Height and position of the Bragg peak in the stopping of charged particles

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We have explored the systematics of the magnitude and location of the Bragg peak in the stopping of ions in matter in terms of the atomic numbers $Z_1$ and $Z_2$ of the projectile and target species, respectively. Extensive analysis of experimental results reveals major deviations from predictions by Bethe stopping theory. In particular, the height of the Bragg peak follows a $\propto Z_1^{4/3}$ dependence rather than $\propto Z_1^2$ following from Bethe theory. We assert this to be due to the fact that, with the exception of H ions, the Bragg peak lies below the energy range where Bethe theory can be expected to be valid. Although experimental values from different sources show significant scatter, especially in the location of the peak on the energy axis, general trends are well described by Bohr stopping theory. Moreover, good agreement is found in the absolute magnitude with computations with PASS, including oscillations as a function of $Z_2$, which are known to be related to the mean excitation energy or $I$-value.

Keywords: Stopping power, LET, Bragg peak, Ion beam, Bethe, Bohr
I. INTRODUCTION

In 1904, W. H. Bragg identified a maximum in the density of ionization generated by an α-particle as a function of the penetrated depth\(^1\). This observation reflects the dependence of the ionization cross section on the beam energy.

The present study addresses the maximum in the stopping cross section as a function of the beam energy, the ‘Bragg peak’. This is of interest where ionization needs to be maximized as in particle therapy or where high sensitivity is needed as in ion beam analysis.

A large amount of experimental and theoretical data on stopping cross sections have accumulated over a time span of more than a century. However, for many frequently-used ion-target combinations an attempt to extract the height and position of the Bragg peak reveals error margins well beyond what is acceptable in applications.

Our study employs both experimental data and theory to explore the dependence of peak height and position on the atomic numbers \((Z_1, Z_2)\) of the projectile and target, respectively. In the first step we establish approximate scaling relations on the basis of Bohr\(^2\) and Bethe\(^3\) stopping theory. In the second step we make an attempt to extract such data from a collection of measured stopping cross sections\(^4\), and finally we compare these data with curves extracted from our Dpass database\(^5\).

It has been found long ago\(^6\) that measured stopping cross sections show oscillations as a function of \(Z_2\). This phenomenon is known to originate in oscillations of the mean excitation energy \(I\), the ‘\(I\)-value’. They are found to affect both the height and the position of the Bragg peak. However, the product of peak height and position turns out to be less sensitive to the \(I\)-value. This feature is useful in the comparison between stopping cross sections originating from different sources.

II. PRELIMINARY ESTIMATES

The stopping cross section is conventionally written in the form\(^7\)

\[
S(E) = \frac{4\pi Z_1^2 Z_2 e^4}{mv^2} L(v),
\]

(1)

where the stopping number \(L(v)\) is a dimensionless function of \(v\), \(Z_1\) and \(Z_2\). If \(L(v)/v^2\) has its maximum at \(v = v_1\), the product of the peak height and the position (in terms of energy
per atomic mass unit) is given by

\[ E_1 \times S_1 = (uv_1^2/2)S(v_1) = 2\pi \frac{u}{m}Z_1^2Z_2e^4L(v_1) \]  

(2)
in the nonrelativistic energy range, where \( u \) denotes the atomic mass unit. The function \( L(v) \) is known to vary logarithmically over a wide velocity range and more slowly at low speed. Therefore, eq. (2) is a useful relation between peak energy and position, since the dependence on the ion-target combination is given primarily by the factor \( Z_1^2Z_2 \), whereas more specific parameters enter into the slowly varying function \( L(v) \).

### A. Bohr theory

Classical Bohr stopping theory operates with a single target electron bound harmonically to the nucleus with a resonance frequency \( \omega_0 \) interacting with a point charge in uniform motion. This leads to a stopping number

\[ L = L_{\text{Bohr}}(\xi), \quad \text{with} \quad \xi = \frac{mv^3}{Z_1e^2\omega_0}. \]  

(3)

Bohr quoted the asymptotic result, \( L_{\text{Bohr}} \to \ln(C\xi) \) with \( C = 1.1229 \). A full tabulation of \( L_{\text{Bohr}} \) has been reported in ref. 8.

After expressing \( v \) in terms of \( \xi \) we may find a maximum at \( \xi = \xi_1 \), where \( \xi_1 \) depends only on the functional form of \( f(\xi) = L(\xi)/\xi^{2/3} \). As a result we find

\[ S_1 = 4\pi \left( \frac{Z_1^4e^8}{m\omega_0^2} \right)^{1/3} f(\xi_1) \]  

(4)
for the height of the Bragg peak and

\[ E_1 = uv_1^2/2 = \frac{u}{2} \left( \frac{Z_1e^2\omega_0\xi_1}{m} \right)^{2/3} \]  

(5)
for its position in energy space.

For a target atom with more than a single resonance frequency these simple dependencies are no longer valid. However, as a rough approximation we may follow the conventional scheme and operate with \( Z_2 \) target electrons with a mean resonance frequency \( \omega = I/\hbar \), where \( I \) denotes the mean excitation energy. This leaves eq. (5) unchanged except for replacement of \( \omega_0 \) by \( \omega \) and adds a factor \( Z_2 \) to eq. (4).
The effective binding frequency $\omega$ enters as $\omega^{2/3}$, implying that both peak height and position vary more rapidly than the stopping cross section at fixed energy. Moreover, the peak height decreases with increasing $\omega$, whilst the position increases, as to be expected from the fact that $\omega$ does not enter into the product according to eq. (2). This is important, since $\omega$ is known to oscillate as a function of $Z_2$, as noted above.

Neglecting these oscillations we can find an average behavior by means of the Bloch formula $^9 I \simeq 10Z_2 \text{ eV}$, so that

$$S_1 \propto Z_1^{4/3}Z_2^{1/3}$$

for the peak height and

$$E_1 \propto Z_1^{2/3}Z_2^{2/3}$$

for the position.

B. Bethe theory

The range of approximate validity of classical stopping theory is given by the Bohr criterion $^{10} 2Z_1v_0/v > 1$, where $v_0$ denotes the Bohr velocity or, in energy units,

$$E_{\text{sep}} \leq 0.1Z_1^2 \text{ MeV/u.}$$

Above this limit, Born approximation in the form of the Bethe theory $^3$ is an appropriate starting point.

Actually, because of the dependence on $Z_1^2$ this limit lies well above the Bragg peak for all ions except hydrogen, where the Bragg peak lies in the transition region. Therefore, Bohr theory is expected to be closer to reality, as far as comparison with experiment is concerned.

Nevertheless we quote results from Bethe theory. For a single electron modelled as a harmonic oscillator one finds

$$L = L_{\text{Bethe}}(B) \quad \text{with} \quad B = \frac{2mv^2}{\hbar\omega_0}. \quad (9)$$

According to Bethe $^3$ the asymptotic result is $L(B) \to \ln B$. A full tabulation of $L(B)$ as been reported in ref.$^{11}$. With this we find

$$S_1 = \frac{8\pi Z_1^2Z_2e^4 L(B_1)}{\hbar\omega B_1} \quad (10)$$
and

\[ E_1 = \frac{u \hbar \omega}{2m} B_1. \]  \hspace{1cm} (11)

where \( B_1 \) is a dimensionless constant dependent only on the functional form of \( L_{\text{Bethe}}(B) \). Here, peak height and position go as \( \omega^{\pm 1} \). For the average behavior the Bloch formula suggests that

\[ S_1 \propto Z_1^2 \]  \hspace{1cm} (12)

and

\[ E_1 \propto Z_2. \]  \hspace{1cm} (13)

C. Connection between peak height and position

According to eq. (2) the product of the height and the position of the Bragg peak is independent of the material constant \( \omega \). Within the validity of the Bohr model we may express the coefficient \( L_{\text{Bohr}}(v_1) \) by the Bohr variable \( \xi \). Inserting the asymptotic Bohr formula, \( L(\xi) = \ln(C\xi) \) we find

\[ L_{\text{Bohr}}(v_1) = \frac{2}{3} f(\xi_1) = 3/2 \]  \hspace{1cm} (14)

and, similarly,

\[ L_{\text{Bethe}}(v_1) = L(B_1) = 1. \]  \hspace{1cm} (15)

The important feature here is the fact that for both Bohr and Bethe theory, the dependences on \( Z_1 \) and \( Z_2 \) are given by the product \( Z_1^2 Z_2 \), independent of the material constant \( I = \hbar \omega \). An essential ingredient in the derivation is, however, the dependence of the stopping number on a single scaling variable. It is not clear from the beginning, to what extent standard corrections to Bohr or Bethe theory such as screening and Barkas-Andersen correction as well as charge exchange and projectile excitation affect this result.

Within its range of validity, (2) may serve at least two purposes:

- a check on experimental data on energy loss, and
- determining the position of the Bragg peak from its height.
III. PASS CODE

A. Recapitulation

The PASS code\textsuperscript{12,13} represents an extension of Bohr stopping theory incorporating a Barkas-Andersen effect and allowing for shell and screening corrections, projectile excitation and charge exchange, as well as an inverse-Bloch term that ensures a smooth transition into the Bethe regime. With the exception of the Barkas-Andersen effect, these additions do not obey the simple scaling relations (3) or (9).

B. Recent changes

PASS data compare, for the majority of ion-target combinations studied, well with experimental stopping cross sections\textsuperscript{14}. Nevertheless, improvements have been made which have resulted in the 2020 version of DPASS, which has been accessible on the internet\textsuperscript{5} from March 2020:

1. The main motivation for a mild revision was the experience that for some ions, especially hydrogen and helium, the height of the Bragg peak was underestimated. This deficiency has been found to originate in the excitation of a partially-stripped projectile ion. PASS allows two options, a bottom-up version where the remaining electrons are in their ground state, and a ‘distributed’ version, where they are distributed uniformly over those levels that are occupied in the neutral projectile. In the second option the effective binding energy is lower, and the resulting increase in projectile excitation is found to cause a noticable increase in the effective stopping cross section.

2. PASS allows to differentiate between conductors and insulators by describing the valence shell either by a free-electron gas or by atomic orbitals. Previously we operated with a classical electron gas characterized by its density via the plasma frequency, much like in Kramers’ model\textsuperscript{15}. We keep this description for ions with \( Z_1 = 2 - 92 \), where the Bragg peak lies within the classical limit, but for \( Z_1 = 1 \), a scheme incorporating quantal effects is preferable. Presently we use the Lindhard model\textsuperscript{16}, which is based on perturbation theory and thus is valid mostly for H ions.
3. Most existing $I$-values are based on analysis of measured stopping cross sections for protons in the MeV region\textsuperscript{17,18}. Because of lacking experimental data, $I$-values for heavy target materials are rather uncertain. Therefore, theoretical values from\textsuperscript{19} have been employed for W, Au, Pb and Bi targets and subsequent interpolation between these elements.

C. Comparison with simple estimates

Figure 1 shows a comparison between output from PASS and the simple scaling relation (2). It is seen that the predicted quadratic dependence on $Z_1$ is well fulfilled. Existing differences are to be expected, since PASS output does not obey the simple scaling law (2). However, these differences decrease with increasing $Z_1$, i.e., for ions deeply in the Bohr regime. At the opposite end, for $Z_1 = 1$ and $Z_2 = 2$, the dashed curve is about factor of 1.5 higher than the full-drawn line, as is to be expected, since the Bethe value $L_{\text{Bethe}} = 1$ is more appropriate for protons. However, this difference increases with increasing $Z_2$, indicating that a description in terms of a single resonance becomes questionable with increasing $Z_2$.

Figure 2 shows a similar graph for $Z_2$ as the independent variable. Again, deviations from the simple scaling relationship (2) increase with $Z_2$ and are most pronounced for He ions while very small (within ±20\%) for Kr, Xe and U ions.

Again, we cannot expect perfect agreement, but noticeable deviations from the expected linearity are confined to the lightest ions on the heavier targets. For hydrogen ions we have adopted the coefficient 1.0 from the Bethe model.

IV. EXPERIMENTAL DATA

A. Stopping Cross Sections

Figure 3 shows measured stopping cross sections compiled in the IAEA database\textsuperscript{4}. Only the energy range around the Bragg peak is shown, and only data files that, one by one, could be used to extract numbers for peak height and position.

The upper graph depicts the He-Si system, which is covered by 21 data sets in\textsuperscript{4}, of which 14 include the Bragg peak and have entered the plot. The peak height is seen to vary between 61.5 and 75 units, while the corresponding number for the lowest position is 0.1
and the highest 0.17 MeV/u. The product varies between 6.15 and 12.75. These numbers are typical for ‘good’ systems.

The lower graph in figure 3 illustrates the O-Au system which is likewise well-covered with experimental data, but with significant scatter not only between different data sets but also within at least one of those shown in the graph. While the height of the maximum varies between 820 and 900 units, one may extract values between 0.55 and 1.15 MeV/u for the position, i.e., differing by more than a factor of two. The product $E_1S_1$ is found to vary between 5.59 and 10.35, while eq. (2) delivers a value of 9.40.

Nevertheless, O-Au is still a ‘good’ system, compared with numerous others. Typically, systems are covered by only one data set comprising the Bragg peak. Even if those data allow extraction of accurate numbers for height and position, their absolute error must be suspected to be comparable with what was found for the two examples discussed here, unless there is strong experimental or theoretical support for better accuracy.

### B. Dependence on the ion

Figures 4 and 5 show peak heights and positions for a sequence of ions in He, Ar, Ag and Au. Plotted are experimental values together with curves extracted from DPASS, as described above. We find agreement between measured and calculated peak heights within the scatter of the experimental data. Moreover, the $Z_1$ dependence is very close to $\propto Z_1^{4/3}$ following the Bohr model.

For peak positions the situation differs dramatically. Although the $Z_1$ dependence is close to $\propto Z_1^{2/3}$ as expected from the Bohr model, a scatter of a factor of two between different data sets, as found above in figure 3 is clearly not exceptional. As is evident from figure 3, this scatter reflects differences between different experiments. The scatter within data from individual experiments is much smaller.

These graphs also include the product $E_1S_1$. Despite significant scatter, caused by the scatter in peak positions, these curves follow the prediction from PASS which, moreover, is close to the expected $\propto Z_1^2$ dependence according to the Bohr model.
C. Dependence on the target

Figures 6 and 7 show corresponding graphs for H, He, Kr and Xe ions in a sequence of target materials. Most striking is the periodic dependence of the peak height on $Z_2$ with an amplitude ratio of up to a factor of two. This is most pronounced for hydrogen ions, less dramatic for He and hardly visible for Kr and Xe ions.

It has been found in sect. II that an expected source of such oscillations is fluctuations in the resonance frequency $\omega$, i.e., the mean excitation energy or $I$-value. This is clearly confirmed by the fact that, as found in eqs. (4) and (5), a maximum in height is accompanied by a minimum in position, and vice versa.

Oscillations in the $I$-value were first reported by Burkig and McKenzie\textsuperscript{6} in measurements of stopping cross sections relative to aluminium with 19.8 MeV protons. Absolute measurements by Andersen et al.\textsuperscript{17} with 5-12 MeV protons and deuterons confirmed their existence. Further evidence was found by Chu et al.\textsuperscript{20} in measurements with 2 MeV He ions. An extensive analysis on the basis of a local-density approach and Lindhard-Winter theory of stopping in a Fermi gas\textsuperscript{21} provided a theoretical basis\textsuperscript{22}. These calculations predicted a dramatic increase in oscillation amplitudes toward lower beam energies.

It is easily verified (but not shown in the graphs for clarity) that the prediction of a $\propto Z_2^{1/3}$ dependence from the Bohr model very well describes the general behavior of the peak height in all four cases. Moreover, the product curve is well described by the expected $\propto Z_2$ dependence, with the exception for H and He ions in H and He targets.

Again we find large scatter between peak positions. Despite this, oscillations in the products $E_1 S_1$ are noticably attenuated.

V. DISCUSSION

Figures 8 and 9 summarize the qualitative behavior of the height and position of the Bragg peak as well as the product of the two quantities, predicted by PASS. As could be expected from figure 1, the upper graph in figure 8 shows a smooth behavior, whereas the lower graph shows structure of the type seen in figure 2. Again, this structure is eliminated in the product of the two quantities, just as in figures 4–7\textsuperscript{23}.

The most important result of this study is the fact that with the new version of the PASS
code we have significantly better control of the behavior of the stopping cross section in and near the maximum.

In the IAEA database\textsuperscript{4}, a significant number of experimental results has been rejected because of deviations from the average behavior reported by various authors. Although we have not found obvious errors in specific ion-target combinations, inspection of PASS results may be useful in an attempt to estimate the relative accuracy of experimental findings that disagree less drastically than the behavior of one of the data sets shown for He-Si in figure 3.

A tabulation of peak heights and positions as well as their product, underlying figures 8 and 9 is available from the DPASS site, www.sdu.dk/DPASS, from where also the latest (2020) version of DPASS may be obtained.

VI. CONCLUSIONS

- The height of the Bragg peak follows a power law close to $\propto Z_1^{2/3}$ as a function of the atomic number of the ion, in contrast to a $\propto Z_1^2$ dependence predicted by Bethe theory.

- The position of the Bragg peak varies approximately as $Z_1^{2/3}$, in contrast to a position independent of $Z_1$ as predicted by the Bethe theory.

- Both results follow from the fact that for $Z_1 \geq 2$ the Bragg peak lies well below the range of validity of the Bethe theory.

- The product of peak height and position goes as $Z_1^2$, as follows from both Bethe and Bohr stopping theory.

- Both peak height and position show an oscillatory structure as a function of the atomic number $Z_2$ of the target. The amplitude of these oscillations decreases with increasing $Z_1$, and the phases are opposite to each other.

- This implies that $Z_2$ structure is less pronounced in the product of height and position.

- Neither experiment nor theory show a completely smooth behavior of the product. This is asserted to be due to deviations from the simple scaling behavior due to shell
and Barkas-Andersen correction, projectile screening and excitation as well as charge exchange, which are ignored in these simple estimates.

- These corrections are all accounted for in the PASS code. Predictions of the peak height by PASS agree very well with available measurements, including $Z_2$ structure.

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The spike at $Z_2 = 85$ is an artifact caused by the numerics in the PASS code. Since we are unaware of stopping measurements in Astatium, we have postponed an attempt to localize its origin.
FIG. 1. Product of peak height and position vs. $Z_1$ for five target materials. Solid lines: Extracted from$^5$; Dashed lines: Eq. (2) with $L_{\text{Bohr}} = 3/2$ according to eq. (14).
FIG. 2. Same as figure 1 versus target number for eight projectile ions. For protons the coefficient in eq. (2) has been chosen to 1, which is appropriate in the Born regime.
FIG. 3. Measured stopping cross sections for He in Si (upper graph) and O ions in Au (lower graph). Experimental data from ref. 4.
FIG. 4. Bragg peak vs. atomic number $Z_1$ of the ion for He and Ar target. Points: Measured peak height $S_1$, position $E_1$ (upright triangle), peak position (downward triangle) and product $E_1S_1$ (crossed squares). Lines: PASS predictions.
FIG. 5. Same as figure 4 for Ag and Au target.
FIG. 6. Same as figure 4 vs. atomic number of the target for H and He ion.
FIG. 7. Same as figure 6 for Kr and Xe ion.
FIG. 8. Contour map for the height (upper graph) and the position (lower graph) of the Bragg peak extracted from PASS output. The letter $k$ represents a factor of 1000.
FIG. 9. Same as figure 8 for the product of peak height and position.