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# Resonant effects in the stopping power of clusters

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**Abstract**

The stopping power of large $(H_2)_n$-clusters in silicon is analyzed as a function of the cluster size and the cluster velocity. A dielectric formalism is used to describe the electronic interaction between the projectile and the target. The intramolecular and intermolecular interference effects in the cluster energy loss are evaluated as a function of the cluster velocity. For high velocities and large clusters the intermolecular collective effects dominate in the cluster stopping power. For each cluster velocity we find a resonant cluster size for which the intermolecular stopping power clearly shows a maximum and it saturates to a constant value for larger clusters. The radius of this resonant cluster and the maximum stopping power are proportional to the cluster velocity.

The study of polyatomic ion beams interacting with matter has increased during the last years [1]. New sources have been developed to produce clusters of different materials and larger kinetic energies. Also, a growing effort has been devoted to understand the basic physical properties underlying the interaction of cluster beams with matter. This renewed interest in cluster research is partially due to the interesting collective effects that appear when correlated particles interact with matter [2,3] and its feasible application in different fields. For instance, the use of cluster ion beams to drive the implosion in an inertial confinement fusion device has been studied due to the high density of deposited energy [4–7]. There are several theoretical works on cluster stopping power in solid materials [3,7–13] and in plasmas [14–17], however only a few experimental results in this field are available [18–20].

In this paper we discuss the stopping power of large molecular hydrogen clusters, $(H_2)_n$, moving at high velocity in a silicon target. We are interested in the evaluation of collective effects in the energy loss due to the simultaneous perturbation of the medium by the correlated motion of the cluster ions. Our main aim is to analyze the dependence of the cluster stopping power with the size and the velocity of the cluster.

The dielectric formalism is used to describe the interaction between the cluster ions and the electrons of the target. We only consider electronic energy loss since at high cluster velocities this is the principal mechanism. The dynamic response of the target to an external perturbation is provided by its dielectric function $\epsilon(k, \omega)$, where $k$ and $\omega$ represent, respectively, the momentum and energy transferred to the electrons of the solid in an inelastic event.

We do not take into account the Coulomb explosion due to the repulsion between the ions that constitute the cluster; this assumption is valid for swift clusters and thin targets. We neglect non-linear effects, which may be important at low velocities, but certainly will be less significant in the high velocity range we plan to discuss. Note that atomic units will be used throughout this paper.

In this scheme, a general expression for the stopping power $S_{cl}$ of a randomly oriented cluster composed by $N$ particles penetrating with velocity $v$ in a target is given by [8,11]:

$$S_{cl} = NZ^{*2}\left[1 + \bar{n}\int d^3r\ g_{cl}(r) I(r)\right] S_p, \qquad (1)$$

where $Z^*$ is the effective charge of each individual particle in the cluster and $\bar{n}$ is the average nuclear density of the cluster. We assume that the equilibrium charge state is reached for each constituent of the cluster and this mean equilibrium charge state or effective charge $Z^*$ is equal for atomic and cluster ions.

The pair correlation function $g_{cl}(r)$ is related to the probability to find two particles at a given distance $r$ in the cluster and satisfies the normalization condition: $N = 1 + \bar{n}\int d^3r\ g_{cl}(r)$. In the dielectric formalism the proton stopping power $S_p$ is expressed as:

$$S_p = \frac{2}{\pi v^2}\int_0^\infty \frac{dk}{k}\int_0^{kv} d\omega\ \omega\ \mathrm{Im}\left[\frac{-1}{\epsilon(k,\omega)}\right]. \qquad (2)$$

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The interference – or vicinage – function $I(r)$ measures the collective effects that appear in the stopping power of correlated particles. Note that $I(r)$ represents an angular average over all cluster orientations and it is given by [8]:

$$I(r) = \frac{2}{\pi v^2 S_p} \int_0^\infty \frac{dk}{k} \frac{\sin kr}{kr} \int_0^{kv} d\omega\ \omega\ \mathrm{Im}\left[\frac{-1}{\epsilon(k,\omega)}\right]. \tag{3}$$

We consider large clusters of molecular hydrogen, $(H_2)_n$, then it is convenient to separate the interference effects due to two hydrogen atoms of the same molecule (called *intra*-molecular terms) from the interference effects produced by two hydrogen atoms from different molecules (called *inter*-molecular effects). Therefore the pair correlation function $g_{cl}(r)$, which contains information about the cluster structure, can be split into two contributions: $g_{cl}(r) = g_{intra}(r) + g_{inter}(r)$. The structure of the $H_2$ molecule is described by $g_{intra}(r)$ through a delta function at 1.40 a.u., the internuclear distance in the $H_2$ molecule [21]. The intermolecular pair correlation function $g_{inter}(r)$ is modeled by a random distribution of intermolecular distances in a spherical cluster, an assumption that is appropriate for large clusters. Due to the repulsive interaction of the $H_2$-molecules at close distances we consider an exclusion volume of radius $r_{ex}$ around each atom; $g_{inter}(r)$ also incorporates the finite size of the cluster. For a more detailed discussion we refer the interested reader to Ref. [12]. With this model for the cluster structure the number of atoms in the cluster, $N$, is related with the radius of the cluster, $R_{cl}$, by $N = \frac{4}{3}\pi \bar{n} R_{cl}^3$.

Now it is possible to separately evaluate the contributions of the intramolecular and intermolecular interference effects to the cluster energy loss. The total reduced cluster stopping power, $S_{cl}/(NZ^{*2})$, can be written as a sum of three different quantities,

$$\frac{S_{cl}}{NZ^{*2}} = S_p + S_{intra} + S_{inter}. \tag{4}$$

The proton stopping power $S_p$ represents the independent-particle term, $S_{intra}$ and $S_{inter}$ are due to the intra- and inter-molecular interference effects, respectively. All the calculations presented in this paper are given for an exclusion radius $r_{ex} = 3.59$ a.u., corresponding to half the nearest-neighbour distance in solid hydrogen and the value of the cluster density we have used is $\bar{n} = 6.27 \times 10^{-3}$ a.u. [21].

In order to evaluate the cluster stopping power using Eqs. (1)–(3) it is necessary to specify the target properties. The response of the electrons in the solid to an external perturbation is expressed in terms of the energy loss function (ELF) of the medium, $\mathrm{Im}(-1/\epsilon(k,\omega))$, a magnitude that is directly related to the probability that an inelastic event with momentum transfer $k$ and energy transfer $\omega$ will take place in the solid. The ELF of silicon measured at zero momentum transfer [24] shows a well defined peak at $\sim 17$ eV. We consider the target as a homogeneous and isotropic system and describe the dielectric properties of silicon by a Mermin [22] dielectric function $\epsilon_M(\omega, k)$. This dielectric function is a generalization of the Lindhard dielectric function [23] for a free electron gas, but it takes into account a finite plasmon lifetime and preserves the local particle number. The main parameters in $\epsilon_M$ are the plasmon energy $\omega_p$ and damping $\gamma$, which are related to the location and width of the peak in the ELF. Fitting [25] the Mermin ELF at $k = 0$ to experimental optical data for silicon taken from Ref. [24] we obtain $\omega_p = 0.62$ a.u. and $\gamma = 0.156$ a.u.

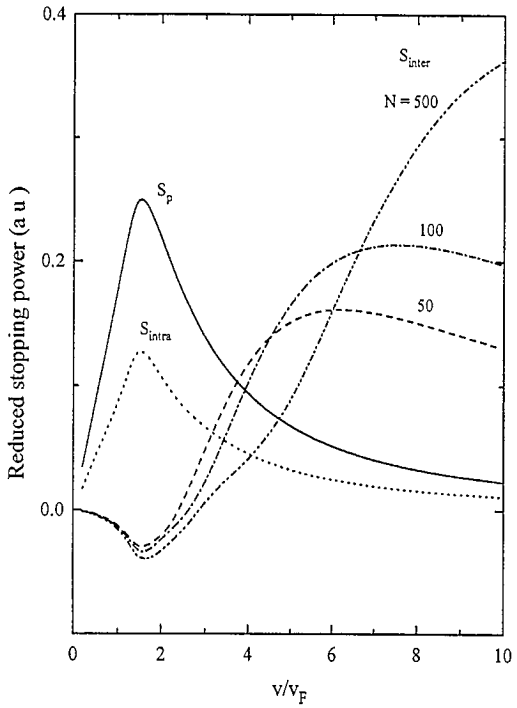


Fig. 1. Reduced cluster stopping power versus cluster velocity for $(H_2)_n \rightarrow$ Si. Solid line: proton stopping power, $S_p$. Dotted line: intramolecular stopping power, $S_{intra}$. Dashed line: intermolecular stopping power, $S_{inter}$, for $N = 50$ (– – –), 100 (–·–·–) and 500 (–···–···–) particles in the cluster.

In Fig. 1 we show the dependence of the different terms in the reduced cluster stopping power, Eq. (4), with the cluster velocity $v$ in units of the Fermi velocity, which is $v_F = 0.9675$ a.u. for silicon. The intramolecular stopping power $S_{intra}$ follows the same behaviour with $S_p$, being $S_{intra}/S_p \simeq 0.5$ in the whole range of velocities considered. The intermolecular energy loss $S_{inter}$ for a cluster with $N = 50$, 100 and 500 particles is also depicted in Fig. 1, and we can observe that it strongly depends on the cluster size. As a result of the coherent interference due to a large number of atoms a sizeable collective effect appears in $S_{inter}$ at high velocities in such a way that $S_{inter}$ can exceed in more than one order of magnitude the proton stopping power, $S_p$. The interference effects are bigger when the

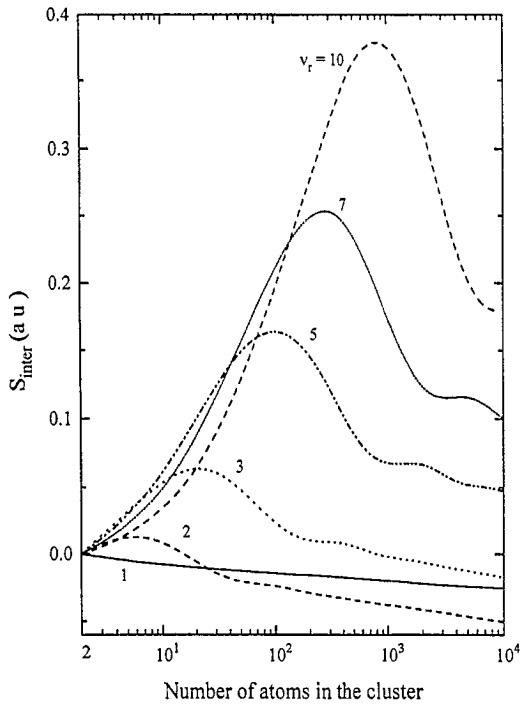


Fig. 2. Intermolecular stopping power $S_{inter}$ as a function of the cluster size for $(H_2)_n \rightarrow Si$. The number in the curves indicate the cluster velocity relative to the silicon Fermi velocity, $v_r = v/v_F$.

plasmon wavelength is similar to the cluster size, and the excitation of large wavelength plasmons is possible only for swift clusters [12]. It is interesting to note that the reduced intermolecular stopping power $S_{inter}$ clearly dominates the collective effects at high velocities, being stronger for larger clusters. The behaviour of $S_{intra}$ and $S_{inter}$ as a function of the cluster velocity for a silicon target shows general trends similar to the results previously obtained for a carbon target [11].

Subsequently we analyze, for a given velocity, the dependence of the cluster stopping power with the cluster size; $S_p$ and $S_{intra}$ are independent of the number of atoms in the cluster and remain constant for a fixed cluster velocity. Also, as we discussed in Fig. 1, the main collective effects for large clusters are given by the intermolecular energy loss $S_{inter}$, then we center our interest in the behaviour of $S_{inter}$ with the cluster size.

In Fig. 2 we present the reduced intermolecular stopping power $S_{inter}$ as a function of the number of atoms in the cluster, for the velocity range $v = (1-10)v_F$. For $v > v_F$ we find a resonant cluster size, characteristic of each velocity, for which $S_{inter}$ displays a maximum, after which the energy loss saturates to a constant value. This resonant cluster configuration is given when all the ions in the cluster produce constructive interference effects, then the integration of the vicinage function $I(r)$ over all the cluster particles (Eq. (1)) add only positive values to the stopping power. When the cluster size increases, destructive interference effects appear, which reduce the cluster stopping power. A saturation in the energy loss is reached when the interference effects produced by the particles in the cluster remain constant, even though more atoms belong to the cluster. This is so because in this case the distances between atoms in the cluster are so large that the new interference effects are negligible.

We remark that the strong enhancement in the interference effects in the intermolecular stopping power is for cluster sizes with an intermediate number of particles and not for the largest cluster. When the cluster velocity increases the resonant cluster size and their corresponding maximum stopping power increase as well as the saturation value of the energy loss.

For cluster velocities of the same order as the Fermi velocity a maximum in the intermolecular stopping power does not appear (see Fig. 2) and a roughly constant value of $S_{inter}$ is found independent of the number of atoms in the cluster. This saturation effect is produced because there is not enough energy to excite plasmons in the solid, which are responsible for the interference effects [12]. Analyzing the dispersion relation of silicon we observe that the plasmon line decays into individual excitation at a critical wave number $k_c = 0.703$ a.u., therefore the minimum velocity to excite plasmons will be $v_c = 1.3$ a.u. Only particles with velocities larger than $v_c$ can produce collective excitations in the target, and therefore will give rise to the interference effects in the cluster stopping power. Saturation effects with the cluster size have been found experimentally in the slowing down of low energy hydrogen clusters in carbon foils [18].

The resonant cluster configuration can be characterized by its radius, $R'_{cl}$, and by its maximum intermolecular stopping power, $S'_{inter}$. The dependence of these two magnitudes on the cluster velocity will be analyzed in what follows. In Fig. 3 we show the resonant cluster radius $R'_{cl}$, for which the interference effect in the cluster energy loss is maximum, as a function of the cluster velocity. As we can see, the critical cluster radius $R'_{cl}$ increases linearly with the velocity of the cluster. The maximum interference effects in $S_{inter}$ will be when the wavelengths of the collective plasmon excitations are similar to the cluster dimension, accordingly we get the scale rule $R'_{cl} = 2v/\omega_p$, where $\omega_p$ is the silicon plasmon energy. As we observe in Fig. 3, this relation perfectly agrees with the theoretical results obtained by our model for the critical cluster size. Then for any given cluster velocity, larger electron densities in the target will correspond to smaller number of particles in the resonant cluster.

The cluster velocity dependence of the maximum values in the intermolecular stopping power divided by the proton stopping power, $S'_{inter}/S_p$, is presented in Fig. 4. It can be seen that the calculated points closely follow the relation $S'_{inter}/S_p \simeq 0.8\bar{n}(v/\omega_p)^3$, which shows that $S'_{inter}$ goes linearly with the cluster velocity, since $S_p$ is proportional to $1/v^2$ at large velocities. Studying the energy loss

of clusters in classical plasmas, Bringa and Arista [17] found an identical behaviour for $R'_{cl}$ and a similar one for $S'_{inter}/S_p$ to those reported here.

A complete description of the cluster stopping power in the target foil would require the inclusion of the Coulomb explosion between the cluster constituents. However due to the different time scale of the electronic interaction between the cluster particles and the target, and the nuclear explosion, both processes can be evaluated separately.

We estimate the consequences of the Coulomb explosion in the dependence of the intermolecular cluster stopping power $S_{inter}$ with the cluster size by decreasing the density $\bar{n}$ of the cluster, which corresponds to an increase in the distance between the cluster components. We evaluate $S_{inter}$ for an expanded cluster with a density $\bar{n}_{exp}$ ranging from $\bar{n}$ to $0.3\bar{n}$; this corresponds to a cluster with a radius from $R_{cl}$ to $1.44R_{cl}$ and we obtain, for all the cluster velocities, a resonant cluster configuration with a similar behaviour to that shown in Fig. 2. According to the scaling rule for the resonant cluster radius $R'_{cl}$ and for the maximum value of the intermolecular cluster stopping power $S'_{inter}$, we find the following relations between the number of atoms $N'$ of the resonant cluster configuration and the maximum value of the intermolecular stopping power for the expanded cluster: $N'_{cl,exp} = (\bar{n}_{exp}/\bar{n})N'_{cl}$ and $S'_{inter,exp} = (\bar{n}_{exp}/\bar{n})S'_{inter}$, respectively. Then, the effect of the Coulomb explosion is to decrease the absolute value of the intermolecular cluster stopping power but maintaining the same behaviour with the cluster size, indicating the existence of a resonant cluster configuration.

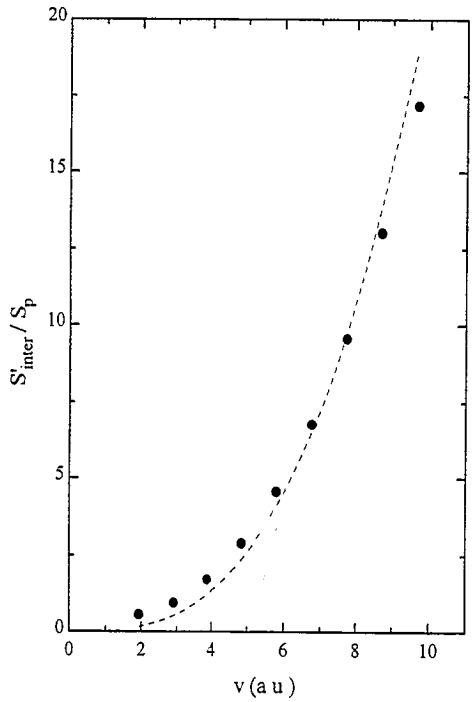


Fig. 4. Maximum reduced intermolecular stopping power divided by the proton stopping power, $S'_{inter}/S_p$, for the critical cluster size as a function of the cluster velocity. Dots represent the results obtained theoretically by our model. The dashed line represents the relation $S'_{inter}/S_p = 0.8\ \bar{n}\ (v/\omega_p)^3$.

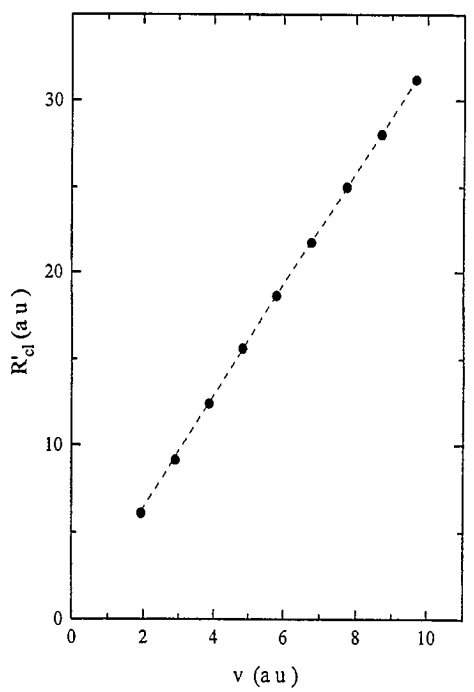


Fig. 3. Critical cluster radius $R'_{cl}$ as a function of the cluster velocity for $(H_2)_n \rightarrow$ Si. Dots represent the results obtained theoretically by our model. The dashed line represents the scale rule $R'_{cl} = 2v/\omega_p$.

In conclusion, in this work, we have evaluated the collective effects in the stopping power of large molecular hydrogen clusters moving at high velocities in a silicon target. The dependence with the cluster velocity and the cluster size were evaluated, and we found that for swift and large clusters the intermolecular interference effects dominate in the cluster stopping power. It is worth to mention that for each velocity there is a critical cluster configuration for which the energy loss of the cluster is maximum, and both the radius of this resonant cluster and the maximum stopping power are directly proportional to the cluster velocity. For cluster sizes bigger than the critical one the intermolecular stopping power goes to a saturation value. This kind of prediction could be applied to experimental situations, like inertial confinement fusion devices, where a maximum deposit of energy is desired.

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