

Anisotropic angular correlation between the γ rays and the K x rays following internal conversion in $^{169}\text{Tm}^\dagger$

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The angular distribution of K x rays emitted following K conversion has been investigated theoretically. It is shown that the static nuclear quadrupole moment may sufficiently perturb the innermost electron shell to cause anisotropy in the angular distribution between the K x rays and the coincident γ rays, as suggested by Sen and subsequently confirmed experimentally by Sen, Salie, and Tomchuk.

[RADIOACTIVITY ^{169}Tm ; calculated $\gamma x(\theta)$ accompanying K conversion when perturbed by the static nuclear quadrupole moment.]

I. INTRODUCTION

It has been reported by Sen, Salie, and Tomchuk¹ that a small anisotropy exists in the angular distribution of K x rays, accompanying K conversion of the 177-keV transition, with respect to the direction of the 131-keV γ rays which are subsequently emitted in the deexcitation of ^{169}Tm (Fig. 1). This new effect is ascribed to the mixing of atomic states by a noncentral nuclear field; it differs from the anisotropy produced by admixture of magnetic quadrupole radiation, as reported by Catz.² The new result is explained as due to a first-order perturbation of the K -shell wave function by the static quadrupole moment of the nucleus.

It has been pointed out by Sen³ that, because K conversion takes place in or near the nucleus, perturbation effects due to the nucleus may influence the internal conversion process. The first-order perturbation (Church and Weneser⁴) of the K -shell wave function by the static nuclear quadrupole moment adds the states $d'_{5/2}, 1s_{1/2}, J=2$ and $d'_{3/2}, 1s_{1/2}, J=2$ to the unperturbed states $1s_{1/2}, 1s_{1/2}, J=0$. When there is ejection of electrons from these states due to the interaction of the nuclear transition field, x rays will be emitted mainly through electron jumps from the $L_{\text{III}}(p_{1/2})$ and the $L_{\text{III}}(p_{3/2})$ levels (Figs. 2 and 3).

The selection rules for electric multipole x-ray transitions of order L_x between the initial state having orbital and total angular momenta l_0 and J_0 , respectively, and the final state having the corresponding momenta l_f and J_f are (a) $L_x + l_0 + l_f$ must be even and (b) L_x, J_0 , and J_f must form a triangle, i.e., $|J_0 - J_f| \leq L_x \leq J_0 + J_f$. These conditions allow only seven *dominant* x ray transitions

as shown in Figs. 2 and 3; a, b, c, d, e, f, and g indicate the electron transitions resulting in the emission of K x rays. In the absence of any perturbation the x ray transitions arise from electron jumps mainly from the $\frac{3}{2}^-$ and $\frac{1}{2}^-$ states to the $\frac{1}{2}^+$ state only (Fig. 2); hence the resulting K x rays have an isotropic angular distribution. However, with perturbation the x rays will in addition be emitted from electron jumps from the $\frac{3}{2}^-, \frac{1}{2}^-, \frac{1}{2}^+$ states to the $\frac{3}{2}^+$ and $\frac{5}{2}^+$ states (Fig. 3).

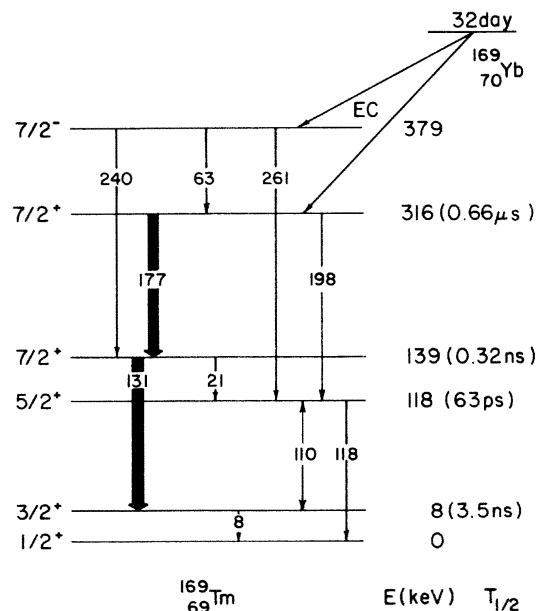


FIG. 1. The partial decay scheme of ^{169}Tm [Lederer, Hollander, and Perlman, *Table of Isotopes* (Wiley, New York, 1967), 6th ed., p. 334]. The transitions of interest in the present work are drawn as heavy lines.

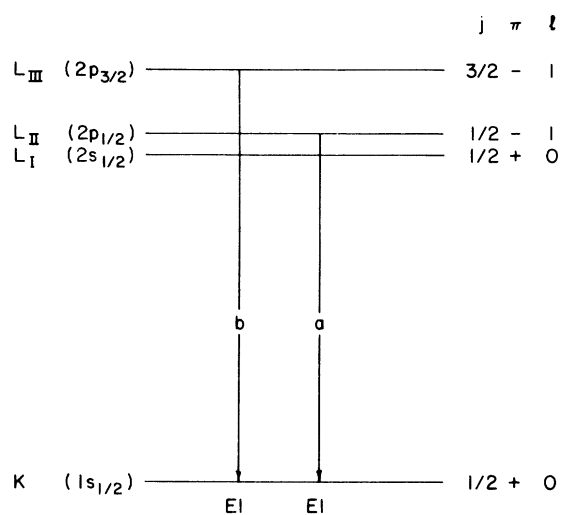


FIG. 2. Electron transitions generating $K\alpha$ x rays (unperturbed situation). The quantum numbers j , π , and l are the total angular momentum, parity, and the orbital angular momentum of the respective atomic states.

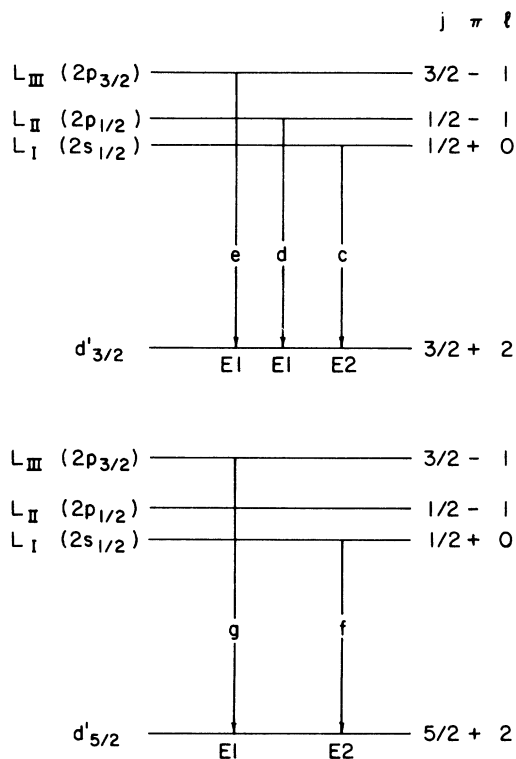


FIG. 3. Additional electron transitions generating $K\alpha$ x rays when the K shell is perturbed by the static nuclear quadrupole moment. Here j , π , and l are the total angular momentum, parity, and the orbital angular momentum of the atomic states, respectively.

These K x rays will not be isotropic with respect to the direction of emission of the subsequent radiation in the cascade.

Sen³ suggested the study of the angular correlation between the K x rays and the corresponding conversion electrons from nuclei with large static deformation and for $E0$ and retarded $E1$ and $M1$ nuclear transitions from long-lived isomeric levels. For $E0$, and retarded $E1$ and $M1$ transitions, the electron penetration in the nuclear volume will be enhanced and the perturbation effect will increase. A long-lived isomeric state will cause the perturbation to act for a long time making the effect larger.

In the present work we analyze theoretically the anisotropic angular correlations observed by Sen, Salie, and Tomchuk.¹ These investigators' experiments were performed using the ^{169}Tm nucleus formed by electron capture decay of ^{169}Yb (Fig. 1). ^{169}Tm is a highly deformed nucleus, uniquely suited for the study of the nuclear quadrupole interaction with the K shell. The 177- and 198-keV (predominantly $M1$) transitions originating from the 316-keV level are strongly forbidden by the K selection rule ($\Delta K=3$). The 316-keV isomeric level ($T_{1/2}=0.66 \mu\text{s}$) isolates most of the K x rays following K capture by the ^{169}Yb nucleus from those below in coincidence measurements with K x rays. No electron capture decay of ^{169}Yb to ^{169}Tm occurs below the 316-keV level. The energy resolution of the x-ray detector, defined by a full width at half maximum of 430 eV at 14.4-keV energy, was good enough to resolve the $K\alpha$ from the $K\beta$ x rays completely. The experimental result¹ for the angular correlation $W(\theta)$ between the K

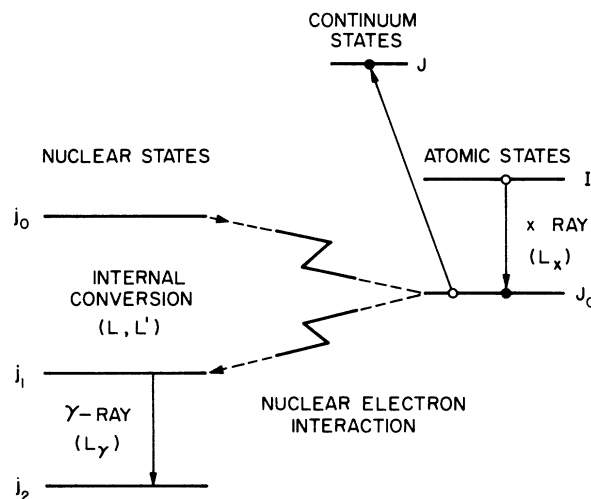


FIG. 4. Atomic and nuclear states involved in the internal conversion process and the deexciting transitions leading to the emission of x rays and γ rays.

x rays following K conversion of the 177-keV γ ray and the 131-keV γ ray in ^{169}Tm (Fig. 1) was

$$W(\theta) = 1 - (0.051 \pm 0.023)P_2(\cos\theta) + (0.011 \pm 0.024)P_4(\cos\theta), \quad (1)$$

where $P_2(\cos\theta)$ and $P_4(\cos\theta)$ are Legendre polynomials.

A theory of directional correlation between the L_3 x rays and γ rays after nuclear electron capture and internal conversion has been developed by Dolginov.⁵ Recently Rupnik and Crasemann⁶ have reviewed Dolginov's work and extended it to calculate the directional correlation between L_3 x rays and γ rays emitted following second forbidden non-unique electron capture.⁷ In the present paper we derive a general expression for the angular distribution between K x rays following K conversion and the coincident γ rays in nuclear decay. We apply the result to show that under normal conditions, i.e., in an unperturbed situation (Fig. 2), the angular distribution is isotropic (as would be expected), while under a perturbed condition (Fig. 3), as for example when the nucleus affects

its own atomic electrons through the quadrupole interaction, the distribution becomes anisotropic, in agreement with the reported experimental results.¹

II. THEORY

As illustrated in Fig. 4, j_0 is the spin of the initial state, j_1 is the spin of the intermediate state after electron conversion, j_2 is the spin of the final state of the nucleus upon γ -ray emission ($j_1 - j_2$) of multipolarity L_γ ; J_0 and $l_0 = J_0 + \lambda_0$ are the total and orbital angular momenta, respectively, of the conversion electron; J_f and $l_f = J_f + \lambda_f$ are the total and orbital angular momenta of the electron which fills the vacancy created by the ejection of the internally converted electron to the continuum state with total angular momentum J and orbital angular momentum $l = J + \lambda$; L_x is the multipole order of the accompanying x ray ($J_0 - J_f$) emission. The parameters λ can have values $+\frac{1}{2}$ and $-\frac{1}{2}$. The angular correlation function between the directions of the x rays following internal conversion and the coincident γ rays is^{5,6}

$$W_{\gamma x} = \sum (-1)^L (2J+1)(2g+1)^{-1} P_{g\eta}^J P_{g\eta}^{J'} |W(J_\sigma J g L', L J_0) W(j_1 j_1 L' L, g j_0) B_{J\lambda L}^{J_0 \lambda_0} B_{J\lambda L}^{J_0 \lambda_0*}|, \quad (2)$$

where L and L' are multipole orders of the conversion transitions; $\eta = 0$ if the quantization direction is chosen as that of either the x ray or the γ ray. The permitted values of g depend on the various angular momenta involved in the expression [$0 \leq g \leq (2L_x, 2L_\gamma, 2J_0, 2j_1)$]. The summation in Eq. (2) is taken over the permissible values of all indices. The γ -ray polarization tensor is

$$P_{g\eta}^{J'} = 2[4\pi(2g+1)(2L_\gamma+1)(2j_1+1)]^{1/2} [1 - g(g+1)/2L_\gamma(L_\gamma+1)] C_{L_\gamma 0 g 0}^{L_\gamma 0} Y_{g\eta}(\theta_\gamma, \phi_\gamma) W(L_\gamma j_2 g j_1, j_1 L_\gamma), \quad (3)$$

while the x-ray polarization tensor is given by

$$P_{g\eta}^{J_0} = 2[4\pi(2g+1)(2L_x+1)(2J_0+1)]^{1/2} [1 - g(g+1)/2L_x(L_x+1)] C_{L_x 0 g 0}^{L_x 0} Y_{g\eta}(\theta_x, \phi_x) W(L_x J_f g J_0, J_0 L_x), \quad (4)$$

where $Y_{g\eta}(\theta, \phi)$ are spherical harmonics, and $\theta_\gamma, \phi_\gamma$, and θ_x, ϕ_x define the direction of the γ rays and the x rays, respectively. The W 's are Racah coefficients while the C 's are Clebsch-Gordan coefficients. The B 's are the matrix elements of the operator which describes the internal conversion process. For electric conversion transitions, we have

$$B_{J\lambda L}^{J_0 \lambda_0} = [(2J_0+1)(2L+1)/L(L+1)]^{1/2} \times \left\{ (2J_0 - 2\lambda_0 + 1)^{1/2} W(L J J_0 - \lambda_0 \frac{1}{2}, J_\sigma J - \lambda) C_{J_0 - \lambda_0 0 L}^{J - \lambda_0} [L R_3 + [L + (2J_0 + 1)\lambda_0 - (2J + 1)\lambda] R_6] \right. \\ \left. + (2J_0 + 2\lambda_0 + 1)^{1/2} W(L J J_0 + \lambda_0 \frac{1}{2}, J_\sigma J + \lambda) C_{J_0 + \lambda_0 0 L}^{J + \lambda_0} [L R_4 - [L - (2J_0 + 1)\lambda_0 + 2(J + 1)\lambda] R_5] \right\} M_L^{g1}, \quad (5)$$

and for magnetic conversion transitions,

$$B_{J\lambda L}^{J_0 \lambda_0} = -[(2J_0+1)(2L+1)/L(L+1)]^{1/2} [\lambda(2J+1) + \lambda_0(2J_0+1)] \times \left\{ (2J_0 + 2\lambda_0 + 1)^{1/2} W(L J J_0 + \lambda_0 \frac{1}{2}, J_\sigma J - \lambda) C_{J_0 + \lambda_0 0 L}^{J - \lambda_0} R_1 \right. \\ \left. + (2J_0 - 2\lambda_0 + 1)^{1/2} W(L J J_0 - \lambda_0 \frac{1}{2}, J_\sigma J + \lambda) C_{J_0 - \lambda_0 0 L}^{J + \lambda_0} R_2 \right\} M_L^{mg}. \quad (6)$$

Here, R_1, R_2, R_3, R_4, R_5 , and R_6 are radial integrals which depend on the multipole orders of the internal conversion transition, on the total (J_0) and orbital ($l_0 = J_0 + \lambda_0$; $\lambda_0 = \pm \frac{1}{2}$) angular momenta of the conversion electron as well as on the total (J) and

orbital ($l = J + \lambda$; $\lambda = \pm \frac{1}{2}$) angular momenta of the electron after conversion. The quantities M_L^{g1} and M_L^{mg} are reduced matrix elements⁵ for the nuclear transition and depend on the structure of the nucleus.⁸ The expressions for R are given by Rose.⁹

TABLE II. Calculated expressions for B and W [Eqs. (11)–(13)] and the anisotropy parameter values in the angular correlation for each of the transitions listed in Table I. The indices in the parentheses associated with the R 's are used to indicate that the radial integrals R_1 and R_2 are not the same for all the transitions. The I 's are the same as in Table I.

I	$B_{J\lambda_1}^{J_0\lambda_0}/M_1^{m\pi}$	$W_{\gamma x}(g=0)/(M_1^{m\pi})^2$	$W_{\gamma x}(g=2)/(M_1^{m\pi})^2$	$W_{\gamma x}(g=2)/W_{\gamma x}(g=0)$
1	$\sqrt{3}[R_1(1)+R_2(1)]$	$-2[R_1(1)+R_2(1)]^2$	0	0
2	$-(\sqrt{3}/2)[R_1(2)+\sqrt{\frac{2}{3}}R_2(2)]$	$-[R_1(2)+\sqrt{\frac{2}{3}}R_2(2)]^2$	0	0
3	Same as for $I=1$	Same as for $I=1$	0	0
4	Same as for $I=2$	Same as for $I=2$	0	0
5	$[R_1(3)+\sqrt{\frac{3}{2}}R_2(3)]$	$-\frac{1}{3}\sqrt{2}[R_1(3)+\sqrt{\frac{3}{2}}R_2(3)]^2$	$-\frac{5}{147}\sqrt{2}[R_1(3)+\sqrt{\frac{3}{2}}R_2(3)]^2$	+0.1020
6	$-2\sqrt{\frac{2}{5}}[R_1(4)+R_2(4)]$	$-\frac{16}{15}\sqrt{2}[R_1(4)+R_2(4)]^2$	$\frac{64}{735}\sqrt{2}[R_1(4)+R_2(4)]^2$	-0.0816
7	$+\sqrt{\frac{3}{5}}[R_1(5)+R_2(5)]$	$-\frac{3}{5}\sqrt{2}[R_1(5)+R_2(5)]^2$	$-\frac{3}{245}\sqrt{2}[R_1(5)+R_2(5)]^2$	+0.0204
8	Same as for $I=5$	Same as for $I=5$	$\frac{5}{49}(\sqrt{2}/3)[R_1(3)+\sqrt{\frac{3}{2}}R_2(3)]^2$	-0.1020
9	Same as for $I=6$	Same as for $I=6$	$-\frac{64}{245}(\sqrt{2}/3)[R_1(4)+R_2(4)]^2$	+0.0816
10	Same as for $I=7$	Same as for $I=7$	$\frac{3}{245}\sqrt{2}[R_1(5)+R_2(5)]^2$	-0.0204
11	Same as for $I=5$	Same as for $I=5$	$-\frac{4}{147}\sqrt{2}[R_1(3)+\sqrt{\frac{3}{2}}R_2(3)]^2$	+0.0816
12	Same as for $I=6$	Same as for $I=6$	$\frac{256}{3675}\sqrt{2}[R_1(4)+R_2(4)]^2$	-0.0653
13	Same as for $I=7$	Same as for $I=7$	$-\frac{12}{1225}\sqrt{2}[R_1(5)+R_2(5)]^2$	+0.0163
14	$-(3/\sqrt{10})[R_1(6)+R_2(6)]$	$-\frac{2}{5}\sqrt{3}[R_1(6)+R_2(6)]^2$	$-\frac{8}{245}\sqrt{3}[R_1(6)+R_2(6)]^2$	+0.0816
15	$3\sqrt{\frac{6}{35}}[R_1(7)+R_2(7)]$	$-\frac{36}{35}\sqrt{3}[R_1(7)+R_2(7)]^2$	$\frac{1152}{12005}\sqrt{3}[R_1(7)+R_2(7)]^2$	-0.0933
16	$-(3/\sqrt{14})[R_1(8)+\frac{1}{3}R_2(8)]$	$-\frac{4}{7}\sqrt{3}[R_1(8)+\frac{1}{3}R_2(8)]^2$	$-\frac{40}{2401}\sqrt{3}[R_1(8)+\frac{1}{3}R_2(8)]^2$	+0.0292
17	Same as for $I=14$	Same as for $I=14$	$\frac{12}{525}\sqrt{3}[R_1(6)+R_2(6)]^2$	-0.0571
18	Same as for $I=15$	Same as for $I=15$	$-\frac{576}{8575}\sqrt{3}[R_1(7)+R_2(7)]^2$	+0.0653
19	Same as for $I=16$	Same as for $I=16$	$\frac{4}{343}\sqrt{3}[R_1(8)+\frac{1}{3}R_2(8)]^2$	-0.0204

that for the angular correlation between the γ ray, of a single multipole order L_γ , for a transition between two nuclear states ($j_1 \rightarrow j_2$), and the coincident x ray of a single multipole order L_x for a transition between two atomic states ($J_0 \rightarrow J_f$). If either the γ ray or the coincident x ray is emitted by alternative transitions, a summation over approximate angular momenta must be taken separately for numerator and denominator in Eq. (11a). A nonzero A_{22} indicates the presence of anisotropy in the γ -ray-x-ray angular correlation.

III. APPLICATION TO ^{169}Tm DECAY

The electron conversion nuclear transition, $j_0 = \frac{7}{2}$ to $j_1 = \frac{7}{2}$, in ^{169}Tm is assumed¹¹ to be pure $M1$ giving $L=L'=1$ (Fig. 1). The succeeding nuclear transition ($j_1 = \frac{7}{2}$ to $j_2 = \frac{3}{2}$) is accompanied by a γ ray, a pure $E2$ transition ($L_\gamma=2$). The possible atomic transitions leading to the emission of x rays are shown in Figs. 2 and 3. The allowed J and corresponding l values are determined from the conditions,

$$J=J_0+L, \quad J_0+L-1, \dots, |J_0-L|$$

with $J=J_0+1, J_0, J_0-1$ in the present case; $l=J+\lambda$ with $\lambda=\pm\frac{1}{2}$, and $(-1)^l(-1)^i=+1$ (no parity change for an $M1$ transition) and since $l_0=0$ or 2, l must be even. The angular momenta of the allowed atomic transitions are listed in Table I. The expressions for $W_{\gamma x}$ [Eqs. (7) and (10)] are then simplified to

TABLE III. The x-ray directional coefficients $A_2^I(K\alpha_{177})$ and the coupling coefficients G_2^I . The I 's are the same as in Tables I and II.

I	$A_2^I(K\alpha_{177})$	I	G_2^I
5, 6, 7	-0.500	5, 8, 11	+0.436
8, 9, 10	+0.500	6, 9, 12	-0.349
11, 12, 13	-0.400	7, 10, 13	+0.087
14, 15, 16	-0.535	14, 17	+0.326
17, 18, 19	+0.374	15, 18	-0.373
		16, 19	+0.117

$$W_{\gamma x}(g=0) = -\frac{8}{3} \sum_{J\lambda} (2J+1)[2(2J_0+1)]^{-1/2} |B_{J\lambda}^{J_0\lambda_0}|^2, \quad (12)$$

$$W_{\gamma x}(g=2) = \frac{20}{49} \sqrt{\frac{5}{3}} \sum_{J\lambda} (2J+1)(2L_x+1)(2J_0+1)^{1/2} [1 - 3/L_x(L_x+1)] C_{L_x 0}^{L_x 0} W(L_x J_f 2J_0, J_0 L_x) \\ \times W(J_0 J 21, 1J_0) |B_{J\lambda}^{J_0\lambda_0}|^2 P_2(\cos\theta), \quad (13)$$

where

$$B_{J\lambda}^{J_0\lambda_0} = -\left[\frac{3}{2}(2J_0+1)\right]^{1/2} [\lambda(2J+1) + \lambda_0(2J_0+1)] \\ \times [(2J_0+2\lambda_0+1)^{1/2} W(1JJ_0 + \lambda_0 \frac{1}{2}, J_0 J - \lambda) C_{J_0 + \lambda_0 0 1}^{J - \lambda_0} R_1 \\ + (2J_0 - 2\lambda_0 + 1)^{1/2} W(1JJ_0 - \lambda_0 \frac{1}{2}, J_0 J + \lambda) C_{J_0 - \lambda_0 0 1}^{J + \lambda_0} R_2] M_1^{mg}. \quad (14)$$

The calculated $B_{J\lambda}^{J_0\lambda_0}$, $W_{\gamma x}(g=0)$, $W_{\gamma x}(g=2)$, and $W_{\gamma x}(g=2)/W_{\gamma x}(g=0)$ are given in Table II for each of the 19 transitions presented in Table I.

The weighted average value¹ of the observed angular correlation coefficient, $A_{22}(K\alpha_{177} - \gamma_{131})$, for the $K\alpha_{177}$ x rays in coincidence with the γ_{131} rays in ¹⁶⁹Tm (Fig. 1) is -0.053 ± 0.023 . As expected, the transitions with $I=1, 2, 3$, and 4 are isotropic. In Tables I and II the I is used as an index to refer to the various transitions. Recalling that only those transitions with a minimum l value (Table I) contribute nontrivially to an anisotropic

correlation, we need consider only those transitions with $I=5, 8$, and 11. Furthermore it is known that x rays with a multipole order $L_x > 1$ are forbidden. Hence we find that the total anisotropy (Eq. 11) is

$$A_{22}(K\alpha_{177} - \gamma_{131}) = \sum_I W_{\gamma x}^I(g=2) / \sum_I W_{\gamma x}^I(g=0), \quad (15)$$

with $I=1, 2, 3, 4, 8$, and 11.

Thus we have

$$A_{22} = -\frac{(\sqrt{2}/147)[R_1(3) + \sqrt{\frac{3}{2}}R_2(3)]^2}{4[R_1(1) + R_2(1)]^2 + 2[R_1(2) + R_2(2)]^2 + \frac{2}{3}\sqrt{2}[R_1(3) + \sqrt{\frac{3}{2}}R_2(3)]^2}. \quad (16)$$

The sign of this A_{22} is the same as that of the observed A_{22} . It is not possible to calculate the numerical value of A_{22} unless the radial integrals are specifically evaluated. However, an upper limit of $A_{22} = -0.0102$ is obtained by neglecting the first two terms in the denominator of Eq. (16). The experimental value is at least 3 times larger than this.

A scrutiny of the various expressions and assumptions involved in the calculation of the anisotropy parameter A_{22} reveals that the present calculations pertain to a much simplified situation. This is particularly true for the transitions from the 316-keV state ($T_{1/2} = 0.66 \mu\text{s}$) of the highly deformed ¹⁶⁹Tm nucleus (Fig. 1) where the penetration of the atomic electrons into the nuclear volume may be very important. A proper theoretical value of A_{22} cannot be obtained until all the relevant radial integrals R_1 and R_2 are calculated using realistic wave functions for both the nuclear and electron states. Nevertheless, the present calculations establish that an anisotropy can arise in the angular distribution of the $K\alpha$ x rays if the K electron shell is sufficiently perturbed by the static quadrupole moment of the nucleus.

The value of A_{22} [Table II, $W_{\gamma x}^I(g=2)/W_{\gamma x}^I(g=0)$]

for each transition can be written in the conventional form for comparison with experimental results:

$$A_{22}^I = A_2(\gamma) G_2^I A_2^I(x). \quad (17)$$

Here, $A_2(\gamma)$ and $A_2^I(x)$ are the directional correlation coefficients of the individual rays, tabulated by Ferentz and Rosenzweig,¹² and G_2^I is the parameter defining the coupling between the two radiations. Now, taking $A_2(\gamma_{131}) = -0.468$ and the values of $A_2^I(K\alpha_{177})$ from the work of Ferentz and Rosenzweig,¹² the G_2^I were calculated (Table III) for $I=5, 6, \dots, 19$. The G_2^I for $I=1, \dots, 4$ are meaningless because the corresponding A_{22}^I vanish. It is evident from Eq. (15) that to obtain the total A_{22} from the A_{22}^I 's, a weight factor involving the radial integral R 's must be calculated for each A_{22}^I . The weight factors are

$$F^I = \frac{W_{\gamma x}^I(g=0)}{\sum_I W_{\gamma x}^I(g=0)} \quad (18)$$

and the total angular correlation coefficient is

$$A_{22}(\gamma - x) = \sum_I F^I A_{22}^I. \quad (19)$$

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⁷Our calculation gave $W_{\gamma x}(\theta) = 1 - 0.142A_I P_2(\cos\theta)$ as against Rupnik and Crasemann's $W_{\gamma x}(\theta) = 1 - 0.187A_I P_2(\cos\theta)$ for ^{207}Bi decay [Eq. (37) of Ref. 6]. In addition, a factor 2 is missing in the expression

for P_{20} [Eq. (16)] given by Rupnik and Crasemann (Ref. 6). These errors have been confirmed by Dr. Rupnik in a letter dated January 21, 1974. (See also erratum in *Phys. Rev. C* **13**, 890 (1976).)

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