Electronic Exchange Effects in p + Ne and p + Ar Collisions

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Total cross sections for net electron loss and ionization in p + Ne and p + Ar collisions are presented with emphasis on their crucial dependence on the qualitiy of the static atomic exchange potential. In order to investigate these electronic exchange effects from different viewpoints, we have applied two theoretical models within an effective one particle picture, the *continuum distorted wave with eikonal initial state* approximation and a coupled-channel approach based on an optimized set of pseudostates. [S0031-9007(97)03994-X]

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Inelastic processes in energetic collisions between bare ions and noble gas atoms have been extensively studied experimentally [1] in order to understand the dynamics of many-electron systems. The quantum mechanical description of scattering systems involving more than two active electrons commonly relies on an effective one particle picture within the impact parameter approximation [2–4]. In this framework the electron-electron interaction is taken into account in terms of a single particle potential which can be expressed on the basis of density functional theory as a functional of the density only [5,6]. Disregarding the response of the density in the functional dependence of this potential in the presence of the projectile, the question remains, which level of accuracy is required for the description of static electronic screening and exchange-correlation effects in the target atom in order to obtain reliable results for an ionizing collision process.

In the present contribution we address this question with regard to total cross sections for net electron loss and ionization in p + Ne and p + Ar collisions, where experimental data are available for impact energies ranging from a few keV up to a few MeV [7,8].

The single particle Hamiltonian under discussion reads ($\hbar = m_e = e = 1$)

$$\begin{aligned}\hat{h}(t) &= \hat{h}_0 + \hat{v}(t) \\ &= \left(-\frac{1}{2}\,\Delta + v_{\mathrm{atom}}(r)\right) + \left(\frac{-Z_P}{|\mathbf{r} - \mathbf{R}(t)|}\right),\end{aligned} \tag{1}$$

where v_{atom} denotes the frozen atomic screening potential with spherical symmetry.

Recently, Gulyás *et al.* [9] reported results obtained in the *continuum distorted wave with eikonal initial state* (CDW-EIS) approximation introduced by Crothers and McCann [10], using a numerical Hartree-Fock-Slater (HFS) potential in v_{atom} and corresponding bound and continuum eigenfunctions to construct the initial and final states within the CDW-EIS model. This procedure improved upon previous calculations that suffered from the nonorthogonality between initial and final states. Good agreement with experimental data was obtained in the case of p + Ne collisions at impact energies larger than 100 keV, but the results for argon targets overestimated the total cross sections even at high energies [9].

These deviations originate from the inadequate Slater exchange potential. We here show that one obtains improved results within the CDW-EIS approximation if one uses a more accurate form of the atomic exchange potential. In order to confirm the observed importance of exchange effects, we furthermore analyze different single particle potentials within a different approach to the scattering problem in terms of a recently proposed coupled-channel model [11]. This approach permits the evaluation of total electron loss cross sections (the sum of ionization and capture) over an extended impact energy region.

Accurate frozen atomic potentials can be based on the Kohn-Sham scheme of density functional theory [5]. This formalism ensures the existence of a local one particle potential that yields the exact one particle density $n(r)$. One can split this potential according to

$$v_{\mathrm{atom}}(r) = -\frac{Z_T}{r} + \int \frac{n(r')}{|\mathbf{r} - \mathbf{r}'|}\, d^3r' + v_{xc}([n], r)\,, \tag{2}$$

where the exchange-correlation potential v_{xc} is a functional of the one particle density. Here we employed two systematic approximations to v_{xc}:

(i) The local density approximation (LDA) relying on the density dependence of v_{xc} found for the homogenous electron gas [12].

(ii) The optimized potential method (OPM), in which the exchange-only part of the exchange-correlation functional is treated exactly [13]. In addition, the OPM potential used contains a correlation contribution within the approximation of Colle and Salvetti [14].

The OPM potential is superior to the LDA and the previously used HFS potentials, mainly because of its more accurate description of attractive exchange effects. Besides the asymptotic $-1/r$ behavior, power-law corrections have been shown to influence the eigenvalues of the highest occupied orbitals [13] which play an important role for total ionization. By contrast, HFS and LDA exchange potentials decrease exponentially in the asymptotic region and the correct $-1/r$ behavior has to be introduced by hand. The HFS and LDA potentials differ in strength of the exchange part (3/2 vs 1), where the LDA value corresponds to the electron gas limit.

Correlation effects are included in the OPM and LDA potentials in an effective form, but are of minor importance for the binding of the outer electrons, which are dominantly ionized.

The calculation of net ionization and loss (sometimes termed "gross" cross sections [8]) is straightforward for any density functional potential, because these quantities are directly related to the well-defined one particle density.

First we shall discuss the effects of different potentials on the net ionization in p + Ne and p + Ar collisions. Figure 1 shows results obtained in the CDW-EIS approximation with OPM, LDA, and HFS potentials, respectively, in comparison with the cross sections recommended by Rudd [8]. The results obtained with the LDA potential considerably overestimate the experimental data for both neon and argon targets in the energy region where the CDW-EIS model should be valid. The smaller ionization cross sections obtained with the HFS potential reflect the difference in strength of the exchange potential. As stated above the HFS potential leads to good agreement in the case of neon, but the ionization of argon is overestimated. However, if one employs the OPM potential, the CDW-EIS model fits the experiment well for both scattering systems in its region of validity. It thus can be concluded that the method is able to describe ionization of multielectronic targets if an adequate effective one particle potential is used.

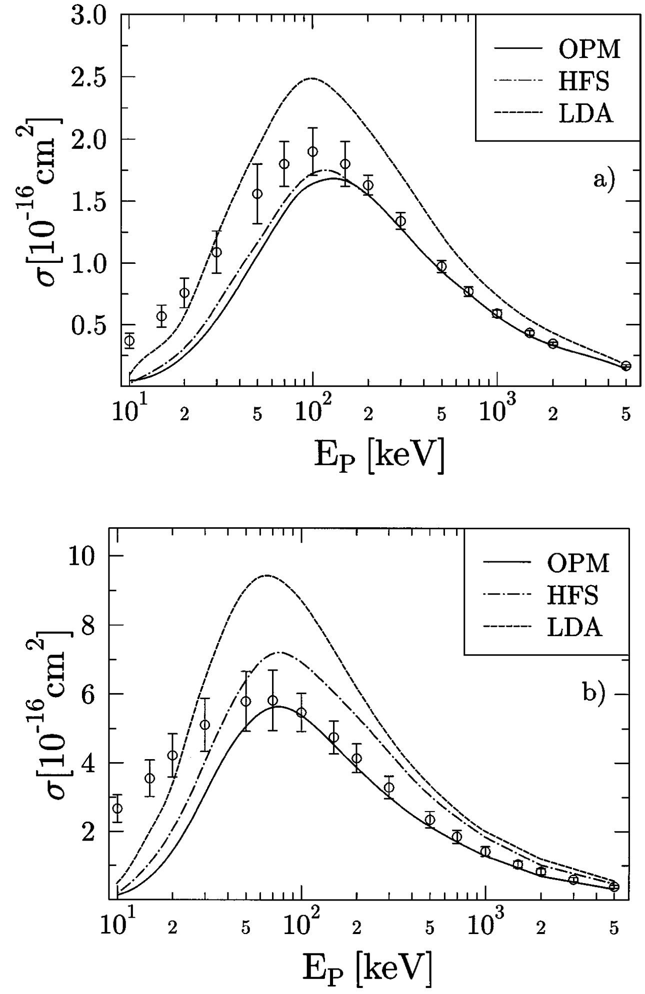

FIG. 1. Net ionization cross section as a function of impact energy for (a) p + Ne and (b) p + Ar. Lines: CDW-EIS calculations with different atomic potentials; symbols: (○) experimental data taken from [8].

In order to extend the investigation to smaller impact energies, we solved the one particle Schrödinger equation with the Hamiltonian (1) for each of the initially occupied orbitals in a coupled-channel representation. According to [11] an optimized set of pseudostates $\Phi_m^M(\mathbf{r},t)$ is generated from the eigenstates $\Phi_l^0(\mathbf{r})$ of the unperturbed atomic Hamiltonian $\hat{h}_0$, with the aim to structurally minimize coupling structures within the Hilbert space obtained in this fashion. Following the arguments given in [11], we used a polynomial ansatz for the pseudostates

$$\Phi_m^M(\mathbf{r},t) = \sum_{\mu=0}^{M} [W(\mathbf{r},t)]^\mu \sum_{l=1}^{N} a_{m,l}^{M,\mu}(t)\Phi_l^0(\mathbf{r}) \qquad (3)$$

with the regularized potential

$$W(\mathbf{r},t) = \frac{-1}{|\mathbf{r}-\mathbf{R}(t)|}\,(1-e^{-|\mathbf{r}-\mathbf{R}(t)|}) \qquad (4)$$

and coefficients $a_{m,l}^{M,\mu}$ that are constructed to guarantee orthogonality between subsets with different superscripts M.

The set of eigenstates $\Phi_l^0(\mathbf{r})$ was chosen to include the important target excitation channels ($1s$-$3d$ for neon, $1s$-$4d$ for argon). For this reason occupation of the pseudostates can be interpreted as single electron loss p_j, the sum of the single particle capture and ionization probabilities from the initially occupied orbital $\Phi_j^0(\mathbf{r})$. Total electron loss is then obtained by summation of the single particle probabilities p_j.

It has been shown [11] that the probability flux between the finite space spanned by the eigenfunctions $\Phi_l^0(\mathbf{r})$ of the undisturbed system and the Hilbert space of the interacting system has to go through a subspace spanned by the pseudostates corresponding to $M = 1$. Therefore, the method should be appropriate for the discussion of electron loss. Moreover, the representation in the pseudobasis is not restricted with respect to the impact energy and has been shown to give reasonable loss cross sections over a wider range of energies in the case of one electron scattering systems [15]. In the present study we included subsets of pseudostates to the order $M = 8$ for both target atoms.

Results obtained with this method for different atomic potentials are presented in Fig. 2. Since capture processes are known to contribute only for impact energies up to 150–200 keV [7], the net electron loss can be identified with ionization at the higher energies. In fact, in this region the results for each atomic potential are in good agreement with those obtained in the CDW-EIS approximation (cf. Fig. 1). This agreement between the two approaches indicates that the differences resulting from different atomic potentials do not depend on the method.

At lower impact energies the situation is more involved. For neon targets the calculations with the OPM potential lead to excellent results down to 5 keV (where capture is the dominant process) [Fig. 2(a)], if one disregards a slight overestimation of the experimental data at around 100 keV. For argon targets [Fig. 2(b)] these deviations are more pronounced and extend over a wider energy range. Only at the lowest energies the theoretical results do again lie within the experimental error bars.

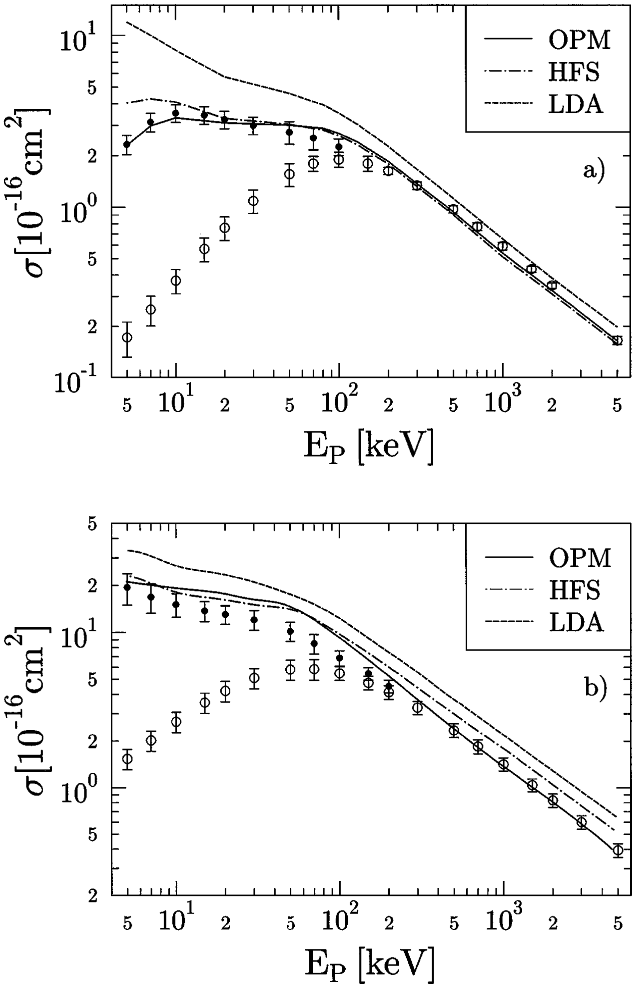

FIG. 2. Net electron loss cross section as a function of impact energy for (a) p + Ne and (b) p + Ar. Lines: coupled-channel calculations with different atomic potentials; symbols: (●) experimental data for net electron loss [7]; (○) experimental data for net ionization [8].

This behavior illustrates the range for which the basic assumption of a frozen atomic potential is valid. The assumption is reasonable at high impact energies where the spatial electronic distribution does not change considerably during the interaction time. Furthermore, it should not lead to significant errors at lower energies if the physics is strongly dominated by *single* electron loss. According to the experimental data [16,17], it can be stated that multiple loss processes hardly contribute at the lowest and highest energies, but are more likely to occur in the intermediate energy region. Generally they are more important in p + Ar than in p + Ne collisions as the valence electrons of argon are more weakly bound.

We therefore conclude that the observed overestimation in the theoretical results based on the OPM potential is due to time-dependent screening effects which are neglected in our model. Qualitatively one can expect a reduction of the net electron loss cross sections as a time-dependent electron density will both screen the projectile potential and lead to a more attractive target potential as ionization sets in. An accurate incorporation of these effects could in principle be attempted if one replaces v_{atom} in the one particle Hamiltonian (1) by a time-dependent optimized potential [6].

Moreover, Fig. 2 mirrors the deficiencies of the LDA and HFS atomic potentials. The LDA description leads for both neon and argon targets to an overall overestimation of the experimental loss cross sections. The HFS potential, however, does not indicate a systematic pattern. In the case of argon the cross sections are in agreement with the OPM results at low energies but deviate in the high energy region as indicated in Fig. 2(b). By contrast, the results for neon show discrepancies at the lowest impact energies. Since time-dependent screening effects are of minor importance here, we attribute this behavior to the inaccurate atomic exchange potential. The partial agreement of the HFS results appears to be accidental.

In summary, we have shown that net electron loss from atomic targets can properly be calculated in an effective one particle picture with a frozen screening potential that accurately accounts for the static exchange. The OPM approach yields the best local representation of the exchange potential, whereas both the HFS and the LDA rely on a local approximation based on the electron gas model. Our calculations indicate that only the OPM potential leads to results which are in quantitative agreement with experimental data in the regions where time-dependent screening effects can be neglected. At these energies the coupled-channel approach based on an optimized set of pseudostates seems to be well suited for the calculation of net electron loss, while the CDW-EIS approximation provides a proper description of ionization at high energies. Both methods give comparable results in the high energy regime if the same atomic potential is used.

The exchange part, which has been shown to be important, reflects a genuine quantum mechanical property of the many-electron system. One therefore has to be careful in interpreting results for collisions involving a quantum many-particle system which are calculated solely with classical methods. It is expected that the quantum character of the atomic potential does not only affect the net cross sections but determines also the details of the collision process. Studies concerning the angle and energy distributions of ionized electrons as well as total cross sections for *multiple* electron capture and ionization will be reported in subsequent publications.

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