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Ionization probability for multiple ionization: An assessment of the geometrical model

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Abstract

As X-ray spectra of multiply ionized atoms are resolved with higher resolution, the number of vacancies in outer shells—used to deduce the probability of ionization at the zero impact parameter $P[0]$—can be determined with increasing accuracy. The $P[0]$ so extracted, or served as input in X-ray data analyses, has been for over two decades and almost universally compared with, or entered from, the predictions of the geometrical model of Sulik et al. [1984. Charge scaling of ionization probabilities in ion-atom collisions for zero impact parameter. J. Phys. B 17, 3239–3244] which is grounded on the binary encounter approximation (BEA). The validity of $P[0]$ employed in the geometrical model is tested with the straight-line semiclassical approximation (SCA) at $p = 0$. It is shown that the $P[0]$ of Sulik et al. is reasonably close to the semiclassical $P[0]$ when the projectile velocity v_1 is in the neighborhood of the velocity of an S-shell electron v_{2S}, but the geometrical model increasingly and significantly under- and overestimates the SCA values when, respectively, $v_1/v_{2S}<1$ and $v_1/v_{2S}>1$. This is consistent with breakdown of the geometrical model as indicated by the recent experiment of Horvat et al. [2005. L X-rays emitted from multiply ionized holmium atoms. Phys. Rev. A 71, 062709].

Keywords: Ionization probability; Multiple ionization; Geometrical model; BEA; SCA

1. Introduction

For over three decades, the X-ray spectra of atoms multiply ionized in their outer shells have been untangled with recipes that call for probabilities of ionization of an electron from a shell that is inner with respect to these outer shells (McGuire and Richard, 1973; Richard 1975 with references therein). As the probabilities of outer shell ionization in a given atom tend to be constant over the range of more compact inner shells, an accurate knowledge of $P[0]$—the ionization probability of the inner-shell at the zero impact parameter—generally suffices in such analysis. Tables of $P[p]$ in K-, L-, and M-shell ionization calculated three decades ago in the straight-line semiclassical approximation (SCA) (Hansteen et al., 1975) could have been extrapolated to $p = 0$. Yet, since the early SCA calculations of Hansteen and Mosebekk (1972) were thought to be inferior to the results of the binary encounter approximation (BEA) and the experiments that were then available (see, for example, Li et al., 1973), the BEA became fashionable and gradually relegated the SCA to an occasional consideration in studies of multiple ionization.

The relative ease with which ionization cross section can be evaluated in the BEA, and the idea that BEA cross sections divided by the geometrical size of an inner-shell gave a reliable measure of $P[0]$, led McGuire and Richard (1973) to close their abstract with "[m]ultiple-ionization cross sections may thus be estimated without the aid of a computer." Despite the phenomenal progress in computer power in the intervening years, estimates of ionization probabilities for analysis of multiple ionization and its ramifications for correction of single-hole fluorescence yields are still mostly grounded on BEA approaches. In particular, Sulik et al. (1984) proposed the so-called geometrical model and—as presented by Sulik and Hock (1984) at the 2nd High-Energy Ion-Atom Collisions Workshop in Debrecen—furnished it with the specific

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formulas for $P[0]$ in K-, L-, and M-shells. After appropriate corrections, for collisions above 1 MeV/u, these formulas appeared to agree with coupled-channel calculations of Becker et al. (1984).

Over the years, a simple formula developed by Sulik et al. (1987)—in terms of the scaling variable of the BEA-inspired geometrical model for $P[0]$—has won over the SCA approach. Yet, a review of papers—where the geometrical model is compared with SCA calculations (see Refs.13,51,54,56,58–60 in a recent article by Lapicki et al., 2005)—suggests that $P[0]$ of that model falls below and above the SCA predictions when, respectively, the projectile velocity v_1 is less or greater than the orbital velocity v_{2S} of the electron ionized from an S shell. With the progress in the analysis of high-resolution X-ray spectra, it became apparent (see Fig. 5 in Horvat et al., 2005) that the experimentally determined fraction of spectator vacancies in collisions with heavy ions saturates by a factor of 2 below what is calculated by the geometrical model of Sulik et al. (1987). This paper focuses on an assessment of the BEA-driven formulas as gauged by the $P[0]$ of the SCA in its simplest straight-line incarnation of Hansteen et al. (1975).

2. Ionization cross section and *P*[*p*] in the binary encounter approximation

Subsequent to the development of the quantum mechanical plane-wave Born approximation (PWBA, Merzbacher and Lewis, 1958) and the SCA (Bang and Hansteen, 1959), as reviewed by Lapicki (1988), Gryziński (1965a–c) revived the classical approach of Thomson (1912) of the BEA in which the ionization cross-section of an S-shell electron by an ion of atomic number Z_1 and velocity v_1 could be calculated in a closed form

$$\sigma^{\mathrm{BEA}} = Z_1^2 4\pi N a_{2S}^2 V^2 G(V)/v_1^2. \tag{1}$$

N is the number of S-shell electrons and G, a function of $V \equiv v_1/v_{2S}\theta_{2S}^{1/2}$, is usually evaluated with the screened ($Z_{2S} = Z_2 - s$ with s being Slater's screening constant) hydrogenic wavefunctions. With these wavefunctions, the S-shell radius $a_{2S} = n^2/Z_{2S}$, the orbital velocity of its electron $v_{2S} = Z_{2S}/n$, and the eigenenergy $\frac{1}{2}(Z_{2S}/n)^2\theta_{2S}$ is set to equal the observed binding energy; θ_{2S} is a ratio of that energy to the binding energy in the screened hydrogenic atom. A careful analysis of the kinematics of the binary encounter between the projectile and an electron of velocity v_2 and the cross section for energy transfer in such a collision, as encoded by Gerjuoy (1966), Vriens (1967) and Garcia et al. (1968, 1970, 1971), led after its average over the velocity distribution of the S-shell electron to a tabular list of $G(V)$ that was reported by McGuire and Richard (1973). For the derivation of $P[p]$, other than a simplistic $P[0] = \sigma^{BEA}/\pi N a_{2S}^2$, the $v_2(r)$-dependent binary encounter cross-sections of Vriens had to be integrated along the z axis of the incoming and outgoing projectile with a *spatial* distribution $\rho(r)$ of the S-shell electron and subject to the condition that the impact parameter $p < (r^2 - z^2)^{1/2}$. By imposing conservation of energy on an electron locally at a radial distance r, McGuire and Richard (1973) limited the available collisions to the electron within the S-shell diameter i.e., $v_2(r) = v_{2S}(2a_{2S}/r - \theta_{2S})^{1/2}$ and $v_2(r) = 0$ for $r > 2a_{2S}$. Although the elimination of the electron from the periphery of the S-shell was hardly critical for a good estimate of $P[0]$, in deriving $P[p]$ we rely on a physically appealing condition set up by Hansen (1973): $v_2(r)$ ought to be such that the velocity electron distribution integrated from this $v_2(r)$ to infinity must be equal to the spatial electron distribution integrated from zero to r. To facilitate further calculations of $P[p]$, for a hydrogenic full-shell electron, we found that an analytical formula

$$v_2(r) = \left(\frac{24}{5\pi r^3}\right)^{1/5} \mathrm{e}^{-2r/3}\left(1 + 0.6r - 0.5r^{1.2} + \left(\frac{5\pi}{24}\right)^{1/5}\left(\frac{3\pi}{32}\right)^{1/3} r^{19/15}\right) \tag{2}$$

has the proper limiting behavior for small and large r values, and satisfies Hansen's condition to within 1% for all r. With it and Vriens' (1967) cross sections for the energy transfer in a binary encounter, we have derived $P^{\mathrm{Vriens}}[p]$ from which $\sigma^{\mathrm{Vriens}} = 2\pi \int P^{Vriens}[p] p\,\mathrm{d}p$ and $P^{\mathrm{Vriens}}[0]$ were obtained.

Also, calculated is what Hansen (1973) called constrained-BEA arising from the requirement that $P[p]$ should not be greater than 1. Hansen enforced this requirement by $P^{Hansen}[p] = 1 - [1 - P'[p]]^N$ with $P'[p] = 1 - \exp[-P[p]/N]$ where $P'[p]$ is the impact-ionization probability per electron. For the $P[p]$ that enters into this prescription, we used $P^{\mathrm{Vriens}}[p]$. Note that when $P'[p] \ll 1$, as it occurs in asymmetric ($Z_1 \ll Z_2$) collisions, $P^{\mathrm{Hansen}}[p] \approx P^{\mathrm{Vriens}}[p]$.

In our figures, $P^{\mathrm{Sulik}}[p]$ is displayed as a step function: $P^{\mathrm{Sulik}}[0]$ from the works of Sulik et al. (1984, 1987) for $p < p_{\mathrm{c}}$ and 0 otherwise where p_{c} is such that $2\pi_{\mathrm{o}}^{\mathrm{pc}} \int P^{\mathrm{Vriens}}[p] p\,\mathrm{d}p = \sigma^{\mathrm{Vriens}}$. Since only $P^{\mathrm{Sulik}}[p]$ at $p = 0$ is germane for its assessment vis a vis $P^{\mathrm{SCA}}[0]$ and data to be utilized or obtained in studies of multiple ionization, this graphical representation of $P^{\mathrm{Sulik}}[p]$ emphasizes its value at $p = 0$ and is immaterial for $p > 0$.

3. BEA cross sections and BEA *P*[0] vs. SCA cross sections and SCA *P*[0] for *K*-shell ionization

Although BEA and SCA impact-parameter ionization probabilities $P[p]$ and related cross sections are given by subshell specific formulas, we assume that the comparison of these approximations should exhibit similar trends as for the full S-shell or for the K-shell, which has the same scaled spatial distribution of an electron in any full S-shell.

3.1. Asymmetric ($Z_1 \ll Z_2$) collisions

BEA and SCA cross sections for ionization of copper by protons appear in Fig. 1 to be in very good agreement over a wide range of proton energies. As indicated by their near overlap with a theory that goes beyond the first-order scattering theory e.g., the ECPSSR theory (Brandt and Lapicki, 1981)—that accounts for the projectile's energy loss and Coulomb deflection as well as for a perturbed stationary-state and relativistic character of the target's *S*-shell—these first-order theories could be sensibly compared with experiments once the energy of the protons exceeds 1 MeV. In Fig. 2, the BEA, SCA, and ECPSSR cross sections are divided by the results of the first Born PWBA (Merzbacher and Lewis, 1958) that in principle should be equal to the straight-line SCA cross sections. [The observed

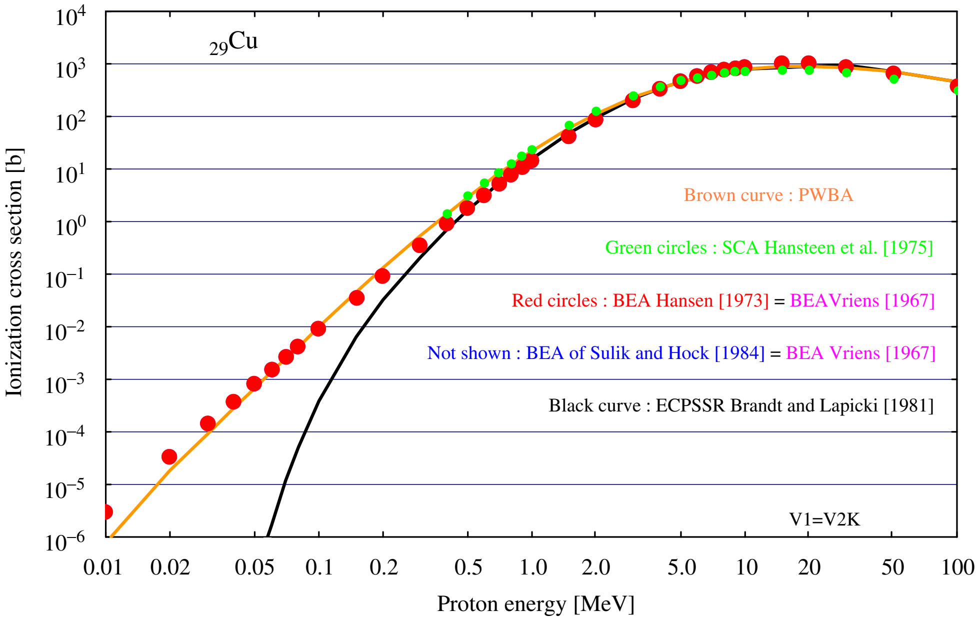

Fig. 1. Cross sections for *K*-shell ionization of copper by protons according to the PWBA, SCA, BEA, and ECPSSR theories as labeled. The PWBA (light brown curve), SCA (small green circles), and BEA (large red circles) are satisfactory as first-order theories to the extent that they are close to the ECPSSR (black curve) when proton energies exceed 1 MeV.

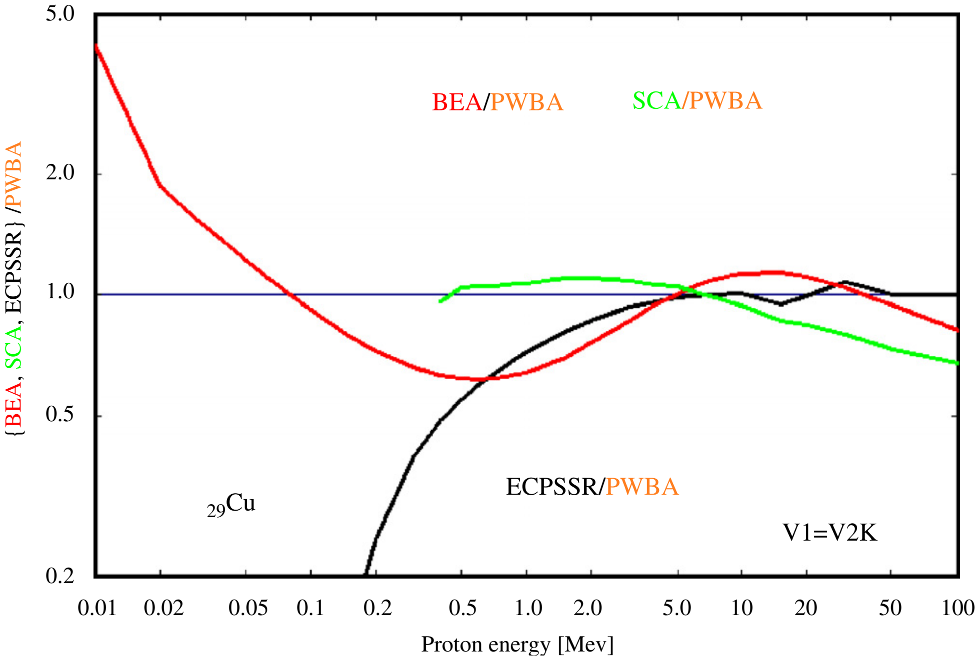

Fig. 2. Ratios of cross sections for *K*-shell ionization of copper by protons according to the SCA, BEA, and ECPSSR theories to the predictions of the PWBA (Merzbacher and Lewis, 1958).

differences between these theories (see light green curve in Fig. 2) are explainable to a large degree by uncertainties of as much as 5% in the SCA tables of Hansteen et al., 1975.] Fig. 2 demonstrates that the apparent agreement between BEA and SCA cross sections is limited; this agreement remains good only for a few-MeV protons. It is in this range that differences among the $P^{\mathrm{BEA}}[0]$, $P^{\mathrm{Sulik}}[0]$ and $P^{\mathrm{SCA}}[0]$ should be primarily attributed to the accuracy of the Vriens' and Sulik's BEA versus SCA results for the $P[0]$—that are critical for multiple-ionization analyses—rather than to their shortcomings in the evaluation of cross sections. While the 124 barn cross section measurement of Leagsgaard et al. (1972) for K-shell ionization of copper by 2-MeV protons is excellently reproduced by the 126 barn calculated in the SCA and is in a reasonable 30% proximity to the 87 barn obtained in the BEA, Fig. 3 demonstrates that $P^{\mathrm{Sulik}}[0]$ is about a factor of 4 below what the data indicate, and a factor of 3 below $P^{\mathrm{SCA}}[0]$. For K-shell ionization of $_{29}$Cu by 1-MeV protons, this comparison is even worse: the $P^{\mathrm{Sulik}}[0]$ is about a factor of 7 below $P^{\mathrm{SCA}}[0]$. Similar findings obtain for the K-shell impact-parameter ionization of $_{34}$Se by 2-MeV protons data of Leagsgaard et al. (1972). As seen in Fig. 4, $P^{\mathrm{Sulik}}[0]$ falls almost a factor of 8 below $P^{\mathrm{SCA}}[0]$ and an extrapolation of the Leagsgaard et al. (1972) data for K-shell ionization of $_{47}$Ag by 2-MeV protons to $p = 0$ shows that at the zero impact parameter $P^{\mathrm{Sulik}}[0]$ would underestimate them by an order of magnitude.

For no apparent reason, the $P^{\mathrm{Sulik}}[0]/P^{\mathrm{SCA}}[0]$ ratio correlates well with the v_1/v_{2K} ratio: $P^{\mathrm{Sulik}}[0]/P^{\mathrm{SCA}}[0] = 0.30$ at $v_1/v_{2K} = 0.30$ (Fig. 3), 0.22 at 0.26 for selenium ionization by 2-MeV protons, 0.15 at 0.21 for copper ionization by 1-MeV protons, and 0.13 at $v_1/v_{2K} = 0.19$ for silver ionization by 2-MeV protons (Fig. 4). To within 10%, $P^{\mathrm{Sulik}}[0]/P^{\mathrm{SCA}}[0] \approx 3.3\ (v_1/v_{2K})^2$ for $v_1/v_{2K} < 0.3$ – $P^{\mathrm{Sulik}}[0]/P^{\mathrm{SCA}}[0]$ *definitely* decreases with the decreasing v_1/v_{2K}, and appears to be less than 1 when $v_1/v_{2K} < 1$.

3.2. Symmetric ($Z_1 \approx Z_2$) collisions

Effects of multiple ionization become significant when $P[0]$ exceeds 0.1 as they do in heavy-ion collisions that fall into a class of symmetric or nearly symmetric collisions. Ionization of H by protons serves as a prototype of such collisions; the prototype that is free—at the level of the first-order perturbation at which the BEA and SCA results are compared here—from ambiguities about the nature of the electron wavefunction.

As shown in Fig. 5, the BEA cross sections are in remarkably good agreement with the PWBA and SCA below $v_1/v_{2K} = 1$. Unfortunately, as indicated by their growing difference with the ECPSSR theory and the data (Shah and Gilbody, 1981; Shah et al., 1987, 1988; Rudd et al., 1992; Pieksma et al., 1994), as v_1/v_{2K} decreases one cannot use these first-order theories in this slow collision regime. Although inexplicably the cross section measurement of Shah et al. (1981) is a factor of 2 smaller than the theoretical predictions of BEA, SCA and even more so below coupled-state calculations of Fitchard et al. (1977), at $v_1/v_{2K} = 1.0$ (i.e., when the cross section peaks for 0.025 MeV protons), BEA's and SCA's $P[0]$ are in a remarkably good agreement. The stars in Fig. 6 were calculated by Trautmann (2005) also in the straight-line SCA. They were to check the numbers obtained from the

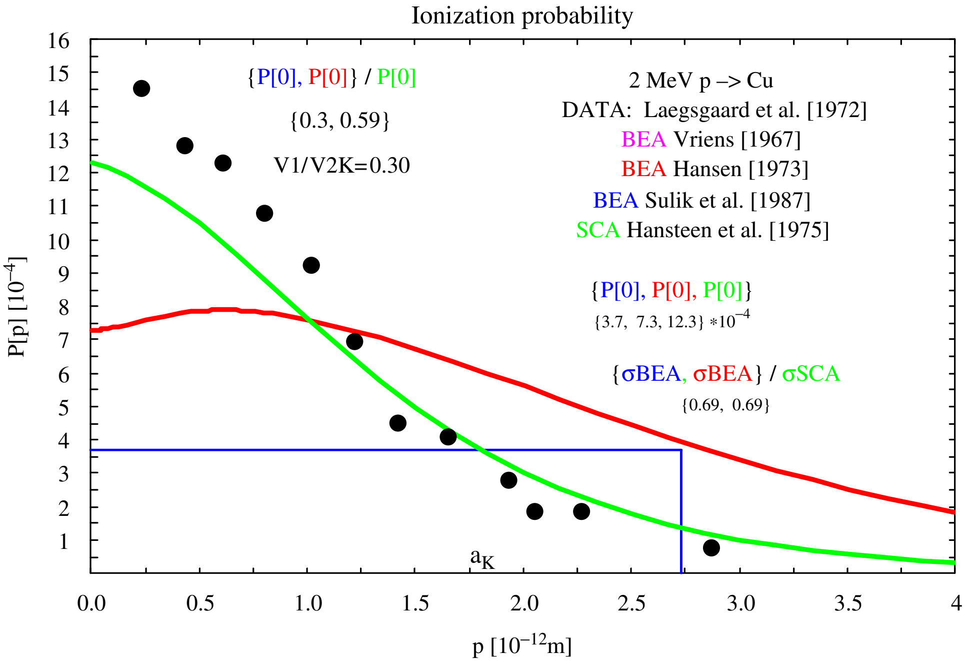

Fig. 3. Impact parameter ionization probability for K-shell ionization of copper by 2-MeV protons according to the geometrical model of Sulik et al. (blue rectangle), SCA (light green curve), and BEA of Hansen (dark red curve) that for $P[p] < 0.001$ overlaps with BEA of Vriens. Note that at $v_1/v_{2K} = 0.30$, $P^{\mathrm{Sulik}}[0]/P^{\mathrm{SCA}}[0] = 0.3$ and $P^{\mathrm{Sulik}}[0]$ underestimates the data extrapolated to $p = 0$ by a factor of 4.

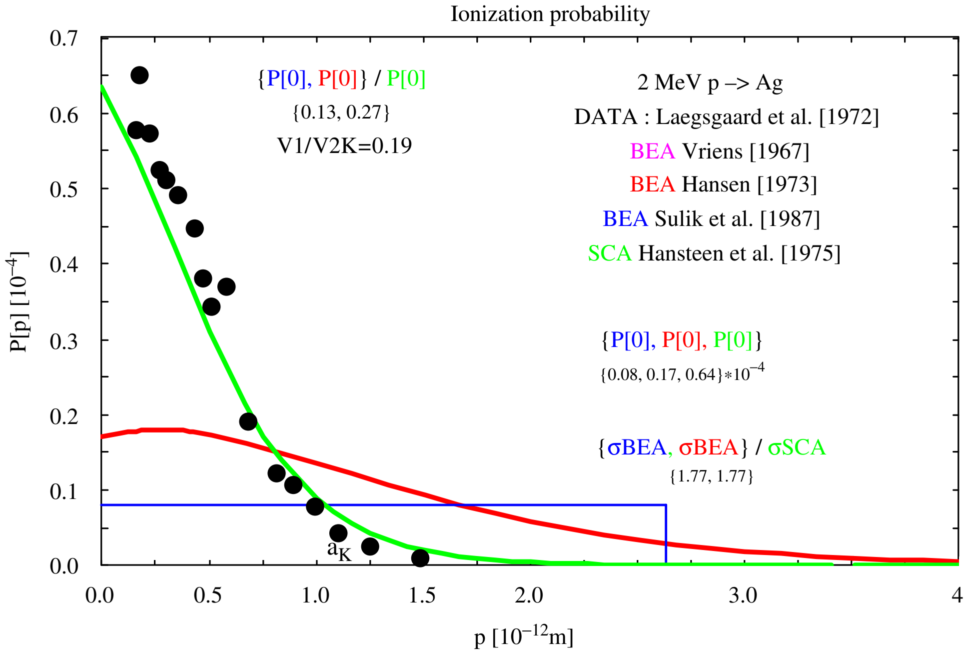

Fig. 4. Impact parameter ionization probability for K-shell ionization of silver by 2-MeV protons. Note that at $v_1/v_{2K} = 0.19$, $P^{\mathrm{Sulik}}[0]/P^{\mathrm{SCA}}[0] = 0.13$ and $P^{\mathrm{Sulik}}[0]$ underestimates the data extrapolated to $p = 0$ by a factor of 10.

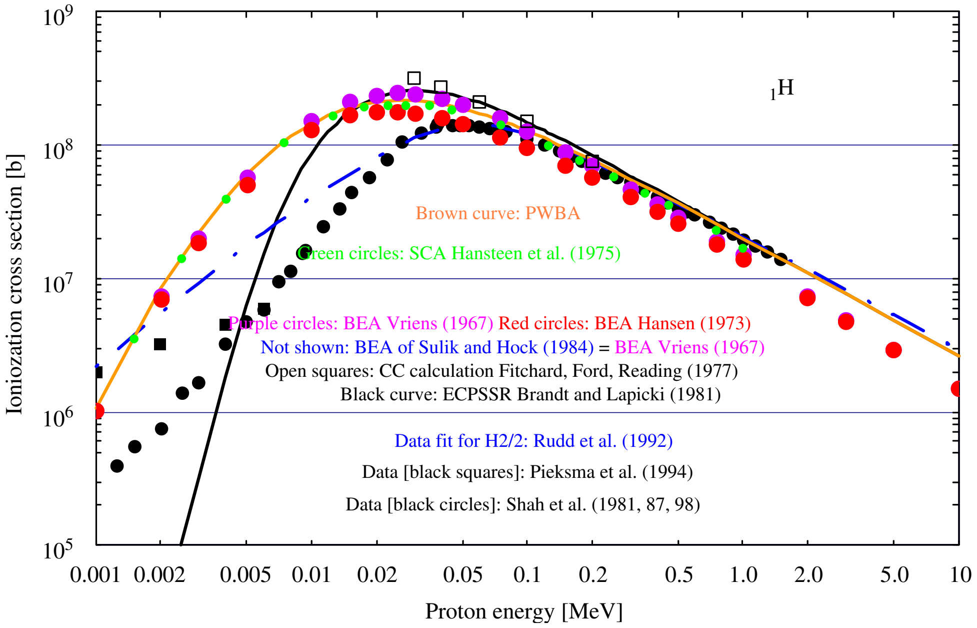

Fig. 5. Cross sections for hydrogen ionization by protons.

tables of Hansteen et al. (1975), and essentially confirmed them: 196 b vs. 200 b for the cross section and 0.94 vs. 0.90 for $P^{\mathrm{SCA}}[0]$ according to Hansteen et al. vs. Trautmann. Either way, at $v_1/v_{2K} = 1.0$ $P^{\mathrm{Sulik}}[0]$ is merely about 30% less than these semiclassical values.

This is not so, however, outside the collision regime where the cross section peaks i.e., where v_1 no longer matches v_{2S}. Fig. 7 exhibits reasonable agreement between BEA and SCA cross sections for $v_1 \leqslant v_{2S}$ while Fig. 8 clearly demonstrates that such agreement for $P[0]$ exists only where these velocities are in a close match. In particular, while generally better than $P^{\mathrm{Vriens}}[0]$ and $P^{\mathrm{Hansen}}[0]$, $P^{\mathrm{Sulik}}[0]$ of the geometrical model is for $v_1 < v_{2S}$ and for $v_1 > v_{2S}$ by as much as a factor of 5, respectively, below and above $P^{\mathrm{SCA}}[0]$. The degree to which $P^{\mathrm{Sulik}}[0]$ falls below $P^{\mathrm{SCA}}[0]$ when $v_1 < v_{2S}$ appears to be only slightly dependent on the Z_1/Z_2 as the $P^{\mathrm{Sulik}}[0]/P^{\mathrm{SCA}}[0]$ calculated for K-shell ionization of $_{29}$Cu, $_{34}$Se, $_{47}$Ag for $Z_1/Z_2 < 0.35$ (see blue points in Fig. 8) are in close proximity of that ratio calculated for $Z_1/Z_2 = 1$ (see blue

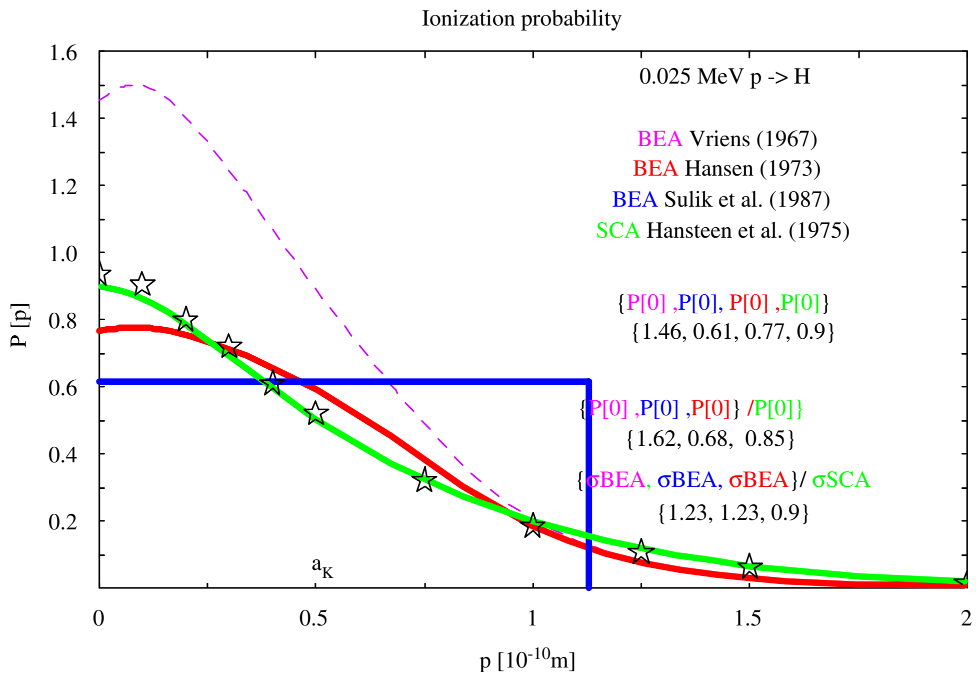

Fig. 6. Impact parameter ionization probability for ionization of hydrogen by 0.025-MeV protons according to the geometrical model of Sulik et al. (blue rectangle), SCA (light green curve), BEA of Hansen (dark red curve), and BEA of Vriens (dashed purple curve). The stars represent the SCA calculated by Trautmann. Note that at $v_1/v_{2K} = 1.0$, $P^{\text{Sulik}}[0]$ is in a 30% agreement with $P^{\text{SCA}}[0]$.

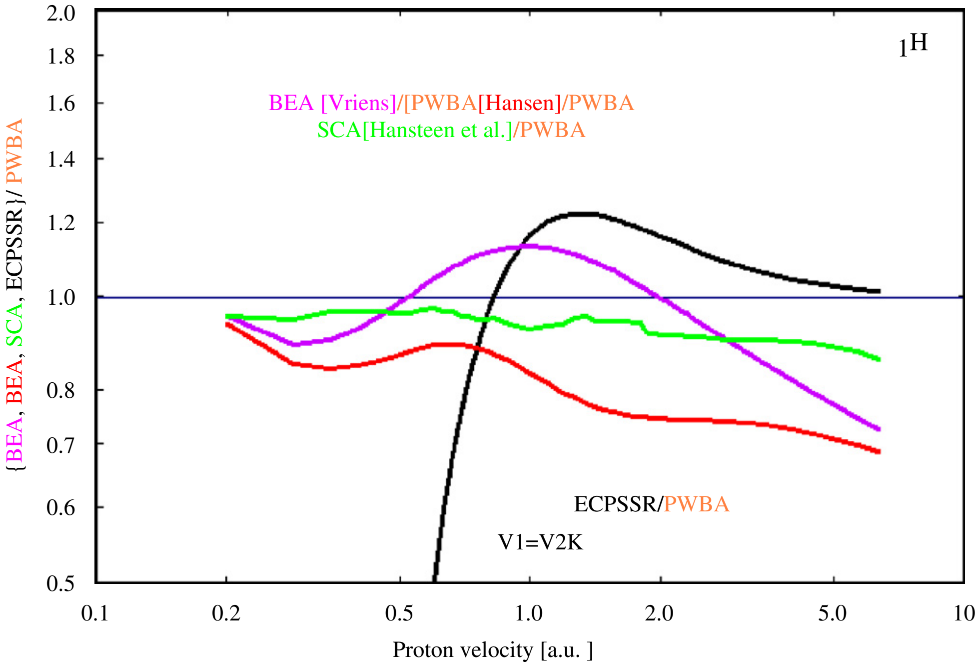

Fig. 7. Ratio of cross sections for ionization of hydrogen by protons according to the SCA, BEA, and ECPSSR theories to the predictions of the PWBA (Merzbacher and Lewis, 1958).

curves in Fig. 8). The underestimate of $P^{\text{SCA}}[0]$ by the geometrical model of Sulik et al. (1984, 1987) when $v_1 \leqslant v_{2S}$ presents a somewhat academic issue because neither of these first-order theories is expected to be satisfactory in this slow collision regime. Nevertheless, it strongly suggests that $P^{\text{Sulik}}[0]$ should not be employed in multiple ionization studies that are performed in this collision regime.

The black curves in Fig. 8 show $P^{\text{Lapicki}}[0]$ developed by Lapicki et al. (1986) on the basis of the classical cross section of Thomson (1912). This formula had an adjustable

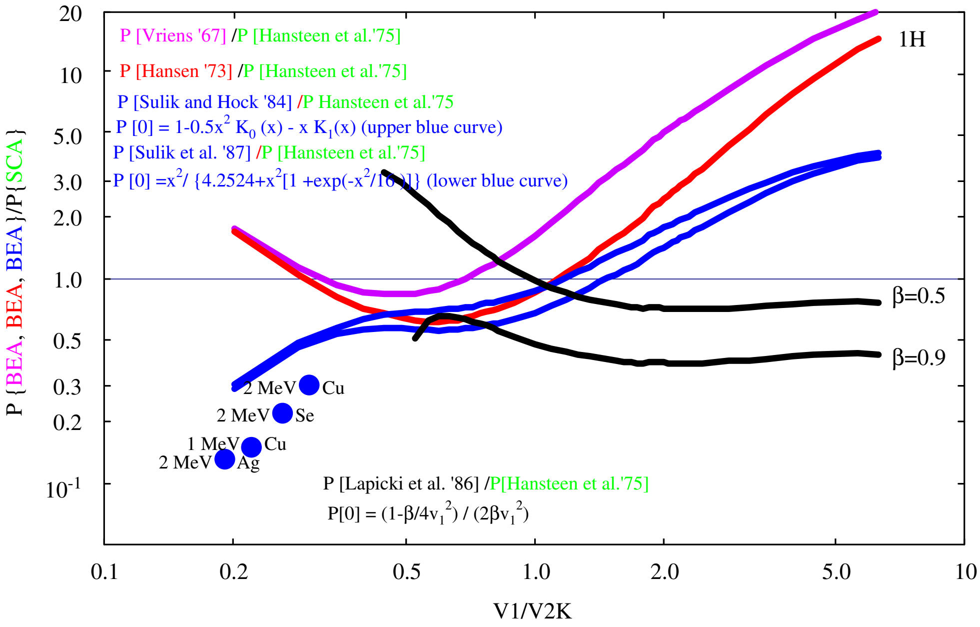

Fig. 8. Ratio of $P[0]$ for ionization of hydrogen by protons according to the BEA of Vriens (top purple curve) and Hansen (second from the top red curve), the geometrical model of Sulik et al. (blue curves that merge together), and Lapicki et al. (black curves for different β) to the predictions of the SCA.

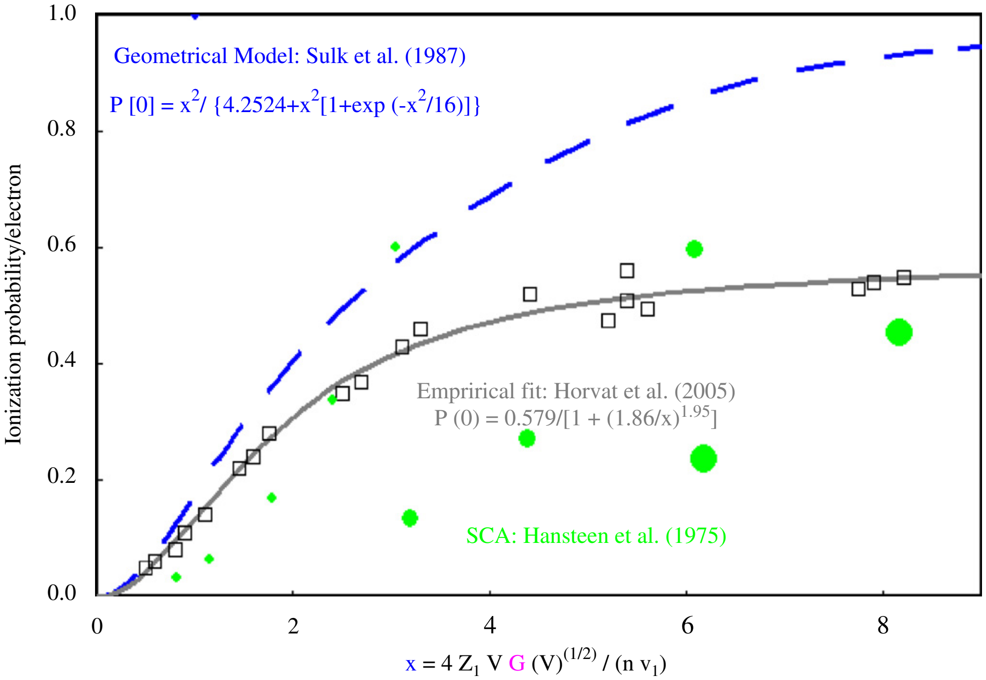

Fig. 9. Ionization probability $P[0]$ as a function of the scaling variable of the geometrical model. The geometrical model (dashed blue curve) overestimates the data of Horvat et al. (2005) (squares and the gray curve for their fit) by as much as a factor of 2 while the calculation based on the SCA of Hansteen et al. (1975) (green circles of increasing size for $Z_1 = 1, 2, 3$) tend to follow the trend of these data.

parameter β that was set at a value of 0.9. For $v_1 > v_{2S}$—as gauged by $P^{\mathrm{SCA}}[0]$ and in contrast to $P^{\mathrm{Sulik}}[0]$—$P^{\mathrm{Lapicki}}[0]$ underestimates the semiclassical result by about a factor of 2.5. It appears that setting β at a smaller value would, at the expense of a deteriorated agreement for $v_1 < v_{2S}$, bring the formula of Lapicki et al. (1986) in very good agreement with $P^{\mathrm{SCA}}[0]$ for $v_1 > v_{2S}$.

4. Conclusions

While the semiclassical theory (the increasing in size circles in Fig. 9 were evaluated in the SCA for $Z_1 = 1, 2,$ and 3 projectiles on hydrogen) tends to mimic the experimental trend seen recently by Horvat et al. (2005), the geometrical model of Sulik et al. fails to agree with these data. The scaling variable x of the geometrical model of Sulik et al. (1984, 1987) increases as v_1/v_{2S} becomes larger. When $v_1/v_{2S}>1$, in $P^{\text{Sulik}}[0]$ progressively exceeds $P^{\text{SCA}}[0]$ with the increasing x. Over 30 years ago (see Li et al., 1973), based on measurements with light ions, Watson and his coworkers—who have now established (see Horvat et al., 2005) the shortcoming of a BEA treatment of multiple ionizations by heavy ions—favored, somewhat ironically, a BEA approach over the SCA of Hansteen and Mosebekk (1972). In hindsight, the data in Fig. 6 of Li et al. (1973) were in fact overestimated by the BEA calculations. Not inconsistent with what is seen in Fig. 8 at such v_1/v_{2S}, they were increasingly overestimated with the increasing $v_1/v_{2L}>1$ as the $P^{\text{BEA}}[0]/P^{\text{SCA}}[0]$ ratio rose to about a factor of 4.

As gauged by the SCA of Hansteen et al. (1975), the success and failure of $P^{\text{Sulik}}[0]$ depends critically on v_1/v_{2S}: *while* $P^{\text{Sulik}}[0]$ *can reliably predict effects of multiple-ionization in the regime of the match of these velocities*, $P^{\text{Sulik}}[0]$ increasingly and significantly under- and over-estimates these effects when, respectively, $v_1/v_{2S}<1$ and $v_1/v_{2S}>1$.

The underestimate of $P^{\text{SCA}}[0]$ by $P^{\text{Sulik}}[0]$ when $v_1/v_{2S}<1$ presents somewhat of an academic issue because neither of these first-order theories is expected to be satisfactory in this slow collision regime. The issue remains, however, that $P^{\text{Sulik}}[0]$ should not be employed in multiple ionization analyses when $v_1/v_{2S}<1$. The overestimate of $P^{\text{SCA}}[0]$ by $P^{\text{Sulik}}[0]$ when $v_1/v_{2S}>1$, which is assumed in the derivation of the geometrical model formula, places its results in jeopardy as evident from their comparison with the data of Horvat et al. (2005).

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