### **DIRAC NOTE 2007-10**

## Direct calculation of probability of the pionium ionization in target

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#### Abstract

The DIRAC analyzes  $\pi^+\pi^-$ -pairs with small relative momenta Q in their center of mass system in order to find out signal from the pionium ionization in target. The pionium ionization is in concurrence with the pionium annihilation which is mainly defined by the charge-exchange process  $\pi^+\pi^- \to \pi^0\pi^0$  with the theoretically predicted lifetime  $(2.9 \pm 0.1) \cdot 10^{-15}$  s. Thus the pionium lifetime can be deduced from the experimentally defined probability of pionium ionization if the dependence of the pionium ionization probability in the target as a function of its lifetime is established. Based on ionization cross-sections of pionium with target atoms we perform the first direct calculation of the pionium ionization probability in the target.

### Introduction

The main task of this work is the direct (based on ionization cross-sections) calculation of the pionium ionization probability in the target. The formalism of pionium dynamics based on a set of the probabilistic kinetic equations is reminded in Sect. 1. Section 2 is devoted to the direct calculation of the ionization probability.

## 1 Dynamics of a pionium in the target

Pionium atoms are Coulomb bound system of two oppositely charged pions. They can be created in a bound state from inelastic proton-nuclei collisions with the probability given by [1]

$$\frac{d\sigma_A}{d\vec{P}_A} = (2\pi)^3 |\Psi(0)|^2 \frac{E}{M} \left. \frac{d\sigma_s^0}{d\vec{p}_+ d\vec{p}_-} \right|_{\vec{p}_+ \approx \vec{p}_-},\tag{1}$$

where  $\frac{d\sigma_s^0}{d\vec{p}_+d\vec{p}_-}$  is the double inclusive cross section of  $\pi^+$  and  $\pi^-$  pairs without interaction in the final state with both pions produced either directly in hadronic processes or through short-lived resonances. The production of the pionium atoms with the angular momentum l > 0 is suppressed. The square of the S-wave function modulus at zero separation can be approximated in the following way [2]

$$|\Psi_{n0}(0)|^2 = (1+\delta_n) \left|\Psi_{n0}^C(0)\right|^2,\tag{2}$$

where  $\Psi_{n0}^{C}(0)$  is the pure Coulomb wave function of the  $\pi^{+}\pi^{-}$  atom at zero distance and the correction factor  $(1 + \delta_n)$  takes into account the effect of finite-size of the pion production region and the two-pion strong interaction in the final state. It was found that this correction shifts the probability of ionization on per mille level, therefore we will use pure Coulomb wave functions hereafter. For them only nS states are non-zero in the origin

$$\left|\Psi_{nlm}^{C}(0)\right|^{2} = \begin{cases} \frac{\left(\alpha m_{\pi}/2\right)^{3}}{\pi n^{3}} & \text{if } l = 0, \\ 0 & \text{otherwise.} \end{cases}$$
(3)

If we normalize probability of atom production to unity then the probability of atom production in the  $|nlm\rangle$  state reads

$$p_{nlm}(0) = \frac{\left|\Psi_{nlm}^{C}(0)\right|^{2}}{\sum_{n=1}^{\infty}\left|\Psi_{nS}^{C}(0)\right|^{2}} = \frac{\delta_{l0}}{n^{3}\sum_{n=1}^{\infty}1/n^{3}} = \frac{\delta_{l0}}{n^{3}\zeta(3)},\tag{4}$$

$$p_{100}(0) = 0.832, \quad p_{200} = 0.104, \quad p_{300} = 0.031.$$
 (5)

After production atom can either annihilate (mainly through  $\pi^+\pi^- \to \pi^0\pi^0$  process<sup>1</sup>) or electromagnetically interact with target atoms.

The partial decay width of the pionium in 1S state is [5]:

$$\Gamma_{2\pi^{0}} = \frac{2}{9}\alpha^{3}\sqrt{m_{\pi^{+}}^{2} - m_{\pi^{0}}^{2} - \frac{1}{4}m_{\pi^{+}}^{2}\alpha^{2}}(a_{0}^{0} - a_{0}^{2})^{2}m_{\pi^{+}}^{2}(1 + \delta_{\Gamma}), \quad \delta_{\Gamma} = (5.8 \pm 1.2) \times 10^{-2}.$$
(6)

The  $(a_0^0 - a_0^2)$  difference of the pion-pion S-wave scattering lengths with isospin 0 and 2 have been calculated [6] within the framework of standard chiral perturbation theory [7]

$$a_0^0 - a_0^2 = (0.265 \pm 0.004) m_{\pi^+}^{-1}.$$
(7)

This difference leads to the predicted value of the pionium lifetime in the ground state

$$\tau_{1S} = (2.9 \pm 0.1) \cdot 10^{-15} \, s. \tag{8}$$

While lifetime in nS states reads  $\tau_{nS} = \tau_{1S} \frac{|\Psi_{1S}(0)|^2}{|\Psi_{nS}(0)|^2} = \tau_{1S} n^3$ . Therefore the probability for a pionium with a laboratory momentum  $p_A$  to annihilate per unit length in target material is

$$W_{nlm}^{\text{anh}} = \frac{1}{\lambda_{nlm}^{\text{anh}}} = \begin{cases} \frac{1}{\gamma\beta c\tau_n} = \frac{2m_\pi}{p_A\tau_{1S}n^3} & \text{in } nS \text{ states,} \\ 0 & \text{in other states.} \end{cases}$$
(9)

While crossing the target a pionium electromagnetically interacts with target atoms. As the result a  $\pi^+\pi^-$  atom can be either ionized or transit from the initial bound state  $|n_i l_i m_i\rangle$  to another bound state  $|n_f l_f m_f\rangle$  (excitation/de-excitation). Hereafter we will denote initial and final bound states as  $|i\rangle$  and  $|f\rangle$ , respectively.

The probability of an ionization per unit length from the state  $|i\rangle$  is given by

$$W_i^{\text{ion}} = \frac{1}{\lambda_i^{\text{ion}}} = \frac{\rho N_A}{A} \sigma_i^{\text{ion}},\tag{10}$$

where  $\rho$  is the target density, A is its atomic weight,  $N_A$  is the Avogadro constant and  $\sigma_i^{\text{ion}}$  is the ionization cross section.

<sup>1</sup>Another annihilation channel  $\pi^+\pi^- \to 2\gamma$  amounts only about 0.3% [3, 4].

The probability of a pionium excitation per unit length from the state  $|i\rangle$  to the final state  $|f\rangle$  is given by

$$W_i^f = \frac{1}{\lambda_i^f} = \frac{\rho N_A}{A} \sigma_i^f = W_f^i, \tag{11}$$

where  $\sigma_i^f$  is the discrete (bound-bound) transition cross section.

The total cross section gives the probability of an atom to undergo an electromagnetic interaction

$$\sigma_i^{\text{tot}} = \sum_f \sigma_i^f + \sigma_i^{\text{ion}}.$$
 (12)

Total cross sections can be calculated owing to the completeness of the eigenstates of the Coulomb Hamiltonian.

Total and transition cross sections for any bound states were initially calculated in the Born approximation with the static potential of target atoms [8]. Later more accurate set of cross sections was derived which takes into account relativistic effects and target excitations [9]. Moreover in the latter work authors calculated ionization cross sections which provides the possibility to perform direct calculation of the pionium ionization probability in the target, which is the subject of this work. Comparison between different sets of cross sections was performed [10], where authors found that uncertainties in most precise sets of cross-sections for Ni target will cause only 1% uncertainty in the pionium lifetime. Uncertainty due to the accuracy of cross sections is expected to dominate precision of the ionization probability dependence on the pionium lifetime.

The dynamics of the pionium interaction with target atoms is supposed to be described by a set of kinetic equations [8] using the probabilities  $p_i(s)$  to find the  $\pi^+\pi^-$  atom in the definite quantum state  $|i\rangle$  at a distance s from the production point. This approach ignores any interference between different pionium bound states. For low n most of interference effects are suppressed at typical pionium momenta in DIRAC ( $3 \div 8 \text{ GeV}/c$ ) as the mean free path between pionium inelastic interactions is usually longer than the formation time of atomic system multiplied by its velocity. Nevertheless even for low n some interference effects can take place due to the accidental degeneracy of energy levels of hydrogen-like atoms. This problem was considered in the framework of the density matrix formalism [11]. It was found that the interference between quantum states with small n does not change the result based on a set of probabilistic kinetic equations (their difference is less than per mille).

Eigenstates of the Coulomb Hamiltonian form a countable set of discrete levels. For numerical calculations we will take into account only levels with principal quantum number  $n \leq n_{\text{max}}$ . For a given principal quantum number n there are  $n^2$  states  $|nlm\rangle$ with different orbital and magnetic quantum numbers. We will denote the total number of discrete bound states taken for the calculation as N. To make the system complete we introduce cross section  $\sigma_i^u$  which stands for the sum of transitions from state  $|i\rangle$  to any discrete state above  $n_{\text{max}}$ :

$$\sigma_i^u = \sum_{f:n_f > n_{\max}} \sigma_i^f = \sigma_i^{tot} - \sigma_i^{ion} - \sum_{f:n_f \leqslant n_{\max}} \sigma_i^f.$$
(13)

It is straightforward to write the probability of the pionium atom to be produced in any bound state above  $n_{\text{max}}$ :

$$p_u(0) = 1 - \sum_{i:n_i \leqslant n_{\max}} p_i(0).$$
(14)

Finally we will write the system of kinetic equations in the matrix form

$$\frac{d}{ds}\begin{pmatrix}p_{1}\\p_{2}\\\dots\\p_{N}\\p_{u}\\p_{\text{ion}}\\p_{\text{anh}}\end{pmatrix} = \begin{pmatrix}W_{1}^{1} & W_{2}^{1} & \dots & W_{N}^{1} & 0 & 0 & 0\\W_{1}^{1} & W_{2}^{2} & \dots & W_{N}^{2} & 0 & 0 & 0\\\vdots & \vdots & \ddots & \vdots & 0 & 0 & 0\\W_{1}^{N} & W_{2}^{N} & \dots & W_{N}^{N} & 0 & 0 & 0\\W_{1}^{\text{ion}} & W_{2}^{\text{ion}} & \dots & W_{N}^{\text{ion}} & 0 & 0 & 0\\W_{1}^{\text{ion}} & W_{2}^{\text{ion}} & \dots & W_{N}^{\text{ion}} & 0 & 0 & 0\\W_{1}^{\text{ion}} & W_{2}^{\text{ion}} & \dots & W_{N}^{\text{ion}} & 0 & 0 & 0\\W_{1}^{\text{ion}} & W_{2}^{\text{ion}} & \dots & W_{N}^{\text{ion}} & 0 & 0 & 0\\W_{1}^{\text{ion}} & W_{2}^{\text{ion}} & \dots & W_{N}^{\text{ion}} & 0 & 0 & 0\end{pmatrix}\begin{pmatrix}p_{1}\\p_{2}\\\dots\\p_{N}\\p_{u}\\p_{ion}\\p_{anh}\end{pmatrix}.$$
(15)

Diagonal terms describe the total decrease of level population

$$W_i^i = -\frac{\rho N_A}{A} \sigma_i^{\text{tot}} - W_i^{\text{anh}}.$$
(16)

System (15) is a system of linear ordinary differential equations with constant coefficients. The rank of the matrix is N, with 3 low lines being a linear combination of first N lines. It is exactly solvable

$$p_i(s) = \sum_k c_k \alpha_i^{(k)} e^{\lambda_k s},\tag{17}$$

where  $\lambda_1, \ldots, \lambda_N$  are eigenvalues and  $\alpha^{(k)}$  their corresponding eigenvectors. Symmetry of the upper left *N*-by-*N* corner guaranties that all its eigenvalues are real [15]. Coefficients  $c_k$  are fixed from initial conditions:

$$p_i(0) = \sum_k c_k \alpha_i^{(k)}.$$
(18)

The probability of ionization is expressed through the solution (17)

$$p_{\rm ion}(s) = \sum_{k} \frac{c_k}{\lambda_k} \left( e^{\lambda_k s} - 1 \right) \sum_{i} W_i^{\rm ion} \alpha_i^{(k)}.$$
 (19)

Expressions for  $p_u(s)$  and  $p_{anh}(s)$  have the same form as (19) if one substitutes  $W_i^{\text{ion}}$  with  $W_i^u$  or  $W_i^{\text{anh}}$  respectively.

DIRAC uses very thin targets (their nuclear efficiency is less than  $10^{-3}$ ), therefore atoms are produced nearly uniformly over the target thickness  $s_0$ . Hence the probability for a pionium to leave the target in the state  $|i\rangle$  reads

$$P_i(s_0) = \sum_k c_k \alpha_i^{(k)} \frac{1}{\lambda_k s_0} \left( e^{\lambda_k s_0} - 1 \right),$$
(20)

while the probability of ionization on the exit of the target is

$$P_{\rm ion}(s_0) = \sum_k \frac{c_k}{\lambda_k} \left( \frac{1}{\lambda_k s_0} \left( e^{\lambda_k s_0} - 1 \right) - 1 \right) \sum_i W_i^{\rm ion} \alpha_i^{(k)}.$$
(21)

Similar expressions can be derived for the probability of annihilation  $P_{\text{anh}}$  and for the probability  $P_u$  to reach any excited state with  $n > n_{\text{max}}$ .

$n_{\rm max}$	$P^A_{\rm dsc}$	$P^A_{\rm anh}$	$P^A_{\rm ion}$	$P_{\mathrm{u}}^{A}$
1	0.0712	0.4320	0.1246	0.3722
2	0.0841	0.4395	0.1742	0.3022
3	0.0889	0.4404	0.2042	0.2664
4	0.0914	0.4406	0.2262	0.2418
5	0.0928	0.4407	0.2438	0.2227
6	0.0937	0.4407	0.2586	0.2070
7	0.0943	0.4407	0.2715	0.1935
8	0.0947	0.4407	0.2828	0.1817

Table 1: Numerical solution with  $n > n_{\text{max}}$  states intact

In table 1 we illustrate this solution for a pionium atom produced in 95  $\mu$ m thick Ni target with momentum  $p_A = 4.6 \text{ GeV}/c$ , corresponding to the average laboratory momentum of produced pioniums in the kinematic range of the DIRAC experiment. Eigenvalues were numerically found by the appropriate function from LAPACK [16]. Numerical precision of the above solution can be estimated from the inequality

$$\left|1 - P_{\rm dsc}^A - P_{\rm anh}^A - P_{\rm ion}^A - P_{\rm u}^A\right| < 1 \cdot 10^{-12},\tag{22}$$

thus round-off errors do not affect the result.

System of equations (15) is constructed in a way that as soon as an atom reach the state with  $n > n_{\text{max}}$  it effectively quits from calculations and is kept intact, though in reality it is expected to undergo further electromagnetic interactions, e.g. it can be ionized or de-excited to the low-lying states. Therefore  $P_{\rm u}^A$  is the probability for atoms to reach states with  $n > n_{\text{max}}$ , which amounts to about 20%. This numerical value is in agreement with the earlier calculations [10] (fig. 3(d)).

As an atom transits to the state  $n_f > n_i$  its effective radius of electromagnetic interactions grows and its characteristic ionization length is getting shorter

$$\lambda_{|n>n_{\max},lm\rangle}^{\text{ion}} < \lambda_{|n_{\max}=8,lm\rangle}^{\text{ion}} \approx 2\,\mu\text{m}.$$
(23)

The target used in DIRAC is  $95\mu$ m thick, therefore highly excited atoms have a chance to leave the target in a bound state only if they were created in the rear part of the target close to its boundary. Otherwise these highly excited atoms will be ionized. These allow us to set the range for the ionization probability in the target:

$$0.2828 = P_{\rm ion}^A < P_{\rm ion} < P_{\rm ion}^A + P_u^A = 0.4645.$$
(24)

Here the upper bound corresponds to the case when all highly excited atoms are ionized, while the lower bound  $P_{\text{ion}}^A$  is at least probability of ionization from states with  $n \leq n_{\text{max}}$ .

From table 1 one can conclude that above upper and lower bounds converge slowly with increase of  $n_{\text{max}}$  and in this way it would be difficult to increase  $n_{\text{max}}$  in order to achieve precision required by DIRAC (per cent level).

# $2 \quad \text{Evolution of states with } n > n_{\max}$

Rather than trying to solve the system (15) directly we will modify it in order to get the *lower* bound of the ionization probability by taking into account dynamics of highly excited states with  $n > n_{\text{max}}$ . We will modify the system (15) in the following form:

$$\frac{d}{ds}\begin{pmatrix}p_{1}\\p_{2}\\\dots\\p_{N}\\p_{u}\\p_{\text{ion}}\\p_{\text{anh}}\end{pmatrix} = \begin{pmatrix}W_{1}^{1} & W_{2}^{1} & \dots & W_{N}^{1} & W_{u}^{1} & 0 & 0\\W_{1}^{2} & W_{2}^{2} & \dots & W_{N}^{2} & W_{u}^{2} & 0 & 0\\\vdots & \vdots & \ddots & \vdots & \vdots & 0 & 0\\W_{1}^{N} & W_{2}^{N} & \dots & W_{N}^{N} & W_{u}^{N} & 0 & 0\\W_{1}^{N} & W_{2}^{u} & \dots & W_{N}^{N} & W_{u}^{u} & 0 & 0\\W_{1}^{\text{ion}} & W_{2}^{\text{ion}} & \dots & W_{N}^{\text{ion}} & W_{u}^{\text{ion}} & 0 & 0\\W_{1}^{\text{ion}} & W_{2}^{\text{ion}} & \dots & W_{N}^{\text{ion}} & W_{u}^{\text{ion}} & 0 & 0\\W_{1}^{\text{ion}} & W_{2}^{\text{ion}} & \dots & W_{N}^{\text{ion}} & W_{u}^{\text{ion}} & 0 & 0\\W_{1}^{\text{ion}} & W_{2}^{\text{ion}} & \dots & W_{N}^{\text{ion}} & W_{u}^{\text{ion}} & 0 & 0\end{pmatrix}\begin{pmatrix}p_{1}\\p_{2}\\\dots\\p_{N}\\p_{N}\\p_{N}\\p_{ion}\\p_{anh}\end{pmatrix}.$$
(25)

Here

$$W_u^{\text{ion}} = \frac{\rho N_A}{A} \sigma_{n_{\max}+1,\min}^{\text{ion}}, \quad \sigma_{n_{\max}+1,\min}^{\text{ion}} = \min_{l'm'} \left\{ \sigma_{|n_{\max}+1,l'm'\rangle}^{\text{ion}} \right\}$$
(26)

is the lower bound of the probability of an ionization per unit length from any state with  $n > n_{\text{max}}$ , because the ionization cross section tends to grow with increasing of the principal quantum number n due to the corresponding expansion of the atomic radius. The minimal and maximal values of the ionization cross-section for different principal quantum numbers are drawn in fig. 1. To find the lower bound of the ionization probability, further we require all probabilities of an ionization per unit length from any state  $|nlm\rangle$  to be smaller, than  $W_u^{\text{ion}}$ :

$$W_{|nlm\rangle,\min}^{\rm ion} = \frac{\rho N_A}{A} \sigma_{|nlm\rangle,\min}^{\rm ion}, \quad \sigma_{|nlm\rangle,\min}^{\rm ion} = \min\left\{\sigma_{|nlm\rangle}^{\rm ion}, \ \sigma_{n_{\rm max}+1,\min}^{\rm ion}\right\}.$$
 (27)

The diagonal term  $W_i^i$ , which describes the level de-population, is changed accordingly to fulfill (12).



Figure 1:  $\min_{l'm'} \left\{ \sigma_{|nl'm'\rangle}^{\text{ion}} \right\}$  and  $\max_{l'm'} \left\{ \sigma_{|nl'm'\rangle}^{\text{ion}} \right\}$  for different principal quantum numbers n.

Upper bound of the probability of the de-excitation per unit length from all states with  $n > n_{\text{max}}$  to a state f with  $n_f \leq n_{\text{max}}$  is obtained from the following inequality

$$\sum_{i:n_i > n_{\max}} W_i^f p_i < \sum_{i:n_i > n_{\max}} W_i^f \cdot \sum_{i:n_i > n_{\max}} p_i = W_u^f p_u = W_f^u p_u.$$
(28)

Finally the diagonal term for the sum of discrete states with  $n > n_{\text{max}}$  is

$$W_u^u = -\frac{\rho N_A}{A} \sigma_{n_{\max}+1,\min}^{\text{ion}} - W_u^{\text{anh}} - \sum_{f:n_f \leqslant n_{\max}} W_u^f, \tag{29}$$

where  $W_u^{\text{anh}} = \frac{2m_{\pi}}{p_A \tau_{1S} (n_{\text{max}} + 1)^3}$  is the upper bound of the probability of an annihilation per unit length from all states with  $n > n_{\text{max}}$ . The rank of the new system is N+1. The system is constructed in the way that ionization is *underestimated* and all competitive processes including de-excitation from high n states (thus transitions to bound states with even lower ionization) are *overestimated*, therefore the solution is the *mathematical lower bound* of the probability of ionization. Numerical results are presented in Tab. 2.

$n_{\max}$	$P^B_{\rm dsc}$	$P^B_{\rm anh}$	$P^B_{\rm ion}$	$P_{\rm u}^B$
1	0.0856	0.5367	0.3241	0.05367
2	0.1273	0.4674	0.3881	0.01714
3	0.1247	0.4479	0.4225	0.00491
4	0.1169	0.4431	0.4383	0.00167
5	0.1109	0.4416	0.4468	0.00067
6	0.1068	0.4411	0.4517	0.00030
7	0.1041	0.4409	0.4548	0.00015
8	0.1023	0.4408	0.4567	0.00008

Table 2: Numerical solution for the lower bound of  $P_{ion}$  as a function of  $n_{max}$ 

Upper and lower bounds effectively squeeze the solution (fig. 2), for  $n_{\text{max}} = 8$  they are

$$0.4567 = P_{\rm ion}^B < P_{\rm ion} < P_{\rm ion}^A + P_u^A = 0.4645, \quad \frac{P_{\rm ion}^{\rm max} - P_{\rm ion}^{\rm min}}{2P_{\rm ion}} \approx 0.8 \cdot 10^{-2}, \quad (30)$$

which is within the precision, required by DIRAC. Range can be further shrunk by extrapolation as shown in fig. 2.

#### 2.1 Very thin target

We have to emphasize that upper and lower bounds of the ionization probability squeeze the solution with the required precision due to the fact, that for atoms with the principal quantum number n > 8 the characteristic ionization length is less than 2  $\mu$ m, which is much shorter than the target thickness of 95 $\mu$ m. If one selects very thin target (e.g. 10 $\mu$ m thick Ni) then the upper and lower bounds will show a worse convergence (fig. 3).



Figure 2: Upper and lower bounds of  $P_{\text{ion}}$  as a function of  $n_{\text{max}}$ , fitted by  $ae^{\alpha n_{\text{max}}} + c$  functions to guide the eye.



Figure 3: Upper and lower bounds of  $P_{\text{ion}}$  as a function of  $n_{\text{max}}$  for 10  $\mu$ m thick Ni target.

### 2.2 Extrapolated value of $P_{\rm ion}$

Upper and lower bounds of  $P_{\text{ion}}$  as a function of  $n_{\text{max}}$  do converge to the value of  $P_{\text{ion}}$  if  $n_{\text{max}} \to \infty$ . Dependence of  $P_{\text{ion}}$  bounds as a function of  $n_{\text{max}}$  can be approximated by hyperbolas

$$f_{\rm hyp}(n_{\rm max}) = \begin{cases} \frac{c_0}{n_{\rm max}^{c_1}} + c_2, & \text{upper limit} \\ -\frac{c_3}{n_{\rm max}^{c_4}} + c_2, & \text{lower limit} \end{cases}$$
(31)

or exponential functions

$$f_{\exp}(n_{\max}) = \begin{cases} c_0 e^{-c_1 n_{\max}} + c_2, & \text{upper limit} \\ -c_3 e^{-c_4 n_{\max}} + c_2, & \text{lower limit,} \end{cases}$$
(32)

which are simple shapes, which have required asymptotic behavior. Parameters  $c_0, \ldots, c_4$  were chosen to minimize the sum of squares of the residuals (points with  $n = [3, n_{\text{max}}]$  were used). Fits by  $f_{\text{hyp}}(n_{\text{max}})$  suggests the asymptotic value of  $\approx 0.464$ , while  $f_{\text{exp}}(n_{\text{max}})$  gives  $\approx 0.459$  (fig. 4).

# Conclusions

We confirm that the contribution of highly-excited states (with  $n > n_{\text{max}} = 8$ ) to the probability of ionization is significant (> 1/3). We derived a mathematical approach to calculate lower and upper bounds for the probability of ionization. These lower and upper bounds effectively squeeze the solution to the value of the probability of ionization with 1% precision, which is within requirements of the DIRAC experiment. Thus the first direct (based on ionization cross sections) calculation of the probability of ionization has been performed.

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## A Comparison to indirect calculations of $P_{ion}$

Before the probability of ionization  $P_{ion}$  was estimated indirectly [8, 10]

$$P_{\rm ion} = 1 - P_{\rm dsc} - P_{\rm anh}.$$
(33)

For  $n_{\text{max}} = 8$  the probability of annihilation  $P_{\text{anh}} \approx P_{\text{anh}}(n < n_{\text{max}})$  and the probability to leave the target in a bound state  $P_{\text{dsc}} = P_{\text{dsc}}(n < n_{\text{max}}) + P_{\text{dsc}}(n \ge n_{\text{max}})$  with the population of highly excited bound states approximated by

$$f_{\text{tail}}(n) = \frac{a}{n^3} + \frac{b}{n^5}.$$
 (34)

Free parameters a and b were chosen in a way that  $f_{\text{tail}}$  interpolates populations of bound states with  $n_{\text{max}} - 2$  and  $n_{\text{max}} - 1$ . Resulting indirect estimation of  $P_{\text{ion}}$  as a function of  $n_{\text{max}}$  is shown on fig. 4.



Figure 4: Asymptotic values of  $P_{\text{ion}}$  as a function of  $n_{\text{max}}$  for 95  $\mu$ m (left) and 10  $\mu$ m (right) thick Ni targets.

### **B** Comment on level populations

As small momentum transfers are preferred, transitions between close states prevail over transitions between more distant levels:

$$\sigma_{|n'l'm'\rangle}^{|nlm\rangle} > \sigma_{|n''l'm'\rangle}^{|nlm\rangle}, \text{ if } |n'-n| < |n''-n|.$$
(35)

Atoms are produced mainly in 1S- and 2S-states. The natural way to reach states with  $n > n_{\text{max}}$  is to follow a sequence of transitions with  $\Delta n = 1$  while increasing the orbital momentum of the pionium at the same time. This can be illustrated with fig. 5, where distribution of number of discrete-discrete transitions has its peak close to  $(n_{\text{max}}+1) - 1 = 8$  and even numbers of transitions are dominated. Fig. 6 shows populations of  $|n = 7, l, m\rangle$  states on the exit of the Ni 95  $\mu$ m target (the quantization axis is along the pionium momentum).



Figure 5: Distribution of number of discrete-discrete transitions before pionium reaches states with  $n > n_{\text{max}} = 8$ .



Figure 6: Population of pionium bound states with n = 7 on the exit of the target according to Eq. (15).

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