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### Collisions between ultracold metastable He atoms

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#### Abstract

We present experimental data on collisions between excited He-atoms occurring in a magneto-optical trap (MOT) at a temperature of 1.1 mK. He( $2^3$ S)-atoms produced in a discharge are pre-cooled and trapped using the He( $2^3$ S)-He( $2^3$ P<sub>2</sub>) transition for laser manipulation. Measurements of the Penning ionization rate as a function of the MOT-laser frequency are presented and theoretically analyzed. The analysis, based on a model which is presented in detail for the first time, leads to a good understanding of the complex nature of optical collisions. Further, first and preliminary measurements of the kinetic energy distributions of He<sup>+</sup><sub>2</sub> and He<sup>+</sup>-ions formed by Penning ionization in optical collisions are presented. © 1999 Elsevier Science B.V. All rights reserved.

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#### 1. Introduction

During recent years, rapid advances in laboratory techniques for laser cooling and trapping of neutral atoms have made it possible to prepare atomic ensembles at "ultracold" temperatures, of the order of 1 mK and below [see e.g. 1]. Neutral atoms that represent an essential "two-level system", with a long lived lower state and a resonant higher state, can be "laser manipulated". The most prominent atoms that fulfill this requirement are the alkalis for which the two levels are, respectively, the ground state and the first excited P-state. But also, the metastable rare gas atoms can be manipulated, because the lifetimes of these atoms in metastable states is long compared to the time necessary to manipulate the atoms, and because there exists a resonant state that forms a closed two-level system with the metastable state. In the case of He atoms, the He\*(1s2s)<sup>3</sup>S state provides the lower level, and the He\*(1s2p)<sup>3</sup>P state the resonant, higher level.

Collisions of the manipulated atoms can be studied either in a "trap" or in "beam-trap" experiments. There are several general aspects that make such studies at ultracold temperatures especially interesting. To be concrete, let us consider the collision of two He-atoms at a temperature of 1 mK, which can easily be reached in a magnetooptical trap (MOT). The average relative kinetic

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energy is then about  $E_{\rm k} = 100$  neV. At these energies, the collisions are dominated by the long range forces. The typical interaction distance, which we obtain by equating the kinetic energy to the interaction potential, is  $R_i = (C_6/E_k)^{(1/6)} = 52$  Å for the case of two  $He(2^{3}S)$ -atoms. If we compare this distance with the deBroglie wave length of the collision system,  $\lambda_{deB} = h/(2^*E_k\mu)^{1/2} = 640$  Å, we notice that,  $R_i \ll \lambda_{deB}$  and therefore only s-wave scattering occurs. If, on the other hand, an atom is in the resonance state  $He(2^{3}P)$ , it interacts with a collision partner in the  $He(2^{3}S)$ -state via the dipole-dipole interaction, which has a much larger range, characterized by  $R_i = (C_3/E_k)^{(1/3)} = 747$  Å. To travel this large distance, it takes the system  $t_{\rm c} = 25$  ns. One of the most interesting aspects of the "ultracold collision" regime is that during these long collision times it is possible to influence the collisions by laser radiation. For instance, a pair of  $He(2^{3}S)$ -atoms at a large distance and a relative velocity that would not lead to a collision, can be excited by absorption of a near resonant photon into a He(2<sup>3</sup>S)-He(2<sup>3</sup>P) pair on an attractive potential curve that induces a close collision. Depending on whether or not during the approach spontaneous re-emission of the photon occurs, the collision occurs in the ground-or the excited state. Another possibility is that, a pair of  $He(2^{3}S)$ -atoms in a kinetic state that would lead to a collision is excited into a repulsive  $He(2^{3}S)$ - $He(2^{3}P)$  state so that the collision is avoided. Collisions that are manipulated in the described way are usually called "optical collisions" [2,3]. The distance region where collision systems are excited, can be controlled by the chosen detuning  $(\delta)$  of the laser radiation from the atomic resonance: the excitation will be maximal at the so called Condon distance  $(R_c)$ , where the transition can occur resonantly, i.e., where the interaction energy is compensated by the detuning. At large distances where only the dipole-dipole interaction in the excited state is significant the Condon distance is given by  $R_c = (C_3/\delta)^{1/3}$ . At negative detuning the excitation will lead to population of an attractive state, and at positive detuning a repulsive state will be populated. With increasing detuning  $(\delta)$ , the distance decreases where resonant excitation can occur.

The experimental method to study collision systems by optical collisions has been mainly restricted to systems with a *stable* lower state [3,4]. In that case, the method may be termed a "dynamical spectroscopy of transient molecules", yielding unique information especially on details of longrange interactions.

Collision systems of atoms with a *metastable* lower level differ in one important characteristic from those with a *stable* lower level: they decay at short distances spontaneously by emission of an electron, i.e., by the so called process of Penning ionization (PI) [5]. In the case of two colliding  $He(2^{3}S)$  atoms, this process may be indicated schematically as:

$$\begin{aligned} \text{He}^{*}(2^{3}\text{S}) + \text{He}^{*}(2^{3}\text{S}) &\to \text{He}^{+}(1^{2}\text{S}) + \text{He}(1^{1}\text{S}) \\ &+ e^{-}(\varepsilon) \to \text{He}_{2}^{+} + e^{-}(\varepsilon') \quad (1) \end{aligned}$$

This process has been studied experimentally at thermal energies [6], and also the general theory of PI is rather well established [7]. Based on this theory, one may safely assume that PI of a He(2<sup>3</sup>S)–He(2<sup>3</sup>P) collision system occurs in a very similar way. For the present context it is sufficient to note that for "close collisions" the probability for Penning ionization is practically 100%, for molecular states of the  ${}^{3}S{}-{}^{3}S$  system as well as for those of the  ${}^{3}S{}-{}^{3}P$  system. The only exceptions are the molecular states with total spin quantum number S = 2, for which PI violates the total spin conservation rule. For collision systems in these states the PI probability is predicted to be suppressed by at least five orders of magnitude [8].

The situation for optical collisions in an ensemble of metastable atoms is schematically shown in Fig. 1. The occurrence of Penning ionization for close collisions, together with the possibility to manipulate collisions by laser radiation at large distances opens up a number of interesting possibilities for experimental studies. First of all, the excitation of collision systems at large distances can easily be monitored by just measuring the rate of ions formed, so that, by measuring the ion rate as a function of the laser detuning, one obtains details on optical collisions for an enormous distance range. Especially for the He-system, where the lifetime for spontaneous emission is comparatively



Fig. 1. Potential curve diagram that demonstrates the situation of optical collisions in the case of two metastable He(2<sup>3</sup>S)-atoms exposed to radiation negatively detuned by ( $\delta$ ) from the He(2<sup>3</sup>S)-He(2<sup>3</sup>P) transition frequency.

long with ca. 100 ns, this range extends up to distances where retardation effects become important. Secondly, the Penning ionization process can be studied in more detail, and under more selective experimental conditions than in "normal collisions". For instance, the number of participating partial waves can be selected by changing the laser detuning, and the occurrence of PI from quintet (S=2) molecular states can be investigated by spin polarization of the target atoms. Further interesting possibilities arise, if the ions formed in PI are mass-analyzed, and the emitted electrons or ions are energy analyzed. To indicate these possibilities we give below a brief outline of the process of PI.

The energy of the ejected electron ( $\varepsilon$ ) is related by energy conservation to the change of electronicand relative kinetic energy in the process. The latter change is given by the kinetic energy in the final state  $E_k^f$ , because, for ultracold collisions, we may neglect the kinetic energy in the initial state. Calling ( $\Delta$ ) the change in electronic energy, we therefore have the following relation:

$$\varepsilon = \varDelta - E_{\mathbf{k}}^{\mathsf{t}}.\tag{2}$$

 $E_{\rm k}^{\rm f}$  can be positive or negative. In the latter case  ${\rm He}_2^+$  is formed in a state with binding energy  $(-E_{\rm k}^{\rm f})$ . The situation for PI, which is a "vertical transition" approximately conserving the instan-

taneous energy of relative motion, is shown schematically in Fig. 2. Ab initio potential curves for the initial and final states relevant for the  ${}^{3}S-{}^{3}S$ collision system are available [6], and allow one to calculate the expected electron- or kinetic-energy distributions for any collision energy. A comparison of measured and calculated electron spectra has been performed for the  ${}^{3}S-{}^{3}S$  system at thermal energies [6]. Although contributions from transitions between several initial- and final states were involved, and of the order of 50 partial waves participated in the collisions, the comparison resulted in a rather stringent test of the theory and of the potential curves.

In optical collisions it is possible to measure spectra for the  ${}^{3}S{}-{}^{3}S$  and  ${}^{3}S{}-{}^{3}P$  potential curves, and to vary the number of participating partial waves. This allows one get information on the unknown  ${}^{3}S{}-{}^{3}P$  potential curves and on the unknown PI-transition probabilities, and to access problems like the dynamic coupling between different curves.

In the present paper we present and discuss experimental data obtained for ultracold optical collisions in an ensemble of  $He(2^{3}S)$  atoms in a MOT. The main result consists of the dependence of the ion rate on the laser detuning. This result has been published earlier [9]. It is shown that this



Fig. 2. Potential curve diagram demonstrating the situation for Penning ionization. Especially it is shown how the kinetic energy in the final state can be positive, or negative, leading to  $He^+$ - or  $He_2^+$ -formation.

dependence can be described by a new semi-classical model, which we present for the first time in detail. We also present first preliminary results on kinetic energy distributions of ions formed in optical collisions.

#### 2. Experimental setup

The experimental set-up is shown in Fig. 3. Since it has been described elsewhere [10], we only give a brief outline here. He metastable atoms are produced in a discharge source cooled by liquid nitrogen [11]. The He(2<sup>3</sup>S) metastables are then slowed down over a distance of 3 m, using the technique of Zeeman-compensated slowing [12], and finally loaded into a magneto-optical trap (MOT). The laser radiation for both, slowing and trapping, is furnished by diode-lasers manufactured especially for the  $He(2^{3}S)$ - $He(2^{3}P)$  transition at 1.083 µm [13]. Since, the loading rate from the slowed beam is rather low, the MOT is operated in a regime, where the capture velocity is large. This is realized by using a large negative detuning  $\delta = -15\gamma$  of the MOT-laser from the atomic resonance (y: natural line-width of the atomic resonance), and a high intensity. We chose an intensity

of 8.3 mW/cm<sup>2</sup> (corresponding to a saturation parameter  $s_0 = 50$ ) for each of the six laser beams. Typical values for the trap are,  $10^5$  for the number of trapped He metastables, and  $2.5 \times 10^7$  atoms/ cm<sup>3</sup> for the density. The temperature of the atoms was measured using a time-of-flight technique: the MOT lasers are interrupted, and the arrival times of the released metastables are measured on a set of micro-channel plates. The measured distribution of arrival times was found to be consistent with a Maxwell-Boltzmann distribution of velocities corresponding to a temperature of 1.1 mK. Ions formed in the trap are extracted by an electric field and mass analyzed in a quadrupole mass filter. He<sup>+</sup>- and He<sup>+</sup><sub>2</sub>-ions that can only be formed in collisions of two excited He-atoms, are taken as the measure of intra-MOT collisions.

In case of the time-of-flight (TOF) measurements of the He-ions that will be presented, a different set-up for cooling and trapping was used, which allowed us to achieve a much higher trap density, around  $2 \times 10^8$  atoms/cm<sup>3</sup>. This was possible with a newly developed discharge source running in a liquid He-cryostat [14] which produced sufficiently slow metastables that could directly be loaded into the trap after slowing in the magnetic field of the MOT.



Fig. 3. Schematic drawing of the experimental set-up (see text).

## 3. Measurements of optical collisions for the He–He system

The following experimental conditions regarding the trapped atoms are important for the measurements. The MOT is operated in such a way, that the atoms are confined to a volume of ca. 4 mm<sup>3</sup> around the center of the "anti-Helmholz" magnetic field configuration, where the field is zero. The field, the trapped atoms experience leads to Zeeman shifts small compared to the detunings applied in the experiment, and may therefore be neglected. Further, the velocities of the trapped atoms are low, so that, after interrupting the MOT-laser, the "ballistic" expansion of the trapped ensemble of atoms is negligible during time spans of the order of milliseconds. And finally, the trap decay by intra-MOT collisions and by collisions with the residual gas has a measured time constant of the order of seconds, and may therefore also be neglected during time spans up to milliseconds. Under these conditions, it is possible to use the MOT-laser in two ways: (1) during repeated time intervals of the order of a few microseconds as a confining MOT-laser at a constant negative detuning of  $\delta = -15\gamma$  to maintain trap conditions, and (2), intermittently – also during intervals of a few microseconds - as a probe laser at various detunings ( $\delta$ ). By measuring the He ion rates during these "probe-laser intervals", optical collisions as a function of the detuning in the sense outlined above can be investigated. In the present experiment the probe-laser-interval is chosen to be 60 µs, and the MOT-laser interval 100 µs, and the detuning range extended from 0 to  $-200\gamma$ . The result is shown in Fig. 4 for the total ion rate. The rates are given on an absolute scale in atomic units (one atomic unit corresponds to  $6.15 \times 10^{-9}$  [cm<sup>3</sup>/ s]) and normalized to unit volume and unit density of the collision partners. In this way the curve in Fig. 4 represents the absolute rate constant as a function of the laser detuning. The absolute normalization is obtained from the theoretical calculations represented by the solid line through the experimental points. This theoretical calibration agrees with the experimental absolute calibration within the rather large limits of error of the latter one (an estimated factor of two). We note a dra-



Fig. 4. The total-ion rate as a function of the detuning ( $\delta$ ) of the MOT-laser from the He(2<sup>3</sup>S)–He(2<sup>3</sup>P) resonance frequency. The rate is given in "atomic units" (a.u.; 1 a.u. = 6.15×10<sup>-9</sup> cm<sup>3</sup>/s) and is normalized to unit reaction volume and unit density of the collision partners. The enhancement of the ion rate is due to optical collisions. The curve through the data points is the prediction of our theoretical model. The intensity is given on the scale of the theoretical rate constant for total ionization.

matic increase of the ion rate when the ensemble of He(2<sup>3</sup>S)-atoms is irradiated by laser light negatively detuned with respect to the atomic  $He(2^{3}S)$ -He(2<sup>3</sup>P) resonance. At detunings beyond ca.  $-300\gamma$ , the ion rate saturates at a low value and may be ascribed to collisions between two  $He(2^{3}S)$ atoms. And in the other extreme case of zero detuning, we may ascribe the ion-rate to collisions between atoms prepared in certain states at certain partial densities. For the high laser power used in the present experiment, the densities of  $He(2^{3}S)$ atoms and He(2<sup>3</sup>P)-atoms are 50% of the total density for both types of atoms. In the whole range of negative detunings between the extreme cases we have to consider optical collisions that cannot be described within the framework of normal collision theory.

#### 4. A semi-classical model for optical collisions

Several models for the description of optical collisions have been proposed and discussed [2,15]. None of them, however, can be directly applied to

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the present case. The main reasons are: (1) only the case of weak coupling by the laser radiation has been treated, while at our rather large laser power and large detuning range, we need a description that is valid in the whole range, from weak to strong interaction; (2) some special aspects arising because of the instability of the system due to Penning ionization have not been considered, and (3) the existing models have been formulated for just one attractive potential curve in the excited state, while in real systems usually several potential curves are relevant. In order to explain our data, we have therefore developed a new model which we will briefly outline below. It is mainly meant to allow us to understand the physics, behind the observed phenomena.

We use a semi-classical description, i.e., we use "trajectories" for the relative motion of the collision partners instead of a formulation in terms of partial waves. The inaccuracies induced are probably not severe, because the large observed rates prove that many partial waves are involved. Although a formulation in terms of partial waves is not more complicated, we choose the semi-classical description because it is more transparent.

The central ingredient of our model is the description of the excitation on a trajectory in terms of a simple Landau–Zener (LZ) crossing [16,17]. This is shown in Fig. 5. The crossing occurs between the potential curve belonging to the electronic "state" ( ${}^{3}S-{}^{3}S+hv$ ), and the potential curve belonging to the excited state ( ${}^{3}S-{}^{3}P$ ). These are the "diabatic" curves of the LZ-model, which corresponds to the omission of electronic coupling between them. Inclusion of the coupling ( $H_{12}$ ) in the electronic Hamiltonian leads to the adiabatic curves which avoid the crossing. According to the LZ-model, the probability for the system to follow the adiabatic path, i.e., to avoid the crossing, is given by [16,17]

$$P_{\rm exc} = 1 - \exp\left\{-2\pi |H_{12}|^2/(\alpha \times v)\right\},$$
 (3)

where ( $\alpha$ ) is the difference of the first derivatives of the potential curves with respect to the distance at the crossing point  $R_c$ , and v is the radial relative velocity at  $R_c$ . We use the symbol ( $P_{exc}$ ) to indicate that in our case "following the adiabatic curve"

Fig. 5. Potential curve diagram demonstrating in which way the Landau–Zener approximation is used to describe the excitation of a pair of  $He(2^3S)$ -atoms into an excited  $He(2^3S)$ –  $He(2^3P)$  pair. Around the Condon distance, where the diabatic states cross, non-crossing adiabatic states are formed by the coupling through the resonant radiation.

means absorption of the photon and formation of the system in the attractive ( ${}^{3}S{}-{}^{3}P$ ) state. The nice feature of relation (3) is that it can be accurately evaluated in the present case. At resonance, i.e., at the Condon point  $R_{c}$ , the electronic coupling between ground- and resonant state can be given in terms of the photon flux which, in turn, can be expressed in terms of the Rabi-frequency  $\Omega$  in the following way [18]:

$$|H_{12}|^2 = \hbar^2 \Omega^2 / 4. \tag{4}$$

The Rabi-frequency is related to the experimentally known saturation parameter  $s_0$  by

$$s_0 = 2\Omega^2 / \gamma^2. \tag{5}$$

For the range of detunings used in our present measurements, the corresponding Condon distances are so large that only the  $(R^{-3})$ -interaction in the excited state needs be considered. The Condon point is then given by  $R_c = (C_3/\delta)^{1/3}$ , and the difference in slopes of the interaction energies at  $R_c$  by  $\alpha = 3C_3/R_c^4$ . The radial velocity at  $R_c$  to be used in relation (3) is determined by the thermal velocities ( $v_{th}$ ) occurring for the ensemble of trapped He(2<sup>3</sup>S)-atoms at 1.1 mK. For a specific velocity vector the corresponding radial velocity is then simply  $v(\theta) = v_{th} \cos(\theta)$ , with ( $\theta$ ) being the



angle between the radius vector and the relative velocity vector at  $R_c$ . An evaluation of relation (3) for our experimental situation is shown in Fig. 6. The excitation probability is calculated for four different radial velocities, the highest one corresponding to a head on collision ( $\theta = 0$ ) at the average thermal velocity at 1.1 mK.

In our quantitative model, which is based on the evaluation of relation (3), we make the following simplifying assumptions:

- For close collisions on both, <sup>3</sup>S-<sup>3</sup>S- and <sup>3</sup>S-<sup>3</sup> P-potentials, the probability for Penning ionization is unity, if not forbidden by total spin selection rule. This is a good approximation as has been found for thermal collisions [6].
- Systems which decay by spontaneous photoemission while approaching can contribute to the ion rate only via a close collision on the <sup>3</sup>S-<sup>3</sup>S-potential. This is reasonable because acceleration in the excited state leads to sufficiently high relative velocities, so that re-excitation and capture is very improbable.
- The influence of photon recoil on the relative motion in the Condon region is neglected.
- Contributions to the ion rate from different 'paths' are obtained by summing intensities, so that possible coherence effects are neglected.
- To obtain ion rates comparable to experiment, the rates for the eight different attractive poten-



Fig. 6. Transition probabilities for systems crossing the Condon region, calculated with the Landau–Zener approximation for the conditions of our experiment ( $s_0 = 300$ , T = 1.1 mK). The different curves belong to different radial collision velocities (indicated fractions of the average thermal velocity).

tials are calculated separately, and the total rate is obtained as a weighted average. The corresponding different  $C_3$ -coefficients and molecular  $\gamma$ -values can be calculated from Ref. [15].

For the calculation of the ion rate constant for a given potential curve we proceed as follows. First, for a given detuning ( $\delta$ ) and a certain velocity ( $v_{th}$ ) the differential,  $\theta$  – dependent flux of collision systems through the Condon region is calculated for unit density. This differential flux (*F*) is given by

$$F(\theta) = 2\pi R_{\rm c}^2 v_{\rm th} \cos(\theta) \sin(\theta) \tag{6}$$

This flux branches at the Condon distance into two parts, depending on whether excitation occurs:

$$F(\theta)_{\rm SP} = F(\theta) \times p_{\rm exc}(\theta),$$
  

$$F(\theta)_{\rm SS} = F(\theta) \times (1 - p_{\rm exc}(\theta)).$$
(7)

Inside the Condon distance the excited collision systems evolve on the  ${}^{3}S{}^{-3}P{}$ -potential, and the unexcited ones on the  ${}^{3}S{}^{-3}S{}$ -potential.

Systems on the  ${}^{3}S{}^{-3}P{}$ -potential can have three fates: (1) A close collision occurs and leads to Penning ionization; (2) A close collision does not occur, so that the system does not contribute to ionization; (3) Spontaneous photo-emission occurs. In that case a close collision on the  ${}^{3}S{}^{-3}S{}^{-3}$ potential can still occur depending on the kinetic conditions, so that some contribution to the ion rate will arise. We thus have two differential ion rates from the initial inward motion of on the  ${}^{3}S{}^{-3}P{}^{-3}P{}$ -potential. For case (1):

$$DR(\theta)_{SP}(\text{inward})$$

$$= F(\theta) \times p_{exc}(\theta) \times C(\theta)_{SP} \times S(\theta), \text{ with }$$

$$C(\theta)_{SP} = 1 \text{ for close collisions,}$$

$$C(\theta)_{SP} = 0 \text{ otherwise,}$$
(8)

and with  $S(\theta)$  the survival probability of the system in the excited state at the time point of the close collision. And for case (3):

$$DR(\theta)_{SP}(inward)_{SS} = F(\theta) \times p_{exc}(\theta) \times C(\theta)_{SP} \times P(\theta),$$
(9)

where  $P(\theta)$  is the probability that spontaneous photo-emission occurs in such a way during par-

ticle approach that a close collision on the  ${}^{3}S-{}^{3}S-$  potential arises.

We now consider the case of systems not excited when first crossing the Condon region. There is again the possibility of a close collision, this time on the  ${}^{3}S$ -potential. This yields the ion rate:

$$DR(\theta)_{SS}(inward) = F(\theta) \times (1 - p_{exc}(\theta)) \times C(\theta)_{SS}.$$
(10)

 $C(\theta)_{SS}$  is defined in an equivalent way as  $C(\theta)_{SP}$  in (8). All systems that do not suffer a close collision, evolve along a straight line trajectory, and will cross the Condon region again in an outward motion. If such a system is not excited at this second crossing, it will separate. But if it is excited, it will be reflected on the attractive the <sup>3</sup>S-<sup>3</sup>P-potential if its radial kinetic energy is smaller than the interaction energy at  $R_c$ , i.e., smaller than the detuning. For the reflected part the same possibilities arise as those described above for the initial inward motion, with the exception that no close collisions on the <sup>3</sup>S-<sup>3</sup>S-potential are possible. These systems form a transient rotating and vibrating molecule which can decay in two ways, by a kind of predissociation during the outward motion, and by Penning ionization (or photo-emission) in close collisions, if it is not de-excited again after the reflection and evolves on the <sup>3</sup>S-<sup>3</sup>P-potential. Under the simplifying assumptions mentioned above, the ionization rate can be obtained by summing the, in principle, infinite number of contributions from the successive vibrations. We obtain in this way the following rates:

 $DR(\theta)_{SP}(outward)$ 

$$= F(\theta) \times p_{\text{exc}}(\theta) \times (1 - C(\theta)_{SS})$$
$$\times \{ (1 - p_{\text{exc}}(\theta)) / (1 + p_{\text{exc}}(\theta)) \}$$
$$\times C(\theta)_{\text{SP}} \times S'(\theta)$$
(11)

 $DR(\theta)_{SP}(outward)_{SS}$ 

$$= F(\theta) \times p_{\text{exc}}(\theta) \times (1 - C(\theta)_{\text{SS}})$$
$$\times \{ (1 - p_{\text{exc}}(\theta)) / (1 + p_{\text{exc}}(\theta)) \}$$
$$\times C(\theta)_{\text{SP}} \times P(\theta).$$
(12)

Relations (11) and (12) are modified with respect to relations (8) and (9) by the factor  $(1 - C(\theta)_{SS})$  to

account for the fact that in the first inward motion the trajectories leading to close collisions on the  ${}^{3}S$ -potential have been taken away by Penning ionization, and by the factor in curled brackets which accounts for the branching of the "outward" systems into close collisions with ionization on the  ${}^{3}S$ - ${}^{3}P$ -potential, and separation on the  ${}^{3}S$ - ${}^{3}S$ -potential, respectively.

The values of the step functions  $C(\theta)$ , of the survival probabilities  $S(\theta)$ , and of the probability function  $P(\theta)$ , are obtained by solving numerically the equations of motion for the trajectory characterized by the known initial conditions ( $v_{\rm th}$ ,  $\theta$ ,  $R_{\rm c}$ ). From the differential rates (8)–(12) the corresponding ion rates for given detuning ( $\delta$ ) and velocity  $(v_{\rm th})$  are obtained by integrating over  $(\theta)$ from 0 to  $(\pi/2)$ . By averaging over the thermal distribution of velocities, finally the total ion rate  $R(\delta)$  is obtained as the sum of the five contributions corresponding to the five distinguished differential rates (8)-(12). The curve in Fig. 4 is calculated by first calculating the curves  $R(\delta)$  for the eight relevant attractive <sup>3</sup>S-<sup>3</sup>P-potentials and then forming a weighted average. In Fig. 7 we show on a logarithmic scale in which way the rate  $R(\delta)$  is composed of the different contributions.

The agreement between the model prediction and the experiment is very good. In addition, the chosen semi-classical formulation affords good





insight into the physics of optical collisions. We would like to emphasize that our model contains no free parameter. This is the first time a successful detailed analysis of optical collisions is presented.

# 5. Measurements of kinetic energy distributions of Penning ions

The TOF-measurements were carried out at a negative detuning  $\delta = -15\gamma$  of the MOT-laser. From our analysis described above we know that under these conditions the main contribution to the ion rate comes from optical collisions, and more specifically, from Penning ionization of collision systems in the various attractive <sup>3</sup>S-<sup>3</sup>P-potentials. The ions were extracted from the MOT by a short low voltage pulse ( $\Delta \tau = 1 \mu s$ ,  $\Delta V = 6 V$ ). The quantitative analysis of the TOF-data is somewhat complicated due to the presence of the MOT magnetic field, and need be further investigated. The result of a straight forward analysis with only a first-order correction of the energy dependent transmission is presented in Fig. 8. The TOF-data, which show clearly separated distributions for He<sub>2</sub><sup>+</sup>- and He<sup>+</sup>-ions, are separately transformed to the energy scale for the two types of ions, and are presented on a scale where zero



Fig. 8. Kinetic energy distributions of He<sup>+</sup>- and He<sup>+</sup><sub>2</sub>-ions formed in optical collisions of two excited He-atoms by Penning ionization. The distributions are obtained from TOF-measurements.

means no initial kinetic energy. The He<sup>+</sup>-energy distribution shows the expected shape, and a width reflecting the involved potentials (see Fig. 2). The He<sub>2</sub><sup>+</sup>-distribution is narrow as expected, but seems to have a finite width and even some structure.

#### 6. Summary

We have presented experimental data for collisions of ultracold excited He-atoms in a MOT. The observed influence of the detuning of the MOT-laser frequency on the ion rate due to Penning ionization was analyzed in terms of a semiclassical model with no adjustable parameter. Good agreement between the model prediction and the experimental data was found. The complex nature of optical collisions became transparent for the first time in the analysis. Further, first preliminary measurements of energy distributions of ions produced in optical collisions of excited He-atoms were presented.

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#### References

- [1] H. Metcalf, P. Van der Straten, Phys. Rep. 244 (1994) 203.
- [2] K.-A. Suominen, J. Phys. B 29 (1996) 5981.
- [3] P.S. Julienne, A.M. Smith, K. Burnett, Adv. At. Mol. Opt. Phys. 30 (1993) 141.
- [4] P.D. Lett, P.S. Julienne, W.D. Phillips, Rev. Phys. Chem. 46 (1995) 423.
- [5] A. Niehaus, Phys. Rep. 186 (1990) 149.
- [6] M.W. Müller, A. Merz, M.W. Ruf, H. Hotop, W. Meyer, M. Movre, Z. Phys. D 21 (1991) 89.
- [7] R.J. Bieniek, Phys. Rev. A 18 (1977) 392.
- [8] G.V. Schlyapnikov, J.T.M. Walraven, U.M. Rahmanov, M.W. Reynolds, Phys. Rev. Lett. 73 (1994) 3247.

- [9] H.C. Mastwijk, J.W. Thomsen, P. van der Straten, A. Niehaus, Phys. Rev. Lett. 80 (1998) 5516.
- [10] H.C. Mastwijk, M. van Rijnbach, J.W. Thomsen, P. van der Straten, A. Niehaus, EPJ D, in press.
- [11] J. Kawanaka, M. Hagiuda, K. Shimizu, F. Shimizu, H. Takuma, Appl. Phys. B 56 (1993) 21.
- [12] P.A. Molenaar, P. van der Straten, H.G.M. Heideman, H. Metcalf, Phys. Rev. A 55 (1997) 605.
- [13] J.S. Major, D.F. Welch, Electron. Lett. 29 (1993) 2121.
- [14] J.W. Thomsen et al. (to be published).
- [15] P.S. Julienne, J. Vigué, Phys. Rev. A 44 (1991) 4464.
- [16] L. Landau, Phys. Z. Sowjet Union 2 (1932) 46.
- [17] P.S. Julienne, A.M. Smith, K. Burnett, Adv. At. Mol. Opt. Phys. 30 (1992) 141.
- [18] B.W. Shore, in: The Theory of Coherent Atomic Excitation, vol. 1, Wiley, New York, 1990, p. 209.