# On the energy-loss straggling of protons in elemental solids: the importance of electron bunching

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**Abstract**

In this work we investigate the energy-loss straggling of low and medium energy protons on available simple materials (C, Al, Si, Ni, Cu, Zn, Ge, Se, Pd, Ag, Sb, Pt, Au, Pb) and demonstrate that the well-known Yang-O'Connor-Wang empirical formula for the energy-loss straggling of protons in matter has a clear physical interpretation. This formula predicts, on empirical grounds, energy-loss straggling values for protons that exceed the values of traditional straggling formulas (e.g., by Lindhard and Chu) at energies around 0.3 MeV/uma by a factor of 2-3. This excess is correlated to the bunching effect, which comes from a non-homogeneous feature of the electron density. Although this effect is specific to each element and can be calculated using e.g. the CasP and PASS programs, a single formula correlates better with overall experimental values. The Yang-O'Connor-Wang formula was refined accounting for subsequent experimental data.

*Keywords:* energy-loss straggling, bunching effect, ion beam analysis, stopping power

## 1. Introduction

The energy-loss straggling is a basic quantity, which quantifies the energy loss fluctuations of ions slowing down into matter [1, 2]. Typically the variance $\sigma^2$ and mean energy-loss $\langle \Delta E \rangle$ are proportional to the ion path length and therefore the effect of energy-loss fluctuations $\sigma/\langle \Delta E \rangle$ is more important for short traveled distances and low-medium projectile energies. For proton energies larger than typically 10 keV the energy-loss straggling due to the interaction with target electrons dominates. This is the so-called *electronic straggling* and is well described at high projectile energies by the Bohr formula [3]. The electronic energy-loss straggling affects the depth resolution in ion-beam analysis techniques such as in Rutherford Backscattering Spectrometry (RBS) [4], Nuclear Reaction Analysis (NRA) [5] and Medium Energy Ion Scattering (MEIS) [6]. Typically the electronic straggling increases with increasing ion velocity and saturates at the value given by the Bohr formula. Different formulas, mainly based on free electron gas model, were proposed [7–10] but their accuracy for medium-energy ions is generally not good enough to describe energy-loss spectra provided by high-resolution ion beam experiments.

In many cases experimental groups had to use empirical values of the electronic straggling obtained from a best fit of the experimental results as provided by the Yang-O'Connor-Wang (YOW) formula [9]. These empirical values can exceed by a large amount the results given by standard theoretical formulas and the corresponding underlying physics has only been marginally discussed. Here we demonstrate the excess of energy-loss straggling predicted by the empirical YOW formula for protons arises from the spatial distribution of the electrons in each atom, the so-called bunching effect [10–12]. This effect can be understood as an intrinsic target roughness and can be obtained from the impact-parameter-dependent stopping power [11] as reviewed in what follows.

## 2. Theoretical procedure

The electronic energy-loss straggling has been investigated for many years and its main formulas and aspects can be found in an extensive literature [1, 2]. Here we summarize its main properties. The energy-loss straggling is defined as the variance of the energy loss distribution of a monoenergetic beam passing through a thin target.

From the theoretical point of view, the cause of energy loss fluctuations for gases are the variation of the number of collisions encountered over the distance $\delta x$ traveled by the ion, and the variation in impact parameters $b$ in each collision. Moreover, even in given collision with a given impact parameter $b$ the energy loss has a distribution mostly due to the quantum nature of the collision. Let $T$ the stochastic energy loss and $\overline{T}(b)$ and $\overline{\Delta T^2}(b)$ the corresponding mean and variance values of this distribution for each collisional impact parameter $b$. By assuming statistical independence of collision events, the variance $\delta\Omega^2$ of the energy distribution is then given by [1, 11, 13]

$$\delta\Omega^2 = N\delta x \int_0^\infty 2\pi b db \left(\overline{\Delta T^2}(b) + \overline{T}^2(b)\right) \tag{1}$$

$$= N\delta x \int_0^\infty 2\pi b db \overline{T^2}(b) \tag{2}$$

where $N$ is the target density in atoms per cm$^3$. The last term in Eq. (1) arises after assuming Poisson statistics to describe the fluctuations in the number of ion-target collisions.

Eq. (2) is quite general and can also be used for condensed matter after some adjustments [1, 11]. Particularly, for a homogeneous electron system the impact parameter $b$ will be relative to the electron and the integral in Eq. (2) is better written in terms of the differential cross-section $\frac{d\sigma}{dT}$ or Differential Inverse Inelastic Mean Free Path (DIIMFP) $N\frac{d\sigma}{dT}$ as

$$\delta\Omega^2 = \delta x \int dT T^2 N \frac{d\sigma}{dT}. \tag{3}$$

This formula has been specially employed for systems described in terms of the dielectric function formalism as the Lindhard and Scharff [7] and Chu [8] models. They treat solids as an ensemble of free electron gases (FEG). The Chu model was parameterized in [9] (called here Chu formula) and is used as a reference in what follows. At high projectile energies all straggling formulas converge towards the Bohr straggling [3] (in atomic units)

$$\delta\Omega^2_{\text{Bohr}} = N\delta x \, 4\pi Z_1^2 Z_2, \tag{4}$$

which is a constant for each projectile-target combination, described by atomic numbers $Z_1$ and $Z_2$, respectively and has been used as a normalization value.

The energy-loss straggling is sensitive to different kind of physical correlations [14], particularly spatial correlations, even in the framework of the independent electron model [1]. According to this model the total energy loss is the sum of energy losses due to each electron $T_i(b)$, independent from the other *passive* electrons. Therefore, using $T(b) = \sum_i T_i(b)$ in Eq. (2) we get

$$\delta\Omega^2 = N\delta x \int_0^\infty 2\pi b db \left( \sum_i \overline{T_i^2}(b) + \sum_{i \neq j} \overline{T_i}(b)\, \overline{T_j}(b) \right). \tag{5}$$

The first term is called *uncorrelated straggling* and was used in the derivation of well-known straggling formulas (Bohr, Lindhard, Chu, Bethe-Livingston, etc [1, 2]). The second term gives rise to *bunching effects* when electron $i$ and j belong to the same atom or *packing effects* when electron $i$ and j belong to the different atoms [2].

### 2.1. Bunching effect

According to the convolution approximation implemented by the CasP code [15] and Binary theory (realized by the PASS code) [2, 16, 17] the individual electron density distribution does not affect directly the stopping cross-section as well as the uncorrelated straggling cross-section (as given by Eq. (3)). However, it does affect the bunching effect for the energy-loss straggling. Indeed, the energy-loss straggling for an inhomogeneous electron system, such as an atom with different shells is not the same as for a homogeneous system with the same average density. The genesis of the effect went back to many years ago when the straggling of molecular gases was investigated [18]. The first calculations for light ions were reported by Besenbacher *et al.* [10], however, as observed later in Ref. [19], the results were not quite correct. The bunching contribution is typically important for projectile energies close to the maximum of the stopping power and has been investigated only in a few systems [2, 11–13, 19, 20] so far. The bunching energy-loss straggling ($\delta\Omega^2_{bunch}$) is obtained from the second term of Eq. (5) and reads

$$\begin{aligned}\delta\Omega^2_{bunch} &= N\delta x\left[\int_0^\infty 2\pi b db\left(\sum_i \overline{T_i}(b)\right)^2\right.\\ &\left.- \sum_i\int_0^\infty 2\pi b db\ \overline{T_i}^2(b)\right].\end{aligned} \tag{6}$$

In what follows, the energy-loss straggling due to the bunching effect ($\delta\Omega^2_{bunch}$) was then calculated directly from Eq. (6) using the shellwise mean energy loss $T_i(b)$ per electron obtained from the CasP program [21]. We did not calculate packing effects since these are typically much smaller than bunching effects [20]. It is noted that bunching effects calculated from the PASS code are usually larger than CasP predictions as observed in Ref. [11]

## 3. Results and Discussion

### *3.1. Experimental straggling excess and bunching calculations*

In the Fig. 1, we present the experimental data for each element: Carbon [9, 22, 23], Aluminum [9, 23–25], Silicon [22], Nickel [9, 23, 25], Copper [9], Zinc [24], Germanium [9], Selenium [9], Palladium [26], Silver [9], Antimony [9], Platinum [12], Gold [9, 23–25] and Lead [9]. All data (excluding those collected directly from the Ref. [9]) were subtracted from the value of Chu's straggling [8, 9] and divided by Bohr's straggling [3]. In this way, it was produced a plot of energy-loss straggling in excess of the Chu formula for each element as shown in Fig. 1. In each graph, it was plotted the empirical formula obtained by Yang, O'Connor and Wang [9] (dash-dot black line) and the blue line (DRF(updated)) will be explained in the following paragraphs. The impact-parameter-dependent energy loss used here to calculate the bunching effect (dash red line, calculated by CasP - CasP-Bunching) in the energy-loss straggling of protons in different elemental solids was determined from the CasP program [21], version 6.0. The following options were used (most of them are default options): screening none, UCA mode, shell corrections, Barkas on and gas target off (muffin-tin on).

In some cases, the CasP-Bunching curve presented a second maximum as predicted by Sigmund [2]. As can be observed in Fig. 1, CasP calculations present a single mode for the bunching effect as assumed in our fit formula Eq. (7) for some elements while bimodal or multimodal structures are also visible. This can be a consequence of the shell structure of each atom, which encompasses shells with different binding energies, mean radius and velocities. Additional CasP calculations were performed to better clarify the origin of these multimodal structures and in fact they disappeared as the shell corrections were turned off and became much more evident by using a simplified model, containing target electron velocities only along the ion velocity.

Fig. 2 presents almost all experimental data of energy-loss straggling for protons on single elemental solid targets up to date. It shows the difference between the reported experimental values and the corresponding predictions of the Chu formula [9], normalized by the Bohr straggling (Eq. (4)). This excess was observed by Yang, O'Connor and Wang in 1991 [9] and fitted using a deformed resonance curve (DRF):

$$\frac{\delta\Omega^2}{\delta\Omega^2_{\text{Bohr}}} = \frac{B_1\Gamma}{(E-B_2)^2+\Gamma^2} \tag{7}$$

with

$$\Gamma = B_3\left(1-\exp\left(-B_4E\right)\right). \tag{8}$$

We updated this procedure by including more recent measurements [12, 22–26] and obtained the new parameters for Eq. (7) and (8) as shown in Table 1. The data without thickness specifications and/or with only one measurement per element were not taken into account. It is pointed out that Kido and Koshikawa [27] have measured the energy-loss straggling for $H^+$ ions in Cu, Ag and Pt, but their results are too small and at variance with all other experimental data and, consequently, the results are not included in the figure presented here. The new fit curve (DRF(updated)) is higher than the DRF(YOW) original formula: 7% in the maximum point and it exceeds the previous formula by a factor of two to three at higher energies.

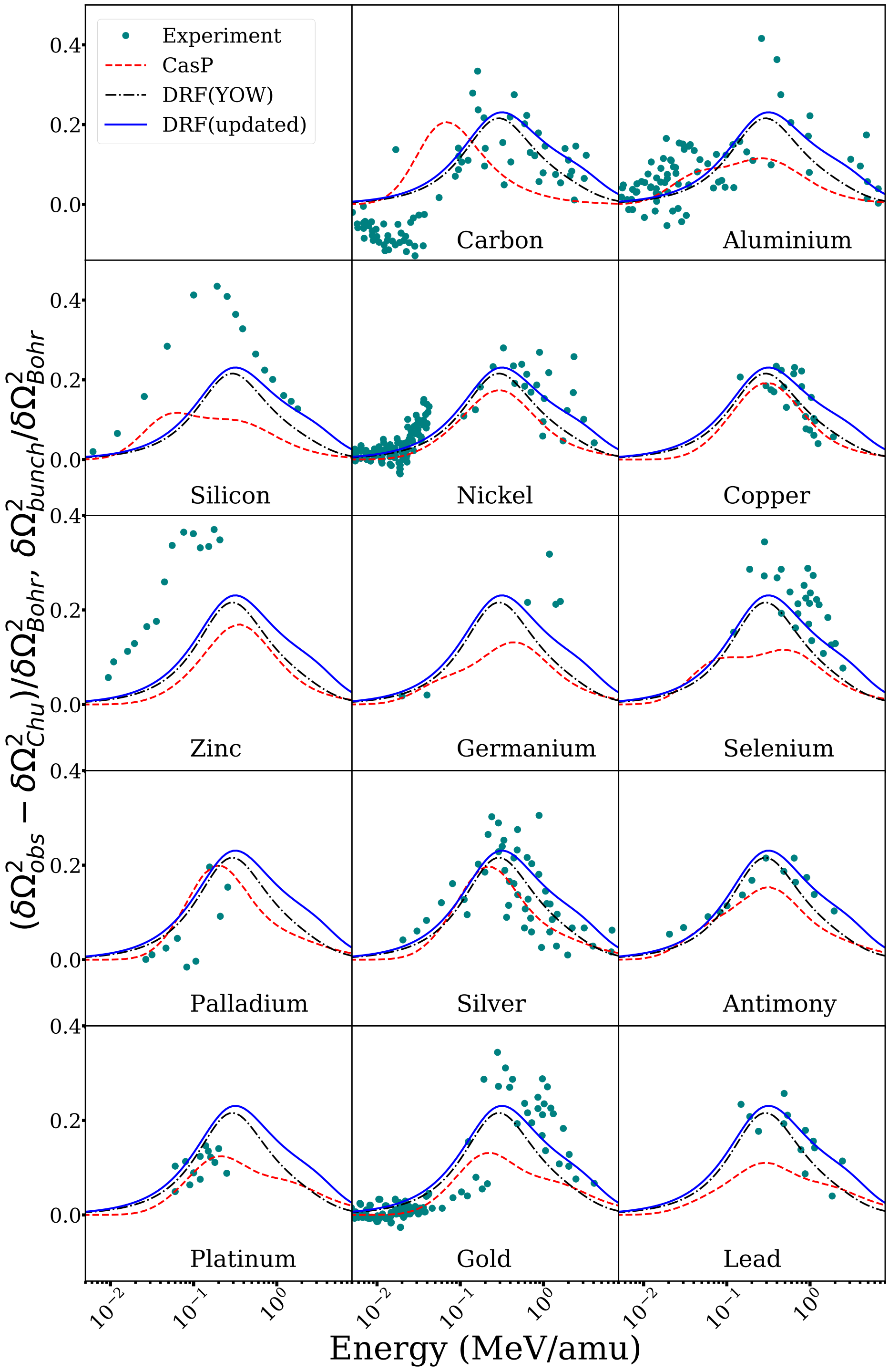


Figure 1: Values of energy-loss straggling subtracted from Chu's calculations and normalized to the Bohr straggling values for protons in elemental compounds. The symbols are experimental data collected from Ref. [9, 12, 22–26]. The dash red line is the CasP curve for each element. The dash-dot black and solid blue curves are the Eq. 7 and 8 fitting with the parameters from Table 1.

Table 1: Parameters in Eq. (7) and (8) for energy straggling of protons in solid and atomic gas targets.

| DRF | $B_1$ (MeV) | $B_2$ (MeV) | $B_3$ (MeV) | $B_4$ (MeV)$^{-1}$ |
|---|---|---|---|---|
| Solid | | | | |
| YOW [9] | 0.195 | 0.694 | 2.522 | 1.040 |
| Updated | 0.412 | 1.165 | 3.759 | 1.176 |
| CasP | 0.115 | 0.574 | 1.460 | 2.176 |
| CasP* | 0.117 | 0.573 | 1.488 | 2.139 |
| Atomic gas | | | | |
| YOW [9] | 0.101 | 0.370 | 0.964 | 3.987 |
| CasP | 0.109 | 0.446 | 2.112 | 1.658 |

* CasP's theoretical curves (using the same elements from Ref. [9]).

In order to show that this straggling excess corresponds to bunching effects ($\delta\Omega^2_{bunch}$) we calculated them using Eq. (6) and $T_i(b)$ from the CasP program for all elements and energies present in the experimental data. These bunching results were also best fitted by a DRF function (Eq. (7)) (fourteen CasP curves in Fig. 1) called DRF(CasP) (solid red curve in Fig. 2). The corresponding parameters are also depicted in Table 1. The bunching CasP results resembles the averaged experimental straggling excess but underestimates the averaged experimental values. In addition the position of the maximum is also at somewhat lower energy. Nevertheless given the large experimental uncertainties, the criterion to subtract the experimental by the Chu formula, and the accuracy of impact parameters dependent energy losses from CasP, we consider this evidence that the bunching effect is the main cause for the observed excess straggling for protons.

In the Yang-O'Connor-Wang work [9] they also provided a DRF formula for the straggling excess for protons in some gases (see Fig. 3). The corresponding magnitude of the straggling excess is similar for solids but its maximum effect shifts to lower energies. This shift is also observed for the DRF curve obtained for CasP calculations of the bunching effects. This confirms the connection between the straggling excess and the bunching effect, which is an atomic feature and therefore less sensitive to gas/solid effects. Indeed the position of the DRF maximum, which best fit the bunching calculations depends specifically on the selected elements and no real gas/solid effect was observed in the CasP bunching calculations.

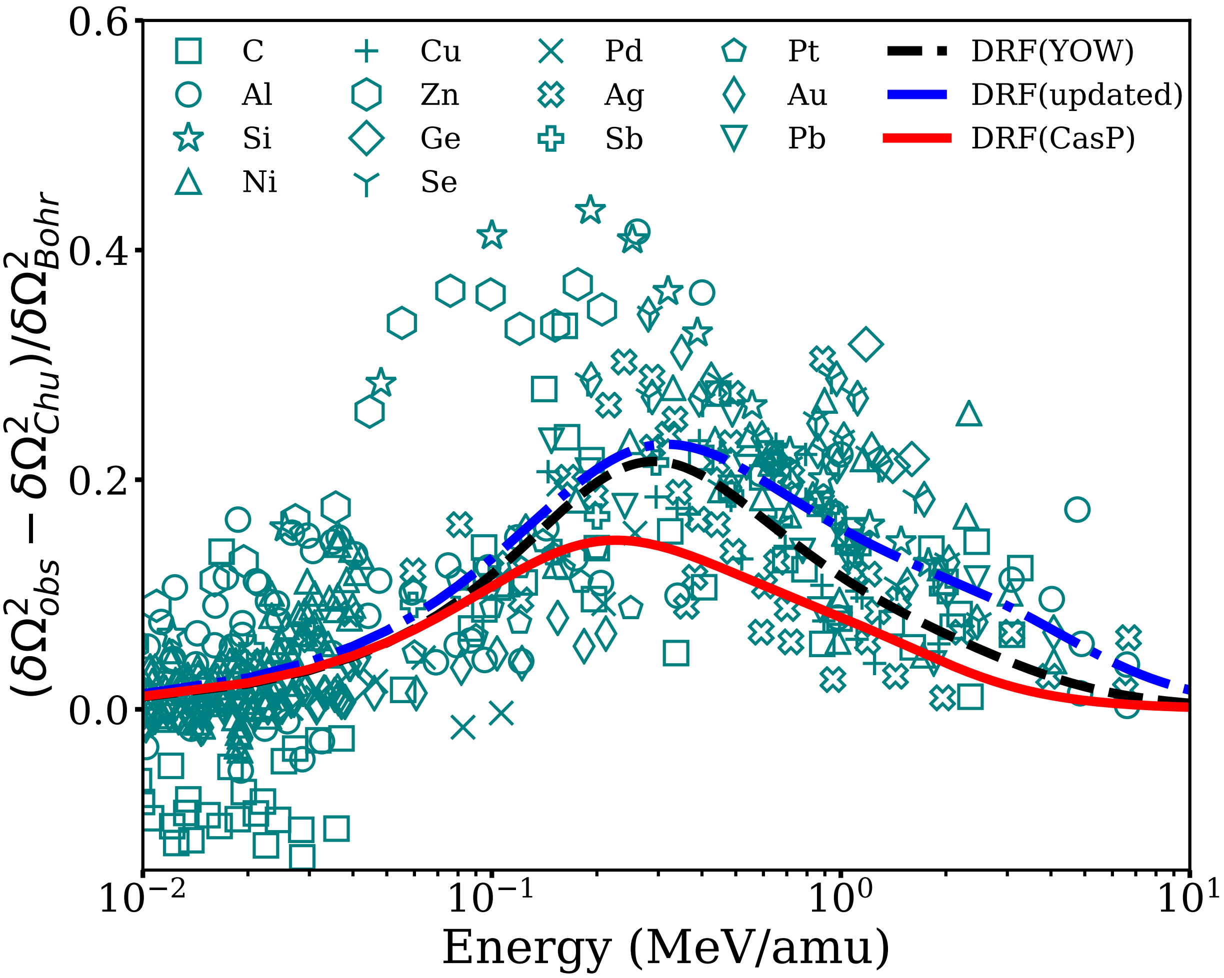


Figure 2: Values of energy-loss straggling subtracted from Chu formula and normalized to the Bohr straggling values for protons in elemental compounds. The open symbols are experimental data collected from Ref. [9, 12, 22–26]. The dash black, dash-dot blue and solid red curves are the DRF fit curve using the parameters from Table 1.

112 *3.2. Statistical correlation*

Another way to evaluate the relation between the experimental straggling excess and the bunching results is through the use of statistical correlations such as the Pearson's correlation coefficient

$$r_{\text{Pearson}} = \frac{\sum_i^n (x_i - \overline{x})(y_i - \overline{y})}{\sqrt{\sum_i^n (x_i - \overline{x})^2 \sum_i^n (y_i - \overline{y})^2}}, \tag{9}$$

where $x_i$ and $y_i$ are two sets of data and $\overline{x} = \sum_i^n \frac{x_i}{n}$ and $\overline{y} = \sum_i^n \frac{y_i}{n}$ are the corresponding mean values respectively ($n$ is the data set length). The Pearson correlation assumes a linear behavior

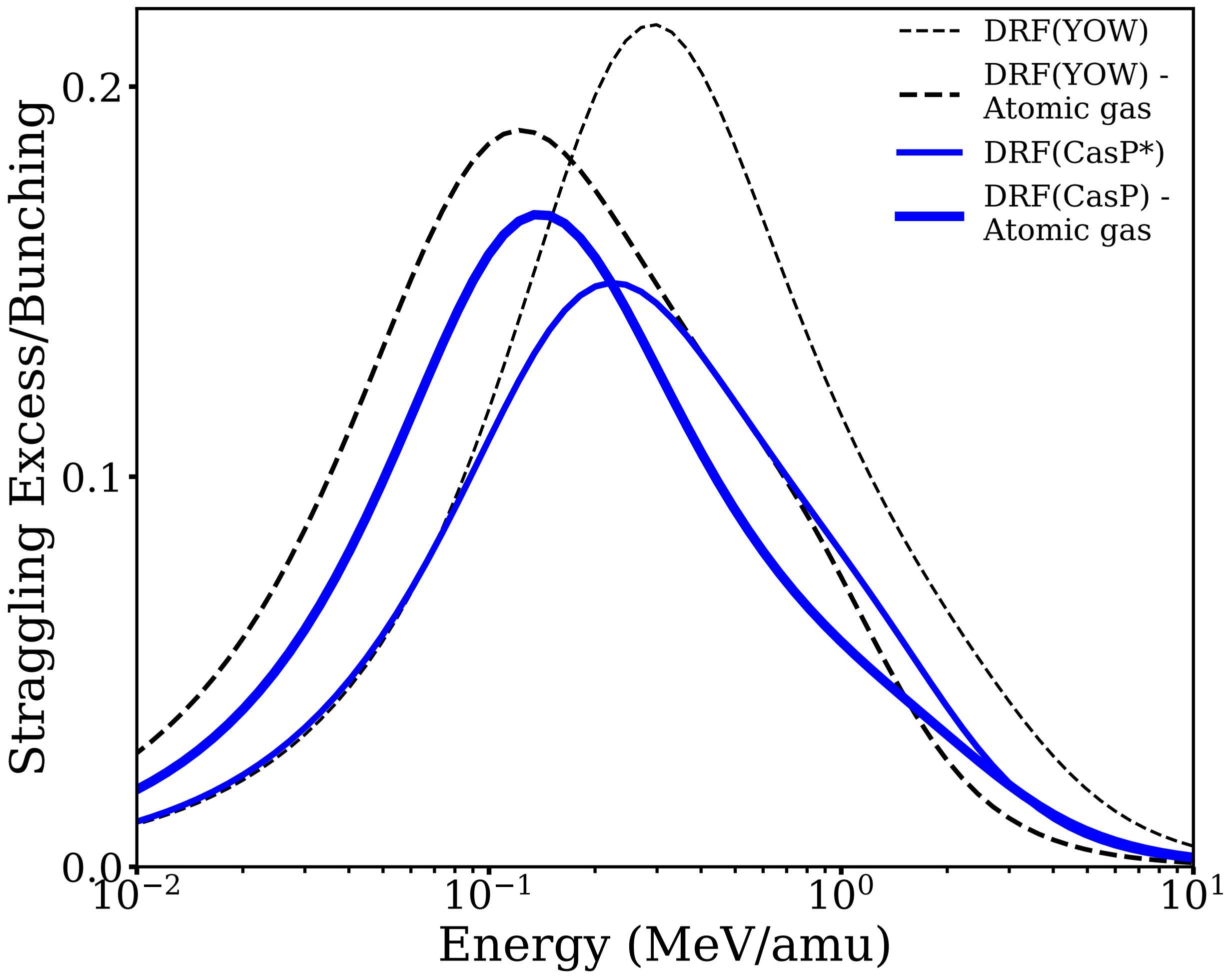


Figure 3: Empirical formulae for solid and noble gas targets obtained by YOW [9] for straggling excess compared by the CasP's theoretical curves (using the same elements from Ref. [9]) for the bunching effect. The curves are the DRF fit curve using the parameters from Table 1.

between the $x_i$ and $y_i$ data sets. On the other hand, the Spearman's rank order correlation coefficient

$$r_{\text{Spearman}} = 1 - \frac{6\sum_i^n d_i^2}{n\left(n^2-1\right)}, \tag{10}$$

where $d_i$ is the difference between corresponding ranks of each variables $x_i$ and $y_i$ [28–32], does not assume any particular dependence between the data sets. For Spearman's rank order correlation, the two data sets ($x_i$ and $y_i$) are ordered in descending order and each element of the reordered data set is assigned a rating between 1 and $n$.

Here, the data sets are the experimental data, bunching calculations and fit curves (displayed

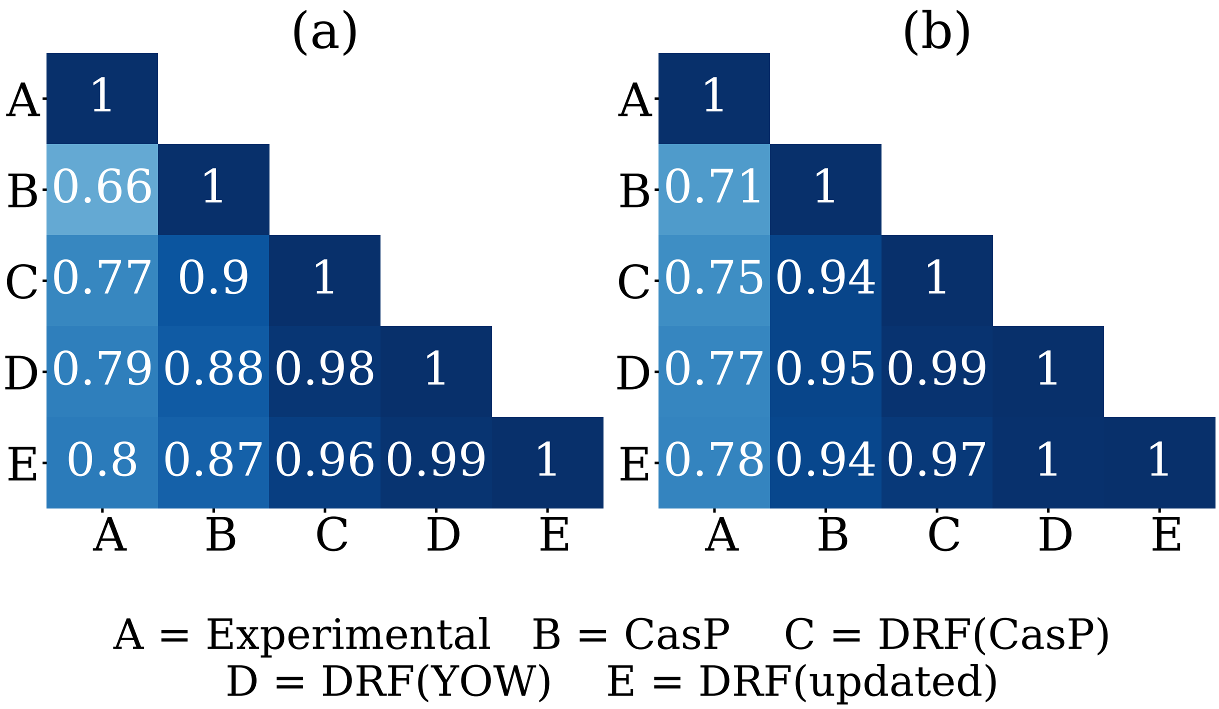


Figure 4: Correlation matrix obtained by: (a) Pearson's correlation coefficient Eq. (9) and (b) Spearman's rank order correlation coefficient Eq. (10).

in Fig. 2). The correlations are calculated for each pair of data sets. First we analyzed the correlations between averaged results and subsequently, we drawn the attention to the correlations for each element. It is pointed out that a correlation coefficient of one only means that the two data set are proportional to each other, but not necessarily identical.

In Fig. 4 we display the statistical correlation matrices for the Pearson's (a) and Spearman's (b) coefficients. The former assumes a linear relationship between two set of data ($x$ and $y$ in Eq. (9)) whereas the Spearman's coefficient (Eq. (10)) is a nonparametric version of Pearson's correlation. Each data set consists of all experimental data for the straggling excess normalized by the Bohr value (Experiment), CasP bunching calculations (CasP) for the same elements and energies in Experiment and the use of the three curves displayed in Fig. 2, namely the averaged bunching (solid red curve), the YOW formula (dash black curve) and this work DRF(updated) (dash-dot blue curve). All statistical correlations are positive and higher than 0.6, which indicates a good correlation between any two data set. The lowest correlation values were found between the experimental data and CasP (0.66), but if we consider the DRF function derived from CasP (DRF(CasP)) (solid red curve in Fig. 2) the correlation increases to 0.77.

Now let us consider the correlations for each element. They are shown in Fig. 5 and 6 for Pearson and Spearman coefficients respectively. Both correlations indicate a good correlation between variables being typically larger than 0.5. Particularly, the DRF(YOW) and the present formulas DRF(updated) are very well correlated with the experimental data. With the exception of carbon, CasP bunching results correlate very well with the experimental data for the straggling excess. For carbon if we consider just straggling data for higher energies the correlation coefficient between Experiment and CasP goes from 0.3 to 0.4 and from 0.22 to 0.36 for Pearson and Spearman coefficients respectively. Still, the carbon case has a poor correlation with CasP curve and show that either the bunching calculation model (here realized by CasP) or the uncorrelated straggling model (here the Chu model) need to be revisited.

Finally, using the Pearson correlation coefficient we were able to find the best correlation between the bunching calculations and a power law of stopping ($S_e^m$) from CasP. The maximum Pearson's correlation coefficient is 0.8 for $m = 4/5$. Here, we use $x_i$ for the CasP stopping calculations and $y_i$ for the CasP bunching calculations, where $i$ is the energy label (ranged from 0.4 keV to 6.7 MeV). This strong correlation indicates that the bunching effect goes with $S_e^{4/5}$ in contrast to the square of stopping power as stated in Ref. [11].

## 4. Conclusion

In this work we show that the well-known Yang-O'Connor-Wang straggling formula has captured bunching effects from the experimental data in an empirical way. This formula was further refined in this work to take into account the subsequent experimental data. The new parameters are depicted in Table 1.

The correlation analysis indicates that the DRF formula from Eq. (7) and (8) describes well the excess of straggling data relative to the Chu formula. Furthermore the strong correlation between the experimental straggling excess to the Chu formula, Eq. (7) and (8) (DRF(YOW) and DRF(updated)), and CasP curves shows the importance of the bunching effect. This effect and the straggling excess are similar and highly correlated for both solid and gas targets and thus, packing effects should be of minor importance. Finally the bunching effect amounts a significant part of the total straggling at 0.2 MeV/amu and calls for more improved straggling calculations and accurate measurements.

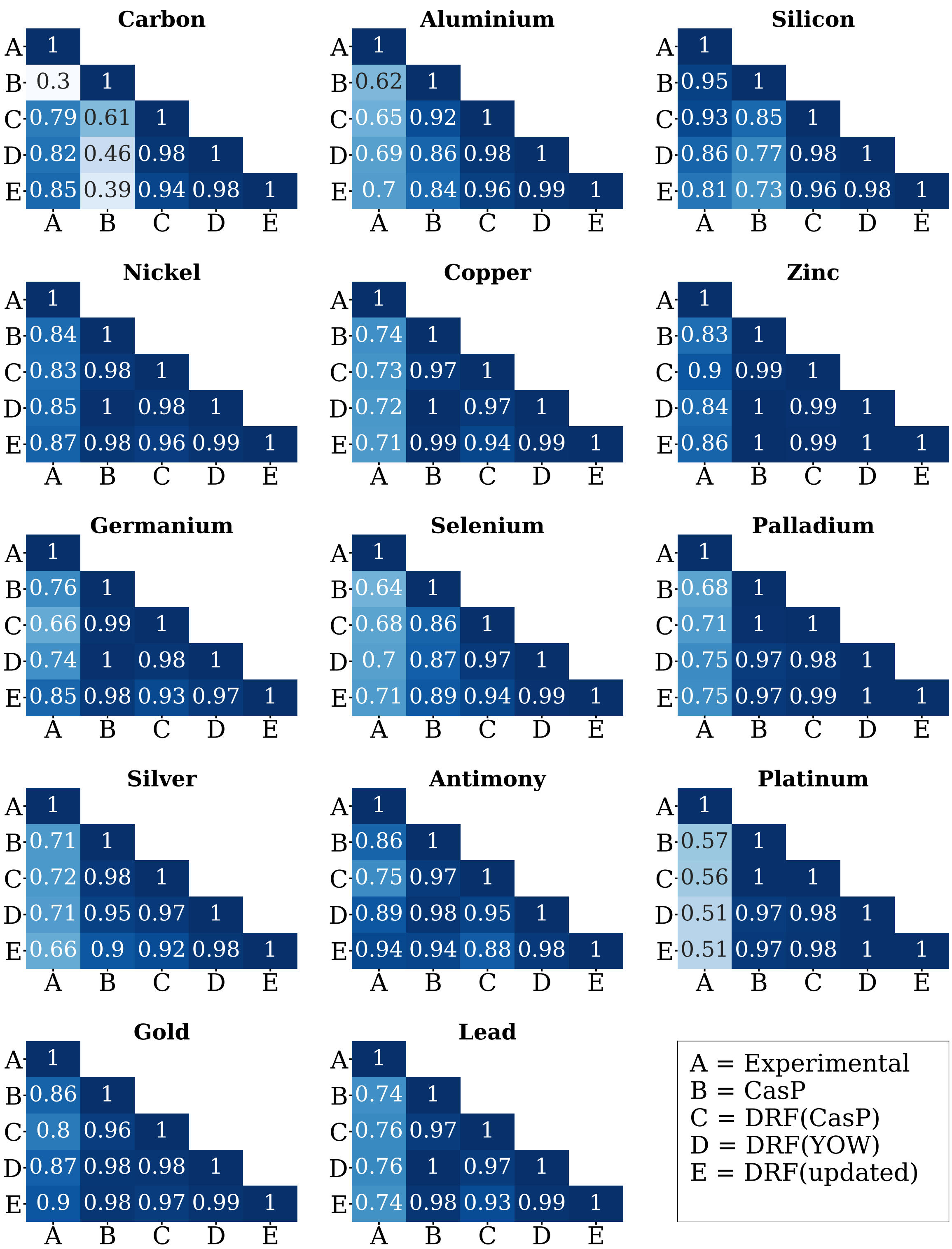


Figure 5: Pearson correlation matrix obtained by Eq. (9) using the same energy range from each experimental data for each element.

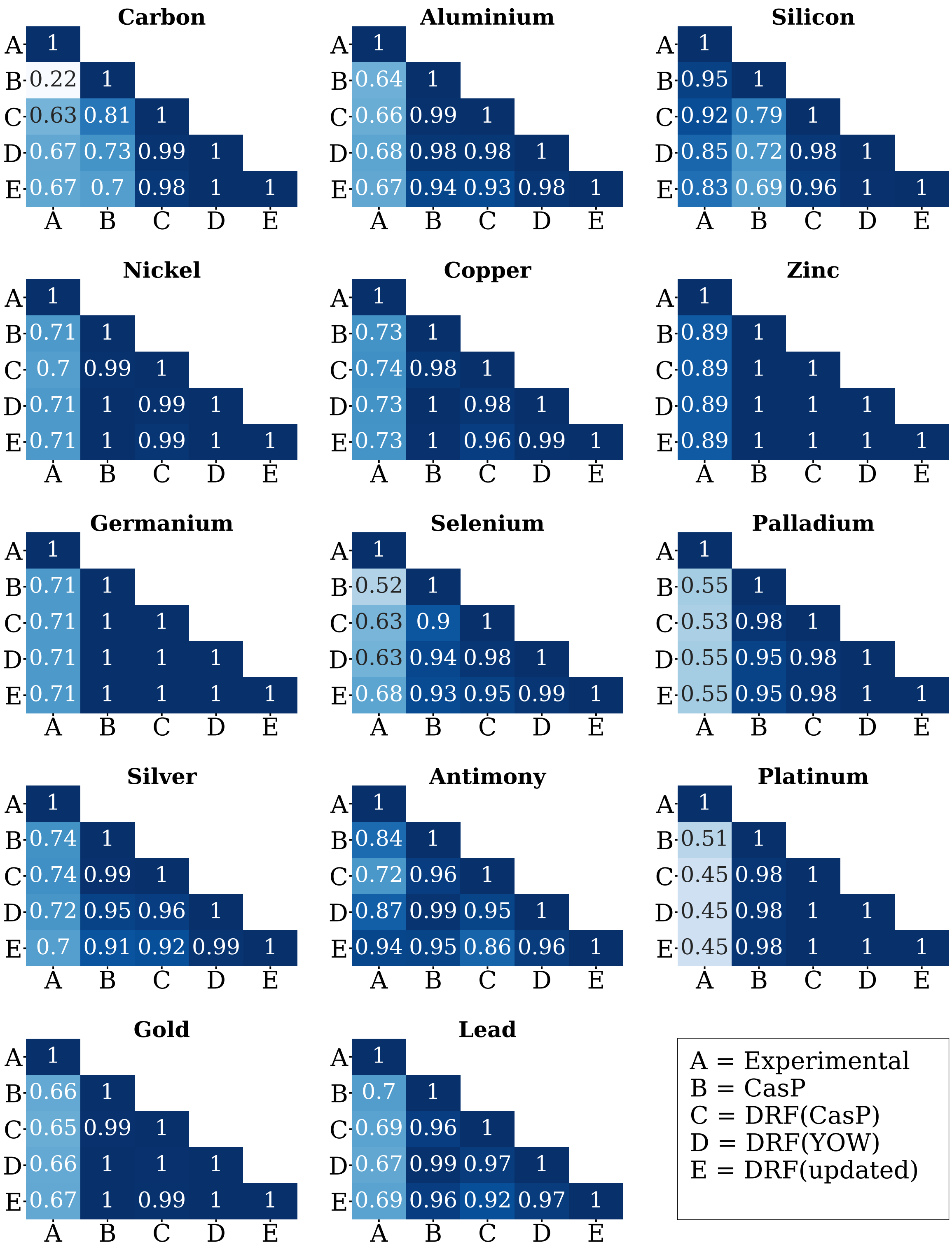


Figure 6: Correlation matrix obtained by Spearman's rank order correlation coefficient Eq. (10) using the same energy range from each experimental data for each element.

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