Applicability of atomic collisional ionization cross sections in plasma environment

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Abstract

Secondary ionization is an important process in a plasma environment. Plasma environment emerges also in matter driven by high intensity X-rays. It is difficult to treat such systems by theory, because of their complex non-equilibrium dynamics. That is also a reason why usually cross section data of isolated atoms are used in existing modeling frameworks. However, these cross sections may be modified by the plasma environment. Our aim is to explore this problem using the classical Molecular dynamics technique.

Contents

1 Introduction: High intensity x-ray induced dynamics in matter

1.1 Ultrabright X-ray sources

X-rays cover the spectrum of electromagnetic radiation between the wavelength 0.01 nm – 10 nm and photon energy 100 eV – 100 keV, correspondingly. This photon energy is enough to ionize atoms and disrupt molecular bonds, while the wavelength is in the range of the atomic distances leading to strong diffraction phenomena due to the atomic structure. These are the reasons for using X-rays widely in materials sciences.
Today there are several types of sources producing X-rays. The rapid progress in the development of power light sources led to the construction of X-ray free-electron lasers (XFEL). These lasers generate coherent X-ray radiation by single pass of a relativistic electron beam through a long undulator. XFEL combines high intensity of the pulse and high photon energy. Hence it offers new possibilities, which were unavailable before. They produce not only radiation of unprecedented brilliance, but radiation that has full transverse coherence and is pulsed with typical ultrashort pulse lengths of less than 100 fs. At the XFEL facilities the radiation is used to study the structure and dynamics of matter. Therefore it is necessary to have a strong theoretical background for describing high-energy X-ray induced processes.

1.2 Characteristics of x-ray - matter interaction

In intense X-rays the predominant process due to the electromagnetic wave-matter interaction is sequential single photon absorption of the atoms. Other processes such as Auger and fluorescent decays then also occur in the inner shell, following the photoionization event.
In a molecular environment all previous processes take place together with the followings:

- collisional (secondary) ionization
- recombination
- valance electron change transfer, when charge imbalance is built up between neighboring atoms [4]

Consequently, plasma formation is possible, even within nanosize samples [5].

1.3 Theoretical simulations of high intensity x-ray induced dynamics

High intensity X-ray driven systems with large number of ions and ionized electrons are difficult to treat by theory, because of their complex structure and their non-equilibrium dynamics. For this purpose special simulation tools XATOM and XMDYN[1] have been developed.

XATOM can calculate cross sections and rates of x-ray-induced atomic processes, based on nonrelativistic quantum electrodynamics and perturbation theory within the Hartree-Fock-Slater model. Such data can be calculated for arbitrary electronic configuration, including multiple-hole states, of any atomic species. These atomic data are used not only for simulating ionization dynamics of atoms within XATOM, but also as input for XMDYN simulations for complex systems.
In XMDYN electronic configurations of atoms and ions are followed by tracking the occupation of the orbitals using a Monte Carlo algorithm. One run generates temporal snapshots of a single trajectory which is a realization of the temporal evolution of the system affected by stochastic damage processes. XMDYN takes into account all processes listed in Sec.1.2, including collisional ionization.

2 Project goal: investigation of collisional ionization in plasma

2.1 Collisional ionization for an isolated atom

Collisional ionization is a process when an incoming electron ionizes an atom or an ion and two (or more) electrons leave the system. The outcome depends on the kinetic energy of the incoming electron K and the electronic configuration of the atom shaped primarily by the atomic potential. This scenario can be described with the cross section parameter σ and with the differential cross section $d\sigma/dE$. Formulae exist for the approximation of the atomic cross section, e.g. the famous one introduced by Lotz or the more complex BEB formula.

2.2 Collisional ionization in modeling of complex systems

Collisional ionization has an important role in an ionized environment in shaping the dynamics. Therefore theoretical models addressing such systems, such as XMDYN, do include this process. In the treatment usually atomic cross sections are used due to the lack of more accurate data. However, in a plasma environment, especially at high electron densities, it cannot be ensured that data derived for isolated atomic system accurately describe the process. The current project aims for investigating this problem in two steps.

1. Deriving accurate cross section data for a simple isolated atomic system.

2. Comparing key properties of collisional ionization dynamics in a plasma yielded from atomic data via kinetic equations to the same properties extracted from numerical simulations tracking the process in its entirety, e.g. without any approximation.

The program is computationally feasible for purely classical systems.

3 Model systems

3.1 Collisional ionization for an isolated classical atom

Here we investigate an isolated classical 'Hydrogen-like' atom with one bound electron and another one, coming from (quasi-)infinity. The objects are treated as point particles and their dynamics is followed using the classical Molecular dynamics technique with Velocity-Verlet integrator.
The divergent Coulomb potential leads to instability in numerical calculations, therefore, instead, we used a soft core Coulomb interaction between charges:

$$V = const * \frac{1}{\sqrt{r^2 + r_0^2}} \tag{1}$$

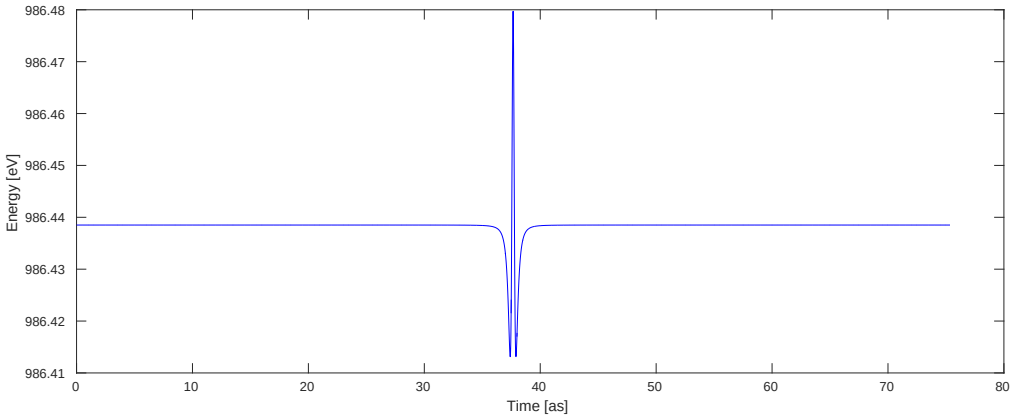
We choose softening parameter values satisfying the following points:

- we set the softening parameter $r_{0,ae}$ used in the electron-ion interaction in a way that a classical electron in rest at $r = 0$ distance has a binding energy equal to 13.6 eV, the electron binding energy in a real Hydrogen atom.
- we set the softening parameter $r_{0,ee}$ used in the electron-electron interaction in a way that at the soft potential does not deviate significantly from the pure Coulomb potential at 1000 eV, the maximum incoming electron energy in our study. This is important to describe electron electron elastic collision correctly, e.g. avoiding direct crossing of an electron through another.

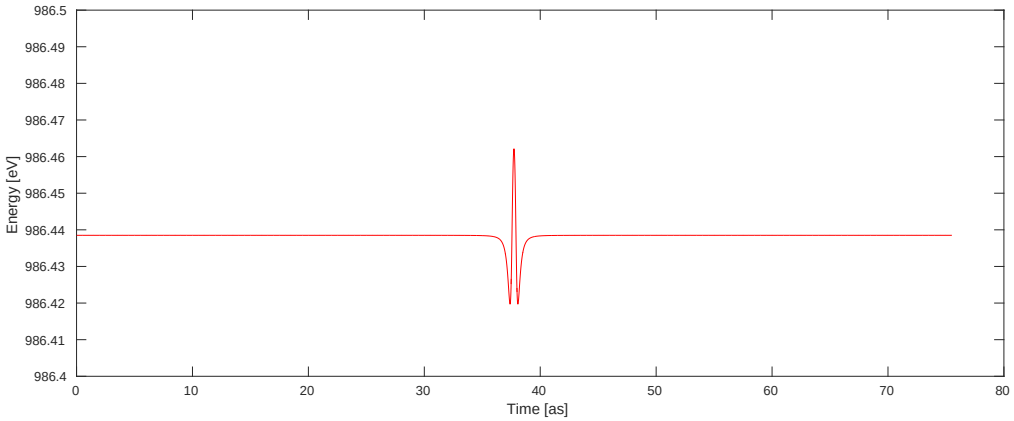
These points are satisfied using $r_{0,ae} = 1.06$Å and $r_{0,ee} = 0.01$Å values.
Further, we had to find a proper dt timestep for the molecular dynamics algorithm that leads negligible violation of the energy conservation. For this we required that at direct collision ($b = 0$) at the maximum incoming electron energy the numerical error ΔE is negligible compared to the binding energy B, e.g.

$$\Delta E << B \tag{2}$$

Using the XMDYN [1], it is found that timestep $dt = 0.01$ as is enough for kinetic energy $K = 1000eV$. We note that going much above 1000 eV kinetic energy the collision properties using soft potential differ from using the pure Coulomb potential, but it does not affect significantly our simulations as (i) at high energies the cross section of collisional ionization is small and (ii) using the previous timestep value the violation of energy conservation is small in the MD propogation.
Although the chosen timestep is also enough for lower energies, below 100 eV we increased the timestep 10 times to speed up calculations without violating much the energy conservation law.
In Fig. 1 we demonstrate the behavior of energy conservation for impact parameter values 0.01 Å 0.02 Å or 0.05 Å.



a)



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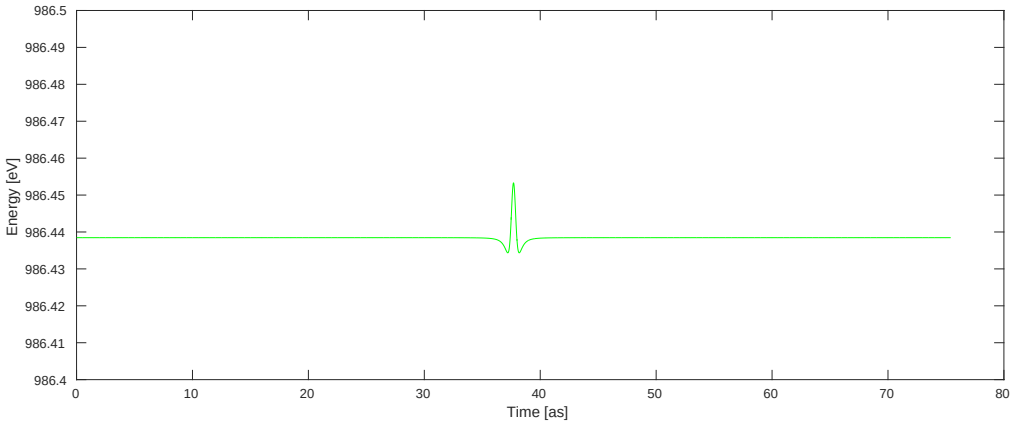


c)

Figure 1: Energy conservation with softening parameter: a) 0.01Å b) 0.02Å c) 0.05Åand kinetic energy 1000 eV

After a scan over the initial distance of the incoming electron from the atom we found 7Å to be appropriate that we used in all simulations.
The outcome of an event in our model depends not only on the kinetic energy K and the atomic potential, but also on the impact parameter b. For example, there will be no perceptible interaction between two electrons, if the impact parameter is too big and Coulomb forces are negligible. On the other hand, it is important to mention that if $b = 0$, there is no secondary ionization too, but there is a replacement of the bound electron by the incoming one. This way we need to introduce minimum and maximum impact parameters for a given kinetic energy, between which secondary ionization occurs. They are b_{min} and b_{max} respectively. In a purely classical system the outcome of a process is fully defined by the initial conditions, therefore the cross section can be connected directly to the impact parameter. The cross section simply read as

$$\sigma = \int_{b_{min}}^{b_{max}} 2\pi b db \tag{3}$$

3.2 Collisional ionization within a plasma environment

For this study our test system is the classical atom introduced in Sec. 3.1, but embedded within a thermalized classical electron plasma. Electrons by themselves do not form a stable plasma as such a system would simply explode due to the repulsive Coulomb forces. Therefore, one may introduce a harmonic potential

$$V_H = -0.5 * D * r^2 \tag{4}$$

acting on all electrons that playes the role of a homogeneous positively charged background to stabilize the plasma. The parameter D is set according to the required electron density n:

$$D = \frac{e}{3\epsilon_0 * n} \tag{5}$$

The phenomenon of 'electron replacement', e.g. when an incoming electron replaces the originally bound electron, makes it difficult to identify 'true' (e.g. 'one electron in, two elecreons out') collisional ionization events in a plasma environment. Therefore, we investigate the process of 'bound electron release', e.g. we are focusing exclusively on the release dynamics of the originally bound electron. In this picture the cross section is defined only through b_{max} (e.g. $b_{min} = 0$ in Eq. 3) as below b_{min} the event of bound electron release does occur.

4 Results

4.1 Cross section for the isolated atom

A dedicated matlab code has been developed for mapping out the cross section for the classical model atom. This codes includes real space propagation, automatic identifica-

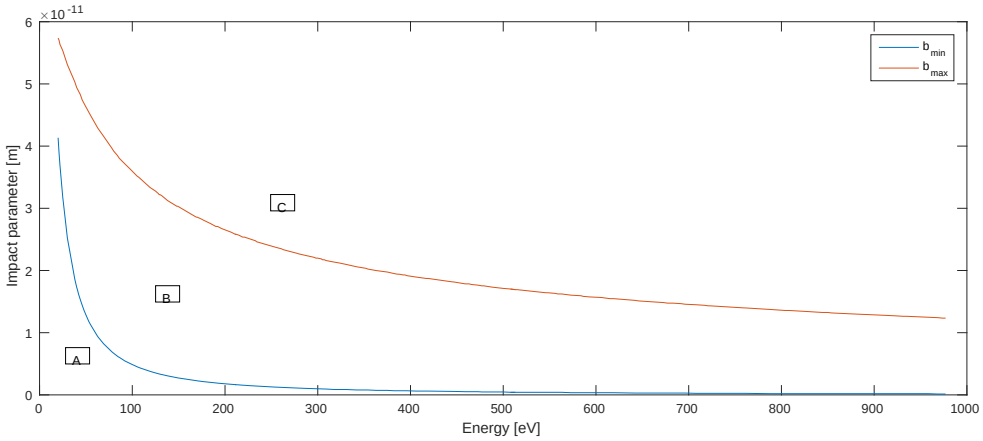
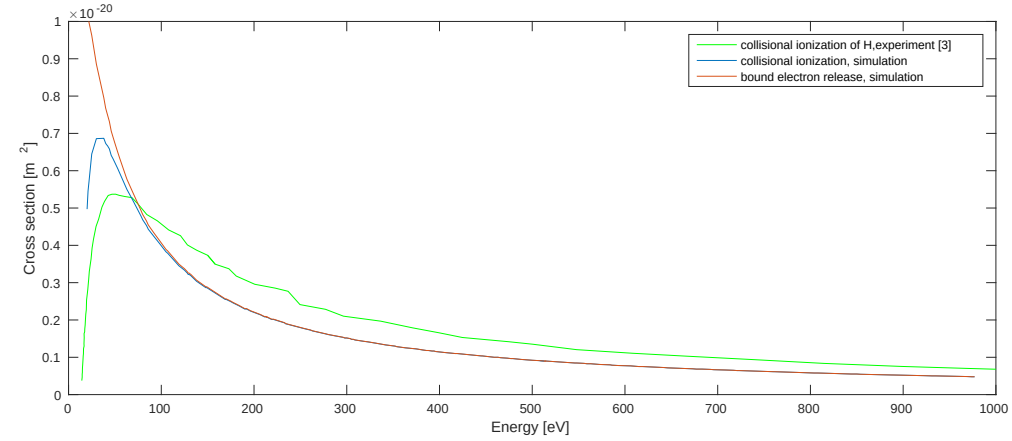


Figure 2: Threshold impact parameters

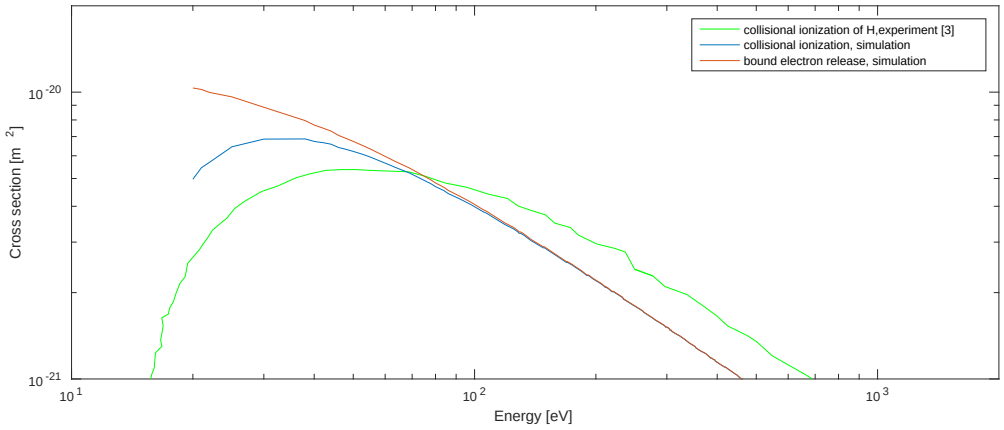
tion of reaching the asymptotic states (needed for the determination if ionization event happened) and an iteration scheme to refine b_{min} and b_{max}.
Fig. 2 shows b_{min} and b_{max} extracted from the simulations. The curves divide the energy – impact parameter space into three regions. In area A there is no collisional ionization, but the replacement of the bound electron by the incoming one. Secondary ionization takes place in the area B between curves $b_{max}(E)$ and $b_{min}(E)$. Finally, in the area C, where impact parameter is bigger than b_{max}, no secondary ionization occurs, but, due to the possibility of transferring energy of arbitrary small amount in a classical system, the excitation of the bound electron happens.
Based on b_{min} and b_{max} one can calculate the cross section, shown in Fig. 3. Although our model atom is a classical system and does not have a pure Coulomb potential, it is interesting to see the reasonable agreement with experimental data on Hydrogen atom [3]. The same qualitative behaviors can be found showing the low energy peak and the decay at high energies.
By plotting the cross section on log-log scale (Fig. 3 b) one finds an asymptotically parallel behavior between our simulation and the experimental data. In this region the Born approximation is valid [3]:

$$\sigma(E) = A\log(E)/E + B/E \tag{6}$$

Based on this the asymptotic value of the slope can be estimated. When $E \to \infty$, the last term can be neglected and the logarithm of the equation (6) is



a)



b)

Figure 3: Cross sections: a)linear scale, b) logarithmic scale.

$$\log(\sigma(E)) = \log(A \log E) - \log(E) = \log(A) = \log(\log(E)) - \log(E) \tag{7}$$

Then if we define $\log(\sigma(E))$ as s, $\log(E)$ as ϵ and take $s\prime$, it will be

$$s\prime = -1 + \frac{1}{\epsilon} \tag{8}$$

And now it is clear, that the larger ϵ, the closer derivative is to the value -1. That is why there is a linear dependence on Figure 3(b).

5 Perspectives for the plasma analysis

The time available did not permit to perform the numerical analysis, however, in this section we outline the program.

5.1 Dynamics of the atom + electron plasma system

– Configurations consistent with a thermalized electron plasma can be generated using XMDYN, starting from a set of electrons with random initial positions and velocities and using the Berendsen thermostat. Thermal equilibrium is reached in time when the thermostat does not change monotonically the total energy of the system. Note that one should transform out the center of mass motion (momentum) as well as the total angular momentum from the starting configuration.
– By running XMDYN with the thermostat option one gets a trajectory that can be sampled for initial configurations for the collisional ionization simulations. By taking a snapshot and placing the classical atom in the center (at $r = 0$) the MD calculation can be run with the timestep and potential parameters used in Sec. 3.1.

5.2 Analysis

5.2.1 Collisional rate

A natural quantity to be used as a basis of comparison is the ionization rate. For a homogeneous electron plasma with constant density ρ_0 the rate w of collisional ionization can be calculated within the kinematic description:

$$w = \int d^3v \sigma(|\mathbf{v}|)|\mathbf{v}|\rho_0 f_v(\mathbf{v}) \tag{9}$$

where $f_v(v)$ is Maxwell - Boltzmann distribution function.
However, in the case of the numerical simulation, it is not straightforward how to identify the moment of electron release within the other plasma electrons. A plausible definition is to choose it as the time t when the energy of the originally bound electron becomes positive in the potential of the ion (disregarding all other particles). We emphasize, that this definition is physically motivated, but its validity is rigorously not proven.

5.2.2 Time evolution of displacement from the ion

A rigorously defined quantity that should reflect the event of electron release in its time evolution is the distance of the bound electron from its initial position, R. Using the kinematic picture the average distance is:

$$< R(T) >= \int_0^\infty dv \int_0^\infty du \int_0^T dt f_v(v) f_u(u,v) f_t(v,t) u(T-t) \tag{10}$$

Where v is electron's initial velocity, u – its velocity after the collision, $f_v(v)$ is Maxwell-Boltzmann distribution function and the others functions are

$$f_u(u,v) = \frac{1}{\sigma(v)} \frac{d\sigma(u,v)}{du}$$
$$f_t(v,t) = w(r) e^{-w(v)t}$$

It is straightforward to extract $< R(T) >$ from the molecular dynamics simulations, therefore the comparison could be performed.
We note that as we have a rate w defined and averaged distance $< R(T) >$, it will be reasonable to look for characteristic changes of $< R(T) >$ at the average release time $1/w$.

6 Summary

In summary, we numerically analyzed a classical collisional ionization scenario, a process when classical model atom is hit by an incoming classical electron, and another electron may be released. We extracted the collisional ionization cross section for this system that compares reasonably well to experimental data on Hydrogen. Our results serve as a basis for a follow-up investigation aiming for the analysis of the atomic collisional ionization cross sections modified by a plasma environment. As part of the project we outlined a program towards this goal.

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