Solution of two–center time–dependent Dirac equation in spherical coordinates: 
Application of the multipole expansion of the electron–nuclei interaction

S. R. McConnell1,2, A. N. Artemyev1,2, M. Mai1,3 and A. Surzhykov1,2

1 Physikalisches Institut, Universität Heidelberg, Im Neuenheimerfeld 226, D–69120 Heidelberg, Germany
2 GSI Helmholtzzentrum für Schwerionenforschung GmbH, Planckstr. 1, D–64291 Darmstadt, Germany
3 Department of Physics, Yale University, 217 Prospect Street, New Haven, Connecticut 06511–8499, USA

(Dated: October 25, 2012)

Abstract: A non–perturbative approach to the solution of the time–dependent, two–center Dirac equation is presented with a special emphasis on the proper treatment of the potential of the nuclei. In order to account for the full multipole expansion of this potential, we express eigenfunctions of the two–center Hamiltonian in terms of well–known solutions of the “monopole” problem that employs solely the spherically–symmetric part of the interaction. When combined with the coupled–channel method, such a wavefunction–expansion technique allows for an accurate description of the electron dynamics in the field of moving ions for a wide range of internuclear distances. To illustrate the applicability of the proposed approach, the probabilities of the K– as well as L– shell ionization of hydrogen–like ions in the course of nuclear α–decay and slow ion–ion collisions have been calculated.

PACS numbers: 31.30.Jv, 34.80.Dp, 34.50.Fa

I. INTRODUCTION

Recent developments in accelerator and storage ring technologies have made it possible to perform a new generation of experiments on collisions between heavy, highly–charged ions. Of special interest in these studies are the low–energy collisions leading to the formation of short–lived quasi–molecular systems in which electrons move in the Coulomb field of two (or more) nuclei. Analysis of the excitation, ionization, charge–transfer, and pair–production processes in such a low–energy domain may reveal important information about the properties and behaviour of few–electron systems and even of the quantum vacuum in the presence of extremely strong electromagnetic fields. To achieve and exploit the strong–field regime, a broad research program is planned to be undertaken at the future Facility for Antiproton and Ion Research (FAIR) in Darmstadt, at which ions up to bare uranium will be produced and decelerated to required energies [1, 2].

In order to better understand the basic atomic processes accompanying slow ion collisions, the experimental findings have to be supplemented by a detailed theoretical analysis. In the simplest case of the collision between bare and hydrogen–like heavy ions, such an analysis can be traced back to the single–electron two–center Dirac problem. For small relative velocities and comparable charges of the nuclei, $Z_1 ≃ Z_2$, the non–perturbative treatment of such a problem is usually required and can be performed by using various coupled–channel techniques. Along this line, the time–dependent electron wave packet is expanded in terms of eigensolutions of the stationary Dirac equation, which describes the two–center system at a fixed internuclear distance $R$. The performance of the coupled–channel methods depends, therefore, on the efficiency of the spectrum generation of the time–independent Hamiltonian at each required $R$.

An accurate solution of the static two–center problem is in general a rather sophisticated task which can benefit from a proper choice of coordinate system. During the last two decades in particular, a number of theoretical methods have been developed which make use of Cassini and prolate spheroidal coordinate systems. Even though these (non–spherical) coordinates are very practical for the computation of quasi–molecular spectra at arbitrary internuclear distance $R$, their employment may be hampered by the lack of established numerical techniques for the evaluation of two–center matrix elements. Consequently, retention of standard spherical coordinates for the treatment of ion–ion (or ion–atom) collisions still attracts much current attention. The use of these, essentially one–center, coordinates for the description of the two–center problem also requires the development of various approximate methods. Within the linear combination of atomic orbitals (LCAO) approaches [8–12], for example, quasi–molecular wavefunctions are constructed from sets of atomic orbitals, centered on each nucleus. Yet another and very promising method relies on the direct solution of the two–center Dirac problem. Such a solution is rather straightforward and well–elaborated if the electron–nuclei potential is approximated by its spherically symmetrical part [13, 14]. This so–called monopole approximation is successfully used for the description of strong–field phenomena in close–ion collisions, but performs poorly when the Coulomb centers are far from each other. The extension of the multipole theory towards accounting for higher terms in the decomposition of the two–center potential is crucial, therefore, for the proper treatment of heavy–ion collisions in spherical coordinates.

*Corresponding author. Email: smcconne@physi.uni-heidelberg.de
II. THEORETICAL BACKGROUND

The electron dynamics in the Coulomb field of two nuclei is described by the time–dependent Dirac equation:

\[
\frac{\partial}{\partial t}\Psi(r, t) = \hat{H}_{TC}\Psi(r, t),
\]

where the Hamiltonian reads, in spherical coordinates, as:

\[
\hat{H}_{TC} = \alpha \cdot \mathbf{p} + V(Z_1, |r - R_1|) + V(Z_2, |r - R_2|) + \beta.
\]

In this expression, \( \mathbf{p} = -i \mathbf{\nabla} \) is the electron momentum operator, \( \beta \) and \( \alpha = \{\alpha_x, \alpha_y, \alpha_z\} \) are the standard Dirac matrices, and the potential generated by the \( i \)th nucleus:

\[
V(Z_i, |r - R_i|) = \int_0^\infty dr' \frac{\rho(r', Z_i)}{\max(r, R_i)}.
\]

is a function of its charge density distribution \( \rho(r, Z_i) \) and charge \( Z_i \). Moreover, \( R_1 \) and \( R_2 \) describe positions of the nuclei with respect to the center–of–mass of the system:

\[
R_1 = \frac{M_2}{M_1 + M_2} R, \\
R_2 = - \frac{M_1}{M_1 + M_2} R,
\]

where the internuclear vector \( R = R(t) \) varies over time.

In what follows, we shall discuss the solution of the time–dependent Dirac equation for relative ion velocities that are much smaller than the bound electron velocity \( v \approx \alpha Z_i \). For such a slow collision regime, the adiabatic approach is justified and requires first the treatment of the static two–center problem. In the next subsection, therefore, we will show how the eigensolutions of the time–independent (two–center) Hamiltonian can be efficiently generated for any internuclear distance.

A. Stationary two–center Dirac problem

For each (instantaneous) position of the nuclei, the spectrum of the two–center system can be obtained by solving the time–independent Dirac equation:

\[
\hat{H}_{TC}\Phi(r) = E\Phi(r)
\]

where \( E \) is the total energy and the Hamiltonian \( \hat{H}_{TC} \) is given by Eq. (2). Analysis of such an eigenproblem, can be significantly simplified by the proper choice of the quantization \( (z–) \) axis. For example, by setting this axis along the internuclear vector \( R \), we can write the multipole expansion of the two–center potential from Eq. (2) in the form:

\[
V_{TC}(r, R) = V(Z_1, |r - R_1|) + V(Z_2, |r - R_2|) = \sum_{l=0}^{\infty} V_l(r, R) P_l(\cos \theta),
\]

Natural units \((\hbar = m_e = c = 1)\) are used throughout the paper.
where \( P_l \) is the Legendre polynomial, \( \theta \) is the polar angle of the vector \( \mathbf{r} \), and the expansion coefficients \( V_l \) are given by:

\[
V_l(r, R) = \frac{2l+1}{2} \int_0^\pi \sin \theta \, d\theta \left( V(Z_1, |\mathbf{r} - \mathbf{R}_1|) + V(Z_2, |\mathbf{r} - \mathbf{R}_2|) \right) P_l(\cos \theta) .
\]

Moreover, if summation over \( l \) in Eq. (7) is restricted to the zeroth term, \( l = 0 \), the electron–nuclear interaction is governed by the spherically symmetric potential \( V_{TC}(\mathbf{r}, \mathbf{R}) = V_0(\mathbf{r}, R) \). The solution of the Dirac equation within such a monopole approximation is well–elaborated and has been discussed in a number of works \[13\,17\]. In particular, the eigenfunctions of the monopole Hamiltonian \( \hat{H}_{TC}^{(0)} = \alpha \cdot \mathbf{p} + V_0(\mathbf{r}, R) + \beta \) can be found in the form:

\[
\phi_{\kappa\mu}(\mathbf{r}) = \frac{1}{r} \left( \frac{G_k(r)\chi_{\kappa\mu}(\mathbf{r})}{F_k(r)} \right) ,
\]

where \( \chi_{\kappa\mu} \) is the standard Dirac spinor and the radial components satisfy the equation:

\[
\begin{bmatrix}
V_0 + 1 + \frac{d}{dr} + \frac{\kappa}{r}

\frac{d}{dr} + \frac{\kappa}{r} - \frac{d}{dr}

V_0 - 1 \end{bmatrix}
\begin{bmatrix}
G_k(r)

F_k(r)
\end{bmatrix} = \epsilon \begin{bmatrix}
G_k(r)

F_k(r)
\end{bmatrix} .
\]

Eq. (13) allows one to determine the vector \( \mathbf{C} = \{ C_1, C_2, ..., C_{N_{\text{max}}} \} \). Here, for the sake of brevity, we use short–hand notations \( C_j \equiv C_{n\kappa \mu}^{\kappa \mu} \), \( \phi_j \equiv \phi_n^{\kappa \mu} \) and \( \epsilon_j \equiv \epsilon_{nk} \).

As seen from the discussion above, the spectrum of the time–independent Hamiltonian \[2\] for each fixed internuclear distance \( R \) can be generated by means of the two–step procedure. In the first step, we employ the DKB finite–basis set approach to find solutions \( \{ \epsilon_{nk}, \phi_n^{\kappa \mu} \} \) of the monopole Hamiltonian. These solutions are used then, in the second step, to solve the generalized eigenvalue problem \[13\] and to obtain both the expansion coefficients \( C_{n\kappa \mu}^{\kappa \mu} \) of the wavefunctions \( \Phi_{n\kappa \mu}(\mathbf{r}) \) and the energies \( E_{\kappa \mu} \) of the electron states in the full two–center potential \[\Phi\]. In the next Section, such a new set of eigenstates \( \{ E_{\kappa \mu}, \Phi_{\kappa \mu} \} \) will be employed for solving the non–stationary Dirac problem.

**B. Time–dependent two–center Dirac problem**

Having generated a (quasi–) complete set of eigenstates of the two–center Hamiltonian \[2\] at each internuclear distance \( R \), we are ready now to solve the time–dependent equation \[11\] using the coupled channel method. Within this approach, the electron wavepacket \( \Psi(\mathbf{r}, t) \) is expanded:

\[
\Psi(\mathbf{r}, t) = \sum_{\kappa \mu} a_{\kappa \mu}(t) \Phi_{\kappa \mu}(\mathbf{r}, t) ,
\]

in terms of the functions \( \Phi_{\kappa \mu} \) which parametrically depend on the internuclear distance and, hence, on time \( t \). In Eq. (15), moreover, \( a_{\kappa \mu}(t) \) are the time–dependent expansion coefficients whose squares, \( |a_{\kappa \mu}(t)|^2 \), provide the occupation probabilities of the states \( \{ \Phi_{\kappa \mu} \} \) at a particular instant in time. In order to find these coefficients,
we substitute the expansion into the Dirac equation and derive the system of coupled channel equations:

\[
\frac{\, i\,}{\, dt\,} a_{k\mu}(t) = E_{k\mu}(t) a_{k\mu}(t) - i \sum_{n \neq k, \mu'} a_{n\mu'}(t) \left( \Phi_{k\mu}(t) \left[ \frac{\partial \Phi_{n\mu'}(t)}{\partial t} \right] \right). \tag{16}
\]

Any further analysis of this system requires the knowledge of how the electron–nuclei potential varies with time. Since the time–dependence enters into the problem solely through the internuclear distance \( R \), the equation of motion of colliding nuclei must be established. In the present work we consider the simplest case of motion along the Rutherford trajectories. In this case the time, the internuclear distance and the tilt angle of the molecular axis can be expressed in terms of the dimensionless parameter \( \xi \) as follows:

\[
t = \frac{a}{v_\infty} (\epsilon \sinh \xi + \xi), \quad R = a(\epsilon \cosh \xi + 1), \quad \theta = 2 \arctan \left( \frac{\sqrt{\epsilon^2 - 1} (\tanh(\xi/2) + 1)}{\epsilon + 1 - (\epsilon - 1) \tanh(\xi/2)} \right). \tag{17}
\]

Here notations are introduced

\[
a = \frac{\alpha Z_1 Z_2}{M_1 v_\infty^2}, \quad \epsilon = \left( 1 + \frac{b^2}{a^2} \right)^{1/2}, \tag{18}
\]

with \( b \) denoting the impact parameter, \( v_\infty \) the asymptotic value of the relative velocity of two particles at \( t = \infty \), and \( M_1 \) the reduced mass. By inserting Eq. (17) into the system of coupled channel equations (16) and re–writing it in terms of the parameter \( \xi \), we derive:

\[
\frac{\, i\,}{\, d\xi\,} a_{k\mu}(\xi) = \left( \frac{\partial t}{\partial \xi} \right) E_{k\mu}(\xi) a_{k\mu}(\xi)
- i \sum_{n \neq k, \mu'} a_{n\mu'}(\xi) \left( \frac{\Phi_{k\mu}(\xi) \left[ \frac{\partial R}{\partial \xi} \frac{\partial V(r, \mathbf{R}, \xi)}{\partial R} \right] \Phi_{n\mu'}(\xi)}{E_{n\mu'}(\xi) - E_{k\mu}(\xi)} \right)
- \frac{\, i\,}{\, d\theta\,} \langle \Phi_{k\mu}(\xi) | j_y | \Phi_{n\mu'}(\xi) \rangle, \tag{19}
\]

where we used the relation

\[
\langle \Phi_{k\mu} | \Phi_{n\mu'} \rangle = \delta_{k,n} \frac{\langle \Phi_{k\mu} | \frac{\partial V(r, \mathbf{R}, \xi)}{\partial R} | \Phi_{n\mu'} \rangle}{E_{n\mu'} - E_{k\mu}}, \tag{20}
\]

which is valid if the collision occurs in XZ–plane. Here \( j_y \) is \( y \)–component of the total momentum projection operator and \( \delta_{k,n} \) is the anti–Kronecker delta symbol. The parametrization of \( t, R \) and \( \theta \) in terms of \( \xi \) is most natural since the differential equation governing the time evolution of \( R \) is autonomous i.e. an exact solution is possible only for \( t(R) \) and not, as required, for \( R(t) \).

In order to solve the system of coupled channel equations and, hence, to find the expansion coefficients \( a_k \), it is convenient to re–write Eq. (19) in matrix form:

\[
\frac{\, i\,}{\, d\xi\,} \tilde{a}(\xi) = M(\xi) \tilde{a}(\xi), \tag{21}
\]

where \( \tilde{a} = (a_1, a_2, \ldots) \), and the individual elements of \( M_{k,n}(\xi) \) are given by

\[
M_{k,n}(\xi) = \frac{\partial t}{\partial \xi} E_{k\mu} \delta_{k,n} \delta_{\mu\mu'}
- \frac{\, i\,}{\, d\theta\,} \langle \Phi_{k\mu} | j_y | \Phi_{n\mu'} \rangle.
\tag{22}
\]

The matrix equation (21) can be integrated numerically on a grid of spacing \( \Delta \xi \) according to:

\[
\tilde{a}(\xi + \Delta \xi) = e^{-iM(\xi+\Delta \xi)\Delta \xi} \tilde{a}(\xi) + O(\Delta \xi^2), \tag{23}
\]

and determines the vector \( \tilde{a}(\xi + \Delta \xi) \) at the “time” \( \xi + \Delta \xi \) provided that the expansion coefficients \( a_{k\mu} \) at the earlier moment \( \xi \) are known. Since the matrix exponential in the right–hand–side of Eq. (23) is unitary, the norm of the vector \( \tilde{a} \) will be preserved at each iteration.

The iteration scheme (23) represents the final step in the numerical treatment of the time–dependent two–center Dirac equation (1). In Section IV we will use this scheme in order to investigate the electron ionization induced by the nuclear \( \alpha \) decay as well as the slow ion–ion collisions. In the present calculations, we shall restrict ourselves to the simplest case of zero–impact–parameter picture, \( b = 0 \). Within this framework, the last term in Eqs. (19) and (20) vanishes and, hence, the matrix elements of the evolution matrix (22) are diagonal in \( \mu \).

III. DETAILS OF COMPUTATIONS

Having discussed the non–perturbative approach to the solution of the two–center Dirac problem, we are ready now to investigate the electron emission accompanying both, the \( \alpha \)–decay of heavy nuclei and the slow ion–ion collisions. Before starting with the presentation and analysis of the numerical results, let us briefly summarize the most important details of our calculations which, as mentioned in Section III, can be split into three stages. In the first step of this procedure, the eigenfunctions of the spherically–symmetric Hamiltonian \( \hat{H}^{(0)}_{TC} \) are obtained by the DKB B–spline basis set method which guarantees the
absence of the non–physical spurious states in the spectrum [20]. In the present work, we used about 200 B–splines of eighth order defined in a box of size \( L \approx 10^3 \) fm in order to construct “monopole” wavefunctions \( \phi_{n\kappa}^a(r) \) with energies in the range \( 0 \leq \epsilon_{n\kappa} \leq 10 \) mc\(^2\). Based on the detailed numerical analysis, we argue that such a truncated basis set allows one to achieve \( \sim 5–10 \% \) accuracy in the prediction of the ionization cross sections.

As the second step of the non–perturbative treatment, the solutions of the full two–center Hamiltonian (2) are expanded in terms of \( \phi_{n\kappa}^a(r) \) (cf. Eq. (10)). Along this line, we obtain about 300 functions \( \Phi_{k\mu}(r; R) \) and corresponding energies \( E_k \) for each internuclear distance \( R \) (or, equivalently, dimensionless parameter \( \xi \)). It is worth mentioning that the solutions of eigenproblem \( H\Phi = \epsilon \Phi \) and, hence, \( \Phi_{k\mu}(r) \) are defined up to an arbitrary sign. In our calculations, this sign is chosen for all \( \Phi_{k\mu}(r) \) from the requirement that their large radial components, calculated for two successive steps over \( \xi \), behave similarly near the origin of the coordinates, i.e. for \( r = 0...500 \) fm.

With the help of generated basis sets \( \{ \Phi_{k\mu}(r; R(\xi)) \}_{k=1,...,N} \) we are finally able to perform the time propagation of the electron wavepacket in the field of moving nuclei. Prior to starting this propagation, one has to define the electron wavefunction in the initial moment of time. Indeed, the initial conditions depend on the particular process under consideration. For the nuclear \( \alpha–\)decay, for example, we assume that the electron is originally in the ground \( 1s \) state of the united nucleus of charge \( Z \). Since the time–propagation begins from the moment when the \( \alpha \) particle leaves the potential barrier at the distance \( R_0 \) from the daughter nucleus, we project, at \( \xi = 0 \) (corresponding to \( t = 0 \)), the wavefunction \( \psi_{1s1/2}(r; Z) \) onto the basis set of eigenfunctions of Hamiltonian (2) describing system of two Coulomb centers with charges \( Z_1 = 2 \) and \( Z_2 = Z – 2 \), placed at distances \( R_1 \) and \( R_2 \) with respect to their center–of–mass (see Eq. (4)). Such a projection procedure allows us to account for the shake–off effect and to obtain the first set of expansion parameters \( \{ a_{k\mu}(\xi = 0) \}_{k=1,...,N} \) which are used then to find the electron wavepacket in subsequent time steps (cf. Eq. (28)). In order to produce results, presented in the Section IV A time propagation was carried out for about 750 such steps of \( \Delta \xi = 0.01 \), this corresponds to the retreat of the \( \alpha \) particle to a distance of about \( 10^4 \) fm.

In contrast to the \( \alpha–\)decay, the time propagation of the electron wavepacket in the field of two colliding uranium ions, studied in Section IV B, was started from the moment when the ions are separated from each other by the distance \( R = 5 \cdot 10^3 \) fm. In this initial moment, the electron finds itself in the ground \( 1s1/2 \) state of one of the projectiles. The wavefunction of such a state is given by the sum of the lowest–lying gerade and ungerade solutions of the (stationary) two–center Hamiltonian, \( \psi_{1s1/2}(r; Z = 92) \approx 1/\sqrt{2}(\Phi_{1s\sigma} + \Phi_{1s\sigma}) \); an approxima-

![Figure 1](attachment:image.png)

**FIG. 1:** \( K\)–shell ionization probability of hydrogen–like xenon (top panel), gadolinium (middle panel) and polonium (bottom panel) ions following the \( \alpha–\)decay. Non–perturbative calculations were carried out within the monopole approximation (dashed line) and by taking the full two–center potential into account (solid line). The probability is scaled \( \times 10^5 \).

IV. RESULTS AND DISCUSSION

A. Ionization following \( \alpha–\)decay of heavy nuclei

The non–perturbative approach presented in Section III can be used to study basic atomic processes accompanying slow collisions of two ions independent of their nuclear charges \( Z_1 \) and \( Z_2 \). In this section, we employ it to re–analyze the nuclear \( \alpha \) decay, which is an example of a (charge–) asymmetric collision, \( Z_1 << Z_2 \), with zero impact parameter and which can be treated also within first–order perturbation theory. As mentioned already, such a perturbative treatment has been successfully ap-
plied over the last decades in a large number of studies \cite{24,26,27}. In order to compare predictions of the non–perturbative and perturbative theories, we consider the decay of $\alpha$–active $^{110}$Xe, $^{148}$Gd and $^{210}$Po isotopes. For these zero–nuclear–spin nuclei, calculations have been performed for the ionization probability $P_K$ of an electron from the ground $1s_{1/2}$ state of an initially hydrogen–like system. In Fig.\(1\) we display the non–perturbative results for the $P_K$ as a function of the internuclear distance. To deduce this probability, we have evaluated the electron wavefunction $\Psi(\mathbf{r}, t)$ at each step of the time propagation (see Eqs. (15)–(23) and related discussion) and projected it onto the positive–energy solutions of the two–center Dirac equation for the instantaneous distance $R = R(t)$

$$P_K(R(t)) = \sum_{E_k > mc^2} |\langle \Phi_{kp} | \Psi(\mathbf{r}, t) \rangle|^2 = \sum_{E_k > mc^2} |a_{kp}(t)|^2.$$ (24)

Calculations have been performed both within the monopole approximation, in which summation over \(l\) in Eq. (6) is restricted to the zeroth term, and by taking the full two–center potential $V_{TC}(\mathbf{r}, \mathbf{R})$ into account. As was expected, these two approaches agree only for relatively small internuclear distances. If $R$ becomes greater than 500 fm, the monopole approximation can significantly underestimate the ionization probability; an effect which becomes most pronounced for the heavy nuclei.

Fig.\(1\) shows that at very large distances, $R > 8000$ fm, the ionization probability $P_K$ converges to some final value which depends only on the charge of the mother nucleus and the initial velocity of the $\alpha$ particle. This “asymptotic” value of $P_K$ is displayed in Table \(1\) for xenon, gadolinium and polonium ions, and compared with the results of our first–order perturbation calculations (see Ref. \cite{28} for further details). Moreover, the previous (perturbative) predictions of Law \cite{22}, and Fischbeck and Freedman \cite{24} obtained for the decay of polonium are given in the third column. As seen from the table, the non–perturbative treatment, based on the full multipole expansion of the two–center potential, reproduces well the ionization probabilities for all three ions. In particular, both perturbative and non–perturbative theories yield results that agree to within 5% if applied to the exploration of the $\alpha$ decay of polonium ions. If, however, the potential $V_{TC}(\mathbf{r}, \mathbf{R})$ is approximated in Eq. (6) by the single monopole term, the non–perturbative calculations may result in approximately a 30% misestimation of $P_K$.

Until now we have discussed the $\alpha$–decay–induced ionization of hydrogen–like ions that have been prepared initially in the ground $1s_{1/2}$ state. In order to verify the performance of the non–perturbative technique, based on the multipole expansion of the two–center interaction operator, it is also worth considering the electron emission from the various $L$ subshells. Even though experimental observation of the $L$–shell ionization of hydrogen–like systems might be hampered by the short lifetimes of excited ionic states, it can be measured for neutral atoms. Theoretically, such an atomic inner–shell ionization can be well described by using the developed approach if the proper screening potential is used in Eq. (6). The analysis of the screening effects in $\alpha$–decay–induced processes in neutral systems is, however, out of the scope of the present work. Instead, we just employ the $L$–shell ionization of hydrogen–like atoms as a testing ground for the non–perturbative theory from Section \(1\) The internuclear–distance–dependent probabilities for the ionization of $2s_{1/2}$ (top panel), $2p_{1/2}$ (middle panel) and $2p_{3/2}$ (bottom panel) states of hydrogen–like polonium are evaluated based on this theory and are presented in Fig.\(2\). Similar to before, calculations have been performed by accounting for the full multipole expansion of the two–center potential (solid line) and by restricting this summation to the monopole term only (dashed line). Agreement between these two approaches can be observed again only for small internuclear distances, while for $R > 600$ fm the monopole calculations underestimate the ionization probabilities by more than 25%. Moreover, the monopole approximation fails to reproduce $P_{2p_{1/2}}$ for the entire range of $R$.

The asymptotic values of $P_{2s_{1/2}}$, $P_{2p_{1/2}}$ and $P_{2p_{3/2}}$ calculated for large distances $R$ are presented in Table \(1\) and compared with the predictions of first–order perturbation theory \cite{24} and data by Law \cite{22}. As in the case of $K$–shell ionization, the full account of the electron–nuclei interaction $V_{TC}(\mathbf{r}, \mathbf{R})$ in Eq. (2) leads here to approximately 5% agreement between the predictions of perturbative and non–perturbative theories for the entire $L$ shell. In contrast, the time propagation of the electron wavepacket in the spherically–symmetric potential $V_0(\mathbf{r}, R)$ yields the probabilities $P_L$ that are 30% smaller.

| Ion         | $T_{kin}$ (MeV) | Perturbative | Non–perturbative |
|-------------|-----------------|-------------|-----------------|
| $^{110}$Xe$^{+54}$ | 3.7             | 3.61        | 2.6             | 3.2             |
| $^{148}$Gd$^{+63}$ | 3.1             | 2.15        | 1.6             | 2.3             |
| $^{210}$Po$^{+83}$ | 5.4             | 2.00        | 1.81$^a$        | 2.03$^b$        |

$^a$Law \cite{22}  
$^b$Fischbeck and Freedman \cite{24}

\(K\)-shell ionization probability of hydrogen–like xenon, gadolinium and polonium ions following the $\alpha$–decay.

The non–perturbative calculations, performed for $R \rightarrow \infty$ by using the monopole as well as exact approximations to the two–center potential, are compared with the first–order perturbation results and predictions by Law \cite{22}, and Fischbeck and Freedman \cite{24}. The asymptotic kinetic energy of a particle $T_{kin} = M_0 v_{\infty}^2 / 2$ from Ref. \cite{28} is given in the second column. All probabilities are of the order of $10^2$.
fully utilized to study the functions in terms of monopole solutions, can be successfully utilized to study the α–decay–induced ionization. Non–perturbative calculations were carried out within the monopole approximation (dashed line) and by taking the full two–center potential into account (solid line). The probability is scaled $\times 10^3$.

comparing to the perturbative results. Again, these findings stress the importance of the higher multipole contributions to the electron–nuclei interaction for the time–dependent analysis [23] of the electron dynamics accompanying ion collisions.

B. Ionization in $^9{U}^{2+}$–$^9{U}^{2+}$ collisions

So far, we have shown that the time–dependent method [19], based on the expansion of the basis wavefunctions in terms of monopole solutions, can be successfully utilized to study the α–decay–induced ionization. Besides this—purely perturbative—problem, the performance of the developed approach has been also examined for slow collisions between two high–Z ions. In contrast to the α–decay, theoretical analysis of such collisions usually can not be carried out within the framework of the perturbation theory and demands the application of non–perturbative techniques. Along this line we have focused, in particular, on the K–shell ionization in $^9{U}^{2+}$–$^9{U}^{2+}$ collisions at zero impact parameter. The ionization probability $P_K$ has been calculated based on Eq. (24), where the electron wavepacket $\Psi(r,t)$ was propagated from a time when the ions were at a distance $R = 5 \cdot 10^3$ fm, through the closest approach $R_0 \approx 50$ fm, to a moment when the internuclear distance increased again to $R = 5 \cdot 10^3$ fm. In Fig. 3 for example, $P_K$ is displayed as a function of the distance $R$ and for the (relative) collision energies $T_p = 1.8$, 2.0 and 2.2 MeV/u. As seen from the figure, the steep rise of the ionization probability appears immediately after the point of closest approach $R_0$ at which the (relative) ionic motion is suddenly reversed and the electron can be “shaken off” into the continuum. Such a behaviour of the $P_K$ as well as its further damped oscillations have been predicted previously in Ref. [12] based on the monopole approximation and now is confirmed by our theory that accounts for the multipole expansion of the electron–nuclei interaction. Moreover, our calculations clearly indicate a rise of the ionization probability with the collision energy. For example, the asymptotic value of the $P_K$ is increased by almost factor of three if the initial (relative) energy changes from 1.8 to 2.2 MeV/u. Further significant enhancement of the $P_K$ is predicted for higher energies at which the “diving” of the ground quasi–molecular state into the Dirac’s negative continuum takes place [12]. However, since the analysis of such strong–field phenomena is out of scope of the present paper, we restrict here our calculations to the “undercritical” energy range, $T_p \lesssim 2.3$ MeV/u.

V. SUMMARY AND OUTLOOK

In summary, we have laid out a theoretical approach to the time–dependent two–center Dirac problem. Within such an approach, the wavefunctions, describing the (sin-
The α \textit{positive} values of \linebreak \textit{line}, and 2.2 MeV/u (dash–dotted \textit{line}). The \textit{negative} and \textit{positive} \textit{values} \textit{of} \textit{R} \textit{correspond} \textit{to} \textit{the} \textit{times} \textit{when} ions \textit{approach} \textit{and move} \textit{away} \textit{from} each other, respectively.

The developed \textit{time–dependent approach} \textit{can} \textit{help} \textit{to} \textit{explore} \textit{various} \textit{atomic} \textit{processes} \textit{accompanying} slow \textit{ion collisions}. In the \textit{present work}, \textit{for} \textit{example}, \textit{we} \textit{used} \textit{this} \textit{theory} \textit{to} \textit{calculate} \textit{the} \textit{electron–loss} \textit{probabilities} \textit{for} the \textit{K}– \textit{and L}–shell α–decay–induced \textit{ionization}, \textit{predictions} of both \textit{perturbative} and \textit{non–perturbative} \textit{methods} \textit{were found} \textit{to} \textit{agree} \textit{to} \textit{within} about 5 \% \textit{if} the \textit{multipole expansion} of the \textit{two–center potential} \textit{is} \textit{taken} \textit{into} \textit{account} \textit{in} the \textit{time–dependent Hamiltonian \textit{2}}. If, \textit{in} \textit{contrast}, \textit{this} \textit{potential} \textit{is} \textit{approximated} \textit{by} its \textit{monopole term}, \textit{our} \textit{calculations} \textit{may} \textit{underestimate} \textit{the} ionization \textit{probabilities} \textit{by} more than 30 \%; \textit{this} \textit{failure} \textit{of} \textit{the} monopole \textit{approximation} \textit{becomes} \textit{most} \textit{pronounced} \textit{for} \textit{large} inter–nuclear \textit{distances}. Based on \textit{these} \textit{findings} \textit{we} \textit{stressed} \textit{the} \textit{vital importance} \textit{of} \textit{the} \textit{proper} \textit{treatment} \textit{of} the electron–nuclear \textit{interaction} \textit{for} the \textit{accurate} \textit{description} \textit{of} slow \textit{ion collisions}. The \textit{rigorous} \textit{“multipole” approach} \textit{has} \textit{been} \textit{employed} \textit{then} \textit{to} \textit{explore} the \textit{K}–shell \textit{ionization} \textit{accompanying} U$^{91+}$–U$^{92+}$ collisions. For \textit{this}—purely \textit{non–perturbative—process}, \textit{we} \textit{qualitatively} \textit{confirmed} \textit{the} \textit{impact–parameter–behaviour} \textit{of} \textit{the} ionization \textit{probability}, \textit{which} \textit{was} \textit{predicted} \textit{previously} \textit{by} Betz \textit{and co–authors \textit{12} within} the \textit{monopole theory}.

Both the α–decay \textit{of} hydrogen–like \textit{ions} \textit{and} the U$^{91+}$–U$^{92+}$ \textit{scattering} \textit{have} \textit{been} \textit{explored} \textit{in} \textit{the} \textit{present work} \textit{for} \textit{the} \textit{case} \textit{of} zero \textit{impact} \textit{parameter}. \textit{Of} \textit{course}, \textit{the} developed \textit{non–perturbative} \textit{method} \textit{is} \textit{not} \textit{limited} \textit{to} \textit{such} \textit{a} \textit{simple} \textit{geometry} \textit{and} \textit{can} \textit{be} \textit{applied} \textit{to} \textit{analyze} heavy–ion \textit{collisions} \textit{at} $b \neq 0$. \textit{For} \textit{these} \textit{collisions}, \textit{the} \textit{last} \textit{term} \textit{of} Eqs. \textit{19}–\textit{20}, 
\textit{that} \textit{accounts} \textit{for} \textit{the} \textit{rotation} \textit{of} \textit{the} \textit{internuclear} \textit{distance}, \textit{does} \textit{not} \textit{vanish} \textit{and} \textit{makes} \textit{the} \textit{elements} \textit{of} \textit{the} \textit{rotation} \textit{matrix} \textit{22} \textit{non}–\textit{diagonal} \textit{in} $\mu$. \textit{The} \textit{impact–parameter dependence} \textit{of} \textit{the} \textit{electron loss} \textit{as} \textit{well} \textit{as} \textit{the} \textit{excitation} \textit{and} \textit{the} \textit{charge} \textit{transfer} \textit{processes} \textit{will} \textit{be} \textit{discussed} \textit{in} \textit{a} \textit{forthcoming} \textit{publication} \textit{and} \textit{will} \textit{help} \textit{in} \textit{planning} \textit{future} \textit{experiments} \textit{on} \textit{slow} \textit{collisions} \textit{between} \textit{two} \textit{high–Z projectiles}. \textit{These} \textit{experiments} \textit{are} \textit{likely} \textit{to} \textit{be} \textit{carried out} \textit{at} \textit{the} \textit{Facility} \textit{for Antiproton and Ion Research (FAIR)} \textit{in Darmstadt} \textit{and} \textit{are} \textit{expected} \textit{to} \textit{reveal} \textit{unique} \textit{information} \textit{about} \textit{the} \textit{quantum} \textit{electrodynamics} \textit{of} extremely \textit{strong} \textit{fields}.

\textbf{VI. ACKNOWLEDGMENTS}

S.M. acknowledges the support of the International Max Planck Research School for Quantum Dynamics (IMPRS–QD). This work is supported by the Helmholtz Gemeinschaft (Nachwuchsgruppe VH–NG–421).

\bibitem{1} FAIR 2001 Conceptual Design Report: An International Accelerator Facility for Beams of Ions and Antiprotons, ed. W. Henning (Darmstadt: GSI)
\bibitem{2} A. Gumberidze \textit{et al.}, Nucl. Instrum. Methods B \textbf{267}, 248 (2009).
\bibitem{3} P. Schlüter, K. H. Wietschorke, and W. Greiner, J. Phys. A: Math. Gen. \textbf{16}, 1999 (1983).
\bibitem{4} K. H. Wietschorke, P. Schlüter, and W. Greiner, J. Phys. A: Math. Gen. \textbf{16}, 2017 (1983).
\bibitem{5} A. N. Artemyev, A. Surzhykov, P. Indelicato, G. Plunien, and Th. Stöhlker, J. Phys. B: At. Mol. Opt. Phys. \textbf{43}, 235207 (2010).
\bibitem{6} O. Kullie and D. Kolb, Eur. Phys. J. D \textbf{17}, 167 (2001).
\bibitem{7} F. Fillion–Gourdeau, E. Lorin, and A. D. Bandrauk,
[8] W.-D. Sepp, D. Kolb, W. Sengler, H. Hartung, and B. Fricke, Phys. Rev. A 33, 3679 (1986).
[9] K. Momberger, N. Grün, and W. Scheid, J. Phys. B: At. Mol. Opt. Phys. 26, 1851 (1993).
[10] M. Gail, N. Grün, and W. Scheid, J. Phys. B: At. Mol. Opt. Phys. 36, 1397 (2003).
[11] I. I. Tupitsyn, Y. S. Kozhedub, V. M. Shabaev, G. B. Deyneka, S. Hagmann, C. Kozhuharov, G. Plunien, and Th. Stöhlker, Phys. Rev. A 82, 042701 (2010).
[12] I. I. Tupitsyn, Y. S. Kozhedub, V. M. Shabaev, A. I. Bondarev, G. B. Deyneka, I. A. Maltsev, S. Hagmann, G. Plunien, and Th. Stöhlker, Phys. Rev. A 85, 032712 (2012).
[13] W. Betz, G. Soff, B. Müller, and W. Greiner, Phys. Rev. Lett. 37, 1046 (1976).
[14] J. Reinhardt, B. Müller, and W. Greiner, Phys. Rev. A 24, 103 (1981).
[15] E. Ackad and M. Horbatsch, Phys. Rev. A 75, 022508 (2007).
[16] E. Ackad and M. Horbatsch, Phys. Rev. A 78, 062711 (2008).
[17] G. B. Deineka, I. A. Maltsev, I. I. Tupitsyn, V. M. Shabaev, G. Plunien, Russ. J. Phys. Chem. B 6, 224 (2012).
[18] W. Greiner, *Relativistic Quantum Mechanics: Wave Equations*, 3rd ed. (Springer, Berlin, 2000).
[19] A. Marsman and M. Horbatsch, Phys. Rev. A 84, 032517 (2011).
[20] V. M. Shabaev, I. I. Tupitsyn, V. A. Yerokhin, G. Plunien, and G. Soff, Phys. Rev. Lett. 93, 130405 (2004).
[21] H. J. Fischbeck and M. S. Freedman, Phys. Rev. A 15, 162 (1977).
[22] J. Law, Nucl. Phys. A 286, 339 (1977). The $L$–shell ionization probabilities, although calculated with the Dirac–Coulomb wavefunctions, are given in this paper per shell. These results, therefore, have been multiplied with the factor $1/(2j + 1)$, where $j$ is the total angular momentum of the electron, to be compared with our predictions.
[23] R. Anholt and P. A. Amundsen, Phys. Rev. A 25, 169 (1982).
[24] S. R. McConnell, A. N. Artemyev, and A. Surzhykov, J. Phys. B: At. Mol. Opt. Phys. 44, 145204 (2011). In the course of the present study, we found that Eq. (11) of this work contains a mistake. The correct form of this expression, that describes the multipole expansion of the Coulomb interaction operator, is given by Eq. (16) in Ref. [25]. Moreover, the non–relativistic form of the recoil operator was used in this work.
[25] K. Rumrich, G. Soff, and W. Greiner, Phys. Rev. A 47, 215 (1993).
[26] J. S. Blair and R. Anholt, Phys. Rev. A 25, 907 (1982).
[27] J. Revai and Y. Nogami, Few–Body Syst. 13, 75 (1992).
[28] R. B. Firestone, 8th ed. *Table of isotopes* (John Wiley and Sons, New Yours, 1996).
[29] H. J. Fischbeck and M. S. Freedman, Phys. Rev. Lett. 34, 173 (1975).