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Electronic stopping of protons in xenon plasmas due to free and bound electrons

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Abstract

In this work, proton stopping due to free and bound electrons in a plasma target is analyzed. The stopping of free electrons is calculated using the dielectric formalism, well described in previous literature. In the case of bound electrons, Hartree-Fock methods and oscillator strength functions are used. Differences between both stopping, due to free and bound electrons, are shown in noble gases. Then, enhanced plasma stopping can be easily estimated from target ionization. Finally, we compare our calculations with an experiment in xenon plasmas finding a close agreement.

Keywords: Bound electrons; Energy loss; Free electrons; Xenon plasmas

INTRODUCTION

Electronic stopping due to free electrons can be analyzed through dielectric formalism. In dielectric formalism, random phase approximation (RPA) has been used extensively, which consist of considering the effect of the incident particle as a perturbation, so that the energy loss was proportional to the square of its charge. Then slowing-down was simplified to a treatment of the properties of the medium only, and a linear description of these properties may then be applied.

But in this work we will study all kinds of plasmas, even coupled plasmas where target electron interactions have to be kept in mind. RPA does not consider these electron interactions, whereas it is well-known that in real materials these interactions exist. Mermin (1970) derived an expression for the dielectric function caring for the plasma electron collisions but only preserving the local particle density. Mermin dielectric function has been successfully applied to solids (dense degenerate electron gas) (Barriga-Carrasco *et al.*, 2004), for classical plasmas (nondegenerate electron gas) (Selchow *et al.*, 1999; Gerike, 2002; Barriga-Carrasco *et al.*, 2006) and also for partially degenerate plasmas (Barriga-Carrasco, 2007).

Recently, we have been able to obtain a dielectric function that includes all conservation laws (density, momentum, and energy) when we take into account plasma electron-electron collisions for plasmas at any degeneracy (Barriga-Carrasco 2010; 2011). This full conserving dielectric function (FCDF) reproduces former RPA and Mermin ones, for not collisions and for collisions only considering electronic density conservation, respectively. Differences are as maximum around 5% between FCDF values and the Mermin ones, and only around 2% between the FCDF ones and RPA ones for plasmas with high enough collision frequency. It is not surprising that as we include more conservation laws the behavior of the dielectric functions yields back the RPA, a model with every conservation laws enforced. The meaning of the fact that FCDF results are similar to the RPA ones, a dielectric function that does not consider electron-electron collisions, is that latter collisions are not important for energy loss calculations. Whether from previous investigations it was inferred the opposite, this was because electron collisions were usually taken into account through a Mermin dielectric function which does not consider momentum and energy conservation. Then we will use the RPA dielectric function to calculate electronic stopping of free electrons in this work.

But, on the other hand, we must also consider stopping power due to electron bound to the target plasma atoms. This study can be performed using the mean excitation energy, I, that appears in the renowned expression of the Bethe logarithm

$$-\frac{dE}{dx} = \frac{4\pi Z^2 e^4}{m_e v^2} n_e \ln \frac{2m_e v^2}{I}. \quad (1)$$

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Mean excitation energies can be determined through Hartree-Fock method or through oscillator strength.

Only for hydrogen atom or hydrogen-like atoms, there are analytical solutions of Schrödinger equation. However, there are no exact solutions for atoms with more than one electron, for this reason, we must use other approximating methods to estimate the atomic properties of many electrons atoms. The main idea of the Hartree-Fock method is to reduce the many-electron problem to one-electron problem. It is called the *independent particle model*, according to which each electron moves in an effective potential that takes into account the attraction of the nucleus and the average effect of the repulsive interactions due to the other electrons. The whole wave-function of the atom is antisymmetric because electron is a fermion that obeys the Pauli exclusion principle. This antisymmetric requirement is the generalization of Hartree's theory known as Hartree-Fock method (Bransden & Joachain, 1983; Haken & Wolf, 2005; Frank *et al.*, 2010).

Alternatively, I can also be considered through atomic oscillator strength. A fast charge passing through an atomic system has an effect like an electromagnetic radiation including the excitation and ionization of atoms and ions. (Dalgarno, 1960; Fano & Cooper, 1968). Within this framework, I, is calculated for every subshell of noble gases (Bell & Dalgarno, 1966; Bell *et al.*, 1972) and for all of the elements from $Z = 1$ to $Z = 36$ (Meltzer *et al.*, 1990). Some authors have studied by means of generalized oscillator strength, proton stopping in aluminum and nickel ions, and in argon, krypton and xenon atoms (McGuire *et al.*, 1982; McGuire 1983; 1991).

Mean excitation energy could also be estimated using the local plasma approximation (Lindhard & Scharff, 1953). The local plasma approximation consists of averaging over density of the inhomogeneous fluid of bound electron around a target ion (Garbet *et al.*, 1987). Then I could be determined using

$$\ln I = \int \ln\left[\gamma\hbar\,\omega_p(r)\,\rho_b(r)\,dr\right], \tag{2}$$

with $\omega_p^2(r) = 4\pi\rho_b(r)e^2/m_e$, $\gamma = \sqrt{2}$ and $\rho_b(r)$ is the bound electron density. A simple analytic formula for I, was proposed through a variational method (Garbet *et al.*, 1987).

$$I = \sqrt{\frac{2K}{\langle r^2\rangle}}. \tag{3}$$

This equation is obtained in the next section.

It is easy to calculate mean ionization energies for noble gases owing to their monatomic nature. Then it allows studying electronic stopping for any degree of ionization from cold gas to plasma state without difficulty. In case of cold gas, only bound electrons contribute to electronic stopping, while in plasma case, stopping of free electrons has to be taken into account. In the results section, we estimate this stopping for all noble gases and we study specifically the case of xenon. We will use atomic units (a.u.), $e = \hbar = m_e = 1$, to simplify formulas.

ELECTRONIC STOPPING DUE TO FREE ELECTRONS

RPA dielectric function is developed in terms of the wave number k and of the frequency ω provided by a consistent quantum mechanical analysis. The RPA analysis yields to the expression (Lindhard, 1954)

$$\varepsilon_{\mathrm{RPA}}(k,\,\omega) = 1 + \frac{1}{\pi^2 k^2}\int d^3k' \frac{f(\vec{k}+\vec{k}') - f(\vec{k}')}{\omega + i\upsilon - (E_{\vec{k}+\vec{k}'} - E_{\vec{k}'})}, \tag{4}$$

where $E_{\vec{k}} = k^2/2$. The temperature dependence is included through the Fermi-Dirac function

$$f(\vec{k}) = \frac{1}{1 + \exp\left[\beta(E_k - \mu)\right]}, \tag{5}$$

being $\beta = 1/k_B T$ and μ the chemical potential of the plasma with electron density n_e and temperature T. In this part of the analysis, we assume the absence of collisions so that the collision frequency tends to zero, $\upsilon \to 0$.

Analytic RPA dielectric function for plasmas at any degeneracy can be obtained directly from Eq. (4) (Gouedard & Deutsch, 1978; Arista & Brandt, 1984)

$$\varepsilon_{\mathrm{RPA}}(k,\,\omega) = 1 + \frac{1}{4z^3\pi\,k_F}[g(u+z) - g(u-z)], \tag{6}$$

where $g(x)$ corresponds to

$$g(x) = \int_0^\infty \frac{y dy}{\exp(Dy^2 - \beta\mu) + 1} \ln\left(\frac{x+y}{x-y}\right),$$

$u = \omega/kv_{\mathrm{F}}$ and $z = k/2k_F$ are the common dimensionless variables (Lindhard, 1954). $D = E_{\mathrm{F}}\beta$ is the degeneracy parameter and $v_F = k_F = \sqrt{2E_F}$ is Fermi velocity in a.u.

Finally, electronic stopping of free plasma electrons will be calculated in the dielectric formalism as

$$Sp_f(v) = \frac{2Z^2}{\pi v^2}\int_0^\infty \frac{\mathrm{d}k}{k}\int_0^{kv} \mathrm{d}\omega\,\omega\,\mathrm{Im}\left[\frac{-1}{\varepsilon_{\mathrm{RPA}}(k,\,\omega)}\right]\text{(a. u.)},$$

where Z is the charge and v is the velocity of the projectile.

ELECTRONIC STOPPING DUE TO BOUND ELECTRONS

In order to determine electronic stopping due to bound electrons, we use analytical formulas in the limit of low and high projectile velocities and, an interpolating expression is derived for intermediate velocities. For a plasma target with atomic density n_{at}, bound electron density for each populated atomic shell is $n_i = P_i n_{\mathrm{at}}$, where P_i is the average electron

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population in the shell of a target atom (Barriga-Carrasco & Maynard, 2005). We can estimate electronic stopping for a proton beam in the form

$$S_p = \frac{4\pi n_{\text{at}}}{v^2} L_b, \tag{7}$$

the stopping number L_b being defined as

$$L_b = \sum_i P_i L_i, \tag{8}$$

where L_b is the stopping number for whole bound electrons of atom or ion and L_i is the stopping number for bound electrons of each shell.

We reckoned L_b by interpolating between the asymptotic formulas valid either for low or for high projectile velocities (Maynard & Deutsch, 1985)

$$L_b(v) = \begin{cases} L_H(v) = \ln\dfrac{2v^2}{I} - \dfrac{2K}{v^2} & \text{for} v > v_{\text{int}} \\ L_B(v) = \dfrac{\alpha v^3}{1+Gv^2} & \text{for} v \le v_{\text{int}} \end{cases}, \tag{9}$$

$$v_{int} = \sqrt{3K + 1.5I}, \tag{10}$$

where G is given by $L_H(v_{\text{int}}) = L_B(v_{\text{int}})$, K is the electron kinetic energy, I is the excitation mean energy, and α is the friction coefficient for low velocities. Eq. (3) is used to determine the mean excitation energy of each shell (Garbet *et al.*, 1987)

$$I = \sqrt{2K / \langle r^2 \rangle},$$

where $\langle r^2 \rangle$ is the average of the square of the radius, for the electron in the i shell. Within the hydrogenic approximation, the friction coefficient of each shell is given by $\alpha = 1.067\sqrt{K}/I$ (Garbet *et al.*, 1987).

Using this approximation, we can easily estimate I from the atomic parameters K and $\langle r^2 \rangle$. These late quantities are been determined by two methods: (1) Hartree-Fock calculations (Fischer, 1987) and (2) oscillator strength sums (Bell *et al.*, 1972).

Hartree Fock Method

The Hartree-Fock equations for an electron i with coordinate $\boldsymbol{R}_i$ is:

$$\left[-\frac{1}{2}\nabla_i^2 - \frac{Z}{\boldsymbol{r_i}} + V_i(\boldsymbol{r}_i)\right]\psi_{Q_i}(\boldsymbol{R}_i) - \sum_{\substack{j \\ j\neq i}} \int \frac{\psi^*_{Q_j}(\boldsymbol{R}_j)\,\psi_{Q_i}(\boldsymbol{R}_j)}{|\boldsymbol{r_i} - \boldsymbol{r_j}|} d\tau_j \cdot \psi_{Q_j}(\boldsymbol{R}_i) = E\psi_{Q_i}(\boldsymbol{R}_i), \tag{11}$$

where the first term in the bracket is the kinetic energy, the second the potential energy due to the nucleus, the third the Coulomb interaction energy with all the other electrons and the last term of the first member is called the *exchange term*, which includes the antisymmetry of the wave-function.

Furthermore, the wave-function of the one electron, could be expressed with all its quantum numbers, including spin (Fischer *et al.*, 1997)

$$\psi_{Q_j}(\boldsymbol{R}_j) = R_{nl}(\boldsymbol{r}_j)\, Y_{lm_l}(\theta,\ \varphi)\, \chi_{m_s}(\sigma_j). \tag{12}$$

It is necessary to obtain kinetic energy and mean square radius from this wave-function, in order to solve Eq. (3). We estimate K and $\langle r^2 \rangle$ through the integration of the following radial functions

$$K_{nl} = -\frac{1}{2}\int_0^\infty P_{nl}(r)\left[\frac{d^2}{dr^2} - \frac{l(l+1)}{r_2}\right] P_{nl}(r)\, dr, \tag{13}$$

where

$$R_{nl}(r) = \frac{1}{r} P_{nl}(r), \tag{14}$$

satisfying normalization condition (Fischer *et al.*, 1997)

$$\int_0^\infty P^2_{nl}(r)\, dr = 1. \tag{15}$$

The general formula to obtain mean powers of radius is (Bransden & Joachain, 1983)

$$\langle r^p \rangle_{nlm} = \int_0^\infty |R_{nl}(r)|^2\, r^{p+2} dr, \tag{16}$$

substituting 2 instead of p in this particular case. These equations are solved for any atom at the ground state using a Fortran 95 code (Fischer & Tachiev, 2009), including K and $\langle r^2 \rangle$.

Oscillator Strength Sum Rules

Several important atomic properties like polarizability, mean excitation energy, stopping power, and straggling are determined from a useful, dimensionless quantity, the oscillator strength (Dehmer *et al.*, 1975). It is defined by

$$f_{0n} = \frac{2E_{0n}}{N} \left\langle \left| \sum_{i=1}^{N} Z_i \right| 0 \right\rangle, \tag{17}$$

where the excitation energy is E_{0n}, calculated for transitions $\{0\} \rightarrow \{n\}$ in a given atom or ion with N bound electrons, and the nuclear charge is Z (Garbet *et al.*, 1987).

Oscillator strengths satisfy the following important identity, known as the Thomas-Reiche-Khun sum rule (Bransden

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& Joachain, 1983)

$$\sum_n f_{0n} = N \tag{18}$$

where N is the total number of atomic electrons.

The momenta $S(\mu)$ and $L(\mu)$ are calculated using the oscillator strength sum rules (Fano & Cooper, 1968)

$$S(\mu) = \sum_n f_{0n} E_{0n}^{\mu}, \tag{19}$$

$$L(\mu) = \sum_n f_{0n} E_{0n}^{\mu} \ln |E_{0n}|. \tag{20}$$

The physical meaning of this sums are given by μ. For example, when $\mu = 0$ Eq. (19) was simplified into Eq. (18)

$$S(0) = \sum_n f_{0n} E_{0n}^{0} = \sum_n f_{0n} = N. \tag{21}$$

When $\mu = -1$

$$S(-1) = \frac{2m_e}{3\hbar^2} a_0^2 \langle \left(\frac{r}{a_0}\right)^2 \rangle, \tag{22}$$

then $S(-1)$ is proportional to square radius. If atomic units are used it is reduced to the expression

$$\langle r^2 \rangle = \frac{3}{2} S(-1). \tag{23}$$

Substituting $\mu = 1$ in Eq. (19), an expression proportional to the kinetic energy is obtained

$$S(1) = \frac{4}{3} \langle 0 \left| \frac{p^2}{2m_e} \right| 0 \rangle, \tag{24}$$

using atomic units

$$2K = \frac{3}{2} S(1). \tag{25}$$

The quantity $L(0)$ is related with the mean excitation energy, I

$$\ln I = \frac{L(0)}{S(0)} = \frac{L(0)}{N}, \tag{26}$$

moreover, ln$I$, $S(0)$, $S(1)$, and $S(-1)$ are related by (Garbet *et al.*, 1987)

$$\ln I = \frac{1}{2} \ln \left[\frac{S(1)}{S(-1)} \right]. \tag{27}$$

Substituting Eq. (23) and Eq. (25) into Eq. (27) Eq. (3) is obtained.

The parameters $S(-1)$ and $S(1)$ have been obtained for all shells of noble gases (Bell *et al.*, 1972). In Table 1, the quantities K, $<r^2>$ and I are listed for all shells of xenon using Hartree-Fock and oscillator strength methods. Both methods show how kinetic and ionization energies decrease when the main quantum number n rises. For n constant, kinetic energies increase when secondary quantum number l ($s = 0$, $p = 1$, $d = 2\ldots$) increases.

In the next section, we will show the differences between electronic stopping of free and bound electrons for all noble gas plasmas, afterward we will estimate the enhanced plasma stopping in the case of xenon. Finally, using results in Table 1, we will compare our calculated electronic stopping of protons in xenon plasmas with experimental data.

RESULTS

Electronic Stopping of Noble Gases Plasmas

To study influence of the number of bound electrons in electronic stopping, we compare plasmas with the same temperature, 10 eV, and ionization, $Q = 1$. For this reason, we choose the same atomic density, n_{at}, as electron density, n_e, both equal to 2×10^{20} e$^-$/cm^3. With this ionization, we can contrast the stopping with noble gases, starting with only one bound electron, $N = 1$, the case of helium, and finishing with $N = 85$, the case of radon. Then, we can estimate how stopping increases due to bound electron when atomic number does, remaining free electron density constant.

Figure 1 show free and bound electron stopping. The stopping of free electrons is the same for all plasmas, because they have the same ionization, $Q = 1$ and electron density, $n_e = 2 \times 10^{20}$ e$^-$/cm^3. As we see, total stopping for low velocities, is mainly due to free electrons, while for high velocities, main contribution is provided by bound electrons. Only for helium, the stopping of bound electrons is below the stopping of free for any velocity. Furthermore, stopping of bound electrons increases when the number of bound electron does. In the case of radon, the maximum stopping for free electrons is very close to the maximum for bound. It means how dominant the stopping of free electrons is, because Rn^+ has 85 bound electrons and only one free.

Enhanced Plasma Stopping

As it is shown in the last section, electronic stopping in plasmas is mainly due to free electrons. Then, when a monatomic gas ionizes into plasma state, increases its electronic stopping (Deutsch *et al.*, 2010). This effect, named enhanced plasma stopping (EPS), cannot be neglected even when a particle beam heat a material upon plasma state.

Figure 2 shows how electronic stopping of xenon hugely increases from cold gas ($N = 54$) to plasma state ($N = 52$ and $Q = 2$). For both cases, cold gas and plasma, we choose the same atomic density, $n_{at} = 1.45 \times 10^{19}$ at/cm^3 and xenon plasma ionization equal to 2 in order to compare with a realistic experimental case (Mintsev *et al.*, 1999). We can see the EPS along all velocities and a maximum at $v = 1$

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Table 1. *Quantities* K, $<r^2>$ *and* I *are listed for every shell of xenon using HF and oscillator strength methods*

XENON	Hartree-Fock (a.u.)			Oscillator Strength (a.u.)		
Shell	*K*	$<r^2>$	*I*	*K*	$<r^2>$	*I*
1s	2859.046	0.001	2391.253	2196.750	0.000	4353.704
2s	605.624	0.017	266.927	533.813	0.001	894.899
2p	1814.592	0.013	528.364	898.163	0.005	580.297
3s	182.406	0.116	56.080	163.433	0.005	247.295
3p	532.038	0.111	97.910	349.354	0.026	164.431
4s	51.220	0.626	12.792	43.466	0.019	68.053
3d	839.280	0.926	42.576	514.238	0.141	85.279
4p	140.070	0.685	20.223	89.618	0.066	52.285
5s	8.114	4.440	1.912	6.012	0.036	18.294
4d	182.170	0.881	20.336	116.651	1.417	12.832
5p	17.580	6.277	2.367	12.291	7.119	1.858

a.u. By $v = 10$ a.u., both, gas and plasma, stoppings are similar. Then, EPS becomes important only at low projectile velocities ($v < 4$ a.u.), since the stopping of free electrons is not so dominant at higher velocities.

Calculations and Experimental Data for Xenon Plasma

They are only few experiments with proton beams through fully characterized, stables and uniform plasmas. We found an experimental case with these characteristics (Mintsev *et al.*, 1999). In this experiment, the authors measure the stopping power of xenon plasmas for a 3 MeV proton beam, $v \approx 11$ a.u. They use two different methods in order to obtain energy loss of the proton beam: time of flight (TF) method and displacement of the position of the spot image (SI) method. The plasma was generated by a shock wave created by explosives. Due to the shock wave, xenon was heated and ionized. Its properties were measured by spectroscopic techniques. Plasma parameters and proton energy loses are placed in Table 2. Cases 2 and 3 repeat conditions of temperature, density and ionization for TF and SI measurement methods. In case 3, authors obtain different stopping for both measurement methods.

Plasma thickness is 10 mm, then it is easy to estimate electronic stopping:

$$Sp\,(a.u.) = \frac{\Delta E(a.u.)}{\Delta x(a.u.)}. \tag{28}$$

Figure 3 show a close agreement between calculated and experimental stopping. Their theoretical model (Basko *et al.*, 1984) is analogous to our oscillator strength method confirming our results. Only for cases 4 and 5 from Table 2, there are significant deviations where experimental stopping is lower than calculated. Mintsev *et al.* (1999) alleged that it is because of non-ideality of the plasma at high densities. We affirm that it could be because at the same time xenon plasma density increases, proton deposited energy also does, then plasma temperature raises and so proton stopping diminishes. Anyway, values using oscillator strength method are lower than values using Hartree-Fock

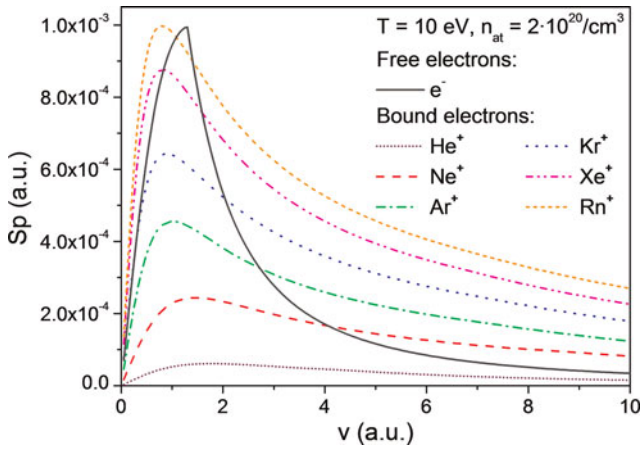

Fig. 1. Electronic stopping as a function of proton velocity. Solid line: stopping of free electrons. Dashed lines: stopping of bound electrons for each noble gas.

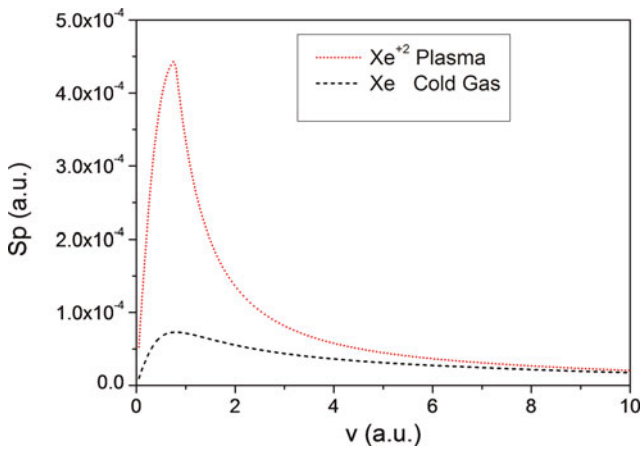

Fig. 2. Electronic stopping as a function of proton velocity for xenon cold gas and plasma.

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Table 2. *Experimental parameters of xenon plasma. There are four experimental measures for TF method and three for SI method*

Cases	ρ (10^{-3} g/cm^{-3})	T (eV)	n_e (10^{19} cm^{-3})	n_e/n_o	ΔE_{exp} (keV)	ΔE_{exp} (a.u.)	Exp. Method
1	1.32	3.42	1.3	2.16	55	2021	TF
2	3.17	3.66	3.1	2.12	90	3308	TF
					90	3308	SI
3	6.15	3.86	5.9	2.08	185	6799	TF
					150	5513	SI
4	9.06	3.98	8.5	2.05	200	7350	TF
5	11.93	4.08	11.9	2.03	260	9555	SI

one, being closer to experimental data. Then, as oscillator strength method is similar to their theoretical results and closer to their experimental data, we can conclude that the oscillator strength technique is a bit more realistic concerning energy loss calculations.

CONCLUSIONS

In this work, proton stopping in plasmas caused by the stopping of free and bound electrons was analyzed. It is shown that the increase of bound electron stopping with atomic number Z in plasmas of noble gas. Free electron stopping has been shown to be more relevant than bound electron one at low velocities with low or medium Z number. But for high proton velocities bound electron stopping has to be taken into account.

Related with the previous matter, EPS has been calculated for xenon. It has been observed an important difference between the stopping in cold gas and plasma state in chosen proton velocity range. The EPS has been estimated to be more important at proton velocities lower than 4 a.u. This is because, in this range, the stopping due to plasma free electrons is higher than the one due to bound electrons in cold gas. However, at high velocities, stopping differences between cold and hot matter become negligible, since free and bound stopping are similar.

Finally, we have compared our calculations with experimental data. Proton stopping was measured in xenon plasmas at high velocities, by $v = 11$ a.u., resulting that bound electrons were responsible of main energy loss. We have found an excellent agreement between our simulation and real data.

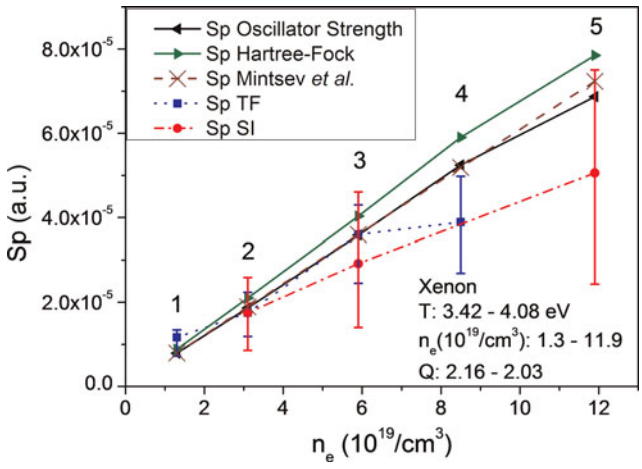

Fig. 3. Proton stopping as a function of plasma electronic density. Experimental: square, Time of Flight, and circle, Spot Image. Calculated: left triangles, Oscillator Strength, right triangles, Hartree-Fock, and cross, Mintsev *et al.* model.

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