Triple Correlations in the $^{60}Ni(p,\gamma\gamma)^{61}Cu$ Reaction

C. R. GOSSETT AND L. S. AUGUST
U. S. Naval Research Laboratory, Washington, D. C.
(Received 9 September 1964)

Triple-directional-correlation measurements have been performed for the $^{60}Ni(p,\gamma\gamma)^{61}Cu$ reaction in order to determine the following assignments for the low-lying states: the excitation energy (in MeV), the spin and probable parity, and the $E2/M1$ amplitude ratio (for the transition from the intermediate state to the ground state) as follows: 0.47, ($\frac{1}{2}^-$), indeterminate; 0.95, $\frac{5}{2}^-$, 0.35±0.03; 1.37, $\frac{5}{2}^-$, $3.78_{-0.13}^{+0.15}$; 1.63, ($\frac{3}{2}^-$), −0.29 to −1.68, or ($\frac{5}{2}^-$), −0.61 to −3.01; 1.89, $\frac{3}{2}^-$, −0.08 to −0.42 or −1.19 to −2.48; and 2.07, ($\frac{1}{2}^-$), indeterminate. Correlations for the first three excited states were observed at each of the resonances at proton energies of 1588-, 1599-, and 1620-keV, while those for the next three excited states were observed only at the 1620-keV resonance. The correlations were all in agreement with previously measured $\frac{3}{2}^-$ assignments to each of these capturing levels and to the ground state of ^{61}Cu. In addition to the above, higher levels for which no spin assignments are made were observed at excitation energies (in MeV) of: (2.4), 2.64, 2.83, and 3.02. All energy measurements of this report are believed to be accurate to ±30 keV. The double sum-coincidence arrangement which allowed the simultaneous measurement of up to 24 correlation functions and which is here described for the first time is discussed in some detail. The results of these experiments are compared with other experimental results and found to be in good agreement. The implications of a reported doublet at about 1.35 MeV are considered with relation to the unusually high quadrupole admixture observed for the 1.37-MeV state in the present experiment. It is shown that the other member of the doublet is not observed in the $(p,\gamma\gamma)$ reaction at the resonances studied, and that the undetected presence of such transitions could not produce the results observed for the 1.37-MeV state. The results of the present experiment are compared with the reported structure of ^{63}Cu and ^{65}Cu and with the core-excitation model which has been applied to these nuclides. This model does not appear to adequately explain the observations for the 1.37-MeV state, if the observed large value of the quadrupole admixture is a proper index of a high degree of collective motion for this state.

INTRODUCTION

TRIPLE-directional-correlation measurements, as applied in the radiative-proton-capture reactions, have proved to be a highly successful spectroscopic tool in the determination of the spin of nuclear excited states and the multipole admixtures of the gamma-ray transitions between these states. Unlike the experimentally simpler double correlation, or angular distribution, the triple-correlation measurements generally permit unambiguous assignments to these quantities if measurements are made in a sufficient number of experimental configurations, called geometries. Nevertheless, triple-correlation measurements in (p,γ) reactions are frequently resorted to only to remove obvious ambiguities from distribution measurements, probably because the necessary coincidence arrangements inherently involve decreased detection efficiency, and because the normal fast-slow coincidence arrangements permit the measurement of the correlation for only a single cascade and in only one geometry at a time. However, it becomes feasible to use the triple-correlation measurements as the principal means of investigation if a sufficient multiplicity is gained for the measurement: that is, if the correlations are measured simultaneously for each two-part cascade present between the capturing level and the ground state, and if these correlations are measured simultaneously in an adequate number of geometries for each cascade. Such an improvement in the multiplicity has been achieved in this laboratory by the use of a double sum-coincidence arrangement. This arrangement has permitted, in the present experiment, the simultaneous measurement of up to 24 correlation functions in a time interval sufficient for a single measurement for the least intense of these transitions. The method also provides a considerable simplification of the pulse-height spectrum. Accordingly, the use of the triple-correlation measurements has been adopted for this experiment as the sole method of determining the spin and admixture parameters.

In the present experiment the reaction $^{60}Ni(p,\gamma\gamma)^{61}Cu$ has been studied to obtain information on the low-lying excited states of one of the odd-A isotopes of copper. Because these isotopes contain a single proton outside a closed shell, they have attracted considerable attention with regard to the application of the core-excitation model. Although speculations on this model have been concerned with the location and spin of the levels and the application of the "center-of-gravity rule" of Lawson and Uretsky,[1] additionally the arguments have been based upon the relationship of the transition probabilities between the presumed members of the expected quartet and the core level. These relationships, discussed by Cumming *et al.*,[2] more recently summarized by Gove,[3] and further treated by Vervier[4] and by Harvey,[5] are based on absolute lifetime measurements with ^{63}Cu and ^{65}Cu, permitted by the stable nature of these nuclides. In this respect, of course, the present

[1] R. D. Lawson and J. L. Uretsky, Phys. Rev. **108**, 1300 (1957).
[2] J. B. Cumming, A. Schwarzschild, A. W. Sunyar, and N. T. Porile, Phys. Rev. **120**, 2128 (1960); J. B. Cumming and N. T. Porile, *ibid.* **122**, 1267 (1961).
[3] H. E. Gove, Phys. Letters **4**, 249 (1963).
[4] J. Vervier, Nuovo Cimento **28**, 1412 (1963).
[5] M. Harvey, Nucl. Phys. **48**, 578 (1963).

experiment does not contribute, as no information on the absolute lifetimes of the low-lying states can be determined by presently available techniques. The present measurements, however, do yield information on relative lifetimes through the measurement of the quadrupole admixture ratio, and abnormally large $E2/M1$ admixtures generally reflect collective motion effects.

Previous information on the low-lying states of ^{61}Cu is contained primarily in a report by Butler and Gossett[6] (hereafter referred to as BG), which provides the preliminary information necessary to the present experiment. In that report, information was developed concerning: the location and intensity of capturing resonances below $Ep=1.8$ MeV, the approximate branching ratios for the more prominent of these resonances, the energies of many of the low-lying excited states in the residual nucleus, and probable spins and parities of some of the capturing levels and of the first excited state from distribution measurements of the primary gamma rays to the ground and first-excited states. Subsequently, in an investigation of the decay of ^{61}Zn, Cumming[7] has observed gamma rays which he has fit into the level scheme reported in BG. On the basis of the intensities of the β^+ transitions inferred from these measurements, some limitations may be imposed on the possible spin values of the levels concerned, although no correlation measurements were made to predict definite values. Very recently Blair[8] has performed an experiment on the ^{60}Ni(^{3}He,d)^{61}Cu reaction. He observes deuteron groups corresponding to the excitation of most of the states observed in the present experiment and makes l-value assignments for the proton stripping on the basis of the forward-angle deuteron distributions. The agreement of these results with the present experiment is apparently good.

A preliminary report concerning the first three excited states of ^{61}Cu was presented at a meeting of the American Physical Society.[9] A detailed account of the experimental instrumentation and technique is given in an unpublished report.[10]

SUM-COINCIDENCE METHOD

In the sum-coincidence method, due to Hoogenboom,[11] an analog differential discriminator (hereafter called the sum gate) is applied at a pulse height corresponding to the excitation energy of the capturing level to the linear sum of the outputs of two detector systems, for which the gains have been adjusted to produce equal response. If this sum condition and an auxiliary fast-coincidence condition between the two detectors is applied to select (gate) events from the total output of one of the detectors, the resulting spectrum provides several advantages over spectra resulting from conventional fast-slow coincidence techniques. First, the sum condition restricts attention to those events in which the full energy of the gamma rays has been deposited in the detectors, thus producing a single nearly Gaussian peak for each of the gamma rays. Second, the single sum gate allows the detection of either member of a two-part cascade under circumstances where the detection of the other member of the cascade in the other detector is assured. Third, because all two-part cascades from the capturing level to the ground state have the same total energy for the sum, the sum gate provides a similar response for each of these cascades and permits the detection of all such cascades present at a given resonance in a single spectrum.

Although the advantages of the sum-coincidence method are apparent, the method has not been widely used in the measurement of triple correlations, largely because its application to measurements necessarily extended over long time periods requires extreme stability in the response of each of the detectors and of the sum gate in order to prevent distortions of the resultant spectra. In the present experiment, this difficulty has been overcome by the use on each detector of a light-source-based gain stabilization system that produces stabilities of the order of 0.1 or 0.2% over a working day. This system, which is based on principles reported by Marlow,[12] is fully described in the detailed report on the present experiment.[10]

A second difficulty, which has been described by Draper and Fleischer,[13] concerns the transfer of energy between the two detectors and arises principally in cases of the escape from one detector and the capture in the other of either an annihilation quantum from the pair-production interaction or a back-scattered quantum from the Compton interaction. These transfer events give rise to satellite peaks that occur for cases which meet the sum-coincidence requirement and which would otherwise have been properly treated, had not the transfer occurred, or to events which satisfy the sum requirement only and for which the transfer itself supplies the fast coincidence condition. This difficulty has been greatly reduced in the present arrangement by the use of conical Pb collimators which almost completely eliminate such low-energy transfer events except those which pass through the restricted collimator opening. The elimination of the satellite peaks enhances the ability of the sum-coincidence method to detect very weak two-part cascades in the presence of much stronger radiation. This ability is further enhanced by the use of a moderately fast coincidence circuit ($2\tau\sim80$

[6] J. W. Butler and C. R. Gossett, Phys. Rev. **108**, 1473 (1957).
[7] J. B. Cumming, Phys. Rev. **114**, 1600 (1959).
[8] A. G. Blair (private communication).
[9] C. R. Gossett, L. S. August, and P. A. Treado, Bull. Am. Phys. Soc. **9**, 472 (1964).
[10] C. R. Gossett, and L. S. August, U. S. Naval Research Laboratory Report No. 6186, 1965 (unpublished).
[11] A. M. Hoogenboom, Nucl. Instr. Methods **3**, 57 (1958).
[12] K. W. Marlow, Nucl. Instr. Methods **15**, 188 (1962).
[13] J. E. Draper and A. A. Fleischer, Nucl. Instr. Methods **9**, 67 (1960).

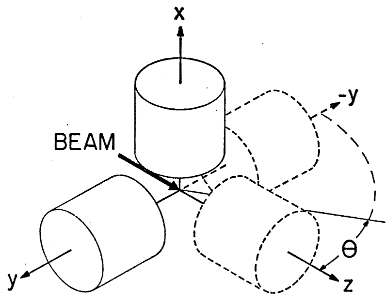

FIG. 1. The relationship of the three detectors used in the present experiment to provide the geometries defined in Table I. The two detectors shown with solid lines are fixed in position, while the third detector moves between the two positions shown with dashed lines.

nsec) and special techniques to prevent pile-up events and false gating (see Ref. 10).

The present experiment uses several of the Chalk River geometries which provide that one of the detectors be placed at an angle of 90° to the beam direction and the other detector be moved on an arc of one of the principal planes. For each such geometry a similar geometry exists, different only in whether the primary member of the cascade is detected in the fixed or moving detector; of course, in the sum-coincidence method correlations are simultaneously measured in both members of such a related pair of geometries. Under the condition of the fixed detector at 90° to the beam, each correlation may be expressed as a simple normalized Legendre expansion in even powers restricted to the fourth order: $W(\theta)=1+A_2P_2(\cos\theta)+A_4P_4(\cos\theta)$. Thus, although each correlation yields two significant ratios, A_2 and A_4, the latter is considerably less sensitive and in most correlations is near zero. This lack of sensitivity effectively reduces the number of sensitive ratios to two for a single sum-coincidence arrangement. Therefore, to provide four independent A_2 values, it has proved expedient to provide an additional pair of geometries for each cascade by supplying a second sum-coincidence arrangement. This has been achieved, as illustrated in Fig. 1, by the use of a single additional detector fixed at 90° to the beam and 90° from the other fixed detector. Thus, with the beam in the $+z$ direction and with detectors fixed on the $+x$ and $+y$ axes, the third detector is moved in the y-z plane on an arc between the $+z$ and $-y$ axes in terms of an angle θ to the $+z$ axis. For convenience, the four geometries used have been labeled: A, B, C, and D. These are defined in Table I in terms of the equivalent Chalk River cases and the spherical angles θ and ϕ. This choice of geometries allows three detectors to operate simultaneously with an angle of no less than 90° between any pair, as required by the collimator arrangements. By placing the moving detector in the horizontal plane the mechanically more complex problem of moving a shielded detector in a vertical plane has been avoided. It should be noted that this

TABLE I. The definition of the geometries (experimental configurations) used in the present experiment in terms of the equivalent Chalk River geometries and the spherical angles θ and ϕ, where the subscript 2 refers to the primary, and the subscript 3 to the secondary members of the two-part cascade from the capturing level to the ground state.

Geometry	Equivalent geometry[a]	θ_2	θ_3	ϕ
A	I	variable[b]	90°	180°
B	II	90°	variable	180°
C	VI	90°	variable	90°
D	VII	variable	90°	90°

[a] Reference 18.
[b] Variable: $0° \leq \theta \leq 90°$.

arrangement produces the equivalent of two legs of the spherical octant used at Chalk River. The degree of overdetermination thus provided has proved adequate in the present experiment, in which the zero-spin ground state for the target nucleus removes the possibility of admixture in the entrance channel.

EXPERIMENTAL APPARATUS

Even with the multiplicity gained by the sum-coincidence method, the reaction yield from this experiment is so low that a large solid angle must be provided for the detectors. The arrangement designed to accomplish this condition is shown in Fig. 2, which is a section through the target in the horizontal plane. Shown are two of the three detectors, the third being normal to the plane of the diagram (see Fig. 1). The three detectors, each a 3-in.-diam×3-in.-long NaI(Tl) crystal, are contained within conical Pb collimators with acceptance half-angles of 26.5°. The front faces of the crystals are placed at 6.0 cm from the center of the target; this distance being the smallest deemed consistent with providing the necessary shielding between detectors, while permitting the detectors to be placed as close as 90° to each other. All three detectors are

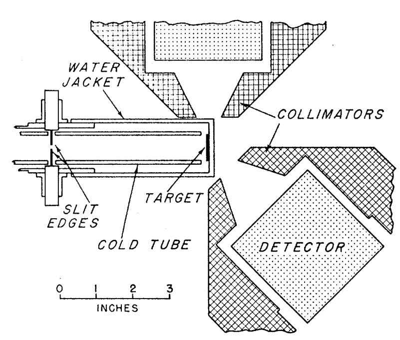

FIG. 2. Simplified section through the horizontal plane at the target showing the relationship of two of the detectors and Pb collimators to the target and beam defining slit edges. The beam enters from the left to pass through the slit and strike the center of the target. One detector is shown fixed at 90° to the beam, while the second (shown at 45°) moves between 0° and 90°. A third detector and collimator (not shown) is fixed normal to the plane (see Fig. 1).

capable of radial motion and the two shown in the horizontal plane are capable also of angular motion between the beam direction and 90° to either side.

The beam of protons enters from the left in Fig. 2, passes through the slit edges (an additional pair is normal to those shown), and illuminates a $\frac{1}{8}$-in.-diam spot at the center of the target. The target consists of a surface layer, approximately 2 keV thick to 1.5-MeV protons, of isotopically enriched ^{60}Ni electrodeposited (as described in BG) on a $\frac{3}{4}$-in.-diam by 0.025-in.-thick Au backing. This backing and the last set of beam defining slit edges are of high purity Au, heavily etched to remove surface contamination. The target chamber (shown schematically) consists of a double-walled water jacket providing sufficient cooling to allow steady bombardment with a 50-μA proton beam. Also shown in Fig. 2 is the cold tube, maintained at liquid-nitrogen temperature, which serves to protect the target and slit edges while under bombardment from the accumulation of contaminants (as described in BG). The details of the apparatus are given in Ref. 10.

The use of a cylindrical rather than spherical geometry in the vicinity of the target is not standard in correlation work; however, placing the target normal to the beam, rather than at 45°, allows the measurement of the correlation to either side in the horizontal plane (as described below in Experimental Procedure). The materials in the vicinity of the target are as light as possible, and correction factors to account for the unequal absorption of gamma rays from the target by the anisotropic distribution of materials have been calculated as a function of the angle of the detector. The results of these calculations have been tested experimentally with isotropic radiations and found to be accurate, even at the lowest energies present in this experiment (0.47 MeV), to an accuracy of better than 2%.

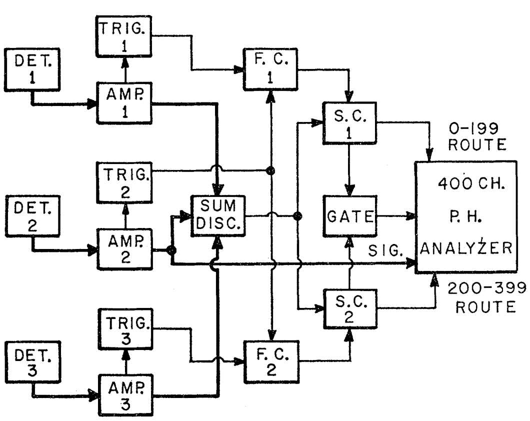

FIG. 3. Simplified functional block diagram of the electronics illustrating the manner in which the two spectra from the double sum-coincidence arrangement are stored in the two halves of the pulse-height-analyzer memory according to the coincidence condition. Identified are: detectors, amplifiers, trigger circuits, sum discriminator, and fast and slow coincidence circuits. The analog signal paths are indicated by heavy lines.

The problem of locating and maintaining the source position (momentary position of the beam on the face of the target) is intensified by the closeness of the front face of the collimators, the openings of which provide the definitive elements of solid angle for the detectors. To achieve alignment, a beam positioning device (not shown) replaces the target chamber of Fig. 2 such that a $\frac{1}{8}$-in.-diam aperture followed by a Faraday cup (both water cooled) is in the position of the target plane and is aligned with respect to the center of the table by a micrometer device. The beam, at the full intensity used in the experiment, is then directed to provide minimum current intercepted by the aperture with maximum current transmitted to the Faraday cup and the slit edges adjusted to define this position.

The electronic arrangements associated with the storage of the two separate spectra evolved in the double sum-coincidence arrangement (described under Sum-Coincidence Method) are illustrated in the simplified functional block diagram of Fig. 3. Here the split-memory feature of the pulse-height analyzer is used to permit the storage of the information from selected pulses from the moving detector (shown here as Det. 2), as routed into one or the other halves of the memory, according to which of the fixed detectors the event in the moving detector is in coincidence. Thus, the pulse amplitude requirement is applied by the differential discriminator (sum gate) to the sum of the adjusted and stabilized outputs of all three detectors; a gate pulse, allowing the analysis of the associated pulse from the moving detector, is produced by a slow coincidence of a sum condition and a fast-coincidence condition between the moving detector and either of the fixed detectors; and the event is stored in the appropriate half of the memory as routed by the particular fast-slow coincidence condition. Double-delay-line amplifiers with associated pulse-crossover-pickoff circuits have been used to generate the trigger pulses for the coincidence arrangements in order to provide minimum walk over the wide range of pulse heights which may be selected by the sum condition. In actual use, either Det. 2 or Det. 3 (both physically in the horizontal plane) may be chosen as the moving detector, and switching arrangements (not shown here) allow this substitution.

EXPERIMENTAL PROCEDURE

The yields of two-part cascades observed in this experiment are too weak to obtain adequate statistics for any but the strongest transitions in the course of a single day. Thus, in addition to the requirement of stability over the course of a day, it is required that the conditions may be reproduced from day to day to allow the summing of individual spectra taken over an extended period. The instrumentation used in this experiment has provided excellent stability. In addition the most pertinent factors are checked daily before the correlation runs and readjusted if necessary.

In the measurement of the correlations, the three-angle approach recently reported by Reich, Merrill, and Klema[14] has been used. It is shown in that report that measurement at three particular angles, with additional observations for the center angle, produces an enhanced statistical precision. In the present experiment the three-angle approach, with slight modification, additionally provides better control of the chief source of systematic error present. Thus, because the beam position at the target (see Experimental Apparatus) may be significantly displaced from the center of the system only *in the plane of the target*, i.e., normal to the beam direction, control of this problem is best made by repeated measurements at a small number of angles, if those angles are observed to *both sides* of the beam axis. Therefore, in the procedure used in the present experiment, a large number of shorter runs are made, distributed among the three angles, and further, such that half of these at each angle are obtained with the fixed detector (in the horizontal plane) to one side and the other half to the other side. A 14-run sequence is used, distributed two, three, and two, to each side at the angles 0, 43.6, and 90°, respectively. The sequence is systematically arranged so that any slow drifts in condition are apt to be distributed properly among the angles.

The monitor counts, used for normalization of the runs at different angles, are produced by a differential discriminator (monitor gate) placed over a portion of the upper half of the pulse-height spectrum from the fixed detector out of the horizontal plane. Difficulties due to drifts are minimized by the gain stabilization of the detector system and the choice of the position of the discriminators. The monitor counts are corrected for background and system live time. The latter correction includes both the effect of a blocker signal in the coincidence circuits[10] and the normal analysis and storage dead time of the pulse-height analyzer. Also monitored are the routing pulses corresponding to the total number of events meeting the sum-coincidence conditions in each geometry set.

Various possible sources of background in the sum-coincidence arrangement have been investigated; of these only two were found to be of any significance: accidental time coincidences, which primarily affect only the extremes of the spectra and which are due largely to the flux of low-energy Coulomb excitation gamma rays from the target backing; and the effect of the next-lowest-energy resonance from the Ni reaction,[15] which depends upon the target thickness and the relative spacing and intensities of the resonances. The effect of the accidentals was evaluated by standard techniques. At only the 1620-keV resonance was correction necessary for the effect of the next-lowest-energy resonance, the nearly equally intense resonance at 1607 keV. For each target that was run at the 1620-keV resonance, the amount of contribution from a resonance 13 keV lower in energy was evaluated to allow correction for the different correlations observed for that resonance. Nonresonant high-energy gamma-ray effects, due to reactions induced by the incident beam with system contaminants, have been reduced to negligible proportions by the cold-tube and target-backing etching procedures. The possible effects of the detection of all members of a three-part cascade were also evaluated by requiring a triple sum-coincidence and found to be generally negligible, as expected, since such detection involves an additional efficiency term of approximately 10^{-2}. It is apparent from the analysis of various sum-coincident spectra that an additional small background (which may at least be approximated as constant) is present although its origin remains undetermined.

DATA ANALYSIS

An example of the data obtained from the present arrangements is shown in Fig. 4, where both an ungated and a sum-coincident-gated spectrum are shown normalized to the same yield. Readily apparent are both the improvement in peak definition and the accompanying loss in intensity in the lower spectrum. A single peak exists for each gamma ray occurring in a two-part cascade to the ground state; and these peaks are nearly Gaussian, although a distortion on the side of the peak

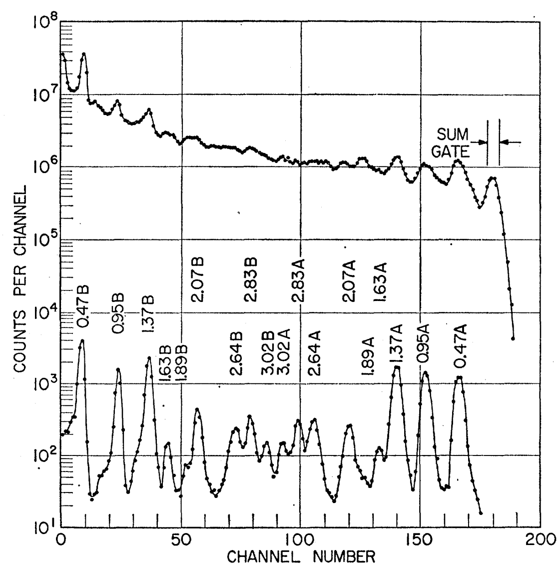

FIG. 4. Ungated (upper) and sum-coincidence gated (lower) spectra, normalized to the same reaction yield, as obtained at the 1599-keV resonance. The peaks of the sum-coincidence spectra are labeled as to the intermediate excited state (energy in MeV), involved in the two-part cascade to the ground state, and as to whether the transition is the primary (A) or secondary (B) member of the cascade.

[14] C. W. Reich, J. A. Merrill, and E. D. Klema, Nucl. Instr. Methods **23**, 36 (1963).

[15] The latter effect is not to be confused with coherent interference of these levels, which is negligible in this case because of the extreme narrowness of the natural widths of the capturing levels (see Ref. 6).

away from the middle of the spectrum is apparent, particularly for the more narrow peaks in the lower portion of the spectrum. As the sources of these distortions are not completely understood, the determination of the yield of gamma rays associated with a given peak has been restricted to fitting the Gaussian portion of the peaks.

Because of the remarkable stability and reproducibility of the system, it has been possible to determine the peak position and width parameters for the Gaussian peaks from data obtained by summing individual spectra taken under varying conditions of angle and geometry and to apply these parameters to the fits of the peaks in individual runs (or sums of small numbers of runs) in order to determine the remaining parameter, amplitude, for these individual runs (or sums). Where necessary, corrections are made for the effects of distortions, background, and incompletely resolved neighboring peaks. The peak yields thus obtained are corrected as a function of the angle of the detector for the unequal absorption of gamma rays in the anisotropic distribution of materials surrounding the target (see Experimental Apparatus), and the runs from a single day normalized on the basis of the corrected monitor counts. The resulting yields as a function of angle for each geometry and transition are subjected to weighted-least-squares fits to the Legendre expansion for both the first two and the first three terms. In the error estimates not only the counting statistics, but also an estimate of the error introduced in the consideration of distortion, background, and unresolved peak effects, are included, based upon the relative contribution of the Gaussian peak to the total counts observed.

PREDICTED CORRELATION FUNCTIONS

In the case of proton capture with a zero-spin target nucleus the relevant parameters (in the notation of Devons and Goldfarb[16]) are the seven quantized variables: a, the initial channel spin; l_1, the incoming orbital momentum; b, c, and d, the spins of the capturing, intermediate, and residual (ground) states, respectively; and L_2 and L_3, the multipolarity of the primary and secondary gamma rays; as well as the two unquantized variables: δ_2 and δ_3, the multipole amplitude ratios of these gamma rays. In the present experiment the channel spin is just that of the incident proton because of the zero target spin, the spin of the ground state of the residual nucleus is known from other measurements,[17] and the spin of the capturing states have been measured (with possible ambiguity) by angular distributions in BG. The orbital angular momentum l_1 of the incoming proton depends upon the spins and parities of the capturing state and the target nucleus. Thus, the principal unknowns to be determined experimentally are the spin of the intermediate state and the multipole admixtures of the gamma rays populating and de-exciting this state. Theoretical correlation functions, yielding the coefficients of the Legendre expansion, are calculated for various possible combinations of the spins of the states and as a function of the two independent admixture ratios by use of the tabulations of Ferguson and Rutledge.[18] That report contains all of the relevant formulation, which will therefore not be repeated here. Because the prediction for the coefficient of the Legendre expansion, A_2 or A_4, for a given set of the quantized variables produces in the general case a ratio of two double quadratic equations in the independent variables, δ_2 and δ_3, the contour plot has been chosen to represent such a function for search and display purposes. Thus, in the comparison of the experiment with theory, the experimental range of values of the Legendre expansion coefficient for each geometry results in a band of allowed values of the δ's in the contour plot representation. Successful agreement between theory and experiment is then indicated by the overlap of all relevant bands. The use of four geometries generally removes ambiguities and allows only one such overlap.

In triple-correlation analysis the correction for the finite solid angle of the detectors must be incorporated in the theoretical prediction. The formulation of this correction for cylindrical detectors was reported by Rose,[19] and several tabulations based upon this analytical approach are in existence. Such analytic calculations, however, are based upon "any interaction" of the incident gamma ray with the detector. Such correction coefficients do not adequately account for the actual physical situation in that events which are accepted for analysis in the sum-coincidence arrangement are not these which have undergone "any interaction" with the detector, but only those which deposited their *full energy* in the detector. As events which interact nearer the walls of the detector are less likely to deposit the full energy of the gamma ray in the detector, the finite acceptance angles subtended by the detectors are less effective in smearing the observed distribution, and the proper correction factors should be less severe than those predicted on the basis of "any interaction." The generation of correction coefficients on the basis of the full-energy absorption is not subject to analytical calculation nor to reasonable approximation, and Monte Carlo techniques have been used to simulate the interaction of the gamma rays with the detector in order to determine the effect of considering only such events. Computer programs prepared by Dr. Charlotte Davisson of this Laboratory were used to determine

[16] S. Devons and L. J. B. Goldfarb, in *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1957), Vol. 42, p. 362.

[17] W. A. Nierenberg, H. A. Shugart, and H. B. Silsbee, Bull. Am. Phys. Soc. **2**, 200 (1957); J. B. Reynolds, R. L. Christensen, D. R. Hamilton, W. M. Hooke, and H. H. Stroke, Phys. Rev. **109**, 465 (1958).

[18] A. J. Ferguson and A. R. Rutledge, Atomic Energy of Canada, Ltd., Report CRP-615, AECL-420, 1957, revised and reprinted 1962 (unpublished).

[19] M. E. Rose, Phys. Rev. **91**, 610 (1953).

correction coefficients under various circumstances of crystal size, source-to-crystal-face distance, and gamma-ray energy.[20] These results have been presented in a manner which readily allows consideration of the limiting effects of the conical collimator and of the fringe penetration of the collimator edges. The corrections used in the analysis of the present experiment include such effects as determined for the experimental arrangement used.

IDENTIFICATION OF STATES

The enhanced sensitivity for the detection of weak two-part cascades in the presence of stronger transitions, as evidenced in Fig. 4, permits the identification of states not readily identified from ungated spectra or, in most cases, from normal fast-slow coincidence work. Thus, all of the low-lying states reported in BG have been identified in the present experiment as well as several additional states. However, the effect of the enhanced sensitivity of the sum-coincidence method is somewhat diluted for states above the first few in that the possibility of further cascade through some of the lower-lying states increases generally with increasing excitation energy. The efficiency for the detection of such three- or four-part cascades is considerably reduced from that for the two-part cascade, and multiple (>2) cascades are not normally observed. Thus, the observation of states with spins differing by more than two units of spin from that of the capturing level is improbable in this experiment, as such states could be populated only by a cascade of at least three parts if it is assumed that octupole transitions from the capturing level will not be observed to compete.

Although necessary to the determination of some of the transitions, the sum-coincidence method does not lend itself to as great precision in the measurement of the energy of gamma rays as can be obtained under ungated but gain stabilized conditions. Nevertheless, the stability of the present system is such that it appears reasonable to assign a probable error of ±30 keV to each of the gamma-ray-energy measurements of this report, and thus to the energy of each of the excited states reported. This assigned error value does not vary with gamma-ray energy in the manner of ordinary measurements, since it arises primarily from partially compensating gain and base-line shifts with respect to conditions at the time of calibration. The energy values quoted in this report may differ slightly from, but are in excellent agreement with, those reported in BG; however, in order to provide a uniform energy scale for the additional states, not identified in BG, the values of the present experiment have been uniformly used in this article. No confusion should exist on the identification of a particular level in the two reports, however, because the differences in the quoted values are small compared to the separation of neighboring states.

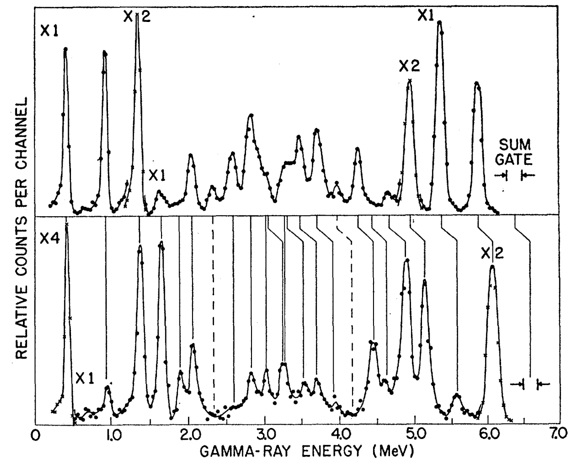

FIG. 5. Sum-coincidence spectra obtained at the 1588-keV (upper) and the 1770-keV (lower) resonances, plotted to the same energy scale. These spectra illustrate the shift in the energy of the primary gamma rays corresponding to the difference in the excitation energy of the capturing levels, and hence serve to identify the secondary gamma rays and the corresponding intermediate excited states. The data points represented by ×'s indicate that the scale should be multiplied by the listed factor for those peaks.

The identification of the energy of an intermediate state involved in a two-part cascade of course requires the establishment of which gamma ray is the primary transition populating the level and which is the secondary transition de-exciting the level. Such an identification is possible in the $(p,\gamma\gamma)$ reactions, where capturing resonances at different excitation energies may be studied. Thus, as observed from resonances sufficiently separated in excitation energy, and for two-part cascades through the same intermediate state: the energy of the secondary transition must remain constant, as it represents the energy difference between the same intermediate level and the ground state in each case; and the energy of the primary transition must change by the same amount as the energy difference in excitation of the capturing levels. This effect is illustrated in Fig. 5 where sum-coincidence spectra obtained at the 1588- and 1770-keV resonances are displayed to the same energy scale. The sum gate for the latter spectrum is displaced by an amount equal to the difference in excitation energy of the two resonances as indicated by the bent vertical line. Thus, secondary transitions are identified by straight vertical lines; while primary transitions are identified by the bent vertical lines.

From Fig. 5, it is quite clear that states exist at energies of 0.47, 0.95, 1.37, 1.63, and 2.07 MeV. The existence of a state at 1.89 MeV, discernible at these resonances, is more clearly seen at the 1620-keV resonance. The two members of a cascade observed at about 2.4 and 4.2 MeV in the 1588-keV resonance spectrum were not observed at any of the other resonances

[20] C. R. Gossett and C. M. Davisson, Bull. Am. Phys. Soc. 7, 9 (1962); C. R. Gossett and C. M. Davisson, U. S. Naval Research Laboratory Quarterly Report on Nuclear Science and Technology, 1961, p. 38 (unpublished). Values for 3-in.-diam×3-in. and 5-in.-diam×6-in. NaI crystals are available from the authors.

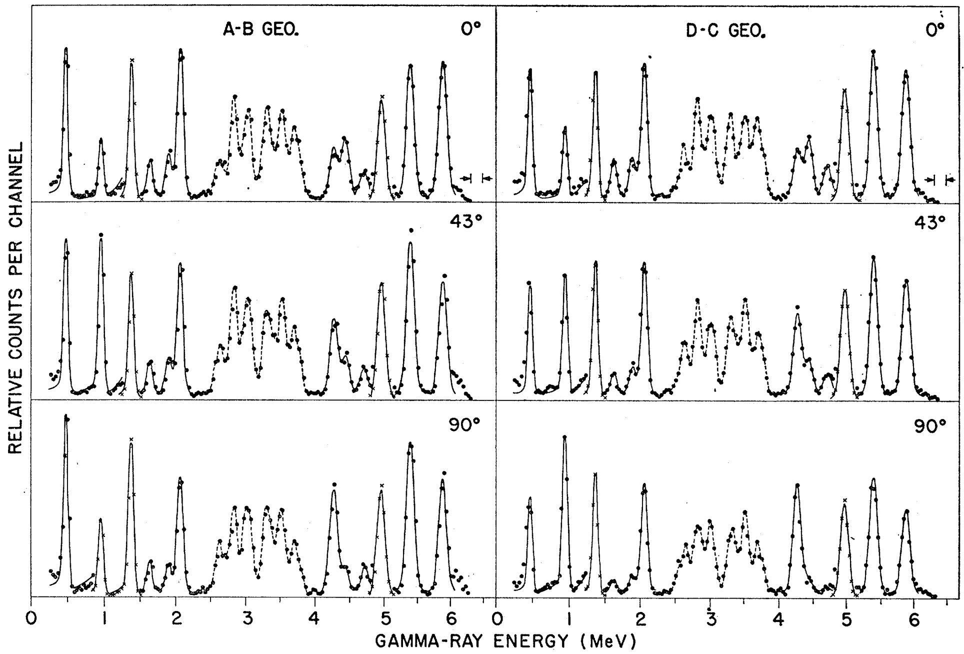

FIG. 6. Sum-coincidence spectra obtained at the 1620-keV resonance for the two sets of geometries simultaneously measured at each of three angles by the double sum-coincidence arrangement. The data have been normalized to equal reaction yield, but have not been corrected for the effects of unequal absorption of the gamma rays at the different angular positions of the detector. The solid curves are the sums of the calculated Gaussian fits made to the peaks and an estimation of the distortion and background effects. The dashed curves merely connect the points for purposes of clarity. The data points represented by ×'s (principally for the peaks at 1.37 and 4.97 MeV) indicate that the scale should be multiplied by a factor of 3 for those peaks.

studied; therefore, identification of the primary and secondary members of the cascade is impossible. The vertical lines for these transitions are shown dashed to indicate the possible existence of a state corresponding to the lower of these energies since all transitions observed below this energy proved to be secondary transitions. The three peaks, corresponding to transitions involving states at 2.64, 2.83, and 3.02 MeV, appear immediately to each side of the median energy (half of the excitation energy of the capturing level) at the 1599- and the 1620-keV resonances. These peaks are not so simple as might be supposed, for analysis indicates that at least those corresponding to states at 2.64 and 3.02 MeV are too wide for single peaks and that the structure in this region is surely complex. Further evidence of this is the apparent filling of the valley between the peaks corresponding to states at 2.83 and 3.02 MeV in the 1588-keV resonance spectrum, and the appearance of a closely spaced doublet at the median energy in the 1770-keV spectrum. The latter peaks correspond to a state which would produce transitions unresolved from those for the 3.02-MeV state when observed at the lower energy resonances due to the inversion of the positions of the primary and secondary transitions. The existence of transitions unresolved from those corresponding to the state at 2.64 MeV is also clear although it has not been possible to determine further information on these transitions.

The congestion of peaks which appears to be occurring near the median energy is perhaps not surprising since both an increase in level density and an inversion of the position of the primary and secondary transitions occur near this energy. It is quite likely that at least some of the weaker unresolved transitions noted above are due to states above the median energy for which such an inversion occurs. It is of course expected that states at higher excitations will not generally be as strongly excited by two-part cascade as lower energy states, both because of the previously mentioned tendency for multiple cascades and because of the strong energy dependence in the transition probability for gamma emission. These factors probably account for the apparent absence of any inversion in the transitions observed in regions other than those near the median energy.

CORRELATION RESULTS

Chronologically, correlations have been run at the three resonances in the order 1599, 1588, and 1620 keV. The strong 1599-keV resonance produced adequate statistics for spin assignment only for the three strongest two-part cascades observed, those involving the first three excited states. The weaker 1588-keV resonance was then run with poorer statistics as a confirmation of the unusual results obtained for the 1.37-MeV state from the 1599-keV resonance data. The 1620-keV resonance, which was chosen because it provided the most intense two-part cascade through the 1.63-MeV state of any of the usable resonances available, was subsequently given a long run in an attempt to provide adequate statistics for assignments to this and other weakly excited states. With these results, it has been possible to make assignments to this state and to the incompletely resolved pair of states at 1.89 and 2.07 MeV in addition to the previously determined first three excited states. No attempt has been made to obtain assignments for any higher excited state contained within the complex structure near the median energy, although some of these peaks provide adequate statistics, because even those peaks which appear to be well resolved almost surely contain contributions in unknown quantities from other transitions which may have quite different correlation functions. It would thus appear that with the resolution restrictions imposed by NaI detectors, this complexity of structure, which appears here near the median energy, may prove to be the limiting factor on the height in excitation energy of states for which spin assignments may be expected with the techniques described in this report.

The sum-coincidence spectra obtained at the 1620-keV resonance are shown in Fig. 6 for the three angles in each of the two geometry sets. The results shown are appropriate sums from the 14-run sequences obtained for each of the 11 days of correlation measurement; the quality of the resultant spectra indicates the effectiveness of the means taken to achieve stability and reproducibility over such an extended period. The peaks at 0.47 MeV in the D-C geometry spectra and those at 2.07 MeV in all spectra are due to transitions for which the correlations are believed to be isotropic; thus, the magnitude of the correction factor for the unequal absorption of the gamma rays in the anisotropic distribution of materials in the vicinity of the target may be inferred from the degree of anisotropy observed for these peaks. In addition to these instrumental effects, it may be noted that rather large but opposite anisotropies are present in peaks at 0.95 MeV in the A-B geometry and at 1.63 MeV in the D-C geometry spectra, and also in the incompletely resolved pair at 4.28 and 4.44 MeV in all spectra.

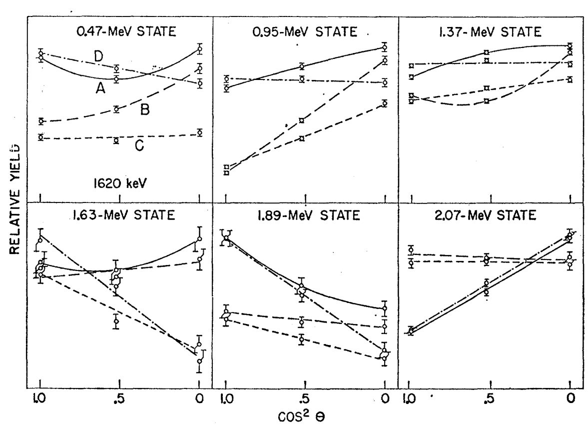

FIG. 7. Corrected and normalized yield versus $\cos^2\theta$ for the 24 correlation functions simultaneously measured at the 1620-keV resonance. The correlations are grouped according to the indicated intermediate excited state, and the four geometries for each state are designated by the pattern of the curves (key shown for first group in upper left hand corner). The points are the sum of all data appropriate to that angle and geometry for each state, and the associated error bars include both purely statistical effects and an estimation of the error introduced in the correction for background, distortions, and incompletely-resolved groups. The curves are calculated from the results of the least-squares fits to the normalized Legendre expansion for either the first two or first three terms, as appropriate to the theoretical predictions in agreement with the results for that state. The values of the coefficients of the normalized Legendre expansion obtained from the least-squares fit are listed in Table II. The correlations for the A and B geometries for the 0.47-MeV state are instrumentally distorted.

The corrected and normalized yields for the 24 correlations measured at the 1620-keV resonance are shown in Fig. 7. The points shown are sums of the individual groups of runs used in the least-squares analysis, and the curves are calculated from the results of that fit for each correlation (see Data Analysis). The results of the fits for the coefficients of the Legendre expansion are listed in Table II. Both the curves and the A_2 listings are for fits of two or three terms in the Legendre expansion, as required by the predicted correlation functions which specify those geometries for which the A_4 must vanish. The curves reflect the vanishing A_4 re-

TABLE II. Results of the least-squares fit to the normalized Legendre expansion for the 24 triple correlations measured simultaneously at the 1620-keV resonance. The correlations are identified as to the intermediate state and geometry, and the coefficients of the expansion, A_2 and A_4, are listed. The value of A_2 is that determined from either a two- or three-term fit as appropriate to the theoretical prediction for the assignment ultimately made to the state. Values of A_2 or A_4 enclosed in parentheses are for quantities required to vanish by these predictions, and are included to indicate the extent to which this requirement is met. Values enclosed in brackets are instrumentally distorted.

State (MeV)	Geometry	A_2	A_4
0.47	A	[−0.043±0.044]	[(0.170±0.036)]
	B	[(−0.314±0.036)]	[(0.106±0.036)]
	C	(−0.048±0.036)	(0.074±0.037)
	D	0.153±0.036	(0.007±0.036)
0.95	A	−0.184±0.028	−0.016±0.026
	B	−0.720±0.028	0.009±0.027
	C	−0.543±0.035	(0.022±0.033)
	D	0.020±0.031	(−0.012±0.029)
1.37	A	−0.112±0.018	−0.058±0.017
	B	−0.318±0.020	0.195±0.018
	C	−0.124±0.020	(−0.021±0.019)
	D	−0.006±0.019	(−0.030±0.018)
1.63	A	−0.17 ±0.09	0.13 ±0.09
	B	−0.09 ±0.08	(0.07 ±0.08)
	C	0.79 ±0.18	(0.25 ±0.15)
	D	1.12 ±0.21	(−0.14 ±0.15)
1.89	A	0.43 ±0.09	0.14 ±0.08
	B	0.15 ±0.11	(0.05 ±0.10)
	C	0.62 ±0.19	(0.04 ±0.15)
	D	0.99 ±0.13	(0.05 ±0.10)
2.07	A	−0.526±0.037	(0.044±0.036)
	B	(0.033±0.038)	(0.060±0.036)
	C	(0.008±0.038)	(−0.045±0.036)
	D	−0.517±0.036	(−0.012±0.036)

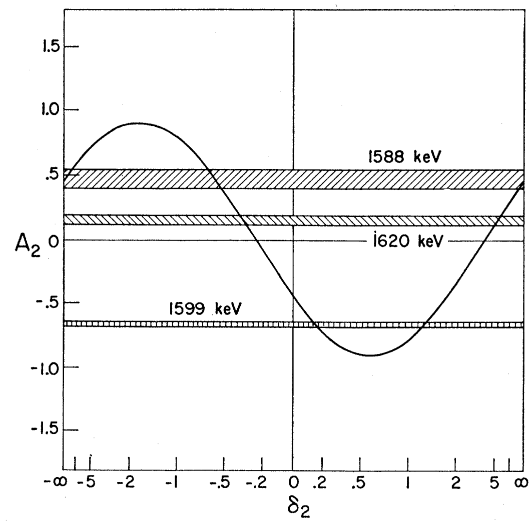

FIG. 8. The theoretical predictions for a $\frac{1}{2}$ assignment to the first excited state at 0.47 MeV, where the coefficient of the normalized Legendre expansion, A_2, is shown as a function of the quadrupole amplitude ratio, δ_2, for the primary transitions (A and D geometries). The sign convention for the admixture ratio is that of Devons and Goldfarb (see Ref. 16). This prediction requires isotropy for the secondary transitions (B and C geometries). The range of the experimental values for A_2 at each of the indicated resonances is shown by the crosshatched band. The resulting predictions for the primary quadrupole admixture are listed in Table IV.

quirement in such cases by appearing as straight lines in the plot against $\cos^2\theta$; while enclosure in parentheses of entries for A_4 in Table II indicates those values required to vanish, the values being included to indicate the extent to which this requirement is met. The occurrence of a number of instances of exceptions to the vanishing requirement by amounts greater than the listed errors is probably due to systematic error, either that associated with the beam alignment problem, or in the case of the weaker transitions, that associated with the background subtraction. However, it will be noted that large values of the A_4 coefficient do not generally occur, even where they are allowed, with the notable exception of the B geometry for the 1.37-MeV state, which will be discussed below in detail.

The predictions for a $\frac{1}{2}$-unit spin intermediate state are isotropy for the secondary transitions in the B and C geometries, and identical anisotropies for the primary transitions in the A and D geometries. These predictions for the 0.47-MeV state are met at the 1599-keV resonance, as will be shown; but they clearly are not met by the A and B geometries at the 1620-keV resonance. This effect is due to cross-talk between detectors (see Sum-Coincidence Method). The effect does not significantly appear in the C and D geometries, where the two detectors are always at 90° from each other (and thus present no unshielded path for the transfer of an annihilation quantum from one detector to the other) or for the $\theta=0°$ data for the A and B geometries, where the relevant detectors are again at 90° to each other; but it does occur for the A and B geometries at $\theta=43°$ and to an even greater extent at $\theta=90°$, as the relative opening between the detectors presented by the collimators increases to its maximum at $\theta=90°$, where the two detectors in the horizontal plane are directly opposite each other.

The collimators used in the present experiment are apparently sufficiently effective that such a cross-talk problem exists only with relation to the total capture peak for the ground state transitions from the capturing level. This is so because transfer of an annihilation quantum may occur for events which should be in this peak and which meet the sum condition but which require only a single detector-efficiency term (the fast-coincidence condition being supplied by the transfer event itself) rather than the two detector-efficiency terms normally required in sum coincidence. The satellite peaks associated with this particular transfer event mimic a state at the energy of the annihilation quantum and provide difficulty in the present experiment because such events are experimentally indistinguishable[21] from true two-part cascades involving the first excited state at 0.47 MeV. Even so, the effect is not appreciable unless the first excited state is rather weakly excited in the branching of the capturing level, as occurs for the 1588- and 1620-keV resonances.

That the transitions associated with the 0.47-MeV state meet the requirements (stated above) for a $\frac{1}{2}$-unit spin assignment will be shown in connection with a discussion of the results for the 1599-keV resonance. However, it is quite clear that the deviations from these

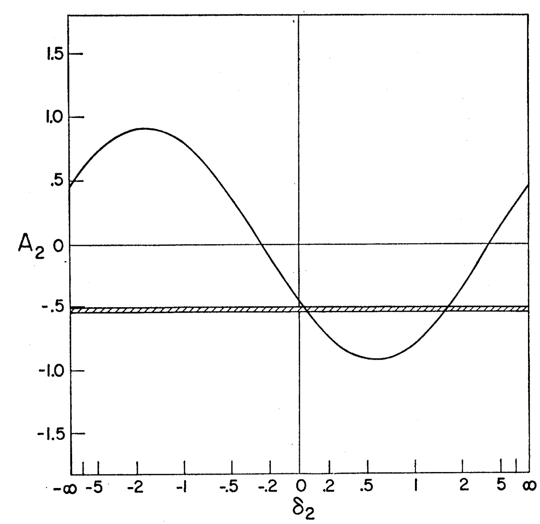

FIG. 9. Theoretical predictions for a $\frac{1}{2}$ assignment to the 2.07-MeV state as observed at the 1620-keV resonance only. See Fig. 8 for details of the representation. The A_2 scale for this figure differs very slightly from that of Fig. 8, because of the effect of the finite-geometry-correction coefficients for the different energies of the transitions involved for the two states.

[21] This is true of individual events; however, there is a noticeable trend for distortion of the 0.47-MeV peak toward higher energy in the $\theta=43°$ and $\theta=90°$ spectra for the A-B geometry in Fig. 6.

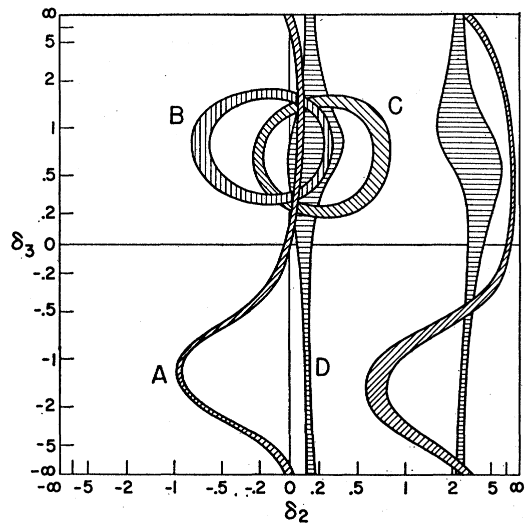

FIG. 10. Contour plot for a $\frac{5}{2}$ assignment to the 0.95-MeV state as observed at the 1620-keV resonance, where the experimental results are shown as a function of the secondary and primary quadrupole admixture ratios, δ_3 and δ_2, respectively. Here the range of values of the admixture ratios allowed by the range of the experimental values of the A_2 coefficient of the normalized Legendre expansion is indicated by different crosshatching for each geometry (keyed in this figure only). The admixture ratio axes are in an arctan representation to allow display of the complete range of the variable. The sign convention for the admixture ratio is that of Devons and Goldfarb (see Ref. 16). Agreement of experiment and the predicted correlation functions is indicated by overlap of all four bands, and the range of the resulting allowed value(s) of the admixture ratios are listed in Table IV.

predictions, observed in the A and B geometries at both the 1620- and 1588-keV resonances, and attributed to instrumental effects, do not contradict the $\frac{1}{2}$ assignment. Thus, whether the isotropy observed in the secondary transitions (B and C geometries) at the 1599-keV resonance is due to a $\frac{1}{2}$ spin assignment or to a higher spin assignment, this isotropy should be observed for these two geometries at *any* other resonance. This is true because even for higher spin assignments the isotropy is due to the particular quadrupole admixture for the secondary transition, and this admixture is the property of the intermediate and final states only and does not depend upon the particular capturing resonance from which the state is excited.

It is an unfortunate consequence of angular correlation theory that degeneracies are associated with low-valued spin states. One such degeneracy is the impossibility by means of angular correlations alone, no matter how many geometries are measured, of differentiating uniquely between a $\frac{1}{2}$-unit spin state and higher spin possibilities because higher spin assignments can always be made to show agreement with the data for some particular values of the admixture ratios. Nevertheless, the $\frac{1}{2}$ assignment is the most probable, when the conditions required by this assignment are met by the data, as this assignment does not require the accidental presence of a particular admixture ratio to fit the data. Another consequence of the degeneracies associated with the $\frac{1}{2}$ assignment is that, although the possibility of quadrupole admixture exists in both transitions, the admixture of the secondary transition is completely indeterminate. Therefore, the comparison of theory and experiment resolves to the representation of Fig. 8, where the predictions for the A_2 of the primary transitions (A and D geometries) are given as a function of the primary admixture only. As a consequence of a further degeneracy associated with the $\frac{1}{2}$ assignment, it is not possible to distinguish between the two predicted values of the primary admixture purely on the basis of the correlation measurements alone.

The results for the 0.47-MeV state from all three resonances at which correlations were run are shown in Fig. 8, the ranges of the experimentally observed values of A_2 being represented by the cross-hatched horizontal bands. The experimental values used are from only the undistorted C and D geometries in those cases where distortions are evident in the A and B geometries. The 2.07-MeV state, for which correlations are observed only at the 1620-keV resonance, produces results which are also consistent with the predictions for a $\frac{1}{2}$ assignment. The prediction for this state is shown in Fig. 9, where the energy dependence of the finite-geometry-correction coefficients provides a very slight difference in scale from the predictions for the 0.47-MeV state.

The 0.95-MeV state, which shows large anisotropies in the B and C geometries, is consistent only with a $\frac{5}{2}$ assignment for the spin of the intermediate state. Here admixtures in both the primary and secondary transitions are allowed, and the theoretical predictions corresponding to the experimental values of A_2 obtained

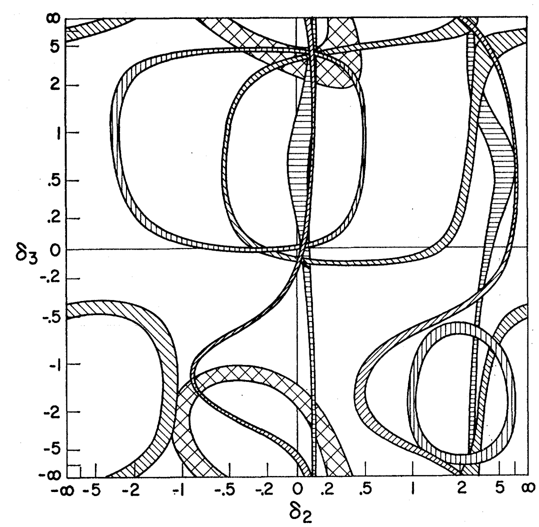

FIG. 11. Contour plot for a $\frac{5}{2}$ assignment for the 1.37-MeV state as observed at the 1620-keV resonance. See Fig. 10 for details of the representation. Here an additional band shown by double crosshatching represents the results for the significant A_4 coefficient for the B geometry. Note that the only possible overlap of all five bands occurs at an unusually high value of the secondary admixture ratio.

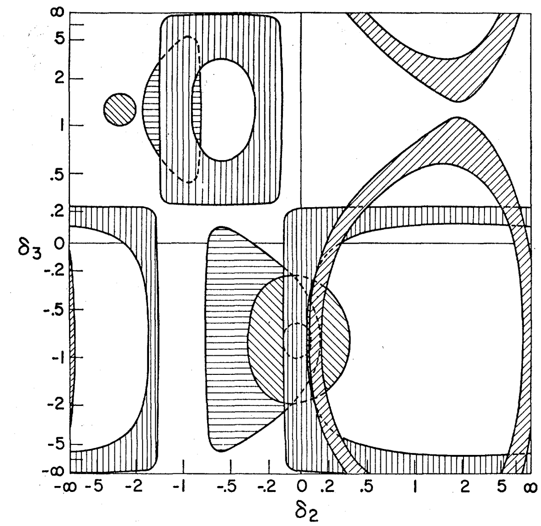

FIG. 12. Contour plot for a possible $\frac{3}{2}$ assignment to the 1.63-MeV state as observed at the 1620-keV resonance. See Fig. 10 for details of the representation.

at the 1620-keV resonance are shown in Fig. 10 in a contour plot representation. In this instance the overlap of the bands is not ideal, and it is also impossible to distinguish which of the two possible regions is appropriate. However, the results at the 1599- and 1588-keV resonances show that only that associated with the lower-valued possibility of the secondary admixture ratio is proper.

The 1.37-MeV state is also found to be consistent only with a $\frac{5}{2}$ assignment, but it produces quite a different aspect, as shown in Fig. 11. Here, better statistics were available for the transitions involving this state, and this fact is reflected in the generally more narrow bands. It may be seen that the *only* possible overlap

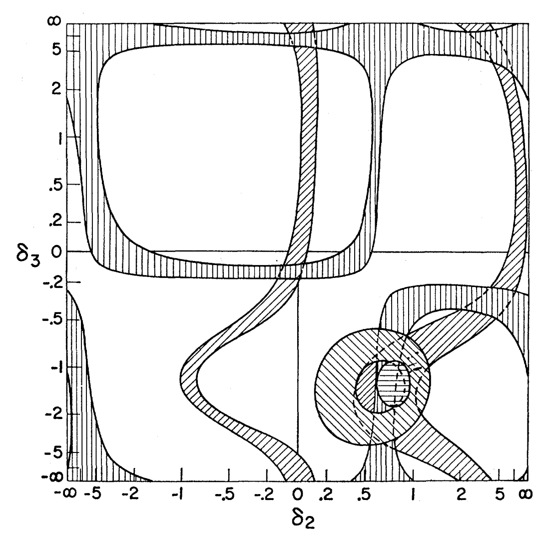

FIG. 13. Contour plot for a possible $\frac{5}{2}$ assignment to the 1.63-MeV state as observed at the 1620-keV resonance. See Fig. 10 for details of the representation.

occurs at a surprisingly high value of the secondary admixture ratio. Such a high quadrupole admixture is generally associated with the observation of a significant A_4 value; and indeed, as previously noted, such an observation was made in this case. An additional band with double crosshatching indicates the range predicted by this value, and it is seen that this band is also consistent with the observed overlap, this consistency providing conclusive evidence for the very large value of the secondary admixture ratio.

The very weak transitions through the 1.63-MeV intermediate state provide the only instance of ambiguity in the spin of the intermediate state which was observed from any of the correlations, other than those associated with the degeneracies for low-valued spin states. Probably because of the poor statistics and the large probability of systematic error in connection with

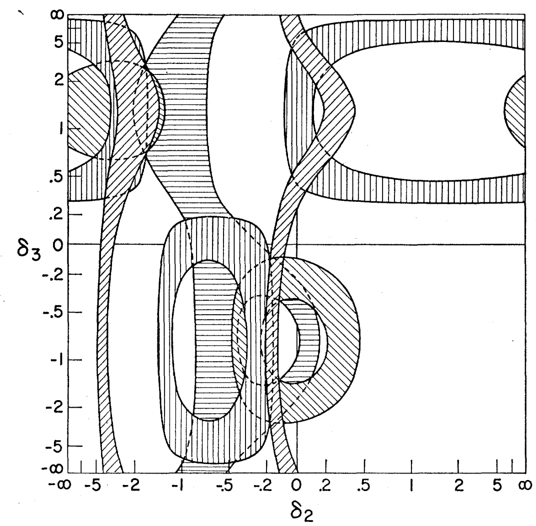

FIG. 14. Contour plot for the $\frac{3}{2}$ assignment to the 1.89-MeV state as observed at the 1620-keV resonance. See Fig. 10 for details of the representation. A possible overlap to the upper left is excluded by the observed A_4 coefficients (not shown).

the background subtraction, the overlap is poor for either a possible $\frac{3}{2}$ or $\frac{5}{2}$ assignment, as shown in Figs. 12 and 13; therefore, it is not possible to distinguish between these two possible assignments on the basis of this comparison with theory alone. There is a tendency to favor the $\frac{3}{2}$ assignment because of the much stronger resemblance to the proved $\frac{3}{2}$ case for the 1.89-MeV state, rather than to those observed for states to which $\frac{5}{2}$ assignments were made (see Fig. 7); but it would probably be inappropriate to make an assignment on this basis due to the present paucity of such examples.

Although the 1.89-MeV state also involved rather poor statistics, and additionally the problem of incomplete resolution from the somewhat stronger transitions associated with the 2.07-MeV state, it does provide a clear exclusion of the $\frac{5}{2}$ possibility and a somewhat better overlap for the allowed bands in the $\frac{3}{2}$ prediction as shown in Fig. 14. The case of a $\frac{3}{2}$ assignment also provides a degeneracy, in this case only in the secondary admixture ratio, this ambiguity only recently having been reported.[22] This ambiguity is evidenced by the symmetry in δ_3, observed in Figs. 12 and 14, which is not present in the predictions for the $\frac{5}{2}$ assignment.

The corrected and normalized yields and the fitted correlation functions are shown in Fig. 15 for the first three excited states, as observed at the 1599- and 1588-keV resonances. The results of the least-squares fits for the coefficients of the correlation functions are listed in Table III. The previously mentioned nondistorted agreement with theory of the correlations associated with the 0.47-MeV state at the 1599-keV resonance may be noted. The comparison with theory for this state at these two resonances has been shown in Fig. 8. The theoretical prediction for the 0.95-MeV state is shown only for the 1599-keV resonance in Fig. 16, where the exclusion of the higher secondary-admixture-

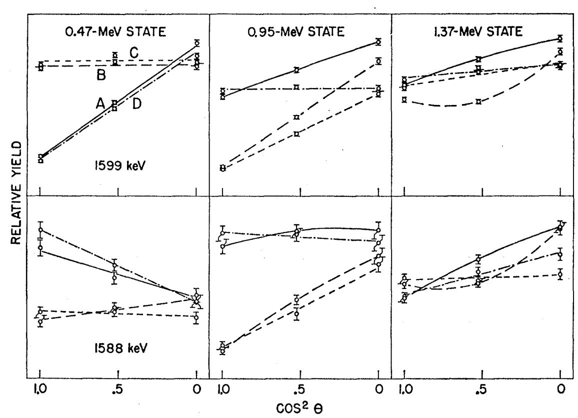

FIG. 15. Corrected and normalized yield versus $\cos^2\theta$ for the indicated excited states as observed at both the 1599- and 1588-keV resonances. See Fig. 7 for details of the representation.

ratio possibility observed at the 1620-keV resonance is apparent. For the 1.37-MeV state, large A_4 terms are again observed in the B geometry at both resonances in Fig. 15. The theoretical predictions for both resonances, shown in Figs. 17 and 18, again indicate the only possible overlap of all five bands occurs at the same large secondary admixture ratio. The poorer statistics obtained at the 1588-keV resonance are evident both in the error bars of Fig. 15 and in the wider allowed bands of Fig. 18.

The spin and parity assignments for the intermediate state, as determined by the predicted correlation functions at each of the resonances, are summarized in Table IV along with the quadrupole-to-dipole amplitude ratios, as determined for the primary and secondary transitions. The error estimates for the latter are based upon the width of the allowed bands at the point of overlap and some consideration of the quality of the

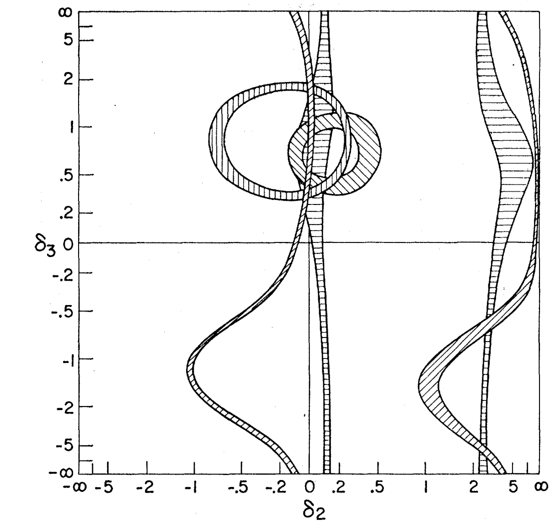

FIG. 16. Contour plot for the $\frac{5}{2}$ assignment to the 0.96-MeV state as observed at the 1599-keV resonance. See Fig. 10 for details of the representation.

overlap. Several factors may be noted: First, the assignment of the spin of the capturing level ($\frac{3}{2}$ in each case) determined for each cascade for which correlations are run at a given resonance must and do agree. Further, these assignments are in agreement with those made on the basis of the angular distribution of the ground state transtions reported in BG. Second, the assignment of spin to an intermediate state made at any one resonance must and does agree with the assignments made to the same state at any other resonance for which correlations for that state were made. Third, the quadrupole admixture ratio for only the secondary transition for a given state must and does agree with those for that

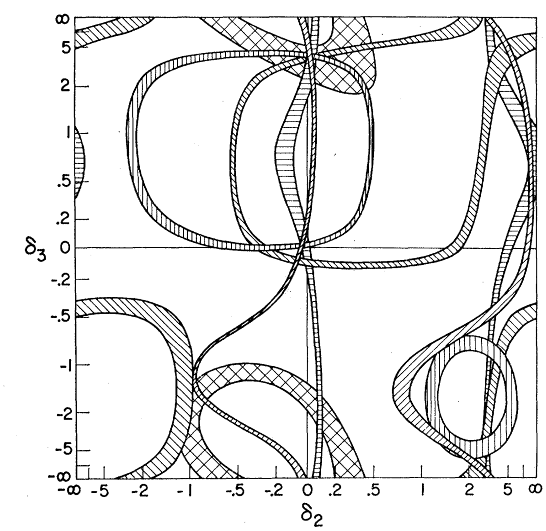

FIG. 17. Contour plot for the $\frac{5}{2}$ assignment to the 1.37-MeV state as observed at the 1599-keV resonance. See Figs. 10 and 11 for details of the representation.

[22] G. I. Harris and L. W. Seagondollar, Phys. Rev. **131**, 787 (1963); H. Van Rinsvelt and P. B. Smith, Physica **30**, 59 (1964).

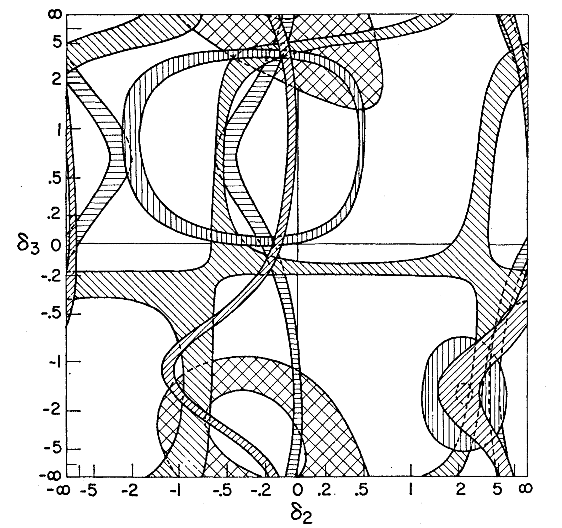

FIG. 18. Contour plot for the $\frac{5}{2}$ assignment to the 1.37-MeV state as observed at the weak 1588-keV resonance. See Figs. 10 and 11 for details of the representation.

state obtained at all other resonances because this property does not depend upon the capturing resonance. The quadrupole admixtures for the primary transitions, on the other hand, may and do show different values as determined at different resonances, even though the resonances are of the same spin and parity because this property does depend on the emitting state which is of course different at each resonance.

TABLE III. Results of the least-squares fit to the normalized Legendre expansion for the 12 triple correlations measured simultaneously at each of the resonances at 1599 and at 1588 keV. See Table II for details.

Resonance (keV)	State (MeV)	Geometry	A_2	A_4
1599	0.47	A	-0.661 ± 0.021	(0.047 ± 0.021)
		B	(-0.010 ± 0.021)	(-0.040 ± 0.021)
		C	(-0.007 ± 0.021)	(-0.057 ± 0.031)
		D	-0.657 ± 0.021	(-0.071 ± 0.021)
	0.95	A	-0.267 ± 0.023	-0.008 ± 0.021
		B	-0.711 ± 0.024	0.002 ± 0.017
		C	-0.651 ± 0.028	(0.013 ± 0.028)
		D	-0.016 ± 0.026	(-0.024 ± 0.029)
	1.37	A	-0.205 ± 0.022	-0.025 ± 0.020
		B	-0.360 ± 0.024	0.194 ± 0.022
		C	-0.128 ± 0.021	(-0.032 ± 0.023)
		D	-0.073 ± 0.023	(-0.038 ± 0.022)
1588	0.47	A	$[0.32\ \pm0.08]$	$[(0.07\ \pm0.09)]$
		B	$[(-0.19\ \pm0.08)]$	$[(-0.06\ \pm0.08)]$
		C	$(0.05\ \pm0.09)$	$(-0.05\ \pm0.08)$
		D	$0.47\ \pm0.08$	$(0.01\ \pm0.08)$
	0.95	A	$-0.05\ \pm0.05$	$-0.05\ \pm0.05$
		B	$-0.65\ \pm0.07$	$-0.06\ \pm0.06$
		C	$-0.63\ \pm0.07$	$(0.08\ \pm0.07)$
		D	$0.05\ \pm0.06$	$(-0.02\ \pm0.06)$
	1.37	A	-0.350 ± 0.040	-0.035 ± 0.039
		B	-0.396 ± 0.044	0.196 ± 0.043
		C	-0.028 ± 0.050	(0.036 ± 0.050)
		D	-0.257 ± 0.046	(-0.043 ± 0.044)

TABLE IV. Summary of the assignments from the present experiment for the spin and parity, J^π, of the intermediate state and the quadrupole admixture amplitude ratios, δ_2 and δ_3, for the primary and secondary members, respectively, of the two-part cascade from the capturing state through the intermediate state to the ground state. The values listed are identified as to the intermediate state and the resonance for which the triple correlation was observed. The spin assignments are all made on the basis of $\frac{3}{2}$ for the spin of both the capturing level (resonance) and for the ground state of ^{61}Cu (see Ref. 17). The three resonances are additionally assigned probable negative parity. Where assignments for the secondary admixture ratio were made at more than one resonance the last value listed is a weighted mean of the other values. The phase convention for δ is that of Devons and Goldfarb (see Ref. 16).

State (MeV)	Resonance (keV)	J^π	δ_2	δ_3
0.47	1588	$(\frac{1}{2}^-)$[a]	-0.60 ± 0.07	
			or	
			<-16 or >26	
	1599	$(\frac{1}{2}^-)$	0.15 ± 0.02	
			or	
			1.25 ± 0.05	
	1620	$(\frac{1}{2}^-)$	-0.35 ± 0.02	
			or	
			$5.8\ \pm0.8$	
0.95	1588	$\frac{5}{2}^-$	0.13 ± 0.06	$0.34^{+0.17}_{-0.12}$
	1599	$\frac{5}{2}^-$	-0.01 ± 0.03	0.39 ± 0.05
	1620	$\frac{5}{2}^-$	0.03 ± 0.04	0.32 ± 0.04
				0.35 ± 0.03
1.37	1588	$\frac{5}{2}^-$	-0.12 ± 0.06	$3.9^{+1.2}_{-0.8}$
	1599	$\frac{5}{2}^-$	0.04 ± 0.03	$3.63^{+0.39}_{-0.33}$
	1620	$\frac{5}{2}^-$	0.12 ± 0.02	$3.92^{+0.55}_{-0.43}$
				$3.78^{+0.15}_{-0.13}$
1.63	1620	$(\frac{3}{2}^-)$[b]	0.03 to 0.16[c]	-0.29 to -1.68[c,d]
		or		
		$(\frac{5}{2}^-)$	0.56 to 1.05[c]	-0.61 to -3.01[c]
1.89	1620	$\frac{3}{2}^-$	-0.12 to -0.26[c]	-0.08 to -0.42[c,d]
				or
				-1.19 to -2.48[c,d]
2.07	1620	$(\frac{1}{2}^-)$[a]	0.05 ± 0.03	
			or	
			1.54 ± 0.09	

[a] Degeneracies associated with the $\frac{1}{2}$ assignment prevent a unique spin assignment, introduce an ambiguity in δ_2, and make δ_3 completely indeterminate.
[b] The 1.63-MeV state was so weakly excited that it was impossible to distinguish on the basis of the correlations alone between a $\frac{3}{2}^-$ and a $\frac{5}{2}^-$ assignment.
[c] The 1.63- and 1.89-MeV states were so weakly excited that the δ's are better represented as a probable range of values rather than a value with associated error.
[d] Degeneracies associated with the $\frac{3}{2}$ assignment introduce an ambiguity in δ_3; however, for the 1.63-MeV state the two regions are sufficiently indistinct that they are represented by a single range.

Angular-correlation measurements do not in principle determine parity, which may be *measured* only by a polarization experiment for gamma-ray transitions and inferred from certain other measurements. However, *probable* negative parity has been proposed for each of the intermediate states for which spin has been assigned in this report (except the $\frac{1}{2}$-unit spin states), on the basis of the observation of significant quadrupole admixtures in the secondary transitions as indicated in Table IV. As the secondary quadrupole admixture for the $\frac{1}{2}$-unit spin states is indeterminate in the correlation measurements, negative parity cannot be assigned to them on this basis. The assignment of negative parity to the three capturing states is perhaps on less certain

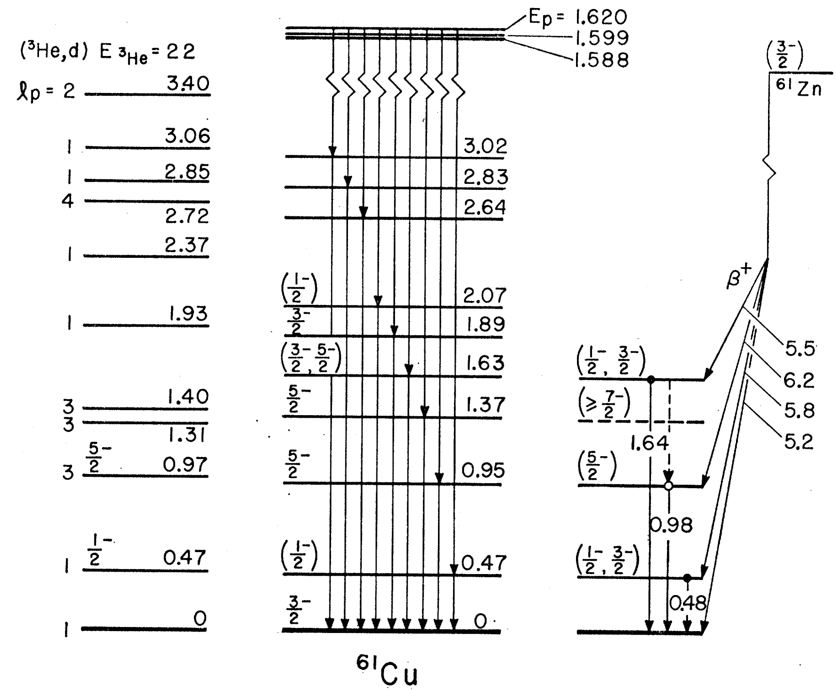

FIG. 19. A level diagram for the nuclide, ^{61}Cu, showing the results of the present experiment in the center. Here, only two-part cascades are shown as the experimental arrangement preferentially selects these. Possible limitations on the spins of some of these states as inferred from the results of Cumming (see Ref. 7) in the decay of ^{61}Zn are shown on the right with the logft values indicated for the β^+ transitions. Recent results by Blair (see Ref. 8) for the (^{3}He,d) reaction are shown on the left with the l values determined from the proton stripping. His spin assignments for the first two excited states are based upon sum-rule considerations. The assignments for the excitation energies of the low-lying excited states are well within experimental errors for all experiments and the agreement for the spin assignments is good. The implications of the doublet at about 1.35 MeV reported by Blair are discussed in the text.

grounds as the quadrupole admixtures for the primary transitions are not so large as those observed for the secondary transitions. However, penetrability arguments (see BG) favor the $\frac{3}{2}^-$ assignment of p-wave capture to the $\frac{3}{2}^+$ of d-wave capture. It is then possible on the basis of the observation of significant admixtures for the primary transitions to the $\frac{1}{2}$-unit spin states to predict *probable* negative parity for them.

DISCUSSION

The conclusions of this report with regard to the positions of levels and their spin and probable parity assignments are summarized in the level diagram of Fig. 19, along with the results of Blair[8] and Cumming.[7] There would appear to be agreement for the assignments to the first two excited states at 0.47 and 0.95 MeV. These spin assignments are also in agreement with assignments to presumably equivalent states in ^{63}Cu and ^{65}Cu, which have received considerable experimental attention. The quadrupole admixture for the 0.95-MeV state observed in the present experiment is in generally the same range of values as those observed for these states in ^{63}Cu (0.961 MeV) and ^{65}Cu (1.115 MeV), although there are some disagreements between different experiments for the same values as shown in Table V. This discrepancy is particularly apparent for the recent resonance fluorescence result[23] compared to those from Coulomb excitation.[24,25]

[23] G. B. Beard, Phys. Rev. **135**, B577 (1964).
[24] R. L. Robinson, F. K. McGowan, and P. H. Stelson, Phys. Rev. **134**, B567 (1964).
[25] B. Elbek, H. E. Gove, and B. Herskind (to be published).

On the other hand, the situation with regard to the third excited state is less clear. In ^{63}Cu and ^{65}Cu the third excited states are reported[24,25] to be $\frac{7}{2}^-$, this assignment being in agreement with the expectations of the core-excitation model. Indeed, this is one of the levels for which absolute lifetime measurements have been made relating to the arguments for this model. Because Cumming observed no evidence of β^+ transitions to the level reported by BG at 1.38 MeV in ^{61}Cu, it has been widely assumed (see Ref. 26) that this level has a J^π of $\frac{7}{2}^-$. The assignment of $\frac{5}{2}^-$ of the present report is of course in disagreement with this conclusion. That the theoretical predictions for a $\frac{7}{2}^-$ assignment to the 1.37-MeV state do not agree with the experimental observations may readily be seen by a

TABLE V. Comparison of the quadrupole-to-dipole amplitude ratios, $\delta=(E2/M1)^{\frac{1}{2}}$, observed for the $\frac{5}{2}^-$ second excited states of some of the odd-A isotopes of Cu.

^{61}Cu	^{63}Cu	^{65}Cu
$+0.35\pm0.03$[a]	$-0.41^{+0.07}_{-0.11}$[b]	-0.22 ± 0.06[b]
	-0.27 ± 0.08[c]	-0.30 ± 0.13[c]
	±0.40[e]	$-0.52^{+0.07}_{-0.05}$[d]

[a] Present result. The difference of the sign of this result from the other measurements is due merely to different conventions, Devons and Goldfarb for the present results and Biedenharn and Rose for the others.
[b] See Ref. 24.
[c] See Ref. 25.
[d] See Ref. 23.
[e] See Ref. 2. Inferred from total lifetime measurement and earlier Coulomb excitation measurement of $B(E2)$.

[26] *Nuclear Data Sheets*, compiled by K. Way *et al.* (Printing and Publishing Office, National Academy of Sciences—National Research Council, Washington, D. C., 1960), NRC 60-5-40, 41.

TABLE VI. The predicted correlation functions for a $\frac{7}{2}^-$ assignment to the 1.37-MeV state for comparison with those actually observed, as listed in Tables II and III. These predictions contain the effects of finite solid angle of the detectors. Because $M3$ transitions are not expected to compete in reaction induced transitions, the predictions for the $\frac{3}{2}^-(E2)\frac{7}{2}^-(E2)\frac{3}{2}^-$ cascade are not a function of multipole admixtures.

Geometry	A_2	A_4
A	−0.166	0.068
B	−0.009	0.145
C	0.410	0
D	0.206	0

comparison of these predictions, listed in Table VI, with the experimentally observed quantities, listed in Tables II and III. Blair's recent experiment offers an interesting possibility: he observes two $l=3$ states at 1.31 and 1.40 MeV. As the $l=3$ stripping leads to states of either $\frac{5}{2}^-$ or $\frac{7}{2}^-$, Blair's result is clearly not inconsistent with the $\frac{5}{2}^-$ assignment of the present report. Indeed, one is tempted to suggest that one member of Blair's doublet is the $\frac{5}{2}^-$ state, and that the other member is the missing $\frac{7}{2}^-$ state expected by analogy with ^{63}Cu and ^{65}Cu and required by the core-excitation model. It is not clear to which member of Blair's doublet the 1.37-MeV state of the present report corresponds, although the 1.40-MeV state may be slightly favored. However, as the $\frac{7}{2}^-$ state is not observed in the $(p,\gamma\gamma)$ reaction, definite evidence for such an assignment is lacking.

The nonobservation of transitions involving a $\frac{7}{2}^-$ intermediate state, which would require a pure $E2$ transition from a $\frac{3}{2}^-$ capturing level, is perhaps not surprising in the present experiment. Although the transition de-exciting the intermediate state (secondary transition) may show an enhancement of the $E2$ transition probability if the state is analogous to those $\frac{7}{2}^-$ states observed in ^{63}Cu and ^{65}Cu, there is no reason to expect such an enhancement for the $E2$ transition from the capturing level to the $\frac{7}{2}^-$ state (primary transition). However, the strengths of the branching of transitions from the capturing level through various intermediate states, and the resultant observation in the present experiment, depend upon the transition probabilities of these primary transitions. Thus the situation of the present experiment is not at all comparable to that of the Coulomb excitation experiments in which the $E2$ transition probability between the $\frac{7}{2}^-$ state and the ground state is controlling.

Although no measurements had been made to determine the spin of the fourth (1.412-MeV) excited state of ^{63}Cu, a value of $\frac{3}{2}^-$ had been assumed[1,3] for this state on the basis of the center of gravity rule. However, a recently published experiment by Blair[27] indicates $l=3$ stripping for this state in disagreement with this assumption. Blair suggests that his observations favor the unified-model calculations of Bouten and Van Leuven[28] which predict $\frac{5}{2}^-$ for this state and allow the greater single particle strengths which Blair states his stripping measurements indicate. If the fourth excited state of ^{63}Cu is $\frac{5}{2}^-$, the situation in that nuclide is perhaps similar (but it is possible that the positions of the levels may be inverted) to that possibly existing in ^{61}Cu (as suggested above). (The small shifts in level positions, which would be required to invert the positions of the levels from the positions observed in ^{63}Cu, are not inconsistent with changes which may occur with the different neutron populations of these isotopes.)

Blair's observation of a doublet in ^{61}Cu, however, may raise the question of whether the presence of an undetected doublet could not so distort the results of the present experiment as to lead to an erroneous $\frac{5}{2}^-$ assignment with the unusually high secondary quadrupole admixture observed. That this is not the case is indicated by three arguments: First, the full width at half-maximum (FWHM) is 89±6 keV for the peak at 1.37 MeV in the 1620-keV spectra, and this value is in good agreement with that observed for neighboring peaks. Further, as may be seen from the various spectra, the high-energy side of the Gaussian distribution for the 1.37-MeV peak is quite clear of evidence for an unresolved weaker transition; although the low-energy side of this peak is not so clear due to the presence of distortions (see Data Analysis). While it is difficult to make a quantitative estimate of the amount that could be present in the 1.37-MeV peak of a transition at either a 90-keV higher or lower energy, it seems unlikely that an amount sufficient to significantly distort the observed correlation could be present and remain undetected. Second, the transitions associated with the 1.37-MeV state are among the strongest at each resonance, and they produce excellent statistics. Thus, the evidence for the $\frac{5}{2}^-$ assignment to this state probably represents better agreement of the predictions and experiment than that obtained for any of the other states of this report. It therefore seems unlikely that a contribution from the other unrelated member of a doublet could accidentally produce such an excellent agreement with the predictions at any one resonance, let alone three different resonances. This is particularly evident, since the branching ratios for transitions to different states generally vary from resonance to resonance, and it would be highly unlikely that precisely the proper contribution be maintained at three different resonances. Third, the observation of a large A_4 term in *only* the B geometry is not what would be expected from the addition of unrelated angular correlations in an arbitrary manner. Thus, a distortion due to the effect of an undetected doublet would very likely also strongly affect the A_4 coefficients for the C and D geometries, whereas the theory requires that these coefficients vanish in any pure case. However, the values observed

[27] A. G. Blair, Phys. Letters **9**, 37 (1964).

[28] M. Bouten and P. Van Leuven, Nucl. Phys. **32**, 499 (1962).

for these geometries do not appear to deviate significantly more from the quoted errors than those observed for other states, and these values certainly are not comparable to that observed in the B geometry.

With regard to the remaining states at higher excitation energy: Blair does not report the observation of deuteron groups populating a state corresponding to that found at 1.63 MeV in the present report and in BG, and supported by the observations of Cumming. Of the two spin possibilities allowed by the present experiment for this state, the $\frac{3}{2}^-$ assignment may be preferred on the basis of Cumming's reported spin limitations. The state found by Blair at 1.93 MeV is in reasonable agreement with that assigned at 1.89 MeV in the present report, and the assignment of $l=1$ is in agreement with the $\frac{3}{2}^-$ assignment of this report. Blair also does not report a state corresponding to the $\frac{1}{2}^-$ state at 2.07 MeV from the present experiment. The $l=1$ state reported by Blair at 2.37 MeV does not correspond to a state populated in the $(p,\gamma\gamma)$ reaction unless possibly it be that state at near this energy observed only at the 1588-keV resonance. Above this energy the agreement of states is problematical, due to the increased density of states and the difficulties of the sum-coincidence method in this region (see Identification of States). It is in particular unlikely that the $l=4$ state reported by Blair at 2.72 MeV corresponds to a state that is observed near this energy in the present experiment, as it is not expected that a $\frac{7}{2}^+$ or $\frac{9}{2}^+$ level would be populated by a double cascade in the $(p,\gamma\gamma)$ reaction.

Thus, the agreement of the present experiment with other experimental evidence appears to be quite good, although the agreement with theory, or at least with some of the existing versions of the core-excitation model, would appear less satisfactory. The assignments to the first two excited states in the present experiment are of course consistent with that model; and the absence of the observation of a $\frac{7}{2}^-$ state may not be serious, in that the possibility that the unobserved member of the doublet reported by Blair could be such a state; however, these versions of the core-excitation model, at least as applied to the neighboring Cu isotopes, apparently do not explain the unusual character of the state which is here reported at 1.37 MeV for ^{61}Cu. Thus, if the state in ^{61}Cu at 0.95 MeV is the $\frac{5}{2}^-$ member of the core excitation quartet, as is to be expected if the apparently equivalent states in ^{63}Cu and ^{65}Cu are the $\frac{5}{2}^-$ members essential to the reduced $E2$ transition probability arguments now being used to support the model, then the 1.37-MeV state cannot be the $\frac{5}{2}^-$ member of this quartet. If the enhancement of the $E2$ transition probability, reflected in the unusually high quadrupole admixture, is indeed an index of collective effects, why then does a model which purports to represent the effects of collective motions in these nuclides not explain the existence of a level which apparently has stronger collective features than those states presumably explained by the model?

ACKNOWLEDGMENTS

The authors are pleased to acknowledge the assistance of Dr. P. A. Treado of Georgetown University and this Laboratory, for his participation in some of the data accumulation and analysis and in helpful discussions of the interpretation of the results. The authors also wish to gratefully acknowledge the assistance of many members of the staff of this Laboratory including: Jack Brotzman, for the design of the gain stabilizer and sum-coincidence circuits used in this experiment; C. C. Martin, W. P. Aleksiewicz, and B. C. Taubersmith, for assistance in the design of some of the physical apparatus; Dr. Charlotte Davisson, for providing the computer programming for the Monte Carlo calculations to generate the finite-geometry-correction coefficients; Miss Doris Ellis for programming the predictions of the correlation functions; Carl Starling, for general assistance with the instrumentation; and Dr. L. A. Beach, for his interest in and support of the work. The authors also wish to express their appreciation to Dr. A. G. Blair, of the Los Alamos Scientific Laboratory, for communicating his results prior to publication.