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}.

PHYSICAL REVIEW VOLUME 90, NUMBER 4 MAY 15, 1953

The Quadrupole Moment of Li^7

EDWARD G. HARRIS,* *Department of Physics, University of Tennessee, Knoxville, Tennessee*

AND

MICHEL A. MELKANOFF, *Department of Physics, University of California, Los Angeles, California*

(Received November 10, 1952)

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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* U. S. Atomic Energy Commission Predoctoral Fellow.

[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).

tron density calculated from the molecular wave function. This quantity should be averaged over the vibrational states of the molecule, but in our work this will not be done, since the effect is expected to be small.

The quantity q' was calculated for several values of R in the neighborhood of the equilibrium distance, $R=2.67$A. The results are given in Table I.

It was found that the inner shell electrons contributed very nearly $-2/R^3$ to q', so the first term in q' is the contribution of the nucleus shielded by its two inner shell electrons. The contribution of the remaining molecular charges is included in the second term. This term is primarily due to the functions representing valence electrons and all cross product terms which appear in the charge density. We shall refer to these terms as the nuclear term and the electronic term, respectively. q' is written in this form to facilitate comparison with the results obtained using the James variational wave function. It should be noted that the electronic term varies much more slowly with R than the nuclear term. It was found that including the ionic terms in the wave function increased the electronic term to 0.00394 for $R=2.70$A.

From these results the value of q' at $R=2.67$A is seen to be about $+0.0040$ atomic units. Using the experimental value of $eqQ/h=+0.060$ Mc/sec reported by Logan, Coté, and Kusch,[2] it is found that $Q(\text{Li}^7)=+3.5\times 10^{-26}$ cm^2.

III. CALCULATION OF q WITH THE JAMES VARIATIONAL WAVE FUNCTION

James has found a variational wave function for Li_2 which leads to a dissociation energy of 0.62 ev.[6] Although the discrepancy between this value and the experimental value of 1.14 ev is rather large, this wave function is the best that has been proposed. It consists of a linear combination of 18 Slater determinants, the coefficients of which were obtained by the variational method. The inner shell electrons were described by the same functions as were used in the Heitler-London treatment, and the valence electrons by function of the form $e^{-\gamma\lambda}\lambda^m\mu^j$, where λ and μ are the elliptic coordinates of the electron and the exponents m and j are integers. The chief defect of this wave function seems to be the absence of terms depending on the distance between the valence electrons.

Because of the labor involved in calculating q' with this function, a simpler 12-term function was found which yields a dissociation energy of 0.48 ev. The calculation of q' with this function was laborious but straightforward. Since the wave function was calculated for only one value of the internuclear distance, $R=2.98$A, q could be calculated for this distance only. The result was[7] $q'=1/R^3-0.0089=-0.0032$ atomic units.

If it is assumed that q' has about the same value at the equilibrium distance, $R=2.67$A, as it has at $R=2.98$A, then it is found that $Q(\text{Li}^7)=-4\times 10^{-26}$ cm^2. Because of the rapid variation of the nuclear term with R, this assumption is questionable.

Perhaps a more reasonable assumption is that the results of the last section are at least qualitatively correct and that the variation of the electronic term with R is negligible in the neighborhood of the equilibrium distance. Making this assumption it is found that $q'(R=2.67\text{A})=-0.0011$ atomic units and $Q(\text{Li}^7)=-12\times 10^{-26}$ cm^2.

IV. DISCUSSION

It is rather surprising that the two wave functions which differ so little in the predicted dissociation energy should lead to such greatly different values of q. It may be taken as an indication of the sensitivity of q to changes in the charge distribution in the molecule. In order to gain some insight into the reason for this difference, the charge density calculated with the variational wave function was plotted along the internuclear axis and compared with that calculated with the Heitler-London function. It was found that the two curves were similar, but that the variational function led to a charge density that was more than twice as great at the midpoint of the molecule and slightly less at the ends than the charge density calculated with the Heitler-London function.

Qualitatively, this is a reasonable result since the variational function is sufficiently flexible to take account of the polarization of each lithium atom by the other. In Nordsieck's work on the hydrogen molecule[8] it was found that the Heitler-London function underestimates the charge density at the midpoint of the molecule, but the error is much smaller than it is in the case of Li_2. This suggests that the Heitler-London function for Li_2 could be considerably improved by the addition of terms representing configurations in which one or both of the valence electrons were in $2p$ states in order to take account of polarization. Apparently the effect of polarization is much more important in the case of Li_2 than it is in H_2. This can perhaps be explained by the fact that in the limiting case of the united atom (i.e., when the nuclei coincide) two electrons of the lithium molecule must be promoted to the $2p$ shell. In the hydrogen molecule such promotion does not occur. It seems reasonable to believe that the effect of promotion would make the polarization terms more important in Li_2 than in H_2.

It may be concluded from these results that there is no longer any reason to believe that $Q(\text{Li}^7)$ is positive. Since the variational wave function is more flexible than the Heitler-London function and gives a better value for the dissociation energy, our calculations tend to favor a negative value for $Q(\text{Li}^7)$. It should be

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[7] The second term in q' is the average of the values obtained independently by the authors whose results agreed to within 5 percent. This small discrepancy was attributed to small differences in the wave functions used.

[8] A. Nordsieck, Phys. Rev. 58, 310 (1940).

pointed out that a negative quadrupole moment is in agreement with the predictions of all the simple nuclear models that have been proposed for Li^7. However the magnitude and even the sign of $Q(\mathrm{Li}^7)$ must still be considered to be in doubt for the following reasons:

(1) The quantity q is the difference between two very nearly equal terms which represent, respectively, nuclear and electronic contributions. The electronic term in q is much more sensitive than the dissociation energy to changes in the wave function. Because of this sensitivity improved wave functions may lead to quite different values of q. One of us (M.A.M.) is repeating the calculations with the more accurate 18-term wave function.

(2) The variational wave function has been determined for only one internuclear distance, $R=2.98\text{A}$. The assumption made about the variation of q with R was based on calculations made with a simpler wave function.

(3) The experimental value of eqQ is rather uncertain, since the satellite maxima could not be resolved. There seems to be little doubt, however, about the sign.

(4) No average was made over the vibrational states of the molecule, and no account was taken of rotational distortion. The resulting errors are probably very small.

(5) In both of the wave functions used in these calculations, the 1s functions are of the form $e^{-\alpha r}$. No account has been taken of the shielding effect due to the quadrupole moment induced in the 1s shell. Sternheimer[9] has calculated a correction factor for this effect in atoms. He finds that for the excited lithium atom the observed nuclear quadrupole moment should be multiplied by the factor 1.148. Although our use of molecular wave functions for the valence electrons precludes a direct application of Sternheimer's result, it seems likely that the correction would be of the same order of magnitude.

It seems unlikely that $Q(\mathrm{Li}^7)$ can be calculated with reasonable accuracy from the observed quadrupole interaction energy until a molecular wave function for Li$_2$ is developed which will compare in accuracy with the James-Coolidge function for H$_2$.

We are indebted to Dr. R. J. Finkelstein and Dr. R. D. Present for their valuable suggestions and discussions concerning this work and to Dr. H. M. James who kindly made available his manuscripts on the Li$_2$ molecule.

Note added in proof.—The calculations with the 18-term James function mentioned previously have been completed, and the following results were obtained: dissociation energy $=-0.51$ ev (James originally gave -0.62 ev due to a slight error in his calculations). Using $R=2.98\text{A}$: $q'=-0.0030$ atomic unit, $Q(\mathrm{Li}^7)=-4.2\times 10^{-26}\ \text{cm}^2$.

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[9] R. Sternheimer, Phys. Rev. **80**, 102 (1950); **84**, 244 (1951); **86**, 316 (1952).

PHYSICAL REVIEW VOLUME 90, NUMBER 4 MAY 15, 1953

Excited States of Even-Even Nuclei*

GERTRUDE SCHARFF-GOLDHABER
Brookhaven National Laboratory, Upton, New York

(Received January 20, 1953)

A general survey of excited states of even-even nuclei yields the following results: The nth excited state has usually a spin $I\leq 2n$. For $n=1$, the assignment $I=2+$ (even parity) is compatible with experimental results for 66 out of 68 nuclei investigated. For $n=2$, of 26 nuclei investigated, about one-third have $I=2+$, one-third $I=4+$, and one-third miscellaneous spins of both even and odd parities. The energy of the first excited state plotted against the number of protons or neutrons in the nucleus varies rather smoothly and reaches maxima at closed shells. Wherever the first excited state is very low, e.g., in the rare earths region and for the heavy elements from thorium up, the one-particle model for odd A nuclei is likely to break down except for the ground state. The lack of isomers of odd proton nuclei below magic number 82 may be due to this fact. The average energy of the first excited state of the even-even core in this region is of the order of 0.1 Mev, whereas this energy is of the order of 0.5 Mev for the core of the corresponding odd neutron nuclei ($N<82$). Isomerism in even-even nuclei is discussed. The results are compared with theoretical predictions derived from an extended j-j coupling model and from the liquid drop model of the nucleus.

I. INTRODUCTION

SINCE the strong spin orbit coupling model[1,2] implying a "shell structure" of the nucleus was suggested several years ago, nuclear physics has gravitated toward the study of odd A nuclei. This model, which received its first impetus from a consideration of the pronounced stability of certain nuclear species, soon scored a series of important successes wherever the prediction of spins and parities of nuclear states entered, e.g., in the fields of beta-decay and of isomeric states. However, at the same time a number of features became apparent which seemed to contradict a rigorous single particle picture, such as the large values found for the matrix elements of a number of $E2$ transitions, the sign and size of quadrupole moments, and the scarcity of odd-proton isomers for elements with $50<Z<82$. Also, the model in its present form does not provide a basis for quantitative prediction of energies of nuclear states.

Obviously, some interaction of the single particle with the even-even core has to be taken into account. Whether the whole core has to be considered[3,4] or, in first approximation, only the "loose" particles with the

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* Work was supported by the U. S. Atomic Energy Commission.

[1] M. G. Mayer, Phys. Rev. **75**, 1894 (1949); **78**, 16 and 22 (1950).

[2] Haxel, Jensen, and Suess, Phys. Rev. **75**, 1766 (1949).

[3] J. Rainwater, Phys. Rev. **79**, 432 (1950).

[4] A. Bohr, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **26**, 14 (1952).