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DOUBLE IONIZATION OF HELIUM BY RELATIVISTIC HIGHLY CHARGED ION IMPACT

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We consider an interesting realization of the fundamental four-body problem: double ionization of helium in superintense electromagnetic fields generated by highly charged ions in relativistic collisions. We show how the simultaneous interaction of such fields with all the three target constituents (which is not described by a first-order theory) strongly influences the collision dynamics even at very high collision energies and how the «genuine» photo-like emission pattern may emerge in collisions at extreme relativistic energies. A very good agreement with available experimental data is found.

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The question of the dynamics of quantum mechanical few-particle systems on various time scales is among the most interesting topics in modern atomic, molecular, and optical physics [1]. One of the fundamental examples of the quantum few-body problem is given by ionization of helium in collisions with fast highly charged ions. During the last decade, there has been remarkable progress in this field [1, 2]. Most of the studies of the helium ionization, however, were performed for single ionization and for nonrelativistic collision velocities.

Whereas single ionization of helium is normally treated as a three-body problem (projectile, «active» electron, and recoil ion), double ionization represents a particularly strong challenge for the theory because it is a pure four-body problem. Indeed, a satisfactory (but still incomplete) understanding of helium double ionization by charged projectiles has been only reached for collisions with fast enough electrons where the first Born approximation (FBA) in the projectile–target interaction is valid. Helium double ionization by highly charged ions is more difficult to describe, and it has attracted much less attention so far. In particular, helium double ionization by relativistic ions with such a high charge Z_p that $Z_p/v_p \sim 1$ even for collision velocities v_p approaching the speed of light c ($c \approx 137$ a.u.) has remained a *terra incognita* to a large extent.

First measurements of differential cross sections for double ionization of helium in relativistic collisions (1 GeV/u U^{92+}, $v_p = 120$ a.u., $\gamma = (1 - v_p^2/c^2)^{-1/2} \approx 2$, and $Z_p/v_p \approx 0.77$) were done in [3]. Detailed experimental studies of helium ionization by highly charged ions in collisions at $\gamma \approx 1.5$–2 are scheduled for 2005 (GSI, Germany) and collision energies up to those corresponding to $\gamma \approx 30$ will become routinely accessible for atomic physics experiments in the near future [4].

Relativistic collisions with ions like U^{92+} may expose helium atoms to extreme conditions. Indeed, rough estimates show that electromagnetic pulses with effective power densities as high as 10^{19} to 10^{23} W/cm^2 can be generated by relativistic highly charged ions in collisions at $\gamma \sim 10$–30 for impact parameters between 2 and 10 a.u. such that the whole target atom is exposed to a nearly homogeneous field. Besides, such pulses are ultrashort and, despite the enormous intensities, may «gently» irradiate the target, making its «snap-shots» on the subatomic time scale.

Only a few attempts have been made to evaluate differential cross sections for double ionization of helium in relativistic collisions with highly charged ions. The estimates in [3] and [5] were based on the Weizsäcker–Williams method of equivalent photons. However, for collisions with light targets, strictly speaking, this method may be applied only at extreme relativistic energies [6]. Besides, the results in [5] were obtained only for a fixed collision impact parameter and cannot there-

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fore be related to experiment. In [7], helium ionization was treated using the classical-trajectory Monte Carlo approach. But the cross sections reported in [7] were too small because this approach fails to properly describe collisions with relatively small momentum transfers, which become of great importance at very high impact energies.

In this paper, we consider helium double ionization in relativistic collisions with very highly charged ions by developing an approach that, for the first time, enables a detailed description of this extraordinary case of the four-body quantum dynamical problem.

We start with the following remarks. First, even in collisions with relativistic projectiles, the overwhelming majority of electrons emitted from light targets have nonrelativistic energies[1]. Therefore, we consider helium ionization in the target frame and use a nonrelativistic description for the electron motion. Second, because the momentum exchange does not actually exceed several atomic units in collisions of interest for the present study, the recoil velocity of the target nucleus and the deflection angle of the projectile are always very small. This allows us to begin the consideration with the semiclassical picture in which (i) only the electrons are treated quantum mechanically; (ii) the target nucleus is assumed to be at rest and is taken as the origin of the target frame; (iii) in this frame, the projectile moves along a straight-line classical trajectory $\mathbf{R}(t) = \mathbf{b} + \mathbf{v}_p t$, where $\mathbf{b}$ is the impact parameter. The corresponding Schrödinger equation is

$$i\frac{\partial\Psi}{\partial t} = \left\{\sum_{j=1}^{2}\left[\frac{1}{2}\left(\hat{\mathbf{p}}_j+\frac{\mathbf{A}_j}{c}\right)^2-\varphi_j\right]+V_{at}+Z_t\varphi\right\}\Psi. \quad (1)$$

Here, $\hat{\mathbf{p}}_j$ is the momentum operator for the jth atomic electron, φ_j and $\mathbf{A}_j$ are the scalar and vector potentials of the projectile field at the position of the jth atomic electron, and φ is the corresponding scalar potential at the origin. Furthermore, $Z_t = 2$ is the charge of the target nucleus and $V_{at} = -Z_t/r_1 - Z_t/r_2 + 1/r_{12}$ is the interaction between the target particles, where $\mathbf{r}_j$ is the coordinate of the jth electron with respect to the target nucleus and $\mathbf{r}_{12} = \mathbf{r}_1 - \mathbf{r}_2$. The spin-flip transitions are suppressed in our case by a factor $\sim v_p/c^2$ compared to the non-spin-flip ones, and the spin terms are therefore ignored in Eq. (1).

[1] Actually, in the target frame, the energies of most emitted electrons do not exceed few atomic units (see Fig. 2).

In the Lorentz gauge, the projectile potentials are given by [8]

$$\varphi_j = \frac{\gamma Z_p}{s_j}, \quad \varphi = \frac{\gamma Z_p}{s}, \quad \mathbf{A}_j = \frac{\mathbf{v}_p}{c}\varphi_j, \quad (2)$$

where $\mathbf{s}_j$ and $\mathbf{s}$ are the coordinates of the jth target electron and the target nucleus with respect to the projectile ion given in the projectile rest frame.

Taking into account that in both the initial and final channels the projectile velocity is much higher than typical electron velocities (1–3 a.u.), we use the symmetric eikonal approximation (SEA). In the SEA, the state Ψ is replaced by Ψ_i and Ψ_f in the initial and final channels respectively, where

$$\begin{aligned}&\Psi_i(t) = \psi_i(\mathbf{r}_1,\mathbf{r}_2)\exp(-i\varepsilon_i t)(vs+\mathbf{v}\cdot\mathbf{s})^{i\eta_p}\times\\&\times(vs_1+\mathbf{v}\cdot\mathbf{s}_1)^{-i\nu_p}(vs_2+\mathbf{v}\cdot\mathbf{s}_2)^{-i\nu_p},\\&\Psi_f(t) = \psi_f(\mathbf{r}_1,\mathbf{r}_2)\exp(-i\varepsilon_f t)(vs-\mathbf{v}\cdot\mathbf{s})^{-i\eta_p}\times\\&\times(vs_1-\mathbf{v}\cdot\mathbf{s}_1)^{i\nu_p}(vs_2-\mathbf{v}\cdot\mathbf{s}_2)^{i\nu_p},\end{aligned} \quad (3)$$

ψ_i and ψ_f are the initial and final states of the three-body target subsystem with energies ε_i, and ε_f, $\nu_p = Z_p/v_p$, and $\eta_p = Z_p Z_t/v_p$. We note that in Eq. (3), the Coulomb boundary conditions (due to the projectile field) are satisfied for all the three target particles.

Within the SEA, the prior form of the semiclassical transition amplitude is

$$a_{fi}(\mathbf{b}) = -i\int_{-\infty}^{\infty} dt\langle\Psi_f(t)|\hat{W}(t)|\Psi_i(t)\rangle, \quad (4)$$

where the distortion interaction $\hat{W}(t)$ is given by

$$\begin{aligned}\hat{W}\Psi_i = (vs_1-\mathbf{v}\cdot\mathbf{s}_1)^{-i\nu_p}(vs_2-\mathbf{v}\cdot\mathbf{s}_2)^{-i\nu_p}(vs-\mathbf{v}\cdot\mathbf{s})^{i\eta_p}\times\\ \times\,\nu_p\exp(-i\varepsilon_i t)\sum_{j=1}^{2}\left(\mathbf{C}_j\cdot\hat{\mathbf{p}}_j+\nu_p D_j\right)\psi_i\end{aligned} \quad (5)$$

with

$$\begin{aligned}&\mathbf{C}_j=-s_j^{-1}\left(s_{j,x}(s_j+s_{j,z})^{-1}; s_{j,y}(s_j+s_{j,z})^{-1}; \gamma^{-1}\right),\\&D_j = (s_j(s_j+s_{j,z}))^{-1} - 0.5v_p^2(cs_j)^{-2},\end{aligned} \quad (6)$$

where $s_{j,z} = \mathbf{s}_j\cdot\mathbf{v}_p/v_p$ and $(s_{j,x}; s_{j,y}) = \mathbf{s}_j - s_{j,z}\mathbf{v}_p/v_p$.

The full quantum dynamics of the collision cannot be treated with the semiclassical amplitude given by Eq. (4). However, for collisions with very small projectile scattering angles and negligible velocities of the target nucleus, the quantum transition amplitude S_{fi} can be obtained from the semiclassical amplitude in (4) as

$$S_{fi}(\mathbf{Q}) = \frac{1}{2\pi}\int d^2b\exp(i\mathbf{Q}\cdot\mathbf{b})a_{fi}(\mathbf{b}), \quad (7)$$

where $\mathbf{Q}$ is the two-dimensional transverse part ($\mathbf{Q}\cdot\mathbf{v}_p = 0$) of the momentum transfer $\mathbf{q}$ to the target. In contrast to the impact parameter $\mathbf{b}$, the momentum transfer is accessible to direct measurement. We have $\mathbf{q} = (\mathbf{Q}; q_{min})$, where $q_{min} = \omega_{fi}/v_p$ with $\omega_{fi} = \varepsilon_f - \varepsilon_i$.

Amplitude (7) is the first term of the symmetric eikonal distorted wave series. The analysis shows that for the most important part of the emission, the expansion parameter of this series is essentially given by $\varsigma = Z_p/v_p^2$. In relativistic collisions, ς does not exceed 0.01 even for the highest possible projectile charge states $Z_p \sim v_p$. Therefore, the first term of this series alone may already be sufficient for a successful treatment of the collision dynamics. This is to be contrasted with the standard Born series, which is generated from Eq. (1) in the usual way and has the expansion parameter $\nu_p = Z_p/v_p$. In collisions with the heaviest bare nuclei, the parameter ν_p is never much less than unity. Therefore, not only might the first Born approximation be insufficient[2)] but also the whole Born series is likely to become meaningless.

The success of distorted wave models for nonrelativistic ion–atom collisions was to a very large extent caused by the facts that (i) the interaction between the projectile and the target nucleus (the n–n interaction) does not affect the electron emission spectra integrated over the projectile deflection angle and (ii) for collisions with hydrogen-like targets, the transition amplitude of type (7) can be evaluated analytically provided the n–n interaction is ignored. The account of the interaction between the projectile and the second «active» target electron tremendously complicates calculations, and the situation is certainly not simplified if the n–n interaction must also be included, for instance, in the case where the full collision dynamics has to be considered.

At $\nu_p \sim 1$, the direct numerical integration of the multiple integral in Eq. (7) faces difficulties because in both the initial and final channels, the motion of the projectile is not bounded in space. Therefore, the integral over $d^3R = d^2b\, v_p\, dt$ in Eq. (7) is not absolutely convergent and should be taken analytically. The result is

$$S_{fi}(\mathbf{Q}) = \frac{i\nu_p}{2\pi v_p \gamma} \times \\ \times \int d^2\boldsymbol{\zeta} d^2\boldsymbol{\xi} \langle \psi_f | \mathbf{G}_1 \cdot \hat{\mathbf{p}}_1 + \mathbf{G}_2 \cdot \hat{\mathbf{p}}_2 + F_1 + F_2 | \psi_i \rangle. \quad (8)$$

[2)] In collisions at very high γ, where very small momentum transfers contribute most to the double ionization, even for $\nu_p \sim 1$, a properly formulated first-order approach may be applied to the total cross section for the double ionization.

Here, $\boldsymbol{\zeta}$ and $\boldsymbol{\xi}$ are two-dimensional vectors perpendicular to $\mathbf{v}_p$,

$$\mathbf{G}_1 = \mathbf{G}(\nu_p, \eta_p; \gamma; \boldsymbol{\zeta}, \boldsymbol{\xi}, \mathbf{q}; \mathbf{r}_1, \mathbf{r}_2),$$

$$F_1 = F(\nu_p, \eta_p; \gamma; \boldsymbol{\zeta}, \boldsymbol{\xi}, \mathbf{q}; \mathbf{r}_1, \mathbf{r}_2),$$

$\mathbf{G}_2 = \mathbf{G}_1(\mathbf{r}_1 \leftrightarrow \mathbf{r}_2)$, and $F_2 = F_1(\mathbf{r}_1 \leftrightarrow \mathbf{r}_2)$, where $\mathbf{G}$ and F are expressions containing the exponential, gamma, and hypergeometric functions. The explicit forms of $\mathbf{G}$ and F are very cumbersome and will be given elsewhere.

We note that the right-hand side of Eq. (5) was written with ψ_i assumed to be an exact state of the free target. If this is not the case, an additional term appears in the right-hand side of Eq. (5). But if $\varepsilon_f \neq \varepsilon_i$, this term gives zero contribution to the transition amplitude. Therefore, there are no formal restrictions imposed on ψ_i and ψ_f by the use of Eq. (8). Because the three-body problem has no exact solution, the actual choice of ψ_i and ψ_f is dictated by two main points: these states should be «sufficiently good» and, simultaneously, allow performing at least the ten-fold integration in Eq. (8) necessary to obtain the fully differential cross section

$$\frac{d\sigma}{d^2Q\, d^3k_1 d^3k_2} = |S_{fi}|^2, \quad (9)$$

where $\mathbf{k}_1$ and $\mathbf{k}_2$ are the electron momenta in the final state.

As was already remarked, the SEA is superior to the FBA at $\nu_p = Z_p/v_p \sim 1$. One would expect that as $\nu_p \to 0$, the results of both approximations converge if the exact target states ψ_i and ψ_f can be used. But even with such states, the ultrarelativistic limits of these two approaches are still different: the symmetric eikonal approximation yields the correct asymptotic behavior for cross sections as $\gamma \to \infty$, but the first Born approximation does not. This point is very important and deserves a separate and detailed discussion. Here, we only note that at $\nu_p \ll 1$, the first Born approximation with exact target states would strongly fail only at $\gamma \sim c/\nu_p^2$ and higher.

The results of both the SEA and FBA using approximate target states ψ_i and ψ_f do not coincide even as $\nu_p \to 0$. Therefore, a consistent way to «highlight» higher-order effects in the projectile–target interaction is as follows. For a given $v_p \sim c$, calculations in the SEA are performed for the actual projectile (Z_p) and for the proton impact. The first-order result for the actual projectile is then obtained from that for the proton using the first-order scaling, i.e., via multiplication with Z_p^2. We call this first-order approach the SEA-1.

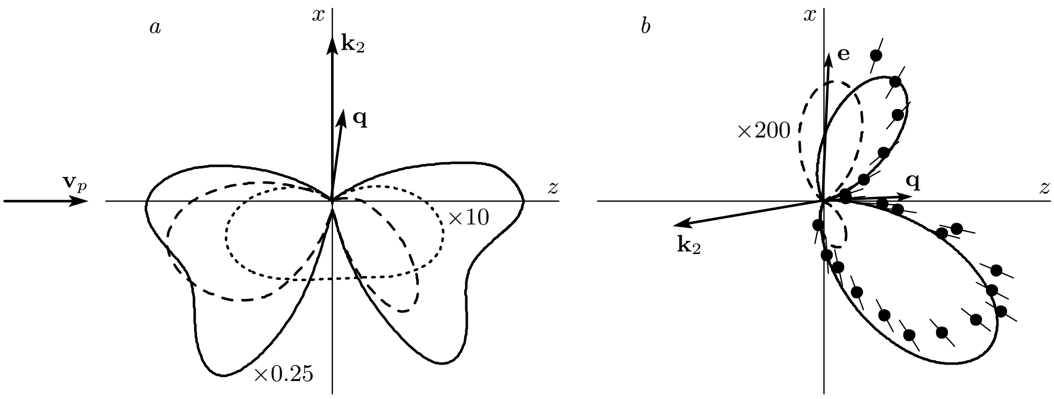

Fig. 1. The fully differential cross section (in arb. units) as a function of the polar emission angle ϑ_1 of the «first» electron, given in the plane defined by $\mathbf{v}_p = (0, 0, v_p)$ and $\mathbf{q} = (Q, 0, q_{min})$. Emission energies $E_1 = E_2 = 10$ eV, azimuthal emission angles $\varphi_1 = \varphi_2 = 0°$. *a*) $v_p = 120$ a.u., $Q = 0.25$ a.u., $\vartheta_2 = 90°$. The solid curve corresponds to the SEA, the dashed curve to the SEA without the n–n interaction, the doted curve to the SEA-1. *b*) $v_p \approx 137$ a.u. ($\gamma = 26$), $Q = 10^{-3}$ a.u., $\vartheta_2 = 192°$. The solid curve corresponds to the SEA, the dashed curve to the nonrelativistic SEA ($c = \infty$), the symbols are the experimental data on double photoionization [9] (the incident real photon is polarized along the x axis) normalized to the SEA results

In calculations of the fully differential cross section given by Eq. (9), we approximate the initial state by the four-parameter Hylleraas wave function

$$\psi_i = N_i \left[\exp(-\alpha r_1 - \beta r_2) + (r_1 \leftrightarrow r_2)\right] \times \times \left[1 - \delta \exp(-\lambda r_{12})\right], \quad (10)$$

where $N_i = 1.638$ is the normalization factor, $\alpha = 1.4096$, $\beta = 2.2058$, $\delta = 0.6054$, and $\lambda = 0.2420$. Wave function (10) yields $\varepsilon_i = -2.902$ a.u., which is close to the exact value of -2.904 a.u. The final state is taken as

$$\psi_f = \psi_{3C} - \langle \psi_i | \psi_{3C} \rangle \psi_i,$$

$$\psi_{3C} = \frac{1}{\sqrt{2}} \left[\psi_{\mathbf{k}_1}(\mathbf{r}_1)\psi_{\mathbf{k}_2}(\mathbf{r}_2)\chi_{\mathbf{k}_{12}}(\mathbf{r}_{12}) + + (\mathbf{r}_1 \leftrightarrow \mathbf{r}_2)\right], \quad (11)$$

where $\mathbf{k}_{12} = (\mathbf{k}_1 - \mathbf{k}_2)/2$ and ψ_{3C} is the so-called $3C$ state, a (symmetrized) product of three Coulomb waves describing all pairwise interactions between the constituents of the target. The above approximations are chosen because they yield good results for helium double ionization due to the photoeffect and by fast electrons in collisions with relatively small momentum transfers. Such collisions become especially important at relativistic impact energies. In addition, with the states in Eqs. (10) and (11), the six-fold integrals over the electron coordinates in Eq. (8) can be reduced to two-fold integrals.

The results for the fully differential cross section in collisions with U^{92+} are shown in Fig. 1. Two important points should be mentioned.

First, within any first-order approach, the projectile may exchange only a single virtual photon with the target and can therefore directly interact with just one electron. Double ionization may then only occur due to electron–electron correlations and/or rearrangement in the target final state. However, the highly charged projectile, due to its strong field, can directly and very effectively interact with all the three target particles simultaneously. Therefore, such (higher-order) effects in the projectile–target interaction, which are properly described within the SEA, may profoundly influence the collision dynamics (Fig. 1*a*). Not only the direct interaction of the projectile with both electrons but also the n–n interaction (which itself does not lead to ionization) may very strongly affect the fully differential emission pattern.

Second, in collisions at very high γ and very low Q, the higher-order effects become of minor importance even at $\nu_p \sim 1$. A very interesting peculiarity of such collisions is that the physics of the impact ionization may become very similar to that of the photoeffect. A certain similarity between impact ionization and photoionization has been the subject of the long-term discussion in studies of the double ionization by fast nonrelativistic electrons. Such discussions, however, are of superficial character and can even be misleading because the fundamental similarity between these processes is only possible if $\gamma \gg 1$. Indeed, the emission pattern in Fig. 1*b* is almost indistinguishable from that due to the photoeffect because it is produced by the

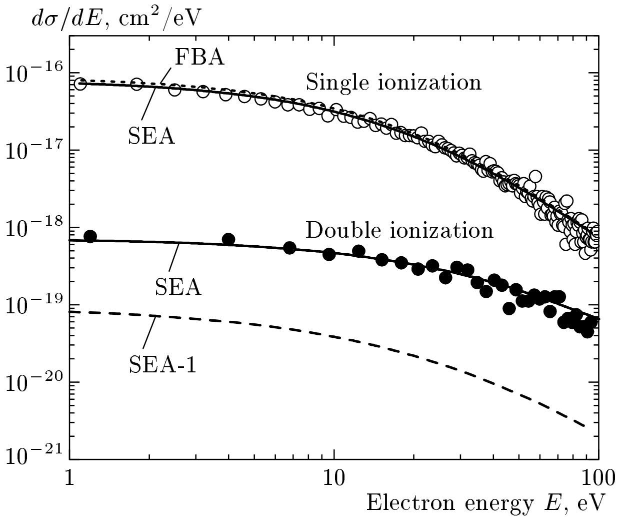

Fig. 2. Energy spectra of electrons emitted in 1-GeV/u U^{92+} + He(1s^2) collisions. Symbols are the experimental data from [3]. See the text for more explanations

absorption of a virtual photon whose properties are very close to those of a real photon [6]. As a result, not the virtual photon momentum $\mathbf{q}$ but its polarization $\mathbf{e} \sim \mathbf{q}/\omega_{fi} - \mathbf{v}_p/c^2$ [6], almost perpendicular to $\mathbf{q}$, determines the shape of the emission cross section in Fig. 1*b*.

In [3], the emission spectrum differential in the energy of one of the ejected electrons has been reported. To produce such a spectrum from the cross section in (9), one has to perform seven additional integrations. This task is not feasible if ψ_i and ψ_f are given by Eqs. (10) and (11), but can be carried out if the terms depending on $\mathbf{r}_{12}$ are neglected in these equation, which allows evaluating the integrals over the electron coordinates in Eq. (8) analytically. Of course, the neglect of the electron correlation would be a very improper approximation in the study of the fully differential cross section given by Eq. (9). Nevertheless, it is known that for collisions with highly charged ions, this approximation can still be used to estimate the total cross section and the energy emission spectrum integrated over the momentum transfer and all emission angles. The basic reasons for this are twofold. First, the double ionization in our case is dominated by the so-called TS-2 process, in which the electrons undergo transitions due to the «direct» interaction between the projectile and each of the two electrons. Second, while the electron–electron interaction in the continuum can strongly affect angular distributions, it cannot change the total energy of the electrons.

The results of such calculations ($\alpha = 1.885$, $\beta = 2.1832$, and $\delta = 0 \Rightarrow \varepsilon_i = -2.876$ a.u. and $\chi_{\mathbf{k}_{12}} = 1$) are shown in Fig. 2. For completeness, the energy spectrum of electrons emitted in singly ionizing collisions is also displayed[3)]. For both single and double ionization, a very good agreement between the SEA results and experimental data is observed[4)]. The overall effect of the higher-order terms in the projectile–target interaction is clearly seen in Fig. 2: it only slightly decreases the single ionization cross section but is very strong for the double ionization. Compared to the first-order result, the energy spectrum for double ionization decreases substantially slower as the emission energy increases and is larger on an absolute scale by a factor of 10–30 due to the large contribution from collisions in which both target electrons are removed simultaneously by their «independent» interactions with the projectile.

In conclusion, using a novel approach that treats, within the SEA, the interaction of the projectile with all the three target constituents on an equal footing, we have considered the double ionization of helium in relativistic collisions with highly charged ions. By exploring the basic dynamics of these collisions for the first time, we have demonstrated how the direct interaction of the projectile with all the three target particles can strongly affect the fully differential cross section. We have further shown that the fundamental similarity between the impact double ionization and double photoionization of helium naturally emerges in extreme relativistic collisions with very small transverse momentum transfers.

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3) Results for this spectrum were obtained by combining the SEA and FBA with the Hartree – Fock description of the «active» electron. We note that within the effective three-body collision model, where the (active) electron moves in the same Hartree – Fock potential in both initial and final states, the results of SEA and FBA nicely converge at $\nu_p \ll 1$.

4) We note that for ionization by 1-GeV protons, the SEA, with ψ_i and ψ_f used to produce the spectrum in Fig. 2, yields the double-to-single ionization ratio $\sigma^{2+}/\sigma^{+} \approx 2 \cdot 10^{-3}$, which is quite close to the established high-velocity limit for this ratio approximately equal to $2.5 \cdot 10^{-3}$ (see, e.g., [10]).

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