_{2}$ state. The width of the observed dip is 20(1) MHz, in good agreement with the calculation. This indicates that Doppler broadening due to improper overlapping was small. We again get a good agreement with the result from optical Bloch equation with a Rabi frequency $\Omega_{32}$ of $120 \cdot 10^{6}$ rad/s, and a fraction of 50% of the atoms passing only through the 553.7 nm laser beam.

The determined Rabi frequency is of the order of $10^{8}$ rad/s. Thus we can expect that we achieve Rabi frequencies larger than $10^{7}$ rad/s with a beam diameter of 5 mm with the available laser power of 10 mW from the fiber laser. For the repumping of the $^{3}D_{2}$ state we expect similar Rabi frequencies at the same power (Table 5.1).

### 5.2 Laser Cooling of Barium

In this section we discuss aspects of laser cooling relevant for our system. A detailed information on laser cooling in general can be found elsewhere [16]. A simple approach to describe the laser cooling principle is to consider a closed two level atomic system. One parameter that plays an important role is the scattering rate $\gamma_p$. It is defined as the ratio of population of the excited state $P_e$ to the life time $\tau$ of the excited state. It is written as

$$\gamma_p = \frac{P_e}{\tau} = \frac{\gamma}{2 \frac{S}{1 + S}} \frac{1}{1 + (\frac{2 \delta}{\gamma})^2}$$  (5.34)

where $\gamma$ is the decay rate and $\gamma' = \gamma \cdot \sqrt{1 + S}$ is the power broadened linewidth of the atomic transition. $\delta = \omega_{laser} - \omega_0$ is the detuning of the laser frequency $\omega_{laser}$ with respect to the $\omega_0$ of the atomic transition. Power broadening becomes important when the intensity of the laser is greater than the saturation intensity. $S$ is the saturation parameter of the atomic transition that is defined as
\[ S = \frac{I}{I_s} = 2 \frac{\Omega^2}{\Gamma^2} \]  
(5.35)

where \( \Omega \) is the Rabi frequency and \( \Gamma \) is the linewidth of the transition and \( I_s \) is the saturation intensity defined as

\[ I_s = \frac{\pi hc}{3\lambda^3 \tau} \]  
(5.36)

where \( h \) is the Planck constant, \( c \) is the velocity of light and \( \lambda \) is the wavelength of the atomic transition.

### 5.2.1 Requirements for Slowing Barium

To demonstrate the slowing of an atomic beam of barium we have chosen the cooling beam to be counter propagating to the atomic beam. The repumpers are also counter propagated to the atomic beam. This limits the range of cooling force to a part of the velocity spectrum from an effusive beam depending on the linewidth of the laser. The central velocity which is effected depends on the laser detuning.

Here we give a rough estimate of the time the atoms spend in the metastable states and compare them with our experimental values reported in Section 5.2.3. For stopping barium atoms of velocity \( v \), assuming a constant acceleration \( a \), over a distance \( d \) a time \( \delta t \)

\[ \delta t = \frac{2d}{v}, \]  
(5.37)

is needed. The constant acceleration can be achieved by chirping the laser frequency or by broadening of the spectral width of the lasers.

During the slowing process the atom needs to scatter \( N_p = v/v_r \) photons, where \( v_r \) is the single photon recoil velocity. The minimum scattering rate \( \gamma_m \) to achieve a sufficient deceleration

\[ \gamma_m = \frac{N_p}{\delta t}, \]  
(5.38)

which has to be smaller than \( \gamma_p \) given in Eq. 5.34. We estimate the fraction of time the atoms have to be in the cooling cycle in order to achieve this scattering rate. Here we assume that all the lasers are on resonance all the time. This could be achieved by chirping the lasers. Assuming that we work at saturation intensity for the cooling transition we find in steady state a fraction of
of the atoms in the ground state. This is the result from a rate equation, which does not describe the coherent Raman transitions which we will discuss in detail below. In addition, driving the cooling transition at saturation intensity results in an excited state population of \( f/4 \). The atom will be in the metastable states for the rest of the time.

With these assumptions we calculate the required repumping rate \( r \), written as

\[
r = \frac{N_p}{A_{\text{leak}} \delta t (1 - 5f/4)}
\]

(5.40)

For a weak excitation the repumping rate \( r_r \) is determined by the Rabi frequency \( \Omega_{32} \) at the repump wavelength and the decay rate \( \Gamma_{21} \) of the cooling transition,

\[
r_r = \frac{\Omega_{32}^2}{\Gamma_{21}}.
\]

(5.41)

As a numerical example, we assume a starting velocity of \( v = 120 \text{ m/s} \) for the atoms. The distance over which atoms are interacting with the lasers in our apparatus is \( d = 0.6 \text{ m} \). This gives a slowing time \( \delta t = 10 \text{ ms} \) (Eq. 5.37). To stop the atoms the number of scattered photons \( N_p \) is then 23000 and the minimum scattering rate \( \gamma_m \) equates to \( 2.3 \times 10^6/\text{s} \) (Eq. 5.38). Assuming that the cooling beam at 553.7 nm is at saturation intensity, we arrive at a fraction of at least \( 5f/4 = 0.1 \) (Eq. 5.39) in which the atoms have to be in the cooling cycle. Thus the repumping rate has to be larger than 8000/s (Eq. 5.40). The minimal required Rabi frequency \( \Omega_{32} \geq 10^6 \text{ rad/s} \) corresponds to an intensity of about 0.1 mW/cm\(^2\) (1500.4 nm). Higher intensities would allow for larger cooling forces. This low intensity would be sufficient if the cooling transition and the repumping transition would not use a common excited state.

For investigating the effect of the coherent Raman transition on the repumping rate we have solved the optical Bloch equations for the three different level configurations (Fig. 5.6). The cooling force is proportional to the number of scattered photons, which is

\[
N_{\text{scatter}} = \frac{\rho_{22}}{\tau}
\]

(5.42)

where \( \rho_{22} \) is the probability to be in the excited state and \( \tau \) is the lifetime of the excited state.
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We compare the requirements for the different situations. A lower repump laser intensity can be used when different excited states are involved. The result for the population of the excited state is shown in Fig. 5.7. The calculation was done for different detunings at $\Omega_{\text{repump}} = 3.5 \times 10^6 \text{ rad/s}$ and on resonance at saturation intensity of the cooling transition. We see that the population $\rho_{22}$ in the excited state is about 0.13. The calculation for the three level $\Lambda$-system was done for different linewidths of the repump laser. A larger linewidth effectively washes out the two-photon resonance, but the area under the curve gets smaller, indicating lower efficiency in recycling to the cooling transition. The population of the ground state is 0.04 for the two photon resonance. Integrating over the full spectrum the repumping is about a factor of two less efficient than in case (a) and the Rabi frequency should be larger by this factor to achieve the same effective cooling.

In a more realistic case of the four-level system with ($\Omega_{23} = 3.5 \times 10^6 \text{ rad/s}$ and $\Omega_{24} = 2.5 \times 10^6 \text{ rad/s}$), the population of the excited state appears to be even less. These Rabi frequencies correspond to 0.75 mW/cm$^2$ and 1.6 mW/cm$^2$ respectively. It is at maximum 0.03, which in the above numerical example was determined as the minimal required value for the population. In order to understand laser cooling with strong repumping requirements a full simulation of the slowing process might be helpful.

Since the detuning of the lasers from the resonance is Doppler sensitive
Figure 5.7: $^1P_1$ state ($\rho_{22}$) population at different linewidths of the repump lasers: (a) non-coherent population probability of $^1P_1$ state, (b) 1 MHz, (c) 5 MHz, (d) 10 MHz and (e) 20 MHz.

the lasers are not always in resonance. While the velocity changes, they move through the resonance. As a result, we expect that we need higher intensities in the experimental realization than from this simple estimate.

5.2.2 Different Slowing Methods

Neutral thermal atomic beams have been cooled by various methods. Of them to mention are Zeeman slower method [172], frequency chirping of lasers [173], low-velocity intense source method [174], white light cooling technique [175, 176] and two step deceleration of an atomic beam [177]. Here we discuss briefly the advantages and disadvantages of these methods for barium atoms.

Zeeman Slowing Technique

During laser cooling, an atom is decelerated as it receives the momentum of photons from a laser beam counter propagating to its direction of motion. While...
slowing an atom with a suitable wavelength laser light each absorbed photon gives a momentum 'kick' of $\Delta p = \hbar k$ to a slowing atom. Subsequently, to bring it to rest tens of thousands of photons have to be scattered as quickly as possible. With the method of decelerating atoms by Zeeman tuned slowing, atoms pass through a spatially varying magnetic field that is designed to keep the atomic transition resonant with the laser by compensating the decelerated atom’s changing Doppler shift with an equal but opposite Zeeman shift[172].

Decelerating barium atoms using Zeeman slower method may not be a feasible option because of the need of repump lasers. The barium $^{138}\text{Ba}$ ($I = 0$) atoms in an external magnetic field experience Zeeman splitting. The $^1P_1$ singlet state has $3\ m_J$ states. The $^1D_2$ state has $5\ m_J$ states and the $^3D_{1,2}$ states have 3 and 5 $m_J$ states respectively. For perfect circular polarization the excited state decays into 3 magnetic substates of $^1D_2$ and $^3D_2$ and 2 states in $^3D_1$. The $g_J$ factors for these transitions are given in Table 5.2.

In practice the polarization is not perfect and the other magnetic substates are populated. The energy level splitting in an external magnetic field is

$$\Delta f = \frac{m_J \ g_J \ \mu_B}{h} \ B \quad (5.43)$$
where \( m_J \) is the magnetic quantum number, \( g_J \) is the Lande \( g \)-factor of the state, \( \mu_B \), Bohr magneton, \( h \) is the Planck constant and \( B \) is the external magnetic field.

The magnetic field \( B_{\text{max}} \) for a Zeeman slower aiming for an initial velocity of \( v_{\text{max}} \) of the atom is

\[
B_{\text{max}} = \frac{v_{\text{max}}}{\lambda \cdot g_J \cdot m_J \cdot \mu_B}
\]  

(5.44)

where \( \lambda = 553.7 \text{ nm} \) is the wavelength of the \( ^1S_0-^1P_1 \) transition. For a barium atom of velocity \( v_{\text{max}} = 350 \text{ m/s} \) we arrive at a magnetic field of 450 G and a frequency splitting of 630 MHz. To cover the entire Zeeman splitting range for all the substates of the metastable \( D \)-levels, the repump lasers need to cover a frequency range of

\[
\Delta f_{\text{rep}} = 2(g_J^P \cdot m_{j_{\text{max}}}^P - g_J^D \cdot m_{J_{\text{max}}}^D) \mu_B \text{ MHz}.
\]

(5.45)

Thus \( \Delta f_{\text{rep}} \) is on the order of twice the Zeeman splitting of the \( ^1P_1 \) state. In a detailed analysis one has to take the selection rules for the dipole transitions in a

| \( g_J \) values of levels in barium |
|---|---|---|---|---|
| Level | \(^1S_0\) | \(^1P_1\) | \(^1D_2\) | \(^3D_2\) | \(^3D_1\) |
| \( g_J \) | 0 | 1 | 1 | 7/6 | 1/2 |

**Table 5.2:** The \( g_J \) values of the levels involved in laser cooling of barium.
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magnetic field into account.

However, the Doppler shift is inversely proportional to wavelength $\lambda$ of the transition. Thus the Doppler shift cannot be compensated for all the transitions at the same time and a broad spectrum of the repump lasers is required. The situation is complicated in the case of isotopes with nuclear spin ($I \neq 0$) because of the large hyperfine structure.

Frequency Broadened Lasers

Another approach to achieve laser cooling of atoms is using a broad frequency spectrum of the laser light for the cooling transition, also called white light cooling [175]. In this method, there is always some portion of the light spectrum that is in resonance to compensate change in the velocity of the atom. For barium this may be an option. The broad frequency spectrum of the repump lasers is necessary to keep the Doppler shifted atoms in resonance.

We estimate the required intensity to $2-4 \text{ mW/cm}^2$ in $19 \text{ MHz bandwidth}$ to achieve a Rabi frequency of $5 \cdot 10^6 \text{ rad/s}$. To cover a range up to the most probable velocity, which is 350 m/s, one needs a bandwidth of around 230 MHz for the 1500.4 nm or 12 times the power, and 310 MHz for the 1130.6 nm or 16 times the power required in a 19 MHz bandwidth.

The frequency broadening of the transition can be achieved by using an EOM with frequency side bands spacing of about the linewidth of the transition. To achieve the same bandwidth by power broadening of the transition one needs much more power, since Rabi frequencies of 1 GHz would be required. This corresponds to intensities for example $\approx 50 \text{ W/cm}^2$ (1500.4 nm), respectively $\approx 250 \text{ W/cm}^2$ (1130 nm). These intensities pose significant limitations in an experiment.

5.2.3 Experimental Results of Laser Cooling of Barium

The estimates for the required intensities show that the best way to achieve slowing of a barium atomic beam is with overlapping cooling and repumping lasers against the atomic beam. The experimental setup is shown in Fig. 5.10. We used enriched barium ($^{138}\text{Ba}$) because fluorescence from slow $^{138}\text{Ba}$ atoms overlaps with the fluorescence of other isotopes at higher velocity which are more abundant. In addition, $^{138}\text{Ba}$ has no hyperfine structure. The 553.7 nm laser provides light for the cooling beam counter propagating to the atomic beam and a probe beam to measure the velocity distribution at 45° relative to the atomic beam.
The frequencies needed for the cooling and probe beams are generated using acousto optic modulators (Fig. 5.10). The acousto optic modulator AOM1 is used for the output power stabilization of the 553.7 nm laser (Section 4.4.1). The acousto optic modulator denoted as AOM2 is used as a variable beam splitter to provide power for cooling and probe beams. With the RF power to AOM2 we regulate the power into the zero order and first order beams. The zero order beam is passed through AOM3. We use ‘+1’ order that is frequency shifted by \( f_3 \) relative to the output of the dye laser. It is utilized as cooling beam with 30 mW power available for the experiment. A fraction of the laser light (1 - 2 mW) is used in a second atomic beam apparatus to provide an absolute frequency reference to the barium transition.

The ‘-1’ order beam, which is frequency shifted by AOM2 \( (f_2 = 60 \text{ MHz}) \), is passed through AOM4 \( (f_4) \) to produce the probe beam. We use AOM4 in double pass configuration for the ‘+1’ order to allow for scanning the frequency without changing the position of the laser beam. The beam crosses the atomic beam at 45° angle as shown in Fig. 5.10.

The difference frequencies \( \delta f \) between the cooling beam and probe beam are given by the AOM frequencies \( f_2, f_3 \) and \( f_4 \) by

\[
\delta f = 2 \cdot f_4 - f_3 - f_2
\]

The two beams probe different velocity classes of the atomic beam. Because of the different angles of the beams relative to the atomic beam they scan at a different rate across the Doppler profile (Fig. 5.11). The detuning for the cooling beam is \( \delta_c = v/\lambda \), while for the probe beam we have \( \delta_p = v \cdot \cos \alpha / \lambda \), where \( \alpha = 45° \). The beams are both on resonance at a characteristic velocity \( v_0 \) depending on the difference frequency \( \Delta f \)

\[
v_0 = \frac{\Delta f \cdot \lambda}{(1 - \cos \alpha)}
\]

where \( v_0 \) is the velocity class of the atoms both cooling and probe laser beams interact with at \( \Delta f \), \( \lambda \) is the wavelength of the \(^1S_0 - ^1P_1\) transition and \( \alpha = 45° \) angle relative to the atomic beam.

To scan across the velocity profile of the atomic beam we scan the dye laser frequency. This changes the detuning of the cooling beam as well as the probe beam at the same time. This method is advantageous over using the double pass AOM3 for tuning the probe beam, because we can scan over a larger frequency range, while keeping alone the probe beam power constant.
5.2. Laser Cooling of Barium

Imaging optics & PMT

PBS

Ba Oven ~ 800°K

M

AOM4 553.7 nm

Pump laser 532 nm

Cooling beam

Repumpers 1500.4 nm 1130.6 nm

BS

M

AOM1 60 MHz

BS

AOM2 60 MHz

140-240 MHz

R= -200

140-280 MHz

+1 order

Doppler-free signal

Coherent 699 Ring Dye laser

To power stabilization

λ/4

Frequency Reference beam

M

AOM3 140-280 MHz

+1 order

Cooling beam shifted by f₃

Figure 5.10: Setup for laser cooling of barium atoms.
Figure 5.11: Velocities class of the atoms probed by the cooling beam and the probe beam. At a specific detuning both beams probe the same velocity class of atoms. This would be $v_0 = 302 \text{ m/s}$ for a $\delta f = 160 \text{ MHz}$ (Eqn. 5.46 AOM frequencies $f_3 = 200 \text{ MHz}$ and $f_4 = 210 \text{ MHz}$).

The two repump lasers for the $^1D_2 - ^1P_1$ (1500.4 nm) and the $^3D_2 - ^1P_1$ (1130.6 nm) transitions are overlapped with the cooling beam. The main power of the two repump lasers is utilized for laser cooling. The laser beams at 1500.4 nm and 1130.6 nm are combined on a dichroic beam splitter. 1-2 mW power of the laser light from the repump lasers is used as a reference to find the Doppler free resonance of the respective repump lasers. The beams cross the atomic beam perpendicularly in the trap chamber and the fluorescence is observed by the same detection system (not shown in the Fig. 5.10).

The two repump laser beams are combined with the 553.7 nm cooling beam on a second beam splitter. At this point we are left with 44% of the output power for the 1500.4 nm laser and 30% for the 1130 nm laser.

The combined beams are counter propagated to the atomic beam to achieve long interaction times. All the three beams are focussed to match the divergence of the atomic beam in order to achieve higher intensities. The atoms can interact with the laser beams over a length of 60 cm. The initial beam size is about 3 mm
5.2. Laser Cooling of Barium

Figure 5.12: The velocity distribution of barium atoms from an effusive oven measured with probe beam at 45° to the atomic beam. From the fit we extract a mean velocity of 250(20) m/s, which corresponds to a temperature of 800(40) °C.

radius for the infrared and about twice the size for the 553.7 nm.

The probe beam at 45° to the atomic beam is aligned to intersect the cooling beam in the middle of the trap chamber. During the measurement the probe beam power is typically between 0.8 to 1.2 mW. The fluorescence from the atoms is observed with the detection system described in Section 4.3.4. The different power changes the overall fluorescence signal but does not effect its shape.

Results

A typical longitudinal velocity distribution of the barium atoms from the atomic oven is measured with the probe beam at an angle of 45°. In Fig. 5.13 we show two examples where the velocity distribution is denoted with (a). This velocity distribution is used as a reference spectrum when we affect the distribution by the cooling lasers. The spectrum agrees with the velocity distribution from an effusive oven (Fig. 5.12). The velocity spectrum is fitted assuming a Maxwell Boltzmann velocity distribution of atoms from an effusive oven and taking into consideration the laser beam size and the linewidth of the transition. This is
because the barium atom scatters maximum $A_{\text{leak}}^{-1}$ photons in the time it stays in the interaction region before it ends up in one of the metastable $D$-states.

The fit to the spectrum agreed with the zero velocity calibration from a Doppler-free spectrum of barium isotopes measured in a second atomic beam setup. The two measurements are in good agreement with in the experimental error of about 10 m/s, which is due to the alignment of the laser beams with respect to the atomic beam.

The fluorescence spectrum measured when both the probe beam and cooling beam interacting with the $^{138}$Ba atomic beam is denoted as (b) in both figures. For velocities below $v_0$, where cooling and probe beams address the same velocity class we get a strong depletion, because of the optical pumping to the $D$-states. For the upper plot $v_0 \approx 230$ m/s and for the lower plot 320 m/s. The fluorescence in the absence of the repump lasers has two origins. First, the cooling beam does not cover the full atomic beam divergence and second a small reflection from one of the windows gives a probe beam at $135^\circ$ relative to the atomic beam. The latter increases the fluorescence for negative velocities, meaning atoms are moving away from the beam.

In order to achieve cooling we overlapped the two repump laser beams with the atomic beam and the cooling beam at a specific detuning relative to the Doppler free frequency. At the right value for the detuning of the repump lasers we see a clear increase of atomic flux at lower velocities, a sign for laser cooling of barium atoms. The two figures denoted with (c) in Fig. 5.13 show optimization for two different velocity classes of the atoms and at different detuning of the repump lasers.

To get a quantitative estimate on the cooling we plot the enhancement of flux versus the difference of velocity excited by cooling and probe beam. In Fig. 5.14 (I) the enhancement of flux is over a wide range of velocity which extend down to zero velocity. From this figure we can estimate the velocity change of the atoms to $\Delta v = 40$ m/s for atoms around 200 m/s. Atoms starting at lower velocity have been slowed further since they spend a longer time in the cooling laser beam. To slow an atom by 40 m/s it has to scatter $N_p = \Delta v/v_r = 7500$ photons. This can only be possible if the atom is pumped back from metastable $D$-states about $n_r = N_p/A_{\text{leak}}^{-1} = 22$ times. The time the atom had spend in the cooling beam over the distance of 0.6 m is $t = d/v = 3$ ms. Thus the average time in a $D$-state is around 140 $\mu$s. The laser intensities used in the experiment are about 18 mW/cm² and 64 mW/cm² corresponding to Rabi frequencies of $19 \times 10^6$ rad/s and $16 \times 10^6$ rad/s respectively. The Rabi frequencies are larger than our estimate above by a factor of 5. The main reason for this difference is probably the overlap
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![Graph showing laser cooling results for two different detuning of the lasers.](image)

**Figure 5.13:** Examples of laser cooling results for two different detuning of the lasers. **I:** $v_0=230\text{m/s}$ and **II:** $v_0=320\text{m/s}$. (a) Longitudinal velocity distribution of the atoms measured with 553.7 nm probe beam at 45° to the atomic barium beam. (b) Velocity distribution of the atoms measured with the cooling beam counter propagating the atomic beam and 553.7 nm probe beam at 45° to the atomic beam. The fluorescence of the atoms with velocities less than $v_0$ is almost equal to the background counts because the atoms decayed to the metastable $D$-states. Only fluorescence from the faster atoms is detected. (c) Velocity distribution of the atoms measured with the 553.7 nm cooling beam and repump laser beams counter propagating to the atomic beam and 553.7 nm probe beam at 45° to the atomic beam.
Enhancement of slow atomic flux relative to MB distribution

Figure 5.14: Increase in the flux of slow atoms after laser cooling. We plot the enhancement of flux over the velocity distribution from the oven versus the difference of velocity with which the cooling beam is on resonance and the probed velocity which is a measure of the slowing of the atoms. The increase is the ratio of spectra (c) to (a) in Fig. 5.13. the enhancement of flux is over a wide velocity range in I whereas it is flat after a certain velocity range in II.

Apart from the geometrical overlap of laser beams and the atomic beam the cooling result depends on several parameters. These are, in particular, the repump lasers power, detuning and the frequency spectrum of the repump lasers. The effects due to a change in these parameters are reflected in the velocity spectrum after the slowing. Thus our observation is always the effect of the changed parameter averaged over the whole velocity distribution of the atomic beam. To extract the effect on a particular velocity class, one would need a full numerical solution of the slowing process. Without that, we can only give a qualitative picture.

We investigated the influence of repump lasers intensity on the enhancement of lower velocity atom flux. The observed spectra are shown in Fig. 5.15. They show the result for 3 different intensities of the repump lasers. A spectrum with-
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Figure 5.15: Velocity distribution measured with cooling laser and at different output power of the repump lasers against the atomic beam and the probe laser at 45° to the atomic beam. (b) with no repump lasers. (c) Enhancement of atom flux at repump lasers intensities of 20(4) mW/cm² (1130.6 nm) and 5(1) mW/cm² (1500.4 nm), (d) 40(8) mW/cm² and 10(2) mW/cm², (e) 60(12) mW/cm² and 15(3) mW/cm² respectively. The bottom plot shows the enhancement at low velocities at different powers. Further increase in the repump lasers power did not increase the slow barium atom flux.
out repumping is added for reference. The slow atom flux in the velocity distribution of atoms increases with higher power of the repump lasers. A further increase of the intensities of the repump lasers did not increase the atoms at lower velocities. This indicated that due to the changing of the Doppler shift, the atoms moved out of resonance and could not be slowed further. The intensities of the lasers were not large enough to cause power broadening and thus allow a larger velocity range to be slowed.

The measurements were repeated for different detuning frequency of the repump lasers (Fig. 5.16). We observe that changing the detuning of the repump lasers by about half the linewidth of the transition influences the repumping and the slowing of atoms. This strong dependance is expected from the calculations of the multilevel system (Fig. 5.7). In particular, the dark Raman resonance can cause that the atoms cannot be slowed further than the velocity, at which it appears, because effective repumping has ceased. One can assume that at higher intensities for the repump lasers the effect of the detuning is less. With our experimental setup we could not explore higher intensities due to the limitation in the power of the fiber lasers.

As a third parameter, we investigated the influence of the spectral width for the repump lasers. For these measurements we introduces an EOM in the pass of the infrared lasers. Depending on the RF-power applied to the resonant EOM the spectral width is broadened (as an example see Fig. 4.19). In Fig. 5.17 several spectra are shown. In the caption the effective spectral width of the repump laser is given. The intensity of the lasers is held constant. The largest enhancement at low velocities is achieved with 60 MHz (1130 nm) and 35 MHz (1500 nm). Increasing the spectral width further lead to a decrease in slow atoms down to the level of no broadening due to the EOM. Here, the power is distributed over a larger frequency range and the effective repumping rates are reduced. Still, at velocities below 50 m/s, there is a small advantage over the case without frequency broadening. The longer time of flight of the atoms can compensate for the smaller repump rates.

The results of the measurements with variations in the parameters fit well with our expectations. In order to get a better picture on the various contributions to the cooling a single velocity barium beam would be advantageous. Such a beam could be produced by a velocity selector with chopping wheels.
Figure 5.16: Velocity distribution of the barium atoms measured with cooling laser, for 3 different detuning of the repump lasers. (a) initial velocity distribution, (b) depletion of velocity spectrum due to optical pumping to dark states, and (c-e) with different detunings of the repumper lasers. In (d) the detuning is -20(1) MHz (1130 nm) and -24 MHz (1500 nm) relative to (c) and in (e) the detuning is -8(1) MHz (1130 nm) and -15 MHz (1500 nm) relative to (c). Thus a change of less than the linewidth of the transition has a large consequence for the slowing process. The effect over no repump lasers in fluorescence is shown in the lower figure.
Figure 5.17: Velocity distribution of atoms measured with cooling laser and frequency broadened repump lasers by an EOM counter propagating the atomic beam and probe laser at 45° to the atomic beam. The repump lasers are detuned by -60 MHz. Enhancement of slow barium atom flux without RF power to infrared lasers EOM, at different RF power to EOM: -2 dBm corresponding to a frequency broadening of 45 MHz (1130.6 nm) and 25 MHz (1500.4 nm), 0 dBm corresponding to 60 MHz and 35 MHz and +2 dBm: 75 MHz and 45 MHz. The enhancement increases with increasing RF power to the EOM but at +2 dBm it decreases because the frequency broadening is large for the repump lasers so that the repump lasers are in resonance with high velocity atoms.
5.3 Cooling of Atoms with Complex Level Structure

In this chapter, we have investigated the possibilities for laser cooling of atoms with a complex level structure. In particular, we discussed the heavy alkaline earth element barium. We deal with two features.

- A strong leak in the singlet-singlet $^1S_0 - ^1P_1$ cooling transition at 553.7 nm to the metastable $D$-states. The leakage is stronger than in any other atom which has been laser cooled so far. Without repumping the velocity can only be changed by less than 2 m/s before the atoms are pumped into one of the metastable states.

- The repumping from the metastable $D$-states has to go via the excited state of the cooling transition. During repumping we see the effects of the coherent Raman transitions in this level system.

We showed that these features do not cause a major limitation for laser cooling of such atoms.

We have discussed the behavior of a $\Lambda$-system barium interacting with two coherent light fields. The properties of the two-photon transition that exhibit coherent Raman resonances are observed and compared to the results from a numerical solution of the optical Bloch equations. The extracted Rabi frequencies are large enough to allow for slowing a barium atomic beam. We demonstrate the first successful slowing of a thermal atomic beam by about 60 m/s. At velocities below 30 m/s we see an enhancement of the flux of atoms of nearly a factor of 5 over the thermal distribution from the atomic beam. In addition, we showed that the detuning of the repump lasers play an important role in achieving efficient slowing. The slow atom flux, which we have achieved was limited by the amount of laser power available for the repump lasers.

It should be possible to capture the slowed atoms from the beam in a magneto optical trap. The results are directly applicable to laser cooling of radium.