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Nuclear Structure Effects in Tl^{203}†

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Angular correlation measurements have been made on the 404-kev and 279-kev transitions following electron capture from 52-hour Pb^{203}. The following directional correlations were measured: gamma-gamma, K conversion electron-gamma, and gamma-K conversion electron. The angular distribution of all measurements were of the form $W(\theta)=1+AP_2(\cos\theta)$. The following results were obtained. For the $404\gamma-279Ke^-$ correlation, $A=-0.052\pm0.015$, for the $279\gamma-404Ke^-$ correlation, $A=-0.036\pm0.010$, and for the $404\gamma-279\gamma$ correlation, $A=-0.151\pm0.010$. No effects due to perturbations of the intermediate state were found.

Using the well measured 279-kev K-conversion coefficient, the 404-kev K-conversion coefficient was determined to be 0.117 ± 0.015 while the 680-kev K-conversion coefficient was determined to be 0.011 ± 0.004. From the gamma-gamma angular correlation measurement result and the mixture ratio in the 279-kev transition determined by Stelson and McGowan, the mixture ratio in the 404-kev transition was determined to be $\delta_1=+0.043\pm0.010$. The Sliv value for the K-conversion coefficient for this mixture ratio is 0.147. The correlations involving conversion electrons rule out a $\frac{3}{2}\rightarrow\frac{3}{2}\rightarrow\frac{1}{2}$ transition. They indicate that the $M1$ particle parameter in the 404-kev transition is about twice the theoretical prediction. The particle parameters for the 279-kev transition also do not agree with the theoretical predictions.

I. INTRODUCTION

It has been proposed by Church and Weneser[1] that the internal conversion process should be sensitive to nuclear structure in certain cases. These structure effects should influence the internal conversion coefficient and angular correlations in which internal conversion electrons are one of the particles detected. The structure effects should be large in the so-called l-forbidden magnetic dipole ($M1$) transitions, i.e., orbital angular momentum forbidden by the single particle shell model which allows only spin flip $M1$ transitions, e.g., $p_{\frac{3}{2}}\rightarrow p_{\frac{1}{2}}$ but not $d_{\frac{3}{2}}\rightarrow s_{\frac{1}{2}}$. The study of Tl^{203} was made because its decay involves an l-forbidden $M1$ transition in which structure effects can be determined by both an internal conversion coefficient measurement and by an angular correlation measurement in which a conversion electron is detected. In addition, since there is also an l-allowed $M1$ transition in the decay of Tl^{203}, one can compare the structure effects in an l-allowed and an l-forbidden $M1$ transition in the same isotope.

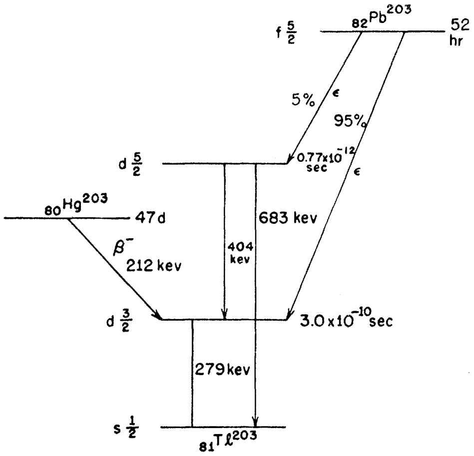

FIG. 1. Decay scheme of Tl^{203}.

The main features of the decay scheme of Tl^{203} as shown in Fig. 1 were established by Prescott[2], Varma[3], Wapstra *et al.*[4] Both the 404-kev and the 279-kev transition have conversion coefficients of the order of 10% making it feasible to measure gamma-gamma, electron-gamma, and gamma-electron correlations. Both transitions are mixtures of $M1$ and electric quadrupole ($E2$) radiation. According to the single particle model assignments for the states, the 404-kev transition is l-allowed while the 279-kev transition is l-forbidden.

II. SOURCE PREPARATION AND APPARATUS

Angular correlation experiments involving electrons require very thin sources to avoid attenuation of the correlation by scattering of the electrons. Because of this, an effort was made to keep the source material to

TABLE I. Gamma-gamma correlations in different chemical environments.

A	Chemical environment
-0.151 ± 0.010	Average of evaporated $PbCl_2$ sources
-0.139 ± 0.015	$PbCl_2$ in H_2O solution
-0.171 ± 0.015	$PbCl_2$ in glycerine
-0.146 ± 0.020	Molten metal. Pb and Sn
-0.180 ± 0.020	Solid metal. Pb and Sn

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‡ Based in part on a thesis submitted for the Ph.D. degree at the University of Pennsylvania.

[1] E. L. Church and J. Weneser, Phys. Rev. **104**, 1382 (1956).

[2] J. R. Prescott, Proc. Phys. Soc. (London) **A67**, 254 (1954).

[3] J. Varma, J. Franklin Inst. **257**, 247 (1954).

[4] Wapstra, Maeder, Nijgh, and Ornstein, Physica **20**, 169 (1954).

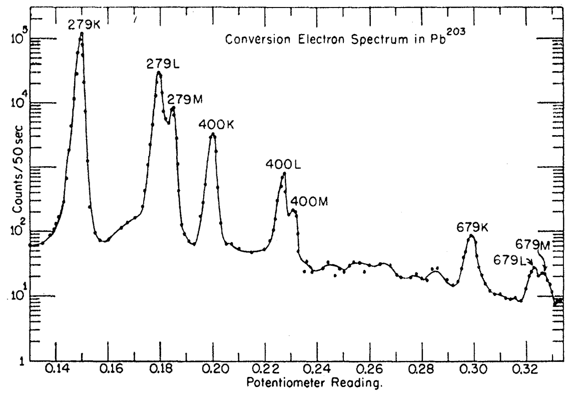

FIG. 2. Conversion electron spectrum obtained with lens spectrometer.

a minimum. Spectroscopically pure thallium (listed by Johnson, Mathey and Company as JM400(Tl) was bombarded with 18-Mev deuterons in the Brookhaven cyclotron producing Pb^{203} through the Tl$^{203}(d,2n)$Pb^{203} reaction. Details of the chemical separation are described in a thesis by Deutch.[5]

The carrier free Pb^{203} was vacuum evaporated from a small Pyrex oven onto a 200μg/cm^2 aluminum foil. Three sources were made in this way. The thickness of all sources was less than 10μg/cm^2.

The gamma-ray detectors were NaI(Tl) crystals mounted on RCA type 6342 photomultipliers. A thin lens beta spectrometer was used for the electron measurements. The detector in the spectrometer was Pilot Plastic Scintillator B mounted on an E.M.I. type 5311 photomultiplier. The coincidence apparatus employed a fast-slow system with a resolving time of 10^{-8} sec. This equipment has been described in greater detail elsewhere.[6]

III. RESULTS

(a) 279-kev Gamma–404-kev Gamma Angular Correlation

Since the intermediate state has a spin $\frac{3}{2}$, the correlation will be of the form $1+AP_2(\cos\theta)$. A number of runs were made at the 90°, 135°, and 180° positions to confirm this. The expansion coefficient A was determined by a least square fit to this data.[7] The remaining data was taken at the 90° and 180° positions only.

Since Van Nooijen and Wapstra[8] had found that the gamma-gamma angular correlation was attenuated in a lead sulphate powder, we decided to search for attenuations in our sources. The results for various sources corrected for finite size of the detectors and source, are shown in Table I. If perturbations on the intermediate state were attenuating the correlation, one would find a smaller correlation in the glycerine solution than in the water solution[9] and a larger anisotropy in the molten source than in the solid source.[10] Our results show no such affect. This results for the evaporated source are in good agreement with the measurements of Lindquist and Markland[11] and Van Nooijen and Wapstra,[8] but not with Varma.[3]

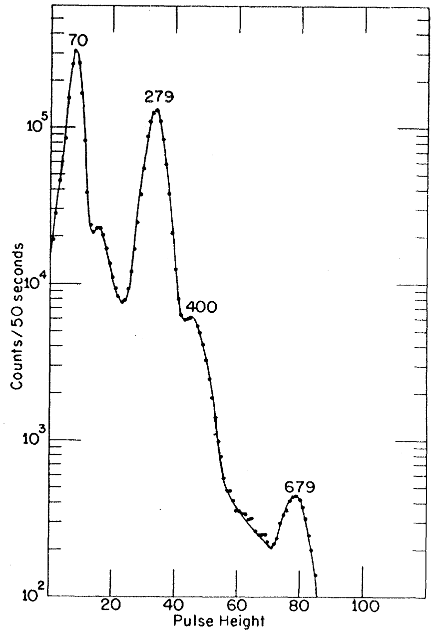

FIG. 3. Gamma-ray spectrum of Tl^{203}.

(b) 404-kev *K*-Conversion Electron–279-kev Gamma Angular Correlation

Vacuum evaporated carrier free PbCl$_2$ sources were used in all measurements involving conversion electrons. Figure 2 shows the conversion electron spectrum obtained with the lens spectrometer. The spectrometer was set to count the 404-kev K-conversion electrons and the gamma detector was set to accept only the 279-kev photopeak. The average value of A determined from runs with three different sources was $A=-0.036\pm0.010$.

(c) 404-kev Gamma–279-kev *K*-Conversion Electron Angular Correlation

Since the weak 404-kev gamma photopeak was riding on the slope of the much stronger 279-kev photopeak (see Fig. 3), shifts in the counting rate due to drifts in the amplifier and/or the pulse-height analyzer did not necessarily represent changes in the number

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[5] B. I. Deutch, thesis, University of Pennsylvania, 1959 (unpublished).

[6] J. V. Kane, thesis, University of Pennsylvania, 1957 (unpublished).

[7] We are indebted to the staff of the Univac at the University of Pennsylvania Computing Center for the use of their facilities and time on the computing machine.

[8] B. Van Nooijen and A. H. Wapstra, Physica **23**, 404 (1957).

[9] P. B. Hemmig and R. M. Steffen, Phys. Rev. **92**, 832 (1953).

[10] H. Frauenfelder *et al.*, Phys. Rev. **92**, 513 (1953).

[11] T. Lindquist and I. Marklund, Nuclear Phys. **3**, 367 (1957).

of 404-kev gamma rays that were being detected. This prevented us from using the usual method for correcting for such shifts and small errors in centering; namely, divide the coincidence rate by the counting rate in the movable counter. Instead we set the window on the pulse height selector wide enough to count all the pulses in the 404-kev photopeak in spite of small shifts in the electronic equipment. Corrections for small errors in centering were made by determining the relative solid angle at each position. The average value of A for three runs was $A=-0.052\pm0.015$.

Because the correlations involving conversion electrons were small, a check for asymmetries in the equipment was made by measuring the 70-kev x-ray-279 K-conversion electron correlation. This correlation should be isotropic, i.e., $A=0$ and was found to be so to within 0.5%. The errors for A reflect this possible asymmetry. Table II summarizes the results of our angular correlation measurements.

(d) Conversion Coefficients

The 404-kev K-conversion coefficient was determined by measuring the relative intensity of the 404-kev K electron and the 279-kev K electron. With a knowledge of the ratio of the gamma rays and the 279 K-conversion coefficient, one can calculate the 404-kev K-conversion coefficient from the relation

$$\alpha_K(404)=\alpha_K(279)\left(\frac{N_\gamma(279)}{N_\gamma(404)}\right)\left(\frac{N_e(404)}{N_e(279)}\right) \tag{1}$$

where $\alpha_K(279)=0.162\pm0.003$,[12] and $(N_\gamma(279)/N_\gamma(404))$ = ratio of 279- to 404-kev gamma rays $=18.5\pm2$.[13] Our value of 0.117 ± 0.015 is in excellent agreement with the value 0.118 of Nijgh *et al.*[12]

In a similar manner we determined the K-conversion coefficient of the 683-kev transition to be 0.011 ± 0.004. We have used $(N_\gamma(279)/N_\gamma(683))=117$.[1–4]

IV. DISCUSSION

(a) Angular Correlation Function

The angular correlation for all the thallium cascades is of the form

$$W(\theta)=1+AP_2(\cos\theta). \tag{2}$$

TABLE II. Summary of all angular correlation measurements.

Particles detected	A
$\gamma-\gamma$	-0.151 ± 0.010
$e-\gamma$	-0.036 ± 0.010
$\gamma-e$	-0.052 ± 0.015

TABLE III. Value of $b_2{}^m$ and b for $Z=81$ for various values of λ.

	λ	-19	-3	$+1$	$+5$	$+21$	Rose
$b_2{}^m$	279 kev	0.040	0.054	0.059	0.066	0.114	0.050
	404 kev	0.084	0.113	0.126	0.142	0.257	0.110
b_2	279 kev	-0.108	-0.119	-0.125	-0.130	-0.171	-0.121
	404 kev	-0.247	-0.260	-0.268	-0.277	-0.342	-0.269
$b_2{}^e$	279 kev						1.43
	404 kev						1.30

The coefficient "A" can be expressed as a product

$$A=A_1A_2, \tag{3}$$

where A_1 depends only on the parameters of the first transition and A_2 depends only on the parameters of the second transition.[14] For a mixed $M1+E2$ transition in a gamma-gamma correlation for the ith transition

$$A_{i\gamma}=[1/(1+\delta_i{}^2)][A^m+2\delta_iA+\delta_i{}^2A^e], \tag{4}$$

where the A^m, A, and A^e are tabulated by Beidenhorn and Rose[14] and δ^2 is the ratio of the $E2$ gamma-ray transition probability to the $M1$ gamma-ray transition probability.

For a mixed transition where the particle detected is a K-conversion electron we have

$$A_{ie}=\{1/[1+(\alpha/\beta)\delta_i{}^2]\}[b^mA^m+2(\alpha/\beta)^{\frac{1}{2}}\delta_ibA + (\alpha/\beta)\delta_i{}^2b^eA^e], \tag{5}$$

where α and β are, respectively, the $E2$ and $M1$ K-conversion coefficients. The particles parameters b^m, b, and b^e have been tabulated for a point nucleus by Beidenharn and Rose.[14]

(b) Nuclear Structure Effects

The original calculations of internal conversion coefficients of Rose *et al.*[15] assumed a point nucleus. Later calculations of Sliv[16,17] included the effect of a finite nucleus. The Sliv calculations differ from the Rose calculations mainly in the use of improved electron wave functions. The effect of electron penetration of the nucleus is only included in an average way in their model which restricts the nuclear transition currents to the nuclear surface.

More recently, Church and Weneser[1] have suggested that electron penetration of the nucleus can in certain cases have a large effect on the conversion coefficient. The effects which depend on details of the nuclear structure, have been characterized in their treatment for $M1$ transitions by a parameter

$$\lambda=m_e/m_\gamma, \tag{6}$$

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[12] G. S. Nijgh and A. H. Wapstra, Nuclear Phys. **9**, 545 (1958/9).

[13] Nijgh, Wapstra, Ornstein, Salomons-Grabben, and Huizenga, Nuclear Phys. **9**, 528 (1958/9).

[14] L. C. Biedenharn and M. E. Rose, Revs. Modern Phys. **25**, 729 (1955).

[15] Rose, Goertzel, Spinrad, Harr, and Strong, Phys. Rev. **83**, 79 (1951).

[16] L. A. Sliv, Zhur. Eksptl i Teoret. Fiz. **21**, 770 (1951).

[17] L. A. Sliv and M. Listingarten, Zhur. Eksptl. i Theoret. Fiz. **22**, 29 (1952).

where the m_e is a new nuclear matrix element which arises from the penetration of the electron into the nucleus and m_γ is the matrix element for $M1$ gamma emission. Sliv's assumption that the currents are restricted to the nuclear surface corresponds to $\lambda=1$. In terms of λ, the corrected $M1$ conversion coefficient is given approximately by

$$\beta(\lambda)\sim\beta(1)[1-(\lambda-1)C(Z,k)]^2, \qquad (7)$$

where the $C(Z,k)$ can be determined from tables in Green and Rose.[18]

The particle parameters, b, appearing in the angular correlation function for conversion electrons (5) are also influenced by the finite size of the nucleus. Sliv has calculated b^m and b^e including finite size effects in the same approximation that he used for the conversion coefficients, namely for $\lambda=1$.[19] Church, Rose, and Weneser[20] have described the procedure for calculating b^m and b as a function of λ. Values of the b's for various values of λ are shown in Table III. These values were interpolated from calculations made for $Z=78$ and $Z=83$.

V. EVALUATION OF THE DATA

(a) Excluding Spin Sequences $\frac{3}{2}\rightarrow\frac{3}{2}\rightarrow\frac{1}{2}$

Assuming the value of δ for the 279-kev transition reported by McGowan and Stelson,[21] $\delta_2=+1.50\pm0.08$, we have calculated $A_{2\gamma}=-0.99\pm0.01$ from (4). This is shown graphically in Fig. 4. This assumption is not an important restriction since $A_{2\gamma}$ is relatively insensitive to δ_2 in the region around $\delta_2=1.5$. Use of the value $\delta_2=1.4$ of Deutch, Wilhelm, and Metzger[22] or even $\delta_2=1.2$ determined by Nijgh and Wapstra[12] would not affect our conclusions significantly. Using this value of $A_{2\gamma}$ and our measurement of $A(\gamma-\gamma)$ we determine $A_{1\gamma}=0.152\pm0.010$ by using (3). Similarly $A(e-\gamma)$ yields $A_{1e}=+0.036\pm0.010$. The sign of these results alone can rule out a suggestion by Nooijen and Wapstra[7] that the spin sequence in the Pb^{203} cascade might be $\frac{3}{2}\rightarrow\frac{3}{2}\rightarrow\frac{1}{2}$. This can be seen as follows:

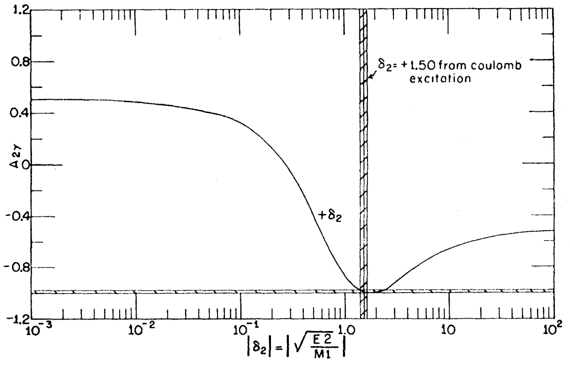

FIG. 4. $A_{2\gamma}$ as a function of δ_2. Vertical hatched lines show value of δ_2 determined from Coulomb excitation. Horizontal hatched lines show our value of $A_{2\gamma}$.

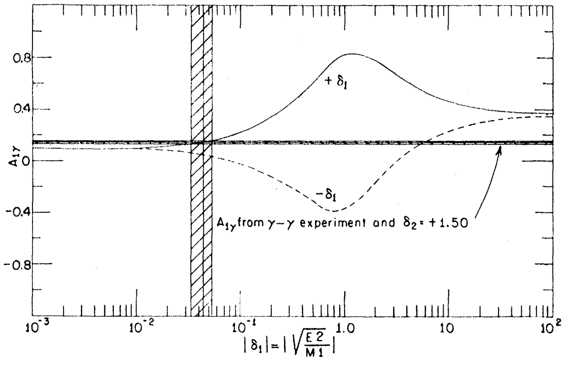

FIG. 5. $A_{1\gamma}$ as a function of δ_1 for a $\frac{5}{2}\rightarrow\frac{3}{2}$ transition. Horizontal lines show our determination of $A_{1\gamma}$ using the results of $\gamma-\gamma$ experiment and $\delta_2=+1.50$. Vertical hatched area shows the resulting value of δ_1.

The gamma and electron angular correlation functions of a mixed $M1+E2$ transition for a $\frac{3}{2}\rightarrow\frac{3}{2}$ spin state are

$$A_{1\gamma}=\frac{1}{1+\delta_1{}^2}[A^m+2\delta_1A+\delta_1{}^2A^e]=\frac{1}{1+\delta_1{}^2}[-0.4-1.55\delta_1],$$

$$A_{1e}=\frac{1}{1+(\alpha/\beta)\delta_1{}^2}\left[b^mA^m+2\delta_1(\alpha/\beta)^{\frac{1}{2}}bA+\delta_1{}^2\frac{\alpha}{\beta}b^eA^e\right]$$

$$=\frac{1}{1+0.226\delta_1{}^2}[-0.0616+0.288\delta_1].$$

A positive $A_{1\gamma}$ requires a negative δ_1, which results in a negative A_{1e} contrary to our results.

(b) Determination of δ for 404-kev Transition

Figure 5 is a graph of $A_{1\gamma}$ as a function of δ_1 for a $\frac{5}{2}\rightarrow\frac{3}{2}$ transition. Our result shown by the hatched area allows two different values of δ_1 but conversion coefficient and K/L measurements exclude the large value (about -5). The result $\delta_1=0.043\pm0.010$ is consistent with the value $\delta_1\leqslant0.05$ determined by McGowan and Stelson[21] by Coulomb excitation.

(c) Structure Effects in the 279-kev Transition

The value of $A_{1\gamma}$ coupled with the $A(\gamma-e)$ measurement determines $A_{2e}=-0.34\pm0.10$. Figure 6 is a plot of A_{2e} as a function of δ_2 in the region of interest for a few values of λ. In calculating A_{2e} we have assumed that the b_e, the particle parameters appropriate to $E2$ transitions, is not dependent on λ. Since it has been found that finite size effects do not significantly affect

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[18] T. A. Green and M. E. Rose, Phys. Rev. **110**, 105 (1958).

[19] Privately circulated table via O. Nathan.

[20] Church, Rose, and Weneser, Phys. Rev. **109**, 1299 (1958). The necessary electron radial matrix elements are tabulated in M. E. Rose, *Internal Conversion Coefficients* (North-Holland Publishing Company, Amsterdam, 1958).

[21] F. K. McGowan and P. H. Stelson, Phys. Rev. **109**, 901 (1958).

[22] Deutch, Wilhelm, and Metzger (private communication).

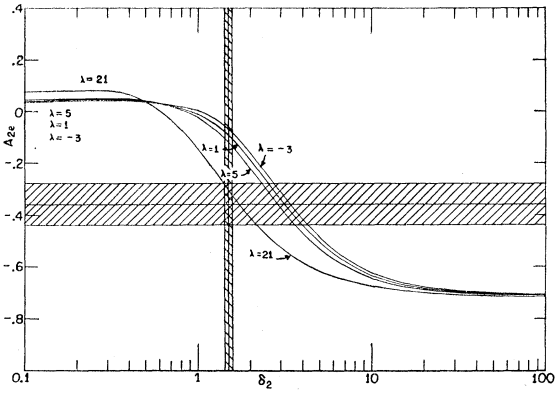

FIG. 6. A_{2e} as a function of δ_2 for various values of λ. The horizontal hatched lines show our determination of A_{2e} using our determination of $A_{1\gamma}$ and $\gamma-e$ correlation measurements.

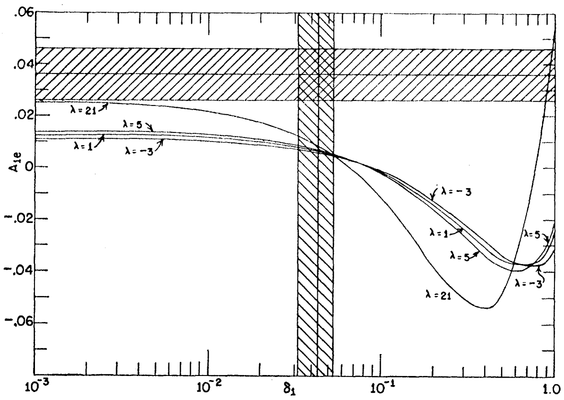

FIG. 7. A_{1e} as a function of δ_1 for various values of λ. The horizontal hatched lines show our determination of A_{1e} from the $e-\gamma$ correlation measurement and using $\delta_2=+1.50$. The vertical hatched lines show our determination of δ_1.

the $E2$ conversion coefficients, this seems to be a reasonable assumption. Figure 6 shows a very large value of λ is required to fit our data. This value disagrees with λ derived from internal conversion coefficient measurements and also the calculation by Kisslinger.[23] We can fit our data for the gamma-electron and the gamma-gamma correlation with $\delta_2=2.5$. This value is appreciably larger than other determinations of δ.[12,21,22]

(d) Structure Effects in the 404-kev Transition

Again using $\delta_2=1.5$ to determine $A_{2\gamma}$, we can use the measurement of $A(e-\gamma)$ to determine $A_{1e}=+0.036 \pm 0.010$. Figure 7 is a graph of A_{1e} as a function of δ_1 for a few values of λ. Again a very large value of λ is required to fit our results. Since the 404-kev transition is not retarded, structure effects should not be important in this transition. It should also be pointed out that the conversion coefficient measurement for this transition $\alpha_K=0.117$ is anomalously low. The Sliv value of the conversion coefficients coupled with our value $\delta_1=0.043$ predicts $\alpha_K=0.147$.

VI. DISCUSSION

It has not been possible to fit our angular correlation measurements involving conversion electrons with the theoretical values of the particle parameters. The required value of λ to fit our data is much larger than one would expect from conversion coefficient and lifetime measurements in both transitions. In both of these correlations, the anisotropy seems to be too large. One would ordinarily expect experimental errors to reduce the correlation. The K-conversion coefficient for the 404-kev transition is 20% lower than the theoretical value. Nijgh *et al.*[12,13] have also reported conversion coefficients in disagreement with theory in this isotope for the 279-kev transition.

VII. ACKNOWLEDGMENTS

The authors gratefully acknowledge many valuable suggestions from Professor Sherman Frankel. We also thank Dr. E. L. Church and Dr. J. Weneser for helpful discussions and Mr. Jose Palathingal for his assistance in calculating Table III.

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[23] L. S. Kisslinger, Bull. Am. Phys. Soc. 2, 358 (1957).