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Classical impact parameter approximation: Application to atomic collisions with n independent electrons and to recoil analysis

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Abstract. We show that a classical Impact Parameter Method may be derived when taking fully into account the smallness of the ratio between the electron and nuclear masses. It allows to calculate, exactly as in the quantum version, projectile scattering and therefore recoil momenta required for the interpretation of recent measurements. We prove an *additivity theorem* which allows, in particular, to reduce the n-non-interacting electron problem to a set of n one-electron problems. Consequences for the interpretation of target recoil measurements are discussed.

PACS. 34.10.+x General theories and models of atomic and molecular collisions and interactions (including statistical theories, transition state, stochastic and trajectory models, etc.) – 34.50.Fa Electronic excitation and ionization of atoms (including beam-foil excitation and ionization)

1 Introduction

The impact parameter method plays a central role in the theory of ion(atom)-atom collision processes: while the relative motion of the nuclei is represented by a straight-line trajectory, the electron motion is treated quantum mechanically. This method brings into the theory the enormous simplification that arises from the smallness of the ratio ϵ of the electron mass m_e to the reduced mass of the nuclei μ [1]. It allows to determine cross-sections differential in projectile scattering angle [2], for small scattering angles, including both electron-nuclei and internuclear interactions. We refer hereafter to this theory as the *quantum* impact parameter method. A similar classical theory can be set up in which the electron motion is treated classically, and we refer to such an approach as the *classical* impact parameter method. It has been used to calculate cross-sections for a variety of one-electron processes (see *e.g.* [3]). In the case of scattering by a central potential the relation between classical and quantum impact parameter methods has been established [2]. It shows that the momentum transfer in the classical theory is linear in the potential.

In the present contribution we show that projectile scattering may also be calculated to first order in $\epsilon = m_\mathrm{e}/\mu$ and that the corresponding limit simplifies considerably the solution of the problem for the case of n non-interacting electrons [4]. This is of particular interest for the application of CTMC calculations to the interpretation of present day measurements of recoil distributions [5,6].

Atomic units are used unless otherwise stated.

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2 Theory

We write down the equations of motion in the center-of-mass (CM) frame. Consider first the one-electron case. Let $\mathbf{R}$ be the projectile position vector with respect to the target nucleus and $\mathbf{r}$ that of the electron with respect to the center of mass of the nuclei. We designate by $\mathbf{P}$ and $\mathbf{p}$ the momenta conjugate to $\mathbf{R}$ and $\mathbf{r}$ respectively. Hamilton function reads

$$H = \frac{P^2}{2\mu} + \frac{p^2}{2m_\mathrm{e}} + V(\mathbf{r},\mathbf{R}). \tag{1}$$

It is convenient to split the potential into two parts

$$V(\mathbf{r},\mathbf{R}) = V_\mathrm{el}(\mathbf{r},\mathbf{R}) + V_\mathrm{nuc}(R), \tag{2}$$

where V_el is the interaction potential between the electron and nuclei whereas V_nuc is the internuclear potential. Hamilton equations are

$$\frac{\mathrm{d}R_i}{\mathrm{d}t} = \frac{P_i}{\mu} \tag{3a}$$

$$\frac{\mathrm{d}P_i}{\mathrm{d}t} = -\frac{\mathrm{d}V(\mathbf{r},\mathbf{R})}{\mathrm{d}R_i} \tag{3b}$$

$$\frac{\mathrm{d}r_i}{\mathrm{d}t} = \frac{p_i}{m_\mathrm{e}} \tag{3c}$$

$$\frac{\mathrm{d}p_i}{\mathrm{d}t} = -\frac{\mathrm{d}V_\mathrm{el}(\mathbf{r},\mathbf{R})}{\mathrm{d}r_i}, \tag{3d}$$

where the index i labels the Cartesian components of the vectors. To solve equation (3) to first order in $\epsilon = m_\mathrm{e}/\mu$, we may determine $\mathbf{R}(t)$ to first order in ϵ. From now on,

we confine ourselves to small angle scattering so that the variation of $\mathbf{P}$ given by (3b) is of order $0(\epsilon)$ with respect to the initial value $\mu\mathbf{v}$. Consequently $\mathbf{R} = \mathbf{R}_0(t) = \mathbf{v}t + \boldsymbol{\rho}$ where $\boldsymbol{\rho}$ is the impact parameter and the origin of time is fixed at the distance of minimum approach. This is simply the usual impact parameter equation, as could be expected. For clarity we designate by $\mathbf{R}_0(t)$ the value of $\mathbf{R}$ when calculated in this way. As a consequence, equations (3c, 3d) for the electron motion are completely decoupled from the equations for the relative motion of the heavy particles once the expression for $\mathbf{R}_0(t)$ is substituted into the potential in (3d), as in the quantum impact parameter approximation. We note also that, exactly as in the quantum impact parameter approximation, the internuclear potential plays no role in the electronic evolution. Until now, no new result has been obtained: this form of solution has been used by many authors to evaluate the CTMC probability of various electronic processes (see *e.g.* [3]). The error associated with the fact that the obtained solution is only correct to first order in ϵ is very small as confirmed in actual calculations.

Once Hamilton equations for the electron motion have been solved, it is possible to determine the variation of $\mathbf{P}$ to first order in ϵ. This is done by solving equation (3b) with $\mathbf{r} = \mathbf{r}_0(t)$ (the solution just derived for the electronic evolution) and $\mathbf{R} = \mathbf{R}_0(t)$. One gets

$$\frac{\mathrm{d}P_i}{\mathrm{d}t} = -\left[\frac{\mathrm{d}V_{\mathrm{el}}(\mathbf{r}_0(t), \mathbf{R})}{\mathrm{d}R_i} + \frac{\mathrm{d}V_{\mathrm{nuc}}(R)}{\mathrm{d}R_i}\right]_{\mathbf{R}=\mathbf{R}_0(t)}. \quad (4)$$

The latter expression shows the additivity of the internuclear and electronic interactions for the determination of the momentum change in the nuclear relative motion. If we call $\Delta\mathbf{P}$ the momentum change over the collision in the relative nuclear motion,

$$\Delta\mathbf{P} = \Delta\mathbf{P}_{\mathrm{el}} + \Delta\mathbf{P}_{\mathrm{nuc}}, \quad (5)$$

where $\Delta\mathbf{P}_{\mathrm{el}}$ is the momentum change due to the interaction with the electron and $\Delta\mathbf{P}_{\mathrm{nuc}}$ that due to the internuclear interaction. We call the latter result *the additivity theorem*: when a calculation is performed to first order in ϵ the momentum change in the nuclear relative motion is the sum of one term due only to the electron and another one due to the internuclear interaction. We note that the linearity in the potential had already been proved in reference [2] for the case of scattering by a central potential when looking at the classical limit of the quantum impact parameter method (see Eq. (38) of [2]).

We are now in a position to understand how the n independent electron problem may be greatly simplified: it is a direct consequence of the additivity theorem for a solution to first order in ϵ. It is not necessary to write down the corresponding equations in details since the generalization is trivial. In fact, in the absence of electron-electron interaction, Hamilton equations for each electron are exactly the same as above. Once the time evolution is determined for each electron, the momentum transfer to the projectile may be calculated by application of the additivity theorem, *i.e.* summing the contribution to the internuclear momentum transfer coming from each electron. This means that one has to solve a sequence of n independent one electron problems and the momentum change for the internuclear motion is that obtained by summing the contribution from all electrons to that of the internuclear interaction. Once the momentum change of the electrons and that associated with the relative motion of the nuclei has been determined in the CM, it is a trivial task to determine the projectile and target final momenta by using conservation of momentum and the transformation from the center of mass to the laboratory frame. Further generalizations may be easily derived (*e.g.* molecular targets and/or projectiles).

In practice, the procedure consists in solving Hamilton equations (3b, 3c, 3d) – one might include as well (3a), the difference being only of order ϵ – for each electron *with the internuclear potential omitted*. The momentum transfer due to the internuclear potential is calculated separately. The set of initial conditions for the n-electron problem corresponds, of course, to the tensorial product of n sets of initial conditions for the one-electron problems. So, the additivity theorem is used to combine the momentum transfers in the n sets of solutions to produce the one associated with the n-electron problem. We see that the classical impact parameter approximation brings a tremendous simplification in the evaluation of the dynamics for the case of n non-interacting electrons. Consider that we have to carry out p calculations per electron to keep the statistical error below some prescribed value. Then, the usual n-CTMC calculations require p^n trajectories and therefore powerful vector computers [5]. With our method only np trajectories are required and the calculations can be performed on any PC, while maintaining the error at the level of ϵ, which is usually negligible. It is gratifying that physical insight may still be in a position to compete with the enormous effectiveness of present day computer industry.

The validity of the classical impact parameter method may be verified through the knowledge of the final transverse momentum. For small angles:

$$\Delta P^{\perp} = \mu v \theta_{\mathrm{CM}}, \quad (6)$$

where θ_{CM} is the center of mass scattering angle. The validity condition for the first order evaluation is $\theta_{\mathrm{CM}} \ll 1$. This is verified in a very large class of ion-atom processes. The approximation breaks down when the scattering angle is appreciable, *i.e.* for small impact parameters, low energies or when a very large number of electrons is involved. One may note that, at high energies, large scattering angles are associated with small impact parameters and the scattering is then usually dominated by the internuclear interaction. Therefore, an obvious generalization could be explored in which $\mathbf{R}(t)$ would be determined by the internuclear potential alone and the corresponding trajectory used to solve for the electron motion. The analogous method is well documented as a generalization of the quantum impact parameter equation.

3 Discussion

Beside its usefulness in performing the calculations for n independent electrons, the additivity theorem brings information which is useful in interpreting the comparison between CTMC results and experiment. We outline briefly some examples. We now work in the Laboratory-frame (Lab) and label any quantity in this frame relative to the projectile (resp. target) by a p (resp. t) subscript. We call $\Delta\mathbf{p}_{\rm L}$ the total momentum change of the electrons in the Lab frame. The momentum transfer in the Lab frame is expressed in terms of the momentum transfer $\Delta\mathbf{P}$, solution of (3), through a linear transformation:

$$\Delta\mathbf{P}_{\rm p} = \Delta\mathbf{P} - m_{\rm p}/(m_{\rm p}+m_{\rm t})\Delta\mathbf{p}_{\rm L} \tag{7a}$$

$$\Delta\mathbf{P}_{\rm t} = -\Delta\mathbf{P} - m_{\rm t}/(m_{\rm p}+m_{\rm t})\Delta\mathbf{p}_{\rm L} \tag{7b}$$

where $m_{\rm p}$ and $m_{\rm t}$ are the projectile and target mass respectively. Therefore, the additivity theorem also works in the Lab frame.

The projectile and target momentum change may be decomposed into an internuclear [$\Delta\mathbf{P}_{\rm p,t}$(nuc)] and "electronic" contribution (or rather a *two-center* interaction: the electron interacts at the same time with the projectile and target). As the internuclear potential is a central potential, it causes a longitudinal momentum transfer of order ϵ with respect to the transverse momentum transfer $\Delta\mathbf{P}_{\rm p}$(nuc) $\simeq$ $\Delta\mathbf{P}_{\rm p}^{\perp}$(nuc) $=$ $-\Delta\mathbf{P}_{\rm t}^{\perp}$(nuc). Therefore, the additivity theorem proves that the internuclear potential gives a negligible contribution to the target longitudinal momentum change: any transfer of longitudinal momentum from the projectile to the recoiling nucleus must be transmitted *via* the electrons.

Consider the case of longitudinal momentum analysis. If we call $\Delta E_{\rm el}$ the change in energy of the electron over the collision, conservation of energy gives:

$$\Delta E_{\rm el} = M_{\rm p}v^2/2 - M_{\rm p}(v+\Delta P_{\rm p}^{\parallel}/M_{\rm p})^2/2 - (\Delta P_{\rm p}^{\perp})^2/2M_{\rm p} - (\Delta P_{\rm t})^2/2M_{\rm t} \tag{8}$$

and to first order in ϵ,

$$\Delta E_{\rm el}/v = -\Delta P_{\rm p}^{\parallel} + 0(\epsilon). \tag{9}$$

Conservation of momentum in the Lab-frame yields

$$\Delta P_{\rm t}^{\parallel} + \Delta p_{\rm L}^{\parallel} - \Delta E_{\rm el}/v = 0. \tag{10}$$

As the internuclear interaction does not contribute, the longitudinal recoil momentum is a direct measure of the electronic longitudinal momentum change. This property is merely a kinematical property and a direct consequence of the smallness of ϵ [6]. It is independent of the form and strength of the projectile-electron interaction. Therefore, its analysis in terms of a similarity between the projectile and a photon (see Sect. IV.A of Ref. [6]) is misleading. We note that the above conclusion is valid for processes involving any number of electrons (interacting or not).

The transverse momentum analysis is made extremely simple when noting that, for small $\theta_{\rm CM}$, the projectile scattering angle in the Lab-frame is such that

$$M_{\rm p}\theta_{\rm p} = \mu\theta_{\rm CM} + 0(\theta,\epsilon). \tag{11}$$

The target transverse momentum is obtained readily by application of momentum conservation in the Lab-frame

$$\Delta P_{\rm t}^{\perp} + \Delta p_{\rm L}^{\perp} - M_{\rm p}v\theta_{\rm p} = 0(\theta,\epsilon). \tag{12}$$

Let us now analyse the situation, observed *e.g.* in reference [6], in which the momentum transfer to the projectile $\Delta P_{\rm p}$ is much smaller than $\Delta P_{\rm t}^{\perp}$ and, therefore, $\Delta p_{\rm L}^{\perp}$. This is the case, for example, for dipolar interactions, as can be checked from quantal calculations. However, the very fact that CTMC calculations, in which dipolar processes are strongly suppressed [7], give rise to similar situations (*i.e.* $\Delta P_{\rm p}^{\perp} \ll \Delta P_{\rm t}^{\perp}$) *proves* that it does not necessarily correspond to a dipolar interaction, as often assumed in the analysis of experiments [6]. Indeed, the two-center nature of the electronic potential allows processes in which a small transverse momentum transfer from projectile to target goes along with a strong electron-target nucleus transverse momentum transfer (see Fig. 6 of Ref. [6]). In fact, equation (7) shows that there is nothing exceptional in this since it corresponds to

$$\Delta P^{\perp} \simeq m_{\rm p}/(m_{\rm p}+m_{\rm t})\,\Delta p_{\rm L}^{\perp} \tag{13}$$

Now, equation (3b) for $\Delta\mathbf{P}$ does not depend on masses while condition (13) depends on $m_{\rm p}/m_{\rm t}$. Therefore, for identical potentials, the satisfaction of condition (13) will depend on nuclear masses. This is a further confirmation that the case of small transverse momentum transfer to the projectile does not imply necessarily a dipolar photon-like interaction.

4 Conclusion

We have shown that the classical impact parameter formulation is completely adequate to describe many particle interactions. It allows a more thorough analysis of multi-electron processes in terms of independent electron excitations and internuclear interaction, as exemplified with a few conclusions that can be reached straightforwardly from our formulation. The CTMC method is not the only model available that includes many particle interactions and the coupling between electronic and nuclear motion in a consistent way. The additivity theorem shows that it is equivalent, for small ϵ, to a separate treatment of the electronic and nuclear interactions, exactly as for the quantum impact parameter case [8]. Exactly as in the quantum case, one should speak of a *two-center* situation (rather than a *three-body*) since the evolution of the electrons within the field of the nuclei may be separated from the evolution involving the internuclear interaction. The *additivity theorem* turns out to be a valuable tool to study more complex situations like collisions involving molecules.

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