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Anisotropic directional correlations between γ rays and K x rays emitted from atoms with deformed nuclei

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Measurements of the directional correlations between K x rays following internal conversion and γ rays in ^{169}Tm and ^{181}Ta have been made. For the cascades in ^{169}Tm the correlation coefficients are $A_{22}(K\alpha_1 - 131\gamma) = -0.032 \pm 0.007$, $A_{44}(K\alpha_1 - 131\gamma) = +0.014 \pm 0.008$, $A_{22}(K\alpha_2 - 131\gamma) = -0.017 \pm 0.007$, and $A_{44}(K\alpha_2 - 131\gamma) = 0.012 \pm 0.007$. For the cascades in ^{181}Ta the correlation coefficients are $A_{22}(K\alpha_1 - 133\gamma) = -0.037 \pm 0.012$, $A_{44}(K\alpha_1 - 133\gamma) = 0.022 \pm 0.017$, $A_{22}(K\alpha_2 - 133\gamma) = -0.038 \pm 0.017$, and $A_{44}(K\alpha_2 - 133\gamma) = 0.037 \pm 0.029$. The anisotropic correlations in ^{169}Tm verify the existence of the effect and serve as a test case for x rays following magnetic-dipole internal conversion. The measurements in ^{181}Ta establish the second known case of this phenomenon and in addition were made with better energy resolution. This case involves several mixed nuclear transitions which result in 38% of the x rays following electric-quadrupole internal-conversion processes.

I. INTRODUCTION

The theoretical and experimental investigation of the directional correlations between x rays and between x rays and γ rays has been the subject of many previous papers. Three theoretical papers sufficiently describe the developments of the currently interesting theoretical considerations.[1-3] The major conclusion of Dolginov's early work[1] was that the K x-γ directional correlations involving x rays following internal conversion in the K shell would be isotropic; however, the experimental results of Perepelkin[4] indicated that an observable anisotropy does exist between K x rays from electron capture, and subsequent γ radiation in the daughter nucleus. Even though later experimental results of Fechner *et al.*[5] did not support the existence of observable anisotropies reported in Ref. 4, the controversy stimulated a great deal of activity and renewed interest in this problem.

The null results reported for K x-γ directional correlations in Ref. 5, involved K x rays from the K capture in ^{54}Mn, ^{139}Ce, ^{153}Gd, and ^{202}Tl. Similarly, null results were also reported by Ramaswamy[6] involving K x rays from the K capture in ^{65}Zn, ^{85}Sr, ^{113}Sn, and by McDonnell and Ramaswamy,[7] in the decay of ^{133}Ba, and by Murty *et al.*,[8] in the decay of ^{114}In. A list of such results was given in 1972 by Ramaswamy.[9] Nonisotropic directional correlations between K and L x rays in Pb were observed by Catz[10] and were in good agreement with theoretical predictions only when small admixtures of $M2$ radiation with the predominantly $E1$ x rays were taken into account. Similar results in ^{181}Ta and in ^{203}Tl were reported by Catz and Macias,[11] in ^{233}U by Catz and Finkel,[12] in ^{239}Pu by Zalutsky and Macias,[13] and in ^{181}Ta by Zalutsky, Macias, and Catz.[14]

In 1972 Sen, Salie, and Tomchuk[15] reported a significant anisotropy in the directional correlation between the K x rays following the internal conversion of the predominantly $M1$, 177-keV nuclear transition and the 131-keV γ ray in ^{169}Tm. They attributed this anisotropy to a new effect caused by the perturbation of the wave functions of the atomic electrons by the static nuclear-quadrupole moment. In particular, this static quadrupole interaction can cause a mixture of the two-coupled two-electron states $|d'_{3/2}, 1s_{1/2}, J=2\rangle$ and $|d'_{5/2}, 1s_{1/2}, J=2\rangle$, with the unperturbed state $|1s_{1/2}, 1s_{1/2}, J=0\rangle$. The K-shell electrons are then in a quantum state which is an admixture of the three states mentioned above, and K-shell internal conversion results in a $d'_{3/2}$ or $d'_{5/2}$ vacancy in the mixed K-shell state. Accordingly, x rays from the transition between either $2p_{3/2}$ or the $2p_{1/2}$ level, to a $d'_{3/2}$ or $d/_{5/2}$ vacancy, following internal conversion, can result in anisotropic x-ray angular distributions relative to the nuclear symmetry axis. This in turn results in anisotropic directional correlations between K x rays nuclear γ rays. A theoretical treatment of these anisotropies was given later by Sen, Gupta, and Tomchuk.[3]

There were two main goals of the present investigation. One was to verify quantitatively and qualitatively the existence of such anisotropic K x-γ directional correlations. The second was to attempt to measure the correlations involving $K\alpha_1$ and $K\alpha_2$ x rays with an energy resolution which would allow sensitive detection of possible experimental interferences which might explain the previously measured strong correlations.[15]

In an earlier letter, we reported measurements of the $K\alpha_1$-x- $K\alpha_2$-x-131-keV γ-ray directional correlations in ^{169}Tm and the results are also reported here. This experiment served to verify the existence of the effect reported by Sen and his co-workers[15]; however, we found a small bump on the high-energy side of the $K\alpha_1$ peak which was consistent throughout the data. In addition, we measured the correlation coefficients of the $K\alpha_1-\gamma$ and $K\alpha_2 - \gamma$ cascades separately and found them to be different. We found that the anisotropy analyzed channel by channel was constant over the energy range covering the $K\alpha_2$ peak but was not constant over the energy range of the $K\alpha_1$ peak. We have reexamined this data and we maintain our earlier conclusion that there was probably a small interference, yet unexplained in the $K\alpha_1$ data, and that the effect is probably about half as strong as that reported in Ref. 15 when the data exclude the high-energy edge of the $K\alpha_1$ line. In other words, we find no reason to question the results reported in Table I for which the $K\alpha_1$ and $K\alpha_2$ lines are reported separately.

We have extended this work to ^{181}Ta using improved experimental techniques. The Ge(Li) detector was replaced by a larger intrinsic Ge detector and new electronic modules were used to improve coincidence timing, γ-ray energy resolution, and stability. In addition, we chose a case with an associated, well-known γ-γ directional correlation with a large anisotropy which allowed the use of a solid source, for better x-ray energy resolution. The strong known γ-γ correlation was used to make the correction for the perturbation of the directional correlations due to the interaction of the nuclear moments with extra nuclear fields. The results of these measurements are far more convincing than those in ^{169}Tm in that they show no evidence of experimental interferences and in addition the resulting correlation coefficients for the $K\alpha_1$-x-γ and $K\alpha_2$-x-γ cascades are not sensitive to the portion of the peaks used in the analysis.

II. EXPERIMENTAL PROCEDURE

The directional correlation measurements were made using a fixed-position 33-cm^3 true coaxial Ge(Li) detector to detect the 131-keV γ ray in the ^{169}Tm measurements, and a 147 cm^3 intrinsic Ge detector to detect the 133-keV γ ray in the ^{181}Ta measurements. In both experiments, the x rays were detected with a 2.44-cm-diameter, 1-cm-deep planar intrinsic Ge x-ray detector with a resolution of 378 eV at 5.9 keV and 634 eV at 122 keV. The x-ray detector was mounted on the moving arm of a 1-m-diameter automatic angular correlation table. The details of this apparatus are given in earlier papers published by one of us (F.T.A.) and will not be repeated here.[17-19]

The coincidence x-ray spectra were obtained from many measurements in which the x-ray detector axis was oriented, relative to the γ-ray detector axis, at angles of 90, 135, and 180° for the ^{169}Tm measurements and at angles of 90, 112.5, 135, and 180° for the ^{181}Ta measurements. The same data positions were performed on both sides of the table, reflected about the 180° axis, to detect possible experimental asymmetries. The source was centered to within 1% singles-counting-rate consistency and a small correction was made for the remaining small experimental asymmetry in the singles-counting rates. Standard coincidence techniques were used in the ^{169}Tm experiments, which involved selectable active filter amplifiers, differential timing, single-channel analyzers, a coincidence circuit, and a linear gate as described in Ref. 16. For the ^{181}Ta experiments, the single-channel analyzers were replaced with units with constant-fraction differential discriminators, more modern amplifiers and a time-to-amplitude converter followed by a single-channel analyzer to produce the gating pulse for the linear gate. In the

TABLE I. Directional correlation coefficients for the indicated K x-γ cascades in ^{169}Tm and ^{181}Ta.

Cascade	A_{22}	A_{44}	Reference
		^{169}Tm	
$K\alpha$-131γ	-0.040 ± 0.019	$+0.045 \pm 0.017$	16
$K\alpha$-131γ	-0.053 ± 0.013	$+0.021 \pm 0.013$	15
$K\alpha_1$-131γ	-0.032 ± 0.007	$+0.014 \pm 0.008$	16
$K\alpha_2$-131γ	-0.017 ± 0.007	$+0.012 \pm 0.007$	16
		^{181}Ta	
$K\alpha_1$-133γ	-0.037 ± 0.012	$+0.022 \pm 0.017$	Present results
$K\alpha_2$-133γ	-0.038 ± 0.017	$+0.037 \pm 0.029$	Present results

^{169}Tm experiment a coincidence time resolution of 100 nsec was used while in the ^{181}Ta case this was increased to 200 nsec to ensure that the apparatus integrated over many times the 11 nsec half-life of the 482-keV intermediate level in the correlation (See Fig. 1). This is necessary if accurate corrections for the perturbation due to extranuclear fields are to be made as discussed below.

The memory of the multichannel analyzer was separated into sections, one corresponding to each angle. The selection of a given section is accomplished with a dc routing system in which the voltage levels are obtained from angle position switches on the correlation table. The movable detector was programmed for repositioning at intervals of 30 min. The total time at each position is recorded using clocks connected to the multichannel analyzer at each section of the memory. The relays and clocks are accurate to one hundredth of a second. This allows easy detection of an artificial asymmetry introduced by a malfunction of the programmer which controls the position of the detector. Accidental coincidence rates were measured after each run and the source strengths were chosen to keep these rates less than 5% of the total coincidence rates. Hundreds of thousands of coincidences were collected at each position.

The preparation of the liquid source used in the ^{169}Tm measurements, the decay scheme of ^{169}Tm and its special features are adequately described in our earlier letter[16] and will not be repeated here. For the ^{181}Ta measurements, it was decided to improve the x-ray energy resolution while also minimizing the scattering within the source by using an evaporated dry source. The radioactive source of ^{181}Hf was obtained by irradiating enriched ^{180}Hf with thermal neutrons. The source was prepared as a liquid by dissolving HfO_2 in a solution of hydrofluoric acid. The solution was deposited on the inside walls of a cylindrical Lucite source holder with a small chamber of radius 2 mm and 3 mm in height. The walls of the chamber were turned to a thickness of approximately 0.25 mm. The improvement in energy resolution can be easily be seen by comparing Fig. 2 with Fig. 2 of Ref. 16. It should be pointed out, however, that the use of a solid source is not advisable except in cases where there is a strong, well known γ-γ directional correlation involving the same intermediate nuclear state as the x-ray-γ correlation of interest. In addition, the assumption is made that the γ-γ correlation and γ-x correlations involving the same intermediate nuclear level are perturbed in the same way.

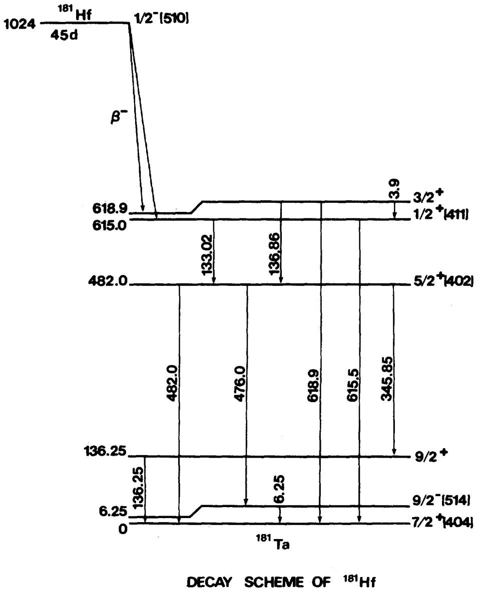

FIG. 1. The decay scheme of ^{181}Hf to the levels of ^{181}Ta.

III. ANALYSIS AND CORRECTIONS TO THE DATA

The data from the ^{169}Tm measurements were analyzed in two ways. First the counts under both $K\alpha$ peaks were summed together for direct comparison to the earlier results given in Ref. 15. Second, the data from the central position of each peak were analyzed separately. The coincidence rates as a function of angle were corrected for the accidental rates and small instrumental asymmetry and fit to a function of the form

$$W(\theta) = 1 + A_{22}P_2(\cos\theta) + A_{44}P_4(\cos\theta), \tag{1}$$

where $P_K(\cos\theta)$ are the Legendre polynomials. In this case it was assumed that in the liquid source, there was no perturbation of the intermediate nuc-

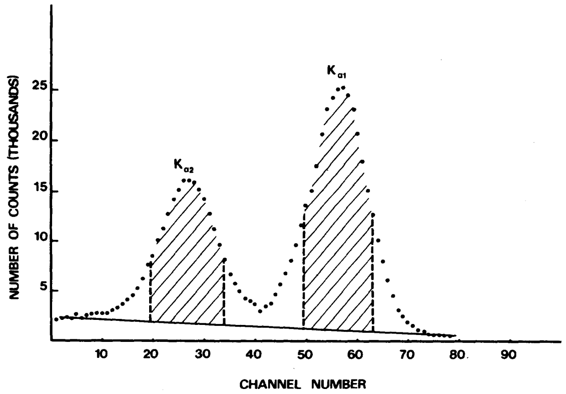

FIG. 2. Typical coincidence spectrum of K x rays in coincidence with the 133-keV γ ray in ^{181}Ta. The shaded areas represent the portion of the data used to compute the correlation coefficients.

lear state due to the interaction of the nuclear moments with extranuclear fields. The resulting A_{KK} correlation coefficients are averaged over the solid angles subtended by detectors. This is usually expressed as

$$A_{KK}(\mathrm{obs}) = Q_K^{(1)}\, Q_K^{(2)}\, A_{KK}(\mathrm{corrected}), \tag{2}$$

where $Q_K^{(i)}$ is the solid-angle correction for the ith detector.[20] The results are presented in Table I along with the results given in Ref. 15.

The corrections to the ^{181}Ta data are complicated by two facts: first, a careful correction must be made for the perturbation of the directional correlation by extranuclear fields and second, the finite solid-angle corrections for the large intrinsic Ge detector do not appear in the literature and had to be calculated from first principles using a complex general-purpose Monte Carlo code.[21]

Directional correlations involving the 11-nsec, 482-keV level in ^{181}Ta, measured in evaporated sources, are well known to suffer serious perturbations due to the interaction of the large quadrupole moment of this state with strong electric-field gradients in the polycrystals formed in evaporation. Fortunately, a very accurate measurement of the strong directional correlation involving the 133-keV–482-keV γ-ray cascade was made earlier.[22] The results of Ref. 22 were obtained with two high-resolution detectors and liquid source. The resulting correlation coefficients were $A_{22} = -0.295 \pm 0.005$ and $A_{44} = -0.069 \pm 0.008$ from which a value of the multipole mixing ratio $\delta(E2/MI) = 5.8(+0.3, -0.2)$ was obtained for the 482-keV transition.[22] These data along with a careful measurement of this correlation with our present source and geometry allow an accurate correction of our K x-γ correlations for both the finite solid angle and extranuclear perturbation effects. The use of the 133-keV-γ–482-keV-γ directional correlation measurement to correct the 133-keV-γ–K-x correlation for the effects of extranuclear perturbations requires that G_{22} be the same for both correlations. While this has not been proven rigorously, the following justification is given on physical grounds. The direction of the 133-keV γ ray forms the quantization ($\theta = 0$) axis for both correlations. In addition, the 482-keV nuclear level is the intermediate state for both the γ-γ and γ-K-x correlations. There is a simple and well-known relationship between a γ-γ and the corresponding γ-e^-(IC) (internal conversion) directional correlations[22] and also a definite correlation between the e_K^- and K x rays which is nonisotropic when the final electron state is not a pure $s_{1/2}$ level. It is well known that the G_{22} factor is the same for a γ-γ correlation and the corresponding γ-e^- correlation, hence we conclude that the same G_{22} factor applies to the corresponding γ-K-x-ray correlation. This is tantamount to considering the triple correlation γ-e^--K x and realizing that the first step is perturbed by G_{22} but that the second step is not effected by the finite lifetime of the intermediate nuclear level.

The γ-γ directional correlation measurement of the 133-keV–482-keV cascade made in the present investigation resulted in the following coefficients:

$$A_{22}^0 \equiv A_{22}G_{22}Q_{22} = -0.077 \pm 0.007, \tag{3a}$$

$$A_{44}^0 \equiv A_{44}G_{44}Q_{44} = -0.012 \pm 0.004, \tag{3b}$$

where G_{KK} is a factor due to the perturbation of the extranuclear fields integrated over the resolving time. The corrected K x-γ directional correlation coefficients can be expressed as follows:

$$A_{KK}(\mathrm{x}\gamma) = A_{KK}^0(\mathrm{x}\gamma)\frac{A_{KK}(\gamma\gamma)Q_K^{(1)}(482)Q_K^{(2)}(133)}{A_{KK}^0(\gamma\gamma)Q_K^{(1)}(133)Q_K^{(2)}(55)} \tag{4a}$$

$$= A_{KK}^0(\mathrm{x}\gamma)C_{KK}(\mathrm{x}\gamma), \tag{4b}$$

where the superscript (1) indicates the 147-cm^3 intrinsic Ge detector and superscript (2) indicates the Planar x-ray detector. The values of $Q_K^{(i)}(E)$ were calculated with the Monte Carlo code and their ratios are far easier to calculate accurately than the absolute values. The results of the Monte Carlo calculations are

$Q_2^{(1)}(0.482) = 0.8645$, $Q_4^{(1)}(0.482) = 0.6007$,

$Q_2^{(2)}(0.133) = 0.9568$, $Q_4^{(2)}(0.133) = 0.8608$,

$Q_2^{(1)}(0.133) = 0.8162$, $Q_4^{(1)}(0.133) = 0.4743$,

$Q_2^{(2)}(0.055) = 0.9509$, $Q_4^{(2)}(0.055) = 0.8426$.

The resulting values of the integrated, extranuclear perturbation factor is $G_{22} = 0.316$. The resulting correction coefficients are $C_{22}(\mathrm{x}\gamma) = 4.08 \pm 0.47$ and $C_{44}(\mathrm{x}\gamma) = 3.7 \pm 1.5$. The resulting corrected correlation coefficients are given in Table I. As a test of the Monte Carlo code, absolute values of several Q_K coefficients were calculated for several sizes of NaI(Tl) detectors for several distances and energies and were found to agree with those of Yates[23] within 1%. The responses to γ rays of both Ge detectors were calculated with the code and agree well with experimental spectra using the relative intensities given in Ref. 22. We conclude from these tests that the quoted errors for the correction coefficients $C_{KK}(\mathrm{x}\gamma)$ are realistic. The much larger uncertainty in $C_{44}(\mathrm{x}\gamma)$ propagates from the larger uncertainties in the A_{44} coefficients in the experimental data.

IV. DISCUSSION AND CONCLUSIONS

We have reported the verification of the anisotropic directional correlations between K x rays and γ rays in ^{169}Tm in a higher resolution measurement[16] than that used in the original discovery of the effect.[15] We have reexamined the data and the analysis and have not changed our conclusions reported earlier.[16] There were two disturbing facts in those results. First, the directional correlation obtained by adding the data under entire $K\alpha$ peak is significantly stronger than that obtained by using only the data from the center of either the $K\alpha_1$ or $K\alpha_2$ peak as seen in Table I. Second, there appeared to be a strange bump on the high-energy side of the $K\alpha_1$ peak which we were unable to explain (see the discussion in Ref. 16). The data from the measurements in ^{169}Tm then, definitely serve to verify the existence of the anisotropic directional correlation, and in addition, the separate measurements of the $K\alpha_1$- and $K\alpha_2$-γ correlations result in data which better serve to test the theory quantitatively. As a further test of the fact that our data were taken under similar conditions to those described in Ref. 15, we summed the coincidence rates under the $K\alpha_1$ and $K\alpha_2$ peaks and obtained correlation coefficients in agreement with those given in Ref. 15.

In the experiment involving the K x-γ directional correlations in ^{181}Ta, it was decided to allow the correlations to be perturbed in a solid source in order to obtain yet higher energy resolution so as to be more sensitive to anomalous peak shapes. In this case no anomalies were noted. This case then serves as the second observation of this phenomenon; however, the interpretation is not as straightforward, since the x rays follow four internally converted transitions from which 38% of the K x rays follow electric-quadrupole internal-conversion processes, while the ^{169}Tm data involve only magnetic-dipole internal-conversion processes.

For the ^{181}Ta data to be used as quantitative test of the theory, it should be clear what fraction of the K x rays is coming from which internal conversions and in particular, what fraction is coming from the conversion of the 482-keV transition, which is known to have a large penetration effect $[\lambda = 175(+7, -4)]$.[22] Using the γ-ray intensities, internal-conversion coefficients, and directional correlation coefficients given in Ref. 22, we can calculate the fraction of the x rays from each of the mixed correlations. There are four γ-γ cascades involved: the (133–482) keV, which is a $\frac{1}{2} \rightarrow \frac{5}{2} \rightarrow \frac{7}{2}$ cascade, the (133–476) and (133–346)-keV cascades, which are both $\frac{1}{2} \rightarrow \frac{5}{2} \rightarrow \frac{9}{2}$ cascades, and the (133–136)-keV triple cascade with a spin sequence $(\frac{1}{2} \rightarrow \frac{5}{2} \rightarrow \frac{9}{2} \rightarrow \frac{7}{2})$. The results are that 71.8% of the K-shell x rays come from the internal conversion of the 136-keV transition with the associated γ-γ directional correlation coefficients $A_{22} = 0.197 \pm 0.012$ and $A_{44} = 0.008 \pm 0.020$,[22] while 19.8% of the K x rays come from the internal conversion of the 482-keV transition with associated γ-γ directional coefficients $A_{22} = -0.295 \pm 0.005$ and $A_{44} = -0.069 \pm 0.008$,[22] and 7.7% of the K x rays come from the 346-keV transition with associated γ-γ correlation coefficients $A_{22} = 0.110 \pm 0.010$ and $A_{44} = +0.022 \pm 0.014$.[22] The remaining 0.7% of the K x rays are due to the pure $M2$, 476-keV transition with theoretically the same associated γ-γ correlation coefficients as the 133–346-keV cascade.

The case of ^{181}Ta is somewhat complicated but should not preclude a careful theoretical analysis. First, the x rays are emitted as a result of four different internally converted transitions. Second, the most intense transition is a mixed $M1 + E2$ multipole transition which is $\simeq 16\%$ $E2$, and third, the $M1$ component of the second most intense transition has a known very large penetration effect. Several facts simplify this situation somewhat so that is should serve as a valuable case for comparison to future theoretical calculations of the atomic radial matrix elements. The mixed $M1 + E2$, 482-keV nuclear transition has a known large mixing ratio[22] $\delta(E2/M1) = 5.8 \pm (0.3,\ 0.2)$ which means that it is $[97.1 \pm (0.3,\ 0.2)]\ \%$ pure $E2$ so that the small ~3% $M1$ mixture can be neglected. This implies that only highly accurate measurements of the properties of the nuclear transitions are sensitive to the small $M1$ component and its inherent penetration effect. Two of the less intense nuclear transitions have the same spin sequence; however, the weak 476-keV transition is magnetic quadrupole while the stronger 346-keV transition is pure $E2$. For practical purposes then about 62% of the x rays arise from magnetic character, internal-conversion processes, and 38% from electric-quardrupole internal-conversion processes.

Also, we have noted that in all cases measured there is a reasonable probability that we have observed A_{44} correlation coefficients different from zero. The valid range of K, the subscript on $P_K(\cos\theta)$, can be no larger than one of several angular momenta and in particular $2L_X$, where L_X is the multipolarity of the x ray. The mixing of the atomic levels does allow transitions from the $2s_{1/2}$ level to both the $d'_{3/2}$ and $d'_{5/2}$ levels. These atomic transitions are of $E2$ character and will contribute to a nonzero A_{44} coefficient. We have carefully examined all of the data from both the K-x-γ and γ-γ directional correlations, and we appear to see the effect of the A_{44} coefficient in the K x-γ correlations, and do not see any anamalous shape in the well-known γ-γ correlation measured in the same geometry. The probable nonzero value of these A_{44} correlation coefficients might serve as further experimental data to help us gain a more complete understanding of the nature of these anisotropic K x-γ directional correlations.

Finally, a complete theoretical prediction of the anisotropic directional correlations, in the cases discussed here, can be made using the formalism presented in Ref. 3 once the appropriate radial integrals describing the atomic transitions are calculated. One of the main motivations for the present experimental investigation was to stimulate such calculations.

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