Projectile scattering and electron-electron interaction in ion-atom collisions

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Abstract. One- and two-electron processes involving electron transfer and excitation are considered for a few basic ion-atom collision systems at intermediate impact energies. We have used the two-center basis generator method in the framework of the independent electron model for orbital propagation and the eikonal approximation for the calculation of scattering-angle differential cross sections. Our results shed some light on the roles of the nucleus-nucleus and the electron-electron interactions in these collisions.

1. Introduction
Recent studies on ion-atom collisions have demonstrated that the role of the nucleus-nucleus interaction is less well understood than one might have thought. The issue appeared on the agenda in 2003, when unexpected discrepancies between experiment and theory were found in kinematically complete data for ionization processes [1]. Quite a few papers offering illuminating analysis and explanations have appeared since then, but none of them was able to clarify the situation completely and solve the problem once and for all. As it seems to date, the nucleus-nucleus interaction poses one of the largest remaining problems in atomic collision theory.

Supportive of this conclusion are investigations of processes involving electron transfer, which is an (almost) unique feature of ion-atom collisions. Even though such processes have been studied for some time for basic collision systems such as p-He and He²⁺-He the state of affairs has become clearer only recently when well-resolved and detailed experimental and theoretical results became available.

In this contribution, we present calculations for one- and two-electron processes involving electron transfer and target excitation [2, 3]. In particular, we address the question whether it is possible to disentangle effects of the heavy-particle motion from those of electronic correlations, which constitute the other unsolved big issue of atomic collision physics.

Our calculations are based on the independent electron model, the two-center basis generator method for orbital propagation, and the eikonal approximation to extract angular-differential cross sections (section 2). Certainly, this framework does not suffice to describe the physics in full, but as shown in section 3 it sheds some light on it and helps answer some of the pending questions.
2. Theory

2.1. Independent electron model (IEM)

The problem is treated in the semiclassical, nonrelativistic approximation. The nuclei are assumed to be moving on classical trajectories with the relative vector $\mathbf{R}(t)$. For the calculation of the Hamiltonian we assume this trajectory to be straight with impact parameter $b$, and therefore $R(t) = \sqrt{b^2 + v^2 t^2}$, where $v$ is the relative velocity. The total Hamiltonian is $\hat{H} = \hat{H}_e + \hat{V}_{nn}$ with the nuclear repulsion $V_{nn}(\mathbf{R}, t) = \frac{Z_p Z_n}{|\mathbf{R}(t) - \mathbf{R}|}$ (atomic units characterized by $\hbar = e^2 = m_e = 1$ are used unless stated otherwise). Within the IEM, $\hat{H}_e$ is approximated as a sum of single-particle Hamiltonians $\hat{h}_i$:

$$\hat{H}_e = \sum_{i=1}^{N} \hat{h}_i. \quad (1)$$

The $\hat{h}_i$ take the electron-electron interaction into account via an effective potential $\hat{V}_{eff}$. We use the helium ground-state potential obtained from the optimized potential method [4], i.e. response effects are neglected. Finally, the single electron Hamiltonian reads in spatial representation

$$h(\mathbf{r}_t, t) = -\frac{1}{2} \Delta + V_{eff}(\mathbf{r}_t) - \frac{Z_p}{|\mathbf{R}(t) - \mathbf{R}_t|}, \quad (2)$$

where $\mathbf{r}_t$ is the relative vector between target nucleus and electron.

In the IEM the wave function can be approximated as a Slater determinant consisting of one-particle states $|\psi_i\rangle$. The resulting time-dependent Schrödinger equations (TDSEs)

$$i\partial_t |\psi_i(t)\rangle = \hat{h}|\psi_i(t)\rangle \quad (3)$$

are solved by the two-center basis generator method (TC-BGM), a problem-adapted, finite basis representation [5, 6]. The TC-BGM basis is constructed from

- target states (all $KLMN$ shell eigenstates of the unperturbed target atom) and from
- projectile states (analogously)

taking Galilean invariance into account by suitable phase factors. The resulting states $|\varphi^0_u\rangle$ are well-suited to describe electrons bound to the target or the projectile. Additionally, we need states that can describe ionization. These so-called pseudo scattering states $|\chi^0_u\rangle$ are constructed by $\mu$-fold application of the (regularized) projectile-electron potential onto the target states.

2.2. Calculation of observables

The single electron transition amplitude $a_{if}$ for the transition of the $i$th electron into the final state $|\varphi^0_f\rangle$ is

$$a_{if} = \lim_{t \to \infty} \langle \varphi^0_f | \psi_i(t) \rangle. \quad (4)$$

Applying the IEM and taking into account that for He targets both electrons start from the same (1s-)orbital (which implies $a_{1f} = a_{2f}$), the 2-electron amplitudes $a^{f'f}$ for transfer of one electron each into $|\varphi^0_f\rangle$ and $|\varphi^0_{f'}\rangle$ are

$$a^{f'f} = \frac{1}{\sqrt{2}} (a_{1f}a_{2f'} + a_{1f'}a_{2f}) = \sqrt{2} a_{1f} a_{1f'} \text{ for } f \neq f',$$

$$a^{f'f} = a^2_{1f}, \quad (5)$$

where the factor $\sqrt{2}$ takes the indistinguishability of the electrons into account.

While the electronic Hamiltonian is calculated under the assumption of a straight-line trajectory
for the projectile, projectile scattering can still be described by the eikonal approximation [7],[8],[9]. The eikonal scattering amplitude \( g^{ff'}(\theta) \) is

\[
g^{ff'}(\theta) = ik \int_0^{\infty} db bJ_{\Delta M} \left( 2kb \sin \left( \frac{\theta}{2} \right) \right) A^{ff'}(b),
\]

where \( k \) is the initial projectile momentum, \( \Delta M \) is the difference of initial and final state magnetic quantum numbers, and \( J_{\Delta M} \) is a Bessel function.

\[
A^{ff'}(b) = a^{ff'}(b) \exp \left( -\frac{2}{\sigma} \int_0^{\infty} dz V_{nn} (\sqrt{b^2 + z^2}) \right)
\]

is the electronic transition amplitude \( a^{ff'}(b) \) multiplied by a phase factor that takes the nucleus-nucleus interaction into account.

Finally, the differential cross section (DCS) in the center-of-mass-system is

\[
\left( \frac{d\sigma^{ff'}}{d\Omega} \right)(\theta) = |g^{ff'}(\theta)|^2.
\]

A technical difficulty consists in the calculation of the integral (6) since the phase factor in (7) becomes singular at \( b \to 0 \), which makes straightforward numerical integration difficult and time-consuming. Two other methods have been tried to overcome this, namely truncated series expansion of the integral and Hankel-transform-Laguerre-expansion [10]. As it turned out, all these methods including direct numerical quadrature yield the same results.

3. Results and Discussion

3.1. p-H system

We have selected the p-H collision system as a test system in our work. It is obvious that there is no electron correlation in this three-body system. Nevertheless, it can help to assess the validity of the straight-line trajectory approximation for the heavy-particle motion. Limitations should be most visible at low projectile energies. However, we obtain good agreement at \( E_P = 1 \) keV with the experiment [11] and an electron nuclear dynamics (END) calculation [12] except for very small scattering angles (figure 1). In the END calculation a classical (Coulomb) trajectory for the nuclear motion was used. Moreover, we were not able to identify interference effects associated with the indistinguishability of the nuclei, because the backward scattering probability is just
Figure 2. Differential capture cross section as a function of scattering angle in p-H collisions at $E_P = 25$ keV. Theory: present TC-BGM calculation for transfer into $n=1$ and $\sum n$, present TC-BGM calculation with classical scattering (TCBGM CL), present TC-BGM calculation without phase factor for the nucleus-nucleus interaction (TCBGM-NN), classical trajectory Monte-Carlo (CTMC) [20], reformulated impulse approximation (RIA), and continuum distorted wave (CDW) calculations [13], close coupling calculation with Sturmian functions (CC-SF) [16, 17, 18]. Experiment: capture into all states [21].

too small. Our calculation with explicit inclusion of these effects did not show any noticeable changes in the DCS.

There are many calculations of the DCS for charge transfer in p-H collisions in the 25–125 keV energy region [13, 14, 15, 16, 17, 18, 19, 20], and they are more or less all successful in describing the experimental DCS reported in [21]. Our own calculation is no exception (figure 2). To illustrate the importance of the nucleus-nucleus interaction we have also included a DCS curve in figure 2 that is obtained by neglecting the corresponding phase factor, i.e. by assuming $A_f' f(b) = a f' f(b)$ in equation (7). It clearly has the wrong shape.

There is another fact we would like to point out here, which was not touched upon in earlier works. Interestingly, if we combine our TC-BGM calculation with the DCS for classical elastic (Coulomb) scattering we reproduce the experimental data in the angular region $\theta > 1/k$, in which this approximation is valid [22]. In contrast, all other, supposedly more refined theories yield smaller cross sections at these relatively large scattering angles. This might be just fortuitous, but new experiments are needed to clarify this case.

3.2. p-He system

For He target atoms the question about the role of the electron-electron interaction comes into play. It has been debated for many years, and it seems that one generally accepted conclusion is that two-electron processes, such as double capture are largely determined by electron correlation effects. The results presented in this and the following subsections indicate that this is not necessarily true.

Recently, the DCS for transfer excitation (TE), in which one electron is transferred to the projectile, while the other one is promoted to an excited target state was measured at intermediate impact energies [23]. When combined with DCSs for single transfer (ST), double excitation (DE), and single excitation (SE) it turned out that the double ratio

$$R = \frac{TE}{ST} \frac{DE}{SE}$$

exhibited a peak structure around the scattering angle $\theta_{LAB} = 0.5$ mrad. A simple analysis shows that if one assumes all DCSs involved in $R$ to be products of one classical DCS for elastic scattering and electronic transition probabilities calculated within the IEM one obtains a constant double ratio $R=2$. When using different potentials for the different processes the
calculated double ratio is no longer constant, but it does not show a peak structure either [23]. At this point it is unclear whether the measured structure is associated with electron-electron interactions beyond the IEM or with the nucleus-nucleus interaction beyond classical scattering or with both.

In order to clarify this issue we have replaced the classical scattering ansatz by the eikonal approximation, but have adhered to the IEM. The results for $R$ are shown in figure 3. Clearly, peak structures in qualitative agreement with experiment are present. We have found that the height of the peak is quite sensitive to the details of how the two-electron transition amplitudes are composed from one-electron amplitudes. The full (black) curve in figure 3 is obtained from the standard IEM according to equation (5). If one uses a slightly modified analysis in which the one-electron processes ST and SE are evaluated in terms of the single-active electron model [2] one obtains the dashed (blue) curve. This might indicate that the peak height is sensitive to electron-electron interaction effects, which are treated differently in both analyses. The fact that it is there, however, is due to nucleus-nucleus interaction effects, or more precisely to quantum mechanical heavy-particle-electron couplings, which are taken into account in the eikonal approximation.

3.3. He$^{2+}$-He system

Finally, we briefly consider the He$^{2+}$-He collision system, in which two-electron transfer is an important reaction channel. Figure 4 shows the DCS for this process at the impact energy $E_P=375$ keV/amu. It was measured some time ago and compared with continuum distorted-wave (CDW) calculations, in which correlation effects in the initial and final states were included in terms of a configuration interaction (CI) ansatz [24, 25]. Furthermore, the calculations were folded with the experimental resolution function. In addition to those previous results the figure includes our (unfolded) DCSs for double capture into the ground state ($n=1$) and into all shells ($\sum n$). The latter calculation is in very good agreement with the measurements at scattering angles $\theta_{LAB} \geq 0.2$ mrad, but lies above the data at smaller angles. In this region, the CDW results show that the convolution with the experimental resolution function reduces the DCS. Assuming that the convolution would have the same effect on our results we would most likely obtain excellent agreement with the experimental data in the entire angular range. In other words, there would be no indication for electron correlation effects for this two-electron cross section. Unfortunately, the experimental resolution function was not available to us so that we were not able to prove this conjecture.

Figure 3. Ratio of transfer-excitation to single transfer versus double to single excitation cross sections as a function of laboratory scattering angle for 50 keV p-He collisions. Theory: TC-BGM calculation for all amplitudes within IEM, calculation within single-active-electron model (SAE) for single transfer and single excitation amplitudes. The constant ratio $R = 2$ corresponds to the simple model described in the text. Experiment: [23].
Figure 4. Differential two-electron transfer cross section as a function of laboratory scattering angle for $E_P = 375$ keV/amu $^4$He$^{2+}$ – $^4$He collisions. Theory: present TC-BGM calculation within IEM for two-electron transfer into the ground state (n=1) and into all ($\sum n$) shells of the projectile; CDW CI calculation [24, 25]; CDW CI folded, same CDW calculation folded with the experimental resolution function [24, 25]. Experiment: two-electron transfer into all shells [25].

4. Conclusions
Fully differential cross sections for one- and two-electron processes in ion-atom collisions exhibit interesting structures, which are not always well understood. Often they have been blamed on electron-electron interaction effects, but not in all cases with good reason. As it turns out, calculations on the level of the independent electron model are able to describe several experimental data sets at intermediate impact energies, if they are based on a reliable propagation method and the eikonal approximation for the extraction of differential cross sections. Our studies provide ample evidence for the importance of the nucleus-nucleus interaction, but not so much for electron correlation effects.

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