LETTER TO THE EDITOR

A quasi classical approach to fully differential ionization cross sections

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Abstract. A classical approximation to time dependent quantum mechanical scattering in the Møller formalism is presented. Numerically, our approach is similar to a standard Classical–Trajectory–Monte–Carlo calculation. Conceptually, however, our formulation allows one to release the restriction to stationary initial distributions. This is achieved by a classical forward–backward propagation technique. As a first application and for comparison with experiment we present fully differential cross sections for electron impact ionization of atomic hydrogen in the Erhardt geometry.

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Classical models and approximations are frequently used for atomic and molecular problems, despite their inherent quantum nature. One of the reasons is that our intuitive understanding is mainly based on classical terms and pictures by which we are surrounded in everyday life. Another reason for not doing (fully) quantum mechanical calculations is the complexity of a problem: fully differential cross sections in higher dimensional atomic systems, e.g., often require a numerical effort still beyond present computing power. Only recently the quantum mechanical Coulomb three–body scattering problem was solved numerically [1, 2].

On the other hand, a remarkably successful classical approach to collisional atomic problems has been developed over the years, the so called Classical–Trajectory–Monte–Carlo method (CTMC). It was introduced as a purely classical model based on a “planetary atom” with a major axis of two meters (!) [3]. This model has produced reasonable results for total or energy differential ionization cross sections on atomic hydrogen [4, 5] and for other few–body Coulomb collision processes [6].

Attempts to reduce the limitation of this classical model aimed at changing the description of the initial state for the hydrogen atom from a microcanonical distribution to one, that is closer to the quantum density [7, 8]. Another idea was to introduce additional ad hoc stabilisation potentials in order to be able to treat multi–electron targets [9]. However, all attempts took as a starting point not the quantum problem but the previously formulated classical model. Hence, the proposed amendments were accompanied by inconsistencies or the need of ”fit parameters” determined from cross sections. In the end, one must say that it is up to now not possible to describe higher differential cross sections or targets with more than one active electron consistently in a classical collision framework.
To achieve progress in this situation we decided to go one step backwards and start with a time dependent quantum mechanical scattering formalism. By following the approximations which lead to the classical description, i.e. the CTMC method, we can identify the source and nature of deviations between the classical and the quantum result which serve as a guide to improve the classical description to a quasi–classical approximation.

We divide the problem into three logically separate steps: (1) preparation of the initial state before the collision, (2) propagation in time, and (3), extraction of the cross section. For a consistent quasiclassical picture each of these steps has to be approximated in the same way. To keep the derivation transparent, we will concentrate on electron impact ionization of one active target electron in the following.

Step (1): The initial wave function for the collision problem is translated first into a quantum phase space distribution by the Wigner transformation. The resulting Wigner distribution is reduced to a classical distribution \(w(p, q)\) which can be propagated classically in phase space by taking the usual limit \(\hbar \to 0\). The difference to an a priori classical approach is the use and interpretation of negative parts of the distribution: Viewed as the \(\hbar \to 0\) limit of a quantum problem they contribute to the observables in the same way as the positive parts since they do not need to be interpreted as weights for real paths of classical particles. Yet, there arise additional problems when using this type of general initial phase space distributions in the usual classical framework: Most of them are not stationary under classical propagation, their Poisson bracket with the Hamilton function does not vanish, \(\{H, w\} \neq 0\). Hence, the initial target distribution will look very different at the time the projectile has approached and the collision actually happens.

Step (2): The formulation of the propagation is crucial since it must resolve the problem of the non–stationary classical initial distribution, as described above. Traditionally, the time dependent scattering is described by calculating the transition amplitude between initial and final state through the S–matrix, which is in turn related to the t–matrix describing directly the cross section, see, e.g., [11]. In a simplified version where the asymptotic initial and final states are eigenstates of the asymptotic Hamiltonians \(H_{0}^{(i)}\) and \(H_{0}^{(f)}\) one normally writes for the transition amplitude
\[
S_{fi} = \lim_{t \to \infty} \langle f | U(t) | i \rangle
\]  
(1)
where \(U(t) = \exp[-iHt]\) denotes propagation with the full Hamiltonian. By a Wigner transform the quantum time evolution operator \(U(t)\) can be directly transformed with the help of the quantum Liouville operator \(\mathcal{L}_q\), which reduces to the classical Liouville operator \(\mathcal{L}_c\) in the limit \(\hbar \to 0\) [12]. The latter describes the evolution of a phase space distribution \(w(r, p, t)\) according to the Poisson bracket
\[
\partial_tw = \{H, w\} \equiv -i\mathcal{L}_cw
\]  
(2)
in analogy to the quantum evolution of the density matrix \(\rho\) generated by the commutator,
\[
\partial_t\rho = -i[H, \rho].
\]  
(3)
Hence, we could directly use the translation of (1) to classical mechanics via the Liouville operator. In connection with the microcanonical initial state distribution this is indeed equivalent to the CTMC formulation [13]. However, using non–stationary initial state distributions is inconsistent with the reduced quantum description of (1).
which relies on the fact that the asymptotic states are eigenstates of $U_0$ and therefore stationary. Instead we have to go back to the full scattering formulation

$$S_{fi} = \langle f|\Omega_+^\dagger\Omega_+|i\rangle,$$

where

$$\Omega_\pm = \lim_{t \to \pm \infty} U_0^\dagger(t) U_0(t)$$

are the Møller operators. The meaning of $\Omega_+$, e.g., is to propagate backwards with $U_0(t)$ using the asymptotic Hamiltonian $H_i^0$ without the projectile–target interaction and then forward again under the full Hamiltonian with $U(t)$. Again, with the help of the Liouville operator we can translate the Møller operators to their classical analogue, thereby obtaining a prescription how to propagate a non-stationary initial phase space distributions $w_i(\gamma)$, where $\gamma = (\vec{p}_1, \varphi_1, \vec{p}_2, \varphi_2)$ is a point in the 12-dimensional phase space:

$$w_f = \lim_{t \to +\infty} \lim_{t' \to -\infty} e^{-i\mathcal{L}_f t'} e^{i\mathcal{L}_c (t-t')} e^{i\mathcal{L}_i t'} w_i \equiv \mathcal{K} w_i.$$  

The difference to (1) are the explicit propagations under $\mathcal{L}_f$ and $\mathcal{L}_c$ in the initial and final channel (which need not be the same). The meaning of (1) becomes very transparent if we insert a discretized distribution, which is used in the actual calculations, $w_i(\gamma) = \sum_n w_n \delta^{12}(\gamma - \gamma_i^n)$. The final distribution reads

$$w_f(\gamma) = \mathcal{K} w_i = \sum_n w_n \delta^{12}(\gamma - \gamma_f^n)$$

where each phase space point $\gamma_f^n$ emerges from $\gamma_i^n$ through solving successively Hamilton’s equations, first with $H_i^0$, then with $H$, and eventually with $H_f^0$. With this propagation scheme a non-stationary initial distribution will spread when being propagated backwards with the asymptotic $\mathcal{L}_c$. However, it will be refocused under the following forward propagation with $\mathcal{L}_c$. Hence, when the actual collision happens for $t \approx 0$ the original target distribution is restored, slightly polarized by the approaching projectile.

Hence, there is no more need for the initial distribution to be classically stationary. We are able to use any phase space distribution as a target in our quasi classical approach. This also includes unstable multi-electron targets, e.g., classical helium.

Step (3): Before we come to the actual evaluation we have to formulate the cross section such that it can make full use of the non-stationary initial phase space distribution $w_i(\vec{p}_1, \varphi_1)$, where “1” refers to the target electron. Without modification the total energy $E$ of the final state forces by energy conservation for each classical trajectory only those parts of the initial phase space distribution to contribute to the cross section which have the same energy $E$. However, this would bring us essentially back to the microcanonical description. In order to make the entire non-stationary initial state distribution “visible” to the collision process, we use the energy transfer $E_1 = E_f^{(f)} - E_i^{(i)}$ to the target electron rather than its energy $E_f^{(f)}$ itself as a differential measure. Of course, as long as the initial state is on the energy shell with well defined energy $E = E_i^{(i)} + E_2^{(i)}$ the new definition coincides with the usual expression for the cross section,

$$\frac{d^5\sigma}{d\Omega_1 d\Omega_2 dE_1} \bigg|_E = \frac{d^5\sigma}{d\Omega_1 d\Omega_2 dE_1} \bigg|_{E_i} = \frac{d^5\sigma}{d\Omega_1 d\Omega_2 dE_1} \bigg|_{E_f^{(f)}}.$$  

where $d\Omega_i$ are the differentials for the solid angles of the two electrons, respectively.
To extract this cross section we have to evaluate the phase space integral
\[
\frac{d^5\sigma}{d\Omega_1 d\Omega_2 dE_1} = \int dx_2 dy_2 d\vec{p}_1 d\vec{q}_1 \prod_{i=1}^2 \delta(\Omega_i^{(f)} - \Omega_i)\delta(\bar{E}_1^{(f)} - \bar{E}_1)w_i, \quad (9)
\]
where the integration is over the initial state variables, namely the impact parameter area \(dx_2 dy_2\) and the phase space of the (bound) target electron \(d\vec{p}_1 d\vec{q}_1\), with initial distribution \(w_i(\vec{p}_1, \vec{q}_1, x_2, y_2)\). The propagated angles \(\Omega_i^{(f)}\) of the electrons as well as the energy transfer \(\bar{E}_1^{(f)}\) have to coincide with the desired values \(\Omega_i\) and \(\bar{E}_1\) to contribute to the cross section (9) which is a generalization of the one derived in [14], e.g., where the initial bound state was assumed to live on a torus, i.e., \(w_i(\vec{p}_1, \vec{q}_1) = \delta(\vec{I}(\vec{p}_1, \vec{q}_1) - \vec{I}_0)\) with a well defined multidimensional action \(\vec{I}_0\).

Finally, we have to respect the Pauli principle for the two identical electrons. Formally, this can be done easily in the Wigner transform for the two ionized electrons in the final state. In the limit \(\hbar = 0\) one is left with the usual classical symmetrization, i.e., an interchange of indices. To keep the notation simple we have omitted symmetrization in the outlined derivation, however, it is included in the actual computation which is carried out by applying standard CTMC techniques to evaluate (9).

As a first application we discuss fully differential ionization cross sections for atomic hydrogen comparing three data sets: a calculation with the
standard microcanonical distribution (CTMC), one with the non-stationary Wigner distribution in our quasi classical framework and experimental data at impact energies of 250eV (fig. 1) and 54.4eV (fig. 2), respectively.

For each impact energy about $10^8$ trajectories have been calculated. The cross section at 250 eV for the Wigner distribution still exhibits negative parts, indicating that this cross section is not yet fully converged. This is not surprising if one takes into account that the fraction of phase space of the final state is so small with the chosen bin sizes for energies and angles, that only between 100 and 300 events finally contribute to the shown cross sections. However, a considerable advantage of the present method is that a sampling of $10^8$ trajectories contains the complete scattering information, not just one specific differential cross section.

The figures show, that the microcanonical distribution, i.e. standard CTMC, is not able to reproduce the binary peak \[17\], whereas with the Wigner distribution it is reproduced fairly well for 250eV and rather well for 54.4eV impact energy. Keeping in mind that in contrast to the microcanonical distribution the Wigner distribution has the correct probability densities in momentum \textit{and} configuration space, one can conclude, that at least for energies between 50 and 250 eV the differential cross sections "image" the initial \textit{phase space} distribution. The present approach is still a classical approximation and cannot reproduce quantum effects. Therefore, features in the cross section, for which coherence is crucial are represented purely or not at all.

To summarize, we have shown, that a consequent classical approximation to a quantum system can give much better results compared to those from an a priori classical model, though both approaches are realized numerically in almost the same way. However, the main difference is conceptual: in the usual classical limit each
individual trajectory represents that of an electron obeying the classical equations of motion, whereas in our classical approximation only the entire phase space distribution is meaningful and individual trajectories are only discretized points of the distribution evolving in time. Hence, there is no problem to deal with "negative probabilities" in the initial distribution, since we regard them not as probabilities but only as weights of the integration which, of course, may be negative.

However, the use of non-stationary distributions like the Wigner distribution as an initial state implies additional difficulties for a scattering description which we have overcome by using a forward-backward propagation scheme akin to the quantum Møller formalism and by a reformulation of the energy differential cross section in terms of the energy transfer during the ionization process. With these modifications all the tools of the standard CTMC technique can be applied straightforwardly.

Moreover, our approach can be in principle generalized to multi–electron targets since we generate our initial phase space distribution from a quantum wave function and we know how to deal with non–stationary initial distributions.

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