

Stopping Power of the Elements $Z=20$ Through $Z=30$ for 5–12-MeV Protons and Deuterons

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The stopping power for 5–12-MeV protons and deuterons of Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, and Zn has been measured. For most of the materials, the accuracy is $\pm 0.3\%$. For Ca, Sc, Cr, and Mn, which were rather difficult to handle, the results are somewhat less accurate. The results have been compared with published experimental data and with semiempirical tables by Barkas and Berger, by Bichsel, by Janni, by Serre, and by Williamson *et al.* Most of these are in agreement with our results within their stated accuracies.

I. INTRODUCTION

THE interest in stopping-power data for all kinds of materials with respect to heavy charged particles such as protons and deuterons has been considerably increasing during recent years. Elementary-particle physicists and nuclear physicists need these data for the evaluation of particle tracks and for the control of their detectors. Health physicists need them for radiation protection purposes. The demand of these people has been met by several tables^{1–5} which are based on a semiempirical approach—partly by using known experimental results and partly by using the Bethe theory of stopping—and show various degrees of accuracy. These tables prove sufficient for materials where experimental data exist, but they are of less applicability for less known substances, because of the uncertainty in extrapolation.

On the other hand, there has been a growing interest by theorists in an increased accuracy of the measurements in order to be able to derive the basic parameters of the stopping-power theory from experiments. We felt, therefore, that a systematic measurement of the stopping power of a series of elements might throw some light upon a problem like the Z variation of these basic parameters.

The measurements were done using 5–12-MeV protons and deuterons at the tandem Van de Graaff accelerator of the Niels Bohr Institute, the University of Copenhagen.

Section II of this paper presents a short discussion of the experimental procedure and of the treatment the

different metals underwent. In Sec. III our results are presented both in diagrammatic and in tabulated form, and in Sec. IV these results are compared with existing data. An attempt to analyze the experimental data with the aim of finding the basic theoretical parameters is presented in a following paper.⁶

II. EXPERIMENTAL PROCEDURE

The method used is the one described previously in detail by Andersen⁷ and by Andersen *et al.*^{8,9} The principle of procedure is shown in Fig. 1. A beam of protons or deuterons passes through a target foil and is stopped in a metal block. Both of these are connected thermally to a liquid-helium reservoir through thermal resistances W_F and W_B . The energy dissipated by the particles causes heating of the foil and block systems giving temperature rises measured by the carbon thermometers R_F and R_B . After the beam is switched off, electric powers P_F and P_B are fed to heaters until the same temperature rises are obtained. The energy loss in the foil is thus

$$\Delta E = E_0 [P_F / (P_F + P_B)], \quad (1)$$

where E_0 is the particle energy immediately in front of the foil. If the foil thickness is t , then

$$\Delta E/t = S(E') \quad (2)$$

is a measure for the stopping power at the energy $E' = E_0 - \frac{1}{2}(\Delta E)$. The errors in S coming from the uncertainties in the determination of E_0 , t , and $P_F/(P_F + P_B)$ are estimated to be 0.2, 0.1–0.15, and 0.1%, respectively, except for Ca, Sc, Cr, and Mn, where the error due to thickness measurements is considerably larger. The accuracy for S is thus 0.3% apart

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¹ W. H. Barkas and M. J. Berger, *Natl. Acad. Sci.—Natl. Res. Council*, Publ. **1133**, 103 (1964).

² H. Bichsel, in *American Institute of Physics Handbook* (McGraw-Hill Book Co., New York, 1963), 2nd ed.

³ J. F. Janni, Air Force Weapons Laboratory Technical Report No. AFWL-TR-65-150, 1966 (unpublished).

⁴ C. Serre, European Organization for Nuclear Research Report No. CERN 67-5 (unpublished).

⁵ C. F. Williamson, J. P. Boujot, and J. Picard, CEN Saclay, Report No. CEA-R 3042, 1966 (unpublished).

⁶ H. H. Andersen, H. Sørensen, and P. Vajda (to be published).

⁷ H. H. Andersen, Atomic Energy Commission, Denmark, Risø Report No. 93, 1965 (unpublished).

⁸ H. H. Andersen, A. F. Garfinkel, C. C. Hanke, and H. Sørensen, *Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd.* **35**, No. 4 (1966).

⁹ H. H. Andersen, C. C. Hanke, H. Sørensen, and P. Vajda, *Phys. Rev.* **153**, 338 (1967).

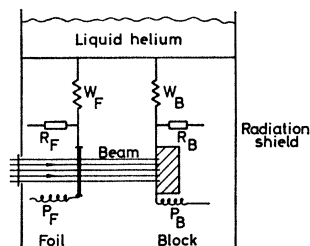


FIG. 1. Diagram of the stopping-power measuring system. W_F and W_B are thermal resistances, R_F and R_B thermometers, and P_F and P_B electrical heaters.

from Ca, Sc, Cr, and Mn, where it is estimated to be 0.5%.

Different precautions have been taken to ensure high thermal conductivity of the foil system, such as high-temperature annealing of foils and good thermal contact to the foil-holder frame. The latter was achieved either by soldering the foil to the frame or where not possible, by fixing it by means of Narmco resin which is very suitable for joints at low temperatures. Since no difference could be seen in the behavior of annealed and unannealed foils, the annealing was later abandoned.

The thickness determination was performed as a weight-per-area measurement; a rectangular piece of the irradiated part of the foil was weighed on a Cahn electrobalance and its area determined in a Leitz Ortholux microscope, unless otherwise stated. The measured area has been corrected for thermal contraction from room temperature to liquid-helium temperature by taking twice the value for the linear contraction.

A detailed description of the metal foils used and the treatment they were given is as follows:

Calcium

Thickness: ~ 11.0 and ~ 11.0 mg/cm². Thermal correction: 1.01%. Purity: 99.6% (+0.3% Mg). Supplier: Oak Ridge National Laboratory, Isotope Target Center. Calcium is very sensitive to oxidation, and special care has thus been taken to protect the foils against the influence of air. Approximately 150 $\mu\text{g}/\text{cm}^2$ aluminum has been evaporated onto both surfaces of the foils; Al has been chosen since the energy loss per mg/cm² is close to that of Ca—the error caused by assuming that Al and Ca have the same stopping power is of the order 0.05%, for the Al thickness used. Most of the treatment was performed in vacuum or in a glove box filled with argon. Handling of the foils in air (already with the Al cover on it) lasted at the most for 6 min. The foils were used unannealed, the thermal contact was achieved by Narmco resin. Some oxidation is of course still possible in spite of the precautions. An oxidation before the irradiation would give a too-high stopping power, while an oxidation taking place between irradiation and weighing would give a too-large thickness and thus a too-low stopping power. In the first

case oxidation would only introduce an impurity with a 20–25% higher stopping power, and 1% oxygen by weight will increase the stopping power by 0.2%. In the second case oxidation would increase the thickness and 1% oxygen will decrease the calculated stopping power by 1%. The probabilities for oxidation are of the same order of magnitude for the two cases, and a too-low stopping power will be the most probable result of an oxidation.

Scandium

Thickness: ~ 18.6 , ~ 18.9 , and ~ 18.9 mg/cm². Thermal correction: 0.40%. Purity: 99.7 (+0.25% O₂). Supplier: Johnson, Matthey and Co., Ltd. Unannealed. Thermal contact with Narmco resin. Considerable thickness gradients were found in these foils, possibly because scandium is rather difficult to roll to thin foils, and this gives larger uncertainty for the thickness determination.

Titanium

Thickness: ~ 6.2 and ~ 16.7 mg/cm². Thermal correction: 0.33%. Purity: 99.5% (+0.25% Fe). Supplier: A. D. Mackay Inc. Unannealed. Thermal contact with Narmco resin.

Vanadium

Thickness: ~ 8.8 and ~ 8.9 mg/cm². Thermal correction: 0.30%. Purity: 99.9%. Supplier: A. D. Mackay Inc. Unannealed. One foil attached to the frame by Narmco resin, the other electroplated with copper at the contact area and soldered.

Chromium

Thickness: ~ 12.9 and ~ 14.4 mg/cm². Thermal correction: 0.19%. Purity 99.5% (+0.4% Al+0.1% Fe). Supplier: Oak Ridge National Laboratory, Isotope Target Center. The foils had been produced by vacuum evaporation and were very brittle. They were cut into a suitable shape with a chisel and soldered to the frame with tin solder. The area was determined by photographing the foil, enlarging the photograph and measuring it with a planimeter. The magnification factor was determined by comparison with a known scale, photographed together with the foil. Unannealed.

Manganese

Thickness: ~ 11.7 and ~ 12.8 mg/cm². Thermal correction: 0.85%. Purity: 99.9%. Supplier: Oak Ridge National Laboratory, Isotope Target Center. The foils had been made by vacuum evaporation and were extremely brittle. They were cut into a suitable shape by a chisel and attached with Narmco resin to a well-annealed Al frame, which itself was soldered to the foil holder. Thereby the heat path became rather long, causing a relatively high thermal time constant. Area determination was done as for the Cr foil. Unannealed.

Iron

Thickness: ~10.0 and ~10.6 mg/cm². Thermal correction: 0.44%. Purity: 99.9%. Supplier: A. D. Mackay Inc. One foil was unannealed, the other annealed at 800°C for 4 h in a vacuum of 10⁻⁵ Torr. Weight determination was performed on a Mettler balance, since the Cahn electrobalance showed sensitivity to the foil's magnetism and yielded unreliable results. Thermal contact by soldering.

Cobalt

Thickness: ~10.8 and ~10.3 mg/cm². Thermal correction: 0.54%. Purity: 99.8% (+0.1% Ni). Supplier: A. D. Mackay Inc. Unannealed. Thermal contact by soldering.

Nickel

Thickness: I~15.1, II~10.1 mg/cm². Thermal correction: 0.48%. Purity: I=99.97%, II=99.5% (+0.5% Co). Supplier: A. D. Mackay Inc. Foil I was annealed at 1000°C for 4 h in a vacuum of 10⁻⁵ Torr, foil II was produced by electrodeposition and was unannealed. Weight determination as for Fe. Thermal contact by soldering.

Copper

Thickness: ~9.9 and ~18.5 mg/cm². Thermal correction: 0.68%. Purity: 99.9 and 99.99%. Supplier: A. D. Mackay Inc. and Heraeus. Annealed at 550°C for 4 h in a vacuum of 10⁻⁵ Torr. The data are taken over from Ref. 9.

Zinc

Thickness: ~9.9 and ~10.0 mg/cm². Thermal correction: 1.19%. Purity: 99.99%. Supplier: A. D. Mackay Inc. Unannealed. Thermal contact by soldering.

III. RESULTS

Several corrections have to be applied before the final results can be presented. Corrections for small-angle and for wide-angle scattering of the particles in the foil are largest for low energies and for thick foils of higher Z, but in our case they never exceed 0.25%. Corrections for x rays escaping from the foils and for δ rays emerging from the surface are most important for thin foils of higher Z and for high energies; the former hardly reach 0.1%, while the latter are ≤0.5%.

Corrections due to nuclear reactions in the foil, low-energy secondary electrons, sputtering of target atoms, stored energy due to radiation damage, and crystallographic effects like channeling are found to be negligible. The energy distribution of the conduction electrons is slightly temperature-dependent and this gives rise to a variation in stopping power with the temperature. Within the range between liquid-helium temperature and room temperature this effect, too, is found to be negligible.

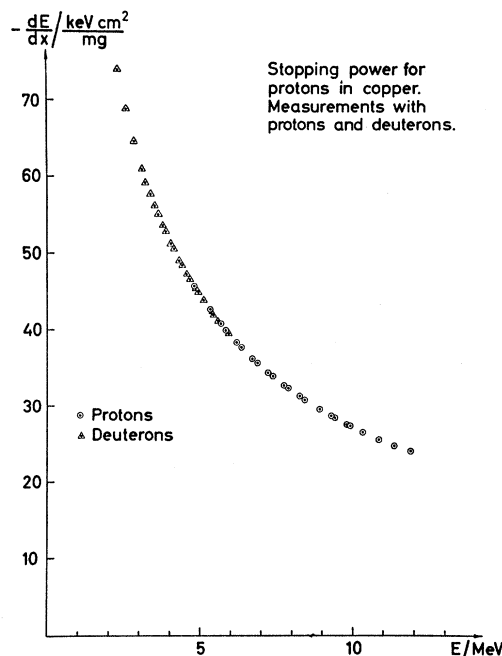


FIG. 2. Measured stopping power of copper. The data have been taken from Ref. 9.

The relativistic Bethe formula¹⁰ for the stopping power is

$$S = -dE/dx = (4\pi e^4 z^2 N_0 Z / m v^2 A) \times [\ln(2mv^2/I) - \beta^2 + \ln(1-\beta^2)^{-1} - (C/Z) - \frac{1}{2}\delta], \quad (3)$$

where *e* and *m* are the charge and the rest mass of the electron, *z* the atomic number of the incident particle, β*c*=*v* the velocity of the incident particle, and *N*₀ Avogadro's number. *Z*, *A*, *I*, and *C/Z* indicate atomic number, atomic weight, mean excitation potential, and shell corrections of the target material, respectively. The so-called density correction δ is only important at higher energies and can be neglected here.

The energy variation of stopping power is mainly given by ln*E*/*E*. This term varies so rapidly with *E* that it is impossible to see small deviations from this dependence, when the stopping power is plotted directly. Figure 2 shows the measured *dE/dx* for copper data from Ref. 9. Deuteron-stopping powers are reduced to proton-stopping powers by plotting the deuteron points at the energy *EM_p/M_d*, where *M_p* and *M_d* are the masses of the protons and deuterons (*M_p*=938.214 MeV and *M_d*=1875.5 MeV, respectively), since particles with the same charge and velocity should have the same stopping power, according to Eq. (3). It is obvious that this plot does not show all the information contained in the data. We have, therefore, used the *X* function, which is

¹⁰ U. Fano, *Ann. Rev. Nucl. Sci.* **13**, 1 (1963).

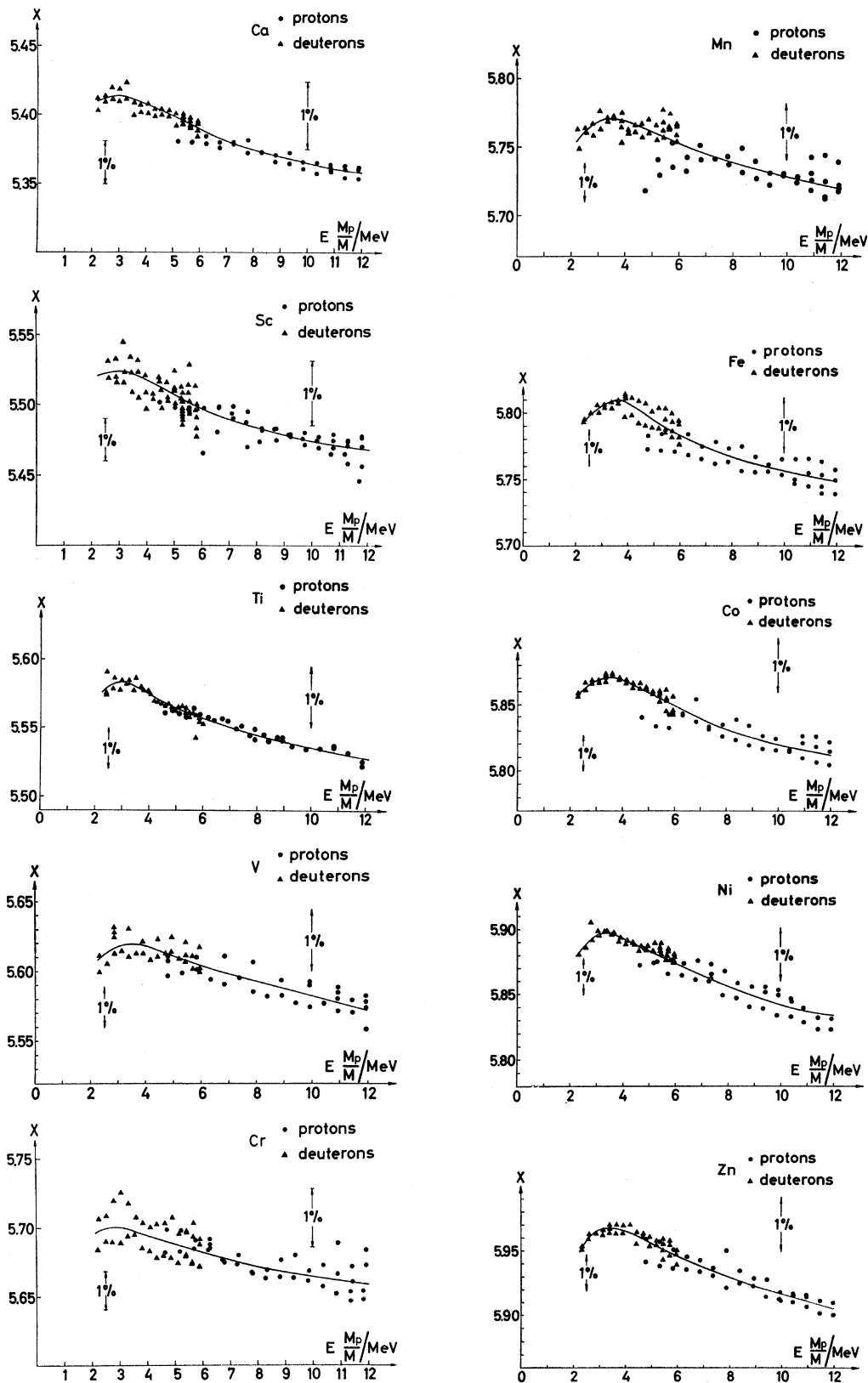


FIG. 3. Measured stopping powers of Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, and Zn in reduced variables defined by Eq. (6). The height of the arrows indicates the change in X caused by a 1% change in stopping power at the respective energies. The full lines are fitted to the experimental points by eye. M_p/M = ratio between proton mass and mass of incident particle (cf. text).

TABLE I. Smoothed values of measured stopping powers of Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, and Zn for 2.25-12.00-MeV protons. Cu is taken over from Andersen *et al.*,^a the other elements are obtained from the full curves of Fig. 3. Estimated accuracy $\pm 0.3\%$; for Ca, Sc, Cr, and Mn $\pm 0.5\%$.

Energy MeV	Stopping power $-dE/dx$ (keV/mg cm ⁻²)										
	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn
2.250	98.91	89.24	86.11	83.73	82.93	80.07	80.83	77.64	80.14	75.19	75.02
2.500	92.03	83.15	80.23	78.02	77.42	74.72	75.45	72.56	74.89	70.28	70.13
2.750	86.15	77.94	75.20	73.14	72.70	70.13	70.85	68.18	70.35	66.07	65.92
3.000	81.09	73.40	70.87	68.91	68.60	66.16	66.84	64.37	66.41	62.44	62.32
3.250	76.65	69.45	67.11	65.22	65.00	62.66	63.32	61.04	62.96	59.25	59.16
3.500	72.76	65.97	63.78	61.93	61.82	59.59	60.21	58.09	59.97	56.43	56.36
3.750	69.30	62.87	60.81	59.04	58.98	56.86	57.45	55.46	57.28	53.91	53.85
4.000	66.18	60.08	58.15	56.44	56.42	54.40	55.00	53.10	54.88	51.65	51.60
4.250	63.38	57.57	55.74	54.09	54.09	52.19	52.79	50.97	52.69	49.60	49.55
4.500	60.83	55.29	53.54	51.95	51.97	50.16	50.78	49.03	50.70	47.72	47.69
4.750	58.51	53.21	51.53	50.00	50.04	48.31	48.94	47.26	48.86	45.99	45.99
5.000	56.37	51.30	49.68	48.21	48.26	46.62	47.24	45.62	47.17	44.42	44.42
5.250	54.41	49.53	47.98	46.56	46.62	45.05	45.67	44.11	45.61	42.95	42.97
5.500	52.59	47.90	46.40	45.03	45.09	43.60	44.22	42.72	44.16	41.59	41.61
5.750	50.91	46.38	44.93	43.61	43.68	42.25	42.87	41.41	42.82	40.33	40.36
6.000	49.35	44.97	43.56	42.29	42.37	40.99	41.60	40.20	41.57	39.15	39.19
6.500	46.53	42.41	41.10	39.90	39.98	38.72	39.31	38.00	39.30	37.01	37.07
7.000	44.04	40.17	38.92	37.80	37.88	36.71	37.28	36.06	37.29	35.12	35.19
7.500	41.83	38.17	36.99	35.93	36.02	34.92	35.47	34.33	35.51	33.44	33.52
8.000	39.85	36.38	35.27	34.26	34.35	33.32	33.85	32.78	33.90	31.93	32.01
8.500	38.08	34.77	33.71	32.75	32.85	31.88	32.39	31.37	32.45	30.57	30.65
9.000	36.47	33.31	32.30	31.39	31.49	30.57	31.06	30.09	31.14	29.33	29.41
9.500	35.00	31.98	31.02	30.15	30.25	29.38	29.85	28.92	29.95	28.21	28.29
10.000	33.66	30.76	29.84	29.01	29.11	28.28	28.74	27.85	28.84	27.17	27.26
10.500	32.43	29.65	28.77	27.97	28.06	27.28	27.72	26.87	27.83	26.22	26.30
11.000	31.30	28.61	27.77	27.01	27.10	26.35	26.78	25.96	26.89	25.35	25.42
11.500	30.25	27.66	26.85	26.12	26.21	25.49	25.90	25.12	26.02	24.54	24.61
12.000	29.28	26.77	26.00	25.30	25.38	24.69	25.09	24.33	25.21	23.78	23.85
Atomic weight	40.08	44.96	47.90	50.95	52.01	54.94	55.85	58.98	58.71	63.54	65.38

^a Reference 9.

defined¹¹ in the following way:

$$S = (Z/A)K(\beta)[f(\beta) - X], \quad (4)$$

where

$$K(\beta) = 4\pi e^4 z^2 N_0 / mc^2 \beta^2,$$

$$f(\beta) = \ln(2mc^2 \beta^2 / 1 - \beta^2) - \beta^2,$$

and

$$X = \ln I + C/Z. \quad (5)$$

$K(\beta)$ and $f(\beta)$ are calculated, and experimental values of X are expressed explicitly by

$$X_{\text{expt}} = f(\beta) - AS_{\text{expt}}/ZK(\beta). \quad (6)$$

The experimental values for $S(E)$ are inserted and we get for each metal a set of points to which a smooth curve is fitted by eye (Fig. 3). For each metal two separate runs with different foils (and often different thicknesses) had been taken. The fitting procedure was made separately for each run and only to the very end the two smooth curves were combined together to yield the single curves of Fig. 3. This explains the apparently poor fit of the curve to the nickel data at the high-energy end. The arrows in the diagrams indicate the change in X , caused by a 1% change in stopping power at the respective energy.

Although the data for Mn scatter somewhat more than usual at the lowest proton energies, where the energy calibration is most difficult, it was still possible to fit a smooth curve to the points for each of the Mn samples. The two X curves obtained did, however, not agree, neither in position nor in slope. The direct thickness determination was somewhat uncertain for Mn, partly because of difficulties in handling the foils and partly because of thickness gradients. The foils were made by evaporation and this can cause serious thickness gradients. The disagreement was thus attributed to errors in the thickness determination. Inspection of Fig. 3 shows that the arrows indicating a change in stopping power of 1% have different lengths at high and low energy. A change in thickness will then change both position and slope of an X curve and as the slope of the X curves varies very slowly with Z , the thickness can be used as a free parameter to determine the position of the X curve. For each of the foils the thickness was then changed until the slopes of the X curves agreed with those of Cr and Fe. This also made the two curves agree. The uncertainty of this thickness determination is estimated to be 0.5%. This method could probably also be used for other materials where thickness determination is difficult, provided the X curves are known for some neighboring elements.

In Fig. 4 all the X curves are shown together. It is seen that the density of curves decreases with decreasing

¹¹ H. Bichsel, Natl. Acad. Sci.—Natl. Res. Council, Publ. 1133, 17 (1964).

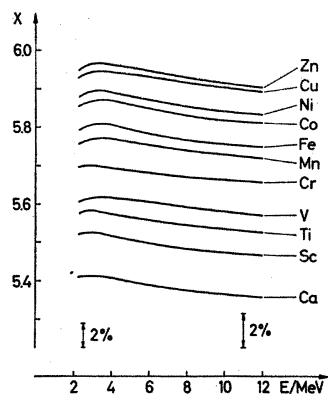


Fig. 4. X curves for the elements $Z=20$ through $Z=30$.

Z value, and the curve for Ca is actually very low lying. A too-high stopping power will give a too-low X curve. An oxidation of the calcium foils is of course possible but the most probable result of an oxidation would, as earlier mentioned, be a too-low stopping power and thus a too-high X curve.

Table I gives the stopping powers for protons of Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, and Zn in keV/mg cm^{-2} from 2.25–12.0 MeV. The numbers in the table were found by taking X values from the fitted curves and calculating the corresponding $S(E)$ using Eq. (4). The copper data are taken from Ref. 9. The errors are 0.3%, except for Ca, Sc, Cr, and Mn, where they are estimated to be 0.5%. For Mn, it was necessary to use the thickness of the foils as a parameter and to match the slope of the X curves to its nearest neighbors in order to get reasonable results as mentioned above. The Sc foils available were not thin enough to allow measurements down to the lowest energies.

IV. DISCUSSION

We have collected several recent tables of stopping power for protons and deuterons of our energy range. They have been transformed to X functions and are in Fig. 5 compared with our results.

The most elaborate recent tables are those of Janni,³ who has calculated stopping powers and ranges for practically all the elements and a number of compounds. Janni's data agree with ours within 1%, except for Ca, Sc, and Mn, where more serious discrepancies are found. The general shape of the X curves coincides, too, although his curves reach a maximum at lower energies than ours do.

The tables by Williamson *et al.*⁵ give simply horizontal lines. For Cu the agreement with our data is better than 1%. For other elements the agreement is not so good, and it is seen that the difference increases with decreasing Z value. The curves by Barkas and Berger¹ cut our curves at two energies, but are otherwise quite different in form. The high-energy tables by Serre⁴ for Cu and

Fe are computed only down to 10 MeV. They coincide with the Barkas-Berger curves and are not shown here. Bichsel's table² for Cu has already been discussed by us in a previous paper on this subject.⁹ It is in very good agreement with our data.

There are several experimentalists who have measured the same elements in our energy range or whose results can be compared with ours after reduction. Gorodetzky *et al.*¹² measured the stopping power of Ca for 1–5.9-MeV protons; their data lie generally above our results, the disagreement is $\sim 2\%$ for the lowest energies and increases to $\sim 5\%$ at higher ones. Wolke *et al.*¹³ used 1–2.7-MeV tritons, corresponding to 0.3–0.9-MeV protons, in Ni; the data scatter less than 1% and provide actually a good extension of our data to lower energies. Comfort *et al.*¹⁴ bombarded Ni with 8.78-MeV α 's and measured stopping powers corresponding to those for 1.15–2.1-MeV protons; their results are generally higher than ours and scatter 2–3%. Perch Nielsen's¹⁵ proton and deuteron measurements on Ni in the energy range of 1–4.5 MeV yield, in general, lower stopping-power results than ours; they scatter 1.5–2%.

Most of the relative stopping-power determinations

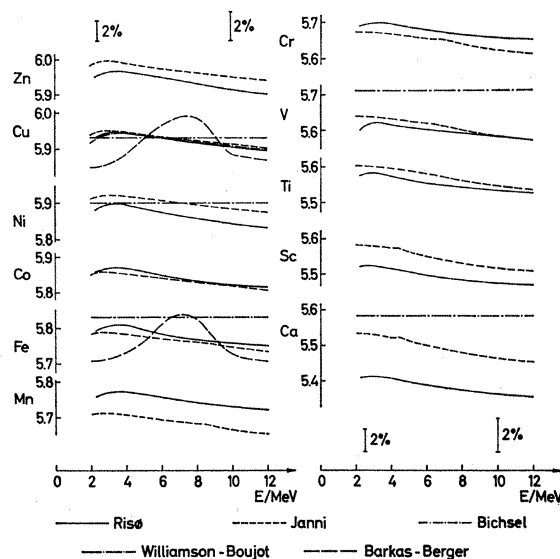


Fig. 5. Comparison of our smoothed X curves to tabulated data. Barkas-Berger (Ref. 1), Bichsel (Ref. 2), Janni (Ref. 3), and Williamson *et al.* (Ref. 5).

¹² S. Gorodetzky, A. Chevallier, A. Pape, J. Cl. Sens, A. M. Bergdolt, M. Bres, and R. Armbruster, Nucl. Phys. **A91**, 133 (1967).

¹³ R. L. Wolke, W. N. Bishop, E. Eichler, N. R. Johnson, and G. D. O'Kelley, Phys. Rev. **129**, 2591 (1963).

¹⁴ J. R. Comfort, J. F. Decker, E. T. Lynk, M. O. Scully, and A. R. Quinton, Phys. Rev. **150**, 249 (1966).

¹⁵ L. Perch Nielsen, Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd. **33**, No. 6 (1961).

were made with respect to Al: Kelly's¹⁶ 22- and 33-MeV α bombardment of Cu gives results that are higher than ours for 5.5- and 8-MeV protons by 0.9 and 1.3%, respectively. Heller¹⁷ used 7.9-MeV deuterons in Ni and Cu. The Ni results are higher than ours by 1.4%, the Cu results lower by 0.4%. Wilson¹⁸ bombarded Fe, Ni, Cu, and Zn with 4-MeV protons. His data are lower than ours by 0.5, 4.6, 3.8, and 3.4%, respectively. Teasdale's¹⁹ 12-MeV proton measurements are lower by 0.2% for Ni, and higher by 0.8% for Cu, than ours.

It is concluded that there are no significant disagreements between our results and the quoted experiments. Although most of the quoted results are considerably less accurate than ours, it is worth noting that there is

no over-all systematic deviation between our data and the quoted ones.

For the tables the agreement is usually quite good for well-known elements, while disagreements exist for elements where experimental data are scarce.

A detailed discussion will be given in a following paper,⁶ where our results are used for the evaluation of shell corrections and of mean excitation potentials.

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¹⁶ E. L. Kelly, Phys. Rev. **75**, 1006 (1949).

¹⁷ Z. H. Heller and D. J. Tendam, Phys. Rev. **84**, 905 (1951).

¹⁸ R. R. Wilson, Phys. Rev. **60**, 749 (1941).

¹⁹ J. G. Teasdale, University of California, Los Angeles, Report No. NP-1368, 1949 (unpublished).

Electron-Nuclear Double Resonance of the Self-Trapped Hole in LiF* †

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An electron-nuclear double-resonance (ENDOR) study of the self-trapped hole in LiF is reported. The results agree with the accepted model, i.e., the hole is shared by two halide-ion lattice sites in the form of a negatively charged diatomic halide molecule (F_2^- in this study). The variations in the ENDOR spectra for different ESR lines is investigated theoretically and experimentally. The observation of negative-contact hyperfine constants for most ENDOR nuclei is explained by exchange polarization of the "closed-shell" molecular orbitals.

I. INTRODUCTION

THE self-trapped hole or V_K center was discovered by Känzig¹ in an electron-spin-resonance (ESR) study of KCl exposed to x rays at liquid-nitrogen temperatures. Castner and Känzig² carried out an ESR investigation of the V_K center in several alkali halides. These ESR studies showed the V_K center to be

a negatively charged diatomic halide molecule (e.g., F_2^-) with the molecular axis oriented in a $[110]$ crystal direction. Also, there was strong indirect evidence that the molecule was located on two halide-ion lattice sites, and that the center was not associated with any other lattice defects such as impurities or vacancies. An excellent introduction to the ESR of the V_K center has been given by Slichter.³

Castner and Känzig tentatively correlated the V_K center with the V_1 optical band. However, Delbecq, Smaller, and Yuster⁴ showed that the defect was

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¹ W. Känzig, Phys. Rev. **99**, 1890 (1955).

² T. G. Castner and W. Känzig, J. Phys. Chem. Solids **3**, 178 (1957).

³ C. P. Slichter, *Principles of Magnetic Resonance* (Harper and Row, New York, 1963).

⁴ C. J. Delbecq, B. Smaller, and P. H. Yuster, Phys. Rev. **111**, 1235 (1958).