Stopping Power of Zr, Gd, and Ta for 5–12-MeV Protons and Deuterons. Further Evidence of an Oscillatory Behavior of the Excitation Potential

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The stopping power of Zr, Cd, and Ta for 5–12-MeV protons and deuterons was measured. The accuracy was 0.3% for Zr and Ta and 0.6% for Gd. The results are compared with Risö data published earlier, and it is shown that the ratio between the mean excitation potential and the atomic number cannot be a monotonically decreasing function of the atomic number. An oscillatory dependence is suggested.

INTRODUCTION

SOME years ago, Burkig and MacKenzie[1] and Nakano, MacKenzie, and Bichsel[2] published measurements of the proton stopping powers of several materials relative to aluminum. Regarded as a function of the atomic number Z_2 of the target material, these results show a smooth and continuous variation, but Burkig and MacKenzie found that the stopping powers of Ca, Ti, V, and Th were higher than indicated by the general behavior of the other elements. For the cases of Ti and V, this was verified by Nakano *et al.*

In a recent publication,[3] which was devoted to an analysis of the theoretical and experimental situation within the field of stopping power, the implications of the above-mentioned data were not really taken into account, the general feeling being that more data were needed for an understanding.[4,5] A smooth variation of the mean excitation potential I with the atomic number Z_2 was suggested,[6] which implies that the Bloch constant I_0, which is the ratio between I and Z_2, decreases monotonically with increasing Z_2.

Recently, stopping-power data for 5–12-MeV protons and deuterons of the elements from $Z_2=20$ to $Z_2=30$ have been published.[7] An analysis shows that I_0 increases with increasing Z_2 for these elements,[8] in agreement with the data of Burkig and MacKenzie and Nakano *et al.*

The new data presented here, together with earlier published data for Ag, Pt, and Au,[9] show that this increase takes place also for higher Z_2 values. In fact, I_0 seems to oscillate.

EXPERIMENTAL DATA AND RESULTS

The measurements were made by the low-temperature calorimetric method, which has been described earlier.[10] An accuracy of 0.3% is normally obtainable by this method. For materials for which the determination of target thickness is less accurate, a somewhat smaller accuracy must be accepted.

Measurements were made with both protons and deuterons, and the energy of the incoming particle was varied from 5 to 12 MeV in steps of 0.5 MeV. Since protons and deuterons with equal velocities are equivalent with respect to stopping power according to theory, the energy range of the deuterons is equivalent to that of 2.5–6.0-MeV protons. For each metal at least two different runs were made, a new foil being used in each run.

The data for the foils used are as follows:

Zirconium. Thickness was ~17.2 and ~16.8 mg/cm². Correction for thermal expansion was 0.26%. Purity was 99.5% (reactor grade). Analysis for O_2 and H_2 was made. The contents were small and would only give rise to corrections less than 0.05%. Supplier was A. D. Mackay Inc.

Gadolinium. Thickness was ~14.0 and ~16.9 mg/cm². Correction for thermal expansion was 0.11%. Purity was 99.9%. Analysis for oxygen content was made by Gulf General Atomic, Inc., the result being 0.44% oxygen by weight for each foil. Because of this, the measured energy losses were approximately 0.5% too high, and corrections for this were made. Supplier was Koch-Light Laboratories Ltd. Gadolinium is difficult to roll into thin foils, and considerable thickness gradients were found. This gives a larger uncertainty in the thickness determination.

Tantalum. Thickness was ~10.8, ~18.9, and ~23.6 mg/cm². Correction for thermal expansion was 0.29%. Purity was 99.9%. Supplier was A. D. Mackay Inc.

All the above-mentioned foils were attached to the target frame with Narmco resin.

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At energies of interest to us here, the stopping power is given by

$$-\frac{dE}{dx}=\frac{4\pi e^4 Z_1^2 N_0 Z_2}{mv^2 A}\left(\ln\frac{2mv^2}{I}-\frac{C}{Z_2}\right), \quad (1)$$

where m and $-e$ are the electron mass and charge, respectively, Z_1 and v are the charge and velocity of the incoming ion, N_0 is Avogadro's number, A and Z_2 are the atomic weight and number of the target material, I is the mean excitation potential, and C/Z_2 is the shell correction. As dE/dx varies rapidly with energy, the experimental results may be depicted by means of a reduced variable defined by

$$X=\frac{dE}{dx}\frac{mv^2 A}{4\pi e^4 Z_1^2 N_0 Z_2}+\ln 2mv^2. \quad (2)$$

From Eq. (1) this is seen to equal the sum $\ln I+C/Z_2$.

The experimental X values are shown in Fig. 1 as a function of the equivalent proton energy $(M_p/M)E$, where M_p is the proton mass and M is the mass of the particle. A smooth curve is drawn through the points in each case. The data for Gd are seen to be less accurate because of uncertainty in foil thickness determinations.

In Table I the stopping powers are given in keV/mg cm^{-2} for the energy range 2.25–12.0 MeV, the values being determined from the smooth curves of Fig. 1 by means of Eq. (1). The accuracy of the tables is 0.3% for Zr and Ta and 0.6% for Gd.

TABLE I. Smoothed values of measured stopping powers of Zr, Gd, and Ta for 2.25–12.00-MeV protons. The data have been obtained from the full curves of Fig. 1. Estimated accuracy ±0.3% for Zr and Ta, ±0.6% for Gd.

Energy (MeV)	Stopping power $-dE/dx$ (keV/mg cm^{-2}) Zr	Gd	Ta
2.250	66.56	51.65	46.20
2.500	62.44	48.58	43.62
2.750	58.83	45.87	41.35
3.000	55.70	43.51	39.35
3.250	52.92	41.42	37.59
3.500	50.42	39.57	36.02
3.750	48.20	37.91	34.60
4.000	46.20	36.42	33.32
4.250	44.40	35.06	32.15
4.500	42.74	33.83	31.07
4.750	41.23	32.69	30.07
5.000	39.84	31.65	29.15
5.250	38.55	30.69	28.29
5.500	37.36	29.80	27.49
5.750	36.25	28.96	26.75
6.000	35.22	28.18	26.06
6.500	33.35	26.77	24.79
7.000	31.69	25.50	23.66
7.500	30.21	24.37	22.65
8.000	28.89	23.36	21.73
8.500	27.69	22.43	20.89
9.000	26.60	21.59	20.13
9.500	25.60	20.82	19.42
10.000	24.69	20.11	18.77
10.500	23.85	19.46	18.17
11.000	23.07	18.85	17.62
11.500	22.35	18.29	17.10
12.000	21.68	17.77	16.62
Atomic weight	91.22	157.26	180.95

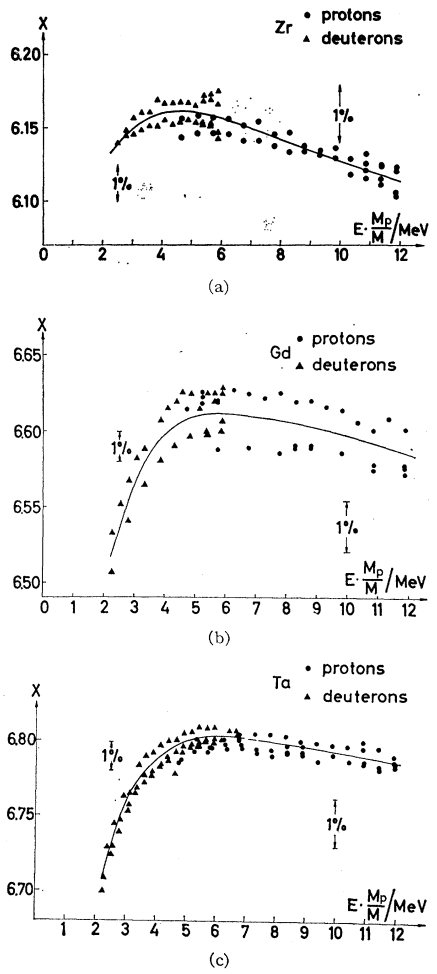

FIG. 1. Measured stopping powers of Zr, Gd, and Ta expressed by the reduced variable X defined by Eq. (2). The height of the arrows indicates the change in X caused by a 1% change in stopping power at the respective energies. M_p/M is the ratio between proton mass and mass of incident particle (see text).

DISCUSSION

In theoretical expression for the stopping power [Eq. (1)] the properties of the target material enter at two places. The factor Z_2/A gives the electron density of the material, while the quantities I and C/Z_2 describe the properties of these electrons. As was mentioned above, the stopping power data give directly an experimental expression for the sum $\ln I+C/Z_2$. The possibility of splitting this sum into its components was discussed in detail in an earlier publication.[8] Use was made of the fact that the mean excitation potential is energy-independent, and only the shell corrections C/Z_2 depend on energy. Within the limited interval $20\leq Z_2\leq 30$, the theoretical knowledge of C/Z_2 was

sufficiently good to serve as a starting point for a separation procedure. This is, however, not the case at higher Z_2 values.

The Z_2 dependence of I is known roughly to be described by the Bloch relation

$$I=I_0Z_2. \tag{3}$$

It is further known that I_0 decreases slowly with increasing Z_2, but that some elements have anomalously low values of I_0[1,2,8] For the interval $20\leq Z_2\leq 30$, we found that I_0 *increased* with *increasing* Z_2.

Since it is not possible to separate $\ln I$ from C/Z_2 for the elements treated here, we look at the expression

$$X_{\text{expt}}-\ln Z_2=\ln I_0+C/Z_2. \tag{4}$$

Here X_{expt} is a function of energy as seen in Fig. 1. We choose the energy 12 MeV which in all cases is well above the maximum in X_{expt} and at which X_{expt} varies slowly with energy. $X_{\text{expt}}-\ln Z_2$ as extracted from the data presented here and those published earlier[7,9,10] is shown in Fig. 2, where the periods of the periodic system are also indicated.

It is seen that there are regions where X-$\ln Z_2$ increases with increasing Z_2, each region being situated in the middle of a period. Or, to put it differently, the indisputable increase from $Z_2=20$ to $Z_2=30$ in period IV is repeated in the next two periods. There are no data to show the behavior on going from one period to the next, but a decrease with minima at the noble gases would probably be the most acceptable suggestion. This implies an oscillatory behavior of $X-\ln Z_2$.

It is seen from Fig. 2 that stopping-power data with an accuracy of 1% or better are needed to show the behavior sufficiently convincingly for higher Z_2 values. The majority of existing data are for elements that should have high values of $X-\ln Z_2$ according to Fig. 2, and we do not know about data for noble gases and neighboring elements of sufficient accuracy in our energy range.

Recently, Duc *et al.*[11] have reported energy-loss and range measurements for 54.4-MeV α particles in Cu, Ag, Au, and the two rare-earth metals Tb($Z_2=65$) and Tm($Z_2=69$). The accuracy of their range measurements is 1%. The data show that ranges in Tb and Tm are, respectively, 4.5 and 5.5% lower than those obtained by interpolation between ranges for Ag and Au found in the same experiment. This is tantamount to stopping powers higher than those which could be obtained by interpolation. The deviations are equivalent to deviations for $\ln I$ of 0.12 and 0.15. This is in agreement with Fig. 2 although the deviation for Tm is somewhat larger than we would expect.

On an earlier occasion[8] we found good agreement with the relative measurements of Burkig and MacKenzie[1] and Nakano *et al.*[2] within the Z_2 interval 20 to 30. For higher Z_2 values only Burkig and MacKenzie have sufficient data to make a comparison possible. Since their measurements were made relative to aluminum at 19.8 MeV, we need the value of dE/dx in aluminum at this energy to calculate X-$\ln Z_2$ for other elements from their data. This value has been taken from Bichsel,[12] and the results obtained are shown in Fig. 3.

For the elements $Z_2=20$ to $Z_2=30$, the behavior is similar to that shown by our data. The elements $Z_2=73$ to $Z_2=79$ again show the same behavior. The point for thorium ($Z_2=90$) is low lying as we would expect. For the elements $Z_2=41$ to $Z_2=50$, the agreement is apparently not good. A second look discloses that only the point for niobium ($Z_2=41$) is really disturbing. Burkig and MacKenzie made cross checks on some of their measurements, i.e., they measured the ratio between two elements directly and compared with the ratios obtained by the measurements relative to aluminum. For Nb a cross check was made between Nb and Rh($Z_2=45$). The experimental uncertainty for the latter element was, however, rather large because of foil inhomogeneity. With this in mind, the lack of agreement is not discomforting.

The oscillatory behavior shown in Fig. 2 is that of $\ln I_0+C/Z_2$. At 12 MeV, C/Z_2 amounts to 0.15 for Cu, 0.2 for Ag, and 0.3 for Au.[13] The increase of $\ln I_0+C/Z_2$ from Ca to Cu is 0.17, which is larger than the shell corrections for Cu. This is only possible with an increasing value of I_0. From Zr to Ag the increase is

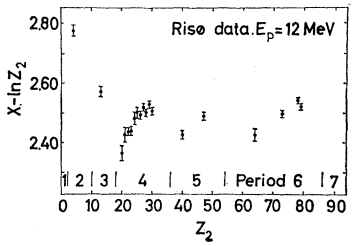

FIG. 2. X-$\ln Z_2$ versus Z_2 for Risö data at 12 MeV.

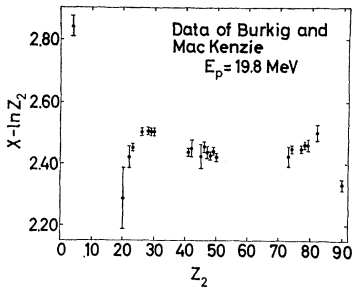

FIG. 3. X-$\ln Z_2$ versus Z_2 for the data of Burkig and McKenzie.

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0.06, and from Gd to Pt and Au it is 0.1. Furthermore, one should remember that the values for Zr and Gd may not be the lowest in these two periods. This may be due either to very large oscillations of the shell corrections or to relatively small oscillations for I_0 The latter is more acceptable and is in agreement with the behavior from Ca to Cu.

The elements with Z_2 below Ca have not been mentioned until now. First, we only have data for two elements here. Secondly, the number of elements in a period is small, and the variation from one element to the next may be great. Also, the mean excitation potential of these elements is strongly affected by chemical-binding effects, and no predictions can be made from the data presented here.

CONCLUSION

It is concluded that the Bloch constant I_0 is oscillating rather than being a monotonically decreasing function of Z_2. This conclusion is in reasonably good agreement with the data of Burkig and MacKenzie. The oscillations cannot be established too well yet, because sufficiently good data for elements at the minima and on the downward slopes do not exist.

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Positron Effective Mass in an Electron Gas

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A calculation is made of the positron effective mass in an electron gas at the sodium electron density, emphasizing terms which are nonlinear in the electron-positron interaction. We exploit a close similarity between the perturbation expansions for the lifetime and the self-energy. Both quantities are approximated by a t-matrix equation, using a two-parameter static effective interaction. The parameters are restricted to values which give agreement with the experimental annihilation rate. It is shown to be possible to obtain agreement with the experimental effective-mass value calculated from the thermal smearing of the 2γ angular correlation by Stewart and co-workers. However, if, in addition, we impose the screening requirement that the total displaced electron charge should exactly compensate the positron charge, we obtain effective-mass values that are too low. This result indicates that the observed thermal smearing of the angular-correlation curves cannot be explained from the effective mass of positrons in an electron gas.

1. INTRODUCTION

IT is generally believed, after the work of Stewart *et al.*[1] and Majumdar,[2] that one can obtain knowledge of the positron effective mass by measuring the thermal smearing in the angular-correlation curve of the annihilation radiation near the angle which corresponds to the c.m. momentum p_F of the annihilating pair. Experimental results based on this method[1] indicate a large effective mass for sodium ($m^*/m=1.8\pm0.2$) and similar values for the other alkali metals.

Several attempts have been made to explain these results theoretically. The band effective mass of the positron in sodium has been computed, among others, by Majumdar[3] and found to be very close to the bare mass. Hamann,[4] on the other hand, has studied the effective mass of a positron in an electron gas due to electron-positron correlations. He evaluated the positron self-energy to the lowest order in the dynamically screened interaction as a function of the electron density. His result for densities corresponding to sodium is again too small. Other authors have suggested that phonon effects,[5] or effects related to the formation of "quasibound" states,[6] might be responsible for the large effective mass. However, neither of these papers gives enough justification for the computational procedure to make one able to judge the validity of the above explanations.

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