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Influence of the electron binding energy in the distortion of the initial state in ion-atom collisions

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In this work, we propose a modified continuum distorted wave (CDW) theory for ion-atom ionization that incorporates an electron effective-mass model for the bound electron-projectile distortion in the entrance channel. Distorted wave theories have been shown to be adequate when the projectile impact energy is high enough but, as the projectile velocity decreases, both CDW and continuum distorted wave–eikonal initial state (CDW-EIS) approximations exhibit remarkable discrepancies when compared with the available experimental data, in particular for highly charged ion impact. Our modified CDW approximation incorporates the active electron binding energy in the distorted wave description of the initial state. We found very good agreement with experimental data in a single ionization doubly differential cross section for multiply charged projectiles and impact energies as low as 50 keV/amu.

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I. INTRODUCTION

The study of electron emission spectra in ion-atom collisions has been a field of intense activity for years [1]. Single ionization for intermediate to high impact energy has been the object of considerable theoretical efforts, particularly focused in the so-called two-center electron emission (TCEE) [2,3]. Improvement in the description of the ionized electron moving in the presence of both residual target and projectile fields after the collision (final state) has been key for the correct description of experimental data [4]. Dynamic correlation effects in the final state of the three particles involved in the collision (electron, target, and projectile) have also been recognized to play an important role in the ionization dynamics [5–11]. But when we deal with this kind of final-state wave function, the integrals resulting from the transition amplitudes are usually nonseparable and involve numerical multidimensional integration. However, even present sophisticated correlated theories have not been shown to be adequate enough when dealing with single ionization by multiply charged projectiles.

On the other hand, the influence of the initial-state quality in atomic ionization by ion impact is not completely clear yet. Within distorted wave approximations, it has been shown that, at least for high impact energy and multiply charged projectiles, the continuum distorted wave (CDW) theory of Belkic [12] used together with a full numerical description of the initial bound and final continuum electron states yields arguably the best results available to date [13]. However, when the projectile impact velocity decreases, the continuum distorted wave–eikonal initial state (CDW-EIS theory of Crothers and McCann [14] yields better results, its only difference being the choice of the initial state. Moreover, the CDW-EIS is formally free of criticisms regarding the initial-state proper normalization [15], and the transition amplitudes do not have the divergent behavior that CDW exhibits (although afterwards it has been demonstrated that the CDW amplitudes are integrable and its doubly differential cross sections are well behaved [16,17]). However, while CDW-EIS outperforms CDW for intermediate energy and simply charged projectiles, its agreement with experimental data is not so good for multiply charged ion-impact ionization [2,4].

In the analysis of proton-hydrogen excitation, Dewangan and Brandsen [18] showed that the use of the CDW wave function for the initial state almost yields a fully closed second-Born term when the scattering amplitude is calculated using the Born approximation for the final state. Accordingly, Dewangan [19], based on the VPS formulation [20], proposed a "two Coulomb waves" (TCW) approximation for the initial state that does lead to a fully closed second-Born term.

Within distorted wave theories, the initial-state projectile–electron distortion is usually derived assuming (a) that the electron is at rest in the target reference frame, and (b) that the electron interacts with the incoming projectile only. In this way, a simple Coulomb type distortion is obtained (CDW or EIS distorsions). However, both hypotheses are high-energy approximations, the first one neglecting the electron orbital velocity when compared with the impinging ion velocity, and the second one ignoring the fact that electron distortion is modified by the binding nature of the electron-nucleus interaction, as well as the fact that the bound state is distorted by the projectile.

Some of these effects have already been considered in the impulse approximation (IA) of Miraglia and Macek [21], which takes into account the velocity distribution of the active electron in the initial-state projectile-electron distortion.

In ionization of atoms by swift electrons, the description of both initial and final collision states has had a more systematic development, probably due to the early experimental data available [22]. Initial-state binding-energy effects have

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been taken into account in an empirical way by considering an effective mass for the active electron. In some models, the whole nucleus mass has been added to the electron's own [23,24]. In these schemes, however, no account has been taken of the projectile velocity dependence of the effective mass. However, it is accepted that binding-energy effects should vanish for increasing impact energy, where the target electron can be considered as quasifree for the purpose of the distorsion calcuation. Recently, electron binding-energy effects have also been taken into account in photo double ionization of He yielding excellent agreement with experimental data [25].

One alternative way to consider the fact that the electron is always under the influence of both projectile and target-ion potential is to explicitly include part of the nonorthogonal kinetic energy operator in the calculation of the initial-state wave functions. This approach leads to the inclusion of dynamic correlation effects between the projectile-electron and target nucleus–electron interactions. Our previous attempts to incorporate these effects in the initial state have shown encouraging results, but the physics involved in this approach must be further tested [26]. One of the main drawbacks of pursuing this approach is that wave-function separability breaks down, and transition amplitudes cannot be analytically calculated.

How can the fact that the electron is *also* in a bound state be taken into account in the initial-state distortion without destroying wave-function separability?

One possible answer to this question is, by *hiding* the target nucleus–electron interaction behind an electron *effective* mass, used in the computation of the distortion. We will use the Wiezsäcker and Williams theory of virtual photons [27], together with the theory of linear response [28], in order to estimate an active electron effective mass for the distortion calculation and its dependence on the projectile velocity. We will use this initial-state distortion in a CDW-type theory and apply it in multiply charged ion impact ionization of helium at intermediate energy.

The paper is organized as follows. In the next section, we describe the theory and models that we use for our calculations. In Sec. III, we compare our model with several sets of experimental data and with standard distorted waves theories. Finally, we draw our main conclusions showing the advantages and disadvantages that we have found in our approximation. Atomic units are used throughout this work unless otherwise stated.

II. THEORY AND MODELS

Let us consider the doubly differential cross section (DDCS) for electron emission in ion-atom collisions. We choose the straight line impact parameter based formalism to describe the collision process [29]. DDCS for ionization arises from the integration of the electronic transition amplitude, $\mathcal{A}_{if}$, over the impact parameter $\boldsymbol{\rho}$,

$$\frac{d^2\sigma}{dEd\Omega} = \int |\mathcal{A}_{if}(\boldsymbol{\rho})|^2 d\boldsymbol{\rho}. \tag{1}$$

$\mathcal{A}_{if}(\boldsymbol{\rho})$ is given in its post version as

$$\mathcal{A}_{if}(\boldsymbol{\rho}) = -\mathrm{i}\int_{-\infty}^{+\infty} dt \left\langle \Phi_f^- \left| \left(H_{el} - \mathrm{i}\frac{\partial}{\partial t}\right)^\dagger \right| \Psi_i^+ \right\rangle, \tag{2}$$

where Ψ_i^+ represents the initial electronic state, *exact* solution for H_{el} with initial conditions

$$\lim_{t\to-\infty} \Psi_i^+ = \Phi_i^+. \tag{3}$$

In the same way, we can define the prior version of $\mathcal{A}_{if}(\rho)$, given by

$$\mathcal{A}_{if}(\boldsymbol{\rho}) = -\mathrm{i}\int_{-\infty}^{+\infty} \mathrm{dt} \left\langle \Psi_f^- \left| \left(H_{el} - \mathrm{i}\frac{\partial}{\partial t}\right) \right| \Phi_i^+ \right\rangle, \tag{4}$$

where Ψ_f^- representsthe final electronic state, *exact* solution for H_{el} with asymptotic conditions

$$\lim_{t\to-\infty} \Psi_f^- = \Phi_f^-. \tag{5}$$

As for the electronic Hamiltonian H_{el}, it reads

$$H_{el} = -\frac{1}{2}\nabla_r^2 + V_T(r_T) - \frac{Z_P}{r_P} + \frac{Z_P Z_T}{R}. \tag{6}$$

Φ_i^+ and Φ_f^- are solutions of the initial and final Hamiltonian, respectively. For hydrogenic targets, we have

$$V_T = -\frac{Z_T}{r_T}.$$

On the other hand, for multielectronic targets, several approximations involving model potentials are usually employed, ranging from effective charge Coulomb potentials that take passive electron screening into account, to fully numerical potentials and other more sophisticated approaches. In this paper, however, we will stick to the simpler Roothan-Hartree-Fock model for helium [30], which, used in distorted wave theories for ion-atom collisions, leads to very good agreement in DDCSs with experimental data for high impact energy, with a remarkable decreasing in the complexity of the expressions involved [31].

By selecting different distorted wave functions and their corresponding distortion potentials, it is possible to obtain a whole range of approximations to the transition amplitude. Making the approximations $\Psi_i^+ \approx \chi_i^+$ in Eq. (2) and $\Psi_f^- \approx \chi_f^-$ in Eq. (4), we get the first order of a distorted wave series. The distorted wave functions χ_i^+ and χ_f^- can be factorized as follows:

$$\chi_i^+(\mathbf{r},t) = \Phi_i(\mathbf{r},t)\mathcal{L}_i^+(\mathbf{r}_p)$$

and

$$\chi_f^-(\mathbf{r},t) = \Phi_f(\mathbf{r},t)\mathcal{L}_f^-(\mathbf{r}_p),$$

where $\Phi_i(\mathbf{r},t)$ and $\Phi_f(\mathbf{r},t)$ are the undistorted asymptotic states [2], and we are free to choose $\mathcal{L}_i^+(\mathbf{r}_P)$ and $\mathcal{L}_f^-(\mathbf{r}p)$, as long as the resulting distorted waves comply with the correct asymptotic conditions (3) and (5), respectively, and the resulting perturbation potential is a short-range potential.

The CDW approximation was originally developed by Cheshire [32] and first applied by Belkic [12] for ion-atom ionization. Within this theory, the initial-state distortion reads

$$\mathcal{L}_i^{+CDW}(\mathbf{r}_P) = N(\alpha_i)\,{}_1F_1(i\alpha_i;1;iv_P r_P + i\mathbf{v}_P\cdot\mathbf{r}_P) \tag{7}$$

with

$$\alpha_i = \frac{Z_P}{v_P} \tag{8}$$

and the corresponding short-range perturbation potential reads

$$W_i^{CDW}\chi_i^{+CDW} = \Phi_i\Big[\vec{\nabla}_{\mathrm{r}_T} ln\varphi_i(r_T)\cdot\vec{\nabla}_{\mathrm{r}_P}\mathcal{L}_i^{+CDW}(\mathbf{r}_P)\Big]. \tag{9}$$

For the final state, we have

$$\chi_f^{-CDW}(\mathbf{r},t) = \Phi_f(\mathbf{r},t)\mathcal{L}_f^{-CDW}(\mathbf{r}_P), \tag{10}$$

where the distortion is

$$\mathcal{L}_f^{-CDW}(\mathbf{r}_P) = N(\alpha_P)\,{}_1F_1(-i\alpha_P;1;-ik_P r_P - i\mathbf{k}_P\cdot\mathbf{r}_P), \tag{11}$$

where

$$\alpha_P = \frac{Z_P}{k_P}. \tag{12}$$

With this election for the final wave function, the Redmond asymptotic conditions for the three-body Coulomb problem, Eqs. (5) and (3), are satisfied [33]. The short-range perturbation potential W_f^{CDW} reads in this case

$$W_f^{CDW}\chi_f^{-CDW} = \Phi_f\Big[\vec{\nabla}_{\mathbf{r}_T}\ln{}_1F_1(-i\alpha_T;1;-ik_T r_T - i\mathbf{k}_T\cdot\mathbf{r}_T)\cdot\vec{\nabla}_{\mathrm{r}_P}\mathcal{L}_f^{-CDW}(\mathbf{r}_P)\Big]. \tag{13}$$

We will use the CDW approximation in its post version for our calculations. In order to take into account the binding-energy effect for the active electron, we propose a change in the initial-state distortion. We note that $\mathcal{L}_i^{+CDW}(\mathbf{r}_P)$ takes into account the projectile-electron interaction as a pure two-body system, i.e., as if the electron were free. Our aim is to explore how the initial-state distortion changes due to the fact that the electron is actually in a bound state, and we will do so by introducing an effective mass for the electron in the initial-state distortion.

Effective-mass model

Let us look again at the collision process for ion-atom ionization, but from a classical electrodynamics point of view. During the collision, the active electron experiences the combined effect of the projectile and nucleus electric fields. It is well known that in a high-energy regime, the electric field produced by the swift incoming projectile at the target position can be assimilated to an equivalent radiation field [34]. The frequency spectra for this virtual photon field can be easily calculated by merely considering the ion's field Fourier transform at the target position.

The electric field $\mathbf{E}$ for an atomic system located at the origin of coordinates produced by a particle with charge Z and velocity v with impact parameter ρ reads

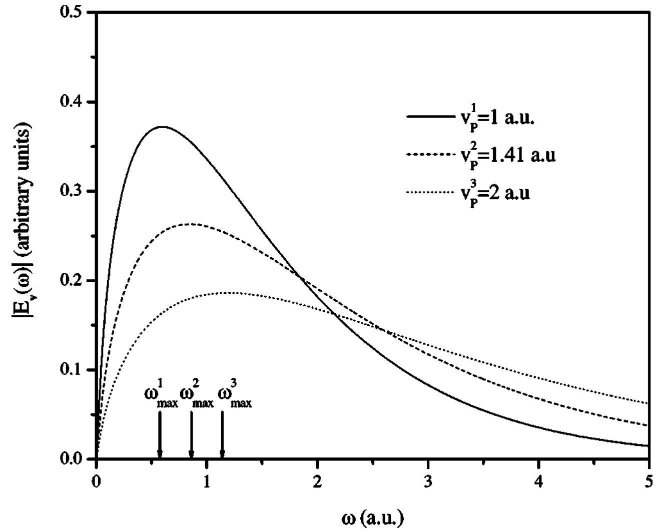

FIG. 1. Spectral descomposition of the incident ion electric field at the target position as a function of its velocity. Impact parameter is taken as 1 a. u. The arrows indicate the positions of the field spectral maximum, showing how it is shifted as the impact velocity increases.

$$\mathbf{E}(t) = -\frac{Z\gamma vt}{(\rho^2+\gamma^2v^2t^2)^{3/2}}\hat{\mathbf{e}}_{\mathbf{v}} + \frac{Z\rho}{(\rho^2+\gamma^2v^2t^2)^{3/2}}\hat{\mathbf{e}}_{\mathbf{v}\perp}, \tag{14}$$

where $\gamma=(1-v^2/c^2)^{-1/2}$ and $\hat{\mathbf{e}}_{\mathbf{v}}$ and $\hat{\mathbf{e}}_{\mathbf{v}\perp}$ are the directions parallel and perpendicular to the velocity vector $\mathbf{v}$, respectively. Since we are working in a nonrelativistic domain, we take $\gamma\approx 1$. The electric field Fourier transform yields

$$\mathbf{E}(\omega) = -i\frac{Z}{v\rho}\left(\frac{2}{\pi}\right)^{1/2}\left[\frac{\omega\rho}{v}K_0\left(\frac{\omega\rho}{v}\right)\right]\hat{\mathbf{e}}_{\mathbf{v}} + \frac{Z}{v\rho}\left(\frac{2}{\pi}\right)^{1/2}\left[\frac{\omega\rho}{v}K_1\left(\frac{\omega\rho}{v}\right)\right]\hat{\mathbf{e}}_{\mathbf{v}\perp}, \tag{15}$$

where K_0 and K_1 are modified Bessel functions. In Fig. 1, we show typical shapes for the longitudinal component (i.e., in the $\mathbf{v}$ direction) of the field spectra. We observe that the field spectrum shifts towards higher frequencies for higher projectile impact velocities. We need now to link the bound electron behavior subject to this field. This we do by using a linear-response approximation for the electron classical motion.

Let us consider the bound electron as a simple harmonic oscillator, characterized by its natural frequency, ω_0. The effect of the impinging ion will be to add an external force described by the electric field considered previously.

The equation governing the classical motion of a harmonically bound electron subject to a time-dependent external electric field is

$$\frac{d^2r}{dt^2} = -\omega_0^2 r - \Gamma\frac{dr}{dt} + E(t). \tag{16}$$

By using a standard linear-response theory, the effective mass β which links the external force to the electron acceleration, reads

$$\beta = 1 + \frac{i\Gamma\omega - \omega_0^2}{\omega^2}. \tag{17}$$

This effective mass will depend on the external force frequency. For high enough frequency, we expect the electron to behave as if it were free, so its effective mass will simply become its ordinary mass (i.e., $\beta = m_e = 1$ a.u.). On the other hand, if we pull the electron with a constant force (i.e., $\omega = 0$), we will actually be pulling the entire atom (at least in this harmonic model of our atomic system) so the electron effective mass would be the whole atom mass, i.e., $\beta \to m_e + M_T$. As we take an infinite mass approximation for the nucleus, in fact, $\beta \to \infty$. For frequency regions close to resonance conditions, strange behaviors are to expected for this classical model, since the effective mass need not be positive, or real, or even finite. Much of this behavior is not expected in our real system. Thus it can be removed from our model by including an "unphysical" drag force which quenches the resonant behavior that might appear. Furthermore, we will only be interested in $|\beta|$, since this describes the inertial properties of the bound electron. β's phase, on the other hand, describes the phase difference between the external force applied and the classical electron acceleration, which is not relevant in our particular system [28].

We must now put together the effective-mass dependence on frequency with the corresponding spectrum for the incident ion electric field. We take here as simple an approach as possible, since our effective-mass model is just a first approximation. As we are interested in the binding effects when the incoming projectile is far away, the transversal component of its electric field at the target position may be neglected, i.e., we will only take into account the longitudinal component of Eq. (14). Furthermore, we will just take the dominant frequency from the whole spectrum thus effectively replacing the incident ion field by a monochromatic photon field. In this way, we are able to build a simple projectile velocity dependent effective mass. In Fig. 2, we show $|\beta|$ as a function of the ion impact velocity, for different values of Γ. We introduce this electron effective mass in the calculation of the initial channel electron-projectile distortion, labeling the resulting theory βCDW,

$$\mathcal{L}_i^{+\beta CDW}(\mathbf{r}_P) = N(\alpha_i)\,{}_1F_1(i\alpha_i;1;i|\beta|v_P r_P + i|\beta|\mathbf{v}_P\cdot\mathbf{r}_P). \tag{18}$$

In this way we take into account, at least partially, the influence of the target nucleus in the projectile-electron interaction in the initial state, while retaining the separability of the standard CDW approach. We are aware of the crudeness of the approximations made in our semiclassical model. But it is interesting to show that even this draft model can describe the fact that the electron effective mass must depend on the projectile velocity, in such a way that for very high impact energy a quasifree model for the bound electron–incoming projectile distortion is recovered and, as the impact energy decreases, the electron binding energy is taken into account in the initial-state distortion in a simple way.

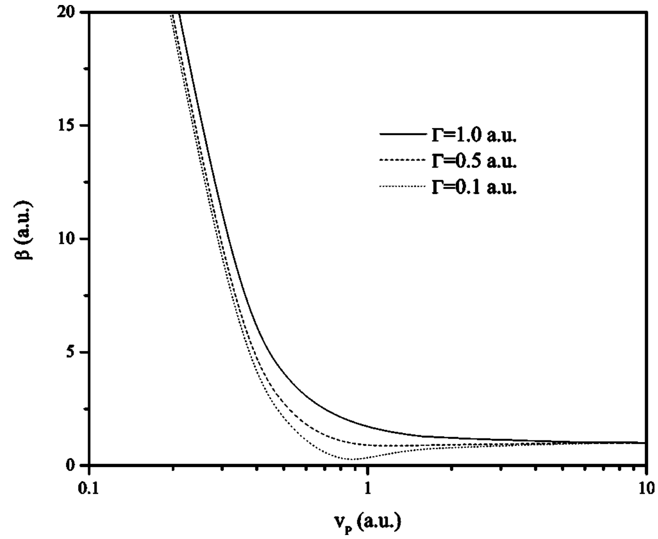

FIG. 2. Absolute value for active electron effective mass $|\beta|$ as a function of the projectile velocity as calculated from linear-response theory, for different damping coefficient Γ. For actual calculations, Γ was taken large enough to quench classical resonant behavior.

III. RESULTS AND COMPARISONS

We have evaluated the DDCS for proton and highly charged ion impact ionization of helium using the βCDW approximation outlined above. We show emission spectra in the forward direction, where we can explore the so-called two-center emission region [2]. This is one of the regions where standard distorted wave theories like CDW and CDW-EIS show larger discrepancies with experimental data. In Fig. 3, we see F^{9+} impinging over He at 1.5 MeV. βCDW reproduces the full numerical CDW results [13], since $|\beta|$

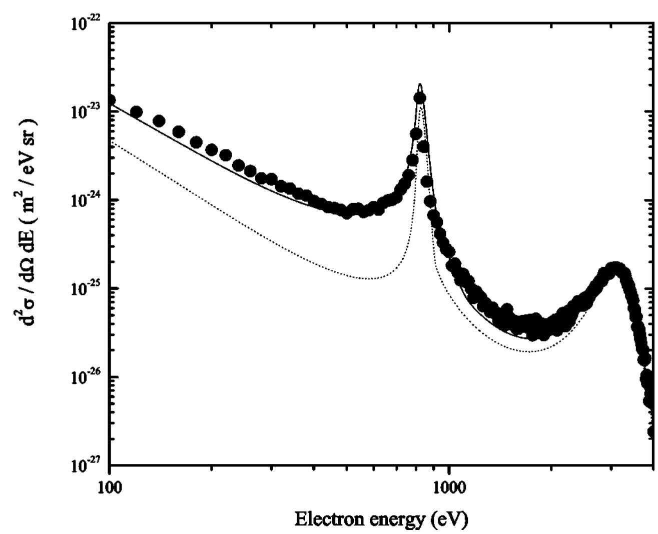

FIG. 3. Doubly differential cross section (DDCS) for single ionization in the forward direction for 1.5 MeV F^{9+} impact on He. Solid line corresponds to present βCDW theory; dotted line corresponds to CDW-EIS calculations; solid circles are experimental data [35].

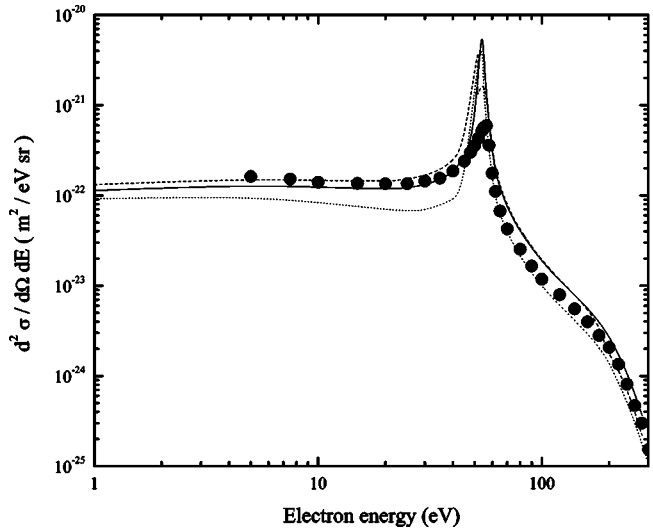

FIG. 4. Same as Fig. 3 for 100 keV H^+ impact on He. Solid line corresponds to present βCDW theory; dashed line corresponds to CDW theory; dotted line corresponds to CDW-EIS calculations; solid circles are experimental data [36].

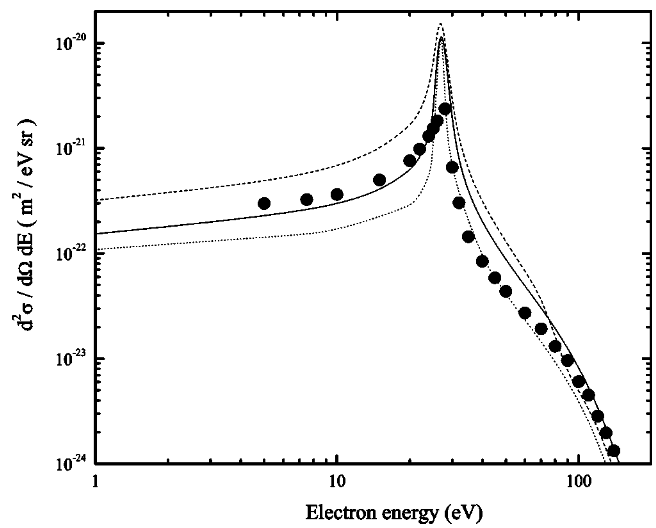

FIG. 6. Same as Fig. 4 for 50 keV H^+ impact on He.

$\simeq 1$ for that impact energy, and agrees very well with experimental data available [35], whereas the CDW-EIS approach underestimates the experimental data, especially in the region between the soft electron (SE) and the electron capture to the continuum (ECC) peaks.

In Figs. 4–7, we compare CDW with CDW-EIS and βCDW for H^+ and $^3He^{2+}$ at 100 and 50 keV impinging over He. For those impact energies, our model yields $|\beta| \simeq 1.5$ and $|\beta| \simeq 2.5$, respectively. We get a very good agreement with experiments, particularly for the two-center emission region. Even when the effective mass does not depend on the projectile charge, our model yields very good results for the He^{2+} projectile. It is known that CDW-EIS theory underestimates the experimental data for that energy range, particularly for emission velocity lower than v_P in the forward direction, while CDW overestimates the experimental data in the same region. We understand that CDW-EIS failure is due to the partial nature of the distortion effects in the initial-state wave function, particularly for multiply charged ions. On the other hand, CDW overestimation can be understood considering that the bound electron is treated always as free in the initial-state projectile-electron distortion, i.e., the resulting distortion is exaggerated when the impact energy decreases. In that sense, βCDW can be considered as an intermediate one between the CDW and CDW-EIS because the intensity of the initial-state distortion is gradually lowered as the impact energy decreases. We have in some way the advantages of both theories, i.e., the correct "weight "for the electron-projectile interaction but using the complete Coulomb function for the distortion, which grants a better description when the charge of the impinging projectile is increased. All approximations tested show similar results for emission velocity higher than v_P. This is to be expected since as we approach the BE peak region, distortion effects are less important.

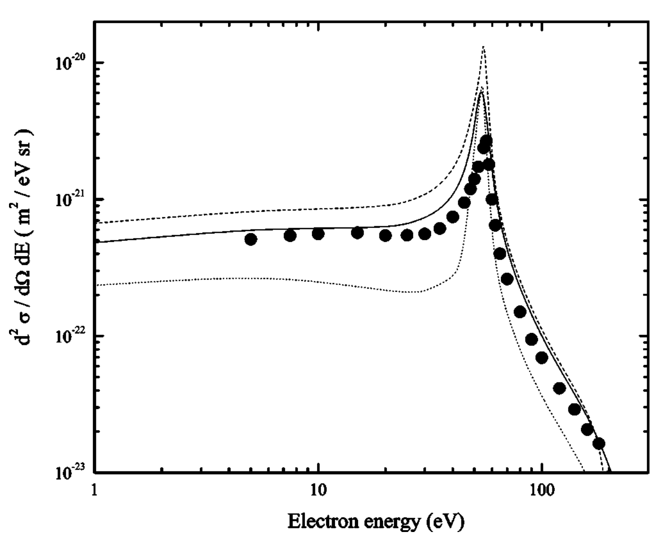

FIG. 5. Same as Fig. 4 for 100 keV $^3He^{2+}$ impact on He.

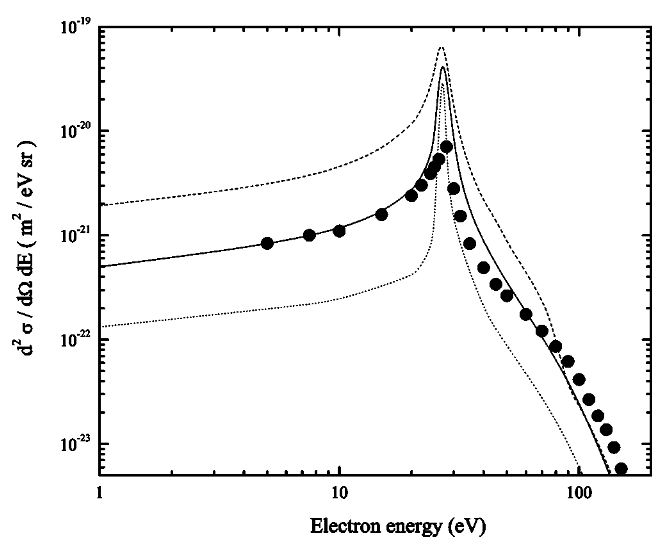

FIG. 7. Same as Fig. 4 for 50 keV $^3He^{2+}$ impact on He.

IV. CONCLUSIONS

We have presented a simple classical model based on linear response to include binding-energy effects in the entrance channel for ion-atom ionization. We have used the resulting theory for helium single ionization by singly and multiply charged ions. Our results are in very good agreement for impact energies as low as 50 keV, stretching the validity region of distorted wave theories. We confirm that distorted wave theories are quite sensitive to the choice of the initial state, and that binding-energy effects for the active electron should be taken into account in initial-state distortions, at least for intermediate impact energies. Our simple model does not include an effective-mass dependence on the projectile charge, but the results presented here do not call for any additional dependence beyond what is already contained in the standard CDW distortion. Energy binding effects should of course be present in other ion-atom collision processes, such as capture and excitation. We are testing now βCDW for capture and excitation. Since in these processes the active electron ends in a bound state, its effective mass needs to be taken into account in both entrance and exit channels.

ACKNOWLEDGMENTS

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