approximation for J(0,k), i.e.,

$$V(\mathbf{0},\mathbf{k}) = I(k^2 a^2) + O(ka)^4,$$
 (B3)

where the nearest neighbor distance has been taken to be  $[6(a^2/z)]^{1/2}$  (z is the coordination number). In this manner we have

$$\frac{1}{N}\sum_{\mathbf{k}}\frac{1}{[V+J(\mathbf{0},\mathbf{k})]J(\mathbf{0},\mathbf{k})}\cong\left(\frac{v}{2\pi^{2}Ia^{3}}\right)\int_{0}^{\infty}\frac{dk}{V+I\mathbf{k}^{2}},\quad(\mathrm{B4})$$

PHYSICAL REVIEW

#### VOLUME 132, NUMBER 2

15 OCTOBER 1963

# Ranges of 5- to 27-keV Deuterons in Aluminum, Copper, and Gold\*

R. L. HINES Northwestern University, Evanston, Illinois (Received 7 June 1963)

A method of measuring ranges is presented which uses the change in optical-reflection coefficient of quartz due to radiation damage by the deuterons as a means of detecting the deuterons. The metal films are deposited on the quartz by vacuum evaporation and their thickness is determined with an accuracy of  $\pm 5\%$ by weighing. By bombarding a given metal film on quartz at a fixed flux and a number of different energies, a number of bombarded areas are formed where the deuterons have penetrated to a corresponding number of different depths in the quartz. After chemically removing the metal film, the optical-reflection coefficients of the bombarded areas are measured and compared with theoretical predictions to determine the bombarding energy for which exactly one-half of the incident deuterons have penetrated the metal films. The ranges obtained agree with theoretical estimates and are consistent with known ranges of protons in aluminum.

## INTRODUCTION

T energies in the keV region, the ranges of ions in A materials are due to a combination of many complex phenomena and can be predicted theoretically in only an approximate manner. The ions may lose energy by excitation of electrons in the material or by displacement of atoms of the material. Furthermore, the incident atom may undergo a number of collisions in which it is deflected through large angles. In this case the incident atom motion is more of a diffusion type motion and the concept of range must be broadened to include the concept of a mean penetration depth. These various factors have been discussed in extensive reviews by Bohr,<sup>1</sup> by Seitz and Koehler,<sup>2</sup> by Dienes and Vineyard,<sup>3</sup> and, more briefly, by Nielsen,<sup>4</sup> and by Lindhard and Scharff.<sup>5</sup> All of the theoretical analyses are approximate because of the complex nature of the interaction between the atoms and their electron clouds. For the

ions of interest in this paper, the main energy-loss mechanism in metals is due to the excitation of only the conduction electrons. The incident atoms are moving too slowly to excite the more tightly bound electrons in the inner shells.

where v is the volume per ion  $(v=a^3, sc; \frac{1}{2}a^3, bcc; \frac{1}{4}a^3, sc; \frac{1}{4}a^3, bcc; \frac{1}{4}a^3, sc; \frac{1}{4}a^3, sc;$ 

 $\times \left[ (\beta_c - \beta) + \frac{3S(S+1)}{\hbar} \{\beta \Gamma(T) - \beta_c \Gamma(T_c)\} \right]^2.$ (B5)

fcc). Therefore, Eqs. (B1) and (6.20) give

 $\chi = \left[\frac{9\mu^2(v/a^3)^2}{[\hbar I(1+X(T_c))]^3 \times 128\pi^2\hbar^4 S^4(S+1)^4}\right]$ 

Experimental determination of ranges in the keVenergy region is hampered by the small depth of penetration ( $\sim 10^{-5}$  cm) and difficulties in detecting the ions. From previous work,<sup>6</sup> it is known that low-energy ion bombardment of quartz alters the optical-reflection coefficient of the crystalline quartz. As a result, the quartz can be used as a detector of low-energy ions. A metal film of the desired thickness can easily be evaporated in a vacuum onto the quartz. When the metal film-quartz combination is bombarded with a fixed flux of ions at a number of different energies, the ions which penetrate the metal film will change the reflection coefficient of the quartz. By chemically removing the metal film and measuring the reflection coefficients of the bombarded areas, it is possible to determine the range of the incident ions in the metal film. This paper presents experimental values of ranges of 5- to 27-keV deuterons in aluminum, copper, and gold as obtained by the above method, and compares the experimental values with theoretical predictions.

Previously experimental work on ranges has been

 <sup>\*</sup> Supported by the U. S. Atomic Energy Commission.
 <sup>1</sup> N. Bohr, Kgl. Danske Videnskab. Selskab, Biol. Medd. 18, 8 (1948).

<sup>&</sup>lt;sup>2</sup> F. Seitz and J. S. Koehler, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1956), Vol. 2.

<sup>&</sup>lt;sup>3</sup>G. J. Dienes and G. H. Vineyard, Radiation Effects in Solids

<sup>&</sup>lt;sup>6</sup> J. Dieles and G. H. Villeyard, Naturation Effects in Solids (Interscience Publishers, Inc., New York, 1957).
<sup>4</sup> K. O. Nielsen, *Electromagnetically Enriched Isotopes and Mass Spectroscopy*, edited by M. L. Smith (Academic Press Inc., New York, 1956), p. 68.
<sup>6</sup> J. Lindhard and M. Scharff, Phys. Rev. 124, 128 (1961).

<sup>&</sup>lt;sup>6</sup> R. L. Hines and R. Arndt, Phys. Rev. 119, 623 (1960).

reviewed by Allison and Warshaw.7 Much of the experimental information is presented in the form of measurements of energy loss per unit path length. In particular, Warshaw<sup>8</sup> gives values of energy loss per unit path length of protons in aluminum, copper, silver, and gold from 50 to 400 keV. Range measurements of  $H^+$ ,  $H_{2^+}$ . and He<sup>+</sup> ions in aluminum have been reported by Young.<sup>9</sup> The exact charge state of the incident ion is not important since ions in the keV-energy region undergo many charge-exchange collisions and reach a chargeequilibrated state in a thickness estimated to be only  $0.01 \ \mu g/cm^2.^{10}$ 

#### THEORY

The various mechanisms for energy loss per unit path length dE/dX are dominant in different energy regions.<sup>3</sup> At energies below

$$E = M_1 \epsilon_F / 16m \tag{1}$$

in metals, where E is the incident atom energy,  $M_1$  is the incident atom mass, m is the electron mass, and where  $\epsilon_F$  is the Fermi energy of the metal, the atom loss energy mainly by displacing atoms from their lattice positions. For an insulator such as quartz, the corresponding relation is

$$E = M_1 I / 8m, \qquad (2)$$

where I (6.4 eV for quartz) is the lowest electronicexcitation energy. The energy loss due to displacements can be estimated theoretically in the region  $\epsilon < 1.0$ , where  $\epsilon$ , the screening distance divided by the distance of closest approach, is given by the relation<sup>5</sup>

$$\epsilon = 0.885 a_0 E M_2 / e^2 Z_1 Z_2 (M_1 + M_2) (Z_1^{2/3} + Z_2^{2/3})^{1/2}, \quad (3)$$

here e is the electronic charge,  $a_0 = \hbar^2/me^2$ ,  $Z_1$  is the atomic number of the incident atom, and  $Z_2$  and  $M_2$  are, respectively, the atomic number and mass of the stopping material. For values of  $\epsilon < 1.0$  the energy loss per unit path length is independent of energy. At energies above that given by (1) the incident atom loses energy in metals by excitation of the conduction electrons. The value of dE/dX is estimated theoretically by considering the energy loss of a low velocity atom moving through an electron gas of constant density. Electron excitation is allowed only if the scattered electron lands in an unoccupied state. Using a Thomas-Fermi treatment, the energy loss per  $g/cm^2$  of stopping material is<sup>5</sup>

$$dE/dX = N_0 \rho 8\pi e^2 a_0 Z_1 Z_2 \xi_{\epsilon} (Z_1^{2/3} + Z_2^{2/3})^{-3/2} (v/v_0), \quad (4)$$

where  $N_0$  is the number of atoms per cm<sup>3</sup> of stopping material,  $\rho$  is the density, v is the velocity of the incident atom, and  $v_0 = e^2/\hbar$ . The parameter  $\xi_{\epsilon}$  contains the details of the interaction and is estimated to be on

the order of 1.0. Equation (4) is valid for atom energies such that

$$v < v_0$$
. (5)

At higher atom energies the excitation of inner shell electrons becomes possible and the problem still more complex.

#### **RANGE DETERMINATION**

The central feature of the method of range determination employed here is that the reflection coefficient of a quartz surface is used as a detector to determine the residual range in the quartz of the atoms which have penetrated the metal film deposited on the surface. The range  $\Re$  is defined as the depth where the atom flux has decreased to one half of its incident value. Starting with the approximation that the incident atoms will come to rest with a Gaussian distribution, the time integrated atom flux density F(X) at a depth X from the metal film surface is given by

$$F(X) = F_{0\frac{1}{2}}\{1 - \operatorname{erf}[(X - \mathfrak{R})/\sqrt{2}\Omega]\} \quad \text{for} \quad X < t, \quad (6)$$

where  $F_0$  is the incident atom flux, t is the film thickness, and  $\Omega$  is the rms fluctuation in particle penetration. The atoms lose most of their energy in the metal film and enter the quartz with a greatly reduced energy,  $E_t$ . The distribution of the atom beam in space and energy and, hence,  $\Omega$  will be determined primarily by the metal film. However, an ion with energy  $E_t$  entering the quartz will travel a distance X' that is different from the distance (X-t) that the ion would have traveled if it had continued on in the metal. For values of  $E_t$  such that  $\epsilon < 1$ , the energy loss per unit path length is constant and the distances are related by the equation

$$X = t + \alpha X', \tag{7}$$

where  $\alpha = \langle dE/dX \rangle_{\text{quartz}} / \langle dE/dX \rangle_{\text{metal}}$  for  $E < E_t$ , where  $\langle dE/dX \rangle$  is the average value of dE/dX for  $0 < E < E_t$ . If d is the depth beneath the quartz surface where the atom flux drops to  $\frac{1}{2}F_0$ , then the atom flux in the quartz is given by the relation

$$F(X') = F_{02} \{1 - \operatorname{erf}[\alpha(X' - d)/\sqrt{2}\Omega]\}.$$
(8)

From previous work<sup>6</sup> it is seen that the change in refractive index,  $\Delta n(F)$ , as a function of the time integrated ion flux density can be approximated by the relation

$$\Delta n(F) = (F/F_{\mathcal{R}}) \Delta n(F_{\mathcal{R}}) \quad \text{for} \quad 0 < F < F_{\mathcal{R}}, \quad (9)$$

where  $F_{\mathfrak{R}}$  is determined by the relation

$$\Delta n(F\mathfrak{R}) = 2\Delta n(\frac{1}{2}F\mathfrak{R}). \tag{10}$$

By substituting (9) into (8) the change in refractive index in the quartz is found, as a function of depth, to be

$$\Delta n(X') = \Delta n(F \mathfrak{R}) \{ 1 - \operatorname{erf}[\alpha(X' - d)/\sqrt{2}\Omega] \}.$$
(11)

The effect on the reflection coefficient of this variation

<sup>&</sup>lt;sup>7</sup>S. K. Allison and S. D. Warshaw, Rev. Mod. Phys. 25, 779 (1953).
<sup>8</sup> S. D. Warshaw, Phys. Rev. 76, 1759 (1949).
<sup>9</sup> J. R. Young, J. Appl. Phys. 27, 1 (1956).
<sup>10</sup> S. K. Allison, Rev. Mod. Phys. 30, 1137 (1958).

in refractive index as a function of depth can be calculated following the method of Rouard.<sup>11</sup> The calculated values of interest are shown in Fig. 1. As can been seen, the reflection coefficient ratios  $R/R_0$  are complicated functions of both d,  $\Omega$ , and  $\lambda$  the light wavelength. The range of wavelengths which are measured here ( $\lambda=450$ to 650 m $\mu$ ) is too small to permit the independent determination of d and  $\Omega$  from a bombardment at a single energy. However, the fact that the different curves coincide at the inflection point provides a test to determine the energy  $E_{\alpha}$  for which the atom range is equal to the metal film thickness (i.e., d=0). If the bombardment energy  $E_0$  exceeds  $E_{\alpha}$ , then the value of d will be given approximately by the relation]

$$E_0 - E_{\Re} = d\langle dE/dX \rangle_{\text{quartz}}.$$
 (12)

The actual value of  $\langle dE/dX \rangle_{\text{quartz}}$  is not required to find



FIG. 1. The reflection-coefficient ratio  $R/R_0$ , for a bombarded surface relative to a nonbombarded surface, at a wavelength  $\lambda$ . The values shown are calculated for quartz (n=1.547) bombarded by ions with a residual range d, a straggling parameter  $\Omega$ , and where  $\Delta n = -0.067$  is the maximum change in refractive index due to the ion bombardment. The parameter  $\alpha$  is the ratio of the energy loss per unit path length of the ions in quartz to the energy loss per unit path length of the ions in the metal film on the quartz surface.

Eq. However,  $(dE/dX)_{\text{quartz}}$  should be approximately constant in order for (12) to be valid. As will be shown later, it can be seen that the energy of the deuterons entering the quartz is  $E_{\text{T}}=3$  keV for  $\lambda=450$  mµ. The energy for which  $\epsilon=1$  is 0.8 keV for a deuteron in quartz. However, Lindhard and Sharff<sup>5</sup> point out that for the larger values of  $\epsilon$  the decrease in dE/dX for displacements is compensated for by the increase in dE/dX due to ionization with the result that the total dE/dX remains approximately constant for values of  $\epsilon$ considerably larger than 1.0.

When a given metal film on quartz is bombarded at a



FIG. 2. Experimental reflection-coefficient ratios as a function of bombarding deuteron energy divided by wavelength for a quartz surface covered by a copper film of  $81-\mu$  g/cm<sup>2</sup> thickness. The copper film is removed chemically before measuring the reflection-coefficient ratios.

number of different energies, a number of bombarded areas are produced with different values of d. If the reflection-coefficient ratios are plotted as a function of  $E_0/\lambda$ , then the experimental information has the form shown in Fig. 2. These curves differ from those shown in Fig. 1 only in that the abscissa is  $E_0/\lambda$  instead of  $d = (E_0 - E_{\text{K}})/\lambda \langle dE/dX \rangle_{\text{quartz}}$ . The range determination now becomes a matter of choosing  $E_{\text{K}}$  such that the experimental curves of  $R/R_0$ , when plotted as a function of  $(E_0 - E_{\text{K}})/\lambda$ , are shifted to a set of curves which overlap at the inflection point as do the curves in Fig. 1. The curves shown in Fig. 3 are the data given in Fig. 2



FIG. 3. The experimental values of  $R/R_0$  given in Fig. 2 are replotted as a function of  $(E_0-5.3)/\lambda$ . Comparison of Figs. 1 and 3 shows that the energy for which deuterons have a range of  $81 \ \mu g/cm^2$  in Cu is 5.3 keV.

<sup>&</sup>lt;sup>11</sup>O. S. Heavens, Optical Properties of Thin Solid Films (Academic Press Inc., New York, 1955), p. 63.

replotted with  $E\mathfrak{a}=5.3$  keV. As mentioned earlier, it can be seen that within the region of interest, up to the first minimum in  $R/R_0$ , the maximum deuteron energy entering the quartz is  $E_0 - E\mathfrak{a} = E_i = 3$  keV for  $\lambda = 450$ m $\mu$ . The parameter  $\Omega/\alpha\lambda$  is different for each value of  $\lambda$ even though  $\Omega$  and  $\alpha$  are constant. The actual procedure used to determine  $E\mathfrak{a}$  is to plot the graphs of  $R/R_0$ versus  $E_0/\lambda$  on separate sheets of paper for each value of  $\lambda$  and to mark the various trial values of  $E\mathfrak{a}/\lambda$  on all the graphs. Then by superimposing the graphs, the value of  $E\mathfrak{a}$  which gives the best fit of the curves at the inflection point can be rapidly chosen.

#### EXPERIMENTAL APPARATUS AND PROCEDURE

The ion-bombardment apparatus consists of a cold cathode-ion source, an analyzer magnet, sweep plates to obtain a uniform current density ion beam, a main accelerating gap, and a target region which can be isolated with a gate value. A more detailed description of the apparatus is given elsewhere.<sup>6</sup> The pressure in the target region during bombardment is on the order of  $2 \times 10^{-5}$  mm Hg.

The target geometry is shown in Fig. 4. The quartz sample is held in place by four pointed screws pressing against its sides. The current contact to the metal film is made with Aquadag. Secondary electrons created by ions striking the edges of the 1-mm-diam hole in the aperture plate are repelled by the shield plate. The number of secondary electrons from the target which pass back through the 1.5-mm-diam hole in the shield plate is negligible with the geometry shown. All adjustments of the ion beam are made with the shutter in place. During bombardment the shutter is moved completely out of the path of the ion beam. Beam current is monitored during bombardment with a dc amplifier. Most of the bombardments employed a beam current of  $D_2^+$  ions of  $0.100 \ \mu A$  for 90 sec. After each bombardment of a given metal film, the quartz is moved to expose a new area of film. Most of the samples are bombarded at eight different energies. Each bombarded spot is identified by its coordinate position on the sample surface.

The metal films are deposited on the quartz in a standard vacuum system using a well baffled oil diffusion pump. Pressures during evaporation are on the order of  $2 \times 10^{-5}$  mm Hg. Evaporation rates are 0.5 µg/cm<sup>2</sup> sec for aluminum and 1.0 µg/cm<sup>2</sup> sec for copper and gold. All samples are bombarded within a few days after the evaporation. The oxide films on the aluminum and the copper<sup>12</sup> (1.8 µg/cm<sup>2</sup>) are negligible for the film thicknesses employed here. The quartz sample is surrounded



<sup>12</sup> S. Roberts, Phys. Rev. 118, 1509 (1960).

in the same plane by four monitor cover glass slides (18 mm×18 mm) and is located 10 cm from the evaporation source. The monitor slides are weighed before and after evaporation to an accuracy of  $\pm 2 \mu g$ . Deviations in the evaporating flux as a function of angle limits the accuracy of the metal film thickness determination to  $\pm 5\%$ . Monitor slides subjected to the usual vacuum cycle, but without metal evaporation showed a weight gain of 2  $\mu$ g/cm<sup>2</sup> which is probably due to oil deposition on the slides. No correction is made for the oil film because it is negligible for all except the thinnest aluminum films. In addition, the stopping power for a given weight per cm<sup>2</sup> should be approximately the same for aluminum and oil. After bombardment the aluminum films are removed with sodium hydroxide and the copper and gold films are removed with aqua regia. The quartz is then scrubbed with a detergent, rinsed thoroughly, and dried with a blast of filtered compressed air. The surfaces are examined visually at low magnification to be sure that all of the metal film is removed.

The reflection coefficients of the quartz samples are measured using a vertical illumination microscope with the eyepiece replaced by a photovoltaic cell. A  $10 \times$ , 0.40 N.A. objective is used for all measurements. Measurements at different wavelengths are obtained using monochromatic interference filters (650, 600, 550, 500, and 450 m $\mu$  with a half-width of 18 m $\mu$ ) and a tungsten filament light source. The complete system is described in detail elsewhere.<sup>6</sup> After locating the bombarded region, and selecting an appropriate region which has not been bombarded, readings are taken of the light intensity reflected from the bombarded region, the nonbombarded region, and the background reading due to light reflected from the optical surfaces in the objective. These readings are then used to calculate  $R/R_0$ . Measurements of  $R/R_0$  are reproducible to within  $\pm 0.5\%$ .

The procedure used to determine the correct bombarding energies and flux for a given metal film is a trial and error process. For the very first bombardments, a copper film of 85  $\mu$ g/cm<sup>2</sup> is bombarded at D<sub>2</sub><sup>+</sup> energies of 13, 16, 19, 22, 25, and 28 keV with a flux of  $9.0 \times 10^{15}$ ions/cm<sup>2</sup> which is the estimated value of  $F_{\mathcal{R}}$  in (10). After measurement of the reflection coefficient ratios, it is found that the  $D_2^+$  energy which gives a residual range in the quartz of d=0 is  $E_{R}=9$  keV. Then a second copper film of about the same thickness ( $t=71 \ \mu g/cm^2$ ) is bombarded with 16-keV  $D_2^+$  ion fluxes of 2.25, 4.50, 6.75, 9.00, 11.3, and 13.6×10<sup>15</sup> ions/cm<sup>2</sup>. Measurement and analysis of the reflection-coefficient ratios of the bombarded areas gives a graph similar to that previously observed for ion bombarded quartz,6 and from this graph the value  $F_{\alpha}=6.75\times10^{15}$  D<sub>2</sub><sup>+</sup> ions/cm<sup>2</sup> is obtained using (10). This value of  $F_{\mathfrak{R}}$  is used in all subsequent bombardments.

For a known thickness of metal film, the value of Ea is estimated and a series of bombardments carried out

at 2-keV intervals from  $E_0 = E\alpha - 4$  to  $E_0 = E\alpha + 10$ . The actual value of  $E\alpha$  for that film thickness is determined using the procedure outlined above. The results are reliable as long as the actual value  $E\alpha$  falls within the range of bombardment energies employed. If  $E\alpha$  does not fall within the bombarding energies employed, then a new metal film is prepared and bombarded in accordance with a revised estimate of  $E\alpha$ . For each of the metals, three or four preliminary trial samples are required to determine the general shape of the range energy curve.

#### **RESULTS AND DISCUSSION**

The experimental results are presented in Table I and in Fig. 5. Actually,  $D_2^+$  ions are used in most of the bombardments to obtain sufficient ion fluxes, especially at low ion energies. However, the binding energy be-

TABLE I. The energy  $E\mathfrak{A}$  required for 50% of the incident ions to penetrate the thickness of stopping-material given. The value of dE/dX is computed by fitting a straight line through the adjacent points.

Stopping material	Thickness (µg/cm²)	Ion	Energy <i>E</i> & (keV/deuteron)	dE/dX (keV/ $\mu$ g cm <sup>-2</sup> )
Al Al Al Al Al Al Cu Cu Cu Cu	$\begin{array}{c} 29.1 \pm 1.5 \\ 38.3 \pm 1.9 \\ 51.5 \pm 2.6 \\ 70.1 \pm 3.5 \\ 72.2 \pm 3.6 \\ 84.3 \pm 4.2 \\ 84.7 \pm 4.2 \\ 81 \pm 4 \\ 120 \pm 6 \\ 156 \pm 8 \end{array}$	$\begin{array}{c} D_{2}^{+} \\ D_{2}^{+} \end{array}$	$\begin{array}{c} 5.3 \pm 0.5 \\ 6.3 \pm 0.3 \\ 10.3 \pm 0.3 \\ 13.8 \pm 0.5 \\ 14.3 \pm 0.5 \\ 20.0 \pm 0.5 \\ 20.8 \pm 0.5 \\ 5.3 \pm 0.5 \\ 9.3 \pm 0.3 \\ 12.3 \pm 0.3 \end{array}$	0.210
Cu Cu Au Au Au Au	$\begin{array}{rrrr} 216 & \pm 11 \\ 283 & \pm 14 \\ 158 & \pm 8 \\ 226 & \pm 11 \\ 357 & \pm 19 \\ 462 & \pm 23 \end{array}$	$D_{2}^{+}$ $D_{2}^{+}$ $D_{2}^{+}$ $D_{2}^{+}$ $D_{2}^{+}$ $D_{2}^{+}$	$17.5 \pm 0.5 \\ 27.0 \pm 0.5 \\ 5.3 \pm 0.3 \\ 8.0 \pm 0.5 \\ 16.0 \pm 0.5 \\ 22.5 \pm 1.0$	0.062

tween the atoms in the  $D_2^+$  ion is very small compared with the energies involved in collisions with atoms of the stopping material and the  $D_2^+$  ion is split as soon as it enters the stopping material. The two atoms in the  $D_2^+$ ion have the same initial velocity and will have equal energies. Thus, the effect of a  $D_2^+$ -ion beam is the same as a D<sup>+</sup>-ion beam of one half the energy and twice the flux. This concept is verified by experiments on the charge and energy distribution of  $H_2^+$  and  $H^+$  beams emerging from thin foils.<sup>13</sup> All of the results in Fig. 5 and Table I are presented in terms of energy per deuterium atom. As an additional check, the energy for a range of  $82 \,\mu g/cm^2$  in aluminum is determined separately using  $D_2^+$  ions and  $D^+$  ions and the results agree within experimental error. The errors shown for ER are estimates of the possible errors in fitting the  $R/R_0$  versus



FIG. 5. Ranges of deuterons in aluminum, copper, and gold as a function of energy.  $D_2^+$  ions are used in the actual bombardment, but the molecular bond is split as soon as the ion strikes the metal and the energy divides equally between the two atoms. Consequently, the results are plotted here with an energy which is exactly one half of the actual  $D_2^+$  bombarding ion energy.

 $E_0/\lambda$  curves at the inflection point to determine  $E\mathfrak{a}$ . As mentioned above, the exact charge state of the incident atom is not important because charge equilibration takes place within a distance of only 0.01  $\mu$ g/cm<sup>2</sup> for ions in this energy range.

Although the effect of straggling is taken into account in the analysis, it is not possible to determine  $\Omega$  with any precision since it enters into the analysis only in the form of  $(\Omega/\alpha\lambda)$  and  $\alpha$  as given by (7) is an unknown constant. However, an estimate of  $\Omega$  can be obtained in the following manner. From Eq. (3) given by Lindhard and Scharff<sup>5</sup> it is seen that the dependence of dE/dX on the stopping material enters only as  $N_0Z_2^{-1/3}$  for  $Z_1 \ll Z_2$ and  $\epsilon < 1$ . With this relation the value of  $\alpha$  is calculated to be 1.2 for aluminum, 1.1 for copper, and 2.2 for gold. By comparison of experimental curves like those shown in Fig. 3 with the theoretical curves shown in Fig. 1, an estimate of  $(\Omega/\alpha\lambda)$  can be made. Using the above values of  $\alpha$ , the values of  $\Omega$  turn out to be  $0.008 \pm 0.008 \mu$  for aluminum,  $0.025 \pm 0.008 \,\mu$  for copper, and  $0.020 \pm 0.008 \,\mu$ for gold. There is some indication that the straggling increases as a function of energy. The experimental reflection coefficient ratios do not give a sensitive determination of the straggling, but it must be taken into account in order to obtain accurate values for the ranges.

The dependence of the range on displacement production, ionization, and large angle scattering events makes any analysis of ranges in this energy region exceedingly complex. However, the data presented here can be used to obtain values of dE/dX in the energy region where (4) should be valid. Table I presents experimental values obtained by fitting a straight line through the

705

<sup>&</sup>lt;sup>13</sup> Ia. M. Fogel, B. G. Safronov, and L. I. Krupnik, Zh. Eksperim. i Teor. Fiz. 28, 711 (1955) [translation: Soviet Phys.—JETP 1, 547 (1955)].

points adjacent to the points for which the value is given. By comparison of (4) and the experimental values of dE/dX given in Table I, experimental values of  $\xi_{\epsilon}$  can be found. These values turn out to be  $\xi_{\epsilon}=0.57$  for aluminum,  $\xi_{\epsilon}=0.50$  for copper, and  $\xi_{\epsilon}=0.82$  for gold which are in reasonable agreement with the theoretical estimate of  $\xi_{\epsilon}=1$ .

The ranges reported here can be compared with values reported for protons since the energy loss due to ionization per unit path length is the same for deuterons and protons of the same velocity. However, the deuteron has twice the energy of a proton of the same velocity. Thus, the range of a deuteron should be twice the range of a proton of one half the deuteron energy. This argument neglects the differences in energy loss due to displacements that might be expected between protons and deuterons. However, from (1) the energy loss due to displacements predominates only below E=1.26 keV for deuterons which is a small fraction of the total deuteron energy employed here. The range of a 15-keV deuteron of  $73 \,\mu g/cm^2$  in aluminum as reported here can be compared with twice the range reported by Young<sup>9</sup> for 7.5-keV protons. The actual values of twice the range in aluminum reported by Young are 76  $\mu$ g/cm<sup>2</sup> for H<sub>2</sub><sup>+</sup> ion bombardment with 7.5 keV per atom, and 62  $\mu$ g/cm<sup>2</sup> for 7.5-keV H<sup>+</sup> ion bombardment. This agreement between the two experiments is as close as can be expected considering the assumptions involved in the comparison.

#### CONCLUSIONS

Ranges of ions in materials that can be evaporated into thin films on quartz can be found using the radiation damage in quartz to detect the ions. Using this method, 5- to 27-keV deuteron ranges in aluminum, copper, and gold are bound to agree with theoretical predictions of energy loss per unit path length and with known experimental ranges of protons in aluminum.

## ACKNOWLEDGMENTS

The author is indebted to R. Arndt for his assistance during the preliminary stages of the experiment, to R. T. Smith for his assistance in preparing the evaporated metal films, and to H. E. Beck for weighing the monitor slides used in the experiment.

PHYSICAL REVIEW

#### VOLUME 132, NUMBER 2

**15 OCTOBER 1963** 

# Electric Quadrupole Interactions in Rare-Earth Ions

A. J. Freeman

National Magnet Laboratory,\* Massachusetts Institute of Technology, Cambridge, Massachusetts, and U. S. Army Materials Research Agency, Watertown, Massachusetts

AND

# R. E. WATSON

Bell Telephone Laboratories, Murray Hill, New Jersey, and Atomic Energy Research Establishment, Harwell, England (Received 10 June 1963)

Electric quadrupole interactions in rare-earth ions arising from electric field gradients due to valence electrons  $(q_{4f})$  and/or crystal fields external to the ion  $(q_{lat})$  are discussed. The contribution of the Sternheimer antishielding factors to both  $q_{4f}$  and  $q_{lat}$  are calculated for the Ce<sup>3+</sup> ion using the method (based on the unrestricted Hartree-Fock formalism) reported earlier. The ionic antishielding factor ( $\gamma_{\infty}$ ) is -73.5, and it is found that the total ionic contribution to the electric field gradient can be comparable in magnitude (and may be opposite in sign) with the valence electron contribution (augmented by the internal antishielding factor). The possible relation of these results to the temperature dependence of measured electric quadrupole interactions is noted.

## I. INTRODUCTION

**M**<sup>EASUREMENTS</sup> of hyperfine interactions in rare-earth ions, by specific heat, electron spin resonance, Mössbauer, and other methods, have revealed very large electric quadrupole contributions.<sup>1</sup> In addition, unusual temperature-dependent electric field gradients, including a change in sign,<sup>2</sup> have been reported. These experiments have pointed out the need for a deeper theoretical understanding of the origin of electric quadrupole interactions in these materials, and more particularly, of the role played by the distor-

<sup>\*</sup> Supported by the U. S. Air Force Office of Scientific Research. <sup>1</sup> See B. Bleaney, Suppl. J. Appl. Phys. **34**, 1024 (1963); J. Phys. Soc. Japan **17**, Suppl. B-I, 435 (1962) for a recent review of these data. See R. Bauminger, S. G. Cohen, A. Marinov, and S. Ofer, Phys. Rev. Letters **6**, 467 (1961); and R. Cohen, V. Hauser, and R. L. Mössbauer, in *The Mössbauer Effect*, edited by D. M. J.

Compton and A. H. Schoen (John Wiley & Sons, Inc., New York, 1962), for some Mössbauer data on the temperature dependence of the electric quadrupole interaction. <sup>2</sup> R. L. Cohen, Ph.D. thesis, California Institute of Technology,

<sup>&</sup>lt;sup>2</sup> R. L. Cohen, Ph.D. thesis, California Institute of Technology, 1962 (unpublished); see in particular, Fig. 18.