THE

PHYSICAL REVIEW

A journal of experimental and theoretical physics established by E. L. Nichols in 1893

SECOND SERIES, VOL. 87, NO. 2 JULY 15, 1952

Angular Correlations in the $Li^6(d,p)Li^{7*}(\gamma)Li^7$ Reaction

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(Received January 30, 1952)

The yield of radiation in the $Li^6(d,p)Li^{7*}(\gamma)Li^7$ reaction has been measured as a function of angle in correlation with protons observed in the direction of the beam and with the beam alone. No anisotropy was detected in either observation. The only plausible theoretical explanation of these results is the assignment of $I=1/2$ to the first excited state of Li^7 in agreement with other recent observations.

I. INTRODUCTION

DIRECTIONAL effects in the emission of particles from nuclei are an important source of information on the properties of nuclear energy levels. The direction of emission may be correlated with the direction of an incoming particle initiating the reaction[1–3] or with the direction of some other emitted particle.[3,4] It has been pointed out[3,5] that the two cases are similar in their formal aspects. General rules have been given[2,3] which govern the complexity of the correlation between any two particles, either incoming or outgoing, in a nuclear process, provided no other directional information is supplied in the experiment.

In a reaction frequently encountered, particle P enters a nucleus to form a compound system, which decays by emission of particle Q to an excited state in a final nucleus and then by γ-emission to the ground state. Angular distributions of the type (P, Q) and (P, γ) have been studied in a number of instances.[6] The angular correlation (Q, γ) has also been investigated in reactions in which particle P enters as an s wave and thus has no influence on the subsequent correlation measurement.[7,8] When the entering wave is not isotropic, however, the correlation (Q, γ) is in reality a correlation (P, Q, γ) among all three particles, since the direction of the incident particle P is generally specified by the experimental arrangement. The situation is equivalent to the (γ, γ, γ) or the (P, γ, γ) correlations which have been treated by Beidenharn, Arfken, and Rose.[9] If the assumption is made that the angular momenta associated with the motion of the particles and with the state of the compound nucleus are all unique, the only additional complexity in the present case arises when particle Q as well as particle P has a spin. The most general case which might be encountered, however, may involve overlapping states in the compound nucleus and multiple values of orbital angular momenta for the particles. The calculation of the correlation function is then exceedingly complex, and the result will include interference terms and a large number of unknown parameters.

The interference effects which involve only magnetic substates and are peculiar to the triple process may be eliminated by observing any two of the radiations in parallel or antiparallel directions.[9,10] The most convenient arrangement for studying states in the final nucleus is to observe the particles Q in the same direction as the beam particles P, a process which we shall designate by $(PQ; \gamma)$. The correlation function may

* Assisted by a contract with the AEC.

[1] R. D. Myers, Phys. Rev. **54**, 361 (1938); E. Gerjuoy, Phys. Rev. **58**, 503 (1940); C. L. Critchfield and E. Teller, Phys. Rev. **60**, 10 (1941); D. R. Inglis, Phys. Rev. **74**, 21 (1948).

[2] E. Eisner and R. G. Sachs, Phys. Rev. **72**, 680 (1947); L. Wolfenstein and R. G. Sachs, Phys. Rev. **73**, 528 (1948).

[3] C. W. Yang, Phys. Rev. **74**, 764 (1948).

[4] D. R. Hamilton, Phys. Rev. **58**, 122 (1940); G. Goertzel, Phys. Rev. **70**, 897 (1946); D. L. Falkoff and G. E. Uhlenbeck, Phys. Rev. **79**, 323 (1950); S. P. Lloyd, Phys. Rev. **80**, 118 (1950); J. A. Spiers, Phys. Rev. **80**, 491 (1950).

[5] L. Eisenbud, Phys. Rev. **76**, 185 (1949).

[6] See the review by Hornyak, Lauritsen, Morrison, and Fowler, Revs. Modern Phys. **22**, 291 (1950).

[7] B. Rose and A. R. W. Wilson, Phys. Rev. **78**, 68 (1950).

[8] W. R. Arnold, Phys. Rev. **80**, 34 (1950); Barnes, French, and Devons, Nature **166**, 145 (1950).

[9] Biedenharn, Arfken, and Rose, Phys. Rev. **83**, 586 (1951).

[10] B. A. Lippmann, Phys. Rev. **81**, 162 (1951).

then be written

$$W(\theta)=\sum_{m_2}\sigma_{m_2}(0)\sum_{m_3}P_{m_2m_3}(\theta), \qquad (1)$$

where $\sigma_{m_2}(0)$ is the partial cross section for the formation of the substate m_2 in the residual nucleus following the emission of particle Q in the direction $\theta=0$; and $P_{m_2m_3}(\theta)$ expresses the relative probability of a transition from the sublevel m_2 of the excited state with angular momentum I_2 to the sublevel m_3 of the final state with angular momentum I_3, by means of 2^{L_2} pole radiation in the direction θ.[11]

Even in those cases in which it seems impractical to calculate the quantities $\sigma_{m_2}(0)$, the correlation function (1) can provide useful information. Since the function

$$U_{m_2}(\theta)=\sum_{m_3}P_{m_2m_3}(\theta) \qquad (2)$$

will be a polynomial in $\cos^2\theta$ of degree not greater than L_2 or I_2, whichever is smaller, a measurement of the correlation function makes it possible to set a lower limit on the value of the spin I_2. A unique assignment of spin may not be possible, even when $L_2>I_2$, since it can happen in rare cases that the function (2) will not attain the maximum allowed complexity.[12] It is also possible that the (P,Q) transition will limit the complexity of the correlation function (1). Except for accidental cancellations, however, it is possible to predict the extent of this limitation for any given case. As has been shown by Biedenharn, Arfken, and Rose,[9] the maximum power of $\cos^2\theta$ in the correlation function cannot exceed either L_0+L_1 or I_1+L_1, whichever is smaller, where L_0 and L_1 are the orbital angular momenta associated with P and Q, respectively, and I_1 is the angular momentum of the compound nucleus.[13] Thus in the triple process the complexity will exceed that allowed by L_1, provided there are no other limitations, and it is possible to have an anisotropic correlation even with $L_1=0$.

A particularly simple application is to the case of an isotropic $(PQ;\gamma)$ correlation. Since $L_2\geqslant 1$, the isotropy probably arises either from the condition $I_2<1$ or from a limitation imposed by the (P,Q) transitions. For the latter to be the case L_1 must equal 0 and either $L_0=0$ or $I_1<1$. In either event the angular distribution of Q relative to P must also be isotropic. If, therefore, the $(PQ;\gamma)$ correlation is isotropic while the (P,Q) correlation is not, the only possible formal limitation is $I_2<1$. In a similar manner the angular distribution of the γ-rays relative to the particles P with Q unobserved is also informative.[14] In this case the complexity of the (P,γ) distribution is limited by the smallest of the quantum numbers (L_0, I_1, I_2, L_2).[9] If again the (P,γ) correlation is isotropic while the (P,Q) correlation is not, the spin I_2 provides the only possible formal limitation.

TABLE I. Angular distribution of radiation emitted in the transition from the substate m_2 of the state I_2 to the state $I_3=3/2$. The functions have the proper relative weight within each group.

I_2	Radiation	m_2	$U_{m_2}(\cos^2\theta)$
1/2	MD	$\pm1/2$	1
1/2	EQ	$\pm1/2$	1
3/2	MD	$\pm3/2$	$6-3\cos^2\theta$
		$\pm1/2$	$4+3\cos^2\theta$
3/2	EQ	$\pm3/2$	1
		$\pm1/2$	1
5/2	MD	$\pm5/2$	$5+5\cos^2\theta$
		$\pm3/2$	$7-\cos^2\theta$
		$\pm1/2$	$8-4\cos^2\theta$
5/2	EQ	$\pm5/2$	$35-45\cos^2\theta+40\cos^4\theta$
		$\pm3/2$	$17+105\cos^2\theta-120\cos^4\theta$
		$\pm1/2$	$32-60\cos^2\theta+80\cos^4\theta$

II. THE $Li^6(d,p)Li^{7*}(\gamma)Li^7$ REACTION

In two reactions it has been established that radiation from the first excited state of Li^7 is emitted isotropically. Littauer[15] has observed an isotropic distribution relative to the incident proton beam in the $Li^7(p,p')Li^{7*}(\gamma)Li^7$ reaction. Unless $L_0=0$ for the entering protons or $I_1=0$ for the compound nucleus Be^8, this result would indicate a spin $I_2<1$, or in the case of Li^7, $I_2=1/2$.† Rose and Wilson[7] have measured the angular correlation between the radiation and the alpha-particles in the $B^{10}(n,\alpha)Li^{7*}(\gamma)Li^7$ reaction, and they find it to be isotropic. In this instance the entering slow neutrons have $L_0=0$ so that the correlation is a simple (α,γ) process, for which quantitative correlation functions can be calculated. The likely possibilities have been examined by Feld[16] and by Devons,[17] and only the case $I_2=1/2$ gives an isotropic correlation function[18] in accordance with the rules discussed above, if the radiation is taken to be either pure magnetic dipole or pure electric quadrupole. Since, however, for the case $I_2=5/2$ (or 7/2) the parity and angular momentum selection rules give $L_1=1$ for the alpha-particles, the correlation function is limited to a $\cos^2\theta$ complexity for

[11] The quantity $\sigma_{m_2}(0)$ may be calculated in the manner used in the references given in 1 and will in general involve many unknown parameters. The calculation of $P_{m_2m_3}(\theta)$ is straightforward and includes only known quantities. See, for example, references in 4.

[12] Arfken, Biedenharn, and Rose, Phys. Rev. **84**, 89 (1951).

[13] If the values of L_0, I_1, and L_1 are not unique, this statement will apply separately to each part of the correlation function corresponding to a particular L_0, I_1, and L_1, but it must be modified appropriately for the terms in which values of L_0, I_1, or L_1 are mixed.

[14] S. S. Hanna, Phys. Rev. **76**, 686 (1949).

[15] R. M. Littauer, Proc. Phys. Soc. (London) **A63**, 294 (1950).

† *Note added in proof:*—A. H. Bethel and R. E. Segel in this laboratory have obtained evidence that the inelastic protons in this reaction are not emitted isotropically, thereby strengthening the assignment of $I_2=1/2$.

[16] B. T. Feld, Phys. Rev. **75**, 1618 (1949).

[17] S. Devons, Proc. Phys. Soc. (London) **A62**, 580 (1949).

[18] See, however, the case $I=3/2$ below.

both types of radiation. Accordingly, it is possible to obtain a cancellation of the $\cos^2\theta$ term by a somewhat unlikely mixture of magnetic dipole and electric quadrupole radiation.[17,19]

In the $Li^6(d,p)Li^{7*}(\gamma)Li^7$ reaction the angular distribution of protons relative to the entering deuterons has been measured at bombarding energies from 0.3 to 1.4 Mev.[20] The measured angular yield functions contain terms as high as $\cos^4\theta$. The odd terms which are observed are indicative of interference between waves of opposite parity. It seems likely therefore that incoming deuterons with $L_0=0$, 1, 2 contribute to the reaction. There is evidence perhaps in these distributions that the (d,p) stripping process contributes partially to the reaction. A forward peak, characteristic of the theory of Butler,[21] begins to form above 1 Mev but is inconspicuous at lower energies, where we shall assume the reaction involves primarily the compound nucleus.

In order to investigate the complexity which may be attained by the yield function of the gamma-rays correlated either to the protons observed in the direction of the deuteron beam or to the beam alone, the distribution functions (2) are listed in Table I for magnetic dipole and electric quadrupole radiation, taking $I_3=3/2$ (odd parity) for the ground state of Li^7 and three choices, $I_2=1/2$, 3/2, 5/2 (all odd parity), for the excited state. It is seen that these functions attain their maximum complexity, except for the case of quadrupole radiation and $I_2=3/2$.[12] With a knowledge of these functions and the rules discussed above it is possible to predict the maximum power allowed in the correlation functions for any particular choice of L_0, I_1, L_1, I_2, and L_2. An examination of all possible cases reveals that a nonisotropic correlation $W(\theta)$ is expected unless $I_2=1/2$ or $I_2=3/2$ (quadrupole). Furthermore, the gamma distribution function relative to the beam is found to be at least as complex as the proton distribution, except for the same two cases. Actual correlation functions $W(\theta)$ have been calculated for a variety of choices of L_0, I_1, and L_1, for $I_2=5/2$ and 3/2 (dipole). In each instance the maximum power of $\cos^2\theta$ was that predicted by the rules. The numerical coefficients ranged from 0.2 to 2.5 times the isotropic term, and in the dipole case were almost uniformly negative for $I_2=5/2$. For all the cases examined involving $L_0=2$, which seems required by the evidence from the (d,p) angular distributions, and for some with $L_0=0,1$, the quadrupole expression involved a term in $\cos^4\theta$. Undoubtedly the real correlation function would include terms corresponding to several choices of L_0, I_1, and L_2, as well as some in which values of these quantities were mixed. Below a bombarding energy of 1 Mev, however,

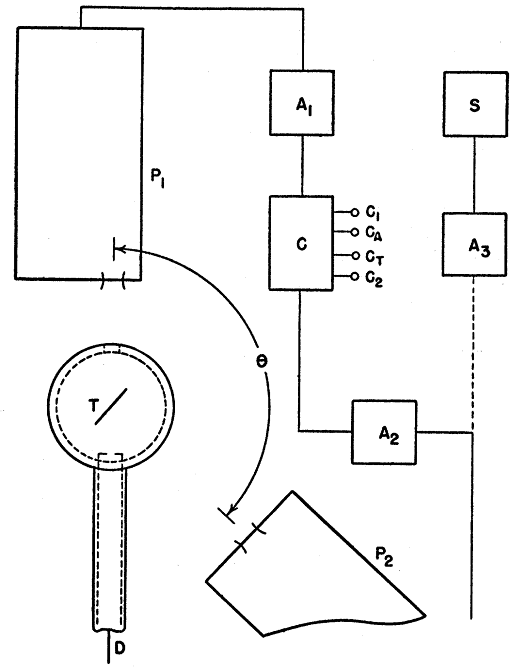

FIG. 1. Schematic diagram of the apparatus: (*D*) deuteron beam; (*T*) lithium target on thin backing; (P_1) fixed proton counter; (P_2) rotating gamma-counter; (θ) correlation angle; (A_1, A_2) 10-mc linear amplifiers; (*C*) coincidence circuit; (C_1, C_2) single channel registers; (C_T, C_A) total and accidental coincidence registers; (A_3) 3-mc linear amplifier; (*S*) single channel discriminator. The last two circuits were used to obtain energy resolution of the radiation observed in the experiment.

the yield of the reaction indicates the presence of a fairly strong resonant state in the compound nucleus.[20] No matter what assignment is made to this resonance, one could expect to find a nonisotropic correlation for the "resonant" part of the reaction, except for $I_2=1/2$ or for pure quadrupole radiation and $I_2=3/2$.

III. EXPERIMENTAL APPARATUS

The energy of the deuteron beam, supplied by the electrostatic accelerator of the Department of Physics, was in the range 600 to 900 kev for which the remarks of the preceding section would apply. In an effort to decrease single channel counting rates enriched Li^6 targets were used in the correlation measurements. One was prepared in a small mass spectrograph,[22] available in the laboratory, with provision for transporting the target without breaking vacuum to the beam tube of the accelerator in order to minimize oxygen contamination. A second target was prepared with lithium sulfate compound[23] having a Li^6/Li^7 ratio of 20. The first target was reasonably thin (less than 0.1 mg/cm^2) and relatively free from oxygen. The

[19] D. R. Inglis, Phys. Rev. **81**, 914 (1951).
[20] Krone, Hanna, and Inglis, Phys. Rev. **75**, 335 (1949); **80**, 603 (1950); W. Whaling and T. W. Bonner, Phys. Rev. **79**, 258 (1950); D. N. F. Dunbar and F. Hirst, Phys. Rev. **83**, 164 (1951).
[21] S. T. Butler, Proc. Roy. Soc. (London) **A208**, 559 (1951).
[22] Built according to the design of Smyth, Rumbaugh, and West, Phys. Rev. **45**, 724 (1934).
[23] Produced by Carbide and Carbon Chemicals Division, Oak Ridge National Laboratory, Y-12 Area, Oak Ridge, Tennessee and obtained on allocation from the Isotopes Division of the AEC.

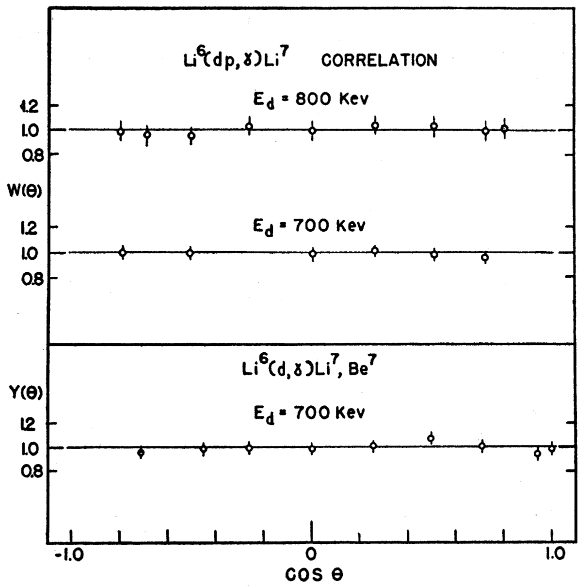

FIG. 2. Measured angular correlations in the $Li^6(d,p)Li^{7*}(\gamma)Li^7$ reaction: (Top) gamma-rays correlated with protons emitted in the direction of the deuteron beam. (Bottom) gamma-rays from Li^{7*} and Be^{7*} correlated only with the deuteron beam.

sulfate target was appreciably thicker (containing roughly the same amount of lithium), but it had relatively less Li^7 (owing principally to a faulty alignment in the lithium separator in the preparation of the first target) and gave a reduced gamma-ray intensity resulting from Li^7 bombardment.

The experimental arrangement is shown schematically in Fig. 1. The aluminum wall surrounding the target was $\frac{1}{8}$ inch thick. Thin aluminum windows were provided for observing charged particles, but one side of the chamber was free for observation of the gamma-rays. The protons and the gamma-rays were detected with anthracene and sodium iodide crystals, respectively, in conjunction with the RCA 5819 photomultiplier tube. In order to reduce gamma and neutron background the anthracene was in the form of a thin, 5-mil flake. Appropriate tests showed that this system, shielded by suitable foils, was an efficient detector of the two proton groups from the Li^6 reaction with unimportant contributions from other radiations. The NaI(TlI) crystal was a $\frac{3}{4}$-inch cube mounted in a Lucite cup, containing mineral oil and making good optical contact with the photomultiplier. Aluminum foil served as a reflector. The efficiency for 0.5-Mev radiation was found to be roughly 50 percent. The proton counter was fixed at zero degrees to the beam, while the gamma-counter was free to rotate in a horizontal plane. Both counters were shielded with lead and aluminum to reduce background. The solid angles of the detectors, about 2×10^{-4} steradian for the gamma-counter and 10^{-4} steradian for the proton counter, produced coincidence rates of about 10/min and total single channel rates on the order of 10^3/sec.

Pulses from the cathode followers of the counters passed through 10-mc amplifiers having gains of about 100. Sensitive trigger circuits were employed in conjunction with blocking oscillators to produce uniform pulses which were mixed on the grids of a pentode for coincidence detection. Single channel rates were obtained directly from the blocking oscillators. Resolving times down to 30 or 40 millimicroseconds were used without excessive loss of true coincidences. In the experiment the background of uncorrelated phenomena in each detector produced an appreciable accidental coincidence rate, which was monitored by feeding the output of each blocking oscillator to a second coincidence detector which received only random pulses. This was accomplished by introducing a 0.5-microsecond delay into the line from one of the blocking oscillators. This procedure is particularly desirable because the unsteadiness of the beam produces an unsteady source of radiation from the target, which results in a higher accidental rate than would be calculated from the resolving time and the single counter rates. In operation it is important to know only the ratio of the resolving times of the two coincidence detectors. This value was measured over long periods of time and found to be subject to only negligible drifts.

IV. THE (dp,γ) CORRELATION[24]

The correlation measurements were obtained by observing the (p,γ) coincidence rate from the $Li^6(d,p)Li^{7*}(\gamma)Li^7$ reaction at angles of the gamma-counter from 35° to 145°. Despite the fact that symmetry about 90° is expected, measurements were obtained in both the front and back quadrants as a check on geometric and scattering effects. The integrated beam intensity was measured with a beam current integrator and also with the total proton count from the fixed proton counter; the two measurements agreed satisfactorily. For each observation the coincidence rate was corrected for slight variations in the distance of the gamma-counter. In all some 100 observations were made at nine angles of the gamma-counter, and approximately 45,000 real coincidences were observed. The measurements were compatible with an isotropic distribution, the observed fluctuations being consistent with the expected statistical uncertainty. The data, presented in Fig. 2, are the results of two separate observations taken several months apart with different targets (see above), somewhat different bombarding energy, and with alterations in counters, circuitry, and geometry. Effects of scattering were investigated by placing lead absorbers in strategic places and by observing the decay of the coincidence counting rate

[24] A brief account of part of the measurements reported in this section has been given in Nature **168**, 429 (1951). Measurements on the (p,γ) correlation in which the protons were observed at 90° have been reported by J. O. Newton, Proc. Phys. Soc. (London) **A64**, 938 (1951). See also W. H. Burke and J. R. Risser, Phys. Rev. **85**, 741 (1952); and Phillips, Heydenburg, and Cowie. Phys. Rev. **85**, 742 (1952).

when various thicknesses of lead were interposed between the target and the gamma-counter.

V. THE (d,γ) ANGULAR DISTRIBUTION

In order to obtain additional evidence for the isotropy of radiation from the excited state under study, a brief investigation was made of the angular distribution of such radiation relative only to the direction of the incident deuteron beam. Energy discrimination was employed in order to discard radiation from target impurities, but the 480- and the 430-kev gamma-rays from Li^{7*} and Be^{7*}, respectively, were not resolved. In these mirror nuclei there is little doubt as to the identical nature of the first excited states.[25] Since the (d,γ) distribution with the proton (or neutron) unobserved will in general differ from the (dp,γ) correlation, unless of course they are both necessarily isotropic, a measurement of the former should provide additional evidence. On the other hand, if the isotropy of the Li^{7*} radiation is conceded, the observation will serve to confirm the expected isotropy of Be^{7*} radiation. The relative yield of Li^{7*} and Be^{7*} radiation was not determined quantitatively in the present experiment, but from observations reported in the literature[6] it can be estimated very roughly that the yields are comparable.

The radiation from the lithium target was detected with a NaI(Tl) crystal similar to the one used in the correlation measurements with minor changes in the mounting to improve the resolving characteristics of the system. All shielding was removed from around the counter assumbly, except for a lead collimator which served greatly to reduce annihilation radiation coming from the walls of the target chamber (arising from the positron activity induced by deuteron bombardment of carbon). The pulses from the photomultiplier after amplification were analyzed in a single channel discriminator[26] with a 1.5 volt channel width. The system was calibrated with annihilation radiation from a Na^{22} source. The angular measurements were obtained by recording the unresolved photopeak of the Li^{7*} and Be^{7*} radiation at several angles with respect to the deuteron beam. No significant angular effects were observed in these measurements, either in the height of the peaks or in the area under the peaks. The data, corrected for geometric effects and small contributions from annihilation radiation, are shown in Fig. 2.

[25] Brown, Snyder, Fowler, and Lauritsen, Phys. Rev. **82**, 159 (1951); D. R. Inglis, Phys. Rev. **82**, 181 (1951).
[26] K. I. Roulston, Nucleonics **7**, 27 (1950).

TABLE II. Coefficients in the series $1+A_1\cos\theta+A_2\cos^2\theta$.

	E_d	A_1	A_2
(dp,γ)	800 kev	0.040	−0.029
(dp,γ)	700 kev	0.009	−0.011
(d,γ)	700 kev	0.023	−0.078

VI. DISCUSSION

As presented the data include the instrumental and geometric corrections. There are in addition small corrections ($\sim$1 percent) arising from the motion of the recoiling Li^{7*} nucleus at the instant of emission of the radiation. After applying these corrections the data were analyzed into series of the form $1+A_1\cos\theta+A_2\cos^2\theta$ by the method of least squares. The results are given in Table II. The coefficients obtained differ from zero by amounts which are easily compatible with the uncertainties in the measurements. In view of the definite and fairly complex asymmetry observed in the (d,p) process, it is striking that in both processes involving the gamma-ray no asymmetry can be detected. Although it is conceivable that an isotropic (or nearly isotropic) correlation function could be constructed for a spin $I_2>1/2$, it seems very unlikely that at the same time the (d,γ) process would not display a detectable anisotropy or vice versa. The most reasonable conclusion is that the lack of anisotropy arises from the nature of the gamma distribution functions, Table I. The choice is between $I_2=1/2$ with either dipole or quadrupole radiation or both and $I_2=3/2$ with pure quadrupole radiation. The latter choice is unlikely in view of the lifetime[27] ($\sim 10^{-13}$ sec) of the radiating state, which indicates that magnetic radiation probably predominates and in addition $I_2=3/2$ seems rather implausible theoretically.[28] The assignment $I_2=1/2$ to the first excited states of Li^7 and Be^7 is the only reasonable explanation for the isotropy of the radiation in the present two observations as well as in the $B^{10}(n,\alpha\gamma)Li^7$ and $Li^7(p,p'\gamma)Li^7$ reactions.

Our thanks are due Paul W. Milich for the construction and care of a considerable part of the equipment used in the experiment, H. M. Watts of the Radiation Laboratory of The Johns Hopkins University for valuable discussions in connection with the circuits employed, and S. M. Shafroth for assistance in measuring the (d,γ) angular distributions.

[27] L. G. Elliott and R. E. Bell, Phys. Rev. **76**, 168 (1949).
[28] H. H. Hummel and D. R. Inglis, Phys. Rev. **81**, 910 (1951).