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Mean excitation energy for the stopping power of metallic aluminum: Comments on an article by McGuire

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McGuire [Phys. Rev. A **28**, 53 (1983)] has suggested that the mean excitation energy I for the stopping power of metallic aluminum should be "in the range of 145–150 eV," as opposed to the currently recommended value of ~ 166 eV. We have reexamined all pertinent information from two independent sources, i.e., measurements of proton stopping power and the semiempirical determination of the dielectric-response function over the entire spectral range. Our conclusion is that the value of 166 eV is essentially correct. Moreover, we point out a number of deficiencies in McGuire's arguments; some of our remarks concern the values of inner-shell corrections to the Bethe stopping-power formula as evaluated by McGuire [Phys. Rev. A **28**, 49 (1983)].

I. INTRODUCTION

A recent article[1] by McGuire advances the suggestion that the mean excitation energy I in the Bethe stopping-power formula[2] for metallic aluminum should be "in the range of 145–150 eV," rather than the widely used value[3,4] of 166 eV. The I value is the crucial attribute of any material for determining its stopping power for charged particles having a wide range of energies, and metallic aluminum is often used as a reference material in measurements of stopping power, particle ranges, and related quantities. Therefore, a revision of the I value as suggested by McGuire would have far-reaching consequences, both in our basic understanding of stopping power and in many applications such as radiation dosimetry. We have examined the logical basis of McGuire's suggestion and reconsidered all pertinent information. In conclusion, we must disagree with McGuire, and argue that the weight of evidence supports the currently recommended value of 166 eV. In what follows, we shall present several points to show deficiencies in McGuire's arguments.

First, in the discussion of stopping-power and range measurements, McGuire puts unwarranted emphasis on a single set[5] of data, while other sets[6–12] of data must be considered with at least the same weight. Indeed, as early as 1964 there were several sets of data, as critically reviewed by Bichsel[13] (who then concluded $I = 163$ eV). McGuire ignored this material and its implications, as we shall further discuss in Sec. II.

Second, in McGuire's data analysis, there is an elementary error (which he has acknowledged in a letter to one of us, H.B.), i.e., in the evaluation of the Bethe formula, he used the atomic mass unit (931 MeV) in place of the proton rest mass (938.26 MeV). To correct for this error, all the I values derived in his article must be increased by about 5 eV; therefore, McGuire's suggestion should really have been a range of 150–155 eV, to be compared with the currently recommended value of 166 eV. This difference in the I value of about 10% leads to a difference in the stopping power of about 2% for particles with energies of 5–100 MeV. This is the size of the issue in question, and is indeed appreciable in comparison with the commonly claimed accuracy of stopping-power measurements.

Third, the determination of the I value from stopping-power data requires knowledge of the shell corrections,[1] viz., the difference between the Bethe asymptotic formula and the precise sum of the cross sections (within the first Born approximation) multiplied by the corresponding energy transfers. There are two earlier lines of work on the evaluation of the shell corrections; one is based on the hydrogenic approximation[14,15] to the form factor and the other uses the free-electron model.[16] The values of the shell corrections resulting from these two radically different descriptions of the electronic structure are quite close in the energy region of interest for the lighter elements such as aluminum. This suggests that the shell corrections are probably insensitive to details of the electronic structure beyond the shell binding energies; indeed, the dominant contributions arise from the K shell, whose properties are much simpler than those of outer shells and insensitive to chemical binding and other valence-shell phenomena. In addition, Sabin and Oddershede[17] applied the Sigmund method[18] of relating energy losses to stationary targets with energy losses to moving targets, and thus estimated the shell corrections. The resulting

values of the total shell corrections for aluminum are close to those by Walske[14,15] and by Bonderup.[16]

Further, earlier studies as reviewed in Refs. 3 and 4 provide a guide to the systematics of the shell correction; in particular, they show a generally smooth variation of shell corrections with atomic number and with particle energy. In contrast, McGuire's[19] evaluation of the stopping powers, through numerical summation of the cross sections multiplied by energy transfers, yields shell corrections which do not vary smoothly with energy. Although his method is correct in principle, there are practical difficulties to be overcome in this formidable task, especially in ensuring the numerical precision of results. As we shall show in Sec. II, McGuire's results leave much room for question.

Finally, McGuire[1] raised doubts about the evaluation[20] of the I value from the dielectric-response function. This evaluation is quite insensitive to minor modifications of the data for the dielectric-response function, provided the data satisfy a few global criteria, e.g., the f sum rule and realistic partition of the oscillator strength among different shells. As a consequence, the resulting I value of about 166 eV is definitive within a few eV. In Sec. III, we shall show that McGuire's doubts about the direct evaluation are unfounded.

II. ANALYSIS OF STOPPING-POWER AND RANGE DATA

We will primarily consider data for protons in the energy range $5 \leq E/\text{MeV} \leq 100$, for which the I value is the dominant parameter. Three methods have been used for energy-loss measurements: (a) determination of total ranges,[6-8] (b) calorimetric measurement of deposited energy,[5,11] and (c) measurement of residual particle energy with Si detectors.[9,10,12] The uncertainty stated by the authors for these data is 1% or less. (We do not discuss other data having larger uncertainties; they have already been considered in Ref. 13.)

From the direct measurements of stopping powers, the quantity

$$X = \ln\left(\frac{2m_e c^2\beta^2}{1-\beta^2}\right) - \beta^2 - B_{\text{ex}} \tag{1}$$

is calculated where

$$B_{\text{ex}} = \frac{\beta^2 Z}{4\pi m_e c^2 r_0^2 N_0 A} S_{\text{ex}} \ , \tag{2}$$

and S_{ex} is the experimentally measured stopping power. Further, $m_e = 511\,004$ eV$/c^2$ is the electron rest mass, βc is the particle speed, $r_0 = e^2/(m_e c^2) = 2.817\,939 \times 10^{-13}$ cm is the classical electron radius, $N_0 = 6.022\,169 \times 10^{23}$/mole is Avogadro's number, Z is the atomic number, A is the atomic weight, and $4\pi m_e c^2 r_0^2 N_0 = 0.307\,08$ MeV cm^2/g. Values of X are given in Fig. 1.

From Fig. 1 we see that three sets[9,11,12] of experimental data agree closely with each other, while the fourth[5] lies about 1% below this grouping. Although this 1% difference is of the order of twice the standard deviations quoted by the authors of Ref. 5, it should be noted that the data of Ref. 11, obtained with the same method, agree with the data of Refs. 9 and 12. We therefore feel that the data of Ref. 11 carry more weight than the data of Ref. 5. In this observation, we differ significantly from McGuire.

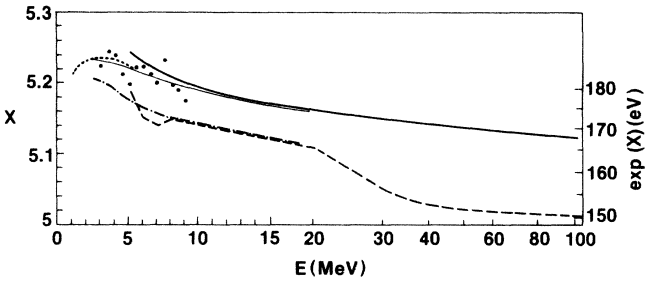

FIG. 1. Experimental data on the stopping power of metallic aluminum for protons. The abscissa represents the proton energy E in MeV. Note that the scale is linear up to $E = 15$ MeV, and is logarithmic thereafter. The ordinate represents X defined in Eq. (1), so long as we use experimental values S_{ex}. The thin solid curve shows the data of Tschalär (Refs. 9 and 10). The dotted curve shows the data of Andersen *et al.* (Ref. 11). The dots show the data of Ishiwari *et al.* (Ref. 12). The dot-dash curve shows the data of Sørensen and Andersen (Ref. 5). For comparison, the figure also includes two plots of the theoretical expression $X_{\text{th}} = \ln I + C(\beta)/Z$. The heavy solid curve shows X_{th} calculated from $I = 166$ eV and from $C(\beta)/Z$ taken from Fig. 3.5 of the ICRU report (Ref. 4). The dashed curve shows X_{th} calculated from $I = 150$ eV and from Table III of McGuire (Ref. 19). The right-hand scale on the ordinate, $\exp(X)$, indicates an effective I value including shell corrections. Note that X_{th} above defined excludes the Barkas (z^3) term and the Bloch (z^4) term. Incorporation of the Barkas term would reduce X_{th} by 0.02 at 6.5 MeV, and by 0.006 at 20 MeV.

Our feeling is also shared by Andersen (a co-author of Refs. 5 and 11), as expressed in a letter to one of us (M.I.). Specifically, Andersen holds it probable that, in the work of Ref. 5, there were a non-negligible number of slit-scattered particles in the beam, causing the stopping-power values to appear too high. In the subsequent study of Ref. 11, greater attention was given to energy calibration and beam quantity, so that this problem did not arise.

It is appropriate to introduce what we consider as the best estimate of the stopping power based on information independent of energy-loss measurements. For the I value we choose 166 eV from experimental dipole oscillator-strength data,[20] as further discussed in Sec. III. To obtain the stopping power we also need values of the shell corrections C/Z. Essentially three sets are available: (a) Walske[14,15] and Bichsel,[3,4] (b) Bonderup,[16] and (c) McGuire.[19] These sets are compared in Fig. 2. While the differences between the Bonderup and the Walske-Bichsel values are negligible for the present purpose, the McGuire values are more than twice as large over the range from 8–20 MeV as the others, and are much smaller for 60–100 MeV. Moreover, the strong fluctuation around 8 MeV indicates problems in the numerical accuracy in the values of the input data used. Since the calculated shell corrections are the difference between two large numbers, we believe that the uncertainty of McGuire's C/Z must be considerable. This belief is further confirmed by the fact that, for $E > 30$ MeV, McGuire's values (as given in Table III of Ref. 19) are roughly proportional to E^{-3}, while general theory[2] indicates C/Z to be proportional to E^{-1}. The shell-correction values by Sabin and Oddershede[17] are similar to other theoretical data and also indicate the E^{-1} dependence at high energies. In conclusion, we use the shell-correction values[4] of Walske (and as modified by Bichsel) in

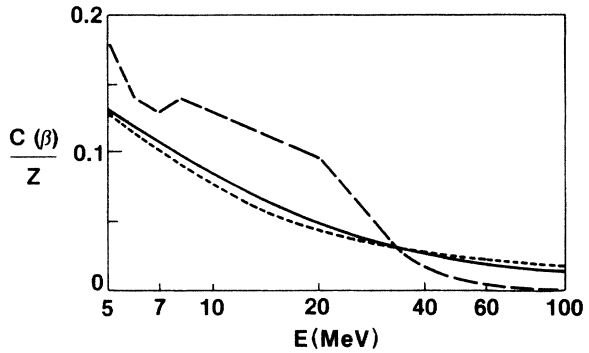

FIG. 2. Total shell corrections $C(\beta)/Z$ plotted against proton energy E. The solid curve represents values given by the ICRU report (Ref. 4) (following the work by Walske and Bichsel). The dotted curve represents values of Bonderup (Ref. 16). The broken curve represents values of McGuire (Ref. 19). For high speeds $\beta c, C(\beta)/Z$ should tend to be proportional to E^{-1}, according to the general theory (Ref. 2). The irregular behavior of the dashed curve, as well as its rapid decline at $E > 20$ MeV, suggest numerical inaccuracies in the evaluation by McGuire (Ref. 19).

arriving at the best estimate, as shown in Fig. 1.

We have omitted the Barkas (z^3) and Bloch (z^4) terms[4] in our consideration, where ze is the charge on the incident ion. They would amount to -0.021 and 0.005, respectively, in X at 6.5 MeV. Their inclusion would increase the I values derived from experimental data by approximately 2 eV.

Figure 1 permits a comparison of the experimental and calculated X values. It is clear that our best estimate, $I \sim 166$ eV, is consistent with the majority of the experimental data,[9-12] while McGuire's values using $I = 150$ eV are compatible only with the data of Ref. 5. (For readers interested in details we may note that the difference between the experimental data of Ref. 5 and those of Ref. 11 leads to a difference in the I value of about 8 eV. The difference in using different shell-correction values accounts for the additional 8 eV difference in the I value.)

For comparison with the range data we have evaluated ranges in the continuous-slowing-down approximation for a series of energies in two ways: one using $I = 166$ eV, and shell corrections from Walske[14,15] and Bichsel,[3,4] and the other using $I = 150$ eV (i.e., the lowest value according to McGuire's suggestion) and total shell corrections given by McGuire.[19] The result is shown in Table I. The average deviation between experimental data and the theory with $I = 166$ eV is $+0.11\%$, while it amounts to -1.2% for $I = 150$ eV. The standard deviations are 1.02% and 1.60%, respectively.

A remark about the accuracy of the data of Ref. 8 is in order. Portner and Moore give no information about the homogeneity of their aluminum absorber. They measured only the average areal density of the 120 circular sheets (0.3 mm thick) they used as the range absorber. The proton beam passed near the edge of these sheets. It is thus possible that the relatively large differences δ between the experimental range R_{ex} and the theoretical range R_{th} in the last two lines of our Table I are due to absorber inhomogeneity.

Some remarks must be made about the range measurements quoted by McGuire in his analysis. The value $I = 150$ eV derived by Wilson[21] is obsolete for the following reason: The differences in straggling and multiple scattering

TABLE I. Proton ranges in aluminum. The experimental values R_{ex} are taken from Bichsel and Uehling (Ref. 7) for $E < 20$ MeV, from Bloembergen and van Heerden (Ref. 6) [corrected for multiple scattering (Refs. 3 and 13)] for $34 < E < 76$ MeV, and from Portner and Moore (Ref. 8) for 100 MeV. The theoretical range R_{th} in the continuous-slowing-down approximation (CSDA) has been calculated from the best estimate of the stopping power by the use of Eqs. (8d)–(14) of Ref. 3. The CSDA range R_M has been similarly calculated from the stopping-power values according to McGuire (Ref. 19) (as represented by the dashed curve in Fig. 1). For the calculations in either case, we have chosen the value of 0.200 g/cm^2 for the range of the 11-MeV proton. In this table, δ represents deviations in percent of theoretical ranges from experimental ranges.

E (MeV)	R_{ex} (g/cm^2)	R_{th} (g/cm^2)	δ (%)	R_M (g/cm^2)	δ (%)
11.82	0.227 12	0.227 11	−0.01	0.227 11	0.01
14.971	0.343 76	0.344 21	0.13	0.343 02	−0.22
17.836	0.468 44	0.468 97	0.11	0.466 56	−0.40
34.96	1.547	1.548 67	0.11	1.531 48	−1.00
37.16	1.757	1.726 47	−1.74	1.706 27	−2.89
39.66	1.928	1.938 72	0.56	1.914 82	−0.68
42.57	2.198	2.199 27	0.06	2.170 73	−1.24
44.86	2.402	2.413 39	0.52	2.382 03	−0.83
47.67	2.634	2.690 25	2.14	2.653 01	0.72
52.08	3.189	3.149 18	−1.25	3.103 82	−2.67
52.33	3.188	3.176 15	−0.37	3.130 32	−1.81
56.68	3.687	3.661 05	−0.70	3.606 76	−2.18
56.96	3.784	3.693 25	−2.40	3.638 40	−3.85
61.79	4.258	4.268 36	0.24	4.203 63	−1.28
62.10	4.280	4.306 48	0.62	4.241 09	−0.91
65.78	4.756	4.770 24	0.30	4.696 98	−1.24
66.10	4.786	4.811 51	0.53	4.737 55	−1.01
72.94	5.773	5.730 11	−0.74	5.640 74	−2.29
73.05	5.714	5.745 44	0.55	5.655 81	−1.02
75.70	6.141	6.120 04	−0.34	6.024 18	−1.90
75.84	6.078	6.140 11	1.02	6.043 92	−0.56
99.58	9.768	9.933 30	1.69	9.775 68	0.08
99.88	9.840	9.986 04	1.48	9.827 59	−0.13

for air and aluminum (amounting to about 0.5% in range) were not considered in the analysis performed by Wilson. Moreover, the I value of aluminum was derived on the basis of an I value for air of 80.5 eV. If the currently adopted value[4] of 85.7 eV is used in Wilson's expression, the I value for aluminum would be 162 eV. Further changes, such as the inclusion of z^3 and z^4 effects should also be made. Wilson's experiments are accurate enough to deserve a full reanalysis.

The two I values derived by Mather and Segrè[22] (quoted by McGuire[1] in support of his case) are probably too low because no correction was made to the measured range for nuclear interactions of the 340-MeV protons, as has been pointed out in Refs. 13 and 23.

Finally, a note about the Bichsel and Uehling[7] data may be made. The analysis was made without the inclusion of the z^3 and z^4 effects. The inclusion of these effects will increase the value of I, which has been obtained without them, especially at small energies. Furthermore, range straggling had been assumed to be Gaussian. This is only approximately correct. With the use of accurate straggling

functions, the path lengths given in Table VIII of Ref. 7 would be slightly different, but the change is insignificant for the conclusions of the present paper.

III. EVALUATION OF THE MEAN EXCITATION ENERGY FROM THE DIELECTRIC-RESPONSE FUNCTION

An independent estimate of I may be obtained from measurements of the dielectric function $\epsilon(\omega)$ using the definition[2]

$$\ln I = \frac{\int_0^\infty \omega \,\mathrm{Im}[\epsilon^{-1}(\omega)] \ln(\hbar\omega)\, d\omega}{\int_0^\infty \omega \,\mathrm{Im}[\epsilon^{-1}(\omega)]\, d\omega} \quad . \tag{3}$$

Equation (3) requires accurate knowledge of $\epsilon(\omega)$ over virtually the entire range of electronic excitations. For aluminum, these data are available from the comprehensive analysis of optical, x-ray, and electron-energy-loss measurements by Shiles *et al.*[20] The resulting composite values[24,25] of $\epsilon(\omega)$ yield $I = 166$ eV with an uncertainty of a few eV.

McGuire[1] pointed out two possible sources of error in the analysis of Shiles *et al.*:[20] (1) uncertainty in a correction of experimental x-ray absorption cross sections, made to compensate for systematic experimental errors brought to light by violations of the f sum rule, and (2) errors arising from oxygen contamination. He suggested that together these led to an excess strength of ~ 0.6 e/atom below the $L_{II,III}$ edge (~ 72.7 eV) and a corresponding deficiency in strength between the $L_{II,III}$ and K edges. Specifically, McGuire suggested a total oscillator strength of ~ 2.4 e/atom below the $L_{II,III}$ edge as opposed to the value 3.1 e/atom, given by Shiles *et al.*[20] This change, however, would *increase,* not decrease, I, since it moves the center of gravity of the spectrum toward higher energy. An estimate using McGuire's proposed redistribution yields I values between 174 and 179 eV, over 20 eV higher than his analysis favors.

Although this eliminates McGuire's specific proposal, it is valuable to consider the sources of error he suggested. To assess possible uncertainties in I introduced by the correction presented by Shiles *et al.*[20] for excess L-shell absorption present in thin-film samples, we note that significant surface effects, possibly associated with contamination, were observed near the L edge by Tomboulian and Pell[26] and by Balzarotti *et al.*[27] Moreover, the corrected data are in good agreement with the analyses of Henke and Elgin[28] and Henke *et al.*,[29] which depended more heavily on measurements at shorter wavelengths, at which thicker samples could be used. Even more to the point, the average excitation energy lies just above the $L_{II,III}$ edge, so that the value of I is insensitive to a rescaling of the thin-film absorption from the L edge to several hundred eV by the very nature of the average. Generally, the correction made to satisfy the f sum rule changed I by less than 1 eV.[30]

Significant effects of oxygen contamination on the optical absorption strength below the $L_{II,III}$ edge are unlikely: Over 93% of this absorption lies below 11.75 eV (see Fig. 9 of Ref. 20), and in this range, optical measurements[31,32] were made on evaporated films prepared in ultrahigh vacuum (uhv) from high-purity starting material, and maintained in an oxygen-free atmosphere during measurement. An independent check is available from the uhv ellipsometric measurements of Mathewson and Myers;[33] agreement with Shiles *et al.*[20] is excellent.

Above 11.75 eV, the dielectric-response function was based on electron-energy-loss or optical measurements[34] made in air on samples prepared by high-vacuum evaporation. These samples are contaminated,[35] but the total absorption strength from 11.75 eV to the $L_{II,III}$ edge is only 6% of the total conduction electron absorption (0.2 vs 3.1 e/atom). Thus, even massive contamination errors at these wavelengths would have far less effect than suggested by McGuire.[1]

A third possible source of error not suggested by McGuire is uncertainty in extrapolating[36] optical data to wavelengths longer than 32 μm. The oscillator strength associated with this extrapolation is ~ 0.7 e/atom. In a subsequent study,[37] the low-energy portion of the spectrum was studied to separate intraband and interband processes. It was found that uncertainties in the models employed correspond to an uncertainty in the oscillator strength in the extrapolated region of only -0.02 to $+0.10$ e/atom.

In summary, aside from gross errors in the optical experiments or their analysis, it seems very unlikely that the f sum up to the $L_{II,III}$ edge could differ by more than ± 0.2 e/atom from the value of 3.1, given by Shiles *et al.*[20] as a result of oxide contamination or of extrapolation errors. This is nowhere near the value of 0.6 e/atom suggested by McGuire.

Finally, we note that McGuire's original concerns were based on his observations[1] that (1) both the $3s$ and $3p$ photoionization cross sections for *atomic* aluminum have minima between 10 and 50 eV, and (2) that "20% of the atomic $3s$ and $3p$ oscillator strength (0.62 of 3.20 e/atom) is at energies higher than the aluminum L edge." However, the analysis of Shiles *et al.*[20] is for *metallic* aluminum, which has a profoundly different absorption below the $L_{II,III}$ edge. In going from atoms to the metal, the discrete atomic spectrum goes over into a continuum with absorption extending to zero energy. For the metal there is no experimental or theoretical evidence for a minimum in the absorption between 1.5 and 72 eV, only a monotonic decrease.

Specifically, the absorption spectrum of the metal is to a good approximation the superposition of intraband and interband terms.[38] The first of these dominates below a few tenths of eV and is well represented[37] by the Drude model[39] for which the absorption decreases monotonically with increasing frequency. The intraband oscillator strength may be estimated from band-structure calculations[40] which yield an optical mass[41] of $m_{\mathrm{opt}}/m_e = 1.45$, corresponding to an intraband strength of 2.07 e/atom for aluminum's three conduction electrons. This theoretical estimate is in good agreement with experimental intraband oscillator strength derived from optical data, which range[37] from 1.88 to 2.0 e/atom. For typical values of the Drude parameters,[24,25,37] all but 1% of this strength is exhausted by 4 eV.

The interband term consists of two strong absorptions[38,40,42] at about 0.5 and 1.5 eV. These have long high-energy tails that go over into a nearly-free-electron absorption spectrum,[43] which also decreases monotonically. Recent calculations[44] of the interband spectrum yield a theoretical strength of 0.97 e/atom between 0 to 7 eV compared to an experimental value of ~ 0.9 e/atom derived from Shiles *et al.*[20]

Combining the theoretical intraband and interband strength yields a total strength for energies between 0 and 7 eV of about 3 e/atom for metallic aluminum. The analysis

of experimental data by Shiles *et al.*[20] yields 2.8 *e*/atom. Again, the agreement of the best available theoretical estimates and experiment as analyzed on the basis of the distribution of oscillator strength presented by Shiles *et al.* is excellent and supports an absorption strength in excess of 3 *e*/atom below the $L_{II,III}$ edge in contrast to the value of 2.4 *e*/atom suggested by McGuire.

IV. CONCLUDING REMARKS

Although we are certain about the essential correctness of the I value of 166 eV, it would be desirable to have further stopping-power measurements for proton energies above 10 MeV. At these energies, the contributions from the z^3 and higher-order terms are inappreciable and the asymptotic form of the shell corrections can be applied, so that the analysis of data with the Bethe formula is straightforward and the result is clear-cut. It will be important to have measurements cover a series of proton energies so that the trend of variation with energy is clearly observed. For energies above 250 MeV, the Fermi density effect[45] will have to be taken into account for analysis of data on aluminum.

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