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Modelling of polarization and correlation effects in the ionization of helium by antiprotons

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Abstract

The ionization of helium atoms by antiprotons is considered in an independent-particle model (IPM) with a model potential that accounts for polarization and binding effects of the electrons in the two-centre Coulomb potential. The model is designed to reproduce the exact ionization potential of the quasimolecular system and to allow for the existence of one bound state in the united-atom limit. We find a single-ionization cross section that is in good agreement with the results of various correlated two-electron calculations, and a double-ionization cross section that is considerably improved compared to a standard IPM calculation with a fixed atomic target potential.

1. Introduction

Collisions between antiprotons and atoms or molecules have received considerable attention since the publication of the first experimental ionization cross sections in 1986 [1]. One point of interest is the shape of the total double-ionization cross section at high impact energies $E_P \geqslant 0.5$ MeV as a function of E_P, and the fact that it differs from the one induced by protons (for a review of the early work on this topic see [2]). It is now agreed upon that the impact energy dependence of the antiproton double-ionization cross section at high E_P can only be described by theory when electron correlation at least in the initial state of the atom is taken into account [3, 4]. The difference between the antiproton and the proton data has also been attributed mainly to electron correlation in the initial and final states [4], an interpretation which was first suggested on the basis of perturbation theory [5]. Very recently this explanation has been put into question [3].

The experimental investigations of antiproton impact ionization were extended to lower projectile energies in recent years, which encouraged theorists to develop nonperturbative descriptions of single and multiple ionization, in which the electron–electron interaction is not only taken into account in the initial and final states, but also *during* the collision. For helium targets it was found that the experimental single-ionization cross section decreases steeply in

the low-energy region [6]. Subsequently, several correlated two-electron calculations were undertaken to describe these data [7–11], but none of them reproduces the experimental cross section below 30–40 keV.

The first such set of calculations was carried out by Reading and co-workers in the framework of the forced-impulse method (FIM), in which the electron–electron interaction is turned on at a few discrete timesteps during the propagation of the electronic wavefunction [7]. In each of the segments between these so-called cuts the time-dependent Schrödinger equation is solved in an independent particle model (IPM) using a single-centred atomic orbital basis. In [8] the authors state that convergence with respect to the number of cuts has been reached in a 16-segment calculation. Their results are in good agreement with experiment in the case of double ionization over the entire range of impact energies covered by the latest and by earlier measurements (see also [12]). However, they are larger than the experimental cross sections for single ionization at energies below 40 keV. Recently, two other groups reported coupled-channel results for single ionization of helium using different single-centred expansions of the two-electron wavefunction [9–11]. Both groups found only a weak dependence of their results with respect to variations in the basis sets and concluded that convergence has been achieved within a few per cent. The calculations essentially reproduce the single-ionization results of the multi-cut FIM reported in [8], thus confirming the discrepancy with the experimental data at low impact energies.

Low-impact-energy ionization in ion–atom collisions can be understood from consideration of the correlation diagram of the corresponding quasimolecular system. In the case of ($\bar{\text{p}}$He) all excited states of the helium atom merge with the single-electron continuum when the distance R between the antiproton and the He nucleus decreases, whereas the adiabatic ground-state energy level lies below the threshold of single ionization for all R [13, 14]. The united-atom limit $R \to 0$ corresponds to the formation of an H^- ion which is stable. Qualitatively, these properties explain why the ionization cross section should be small at low collision energies. It is, however, not clear, up to which energies the adiabatic stabilization of the electrons in the two-centre Coulomb potential is relevant for the collision dynamics.

In this paper we address this question in an IPM calculation designed to account for the adiabatic properties described above. We have constructed an effective single-particle potential that includes polarization and binding effects as a function of R and allows for the existence of one bound state in the united-atom limit $R \to 0$. The model potential is then used in time-dependent collision calculations for single and double ionization in the usual IPM framework. Our motivation is two-fold: on the one hand the accurate propagation of single-particle dynamics is easier to achieve than the correct evolution of a time-dependent two-electron system; on the other hand we wish to investigate which aspect of the two-electron correlation problem dominates the physics. The details of our procedure are described in section 2. In section 3 we discuss our results, and we offer some concluding remarks in section 4. Atomic units ($\hbar = m_e = e = 1$) are used throughout.

2. Theory

We assume that the antiproton moves on a classical straight line trajectory $\boldsymbol{R}(t) = (b, 0, v_P t)$ with impact parameter b and constant velocity v_P, and solve the single-particle equation

$$\mathrm{i}\partial_t \psi(\boldsymbol{r}, t) = \hat{h}(t)\psi(\boldsymbol{r}, t) \tag{1}$$

with the Hamiltonian

$$\hat{h}(t) = -\frac{1}{2}\Delta + v_{\text{atom}}(r) + v_{\text{mod}}(\boldsymbol{r}, R(t)) + \frac{1}{|\boldsymbol{r} - \boldsymbol{R}(t)|}. \tag{2}$$

In equation (2) $v_{\rm atom}$ denotes the Hartree–Fock ground-state potential of the helium atom[1]

$$v_{\rm atom}(r) = -\frac{2}{r} + \int \frac{|\varphi_{1s}(r')|^2}{|\boldsymbol{r} - \boldsymbol{r}'|}\,{\rm d}^3r', \tag{3}$$

and $v_{\rm mod}$ is the model potential that accounts for the adiabatic response of the two-electron system in the presence of the antiproton. We assume that the electron cloud tries to avoid the antiproton by occupying a superposition of p states, thereby acquiring a non-zero dipole moment. This gives rise to a dipole potential. At the same time the spherical part of the potential is modified to account for the increased binding compared to the electrostatic potential of the atomic ground state. We make the ansatz

$$v_{\rm mod}(\boldsymbol{r}, R) = v_{\rm mono}(r, R) + v_{\rm dip}(\boldsymbol{r}, R) = -a(R)\exp(-r) - \frac{p(R) r\cos\theta}{[d(R)]^3 + r^3}, \tag{4}$$

and choose the parameters $p(R)$, $d(R)$, and $a(R)$ such that the binding energy of the lowest single-particle level in the adiabatic eigenvalue problem

$$\hat{h}(R)|\psi_0(R)\rangle = \varepsilon_0(R)|\psi_0(R)\rangle \tag{5}$$

reproduces the ionization potential of the quasimolecular system as a function of R. We note that such a procedure cannot be unique, since we use only one condition to specify three parameters for each value of R. However, our results to be discussed in section 3 indicate that the ionization potential is mainly determined by the monopole potential $v_{\rm mono}$ and thus by $a(R)$, whereas it is relatively insensitive to the dipole potential $v_{\rm dip}$.

The eigenvalue problem (5) and the time-dependent single-particle equation (1) are solved in the same basis representation obtained from the basis generator method (BGM) [15]. In the BGM the time-dependent orbital $\psi(t)$ (and the adiabatic state $\psi_0(R)$) is expanded in a basis that is adapted to the specific collision problem at hand. The method has been applied successfully for positive ion impact collisions (see [16] and references therein), and also for antiproton collisions with atomic hydrogen [17] and neon [18]. The basis set employed in the present study consists of a set of bound eigenfunctions of the undisturbed helium atom $\{\varphi_v^0(\boldsymbol{r})\}$ which account for the elastic and target excitation channels, and a set of pseudo-states $\{\chi_v^\mu(\boldsymbol{r})\}$, which are generated by repeated application of the suitably regularized target potential W_T onto the set $\{\varphi_v^0(\boldsymbol{r})\}$, and which describe the ionized part of the electron cloud

$$\chi_v^\mu(\boldsymbol{r}) = [W_T(\boldsymbol{r})]^\mu \varphi_v^0(\boldsymbol{r}). \tag{6}$$

We note that in our earlier work we have used a hierarchy of pseudo-states based on the regularized projectile potential W_P instead of W_T. This previous choice is mandatory for positive ion impact collisions, in which the pseudo states not only account for the ionized part of the electron cloud but also for capture into bound projectile states. In the present case of antiproton impact the capture channel is closed, and one can expect that the continuum contribution is mainly centred around the target since the electrons are repelled by the projectile (see, e.g. the density distributions for $\bar{\rm p}$ + H collisions shown in [19]). This feature is better described by the pseudo-states defined by equation (6). In fact, we have found that both types of basis sets give very similar *total* ionization cross sections for antiproton impact, but the states of equation (6) yield more reasonable coordinate space density distributions than a basis generated by W_P [20].

For the results presented below we have included all undisturbed target eigenstates $\varphi_v^0(\boldsymbol{r})$ of the KLMNO shells, and 132 functions of the set $\{\chi_v^\mu(\boldsymbol{r}), \mu \geqslant 1\}$ up to order $\mu = 8$, which have been orthogonalized to the set $\{\varphi_v^0(\boldsymbol{r})\}$. The relatively large number of eigenstates $\varphi_v^0(\boldsymbol{r})$

[1] We have used the *optimized potential method* calculation of [21], which equals the Hartree–Fock result in the case of the He ^{1}S ground state.

was used to ensure that all target excitation channels that are populated noticeably during the collision are taken into account explicitly. Therefore, the population of the pseudo-states is not contaminated with transitions to highly excited states and can be interpreted as ionization at a time t_f after the collision

$$p_{\rm ion} = 1 - \sum_{v=1}^{5g} |\langle \varphi_v^0 | \psi(t_f) \rangle|^2. \tag{7}$$

We found it sufficient to stop the propagation of the orbital $\psi(t)$ at times t_f that correspond to a separation of 90 au between the antiproton and the helium nucleus.

From the single-particle ionization probabilities $p_{\rm ion}$ we calculate probabilities for single (P_1) and double (P_2) ionization by the usual binomial formulae

$$P_1 = 2p_{\rm ion}(1 - p_{\rm ion}) \tag{8}$$

$$P_2 = p_{\rm ion}^2. \tag{9}$$

One can also define a net ionization probability $P_{\rm net}$ as the weighted sum of P_1 and P_2

$$P_{\rm net} = P_1 + 2P_2 = 2p_{\rm ion}, \tag{10}$$

which equals the average number of removed electrons.

3. Results and discussion

We have diagonalized the Hamiltonian (2) in the BGM basis described above to obtain the single-particle energy levels of the quasi-molecular system ($\bar{\rm p}$He). The lowest level $\varepsilon_0(R)$ is interpreted as the (negative) ionization potential of the quasi-molecule, which is given as the difference between the (correlated) two-electron ground-state energy of ($\bar{\rm p}$He) and the one-electron ground-state energy of the singly ionized system ($\bar{\rm p}$He)$^+$. The binding energies of both quasi-molecules were calculated by Ahlrichs and co-workers with inclusion of configuration interaction in the case of ($\bar{\rm p}$He) as a function of the internuclear distance R [13]. We have used their results to adjust the parameters $p(R)$, $d(R)$, and $a(R)$ of the model potential $v_{\rm mod}$ (4). Figure 1 shows the energy level $\varepsilon_0(R)$ obtained in this way. As a consequence of the repulsion between the electrons and the antiproton $\varepsilon_0(R)$ rises steeply toward the continuum at small R and approaches the binding energy of the negative ion H^- ($\varepsilon_{H^-} = -0.0276$ au) in the united atom limit $R \to 0$. For large separations $\varepsilon_0(R)$ approaches the ground-state single-particle energy level of helium in the Hartree–Fock limit ($\varepsilon_{\rm He} = -0.918$ au). We note that in contrast to the ground-state level all excited states of the helium atom merge with the continuum below some critical internuclear distance [14].

We have also included in figure 1 results obtained with the dipole correction $v_{\rm dip}$ in $v_{\rm mod}$ turned off and with $v_{\rm mod} = 0$. While the calculation with $v_{\rm dip} = 0$ gives very similar results as the calculation with the full model potential, the ground-state energy level $\varepsilon_0(R)$ merges with the continuum for $R \leqslant 0.5$ au when the model potential is set equal to zero. This demonstrates that the stabilization of the two-electron system in the two-centre Coulomb potential cannot be described in an IPM with a frozen atomic target potential. As a consequence, one can expect that the ionization cross section at low projectile energies will be considerably overestimated in such a calculation.

The parameters $p(R)$, $d(R)$, and $a(R)$ that have been used in $v_{\rm mod}$ to obtain $\varepsilon_0(R)$ are displayed in figure 2. The monopole contribution is relatively short-ranged, while the dipole potential is effective over a wider range of internuclear distances. This is also demonstrated in figure 3, in which we show impact-parameter-weighted net ionization probabilities $bP_{\rm net}(b)$ [cf equation (10)] for the energies 20 and 200 keV obtained with different choices for the model

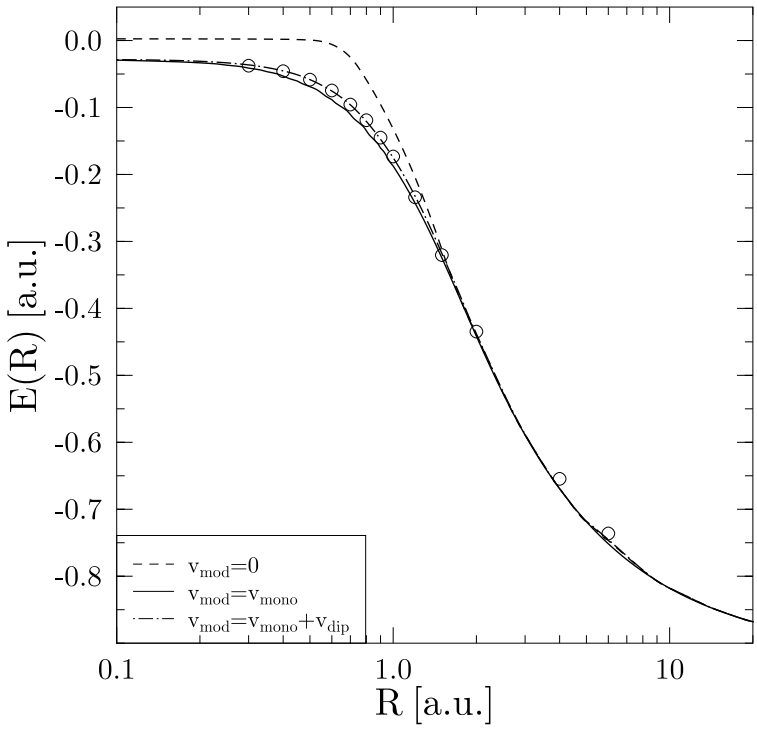

Figure 1. Adiabatic single-particle ground-state energy level of the quasimolecule ($\bar{p}$He) obtained with different choices for the model potential in equation (4). (○): configuration-interaction calculations of [13].

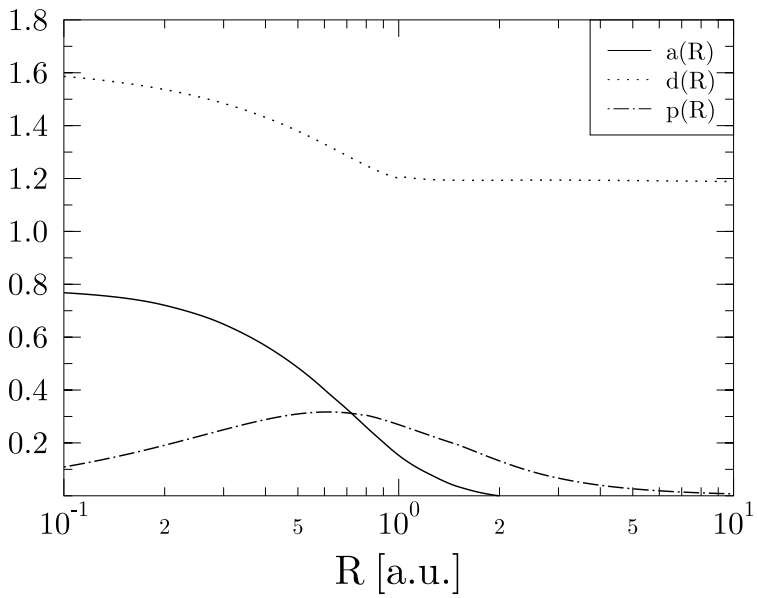

Figure 2. Parameters $p(R)$, $d(R)$, $a(R)$ of the model potential in equation (4).

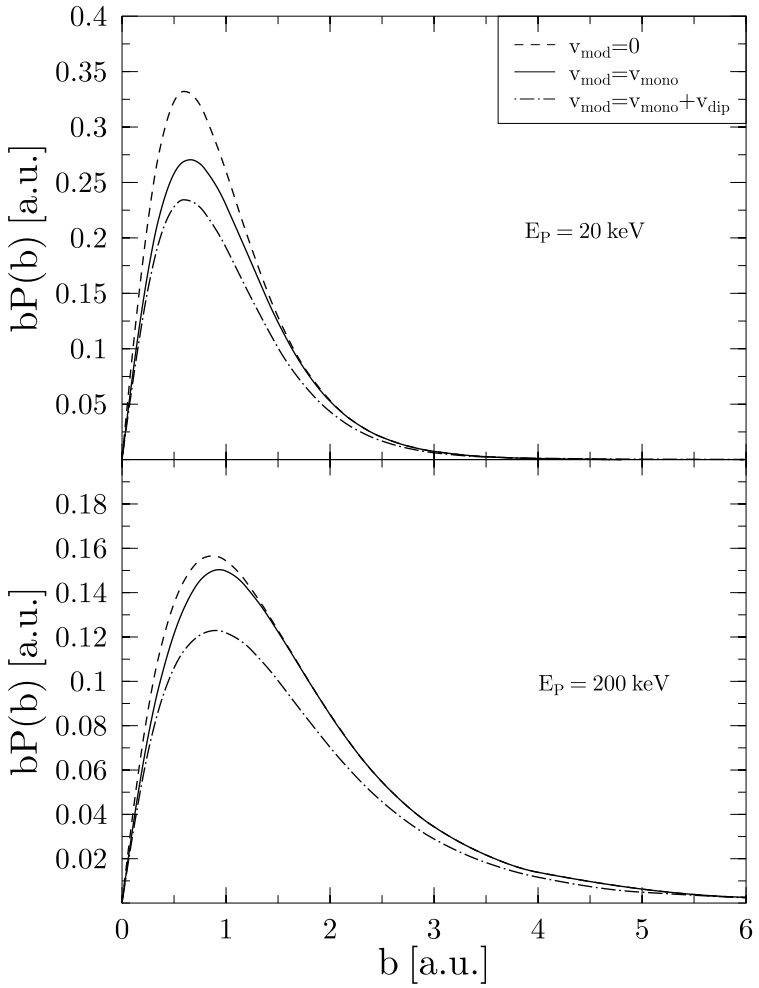

Figure 3. Impact-parameter-weighted net ionization probabilities as a function of the impact parameter b for $\bar{p}$ + He collisions at $E_P = 20$ and 200 keV obtained with different choices for the model potential in equation (4).

potential v_{mod}. Generally we observe that the weighted probabilities extend to larger impact parameters b with increasing projectile energy. The monopole correction alone ($v_{mod} = v_{mono}$) affects the results only at $b \leqslant 1.5$ au, and becomes unimportant at higher impact energies. In contrast, the additional dipole correction ($v_{mod} = v_{mono} + v_{dip}$) leads to a substantial reduction of the weighted ionization probabilities over the entire b region in both cases shown. For projectile velocities above the matching velocity (approximately at $E_P = 50$ keV) one would not expect that polarization effects influence the ionization cross section considerably, as the electron cloud does not have enough time to adjust to the two-centre potential of the nuclei. Hence, the polarization potential v_{dip} becomes unphysical for $E_P \gtrsim 50$ keV and should be switched off.

This observation is supported by a comparison of the theoretical results with experimental ionization cross sections. In figure 4 we show the single ionization as a function of the projectile energy. At low energies the calculations with $v_{mod} = v_{mono}$ and $v_{mod} = v_{mono} + v_{dip}$

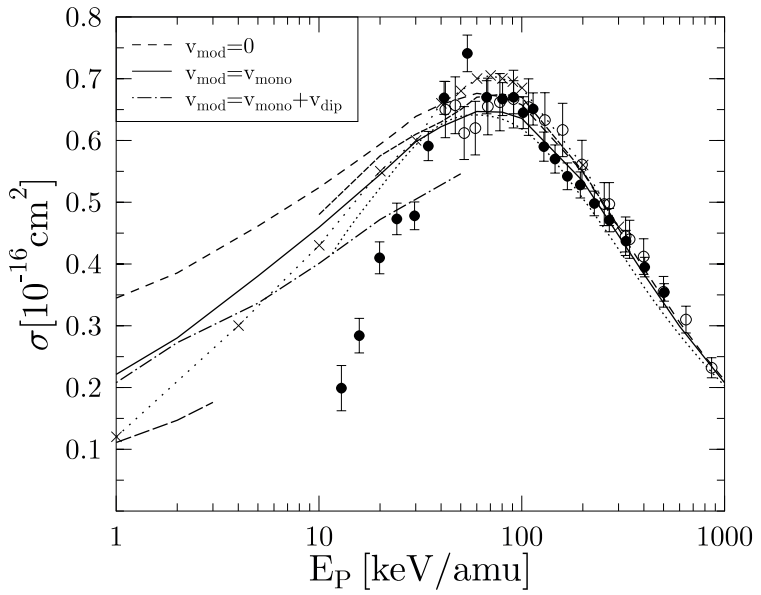

Figure 4. Total cross section for single ionization as a function of impact energy for $\bar{p}$ + He collisions. Theory: present results obtained with different choices for the model potential in equation (4); short-dashed curve [10], long-dashed curve [14], short-dotted curve, multi-cut FIM [8], long-dotted curve with crosses [9]. Experiment: (•) [6], (○) [22].

give comparable results, but above $E_P \approx 40$ keV the latter underestimates significantly the experimental and the other theoretical cross sections, which are in good agreement with each other in the medium- to high-energy region. The calculation with $v_{\mathrm{mod}} = v_{\mathrm{mono}}$ approaches the standard IPM result obtained without a model potential ($v_{\mathrm{mod}} = 0$) at high energies. We have checked that the data of the latter model are in close agreement with conceptually similar IPM calculations reported in [10], which in turn were found to agree well with earlier calculations of the same type [10]. Remarkably, our $v_{\mathrm{mod}} = v_{\mathrm{mono}}$ calculations lead to a significantly reduced single-ionization cross section below $E_P \approx 40$ keV, where the discrepancies between theory and experiment are unexplained to date. The results of our relatively simple model are in good agreement with the computationally much more expensive two-electron calculations of [8–10] in the 10 keV $\leqslant E_P \leqslant$ 40 keV range. This demonstrates that the reduction of the standard IPM cross section is not caused by explicit correlation effects, such as the deviation of the two-electron wavefunction from a simple product state, but is mainly due to a global response of the electron cloud in the presence of the antiproton. Only below 10 keV do the coupled-channel two-electron calculation of [9] and the two-electron hidden crossing calculation of [14] predict a significantly smaller cross section than our model. However, their mutual agreement is poor.

In figure 5 we compare our results obtained with $v_{\mathrm{mod}} = v_{\mathrm{mono}}$ with data of [9] on the level of weighted impact-parameter-dependent probabilities. We observe that our weighted single-ionization probability is shifted toward larger b as compared to the results of [9], and exhibits a slightly smaller peak value. We have also included our result for double ionization, which has not been considered by the authors of [9].

The total cross section for double ionization is displayed in figure 6 in comparison with experiment, the FIM results reported in [8], the results of the so-called converged frozen-correlation approximation of [3] at energies $E_P \geqslant 200$ keV, and the two-electron hidden

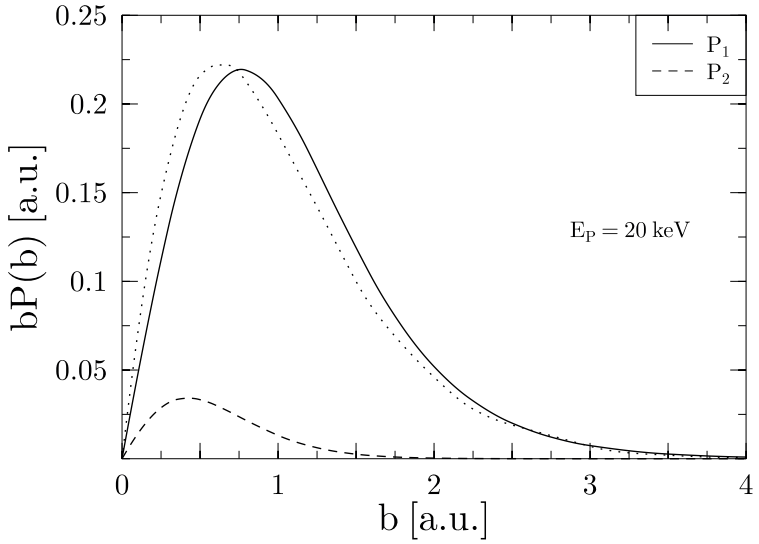

Figure 5. Impact-parameter-weighted single- and double-ionization probabilities as a function of the impact parameter b for $\bar{\mathrm{p}}$ + He collisions at $E_P = 20$ keV obtained with $v_{\mathrm{mod}} = v_{\mathrm{mono}}$. Dotted curve [9].

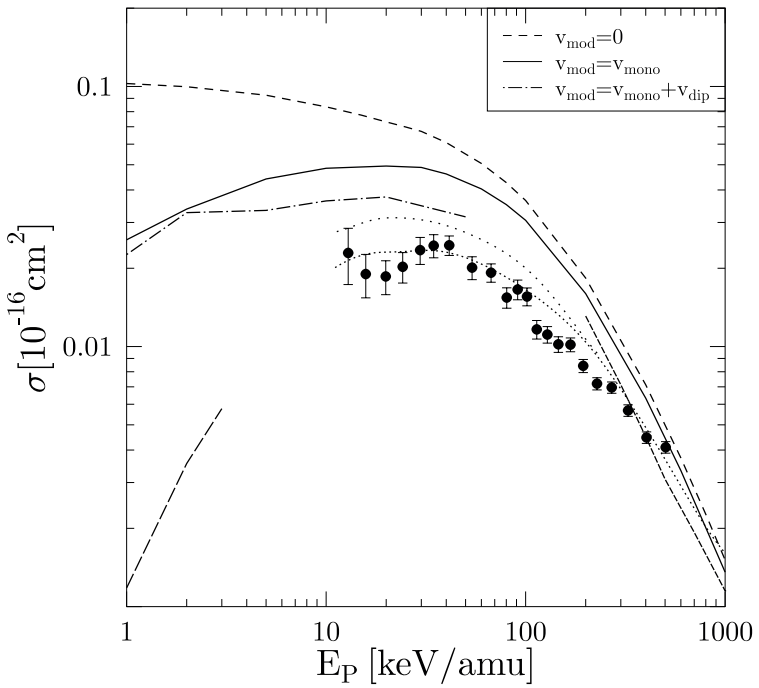

Figure 6. Total cross section for double ionization as a function of impact energy for $\bar{\mathrm{p}}$ + He collisions. Theory: present results obtained with different choices for the model potential in equation (4); short-dashed curve [3], long-dashed curve [14], short-dotted curve, multi-cut FIM [8], long-dotted curve, no-cut FIM [8]. Experiment: (•) [6].

crossing data of [14] at $E_P \leqslant 3$ keV. Similar to the case of single ionization one observes a considerable reduction of the double-ionization cross section, when v_{mono} is included in the IPM calculation. The additional inclusion of v_{dip} has only a small effect in the impact energy range, in which this potential is meaningful. Despite the reduction of the standard IPM cross section our results still lie considerably above the experimental data and the results of the two-electron calculations. We note that the relative reduction due to v_{mono} is similar to the difference between the multi-cut FIM and the no-cut FIM results. In the latter calculation electron–electron correlation is only considered in the initial and the final states, but not during the collision. This indicates that the *dynamical* aspects of the process are taken into account in our model, but the inclusion of initial and final two-electron continuum state correlation is important in order to describe the double-ionization cross section accurately.

4. Conclusions

We have considered the $\bar{\text{p}}$ + He problem in the IPM with a model potential that accounts for polarization effects and for the increased binding of the electrons, which is a consequence of the electronic response in the two-centre Coulomb potential at small internuclear distances. We find that the binding effect represented by v_{mono} reduces the single- and double-ionization cross sections at low and intermediate impact energies significantly compared to a standard IPM calculation without such a model potential. By contrast, the dipole correction has only a small effect in this energy range, but it reduces the cross sections wrongfully at higher impact energies. Hence, the dipole potential v_{dip} should be turned off for energies above the matching velocity, or omitted entirely.

The single-ionization cross section obtained with $v_{\text{mod}} = v_{\text{mono}}$ is comparable with the results of the computationally costly two-electron calculations except at impact energies below 10 keV. This demonstrates that the correlation included in the two-electron calculations can be modelled with good accuracy in a rather simple way for the description of this one-electron process. The discrepancy between theory and experiment in the 10–40 keV region persists. In the case of double ionization we obtain a cross section that is considerably improved compared to a standard IPM calculation. The remaining discrepancies with experiment and the multi-cut FIM calculation of [8] indicate that initial and final state correlations, which lie beyond our model are important for this two-electron process.

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