

tion of the incident beam and for compound-nucleus states well resolved in energy, an analysis of the interferences between compound-nucleus decay and direct processes could lead to information concerning the spins and parities of the compound-nucleus states involved in the reaction.

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Excitation Function of the Reaction $Zn^{64}(n,p)Cu^{64}$ with Neutrons of Energies between 2 and 3.6 Mev

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The relative cross section for $Zn^{64}(n,p)Cu^{64}$ has been measured by an activation method for neutron energies of 2.0 to 3.6 Mev. The absolute cross section at 3.55-Mev neutron energy is found to be 56.4 ± 9.0 mb by comparing the Cu^{64} positron activity with the amount of Si^{31} formed in the $P^{31}(n,p)Si^{31}$ reaction, which has a previously known cross section of 96.2 ± 9.0 mb. The cross section rises monotonically with increasing energy from a value of 12 mb at 2.0 Mev. The experimental results are compared with the predictions of statistical theory; experimental level densities are used in the calculations, but even so the predicted yield is too small. An upper limit of 2 mb is assigned to the cross section for $Zn^{68}(n,\gamma)Zn^{69*}$ at 3 Mev.

INTRODUCTION

THE excitation function of the reaction $Zn^{64}(n,p)Cu^{64}$ is of interest from the various standpoints of fast-neutron detection, reactor design, and nuclear theory. Previously the reaction has been investigated by Bretscher and Wilkinson, who measured the relative yield of the Cu^{64} beta activity at neutron energies between 2 and 3.5 Mev.¹ However, no attempt was made to determine the cross section. The reaction cross section was measured by a number of authors² at 14-Mev incident neutron energy.

The reaction $Zn^{64}(n,p)Cu^{64}$ has a positive Q value of 0.20 Mev, and the product Cu^{64} decays with a half-life of 12.8 hours.³ The decay of Cu^{64} consists of 19% positron emission and 42% of K -capture to the ground state of Ni^{64} , and 39% electron emission to the ground state of Zn^{64} . This paper reports positron activation measurements of the relative yield of the $Zn^{64}(n,p)Cu^{64}$ reaction at neutron energies in the range from 2.0 to 3.6 Mev. The absolute cross section is obtained by comparing the Cu^{64} positron activity with the amount of Si^{31} formed in the $P^{31}(n,p)Si^{31}$ reaction, which has a cross section of 96.2 ± 9.0 mb at 3.56 Mev neutron energy.⁴

EXPERIMENTAL PROCEDURE

The 800-kev Philips cascade generator of the University of Chile delivering an ion beam of about 1 milliampere was used to obtain neutrons by means of the reaction $H^2(d,n)He^3$. The accelerating voltage of the deuterons is controlled by a rotating voltmeter, the scale of which was calibrated previously in terms of energy through the resonances in the $Al^{27}(p,\gamma)Si^{28}$ reaction.⁵

The target was a deuterium self-regenerating target, water-cooled at a speed of two liters per minute. From a systematic study of deuterium self-regenerating targets, Fiebiger concluded that the highest neutron yield was given by a gold target.⁶ Our experience is that the best results could be obtained with a sandwich-type target, in which consecutive thin layers of gold, palladium, and gold are evaporated onto a 1-mm copper backing. Saturation in the neutron yield from a freshly prepared target occurred after $1\frac{1}{2}$ hours of continuous bombardment giving total neutron outputs up to 3.5×10^8 neutrons/sec at 600 kev deuteron bombarding energy. These targets appeared to be very stable and bombardment with currents up to 1 milliampere hardly deteriorated the target. In order to make an estimate of the target thickness, a similar target has been prepared in which the lower gold layer was replaced by one of aluminum. This target was bombarded with protons and from the displacement of the resonance in the $Al^{27}(p,\gamma)Si^{28}$ reaction, we obtained an estimate of the

¹ E. Bretscher and D. H. Wilkinson, Proc. Cambridge Phil. Soc. **45**, 141 (1949).

² E. B. Paul and R. L. Clarke, Can. J. Phys. **31**, 267 (1953); L. Rosen and A. H. Armstrong, Bull. Am. Phys. Soc. Ser. II, **1**, 224 (1956); H. G. Blosser (private communication); D. L. Allan, Proc. Phys. Soc. London **A68**, 925 (1955).

³ Strominger, Hollander, and Seaborg, Revs. Modern Phys. **30**, 585 (1958).

⁴ Grundl, Henkel, and Perkins, Phys. Rev. **109**, 425 (1958).

⁵ P. M. Endt and C. M. Braams, Revs. Modern Phys. **29**, 683 (1957).

⁶ K. Fiebiger, Z. Naturforsch. **11a**, 607 (1956).

average energy loss of the bombarding particles in the target.

Throughout the experiment the neutron yield was monitored by a Hanson and McKibben type long counter⁷ placed at 90° with respect to the deuteron beam, and at a distance of 1 meter from the target. At each deuteron energy, the corresponding neutron energy at each angle in the laboratory system was taken from the tables of Fowler and Brolley.⁸ A check on the neutron energy was obtained by means of a transmission experiment with good geometry on the well-known 2.95-Mev resonance in carbon.⁹ In order to cover the neutron energy range between 2.7 and 3.5 Mev, points were taken at five different deuteron energies between $E_d=200$ kev and $E_d=600$ kev at angles of 0° and 55° with respect to the deuteron beam. From the displacement of the peak in the transmission curve, the spread in neutron energy due to target thickness was found to be about 100 kev at 3 Mev neutron energy, in agreement with the average energy loss of the bombarding particles.

The excitation function was determined by placing seven zinc samples at various angles around the *d*-D neutron source, irradiating them simultaneously for eight hours, and measuring the induced Cu⁶⁴ positron activities afterwards. Since the neutrons produced by this source have energies varying greatly with angle, the energy range from 2.0 to 3.6 Mev could be covered in steps of about 200 kev by irradiations at 400 and 600 kev incident deuteron energy. In order to obtain relative cross sections, the angular distribution of neutrons from the source must be known. The sets of data obtained at the different deuteron energies can then be internormalized if the integrated neutron flux is known at one angle for each irradiation.

The relative angular distributions at the two energies were measured by use of a methane-filled proportional counter (R.C.L., model 203). The counter subtended about the same angle with respect to the neutron source as the zinc samples; thus the measured angular distributions were directly applicable to the zinc irradiations. A correction was made for the variation in counting efficiency of the proportional counter with neutron energy, using the total neutron-proton cross section.¹⁰ The counting rate due to neutrons scattered from the target tube, the walls, and the floor was measured at each angle by placing a paraffin cone of 25-cm length between the counter and the neutron source. The counting rate due to degraded neutrons increased from 5 to 10% with increasing angle.

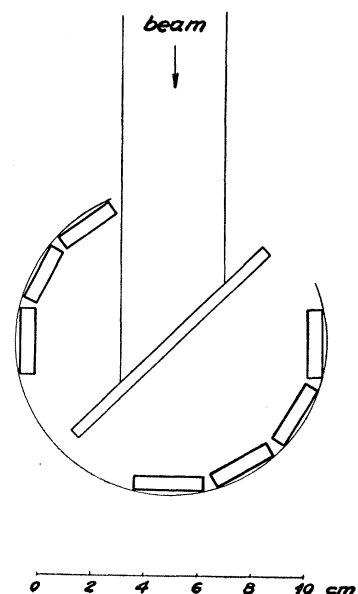
The integrated neutron flux at each deuteron energy

was obtained by the simultaneous irradiation of the zinc samples, and a red phosphorus sample of about 100 mg/cm² thickness placed on top of the Zn at the zero-degree position. The 2.65-hour activity, Si³¹, formed by the $P^{31}(n,p)Si^{31}$ reaction, was measured with a thin-walled, end-window Geiger-Müller Counter (20th Century Electronics Mark EW3H) over a few half-lives. No other activity was found to be present in the phosphorus. The G.-M. counter was calibrated with a set of standard beta-ray sources, which enabled us to derive the absolute activity of the phosphorus sample.

The samples irradiated were disks of spectroscopically pure zinc, about 25 mm in diameter and 6 mm thick. During irradiation the samples were held in well-defined positions, at 6.0 cm from the target inside an aluminum sample-holding ring which was carefully aligned about the target. The positions of the different samples were respectively 0°, 30°, 60°, 90°, 115°, and 143° as illustrated in Fig. 1. In order to reduce the effect of errors due to possible improper alignment, two samples were placed at 90° on either side of the neutron source. A small correction had to be applied in order to account for a slight displacement of the beam off center.

The positron activities were measured by means of the photopeak of the 0.51-Mev annihilation radiation in a NaI(Tl) scintillation spectrometer. The zinc sheets, placed directly on top of the NaI crystal, 4.4 cm in diameter and 5 cm long, were covered with a very thin aluminium foil in order to make sure that all the positrons were annihilated at the sample. The NaI detector was calibrated by using a set of standard gamma-ray sources made up as disks the same diameter as the Zn samples. A source of Na²² was used for the energy calibration; sources of Hg²⁰³, Cs¹³⁷, Sc⁴⁶, and Co⁶⁰ were used for the determination of the photopeak efficiency. The

FIG. 1. Schematic representation of target and sample arrangement.



⁷ A. O. Hanson and J. L. McKibben, *Phys. Rev.* **72**, 673 (1947).

⁸ J. L. Fowler and J. E. Brolley, Jr., *Revs. Modern Phys.* **28**, 103 (1956).

⁹ Bockelman, Miller, Adair, and Barschall, *Phys. Rev.* **84**, 69 (1951).

¹⁰ D. J. Hughes and J. A. Harvey, *Neutron Cross Sections*, Brookhaven National Laboratory Report BNL-325 (Superintendent of Documents, U. S. Government Printing Office, Washington, D.C., 1955).

latter sources were calibrated by comparison with point sources, whose strength was determined by passing a narrow collimated gamma-ray beam through a NaI crystal 5 cm long. The complete pulse-height spectrum and the solid angle subtended, at the source, by the collimation diaphragm were sufficient to determine the absolute gamma-ray yield of the sources. The Co^{60} source-strength was also determined by β - γ coincidence technique, and good agreement was obtained between the two methods.

RESULTS

Two or more irradiations were performed for each of the neutron energies with essentially the same results each time. The results of the irradiations performed under the best conditions are given in Table I. The $Zn^{64}(n,p)Cu^{64}$ absolute cross section of 56.4 ± 9.0 mb at $E_n = 3.55$ Mev has been based on the absolute cross section for the $P^{31}(n,p)Si^{31}$ reaction at 3.56-Mev neutron energy, recently measured at Los Alamos.⁴ The normalization of the neutron flux in the forward direction at $E_d = 400$ kev with that at $E_d = 600$ kev was based on comparing the activities of the phosphorus samples, and interpolating the $P^{31}(n,p)Si^{31}$ cross section from the data of Grundl *et al.* An independent check of this cross section was made using a Lamphere-type of fission chamber¹¹ with natural uranium. The amount of uranium on the foil was determined by alpha counting in a 2π proportional flow counter. A simultaneous irradiation was made with a phosphorus sample mounted on top of the fission chamber. The neutron flux was measured by observing the fission disintegration rate in the uranium and using the known fission cross section for natural uranium. The resultant flux was in excellent agreement

TABLE I. Cross sections for $Zn^{64}(n,p)Cu^{64}$.

Source reaction $D(d,n)He^3$	θ_{lab} from neutron source	Relative angular distribu- tion of source (measured)	Neutron energy (Mev) ^a	Initial counting rate (counts/ min) ^b	$Zn^{64}(n,p)Cu^{64}$ cross section (mb) ^c
$E_d = 600$ kev	0°	1.00	3.55 ± 0.10	10940	56.4 ± 4.0
	30°	0.69	3.40 ± 0.13	6450	53.5 ± 3.8
	60°	0.34	3.04 ± 0.20	2560	46.4 ± 3.4
	90°	0.25 ^d	2.58 ± 0.19	1280	26.4 ± 2.0
	115°	0.29	2.21 ± 0.16	896	17.1 ± 1.3
	143°	0.42	1.99 ± 0.10	728	10.5 ± 0.8
$E_d = 400$ kev	0°	1.00	3.27 ± 0.09	5600	51.3 ± 5.2
	30°	0.73	3.15 ± 0.12	3600	48.8 ± 4.9
	60°	0.37	2.88 ± 0.14	1630	46.4 ± 4.7
	90°	0.27	2.53 ± 0.14	850	30.4 ± 3.1
	115°	0.32 ^d	2.24 ± 0.13	614	19.8 ± 2.0
	143°	0.46	2.05 ± 0.07	672	15.0 ± 1.5

^a The neutron energy spread at the sample due to target thickness and angular spread.

^b Counting rate at the photopeak corrected for background.

^c Standard deviations are relative, including all uncertainties, except the normalization to the cross section of $P^{31}(n,p)Si^{31}$ at 3.55-Mev neutron energy (reference 4).

¹¹ R. W. Lamphere and R. E. Greene, Phys. Rev. **100**, 763 (1955).

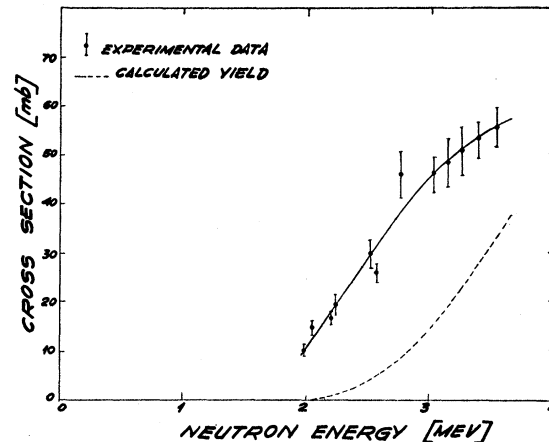


FIG. 2. Excitation function for $Zn^{64}(n,p)Cu^{64}$. The experimental cross sections are given with their standard deviations. The calculated yield is obtained from statistical theory, using experimental level densities of Zn^{64} and Cu^{64} . The nuclear radius used is $r_0 = 1.50 \times 10^{-13}$ cm.

with the one obtained from the Si^{31} activity, the latter being based on the cross section for $P^{31}(n,p)Si^{31}$.

In Table I the energy spread is indicated for each neutron energy; this spread arises primarily from the target thickness and from the angular spread caused by the sample diameter. Also tabulated are the relative angular distributions used at each deuteron energy. These distributions are consistent with those in the literature,¹² taking the finite acceptance angle into account. The angular distribution data are estimated to be uncertain to 3% or 4% at 600-kev deuteron energy, and 5% or 6% at 400-kev deuteron energy. Normalization of the 400-kev data to the 600-kev data involves an additional uncertainty of 5%, to allow for an error in the neutron flux determination at the lower deuteron energy.

Corrections for gamma-ray attenuation, neutron self-absorption, and multiple scattering in the zinc sample have to be considered. These corrections were calculated following the same procedure as described by van Loef and Lind.¹³ The correction factor, defined as the ratio of the observed yield to the true yield, was 0.75 for zinc of 6 mm thickness. An experimental check was made by irradiating a zinc disk sandwiched between two phosphorus samples of 100 mg/cm² thickness each. From the two silicon activities we estimated an average neutron flux in the zinc. Using an average attenuation for the 0.51-Mev gamma radiation, we obtained a correction factor, which within the experimental uncertainties was in agreement with the calculated one.

Small corrections have been applied to all data for decay during irradiation, counter background, and effects due to a slight displacement of the beam. No

¹² N. Jarmie and J. D. Seagrave, Los Alamos Scientific Laboratory, Report LA-2014, 1956 (unpublished).

¹³ J. J. van Loef and D. A. Lind, Phys. Rev. **101**, 103 (1955).

corrections were considered necessary for backscattering from the sample-holding ring, nor for the effect of degraded neutrons from wall scattering. Counting statistics (in most cases 1% or better), uncertainties in the angular distributions, absolute flux determinations, and geometry have all been taken into account in assigning relative standard deviations to the $Zn^{64}(n,p)Cu^{64}$ cross sections. The results are shown in Fig. 2 where the experimental cross sections with their relative standard deviations are given as function of neutron energy. The standard deviation in the absolute cross section includes the efficiencies of the counters, and the absolute error in the cross section of the $P^{31}(n,p)Si^{31}$ reaction at 3.56-Mev neutron energy.

No half-lives other than that of Cu^{64} were observed in the course of this work. Our best value for its half-life is 12.88 ± 0.08 hours (probable error), which is in excellent agreement with 12.82 hours which is an average over the four best values given by Seaborg and co-workers.³ One other known activity produced by neutrons on zinc has a half-life which could interfere with the measurement of the Cu^{64} positron activity. This is the Zn^{69} isomeric state produced by $Zn^{68}(n,\gamma)Zn^{69*}$, which decays with a 13.8-hour half-life and emits a gamma ray of 435 keV on decaying to the ground state of Zn^{69} . In the neutron cross-section compilation¹⁰ Hughes and Harvey give a cross section for fission neutrons of 15 mb. We have established the presence of this activity in an experiment, in which Zn samples were placed behind a paraffin body put around the neutron source. Since fast and slow neutrons were present at the same time, the gamma-ray spectrum showed two peaks at 435 and 510 keV. From the fact that the 435-keV photopeak is absent in the fast-neutron experiments, we can assign an upper limit to the cross section of the $Zn^{68}(n,\gamma)Zn^{69*}$ reaction of 2 mb at 3 MeV neutron energy.

DISCUSSION

The results of the cross section measurements of the $Zn^{64}(n,p)Cu^{64}$ reaction may provide a stimulus for the reinvestigation of the mechanism of neutron counting in ZnS(Ag) phosphors. As both sulphur and zinc show appreciable (n,p) cross sections above 2 MeV neutron energy, neutron detection with these phosphors may be more complicated than was thought.¹⁴

Although many studies have been made of (n,p) reactions by medium-weight isotopes,¹⁵ all but one have been confined to neutron energies higher than 12 MeV. The exception is the reaction $Fe^{56}(n,p)Mn^{56}$ recently reported by Terrell and Holm.¹⁶ These authors find a rapid increase in cross section at about 2 MeV above the energetic threshold of this reaction at 2.9 MeV neutron energy. Taking into account a Q value of $+0.20$ MeV

for the $Zn^{64}(n,p)Cu^{64}$ reaction, the cross section of this reaction shows the same trend. This is no surprise, since the Coulomb barrier penetration factor in both reactions should be very similar. Furthermore the two reactions are equivalent in the sense that the initial nucleus is even-even, which through the (n,p) reaction leads to an odd-odd residual nucleus.

The most probable reaction in the neutron energy range 2 to 4 MeV is inelastic scattering of neutrons. Besides this reaction, competition is possible from the $Zn^{64}(n,\alpha)Ni^{61}$ reaction which has a positive Q value of 3.92 MeV according to the nuclear disintegration energy data.¹⁷ However, it would be expected that its yield is much lower than that of the $Zn^{64}(n,p)Cu^{64}$ reaction, and would, therefore, have little effect on the excitation function. No other reaction is considered to be important.

There is no evidence so far that (n,p) reactions below 12 MeV proceed by a different process than through the compound nucleus. For this reason a comparison of the observed yield of protons with that predicted by the statistical model of nuclear reactions as formulated originally by Blatt and Weisskopf seems to be justified.¹⁸

In order to calculate the cross section for the $Zn^{64}(n,p)Cu^{64}$ reaction, we applied the same expression as Terrell and Holm. These authors used level densities which are based on a Fermi degenerate gas model of the nucleus, and normalized the calculated yield to the experimental cross section at 14 MeV in order to obtain the unknown ratio of level densities between the odd-odd and even-even nucleus. With this normalization, they extended the calculated yield curve to lower energies and obtained cross sections, which are considerably smaller than the experimental ones at neutron energies below 12 MeV.

In our calculations we did not follow this normalization procedure, but, instead, we took level densities which were derived¹⁹ from the results of charged-particle

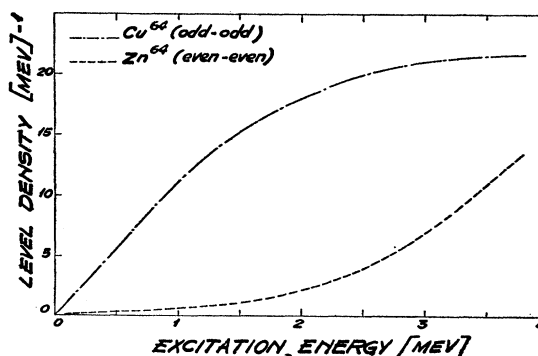


Fig. 3. Experimental level densities. The data for Cu^{64} are obtained from the $Cu^{63}(d,p)Cu^{64}$ results, and the data for Zn^{64} are taken from the known level densities of the adjacent even-even nuclei $Ni^{58,60,62}$ and $Fe^{54,56,58}$.

¹⁷ D. M. van Patter and W. Whaling, *Revs. Modern Phys.* **29**, 757 (1957).

¹⁸ J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley & Sons, Inc., New York, 1952).

¹⁹ J. J. van Loef (to be published).

¹⁴ G. R. Keepin, *Rev. Sci. Instr.* **25**, 30 (1954).

¹⁵ B. B. Kinsey, *Handbuch der Physik* (Springer-Verlag, Berlin, 1957), Vol. 40.

¹⁶ J. Terrell and D. M. Holm, *Phys. Rev.* **109**, 2031 (1958).

scattering data obtained by Buechner *et al.* at the Massachusetts Institute of Technology.²⁰ It turns out that the ratio of the level densities between odd-odd and even-even nuclei varies strongly with excitation energy.

In Fig. 3 the level density for odd-odd Cu^{64} obtained from $Cu^{63}(d,p)Cu^{64}$ results²¹ is shown as a function of excitation energy. The level density rises very rapidly with increasing excitation energy, after which it tends to level off. On the other hand, the level densities of even-even nuclei in general show a more exponential rise with energy. Although no direct data are available for even-even Zn^{64} , the level densities of adjacent even-even nuclei, such as $Ni^{58,60,62}$ and $Fe^{56,58}$, all show a striking resemblance. Thus we feel rather confident that the level density of Zn^{64} changes in the same way. It should be noted here, that the level densities of odd-odd Mn^{56} and Cu^{66} are very similar to Cu^{64} .

The results of the statistical model calculation are given in Fig. 2 along with the experimental data for the

²⁰ Buechner, Browne, Enge, Mazari, and Buntschuh, *Phys. Rev.* **95**, 609 (1954). Further references in 19.

²¹ Figueiredo, Mazari, and Buechner, *Bull. Am. Phys. Soc. Ser. II*, **3**, 38 (1958).

yield of $Zn^{64}(n,p)Cu^{64}$. For the calculations performed, σ_{cn} and σ_{cp} , the cross section for the formation of the compound nucleus by an incident neutron and an incident proton, respectively, were taken from tables given by Blatt and Weisskopf, based on a black square-well model for the nucleus, with the radius constant $r_0=1.50$ fermis. The cross sections were calculated using the experimental level densities of Zn^{64} and Cu^{64} shown in Fig. 3; level densities based on the Fermi degenerate gas model resulted in much lower yields.

Even if we use experimental level densities, there still remains a considerable difference between the experimental and the calculated cross sections. It would be of some interest, therefore, to know if optical model cross sections σ_{cn} and σ_{cp} for a potential well with rounded-off edges would lead to a better agreement, or if another nuclear mechanism should have to be considered at this energy range.

ACKNOWLEDGMENTS

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515-kev and 679-kev Resonances in the Reaction $Na^{23}(p,\gamma)Mg^{24}$

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The 515-kev and 679-kev resonance levels have previously been assigned $J=1^+$ and $J=3^+$, respectively, the latter being formed by LS coupling. An alternative assignment is presented in this paper, according to which the 515-kev level has $J=2^-$ formed by jj coupling with $j_p=\frac{1}{2}^-$ or $j_p=\frac{3}{2}^-$. $J=3^+$ is confirmed for the 679-kev resonance level, assuming jj coupling with $j_p=\frac{3}{2}^+$.

AS is known from angular distribution measurements,¹ the 515-kev and 679-kev resonance levels are assigned $J=1^+$ and $J=3^+$, respectively, the latter being obtained by LS coupling. An alternative assignment is obtained in the present paper, according to which the 515-kev level has $J=2^-$, obtained by jj coupling with $j_p=\frac{1}{2}^-$ or $j_p=\frac{3}{2}^-$. For the 679-kev resonance level, it is confirmed that $J=3^+$, if one assumes jj coupling with $j_p=\frac{3}{2}^+$.

In (p,γ) reactions one has frequently two entrance channels s_1, s_2 , with $s_2=s_1+1$. If we assume an orbital angular momentum l , with a channel spin mixture and multipole mixture of the γ radiation, the angular distribution of the γ radiation can be written

$$W(\theta) = 1 + A_2 P_2(\cos\theta) + A_4 P_4(\cos\theta) + \dots \quad (1)$$

¹ Grant, Rutherglen, Plack, and Hutchinson, *Proc. Phys. Soc. (London)* **A68**, 369 (1955).

where A_ν (ν even) contains, as a factor,

$$F_\nu(ls_1J) + FF_\nu(ls_2J), \quad (2)$$

in which F is the channel spin ratio and J the spin of the compound state. The same is also true for the angular distribution of the later radiation in a $(p,\gamma\gamma)$ reaction in which the first γ radiation is unobserved. A consequence of this is that for every such γ radiation which originates from the compound state, $A_\nu=0$ if

$$F = -\frac{F_\nu(ls_1J)}{F_\nu(ls_2J)} = \frac{W(LJL; s_1\nu)}{W(LJL; s_2\nu)}, \quad (3)$$

which is possible if the expression (3) is ≥ 0 .

This kind of exceptional vanishing of a term can occur when $l=J-s_1$ and $l=s_1+1-J$. It is of interest in cases where A_ν normally vanishes for larger values of ν . Of