Calorimetric measurement of electron stopping power of aluminum and copper between 11 and 127 keV

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The stopping powers of aluminum and copper have been measured for electrons between 11 and 127 keV. Evaporated foils of about 200-nm thickness were stretched across rings and inserted into a short cylinder which was supported by thermocouple wires. Peltier heating and cooling by one thermopile were used to control foil temperature, while another thermopile measured its temperature. Calibration was accomplished by reducing beam energy until complete absorption was obtained. Delta rays produced in the foil were returned to it by repelling electrodes. Corrections were made for increased path length in the foil due to multiple elastic small-angle scattering and for spurious heating by scattered electrons striking the ring holding the foil. After correcting for these effects, good agreement with the Bethe-Bloch formula was found.

I. INTRODUCTION

Stopping power, the space rate of energy loss dE/dx by a fast atomic particle in traversing matter, has been the subject of a great deal of theoretical and experimental study for the past 80 years. Most experimental work has been done with protons or heavier particles at energies of over about 1 MeV. The lack of much good data for electron stopping powers in the region from 10 to 125 keV led us to make such measurements some time ago on aluminum[1] and copper[2] foils by a calorimetric method. Because of the continuing interest in the field, we have now recomputed the data using corrections which were omitted in the original work and using improved, recent values of fundamental physical constants; we now publish it in the open literature to give the results wider circulation. Studies of this kind are useful in confirming the accuracy of the theory, so that such measurements and calculations may be used both to give information about atomic projectiles and the damage they do to matter and for radiation dose calculations in fields such as health physics.

The calorimetric method has the great advantage of measuring directly the quantity sought, the energy loss of a beam of particles in a thin foil, as opposed to the more common method of measuring the beam-energy distributions before and after passage through a foil. In the latter case one must either find the small differences between large beam energies, in the case of a thin foil, or use a thick foil and make very large corrections for the effects of scattered particles which may escape the detector completely or have substantially increased path lengths in the foil.

In brief, our method consisted of bombarding the foil with an electron beam in a small accelerator. The foil was supported by a small ring whose temperature was measured by a set of six thermocouples in thermal contact with it. The ring could be cooled by the Peltier effect of a second set of six thermocouples intercalated among the measuring set. The system was calibrated by lowering the beam voltage till the electrons were completely absorbed in the foil. Energetic secondary electrons were returned to the foil by repelling electrodes, and corrections were made for electrons scattered into the supporting ring and for the increase in path length of the primary electrons resulting from multiple small-angle scattering.

In this paper the results of the basic Bethe-Bloch theory of stopping power will be stated, with emphasis on the theoretical aspects of correcting for scattering; then the calorimetric method and the experimental corrections will be outlined. The features of the apparatus and method will be mentioned and, finally, the results will be compared to the theory and to other related experiments.

II. THEORY OF STOPPING POWER

The theory of stopping power originally formulated by Bohr[3] has been highly developed by many

workers in the past 70 years. It is generally accepted that the theory of Bethe,[4] augmented by the statistical arguments of Bloch[5] for the average excitation potential, adequately describes the slowing down of electrons along their paths in the energy region of interest here, 10 to 125 keV. However, modern semiempirical values for the average excitation potentials I,[6,7] derived from experiments on the stopping of heavy ions and theoretical calculations of electron shell effects, are preferred to the original estimate that $I=13.5Z$. Experiments with heavy particles do not suffer so much from scattering problems as electron experiments do. Further developments are summarized by Birkhoff[8] and Bethe and Ashkin,[9] and a more recent summary has been given by Sigmund.[10]

According to Bethe, the spatial rate of energy loss, or relativistic stopping power using the Möller[11] electron-electron scattering cross section, is given by

$$-\frac{dE}{dx}=2\pi N_A r_0^2 m_0c^2\left[\frac{\rho Z}{A}\right]\frac{1}{\beta^2}\left\{\ln\left[\left[\frac{m_0c^2}{2I}\right]^2\frac{\beta^4}{\gamma^3}\right]-(2\gamma-\gamma^2)\ln 2+\gamma^2+[(1-\gamma^2)/8]\right\}, \quad (1)$$

where the symbols are as follows[12]: N_A is Avogadro's number equal to $6.022\,045\times10^{23}$ mole^{-1}; r_0 is e^2/m_0c^2, the "classical radius" of the electron equal to $2.817\,9380\times10^{-13}$ cm; m_0c^2 is the rest energy of the electron equal to 0.511 0034 MeV; ρ,Z,A are the density, atomic number, and atomic weight of foil; β is v/c (electron velocity/velcocity of light); γ is $(1-\beta^2)^{1/2}$ (not the usual definition of γ); and I is the average ionization potential of foil material—163 eV for Al, 322 eV for Cu.[6,7] β is readily calculated for an electron from the usual relativistic relation

$$\beta=(T^2+2T)^{1/2}/(T+1)\,, \quad (2)$$

where T is the kinetic energy of the electron in m_0c^2 units:

$$T=E(\text{keV})/511.0034\,. \quad (3)$$

Equation (1) gives the *average* rate of energy loss as compared to the *most probable* rate given by Landau[13] and corrected by Blunck and Leisegang[14]for binding effects. These other workers give also the complete *distribution* of energy-loss events, basically a Gaussian curve with a long tail due to the few collisions in which large energy losses occur in producing δ rays. The calorimetric method gives only the average loss, so we need not concern ourselves here with more detailed descriptions of the energy-loss processes nor with energy-loss spectroscopy.

The energy losses of electrons by bremsstrahlung radiation and the "density effect" due to polarization of the stopping medium by the fast electron are both negligible for the electron energies (11—127 keV) and elements (Al and Cu) which we studied. (See Ref. 8, pp. 63 and 70.)

Other work on corrections to the Bethe-Bloch formula[15,16] (and references therein) has been concerned with stopping of electrons with less than 10-keV energy, down to a few eV, and is not applicable in the energy range we used.

We calculated the stopping powers from the Bethe-Bloch formula, Eq. (1), using the fundamental constants given above. The results were compared with the tabulations of Berger and Seltzer[17] and of Pages *et al.*[18] The differences among the three sets of stopping-power results were only a few percent at worst and were due to the slightly different values of fundamental constants and I values used. These results are plotted as solid curves in Figs. 2 and 3 which are discussed below.

III. ELECTRON-SCATTERING CORRECTIONS

Although no measurements of electron scattering are made here, an understanding of scattering was necessary for proper design and interpretation of our calorimetric measurements. Electrons scatter *elastically* from nuclei with a cross section first given by Rutherford.[19] Electrons scatter *inelastically* from the electrons with a cross section given by Thomson[20] and Bohr[3] and improved by Möller.[11] The ratio of the elastic and inelastic macroscopic cross sections per unit solid angle is approximately Z; thus, the importance of considering the nuclear scattering in this work is evident. We will discuss the scattering in three parts: First, the use of reflecting disks to redirect δ rays back into the absorber; second, the correction of the data for elastic scattering of electrons into the foil support ring *with complete absorption;* and third, the effect of multiple small-angle scattering in increasing the electron path length in the foil.

A. δ rays

Wire-mesh biased disks with central holes to pass the beam were mounted on polytetrafluoroethylene (Teflon) insulators on each side of the foil. It was found that a potential of -1000 V on the disks was sufficient to reduce to zero the current pickup from δ rays and scattered electrons leaving the foil and striking the biased disks. Since the theory of energy loss assumes that the electron having *less* than half the energy is the secondary, these bias disks were important in reflecting the energetic secondaries back into the foil where they could contribute to the heating. Actually the bias disk potential should be $E/2$, but this was not possible in our design.

B. Electrons scattered into support ring

The foils were mounted on short cylindrical rings which were designed to make good thermal contact inside the thermocouple support cylinder. Some elastically scattered electrons might strike this ring, losing all their energy. To minimize this effect, the axial dimensions of the rings were kept minimal, intercepting electrons scattered only between 90° and 112° to the forward beam direction. For the Cu foil, a calculation from the Mott[21,22] nuclear scattering formula indicated that 34% of the heating at 32-keV beam energy was due to singly scattered electrons striking the ring, with lower percentages at higher energies. This extraneous heating was due to only 2% of the beam being deflected onto the ring.

The correction for this heating of the calorimeter by large angle Coulomb nuclear scattering in the foil was made as follows (this correction was not made in the original report[1]): We first find the fraction f of the electrons in the beam, singly scattered into the solid angle between angles θ_1 and θ_2 to the forward beam direction, so that they strike the foil support ring. This is given by the Rutherford[19] cross section integrated between θ_1 and θ_2 and multiplied by the correction factor $F=\sigma_{\text{Mott}}/\sigma_{\text{Rutherford}}$ using σ_{Mott} calculated by Mott[21,22] for a Dirac electron. This correction F has been evaluated numerically by McKinley and Feshbach,[23] Feshbach,[24] and others for high energies, and for the Z and energy range of interest here by Doggett and Spencer[25] and Sherman.[26] We have interpolated in the tables of the last two papers to obtain F for our cases. Since F varies slowly with scattering angle θ, we used the mean value between $\theta_1=90°$ and $\theta_2=112°$, the angles subtended by the support ring, namely, $\theta=101°$. The result for a foil of thickness x cm is

$$f=\frac{\pi F N_A r_0^2 Z^2 \rho x (1-\beta^2)}{A\beta^4} \times\left[\frac{1}{\sin^2(\theta_1/2)}-\frac{1}{\sin^2(\theta_2/2)}\right]. \tag{4}$$

Since the heating of the foil and ring is due to the sum of the fraction of the beam energy lost in the foil by the normal stopping-power effect plus all the energy of the electrons scattered into the ring, it is easily shown that the true average energy ΔE which should be deposited in the foil is related to the observed average energy deposited ΔE_0 by

$$\Delta E=\frac{\Delta E_0-fE}{1-f}. \tag{5}$$

C. Path straggling

The problem of multiple scattering of fast charged particles in matter has challenged a great many people, and the literature on both theoretical and experimental aspects is extensive with somewhat discordant results, depending on the assumptions and approximations made.[8,27–48] The theory of path straggling was developed for multiple small-angle scattering by Rossi and Greisen[47] and Yang.[37] Although these authors' derivations are for high-energy electrons (>6 MeV), the same logic may be applied in our energy range, but with a different scattering-angle formula. Yang shows that if one takes y and z axes perpendicular to the beam direction and if all scattering angles θ_y and θ_z are small, then the average increase in path length for the electrons is

$$\langle\Delta t\rangle_{\text{av}}=\tfrac{1}{2}\int_0^t(\langle\theta_y^2+\theta_z^2\rangle_{\text{av}})dt'. \tag{6}$$

He also shows that by symmetry

$$\langle\theta_y^2\rangle_{\text{av}}=\langle\theta_z^2\rangle_{\text{av}}. \tag{7}$$

But the average mean-square polar scattering angle is easily shown to be [see Rossi and Greisen,[47] p. 265, after Eq. (157)]

$$\langle\Theta^2\rangle_{\text{av}}=\langle\theta_y^2\rangle_{\text{av}}+\langle\theta_z^2\rangle_{\text{av}}. \tag{8}$$

Thus

$$\langle\Delta t\rangle_{\text{av}}=\tfrac{1}{2}\int_0^t\langle\Theta^2\rangle_{\text{av}}dt. \tag{9}$$

Ritchie, Ashley, and Emerson[48] give an expression for $\langle\Theta^2\rangle_{\text{av}}$ per unit thickness [their Eq. (9)] which they state is derived from Eq. (56a) of Nigam, Sundaresan, and Wu.[44] For a foil of thickness t, substituting the usual physical constants, we get

$$\langle\Theta^2\rangle_{\rm av}=t\frac{8\pi N_A r_0^2 Z(Z+1)(1-\beta^2)\rho}{A\beta^4}\left[\ln\left[\frac{\beta}{\alpha Z^{1/3}(1-\beta^2)^{1/2}}\right]+\ln\frac{1.76}{\nu}-(1+\beta^2/4)\right]. \quad (10)$$

Ritchie *et al.* state that the mysterious quantity ν is $O(1)$. We shall take $\nu=1$. (See note added in proof.) α is the fine-structure constant 1/137.036.[12]

Since $\langle\Theta^2\rangle_{\rm av}$ is proportional to t, the integration of Eq. (9) using (10) gives a factor of $t^2/4$ in the result for $\langle\Delta t\rangle_{\rm av}$. Hence the path-length correction (in g/cm^2) becomes

$$\langle\Delta t\rangle_{\rm av}=2\pi N_a r_0^2\frac{Z(Z+1)\rho t^2}{A}\frac{(1-\beta^2)}{\beta^4}\left[\ln\left[\frac{\beta}{\alpha Z^{1/3}(1-\beta^2)^{1/2}}\right]+\ln 1.76-(1+\beta^2/4)\right]. \quad (11)$$

The correction is a function of beam energy through β. Hence, the measured foil thickness in g/cm^2 was increased by the calculated $\langle\Delta t\rangle_{\rm av}$ at each energy in obtaining stopping power. For example, the fractional path increases $\langle\Delta t\rangle_{\rm av}/t$ was 0.099 for the thinnest Al foil at the lowest energy 11.9 keV. The total average path was thus 54.9 μg/cm^2 for a foil of 49.9 μg/cm^2 actual thickness. The corrections were less for higher energies.

IV. CALORIMETRIC METHODS

A. History

Calorimetric methods for measuring the energy deposited in matter by various radiations have been used at least since 1903 when Curie and Laborde[50] used a calorimeter to measure the heat generated by the decay of radium. Leithauser[51] in 1904 noticed that electrons lose energy in traversing thin layers of matter. A summary of early work on microcalorimetry for the measurement of radiation is given by Myers,[52] and more recent work has been reviewed by Gunn.[53,54]

Our application[1,2] seems to have been the first use of calorimetry in measuring electron stopping power. Andersen and his colleagues[55–60] later developed the calorimetric method into a technique for obtaining very precise stopping-power values ($<0.5\%$) for heavy ions at MeV energies. They operated the calorimeter at liquid helium temperature, measured both ΔE and beam energy E by the heating of a thin foil and the beam stopping block, respectively. They measured temperature by the resistance changes in a carbon resistor, a method which is quite sensitive at 4 K. They calibrated the systems with electric-heating coils.

Pugachev and Volkov[61] measured calorimetrically the stopping powers of thin films of Al, Ag, and Au bombarded by electrons, using the resistivities and their temperature coefficients as given in the literature to calculate the energy deposited in the films. They made corrections using data from the literature for the differences in resistivity and its temperature coefficient between bulk metals and thin films. Their results will be discussed below.

Berlyand, Generalova, and Gurskii[62] used a thin film of conducting polyamide as a calorimeter for measuring electron-beam dose, but only for much larger beam power than we are concerned with. They did not measure stopping power.

Lockwood *et al.*[63] used a calorimetric method with a thermocouple as a detector and a modulated beam to measure electron energy deposition in extended media, but did not measure stopping power as such.

B. The heat equation

Our calorimeter contained two Cu-constantan thermopiles, one for measuring the temperature and the other for providing Peltier heating and cooling to the foil and support system. Therefore, the heat exchange between the calorimeter and its surroundings can be described as follows. The beam-power input added to the Peltier heating (or cooling, depending on the Peltier current direction) plus Joule heating and the heat gain or loss to the environment by radiation and conduction produces a rate of temperature change inversely proportional to the heat capacity of the calorimeter:

$$i_B\Delta E+(n\Pi T i_p+\tfrac{1}{2}i_p^2R)+L(T_0-T)=\sum_i m_i c_i J\frac{dT}{d\tau}, \quad (12)$$

where i_B and ΔE are the beam current and energy loss in the foil; n is the number of Peltier thermocouples (here, six), Π is the thermoelectric power of the Cu-Constantan couple, about 40 μV/°C; T is the absolute temperature of the ring and T_0 of the surroundings; i_p is the Peltier current; R is the resis-

tance of the Peltier thermopile; m_i and c_i are the masses and specific heats of the foil, ring, and thermopile system; J is the mechanical equivalent of heat; and L is the heat-transfer coefficient from Newton's law of cooling. Since $T-T_0$ was never more than 0.1 °C, Newton's law of cooling was sufficiently accurate. It could easily be shown that one-half of the Joule heating of the thermocouple wires which supported the ring flowed to the ring system, independent of T.

Although a null method of balancing Peltier cooling against beam heating could have been used, it was found to be more accurate and reproducible to precool the ring, turn off the Peltier current, and measure the time rate of temperature drift $dT/d\tau$ with the beam on as T passed through T_0, the ambient temperature.

For zero Peltier cooling, Eq. (12) can be written

$$dT/d\tau = K_1 i_B \Delta E + K_2(T_0 - T)\,, \tag{13}$$

where $K_1 = 1/\sum m_i c_i J$ and $K_2 = LK_1$. The constant K_1 was determined empirically by cooling the calorimeter below ambient temperature, setting the beam voltage so low that the beam was totally absorbed in the foil, and then timing the temperature rise as the foil and ring warmed through ambient temperature. The value of K_1 using six thermocouples and a galvanometer was 38 μW cm^{-1} min, where the dimension cm^{-1} min was the inverse of the galvanometer drift rate. The constant K_2 was determined by again precooling the calorimeter and again letting it drift toward ambient temperature, but this time with the beam off. Values of K_2 were about 0.2 min^{-1}. Both of these values were in reasonable agreement with values calculated from handbook data for the specific heats and heat conductivities of the system and using dimensions and masses of the supporting wires and ring.

We calculated that the relaxation time for a heat pulse to travel from the center of the foil to the ring was about 0.2 sec, which is short compared to the response period of the thermopile galvanometer (6 sec). If the beam was concentrated at a hot spot at the center of the foil, for a typical power input to the foil of about 250 μW, the hot spot would be less than 0.5 °C above the ring temperature.

V. APPARATUS AND EXPERIMENT

A. Apparatus

The electron beam was obtained in a small Cockcroft-Walton-type accelerator about 1.5 m long, with a gun from a cathode-ray oscilloscope.[49] The gun supplies and controls were insulated and at high voltage. The beam was collimated by an 0.8-cm-diam diaphragm about 75 cm from the gun and two more similar ones about 5-cm apart 75-cm farther along the beam path, just in front of the calorimeter. All acceleration occurred in the first 75 cm; the second half of the beam path was field free. Three degaussing coils permitted canceling the earth's field and steering the beam.

The calorimeter, or foil mount, and thermopile system are shown in cutaway view in Fig. 1. Only one thermopile is shown, but the Peltier thermopile was identical, the hot junctions being mounted on alternate tabs of the insulating plastic ring holder. Fine mesh screen diaphragms were placed on each side of the foil ring (only one is shown in the drawing) and charged to -1000 V to repel secondary electrons back to the foil so that they would deposit their energy there, as required by the definition of stopping power. After passing through the foil, the beam went into a deep Faraday cup, from which the beam current was measured with a calibrated galvanometer. Except during calibration, when all the beam stopped in the foil, the currents to the bias disks and foil were negligible compared to the Faraday cup current. The calorimeter was enclosed in a thermally insulated enclosure kept at about 29 °C and thermostated within less than 0.1 °C, so that the foil temperature inside the vacuum enclosure probably varied less than about 0.01 °C. The beam voltage was obtained from a rectified and filtered x-ray power supply and was measured by a calibrated string of 41 4-MΩ wire-wound resistors immersed in oil and a meter whose calibration checked within 2% or less. Below 30 kV, a 1% electrostatic voltmeter was also used and checked the first method within 0.5 kV.

Foils were made by evaporating Al or Cu onto microscope slides which were first coated by evaporation with a soluble parting agent, such as Formvar, Victawet, or water-soluble salts such as NaCl or KCl. The films were floated off the slide in a dish of solvent, ethylene dichloride for Formvar, water for most other cases, and mounted on copper rings about 1.5 cm/diam, 0.2 cm thick and 0.3 cm long. The rings were tapered slightly and fitted snugly inside the plastic ring holder which carried the thermocouple junctions.

The average foil thickness was measured after the stopping-power run by cutting a circular piece with a very sharp, accurately machined punch and weighing the foil piece punched out. For weighing

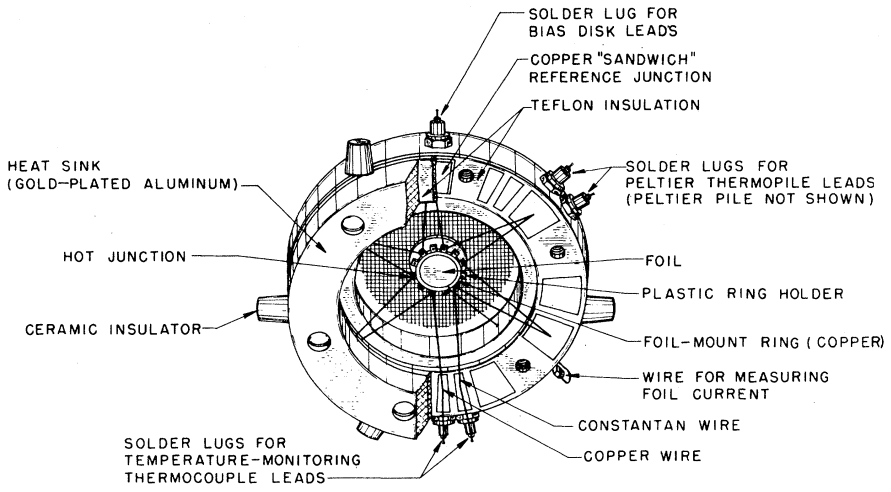

FIG. 1. Cutaway view of foil mount and heat sink for measuring electron stopping power, showing wiring for one thermopile. Rear screen bias disk with a center hole to pass the beam to the Faraday cup is visible. An identical disk in front is not shown. Disks are biased to repel the secondary electrons back into the foil. The second or Peltier cooling thermopile is identical to the one shown, with its inner junctions fastened to the other set of tabs on the foil support ring, which are shown here as empty. Foil is about 1.5 cm diam, and the heat sink about 12 cm diam.

we used a Cahn Electrobalance which was improved by using a standard resistor and Leeds-Northrup-type K2 potentiometer to read the current in the torque motor, a low-power microscope to observe the pointer, and a small ac current in the torque motor to prevent sticking. The balance was calibrated with NBS class-*M* weights.

Data on three foils are reported here—aluminum of 49.9 and 107.8 $\mu g/cm^2$ and copper of 191.8 $\mu g/cm^2$. The stopping powers, after correction for path increase due to straggling and spurious heating due to electrons scattered into the support ring, are plotted as points in Figs. 2 and 3. Also plotted are the Bethe-Bloch stopping-power curves, as discussed in Sec. II.

The error bars represent estimated total probable error, obtained as the square root of the sum of the squares of the errors of each part of the measurement. The experimental points are within three times or less their probable errors (the usual "limit of error") of the theoretical curve.

The only comparable calorimetric measurements we could find were those of Pugachev and Volkov[61] referred to above and plotted with $\times$ on Fig. 2. Their experimental values for aluminum differ from ours by about 10 to 15%, probably because they do not seem to have made any of the three necessary corrections discussed here. They also used only the simple Bethe formula to calculate the stopping powers, instead of the more exact relativistic, quantum-mechanical result as given by Bethe and Ashkin[9] or Rohrlich and Carlson[41] and quoted here as Eq. (1). Our theoretical calculations give stopping-power values about 10% less than Puga-

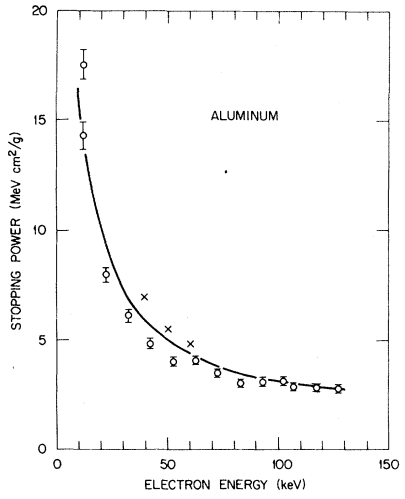

FIG. 2. Stopping power of aluminum for electrons. Points are our experimental results. $\times$ are Pugachev and Volkov's data (Ref. 61). Curve is the Bethe-Bloch formula as calculated by Pages *et al.* (Ref. 17), Berger and Seltzer (Ref. 18), or Eq. (1).

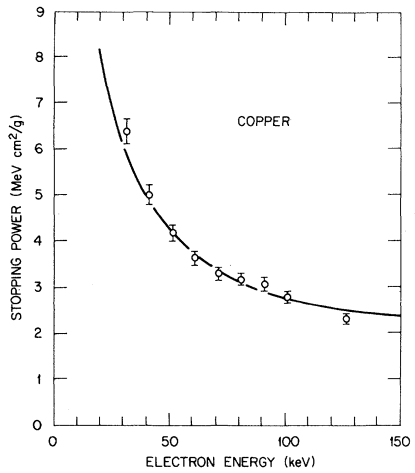

FIG. 3. Stopping power of copper for electrons. See caption to Fig. 2.

chev and Volkov's "theoretical values."

We conclude that the agreement with the Bethe-Bloch stopping-power formula as expressed by Eq. (1) is exact within experimental error over the range of electron energies studied, namely, 11—127 keV, provided proper corrections are made in the experimental data for scattered electrons and path straggling.

Note added in proof. The referee kindly pointed out to us that in Eq. (10) the quantity ν enters as a small correction to $\langle \Delta t \rangle_{av}$, which in turn is a small correction to the path length. Dr. R. H. Ritchie (private communication) suggested to us that a reasonable range of uncertainty in ν is $\pm 10\%$, which produces in the worst case, for Al at 11 keV, an uncertainty of less than 1% in the stopping power, or much less than the probable errors of the experiment. At higher energies the effect of changes in ν is still smaller.

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