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# Impact-parameter dependence of the electronic energy loss of fast cluster projectiles

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**Abstract**

Electronic energy loss of molecular clusters as a function of impact-parameter is less understood than atomic energy loss. Vicinage effects due to mutual interference between cluster fragments play a key role in the determination of the cluster electronic energy loss. In this work, we describe a molecular extension of the perturbative convolution approximation (PCA) energy loss model, namely MPCA (molecular PCA), which yields remarkable agreement with first-order Born semiclassical approximation (SCA) results. The physical inputs of the model are the oscillators strengths of the target atoms and the projectile electron density. A very good agreement is obtained with time consuming full first-order calculations for bare incident molecular clusters for several angles between cluster axis and velocity direction.
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## 1. Introduction

Beams of molecules and ionic clusters are useful tools in both fundamental research and in material science and plasma physics. The effects of a cluster clearly deviate from the sum of individual effects of each cluster component.

In particular, cluster-beam experiments were reported in mid 70s by Poizat and Remillieux [1] and, not much time after, the first evidence of the vicinage effect was reported by Brandt et al. [2]. Since then, it is established that the cluster energy loss is different from the sum of energy losses of its separated components. An increased energy transfer due a cluster may even be used in inertial nuclear fusion processes [3,4].

If the ions enter along a principal axis of a crystalline target, their motion will be guided due the

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correlated collisions with the target atoms. These ions, then, are said to be channeled. The channeling motion of a molecule (or a cluster) will also depend on the Coulomb heating phenomenon (i.e. an increased transverse energy of the cluster fragments due to the mutual Coulomb repulsion), discovered in mid 70s by Caywood et al. and Poizat and Remillieux [5,6]. Recently, the Coulomb heating was simulated [7,8] and experimentally determined in a quantitative way [8].

The cluster stopping-power can be theoretically treated by the united-atom model [9,10], that describes the cluster as an equivalent single atom, with atomic number and mass being the sum of atomic number and mass of each component. However, that model is limited to the very beginning of the interaction between the cluster and the target, therefore, it cannot be used to understand several channeling key effects, for instance, the Coulomb heating effect. An important theoretical treatment is the dielectric formalism in a homogenous electron gas target [2,11] (a detailed review about vicinage effect and dielectric formalism for clusters can be found in [12]) and, to account for the target-core effect, the local density approximation (LDA) model (used for clusters in [13]). However, although successful for homogeneous targets, the dielectric formalism cannot be used to easily describe the cluster energy loss under channeling conditions, where the target cannot be treated as being homogeneous. Then, a theoretical investigation of the cluster stopping-power under channeling conditions, considering the Coulomb heating, requires the use of the impact-parameter method, as presented by Jensen et al. [14] but for distant-collisions only.

This work describes an extension of the perturbative convolution approximation (PCA) model [15,16], based on the impact-parameter method, for molecules and clusters, namely MPCA (molecular perturbative convolution approximation). MPCA gives the energy loss as a function of impact-parameter without time consuming first-order calculations using a set of thousands of final states for both distant- and close-collisions. The starting point of the model is the diatomic molecule. The physical inputs of the model are the target oscillator strengths, the target electronic density, the projectile screening function and the molecular alignment angles.

## 2. Model

The MPCA (molecular perturbative convolution approximation) model is an extension of PCA model [15,16] made for cluster projectiles. Here, we present only a short outline of the PCA method but special attention will be draw to the interference terms that arise from the sum of all ionic potentials. The electronic energy loss is calculated from the expression

$$Q(b) = \sum_{\beta} \left| a_{\beta}(\vec{b}) \right|^2 (\varepsilon_{\beta} - \varepsilon_0), \tag{1}$$

which involves a sum of all final target states with energy $\varepsilon_\beta$ ($\varepsilon_0$ is the ground-state energy) and the corresponding calculation of all transition amplitudes $a_\beta$ for each cluster impact-parameter $b$ (the minimum distance between the center of the cluster and the target nucleus). In order to calculate the energy loss due to target ionization and excitation in a first order perturbation framework, we have to consider the amplitudes for each electronic transition between the initial state $|0\rangle$ to a final state $|\beta\rangle$ due to the cluster with $N$ ions.

$$a_{\beta}(\vec{b}) = -i \int_{-\infty}^{+\infty} \mathrm{d}t \, e^{i(\varepsilon_\beta - \varepsilon_0)t} \left\langle \beta \left| \sum_{i=1}^{N} V_i(\vec{r} - \vec{R}_i(t)) \right| 0 \right\rangle, \tag{2}$$

where $V_i$ is the interacting potential between the $i$th ion (whose charge is $Z_i$) in the cluster and the target electron. The $i$th ion position in space is $\vec{R}_i(t)$ and $\vec{r}$ is the target electron coordinate, both relative to the target nucleus. In the first-order treatment the transition amplitude is only a coherent sum of amplitudes due to each ion of the cluster. If not indicated otherwise, all calculations throughout this work are in atomic units ($\hbar = m_e = e = 1$).

The characteristic time of interaction between the cluster and the target is much smaller than the characteristic times of vibrational and rotational cluster transitions. Therefore, the corresponding degrees of freedom can be neglected.

Taking these assumptions into account, all calculations shall be done for a cluster projectile with impact-parameter $\vec{b}$ with respect to the cluster center. For a straight-line projectile motion without vibrational and rotational degrees of freedom, the time dependent position of the $i$th projectile nucleus is given by $\vec{R}_i(t) = \vec{b}_i + \vec{v}t + \vec{d}_{iz}$, where $\vec{d}_i$ is the distance between the molecule center and the $i$th ion, $d_{iz}$ is the $z$-component of $\vec{d}_i$ (being $d_{iz} = \|\vec{d}_{iz}\|$), $\vec{d}_{i\rho}$ is the transversal component of $\vec{d}_i$ (being $d_{i\rho} = \|\vec{d}_{i\rho}\|$), $\vec{b}_i$ is given by $\vec{b}_i = \vec{b} + \vec{d}_{i\rho}$, which is the $i$th ion impact-parameter and $\vec{v}$ (parallel to the $z$ direction) is the cluster velocity. Fig. 1 shows these vectors for a diatomic molecule. The angle $\theta$ in Fig. 1 refers to the angle between the diatomic molecule axis and the $z$-axis. Finally, for the same molecule, the angle $\phi$ refers to the angle between $\vec{d}_{i\rho}$ and the $x$-axis.

The interacting potential $V_i$ may be one of the following (more details about these potentials are given in [16]):

(a) the Coulomb potential, that describes the potential induced by a moving point charge in vacuum;
(b) the Bohr potential, that describes a potential produced by a external point charge immersed in a homogenous electron gas. The screening parameter ($\alpha_i$) can be obtained either from the Debye screening length [17] or from the generalization of the Friedel sum rule for finite velocities derived by Lifschitz and Arista [18];
(c) the single-zeta potential, that describes the potential due a projectile carrying one or two bound electrons ($n_i = 1, 2$) in hydrogen-like 1s orbitals.

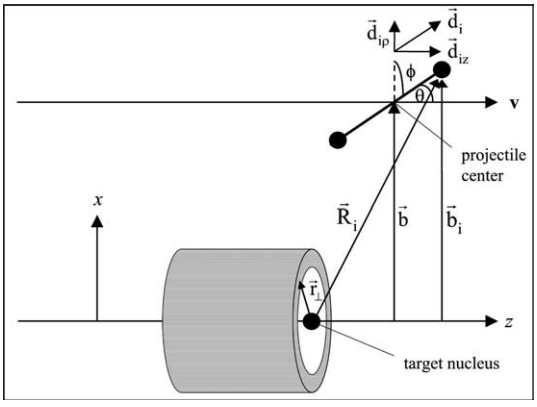


Fig. 1. Representation of the collision geometry, showing the target nucleus, the target electronic distribution, the projectile nuclei, the impact-parameter vectors and the projectile velocity.

It is important to point out that not all ions of the cluster are necessarily generating the same kind of potential. Due to dynamic capture-loss processes, it is possible to find one ion of projectile cluster completely ionized, while its neighbors, after capturing an electron during ion–matter interactions, can have a single-zeta potential. This possibility must be taken into account in computer simulation codes.

According to the atomic PCA model, in a first step we shall find approximations for $Q(b)$ (Eq. (1)) that are valid for a limited range of impact-parameters and in a second step these approximations should be linked. At large impact-parameters the dipole-approximation for $V_i$ can be used, and thus, an analytical expression [19,20] for $Q(b)$ may be obtained. Inserting the cluster interacting potential, we have, for large impact-parameters, an expression of the form

$$Q^{\text{dipole}}(b) = \sum_{i=1}^{N} Q^{\text{dipole}}_{\text{atomic}}(\vec{b}_i) + \sum_{i=1}^{N}\sum_{j>i}^{N} Q^{\text{dipole}}_{\text{int}}(\vec{b}_i, \vec{b}_j), \tag{3}$$

where

$$Q^{\text{dipole}}_{\text{int}}\left(\vec{b}_i, \vec{b}_j\right) = \sum_{\beta} f_\beta \frac{2Z_iZ_j}{v^2} \cos\left(\frac{\omega_{\beta 0} d_{ijz}}{v}\right) \times \left[\frac{2\vec{b}_i \cdot \vec{b}_j}{(b_i b_j)^2} g_\perp(b_i) g_\perp(b_j) + \frac{2 g_\parallel(b_i) g_\parallel(b_j)}{b_i b_j}\right], \tag{4}$$

where, for the Coulomb potential, the functions $g_\parallel(b_i)$ and $g_\perp(b_i)$ read

$$g_\parallel(b_i) = \left(\frac{\omega_{\beta 0} b_i}{v}\right) K_0\left(\frac{\omega_{\beta 0} b_i}{v}\right) \tag{5}$$

and

$$g_\perp(b_i) = \left(\frac{\omega_{\beta 0} b_i}{v}\right) K_1\left(\frac{\omega_{\beta 0} b_i}{v}\right), \tag{6}$$

where $K_0(x)$ and $K_1(x)$ are second kind Bessel functions (for the Bohr and single-zeta potentials, expressions are given in [16]), $f_\beta = 2|\langle\beta|z|0\rangle|^2(\varepsilon_\beta - \varepsilon_0)$ are the oscillator strengths, $d_{ijz} = d_i\cos\theta_i - d_j\cos\theta_j$ and $\theta_i$ is the angle between the $z$-axis and $\vec{d}_i$. The first term in Eq. (3) corresponds to the individual $Q^{\text{dipole}}_{\text{atomic}}(b)$ associated to each ion (see expressions in [16]) and the last one is associated to interference effects (vicinage). The first interference term in Eq. (4), the one associated with $g_\perp$, corresponds to the classical sudden approximation. It is important to point out that for small $b_i$ the function $g_\perp$ ($b_i$) approaches 1 and $g_\parallel$ ($b_i$) approaches 0.

For small impact-parameters, the influence of the target potential can be neglected at high projectile energies, allowing for an analytical expression for $Q^{\text{close}}(b)$ by replacing the final target-continuum states by plane waves. Thus, the energy transfer reads

$$Q^{\text{close}}(b) = \sum_{i=1}^{N} Q^{\text{close}}_{\text{atomic}}(\vec{b}_i) + \int \mathrm{d}^2 r_\perp K^{\text{close}}_{\text{int}}\left(\vec{r}_\perp - \vec{b}\right) \int_{-\infty}^{\infty} \mathrm{d}z\ \rho(\vec{r}_\perp, z), \tag{7}$$

where again the first term corresponds to a incoherent sum of energy losses due to each ion from the cluster (already defined in [15]) and

$$K^{\text{close}}_{\text{int}}(\vec{b}) = \frac{2}{v^2}\sum_{i=1}^{N}\sum_{j>i}^{N} Z_i Z_j h_{\text{int}}\left(\vec{b}_i, \vec{b}_j\right) \tag{8}$$

is the interference term with

$$\begin{aligned} h_{\text{int}}\left(\vec{b}_i, \vec{b}_j\right) = {} & 4v^2 \int_0^1 \mathrm{d}q\, q^2 \\ & \times \cos\left(2vq^2 d_{ijz}\right)\Big\{ q\Big[J_0\left(2vqb_i\sqrt{1-q^2}\right)K_0\left(2vq^2 b_j\right) \\ & + J_0\left(2vqb_j\sqrt{1-q^2}\right)K_0\left(2vq^2 b_i\right)\Big] \\ & + \sqrt{1-q^2}\,\frac{\vec{b}_i}{b_i}\cdot\frac{\vec{b}_j}{b_j}\Big[K_1\left(2vq^2 b_j\right)J_1\left(2vqb_i\sqrt{1-q^2}\right) \\ & + K_1\left(2vq^2 b_i\right)J_1\left(2vqb_j\sqrt{1-q^2}\right)\Big]\Big\}, \end{aligned} \tag{9}$$

where $J_0$ and $J_1$ are first kind Bessel functions.

The function $h_{\text{int}}(\vec{b}_i, \vec{b}_j)$ approaches zero for $b_i \ll 1/v$ or $b_j \ll 1/v$ and, for large values of $b$ (i.e. large values for both $b_i$ and $b_j$), it reaches

$$h_{\text{int}}\left(\vec{b}_i, \vec{b}_j\right) \approx 2\frac{\vec{b}_i \cdot \vec{b}_j}{\left(b_i b_j\right)^2}, \tag{10}$$

recognized as the interference part of the classical sudden approximation result [20] for a diatomic molecule.

In what follows, we propose the following general formula, applicable to all impact-parameters, namely

$$Q(b) = \int \mathrm{d}^2 r_\perp K_{\text{MPCA}}\left(\vec{r}_\perp - \vec{b}\right) \int_{-\infty}^{\infty} \mathrm{d}z\ \rho(\vec{r}_\perp, z), \tag{11}$$

where the kernel is defined as

$$K_{\text{MPCA}}(\vec{b}) = K^{\text{MPCA}}_{\text{atomic}}(\vec{b}) + K^{\text{MPCA}}_{\text{int}}(\vec{b}), \tag{12}$$

where $K^{\text{MPCA}}_{\text{atomic}}$ corresponds to the sum of the energy losses due to each individual ion from the cluster as presented in [15] for a single ion projectile and

$$K^{\text{MPCA}}_{\text{int}}(\vec{b}) = \sum_\beta f_\beta \sum_{i=1}^{N}\sum_{j>i}^{N} \frac{2Z_i Z_j}{v^2}\cos\left(\frac{\omega_{\beta 0} d_{ijz}}{v}\right) \times \left[h_{\text{int}}\left(2v\vec{b}_i, 2v\vec{b}_j\right) g_\perp(b_i) g_\perp(b_j) + \frac{2g_\parallel(b_i) g_\parallel(b_j)}{\sqrt{b_i^2 + b_{\min}^2}\sqrt{b_j^2 + b_{\min}^2}}\right], \tag{13}$$

where $b_{\min} = 1/v^2$ is defined in [20]. This is the molecular perturbative convolution approximation (MPCA).

As can be observed the kernel function in Eq. (13) is based on the expression for large impact-parameters (Eq. (3)) by replacing the interference term $\frac{2\vec{b}_i\cdot\vec{b}_j}{(b_i b_j)^2}$ by $h_{\text{int}}(b_i, b_j)$. In this way, according to Eq. (10), the above energy loss ansatz interpolates smoothly small and large impact-parameters.

In Fig. 2, we compare the dipole and close-collision interference terms with the corresponding MPCA term for two bare diatomic molecule orientations. For both orientations, we can see that MPCA and close-collision interference terms (from Eqs. (13) and (8), respectively) agree with each other for small impact-parameters and the same is observed between MPCA and dipole-approximations (from Eqs. (13) and (4) for large impact-parameters, thus reinforcing the validity of our proposed general formula (11). It should be stressed that the sudden approximation, which

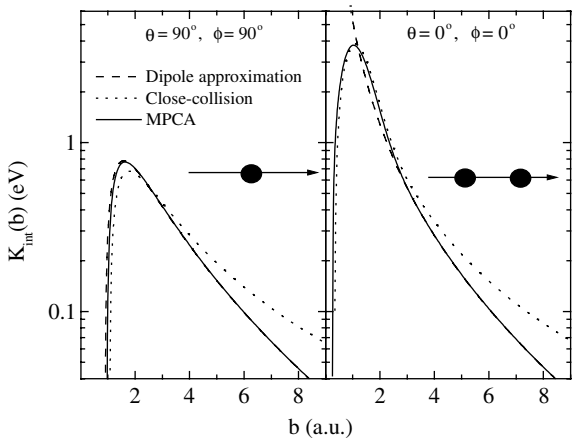


Fig. 2. Comparison between the MPCA model (solid lines), close-collision (dotted lines) and dipole-approximation (dashed lines) interference terms for bare molecules for two possible molecule orientations. In both orientations, we can see an accordance between MPCA and close-collision approximations for small impact-parameters as well as between MPCA and dipole-approximations for large impact-parameters.

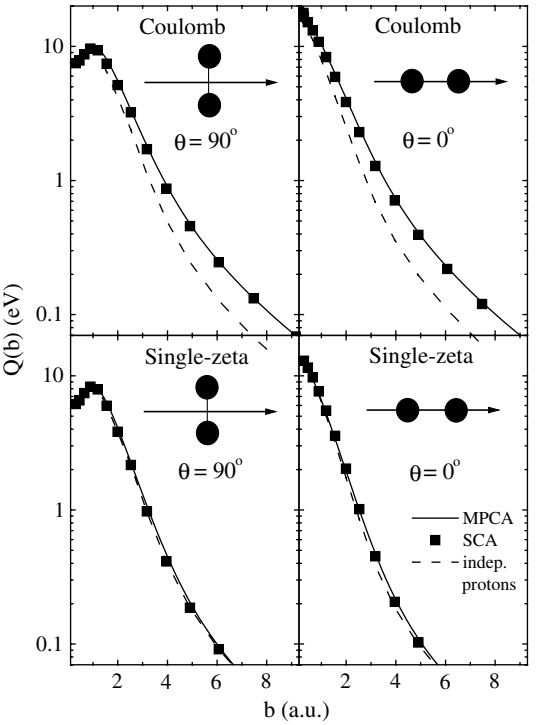


Fig. 3. Comparison between MPCA model and full first-order SCA calculations for 500 keV/amu bare (on top panel) and single-zeta screened (on bottom panel) $H_2$ projectile colliding with H target. The full lines stand for energy loss as a function of impact-parameter as given by the MPCA (Eq. (11)) model. The squares stand for molecular SCA calculations and, to show the interference effect, the dashed lines stand for independent protons SCA calculation.

was used in [15] to link close- and distant-collisions for atomic projectiles, in fact does not link the interference terms for close- and distant-collisions properly. This comes from the fact that the classical sudden approximation does not contain the interference terms due to the phase difference along the $z$-direction (e.g. the cosine term in Eq. (4) and in Eq. (9)).

It is important to point out that Eq. (11) is valid only for a one-electron system. In the framework of the independent particle model, however, it is possible to use Eq. (11) considering the electronic density and the dipole oscillator strengths for each electron of all occupied target shells.

In what follows, only an analysis of the interference term will be performed. The corresponding analysis of the monoatomic terms was already done in [15]. The cluster under consideration will be the diatomic hydrogen molecule. The angles $\theta$ and $\phi$ shown in Fig. 1 will fix the diatomic molecular orientation.

## 3. Discussions and conclusions

In Fig. 3 we see the results of the present model, for two molecule orientations (where $\phi = 0°$), for the impact-parameter dependence of the mean energy loss of bare (top) and single-zeta screened (bottom) $H_2$ molecular projectiles, both at 500 keV/amu, colliding with atomic H (full line). We compare our results with full first-order molecular SCA (semiclassical approximation), similar to the numerical procedure seen in [21] calculations (squares) and with full first-order SCA for two independent protons with the same screen function and impact-parameters as used in molecular SCA (dashed line). In our tests, the interatomic distance was set to 2 a.u. (about 1.06 Å).

About 3500 target states were used in SCA calculations, to ensure an adequate number of partial waves, necessary to calculate $Q(b)$ accurately. Here we have considered two cases. The first one the

molecule has no bound electrons (two protons traveling together, interacting with Coulomb forces) and the second one where one of the proton has captured one electron from the medium ($H^+$ and $H^0$ traveling together).

Fig. 3 (on the top panel) shows a fairly good agreement between molecular SCA and MPCA model. Moreover, it is possible to appreciate the interference terms effect, shown by the difference between MPCA and independent protons SCA. For $\theta = 90°$ and $\phi = 0°$ (i.e. the molecule has its axis orthogonal to its motion and parallel to the impact-parameter direction), the increase of energy loss up to impact-parameter about 1 a.u. is due the choice of the coordinated system. In that orientation and for $b = 1$ a.u., one of the ions (namely the second) will have a head-on-collision with the target. In all orientations, the effect of interference terms leads to an increase in energy loss of about 50% for distant-collisions and less than 10% for close-collisions. That result agrees with the united-atom model for distant-collisions, where the energy loss is proportional to $(Z_1 + Z_2)^2$ (4, for $H_2^+$) and the independent atom model for close-collisions, where $S_c$ is proportional to $Z_1^2 + Z_2^2$ (2, for $H_2^+$).

Fig. 3 (on the bottom panel) shows similar results for a molecule projectile, whose first ion has a single-zeta screening with $\alpha = 2$ ($Z_{\text{eff}} = 1$). It is pointed out that the interference between the projectile components is notably reduced, since the characteristic screening length $1/\alpha$ is only one quarter of the molecule length. Then, the interaction between the bare ion and the target electron is much larger than the one from the screened partner. This explains the significantly reduced difference between molecular SCA and independent proton SCA calculations for screened projectiles. Qualitatively similar results were found for different values of $\phi$ and for the case of Bohr screening for $\alpha = 2$ (not shown).

In conclusion, we have developed a simple formula (Eqs. (11)–(13)) to evaluate the electronic energy loss as a function of impact-parameter for cluster projectiles, valid for high clusters energies and for a wide range of impact-parameters, including the effect of screening. The input parameters are only the target density and the oscillator strengths, as well as the projectile screening parameter for all cluster components. This model reproduces the results of full SCA calculations and is much less time consuming. Thus, the MPCA model is very adequate for use in computer channeling simulations.

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