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Multipole mixing ratios of the 198 and 177 keV γ -ray transitions in ¹⁶⁹Tm

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The multipole mixing ratios for the 198 and 177 keV γ -ray transitions in ¹⁶⁹Tm are determined as $\delta(198) = -6.1^{+0.4}_{-0.6}$ and $\delta(177) = -0.20^{+0.04}_{-0.03}$ from new measurements of angular correlation of the 63-198 and 63-177 keV cascades in addition to measurements on the cascades passing through the 118 and 139 keV levels. The present value of $\delta(198)$ indicates an almost pure E2 character for the 198 keV γ ray while from earlier measurements an admixture of only 9% E2 was reported for this γ ray. For both the 198 and 177 keV transitions the results do not agree with the conversion electron measurements and indicate that the conversion processes in these transitions are not normal. From time-differential measurement on the 63-198 keV cascade, the relaxation constant $\lambda_2 = (0.31 \pm 0.21) \times 10^7 \text{ sec}^{-1}$ is obtained using the source in acetic acid solution. The integral attenuation coefficients for the 316 keV level are found to be $G_2 = 0.19 \pm 0.04$ in liquid chloride source and $G_2 = 0.57 \pm 0.06$ in liquid acetate source.

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The multipole mixing ratios of γ transitions in a nucleus can be determined from internal conversion as well as from γ - γ angular correlation measurements. Due to the finite-size effect of the nucleus, some conversion coefficients (particularly M1), however, deviate from the calculation of Rose et al. [1] in the point nucleus approximation. The deviation from the point nucleus approximation is due to the fact that in these cases the electronnuclear interaction takes place in the immediate vicinity of the nucleus where the electron wave function is mostly modified and, also, within the nucleus itself. In the interior of the nucleus the interaction is not of the same form as in the outside region. Thus in addition to normal conversion matrix elements there appear "penetration matrix elements." It was found that significant anomalies in the conversion coefficient should appear only in transitions highly hindered from the single-particle transition value. The K-shell electrons are mostly affected by the finite size of the nucleus because they have the maximum probability to penetrate into the nucleus itself. On the other hand, the γ - γ angular correlation coefficients are not sensitive to the finite-size effect of the nucleus.

Considering the 198 and 177 keV transitions in the deformed nucleus ¹⁶⁹Tm, we find that these are strongly retarded transitions depopulating the 316 keV level (Fig. 1). For the 198 keV transition the mixing ratio obtained from subshell ratio measurements ($\delta^2 = 0.107 \pm 0.006$) [2] was in good agreement with that reported from γ - $\gamma(\theta)$ measurement on the 198-118 keV cascade ($\delta = -0.306 \pm 0.017$, i.e., $\delta^2 \approx 0.09$) [2]. But in the case of the 177 keV transition, disagreement in the values of $\delta(177)$ were reported from internal conversion coefficient (ICC) [2,3] and γ - $\gamma(\theta)$ measurements on the 177-130 keV cas-

cade [4-6]. For this γ transition Gûnther et al. [2] and Grabowski et al. [3] reported a value of $\delta^2 \approx 0.19$ from subshell ratio measurements. However, γ - $\gamma(\theta)$ measurements (Table I) suggest a value of $\delta^2(177) \approx 0.04$. Due to penetration effects, as mentioned above, the $L_{\rm I}$ and $L_{\rm II}$ conversion may be influenced. Thus for the 198 and 177 keV transitions the measured subshell ratios [2,3] will be sensitive to penetration effects. For the 198 keV transition definite penetration effects in the internal conversion process ($\lambda = 6-9$) were reported by Agnihotry *et al.* [4]. In case of the 177 keV transition, although no such definite penetration parameter was reported, the discrepancy in the results of $\delta(177)$ obtained from ICC and γ - $\gamma(\theta)$ measurements indicates the presence of penetration effects in this transition also. Therefore, for these transitions the ICC results may not give the correct values of mixing ratios. Considering the above facts it seems to



FIG. 1. The relevant portion of the decay scheme of ¹⁶⁹Yb.

be useful to study the angular correlations of the 63-198 and 63-177 keV cascades, not studied earlier, to establish the correct values of mixing ratios of the 198 and 177 keV transitions. Study of these cascades will be helpful also to determine the presence or absence of penetration effects in these transitions. Since these cascades pass through the 316 keV level having a rather long half-life $(T_{1/2} = 660 \text{ nsec})$ (Fig. 2), it is to be expected that the angular correlation coefficients will be attenuated within the lifetime of the intermediate level, even in liquid sources. The time-dependent form of the attenuation in liquid sources is given by $A_2(t) = A_2(0)e^{-\lambda_2 t}$, where λ_2 is the relaxation constant. Thus from time-differential angular correlation study it is possible to find out the unattenuated value of A_2 from which the mixing ratio of the corresponding γ transition can be determined. In the present paper we have performed the differential and integral measurements on the 63-198 and 63-177 keV cascades in liquid chloride and acetate sources using a highresolution LEPS-HPGe system. Measurements of angular correlation on the cascades passing through the 118 and 139 keV levels have also been repeated to check the consistency of our experimental setup.

The radioactive source ¹⁶⁹Yb $(T_{1/2} = 32 d)$ is obtained from BRIT, Bombay, India, in the form of YbCl₃ solution in HCl. For the measurements of angular correlation of various cascades passing through the 118 and 139 keV levels (Fig. 1) a standard slow-fast coincidence setup is used. The coincidence spectra are taken in the LEPS detector gated by the 198 keV γ ray for cascades passing through the 118 keV level and gated by the 177 keV γ ray for cascades passing through the 139 keV level. The measurements are done at five angles and data are stored in five different groups of a multichannel analyzer (MCA). The results of measurements on these cascades are listed in Table I.

Measurements on the 63-198 keV cascade have been performed in two different liquid sources, viz., YbCl₃ solution in HCl and Yb($C_2H_3O_2$)₃ solution in CH₃COOH. For preparation of the acetate source, the original source is evaporated to dryness and is dissolved in glacial acetic acid. For time-differential measurements on this cascade



FIG. 2. The time-to-amplitude conversion spectrum of the 63-198 keV cascade showing the lifetime of the 316 keV level in 169 Tm.

the time-to-amplitude conversion (TAC) spectra (Fig. 2) are stored in different groups of the MCA for different angles. The integral measurement on the 63-177 keV cascade is performed also from the TAC spectra using the liquid source of YbCl₃. Measurement on this cascade is not performed in the acetate source. The results of integral measurements on the 63-198 and 63-177 keV cascades are given in Table II.

From the measurement on the 198-118 keV cascade we obtain the value of A_2 , in agreement with the results of earlier measurements (Table I). Since the result of Gûnther *et al.* [2] has less error, we consider their value of $A_2 = -0.288 \pm 0.010$ for our subsequent analysis. This value of A_2 corresponds to two possible values of $\delta(198)$, viz., $\delta_1(198) = -0.306 \pm 0.017$ and $\delta_2(198) = -6.1^{+0.4}_{-0.6}$ for the spin sequences of 7/2(1,2)5/2(2)1/2. For the 198-110 keV cascade we obtain the value of $A_2 = 0.340 \pm 0.007$, in good agreement with the results of other measurements (Table I). Now, considering the 63-198 keV cascade (Fig. 1) we find that theoretical values of angular correlation

TABLE I. Results of integral angular correlation measurements on the cascades passing through the 118 and 139 keV levels in 169 Tm.

Cascade			
(keV)	A_2	A_4	Reference
198-118	$-0.325{\pm}0.027$	$0.011{\pm}0.038$	This work
7/2(1,2)5/2(2)1/2	$-0.288{\pm}0.010$	$0.002{\pm}0.016$	[2]
	$-0.280{\pm}0.060$	$0.069{\pm}0.088$	[6]
	$-0.295{\pm}0.016$		[12]
198-110	$0.340{\pm}0.007$	$-0.006{\pm}0.011$	This work
7/2(1,2)5/2(1,2)3/2	$0.341{\pm}0.003$	$-0.004{\pm}0.005$	[2]
	$0.345{\pm}0.009$	$-0.049{\pm}0.013$	[6]
	$0.347{\pm}0.006$		[12]
177-130	$0.259{\pm}0.007$	$0.018 {\pm} 0.011$	This work
7/2(1,2)7/2(2)3/2	$0.263{\pm}0.009$	$0.010{\pm}0.014$	[4]
	$0.253{\pm}0.008$	$0.036{\pm}0.010$	[5]
	$0.254{\pm}0.013$	$0.022{\pm}0.023$	[6]

Cascade	Chemical form of the source	G_2A_2	A_2 (theoretical)	G_2
63-198	YbCl ₃ in HCl Yb(C ₂ H ₃ O ₂) ₃ in CH ₃ COOH	$-0.024{\pm}0.005 \\ -0.058{\pm}0.003$	$-0.102\substack{+0.008\\-0.012}$	$0.23{\pm}0.05 \\ 0.57{\pm}0.06$
63-177	YbCl ₃ in HCl	$0.014{\pm}0.007$	$0.115\substack{+0.015\\-0.013}$	$0.12{\pm}0.06$

TABLE II. Results of integral angular correlation measurements on the cascades passing through the 316 keV level in ¹⁶⁹Tm.

coefficients are $A_2 = -0.36 \pm 0.01$, corresponding to the value of $\delta_1(198)$, and $A_2 = -0.102^{-0.008}_{+0.012}$, corresponding to the value of $\delta_2(198)$. The 63 keV transition is a pure E1 γ transition. Our measurement on this cascade yields the integral values of $G_2A_2 = -0.024 \pm 0.005$ in liquid chloride source and $G_2A_2 = -0.058 \pm 0.003$ in liquid acetate source. Since the attenuation of A_2 is less in the acetate source, we have performed the timedifferential measurement on this cascade in liquid acetate source. The values of $A_2(t)$ at different time channels show that the extrapolated value at zero time channel, i.e., $A_2(0)$, converges very near to -0.102 at zero time channel (Fig. 3), corresponding to the value of $\delta_2(198)$. Therefore, from our differential angular correlation data the value of $\delta_1(198)$ can be ruled out and the acceptable value of $\delta(198)$ is $-6.1^{+0.4}_{-0.6}$. A least-squares fitting of the $A_2(t)$ values by an exponential function yields $A_2(0) = -0.097 \pm 0.010 \text{ and } \lambda_2 = (0.31 \pm 0.21) \times 10^7 \text{ sec}^{-1}$ in the acetate source. The geometry-corrected value of $A_2(0)$ is found to be $A_2(0) = -0.102 \pm 0.010$. There are no reported measurements of λ_2 in ¹⁶⁹Tm in any chemical environment of the source.

For the 177-130 keV cascade we obtain the value of A_2 , in good agreement with the values found from earlier measurements (Table I). Correction for attenuation in liquid sources is expected to be very small for the cas-



FIG. 3. Results of the differential angular correlation measurement on the 63-198 keV cascade in liquid acetate source. The value of $A_2(0)$ found and that expected from ICC results are indicated in the figure.

cades passing through the 139 keV level $(T_{1/2} = 302)$ psec [7]). Our measured value of A_2 on this cascade corresponds to two possible values of $\delta(177)$, viz., $\delta_1(177) = -0.20^{+0.04}_{-0.03}$ and $\delta_2(177) = -0.75 \pm 0.05$. The value of δ_2 , however, can be ruled out from the result of our $\gamma \cdot \gamma(\theta)$ measurement on the 63-177 keV cascade (Fig. 1). For this cascade the values of G_2A_2 have been measured to be $G_2A_2 = 0.014 \pm 0.007$ in the chloride source. The theoretical values of A_2 for the 63-177 keV cascade are $A_2 \approx 0.115$ for the value of $\delta_1(177)$ and $A_2 \approx -0.075$ for the value of $\delta_2(177)$. The sign of G_2A_2 measured therefore supports the value of $\delta(177) = -0.20^{+0.04}_{-0.03}$.

If we consider our measured value of $A_2(0) = -0.102 \pm 0.010$ for the 63-198 keV cascade and our measured values of G_2A_2 , we can calculate G_2 for the 316 keV level. In liquid chloride source we find the value of $G_2 = 0.23 \pm 0.05$ and in liquid acetate source this has been found to be $G_2 = 0.57 \pm 0.06$ (Table II). The value of G_2 in liquid chloride source can also be obtained from our measurement on the 63-177 keV cascade. For this cascade the measured value of $G_2A_2 = 0.014 \pm 0.007$ corresponds to $G_2 = 0.12 \pm 0.06$ (Table II). A weighted average value of G_2 in liquid chloride source can be calculated from measurements on these two cascades and is found to be $G_2 = 0.19 \pm 0.04$.

For both the 198 and 177 keV γ transitions we find that the mixing ratios obtained from $\gamma - \gamma(\theta)$ measurements are not in agreement with those found from conversion electron measurements. From ICC measurements Gûnther et al. [2] and Grabowski et al. [3] reported predominantly M1 character ($\approx 9\%$ E2 admixture) for the 198 keV transition. This is found to be in disagreement with present measurements of $\gamma - \gamma(\theta)$ and $\gamma - \gamma(\theta, t)$ on the 198-118 and 63-198 keV cascades. Gunther et al. [2], from $\gamma - \gamma(\theta)$ measurement on the 198-118 keV cascade, reported a value of $\delta(198)$, in agreement with that obtained from their subshell ratio measurements. In fact, out of two possible values of $\delta(198)$ obtainable from γ - $\gamma(\theta)$ measurement on the 198-118 keV cascade, Gûnther et al. [2] accepted that value which agrees with the value of their subshell ratio measurements. But this value of $\delta(198) = -0.306$ is not consistent with the angular correlation result of the 63-198 keV cascade, whereas the other value, viz., $\delta(198) = -6.1$, is consistent with both the angular correlation results. The present value of $\delta(198) = -6.1^{+0.4}_{-0.6}$ corresponds to 97% E2 for the 198 keV transition, while from earlier measurements of ICC and $\gamma - \gamma(\theta)$ only 9% E2 admixture was found. For the 177 keV transition the result of ICC measurements [2,3] suggest $|\delta(177)| \approx 0.44$, corresponding to 17% E2 admixture, whereas the present result and also the earlier results of $\gamma \cdot \gamma(\theta)$ measurements on the 177-130 keV cascade (Table I) give a different value of $\delta(177) \approx -0.20$, corresponding to almost pure *M*1 transition, *E*2 admixture being only 4%.

The discrepancies in the results of $\delta(198)$ and $\delta(177)$ obtained from ICC and $\gamma \cdot \gamma(\theta)$ measurements may be attributed to the penetration effects. As a result of penetration effects the M1 conversion coefficients are modified according to [8]

$$\alpha(M1,\lambda) = \alpha(M1,\lambda=0)(1+B_1\lambda+B_2\lambda^2)$$

where λ is the penetration parameter and is conventionally expressed in terms of the ratio of the penetration matrix element $\langle ||M1|| \rangle_e$ to the γ -ray matrix element $\langle ||M_1|| \rangle_{\gamma}$. The coefficients B_1 and B_2 are tabulated by Hager and Seltzer [8]. The experimental values of α_K for the 198 and 177 keV γ transitions were reported [9] as $\alpha_K(198) = 0.376 \pm 0.014$ and $\alpha_K(177) = 0.49 \pm 0.03$. The theoretical values of α_K without penetration effects, as tabulated by Rösel *et al.* [10], are found to be $\alpha_k(M1) = 0.39$ and $\alpha_K(E2) = 0.17$ for the 198 keV transition. Since from our measurement an almost pure E2 character for the 198 keV γ transition is obtained,

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the experimental value of $\alpha_K(198) = 0.376$ can be taken approximately as the theoretical value of $\alpha_K(E2)$ with penetration effects. For the 177 keV transition the theoretical values of α_K without penetration effects are found to be $\alpha_K(M1) = 0.53$ and $\alpha_k(E2) = 0.23$ from the table of Rösel *et al.* In this case the experimental value of $\alpha_K(177) = 0.49$ can be taken approximately as the theoretical value of $\alpha_K(M1)$ with penetration effects, since an almost pure M1 character for this γ transition is obtained from our measurement.

In ¹⁶⁹Tm the 316 keV level is assigned 7/2, $7/2^+$ [404] and it decays by K-forbidden ($\Delta K = -3$) 177 and 198 keV transitions to states of the ground rotational band, $7/2^+$ and $5/2^+$, K = 1/2 [411] (Fig. 1). For these transitions anomalies in the internal conversion process should not be appreciable normally since these are K-forbidden retarded transitions [11]. This means that there should not be any discrepancy between the values of mixing ratios obtained from ICC and $\gamma - \gamma(\theta)$ measurements. However, large disagreements obtained from ICC and $\gamma - \gamma(\theta)$ measurements for both the 198 and 177 keV transitions indicate that the internal conversion processes in these cases are not normal and seem not to follow the theoretical prediction of Nilsson *et al.* [11].

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