conversion electrons observable in our spectrometers. There is evidence from an observation of the absorption in aluminum that some very low energy gamma-radiation is present.

The ground state of the molybdenum nucleus has been found to be $d_{5/2}$. For the first excited level of the odd-even nucleus with 53 neutrons, from shell theory, a $g_{7/2}$ level is expected. This is in good agreement with present observations if the 768-kev transition is magnetic dipole, and it also satisfies the allowed 165-kev beta-transition from a $g_{9/2}$ level. The 235-kev radiation in niobium is by virtue of its long radiation lifetime and its high conversion coefficient interpreted as an $M4$ transition. If, then, the upper level is a $p_{1/2}$ state and the initial zirconium level, like the molybdenum ground state, is characterized as $d_{5/2}$, it would follow that the 910-kev beta-transition would be forbidden, as observed. No transition from the initial $d_{5/2}$ level to the niobium $g_{9/2}$ level would be expected.

Note added in proof:—A recent report by V. Shpinel (USSR) Zhur. Eksptl. i Teort. Fiz. **21**, 1370 (1951), treats these radioactivities. Two gamma-rays are evaluated from photoelectrons and others are postulated to exist from differences in observed beta-energies. These are not in agreement with our observed energies.

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Radiochemical Studies on the Photofission of Thorium*

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The relative yields of thirteen products resulting from the photofission of Th^{232} have been determined radiochemically. Photofission was induced by a 69-Mev synchrotron bremsstrahlung beam. The photofission yield curve was shown by these data to have two peaks, occurring approximately at nucleon numbers 91 and 138 with values of 6.9 percent. The peak-to-valley ratio was 10 and the half-width of the peaks was 12. Symmetry of the curve about nucleon number of 114.5 indicated the average emission of approximately three neutrons. The results were compared with other photofission data and the yield curves of other fission processes.

I. INTRODUCTION

In the photoexcitation of heavy nuclei at moderate energies, fission may occur in excited states of target nuclei. At higher energies, the fission may be preceded by the loss of one or more neutrons. In any event, the nuclei undergoing fission are generally different from those which are excited by neutron capture or by charged-particle bombardment. A study of photofission, therefore, extends the knowledge concerning the general fission process. This is of particular interest in the case of Th^{232} and U^{238} because neutron yields from spontaneous fission have been determined for the ground states of these two nuclides.[1–3]

Present information of photofission product yields from various fissionable materials is not nearly so comprehensive as that available for the yields from neutron fission reported by the Plutonium Project[4] and by subsequent workers.[5,6] Photofission yield curves have been estimated for Bi^{209} irradiated by 85-Mev x-rays[7] and for U^{235} irradiated by 22-Mev x-rays.[8] Also available are some preliminary results of a current study by Richter[9] and Coryell[10] involving the irradiation of natural uranium with 16-Mev x-rays.

In this paper are reported the radiochemically determined yields of thirteen nucleon (mass) numbers produced by the irradiation of thorium with 69-Mev bremsstrahlung. From the photofission cross section curve of thorium given by McElhinney[11] and Ogle[12] and the characteristics of the bremsstrahlung spectrum, it is apparent that the preponderant majority of fission events resulted from the relatively moderate excitation energies of 10–20 Mev. Therefore it is not surprising that a yield curve of the familiar "twin-peaked" form was produced. The data were sufficient to give a fairly reliable indication of the width of the peaks at half-height, the ratio of the yield of the most probable mode

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[11] J. McElhinney and W. E. Ogle, Phys. Rev. **81**, 342 (1951).

[12] W. E. Ogle and J. McElhinney, Phys. Rev. **81**, 344 (1951).

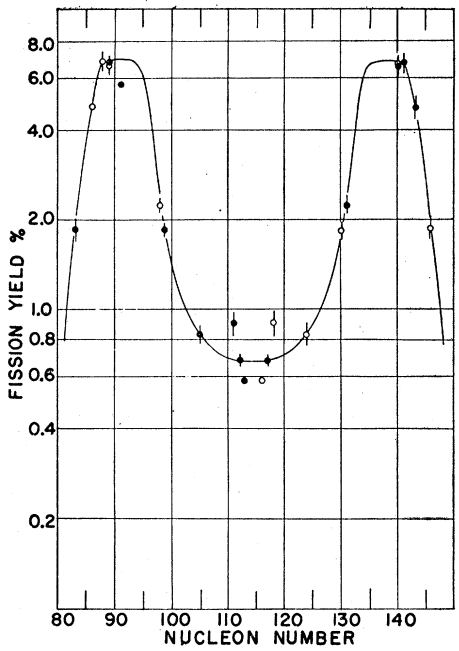

FIG. 1. Absolute yield curve from photofission of thorium with 69-Mev bremsstrahlung. Solid circles represent experimental data at nucleon number A. Open circles represent corresponding mirror images at (229-A).

of fission to that of the symmetrical mode (the peak-to-valley ratio) and the symmetry of the fission yield curve.

II. PROCEDURE

Thorium metal, weighing 13–20 grams, in the form of either pellets or powder, was placed in a glass tube having an 8 mm inside diameter. The sample was aligned with the x-ray beam, 24 cm from the tungsten target of the Iowa State College 69-Mev synchrotron. The specific activation of the sample in the tube decreased by a factor of two for every 7 grams of thorium. A standard irradiation lasted for five hours at an intensity of about 125 roentgens per minute as measured by a Victoreen thimble chamber enclosed in a 0.125-inch lead cylinder fitted with 0.125-inch base, placed one meter from the tungsten target. Deviations from the average intensity during any one bombardment, as monitored by a continuously-recording ionization chamber, were shown to be unimportant.

The freshly-irradiated thorium pellets were dissolved in hydrochloric acid, a small amount of fluosilicate ion being added as catalyst to aid in dissolving small quantities of thorium oxide and thorium carbide, and aliquots were taken for the determination of the various fission products. When powdered metal was irradiated, the target material was transferred to a dry flask and mixed thoroughly after which samples were weighed out and dissolved separately in hydrochloric or nitric acid plus fluosilicate catalyst. Determinations were made in duplicate for each element. In only two cases (strontium-barium and iodine-molybdenum) were analyses made for more than one element on the same aliquot.

A 10- or 20-mg portion of carrier of the element being measured was added to an aliquot of the acidic matrix solution and steps were taken to insure chemical exchange. The element was separated from the matrix solution, purified from all others, and precipitated as a weighable compound. Finally the precipitate, usually weighing from 5 to 30 mg, was filtered in an area of 2 cm^2 on a tared filter-paper disk which was then weighed, mounted on cardboard, and covered with Cellophane.

The chemistry was based on the fission product procedures reported by the Plutonium Project[4] and by Newton,[13] with modifications necessitated by the fact that aliquots of the matrix solution contained 3–5 grams of thorium. Specific modifications will be noted in the appendix which summarizes chemical procedures.

The radioactivity of the samples was measured with a thin mica end-window-type Geiger-Müller tube. The activity of the samples was generally too low to permit complete absorption curves to be taken. Therefore identification of isotopes was made almost exclusively by the interpretation of complete decay curves. Brief aluminum absorption curves were usually taken. These were then back-extrapolated to zero total absorber to correct the counting data for the absorption in the Cellophane, the counter window and the intervening air. Other corrections applied to the counting data were essentially those of Zumwalt,[14] except that because the samples were mounted on filter paper and cardboard, the corrections of Engelkemeir *et al.*[15] were applied for back-scattering, self-scattering and self-absorption. Using the refinements of Zumwalt, the maximum total

TABLE I. Absolute yields from the photofission of Thorium with 69-Mev bremsstrahlung.

Nucleon number	Nuclides isolated and measured	Yields, percent	Estimated reliability percent
83	2.4-hr Br^{83}	1.89	±0.15
89	53-day Sr^{89}	6.7	±0.1
91	9.7-hr Sr^{91}→51-min Y^{91}	5.7	±0.1
99	67-hr Mo^{99}	1.85	±0.10
105	4.5-hr Ru^{105}	0.83	±0.07
111	7.5-day Ag^{111}	0.90	±0.09
112	3.2-hr Ag^{112}	0.68	±0.02
113	5.3-hr Ag^{113}	0.58	±0.01
117	2.72-hr Cd^{117}→1.95-hr In^{117}	0.68	±0.02
131	30-hr Te^{131}→2.5-min. Te^{131}→ 8-day I^{131}	0.81	±0.06
	Total	2.25	±0.10
140	13.4-day Ba^{140}→41.4-hr La^{140}	6.6	±0.5
141	32.5-day Ce^{141}	6.8	±0.5
143	33-hr Ce^{143}	4.8_5	±0.5

[13] A. S. Newton. Numerous fission product separations from Thorium in U. S. Atomic Energy Commission Report AECD-2738, W. W. Meinke, Ed., 1949 (unpublished).

[14] L. R. Zumwalt, U. S. Atomic Energy Commission Report AECU-567, 1950 (unpublished).

[15] Engelkemeir, Seiler, Steinberg, and Winsberg, see reference 4, Paper 4, p. 56.

error is estimated to be 10 percent, most of which occurs in the backscattering corrections. In a few cases where interpolations of the data of Engelkemeir *et al.* were made, errors could have been even larger, but in this case the size of the error could not be confidently predicted.

Either Sr^{91} or Mo^{99} was isolated from each irradiated target and used as a comparative standard for the other isotopes. The yields of Sr^{91} and Mo^{99} were compared in two independent experiments.

III. RESULTS

The experimentally determined relative yields were plotted as a function of nucleon number and the relative yield curve was drawn. Then, since a negligible number of photofission events in thorium result in more than two major fragments,[16,17] the experimental yield points were multiplied by the necessary normalization factor to give an absolute yield curve with an integrated yield of 200 percent.

In drawing this curve, shown in Fig. 1, symmetry about some central nucleon number was assumed. It was further assumed that each peak was roughly symmetrical in the neighborhood of the most probable yield. The normalized fission yield values are shown in Table I.

The curve as drawn is symmetrical about nucleon number 114.5. In addition to the experimental data for nucleon numbers A, points corresponding to the mirror images at 229-A were plotted. Moving the line of symmetry to either 114 or 115 resulted in points which did not fit a smooth curve nearly so well. Therefore the curve seems to indicate that an average of 3 ± 0.5 neutrons was emitted from each thorium nucleus which underwent photofission.

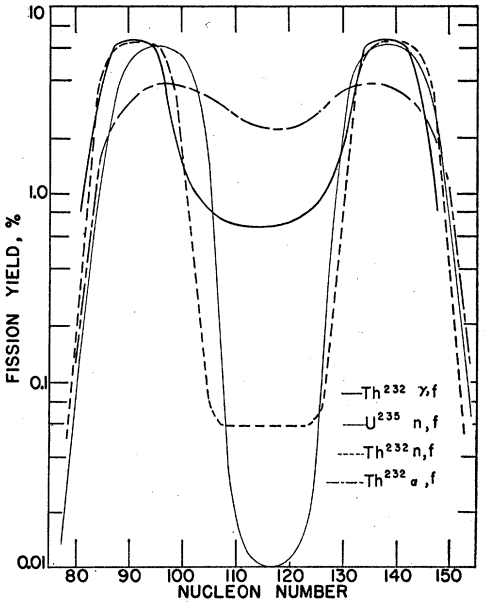

FIG. 2. Yield curves for thermal neutron fission of U^{235} (reference 19), and of thorium fission induced by 69-Mev bremsstrahlung, by pile neutrons (reference 5) and by 37.5-Mev α-particles (reference 18).

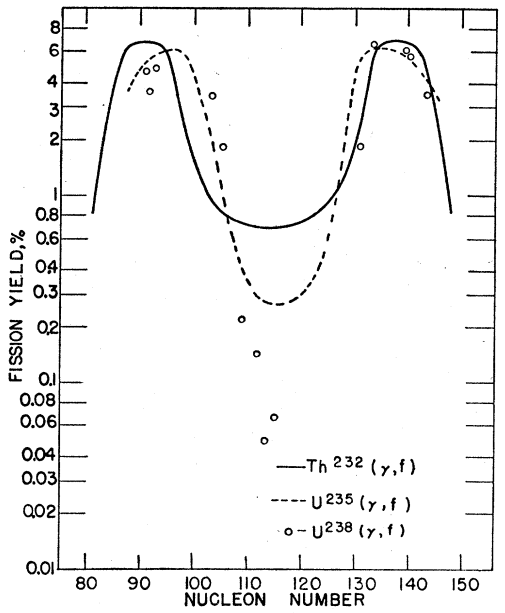

FIG. 3. Photofission yields of Th^{232}, U^{235} (reference 8), and U^{238} (references 9 and 10).

The most probable yields occurred with a magnitude of 6.9 percent at nucleon numbers 91 and 138, giving a value of 1.52 for the most probable mass ratio. The width of each peak at half-height is 12 nucleon units. The peak-to-valley ratio is 10.

IV. DISCUSSION

The yield curves for the fission of thorium by photons, by pile neutrons,[5] and by 37.5-Mev alpha-particles[18] are shown in Fig. 2 along with the well-known curve for thermal neutron fission of U^{235}.[19] The most noteworthy feature of this comparison is that symmetrical fission is ten times more probable in photofission than it is in pile neutron fission, but only about one-third as probable as in fission induced by alpha-particles.

Data are also available on the photofission of U^{238},[9,10] U^{235},[8] and Bi^{209}.[7] In all cases, including the present work, the number of points determined have been so few in number that the fission yield curves, when they can be drawn at all (see Fig. 3), are not sufficiently precise to be subject to fine interpretation. However certain trends can be detected which seem to justify some general statements.

The half-width of the photofission yield curve is 14 in the case of U^{235} and 12 in the case of Th^{232}. The thermal neutron fission yield peaks of U^{235} have a

[16] E. W. Titterton and T. A. Brinkley, Phil. Mag. **41**, 500 (1950).
[17] Goward, Titterton, and Wilkins, Nature **164**, 661 (1949).
[18] A. S. Newton, Phys. Rev. **75**, 17 (1949).
[19] E. P. Steinberg and M. S. Freedman, see reference 4, Paper 219, p. 1378.

half-width of 15.5, whereas the pile neutron fission yield peaks of Th^{232} have a half-width of 14. Thus it appears that, as in the case of neutron fission, the half-width increases with the mass of the compound nucleus. In addition, the cases of U^{238}, U^{235} and Th^{232} seem to indicate that photofission results in definitely smaller half-widths than does low energy neutron fission.

The most probable mode of Bi^{209} photofission is symmetrical.[7] The photofission peak-to-valley ratio is 10 in the case of Th^{232}, over 20 in the case of U^{235},[8] and much greater in the case of U^{238}.[9] The high ratio reported for U^{238} may be partly caused by the fact that photofission was induced by bremsstrahlung with a maximum energy of only 16 Mev, so that this result is not strictly comparable with others. In spite of this qualification, the above data seem to justify the general statement that the tendency towards symmetrical photofission decreases as the nucleon number of the excited nucleus increases.

Further comparison of the photofission yield curves of U^{235} and Th^{232} with their respective neutron fission yield curves reveals that in both cases the heavy peak of the curve predominantly is shifted in the light direction by an amount which, although small, appears to be too large to be accounted for merely by the difference of one neutron in the composition of the respective compound nuclei. The present preliminary data indicate that the photofission yield curve of U^{238} also follows this trend.

Previous reports that the Th^{232} photofission cross section has its peak at 17–18 Mev and practically vanishes above 30 Mev[12,20] led to the belief that in the present experiments the average amount of photoexcitation leading to fission was about 15 Mev, an excitation which is too low to cause emission of many neutrons from the excited nuclei prior to fission. This prediction is corroborated by the fact that the heavy branch of the radiochemically determined experimental yield curve is symmetrical with the light branch, which would not be the case if the neutron yield were high. In addition, comparison of the reported neutron yield of 2.6 per spontaneous Th^{232} fission event[1] with the value of 3 ± 0.5 neutrons per Th^{232} photofission event obtained in the present investigation, shows that the difference is small at most and may in fact not exist at all. If photofission actually does result in a higher neutron yield than does spontaneous fission, the increase is small and may be attributed to that fraction of fission events resulting from relatively high excitation. Further experiments would be required to resolve the doubt which remains as to whether or not photoexcitation ever causes Th^{232} to emit neutrons prior to fission.

This work is described in greater detail in the U. S. Atomic Energy Commission Report, ISC-227 (unpublished).

We are grateful to Dr. P. Chiotti and Dr. D. Peterson for their kind assistance in the preparation of the metallic thorium samples and to Dr. L. J. Laslett, Dr. D. J. Zaffarano and the entire synchrotron crew for their cooperation in performing the bombardment work.

[20] G. C. Baldwin and G. S. Klaiber, Phys. Rev. **71**, 3 (1947).

V. APPENDIX. CHEMICAL PROCEDURES

For each element listed below, the essentials of the chemistry involved in this investigation have been summarized. In the analytical procedures, the practice has been to repeat purification steps two or more times in order to achieve radiochemical purity.

Thorium—The metal sample was dissolved in HCl with a small amount of fluosilicate ion present to clear up the solution. If a minimum of three moles of HCl per mole of Th was present, a speed of solution was obtained which was much higher than that attainable with any other acid or combination of acids. The remaining acid necessary to dissolve the Th could equally well be HCl, $HClO_4$, or H_2SO_4.

Bromine—I_2 was extracted into CCl_4 after the oxidation of I^- to I_2 by dropwise addition of $2M$ cerium(IV). Further dropwise addition of $2M$ cerium(IV) oxidized Br^- to Br_2, which was then extracted into fresh CCl_4. Br_2 was then stripped from the CCl_4 with $NaHSO_3$ solution. AgBr was precipitated and weighed.

The samples decayed with the 2.4-hr half-life of Br^{83}.

Strontium—$Sr(NO_3)_2$ was precipitated with cold fuming HNO_3 after the HCl was displaced from the matrix solution by boiling with HNO_3. Separations of $BaCrO_4$ and $Fe(OH)_3$ were made. Sr was finally precipitated and weighed as $SrC_2O_4\cdot H_2O$.

Activities in the samples were resolved into the 9.7-hr decay of Sr^{91} in equilibrium with 51-min Y^{91}, the 53-day decay of Sr^{89} and the growth and subsequent decay of 57-day Y^{91}.

Molybdenum—Mo was extracted into ether from $6N$ HCl after Br_2 oxidation, precipitated with α-benzoinoxime from $3N$ HNO_3 containing $H_2C_2O_4$, oxidized to MoO_3 with HNO_3 and $HClO_4$, scavenged with $Fe(OH)_3$ and finally precipitated and weighed as $PbMoO_4$.

The samples decayed with the 67-hr half-life characteristic of Mo^{99} in equilibrium with its 6.0-hr Tc^{99} daughter.

Ruthenium—RuO_4 was distilled from the fuming $HClO_4$ into $6N$ NaOH containing Mo holdback carrier, precipitated as a lower oxide with EtOH, dissolved in weak HCl and precipitated as Ru^0 with Mg metal.

The decay curves resolved into the 4.5-hr decay characteristic of Ru^{105} and the growth and subsequent decay of its 37-hr Rh^{105} daughter.

Silver—Ag was precipitated as AgCl and Ag_2S, with intermediate scavengings with $Fe(OH)_3$. It was finally weighed as AgCl.

The decay curves were resolved into three components which were assigned to the 7.5-day Ag^{111}, 3.2-hr Ag^{112} and 5.3-hr Ag^{113}.

Cadmium—Cd was precipitated from cold HCl solution which was 1 percent in thiourea (shown as TH) as $Cd(TH)_2[Cr(NH_3)_2(SCN)_4]_2$ by adding a saturated solution of Reinecke's salt, $NH_4[Cr(NH_3)_2(SCN)_4]\cdot H_2O$. The precipitate was dissolved in weak HCl and precipitated as CdS and $Cd(OH)_2$, scavenged with $Fe(OH)_3$ and scavenged with basic indium acetate. 1.95-hr In^{117} was milked out as basic indium acetate, ignited to In_2O_3, slurried, dried and weighed. Cd was later quantitatively determined as the "Reineckate."

The decay curves of the counting samples resolved into the 1.95-hr decay characteristic of In^{117} and the 4.5-hr decay of In^{115}.

Tellurium—Te was precipitated as Te^0 by SO_2 in $3N$ HCl solution and scavenged repeatedly with $Fe(OH)_3$. The samples were weighed as Te^0.

The decay curves showed the presence of 77-hr Te^{132} in equilibrium with its daughter, 2.4-hr I^{132}, and 30-hr Te^{131} plus its 8.0-day I^{131} daughter.

Iodine—I_2 was extracted into CCl_4 after the oxidation of I^- to I_2 by dropwise addition of $2M$ cerium (IV). The I_2 was reduced to I^- and stripped from CCl_4 with aqueous $NaHSO_3$, and was finally precipitated and weighed as AgI.

The sample, separated 10 days after the end of the irradiation, decayed with the 8.0-day half-life characteristic of I^{131}.

Barium—$Ba(NO_3)_2$ was precipitated with cold fuming HNO_3 after the HCl was displaced from the matrix solution by boiling with HNO_3. Ba was precipitated as $BaCrO_4$ after scavenging with $Fe(OH)_3$, precipitated as $BaCl_2$ with HCl-ether mixture and redissolved in H_2O. The solution was milked of 41.4-hr La^{140} by a $La(OH)_3$ precipitation. The $La(OH)_3$ was filtered, ignited and counted. Ba was later quantitatively determined as $BaSO_4$.

The decay curve of the La sample was resolved to show a 40-hr decay attributed to La^{140}, as well as a 10.6-hr decay assigned to Pb^{212}.

Cerium—Th was extracted into mesityl oxide from the HCl matrix solution containing La, Y and Ce carriers and saturated with $Al(NO_3)_3$. The rare earths were precipitated as hydroxides with NaOH, extracted into tributyl phosphate from a 1*N* HNO_3 solution saturated with $Al(NO_3)_3$, stripped with H_2O and reprecipitated as hydroxides with NaOH. The Ce was oxidized with $HBrO_3$, precipitated as $Ce(IO_3)_4$, dissolved in HCl and H_2O_2 and scavenged with $Zr(IO_3)_4$. Ce was finally precipitated and weighed as $Ce_2(C_2O_4)_3 \cdot 10\ H_2O$.

The decay curves resolved into a 32.5-day decay characteristic of Ce^{141}, a 33-hr decay assigned to Ce^{143}, and an activity which demonstrated the growth and subsequent decay of Pr^{143}.

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The Quadrupole Moment of Li^7

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The quadrupole interaction energy eqQ for Li^7 in Li_2 has been reported to be positive. In an attempt to determine $Q(Li^7)$ a calculation of q has been made using two approximate wave functions for Li_2. One of these is a Heitler-London function including inner shells and the other a twelve term variational function. The two functions lead to dissociation energies of 0.27 ev and 0.48 ev, respectively. The experimental value is 1.14 ev.

The results of the two calculations differ in sign. The more accurate variational function leads to a negative q and consequently a negative value of $Q(Li^7)$. Unfortunately, the electronic and nuclear parts of q are nearly equal in Li_2 so that the magnitude and even the sign are still uncertain.

I. INTRODUCTION

RECENT advances in experimental techniques have made possible the determination of the sign as well as the magnitude of the quadrupole interaction energy eqQ in diatomic molecules.[1,2] Unfortunately, in order to calculate the nuclear quadrupole moment from this data it is necessary to know q, the electric field gradient at the position of the nucleus.[3] This must be calculated from molecular wave functions. A calculation by Foley[4] using the Bartlett-Furry wave function[5] for Li_2 indicates that the quadrupole moment of Li^7 is about $+2\times10^{-26}$ cm^2. The Bartlett-Furry function is a Heitler-London-type wave function in which the inner shell electrons are neglected. James has shown that the good agreement that Bartlett and Furry found between their calculated value of the dissociation energy and the observed value is completely destroyed when the effect of the inner shell electrons is included.[6] Since the value of q calculated with this function is of doubtful accuracy, it was felt that the calculation should be repeated with more accurate wave functions.

II. CALCULATION OF q WITH THE HEITLER-LONDON TYPE WAVE FUNCTION

James has discussed a wave function of the Heitler-London type which leads to a dissociation energy of 0.27 ev.[6] The experimental value of the dissociation energy is 1.14 ev. All electrons are included in this wave function and are described by single particle functions of a form that was used in the treatment of the lithium atom problem. The electronic configurations of the molecule are represented by Slater determinants constructed from these functions. Terms representing the lowest ionic configurations are also considered, and the percentage admixture of these terms, as determined by the variational method, is about 3 percent for an internuclear distance of 2.98A.

For Li_2 the expression for q becomes

$$q=2eq'=2e\left[\frac{3}{R^3}-\frac{1}{2}\int\rho\frac{(3\cos^2\theta-1)}{r^3}d\tau\right],$$

where R is the internuclear distance and ρ is the elec-

TABLE I. Values of $q'(R)$ in Atomic Units.

R (A)	q' (atomic units)
2.49	$1/R^3-0.00388$
2.70	$1/R^3-0.00385$
2.91	$1/R^3-0.00375$

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[1] P. Kusch, Phys. Rev. **76**, 138 (1949).

[2] Logan, Coté, and Kusch, Phys. Rev. **86**, 280 (1952).

[3] The q used here corresponds to that used in reference 2.

[4] A calculation by H. M. Foley quoted in reference 1.

[5] J. H. Bartlett, Jr., and W. H. Furry, Phys. Rev. **38**, 1615 (1931).

[6] H. M. James, J. Chem. Phys. **2**, 794 (1934).