Cold Collisions of
Metastable Helium Atoms

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Metastable Helium Atoms

Koude Botsingen met
Metastabiele Helium Atomen

(met een samenvatting in het Nederlands)

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TER VERKRIJGING VAN DE GRAAD VAN DOCTOR
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Chapter 1

Introduction

1.1 Cold Collisions

By the application of ‘laser cooling’ in the field of atomic physics, the ultra-low energy regime of atom-atom collisions in dilute gases has become accessible. In low-energy collisions, or cold collisions, the atomic De Broglie wavelength exceeds typical distances for atomic interactions. Therefore a classical, localized description of the atomic interaction fails. In cold collisions the long range behavior of the atomic potentials determines whether slowly approaching atoms are attracted towards or repelled from each other. Typically, collision energies are in the order of 100 neV and therefore very weak structures in the interatomic potential, appearing at very large distances, can affect the dynamics of collisions dramatically.

The field of cold collision physics is still very young and only a few diatomic systems have been studied [1–6] and in particular the importance of the presence of resonant light in collisions has been investigated. In the first studies it was demonstrated that, due to the slow relative motion of the atoms, it is possible to influence the collision by absorption of light that is detuned slightly below the resonance of a particular atomic transition. Light absorption by systems of two separated atoms occur at that distance where the interaction energy matches the laser detuning. Depending on the laser detuning, molecular excitations can occur at internuclear distances ranging from distances characteristic for diatomic molecules up to several optical wavelengths. The molecular state formed by absorption can be either a bound state or a continuum state. In the case of negative detunings that exceeds the kinetic energy, absorption leads to the temporary formation of an excited bound state molecule. The lifetime of the molecule depends on the re-emission time of the absorbed photon. If this time is shorter than the time the atoms need to approach small distances, the temporary excitation leads to a modification of the collision of the ground state atoms, otherwise a collision on the excited molecular potential
Introduction

Collisions that are modified by light absorption in the way indicated above are called optical collisions. Experimental investigations of optical collisions provides the opportunity to observe atomic interactions over a large range of distances with spectroscopic precision.

1.2 The He* System

Until recently, in most of the cold collision experiments alkali systems have been studied because they can be easily handled experimentally. A disadvantage of using alkali systems is that the hyperfine structure in such systems significantly complicates the analysis of the collision dynamics. The problem in this respect is that a large number of molecular states has to be considered because transitions between hyperfine states can occur. The complication of hyperfine structure is not present in systems consisting of metastable rare gas atoms since the most abundant isotopes do not have a nuclear spin. The metastable states of rare gas atoms have internal energies between 12 to 20 eV, and the first excited states have lifetimes in the order of milliseconds to seconds. Since the interaction time during collisions is in the order of nanoseconds, the lowest metastable state level may be viewed as an effective ground state.

An additional advantage using metastable state systems for cold collisions studies is that Penning Ionization (PI) occurs. The ions produced by such reactions can be used to directly probe the dynamics in binary collisions. A difficulty with metastable atoms is how to produce them. Because of the lack of efficient sources, only a few preliminary cold collision experiments have been reported, where metastable systems have been studied [2, 3, 7]. The recent development of novel, intense metastable atom sources allows advanced studies on cold collisions of two triplet metastable helium atoms. Of all metastable systems, the helium collision system is particularly interesting because of its small mass and because of the prototype character of its electronic system containing only four electrons. Because of the small mass the quantum regime of s-wave scattering, where only central collisions lead to atom scattering, is already reached if a cold sample of He* atoms is prepared by using standard laser cooling techniques.

Very little is known about the He(2S)+He(2S) collision system at extremely low collision energies. In the single experiment with cold metastable helium atoms reported by Bardou et al. [7], the importance of resonant light in He(2S)+He(2S) collisions was demonstrated. The He(2S)+He(2S) collisions in absence of laser light could not be observed probably because the major source of ions was due to thermal PI reactions of He(2S) with background gas atoms leading to a huge back-
1.3 Contents

In chapter 2 we will review what is currently known about the PI process for He(2^3S) atoms at thermal, subthermal and cold temperatures. In addition we will discuss the remarkable features of the He(2^3S)+He(2^3P) system in connection with the theoretical models for optical collisions proposed in the literature [18, 19]. As it turns out, the two existing models for low energy collisions fail to describe the helium system. We therefore have developed a semi-classical model, based on a two-state Landau-Zener description.

In the collision experiments presented in this thesis we have used a cold sample of triplet metastable helium atoms contained in a so-called magneto-optical trap (MOT), for which we have developed an atomic beam apparatus supporting slowing, cooling and trapping of metastable helium atoms. The experimental setup is discussed in chapter 3, together with the results of the slowing of a thermal beam of atomic He(2^3S).

In chapter 4 we present a simple model for magneto optical trapping in the regime of large laser detunings and high laser intensities to explain the specific features of our He(2^3S) trap. The experimental conditions and results will be discussed. Furthermore, we will study the loss of atoms from the trap in inelastic processes by using ion mass spectrometry, which will turn out to be a powerful technique to analyze reactive collisions in atomic traps.

The trap loss due to elastic scattering on background gas atoms is studied in detail in chapter 5. In this chapter we determine the elastic scattering cross sections of several rare gases and diatomic molecules colliding with trapped He(2^3S) atoms. It will be demonstrated that the use of atomic traps provides a technique to measure absolute scattering cross sections, thereby avoiding severe technical difficulties and sources of systematical errors present in traditional atomic beam experiments.

In chapter 6 the process of Penning Ionization in the system of two triplet metastable helium atoms is investigated at a temperature of 1 mK. Both the He(2^3S)+He(2^3S) “ground state collisions” and the He(2^3S)+He(2^3P) “optical collisions” are studied by measuring the PI cross sections as well as the AI cross sections.

In contrast to chapter 6, where optical excitation of the system preceeds the collision, in chapter 7 we will investigate the excitation of He(2^3S)+He(2^3S) systems during the collision, i.e. we will study the molecular properties of the system. This
Introduction

has been realized by optical excitation of the molecular complex by laser light, its frequency detuned below the $2^3S \rightarrow 2^3P_2$ transition, which is followed by ionization of the He($2^3S$)+He($2^3P$) complex.
Chapter 2
Atom-Atom Interactions

The central problem in atomic collision theory is the calculation of the scattering cross section, which essentially is the effective size of a binary collision system in collision or reaction process. The calculated cross section can be compared with values deduced from experimental data to test and validate theoretical models. To understand the results of the experiments presented in this thesis basic knowledge of potential scattering is needed. We therefore review the relevant formulas used in atomic scattering theory, which can be found in many textbooks on the subject (see for example ref. [8, 9]).

2.1 Potential Scattering

The collision dynamics for a system of two colliding atoms is most conveniently studied in the center of mass (CM) frame. In this frame the relative motion of the atoms is given by the motion of a particle with reduced mass $\mu$ in an external potential $V(R)$, given by the interaction energy of the two atoms when separated by a distance $R$. We will assume that this potential is spherical symmetric, so that the particle is exposed to a central force.

In the case of elastic scattering the time-independent Schrödinger equation for the motion of the particle in this potential is given by

$$\left[\frac{\hbar^2}{2\mu} \nabla^2 + V(R)\right] \psi_k(R) = E \psi_k(R),$$

(2.1)

where $E = \hbar^2 k^2 / 2\mu$ is the energy of the particle and $\psi_k(R)$ is the wavefunction for the position of the particle.

At large distances, where the interaction energy $V(R)$ can be neglected, the wavefunction must satisfy the free-particle Schrödinger equation. In this limit the
total wavefunction can be written as the sum of an incident wave $\psi_k^{inc}(R)$ with wavenumber $k$ and the scattered wave $\psi_k^{sc}(R)$

$$\psi_k(R) = \psi_k^{inc}(R) + \psi_k^{sc}(R).$$ \hspace{1cm} (2.2)

The scattered wave can be written as an outgoing spherical wave which represent the outgoing flow of scattered particles, when the incident (plane) wave is scattered by the central potential $V(R)$:

$$\psi_k^{sc}(R) = f(k, \theta, \phi) \frac{e^{ikR}}{kR}.$$ \hspace{1cm} (2.3)

The function $f(k, \theta, \phi)$ is known as the scattering amplitude and depends on the energy $E$ and the polar angle $\theta$. It can be shown [9], that this function defines the differential scattering cross section $d\sigma/d\Omega$ by

$$\frac{d\sigma}{d\Omega} = |f(k, \theta, \phi)|^2.$$ \hspace{1cm} (2.4)

The total scattering cross section is obtained by integration over all scattering angles.

For a central force the wavefunction can be expanded in a so-called partial wave series of Legendre polynomials $P_\ell(x)$

$$\psi(k, R) = \sum_{\ell=0}^{\infty} A_\ell(k, R) P_\ell(\cos \theta),$$ \hspace{1cm} (2.5)

where the radial functions $A_\ell(k, R)$ are the coefficients for the different polynomials. If this expansion is used in Eq. (2.1) the equation for the radial wavefunctions $u_\ell(k, R) = RA_\ell(k, R)$ is given by

$$\left[ \frac{\hbar^2}{2\mu} \frac{d^2}{dR^2} - \frac{\hbar^2}{2\mu} \frac{\ell(\ell + 1)}{R^2} - V(R) + E \right] u_\ell(k, R) = 0,$$ \hspace{1cm} (2.6)

where $\ell$ is the quantum number of the orbital angular momentum. The term proportional to $\ell(\ell + 1)/R^2$ represents the rotational barrier in the collision reflecting the conservation of total angular momentum in a collision. The barrier is important in collisions, since in a semi-classical description it determines the maximum impact parameter in a collision, that leads to a close collision (see figure 2.1). Collisions with impact parameters larger than a certain $b_c$ are reflected by the rotational barrier, if the height $V_c$ of the barrier exceeds the kinetic energy. In this case the collision cannot lead to a close approach of the two atoms and therefore the collision does not lead to a reaction.
2.1 Potential Scattering

The solution for the radial functions $u_\ell(k, R)$ is given by linear combinations of spherical Neumann and Bessel functions. When these solutions are evaluated, the scattering amplitude $f(k, \theta, \phi)$ and hence the total elastic scattering cross section $\sigma_{\text{tot}}$ is given by [9]

$$\sigma_{\text{tot}}(k) = \frac{4\pi}{k^2} \sum_{\ell=0}^{\infty} (2\ell + 1) \sin^2 \delta_\ell(k),$$  

(2.7)

where $\delta_\ell(k)$ are the phase shifts of the scattered waves relative to the incident partial waves. By using partial wave analysis the calculation of scattering cross sections is reduced to the problem of calculating the phase shifts for the different partial waves in a collision. The shifts depend on the potential $V(R)$ and have in general to be obtained by numerical methods. If a large number of partial waves contribute to scattering the contribution of many phase shifts can be approximated by an average

---

Figure 2.1 (a) The interaction potential $V_n = -C_n/R^n$, which has an attractive long range behavior. (b) Classical trajectories of an atom for different impact parameters when scattered by this potential. For impact parameters larger than $b_c$ the atom cannot cross the rotational barrier. For collisions with smaller impact parameters the atom can cross the rotational barrier, which then can lead to a reactive collision (figure from [10]).
Atom-Atom Interactions

of $\langle \sin^2 \delta_{\ell} \rangle_\ell = 1/2$. In this approximation the summation over the different partial waves up to a certain $\ell_{\text{max}}$ can be performed explicitly resulting in

$$\sigma_{\text{CC}} = \frac{\pi \hbar^2}{\mu E_{\text{rel}}} (\ell_{\text{max}} + 1)^2,$$

which corresponds to the classical expression for the close collision cross section. In classical mechanics, $\ell_{\text{max}}$ corresponds to the largest impact parameter $b_{\text{max}}$, that for a given energy leads to a close collision. The close collision cross section is given by $\sigma_{\text{CC}} = \pi b_{\text{max}}^2$. For long-range potentials $V_n = -C_n/R^n$ the maximum impact parameter leading to a close collision is easily calculated and the cross section in this case is given by

$$\sigma_{\text{CC}} = \frac{\pi n}{n-2} \left[ \frac{(n-2)C_n}{2E} \right]^{2/n}.$$

For example, the long range interaction of neutral S-state atoms is given by the van der Waals attraction, corresponding to $n=6$. For this type of interaction the close collision cross section scales with energy as $E^{-1/3}$.

2.1.1 Complex Potentials

If electronic transitions occur, e.g. ionization or chemical reactions, the flux of incident atoms is not conserved in a collision. These effects can be included in the method of partial waves by introducing a complex potential [9]

$$V(R) = V_{re}(R) - i V_{im}(R).$$

The imaginary part of this potential leads to an attenuation of the incident wave. This reflects that particles are removed from the incident wave by inelastic processes. We will assume that the electronic transitions in the inelastic processes occur on such a short time scale, that the position of the two atomic nuclei can be considered as fixed during this time. This is called the local transition approximation.

If such a complex potential is taken into account the cross section $\sigma_r(k)$ for inelastic processes is given by

$$\sigma_r(k) = \frac{\pi}{k^2} \sum_{\ell=0}^{\infty} (2\ell + 1)(1 - \eta_\ell^2(k)),$$

where

$$\eta_\ell(k) = \exp(-2\Im \delta_\ell(k))$$

(2.12)
are the so-called absorption factors, which are defined by the imaginary part of the complex phase shifts. In general, the elastic scattering cross section of a real potential is changed, when an imaginary term is introduced.

2.1.2 S-Wave Scattering

Because of the rotational barrier in Eq. (2.6) the number of partial waves that contribute to the scattering is reduced in the case of low energy collisions. In the case, that the height of the $\ell=1$ rotational barrier is higher than the amount of kinetic energy in a collision, only the $\ell=0$ partial wave in Eq. (2.7) contributes. This is the quantum regime of $s$-wave scattering, where only central collisions lead to scattering. In the low energy limit of elastic collisions the so-called scattering length $\alpha$ is defined according to

$$\alpha = -\lim_{k \to 0} \frac{\tan \delta_0(k)}{k}. \quad (2.13)$$

The physical significance of the scattering length is that the total elastic scattering cross section in the limit of vanishing relative momentum is given by

$$\sigma(k = 0) = 4\pi \alpha^2. \quad (2.14)$$

The characteristic energy, where the cross section becomes constant, is in the regime where only $s$-wave scattering occurs. The zero energy cross section in Eq. (2.14) determines the thermalization time of a dense cold sample of atoms by elastic collisions. Thermalization by atom-atom collisions is only important at very high atomic densities. In the atomic trap discussed in this thesis the density is too small to reach a thermal equilibrium by collisions. The thermal equilibrium that is reached in the atomic trap of chapter 4 is by absorption and spontaneous emission of photons.

2.2 Penning Ionization

In a collision, the force that acts at large distances between two neutral ground state atoms, is given by the van der Waals attraction [8]. In this second order dipole-induced dipole interaction the potential energy of two atoms at internuclear distance $R$ varies as $V(R) = -C_6/R^6$, where $C_6$ is known as the van der Waals constant and which depends on the system. The coefficient is positive, meaning that the force between two atoms is attractive. If the attraction of the atoms leads to a close approach electronic transitions may occur.
Atom-Atom Interactions

Figure 2.2  Electron spectrum produced in a PI reaction \( A^* + B \rightarrow A + B^+ \). Electrons are produced with energies \( \epsilon (R) \) by transitions in the region of small internuclear distances (shaded area). The energy \( \epsilon (R) \) is given by the potential energy of the \( A^* + B \) molecule minus the energy of the \( A + B^+ \) ion (dashed line). If transitions occur in the dark shaded area bound state molecular ions \( AB^+ \) are produced (AI).

An excited state collision system \( A^* + B \) can undergo an ionization reaction, if the ionization energy of \( B \) is less than the internal energy of \( A \). This type of reaction, in which the kinetic energy of the atoms is small compared to the internal energy, is called Penning Ionization (PI)\(^1\) and the reaction is schematically given by

\[
A^* + B \rightarrow \begin{cases} 
A + B^+ + e^- \\
AB^+ + e^-
\end{cases}.
\]

In a PI reaction two distinct exit channels can be accessed and either a molecular ion or an atomic ion is produced. The total ionization process will be referred to as Penning Ionization (PI), whereas the process on the last line of the equation is called Associative Ionization (AI). The PI process is triggered by electronic transitions occurring at short internuclear distances, where the electron clouds of the two atoms overlap. Usually PI is included in the theory in terms of an imaginary part added to the potential (see section 2.1.1 and ref. [12, 13]). This is the approach we will also adapt here. The imaginary part of the potential is usually described in terms of a

\(^1\)The Penning Ionization reaction is named after the Dutch physicist F.M. Penning, who suggested this type of ionization process in 1927 [11].
2.2 Penning Ionization

“width function” $\Gamma(R)$ as $V_{im} = \Gamma(R)/2$, with

$$\Gamma(R) = \Gamma_0 e^{-R/R_0}.$$  \hspace{1cm} (2.16)

Here $\Gamma_0$ and $R_0$ are characteristic parameters for the strength of the ionization process. Using this approach PI cross sections and electron distributions can be calculated, both semi-classically and quantum-mechanically. Because the width function depends on the distance $R$, the electron spectrum in the PI process contains the structure of the continuum electronic wavefunction of the AB* molecule and of the continuum and bound state waves functions of the AB+ ionic states. In figure 2.2 the origin of a typical electron spectrum is depicted schematically. In the remainder of this section we give an outline of the present knowledge on the reactive collisions where at least one atom is triplet metastable helium.

2.2.1 Metastable Helium

The He$(2^3S)$ state is the lowest triplet metastable state of helium. In the triplet state the spins of the two electrons in the atom are aligned. The part of the level structure of the helium atom that is relevant in this thesis is given in figure 2.3. The He$(2^3S)$ state can be reached from the He$(1^1S)$ ground state by electron impact. The He$(2^3S)$ state is a metastable state with an extremely long lifetime of more than two hours, which is the result of strict selection rules for radiative decay back to the singlet ground state. Since the lifetime is much longer than the typical duration of a collision, the He$(2^3S)$ level may be considered as the ground state in collisions. For laser cooling and trapping we use the $2^3S \rightarrow 2^3P_2$ optical transition at $\lambda=1083$ nm. The He$(2^1P)$ state has a radiative lifetime of 98.8 ns and later on we will see that this time is also long compared to the typical duration of a collision. The He$(2^2S)$ level is also included, since this level is in energy just below the He$(2^1P)$ level and therefore may be accessed in He$(2^3S)$+He$(2^1P)$ collisions.

2.2.2 X+He$(2^3S)$

The processes of PI and AI in the singly excited systems X + He* have been studied in atomic He* beams by Illenberger and Niehaus [14] in the energy range of 25–200 meV:

$$X + He(2^3S) \rightarrow \left\{ \begin{array}{l} X^+ + He(1^1S) + e^- \\ HeX^+ + e^- \end{array} \right.$$

(2.17)

A typical ionization cross section for the reaction with $N_e$ is 3 Å² at 26 meV [14]. The cross sections for the rare gases and other diatomic molecules have the same
Figure 2.3 Level structure of helium. Only the energy levels, which are relevant for the contents of this thesis, are included. In laser cooling and trapping experiments the He(\(2^3\)S) state can be treated as an effective ground state. The He(\(2^3\)S) atoms are produced by electron impact on He(\(1^1\)S) ground state atoms. Radiative decay from the He(\(2^3\)S) level back to the He(\(1^1\)S) ground state is forbidden by spin selection rules. The \(2^3\)S \(\rightarrow\) \(2^3\)P\(_2\) transition is used to study optical collisions. The He(\(2^1\)S) state lies in energy between the two triplet states and may be accessed in optical collisions.

order of magnitude. However, for the case of H\(_2\)O, the ionization cross section has the large value of 70 Å\(^2\) at 70 meV energy [15]. This is due to the fact that the water molecule has a dipole moment which leads to a strong attraction in a collision. The relatively large cross section for He\(^*\)+H\(_2\)O is confirmed by our experiments where it has been found that the major ion production in a low density metastable helium trap is due to this thermal PI reaction with residual water molecules (see chapter 4).
2.2 Penning Ionization

2.2.3 He(23S)+He(23S)

Penning ionization in the doubly excited systems of He*+He* has been investigated in detail by Müller et al. [16] at energies ranging from 1 meV to 60 meV. Both the PI process and the AI process in

\[
\text{He}(2^3\text{S}) + \text{He}(2^3\text{S}) \longrightarrow \begin{cases} 
\text{He}^+ + \text{He}(1^1\text{S}) + e^- \\
\text{He}_2^+ + e^- 
\end{cases}
\]  

(2.18)

were studied in experiments and in theory. The exit channel of the ionization reaction was measured state selective by application of electron spectroscopy. By using this technique it was possible to resolve the electron contributions from the different molecular states, which are accessed in a He(23S)+He(23S) collision.

In principle, three molecular states in this system can couple to the He(23S)+He(23S) asymptotic state. The states have singlet, triplet and quintet symmetry and correspond to the \(1^1\Sigma_u^+, 1^3\Sigma_u^+\) and \(1^5\Sigma_u^+\) molecular states. The quintet state corresponds to the asymptotic He(23S)\(\uparrow\)+He(23S)\(\uparrow\) spin polarized state. It has been shown by Müller et al. [16], that no ionization from the quintet state takes place. This is attributed to the Wigner spin rule, stating that in transitions between different molecular states the spin quantum number is conserved. In the case of PI in the He(23S)+He(23S) system a transition is made from the initial molecular states mentioned above to the \(2^1\Sigma_u^+\) or \(2^3\Sigma_u^+\) ionic states. The final ionic doublet states combined with the spin state of the ejected electron is either a singlet or triplet state. Applying the Wigner spin rule, ionization from the quintet state has to be excluded. Müller et al. [16] calculated an ionization cross section of 318 Å² at 1 meV and measured a branching ratio for AI of 0.07(3). They have also demonstrated, that the measured electron spectra are in good agreement with state-of-the-art theory, which includes a full quantum treatment of both the electronic states and the relative motion of the atoms.

2.2.4 Ionization in Cold Collisions

When He(23S)+He(23S) collisions are studied in a MOT, two features will be important. First, the collision energy of typically 100 neV will lead to a strong suppression of the number of partial waves that contribute to the scattering. Secondly, the near resonant laser light in the trap will induce He(23S)+He(23P) collisions. If the collision energy in the He(23S)+He(23S) system is less than 10 μeV, elastic collisions occur in the quantum domain of pure s-wave scattering. It is expected [16], that then only molecular states with an electronic gerade symmetry contribute to the cross section. Since in s-wave scattering only the \(\ell=0\) partial wave contributes
and because the total wavefunction of the boson system is symmetrical, only gerade states can lead to s-wave scattering. Since the Wigner spin rule has to be obeyed as well, the only molecular state that is accessed in reactive collisions is the $^1\Sigma_g^+$ state. If the collisions occur in the presence of a resonant light field, He(2S)+He(2S) pairs can be optically excited to a He(2S)+He(2P) complex. In this excited state the atoms will experience a strongly attractive potential (see figure 2.6), in which they will be accelerated towards each other. If the atoms reach short distances PI can occur. Bardou et al. have been reported enhanced ionization for He(2S)+He(2S) collisions in a resonant light field [7]. Bardou et al. have also found that the main ionization is induced by reactive S+P collisions [7]:

$$\text{He}(2^3\text{S}) + \text{He}(2^3\text{P}) \rightarrow \begin{cases} \text{He}^+ + \text{He}(1^1\text{S}) + e^- \\ \text{He}_2^+ + e^- \end{cases}$$  \hspace{1cm} (2.19)

In ref. [7] it is estimated that the PI cross section for this reaction lies in the range of $1.5 \times 10^6 - 2.4 \times 10^7$ Å² for a collision energy of 0.1 µeV. This reported cross section is, however, at least a factor of two too large, because the authors did not distinguish S+P pairs from P+S pairs in their equation for the ion production.
In this section we will discuss optical collisions by considering low energy collisions of two S state atoms in a laser field with wavelength $\lambda$. The laser frequency is detuned slightly below resonance of an optical S-P transition in the atom. A characteristic distance for molecular excitation is given by the distance, where the interaction energy of a S+P complex is equal to the linewidth of the optical transition. This distance $R_0$ is given by [17]

$$R_0 = \frac{\lambda}{2\pi}. \quad (2.20)$$

For distances smaller than $R_0$ the optical excitation has the character of a molecular excitation. At larger distances the excitation occurs in the limit of two non-interacting S-state atoms, where one atom is excited to the P state. We define the critical distance $R_{max}$ as the average distance that excited state atoms travel before decaying by spontaneous emission. This distance is given by

$$R_{max} = v\tau\beta, \quad (2.21)$$
Figure 2.6  Long range potential energy curves for the He(2\(^3\)S)+He(2\(^3\)S) potential \(V(R) = C_6/R^6\) and the He(2\(^3\)S)+He(2\(^3\)P) potential \(V(R) = C_3/R^3\) with \(C_3 = 19.25\) au and \(C_6 = 3291\) au. The curves are shifted in energy such that the asymptotes coincides. The excited state potential is more attractive at large distances, since it scales with \(1/R^3\). For internuclear distances in the range of 20–50 \(\text{Å}\) the ground state potential can be considered flat, whereas the excited state potential is attractive.

where \(v\) is the relative velocity of the atoms and \(\tau_{\beta}\) the lifetime of the molecule in the state \(\beta\). At large distances \(\tau_{\beta}\) corresponds to the atomic lifetime \(\tau = 1/\Gamma\) of the P state. In the experiments, where optical collisions have been studied so far, the interaction distances that have been accessed are much smaller than \(R_\lambda\), as is shown in table 2.1. The He*+He* system is the only system, where optical collisions have been studied both in the molecular regime as well as in the regime of two separated atoms. The change of a molecular character to a more atomic character occurs in the intermediate regime of molecular and collision physics and is very interesting, since e.g. retardation effects in the atomic potentials are pronounced.

Molecular excitations in cold collisions are treated by the Gallagher-Pritchard (GP) model [18,20], which is a simple model to understand the dynamics of optical collisions. However, the high temperature limit of the model is not correct and therefore excitations at distances in the order of \(R_\lambda\) are not treated correctly. It has been demonstrated by Julienne and Vigué [19], that this is due to the oversimplified pair distribution in the GP-model. Julienne and Vigué demonstrated that the GP-model can be extended to a more general model by using a more realistic pair distribution function. By analogy, this model is called the JV-model [21]. It has been shown, that the JV-model correctly predicts the close collision cross section
2.3 Optical Collisions

<table>
<thead>
<tr>
<th>System</th>
<th>λ [nm]</th>
<th>τ [ns]</th>
<th>⟨v⟩ [m/s]</th>
<th>R_{max} [Å]</th>
<th>R_{λ} [Å]</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>He*+He*</td>
<td>1083</td>
<td>98</td>
<td>2.0</td>
<td>2744</td>
<td>1724</td>
<td>This thesis</td>
</tr>
<tr>
<td>Kr*+Kr*</td>
<td>811</td>
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<td>0.2</td>
<td>87</td>
<td>1290</td>
<td>[2]</td>
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<tr>
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<td>882</td>
<td>33</td>
<td>0.1</td>
<td>42</td>
<td>1404</td>
<td>[3]</td>
</tr>
<tr>
<td>Na+Na</td>
<td>589</td>
<td>16</td>
<td>0.3</td>
<td>65</td>
<td>940</td>
<td>[4–6]</td>
</tr>
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<td>780</td>
<td>27</td>
<td>0.1</td>
<td>45</td>
<td>1240</td>
<td>[1]</td>
</tr>
</tbody>
</table>

Table 2.1: Critical distances R_{max} of molecular excitation that have been accessed in optical collision experiments. In this thesis reactions are studied for molecular excitations in the He*+He* system far beyond the point of R_{λ}.

![Diagram](image)

**Figure 2.7** He(2\textsuperscript{3}S) + He(2\textsuperscript{3}P) optical collisions. Laser light tuned to resonance of the 2\textsuperscript{3}S → 2\textsuperscript{3}P\textsubscript{2} optical transition opens a second channel for ionization. At large distances He(2\textsuperscript{3}S)+He(2\textsuperscript{3}S) ground state molecules are excited to an He(2\textsuperscript{3}S)+He(2\textsuperscript{3}P) state, which has attractive S+P branches. In this state the atoms are accelerated in the V(R) = -C_3/R\textsuperscript{3} potential followed by a PI reaction at close range. Since the lifetime of the He(2\textsuperscript{3}P) state is 98 ns, the optically excited molecule will ionize before it decays by spontaneous emission of a photon.

of S+P collisions at higher energies. The JV-model, however, is not as transparent as the GP-model. In order to gain some insight in the physics of optical collisions we will therefore first discuss the features of the basic GP-model before we switch to the more complicated JV-model. In chapter 7 we will describe our own model, which combines the simplicity of the GP-model and the correct behavior for high energies of the JV-model.
2.3.1 The GP-model

The problem that is treated in the GP-model is the calculation of rate coefficients for reactions induced by optical collisions. The starting point is the evaluation of the two atom pair distribution \( N(R) \):

\[
dN(R) = n4\pi R^2 dR,
\]

which is the average number of atom pairs in the S+S state within a spherical shell ranging from \( R \) to \( R + dR \). Here \( n \) is the density of atoms in the ground state. If the laser frequency is detuned below resonance of an atomic transition, atom pairs can be excited by the laser light to an S+P state, if the interaction energy \( V(R) = C_3/R^3 \) equals \( \hbar \delta \).

The excitation rate of molecules at a distance \( R \) is assumed to be given by a Lorentz line shape

\[
L(R) = \frac{\lambda^2 I}{2\pi \hbar \omega} \left[ \frac{1}{1 + (2\Delta/\Gamma_\beta)^2} \right],
\]

where \( I \) is the laser intensity, \( \hbar \omega \) the photon energy, \( \Gamma_\beta = 1/\tau_\beta \) the molecular linewidth and \( \Delta = \delta - V(R)/\hbar \) the effective detuning. The total number of molecular excitations by the laser light per unit of time is obtained by multiplication of the number of pairs \( dN(R) \) and the line shape \( L(R) \) followed by integration over \( R \).

After the excitation has taken place the atoms in the molecule are accelerated in a \( C_3/R^3 \) potential. If the lifetime of the excited state molecule is less than the time it takes for the atoms to react, the system will decay back to the ground state by emission of a photon. Due to the spontaneous decay only a fraction of the excited state molecules survives, before a reaction takes place. The survival probability for an excitation, that occurs at \( R \) and leads to a reaction is given by

\[
S(R) = \exp \left( -\int \Gamma_\beta dt \right),
\]

where the time integral is over the classical path of an atom. The atom accelerates in a \( C_3/R^3 \) potential starting at the Condon point \( R_c \) and ending near \( R=0 \), where the reaction takes place. For acceleration in a \( C_3/R^3 \) potential the survival factor can be written more conveniently as a function of the laser detuning \( \delta \):

\[
S(R) = \exp \left( -(\delta_\tau/\delta)^{5/6} \right),
\]

with \( \delta_\tau \) a characteristic detuning depending on the collision system.
2.3 Optical Collisions

If the survival probability is taken into account, the rate constant $K(\delta)$ for optical collisions leading to a reaction is given by

$$K(\delta) = \int_{0}^{\infty} 4\pi R^2 L(R) S(R) dR = \frac{\lambda^2}{2\pi \hbar \omega} \frac{I}{6\pi \hbar^2} e^{-(\delta \tau / \delta)^{5/6}}.$$  \hfill (2.26)

The integration over $R$ is performed by assuming large laser detunings, so that the excitation probability $L(R)$ of the ground state molecule only contributes at the Condon point $R_c$.

The procedure outlined above is the basic GP-model for optical collisions, where the rate constant has been calculated. It has been demonstrated that for excitations at distances much less than $R_\lambda$, i.e. for large detunings, the GP-model describes the experimental data in systems of Rb, Xe* and Kr* rather well (see table 2.1 for the references). For small detunings, however, the lineshape of Eq. (2.26) does not fit to the observed data even though excitations occur at distances much smaller than $R_\lambda$.

2.3.2 The JV-model

The paper of Julienne and Vigué [19] describes a method to include molecular excitations at higher energies, which occurs at large distances. Here the importance of the energy and angular momenta dependence of the pair distribution is being discussed, which is not taken into account in the GP-model. By using a more realistic pair distribution Julienne and Vigué calculated the reaction constant for optical collisions, taking into account the retarded lifetimes of the molecular states and performing the evaluation of the survival factor for the different partial waves $\ell$. The rate constant $K(E, \delta)$ derived in the JV-model is given by

$$K(E, \delta) = K_{CC}(E) S(E, \delta).$$  \hfill (2.27)

Here $K_{CC}$ is the close collision cross section for S+P collision in the absence of spontaneous emission, which can be calculated from potential scattering. The excitation and survival correction factor $S$ describes all the effects of cold collisions, namely the change of excitation probability due to transitions in molecular states compared to atomic states at thermal collisions, and the survival factor of the molecular complex due to spontaneous emission. In the JV-model this factor is a weighted average over all different partial waves $\ell$, that contribute to the scattering cross section.

The averaging over $\ell$ has to be done carefully, since it depends on the laser detuning and collision energy. For large laser detunings, when molecular transitions occur at small distances, the averaging should be over all partial waves that
Atom-Atom Interactions

can come within the Condon radius in the ground state He(2S)+He(2P). At small laser detunings, when S+P collisions are induced at large distances, the averaging is over all partial waves, that are not reflected by the rotational barrier on the S+P potential.

It has been shown [19], that in the high energy limit \( E \gg \hbar \Gamma \) the rate coefficient in the JV-model is determined by the close collision cross section in Eq. (2.8), which is the correct limit, if collisions at thermal energies are considered. In the case, that \( E \ll \hbar \Gamma \) the rate constant in the JV-model is equal to the rate constant in the GP-model [19]. Therefore the JV-model is expected to be valid for energies in the intermediate regime corresponding to \( E \approx \hbar \Gamma \), where atomic interactions at distances in the order of \( R \), are important.
Chapter 3

Slowing an Atomic Beam of Metastable Helium

3.1 Introduction

In this section the experimental setup of our He* atomic beam line is presented. In this setup atoms are slowed and cooled using the principle of Zeeman compensated slowing [22, 23]. In our beam apparatus triplet metastable He atoms are decelerated by scattering photons absorbed from a counterpropagating laser beam, whose frequency is tuned on resonance of an atomic transition. Since we will use the slow atoms to load a Magneto Optical Trap (chapter 4), the flux and the velocity distribution of the slowed atoms are of importance. A schematic overview of the atomic beam apparatus is given in figure 3.1. The vacuum setup consists of three parts, which will be referred to as the Metastable Atom Source, the Zeeman Slower and the Magneto Optical Trap (MOT). The first two parts will be discussed in this chapter. The last part is the subject of the next chapter.

3.2 Metastable Atom Source

In our experiment an atomic beam of He(2S) metastable atoms is produced in a DC glow discharge. Our source is constructed following the design of the Ne* source reported by Kawanaka et al. [25]. A small part of a continuous gas flow expands through an aluminum nozzle of 0.5 mm in diameter into a high vacuum environment. The whole source is cooled by liquid nitrogen to about 77K. The main part of the gas flow is reversed and is used to sustain a DC glow discharge between a negative biased tungsten needle and the grounded nozzle. The discharge region is spatially separated from the nozzle thereby reducing the heat transfer to the atoms in the beam. The electrons produced in the discharge are dragged through
Slowing an Atomic Beam of Metastable Helium

the nozzle and excite He\(^{1}\)S) atoms in the expansion to the He\(^{2}\)S) state. The pressure near the aluminum nozzle is about 2 mbar and a typical voltage over the discharge is 700 V at an electrical current of 3 mA. Degradation of the tungsten needle (Ø 3 mm) at these small currents is rather low. If the source is cooled by liquid nitrogen, and if the current does not exceed 3 mA, the lifetime of the needle is over 4000 hours of operation. Therefore, this type source is practically maintenance free.

Kawanaka et al. tested this type of source for Ne and reported intensities of \(1.0 \times 10^{15}\) Ne\(^{*}\) sr\(^{-1}\)s\(^{-1}\) at a mean velocity of 390 m/s [25]. We have tested our source for Ne where we found a yield of \(5.0 \times 10^{14}\) Ne\(^{*}\) sr\(^{-1}\)s\(^{-1}\) at a mean velocity of 380 m/s. This is comparable to the results reported by Kawanaka et al. When our source runs on He we have an intensity of \(1.0 \times 10^{14}\) He\(^{2}\)S) sr\(^{-1}\)s\(^{-1}\) at a mean velocity of 900 m/s (FWHM 450 m/s) corresponding to a temperature of about 80 K. We have measured the intensity of the source using a stainless steel Faraday cup by current measurements. In the first experiments we have used a mechanical chopper in the vacuum to measure the velocity distribution by a Time Of Flight technique. Later on we have used atomic beam deflection spectroscopy (see section 3.5) to measure TOF spectra. In the latter technique He\(^{2}\)S) atoms in the beam can be spatially separated from He\(^{1}\)S) atoms. By using this feature we have found that over 90% of the metastable atoms are in the He\(^{2}\)S) state, the residual 10% in the He\(^{2}\)S) state.

3.3 Zeeman Compensated Slowing

The most common technique to produce slow atoms is Zeeman compensated slowing, this generates a continuous flux of slow atoms. In our experiments we have implemented Zeeman compensated slowing based on the \(2^{3}\)S\(^{\text{a}}\) \(\rightarrow\) \(2^{3}\)P\(^{\text{a}}\) optical transition. An advantage using this transition is that single mode diode lasers are available for optical excitation [26]. It is, however, not a very efficient “slowing transition” due to the long lifetime of the He\(^{2}\)P) state. Therefore a long flight path in the vacuum setup is needed to slow the atoms down to zero velocity. For the \(2^{3}\)S\(^{\text{a}}\) \(\rightarrow\) \(2^{3}\)P\(^{\text{a}}\) transition the stopping length for atoms at thermal velocities of 1200 m/s is 1.4 meter, if maximum deceleration can be applied over the whole flight path. To avoid losses of atoms in the slowing process, caused by fluctuations of photon absorption cycles, the stopping length in our experiment has been increased to 2.3 meter. The atomic beam source is mounted in a vacuum chamber at a distance of 20 mm from a diaphragm (Ø 0.5 mm), which separates the primary vacuum container from a differential pumping stage where the residual gas load of the source is
3.3 Zeeman Compensated Slowing

Figure 3.1 Schematic overview of the atomic beam line (top view). The apparatus is evacuated by turbo molecular pumps. At the different vacuum stages, the pressure is indicated. The vacuum setup is designed to minimize the pressure in the backside near the MOT chamber in which the collision experiments are performed.

<table>
<thead>
<tr>
<th>He* Source</th>
<th>Differential pumping Stage</th>
<th>Zeeman Slower</th>
<th>MOT Chamber</th>
</tr>
</thead>
<tbody>
<tr>
<td>Coils $\phi$ 200 mm, $N=600$</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Solenoid 1
Solenoid 2

deflection laser beam

slowing laser beam
trapping laser beams

600 mm 1950 260 1100 800

1e-3 mbar 2e-6 mbar 1e-8 mbar 5e-9 mbar

240 l/s 240 l/s 180 l/s 180 l/s
slowed away. The connection of the differential pumping section to the entrance of the Zeeman slower is separated by a second diaphragm (Ø2 mm) to reduce the gas load in to the slowing section. The Zeeman slower is split in two parts and a vacuum pump has been installed between the two sections to reduce the pressure at the end of the slower to less than \(1 \times 10^{-8}\) mbar. At the exit of the slower a six-fold vacuum cross piece is mounted containing a Magneto Optical Compressor (MOC), which is designed to compress the atomic beam by laser light. This device is currently being tested and was not used in the experiments presented in this thesis. At the exit of this device the MOT chamber is connected by a 200 mm long connection piece of 40 mm in diameter. A laser beam with the dimensions of 15x8 mm\(^2\) and a power of 10 mW counterpropagates with respect to the atomic beam to provide slowing. The two solenoids in the Zeeman slower produce a magnetic field to compensate for the changing Doppler shift during the deceleration. The polarization of the laser beam is circular with a helicity such that atoms are slowed in a two level subsystem of the \(2^{3}S \rightarrow 2^{3}P_2\) transition.

### 3.4 Diode Lasers

For optical excitation of the \(2^{3}S \rightarrow 2^{3}P_2\) transition in helium we use free running single mode diode lasers (SDL-6702-H1) \cite{26} supplied by Spectra Diode Lab. The diode lasers are driven by home-made current supplies at a maximum current of 250 mA. For temperature regulation the internal NTC resistor at the laser diode head is monitored. The temperature is actively regulated using the internal Peltier element inside the laser diode package. In front of the emitting aperture we have put an AR coated lens (Thorlabs C240TM-C) at 8 mm and collimate over 50% of the emitted light into a elliptical laser beam with the dimensions of 8x2 mm\(^2\). To shape the laser beams we use cylindrical and spherical telescopes. All optics like lenses, windows and polarizers are supplied by Eksma Co. (Lithuania) and have AR coatings that have less than 0.5% residual reflection per surface.

The linewidth of the diode lasers has been measured in beat experiments using two diode lasers from which we find a linewidth of \(3.0 \pm 0.2\) MHz. The laser frequency is coarse tuned to the \(2^{3}S \rightarrow 2^{3}P_2\) transition using a Philips discharge lamp (93098E) filled with helium. Within the Doppler broadened absorption line of the lamp the laser is tuned to resonance by Doppler free ‘deflection spectroscopy’. We have therefore have split a part of the primary laser beam into a second beam which is used to calibrate the laser frequency. This ‘reference’ beam is aligned perpendicular to the atomic beam and monitors the absorption of laser light by detecting the atoms that are deflected out of the laser beam due to momentum exchange by
3.5 Results

The laser frequency can be calibrated in this way and is known within an uncertainty of about 3 MHz which is limited due to the linewidth of the laser diodes. The long term frequency stability of the diode lasers is compromised by thermal induced drift (typically 100 MHz per hour) caused by ambient temperature changes and air flow. The laser frequency can be modulated by current injection into the diodes. The laser controllers have two current modulation entries: one for quasi-continuous operation (10 kHz bandwidth) and one fast modulation entry (bandwidth > 10 MHz). We use digital programmable waveform generators (Hewlett Packard-HP33120A) to control and manipulate the laser frequency.

3.5 Results

In the slowing experiments we have removed the mechanical chopper to obtain a continuous flow of slowed atoms. The velocity distribution of the slowed atomic beam has been measured using a separate diode laser (see figure 3.1) tuned near resonance of the $^{23}\text{S} \rightarrow ^{23}\text{P}_2$ transition and its beam aligned perpendicular to the atomic beam. The frequency of this laser is periodically switched on resonance (duty cycle 1–5%) to deflect atoms out of the beam into a channeltron positioned 0.9 m down stream, 5 mm off axis. The metastable atoms have sufficient energy to produce a pulse on the channeltron which are analyzed using a standard Multi Channel Scaling expansion board implemented on a micro-computer. The deflection scheme is implemented just after the Zeeman slower stage. By monitoring the drift time of the deflected atoms the velocity distribution in the atomic beam has been measured. We have used this TOF technique for an on-line study of the slowing process. In figure 3.2 the velocity distributions of atoms are shown for the case that (a) no slowing is applied and for the case that (b) all atoms in the beam are slowed to about 400 m/s. The atoms in the beam are not only slowed but the spread of velocities is also strongly reduced due the effect of laser cooling. From the figure one can see that the total intensity of the atomic beam is reduced by about a factor of four. This loss is caused by the spatial spread of the slowed atomic beam in the transverse direction.

3.5.1 Beam Spread

In figure 3.3 the intensity of the slowed atomic beam is plotted for different selected final velocities. The reduction of the intensity is due to the beam spread near the exit of the Zeeman slower: during the slowing process the velocity component of an atom along the beam axis is reduced while the transverse component is not .
Figure 3.2 Measurements of the velocity distribution of a He(2\(^3\)S) atomic beam. (a) The thermal velocity distribution of the beam when the source is cooled by liquid nitrogen. The total intensity of the thermal beam is \(3.0 \times 10^7\) He(2\(^3\)S) atoms cm\(^{-2}\)s\(^{-1}\). (b) Velocity distribution in the beam for the case that the atoms are slowed and cooled by laser light.
3.5 Results

reduced. The atoms will therefore leave the Zeeman slowing stage under a larger angle than they entered the solenoid, leading to a highly divergent atomic beam. The beam spread is even enhanced by an increase of transverse velocity due to the spontaneously emitted photons. The effect of transverse heating can be modeled by a random walk induced by the recoil kicks of scattered photons. The width of the transverse velocity distribution then increases with the square root of the number of steps in this process i.e. the number of photons used in the slowing process to bring the velocity down from some initial velocity to the final velocity. If both effects of slowing and transverse heating of the atoms are taken into account, the central beam intensity can be calculated in a straightforward manner. In our geometry the distance $L$ that atoms have to travel to reach the detector is 0.9 m. Since this distance is much larger than the atomic beam radius $r_0$ of 0.5 cm we can consider the exit of the slowing stage as a source which emits a highly divergent beam of slow atoms. The intensity of the beam as a function of the velocity $v_f$, with the restriction that $v_f$ is less than 700 m/s, can be approximated in our setup by

$$I(v_f) = I_0 \frac{r_0^2}{L^2} \frac{v_f^2}{(v_0 - v_f)v_r},$$

(3.1)

where $I_0$ is the atomic beam intensity if no slowing is applied, $v_0$ is the initial velocity of the atoms in the beam and $v_r$ is the photon recoil velocity, which is 0.1 m/s for the $2^3S \rightarrow 2^3P_2$ optical transition. The dominant effect of the loss is the square dependence on the final velocity $v_f$ induced by the slowing process. This limits the atomic beam intensity at low velocities and therefore the loading rate of slow atoms into the MOT.

3.5.2 Beam Temperature

In the slowing process the width of the longitudinal velocity distribution is reduced by the effect of laser cooling. The width of the distribution in figure 3.2b is about 50 m/s and is broadened by the high laser power used in the slowing experiments due to the effect of power-broadening. We have studied the cooling effect by reducing the laser power. In figure 3.4 the measurement is showed when the laser intensity of the slowing laser was reduced from a saturation level of $s_0=50$ (8 mW) to $s_0=10$ (1.6 mW). The solid line through the data set is a fit of a displaced Gaussian function with a width of 8.6 m/s. This width is well above the so-called Doppler limit (see next chapter) which is 0.3 m/s for this system. The large observed width is mainly due to the relative long duration of the gate time of the deflection laser which was 50 $\mu$s in these experiments. If we take this finite gate time into account, we find
Figure 3.3  Intensity of the slowed atomic beam for different final velocities. As atoms are slowed to lower velocities the intensity is reduced. The solid line through the data points is a fit of the function given by Eq. (3.1) with \( v_0 = 800 \text{ m/s} \).

An resolution of about 6 m/s. The resolution in the experiment is thus too poor to resolve the small velocity spread of 0.3 m/s predicted by Doppler cooling theory.

3.6 Conclusions

In this chapter our experimental setup for slowing of a triplet metastable He* beam has been discussed. Our He* beam source produces \( 1 \times 10^{14} \text{ He}(2^3S) \text{ sr}^{-1} \text{ s}^{-1} \) at an average velocity of 900 m/s and with a velocity spread (FWHM) of 450 m/s. By using a Zeeman compensated slowing setup it has been demonstrated that the complete thermal velocity distribution can be slowed. The intensity loss of the slowed atomic beam is induced by spatial beam spread. The measured longitudinal velocity spread of atoms in the beam is less than 8.6 m/s.
Figure 3.4  High resolution measurement on the velocity spread of the laser cooled atomic beam. The solid line is a Gaussian fit through the data set. The velocity spread of 8.6 m/s is well above the Doppler limit of 0.3 m/s due to the limited experimental resolution.
Slowing an Atomic Beam of Metastable Helium
Chapter 4

Trapping Metastable Helium

4.1 Theory

The cold and dense atomic samples needed for an experimental study on low energy collisions can be obtained by laser cooling and trapping techniques. In a magneto optical trap (MOT) atoms are cooled and trapped in a laser light field combined with an inhomogeneous magnetic field [27]. The density, temperature and volume of the atomic samples are important parameters in collision studies. The parameters depend on the trapping conditions such as the laser frequency and intensity, but also on the loading rate of atoms into the trap as well as on the loss rate of trapped atoms. The traps have to be loaded with atoms that are not too fast since typical trap depths are only on the order of 100 mK. The final temperature of the atomic samples obtained by laser cooling is usually close to the so-called “Doppler temperature” which is the equilibrium temperature determined by the effect of cooling and heating by laser light [28].

In this chapter we will first discuss the basic ingredients of standard Doppler theory and analyze the motion of trapped atoms in a 1D model. Secondly, the model for MOT operation will be applied to the specific case of trapping metastable helium and it will be demonstrated that, depending on the laser intensity and detuning, three different types of traps can be distinguished. Third, the experimental setup of our MOT is presented and the results on trapping He(2$^3$S) atoms are discussed. Finally, the trap loss processes in the MOT will be analyzed in detail.

4.1.1 Motion of Trapped Atoms

For two-level systems the principle of MOT operation is well understood. We will use the well-known standard model of Doppler cooling and trapping (see figure
Trapping Metastable Helium

4.1). The force in the trap that exerted on the atoms is induced by cycles of absorption and spontaneous decay. From the Optical Bloch Equations (OBE) the excitation process of atoms in a laser light field is well known [29]. In this discussion the coherence terms in the OBE are eliminated, i.e., optical excitation will be treated by rate equations. In this approximation the force on an atom in a light field of two counter-propagating laser beams and an inhomogeneous magnetic field, is given by [30]

\[
F(x, v) = \frac{\hbar k}{\Gamma} \left( \frac{s_0}{1 + 2s_0 + (\frac{2\delta}{\Gamma})^2} - \frac{s_0}{1 + 2s_0 + (\frac{2\delta}{\Gamma})^2} \right).
\]

(4.1)

with

\[
\delta_{\pm} = \delta \pm \omega_z(x) \pm kv,
\]

(4.2)

and where \(\omega_z(x)\) is the local Zeeman shift induced by an inhomogeneous magnetic field given by \(B(x) = -\alpha x\) (\(\alpha\) the magnetic field gradient), \(k\) is the wavenumber of the light, \(\delta\) is the laser detuning relative to the optical transition, \(\Gamma\) is the decay rate of the excited state (\(\Gamma = 1/\tau\)) and \(s_0\) is the saturation intensity. The dynamics of the atomic motion is non-linear and can only be solved by numerical integration of Eq. (4.1). In the limit of small displacements and velocities Eq. (4.1) can be approximated by the zero point motion of trapped atoms

\[
F(x, v) = -\beta v - \kappa x,
\]

(4.3)

where the damping parameter \(\beta\), and the spring constant \(\kappa\), are given by

\[
\beta = \frac{-8s_0\hbar k^2\delta/\Gamma}{(1 + 2s_0 + (\frac{2\delta}{\Gamma})^2)^2},
\]

(4.4)

and

\[
\kappa = \frac{-8s_0\hbar k\mu_0\alpha\delta/\Gamma}{(1 + 2s_0 + (\frac{2\delta}{\Gamma})^2)^2}.
\]

(4.5)

Eq. (4.3) is the damped harmonic oscillator equation. The heating of atoms induced by the spontaneous decay is omitted here but will be included afterwards. For realistic parameters of the detuning, the laser intensity and the magnetic field gradient, the eigenfrequency \(\omega_0 = \sqrt{\kappa/m}\) of the oscillation is less than the damping frequency \(\gamma = \beta/m\). In this case the atomic motion is overdamped meaning
4.1 Theory

Figure 4.1 Principle of Magneto Optical Trapping for a $J = 0 \rightarrow J = 1$ transition. In an inhomogeneous magnetic field the energy levels are split by the Zeeman effect. Atoms at the right of the origin ($x'$) will preferentially absorb photons from the laser beam with the $\sigma^-$ polarization since the $m = -1$ sublevel is shifted into resonance. By cycles of photon absorption and spontaneous decay momentum is exchanged by which the atoms are driven towards the origin.

that atoms in the trap will creep to the center. The characteristic damping time $\tau_D$ of this motion is given by

$$\tau_D = \frac{2\omega_0^2}{\gamma}. \quad (4.6)$$

For standard trapping conditions (see next section) this time is on the order of a millisecond.

4.1.2 Trapping He*

We will apply the result of Eq. (4.3) to the case of trapping He(2$^3$S) using the $2^3S \rightarrow 2^3P_2$ transition at $\lambda=1083$ nm. This $J = 1 \rightarrow J = 2$ system is a closed system when driven by laser light and can be treated as a two level system. For the magnetic field gradient we take $\alpha=7.5$ Gauss/cm and we choose three combinations of laser frequency-intensity for which we will analyze the damping in the well and the trapping potential $U$, determined by $F = -dU/dx$, and where $F(x)$ is given by Eq. (4.3).
Figure 4.2 (a) Transient atomic motion in a He* MOT and (b) the spatial profile of the confining potentials in three regimes I: $\delta = -1 \Gamma$ and $s_0 = 2$ II: $\delta = -15 \Gamma$ and $s_0 = 2$ III: $\delta = -15 \Gamma$ and $s_0 = 50$.  

Trapping Metastable Helium
4.1 Theory

(I) Standard Trap: Small Detunings and Low Laser Power

Most MOT based atomic traps operate at modest laser power of typically $s_0=2–10$ and at small laser detunings of $|\delta|=1–3 \Gamma$ to achieve optimal damping of the atomic motion. This is the regime where cooling in optical molasses is most efficient and hence where the coldest and most dense atomic samples are produced. In figure 4.2 the motion of an He* atom in such a trap is plotted together with the confining potential at these conditions. The atomic motion in this type of trap is strongly overdamped at time scales of a millisecond. A major disadvantage using small laser detunings is that the capture velocity of the trap is limited to about 10 m/s which corresponds to a trap depth of only 30 $\mu$eV. The small depth will limit the loading rate of atoms into the trap. Therefore traps operating at these conditions will contain a small number of atoms at relatively high densities.

(II) Hard Sphere Trap; Large Detunings and Low Laser Power

The capture velocity of the standard type MOT can be increased by operating the trap at large laser detunings of about $|\delta|=10–50 \Gamma$. A helium trap operating at such detunings can have capture velocities up to 300 m/s. At these velocities the loading rate of atoms into the trap is dramatically increased. For the chosen laser parameters the confining potential resembles a square well [31]. In figure 4.2 the shape of the potential is plotted together with the atomic motion in the well. From the simulations of the atom trajectories it follows that the damping time of the trap is on the order of 100 milliseconds. Due to the small damping the atomic motion is oscillatory so that the atoms will be distributed over the entire trap volume. The density of the atomic sample in this kind of trap is therefore limited.

(III) Deep Trap; Large Detunings and High Laser Power

The damping of trap II can be enhanced if the laser intensity is increased as well. Under the conditions of large detunings and high laser power the overdamped motion is restored. Cooling is enhanced at high laser intensities due to the effect of power broadening: when the intensity increases the Lorentzian line shape for absorption broadens. If the laser intensity is chosen such that the power-broadened line width $\Gamma \sqrt{s_0 + 1}$ equals the detuning, the maximum damping is obtained. This results in a broad and deep confining potential which is plotted in figure 4.2. From the transient atomic motion in the trap it can be seen that the atoms are rapidly stored to the origin, similar to case I. Because of the strong damping in the trap, atoms are cooled down to low temperatures, thereby reducing the volume of the atomic sample in the confining well.
Trapping Metastable Helium

Trap III can be regarded as an intermediate case of trap type I and II, supporting efficient cooling and a deep confining potential. If the atoms have to be loaded at relative high velocities, trap type III will contain atomic samples with the highest densities.

4.1.3 Temperature, Volume and Density

In traps of type I and III the motion of the atoms is rapidly damped and a stationary state is reached before atoms are ejected out of the trap by collisions with background atoms. The equilibrium velocity of an atom that is reached, is given by the balance of cooling and heating. The cooling rate is determined by the damping term $\beta$ in Eq. (4.4) whereas the heating of the atoms is caused by diffusion induced by spontaneous emitted photons [28].

The mean kinetic energy $\frac{1}{2}m\langle v^2 \rangle$ at equilibrium can be calculated for these processes and is related to a temperature $T$ according to

$$\frac{1}{2}m\langle v^2 \rangle = \frac{1}{2}k_b T$$  \hspace{1cm} (4.7)

In figure 4.3 the trap temperature is plotted as a function of detuning for several laser intensities. For large detunings, ($\delta \gg \delta_0 \Gamma$), the temperature increases linearly with detuning and the temperature dependence is given by

$$\frac{1}{2}k_b T = \frac{1}{2}\hbar \delta.$$  \hspace{1cm} (4.8)

The size of atomic sample is given by the mean position spread $\langle x^2 \rangle$ and can be obtained by realizing that in a harmonic potential the size of the atomic ensemble at temperature $T$ is given by

$$\frac{1}{2}k_b T = \frac{1}{2}\kappa \langle x^2 \rangle,$$  \hspace{1cm} (4.9)

where $\kappa$ is the spring constant given by Eq. (4.5).

In figure 4.4 the size of the cold He* atomic sample is plotted as a function of the laser detuning for several laser intensities. For increasing laser intensity the volume is minimized at detunings of half the power broadened linewidth. This is because under these circumstances the coldest atomic samples are obtained and these occupy the least amount of space in the trap.

In table 4.1 we give an overview and comparison of the four metastable helium traps that have been reported in literature. The temperature of traps are well explained by figure 4.3. The radius of the trap reported by Bardou et al. of 0.3 mm (experiment) is in good agreement with our model calculations given in figure 4.4.
4.2 Experimental Setup

The minimum temperature is reached for $\delta = -\Gamma/2$ at laser intensities of $s_0 \ll 1$. This is where cooling in optical molasses is most efficient and where the Doppler temperature of 40 $\mu$K can be reached. The trap in this thesis operates at a laser detuning of $\delta = -15\Gamma$ and at an intensity of $s_0=50$ and a temperature of 1.1 mK is reached. The temperatures reported by Bardou et al. [7] and Rooijakkers et al. [33] are well explained by our model.

4.2 Experimental Setup

Our MOT is built around a spherical stainless steel vacuum chamber with a diameter of 250 mm, supporting six conflate flanges of 100 mm and twelve conflate flanges of 40 mm. These vacuum connections are used to connect a turbo molecular pump, optical windows, a pressure gauge, a needle valve, two mass spectrometers (one used for residual gas analysis and one to monitor the MOT), a He* metastable detector and an ion detector. The turbo molecular vacuum pump is supplied by Balzers (TPM 180) and is very efficient in evacuating light elements such as helium. The pressure that can be reached in the MOT chamber is $4.0 \times 10^{-9}$ mbar and is limited due to the direct gas load of the atomic beam into the chamber.

Home made window mounts are connected to the chamber containing optical grade quartz windows of 80 mm in diameter. These windows provide access for
### Table 4.1 Comparison of the four reported helium metastable MOT’s. All traps are based on the He(23S)-He(23P2) transition and are loaded with atoms from a Zeeman slowed atomic beam.

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Number of trapped Atoms</td>
<td>$10^4$</td>
<td>$10^4$</td>
<td>$10^7$</td>
<td>$4 \times 10^5$</td>
</tr>
<tr>
<td>Temperature (mK)</td>
<td>-</td>
<td>1</td>
<td>2</td>
<td>1.1</td>
</tr>
<tr>
<td>Volume (mm$^3$)</td>
<td>-</td>
<td>0.1</td>
<td>100$^\dagger$</td>
<td>4.0$^\dagger$</td>
</tr>
<tr>
<td>Density (cm$^{-3}$)</td>
<td>-</td>
<td>$10^8$</td>
<td>$10^8$</td>
<td>$10^7$</td>
</tr>
<tr>
<td>Lifetime (s)</td>
<td>$19 \times 10^{-3}$</td>
<td>0.9</td>
<td>0.8</td>
<td>4.5</td>
</tr>
<tr>
<td>Magnetic field Gradient (G/cm)</td>
<td>-</td>
<td>20</td>
<td>32</td>
<td>15</td>
</tr>
<tr>
<td>Laser beam diameter (mm)</td>
<td>15</td>
<td>15</td>
<td>20 (x,y)</td>
<td>35</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>30 (z)</td>
<td></td>
</tr>
<tr>
<td>Laser detuning ($\Gamma$)</td>
<td>-</td>
<td>3</td>
<td>25</td>
<td>15</td>
</tr>
<tr>
<td>Laser power ($s_0$)</td>
<td>16-100</td>
<td>25</td>
<td>25 (x,y)</td>
<td>50</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>62 (z)</td>
<td></td>
</tr>
<tr>
<td>Ion rate (kc/s)</td>
<td>-</td>
<td>-</td>
<td>180</td>
<td>$20 \text{H}_2\text{O}^+$</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>$2 \text{He}_2^+$</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>$1 \text{He}^+$</td>
</tr>
</tbody>
</table>

$^\dagger$ Calculation

Trapping Metastable Helium

the six orthogonal trapping laser beams needed in a MOT setup. Two water cooled coils, placed under and above this chamber are connected in a anti-Helmholtz configuration to produce an inhomogeneous magnetic field of 15 G/cm in the center of the trap. This gradient is in the z-direction and for a current of 10 A. The MOT operates at the $2^3S \rightarrow 2^3P_2$ transition at $\lambda=1083$ nm for which we use a diode laser (chapter 3). The elliptical beam shape of the laser diode is tailored by cylindrical
4.2 Experimental Setup

![Graph showing MOT size for different laser detunings and intensities. The minimum volume is reached at small detunings and small intensities. Our trap operates at a detuning of $-15\Gamma$ and at a laser intensity of $s_0=50$. For these parameters the radius of the atomic sample is about 1 mm. The radius of the MOT (0.3 mm) reported by Bardou et al. [7] is in good agreement with the model calculations.](image)

**Figure 4.4** MOT size for different laser detunings and intensities. The minimum volume is reached at small detunings and small intensities. Our trap operates at a detuning of $-15\Gamma$ and at a laser intensity of $s_0=50$. For these parameters the radius of the atomic sample is about 1 mm. The radius of the MOT (0.3 mm) reported by Bardou et al. [7] is in good agreement with the model calculations.

optics and a spherical beam expander to obtain a circular laser beam of 30 mm in diameter. The laser beam is recycled by reflecting it 18 times thereby producing six laser beams in an orthogonal cross. The principle of the optical arrangement is shown in figure 4.5. We have found that if the laser beams are reflected under angles less than $45^\circ$ the circularly polarization is preserved. The helicity is adjusted to the direction of the magnetic field such that a restoring force is obtained, as required for MOT operation.

### 4.2.1 Ion Detection

The most common way to observe trapped atoms is by detection of the fluorescent light emitted by the excited atoms [1, 2, 34]. However, detection of fluorescence at a wavelength of $\lambda=1083$ nm is rather complicated because at that wavelength the quantum efficiency and noise characteristics of photo multipliers and diodes compromise the detection of the low light levels. A different way to detect whether
Figure 4.5 Principle of MOT laser beam handling. A single laser beam is recycled to provide six orthogonal beams with the proper circular polarization. The attenuation of the intensity of the laser beam, caused by residual reflections and absorption of the optical windows, is compensated by slightly focusing the incident laser beam.

Atoms are trapped or not is by detection of ions produced in the MOT. An advantage of this technique is that ion detection is a very sensitive and direct probe for reactive collisions. In metastable atom traps there are two inelastic processes in which ions are produced. The first is by PI in collisions with background atoms and the second is by PI in intra-MOT collisions at low energies. In the case of metastable helium PI reactions can occur in collisions with all background gas atoms with the exception of He and Ne, because of the high internal energy of the He(2S) level. This type of inelastic trap loss is most important at low densities. In our setup we use two ion detectors which are positioned in the MOT chamber as shown in figure 4.6. The first detector consists of a channeltron and a negative biased grid in front of it. This detector can collect ions produced within a volume of about 1 cm³ near the center of the MOT container. With this detector the total ion yield from the MOT can be monitored. From ion trajectory calculations we have found that all ions produced within the MOT, irrespective of their mass and kinetic energy, are accelerated towards the channeltron.

The second detector is a quadrupole mass spectrometer mounted opposite to the first detector. The electron impact ionization section of the spectrometer has been
4.2 Experimental Setup

Figure 4.6  Ion detectors used to monitor and study the MOT. (1) A channeltron with a biased grid in front of it extracts all ions produced in the MOT. (2) A mass spectrometer biased at $-200 \text{ V}$ extracts ions from the trap. The ions are guided through an entrance hole and transported along the directions of the rods of the electrical quadrupole section before they are detected at the exit using a channeltron. One should note that the two detectors can not operate simultaneously.

removed such that ions produced in the MOT can be detected. The spectrometer is mounted in the vacuum chamber with its long axis in the plane of symmetry. The head of the device is placed at 50 mm from the center of the trap. The spectrometer is electrically isolated from the vacuum flange by teflon spacers and is biased $-200 \text{ V}$ to extract ions from the MOT. A very fine copper grid (95% transparency) is put in front of this entrance hole to produce a well defined electrical field in which the ions are directed through the spectrometer. The mass selecting section is a 20 cm long oscillating electric quadrupole, operating at a frequency of 3 MHz. At the exit of the quadrupole the ions are detected by a channeltron. The mass resolution of the spectrometer is 1 amu, which is more than sufficient to distinguish He$^+$ from He$_2^+$ ions.

The mass spectrometer operates in the inhomogeneous magnetic field of the MOT. The spectrometer is put in the plane of symmetry of the trap such that the magnetic field is always in the direction of the velocity of the ions. In this way the ions on the axis of the spectrometer are not deflected by the magnetic field. To rule out any effect of the magnetic field on the transfer of ions to the spectrometer we
have carried out the following test: in the center of the chamber ions were produced by PI reactions of He* atoms in the atomic beam with background gas atoms D₂ and N₂. The ion production of these atoms was analyzed by the spectrometer with the magnetic field turned ‘on’ and ‘off’. We did not find any influence of the magnetic field of the MOT and conclude that in our setup the magnetic field does not perturb the ion transfer through the mass spectrometer.

4.2.2 Results

Our MOT runs at a laser detuning of $\delta = -25$ MHz relative to the $^2S \rightarrow ^2P_2$ transition. The Zeeman slower produces He($^2S$) atoms at a velocity of about 70 m/s, which is sufficiently slow for trapping. When the MOT is turned ‘on’ the ion yield increases from 100 c/s to about $2 \times 10^4$ c/s. When the loading of atoms into the trap is interrupted the MOT decays exponentially in time (figure 4.7). From this we conclude the main trap loss mechanism is by thermal collisions with background atoms and not due to cold collisions. The time constant of the decay process is about 5 seconds at a background pressure of $4.0 \times 10^{-9}$ mbar. We have analyzed the trap loss processes by investigating the mass spectrum of the ions produced in the MOT. A typical mass spectrum is shown in figure 4.8. The major ion production is at 18 amu due to H$_2$O$^+$. There are also ions at 17 amu which have to be attributed to OH$^+$. The ions at 18 amu are produced in the thermal PI reaction of He*+H$_2$O→H$_2$O$^+$+He($^1S$)+e$^-$. The peaks at 4 amu and 8 amu can be identified as He$^+$ and He$^+_2$. These ions are produced in the PI reaction of He*+He* (intra-MOT collisions). The very small ion production rate at 5 amu reproduces well in the experiments and may be due to molecular HeH$^+$ ions produced in a AI reaction of He*+H$_2$. The structure near zero mass is due to an artifact of the mass spectrometer. If no mass selection is applied a peak will appear in the spectrum near zero mass. Because of this artifact we are unable to resolve the production of H$_2^+$. The ion spectrum has also been measured without the operating the MOT. We detected a rate of 0.1 c/s at mass 4 and 8 amu and a rate of 1.5 c/s at mass 18. The background rates at 4 and 8 amu are due to residual effects such as electrical noise and cosmic radiation, whereas the background at 18 amu is due to the PI reaction of residual water with He* in the atomic beam: when the He* atomic beam was turned ‘off’ the 1.5 c/s ion rate disappeared.

Knowing the dominant trap loss process, the total number of trapped atoms $N_{stat}$ can be estimated from the steady state ion production $\dot{I}$ by using

$$N_{stat} = \frac{\dot{I}}{\eta \gamma_w}, \quad (4.10)$$
4.2 Experimental Setup

Figure 4.7 MOT decay when the loading of atoms into the trap is interrupted. The trap is monitored by detecting ions produced by PI reactions. The decay is exponential in time to a very high degree (see lin-log inset) indicating that the main ion production is from PI reactions with background gases.

where $\eta$ is the ion detection efficiency of the channeltron and $\gamma_w$ is the decay rate of the trap induced by the process of PI with water. A conservative estimate for the decay rate $\gamma_w$ is that all trap loss is by the PI reaction of He* and water. Using $\dot{I}=30\times10^3$ c/s, $\eta=0.4$ and $\gamma_w=0.2$ Hz we find for the number of trapped atoms $N_{stat}=4.0\times10^5$.

4.2.3 Temperature Measurements

The temperature of the atoms in the trap has been measured by a ballistic flight technique. The trapped atoms were released from the trap by suddenly shifting the trapping laser far from resonance. A micro-channel plate detector positioned 70 mm away from the trap was used to detect the He* after their ballistic flight. The detector was put parallel to the z-direction to reduce the effect of gravitational acceleration. In figure 4.9 the TOF data set is plotted. The mean velocity of the atoms is 2.3 m/s corresponds to a MOT temperature of 1.1 mK and is good agreement with the model calculation in section 4.1.3 which predicts 1 mK.
4.3 Conclusions

Magneto optical trapping of He* is favored at large laser detunings and at high laser intensities. The cooling mechanism in this regime is still explained by Doppler cooling theory. In a trap that is operated at detunings of $\delta = -15\Gamma$ and high intensities of $s_0=50$, the confining potential has a depths of 250 $\mu$eV and a capture volume of several cm$^3$. Such a trap can be loaded with atoms at high initial velocities of 80 m/s. The cooling mechanism in a large detuned trap is the same as in optical molasses, although not as efficient. Temperatures of 1 mK can be reached, which is well above the Doppler temperature of 40 $\mu$K. Our He* MOT contains about $4.0\times10^5$ He* atoms when it is operated at a detuning $\delta=-25$ MHz and a laser intensity of $s_0=50$. The temperature of the atomic sample is 1.1 mK which has been measured by ballistic time of flight. This temperature is well explained within the framework of Doppler cooling theory, predicting 1 mK for the
4.3 Conclusions

Figure 4.9 Ballistic flight of He(2^3S) atoms. At t=0 the atoms in the MOT are released by sweeping the trapping laser frequency far below resonance. The Time Of Flight of He^* atoms is measured by a microchannel plate mounted 70 mm from the trap. A grid in front of the channelplate is biased at +100 V to repel the ions produced in the MOT. The solid line is a fit of a Maxwell-Boltzmann velocity distribution at a temperature of 1.1 mK.

given laser parameters. The trap decay is exponential in time indicating that trap loss by intra-MOT collisions is small. This is confirmed by the analysis of the inelastic trap loss processes using ion mass spectrometry by which we have been able to separate the ion production of thermal processes from low-energy collision processes. The main ion production of about $2 \times 10^4$ c/s is due to the thermal PI reaction He^*+H_2O→H_2O^++He(1^1S)+e^- . The ion production due to PI in cold collisions is about $2 \times 10^3$ c/s.
Trapping Metastable Helium
Ejection of Trapped Atoms by Elastic Scattering at Thermal Energies

In an atomic trap operated at low densities elastic collisions with background gas atoms have been studied by measuring the loss of atoms from the trap. The use of an atomic trap provides an alternative technique to measure elastic scattering cross sections in addition to “traditional” atomic beam experiments. One advantage of using atomic traps over atomic beam experiments is that absolute elastic cross sections can be measured without the need to determine detector efficiencies or overlap integrals, thereby eliminating a large number of possible systematic errors. We have explored this technique by measuring decay of a metastable helium trap in environments of the different buffer gases He, Ne, Ar, Kr, Xe, H₂, D₂, O₂ and N₂.
Ejection of Trapped Atoms

5.1 Introduction

Nowadays atomic traps are widely used as a research tool in the field of atomic physics and quantum optics. Of particular interest is the magneto optical trap (MOT) which enables continuous cooling and trapping of large amounts of neutral atoms. The total number of atoms in a trap is determined by the loading rate and the loss rate of atoms. In a low density trap the loss of atoms is determined by elastic collisions with thermal background gas atoms. At high atomic densities, however, the main trap loss is by inelastic collisions of two trapped atoms at low collision energies. Thermal elastic scattering is a loss mechanism in spin polarized traps, e.g. static magnetic traps or spin polarized MOT’s, where scattering by background gas atoms limits the trap density and temperature.

Elastic scattering of neutral atoms has been investigated thoroughly the last decades in atomic beam experiments [35]. In “traditional” scattering experiments a well-collimated atomic beam crosses a volume containing dilute target gas. Scattering is usually observed by detecting atoms scattered out of the primary beam in the forward direction. Angle resolved detection is performed by using a narrow slit in front of a particle detector which can be rotated with respect to the incident beam to detect the scattered atoms. Typical angular resolutions in these measurements are in the range of 1–10 mrad. By measuring the intensity of scattered particles as a function of the angle, the small angle scattering regime and the rainbow scattering regime have been studied in detail [35].

Trap loss measurements in atomic traps provide an alternative way to study elastic atom scattering. When atoms in the trap are scattered in collisions with background gas the initially trapped atom can be ejected from the trap if sufficient energy is exchanged. Elastic scattering can be studied by monitoring the loss rate of trapped atoms when background gas atoms are introduced. There are a few special features when atomic traps are used to study elastic scattering. The first is that absolute scattering cross sections can determined without the need to measure detector areas, quantum efficiencies or beam overlap integrals [36] which is due to the specific collision geometry. So, by using atoms traps, a large number of systematic errors can be eliminated that limits the accuracy and traceability of absolute cross section measurements in traditional beam experiments. Secondly, collisions can be studied at sub-thermal energies because the trapped atom is initially at rest. This provides a check to see whether the interaction potentials is still applicable in the sub-thermal collision regime where interactions at larger distances dominates. A drawback of using an atomic trap is that no differential cross sections can be determined since collisions leading to trap loss occur over all angles.
5.2 Trap Decay

If an atom in the trap collides with an atom in the background gas, there is a large probability that the trapped atom will lost. The trap loss in a low density metastable helium trap can occur by either elastic or inelastic processes. In the inelastic process of Penning Ionization (PI) background gas atoms will de-excite metastable helium atoms in reactive collisions by which atoms are removed from the trap. Elastic scattering can lead to trap loss if due to the collision sufficient momentum is transferred from the “hot” background atom to the “cold” metastable helium atom. Since the trap depth in a typical MOT is only of the order of few Kelvin, and because of the relatively large cross sections for scattering the trap loss due to elastic collisions is the dominant loss process.

The amount of momentum transfer during the collision increases if the scattering angle of the collision increases. Trap loss occurs if the metastable helium atom acquires due to the collision a velocity larger than the escape velocity $v_e$. One can show from conservation of energy and momentum in the collision, that the minimum scattering angle $\theta_e$ (CM-frame) required to transfer a momentum of $m_{he}v_e$ to the cold atom is given by

$$\theta_e = 2 \arcsin \left( \frac{m_{he}v_e}{2\mu v} \right) \approx \frac{m_{he}v_e}{\mu v},$$ (5.1)

with $v$ the relative velocity of the collision, $\mu$ the reduced mass and $m_{he}$ the mass of the helium atom. The approximation is valid for small angles $\theta_e$.

The average collision rate $R$ is given by

$$R = n\bar{v}\sigma(T),$$ (5.2)

with $n$ the density of the atoms in the background gas, $\sigma(T)$ the elastic scattering cross section, averaged over the thermal distribution of the background atoms, and $\bar{v}$ the average collision velocity. The relative collision velocity is equal to the velocity of the background gas atom, since the metastable helium atoms are initially at rest. The thermal average of this velocity is given by

$$\bar{v} = \sqrt{\frac{8k_BT}{\pi m}},$$ (5.3)

with $m$ the mass of the background gas atom. The average relative energy $\bar{E}_{rel} = \frac{1}{2}\mu\bar{v}$ is proportional to the reduced mass and the collision energy is in most cases below thermal energies, since helium is in most cases much lighter than the background gas.
Not all elastic scattering processes lead to trap loss and we can split the total rate $R$ into two parts:

$$R = R_{\text{loss}} + R_{\text{recap}} = n\bar{v}(\sigma_{\text{loss}} + \sigma_{\text{recap}}),$$

(5.4)

where the first part describes the loss rate and the second part the “recapture” rate. By measuring the lifetime $\tau$ of the trap the loss rate $R_{\text{loss}}$ can be measured directly since $R_{\text{loss}} = 1/\tau$. So apart from the lifetime only the density of the background gas has to be measured in order to determine the elastic cross section. This is a large advantage compared to “traditional” scattering experiments with molecular beams, since the cross section depends in that case on the velocity and spatial distribution of both target and projectile and on the overlap of the beams, which are sources of systematic errors in these experiments [37]. Such systematic errors are avoided if an atomic trap is used instead, since for the determination of a cross section only time and density measurements are needed (see Eq. (5.4)), for which accurate standards are available.

In order to calculate the loss rate different methods can be used. We will first describe a classical model for the scattering process, since it rather directly shows how the loss rate depends on the experimental conditions. Then we will describe a quantum-mechanical model to evaluate the loss rate. In the last section we will describe the model potentials we have used.

### 5.2.1 The Classical Deflection Function

In classical mechanics the scattering angle in the center of mass frame is given by [8]

$$\theta = \pi - 2\int_{R_0}^{\infty} \frac{L}{\mu R^2} \left( \frac{2}{\mu} [E_{\text{rel}} - V(R)] - \frac{L^2}{\mu^2 R^2} \right)^{-1/2} dR,$$

(5.5)

with $E_{\text{rel}}$ the relative energy in the collision, $L = \mu v b$ the orbital angular momentum, $b$ the impact parameter, $V(R)$ the interaction potential and $R_0$ the distance of closest approach. We will consider the case, that the potential can be approximated by the long-range part given by $V(R) = -C_n/R^n$. For two neutral atoms the smallest value of $n$ contributing to the potential at long range is $n = 6$, i.e. a van der Waals interaction. If $E_{\text{rel}} \gg V(R)$ we can approximate the square root in Eq. (5.5). The deflection angle becomes

$$\theta = \frac{15\pi C_6}{16E_{\text{rel}} b^6},$$

(5.6)
Applying the relation for the scattering angle given in Eq. (5.1) we find the maximum impact parameter $b_{\text{max}}$. If we use the classical relation $\sigma = \pi b_{\text{max}}^2$ between the cross section and the maximum impact parameter we find for the loss cross section:

$$\sigma_{\text{loss}} = \left( \frac{15\pi^4 C_6}{8m_{\text{he}} v_e v} \right)^{1/3}.$$ (5.7)

Note, that the cross section is only weakly dependent on the $C_6$ coefficient, the escape velocity and the relative velocity. In order to change the cross section by a factor 2 one has to change one of these parameters by a factor 8. Furthermore, the classical cross section will diverge if the escape velocity goes to zero, which is due to the fact that the classical cross section diverges for small scattering angles. This is repaired by using the quantum-mechanical cross section, which we will discuss in the next section.

### 5.2.2 The Method of Partial Waves

In order to calculate the cross section for trap loss quantum mechanically the method of partial waves is employed. The total cross section is given by (chapter 2)

$$\sigma = \frac{4\pi}{k^2} \sum_{\ell} (2\ell + 1) \sin^2 \delta_{\ell}(k),$$ (5.8)

with $\delta_{\ell}(k)$ is the phase shift for scattering in the potential with a well-defined angular momentum $\ell(\ell + 1)\hbar$ and $k = \sqrt{2\mu E_{\text{rel}}}$ the wavenumber. The phase shift can be calculated by numerically integrating the Schrödinger equation for a particle with reduced mass $\mu$ in a potential $V(R) + \ell(\ell + 1)\hbar^2/2\mu R^2$. Numerical integration of this second order differential equation can be performed using the Numerov-method [38]. The solution for large range are matched with the analytical solutions of the Schrödinger equation in absence of the potential $V(R)$, which determines the phase shift $\delta_{\ell}(k)$.

The differential cross section $d\sigma/d\theta$ is given by [9]

$$\frac{d\sigma}{d\theta} = \frac{1}{k^2} \left| \sum_{\ell} (2\ell + 1)e^{i\delta_{\ell}(k)} \sin \delta_{\ell} P_{\ell}(\cos \theta) \right|^2,$$ (5.9)

with $P_{\ell}(\cos \theta)$ the Legendre polynomial for order $\ell$. Note, that contrary to the total cross section the differential cross section contains cross terms between different values of $\ell$, which causes constructive and destructive interference in the differential cross section. These interferences represent the effects of rainbow oscillations and glory scattering.
Ejection of Trapped Atoms

For the loss rate we have to integrate the differential cross section over the angle $\theta$ from the minimum angle $\theta_e$ to $\pi$. Furthermore, we have to integrate this rate over the velocity distribution of the background gas atoms. We find

$$R_{\text{loss}} = 2\pi n \int_0^\infty dv \int_{\theta_e}^\pi d\theta f(v) v \frac{d\sigma(\theta, v)}{d\theta} \sin \theta.$$  \hfill (5.10)

Here $f(v)$ is the distribution of the incoming velocities of the background gas atoms with mass $m$, which is given by the Maxwell-Boltzmann distribution at temperature $T$:

$$f(v) = 4\pi \left(\frac{m}{2\pi k_b T}\right)^{3/2} v^2 e^{-mv^2/2kT}$$  \hfill (5.11)

The loss cross section is then given by $\sigma_{\text{loss}} = R_{\text{loss}} / n \bar{v}$. In the quantum-mechanical calculation the integration over the velocity distribution will average out the interferences. Also in the classical case we have to average the loss rate over the Maxwell-Boltzmann distribution. However, since the classical cross section depends so weakly on the relative energy the averaging does not change the rate appreciably.

5.2.3 Interaction Potential

For both the classical and quantum-mechanical calculations we need to know the interaction potential. Since we will be discussing scattering of metastable helium on a large variety of different gases, we will construct a model potential with only a few free parameters, which can be used for all these systems. For the short range we will be using a Born-Mayer potential and add dispersion terms to describe the long range:

$$V(r) = a e^{-bR} - \sum_{n=3}^{8} \frac{\gamma_{2n+1}(bR)}{2n!} \frac{C_{2n}}{R^{2n}}.$$  \hfill (5.12)

In order to avoid divergence of the dispersion terms at short range we will cut-off these terms with the incomplete gamma function $\gamma_n$ [39] as discussed by Tang and Toennies [40]. The parameters $C_n$ are available in the literature for a large number of systems, which we will discuss later in this section. The parameters $a$ and $b$ can be determined by demanding that our model potential has a well depth $\epsilon_D$ and equilibrium position $r_D$ as given in the literature. A procedure to calculate the values of $a$ and $b$ for given values of $\epsilon_D$, $r_D$ and $C_n$’s is given by Tang and Toennies [40].
5.3 Experimental Setup

The parameters $C_6$, $C_8$ and $C_{10}$ are calculated for He* on He, Ne, Ar, Kr and Xe by Proctor and Stwalley [41]. They show, that for a system of two interacting particles, where one particle has a closed shell (in our case the rare gas atoms) and the other particle has only one active electron (in our case He*) the $C_n$ coefficients can be expanded in a term expansion, where the higher order terms decrease rapidly [42]. In this way they are able to determine the $C_n$ coefficients with an accuracy of 1–10%. In the case of $C_6$ the accuracy is 1–4%, where the lowest uncertainty is for He and the highest for Xe. The values for He* on rare gas atoms are given in table 5.1. Values for He* on diatomic gases H$_2$, D$_2$, N$_2$ and O$_2$ are not known. However, Proctor and Stwalley [43] have shown, that the $C_n$ parameters are proportional to the polarizability $\alpha$ of the gas. The proportionality constants can in our case be found by using the values for He* on the rare gas atoms. The values found this way are also shown in table 5.1. For higher order $C_n$ parameters we scale from lower order parameters according to [40]

$$C_{2n+6} = \left( \frac{C_{2n+4}}{C_{2n+2}} \right)^3 C_{2n}.$$  (5.13)

In order to test the validity of our model potential we have compared the results of the calculations for the loss rate with calculations performed with potentials published in the literature for He*-Ar [44], He*-Kr and He*-Xe [45]. In all cases we find that the rates calculated this way deviate not more than 5–10% from the values found by using the model potential. From this we conclude that we can obtain sufficient accuracy by using our model potential. The case of He*-He is special, since in that case the long range potential is not only determined by dispersion forces, but also by chemical binding. Therefore, we cannot accurately calculate the loss rate by using a model potential, but we will employ the potential published in ref. [46].

5.3 Experimental Setup

For the scattering experiments we have used a metastable He(2$^3$S) MOT (chapter 4). The trap is loaded with atoms from a slowed atomic beam using the technique of Zeeman compensated slowing. Both the slower and the MOT operate at the $2^3{\text{S}} \rightarrow 2^3{\text{P}}_2$ transition at 1083 nm. The laser light needed for slowing and trapping is generated by diode lasers. The trap is monitored by counting H$_2$O$^+$ ions produced in the Penning Ionization (PI) reaction of He(2$^3$S) and residual water (chapter 4). In figure 5.1 the ion detector setup is shown. Typically, we have an ion yield of $2 \times 10^4$ c/s. The trap is operated at low densities such that the ion production in cold collisions can be neglected. Under these circumstances the ion rate is
Ejection of Trapped Atoms

proportional to the number of trapped atoms, so the trap decay can be measured by probing ions produced in thermal processes. The trap contains about $4.0 \times 10^5$ He* atoms of which 80% are in the He($2^3S$) state and 20% are in the He($2^3P$) state. The lifetime of the trap is about 5 seconds at a background pressure of $4.0 \times 10^{-9}$ mbar.

The temperature of the MOT is 1.1 mK and has been measured by ballistic time of flight. The escape velocity of the trap, estimated by 1D simulations of the atomic motion in the trap, is about 50 m/s (see chapter 4).

A calibrated gas inlet and monitoring system is connected to the MOT chamber. The system allows us to inject gas into the experiment at well-defined pressures (uncertainty 5%) in the range of $10^{-9}$ to $10^{-6}$ mbar. The gas inlet system consists of a calibrated volume ($V_{ref}=778\pm5$ ml) connected to a capacitive membrane manometer (Baratron). The pressure in the meter is determined by the force which is exerted on the membrane by the gas. Therefore the pressure reading is independent of the atomic mass of the gas. The Baratron has been calibrated on an absolute scale using the vapor pressure of triple point water. The linearity of the meter is better than 1% in the pressure range of 0.1 mbar to 50 mbar, whereas the pressure can be conveniently monitored by a voltage over the membrane. The manometer we have used has a resolution of $1\times10^{-6}$ mbar by which injection of small but well-defined amounts of gas is possible. The system is connected to the MOT chamber via a precision needle valve which allows us to control the injection of gas into the experiment. The gas contents is monitored by a mass spectrometer (Balzers QMS 311). The calibration of the pressure in the MOT chamber was car-

### Table 5.1

<table>
<thead>
<tr>
<th>gas</th>
<th>$R_m$ [Å]</th>
<th>$\epsilon_m$ [meV]</th>
<th>$\alpha$ [a.u.]</th>
<th>$C_6$ [a.u.]</th>
<th>$C_8$ [$10^3$ [a.u.]]</th>
<th>$C_{10}$ [$10^6$ [a.u.]]</th>
<th>$a$ [a.u.]</th>
<th>$b$ [a.u.]</th>
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<td>0.35</td>
<td>0.80</td>
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<td>0.590</td>
<td>5.37</td>
<td>111</td>
<td>6.8</td>
<td>0.55</td>
<td>0.35</td>
<td>0.80</td>
</tr>
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<td>60.2</td>
<td>1.14</td>
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</tbody>
</table>

**ref. [12], [41, 43], [47], [41, 43], [41, 43]**
5.3 Experimental Setup

Figure 5.1  Ion detection in the MOT setup. The ions produced in the trap are accelerated in the electrical field produced by the biased grid in front of the channeltron. In the case of low trap densities, ions are mainly produced by the thermal PI reaction He*+H2O→H2O+He(1S)+e−. Therefore, the decay of a He* MOT can be monitored by ion detection instead of fluorescence.

Figure 5.2  Evacuation time of the MOT chamber for different gases. The solid line is a fit to the experimental data obtained by using elementary molecular flow dynamics taking into account our specific vacuum setup. The flow rate is mass dependent due to the connecting vacuum tubing between the turbo molecular pump and the MOT chamber. The data point for H2 is of the line because of the reduced pumping rate of the turbo molecular pump for H2.
Ejection of Trapped Atoms

ried out by means of volumetric expansion. A well-known flow of gas is injected into the UHV environment of the MOT chamber. The pressure in the chamber is determined by the gas flow in and the flow rate out of the chamber. The gas flow into the chamber was determined by injection gas at a constant rate set by the needle valve. In this way a leak with volume flow rate \( C_{\text{leak}} \) is introduced and gas will flow from the calibrated volume into the MOT chamber. A typical gas load in the MOT chamber is \( 10^{-4} \) mbar l/s. By monitoring the change of the pressure in the reference volume the gas load is known with an uncertainty of 1%.

The volume flow rate out of the chamber is determined by the capacity of the pump evacuating the chamber. The turbo molecular pump used (Balzers TPM 180) has a specified pump rate of 170 l/s for N\(_2\) and He and a rate 130 l/s for H\(_2\). The volume flow rate out of the MOT chamber has been determined independently by measuring the pressure response time after pressure changes were introduced in the MOT chamber (V=15.7 l). The measured volume flow rates for various gases are plotted in figure 5.2. The measured volume flow rate \( C_{\text{mot}} \) out of the MOT chamber is mass dependent and smaller than the volume flow rate \( C_{\text{pump}} \) of the turbo pump. This is because the turbo pump is mounted to the chamber using an extra vacuum cross piece, by which an extra pump resistance is introduced. As a consistency check \( C_{\text{pump}} \) has been determined using the measured rates and by modeling the vacuum tubing by elementary molecular flow dynamics (solid line in figure 5.2). We find \( C_{\text{pump}} = 172 \pm 3 \) l/s for all gases except H\(_2\). For H\(_2\) we have found \( C_{\text{pump}} = 131 \pm 2 \) l/s. These values agree very well with the manufacturers specifications. The pressure in the MOT chamber is given by the gas load \( C_{\text{leak}} \) into the MOT chamber and the volume flow rate \( C_{\text{mot}} \) out of the chamber according to

\[
P_{\text{MOT}} = \frac{C_{\text{leak}}}{C_{\text{mot}}} P_{\text{ref}}.
\]

Under steady state conditions the ion yield of the mass spectrometer has been calibrated to this absolute pressure. The mass spectrometer has been calibrated independently for all gases under consideration so that the gas contents and partial pressures during the decay measurements are known on an absolute scale, within an uncertainty of about 10%.

### 5.4 Results

The gases He, Ne, Kr, Xe, H\(_2\), D\(_2\), O\(_2\) and N\(_2\) were injected as buffer gases into the MOT chamber to study trap decay as a function of the density. For all these elements the partial pressure was varied in the region of \( 10^{-8} \) to \( 10^{-6} \) mbar. The decay time of the MOT was measured by interrupting the loading of atoms into the
5.4 Results

trap. This was realized by blocking the slowing laser beam. During the interruption the decay rate of the MOT was determined by measuring the ion production as a function of time. For every decay curve the background gas spectrum was analyzed to determine the background gas contents and partial gas densities. In figure 5.3 a measurement is shown for the cases that no gas was introduced and for the case that He was introduced as a background gas. The MOT chamber was not baked out in these measurements so that the background pressure of about $5 \times 10^{-8}$ mbar was due to residual water. Because of the large PI cross section of the He$^*$+H$_2$O$\rightarrow$H$_2$O$^+$+He(1$^1$S)+e$^-$ reaction an large ion yield was obtained which was used to determine the trap loss. From these figures we see that if the background density is increased both the lifetime of the trap and the stationary ion yield are reduced. Both are reduced because the collision rate is increased at higher densities Eq. (5.4). The small ion production of about 100 Hz, which is visible in figure 5.3 after the MOT signal has vanished, is from the PI reaction of He$^*$ in the atomic beam and residual water.

The decay rate of the MOT has been determined by a fit of the ion curves with an exponential decreasing function. Typical values for the decay rate lie in the range of 0.5 to 3 Hz, corresponding to lifetimes of the trap of 2 to 0.3 seconds. The uncertainty in the decay rate is typically 3% which is due to the statistical spread of the ion yield data. The partial pressure measurements is reproducible within 5% and is limited by gas contamination during the gas inlet. The decay rates as a function of the gas densities have been fitted for each collision system to obtain the trap loss cross section given by Eq. (5.4). In figure 5.4 the decay rate for different partial pressures of He is plotted. The decay rate is linear in the background gas density indicating that the trap loss mechanism is due to binary collisions. The solid line in the plot is a linear fit through the data set. The slope of the line is given by the thermal average rate constant for trap loss $\sigma_{loss} \bar{v}/kT$. Trap decay measurements at different background pressures have been performed for the rare gases and diatomic molecules He, Ne, Kr, Xe, H$_2$, D$_2$, N$_2$ and O$_2$ to determine the trap loss cross sections for collisions with He(2$^3$S). The decay rate and background density in Eq. (5.4) have been obtained as discussed above for the case He+He(2$^3$S). For the calculation of $\sigma_{loss}$ the collision rates are divided by the mean velocity of a Maxwell-Boltzmann distribution at 300 K given by $\bar{v} = \sqrt{8kT/\pi m}$. The experimentally determined trap loss cross sections for the various background gases are given in table 5.2.
Figure 5.3 Trap decay measurements for the case that (a) no buffer gas used and (b) when helium is introduced in the background. Each data set is fitted to an exponential function (solid lines) from which the decay rate of the MOT is obtained. The trace gas spectra are plotted in the insets, demonstrating that only the pressure of helium is increased.
5.5 Analysis

For the analysis of the data we have performed calculations based on the method of partial waves. For a given system we have calculated for a number of energies in the Maxwell-Boltzmann distribution the differential cross section $\frac{d\sigma}{d\theta}$ and integrated the part of the differential cross section above the minimum angle $\theta_e$. Note, that $\theta_e$ depends on the relative energy through the incoming velocity $v$. As an example we have plotted the differential cross section for the system He*+Kr in figure 5.5 for an energy of 1.5 meV. The minimum angle $\theta_e = 14^\circ$ for this case is indicated with an arrow. Here we have assumed that the escape velocity is 50 m/s, which follows from a simulation of the motion of the atoms in the trap. As can be seen from figure 5.5 the loss rate is not critically dependent on the escape velocity, since apart from atoms scattered in the forward peak all elastic scattering processes lead to trap loss. The results from this analysis are shown in table 5.2, where we have calculated both the total and the loss cross sections. As can be seen from the table almost 45–60% of the elastic collisions lead to trap loss, where the lowest percentage is for the heavy systems and the highest values for the light systems. For comparison we have also shown the results of the classical calculations. The
Ejection of Trapped Atoms

<table>
<thead>
<tr>
<th>gas</th>
<th>cross section [Å²]</th>
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<tr>
<td></td>
<td>loss</td>
</tr>
<tr>
<td></td>
<td>loss</td>
</tr>
<tr>
<td>H₂</td>
<td>140 ± 13</td>
</tr>
<tr>
<td>D₂</td>
<td>133 ± 8</td>
</tr>
<tr>
<td>He</td>
<td>114 ± 11</td>
</tr>
<tr>
<td>Ne</td>
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</tr>
<tr>
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<tr>
<td>O₂</td>
<td>195 ± 25</td>
</tr>
<tr>
<td>Ar</td>
<td>165 ± 18</td>
</tr>
<tr>
<td>Kr</td>
<td>400 ± 24</td>
</tr>
<tr>
<td>Xe</td>
<td>321 ± 20</td>
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Table 5.2 Absolute trap loss cross sections for the process of elastic scattering of trapped He(2³S) atoms by different background gas atoms and molecules at 300 K.

<table>
<thead>
<tr>
<th>gas</th>
<th>μ [amu]</th>
<th>v [m/s]</th>
<th>E_{rel} [meV]</th>
<th>θ_e [mrad]</th>
<th>θ_c [mrad]</th>
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</thead>
<tbody>
<tr>
<td>H₂</td>
<td>1.33</td>
<td>1761</td>
<td>22.9</td>
<td>85</td>
<td>38</td>
</tr>
<tr>
<td>D₂</td>
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<tr>
<td>He</td>
<td>2.00</td>
<td>1245</td>
<td>16.5</td>
<td>80</td>
<td>39</td>
</tr>
<tr>
<td>Ne</td>
<td>3.33</td>
<td>557</td>
<td>5.5</td>
<td>108</td>
<td>50</td>
</tr>
<tr>
<td>N₂</td>
<td>3.50</td>
<td>471</td>
<td>4.1</td>
<td>121</td>
<td>39</td>
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<tr>
<td>O₂</td>
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<td>Ar</td>
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<td>192</td>
<td>50</td>
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<td>218</td>
<td>1.0</td>
<td>236</td>
<td>69</td>
</tr>
</tbody>
</table>

Table 5.3 Collision parameters for the various systems. The escape velocity is 50 m/s.

The correspondence between classical and quantum mechanical calculation is rather good, indicating that the classical result provides a good estimate for the loss cross section. This is to be expected, since the minimum angle θ_e is well above the so-called critical angle θ_c, which forms the transition angle from classical scattering to quantum mechanical scattering [8]. In our case θ_e is in the order of 40–70° (see table 5.3).
5.6 Discussion

Figure 5.5 Calculated differential elastic scattering cross section for He\((2^3\text{S})+\text{Kr}\) (CM-frame) at 1.5 meV energy. The strong increase for angles near 5° is due to small angle scattering. Trap loss is only possible for scattering angles larger than 11° (192 mrad), meaning that the trap loss cross section is about half of the total elastic scattering cross section.

5.6 Discussion

When we compare the experimental results with the theoretical trap loss cross sections in table 5.2, we observe that the cross sections of the light collision systems are in reasonable agreement. For Ne to Xe, but with the exception of Kr, the calculated scattering cross sections are systematically larger than the experimental values. For the case of Ar the calibration of the pressure may contain a systematic error. For unknown reasons the calibration of the pressure for Ar was only reproducible within a factor of two. We will discuss a number of complications that might explain the observed discrepancy for the heavy collisions systems.

In principle the calibration procedure may contain systematic errors, if the gas density in the MOT chamber is not distributed homogeneously. Since the gas is pumped out of the chamber through a hole from one side, there is a density gradient in the MOT chamber present. This effect should be most pronounced for gases that are rapidly pumped out of the chamber i.e. for light gases. However, since we have reasonable agreement between the experiment and calculations for the light systems, it is obvious that the effect mentioned here can not explain the discrepancy.
found in the experiments.

Another complication is that we have neglected that a trapped atom is 20% of the time in the He(2^3P) state. If we take this into account the actual collision rate on a single trapped atom is given by

\[ R = n \bar{\nu} (1 - \pi_p) \sigma_e + \pi_p \sigma_p \]

(5.15)

where \( \pi_p \) is the fraction of He(2^3P) atoms, \( \sigma_e \) the elastic scattering cross section in He(2^3S)+X collisions and \( \sigma_p \) the elastic scattering cross section in He(2^3P)+X collisions. If \( \sigma_e \approx \sigma_p \), than the error caused by neglecting the \( \pi_p \) population is small, but if \( \sigma_p \) deviates substantially from \( \sigma_e \) this might cause a large discrepancy between the calculated and measured trap loss cross section. Only the case that \( \sigma_p \) is smaller than \( \sigma_e \) can explain the observation of a smaller scattering cross section. We have, however, no reason to assume that the elastic scattering cross section for He(2^3P)+X systems is less than for He(2^3S)+X systems.

Up to this point we have neglected the effect of reactive collisions (PI) on the elastic cross section. The modification of the total scattering cross section by PI should be most pronounced in the collision systems where atoms reach a close approach. This is because the auto-ionization probability (see chapter 2) decreases exponentially with distance. In our experiments the heavy gas atoms have to approach the He(2^3S) atom to short distances to initiate trap loss. This is due to the low relative velocities in these collisions only in head-on collisions sufficient energy can be exchanged to induce trap loss. We have investigated the modification of the elastic cross section by including the process of PI in our Xe+He(2^3S) calculations (chapter 2). We find that the trap loss cross section is lowered, but only by 20%. We therefore conclude that an effect is present, but it cannot account for the large discrepancy found. Motivated by this we are currently investigating the effect of reactive X+He(2^3P) collisions. Such light induced collisions are expected to enhance the ionization cross section dramatically (see chapter 6). The reduction in the calculated elastic trap loss cross section can therefore be much more severe.

5.7 Conclusions

An alternative way to study elastic scattering of neutral atoms at thermal energies is by using atomic traps. An advantageous feature of using trapped atoms is that cross sections can be determined on an absolute scale. By monitoring trap decay in a He* MOT we have determined absolute elastic scattering cross sections for He(2^3S)+X collisions at a temperature of 300 K. We have measured absolute trap loss cross sections for He, Ne, Ar, Kr, Xe, H₂, D₂, O₂, N₂. We find that the scattering cross
sections found both classical and quantum theory are in good agreement with the measurements of He, H\(_2\) and D\(_2\). The measured cross sections for the more heavy elements, but with the exception of Kr, are systematically too low. The discrepancy for the heavy collision systems is probably due to the fact that the collisions occur in the near resonant light field of the MOT.
Chapter 6

Penning Ionization of Metastable Helium at mK Temperatures

In this chapter we report our study of the Penning Ionization (PI) process in low energy He(2S)+He(2S) and He(2S)+He(2P) collisions. We have used a magneto optical trap containing He(2S) atoms at a temperature of 1.1 mK. The He(2S)+He(2S) collisions have been studied by switching the laser beams in the trap ‘off’, whereas the He(2S)+He(2P) collisions are studied in a strong laser field, resonant with the 2S→2P transition. We have detected the ions produced in PI and AI reactions and find that the total PI cross section for He(2S)+He(2P) collisions is about 7 times larger than for He(2S)+He(2S) collisions. The AI fraction in He(2S)+He(2P) collisions of 0.16 is 5 times larger than the AI fraction of 0.03 in He(2S)+He(2S) collisions. The large AI fraction is explained by the fact that ionization occurs directly in the He(2S)+He(2P) excited state channel.
6.1 Introduction

Low energy collisions can be studied in magneto optical traps (MOT) at typical temperatures in the range of $10 \mu K$ to $10 mK$. In a trap atom-atom collisions can be investigated at low kinetic energy for which the collision dynamics is sensitive to the interatomic potential at large distances. Of particular interest are the collision systems consisting of two metastable state atoms such as He*, Ne*, Kr* or Xe*. The electronic spectra of metastable state atoms is easier compared to alkali systems due to the absence of hyperfine structure. For He* the fine structure is relatively simple and it resembles the ground state of a pseudo single electron atom to a large extend. Therefore this system can be considered as a model system for low-energy atom-atom scattering. From an experimental point of view metastable state systems are interesting because of the Penning Ionization (PI) process allows direct detection of the outcome of a collision. Monitoring trap loss by detection of reactive products such as ions seems to be an excellent approach to study collisions in cold samples of atoms [4–6].

The collision system consisting of two triplet metastable helium atoms is especially interesting due to its simple electronic structure and its small mass. Concerning the electronic structure, it is well-known that the spin states in atomic helium are well defined [8] and that transitions between triplet and singlet states are forbidden to a very high degree. This is reflected by the extremely long lifetime of the He(23S) state which is in excess of two hours. The electronic structure of helium molecules also have interesting features. The total spin of the molecular states of the He(23S)+He(23S) system is expected to be a ‘good’ quantum number due to the Wigner spin rule stating that the total spin in molecular transitions is conserved. The consequence for the He(23S)+He(23S) molecule is that PI from the $1^3\Sigma_g^+$ state is forbidden since this doublet state can not couple to the singlet $1^1\Sigma_g^+$ and triplet $1^3\Sigma_u^+$ ionic states (see chapter 2). That the Wigner spin rule holds for the $1^3\Sigma_g^+$ is confirmed in the experiments by Müller et al. [16] for collisions at thermal energies. The small reduced mass of the He(23S)+He(23S) system leads to the special situation that scattering is already reduced to s-wave at a relatively high temperature of $10 mK$ [16] whereas typical temperatures of $1 mK$ can be reached in experiments using standard laser cooling techniques [7]. Therefore, the quantum regime of s-wave scattering can be accessed experimentally without much effort.

In the experiments discussed in this chapter we have used a MOT containing He(23S) atoms at a temperature of 1.1 mK (see chapter 4) to study PI and AI in
6.2 Experiments

He\(^{23}\text{S}\) + He\(^{23}\text{S}\) collisions by the reaction

\[
\text{He}^{23}\text{S} + \text{He}^{23}\text{S} \rightarrow \begin{cases} 
\text{He}^+ + \text{He}^{1\text{S}} + \text{e}^- \\
\text{He}_2^+ + \text{e}^-
\end{cases},
\]

which we will refer to as \(S+S\) or ground state collision. When in the exit channel a molecular ion is produced the process is called Associative Ionization (AI). We define the AI fraction as that fraction of the total ionization process that leads to the formation of molecular ions. At mK temperatures it is expected that only the \(1^1\Sigma_g^+\) molecular state is accessed in PI process (see chapter 2).

When two He\(^{23}\text{S}\) atoms collide in a near resonant light field excitations to He\(^{23}\text{S}\)+He\(^{23}\text{P}\) states can occur during the collision. This kind of collision is called an optical collision. Such collisions have been studied in a number of systems [1–7], where enhanced trap loss has been observed when laser light was introduced detuned slightly below atomic resonance. In this work we have studied ionization in He\(^{23}\text{S}\)+He\(^{23}\text{P}\) collisions, which we will refer to as \(S+P\) or optical collisions, by the PI reaction

\[
\text{He}^{23}\text{S} + \text{He}^{23}\text{P}_2 \rightarrow \begin{cases} 
\text{He}^+ + \text{He}^{1\text{S}} + \text{e}^- \\
\text{He}_2^+ + \text{e}^-
\end{cases}.
\]

This reaction is essentially the same as the reaction in ground state collisions, except that one atom in the entrance channel is excited to the He\(^{23}\text{P}\) state by a resonant laser light. The change of the atomic states in the entrance channel will in general lead to a change in the PI rate and AI fraction. The reactive He\(^{23}\text{S}\)+He\(^{23}\text{P}\) collisions in the system are in some respects different from similar type of collisions which have been observed in systems of Rb [1] and Xe* [3]. This is because the critical distance, \(R_{\text{max}}\), over which atoms in the excited state can travel before decaying back to the ground state is large compared to \(R_\lambda\), which is the typical interatomic distance of molecular excitation (see chapter 2). Our experimental conditions are such that \(R_{\text{max}}\) is about 3000 Å whereas \(R_\lambda=1700\) Å. It is therefore to be expected that in a He\(^{23}\text{S}\) trap, S+P collisions can lead to reactive collisions and thus trap loss, if the laser light is detuned on resonance of the \(2^3\text{S}\rightarrow 2^3\text{P}_2\) transition (see figure 6.1). The feature of non-vanishing trap loss at atomic resonance is different from other traps [1–3], where the trap loss by S+P collisions is zero due to the short molecular lifetimes.

6.2 Experiments

The He\(^*\) MOT used in the experiments is described in chapter 4. Atoms are loaded into the trap using a Zeeman compensated slowed atomic beam. Both the slower
PI of He* at mK Temperatures

Figure 6.1 (solid arrows) S+P collisions in the He*+He* system induced at large distances. At zero laser detuning one atom of a He(2 3S)+He(23S) pair is excited to the P-state at an interatomic distance in the order of an optical wavelength. Because of the long lifetime of the He(2 3P) state the He(23S)+He(23P) collision complex lives sufficiently long, so that the collision complex can reach short interatomic distances where ionization takes place. (dashed arrow) S+S collisions in the He*+He* system. Since the PI reaction is also possible from the He(23S)+He(23S) complex, ionization along this route is possible as well.

and the MOT operate at the 2 3S → 2 3P2 transition at λ=1083 nm. The line width of the optical transition is 1.6 MHz, corresponding to a radiative lifetime of 98 ns for the He(23P) state. The saturation intensity of the 2 3S → 2 3P2 transition is 160 µW/cm². For slowing and trapping we use two single mode diode lasers, generating 20 mW optical power each. We monitor He* atoms in the trap by detecting ions which are produced in PI reactions. A typical ion yield is 2×10⁴ c/s. The lifetime of our trap is about 5 seconds at a background pressure of 4.0×10⁻⁹ mbar. The temperature of the atoms in our trap is 1.1 mK and has been measured by ballistic TOF. Our MOT contains about 4.0×10⁵ He* atoms, 80% are in the He(2 3S) state and 20% are in the He(2 3P2) state. The ions produced in the trap are analyzed by their mass using a quadrupole mass spectrometer, which is incorporated in the MOT setup (see figure 6.2). The resolution of 1 amu is sufficient to resolve He⁺ from He₂⁺. With this device we have demonstrated that the main ion production in the trap is by the PI reaction of H₂O+He* (see chapter 4). The mass spectrum of ions produced in a He* MOT is plotted in figure 6.3. In this chapter we focus on cold collisions in our trap, so we only consider the production of He⁺ and He₂⁺.
6.2 Experiments

Figure 6.2 Top view on the ion mass spectrometer monitoring the He* MOT. The head of the device is biased to $-200 \text{ V}$ to extract ions out of the trap. The ions are detected at the exit of the spectrometer with a channeltron. Mass selection is obtained by an electrical quadrupole section. The spectrometer is put in the plane of symmetry of the inhomogeneous magnetic field of the MOT to minimize the magnetical deflection of the ions.

6.2.1 Optical Manipulation of Collisions

The MOT is operated at a detuning of $\delta=-25 \text{ MHz}$ at a laser intensity of 8 mW/cm$^2$ to obtain a dense sample of cold atoms (see chapters 4). The on-resonance saturation parameter is $s_0=50$ for each of the six trapping beams. The trapping laser frequency is periodically switched with a frequency of 50 kHz and a duty cycle of 10% (see figure 6.4), while the MOT operates 90% of the time at a fixed density and temperature. By the frequency switching we are able to manipulate the atomic S- and P-state population. The switching procedure is explained in figure 6.4. In a first interval of 20 $\mu$s the laser is detuned 350 MHz below the $^2S\rightarrow ^2P_2$ transition. During this non-trapping time the expansion of the atomic sample by ballistic flight can be neglected. Since this non-trapping time is much longer than the lifetime of the P-state, all atoms will be in the S-state in this period. We assume that in this time interval only reactive S+S collisions can contribute to the ionization signal, i.e., ionization by molecular excitations is neglected at this large laser detuning. In
Figure 6.3 Mass spectrum of the ions produced in the He* MOT operating at $\delta=-25$ MHz. The background pressure in the MOT was $4.0 \times 10^{-9}$ mbar. The 19.6 eV internal energy of He(2^3S) is sufficient to ionize H$_2$O contained in the background gas, which has an ionization potential of 12.6 eV. The He$^+$ and He$_2^+$ ions in the spectrum are due to cold collisions. The background level of ion production is less than 0.1 c/s per amu and has been measured with the MOT lasers turned off.

This period the ionization rate $I_{ss}$ is given by

$$I_{ss} = \frac{1}{2} n^2 \sigma_{ss} v V,$$

where $v$ is the relative velocity of the atoms in the entrance channel, $V$ is the volume of the atomic sample, $n$ is the density of the trapped atoms and $\sigma_{ss}$ is the PI cross section if the He$^+$ and He$_2^+$ productions are added and it is the AI ionization cross section if only He$_2^+$ is considered. In a second interval of 20 $\mu$s the laser frequency is ramped to resonance. By saturated optical pumping half of atoms are in the S-state and the other half in the P-state. The total ionization rate is given by the sum of the rates for S+S, P+P and S+P collisions according to

$$I_{tot} = \left( \frac{1}{2} n^2 \sigma_{ss} + n_s n_p \sigma_{sp} + \frac{1}{2} n^2_p \sigma_{pp} \right) v V$$

At resonance half of the population is in the S state and the other half in the P-state: $\pi_s = \pi_p = 1/2$. Because of the large ionization cross section of the S+P process the
6.2 Experiments

Figure 6.4  (a) The frequency of the trapping laser is modulated in time. The MOT operates continuously while the laser frequency is periodically switched to \(-350\) MHz for a duration of 20 \(\mu\)sec and then ramped through resonance in a second time interval. (b) \(\text{He}_2^+\) production when the switching procedure is applied. The \(\text{He}_2^+\) yield is suppressed when the laser is detuned far off resonance. In this time interval the ionization rate \(I_{ss}\) is measured. At zero detuning the ionization level reaches a maximum of \(I_{sp}\).
total ionization rate at zero detuning can be approximated by

\[ I_{sp} = \frac{1}{4}(\sigma_{ss} + \sigma_{sp})n v V, \quad (6.5) \]

where we have implicitly assumed that \( \sigma_{pp} = \sigma_{ss} \). The assumption that the S+P process dominates the PI process over the P+P process has been verified experimentally by Bardou et al. [7]. In figure 6.4 the He\(^+\) \( P \) ionization rate in the MOT is plotted when the switching procedure described above is applied. The ionization rate reaches a minimum in the time interval when the light is strongly detuned. When the frequency is ramped to resonance the ionization rate increases by about 50\% with respect to the steady state ionization level. From this we can infer, that for a laser detuning of \(-25 \) MHz the P-state population is about 20\%. The ionization rate reaches a maximum at zero detuning with an uncertainty of about 3 MHz. When the laser frequency is switched from \(-350 \) MHz to \(-25 \) MHz, some time is needed (30 \( \mu s \)) before the steady state ionization rate is recovered. This feature is still under investigation. We can rule out that the effect is caused by an instant change of the density or the temperature of the MOT since the response time of the trap is 2 ms (see chapter 4).

### 6.2.2 Results

The ionization rates \( I_{ss} \) and \( I_{sp} \) have been measured for the production of He\(^+\) and He\(\_2\)\(^+\) by selecting ions with masses 4 and 8 amu similar to the measurement in figure 6.4. A typical He\(\_2\)\(^+\) ion rate in optical collisions is about 200 c/s. To correct for the slowly changing density of the atomic sample, the relative production of He\(\_2\)\(^+\) and He\(^+\) in the different measurements have been normalized to the peaks in the ion mass spectrum of figure 6.3, where the ratio of He\(\_2\)\(^+\) to He\(^+\) is accurately known. In this way we have obtained four ionization rates which are given in table 6.1 and which can be used to determine PI and AI cross sections for the different processes. From this table we first remark that the ionization rate dramatically increases when resonant light is introduced. This is due to the large ionization cross section for He(2\(^3\)S)+He(2\(^3\)P) collisions, which already has been reported in [7] and has also been observed for the same type of collisions in other systems as well [1–3]. Secondly, the ionization ratio He\(\_2\)\(^+\)/He\(^+\) changes when the collisions are manipulated by light. This effect has not been observed before and is due to a change of the ionization channel, which will be discussed later.
6.2 Experiments

<table>
<thead>
<tr>
<th>Collision process</th>
<th>Ionization rate [c/s]</th>
<th>$\text{He}^+$</th>
<th>$\text{He}_2^+$</th>
<th>$\text{He}_2^+/\text{He}^+$</th>
</tr>
</thead>
<tbody>
<tr>
<td>S+S</td>
<td>10 ± 1</td>
<td>11 ± 2</td>
<td>1.1 ± 0.2</td>
<td></td>
</tr>
<tr>
<td>S+P</td>
<td>75 ± 6</td>
<td>225 ± 10</td>
<td>3.0 ± 0.3</td>
<td></td>
</tr>
</tbody>
</table>

Table 6.1 Measured ionization rates in S+S and S+P collisions. The total ionization rates are enhanced in optical collisions by the large PI cross section for S+P collisions. The $\text{He}_2^+/\text{He}^+$ ratio changes by about a factor of three indicating that a different ionization channel is accessed when collisions take place in a resonant light field.

![Figure 6.5](image)

Figure 6.5 Ion extraction by the mass spectrometer as a function of the energy. All ions with kinetic energies below 10 meV are detected. At higher energies the ion trajectories spread out and ions will be lost.

6.2.3 Ion Detection

From the measured ionization rates the PI and AI cross sections in ground state and optical collisions can be determined by using Eq. (6.3) and (6.5). However, there is a complication for the detection of $\text{He}^+$. Due to the relative high kinetic energy, $\text{He}^+$ ions will spread out during the extraction and a large number of ions will therefore not enter the spectrometer. We have studied this effect in numerical simulations of the ion trajectories, taking into account the static electrical field configuration of the MOT. The ‘transmission’ function of the spectrometer for ion energies in the range of 0 to 100 meV is plotted in figure 6.5. It can be seen that the transmission strongly decreases for energies above 10 meV. Although we study low
energy collisions He$^+$ ion fragments can have considerable energies (0–500 mV) since part of initial potential energy in the reaction is transferred to kinetic energy by the PI process. The transmission of He$^+_2$ is unity because the kinetic energy that is gained by molecular ions is negligible in cold collision reactions.

In order to calculate the fraction of He$^+$ ions that is detected in our experiments, we have calculated the electron spectra for collisions at 100 neV kinetic energy to determine the energy distribution of the ions. The kinetic energy distribution of the ions is directly related to the electron energy in low energy collisions: since
6.3 PI and AI Cross Sections

We have corrected the measured ion yield in table 6.1 for the transmission efficiency of the spectrometer to obtain the corrected ion yields. We have assumed that the ionization process in S+P collisions is initiated directly from the S+P potential. By taking into account the different S- and P-state populations the PI and AI cross sections have been obtained for S+S and S+P collisions. Because the transmission

<table>
<thead>
<tr>
<th>process</th>
<th>ion mass [amu]</th>
<th>transmission fraction</th>
<th>correction factor</th>
</tr>
</thead>
<tbody>
<tr>
<td>S+S</td>
<td>4</td>
<td>0.03</td>
<td>33.3</td>
</tr>
<tr>
<td>S+P</td>
<td>4</td>
<td>0.064</td>
<td>15.6</td>
</tr>
<tr>
<td>S+S</td>
<td>8</td>
<td>1</td>
<td>1.0</td>
</tr>
<tr>
<td>S+P</td>
<td>8</td>
<td>1</td>
<td>1.0</td>
</tr>
</tbody>
</table>

Table 6.2 Correction factors for the detection efficiency of ions in S+S and S+P collisions. Molecular He$^+_2$ ions which are ejected in the process of AI and carry only the electron recoil energy, which is 0.1 eV. At these low kinetic energies the transmission is 100%.

the initial kinetic energy in the collision is zero the energy of a He$^+$ ion is the same as the energy of the He(1$^1$S) atom. The energy is then the nominal energy of the He$^+$+He$^+$ state minus the energy of the ejected electron. The PI process is modeled using theory similar to ref. [16]. For the auto-ionization width function (see chapter 2) for PI in ground state collisions we have used

$$\Gamma(1^1\Sigma^+_g) = \Gamma_0 e^{(-R/R_0)},$$

(6.6)

with $\Gamma_0 = 8.163$ eV and $R_0 = 0.487 \text{ Å}$.

The process of PI from the molecular states of the He(2$^3$S)+He(2$^3$P) system has neither been investigated in experiments nor in theory. For the auto-ionization width of the S+P potential we used the same width function as for the S+S potential, which thus is treated as an empirical width. The $1^1\Sigma^+_g$ and $2^1\Sigma^+_u$ ionic potentials used in the calculations are taken from ref. [16]. The He(2$^3$S)+He(2$^3$P) excited state potential is from ref. [49] and is plotted in figure 6.7. In figure 6.6 the calculated electron spectra for the electronic transitions to the $2^1\Sigma^+_g$ ionic state are given for S+S and S+P collisions. We assume that 50% of the total ionization is by transitions to the repulsive $2^1\Sigma^+_g$ ionic state [16]. We have folded our electron spectra with the transmission function to obtain the overall transmission efficiency of He$^+$ ions. The transmission coefficients are given in table 6.2.

6.3 PI and AI Cross Sections

We have corrected the measured ion yield in table 6.1 for the transmission efficiency of the spectrometer to obtain the corrected ion yields. We have assumed that the ionization process in S+P collisions is initiated directly from the S+P potential. By taking into account the different S- and P-state populations the PI and AI cross sections have been obtained for S+S and S+P collisions. Because the transmission
function for \( \text{He}^+ \) is unity, we can estimate the AI cross section from the measured steady state molecular ion production in ground state collisions, which is given by

\[
I_{ss}(\text{He}^+) = \frac{1}{2} \eta n^2 V \sigma_{AI} v,
\]

where \( \eta \) is the efficiency of the channeltron, \( n \) is the density of trapped atoms, \( V \) is the volume of the atomic sample and \( v \) the relative velocity. If we use \( \eta=40\% \), \( n=1.0 \times 10^8 \) atoms/cm\(^3\), \( V=4.0 \times 10^{-3} \) cm\(^3\) (chapter 4) and \( I_{ss}=11 \) c/s we obtain an AI cross section of 45 Å\(^2\). By using the experimentally determined AI fraction of 0.03, we obtain a total PI cross section of \( (1.5\pm0.7) \times 10^3 \) Å\(^2\). The large uncertainty is due the uncertainty in \( n \) and \( V \). This AI cross section is used to calibrate the other ionization cross sections for S+S and S+P collisions using the relative ion productions in the experiments correct for the transmission effect. The ionization cross sections we have obtained are given in table 6.3.

### 6.4 Discussion

In table 6.3 we have included all published PI and AI cross sections for the He(2\(^3\)S)+He(2\(^3\)S, 2\(^3\)P) systems in low energy collisions. We have been able to measure the AI and PI cross sections for He(2\(^3\)S)+He(2\(^3\)S) collisions at 100 neV collision energy and have found an ionization cross section for this process of 1500 \( \pm \) 700 Å\(^2\). We have determined the PI and AI cross sections for the different processes by calibrating the relative ionization cross sections to the measured PI cross section in He(2\(^3\)S)+He(2\(^3\)S) collisions. The cross section we have obtained is consistent with ionization cross section for the process calculated by Müller et al. [16] at 1 µV.

The ionization cross section in S+P collisions is \( (1.2\pm0.5) \times 10^4 \) Å\(^2\). There is an extreme large discrepancy with the result reported by Bardou et al. [7] who found an ionization cross section at 100 neV in the range of 0.15–2.4 \( \times 10^7 \). We can not explain the extremely large discrepancy between the results.

In our work we have observed that the AI fraction for He(2\(^3\)S)+He(2\(^3\)S) collisions that occur in a resonant light field is 0.16 and is about 5 times larger than in absence of the light. We have found that this is due to the fact that ionization process proceeds to the He(2\(^3\)S)+He(2\(^3\)P) channel. When the light is turned ‘on’ the He(2\(^3\)S)+He(2\(^3\)S) system is excited to He(2\(^3\)S)+He(2\(^3\)P) complex. Since the P-state atom can travel over a distance of about 3000 Å before it decays (chapter 2) a major part of the excited collision complexes will not decay by spontaneous emission before ionization takes place. As a result an major fraction of ionization is directly through the S+P channel. We have investigated what the actual mechanism is that is responsible for the large AI fraction in the S+P channel. For this we have
6.4 Discussion

<table>
<thead>
<tr>
<th>Reference</th>
<th>Energy</th>
<th>Reaction</th>
<th>PI cross section [Å²]</th>
<th>AI fraction</th>
</tr>
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<tr>
<td>Present Work (experiment)</td>
<td>100 neV</td>
<td>He(2³S)+He(2¹S)</td>
<td>(1.5±0.7)×10³</td>
<td>0.030(3)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>He(2³S)+He(2³P)</td>
<td>(1.2±0.5)×10⁴</td>
<td>0.16(2)</td>
</tr>
<tr>
<td>Present Work (theory)</td>
<td>100 neV</td>
<td>He(2³S)+He(2¹S)</td>
<td>-</td>
<td>0.11</td>
</tr>
<tr>
<td></td>
<td></td>
<td>He(2³S)+He(2³P)</td>
<td>-</td>
<td>0.055</td>
</tr>
<tr>
<td>Bardou et al. [7]</td>
<td>100 neV</td>
<td>He(2³S)+He(2¹S)</td>
<td>0.15–2.4×10⁵ †</td>
<td>-</td>
</tr>
<tr>
<td>(experiment)</td>
<td></td>
<td>He(2³S)+He(2³P)</td>
<td>0.15–2.4×10⁷</td>
<td>-</td>
</tr>
<tr>
<td>Müller et al. [16]</td>
<td>1µeV</td>
<td>He(2³S)+He(2¹S)</td>
<td>561††</td>
<td>0.25††</td>
</tr>
<tr>
<td>(theory)</td>
<td>1.6 meV</td>
<td>He(2³S)+He(2¹S)</td>
<td>318</td>
<td>0.09</td>
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<tr>
<td></td>
<td>61 meV</td>
<td>He(2³S)+He(2¹S)</td>
<td>92</td>
<td>0.023</td>
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<tr>
<td>Müller et al. [16]</td>
<td>1.6 meV</td>
<td>He(2³S)+He(2¹S)</td>
<td>-</td>
<td>0.07(3)</td>
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<tr>
<td>(experiment)</td>
<td>61 meV</td>
<td>He(2³S)+He(2¹S)</td>
<td>-</td>
<td>0.03(3)</td>
</tr>
</tbody>
</table>

† The authors in [7] estimate that $\sigma_{sp}/\sigma_{ss} \approx 100$

†† Estimated from the $1^1\Sigma_g^+\rightarrow 2^1\Sigma_g^+, 2^1\Sigma_u^+$ electron spectrum in [16].

Contributions from $1^1\Sigma_u^+$ states are not included.

Table 6.3 Ionization cross sections in the triplet He⁺+He⁺ system for ground state and optical collisions at low collision energies. The numbers in the parenthesis in the last column refer to the uncertainty in the last digit.

used the calculations of the electron spectra (see section 6.2.3). By changing the interatomic potentials in the calculations we have found that the AI fraction in the process of PI is mainly determined by the structure of the interatomic potential near the inner turning point. The reason for this is that the AI process is only possible at short internuclear distances where transitions to the bound states of the $2\Sigma_u^+$ ionic potential can occur (see chapter 2). More specific, the AI fraction depends on the slope $dV/dR$ near the inner turning point of the interaction potential. In a classical picture, the slope at the inner turning point determines the interaction time of atoms at their closest approach and for smaller slopes the time is longer. Hence the probability for AI is larger. Due to the different electronic coupling at short internuclear distances, the slope of the S+P interaction potential is substantially smaller than the slope of the S+S potential (see figure 6.7) which is the direct cause that we find a
large fraction of AI in S+P collisions.

6.5 Conclusions

In this chapter we have studied the Penning Ionization process in the collision systems He(23S)+He(23S) and He(23S)+He(23P) at mK temperatures. We have used a MOT containing He(23S) atoms and have applied ion mass spectrometry to study trap loss by reactive collisions. We have measured the PI and AI cross section in He(23S)+He(23S) collisions and He(23S)+He(23P) collisions. The Penning Ionization cross section in He(23S)+He(23P) collisions is 7 times larger than for collisions without light. The AI fraction for He(23S)+He(23S) collisions is 0.03 and in He(23S)+He(23P) collisions 0.16. The change in the AI fraction in the presence of resonant light is explained by the fact that ionization in S+P collisions proceeds directly through the S+P channel.
Chapter 7

Dynamical Spectroscopy of Transient He$_2^*$ Molecules

7.1 Introduction

In the previous chapter we have studied the Penning Ionization (PI) process of two metastable helium atoms in cold collisions. The reactions we have studied were induced by He(2$^3$S)+He(2$^3$S) collisions in the presence of a near resonant light field. By tuning the laser light on resonance with the $2^3S \rightarrow 2^3P_2$ transition we have prepared He(2$^3$S)+He(2$^3$P) collision systems by exciting one He(2$^3$S) atom in the entrance channel to the He(2$^3$P) state. In this chapter the laser light will be detuned below resonance such that He(2$^3$S) pairs can be excited in the molecular regime (see chapter 2). The interatomic distance where He(2$^3$S)+He(2$^3$S) molecules are resonantly excited to the He(2$^3$S)+He(2$^3$P) manifold is called the ‘Condon’ point $R_c$. It is defined more precisely as the point where the potential energy $V(R) = C_3/R^3$ of the He(2$^3$S)+He(2$^3$P) complex is equal to the laser detuning $\delta$:

$$R_c = (C_3/h\delta)^{1/3}.$$  \hfill (7.1)

By using light detuned below atomic resonance the formation of molecules can be studied. After the complex has been excited to the He(2$^3$S)+He(2$^3$P) state, it is very likely to ionize in the PI reaction (see chapter 6) given by
Dynamical Spectroscopy of Transient $\text{He}_2$ Molecules

\[
\begin{align*}
\text{He}(2^3\text{S}) + \text{He}(2^3\text{S}) + h\omega & \rightarrow \left[\text{He}(2^3\text{S}) + \text{He}(2^3\text{P})\right] \\
& \rightarrow \begin{cases} 
\text{He}^+ + \text{He}(1^1\text{S}) + e^- \\
\text{He}^+_2 + e^- 
\end{cases}
\end{align*}
\] 

(7.2)

Thus by photon absorption of the molecular complex and the subsequent ionization, the formation of ‘transient’ $\text{He}(2^3\text{S})+\text{He}(2^3\text{S})$ molecules can be studied.

7.2 Experiments

We have used a MOT containing $\text{He}(2^3\text{S})$ atoms at a temperature of 1.1 mK (see chapter 4). The mean velocity of $\text{He}(2^3\text{S})$ atoms in the trap is about 2 m/s. The time is therefore sufficiently long to make an excitation, when two $\text{He}(2^3\text{S})$ atoms approach. We use the $2^3\text{S} \rightarrow 2^3\text{P}_2$ optical transition to excite the $\text{He}(2^3\text{S})+\text{He}(2^3\text{S})$ molecules to the $\text{He}(2^3\text{S})+\text{He}(2^3\text{P})$ state. If the excitations are to the bound states of the $\text{He}(2^3\text{S})+\text{He}(2^3\text{P})$ molecule, the probability for ionization is about 90%. We detect the ion production as a function of the laser detuning using the ion mass spectrometer fixed at 8 amu to detect $\text{He}_2^+$. We probe the molecular ions produced in the Associative Ionization (AI) reaction since this reaction has the highest ion yield in the experiment (chapter 4). Similar as in the experiments of chapter 6 we have used the trapping laser as a ‘probe’ laser by modulating the trapping laser frequency in a short time interval such that the MOT is not destroyed.

The frequency cycle of the trapping laser was as follows (see figure 7.1a): In the first time interval from $t=0-20$ $\mu$s the trapping laser frequency is fixed to $-25$ MHz which is the detuning at which the MOT normally works. From $t=20-50$ $\mu$s the laser is shifted into resonance and linearly ramped to $-300$ MHz. From $t=50-60$ $\mu$s the frequency is fixed to $-300$ MHz and for $t=60-100$ $\mu$s the frequency is shifted back to $-25$ MHz. In the non-trapping time interval $t=20-60$ $\mu$s the expansion of the cold atomic sample by ballistic flight can be neglected. In figure 7.1b the ion yield is plotted for the time intervals discussed above. The measurement is integrated over $4 \times 10^6$ cycles. The time delay in the ion signal of about 5 $\mu$s after the laser is switched to resonance is due to the flight time of the ions from the MOT to the detector. In the time interval of 20–60 $\mu$s the ion production is acquired as a function of laser detuning. The slow recovery of the ion signal after the laser is instantaneously switched from $-300$ MHz to $-25$ MHz has also been observed in the experiments of chapter 6 and is still under investigation. We have collected several ion curves as shown in figure 7.1 using different detunings and time intervals for modulation in the range of 20–40 $\mu$s. We have transformed the data in the time
interval from 20 µs to 50 µs in each measurement to obtain the ion production as a function of the detuning. In figure 7.2 we present our results.

7.3 Analysis

7.3.1 Initial considerations

Qualitatively, the result is explained as follows. At a given detuning, in a first step, pairs of atoms are excited that happen to be at distances around the Condon distance \( R_c = (C_3/\hbar\delta)^{1/3} \), where they can resonantly absorb the radiation. The excited quasi-molecule is then in an excited S+P state. In this state it “lives”, on the average, for a period given by the molecular lifetime for spontaneous emission. These lifetimes can be derived from the atomic lifetime. While being in the excited state, the quasi-molecule will dynamically evolve depending on the initial conditions at the instant of its formation. These conditions may be characterized by the angular momentum, by the radial velocity, and by the distance. At small transition distances, i.e. large detunings, the radial velocity will be too small for the complex to break up.

All excited molecules will live until they either spontaneous decay by photon emission, or approach to small distances, where Penning ionization occurs with nearly unit probability. At large distances, i.e. small detunings, there are three possible fates of the excited complex (see figure 7.3):

1. The complex breaks up, since the angular momentum in the complex is too large to overcome the potential barrier.

2. The complex overcomes the potential barrier and may approach to small distances in the excited state, leading to Penning ionization.

3. Even if the complex overcomes the barrier, the complex may spontaneously decay by photon emission, leading to escape in the weak S+S potential.

Decreasing the detuning from large detunings, at first the number of pairs that can absorb the radiation increases, because the number of pairs having distances around \( R_c \) increases proportional to the square of its value. As the detuning is further decreased, more and more excited pairs will either break up or decay spontaneously, leading to a maximum rate at medium detunings, and finally to a decrease of the rate. At very small detunings, of the order of the natural linewidth and below, there is a transition from the “molecular situation” to the “atomic situation”. At resonance, i.e. in the “atomic situation”, there is no relation between atom-atom
distances and the probability of excitation, so that S+P collisions will be accidental, leading to an ion rate that depends critically on the lifetime of the P-state. In the limit of infinite lifetime, and for an average probability for population of the P-state, the ion rate is given by the rate for “close S+P collision”. For a finite lifetime, as in the present case, the ion rate will be lower than this limiting value. We see that the features of the measured data points in figure 7.2 are nicely explained by the qualitative considerations above.

As compared to the other systems for which similar experimental data are available (see table 2.1), a special feature of the present data is the large ion rate at zero detuning. In case of the other systems, this rate is zero. The reason for this difference is the combined effect of the rather long lifetime of the He(2P) state, and the small mass of the helium atoms, leading to large distance the excited He(2P) state atoms can cover during their lifetime. The large ion rate at zero detuning of our system is very interesting in connection with theoretical models designed to describe optical collisions, because it implies that the data cover the whole range, from the pure molecular to the pure atomic situation, allowing one to test the models in the interesting transition region.

### 7.3.2 The Semi-Classical Model

The two models that have been developed up until present for the description of optical collisions have already been introduced in chapter 2. The GP-model, designed to describe the molecular regime, clearly cannot be used for our system. The JV-model, on the other hand, attempts to describe also the transition region. Unfortunately, however, the JV-model is formulated for the case of low excitation probability of the atomic pairs in the whole region of detunings, whereas in our experiment the S+P transition is strongly saturated for small detunings.

Several authors have extended the GP-model to go beyond these restrictions. In the case of saturation one has to deal with the molecule-light interaction in the regime, where the velocity in the two coupled channels are not identical. Several methods have been proposed to accommodate this problem: optical Bloch equations (OBE) on one reference trajectory [53], complex potential method [54], Landau-Zener curve crossing [21,51,52], and Monte-Carlo simulations (MCWP) [55, 56]. Especially the MCWP-method yields correct results under various conditions, however, the method is very demanding in computer time and space. Therefore results for limited number of situations can be obtained and the model is not as transparent as for instance the GP-model.

In order to regain the transparency of the GP-model and go beyond its restrictions, we have developed our own model, which is based on a semi-classical treat-
7.3 Analysis

ment of the problem. We use the dynamical Landau-Zener level crossing model to describe the S+P excitation at a certain detuning (see e.g. [51]). At the Condon distance corresponding to this detuning $\delta$, the “dressed” S+S potential crosses the S+P potential, leading to the usual curve crossing picture. The excitation probability for an S+S atomic pair whose distance happens to vary across the Condon point with a velocity $v$, is given by [51]

$$P_{ex} = 1 - \exp(-\pi \Lambda),$$

(7.3)

where

$$\Lambda = \frac{2\hbar \Omega^2}{\alpha v},$$

(7.4)

is the Landau-Zener parameter. Here $\alpha$ is the gradient of the potential difference between the ground and excited state at $R_c$. Since the ground state is practically flat at the Condon point we have, given the excited potential $V(R) = -C_3/R^3$,

$$\alpha = \left| \frac{dV}{dR} \right|_{R_c} = \frac{3C_3}{R_c^4}.$$  

(7.5)

The electronic coupling strength is given in terms of the Rabi-frequency for the atomic transition. The Rabi-frequency $\Omega$ in turn is related to the photon flux ($\phi$) via the relation [57]

$$\Omega = \left( \frac{\lambda^2 \Gamma \phi}{8\pi} \right).$$

(7.6)

The photon flux we can relate to the experimentally known saturation parameter defined by $s_0 = \phi/\phi_0$, with the saturation flux $\phi_0$, by definition given by,

$$\phi_0 \frac{\lambda^2}{\pi} = \Gamma.$$  

(7.7)

In this way we obtain for the Landau-Zener parameter $\Lambda$,

$$\Lambda = \frac{2\hbar s_0 \Gamma^2}{\alpha v}.$$  

(7.8)

The radial relative velocity $v_r$ of the S+S pair may be expressed by the average value of the relative velocity, $v_0$, and the angle $\theta$ between by the velocity vector and the internuclear axis. The excitation probability thus becomes a function of three variables: $\delta$, $v_0$ and $\theta$. We are interested in the excitation rate, which is given by the
flux of systems crossing the Condon-distance times the probability for excitation, i.e.

\[ R_{\text{ex}} = 4\pi R_c^2 v_r \left(1 - e^{-\pi/\Lambda} \right). \] (7.9)

It is interesting to note that the excitation rate becomes independent of \( \delta, \upsilon_0 \) and \( \theta \) in the limit of weak coupling. For large detunings, where the excited pairs cannot dissociate, and where no radiative decay occurs, the above expression becomes

\[ R = \frac{8\pi^2 s_0 C_3 \Gamma^2}{3\delta^2}, \] (7.10)

and is therefore already a valid description of the absolute ion rate.

In order to incorporate the possibility for decay and for breaking up, one has to follow the evolution of the excited system for the initial conditions given by specific values of \( \delta, \upsilon_0 \) and \( \theta \). This we do by numerical integration of the classical trajectory, from which we obtain the time span \( \tau_0 \) necessary for the system to approach to small distances, and a capture probability function \( w \), which can have the values 0 and 1, depending on whether the system dissociates or not, respectively. From the traveling time \( \tau_0 \) we construct the survival factor \( S(\tau_0) = \exp(-\Gamma\tau_0) \). The detailed contribution to the ion rate from an excitation is specified by \( \delta, \upsilon_0 \) and \( \theta \) and thus becomes

\[ R_{\text{ion}} = R_{\text{ex}} S(\tau_0) w. \] (7.11)

which is our central result. By integration over theta, we finally obtain the ion rate corresponding to a certain detuning \( \delta \)

\[ \tilde{R}_{\text{ion}} = \int_{-\pi/2}^{\pi/2} R_{\text{ion}} \sin(\theta) d\theta / 2. \] (7.12)

In order to test the expression for the limiting case of zero detuning and infinite lifetime, we calculated the ion rate for a \( C_3/R^3 \) potential for the case of saturation near zero detuning (s_0=50). We found the ion rate approaches the close collision rate given by \( \sigma_{CC} = \pi b_{\text{max}}^2 \) (see chapter 2). We emphasize that our model predicts absolute rates, has the right behavior for zero detuning, and describes the subtle transition from molecular to atomic behavior as a function of detuning.

In order to carry out a calculation that can be compared to the experimental data of figure 7.2, one has to take into account the following complications:

1. There are several potential curves that correlate asymptotically to the atomic S+P states.
2. These potentials have different $C_3$ coefficients and correspondingly different molecular lifetimes.

3. At small distances the potentials correlate to three total molecular spin states leading to a certain reduction of the ionization contribution because of the Wigner spin rule.

4. The relevant distances for small detunings are already of the order of the wavelength of the radiation, so that $R$-dependent, retarded, values for $C_3$ coefficients and lifetimes have to be used.

5. Since only even or only uneven rotational states are allowed for our boson system, the rate has to be multiplied by a factor of $1/2$.

6. One has to average over the Maxwell-Boltzmann distribution of velocities.

The final ion rate is obtained as a sum over eight contributions from the attractive S+P potentials, each of the contributions being obtained using Eq. 7.12. The result is shown as the solid line in figure 7.2, where one parameter has been adapted to bring the calculation on the same absolute scale as the experiment. The agreement is very satisfying.

7.4 Conclusions

We have studied the formation of He(2$^3$S)+He(2$^3$S) molecules by optical excitation on the 2$^3$S→2$^3$P$^2$ transition followed by ionization. By rapidly scanning the trapping laser frequency we have been able to study He(2$^3$S)+He(2$^3$S) collisions in the cold atomic sample. We have studied excitations for the laser frequencies from 300 MHz below resonance to resonance such that the Condon point is shifted from distances, where the complex is in the molecular regime, to distances, where the complex is in the atomic regime.

The ionization yield for large detunings (molecular regime) is well explained by the standard GP model. At smaller detunings (atomic regime), however, the model can not applied anymore because of the dynamical character (atomic regime) of collisions in the He(2$^3$S)+He(2$^3$S) system at large interatomic distances. We describe a semi-classical model for the He system that takes into account both the molecular aspects as well as the collisional aspects of the system. The model is based on a two level Landau-Zener system in which excitations can take place between S and P states by laser light. Both the lineshape of the model and the absolute ion production are in good agreement with the experimental results.
Figure 7.1  Spectroscopy of He(2\(^3\)S)+He(2\(^3\)S) molecules. (a) The trapping laser frequency is periodically scanned from zero detuning to \(-300\) MHz within a time period of 40 \(\mu s\), while the atoms in the MOT are nearly unperturbed. (b) He\(^+\)\(_2\) yield integrated over \(4 \times 10^6\) cycles.
7.4 Conclusions

Figure 7.2 Ion production in a He(2\(^3\)S) MOT at 1 mK as a function of the laser detuning relative to the \(2^3S \rightarrow 2^3P_2\) transition. The background ion level is 0.1 Hz and is not visible on this scale. At laser detunings below \(-30\) MHz ionization is due to molecular excitation of He(2\(^3\)S)+He(2\(^3\)S) pairs. The solid line is the lineshape of the model calculation discussed in section 7.3.2.

Figure 7.3 Two possible fates of the complex. For small angles the angular momentum is small enough for the complex to overcome the rotational barrier (\(w=1\)). For larger angles the complex does not overcome the barrier and the complex breaks up (\(w=0\)).
References


References


Summary

Low energy collisions of two neutral atoms can be studied using modern atomic traps such as the magneto optical trap (MOT), in which a large number of atoms can be simultaneously cooled and trapped by laser light. The interesting feature in low energy collisions is that weak structures in the interatomic potentials, appearing at very large distances, affect the dynamics of collisions dramatically. In this respect the system of two triplet helium atoms is a special case. Because of the small mass of the helium atoms their collisions at low energies are dominated by interactions at distances in the order of an optical wavelength, i.e. at distances of about 1000 Å. So far, atomic interactions at these long distances could not be studied. This thesis presents a novel study of the He(2^3S)+He(2^3S) system and gives insight in the interaction processes that occur at these large distances.

For an experimental study on low energy collisions a cold and dense sample of He(2^3S) atoms is needed. Therefore we have built a MOT in which He(2^3S) atoms are cooled and confined. We have found that cooling and trapping of He(2^3S) atoms is only efficient if the trap is operated at considerable laser detunings and intensities. The large laser detuning is needed to obtain a trap with a sufficiently large depth so atoms with relative high initial energy can be loaded. By operating the MOT at large laser detunings the “laser cooling” effect is compromised due to the small friction force but it has turned out that the cooling effect is restored if the laser intensity is increased. When we operate our He(2^3S) MOT in this regime, $4 \times 10^5$ atoms are confined in a volume of about $4 \times 10^{-3}$ cm$^3$ at a temperature of 1 mK.

The atomic density in the trap is determined by the loading rate of atoms into the trap and the loss rate of atoms by collisions. In principle there are two distinct loss processes: the first is the loss of a single trapped atom by a collision with a background atom, which can be either elastic or inelastic (reactive) and the second process is the loss of two trapped atoms in a reactive low energy collision. By analyzing the mass spectrum of ions produced in the MOT, we have found that the most important inelastic trap loss process by background gas atoms is due the ther-
mal Penning Ionization reaction $\text{He}(2^3S)+\text{H}_2\text{O} \rightarrow \text{H}_2\text{O}^++\text{He}(1^1S)+e^-$. This rather unexpected result is explained by the large ionization cross section of the process of about $100 \text{ Å}^2$. The trap loss due to elastic scattering of background gas atoms has been studied by lifetime measurements of the $\text{He}(2^3S)$ sample in environments of different rare gas atoms and diatomic molecules. By using the atomic trap the elastic scattering cross sections for several systems $\text{X}+\text{He}(2^3S)$ have been measured on an absolute scale. In contrast to traditional atomic beam experiments, the absolute scattering cross sections, measured by using a MOT, are accurate to a very high degree since only the independent variables time and density are needed to determine the cross section.

Cold collisions of two triplet metastable helium atoms have been studied at mK temperatures by observing ‘trap loss’ of two trapped atoms in the Penning Ionization (PI) reaction

$$\text{He}(2^3S) + \text{He}(2^3S) \rightarrow \begin{cases} \text{He}^+ + \text{He}(1^1S) + e^- \\ \text{He}_2^+ + e^- \end{cases},$$

where the second exit channel is called Associative Ionization (AI). One of the results we have found is that if the $\text{He}(2^3S)+\text{He}(2^3S)$ collision takes place in a laser light field in resonance with the $2^3S \rightarrow 2^3P_2$ optical transition, the AI reaction is favored. This is due to the ionization of the $\text{He}(2^3S)+\text{He}(2^3P)$ complex, for which the ionization proceeds directly through the S+P channel.

If the $\text{He}(2^3S)+\text{He}(2^3S)$ complex is excited by laser light detuned below resonance the molecular properties of the systems can be studied. In this thesis we have studied the formation of $\text{He}(2^3S)+\text{He}(2^3S)$ molecules by absorption of nearly resonant light. The $\text{He}(2^3S)+\text{He}(2^3P)$ molecules that are formed will ionize in the PI reaction and the ions of this process have been used to probe the formation of $\text{He}(2^3S)+\text{He}(2^3S)$ molecules. The absorption line we have obtained in this way has both a ‘molecular’ and an ‘atomic’ component. To understand the lineshape and the dynamics of the $\text{He}(2^3S)+\text{He}(2^3S)$ collision in the intermediate regime of molecular physics and atomic collision physics, we have modeled the $\text{He}(2^3S)+\text{He}(2^3S)$ system. Our semi-classical model describes the optical excitation of the $\text{He}(2^3S)+\text{He}(2^3S)$ complex as a dynamical process and fits well to the experimental lineshape.
Samenvatting

Botsingen van atomen bij lage energieën kunnen worden bestudeerd in atomaire valen zoals de magneto optische val (MOT). Een MOT heeft als eigenschap dat grote hoeveelheden atomen kunnen worden opgesloten in een laser lichtveld, waarbij de atomen bovendien door het laserlicht worden afgekoeld tot temperaturen in de orde van 1 mK. In laag-energetische botsingen, ook wel koude botsingen genoemd, wordt de dynamica sterk bepaald door de atomaire interacties op grote afstanden. Dit geldt in hoge mate voor het systeem bestaande uit twee triplet helium atomen, waar de botsingsdynamica wordt bepaald door atomaire wisselwerking op afstanden in de orde van de golflengte van licht. Interacties op zulke grote atomaire afstanden zijn nog niet eerder bestudeerd en zijn van belang omdat deze plaatsvinden in het intermediaire gebied van de molecul- en botsingsfysica. In dit proefschrift worden koude botsingen in het He*+He* systeem bestudeerd om meer inzicht te krijgen in de mechanismen van interacties op grote afstanden.

Om koude botsingen van He(23S) atomen experimenteel te bestuderen is een MOT geconstrueerd waarin een koud ‘sample’ van He(23S) atomen zit opgesloten. Het blijkt dat de dichtheid van het sample atomen in de MOT een maximaal is bij grote verstemmingen van de laserfrequentie en bij een hoog laservermogen. De grote frequentieverstelling is nodig om atomen met een relatief hoge snelheid in te kunnen vangen. Het grote laservermogen blijkt nodig om ervoor te zorgen dat het effect van ‘laserkoeling’ onder deze omstandigheden optimaal is. Als de MOT onder bovengenoemde omstandigheden werkt zitten er $4 \times 10^5$ atomen in de val bij een temperatuur van 1 mK. Het sample heeft een volume van $4 \times 10^{-3} \text{ cm}^3$. Het verliesproces van atomen uit de MOT wordt bij lage dichtheden bepaald door het eerste orde proces van elastische en inelastische botsingen met achtergrondgas atomen. Door de ionen te bestuderen die in de MOT worden geproduceerd in Penning-Ionisatie reacties, blijkt het belangrijkste inelastische verliesproces door de He(23S)+H2O→H2O^++He(23S)+e^- reactie te worden bepaald. Dit onverwachte resultaat wordt verklaard door de grote werkzame doorsnede van dit proces, die namelijk ongeveer 100 Å² is.
Het verliesproces door elastische botsingen is bestudeerd door te kijken hoe lang ‘leeglopen’ van de MOT duurt als het laadproces van atomen wordt onderbroken in een omgeving van bekende achtergrondgas atomen. Door de gunstige botsingsgeometrie in een MOT en het specifieke meetproces kunnen werkzame doorsneden voor elastische verstrooiing op een absolute schaal worden gemeten. Het gebruik van een MOT is een goede aanvulling op de traditionele atomaire bundlexperimenten waar de noodzakelijke bepaling van detector efficiënties en atoomaire bundeloverlap aanleiding geven tot systematische fouten in absolute metingen.

Koude botsingen in het He(23S)+He(23S) systeem zijn bestudeerd aan de hand van de volgende Penning-Ionisatie (PI) reactie:

\[
\text{He}^{(23S)} + \text{He}^{(23S)} \rightarrow \begin{cases} 
\text{He}^+ + \text{He}^{(1S)} + e^- \\
\text{He}^+_2 + e^- 
\end{cases}
\]

Het deelproces, waarin moleculaire ionen worden gevormd, wordt Associative Ionisatie (AI) genoemd. De He(23S)+He(23S) reacties noemen we ‘grondtoestand botsingen’. Ook mogelijk zijn He(23S)+He(23P) reacties, ‘optische botsingen’ genaamd, waarbij het He(23P) atoom in het ingangskanaal door laserlicht is aangeslagen. Het blijkt ondermeer dat de AI fractie in He(23S)+He(23P) botsingen groter is dan in He(23S)+He(23S) botsingen. De reden is, dat het PI proces in een He(23S)+He(23P) botsing direct in het aangeslagen kanaal plaatsvindt.

Als He(23S)+He(23S) botsingen plaatsvinden in een lichtveld dat beneden atoomaire resonantie is verstemd, dan kan het moleculaire complex worden aangeslagen. We hebben met behulp van laserexcitatie de moleculaire dynamica van het He(23S)+He(23S) complex bestudeerd. Het principe is een overgang te maken met het laserlicht naar de He(23S)+He(23P) toestand, die vervolgens ioniseert. Door dit principe toe te passen, hebben we de moleculaire absorptielijn van het He(23S)+He(23S) molecuul gemeten in de buurt van de 23S → 23P2 overgang. Nabij resonantie heeft het He(23S)+He(23S) complex een atomair karakter en bij grote frequentieverstemmingen een moleculair karakter. Het proces van excitatie en ionisatie is een dynamisch proces. Om de dynamica en de moleculaire vorming van het systeem te begrijpen hebben we een semi-klassiek model ontwikkeld. Dit model is gebaseerd op een Landau-Zener overgang, waarin de absorptie van het licht in het complex dynamisch wordt beschreven. De lijnform, die met het model kan worden berekend, komt goed overeen met de gemeten absorptielijn.
Nawoord

Met een zak vol geld en veel goede ideeën ging vier jaar geleden het ‘Gemini project’ van start. De eerste ‘Utrechtse’ resultaten vormen de basis van dit proefschrift. Het idee van het project is door de toepassing van ‘laser koeling’ atomen af te koelen om atomaire botsingen te bestuderen bij extreem lage energieën. Op het gebied van botsingsfysica en reactie-onderzoek is binnen de vakgroep Atoom- en Grenslaagfysica al jarenlang ervaring, vooral met natrium. Ook op het gebied van laserkoeling was de nodige expertise aanwezig door het werk van Peter Molenaar. Natuurlijk is een botsingssysteem met metastabiel helium atomen veel interessanter dan van twee natrium atomen. Het was dan ook mijn taak ‘iets’ aan laserkoeling te doen van metastabiel helium. Uit technologisch oogpunt gezien is dat niet eenvoudig maar al snel kon na de ontwikkeling van een nieuwe metastabiele bron naar een wat ‘ruimere’ experimenteerkamer worden verhuisd waar uiteindelijk een heuse bundellijn is ontstaan. Tijdens de ontwikkeling konden profiteren van de nieuwste generatie vacuüm-pompen die heel efficiënt helium kunnen afpompen en van een nieuw type diodelaser die ons uiteindelijk een hoop ongemakken heeft bespaard. Zulke en andere gebeurtenissen maakte de ontwikkeling van het nieuwe experiment een stuk eenvoudiger.

Het bouwen van een atomaire bundellijn doe en kun je natuurlijk nooit alleen; een klein leger van mensen is hiervoor ingeschakeld. Allereerst zijn dat Arend Niehaus en Peter van der Straten die ik graag wil bedanken voor het leuke onderzoek, de ruime mogelijkheden die ze hebben geboden, voor alle vriendelijke hulp van buitenaf en voor de samenwerking de afgelopen tijd. Speciaal wil ik hen beiden ook bedanken voor wat ze niet hebben gedaan. In het bijzonder wil ik hierbij noemen dat zij niet moeilijk deden over de muur die uit het lab verwijderd moest worden. Graag wil ik Jan Thomsen bedanken voor de enorme versnelling die hij in het botsingsonderzoek heeft bracht. We hebben daardoor het afgelopen jaar al kennis kunnen maken met echt nieuwe fysica. Van grote waarde was natuurlijk Peter Molenaar die als eerste ‘het kunstje van laserkoeling’ binnen onze vakgroep demonstreerde. Peter, het was erg handig en ook leuk om jouw als begeleider en
later als collega en kamergenoot te hebben.

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Een nieuw experiment is nooit eenvoudig, zeker niet als je als student binnenkomt. Gerlene Oudenaarden, Ivo van Vulpen, Mike van der Poel, Jeroen Bongaerts, Femius Koenderinck, Michiel van Rijnbach en Jeroen van Driel hebben allemaal als student een belangrijke bijdrage geleverd aan het onderzoek. In vier jaar tijd maak je kennis met veel collega onderzoekers: Tycho Sonnemans, Arnaud Kok, Geraldine Woestenenk, Roel Knops, Bob van Someren, Arjen Amelink, Ton-nies Nuver, Michael Benndorf, Wilco Planje, Morten Lunsgaard, Jan-Jette Blangé, Geert Spierings, Peter Molenaar, Kees Uiterwaal, Marc Piekema en Jasper Boessenkool wil ik allemaal bedanken voor het uit(lenen) van apparatuur en ideeën en voor de ontspannen werksfeer.
Zomerscholen en Conferenties

- 15th International Conference on Atomic Physics, Amsterdam (1996)

Publicaties

- ‘Ejection of Trapped Atoms by Elastic Scattering at Thermal Energies’, in voorbereiding, als hoofdstuk 5
- ‘Penning Ionization of Metastable Helium at mK temperatures’, in voorbereiding, als hoofdstuk 6