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Angular correlation studies in Tc^{99}

By P. Jagam and V. Lakshminarayana

The Laboratories For Nuclear Research, Andhra University, Waltair, India.

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The combined angular correlation pattern of the cascades 740-180 and 740-(40)-140 keV is studied by the sum-peak-coincidence technique using a fast-slow coincidence scintillation spectrometer and a 100 channel analyser. Assuming equal admixture of the cascades and the spins of the ground 140 and 180 keV states in Tc 99 to be 9/2+, 7/2+ and 5/2+ respectively, the observed correlation pattern indicated a spin assignment of 1/2+ for the 920 keV state.

Introduction

Cappeller & Klingelhofer (1954), Raboy & Krohn (1958), Estulin *et al* (1958), Bodenstedt *et al* (1959), and Andrade *et al* (1965) carried out investigations on angular correlations of the cascades connecting the excited state at 920 keV and the ground in Tc^{99}. The 920 keV state decays to the ground state through two prompt cascades 740-180 keV and 740-(40)-140 keV. The transitions 140 and 180 keV could not be completely separated by scintillation spectrometers. As a result, the angular correlation studies are beset with the difficulty of interference between the two cascades. All the investigators tried to separate these cascades. The result of the study of Andrade *et al* (1965) in which sufficient pracautions are reported to have been adopted to minimise the admixture of the cascades, differs from the theoretical value by as much as 20%, although the error in estimation is much smaller. It thus appears that instead of attempting to separate out the contributions of the 140 and 180 keV transitions, it is proper to accept them collectively and separate the effect of one of them estimated theoretically. Studies on angular correlations are therefore undertaken with a sum-peak-coincidence scintillation spectrometer.

Experimental details

The sum-peak-coincidence technique was first described by Kantele & Fink (1962), the characteristics of the high efficiency 4π sum-peak-coincidence arrangement being studied in detail by Kantele (1962). In this work he gave a detailed account of the methods of sum-peak-coincidence spectrometry. They also referred to the possibilities of angular correlation studies using this technique. In the present investigation, the detectors are symmetrically situated at a distance of 7 cms from the central source. The dectors are enclosed in anti-Compton shields, the radiations from the source being accepted via a narrow conical opening. In this arrangement, the attenuation factor, defined by Kantele, for monochromatic gamma rays is found to be zero. This proves that 'the cross-talk' (Kantele) is completely eliminated. As such this arrangement is extremely useful for studying weak cascades in the presence of intense singles radiations.

The block diagram of the experimental arrangement is shown in figure 1. The scintillation heads consists of two well matched hermetically sealed Harshaw Type $1\frac{3}{4}''$ dia x 2" thick NaI(Tl) crystals attached to two DuMont 6292 photomultipliers. The heads contain the lead shielding and the electrical connections. The heads are fixed to wooden V-grooves by aluminium collars and mounted on a circular glass plate such that they could be rotated about one another in the horizontal plane with respect to a vertical pivot fixed at the centre of the glass plate. The circumference of the glass plate is divided into 5° intervals to facilitate the measurement of the angle between the axes of the two detectors. A 20° interval is also divided into unit angular intervals for angular resolution experiments. The central pivot about which the dectectors rotate, supports a perspex block for holding the source containers. The source container is a thin walled perspex tube of internal dimensions 4 mm diam × 10 mm high. The two scintillation detectors are connected in a fast-slow type of electronic summing arrangement (figure 1). This arrangement

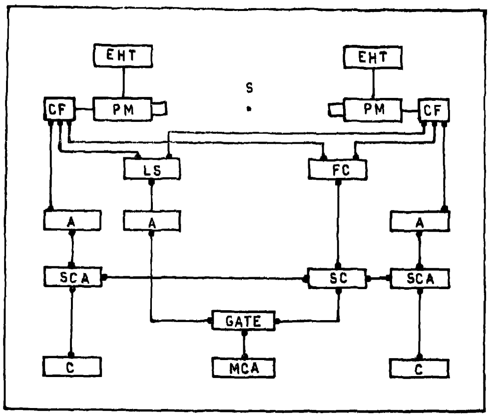

Figure 1. Block diagram of the experimental arrangement : *S* : Source, *PM* : Photomultiplier, *EHT* : High Tension Unit, *CF* : Cathode Follower, *LS* : Linear Summing Network, *A* : Amplifier, *FC* : Fast Coincidence Unit, *SCA* : Single Channel Analyser, *SC* : Slow Coincidence Unit, *C* : Counter, *MCA* : 100 Channel Analyser.

is similar to that of Kantele & Fink except for the inclusion of the fast channel. The fast channel of the present arrangement is based on blocking oscillators and the effective resolving time of the arrangement is experimentally determined to be 100± 10 ns. The sum peak-coincidence spectra are recorded on a 100 channel analyser (Tvpe 2A/NISS/ED) supplied by Bhaba Atomic Research Centre, India.

Results

Scintillation Spectrometer Characteristics : The performance of the electronic summing scintillation spectrometer thus assembled is tested with a Co^{60} source. A few drops of the source are placed in the source container. The position of the source container is adjusted such that the count rate in the movable counter situated in different angular positions is constant to within 0.5%. The sum spectrum is recorded and is shown in figure 2. It can be seen from the figure that

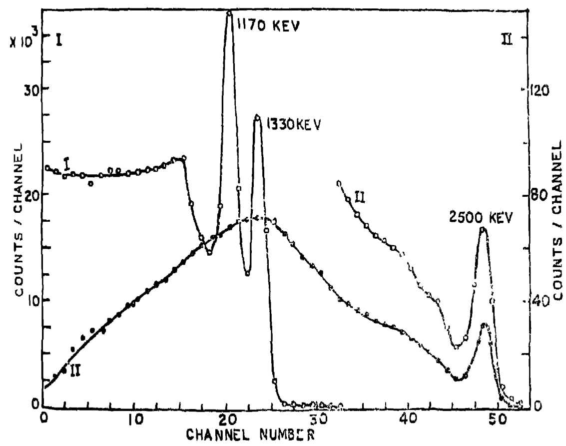

Figure 2. The Sum and Sum-peak Coincidence Spectra for Co^{60}. The curve with O (open circles) represents the sum spectrum and the curve with. O (full circles) represents the zero-bias sum-peak coincidence spectrum.

peaks appear at energies 1.17, 1.33 and 2.5MeV corresponding to single gamma components and the coincident sum energy. The zero-bias sum-peak-coincidence spectrum is recorded by setting the side channels integrally at zero-bias (approximately 80keV in the present work). This spectrum is also shown in figure 2 for comparison. It can be seen from this figure that the sum-peak-coincidence spectrum consists of the sum energy peak at 2.5MeV, the singles peaks being completely attenuated. The area above the continuous distribution under the peak at 2.5 MeV is representative of the coincidence count rate. The coincidence counting efficiency is thus double that of an identical conventional set up in which narrow differential channels select 1.17 and 1.33 MeV for coincidence counting, The background is also completely eliminated from the coincidence spectrum.

The sum-peak-coincidence spectrum is recorded on the 100 channel analyser for angles 90°, 135°, 180°, 225° and 270° between the detectors. These angular positions are changed once in every fifteen minutes to smooth out time dependent fluctuations. A pooled total of 30,000 counts are collected at each angle under the peak at 2.5 MeV. The correlation coefficients are obtained employing White's (1963) method. The values of the coefficients thus obtained are

$$A'_2 = 0.099 \pm 0.006$$

$$A'_4 = 0.008 \pm 0.006$$

These are to be corrected for the angular resolution of the detectors as suggested by Frackel (1951) while neglecting the source size effects. The attenuation factor is given by

$$S_{2\nu} = J_{2\nu}/J_0 \qquad \ldots(1)$$

where

$$J_{2\nu} = \int_0^{\theta_{max}} P_{2\nu}(\cos\theta)\,\varepsilon(\theta)\sin\theta d\theta \qquad \ldots(2)$$

J_0 is the value of $J_{2\nu}$ for $\nu = 0$

$P_{2\nu}(\cos\theta)$ is the Legendre Polynomial

$\varepsilon(\theta)$ is the relative efficiency

θ_{max} is the engle of the conical opening in the shielding of the detector.

The relative efficiencies $\varepsilon(\theta)$ are obtained from the angular resolution curves plotted for different energies (In^{114m} : 192 keV, Cs^{137} : 662 keV, Co^{60} : 1170 keV and 1330 keV, Sb^{124} : 1690 keV) by the method of Lawson & Frauenfelder (1953). The correction factors S_2 and S_4 for the experimental arrangement in the present work are shown in figure 3. For the case of Co^{60}, the attenuation factors for the 1.17 and 1.33 MeV energies are employed to obtain the corrected coefficients $A_{2\nu}$. The corrected coefficients are obtained from the experimental coefficients $A'_{2\nu}$ from the relatation

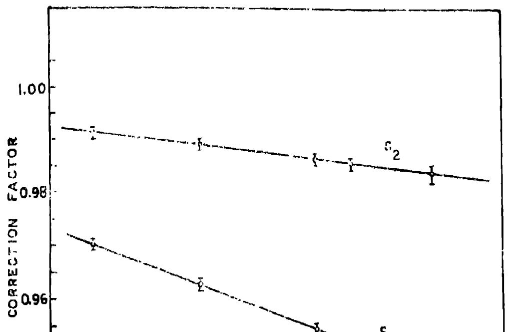

Figure 3. Plots of the attenuation factors S_2 (Upper line) and S_4 (lower line) as functions of energy. They are experimentally determined for the present arrangement.

$$A_{2v}=\frac{A'_{2v}}{S_{2v}(\gamma_1).\,S_{2v}(\gamma_2)} \qquad \ldots(3)$$

where $S_{2v}(\gamma_1)$ and $S_{2v}(\gamma_2)$ are the attenuation factors for the coincident gamma rays γ_1 and γ_2 of the cascade under investigation. In the present case of Co^{60}, the corrected coefficients are obtained to be

$A_2 = 0.101 \pm 0.006$

$A_4 = 0.008 \pm 0.006$

The value of A_2 and A_4 thus obtained to fit the correlation pattern expected for a $E2-E2$ cascade in the spin sequence $4+\to 2+\to 0+$ thereby showing the suitability of the present arrangement for undertaking angular correlation studies.

Note on figure 3 for the correction factors : It is mentioned in the experimental details that the scintillation detectors are enclosed in lead shielding, the radiations from the source being accepted *via* an axial conical opening in the front, which is contrary to normal practice. However, the lead shields are essential because of the sum-peak-coincidence technique in the present investigation. The reason is that, because of the summation, the single gamma rays also appear in the coincidence spectrum of crystal to crystal scattering (referred to as "cross-talk" by Kantele 1962). The lead shielding is designed to avoid all the cross-talk and to provide good geometry with maximum efficiency as illustrated in figure 2 with a Co^{60} source. The absence of the well resolved peaks at 1170 and 1330 keV in the coincidence spectrum proves that the cross-talk is completely by eliminated.

Because of the shielding of the crystals, the correction to be applied for the finite size of the detectors with the shielded crystal geometry is not the same as that with open crystal geometry. Hence, the correction factors quoted in literature for the open crystal geometries are not valid in the present case. As in the case of open crystal geometries, the angular resolution corrections must be determined by the method of Lawson & Frauenfelder (1953) for the particular shielded crystal geometry.

In the present investigation, the $1\frac{3}{4}'' \times 2''$ detectors are provided with lead shields with a frontal thickness of 1.6 cm. The axial conical opening has an opening diameter of 1.4 cm and a base diameter of 2 cm in the lead shield. The detector almost touches the lead shield when assembled in the detector head. For a source situated at the apex of the cone, i.e. at 7 cm from the face of the crystal along the axis, the detector has an acceptance angle of approximately 15°. Because of the conical opening in the front the effective acceptance angle of the detector is not the same at all the energies. Due to the penetration of the higher energy gamma rays, the detector has a larger opening angle at higher energies than at lower energies. In the angular resolution experiment performed with these detectors, on the lines of Lawson & Frauenfelder (Jagam), it is found that the acceptance angle increases from 15° at 192 keV to 19° at 1690 keV. As a result of the increase in the opening angle of the detector, the value of the correction factor decreases with the increase of energy. As such the attenuation for the angular correlation coefficients increases at higher energies contrary to the trend observed with open crystal geometries as reported in the literatute. It is therefore evident that one has to be careful in applying the angular resolution corrections with shielded crystal geometries.

Study of Angular Correlations in Tc^{99} : The radioactive source Mo^{99} is obtained from the Isotope Division of Bhabba Atomic Research Centre, India. It is produced by the fission of uranium with neutrons from the reactor. The Mo^{99} fraction is separated from the fission fragments by alumina-gel chromatography. Carrier free Mo^{99} is finally obtained as ammonuim molybdate in dilute ammonium hydroxide solution the activity being 3mC/ml. The decay scheme of Mo^{99} is well established (Cretzu & Hohmuth 1965, Jagam & Lakshinarayana 1967) and is shown in figure 4.

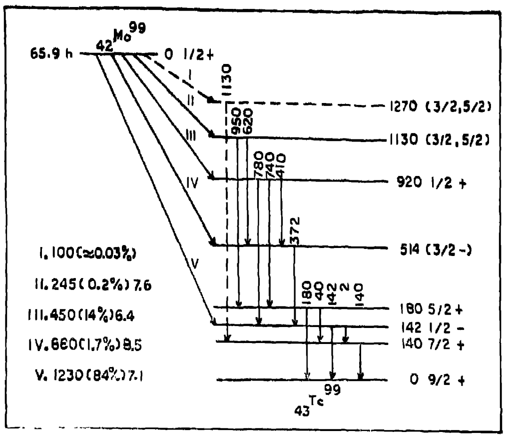

Figure 4. The decay scheme of Mo^{99}. The dashed lines represent the transitions and levels introduced from the work of Jagam & Lakshminarayna.

A few drops of the source sample are placed in the source container. After making the usual adjustments, the zero-bias sum-peak-coincidence spectrum is recorded for fifteen minutes and shown in figure 5. The peak at 920 keV contains predominant contributions from the two prompt cascades 740-180 and 740-(40)-140 keV. A small amount of contribution from the 620-370 keV cascade on the high energy side of the 920 keV peak can be neglected. It can be seen from the figure that the intensity of the peak at 920 keV above the continuous distribution is large and can be measured accurately. The angular correlation studies are carried out by recording the coincidence spectrum at angles 90°, 135°, 180°, 225° and 270° between the detectors. The position of detectors is changed every fifteen minutes and a pooled total of about 30,000 counts is collected at each angular position under the peak at 920 keV. The counts are fitted by White's analysis to the standard polynomial.

$$W(\theta) = 1 + A'_2 P_2 (\cos\theta) + A'_4 P_4 (\cos\theta)$$

where

$$A'_2 = -0.030 \pm 0.006$$
$$A'_4 = +0.009 \pm 0.004$$

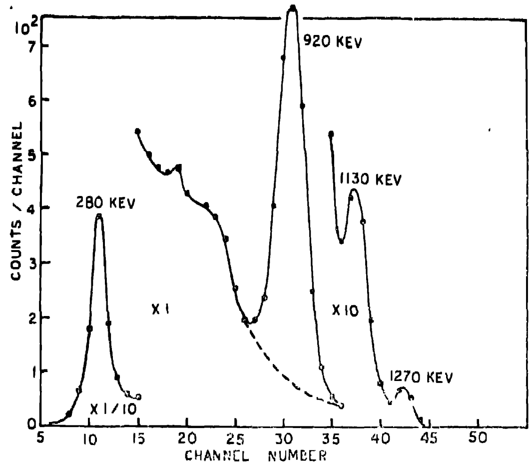

Figure 5. The zero-bias sum-peak-coincidence spectrum recorded for 15 minutes. It can be seen that the 920 keV peak can be well resolved from the distribution.

are the correlation coefficients determined experimently. These are to be corrected for the angular resolution of the detectors. For this purpose, the correction factors for energies 140, 180, and 740 keV are obtained from the figure 3 by interpolation. The angular resolution correction is effected by employing equation (3). The corrected values of the correlation coefficients are given by

$$A_2 = -0.031 \pm 0.006$$
$$A_4 = +0.009 \pm 0.004$$

The peak at 920 keV occuring in the sum-peak-coincidence spctrum accepts both the cascades 740-180 and 740-(40)-140 keV. The 180 keV state decays in equal proportions (after allowing for internal conversion) *via* the two branches. Thus the observed correlation is the result of admixture of the two cascades in equal proportions. However, the first one is a double cascade and the second one is a triple cascade with the intermediate gamma (40 keV) unobserved. Thus the correlation patterns are given by

$$W(\theta)_{740-180} = 1 + A_2 P_2 (\cos\theta) + A_4 P_4 (\cos\theta)$$
$$W(\theta)_{740-(40)-140} = 1 + U_2 A_2 P_2 (\cos\theta) + U_4 A_4 P_4 (\cos\theta)$$

The factor U_{2k} is given by

$$U_{2k} = \frac{-\begin{Bmatrix} I_a & I & 2k \\ I_b & I_b & 1 \end{Bmatrix} + \delta^2 \begin{Bmatrix} I_a & I_a' & 2k \\ I_b & I_b & 2 \end{Bmatrix}}{1+\delta^2} \times$$

$$(-1)^{I_a+I_b}[(2I_a+1)(2I_b+1)]^{\frac{1}{2}}$$

where I_a and I_b are the initial and final spins connected by the unobserved gamma-ray and δ^2 is the mixing ratio of the transition. In the present case, since equal admixture of the two cascades are involved, the final values of A_2 and A_4 are given by

$$A_2 = \tfrac{1}{2}\,[\,A_2\,(740\text{-}180) + U_2\,A_2\,(740\text{-}140)]$$

$$A_4 = \tfrac{1}{2}\,[\,A_4\,(740\text{-}180) + U_4\,A_4\,(740\text{-}140)\,]$$

The theoretical values of A_2 and A_4 are estimated using the above relations for accepted spins of the ground, 140 and 180 keV states (viz. 9/2+, 7/2+, and 5/2+ respectively) and possible spin values of 1/2+ and 3/2+ for the 920 keV state. In this estimation, however, there is an uncertainty arising out of the errors in the relative intensity estimates of the cross-over and the cascade from the 180 keV state. These errors are also estimated for an assumed 10% deviation in the relative intensity of the cascade and cross-over transitions (i.e. 40-140 and 180 keV). If the spin of the 920 keV state is assumed to be 1/2+, the 740 keV transition is pure $E2$. The 40 and 140 keV transitions may be of $M1 + E2$ types, while the 180 keV transition is pure $E2$. From the experimental evidence (Estulin *et al* 1958, Ravier *et al* 1961) the 40 keV transition is assumed to be pure $M1$ and the correlation coefficients are estimated using the relations given above, for various values of the mixing amplitude δ in the 140 keV transition. The total values of the correlation coefficients for the two cascades weighted as detailed above, are given in table 1. It can be seen from table 1 that the present experimental value of A_2 falls in between the theoretical values for quadrupole contents between 1.2% and 5.9% or above 98.8% for the 140 keV transition. However, the latter value for the quadrupole content requires a large negative A_4 coefficient. The present experimental value for A_4, however, is small and positive. It therefore appears reasonable to accept the first possibility (quadrupole content in the 140 keV transition between 1.2 and 5.9% with δ positive). The result is also in agreement with the experimental value of Ravier *et al* (1961) (2% for the quadrupole content in the 140 keV transition). A spin assignment of 1/2 + for the 920 keV state therefore seems appropriate.

However, in order to find out whether other possible spin assignments can be made, theoretical values of the correlation coefficients are estimated

TABLE 1. THEORETICAL VALUES OF THE TOTAL CORRELATION COEFFICIENTS FOR THE 740-(40)-140 KEV TRANSITIONS. A SPIN SEQUENCE OF 1/2+→5/2+→7/2+→9/2+ IS ASSUMED. THE 40 KEV TRANSITION IS ASSUMED TO BE PURE $M1$. THE VALUES AND SIGNS OF δ GIVEN IN THE TABLE REFER TO THE 140 KEV TRANSITION

No	δ	A_2 (total) for δ positive	A_2 (total) for δ negative	A_4 (total)	Quadrupole content % $\delta^2/1+\delta^2$
1	0	+ 0.015±0.006	+ 0.015±0.006	+ 0.0046±0.0005	0.00
2	1/9	− 0.024±0.009	+ 0.053±0 005	+ 0.0043±0.0005	1.22
3	1/4	− 0.068±0.013	+ 0.095±0.006	+ 0.0028±0.0005	5.88
4	3/7	− 0 116±0.018	+ 0.135±0.009	− 0.0003±0.0007	15.51
5	2/3	− 0.156±0.020	+ 0.165±0.012	− 0.0050±0.0011	30.75
6	1	− 0.176±0.023	+ 0.172±0.013	− 0.0110±0.0016	50.00
7	3/2	− 0.169±0.023	+ 0.153±0.011	− 0.0169±0.0021	69.23
8	7/3	− 0.139±0.020	+ 0.113±0.008	− 0.0217±0.0027	84.48
9	4	− 0.098±0.016	+ 0.066±0.005	− 0.0247±0.0029	94.14
10	9	− 0.056±0.011	+ 0.020±0.006	− 0.0261±0.0031	98.77
11	∞	− 0.019±0.009	− 0.019±0.009	− 0.0265±0.0031	100.00

for a spin assignment of 3/2 + for the 920 keV state. In this estimation, as in the previous case, the 40 keV transition is assumed to be pure $M1$. In addition, the 140 keV transition is assumed to be $M1$ with 2% $E2$ admixture ; but both signs of δ are considered in the estimates. Assuming at first the sign of δ in the 140 keV transition to be positive the correlation coefficients are estimated for different values of δ (as well as the two signs) in the 740keV transition (920keV3/2 + → 180keV5/2 +). The procedure is repeated for the negative sign of δ in the 140 keV transition. As in the previous case, the two cascades are weighted together with an error of 10% in their admixtures. The values of the total correlation coefficients are given in tables 2 and 3. It can be seen from tables 2 and 3 that there are four cases of agreement between the theoretical and experimental values of the A_2 coefficients. However, in none of these cases the theoretical values of A_4 is of correct sign. In all these cases the theoretical values for the A_4 coefficients are negative, while the experimental value is positive, It appears unlikely that the spin of 920 keV state is

TABLE 2. THEORETICAL VALUES OF THE TOTAL CORRELATION COEFFICIENTS FOR THE 740-(40)-140 +740-180 KEV TRANSITIONS. A SPIN SEQUENCE OF $3/2+ \rightarrow 5/2+ \rightarrow 7/2+ \rightarrow 9/2+$ IS ASSUMED. THE 40 KEV TRANSITION IS ASSUMED TO BE PURE $M1$. THE 140 KEV TRANSITION IS ASSUMED TO BE $M1+2\%$ $E2$ WITH δ POSITIVE. THE VALUES AND SIGNS OF δ GIVEN IN THE TABLE REFER TO THE 740 KEV TRANSITON.

No.	δ	A_2 (total) for δ positive	δ negative	A_4 (total)	Quadrupole content % $\delta^2/1+\delta^2$
1	0	$+0.024\pm0.007$	$+0.024\pm0.007$	-0.00000 ± 0.00000	0.00
2	1/9	$+0.011\pm0.004$	$+0.037\pm0.011$	-0.00006 ± 0.00001	1.22
3	1/4	-0.007 ± 0.002	$+0.050\pm0.015$	-0.00030 ± 0.00002	5.88
4	3/7	-0.025 ± 0.008	$+0.062\pm0.018$	-0.00070 ± 0.00008	15.51
5	2/3	-0.043 ± 0.013	$+0.069\pm0.020$	-0.00140 ± 0.00020	30.75
6	1	-0.055 ± 0.016	$+0.066\pm0.020$	-0.00230 ± 0.00030	50.00
7	3/2	-0.057 ± 0.017	$+0.055\pm0.016$	-0.00320 ± 0.00040	69.23
8	7/3	-0.050 ± 0.015	$+0.037\pm0.011$	-0.00390 ± 0.00050	84.48
9	4	-0.038 ± 0.012	$+0.019\pm0.006$	-0.00440 ± 0.00050	94.14
10	9	-0.025 ± 0.007	$+0.002\pm0.001$	-0.00450 ± 0.00050	98.77
11	∞	-0.012 ± 0.004	-0.012 ± 0.004	-0.00460 ± 0.00050	100.00

TABLE 3. THEORETICAL VALUES OF TOTAL CORRELATION COEFFICIENTS FOR THE 740-(40)-140 +730-180 KEV TRANSITIONS. A SPIN SEQUENCE OF $3/2+ \rightarrow 5/2+ \rightarrow 7/2+ \rightarrow 9/2+$ IS ASSUMED. THE 40 KEV TRANSITION IS ASSUMED TO BE PURE $M1$. THE 140 KEV TRANSITION IS ASSUMED TO BE $M1 + 2\%$ $E2$ WITH δ NEGATIVE. THE VALUES AND SIGNS OF δ GIVEN IN THE TABLE REFER TO THE 740 KEV TRANSITION.

No	δ	A_2 (total) for δ positive	δ negative	A_4 (total)	Quadrupole content % $\delta^2/1+\delta^2$
1	0	-0.044 ± 0.004	-0.044 ± 0.004	-0.00000 ± 0.00000	0.00
2	1/9	-0.019 ± 0.002	-0.068 ± 0.005	-0.00006 ± 0.00001	1.22
3	1/4	$+0.013\pm0.001$	-0.093 ± 0.008	-0.00030 ± 0.00002	5.88
4	3/7	$+0.048\pm0.004$	-0.115 ± 0.009	-0.00070 ± 0.00008	15.51
5	2/3	$+0.080\pm0.006$	-0.127 ± 0.010	-0.00140 ± 0.00020	30.25
6	1	$+0.102\pm0.008$	-0.123 ± 0.010	-0.00230 ± 0.00030	50.00
7	3/2	$+0.106\pm0.009$	-0.102 ± 0.008	-0.00320 ± 0.00040	69.23
8	7/3	$+0.092\pm0.008$	-0.068 ± 0.005	-0.00390 ± 0.00050	84.48
9	4	$+0.067\pm0.006$	-0.034 ± 0.003	-0.00440 ± 0.00050	94.14
10	9	$+0.047\pm0.004$	-0.003 ± 0.001	-0.00450 ± 0.00050	98.77
11	∞	$+0.023\pm0.002$	$+0.023\pm0.002$	-0.00460 ± 0.00050	100.00

3/2 +. The present experimental results therefore favour a unique assignment of 1/2 + for the spin of 920 keV state.

REFERENCES

Andrade, P. Da R., Maciel, A., Muller, C. S., Wirth, J. & Zawislak, F. C. 1965 *Nucl. Phys.* **66**, 545.

Bodenstedt, E., Matthias, E. & Korner, H. J. 1959 *Z. Phys.*, **153**, 423.

Cappelier, U. & Klingelhoffer, R. 1954 *Z. Phys.*, **139**, 402.

Cretzu, T. & Hohmuth, K. 1965 *Nucl. Phys.* **66**, 391.

Estulin, I. S., Cherov, G. M. & Pastukhovn, Z. N. 1958 *Soviet Physics JETP*, **35**, 71.

Frankel, S. 1951 *Phys. Rev.*, **83**, 673.

Jagam, P. 19 Ph. D. thesis, Andhra University, 95, 67, Andhra.

Jagam, & P. Lakshminarayana, V. 1967 *Proceedings of the Nuclear Physics and Solid State Physics Symposium*, Kanpur, 477.

Kantele, J. & Fink, R. W. 1962 *Nucl. Instr. and Meth.*, **15**, 69.

Kantele, J. 1962 *Nucl. Instr. and Meth.* **17**, 33.

Lawson, J. S. & Frauenfelder, H. 1953 *Phys. Rev.*, **91** 649.

Raboy, S. & Krohn, V. E. 1958 *Phys. Rev.*, **111**, 579.

Ravier, J., Marguin, P. & Moussa, A. 1961 *J. Phys. Rad.*, **22**, 249.

White, D. H. 1963 *Nucl. Instr. and Meth.*, **21**, 209.