Excited-State Spin Assignments Based on $(n, \gamma\gamma)$ Angular-Correlation Measurements on Medium-Weight Nuclei*

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(Received 20 February 1964)

The angular correlations of cascade gamma rays were measured for Ca^{45}, Fe^{55}, Ni^{59}, Ni^{61}, and Ni^{63} formed by thermal-neutron capture. The observation of large anisotropies allows an unambiguous assignment of $J^\pi = \frac{3}{2}^-$ to be made for states at excitations of 2.24 MeV in Ca^{45}, 0.870 MeV in Ni^{59}, and 0.158 and 0.526 MeV in Ni^{63}—all of which exhibit an $l=1$ stripping pattern in (d,p) reactions. The data for the $l=1$ levels at 0.413 MeV in Fe^{55}, 0.470 MeV in Ni^{59}, and 0.284 MeV in Ni^{61} indicate isotropy to within the statistical errors of less than 1.5%, a result that is consistent with an assignment of $J^\pi = \frac{1}{2}^-$ for these levels. The levels at 1.310 MeV in Ni^{59} and 1.008 MeV in Ni^{63} have also been assigned $J^\pi = \frac{1}{2}^-$ on the basis of an observed isotropy, although this assignment is somewhat uncertain because the statistical errors are about 12 and 7%, respectively. The results definitely show that the alleged separation of levels associated with the $p_{3/2}$ and $p_{1/2}$ gross-structure groups is incomplete; $J = \frac{1}{2}$ levels exist within the $p_{3/2}$ group.

INTRODUCTION

THE nuclides in the region from calcium to zinc may be expected to be described rather well by a relatively simple shell-model picture. The $2s$ and $1d$ shells close at Ca^{40} for both protons and neutrons; for nuclides heavier than calcium, both protons and neutrons should fill the $1f_{7/2}$, $2p_{3/2}$, $1f_{5/2}$, $2p_{3/2}$, $2d_{5/2}$, etc., shells in roughly that order. The fact that these nuclides are also expected to have good spherical symmetry further suggests that a simple shell-model picture may be adequate to describe their properties. One might expect, therefore, that careful study of these nuclides, by means of single-nucleon transfer reactions will yield considerable information about their structure in terms of the shell model.

Some of this structure has been studied in a number of experiments performed in recent years. Early poor-resolution experiments on gross structure in (d,p) reactions were interpreted in terms of a simple single-particle model.[1] In particular, it was observed that in

FIG. 1. The computed anisotropy as a function of the multipole mixture for a typical case. Effects of the size, shape, and relative positions of the sample and crystals have been neglected.

several nuclides the $l=1$ strength was split into two groups that had relative intensities of about 2:1, the more intense group being at lower excitation energy. It was concluded that the intense lower group corresponded to the $2p_{3/2}$ single-particle state and the upper group to the $2p_{1/2}$ state. It is known from high-resolution experiments[2] that these "gross structure" states are actually groups of states, all of which have the same value of l. It is important, therefore, to measure the total angular momenta of these individual levels to see if the simple association of the groups with the shell-model states is correct.

Several workers have observed that thermal-neutron capture corresponds very closely to the $l=1$ (d,p) reaction. Capture in a zero-spin target is always into $\frac{1}{2}^+$ states that decay by strong $E1$ gamma transitions to low-lying levels with $J = \frac{1}{2}^-$ or $\frac{3}{2}^-$ that make up the $l=1$ gross-structure states. Measurement of the characteristics of these gamma rays can therefore provide information on these low-lying p states. For instance, a measurement[3] of the circular polarization of the strong gamma rays that follow the capture of thermal neutrons in Ti^{48} indicated the presence of a strong spin $-\frac{1}{2}$ state in the gross-structure peak assigned to the $p_{3/2}$ state. (This assignment was confirmed by subsequent measurements[4] of the angular correlations of the neutron-capture gamma rays emitted by Ti^{49}.) In addition, spin assignments based on the measurement[5] of the inelastic scattering of neutrons by Cr^{53} also lead to the suggestion that spin$-\frac{1}{2}$ states exist within the "$p_{3/2}$" gross-structure group in that nuclide. These results indicate that the nuclear structure is more complex than the early measurements had suggested. It is
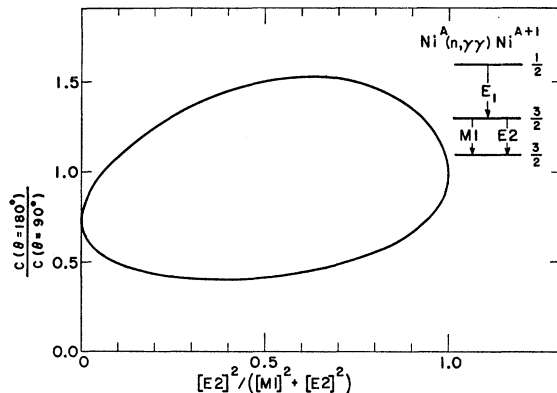
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therefore desirable to obtain as much information as possible about the spins of the states involved.

The angular correlations of the strong cascade gamma rays that follow the capture of thermal neutrons by nuclei by zero spin can be used to determine the spins of the low-lying states in the final nucleus. This correlation usually will be difficult to interpret in terms of unambiguous spin assignments. The possibility of considerable multipole mixing will ordinarily complicate the correlation enough to prohibit the unique determination of spin assignments. In the particular case of low-lying p states, however, unique assignments can be made for many cases. The (d,p) result that $l=1$ for the states in question limits the total angular momentum to $\frac{1}{2}^-$ or $\frac{3}{2}^-$. These states are frequently reached by $E1$ radiation from the $\frac{1}{2}^+$ capturing state. If the state in question is the intermediate state in a γ-γ cascade, a $\frac{1}{2}^-$ state will always produce an isotropic angular correlation. A $\frac{3}{2}^-$ state usually leads to an anisotropic angular correlation of the form $a_0+a_2P_2(\cos\theta)$ in which $P_2(\cos\theta)$ is the second Legendre polynomial, although an isotropic correlation can be obtained for a few particular multipole mixtures. The measurement of an anisotropy in the $(n,\gamma\gamma)$ angular correlation thus indicates an unambiguous assignment of $\frac{3}{2}^-$ for the intermediate state. An isotropic angular correlation strongly suggests an assignment of $\frac{1}{2}^-$, but such an assignment cannot be made with certainty on this basis alone. Often additional information, such as a measured lifetime of the state, can make the assignment definite. The expected anisotropy $C(\theta=180°\mathrm{C})/C(\theta=90°)$ as a function of the $E2/M1$ mixing ratio $[E2]^2/([E2]^2+[M1]^2)$, where the bracketed quantities are the amplitudes of the indicated transitions, is shown in Fig. 1 for a typical case. The present paper reports the measurement of several of these correlations taken to obtain the spins of some of the states that make up the peaks of the p-wave structure.

EXPERIMENTAL DETAILS

Since the aim of the present series of experiments was limited to a quick survey of the extent of the mixing of

FIG. 2. Schematic diagram of the experimental arrangement. Crystal A was 16 in. from the center of the samples, crystal N was 12 in., and crystal B was 9 in. The lead in front of crystal A was about $\frac{1}{4}$ in. thick.

FIG. 3. Typical spectra that illustrate the two methods of analysis. In case (a) the pulse heights in the high-energy crystal were restricted to the shaded region to obtain the accompanying low-energy γ-ray spectrum from crystal B which was used for analysis of the intensity of the transition. In case (b), the pulse height in the low-energy crystal N was restricted to the shaded region to obtain the spectrum of the accompanying high-energy γ ray. In this case the intensity of the transition was determined from an analysis of the high-energy spectrum. All spectra are coincidence spectra; hence ground-state transitions are present at all only because of accidental coincidences.

states having $J=\frac{1}{2}^-$ and $\frac{3}{2}^-$ in the gross-structure peaks, an optimum experimental arrangement was considered secondary to an expedient one. We therefore elected to do the present experiment at the beam port normally reserved for the fast chopper at the research reactor CP-5. The chopper rotor was removed and the beam emerging from the slits of the entrance collimator was further defined by a second collimator designed for experiments with capture gamma rays. This collimator, placed 15 m from the chopper housing, restricted the beam at the sample position to a size comparable to that of the sample. The intensity of the neutron beam was measured to be uniform within a few percent over the width of the samples. The samples were, with one exception, right circular cylinders with diameters from $\frac{1}{4}$ to $\frac{3}{4}$ in. and heights of 5–8 in. All of the samples were enriched to at least 94% in the isotope of interest. The
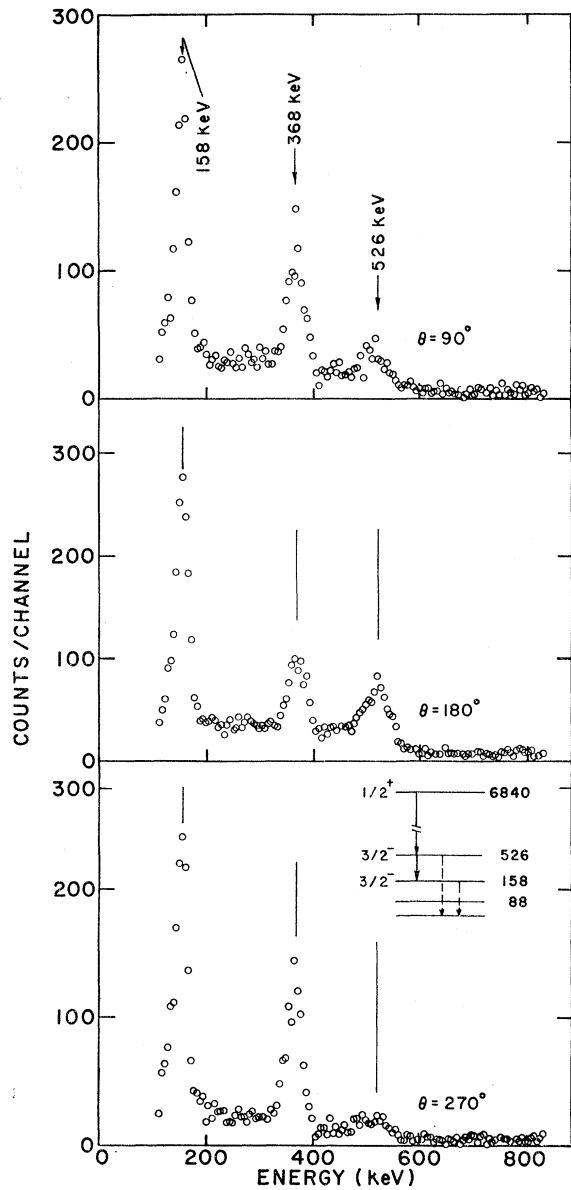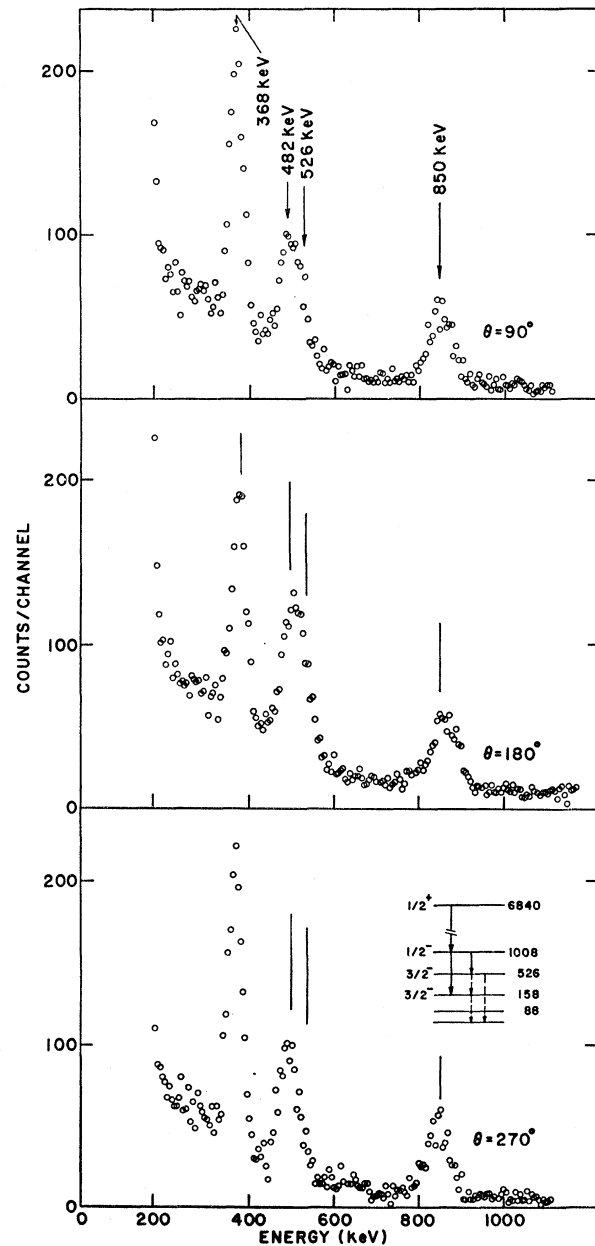
FIG. 4. Examples of the spectra in coincidence with gamma rays of about 6.31 MeV. These were used to determine the spin of the level at 0.526 MeV in Ni[63]. The level scheme is based on the work of Fulmer and McCarthy (Ref. 2). The solid lines indicate the cascade that was studied to determine the spin. The dashed lines indicate the origin of the other lines that appear in the spectrum. It should be noted that it was the intensity of the line at 368 keV that was analyzed in this case and not the transition (526 keV) to the ground state.

NaI(Tl) crystal detecting the primary gamma was 6 in. high by 8 in. in diameter, the two for the de-excitation gammas were 4×4 in. Approximately $\frac{1}{4}$ in. of lead was placed in front of the 6-×8-in. crystal to reduce coincidences between high-energy gamma rays and annihilation quanta that escape from this crystal and are detected by the smaller ones. The coincident pairs of pulse heights were recorded on magnetic tape of the

FIG. 5. Examples of the spectra in coincidence with gamma rays of about 5.83 MeV. These were used to determine the spin of the level at 1.008 MeV in Ni[63]. For comments about the level scheme, see the caption for Fig. 4. An analysis of the line at 850 keV was used to determine the spin of the level at 1.008 MeV.

nickel and iron samples were in the form of metal or oxide, the calcium in the form of the carbonate. The samples were continuously rotated to avoid any systematic errors that might be caused by inhomogeneities.

The experimental arrangement is shown in Fig. 2. In all cases a high-energy primary gamma ray to one of the $l=1$ levels was detected in coincidence with a gamma ray resulting from de-excitation of the $l=1$ state. The

TABLE I. The measured angular correlations and the inferred spins. The quantities a_2 and a_0 are the coefficients of the Legendre polynomials that describe the angular dependence. The errors are standard statistical errors.

Nuclide	Spin	E (MeV)	$C(\theta=180°)/C(\theta=90°)$	a_2/a_0 Uncorrected	a_2/a_0 Corrected[a]
Ca^{45}	$\frac{3}{2}^-$	2.24	1.39±0.07	+0.22±0.04	+0.26 ±0.04
Fe^{55}	$\frac{1}{2}^-$	0.413	1.01±0.01	+0.01±0.007	+0.01 ±0.007
Ni^{59}	$\frac{1}{2}^-$	0.470	1.01±0.01	+0.01±0.007	+0.01 ±0.007
	$\frac{3}{2}^-$	0.870	0.74±0.03	−0.19±0.025	−0.21 ±0.025
	$\frac{1}{2}^-$	1.310	0.92±0.11	−0.06±0.08	−0.06 ±0.08
Ni^{61}	$\frac{1}{2}^-$	0.284	1.02±0.015	+0.01±0.01	+0.02 ±0.01
Ni^{63}	$\frac{3}{2}^-$	0.158	1.62±0.10	+0.34±0.04	+0.39 ±0.05
	$\frac{3}{2}^-$	0.526	0.69±0.06	−0.23±0.04	−0.26 ±0.05
	$\frac{1}{2}^-$	1.008	0.98±0.05	−0.02±0.05	−0.014±0.035

[a] The standard corrections (see Ref. 7) have been made for the finite size of the samples and detectors.

Argonne 3-parameter analyzer.[6] The analyzer was triggered by a pulse formed by a coincidence between either 4-×4-in. crystal and the 6-×8-in. crystal in the manner shown in Fig. 2. The crystal labeled B in the figure served as a monitor and was not moved during a series of measurements on one nuclide. Crystal N was set at 90°, 180°, and 270° with respect to a line passing through the sample and extending along the axis of the 6-×8-in. crystal. Ordinarily one set of data consisted of those data taken at 90°, 180°, 270°, and again 180°. When near isotropy was observed, such a series was re-repeated. The reproducibility of the results for any angle was demanded as a minimal criterion for the absence of systematic error. Monitoring by use of fixed-angle co-incidences reduced the likelihood of experimental errors that might be caused by variations in the neutron flux, changes in the gain of parts of the system, changes in co-incidence resolving time, and systematic errors caused by the placement of the limits that define the areas un-der the gamma-ray peaks. In the analysis it was some-times convenient to place restrictions on the high-energy gamma ray and measure the area associated with the lower energy gamma ray; sometimes the opposite approach was found to be better. The analyzer provided the freedom to make this choice after the data were re-corded and it became clear which was the better way for each cascade. Some examples of the spectra for Ni⁵⁹ and Ni⁶¹ are shown in Fig. 3 to illustrate these two approaches; examples of the spectra analyzed to obtain the spins of the levels at 0.526 and 1.008 MeV in Ni⁶³ are shown in Figs. 4 and 5. The relative weakness of the annihilation peak (511 keV) for the case under consider-ation and the dependence of its strength on angle can be seen in Fig. 4. The Compton tail for a gamma ray of about 500 keV, when observed in a 4-×4-in. NaI(Tl) crystal, has only about 1/20 of the intensity of the photopeak; hence the angular dependence on the annihi-lation peak has little influence on the measurement of the angular correlations studied for the levels at 158, 284, and 368 keV. The lines at 413 keV in Fe⁵⁵ and at

470 keV in Ni⁵⁹ are sufficiently well resolved from (and, in fact, are so much stronger than) the annihilation peak that the variation of the intensity of this peak with angle is not an important factor for these cases either— but somewhat more care is required in the analysis than for the lines at lower energies.

RESULTS

The ratios of the counting rates at 180° to those at 90° and the ratios a_2/a_0 of the Legendre polynomial co-efficients for which the standard corrections[7] have been made are shown in Table I along with the values of the spins inferred from these measured quantities. The large observed anisotropies associated with the level at 2.24 MeV in Ca⁴⁵, the 0.870-MeV level in Ni⁵⁹, and the 0.158- and 0.526-MeV levels in Ni⁶³ allow an unam-biguous assignment of $J=\frac{3}{2}^-$ to be made for these levels. The observation that the level at 0.413 MeV in Fe⁵⁵, the level at 0.470 MeV in Ni⁵⁹, and the level at 0.284 MeV in Ni⁶² are isotropic within 1.5% very strongly suggests that the spin of each of these levels is $\frac{1}{2}$. The assignment is made on this basis, although it should be emphasized that, as shown in Fig. 1, it is possible to ob-serve isotropic correlations for the $\frac{3}{2}$ case for particular multipole mixtures. In the case of Ni⁶¹, there is addi-tional evidence that allows a perfectly definite assign-ment to be made. Lynch and Shipley[8] have measured the lifetime of the state at 0.284 MeV to be less than 0.2 nsec; and from this and the Coulomb-excitation meas-urements of Fagg, Geer, and Wolicki,[9] it follows that $[E2]^2/([E2]^2+[M1]^2) \approx 4\times10^{-3}$. The isotropy ob-served in our measurements is consistent with this re-sult only if the spin of this state is $\frac{1}{2}$. The levels at 1.310 MeV in Ni⁵⁹ and 1.008 MeV in Ni⁶³ have been assigned $J=\frac{1}{2}$ on the basis of observed isotropies, but these must be considered to be more uncertain since the statistical errors associated with these measurements are 12% and

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7%, respectively. Bartholomew and Gunye,[10] who have reported values of the spins for the same levels we have studied in Ni[59], agree with our assignments for the levels at 0.470 and 0.870 MeV, but disagree in assigning the spin $J=\frac{3}{2}$ to the level at 1.31 MeV. However, the disagreement is not as large as might be inferred from these different assignments, since the measured correlations[11] from which these spins are deduced are the same within the statistical errors.

DISCUSSION

The levels in Fe[55], Ni[59], and Ni[61] studied in this experiment all fall within the gross structure in the (d,p) spectrum which was interpreted earlier as the $2p_{3/2}$ single-particle neutron state. The results of the present experiment indicate strongly that the first excited p state in all of these nuclides does, in fact have $J=\frac{1}{2}$, inconsistent with the earlier interpretation. In Ni[63] our results indicate $J=\frac{3}{2}$ and $J=\frac{1}{2}$ states so close to each other in excitation that they would earlier have been interpreted as part of the same gross-structure group. In fact, a low-lying $p_{3/2}$, $p_{1/2}$ doublet at very low excitation seems to be characteristic of all the odd-neutron nuclei in this mass range. This has been discussed in more detail in an earlier note.[12]

It is then apparent that there is considerable mixing of spins within the (d,p) gross structure, at least for the $l=1$ groups. Each p-wave gross-structure peak contains states with both $p_{3/2}$ and $p_{1/2}$ strength and the simple interpretation advanced earlier[1] is not correct. It is disappointing that the simple and naive interpretation is not the correct one. However, it is now possible, with the aid of fast computers, to calculate the details of the fine structure for particularly favorable cases. A recent calculation by Ramavataram,[13] for instance, predicts our value of $J=\frac{1}{2}$ for the 413-keV first excited state of Fe[55]. The calculation does not, however, produce the correct spins for some of the higher excited states which are populated strongly in the (d,p) reaction.[14] Better success was achieved for Cr[53], where the calculation did remarkably well in fitting the known spins for a number of levels.

There remains, however, the question of what interaction is responsible for the experimentally observed splitting of the p-wave gross-structure peaks. It is evidently not the spin-orbit force, which had been suggested earlier. Nor is it apparently an isotopic-spin splitting, which should not affect the results of (d,p) reaction.[15] It will be interesting to see if more sophisticated nuclear-structure calculations can reproduce these unexplained effects.

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Exact Calculation of Bremsstrahlung from Polarized Electrons*

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(Received 27 January 1964)

The production of bremsstrahlung by the interaction of polarized electrons with the Coulomb field of a nucleus is considered. An exact calculation of the angular distribution of the outgoing photons, and the azimuthal asymmetry in this distribution is presented. Numerical calculations were done for an incident electron energy $W_1=1.25m$, a photon energy $k=0.75$ (W_1-m), and a nuclear charge $Ze=79e$.

I. INTRODUCTION

APPROXIMATE expressions for the asymmetry in the distribution of photons in bremsstrahlung production from polarized electrons have been developed by the authors and others.[1,2] The numerical results of these calculations indicate the asymmetry to be a maximum for an incident electron energy $W_1 = 1.25m$ and for a photon energy $k=0.75(W_1-m)$. At these energies the validity of the Born approximation is doubtful since $\alpha Z W_1/p_1$ for the incident electron is of order one for gold. For this reason a more detailed analysis of the asymmetry seems desirable. Using a method similar to that used by Jaeger and Hulme, one can compute this asymmetry exactly.[3]

An exact calculation of the differential cross section

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