Chapter 6

Summary and Outlook

This thesis is written in the framework of the research of the TRIμP group at KVI. Described are the first steps toward the search for a permanent electric dipole moment (EDM) of the radium atom, which violates simultaneously parity (P) and time reversal symmetry (T). Any observation of an EDM would be an unambiguous signature for physics beyond the Standard Model of the electroweak interactions.

We have presented the Standard Model prediction of EDMs for fundamental particles which are well below the limits set by the current experimental efforts with various systems. The Radium atom has drawn in recent years considerable attention by both the theoretical and the experimental physics community for studying discrete symmetries like parity, time reversal and charge conjugation. The high sensitivity of radium arises from the large polarizability due to the near degeneracy of the \( 7s7p \, ^3P_1 \) and \( 7s6d \, ^3D_2 \) levels, which are of opposite parity. An energy difference of only 5 cm\(^{-1}\) between these levels is indicated from the available spectroscopic data. To confirm this, improved spectroscopic studies are required as an input for theoretical studies and for the design of the EDM experiment. In particular, the term energies and the lifetimes of the singlet and triplet \( P \) and \( D \)-states are needed to identify a suitable laser cooling scheme.

Short lived radium isotopes with nuclear spin \( I \neq 0 \), are not readily available for the experiments. They have to be extracted from radioactive sources or produced by nuclear reactions at accelerator facilities like the TRIμP facility at KVI.

We have presented the various stages for the production, separation and slowing of the short lived radioactive isotopes of our interest at TRIμP. Since only small quantities of short lived radioactive atoms are available and because of
long observation times sensitive experiments will be based on laser cooled and trapped atoms.

To develop spectroscopic techniques and laser cooling schemes we used stable isotopes of barium, because of its similarity in its energy level diagram to radium. For spectroscopy, metastable singlet and triplet $D$-states were populated by optical methods rather than using the conventional method of atomic discharges. The singlet $D$ state is populated by driving the $6s^2 \, ^1S_0 - 6s6p \, ^1P_1$ transition. To populate the triplet $6s5d \, ^3D_{1,2}$ states efficiently we exploited the branching ratio of the $6s6p \, ^3P_1$ level by optical pumping with the narrow intercombination line. To excite this $6s \, ^1S_0 - 6s6p \, ^3P_1$ transition at 791.3 nm we built diode lasers. We have achieved long term frequency stability of these lasers by referencing the frequency to a nearby transition in molecular iodine $I_2$, an often used frequency standard. With this setup we have performed several measurements.

We have reported the first direct optical spectroscopy of the $6s5d \, ^1D_2 - 6s6p \, ^1P_1$ (1500.4 nm), the $6s5d \, ^3D_2 - 6s6p \, ^1P_1$ (1130.6 nm) and the $6s65d \, ^3D_1 - 6s6p \, ^1P_1$ (1107.8 nm) transitions respectively.

We optically resolved the hyperfine structure of the $6s5d \, ^3D_{1,2}$ levels for the $^{137,135}\text{Ba}$ isotopes. The intensities of the measured hyperfine spectra are compared with the transition strengths calculated using Clebsch-Gordan coefficients. The hyperfine structure splitting is compared to previous microwave measurements for the $^3D_1 - ^1P_1$ and $^3D_2 - ^1P_1$ transitions, which are of higher precision. These are used for the calibration of the frequency scanning of the fiber lasers at 1107.8 nm and 1130.6 nm lasers. In addition, we used frequency side bands generated with an EOM and Zeeman splitting in a magnetic field to calibrate the fiber laser frequencies scanning.

In an atomic beam of natural barium we find the majority of the atoms distributed over the isotopes $^{138-134}\text{Ba}$. This allowed us to perform the first measurement of isotope shifts of the $6s5d \, ^3D_1 - 6s6p \, ^1P_1$ transition. The observed shift between $^{138}\text{Ba}$ and $^{136}\text{Ba}$, $^{136}\text{Ba}$ and $^{134}\text{Ba}$ isotopes and between the $^{137}\text{Ba}$ and $^{135}\text{Ba}$ isotopes are given in Table 6.1. The analysis with a King plot shows that electron-electron correlations may play a larger role for the values of the isotope shift. A better theoretical understanding would be desirable because of possible consequences for an EDM measurement utilizing atoms in one of the metastable $D$-states.

We report the experimental observation of coherent Raman resonances in barium for the first time. To understand these resonances quantitatively we solved the optical Bloch equations and compared the results with the experimental observation. We extracted the relevant Rabi frequencies for the $\Lambda$-system and estimated
<table>
<thead>
<tr>
<th>Isotope pair</th>
<th>138-136</th>
<th>136-134</th>
<th>137-135</th>
</tr>
</thead>
<tbody>
<tr>
<td>Isotope shift (MHz)</td>
<td>-71(5)</td>
<td>-93(6)</td>
<td>-75.3(0.5)</td>
</tr>
</tbody>
</table>

Table 6.1: Measured isotope shifts of the $6s5d^3D_1 - 6ssp^1P_1$ transition in barium.

the transition rates, relevant for the laser cooling experiment.

We have achieved the first slowing of a barium atomic beam using enriched $^{138}$Ba. These results have a far reaching influence for atoms, in which a closed cooling cycle can only be found with several laser fields at different wavelengths, adding to the complexity of the experimental setup. The results from this study can easily be extended to other atoms with similar level structure, in particular radium.

The only option for laser cooling of barium is offered by the $6s^2^1S_0 - 6s6p^1P_1$ transition. Unlike the other atoms that have been laser cooled and trapped, the barium atoms are easily pumped into metastable $D$-states. Without repumping no significant change in the velocity of the atoms can be achieved. They have to be pumped back through the $6s6p^1P_1$ state used in the cooling transition. This makes barium a multiple $\Lambda$-system. The repumping of atoms through a common excited state results in coherent Raman resonances.

In our experimental setup for slowing of barium atoms we employed a co-propagating cooling laser and infrared repump lasers travelling against the atomic beam. We have measured the velocity distribution from our atomic beam source with a 553.7 nm probe laser at 45° to the atomic beam at a distance of 0.6 m from the oven. We see a clear sign of cooling with cooling laser and repump lasers at appropriate frequencies. The shift in the velocity of the atoms can be larger than 50 m/s. At velocities below 30 m/s we see an enhancement of the thermal beam of flux of atoms nearly a factor of 5.

The next step is to capture the slowed atomic beam into a magneto-optical trap. In the future, we need to expand cooling to isotopes with hyperfine structure. In this case, the repump lasers should cover the frequency spectrum of the hyperfine structure splitting. This can be achieved by electro-optical modulators.

In the future, the slowing of atoms have to be optimized. Frequency chirping of the lasers look like a promising option as well as white light cooling. This can improve the accessible range of velocities from an atomic beam and it shows advantages over Zeeman slowing techniques, especially for isotopes with hyperfine structure.

In addition, the geometry of the experiment can be optimized. A critical parameter is the overlap of the infrared repump lasers with the atomic beam.
Furthermore, the infrared repump lasers should be frequency stabilized. The first steps were taken to lock them to Fabry-Perot cavities.

A theoretical treatment of the laser cooling for barium should be done by solving the equation of motion and the optical Bloch equation simultaneously for the whole cooling process. This can be based on our treatment of the multilevel system. The results of the calculation will go beyond our simple estimates and allow to design the laser system for radium laser cooling.

We have demonstrated the spectroscopy of metastable $D$-states in barium and have used the results to set up the first slowing of a thermal barium beam. All these results will be used for the setup of a radium spectroscopy and cooling experiment at KVI, where we are developing the online production of radium as well as a radium atomic beam. Within the next few years we expect to report similar results on spectroscopy and trapping with radium.