Explanation of the observed trend in the mean excitation energy of a target as determined using several projectiles

R. Cabrera-Trujillo and J. R. Sabin
Physics Department, University of Florida, Gainesville, Florida 32611-8435, USA

J. Oddershede
Department of Chemistry, University of Southern Denmark, DK-5230 Odense M, Denmark

(Received 22 May 2003; published 23 October 2003)

Recently, Porter observed [L.E. Porter, Int. J. Quantum Chem. **90**, 684 (2002)] that the mean excitation energy and stopping cross section of a target, obtained from fitting experimental data at given projectile charge to a modified Bethe-Block theory, gives projectile dependent results. The main result of his work is that there is a trend for the inferred target mean excitation energy, to decrease as the projectile atomic number increases. However, this result is inconsistent with the usual definition of the mean excitation energy as a function of target excitation properties only. Here we present an explanation of Porter's results based on the Bethe theory extended to take projectile electronic structure explicitly into account.

DOI: 10.1103/PhysRevA.68.042902 PACS number(s): 34.50.Bw

I. INTRODUCTION

A topic that has intrigued theoretical and experimental physicists since the beginning of quantum mechanics is the energy loss ΔE and stopping power $-dE/dx$ of charged particle penetrating matter. The understanding of energy loss is of importance in applications involving the probing of materials with beams of energetic ions, such as radiology, plasma physics, and astrophysics, to mention but a few. From the theoretical point of view, there are several formalisms aimed at understanding the energy-loss process of partially stripped ions. For example, the effective charge theory of Brandt and Kitagawa (BK) [1] is based on the dielectric-response approximation [2] for an electron gas and calculates the stopping power under the assumption of proportionality between the effective charge and the stopping power of a bare ion. Within the spirit of the Bethe theory, the scheme suggested by Kim and Cheng [3] proposes an effective projectile charge Z_{eff} and mean excitation energy I_{eff} in the standard Bethe formula. The effective parameters Z_{eff} and I_{eff} can be derived from first principles. Lately, we have extended the Bethe theory to include the projectile electronic structure as well as to provide an analytical formula for the ionization fraction in the manner of BK for a Thomas-Fermi model of the atom [4]. This theory is used in this work and an overview is presented in Sec. II

In the course of several papers on the analysis of stopping power data for several target and projectile systems [5–8], Porter has observed certain trends in the behavior of target mean excitation energy. The method used to extract the stopping parameters from measurements is referred to as ''modified Bethe-Bloch theory'' [9], and consists of writing the stopping power as

$$-\frac{dE}{dx}=n_2S(v)=n_2\frac{4\pi e^4Z_2Z_{1,eff}^2}{m_ev^2}L(v), \qquad (1)$$

where $S(v)$ is the stopping cross section, $L(v)$ is the stopping number, $Z_{1,eff}$ is the projectile effective charge, Z_2 is the number of electrons associated with each scattering center, and n_2 is the density of scattering centers of the target. Neglecting relativistic terms and charge exchange, the stopping number consists of Bethe, Barkas, and Bloch terms, and is written as

$$L(v)=\ln\left(\frac{2m_ev^2}{{}_2I_0}\right)-\frac{C}{Z_2}+\xi F(b/x^{1/2})/Z_1^{1/2}x^{3/2}+L_2. \qquad (2)$$

Of the numerous terms in this expression, the target mean excitation energy ${}_2I_0$ and the Barkas parameters ξ and b [10] are projectile velocity independent. The shell corrections C/Z_2 and the Block term L_2 depend on velocity and can be calculated [9]. The experimental data is then fit to Eqs. (1) and (2) using well-documented fitting techniques [11], and the mean excitation energy and Barkas parameter b are found by minimizing root-mean-square deviations between calculated and measured stopping powers (for details of the fitting procedure, see Ref. [11]).

Of particular interest is the fact that, in general, a larger value of the mean excitation energy ${}_2I_0$ for a given target is extracted from proton measurements than the value extracted from α particles or heavier projectiles [7]. In general, the observed trend is that increasing the atomic number of projectiles is associated with decreasing target mean excitation energy. In a simple Bethe theory this would correspond to an increasing stopping power per unit charge with increasing projectile atomic number, which is contrary to observations (cf., e.g., Ref. [12]). In the same work [7], a proposed explanation of the observed trend is presented, which is based on the argument that ''As a consequence of (modified) Coulomb repulsion, a projectile of higher-z will have less access to collisions with inner shell electrons than would a lower-z projectile of the same initial kinetic energy, so that those electrons will have some of the prospective theoretical excitations unavailable to them in the course of the collision and the actual mean excitation energy will be lower than the calculated value'' [7]; a screening argument.

This explanation is made in the context of Bethe theory which assumes that the electronic structure of the projectile

does not play a role in the energy loss process, except through the effective charge of the projectile. Thus, the difference in the stopping power that results when several different ions colliding with the same target should come from the excitation spectrum of the target only. The object of study presented in this work is to provide an alternative explanation for Porter's observation.

The approach we present here is based on an extended version of Bethe theory [4], which takes explicit account of the electronic structure of both the projectile and the target.

In Sec. II, we present a review of the extended Bethe theory. In Sec. III we discuss our results and finally in Sec. IV we summarize our conclusions.

II. REVIEW OF EXTENDED BETHE THEORY

Since the full description of the extended Bethe theory has been presented elsewhere [4], we provide only a summary of its implementation here.

For a projectile with mass M_1, atomic number Z_1, and N_1 electrons that collides with a target of mass M_2, atomic number Z_2, and N_2 electrons, with a velocity v, the stopping cross section, within the Bethe approximation and ignoring charge exchange, is given by [4]

$$S_e=\frac{4\pi e^4}{m_e v^2}\int_{q_{min}}^{q_{max}}\{N_2[Z_1-{}_1M_{00}(\mathbf{q})]^2 + N_1[Z_2-{}_2M_{00}(\mathbf{q})]^2\}\frac{dq}{q}. \tag{3}$$

Here $q_{min}=\epsilon/\hbar v$ and $q_{max}=2m_e v/\hbar$. The effective mean excitation energy ϵ for the system is given by

$$\epsilon={}_2I_0^{1/(1+\alpha)}{}_1I_0^{\alpha/(1+\alpha)} \tag{4}$$

with

$$\alpha=\frac{(Z_2-N_2)^2N_1}{(Z_1-N_1)^2N_2} \tag{5}$$

and where

$$N_i\ln{}_iI_0=\sum_s {}_if_{s_0s}\ln(E_s-E_{s_0}) \tag{6}$$

defines the mean excitation energy of the projectile ($i=1$) or the target ($i=2$). The ${}_if_{s_0s}$ are the appropriate projectile and target dipole oscillator strengths. Thus, the mean excitation energy depends only on the electronic properties of the atom under consideration, as is the usual case [13].

If we employ the Thomas-Fermi model for the electronic structure of an atom, in the same spirit as the BK model [1], the electronic atomic form factor can be written as

$${}_iM_{00}(q)=N_i[1-(q\Lambda_i b)^2 g(q\Lambda_i b)], \tag{7}$$

where

$$g(z)=\frac{A'}{z^2+B'}+\frac{C'}{z^2+D'} \tag{8}$$

with

$$A'=0.370\,93,\quad B'=13.881\,29,$$
$$C'=0.629\,07,\quad D'=0.967\,375, \tag{9}$$

and

$$\Lambda_i=\frac{ca_0}{Z_i^{1/3}b\left[1-\frac{\lambda}{5}\frac{N_i}{Z_i}\right]}\left(\frac{N_i}{Z_i}\right)^{2/3}. \tag{10}$$

Here $\lambda=5/7$, $c=0.969\,376$, and $b=(8/\pi)^{2/3}$ [4].

The number of electrons associated with the projectile (N_1) depends on the projectile velocity v. According to the Bohr criterion [14,15], in the Thomas-Fermi model of the atom, N_1 is given by [4]

$$N_1(v)=Z_1\left(1-\frac{b^2[3x_c(v)+b]}{[x_c(v)+b]^3}\right) \tag{11}$$

with

$$x_c(v)=-2\left(\frac{b}{3}\right)+\frac{1}{h(v)}\left(\frac{b}{3}\right)+h(v) \tag{12}$$

and

$$h(v)=\left[\frac{a(v)}{2}+\left(\frac{b}{3}\right)+\sqrt{\left(\frac{a(v)}{2}\right)^2+a(v)\left(\frac{b}{3}\right)^3}\right]^{1/3}. \tag{13}$$

Here $a(v)=2b^2/0.606\,47y^2(v)$, and $y(v)=v/v_0Z_1^{2/3}$ is the reduced velocity of the projectile.

Using these results in Eq. (3), one obtains separate contributions to the stopping due to electronic excitations in both the projectile ($i=1$, $j=2$) and in the target ($i=2$, $j=1$), i.e,

$$S_e=S_{e,1}+S_{e,2}, \tag{14}$$

where

$$S_{e,i}=\frac{4\pi e^4}{m_e v^2}N_iZ_j^2\left\{i_{f,j}^2\ln\left(\frac{q_{max}}{q_{min}}\right)+(1-i_{f,j})\left[A(i_{f,j}+B)\times\ln\left(\frac{(q_{max}\Lambda_j)^2+C}{(q_{min}\Lambda_j)^2+C}\right)+D(i_{f,j}+E)\times\ln\left(\frac{(q_{max}\Lambda_j)^2+F}{(q_{min}\Lambda_j)^2+F}\right)\right]+(1-i_{f,j})^2\times\left[G\left[\frac{1}{(q_{max}\Lambda_j)^2+C}-\frac{1}{(q_{min}\Lambda_j)^2+C}\right]+H\left[\frac{1}{(q_{max}\Lambda_j)^2+F}-\frac{1}{(q_{min}\Lambda_j)^2+F}\right]\right]\right\} \tag{15}$$

with

$$A=0.448\,685, \quad B=0.402\,031, \quad C=0.278\,19,$$

$$D=0.051\,315\,1, \quad E=6.228\,48, \quad F=3.991\,87,$$

$$G=0.055\,043\,9, \quad H=0.274\,62. \tag{16}$$

Here $i_{f,j}=1-N_i/Z_i$ is the ionization fraction of each collision partner. Also note the dependence of the screening radius of the projectile, Λ_1, with $i_{f,1}$ through $N_1(v)$.

Thus, for a bare projectile ($i_{f,1}=1$) colliding with a neutral target, ($i_{f,2}=0$) the stopping cross section reduces to the standard Bethe result, i.e.,

$$S_e=\frac{4\pi e^4}{m_e v^2}N_2 Z_1^2 \ln\left(\frac{2m_e v^2}{{}_2I_0}\right), \tag{17}$$

which results from the first term in Eq. (15). The extra nonlinear terms in Eq. (15) represent the contribution of the electronic structure of the colliding partner.

III. ANALYSIS

The scheme summarized above, when applied to a stripped projectile of fixed charge Z_1 ($N_1=0$) colliding with a neutral target yields the standard Bethe result, [Eq. (17)], which is applicable only to projectiles with larger velocity compared to that of the target orbital electrons. In this case, the stopping cross section per Z_1^2 should be independent of the type of the projectile. However, for dressed projectiles, Eqs. (14) and (15) demonstrate that there are additional contributions to the stopping resulting from the projectile electronic structure.

From the equations in the forgoing section, we see that there are two effects in play: one due to the electronic structure of the projectile itself, and the other due to the introduction of an equilibrium charge state of the projectile through the Bohr criterion [Eq. (11)].

We consider the electronic structure case first. Equations (14) and (15) show that there are effects from the electronic structure of both projectile and target that enter into the stopping through the mean excitation energies of both collision partners, through the effective mean excitation energy ϵ [Eq. (4)]. In the case we are dealing with here, that of a neutral target, then from Eq. (5) $\alpha=0$, and therefore the effective mean excitation energy becomes $\epsilon={}_2I_0$. However, there is still a contribution from the electronic structure of the projectile due to the terms in Eq. (15) involving $q_{max}\Lambda_1$ and $q_{min}\Lambda_1$.

A second effect stems from the application of the Bohr criterion, which results in a velocity dependent charge on the projectile: the electrons are successively stripped from the projectile as the projectile velocity increases. If one considers Eqs. (2.22) and (2.23) of Ref. [4] [see Eqs. (11) and (12) of this work], it is apparent that the projectile fractional charge has a rather complicated dependence on both the projectile velocity and nuclear charge. This is graphed in Fig. 1 of Ref. [4] as the fractional number of electrons on the projectile, namely, the charge fraction N_1/Z_1 as a function of reduced velocity $v/Z_1^{2/3}$. Furthermore, the stopping cross section is not proportional to the projectile charge $Z_1 i_{f,1}$ as assumed in the modified Bethe-Bloch theory [see Eq. (1) or first term in Eq. (15)], but a dependence on $i_{f,1}$ also appears in the terms involving the electronic structure of the target [terms with $(1-i_{f,1})$] in Eq. (15).

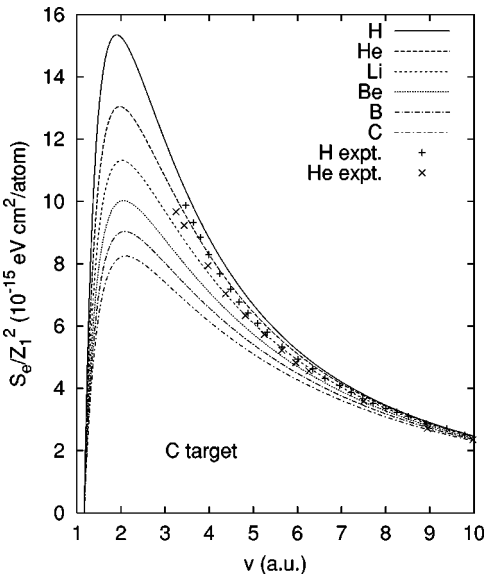

FIG. 1. Carbon stopping cross section per Z_1^2 for projectiles from H to C as a function of the projectile velocity. The experimental data are from Trzaska *et al.* [16].

Since the stopping depends on projectile velocity, not reduced velocity, then as Z_1 increases the curve would "move" left, so at the same velocity, a larger Z_1 atom would have a larger charge fraction, thus a smaller absolute charge, thus the stopping cross section at given velocity would be expected to be smaller for larger Z_1 projectiles, for a common target. If the stopping is described by the single material constant ${}_2I_0$ for the target, then we would expect a "measured" ${}_2I_0$ for a target to increase with the nuclear charge of the projectile.

In Fig. 1, we present the stopping cross section of carbon per Z_1^2 for H, He, Li, Be, B, and C projectiles as obtained by means of the extended Bethe theory [4]. Also, for comparison of the observed trend, we present the experimental data for H and He projectiles [16]. In Fig. 2, we show the stopping cross section per Z_1^2 for H to O projectiles colliding with polystyrene and compare with the experimental data of Leblanc *et al.* [17].

Similarly, in Fig. 3, we present the results for nickel stopping cross section per Z_1^2 for projectiles from H to O as a function of the projectile energy, and compare with available experimental data [12].

For light projectiles, we note a good agreement with the experimental data. Furthermore, the results for hydrogen projectiles are smaller than those for helium and lithium ions when colliding with nickel, as the experiment shows at high energies. However, we should mention that the Bethe approximation reduces the validity of the present results for

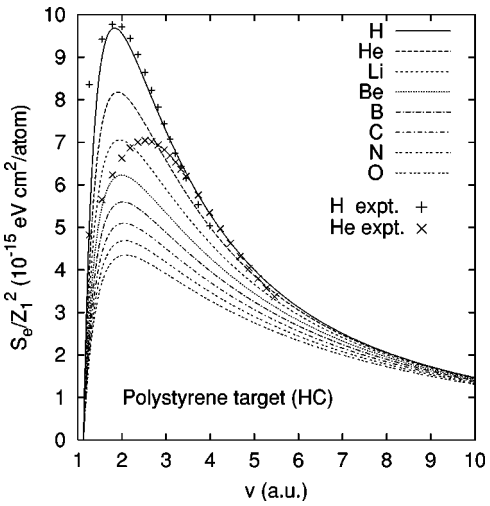

FIG. 2. Polystyrene stopping cross section per Z_1^2 for projectiles from H to O as a function of the projectile energy. The experimental data is from Ref. [17].

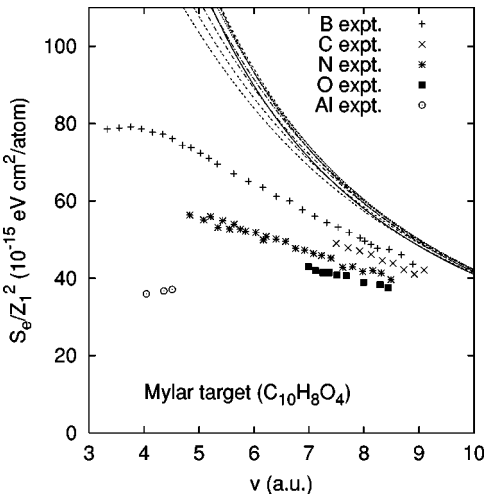

FIG. 4. Mylar stopping cross section per Z_1^2 for projectiles from B to O and Al as a function of the projectile energy. The lines are labeled as in Fig. 2. The experimental data is from Ref. [12].

low projectile energies. We thus expect better agreement with experiment for high projectile energy, as observed.

To illustrate the treatment of a molecular target, we present in Fig. 4 the results obtained for Mylar under the assumption of the validity of the Bragg rule, i.e., neglecting the contribution of the molecular bond to the stopping cross section. From the results, we note the same trend for the stopping cross section as in the previous cases, that is, the importance of projectile electronic structure and its effects on the stopping cross section of a given material target. As expected, however, the results do not compare quantitatively well with the experimental data in the low-energy region, but show the correct trend produced by the electronic structure of the projectile for several projectiles when colliding with a given target.

The values of the mean excitation energy ${}_2I_0$ used for these illustrations where ${}_2I_0(\mathrm{C})=73.8$ eV, ${}_2I_0(HC)=68.7$ eV, ${}_2I_0(\mathrm{Ni})=302.3$ eV, and ${}_2I_0(\mathrm{Mylar})=78.7$ eV, as reported in the ICRU-44 Report [18] and in Janni's tables [19].

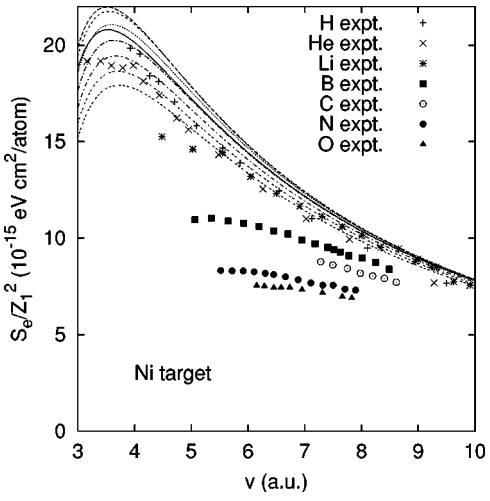

FIG. 3. Nickel stopping cross section per Z_1^2 for projectiles from H to O as a function of the projectile energy. The lines are labeled as in Fig. 2. The experimental data is from Ref. [12].

In all cases, the stopping cross section decreases with increasing projectile atomic number, which is in agreement with the experiments. As expected, the effects are greatest near the peak in the stopping curve, and nearly disappear at large projectile velocities.

IV. CONCLUSIONS

Fitting experimental stopping cross sections for several projectiles impinging on a common target to the modified Bethe-Bloch formula [9] leads to target mean excitation energies, which generally decrease and to Barkas correction parameters, which is generally found to increase [8] with increasing projectile atomic number.

In simple Bethe theory, decreasing the target mean excitation energy, defined as the first moment of the dipole oscillator strength distribution, ought to give an increase in the stopping cross section. However, as the examples presented in Sec. III show, increasing the projectile atomic number leads to decreasing stopping cross section per unit charge in the data analyzed by Porter.

As the only differences among the various cases are rooted in the projectile electronic structure, this must be the origin of the different stopping cross sections. If the projectile electronic structure is taken into account explicitly as it is in the extended Bethe theory, the correct behavior, namely, decreasing stopping cross section per unit charge with increasing projectile atomic number, is obtained. Why then, does the fitting by using the modified Bethe-Bloch theory gives such good fits to experimental data, when it results in counter intuitive trends for the mean excitation energy?

The answer clearly lies in the electronic structure of the projectile, which, as can be seen from the extended Bethe theory calculations, is critical. Fitting the experiments to a Bethe-like theory forces all the electronic effects of both tar-

get and projectile into the free parameters of the fit, namely, the target mean excitation energy and the much less important Barkas parameter. Consequently, the mean excitation energy obtained from the fitting procedure should be viewed as a parameter of the fit that does not corresponds directly to theoretical mean excitation energies determined from calculated dipole oscillator strength distributions.

A corollary to this is that the most reasonable comparisons between experimental mean excitation energies and theoretical ones should be for fast proton projectiles, where there is no projectile electronic structure. A similar argument can be made concerning the Barkas strength factor, which describes the mutual polarization of the projectile and target electron clouds.

It should be emphasized that it is the trends in mean excitation energies and stopping cross sections that we are concerned with here, not the absolute values. We have emphasized before [20] that an experimentally determined mean excitation energy should not be considered ''correct,'' but only consistent with the choices of shell corrections and the forms and parameters for the Barkas and Bloch corrections used to determine it. Using a consistent set of choices when fitting various data should validate the observed trends.

Use of the extended Bethe theory maintains the concept of mean excitation energy as a materials property, for both target and projectile, and provides for their interaction. In addition, the correct trends in stopping cross section per unit charge are reproduced and explained.

Although we have addressed few examples here, the principles should be the same for collisions of dressed ions with complex molecules. If the simple Bragg rule [21] were to be applied to the complicated organic targets measured by Porter, the same trend would be expected to appear, namely, that the stopping cross section would decrease with increasing projectile nuclear charge.

Finally, although qualitative agreement is established for all targets and projectiles, good quantitative agreement is only found for light projectiles colliding on a light target. A more close accord between theory and experiment is desirable for heavy ions or molecular complexes. Dynamical effects [22] as charge exchange, electron capture and loss, bond breaking and/or making, appearance of rovibrational channels, high-order nonlinear effects [23,24], to mention a few, are not considered in this model and which are of extreme importance in the low-to-medium projectile energies. Inclusion of these effects might render a better quantitative agreement.

Note added in proof. Recently, Porter [25] published an analysis of additional data indicating a dependence of inferred mean excitation energies on projectile atomic number. This analysis is consistent with his previous work referenced here, and further supports our conclusions.

ACKNOWLEDGMENTS

This work was supported in part by a grant from the Office of Naval Research (Grant No. N0014-86-1-0707 to JRS). We thank Professor Porter for providing us with a copy of Ref. [8] prior to publication.

[1] W. Brandt and M. Kitagawa, Phys. Rev. B **25**, 5631 (1982).
[2] J. Lindhard and M. Scharff, K. Dan. Vidensk. Selsk. Mat. Fys. Medd. **27**, 15 (1953).
[3] Y.K. Kim and K. Cheng, Phys. Rev. A **22**, 61 (1980).
[4] R. Cabrera-Trujillo, S.A. Cruz, J. Oddershede, and J.R. Sabin, Phys. Rev. A **55**, 2864 (1997).
[5] L.E. Porter, E. Rauhala, and J. Räisänen, Phys. Rev. B **49**, 11543 (1994).
[6] L.E. Porter, Nucl. Instrum. Methods Phys. Res. B **159**, 195 (1999).
[7] L.E. Porter, Int. J. Quantum Chem. **90**, 684 (2002).
[8] L. E. Porter, Int. J. Quantum Chem. **95**, 504 (2003).
[9] L.E. Porter, Int. J. Quantum Chem. **75**, 943 (1999).
[10] J.C. Ashley, R. Ritchie, and W. Brandt, Phys. Rev. B **5**, 2393 (1972).
[11] L.E. Porter, Int. J. Quantum Chem. **65**, 997 (1997).
[12] J. Räisänen and E. Rauhala, Phys. Rev. B **41**, 3951 (1990).
[13] M. Inokuti, Rev. Mod. Phys. **43**, 297 (1971).
[14] N. Bohr, Phys. Rev. **58**, 654 (1940).
[15] N. Bohr, Phys. Rev. **59**, 270 (1941).
[16] W.H. Trzaska, V. Lyapin, T. Alanko, M. Mutterer, J. Räisänen, G. Tjurin, and M. Wojdyr, Nucl. Instrum. Methods Phys. Res. B **195**, 147 (2002).
[17] L. Leblanc, G.G. Ross, and W.E. Wallace, Nucl. Instrum. Methods Phys. Res. B **95**, 457 (1995).
[18] International Commission on Radiation Units and Measurements, Report No. 44 (Bethesda, MD, 1989).
[19] J.F. Janni, At. Data Nucl. Data Tables **27**, 341 (1982).
[20] J.R. Sabin and J. Oddershede, Nucl. Instrum. Methods Phys. Res. B **44**, 253 (1990).
[21] W.H. Bragg and R. Kleeman, Philos. Mag. **10**, 305 (1918).
[22] R. Cabrera-Trujillo, Y. Öhrn, E. Deumens, and J.R. Sabin, J. Chem. Phys. **116**, 2783 (2002).
[23] N.R. Arista, Nucl. Instrum. Methods Phys. Res. B **195**, 91 (2002).
[24] P.L. Grande and G. Schiwietz, Nucl. Instrum. Methods Phys. Res. B **195**, 55 (2002).
[25] L.E. Porter, J. Electron. Spectrosc. Relat. Phenom. **129**, 273 (2003).