Binary-encounter dipole model for positron-impact direct ionization

Kamil Fedus* and Grzegorz P. Karwasz†
Institute of Physics, Faculty of Physics, Astronomy and Informatics, Nicolaus Copernicus University, Grudziadzka 5/7, 87-100 Toruń, Poland

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In the present work we study the applicability of the binary-encounter-dipole (BED) model and its simpler version the binary-encounter-Bethe model for positron-impact direct ionization. We show that ignoring the exchange and interference effects in the theory of Kim and Rudd [Phys. Rev. A **50**, 3954 (1994)] yields simple analytical formulas with no fitting parameters that can be used to estimate cross sections over a wide energy range except near-threshold region. To correct this deficiency we combine BED theory with the Wannier-type threshold law derived by Klar [J. Phys. B **14**, 4165 (1981)] for positron-impact ionization. We show that such combination is necessary in order to predict cross sections at low positron energies where strong polarization-correlation effects are present during collision and the positronium formation is a dominant scattering process. The present theory is tested for a wide range of targets including helium (He), atomic hydrogen (H), neon (Ne), argon (Ar), molecular hydrogen (H_2), nitrogen (N_2), oxygen (O_2), carbon monoxide (CO), carbon dioxide (CO_2), and methane (CH_4). An extensive comparison with available experiments and theories is done.

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I. INTRODUCTION

The binary-encounter-dipole (BED) model and its simpler version, the binary-encounter-Bethe (BEB) model, for electron-impact ionization of atoms and molecules were developed by Kim and Rudd in 1994 [1]. The theory combines the modified Mott cross sections [2] describing hard (close) collisions between two electrons with the Bethe theory [3] for soft (distant) collisions due to a long-range dipole interaction important at high incident energies. Both BED and BEB were found to reproduce known ionization cross sections accurately for small atoms and a variety of large and small molecules (e.g., see [4–14]), and they have been shown to be useful for estimating unknown cross sections. In particular, the BEB model provides a simple and easy-to-use analytic formula with no fitting parameters for the total ionization cross section. Different modifications of original models have been proposed later in order to extend their range of applicability [15–19] including autoionization processes [20,21] and relativistic effects [22]. Helpful references on this theory and BEB cross sections for electron-impact ionization of many targets (compared with experiments) can be found online in the database maintained by the American National Institute of Standards and Technology [23].

Surprisingly, despite a great success of the theory for electron-impact ionization (the original work in Ref. [1] has been already cited more than 500 times), this approach has never been tested for ionization by positron impact. Studies on positron-impact ionization are still in the nascent stage and they are more sparse than corresponding research on electrons which have been actively studied by many research groups since the 1920s. The lack of bright positron beams with good energy resolution hindered the advance of experiments for many years and the oldest reliable ionization cross sections can be dated only to the late 1980s (see the most recent reviews on positron scattering cross sections [24,25]). Moreover, the positron-impact ionization is more challenging experimentally, particularly at low positron energies, where the direct ionization is always accompanied by the competing process of positronium (Ps) formation. Ps is formed whenever an incident positron possesses enough kinetic energy to knock out an electron from the target and bind to it, leaving the target as a positive ion. The energy threshold for Ps formation is lower by 1/2 Rydberg than the corresponding threshold for direct ionization and at low positron energies the Ps formation could be a dominant ionization process. Since both processes lead to the production of ionized targets, experiments have to be designed carefully in order to separate both effects (Ps and direct ionization) [26,27].

From a theoretical point of view, the positron interactions with atoms and molecules are also very difficult to describe despite the absence of quantum exchange effect. In general, computational methods are found to be very sensitive to the delicate balance between the repulsive and attractive forces of the positron-target interaction and an accurate representation of the target becomes crucial. In addition, coupling effects between positronium formation, electronic excitation, and direct ionization channels may also play an important role [28]. In the case of molecular targets, the dissociative ionization channel should be included, which itself can proceed via direct ionization or positronium formation. Consequently, positron-impact direct ionization has not been studied so intensively as other scattering channels. In the past only a few different approaches were analyzed in detail for a wide range

*kamil@fizyka.umk.pl
†karwasz@fizyka.umk.pl

of targets (see [28–31] and references therein). Moreover, the most advanced contemporary theories involve such large basis sets, complicated equations, and complex calculation algorithms that their use is time consuming and they cannot be easily applied to each specific target for which data are needed urgently. For these reasons the search for systematic dependencies in positron cross sections becomes very important. For example, recently Machacek *et al.* [32] analyzed carefully experimental data of Ps formation cross sections. They found that simple parametrized surge function can be used to describe with some accuracy both the shape and the magnitude of this indirect ionization channel for a wide set of atoms and molecules. Such a simple approach can be used to estimate cross sections in media which are inaccessible to experiments. Reliable positron scattering cross sections are required as inputs for modeling and studying of positron transport in biological media and dense gases [33–37], as well as positron behavior in astrophysical conditions [38] and in positron plasmas [39].

In this paper we study the applicability of the binary-encounter-dipole (BED) and the binary-encounter-Bethe (BEB) models for positron-impact direct ionization. We show that ignoring the exchange and interference effects in the original theory of Kim and Rudd [1] yields simple analytical formulas that can be used to estimate positron direct ionization cross sections for many atoms and molecules. Moreover, we show that such straightforward simplification of the original theory is not enough to account for a contribution of strong polarization-correlation effects present just above the ionization threshold where the positronium formation is a dominant scattering process. To correct this deficiency we combine BED theory with the Wannier-type threshold law derived by Klar [40] for positron-impact ionization. The latter law was obtained using similar classical ideas to those of Wannier [41] for electron-impact ionization. The validity of both semiclassical laws was confirmed recently by pure quantum-mechanical calculations [42]. More detailed discussion on the applicability of Wannier-type laws can be found in Ref. [43].

This paper is organized as follows. In Sec. II we describe briefly the principles of the BED and BEB models for positrons. Section III includes results and discussions for selected targets: He, H, Ne, Ar, H_2, N_2, O_2, CO, CO_2, and CH_4. We limit our analysis to targets for which both the experimental data and the necessary physical parameters, such as binding energies and average electron kinetic energies on different orbitals of target, are available in the literature. The main conclusions are summarized in Sec. IV including the advantages and the limitations of BED and BEB for a description of positron-impact direct ionization.

II. BINARY-ENCOUNTER MODEL FOR POSITRONS

Original BED [1] combines Vrien's symmetric binary-encounter theory [2] including interference between the direct and the exchange interaction terms and the Bethe theory [3] for fast incident electrons. Since both models are insensitive to the sign of the projectile charge, they should be also applicable to positron collision with neutral atoms and molecules. Moreover, since there is no exchange in positron interaction with neutral targets, we can ignore all terms related to this effect in Vrien's model. Consequently, Vrien's differential cross sections on the energy distribution of electrons ejected from the atomic-molecular orbital takes the following form in the reduced variables:

$$\frac{d\sigma_i(w,t)}{dw} = \frac{S}{t+u+1}\left[\frac{1}{(w+1)^2} + \frac{4u}{3(w+1)^3}\right], \qquad (1)$$

where $t = T/B$ and $w = W/B$ are kinetic energies of incident positron (T) and ejected electron (W) normalized to electron binding energy (B). Here $u = U/B$, where U is the average orbital kinetic energy of a bound electron. Both B and U can be calculated from any quantum-mechanical calculations of the ground-state wave function of the target atom or molecule. The first term inside the square brackets of Eq. (1) is eqivalent to the modified Rutherford cross sections for collision of free charged particle with bound particle, while the second term takes into account momentum distribution of the target electron in the bound state. The constant prefactor $S = 4\pi a_0^2 N(R/B)^2$, where $a_0 = 5.29 \times 10^{-11}$ m is the Bohr radius, $R = 13.6$ eV is the Rydberg energy, and N is the number of bound electrons in the orbital.

The scaling factor $t + u + 1$ appearing in the denominator before the square brackets was introduced *ad hoc* into the classical binary-encounter theory [2] and it is considered as the "effective" kinetic energy of the projectile seen by the target electron. For incident electron the term $u + 1$ represents its "acceleration" due to the nuclear attraction. The applicability of similar scaling factors for a description of electron-impact excitations of neutral atoms and molecules was studied in detail by Kim [44,45] and it was reviewed for all inelastic processes more recently by Tanaka *et al.* [46]. Probably this acceleration term is related to the shielding of the nuclear charge as seen by the incident projectile [18]. For a positron we cannot use the same interpretation of the acceleration term as for electron impact due to the repulsive nature of the positron-nucleus interaction. However, it is well known that due to the absence of repulsive exchange interaction, the distortion of a target electron cloud caused by an incoming positron could be so strong that the resulting attractive polarization potential can overcome locally the nuclear repulsion [47,48]. This net attractive interaction may even lead to virtual positronium formation at low incident energies [49] and some special enhancement factors have to be used [50] in order to adapt well-established electron scattering models for positron collisions. Hence we can consider the $u + 1$ term as "the acceleration" of the incident positron due to attractive polarization interaction. However, we have found (see results and discussion in Sec. III) that in the near-threshold region the $u + 1$ term is too weak to reproduce experimental data. To account for strong polarization-correlation effects (leading also to the effective Ps formation at low energies) we have to include an additional term in the denominator of Eq. (1). To find this extra term we decided to consider the Wannier-type threshold law proposed by Klar [40] for positron impact ionization. According to this semiclassical theory the near-threshold energy dependence of the total cross section ($\sigma_{\rm tot}$)

for direct ionization by positron impact is predicted to be

$$\sigma_{\rm tot} \propto (T - B_{th})^{2.65}, \tag{2}$$

where $T - B_{th}$ is the energy of the positron in excess of the ionization threshold energy B_{th} (the latter physical quantity corresponds to the binding energy B of the target electron at the outermost orbital). The Wannier-type theory cannot predict the value of the constant of proportionality and the energy range over which this equation is valid (though it is expected to be valid over several eV above the threshold [40]). On the contrary to Eq. (2) the original BED [1] predicts linear proportionality between the total ionization cross sections and the projectile excess energy. To overcome this inconsistency we found that the following acceleration term should be used in Eq. (1) to fulfill the Wannier law:

$$\gamma = u + 1 + \frac{C}{(t-1)^{1.65}}, \tag{3}$$

where C is the unknown positive constant. We found that $C \approx 1$ gives relatively good agreement with experimental data, though the exact values of C for each specific target can be derived once the agreement on total ionization cross sections is achieved. The detailed derivation of the modified acceleration term in Eq. (3) is described in Appendix A.

In order to combine classical binary-encounter theory [2] with a quantum Bethe model [3], Eq. (1) (with new acceleration factor γ) can be rewritten in a more compact form by separating t and w variables:

$$\frac{d\sigma_i(w,t)}{dw} = S\sum_{n=1}^{3} F_n(t) f_n(w), \tag{4}$$

where $F_1 = 0$, $F_2 = 1/(t+\gamma)$, $F_3 = 4u/3(t+\gamma)$, and $f_n = 1/(w+1)^n$. The fact that $F_1 = 0$ is related to the absence of exchange effects. Integral ionization cross section is obtained by integrating Eq. (4) over the entire kinetic-energy range of ejected electrons, i.e., from $w = 0$ to $w = t - 1$:

$$\sigma_i(t) = S\int_o^{t-1} \frac{d\sigma_i}{dw} dw = S\left[F_2\left(1 - \frac{1}{t}\right) + \frac{1}{2}F_3\left(1 - \frac{1}{t^2}\right)\right]. \tag{5}$$

The asymptotic behavior ($t \gg 1$) of the integral cross section in Eq. (5) does not agree with the Bethe theory [3], viz. $\sigma_i \sim t^{-1}\ln t$, which is valid for both electrons and positrons at high incident energies. To correct this deficiency we can follow step by step the procedure described by Kim and Rudd in Ref. [1] (see Appendix B). As a final result the following changes have to be made inside Eq. (4):

$$F_2 = \frac{2 - N_i/N}{(t+\gamma)}, \quad F_3 = \frac{\ln t}{(t+\gamma)}, \quad f_3 = \frac{1}{N(w+1)}\frac{df_i(w)}{dw}, \tag{6}$$

where $df_i(w)/dw$ is the differential oscillator strength and

$$N_i = \int_0^{\infty} \frac{df_i(w)}{dw} dw. \tag{7}$$

Consequently, the differential and integral ionization cross sections take the following forms:

$$\frac{d\sigma_i(w,t)}{dw} = \frac{S}{(t+\gamma)}\left[\left(2 - \frac{N_i}{N}\right)\frac{1}{(w+1)^2} + \frac{\ln t}{N(w+1)}\frac{df_i(w)}{dw}\right], \tag{8}$$

$$\sigma_i^{\rm BED}(t) = \frac{S}{(t+\gamma)}\left[\left(2 - \frac{N_i}{N}\right)\left(1 - \frac{1}{t}\right) + D_i(t)\ln t\right], \tag{9}$$

where

$$D_i(t) = N^{-1}\int_0^{t-1} \frac{1}{w+1}\frac{df_i(w)}{dw} dw. \tag{10}$$

Equation (9) is the positron equivalent of the binary-encounter-dipole (BED) model introduced for electron-impact ionization in Ref. [1]. This model can be used to estimate ionization cross sections when the differential oscillator strengths $[df_i(w)/dw]$ are known for each subshell of the target. The latter can be derived from either theoretical or experimental photoionization cross sections. Note that the integration range used in parameter $D_i(t)$ [defined in Eq. (10)] is twice as wide as the corresponding range for electron-impact ionization. In the latter case we integrate from $w = 0$ to $w = (t-1)/2$ due to indistinguishability of ejected and scattered electrons after collision; see Ref. [1].

Unfortunately, the differential oscillator strengths are available only for a limited number of atoms and molecules. When no information on df_i/dw is available we can use a simplified version of the model assuming hydrogenlike df_i/dw in f_3 of Eq. (6) for all target orbitals [1]:

$$\frac{df_i}{dw} = \frac{b}{(w+1)^2}, \tag{11}$$

where b is a constant. For such a specific choice of df_i/dw, the following relations hold: $b = N_i = QN$, where Q is the dipole constant defined as (see also Appendix B)

$$Q = \frac{2}{N}\int_0^{\infty} \frac{1}{w+1}\frac{df_i(w)}{dw} dw. \tag{12}$$

Consequently, we can set $b = N_i$ and $N_i/N = Q$ in all terms of Eq. (6). As a result, the integral cross section [i.e., Eq. (4) integrated over an entire range of energy w] is simplified to the BEB expression:

$$\sigma_i^{\rm BEB}(t) = \frac{S}{(t+\gamma)}\left[\left(2 - Q\right)\left(1 - \frac{1}{t}\right) + \frac{1}{2}Q\left(1 - \frac{1}{t^2}\right)\ln t\right]. \tag{13}$$

We can set $Q = 1$ as a further approximation in order to rule out df_i/dw (if not available). Such simplification was introduced in Ref. [1] and it was found to work very well for electron-impact ionization of many molecular targets and simple atoms. The total cross sections for positron-impact direct ionization ($\sigma_{\rm tot}$) can be calculated by summing up $\sigma_i^{\rm BED}$ or $\sigma_i^{\rm BEB}$ over all orbitals of target atom or molecule. In the near-threshold region the total cross sections calculated using both models follow the Wannier-type threshold law given by

Eq. (2) due to the introduction of the modified scaling factor defined by Eq. (3) (for $C > 0$).

When we compare positron BED [Eq. (9)] and BEB [Eq. (13)] formulas to corresponding versions for electron impact [i.e., Eqs. (55) and (57) in Ref. [1]], we find that they are very similar to each other except the "$-\ln t/(t+1)$" term that is present in the expressions for electrons. This term comes from the interference between scattered and ejected electrons. Since this additional term is negative, it lowers electron-impact ionization cross sections when compared to corresponding positron cross sections. In general, this is consistent with experimental observations.

III. RESULTS AND DISCUSSION

In this section we apply BED [Eq. (9)] and BEB [Eq. (13)] models to calculate total cross sections for positron-impact direct ionization of different atomic and molecular targets. We consider both cases—the models with original acceleration factor, i.e., $C = 0$ in Eq. (3) and with modified factor, i.e., $C = 1$. The former results are labeled BED and BEB, while the latter are named BEB-W and BED-W (i.e., binary-encounter-dipole-Wannier) in the presented figures. We use binding energies (B), average orbital kinetic energies of bound electrons (U), and electron occupation numbers (N) published at NIST database [23]. For self-consistency of this paper the values used for all three quantities are also presented in Table I.

BED and BED-W models [Eq. (9)] are tested only for He, H, Ne, Ar, and H_2, where reliable differential oscillator strengths for each subshell of the target are available. For atomic targets we use analytical fits to photoionization cross sections calculated with the R-matrix method at low photon energies and Hartree-Dirac-Slater approach at high energies as reported in Refs. [51,52]. For molecular hydrogen (H_2) the power-series fit found by Kim and Rudd [1] is used. Numerical procedures used to calculate df_i/dw are described in more detail in Appendix C.

TABLE I. Atomic and molecular orbital constants: binding energy B, average kinetic energy U, and electron occupation number N (from Refs. [1,4,5,53]).

Target	Orbital	B (eV)	U (eV)	N
He [1]	$1s$	24.5870	39.5100	2
H [1]	$1s$	13.6057	13.6057	1
H_2 [4]	$1\sigma_g$	15.43	15.98	2
	$2\sigma_g$	41.72	71.13	2
N_2 [4]	$2\sigma_u$	21.00	63.18	2
	$1\pi_u$	17.07	44.03	4
	$3\sigma_g$	15.58	54.91	2
Ne [53]	$1s$	866.9	1259.1	2
	$2s$	48.47	141.88	2
	$2p$	21.60	116.02	6
Ar [53]	$1s$	3202.9	4192.9	2
	$2s$	326	683.1	2
	$2p$	249.18	651.4	6
	$3s$	29.24	103.5	2
	$3p$	15.82	78.07	6
O_2 [4]	$2\sigma_g$	46.19	79.73	2
	$2\sigma_u$	29.82	90.92	2
	$1\pi_u$	19.64	59.89	4
	$3\sigma_g$	19.79	71.84	2
	$1\pi_g$	12.07	84.88	2
CO [4]	3σ	41.92	79.63	2
	4σ	21.92	73.18	2
	1π	17.66	54.30	4
	5σ	14.01	42.26	2
CO_2 [4]	$3\sigma_{1g}$	42.04	75.72	2
	$2\sigma_{2u}$	40.60	78.38	2
	$4\sigma_{1g}$	21.62	74.66	2
	$3\sigma_{2u}$	20.27	71.56	2
	$1\pi_u$	19.70	49.97	4
	$1\pi_g$	13.77	64.43	4
CH_4 [5]	$1a_1$	290.70	436.07	2
	$2a_1$	25.73	33.05	2
	$2t_2$	14.25	25.96	6

A. Helium

The present results for He are compared with experiments and other theories in Fig. 1. For comparison we take into account experimental data by Fromme *et al.* [54] from University of Bielefeld, Knudsen *et al.* [55] and Jacobsen *et al.* [56] from University of Aarhus, and Mori and Sueoka [57] from University of Tokyo. Other experimental cross sections were measured at University College London in broad energy range by Moxom *et al.* [58] and in near-threshold range by Ashley *et al.* [59]. Both data sets were corrected later ($\times 0.904$) by Murtagh *et al.* [60] and hence only the latter results are presented in Fig. 1.

The latest theoretical calculations of the direct ionization cross section for He include the modified spherical complex optical potential (SCOP) formalism of Singh and Antony [61], the convergent close-coupling (CCC) computations of Utamuratov *et al.* [62], the distorted-wave model of Campeanu *et al.* [63] and Moores [64] (the latter with close-coupled target states), and the coupled-state calculations of Campbell *et al.* [65] and Chen and Msezane [66].

As shown in Fig. 1 the level of agreement between the various experimental cross sections and a number of theoretical approaches is generally good. Present BED-W and BEB-W models remain consistent with other results over a wide energy range. On the other hand, BED model (and BEB—not shown here for figure clarity reasons) overestimates cross sections at low energies—approximately from ionization threshold to around 60 eV. This is the energy region where the positronium (Ps) formation dominates over direct ionization: integral Ps cross sections peaking with value $\sim 0.45 \times 10^{-20}$ m^2 at around 40 eV [25]. Moreover, polarization-correlation effects play a very important role in a direct ionization at such low energies. These results demonstrate clearly the importance of an additional acceleration term in the BED theory [see Eq. (3)] allowing one to combine it with the Wannier-type threshold law. A similar effect is observed for almost all other targets studied in this work. Note that, in experiment, the near-threshold region is tedious

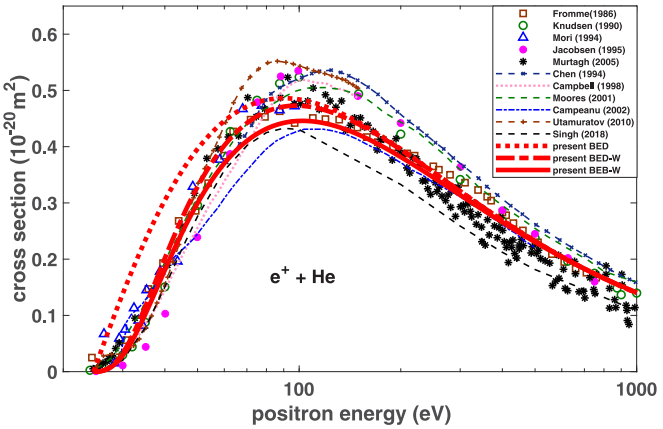

FIG. 1. Direct ionization cross section for positron scattering from helium (He). BEB-W (solid line), BED-W (thick dashed line), and BED (dotted line) are compared with (i) experiments, Fromme *et al.* [54], Knudsen *et al.* [55], Mori and Sueoka [57], Jacobsen *et al.* [56], and Murtagh *et al.* [60], and (ii) theories, Chen and Msezane [66], Campbell *et al.* [65], Moores [64], Campenau *et al.* [63], Utamuratov *et al.* [62], and Singh *et al.* [61].

to characterize due to low resolution of positron beams and low kinetic energies of ions produced, which makes it difficult to complete their collection. Hence the experimental data in the near-threshold region are characterized by the highest uncertainties.

B. Atomic hydrogen

The present results for atomic hydrogen (H) are compared with experiments and other theories in Fig. 2. Experimental results of Hofmann *et al.* [67] obtained by collaborating groups from University of Bielefeld and Brookhaven Laboratory superseded data sets of Spicher *et al.* [68] and Weber *et al.* [69] measured earlier by the same experimental systems.

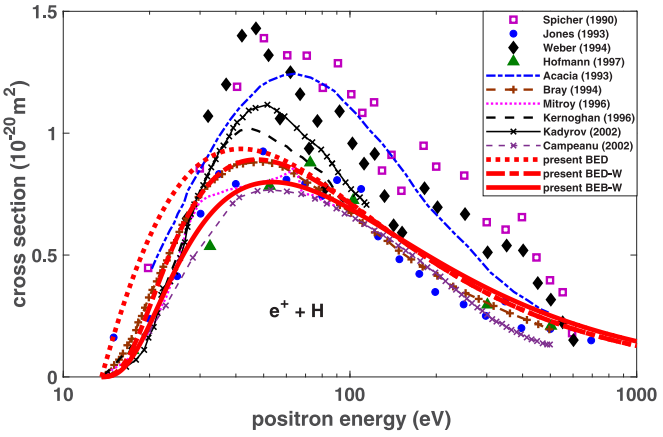

FIG. 2. Direct ionization cross section for positron scattering from atomic hydrogen (H). BEB-W (solid line), BED-W (thick dashed line), and BED (dotted line) are compared with (i) experiments, Spicher *et al.* [68], Jones *et al.* [70], Weber *et al.* [69], and Hofmann *et al.* [67], and (ii) theories, Acacia *et al.* [71], Bray and Stelbovics [72], Mitroy [73], Kernoghan *et al.* [74], Kadyrov and Bray [75], and Campeanu *et al.* [63].

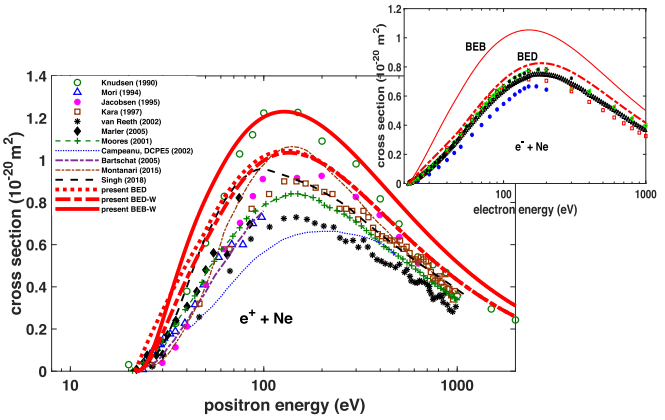

FIG. 3. Direct ionization cross section for positron scattering from neon (Ne). BEB-W (solid line), BED-W (thick dashed line), and BED (dotted line) are compared with (i) experiments, Knudsen *et al.* [55], Mori and Sueoka [57], Jacobsen *et al.* [56], Kara *et al.* [76], van Reeth *et al.* [77], and Marler *et al.* [78], and (ii) theories, Moores [64], Campeanu *et al.* [63], Bartschat [79], Montanari and Miraglia [80], and Singh *et al.* [61]. The inset shows BEB and BED calculations for electron-impact ionization compared with compilation of different experimental and theoretical data sets (from LxCat database [82]).

The latest data [67] are in good agreement with the results of Jones *et al.* [70] from the University College London.

Except for the distorted-wave calculations of Acacia *et al.* [71], all other available theories are quite consistent with Refs. [67,70]. These theories include CCC calculations of Mitroy [73], Bray and Stelbovics [72], and Kadyrov and Bray (total breakup cross section) [75], the distorted wave model of Campeanu *et al.* [63], and computations of Kernoghan *et al.* [74]. Both BEB-W and BED-W remain consistent with these theories supporting the experimental data of Refs. [67,70], while BED (and BEB) predicts higher cross sections in the near-threshold region.

C. Neon

BED and BEB were proved to describe very well cross sections for electron-impact ionization of small atoms He and H [1]. The inset of Fig. 3 shows BED and BEB calculations for ionization of neon (Ne) by electron impact (using the electron version of the theory [1]). Clearly, BEB overstimates cross sections when compared to experimental data. On the other hand, the more accurate BED model is in good agreement with experiments. In other words, the approximation of hydrogen-like differential oscillator strength, see Eq. (11), does not work for neon and exact df_i/dw are needed for each orbital.

This observation remains valid for positrons, see Fig. 3, where both BEB-W and BEB (not shown here) give much higher cross sections when compared with available data. Although the maximum of BEB cross section agrees with the old experiment from Aarhus by Knudsen *et al.* [55], the latter data were corrected later by Jacobsen *et al.* [56] using the same experimental system. The BED-W and BED models are more consistent with updated results from Aarhus, though still the calculated cross sections are higher. Once again BED-W works better in the near-threshold region. In general, there is a

large spread in the available experimental data, particularly in the vicinity of the cross section peak at around 150 eV. Interestingly, the first measurements from UCL group by Kara *et al.* [76] are compatible with the latest Aarhus results. However, van Reeth *et al.* [77] renormalized UCL data using more recent reference cross sections from electron impact ionization. As a result, the positron cross sections were reduced by 19% and became comparable with corresponding data for electrons. No difference between electron and positron impact ionization would mean that the exchange effect lowering electron cross sections either is very small or is recompensated by some indirect ionization phenomenon which is different for incoming electrons and positrons. It could be a contribution from double ionization where either the inner-shell electron is excited to an unstable state which decays by electron emission or two electrons are ejected simultaneously by a single impact. However, at present, this effect seems to be small for neon.

Significant spread in experimental data is followed by similar discrepancies in theoretical calculations. Campeanu *et al.* [63] tested different Ne wave functions within the frame of his distorted-wave model. It was found that the most elaborate DCEP5 shown in Fig. 3 gives the best agreement with experiments for all noble gases except neon, where the agreement was found to be the worst among all tested models. Other theories, such as the distorted-wave method with closed-coupled states of Moores [64], the continuum distorted-wave approach of Montanari and Miraglia [80], the *R* matrix of Bartschat [79], and spherical complex optical potential formalism of Singh *et al.* [61], all support rather the experiments by Jacobsen *et al.* [56] and Kara *et al.* [76].

In the light of large spread in theoretical and experimental data it is difficult to judge the credibility of BED theory for positron-impact ionization of neon; nonetheless this simple approach still gives reasonable values. Definitely, the BEB approximation in the present form overestimates Ne cross sections. Maybe some additional corrections in the empirical scaling factor can improve the BEB approach as was done for metastable noble gases [17].

D. Argon

The inset of Fig. 4 presents BEB and BED calculations for electron-impact ionization (using the electron version of the theory [1]). Theory is compared with a compilation of different experimental and theoretical total ionization cross sections (from [82]). Unlike for neon both BEB and BED give very similar results and they do not reproduce the very rapid increase of the total ionization cross section between threshold and about 40 eV, with a subsequent peak between 40 and 300 eV. Since the double ionization is quite important in electron collision with the argon atom, the experimental cross section for Ar^+ production by Rejoub *et al.* [84] is also included. BED is consistent with this experiment at energies higher than 200 eV, while BEB overlaps with these data at $T > 500$ eV. Therefore, both models describe pretty well direct ionization at high energies where the quantum theory of Bethe [3] is valid. Interestingly, the present BED almost overlaps in the entire energy range with the large-scale *R*-matrix-with-pseudostates calculations by Zatsarinny *et al.* [85]—not shown here for figure clarity reasons. Moreover, we

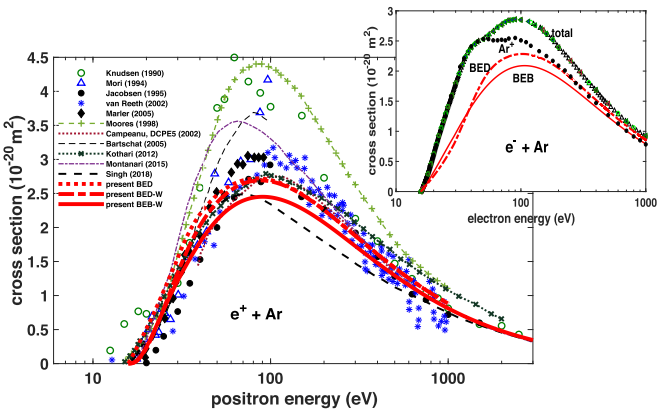

FIG. 4. Direct ionization cross section for positron scattering from argon (Ar). BEB-W (solid line), BED-W (thick dashed line), and BED (dotted line) are compared with (i) experiments, Knudsen *et al.* [55], Mori and Sueoka [57], Jacobsen *et al.* [56], van Reeth *et al.* [77], and Marler *et al.* [78], and (ii) theories, Moores [81], Campeanu *et al.* [63], Bartschat [79], Kothari and Joshipura [83], Montanari and Miraglia [80], and Singh *et al.* [61]. The inset shows BEB and BED calculations for electron-impact ionization compared with compilation of different experimental and theoretical data sets for total ionization cross sections (from LxCat database [82]). Experimental cross section for Ar^+ production (filled dots) [84] is also shown.

checked that it is possible to improve the electron version of BED [1] using a modified scaling factor: $1/\{t + (u + 1)/n\}$ for $n > 2$, where n is the principal quantum number of an orbital. Such modification was proposed for large atoms and singly charged molecular ions in Ref. [17]. We found here that BED with scaling factor modified in such a way allows one to reproduce perfectly experimental total cross sections for argon at low energies from threshold to about 40 eV and at energies higher than 200 eV. However, it overestimates experimental cross sections in the peak region by 20%.

For positrons, see Fig. 4, with few exceptions, most of the available experimental and theoretical cross sections are comparable in magnitude but not in shape with electron data. Regarding the shape—the cross sections do not increase so quickly in the near-threshold region as it happens for electrons. Like for other targets the experimental data of Knudsen *et al.* [55] are much higher than later results of Jacobsen *et al.* [56] measured using the same Aarhus system. High cross sections of Knudsen are supported by experiment of Mori and Sueoka [57] and the distorted-wave calculations of Moores [64] and Montanari and Miraglia [80], as well as the *R*-matrix computations of Bartschat [79]. Other experiments and theories (see Fig. 4), including the present BED-W and BED, are lower in magnitude but they are in relatively good agreement with each other. On the other hand, the present BEB-W and BEB (not shown here) underestimate cross sections in the peak region.

E. Molecular hydrogen

The main processes by which a molecular hydrogen (H_2) and other molecules can be ionized by positron are positronium formation, direct ionization, and dissociative ionization (which itself can proceed via direct ionization or positronium

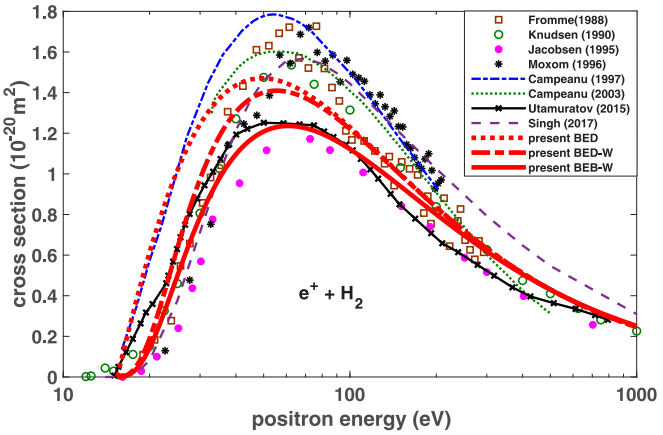

FIG. 5. Direct ionization cross section for positron scattering from molecular hydrogen (H_2). BEB-W (solid line), BED-W (thick dashed line), and BED (dotted line) are compared with (i) experiments, Fromme *et al.* [86], Knudsen *et al.* [55], Jacobsen *et al.* [87], and Moxom *et al.* [58], and (ii) theories, Campenau *et al.* [88,89], Utamuratov *et al.* [90], and Singh and Antony *et al.* [91].

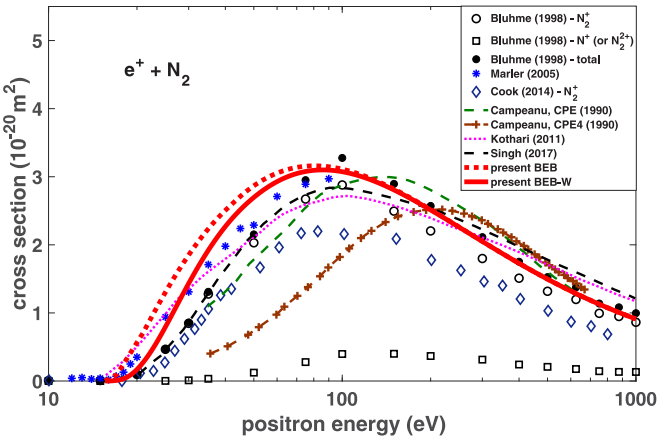

FIG. 6. Direct ionization cross section for positron scattering from molecular hydrogen (N_2). BEB-W (solid line), and BEB (dotted line) are compared with (i) experiments, Bluhme *et al.* [92], Marler and Surko [93], and Cook *et al.* [94], and (ii) theories, Campeanu *et al.* [95], Kothari and Joshipura [96], and Singh and Antony [97].

formation). The latter process may lead to the production of a H^+ ion in e^+-H_2 collisions. Both BEB and BED cross sections are simple sums of cross sections for ejecting one electron from each molecular orbital, so the theory cannot give a detailed account of dissociative ionization. Nevertheless, it was found [4] that the BEB model describes very well total cross sections for electron-impact ionization for a wide range of molecules, though it is still unclear why. The total cross section is understood as the simple sum of all partial cross sections for a production of specific ions.

For positron-impact ionization of molecular hydrogen (H_2) the agreement between different available experimental and theoretical data is rather poor. Though most of the data sets exhibit the same energy dependence, the differences in magnitudes are quite significant as shown in Fig. 5. The direct ionization cross sections for H_2 were characterized experimentally by Fromme *et al.* [86] from University of Bielefeld, Knudsen *et al.* [55] and Jacobsen *et al.* [87] from University of Aarhus, and Moxom *et al.* [58] from University College London. All mentioned works report only nondissociative ionization cross sections since an H^+ ion signal was found to be very small when compared to $H_2{}^+$. The latter observation indicates that positron-impact dissociative ionization is negligible for H_2. The latest data from Aarhus [87] superseded the older one [55] due to careful discrimination against false signals caused by positronium formation and these corrected cross sections are the lowest among all available experiments. On the other hand, there is much better agreement between measurements of University of Bielefeld and University College London, though the latter data were determined with the similar procedure as the old cross sections for He, Ar [58], and Ne [76] corrected later (reduced in magnitude) by the same group [60].

The wave-distorted calculations of Campenau *et al.* [88,89] with different representations of a H_2 wave function and the spherical complex optical potential formalism of Singh and Antony [91] support rather the experiments of Fromme *et al.* [86] and Moxom *et al.* [58]. On the other hand, the present BEB-W (see Fig. 5) supports experiment of Jacobsen *et al.* [87] and it is quite consistent with the latest two-center CCC calculations of Utamuratov *et al.* [90]. Both BED-W and BED predict higher magnitude than BEB-W and they are closer to experimental data by Knudsen *et al.* [55]. Once again the BED model overestimates cross sections in the region where Ps formation is a dominant inelastic process (peak of Ps cross section $\sim 2.8 \times 10^{-20}$ m^2 is located at around 20 eV [24]).

F. Molecular nitrogen

The positron-impact direct ionization cross sections for N_2 were studied experimentally by Bluhme *et al.* [92] from Aarhus University, Marler and Surko [93] from University of California in San Diego (UCSD), and Cook *et al.* [94] from University College London (UCL). Aarhus group [92] measured partial ionization cross section for the production of specific ions in direct ionization, $N_2{}^+$ and N^+, while UCL reported similar measurements only for $N_2{}^+$ since no N^+ signal was detected. The measurement method developed by Marler and Surko [93] did not rely on the collection of slow ions. Consequently, UCSD data may comprise contributions from both nondissociative and dissociative ionizations.

Theoretical calculations: the wave-distorted approach of Campeanu *et al.* [95] and the complex spherical potential formalisms of Kothari and Joshipura [96], and Singh and Antony [97], are generally in good agreement with nondissociative ($N_2{}^+$) measurements. Present BEB-W and BEB (see Fig. 6) overlap with total cross sections (i.e., sum for $N_2{}^+$ and N^+ production) of Bluhme *et al.* [92] at higher energies (>90 eV) and they predict slightly higher cross sections than other works at lower energies, though the BEB-W is quite consistent (within the experimental uncertainties) with the measurements of Marler and Surko [93].

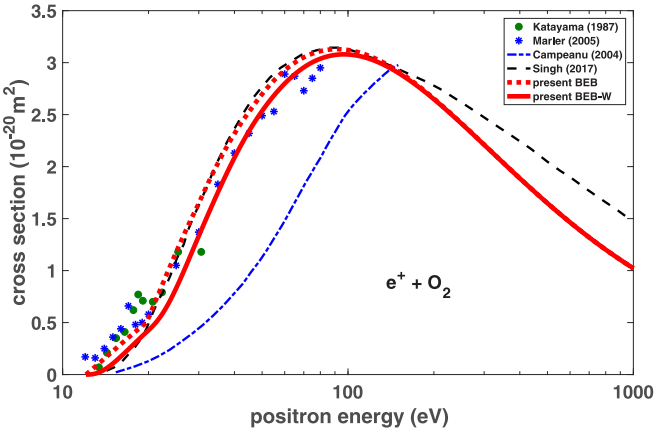

FIG. 7. Direct ionization cross section for positron scattering from molecular oxygen (O_2). BEB-W (solid line), and BEB (dotted line) are compared with (i) experiments, Katayama *et al.* [98] and Marler and Surko [93], and (ii) theories, Campeanu *et al.* [99] and Singh and Antony [97].

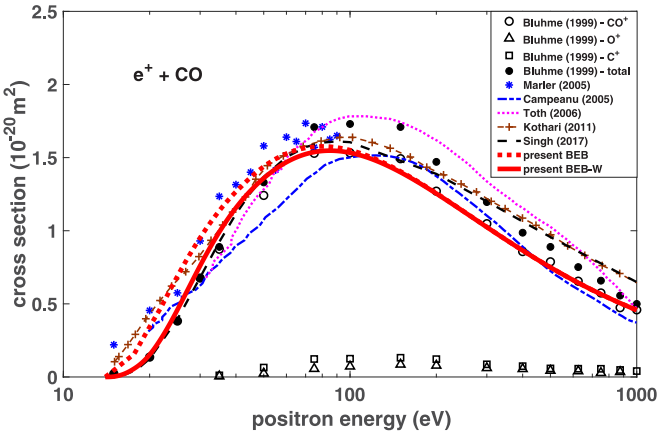

FIG. 8. Direct ionization cross section for positron scattering from carbon monoxide (CO). BEB-W (solid line) and BEB (dotted line) are compared with (i) experiments, Bluhme *et al.* [100] and Marler and Surko [93], and (ii) theories, Campeanu *et al.* [101], Toth *et al.* [102], Kothari and Joshipura [96], and Singh and Antony [91].

G. Molecular oxygen

Direct ionization cross sections for positron scattering from molecular oxygen (O_2) were studied experimentally by Marler and Surko [93] from UCSD and Katayama *et al.* [98] from University of Tokyo. Both data sets may comprise contributions from both nondissociative and dissociative ionizations. Unlike for targets analyzed in previous sections, this time both BEB and BEB-W are in very good agreement with both experiments at low positron energies; see Fig. 7. Interestingly, the positronium formation channel is relatively weak for O_2 when compared with other studied molecular targets (peak of Ps cross section $\sim 1.6 \times 10^{-20}$ m^2 is located at around 20 eV [24]). Moreover, BEB almost coincides with the spherical complex optical potential formalism of Singh and Antony *et al.* [97] for an energy range between 20 eV and 130 eV. BEB works also pretty well in the near-threshold region ($E < 20$ eV) when compared with both experiments. This time the wave-distorted calculations of Campeanu *et al.* [99] disagree with other data.

H. Carbon monoxide

Direct ionization cross sections for positron scattering from carbon monoxide (CO) were studied experimentally in Aarhus [100] and in UCSD [93]. The first group studied partial ionization cross sections for the production of specific ions: CO^+, C^+, and O^+, while UCSD reported total cross sections for direct ionization; see Fig. 8. At low energies (<70 eV) present BEB is consistent with UCSD data. On the other hand, BEB-W almost reconstructs nondissociative cross sections (i.e., for CO^+ production) from Aarhus in the entire energy range.

I. Carbon dioxide

To the best of our knowledge, so far the positron-impact direct ionization of carbon dioxide (CO_2) was studied experimentally only by Bluhme *et al.* [100]; see Fig. 9. This target attracted also little attention from a theoretical part—available works include the wave-distortion calculations of Campeanu *et al.* [101] and Toth *et al.* [102] using different representations of the CO_2 wave function and the spherical complex optical potential formalism of Singh and Antony [91]. Only the latter calculations are consistent with nondissociative (CO_2^+) experimental cross sections which are a dominant process in direct ionization. At energies >70 eV the present BEB and BEB-W calculations are in very good agreement with experimental data summed up over all partial cross sections for a production of specific ions (i.e., $CO_2^+ + CO^+ + C^+ + O^+$). Again at lower positron energies the BEB is slightly higher than experiment.

J. Methane

Direct ionization cross sections for methane (CH_4) were measured by Bluhme *et al.* [100] using the time of flight

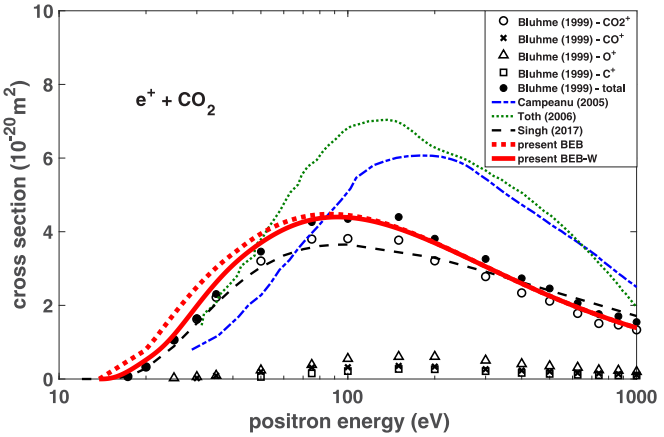

FIG. 9. Direct ionization cross section for positron scattering from carbon dioxide (CO_2). BEB-W (solid line), and BEB (dotted line) are compared with (i) experiments, Bluhme *et al.* [100], and (ii) theories, Campeanu *et al.* [101], Toth *et al.* [102], and Singh and Antony [91].

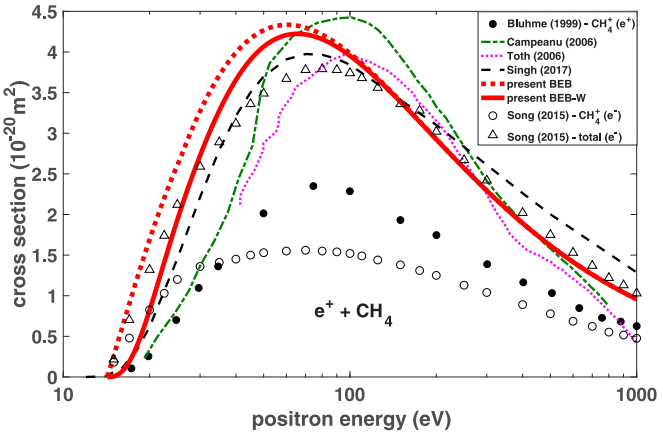

FIG. 10. Direct ionization cross section for positron scattering from methane (CH_4). BEB-W (solid line) and BEB (dotted line) are compared with (i) experiments, Bluhme *et al.* [100], and (ii) theories, Campeanu *et al.* [103], Toth *et al.* [102], and Singh and Antony [91]. For comparison total ionization cross sections and partial cross sections for CH_4^+ production by electron impact (recommended by Song *et al.* [104]) are presented.

technique (TOF). Unfortunately, their experimental system did not allow for deducing count rates for CH_3^+, CH_2^+, and H^+ ions from acquired TOF spectra. Consequently, only nondissociative cross sections for a production of CH_4^+ ions were reported; see Fig. 10.

Available theories [91,102,103] as well as the present models predict very similar magnitude of total ionization cross sections being almost twice larger at maximum than experimental data for CH_4^+. This difference can be tentatively understood if we compare cross sections for a production of CH_4^+ by electron impact with corresponding total cross sections [104]; see open symbols in Fig. 10. The partial cross sections for CH_4^+ production (open circles) by electron impact are more than two times lower than the total cross sections (open triangles)—in other words, the contribution of dissociative ionization to total ionization is higher than 50%. We expect that a similar trend should also hold for positrons. This could explain the present results of BEB, which was proved to be usually consistent with total ionization cross sections, i.e., the sum of all partial cross sections for a production of specific ions. On the other hand, it is quite surprising that both the wave-distorted models [102,103] and the spherical complex optical potential formalism [91] coincide with total ionization cross sections because neither of these theories include dissociation channels (i.e., processes leading to the production of ions other than CH_4^+). When compared to other theories [91,102,103] the maximum of BEB and BEB-W cross sections are clearly shifted towards lower energies.

IV. CONCLUSIONS

In this paper we introduced a positron version of the well-known binary-encounter-dipole (BED) theory proposed originally for electron-impact ionization. We showed that ignoring the exchange and interference effects in the original approach yields a simple analytical formula with no fitting parameters for total direct ionization cross sections. The applicability of the model has been tested for a wide range of atoms and molecules. Through the extensive comparison with experimental and theoretical data we showed that straightforward simplification of the original electron-impact theory is not enough to account for a contribution of strong polarization-correlation effects present in positron scattering just above the ionization threshold. To correct this deficiency we combined the BED model with the Wannier-type threshold law derived by Klar [40] for positron-impact ionization. The binary-encounter-dipole-Wannier (BED-W) model was found to be consistent with experiments over the entire energy range including the near-threshold region.

Like for electron impact the simplified version of the theory—the binary-encounter-Bethe-Wannier (BEB-W) model—predicts reasonable cross sections for small atoms (H and He) and all studied molecules. This means that the hydrogenlike approximation for energy distribution of dipole oscillator strength gives averaged partial cross sections which summed up (over all target orbitals) provide total cross sections of the correct order of magnitude. On the other hand, for larger atoms Ne and Ar the exact energy distribution of oscillator strength is needed; thus a more accurate BED-W method should be rather used. It seems that BED-W describes much better the positron-impact ionization of Ne and Ar than the corresponding model for electron impact. Nevertheless, the large spread in available experimental and theoretical cross sections does not allow one to judge definitely the applicability of the present approach. It has to be added that for each specific target the BED-W model can be used to find an unknown constant of proportionality in the Wannier-type threshold law of Klar [40] and the energy range over which this law is valid once the reliable positron cross sections are available for comparison.

When comparing BED and BEB models (or BED-W with BEB-W) for the same target we found that in general BED predicts larger cross sections in the peak region than BEB. An exception to this rule is Ne, for which the BEB cross sections are higher. Both models approach each other at high incident energies where the dipole interaction is dominant. All these conclusions hold for both electron and positron-impact ionization.

The present approach still could be tested and possibly improved using different quantum-mechanical descriptions of studied targets—it is well known that both electron BED and BEB are very sensitive to the choice of the representation of target wave function [13], particularly to the value of average orbital kinetic energy. For positrons the sensitivity of the model to an accurate description of the target may be even more important than for electrons due to a delicate interplay between repulsive nuclear forces and attractive polarization interaction. Furthermore, all electron models following the original BED theory (e.g., Refs. [15–22]) can be also tested for positron-impact ionization using similar modifications as here.

From a practical point of view, the proposed model can be used to estimate positron direct ionization cross sections in media which are inaccessible to experiments, but important, for example, in modeling positron behavior in human tissues [36]. In particular, the BEB-W formula can greatly simplify extensive semiempirical procedures used to find

unknown ionization cross sections necessary for Boltzmann equation analysis or Monte Carlo modeling (see, for example, Refs. [36,37,105]). Finally, the positron and electron BED models provide simple tools to carry out tentative comparative studies on ionization by equivelocity particle and antiparticle impact.

ACKNOWLEDGMENTS

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APPENDIX A: BINARY-ENCOUNTER-DIPOLE-WANNIER MODEL

In order to account for strong polarization-correlation effects present just above the threshold in positron-impact ionization we have to include additional term $f(t)$ in the scaling denominator $t+u+1$ used originally in the BED equation [1]:

$$\sigma_i^{\mathrm{BED}}(t) = \frac{S}{[t+u+1+f(t)]}\left[\left(2-\frac{N_i}{N}\right)\left(1-\frac{1}{t}\right)+D_i(t)\ln t\right]. \tag{A1}$$

To find this extra term we consider the Wannier-type threshold law in Eq. (2) derived by Klar [40]. To match BED with this law we can look at BED behavior as $t \to 1$:

$$\sigma_i^{\mathrm{BED}}(t) = \frac{S}{[t+u+1+f(t)]}\left[\left(2-\frac{N_i}{N}\right)(t-1)+C'(t-1)^2\right], \tag{A2}$$

where we used the Taylor expansions around 1 of $(t-1)/t$ term inside the square bracket and numerical calculations showing that $D_i(t)\ln(t) \approx C'(t-1)^2$ at t close to 1. Here C' is a target-dependent constant. Since $C' < 1$ and $2 - N_i/N > 1$ for ionization from outer shells of all studied targets, the quadratic term inside the bracket becomes negligible as $t \to 1$. After such simplifications we can match the near threshold BED with the Wannier-type law [Eq. (2)]:

$$\frac{S}{[t+u+1+f(t)]}\left[\left(2-\frac{N_i}{N}\right)(t-1)\right] = C''(t-1)^{2.65}, \tag{A3}$$

where C'' is the unknown constant. From the latter equation the extra term $f(t)$ takes a form

$$f(t) = \frac{C}{(t-1)^{1.65}} - (t+u+1), \tag{A4}$$

where $C = S(2-N_i/N)/C''$. We found numerically that $(t+u+1)$ becomes negligible as $t \to 1$ for all studied targets when $C \approx 1$. Consequently, we can use the following form of the correction term:

$$f(t) = \frac{C}{(t-1)^{1.65}}. \tag{A5}$$

We found that, for such a choice of $f(t)$, the near-threshold BED (and BEB) model follows almost perfectly the Wannier-type law for energies up to 1 eV above the ionization threshold for all targets studied in this work.

APPENDIX B: COMBINING BINARY-ENCOUNTER THEORY WITH BETHE MODEL

In order to combine the semiclassical binary-encounter theory (BE) [2] with the quantum theory of Bethe [3] for the dipole interaction between target and fast incident charged projectiles, we assume that the differential cross sections (for ejecting an electron with kinetic energy w from an atomic-molecular orbital) has a general form given by Eq. (4):

$$\frac{d\sigma_i(w,t)}{dw} = S\sum_{n=1}^{3} F_n(t) f_n(w), \tag{B1}$$

where $f_n(w) = 1/(w+1)^n$. The integral (σ_i) and stopping cross sections (σ_{sti}) for ionization are defined as [1]

$$\sigma_i = \int_0^{t-1} \frac{d\sigma_i}{dw} dw, \tag{B2}$$

$$\sigma_{sti} = B\int_0^{t-1} (w+1)\frac{d\sigma_i}{dw} dw. \tag{B3}$$

In the BE theory, the $n=1$ term represents interference between scattered and ejected particles. The $n=2$ term arises from close collisions, while the $n=3$ term accounts for the broadening of the energy distribution due to the intrinsic momentum distribution of a bound electron being ionized. Kim and Rudd [1] showed that it is possible to find appropriate forms of F_1, F_2, F_3, and f_3 terms that allow one to match the asymptotic behavior (for $t \gg 1$) of $d\sigma_i/dw$, σ_i, and σ_{sti} with the Bethe theory. For positron-impact ionization the first term in the sum of Eq. (B1) disappears since no exchange effect is present, i.e., $F_1(t) = 0$.

Kim and Rudd [1] assumed that the third term dominates the asymptotic behavior of $d\sigma_i/dw$ in Eq. (B1). Under such assumption, both $F_3(t)$ and $f_3(w)$ can be found comparing asymptotic $d\sigma_i/dw$ with the Bethe theory:

$$\begin{aligned} d\sigma_i/dw &= SF_3(t)f_3(w), \quad \text{Eq. (B1) at high } t, \\ d\sigma_i/dw &= S\frac{\ln t}{t}\frac{1}{N(w+1)}\frac{df_i}{dw}, \quad \text{Bethe theory}, \end{aligned} \tag{B4}$$

where df_i/dw is the differential oscillator strength and S and N are defined in Eq. (1). Equating both expressions in Eq. (B4) gives

$$F_3(t) = \frac{\ln t}{t}, \quad f_3(w) = \frac{1}{N(w+1)}\frac{df_i}{dw}. \tag{B5}$$

The asymptotic ($t \gg 1$) forms of σ_i and σ_{sti} and the corresponding Bethe expressions are the following:

$$\begin{aligned} \sigma_i &= S(F_2 + F_3 G), \quad \text{Eq. (B2) at high } t, \\ \sigma_i &= S\frac{Q}{2}\frac{\ln t}{t}, \quad \text{Bethe theory}, \end{aligned} \tag{B6}$$

TABLE II. Fit parameters for partial photoionization cross sections [51,52].

Target	Orbitals	E_{th} (eV)	E_{max} (eV)	E_0 (eV)	σ_0 (10^{-22} m^2)	y_a	P	y_w	y_0	y_1
H	$1s$	13.6	5000	4.298×10^{-1}	5.475×10^{4}	3.288×10^{1}	2.963×10^{0}	0.000×10^{0}	0.000×10^{0}	0.000×10^{0}
He	$1s$	24.59	5000	1.361×10^{1}	9.492×10^{2}	1.469×10^{0}	3.188×10^{0}	2.039×10^{0}	4.434×10^{-1}	2.136×10^{0}
	$2p$	21.60	100000	2.000×10^{1}	1.691×10^{4}	2.442×10^{0}	1.043×10^{1}	0.3345×10^{0}	0.000×10^{0}	0.000×10^{0}
Ne	$2s$	48.47	100000	3.204×10^{1}	5.615×10^{1}	5.808×10^{0}	6.678×10^{0}	0.000×10^{0}	0.000×10^{0}	0.000×10^{0}
	$1s$	866.9	100000	3.144×10^{2}	1.664×10^{1}	2.042×10^{5}	0.845×10^{0}	0.000×10^{0}	0.000×10^{0}	0.000×10^{0}
	$3p$	15.82	249.2	1.709×10^{1}	2.106×10^{1}	2.645×10^{2}	4.796×10^{0}	4.185×10^{-1}	1.688×10^{0}	8.943×10^{-1}
		249.2	100000	3.854×10^{1}	4.872×10^{1}	2.640×10^{1}	6.662×10^{0}	0.2355×10^{0}	0.000×10^{0}	0.000×10^{0}
Ar	$3s$	29.24	100000	2.525×10^{1}	6.394×10^{0}	1.700×10^{2}	04.223×10^{0}	0.000×10^{0}	0.000×10^{0}	0.000×10^{0}
	$2p$	249.18	100000	1.647×10^{2}	8.372×10^{1}	5.452×10^{1}	3.328×10^{0}	0.627×10^{0}	0.000×10^{0}	0.000×10^{0}
	$2s$	326	100000	1.302×10^{2}	9.185×10^{0}	2.693×10^{1}	4.021×10^{0}	0.000×10^{0}	0.000×10^{0}	0.000×10^{0}
	$1s$	3202.9	100000	1.135×10^{3}	4.280×10^{0}	3.285×10^{7}	0.7631×10^{0}	0.000×10^{0}	0.000×10^{0}	0.000×10^{0}

$$\sigma_{sti} = S\frac{B}{R}(F_2\ln t + F_3 H), \quad \text{Eq. (B3) at high } t,$$
$$\sigma_{sti} = S\frac{2B}{R}\frac{\ln t}{t}, \quad \text{Bethe theory,} \tag{B7}$$

where

$$Q = \frac{2BM_i^2}{NR}, \quad \text{with} \quad M_i^2 = \frac{R}{B}\int_0^\infty \frac{1}{(w+1)}\frac{df_i}{dw}dw,$$
$$G = \int_0^\infty f_3(w)dw, \quad H = \int_0^\infty (w+1)f_3(w)dw. \tag{B8}$$

Here Q is the dipole constant appearing in Eq. (12). The upper limits of the integration for G and H have been extended to ∞ in the anticipation that $f_3(w)$ diminishes rapidly enough as $w\to\infty$ such that the asymptotic part of $f_3(w)$ does not contribute to the asymptotic t dependence of Eqs. (B6) and (B7).

Using $f_3(w)$ from Eq. (B5) we can express G and H as

$$G = \frac{1}{N}\int_0^\infty \frac{1}{w+1}\frac{df_i}{dw}dw = \frac{BM_i^2}{RN},$$
$$H = \frac{1}{N}\int_0^\infty \frac{df_i}{dw}dw = \frac{N_i}{N}. \tag{B9}$$

Equating both expressions for σ_{sti} in Eq. (B7) and using the results of Eqs. (B8) and (B9), we can obtain the following form of F_2:

$$F_2(t) = \frac{2 - N_i/N}{t}. \tag{B10}$$

The latter expression diminishes more rapidly than $F_3 G$ as $t\to\infty$ and consequently also the asymptotic form of σ_i in Eq. (B6) becomes compatible with the Bethe theory at high incident energies. Finally, we can combine F_n and f_n with the symmetric binary-encounter cross section by replacing the kinetic energy in the denominators of F_2 and F_3 with the effective energy seen by the target electron, i.e., $t\to(t+\gamma)$, where γ is defined by Eq. (3):

$$F_2(t) = \frac{2 - N_i/N}{(t+\gamma)}, \quad F_3(t) = \frac{\ln t}{(t+\gamma)},$$
$$f_3(w) = \frac{1}{N(w+1)}\frac{df_i}{dw}. \tag{B11}$$

APPENDIX C: DIFFERENTIAL DIPOLE OSCILLATOR STRENGTH

Binary-encounter-dipole model [BED; see Eq. (9)] requires reliable differential dipole oscillator strengths (df_i/dw) for each subshell of the target. In this paper we use analytical fits to partial photoionization cross sections averaged over resonance structures reported by Verner *et al.* [51,52] for atomic targets (H, He, Ne, and Ar). In both papers the following fitting expression was used:

$$\sigma(E) = \sigma_0\left[(x-1)^2 + y_w^2\right]y^{0.5P-5.5l} \times\left(1+\sqrt{1+\frac{y}{y_a}}\right)^{-P}[10^{-22}\text{ m}^2],$$
$$x = \frac{E}{E_0} - y0, \quad y = \sqrt{x^2+y_1^2}, \tag{C1}$$

where E is the photon energy in eV, $l = 0, 1, 2$ (or s, p, d) is the subshell orbital quantum number, and E_0, σ_0, y_a, P, y_0, and y_1 are the fit parameters.

The first Verner paper [51] provides fits to the partial Hartree-Dirac-Slater (HDS) photoionization cross sections for the ground-state shells of all atoms. The second paper [52] corrects the previous parametrization for outer shells using more accurate R-matrix calculations at low energies, just above ionization thresholds. R matrix is considered to be more reliable than HDS in the near threshold region where resonances are present. At higher energies, the latter fits reproduce calculated HDS photoionization cross sections. We use the parametrization from the second paper for all atomic targets: H, He, and Ar except Ne. In the latter case we found that there must be an unintended error in the fit parameters published in Ref. [52] because they do not give [using Eq. (C1)] correct photoionization cross sections when compared with literature data [106]. Therefore, we use fit parameters from Ref. [51] for the Ne atom, which gives cross sections in good agreement with experiments over the entire energy range except a very narrow range near the ionization threshold. Values of all fit parameters used in this work are given in Table II.

Photoionization cross sections (σ) are related to the differential dipole oscillator strength by the

expression [107]

$$\frac{df_i}{dE}[\mathrm{eV}^{-1}] = \frac{\sigma}{109.75}[10^{-22}\ \mathrm{m}^2]. \tag{C2}$$

The dipole oscillator strength (df_i/dw) as a function of normalized kinetic energy of ejected electrons $w = E/B - 1$, as it occurs in Eq. (9), is obtained by multiplying df_i/dE by orbital binding energy B:

$$\frac{df_i}{dw} = B\frac{df_i}{dE}. \tag{C3}$$

The dipole oscillator strength for ionization of $1\sigma_g$ orbital of molecular hydrogen (H_2) is calculated using the power-series fit by Kim and Rudd [1]:

$$\frac{df_i}{dw} = \frac{1.1262}{(w+1)^3} + \frac{6.3982}{(w+1)^4} - \frac{7.8055}{(w+1)^5} + \frac{2.1440}{(w+1)^6}. \tag{C4}$$

This formula is in good agreement with experimental total photoionization cross sections [106].

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