

These equations form together a linear system of order $(p+1)$ in the unknown coefficients $c_0, c_1, c_2, \dots, c_p$, and this system has a nontrivial solution only if its determinant vanishes:

$$\begin{vmatrix} g_n & g_{n+1} & g_{n+2} & \cdots & g_{n+p} \\ g_{n+1} & g_{n+2} & g_{n+3} & \cdots & g_{n+p+1} \\ \cdots & \cdots & \cdots & \cdots & \cdots \\ g_{n+p} & g_{n+p+1} & g_{n+p+2} & \cdots & g_{n+2p} \end{vmatrix} = 0. \quad (5)$$

In general a symmetric determinant of the special type

$$H_n(m) = \begin{vmatrix} g_n & g_{n+1} & g_{n+2} & \cdots & g_{n+m} \\ g_{n+1} & g_{n+2} & g_{n+3} & \cdots & g_{n+m+1} \\ \cdots & \cdots & \cdots & \cdots & \cdots \\ g_{n+m} & g_{n+m+1} & g_{n+m+2} & \cdots & g_{n+2m} \end{vmatrix}, \quad (6)$$

formed by $(2m+1)$ consecutive entries, is called a *Hankel determinant*³; it has the order $(m+1)$. We can now write Eq. (5) in the simple form

$$H_n(p) \equiv 0, \quad (7)$$

and a condition for the possibility of expressing the series of given entries $g_0, g_1, g_2, \dots, g_n, \dots$ as a sum of exactly p geometrical series is then that *all* the Hankel determinants $H_n(p)$ associated with the material should vanish and that at least one of the determinants $H_n(p-1)$ should be different from zero.

We note that, in such a case, the determinants $H_n(m)$ vanish also for $m=p+1, p+2, \dots$ and that none of them have a *rank* higher than p . In fact, the number p of exponentials associated with the given series $g_0, g_1, g_2, \dots, g_N$ is expressed by the rank of the highest Hankel matrix which may be constructed; this matrix has $[\frac{1}{2}N]$ columns and $[\frac{1}{2}(N-1)]$ rows. In practice, it would be possible to determine the rank for instance by a semidiagonalization procedure⁴ which leaves the rank of the matrix invariant,⁵ but the numerical process is influenced by the occurrence of approximations in the entries in the form of rounding-off errors, experimental uncertainties, etc. In order to fix a practical value for the rank p , it may therefore be necessary to neglect certain small quantities, which introduces corresponding errors in the final result. Suitable values of the coefficients $c_0, c_1, c_2, \dots, c_p$, which are good in "average" have then to be determined.⁶ For the sake of simplicity, however, we will here assume that the rank p could be exactly evaluated.

Even the Hankel determinants of order p and lower are of interest, and it should particularly be observed

³ H. Hankel, Leipziger Dissertation, Göttingen, 1861; see also G. Kowalewski, *Determinantentheorie* (Veit und Compagnie, Leipzig, 1909), p. 112.

⁴ See, e.g., the semidiagonalization technique described in W. E. Milne, *Numerical Calculus* (Princeton University Press, Princeton, 1949), pp. 17–26, which is directly applicable also to matrices.

⁵ See, e.g., G. Kowalewski, reference 3, p. 49.

⁶ Instead of the "least-squares method" described by Runge and König, reference 2, we would for optimum accuracy propose a minimization procedure leading to an eigenvalue problem; this technique will be further described in a forthcoming paper.

that, if the series $H_n(p)$ contains only vanishing elements for $n=0, 1, 2, \dots$, the quantities $H_n(p-1)$ form a single geometric series. In order to evaluate the lower determinants, let us introduce the generalized forward-difference operator $\Delta(q) = E - q$, where E is the step-operator defined by $Eg_n = g_{n+1}$ and q is an arbitrary constant. Hence we have

$$\begin{aligned} \Delta(q)g_n &= g_{n+1} - qg_n, \\ \Delta^2(q)g_n &= g_{n+2} - 2qg_{n+1} + q^2g_n, \\ &\cdots \quad \cdots \quad \cdots \quad \cdots \end{aligned} \quad (8)$$

and, by carrying out suitable rearrangements³ of rows and columns in the determinant (6), we obtain the *Hankel transformation*:

$$H_n(m) = \begin{vmatrix} g_n & \Delta g_n & \Delta^2 g_n & \cdots & \Delta^m g_n \\ \Delta g_n & \Delta^2 g_n & \Delta^3 g_n & \cdots & \Delta^{m+1} g_n \\ \cdots & \cdots & \cdots & \cdots & \cdots \\ \Delta^m g_n & \Delta^{m+1} g_n & \Delta^{m+2} g_n & \cdots & \Delta^{m+m} g_n \end{vmatrix}, \quad (9)$$

where the Hankel-type of the determinant is preserved. We can now easily evaluate the determinants $H_n(m)$ for $m=1, 2, \dots, p-1$ with the elements (2) by successively carrying out a series of Hankel transformations (9) with $q=k_1, q=k_2-k_1, q=k_3-k_2, \dots$ respectively, which procedure makes it possible to separate the contributions coming from $\alpha_1, \alpha_2, \alpha_3, \dots$ etc. This gives the formulas

$$\begin{aligned} H_n(1) &= \sum_{r < s} \alpha_r \alpha_s (k_r - k_s)^2 (k_r k_s)^n, \\ H_n(2) &= \sum_{r < s < t} \alpha_r \alpha_s \alpha_t (k_r - k_s)^2 (k_s - k_t)^2 (k_r k_s k_t)^n, \\ &\cdots \end{aligned} \quad (10)$$

$$H_n(p-1) = \alpha_1 \alpha_2 \cdots \alpha_p (k_1 k_2 k_3 \cdots k_p)^n \prod_{r < s} (k_r - k_s)^2,$$

where the last one shows that the quantities $H_n(p-1)$ form a single geometric series with the quotient equal to the product $k_1 k_2 \cdots k_p$.

Evaluation of coefficients and exponents.—After determining the number p of exponentials, we will now say a few words about the evaluation of the quotients k_1, k_2, \dots, k_p and the corresponding coefficients $\alpha_1, \alpha_2, \dots, \alpha_p$ in (2). Instead of explicitly calculating the quantities $c_0, c_1, c_2, \dots, c_p$ in (3) from the system (4), it may be more convenient to combine (3) with the first p relations (4) to a form a system with $(p+1)$ equations with the determinant

$$\begin{vmatrix} 1 & k & k^2 & \cdots & k^p \\ g_n & g_{n+1} & g_{n+2} & \cdots & g_{n+p} \\ g_{n+1} & g_{n+2} & g_{n+3} & \cdots & g_{n+p+1} \\ \cdots & \cdots & \cdots & \cdots & \cdots \\ g_{n+p-1} & g_{n+p} & g_{n+p+1} & \cdots & g_{n+2p-1} \end{vmatrix} = 0. \quad (11)$$

This is an implicit form of the algebraic equation (3), which may be expanded by the standard semidiagonalization procedure.

The coefficient α_1 may be found by considering the cofactor of g_{n+2p-1} in the left-hand side of (11) for $k=k_1$. Carrying out a series of Hankel transformations, we obtain finally

$$\alpha_1(k_2-k_1)(k_3-k_1)\cdots(k_p-k_1)k_1^n$$

$$=H_n(p-1) \begin{vmatrix} 1 & k_1 & k_1^2 & \cdots & k_1^{p-1} \\ g_n & g_{n+1} & g_{n+2} & \cdots & g_{n+p-1} \\ \cdots & \cdots & \cdots & \cdots & \cdots \\ g_{n+p-2} & g_{n+p-1} & g_{n+p} & \cdots & g_{n+2p-3} \end{vmatrix}. \quad (12)$$

Similar formulas hold for the other coefficients.

So far we have assumed that our material is non-degenerate, i.e., that all the quotients are different. A degeneracy may be considered as a limiting case where two or more exponents tend to be the same and, as in the theory of differential equations, this corresponds to the introduction of terms containing powers of r of the same degree as the order of the degeneracy. The occurrence of terms of the type

$$\exp(-a_v r) \{A_v + A_v' r + A_v'' r^2 + \cdots\} \quad (13)$$

is therefore indicated by the appearance of *multiple roots* in the k equation (11), and a useful formula for the coefficient for the highest power r may be obtained by carefully carrying out the limiting procedure in (12).

In conclusion it should be observed that the exponents a_v also may be complex, which implies that the method described could be used for expressing a given material in terms of exponentials, sine, and cosine functions combined with each other and with powers of the variable r .

II. PRACTICAL METHOD FOR DETERMINING AN ANALYTIC FIT TO A SCF FUNCTION

(a) Effect of Rounding-Off Errors in the Exact Method

From our previous experience¹ we know that the SCF function for $\text{Na}^+(2p)$ may be approximated by three exponentials with almost perfect accuracy. According to the exact method, one could therefore expect that the six parameters involved could be determined from six consecutive entries of the function $g_{2p}(r) = f_{2p}(r)/r^2$ giving rise to a six-point analysis. However, there are some additional complications which will here be illustrated by an example.

Let us start from the numerical material for $\text{Na}^+(2p)$ given by Hartree and Hartree,⁷ and let us select the six points $r=0.2(0.2)1.2$; the entries are given in the first part of Table I. From the algebraic equation (11), we get the following three quotients:

$$k_1=1.670, \quad k_2=2.796, \quad k_3=8.455, \quad (14)$$

but, even if the corresponding analytic function passes

TABLE I. Numerical values for the function $g(r)=f(r)/r^2$ for $\text{Na}^+(2p)$; (a) values taken from Hartree-Hartree, reference 7; (b) values taken from the analytic function (15) correctly rounded to four decimals.

r	(a)	(b)
0.2	14.45	14.4882
0.4	6.231	6.2232
0.6	2.947	2.9455
0.8	1.494	1.4971
1.0	0.7990	0.8008
1.2	0.4444	0.4451

nicely through the six given points, it deviates considerably in a large number of other points.

In order to test the value of the six-point analysis itself, we will then consider a function $g_{2p}^*(r)=f_{2p}^*(r)/r^2$ which is exactly defined as the sum of three exponentials:

$$g_{2p}^*(r) = 3.6164e^{-2.1880r} + 15.660e^{-3.7288r} + 18.729e^{-6.8864r}. \quad (15)$$

The function $f_{2p}^*(r)$ which is actually taken from Table XII of Part I, does not deviate from the Hartree-Hartree function $f_{2p}(r)$ by more than ± 0.002 . The three quotients for $h=0.2$ corresponding to the exponentials in (15) are

$$k_1=1.549, \quad k_2=2.108, \quad k_3=3.964. \quad (16)$$

In order to see whether the six-point analysis would accurately reproduce these quotients, we have considered the values of (15) in the same six points as before: $r=0.2(0.2)1.2$; the numerical values of g_{2p}^* correctly abbreviated to four decimals are given in the second part of Table I. The solution (11) gave the three roots:

$$k_1=1.500, \quad k_2=2.031, \quad k_3=3.877, \quad (17)$$

which shows that the exact quotients (16) are only roughly obtained. This effect depends on the fact that, in evaluating the coefficients c_v in the algebraic equation (3) or (11), there is a certain cancellation of significant figures which correspondingly raises the importance of the rounding-off errors.

In general, we have found that the lower Hankel determinants are rather sensitive to the rounding-off errors. As an example, we have given in Table II the values of $H_n(1)$ associated with the function $g_{2p}^*(r)$ defined exactly by (15); if $f_{2p}^*=r^2g_{2p}^*$ is expressed with six decimals, the quantities $H_n(1)$ show a regular behavior and their quotients tend to a definite limit equal to $k_1k_2=3.26533$. However, if f_{2p}^* is correctly abbreviated to only three decimals as in most SCF tables, the quantities $H_n(1)$ will be appreciably influenced with a highly irregular quotient series without any obvious limit, as is shown by the two last columns of Table II. The higher Hankel determinants for $m=2, 3, \dots$ are still more irregular, and it is immedi-

⁷ D. R. Hartree and W. Hartree, Proc. Roy. Soc. (London) A193, 299 (1948).

ately clear that if the exact method should be of any use, it is necessary to take the influence of the rounding-off errors into full account. The resulting exponents and coefficients for the exponentials involved will apparently change appreciably if the last figure in the SCF function $f_{nl}(r)$ is changed by half a unit. In principle, there may exist a certain "best" analytic fit to the SCF function but, from the practical point of view, there exists a large family of functions with coefficients and exponents varying over a rather large domain which are almost just as good. The instability problem occurring here is of essential interest and will be treated further in another paper. Here we will instead confine ourselves to giving a single analytic approximation of almost perfect accuracy, i.e., one which deviates from the given SCF function by quantities smaller than 0.001–0.002.

(b) Method of Successive Approximations Based on 4-Point Analyses

We have seen that the numerical material associated with the SCF functions is not so easily handled by the exact method described above. Instead we will now describe a method of successive approximations, where we have borrowed certain elements from the exact theory. Such a method was described in Part I: one works inwards from the tail $r \approx \infty$ towards the origin $r=0$ and, in each step, a single exponential is determined. This method can now be essentially improved by determining a pair of exponentials in each step by means of a 4-point analysis based on Eq. (11) for $p=2$. Such a 4-point analysis is algebraically simple, and it is not so strongly affected by rounding-off errors as the 6-point analysis, which is also considerably more time-consuming. The introduction of this 4-point analysis in the method of successive approximations seems therefore to be a reasonable compromise between the method of Part I and the exact theory.

TABLE II. Influence of rounding-off errors on the second-order Hankel determinant $H(1)$. As SCF function $f^*(r) = r^2 g^*(r)$ we use the analytic fit (15) for $\text{Na}^+(2p)$. 2.4940 (–2) means 2.4940×10^{-2} .

r	f_{2p}^*	Six decimals in f_{2p}^*		Three decimals in f_{2p}^*	
		$H_n(1)$	Quo- tient	$H_n(1)$	Quo- tient
0.2	0.579521	3.9463 (0)		3.9438 (0)	
0.4	0.995692	6.4080 (–1)	6.1584	6.4830 (–1)	6.0833
0.6	1.060341	1.1774 (–1)	5.4426	1.1786 (–1)	5.5003
0.8	0.958100	2.4940 (–2)	4.7209	2.4716 (–2)	4.7687
1.0	0.800858	6.0275 (–3)	4.1377	6.1881 (–3)	3.9942
1.2	0.640930	1.6075 (–3)	3.7496	1.5199 (–3)	4.0713
1.4	0.499589	4.5632 (–4)	3.5227	4.5024 (–4)	3.3758
1.6	0.382930	1.3424 (–4)	3.3993	1.4235 (–4)	3.1628
1.8	0.290213	4.0252 (–5)	3.3351	4.4112 (–5)	3.2271
2.0	0.218137	1.2193 (–5)	3.3012	1.0691 (–5)	4.1259
2.2	0.162874	3.7126 (–6)	3.2842	2.0982 (–6)	5.0956
2.4	0.120897	1.1337 (–6)	3.2747	3.5091 (–6)	0.5979
2.6	0.089243				
2.8	0.065522				

The method will now be described in greater detail. Let us start by dividing the tabulated SCF function by a polynomial $r^{l+1}(r_0' - r)(r_0'' - r) \cdots$, where r_0' , r_0'' , \cdots are the $(n-l-1)$ zero points of the function itself determined by the procedure described in Part II, p. 1606; the quotient

$$g_{nl}(r) = f_{nl}(r)/r^{l+1}(r_0' - r)(r_0'' - r) \cdots \quad (18)$$

is nodeless and monotonically decreasing with increasing r . In order to expand $g_{nl}(r)$ into exponentials, we observe that in the tail region ($r \approx \infty$) only two exponentials are important, in the next inner region three terms, in the following region four terms etc.; in general $(n+1)$ exponentials seem to be necessary for an almost perfect fit with a tolerance of the order of magnitude 0.001–0.002. The main problem in the tail region is to find four equidistant points which are suitable for a 4-point analysis, i.e., where the distribution of rounding-off errors is such that the two exponentials obtained will give a good "average" fit to the entire outer region. Several methods for selecting these four points are possible in practice, and here we will only describe a method based on the Hankel determinants $H_n(1)$.

Let us form the "centralized" Hankel determinants

$$D(r) = g(r-h)g(r+h) - \{g(r)\}^2. \quad (19)$$

According to (10), these quantities should form a single geometric series in the outer region where only two exponentials are important. As pointed out in (a), the effect of rounding-off errors is appreciable, and the "quotient series" is often quite irregular. However, if we plot $\log_{10} D(r)$ as a function of r , the points are distributed irregularly around a smooth curve which theoretically tends to a definite straight line in the region where only two exponentials are important. In practice this straight line is often rather undetermined (see Fig. 1) because of the irregularities of the distribution, and there is a certain relation between the uncertainty of this line and the average rounding-off errors. Let us now select two values of D at the distance h such that the straight line between them falls within the irregular distribution of points of the tail region. A good "average" choice will lead to a good fit in the tail region. The two values of D selected are associated with four points g_0, g_1, g_2, g_3 , which should be expressed as the sum of two geometric series: $g_n = \alpha_1 k_1^n + \alpha_2 k_2^n$. The quotients and coefficients are determined by the general formulas (11) and (12), which will take the simplified form

$$\begin{vmatrix} 1 & k & k^2 \\ g_0 & g_1 & g_2 \\ g_1 & g_2 & g_3 \end{vmatrix} = 0; \quad (20)$$

$$\alpha_1 = (g_1 - k_2 g_0)/(k_1 - k_2).$$

The 4-point analysis determines two exponentials $A_1 \exp(-a_1 r)$ and $A_2 \exp(-a_2 r)$, but we will fix only the one with the smaller exponent a_1 , which corre-

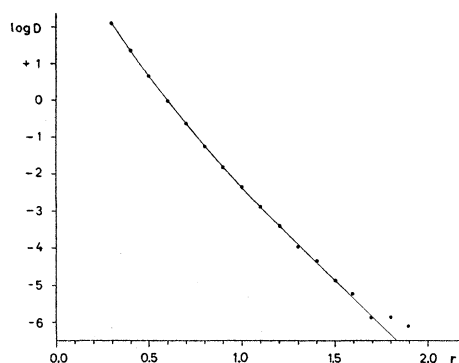


FIG. 1. The behavior of the second-order Hankel determinant ($m=1$) for the $2p$ function of argon with $D(r)$ defined by (19). The small circles (●) correspond to the numerical material given by Hartree and Hartree, reference 8, and the irregularities in the outer region ($r>1.2$) depend on the influence of the rounding-off errors. The solid curve corresponds to the final analytic fit given by (26).

sponds to the outmost part of the tail. This exponential is then subtracted from $g_{nl}(r)$ and the analysis is then repeated for the function

$$\bar{g}_{nl} = g_{nl}(r) - A_1 e^{-a_1 r}. \quad (21)$$

A special fit may sometimes be needed for the last exponential describing the region closest to $r=0$. Let us plot the error left after the last 4-point analysis graphically as a function of r ; if the points are essentially situated on one side of the r axis, the maximum error is often outside the tolerance prescribed (see Fig. 2). However, the error is a continuous function of the coefficient and the exponent of the last exponential and, by a suitable selection and modification of two critical function values determining this exponential, we may successively move the error curve so that the points will finally be equally distributed on both sides of the r axis. The absolute value of the error is now brought to a certain minimum, and, if the accuracy is not good enough, it may be necessary to repeat the analysis starting over from the tail. After some experience, this seems hardly necessary, however.

(c) Practical Example: A($2p$)

As an example of the method, we will give some data from an analysis of the f_{2p} function for argon tabulated by Hartree and Hartree.⁸ We have determined our first interval h from the following "rule of thumb": evaluate the point r_1 where $f_{nl}(r_1) \approx 0.020$ and choose $h \approx \frac{1}{8}r_1$; for A($2p$) this leads to $h=0.3$. Additional entries may be added to the table by interpolation in $\log g(r)$; for the tail region linear interpolation is sufficient, but the inner parts must be more carefully treated. In Fig. 1 the values of $\log D$ are plotted (circles), and as basic points for our straight line we have then chosen $r=1.5$ and $r=1.2$. We note

⁸ D. R. Hartree and W. Hartree, Proc. Roy. Soc. (London) **A166**, 450 (1938).

that the solid line in Fig. 1 corresponds to the final analytic fit. The 4-point analysis of the entries for $r=1.8(0.3)0.9$ gives the following result:

$$\begin{aligned} k_1 &= 4.0933, & k_2 &= 7.2377, \\ a_1 &= 4.6978, & a_2 &\approx 6.5977, \end{aligned} \quad (22)$$

and the outermost exponential is then $10.043 \times \exp(-4.6978r)$. We then repeat the procedure with the function $\bar{g}_{nl}(r)$ obtained according to (21) by subtracting this exponential; the values of $\log \bar{D}$ fall very closely on a straight line in the remaining region, and it seems hence possible to express $\bar{g}_{nl}(r)$ by means of only two exponentials. For a 4-point analysis we choose $r=0.8(0.2)0.2$ and obtain

$$\begin{aligned} k_2 &= 3.7219, & k_3 &= 9.5187, \\ a_2 &= 6.5712, & a_3 &= 11.2663, \end{aligned} \quad (23)$$

where we note the small change in the value of a_2 from the preliminary value in (22). The corresponding exponentials have the coefficients

$$A_2 = 89.062, \quad A_3 = 77.541. \quad (24)$$

In order to check the analytic fit obtained, we have plotted the error

$$\Delta f_{2p}(r) = r^2 \left\{ g_{2p}(r) - \sum_{\nu=1}^3 A_\nu \exp(-a_\nu r) \right\}, \quad (25)$$

in Fig. 2 (crosses). We see immediately that the fit is good enough in the outer region ($r>0.8$); otherwise it had been necessary to make a new 4-point analysis of $\bar{g}_{nl}(r)$ in the neighborhood of the "critical points." The first and second exponentials are from now on considered as fixed; it remains to vary a_3 and A_3 in order to get the best result. We note that the "critical points" of the error curve appear for $r=0.05, 0.20, 0.55$ and that the maximum error is about -0.0043 . In a first trial, we fix two new entries in the points $r=0.05$ and $r=0.55$ with the definite error $\Delta f = -0.0020$ equal to the maximum tolerance permitted. The two points determine a new last exponential, where $\Delta f = +0.0012$

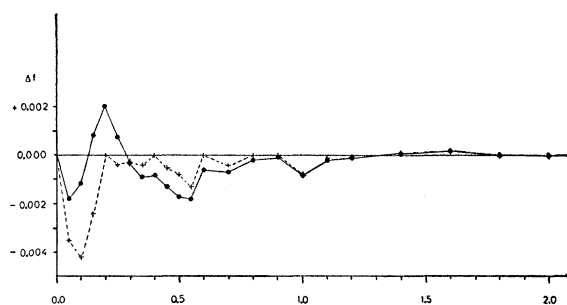


FIG. 2. The error function Δf defined by (25) for the analytic fit of the $2p$ function of argon. The crosses (+) indicate the preliminary result which is then changed to the final form (circles ●) by modifying the last exponential by a suitable choice of two "critical points."

for $r=0.20$. In the next trial, we choose instead $\Delta f(0.05) = \Delta f(0.55) = -0.0018$ and obtain $\Delta f(0.20) = +0.0020$, and the error is then rather evenly distributed on both sides of the r axis. We fix our third exponential and obtain the final result

$$\begin{aligned} a_\nu: & 4.6978, \quad 6.5712, \quad 11.3275, \\ A_\nu: & 10.043, \quad 89.062, \quad 78.967. \end{aligned} \quad (26)$$

The error curve is plotted in Fig. 2 (circles); we note that $|\Delta f| \leq 0.0020$. Finally the normalization is checked and gives the value 0.99993. The contributions from the three exponentials to $f_{2p}(r)$ is illustrated in Fig. 3.

We have tried to give a description of our method which is as complete as possible, since we think that the method is very convenient for a solution of the fitting problem by means of ordinary desk machines only. On our part, we have never felt that the use of a high-speed electronic computer⁹ would be of any essential help to us in solving this particular problem.

III. APPLICATIONS TO THE ARGON-LIKE IONS AND TO COPPER

The SCF functions with exchange for the A-like ions and for Cu^+ have been tabulated in the literature by Hartree and Hartree.¹⁰ In order to test our interpolation procedures, we considered first the series Cl^- , A, K^+ , and Ca^{+2} , where we determined analytic fits for Cl^- and Ca^{+2} ; the results are listed in Table III. Analytic functions for A and K^+ were then interpolated between Cl^- and Ca^{+2} by the analytic method described in Part I, i.e., by assuming that the exponents $a_\nu = a_\nu(Z)$ are linear functions of the atomic number Z and that the coefficients A_ν of $g_{nl}(r)$ are some powers of the exponents

$$A_\nu(Z) = \kappa_\nu \{a_\nu(Z)\}^p. \quad (27)$$

One can expect that an interpolation will give good results if the exponents p_ν in (27) are of the order of

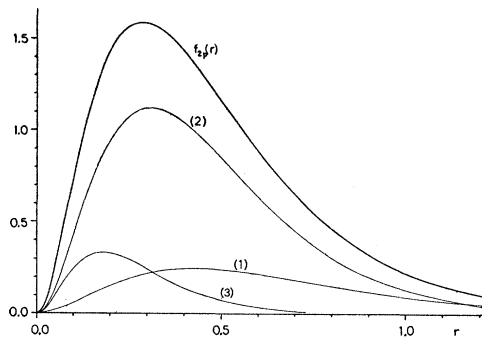


FIG. 3. Contributions to the SCF function $f_{2p}(r)$ for argon from the three exponential terms obtained in (26).

⁹ F. J. Corbató, *Quarterly Progress Reports of Solid-State and Molecular Theory Group of Massachusetts Institute of Technology*, July 15, 1954, p. 49, and October 15, 1954, p. 28. (unpublished),

¹⁰ D. R. Hartree and W. Hartree, *Proc. Roy. Soc. (London)* **A156**, 45 (1936), Cl^- ; **A166**, 450 (1938), A and K^+ ; **A164**, 167 (1937), Ca^{+2} ; **A157**, 490 (1936), Cu^+ .

TABLE III. Analytic wave functions for the argon-like ions Cl^- , A, K^+ , Ca^{+2} , and for Cu^+ , in the form $f_{nl}(r) = r^{l+1}(r_0' - r) \cdots \times \sum_\nu A_\nu \exp(-a_\nu r)$ with maximum error $\Delta f = f_{\text{SCF}} - f_{\text{analytic}}$.

Atom		Cl^-	A	K^+	Ca^{+2}	Cu^+
1s:	A_1	49.235	108.58	75.554	90.328	109.68
	a_1	14.329	16.410	16.564	17.681	25.260
	A_2	87.030	40.542	85.924	84.220	197.01
	a_2	18.381	22.558	21.048	22.381	30.957
	$ \Delta f $	0.0016	0.0023	0.0013	0.0020	0.0014
2s:	r_0	0.12480	0.11765	0.11107	0.10530	0.071612
	A_1	51.335	55.121	56.963	59.233	102.01
	a_1	5.3545	5.6995	6.0090	6.3362	9.3363
	A_2	156.77	197.44	236.27	283.13	913.00
	a_2	7.6112	8.0628	8.4701	8.8995	13.1866
2p:	A_1	97.310	105.42	124.69	138.39	305.05
	a_1	12.6846	13.8773	14.4383	15.3151	22.5215
	$ \Delta f $	0.0015	0.0018	0.0021	0.0020	0.0023
2p:	A_1	22.594	10.043	23.274	23.269	48.720
	a_1	4.7395	4.6978	5.3904	5.7158	8.6425
	A_2	77.535	89.062	113.53	133.74	498.79
	a_2	6.8437	6.5712	7.5418	7.8908	12.5471
	A_3	52.344	78.967	75.717	88.780	164.66
3s:	A_1	11.9160	11.3275	13.0179	13.5689	22.5235
	a_1	0.0016	0.0020	0.0038	0.0008	0.0017
	r_0'	0.12160	0.11437	0.10800	0.10233	0.069238
	r_0''	0.55499	0.51572	0.48189	0.45162	0.29206
	A_1	1.6874	3.0270	6.4608	11.020	24.339
3p:	A_1	1.7189	2.0280	2.4058	2.7492	3.9560
	a_1	27.507	33.231	56.770	77.566	336.08
	A_2	3.0630	3.3032	3.8013	4.1704	6.1429
	a_2	80.475	92.692	142.29	183.93	953.80
	A_3	5.4822	5.4140	6.3818	6.8316	9.8053
3d:	A_4	57.540	96.510	89.050	108.51	428.64
	a_4	11.1332	10.4864	12.8927	13.7724	18.1689
	$ \Delta f $	0.0030	0.0020	0.0032	0.0020	0.0021
3d:	A_1	0.53153	0.48869	0.45199	0.42042	0.26263
	a_1	0.13472	0.81754	2.2951	4.6377	11.625
	A_2	0.9958	1.5080	1.9297	2.3026	3.3778
	a_2	3.2595	10.093	19.932	33.881	155.51
	A_3	1.9059	2.5775	3.1068	3.5717	5.4371
Ca ⁺ :	A_4	22.402	40.354	62.476	90.605	532.99
	a_4	3.4929	4.4182	5.2007	5.9832	8.8262
	A_5	41.722	51.754	61.135	67.642	297.43
	a_5	7.3684	9.1491	10.8938	12.6386	16.9724
	$ \Delta f $	0.0020	0.0014	0.0013	0.0014	0.0013
Ca ⁺ :	A_1				0.12839	0.63429
	a_1				0.8896	1.6879
	A_2				0.57095	11.439
	a_2				1.4568	3.1761
	A_3				4.9228	92.512
Cu ⁺ :	A_4				2.8257	6.0330
	a_4				12.498	116.04
	A_5				5.7048	10.7712
	a_5				0.0027	0.0021
	$ \Delta f $					

magnitude $n+\frac{1}{2}$, $n-\frac{1}{2}$, $n-\frac{3}{2}$, \cdots etc., and if the normalization integral

$$N_{nl} = \int_0^\infty f_{nl}^2(r) dr \quad (28)$$

is only slightly different from unity, i.e., $|N_{nl}-1| \leq 0.05$; the final result should, of course, be renormalized. We would like to emphasize the existence of these conditions for, depending on the ambiguities in the choice of a_ν and A_ν , there may be ill-behaved cases in which they are not satisfied. In order to carry out a good interpolation it may then be necessary to adjust the values of the basic quantities a_ν and A_ν by a new analytic approximation of the given functions.

The results of our interpolation for A and K^+ are listed in Table IV; we note that there is a certain correlation between the normalization integrals N_{nl} and the errors $\Delta f = f_{\text{SCF}} - f_{\text{anal}}$. We have also listed the errors Δf for the functions which are numerically interpolated by means of the characteristic ω functions

TABLE IV. Normalization integrals N_{nl} and maximum errors $|\Delta f|$ for A and K^+ in the analytic interpolation between Cl^- and Ca^{+2} ; for comparison the quantities $|\Delta f|$ in the corresponding numerical interpolation based on the ω functions [Part II, Eq. (33)] are also listed.

nl	A			K^+		
	Analytic interpolation N_{nl}	$ \Delta f $	ω inter- polation $ \Delta f $	Analytic interpolation N_{nl}	$ \Delta f $	ω inter- polation $ \Delta f $
1s	0.9792	0.007	0.008	0.9806	0.001	0.001
2s	0.9863	0.003	0.003	0.9881	0.002	0.001
2p	0.9795	0.005	0.002	0.9813	0.004	0.001
3s	1.0038	0.002	0.006	1.0017	0.003	0.003
3p	0.9801	0.016	0.006	0.9844	0.013	0.004

described in Part II; it turns out that the numerical procedure is considerably more accurate than the analytic method.

For K^+ the results for 1s, 2s, 2p, and 3s were sufficiently accurate to let us include the interpolated analytic functions in Table III. The 3p function had to be treated independently, and the results are listed in Table III.

For A, there was a complication for the 1s function. It was found that our analytic function with the parameters

$$A(1s): \quad a_1 = 15.446, \quad a_2 = 19.714, \quad (29) \\ A_1 = 61.796, \quad A_2 = 86.930,$$

was in almost perfect agreement with the numerical function interpolated by the ω technique, whereas both of them deviated considerably ($|\Delta f| \approx 0.007$) from the numerical function tabulated by Hartree and Hartree.¹⁰ Their results were obtained by a transformation based on McDougall's result for argon without exchange. Since the latter have not been available to us, we have not been able to check this transformation, which would certainly be desirable because of the above-mentioned large deviation. In Table III we have therefore also listed an accurate analytic fit to the 1s function given by Hartree and Hartree; this function was found independently.

In addition to the interpolated functions for A, we have also carried out independent calculations for the other SCF functions by starting from the Hartree-Hartree tables for argon. The results are slightly better than the interpolated ones and are listed in Table III.

All the functions for Cu^+ and the 3d function for Ca^+ were treated separately, and the results are also condensed in Table III.

IV. INTERPOLATED ANALYTIC WAVE FUNCTIONS FOR THE FIRST ROW OF TRANSITION ELEMENTS

The elements with the atomic numbers between $Z=21$ and $Z=28$ (Sc, Ti, V, Cr, Mn, Fe, Co, Ni) are of considerable interest in physics and in chemistry, and it is therefore desirable to have accurate analytic SCF functions with exchange for them. A few functions *without exchange* for elements of this series have previ-

ously been tabulated,¹¹ but the use of the results are often complicated by the fact that the tables are not given in the conventional form developed by Hartree and Hartree.

By using the data available for Ca^+ and Cu^+ with exchange, we have interpolated analytic SCF functions for the first row transition metals. The nodes have been interpolated by the technique developed in Part II¹² and the exponentials by the method described in the previous section. The results have been condensed in Table V.

It should be observed that the interpolation is comparatively well behaved. The normalization integral $N_{nl}(Z)$ is a slowly varying function of Z with a minimum for $Z=24$ with the following values:

$$\begin{array}{ll} 1s: & 0.9684, & 3s: & 0.9670, \\ 2s: & 0.9802, & 3p: & 0.9671, \\ 2p: & 0.9834, & 3d: & 0.9498. \end{array} \quad (30)$$

In order to get at least some idea of the accuracy of the interpolated material, we have considered the SCF functions for Fe ($Z=26$) in somewhat greater detail. From previous experience we know that the numerical interpolations based on the ω functions [Part II, Eq. (33)] are considerably more accurate than the analytic interpolations. In our case, there were two ways of carrying out this numerical procedure: either by linear interpolation between the SCF functions for Ca^+ and Cu^+ with exchange in the function $\omega(r)$ itself, or by linear interpolation in the quantity $\Delta\omega(r)$, which is the difference between the two ω functions for fields with and without exchange, respectively, by using also the SCF material available for Ca, Fe, and Cu^+ *without exchange*.¹³ The two different sets of ω functions for Fe with exchange obtained in this way were practically the same, and we have therefore used only the latter type which may be slightly more accurate. We have below listed the maximum difference between the results of the analytical and numerical interpolations for Fe:

$$|f_{\text{anal}} - f_{\omega}| \\ \begin{array}{cccccc} 1s & 2s & 2p & 3s & 3p & 3d \\ \leq 0.002, & 0.006, & 0.004, & 0.023, & 0.035, & 0.078. \end{array} \quad (31)$$

Since the values of f_{ω} are expected to be fairly accurate, we believe that the figures in (31) will give also the order of magnitude of the actual error in the analytic function.¹⁴

¹¹ See the survey in D. R. Hartree, Repts. Progr. Phys. **11**, 113 (1946).

¹² In Part II, Table XVI, there is an error: the values of r_0'' and $Z(SCF|r_0'')$ for Cu^+ should be 0.29206 and 24.303 instead of 0.29238 and 24.277, respectively.

¹³ D. R. Hartree and W. Hartree, Proc. Roy. Soc. (London) **A149**, 210 (1935), Ca; M. F. Manning and L. Goldberg, Phys. Rev. **53**, 662 (1938), Fe; D. R. Hartree, Proc. Roy. Soc. (London) **A141**, 282 (1933), Cu^+ .

¹⁴ A comparison between the functions for Fe with exchange obtained by analytic and numerical interpolation is given in a table which also contains Manning and Goldberg's functions

TABLE V. Interpolated analytic SCF functions with exchange for the first row of transition elements.

Atom	Sc	Ti	V	Cr	Mn	Fe	Co	Ni
1s: A_1	93.299	96.058	98.594	100.92	103.04	104.96	106.70	108.27
a_1	18.523	19.365	20.207	21.049	21.892	22.734	23.576	24.418
A_2	94.607	105.59	117.14	129.22	141.81	154.91	168.49	182.52
a_2	23.334	24.287	25.240	26.193	27.145	28.098	29.051	30.004
2s: r_0	0.10013	0.09543	0.09114	0.08721	0.08359	0.08025	0.07716	0.07429
A_1	63.948	68.689	73.442	78.204	82.969	87.732	92.491	97.244
a_1	6.6695	7.0029	7.3362	7.6696	8.0029	8.3363	8.6696	9.0030
A_2	332.24	386.27	445.34	509.59	579.14	654.12	734.64	820.83
a_2	9.3758	9.8522	10.3285	10.8049	11.2812	11.7576	12.2339	12.7103
A_3	154.35	171.01	188.34	206.30	224.88	244.06	263.81	284.12
a_3	16.1158	16.9165	17.7172	18.5179	19.3187	20.1194	20.9201	21.7208
2p: A_1	25.792	28.402	31.089	33.851	36.685	39.589	42.560	45.597
a_1	6.0410	6.3662	6.6914	7.0166	7.3417	7.6669	7.9921	8.3173
A_2	160.80	191.01	224.44	261.20	301.40	345.14	392.51	443.62
a_2	8.4082	8.9255	9.4429	9.9603	10.4776	10.9950	11.5124	12.0297
A_3	97.169	105.60	114.05	122.49	130.93	139.37	147.79	156.20
a_3	14.5638	15.5588	16.5537	17.5486	18.5436	19.5385	20.5334	21.5284
3s: r_0'	0.09725	0.09264	0.08842	0.08456	0.08100	0.07772	0.07469	0.07186
r_0''	0.42714	0.40488	0.38456	0.36593	0.34879	0.33297	0.31832	0.30472
A_1	12.316	13.669	15.072	16.520	18.010	19.541	21.107	22.706
a_1	2.8833	3.0174	3.1515	3.2856	3.4196	3.5537	3.6878	3.8219
A_2	94.876	114.68	137.15	162.43	190.66	222.00	256.59	294.57
a_2	4.3896	4.6087	4.8279	5.0471	5.2662	5.4854	5.7046	5.9237
A_3	229.80	283.68	346.33	418.57	501.27	595.24	701.38	820.56
a_3	7.1620	7.4924	7.8228	8.1532	8.4837	8.8141	9.1445	9.4749
A_4	129.95	154.38	182.00	213.05	247.77	286.44	329.28	376.57
a_4	14.2609	14.7494	15.2379	15.7264	16.2149	16.7034	17.1919	17.6804
3p: r_0	0.39621	0.37420	0.35410	0.33568	0.31873	0.30309	0.28860	0.27515
A_1	5.2774	5.9528	6.6653	7.4127	8.1928	9.0053	9.8476	10.717
a_1	2.4221	2.5415	2.6610	2.7805	2.8999	3.0194	3.1389	3.2583
A_2	41.902	51.108	61.601	73.452	86.719	101.49	117.82	135.76
a_2	3.7790	3.9862	4.1935	4.4008	4.6080	4.8153	5.0226	5.2298
A_3	115.46	145.03	179.91	220.64	267.78	321.96	383.73	453.70
a_3	6.2991	6.6150	6.9309	7.2468	7.5626	7.8785	8.1944	8.5103
A_4	82.262	99.089	118.35	140.25	165.02	192.86	224.01	258.71
a_4	13.1201	13.6017	14.0832	14.5647	15.0463	15.5278	16.0093	16.4909
3d: A_1	0.16510	0.20661	0.25301	0.30428	0.36043	0.42147	0.48741	0.55828
a_1	0.9783	1.0670	1.1557	1.2444	1.3331	1.4218	1.5105	1.5992
A_2	0.93030	1.4299	2.0981	2.9653	4.0634	5.4243	7.0825	9.0748
a_2	1.6478	1.8389	2.0299	2.2209	2.4120	2.6030	2.7940	2.9851
A_3	7.9065	12.013	17.471	24.515	33.387	44.350	57.668	73.614
a_3	3.1821	3.5384	3.8948	4.2512	4.6075	4.9639	5.3203	5.6766
A_4	17.635	24.031	31.829	41.171	52.198	65.050	79.870	96.809
a_4	6.2677	6.8307	7.3936	7.9565	8.5195	9.0824	9.6453	10.2083

It is immediately clear that we can expect that the inner functions (1s, 2s, 2p) are interpolated with a good accuracy, whereas there must be considerable uncertainties in the outer functions (3s, 3p, 3d). A comparison between our analytic functions and the functions without exchange for Fe shows further that the inner functions reveal the characteristic contraction towards origin obtained in including exchange effects, whereas this contraction is far less pronounced for the outer analytic functions, the tails of which are situated about

(without exchange) transformed to the standard Hartree-Hartree form. The table, supplementary to this article, has been deposited as Document number 4961 with the ADI Auxiliary Publications Project, Photoduplication Service, Library of Congress, Washington 25, D. C. A copy may be secured by citing the Document number and by remitting \$1.25 for photoprints, or \$1.25 for 35-mm microfilm. Advance payment is required. Make checks or money orders payable to: Chief, Photoduplication Service, Library of Congress.

halfway between the values without exchange and those obtained by numerical interpolation. The accuracy of the outer functions is hence definitely low; if higher accuracy is required, one must instead first carry out a numerical interpolation by means of the ω technique and then transform the result to analytic form. However, we observe that we have not specified the quantum state of the atom under consideration, and that there are actually different SCF functions for different states. The interpolated functions belong to some "average" state of still unknown nature and, in order to achieve full accuracy for the outer functions, it is thus necessary to take the problem of the different states into full account. The functions listed in Table V may therefore be of value only in a first rough investigation of the "average" behavior of the elements concerned.

The 3d functions are of particular interest, and we

TABLE VI. Interpolated Taylor coefficients $c_{l+1}=f_{nl}(r)/r^{l+1}$ for $r=0$ for the first row of transition elements.

nl	Sc	Ti	V	Cr	c_{l+1} Mn	Fe	Co	Ni
1s	188.20	201.97	216.07	230.48	245.19	260.21	275.52	291.12
2s	55.137	59.664	64.310	69.070	73.941	78.922	84.011	89.204
2p	285.78	326.47	370.45	417.80	468.61	522.96	580.92	642.58
3s	19.222	21.046	22.923	24.853	26.835	28.867	30.947	33.075
3p	95.41	110.84	127.69	145.98	165.75	187.06	209.93	234.40
3d	33.41	46.01	61.68	80.85	103.92	131.36	163.61	201.16

have also tried to express them as scaled copper functions, i.e., in the form

$$f_{3d}(Z; r) = \lambda^{\frac{1}{2}} f_{3d}(\text{Cu}^+; \lambda r). \quad (32)$$

A value of the scale factor was obtained for Ca^+ from the condition that the scaled function (32) should have the same r_{max} as the numerically given SCF function with exchange; this gave $\lambda = 0.39529$ for $\text{Ca}^+(3d)$ and the deviation of the scaled function (32) from the actual SCF function did not exceed 0.048. The values of λ for different elements could then be obtained by linear interpolation leading to the formula

$$\lambda = 0.06719Z - 0.94851. \quad (33)$$

For Fe, the scaled function (32) was in rather close agreement with the function obtained by analytic interpolation; the difference did not exceed 0.015 but was usually much smaller and the scaled function was situated between the results of the analytic and numerical interpolations. For elements close to Cu^+ , the general accuracy of (32) will be still better, but, for elements close to Ca^+ , it is recommended to use a scaled Ca^+ function instead of (32).

The region around the origin ($r=0$) is nowadays of particular importance because of the interest in the interaction between the nucleus and the electronic cloud. It should be observed that, in the analytic SCF functions in Table V, we have not tried to get any higher degree of accuracy in this region, since this would require one more exponential in the functions. However, the behavior of the functions at the origin has been investigated in greater detail in Part II, and the value of the first nonvanishing Taylor coefficient,

$$c_{l+1} = [f_{nl}(r)/r^{l+1}]_{r=0}, \quad (34)$$

may be found by considering the corresponding "effective charge" K_{nl} [Part II, Eq. (19)] which is an almost perfectly linear function of the atomic number Z . By using the data available for Ca^+ and Cu^+ , we have obtained values of c_{l+1} for the first row of transition elements which are condensed in Table VI. Our data have also been checked by using the information available for SCF functions *without* exchange by carrying out a similar interpolation in the quantities ΔK_{nl} , which are the differences in the values of K_{nl} for the fields with and without exchange, respectively; the agreement was good.

In addition, we observe that, close to the origin, the

function f_{nl}/r^{l+1} behaves like an exponential [Part II, Eq. (22)] so that

$$f_{nl}(r) = c_{l+1} r^{l+1} \exp[-rZ/(l+1)]. \quad (35)$$

This means that we know the values of $f_{nl}(r)$ fairly well in the innermost region.

V. DISCUSSION

The problem of finding an analytic fit to the numerically given SCF functions is complicated by the fact that, at least from the practical point of view, there does not seem to exist a uniquely defined "best approximation" but instead a whole family of analytic functions of about the same accuracy. In the numerical work based on the exact method for expanding a function into exponentials, this corresponds to a considerable cancellation of significant figures, which greatly increases the influence of even the rounding-off errors. We have avoided this complication by a method of successive approximations where, by means of the 4-point analysis of the exact theory, a pair of exponentials are determined in each step.

We have first evaluated analytic fits for the SCF functions for Cl^- , A, K^+ , and Ca^{+2} with exchange. By using the data available for Ca^+ and Cu^+ , we have further carried out an analytic interpolation to obtain the SCF functions for the first row of transition metals. These functions belong to some "average quantum state" of the elements under consideration, and an investigation of the results for Fe shows that, whereas the accuracy of the inner functions for $n=1$ and $n=2$ is definitely good, the uncertainty of the outer functions for $n=3$ is comparatively large.

It should here be emphasized that much more accurate *numerical* SCF functions with exchange for the first row of transition metals can be obtained by interpolation by means of the ω functions [Part II, Eq. (33)]; the basic ω functions for Ca^+ and Cu^+ are also available (Part II, reference 16). Anyone interested in getting good starting functions for SCF calculations for the elements concerned should therefore definitely use this technique. Better *analytical* SCF functions could then also be obtained by evaluating analytic fits to the numerically interpolated functions, but such a procedure will of course require much more computation than the simple approach used here. These questions will be treated further in forthcoming papers.

In applications to molecular and crystal theory, analytic SCF functions containing several exponentials will lead to rather cumbersome calculations. We have therefore felt that it would be desirable to have also less accurate analytic approximations containing only one or two exponentials, and numerical computations on this problem are therefore now in progress.

In conclusion, it should be stated that the success of a numerical analysis of the type carried out here is highly dependent on the given SCF material. In this connection, we would like to express our sincere appreci-

ation for the numerical SCF tables given by Professor D. R. Hartree and his late father, Dr. W. Hartree; we have found that their numerical data are characterized by an excellent quality: their functions are perfectly smooth and the variation from element to element indicates an outstanding reliability. We sincerely hope that their pioneer work of calculating SCF functions without and with exchange will be continued also in the future by scientists interested in the atomic field.

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Angular Distribution of Photoneutrons from Carbon and Beryllium*

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The photoneutron angular distributions from carbon and beryllium have been measured by using bremsstrahlung of maximum energies of 23 Mev and 18 Mev, respectively. The results are $1 + (1.35 \pm 0.88) \sin^2\theta$ for carbon and $(1.26 \pm 0.11) + \sin^2\theta$ for beryllium.

INTRODUCTION

PREVIOUS measurements of the photoproton angular distribution from C^{12} using bremsstrahlung of 23-Mev maximum energy have been made by Halpern, Mann, and Rothman.¹ The distribution is of the form $1 + a(\sin\theta + 0.25 \sin\theta \cos\theta)^2$, with a forward peak at about 80° relative to the incident γ -ray beam resulting from dipole and quadrupole interference terms. These results have been interpreted² as evidence for an independent-particle description of the giant resonance and LS coupling in the carbon nucleus.

On the assumption of charge independence of nuclear forces, Gell-Mann and Telegdi³ have shown that the (γ, p) and (γ, n) cross sections in nuclei with $J=0$ and $T_z=0$, for each residual state, must be identical with each other as functions of energy and angle. This prediction is valid if there is no interference between electric dipole absorption and other multipole absorption arising from that part of the interaction Hamiltonian that is a scalar in isotopic spin space.

In this paper a measurement of the angular distribution of photoneutrons from the giant-resonance region of the reaction $C^{12}(\gamma, n)C^{11}$ using 23-Mev bremsstrahlung from the University of Pennsylvania betatron is reported. It was hoped in this way to study the direct emission of photoneutrons and also to check the charge-

independence hypothesis. A preliminary measurement⁴ indicated a dip in the angular distribution at 90° .

Also reported is a measurement of the photoneutron angular distribution from Be^9 using bremsstrahlung of maximum energy 18 Mev which is below the giant resonance. Guth and Mullin⁵ and Czyz⁶ have calculated the angular distribution for the reaction $Be^9(\gamma, n)Be^8$, assuming that the Be^9 nucleus consists of a Be^8 core plus a valence neutron in a $P_{3/2}$ ground state. They have obtained a distribution of the form $a + b \sin^2\theta$, arising from electric dipole transitions to S and D states, with the ratio a/b depending on the respective transition probabilities.

EXPERIMENTAL PROCEDURE

The details relating to the formation of a collimated x-ray beam have been presented previously.⁷ In this experiment, an additional concrete wall 16 inches thick has been placed between the betatron and the target, about 2 feet from the target and 18 feet from the betatron. This new wall has cut the neutron background by a factor of 5.

The carbon and beryllium targets were cut in the form of 1-inch cubes. Neutrons were detected by a ZnS—paraffin plastic button⁸ mounted on an RCA photomultiplier tube. The apparatus was arranged so that the photomultiplier tube could rotate about a center post on which was set the target. There was a

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