Effective charge and the mean charge of swift ions in solids

A. F. Lifschitz and N. R. Arista
División Colisiones Atómicas, Comisión Nacional de Energía Atómica, Centro Atómico Bariloche, Instituto Balseiro, RA-8400 San Carlos de Bariloche, Argentina

(Received 22 November 2002; published 23 January 2004)

We perform a nonlinear study of the energy loss of light and heavy ions in solids using the quantum transport-cross-section method based on numerical solution of the Schrödinger equation. The charge of the ion—the relevant parameter in this calculation—is considered equal to the average ion charge measured after emergence from solid foils. When the calculated energy loss values are analyzed within the framework of the effective charge approach, the results of the present calculations are in excellent agreement with the empirical values and also show the expected scaling properties. Through this analysis, we find a relationship between the charge state inside the solid and the effective charge related to energy loss studies, which solves a seaming contradiction between these values. The implications of this study with respect to the Bohr-Lindhard and Betz-Grodzins models are discussed.

DOI: 10.1103/PhysRevA.69.012902 PACS number(s): 34.50.Bw, 34.50.Fa

I. INTRODUCTION

The problem of the charge state of ions penetrating matter is one of the most relevant questions for studies on ion penetration in solid targets. A large number of studies have been produced in this area, both theoretically and experimentally. It is known that after some penetration distance the ions reach a state of charge equilibrium determined by the competition between capture and loss processes [1–3]. As a result of this equilibrium the ions acquire a mean ionization charge $\overline{q}$.

However, a direct determination of the $\overline{q}$ values inside the solid is not possible, since only the charge state after emerging from a foil can be measured. From these type of measurements the values of the mean charge of the emerging beam $\overline{q}_{exit}$ is determined, but the relation between $\overline{q}_{exit}$ and $\overline{q}$ is generally unknown.

Two main models to describe the charge state inside the solid have been proposed: the Bohr-Lindhard [4] and the Betz-Grodzins [5] models, which provide very different views of the problem.

A second and more indirect method to infer the charge state of the ions inside the solid is from its energy loss. This has lead to the concept of the *effective charge* [2,3,6,7] which is supposed to yield information on the equilibrium charge state of the ions. According to perturbation theory, the stopping power of a bare ion with atomic number Z and velocity v is proportional to Z^2. Based on this, the effective charge Z_{eff} of an ion with atomic number Z_1 and velocity v is operationally defined through the stopping power ratio

$$\frac{Z_{eff}}{Z'_{eff}}=\left[\frac{S_{\exp}(v,Z_1)}{S_{\exp}(v,Z'_1)}\right]^{1/2}, \qquad (1)$$

where $S_{\exp}(v,Z_1)$ is the experimental stopping power of the ion Z_1 and $S_{\exp}(v,Z'_1)$ is the corresponding stopping of a chosen reference ion with atomic number Z'_1 (usually hydrogen or helium ions) with the same velocity v.

It has been known for many years that the values of $\overline{q}_{exit}$ for swift ions exceed those of Z_{eff} (we will not consider here the low-energy range where this relation changes). These differences are shown in Fig. 1 for various ions according to the fitting expressions described below. As it may be observed, large differences are obtained for swift heavy ions for energies in the range of a few MeV/u.

The Betz-Grodzins (BG) [5] model claims that the effect of repeated collisions within the solid produces ions with several excited electrons in outer shells, but these electrons remain mostly attached to the ion until it emerges into vacuum and, after this, the ion decays to the ground state by giving up its excess energy by electron emission through Auger processes. Hence, the model predicts a significant number of emitted electrons in the case of swift heavy ions (cf. Fig. 1). These multiple emission processes have been sought for many years [3,8–10], but the number of electrons actually observed was much lower than the predictions.

The Bohr-Lindhard (BL) model [4], on the other hand,

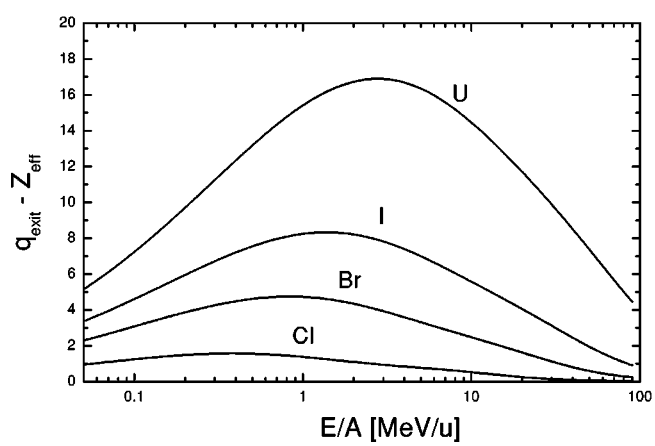

FIG. 1. Differences between the mean charge of ions emerging from solid targets $\overline{q}_{exit}$ (using the empirical fit by Schiewietz and Grande) and the effective charges Z_{eff} (using the fitting equation given in the text) for Cl, Br, I, and U ions, in carbon targets, as a function of energy per unit mass.

considers that the fast sequence of collisions experienced by the ion within a solid produces an enhancement in the excitation and ionization probabilities, leading to an increased equilibrium charge. In this way the BL model would explain the higher values of the ion charge measured after a foil. But the remaining open question is how to explain the difference between $\overline{q}_{exit}$ and the values of the effective charges Z_{eff} determined experimentally from energy loss measurements.

The effective charge concept has proved to be a very useful parameter to condense a very large body of experimental data [7], particularly for swift heavy ions, also providing a simple and practical scheme, and its use in the literature has been generalized. However, it should be noted that the relation (1) is motivated by the property, arising from perturbation theory that the stopping of a bare ion with charge Z is proportional to Z^2. But one should be aware that the usual condition for the applicability of the perturbative picture, $Ze^2/\hbar v \ll 1$ is not fulfilled in many cases, and so the physical meaning of the effective charge obtained from the scaling property $S \propto Z^2$ may be misleading [11,12]. For instance, to represent some cases dealing with swift heavy ions it may be necessary to include higher-order (nonlinear) terms, and still for stronger interactions or low velocities the whole perturbative picture breaks down. One example where this occurs is the phenomenon of Z_1 oscillations in the stopping of slow ions [13–15], a feature that the perturbative or statistical models cannot explain [16], and a proper treatment requires full quantum calculations [17,18]. Recently, various nonperturbative calculations have been carried out [19–21]. These developments may lead to significant advances exploring different approaches.

In addition, various approaches have been developed over the years to calculate the energy loss of swift ions in solids, which use different assumptions on the charge state of the ions. Some of these models use ion-charge values which are very similar to the effective charge fittings, while others are based on the equilibrium ion charge measured after foil strippers. Although there exist significant evidence that the effective charge does not represent the charge state of ions moving inside the solid [19,22], in practice one finds that in the case of swift ions some confusion still remains, and also that the use of fitting q values in widely used computer programs [7,23,24] is sometimes assumed to represent the internal ion charge values.

The purpose of this paper is to show that an adequate nonlinear calculation of the energy loss of heavy ions in solids using quantum theory to evaluate the transport cross-section by numerical methods provides a consistent picture of the charge *vs.* stopping relation which is free from the effective charge ansatz, and where the mean ion charge $\overline{q}$ enters as the only physically relevant ion charge parameter. Through this analysis we will give strong evidence that the ion charge within the solid is reasonably well represented by the emerging ion-charge, $\overline{q} \cong \overline{q}_{exit}$, contrary to the predictions of the BG model. The implication of these results for the interpretation of the effective charge will be discussed.

II. THE NONLINEAR STOPPING APPROACH

The self-consistent nonlinear model to represent the energy loss of nonrelativistic ions was derived in previous papers, both for light [25–27] and heavy ions [28]. The target is considered as a free-electron-like medium. Here we consider the case of carbon, where valence electrons are represented by a uniform electron gas with density *n*, Fermi velocity v_F and Wigner-Seitz radius $r_s = 1.919/v_F$, with $r_s = 1.66$ corresponding to amorphous carbon. Corrections due to *K*-shell excitations are also applied [28] since they are important in the present energy range, but we do not consider the energy losses associated to projectile excitations or electron exchange processes which may yield contributions at lower energies.

The potential of a moving ion with atomic number Z_1 and velocity v is modeled as a sum of two components:

$$V_{ion}(r) = V_{core}(r) + V_s(r) = -\frac{N_e e^2}{r}\phi_{core}(r) - \frac{qe^2}{r}\phi_s(r), \tag{2}$$

where the core term $V_{core}(r)$ includes the potential of the nucleus and atomic screening by the bound electrons (with core screening function ϕ_{core}) and the screening potential $V_s(r)$ is represented by an exponential screening function, $\phi_s(r) = \exp(-\alpha r)$, where the value of α is adjusted for each ion velocity [28]. $N_e = Z_1 - q$ is the number of bound electrons attached to the ion and q is its charge (which depends on the ion velocity).

The ion-potential model $V_{core}(r)$ used here is the so-called Molière ion potential [28], which may be simply obtained from the usual Molière potential for neutral atoms using an appropriate screening function $\phi_{core}(r)$.

The mean energy loss is evaluated in a nonlinear (or nonperturbative) way starting from numerical integrations of the radial Schrödinger equation, which describes the scattering of electrons in the field of the moving ion, Eq. (2). From these integrations we determine the phase shifts $\delta_l(v_r, v)$ (with $l = 0, 1, \ldots$), which depend on the *relative* electron-ion velocity v_r (with $\vec{v}_r = \vec{v}_e - \vec{v}$) and on the ion velocity v (because the scattering potential $V_{ion}(r)$ depends parametrically on v). From the δ_l values we can calculate the transport-cross-section (TCS) $\sigma_{tr}(v_r, v)$ as a function of v_r and v,

$$\sigma_{tr}(v_r, v) = \frac{4\pi}{{v_r}^2}\sum_{l=0}^{\infty}(l+1)\sin^2[\delta_l(v_r, v) - \delta_{l+1}(v_r, v)]. \tag{3}$$

Finally, we obtain the stopping power $S = -\langle dE/dx \rangle$ integrating the TCS over relative electron-ion velocities: $|v - v_e| \leq v_r \leq v + v_e$, and over the distribution of initial electron velocities within a Fermi sphere ($0 \leq v_e \leq v_F$) [25–28], namely,

$$S(v) = \frac{1}{4\pi v^2}\int_0^{v_F} v_e dv_e \int_{|v-v_e|}^{|v+v_e|} dv_r v_r^4 \sigma_{tr}(v_r, v) \times\left[1 + \frac{v^2 - v_e^2}{v_r^2}\right]. \tag{4}$$

The relevant parameter required by the present calculations is the equilibrium charge state of the ion within the solid. The main assumption here is that the charge state inside the solid may be well represented by $\overline{q}_{exit}$ (the ion-charge measured after emerging from the solid). Therefore, we have used as input values the empirical fitting of $\overline{q}_{exit}$ given by Nikolaev and Dmitriev [29] (ND) and the recent one given by Schiwietz and Grande [30] (SG), which agree closely with the experiments [31].

III. RESULTS AND DISCUSSION

We have performed a series of calculations for incident ions with atomic numbers in the range $1 \leqslant Z_1 \leqslant 92$ and with energies of 1, 2, 5, and 10 MeV/u. The ion charges within the solid are represented by the $\overline{q}_{exit}$ values given by the ND and SG fittings [29,30]. From these results we have evaluated the stopping power ratios relative to protons, $S(Z_1,v)/S(1,v)$. These ratios were used to obtain theoretical values of the effective charges according to the definition of Eq. (1), namely; $Z_{eff}=[S(Z_1,v)/S(1,v)]^{1/2}$.

The results of these calculations are presented in Figs. 2 and 3. First, we show in Fig. 2 a set of calculations for fixed velocities, corresponding to 1, 2, and 5 MeV/u. The two curves indicated by A are the $\overline{q}_{exit}$ values given by the ND and SG fittings (our input charge values $\overline{q}$) while the two curves denoted by B are the equivalent "effective charges" theoretically obtained for each case. The circles in the figure are the empirical effective charge values determined from energy loss measurements [32,33]. We find a remarkable agreement with the empirical values in nearly all the cases [except for some discrepancies in panel (a) for the heaviest ions]. It may be observed that the lines A and B start to diverge for $Z_1 > 20$. This is the relevant range of Z_1 where the differences between $\overline{q}_{exit}$ and Z_{eff} are important. Also, as observed in Fig. 1, the most adequate energy range is the one covered by these calculations. The results for 1 MeV/u show the largest differences between the input charge $\overline{q}=\overline{q}_{exit}$ used in the calculations (curves A) and the deduced effective charge values (curves B). The differences between both curves B show the sensitivity of the calculations with respect to the input charge values. It may also be observed that if we had used $\overline{q}$-values similar to Z_{eff} as the input ion-charge within the solid, the results of the analysis would produce output curves well below those of curves B, in wide discrepancy with the experimental results. Hence, the possibility of assuming ion-charge values within the solid close to Z_{eff} is clearly ruled out by these nonlinear calculations. It is of interest to note that a somewhat similar analysis has been done by Maynard *et al.* [19] using a different (but also nonlinear) kinetic-theory type of approach.

In Fig. 3 we have collected the results of numerous calculations, for all Z_1 values, and for the range of energies between 1 and 10 MeV/u. Here we have merged all the calculated stopping values, represented in the form of effective charges using Eq. (1), and we have rescaled all the velocities according to the Thomas-Fermi prescription [6], in the form

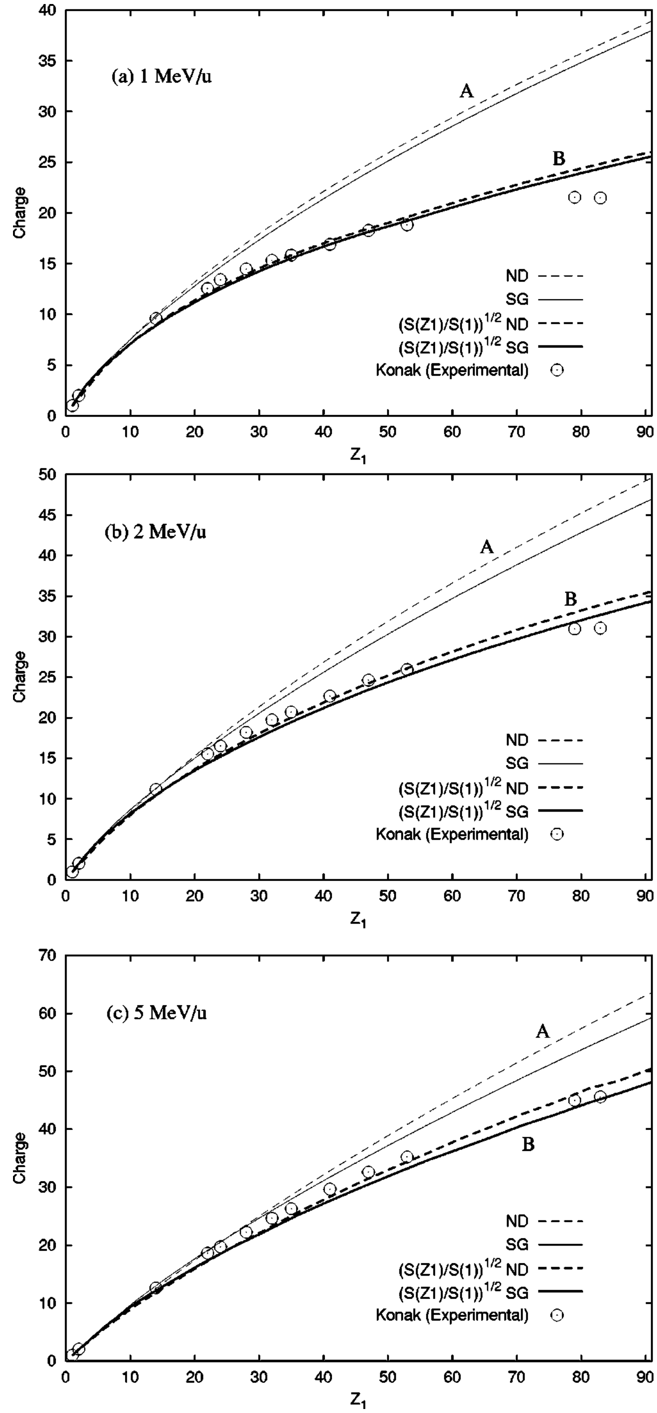

FIG. 2. Curves A: mean ion charges used as input values in the present calculations according to the Nikolaev-Dmitriev (ND) and Schiwietz-Grande (SG) fittings (shown with dashed and continuous thin lines respectively). Curves B: theoretical "effective-charge" values deduced from the no-linear stopping calculations using the previous ND and SG input charge values (results shown with dashed and continuous thick lines respectively). The circles indicate the empirical values of effective charges obtained from energy loss measurements. The results for ion energies of 1, 2, and 5 MeV/u are shown separately in panels (a), (b), and (c).

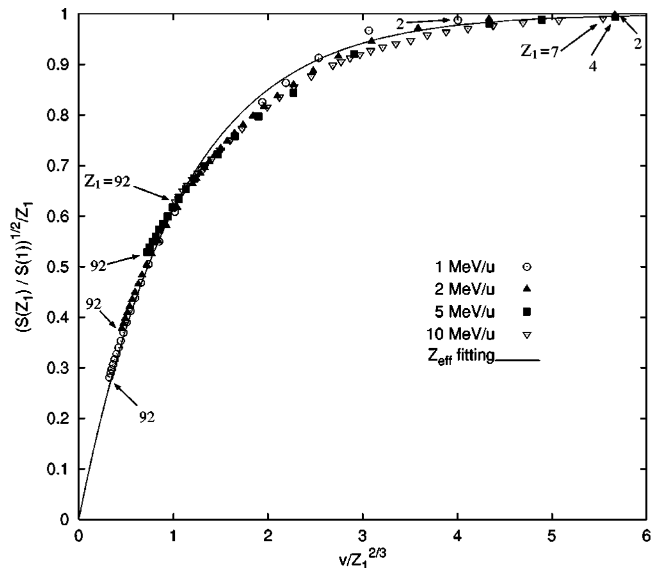

FIG. 3. Joint representation of all the "effective charge" values theoretically obtained from Eq. (1) using the results of energy loss calculations for all incident ions with $1 \leqslant Z_1 \leqslant 92$ and for energies of 1, 2, 5, and 10 MeV/u, as a function of the reduced velocity parameter $v/Z_1^{2/3}$. The calculated values are indicated here by symbols. The curve represents the universal fitting by Ziegler *et al.* to the empirical effective-charge values, corresponding to a large collection of measurements for many different cases.

$v/Z_1^{2/3}$. The calculated results are shown here with symbols. We also show in the figure the widely used fitting formula: $Z_{eff}/Z_1 = 1 - \exp(-0.92v/Z_1^{2/3})$. As shown by previous compilations of data [7], this formula represents within 10% the universe of experimental results for $v > 3$ a.u. Hence, this figure shows quite conclusively the scaling of the present theoretical results, as well as a remarkable agreement with the whole body of experimental data represented by the fitting curve.

It should be stressed that our calculations are based on the assumption that the $\overline{q}$ values inside the solid may be approximated by $\overline{q}_{exit}$. These values have been considered for a long time to be inconsistent with the empirical Z_{eff} values. Our calculations show that there is no contradiction between these quantities, and moreover there is a relationship between them which emerges from a full nonlinear calculation of the stopping power. This explains also why this relationship could not be found in previous studies which were based on perturbative models. We should also note that in the range of high ionization explored here the values of Z_{eff} are systematically smaller than those of the real charge $\overline{q}$. This is because the nonlinear effects in this range are strong enough to reduce the stopping values with respect to those expected in a perturbative picture (*saturation effect*) [28]. But this relation ($Z_{eff} < \overline{q}$) may be reversed when the charge $\overline{q}$ is small, like at low energies [18]. According to our calculations, in the high-energy regime the condition $Z_{eff} > \overline{q}$ may only be fulfilled for weakly ionized projectiles (i.e, out of charge equilibrium).

To illustrate the different ion-charge models currently used in the literature we compare in Fig. 4 the fitting values q_{ND} and q_{SG}, representing the equilibrium ionization of ions emerging from solid foils [29,30], together with the effective-charge fitting, Z_{Betz}, and with the q values assumed in various computer programs, such as the TRIM and SRIM programs [23], the GEANT program [24], and the binary theory [21]. As explained before, our nonlinear stopping calculations are consistent with the equilibrium ionization values given by the q_{ND} and q_{SG} fittings, which we consider to represent in a better way the charge state of ions moving inside the solid.

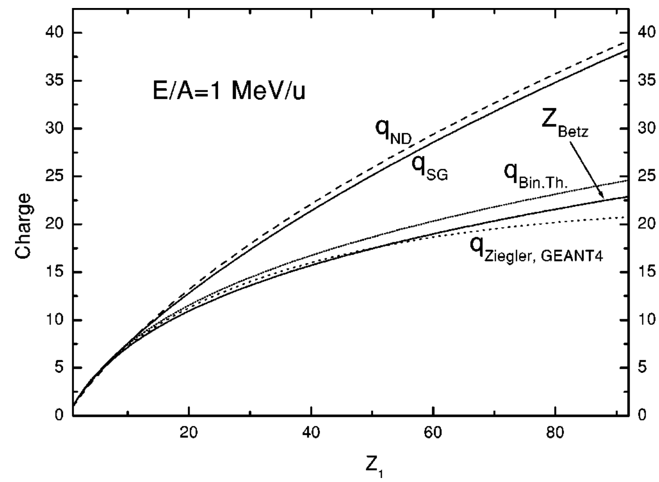

FIG. 4. Different ion-charge models currently used in the literature: fitting values q_{ND} and q_{SG} [29,30] representing the equilibrium ionization of ions emerging from solid foils, effective-charge fitting, $Z_{Betz} = Z_1[1 - \exp(-0.92v/Z_1^{2/3})]$; q values used in various computer programs: TRIM, SRIM [23] and GEANT program [24], and q values used in the Binary Theory [21].

Finally, we can extract from this study a further conclusion on the seemingly controversial question of the postulated differences between the charge states inside or outside solids for emerging ions [3]. In earlier analyses the charge state inside the solid was assumed to be significantly smaller than that of the emerging ions (and similar to Z_{eff}), and in order to justify this difference it was proposed [5] that a significant number of excited electrons (attached to the ion) would be emitted in the form of Auger electrons after the ion leaves the solid. In contrast with this, our study provides strong evidence that the charge state inside the solid should be quite close to the one observed after emergence into vacuum. Therefore, no significant electron emission through this process should be expected. We think this explains why in all the experiments designed to detect these electrons [8–10] the yield actually measured was always much less than the values predicted by the BG model.

IV. SUMMARY AND CONCLUSIONS

In summary, by performing extensive nonlinear stopping power calculations we have demonstrated the consistency between the charge state of ions in solids and the empirical effective charge values. The seemingly paradoxical discrepancies between both magnitudes pointed out over the years, is only a consequence of performing the analyses of experimental stopping values within the framework of the Z^2 scaling predicted by perturbation theories, and by the assumption that the Z_{eff} values so extracted represent to a first approximation the charge state of the ions.

The more exact nonlinear representation of the stopping phenomena applied here is fully consistent with the view of the emerging ion charge as a realistic approximation to the value of the ion charge within the solid ($\overline{q} \cong \overline{q}_{exit}$). The present analysis does not exclude the possible influence of charge exchange effects at the exit surface, although as a rather minor effect and without much influence in the case of swift heavy ions (we estimate that this effect may be at most of the order of a few units of charge according to the variation of the curves shown in Fig. 2). But this analysis excludes the possibility of large Auger electron emission processes in the range of values that would be required by the BG model, and is therefore in accord with the negative results that several experiments have produced with respect to this prediction.

ACKNOWLEDGMENTS

The authors are grateful to P. L. Grande, P. M. Echenique, and L. Toburen for stimulating comments. Critical comments by P. Sigmund are also acknowledged. This work was partially supported by ANPCYT of Argentina, Project No. PICT 0303579.

[1] N. Bohr, K. Dan. Vidensk. Selsk. Mat. Fys. Medd. **18**, 8 (1948).
[2] L.C. Northcliffe, Annu. Rev. Nucl. Sci. **13**, 67 (1963).
[3] H.D. Betz, Rev. Mod. Phys. **44**, 465 (1972).
[4] N. Bohr and J. Lindhard, K. Dan. Vidensk. Selsk. Mat. Fys. Medd. **28**, 7 (1954).
[5] H.D. Betz and L. Grodzins, Phys. Rev. Lett. **25**, 903 (1970).
[6] B.S. Yarlagadda, J. E Robinson, and W. Brandt, Phys. Rev. B **17**, 3473 (1978).
[7] J. Ziegler, J.P. Biersack, and U. Littmark, *The Stopping and Range of Ions in Solids* (Pergamon, New York, 1985).
[8] R.A. Baragiola, P. Ziem, and N. Stolterfoht, J. Phys. B **9**, L447 (1976).
[9] H.D. Betz, R. Schramm, and W. Oswald, in *Interaction of Charged Particles with Solids and Surfaces*, edited by A. Gras-Martí, H.M. Urbassek, N.R. Arista, and F. Flores (Plenum, New York, 1991).
[10] R. Schramm and H.D. Betz, Nucl. Instrum. Methods Phys. Res. B **69**, 123 (1992).
[11] P. Sigmund, Phys. Rev. A **56**, 3781 (1997).
[12] P. Sigmund and A. Schinner, Nucl. Instrum. Methods Phys. Res. B **174**, 535 (2001).
[13] J.H. Ormrod and H.E. Duckworth, Can. J. Phys. **41**, 1424 (1963); J.H. Ormrod, J.R. Macdonald, and H.E. Duckworth, *ibid.* **43**, 275 (1965).
[14] B. Fastrup, P. Hvelplund, and C.A. Sautter, K. Dan. Vidensk. Selsk. Mat. Fys. Medd. **35**, 10 (1966); P. Hvelplund and B. Fastrup, Phys. Rev. **165**, 408 (1968).
[15] W.N. Lennard, H. Geissel, D.P. Jackson, and D. Phillips, Nucl. Instrum. Methods Phys. Res. B **13**, 127 (1986).
[16] W. Brandt and M. Kitagawa, Phys. Rev. B **25**, 5631 (1982).
[17] J.S. Briggs and A.P. Pathak, J. Phys. C **6**, L153 (1973); **7**, 1929 (1974).
[18] P.M. Echenique, R.M. Nieminen, J.C. Ashley, and R.H. Ritchie, Phys. Rev. A **33**, 897 (1986); P.M. Echenique, F. Flores, and R.H. Ritchie, Solid State Phys. **43**, 229 (1990).
[19] G. Maynard, M. Chabot, and D. Gardes, Nucl. Instrum. Methods Phys. Res. B **164-165**, 139 (2000).
[20] P. Grande and G. Schiwietz, Nucl. Instrum. Methods Phys. Res. B **195**, 55 (2002).
[21] P. Sigmund and A. Schinner, Nucl. Instrum. Methods Phys. Res. B **195**, 166 (2002).
[22] L.H. Toburen, Report No. IAEA-TECDOC-799, 1995 (unpublished).
[23] J.F. Ziegler, TRIM-92, http://www.research.ibm.com/ion-beam (1992); SRIM-2000, http://www.research.ibm.com/ion-beam (2001)
[24] S. Agostinelli *et al.*, Nucl. Instrum. Methods Phys. Res. A **506**, 250 (2003); see, http://geant4.web.cern.ch/geant4
[25] A.F. Lifschitz and N.R. Arista, Phys. Rev. A **57**, 200 (1998).
[26] A.F. Lifschitz and N.R. Arista, Phys. Rev. A **58**, 2168 (1999).
[27] N.R. Arista and A.F. Lifschitz, Phys. Rev. A **59**, 2719 (1999).
[28] N.R. Arista, Nucl. Instrum. Methods Phys. Res. B **195**, 91 (2002); **207**, 232(E) (2003).
[29] V.S. Nikolaev and I.S. Dmitriev, Sov. Phys. JETP **20**, 409 (1965); Phys. Lett. A **28**, 277 (1968).
[30] G. Schiwietz and P.L. Grande, Nucl. Instrum. Methods Phys. Res. B **175-177**, 125 (2001).
[31] K. Shima, T. Mikuno, and H. Tawara, At. Data Nucl. Data Tables **34**, 357 (1986); K. Shima, N. Kuno, M. Yamanouchi, and H. Tawara, *ibid.* **51**, 173 (1992).
[32] G. Konac, Ch. Klatt, and S. Kalbitzer, Nucl. Instrum. Methods Phys. Res. B **146**, 106 (1998).
[33] F. Hubert, R. Bimbot, and H. Gauvin, At. Data Nucl. Data Tables **46**, 1 (1990).