The Directional Correlation of the Pb^{204} $\gamma-\gamma$ Cascade*

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(Received August 3, 1953)

The directional correlation of the Pb^{204} $\gamma-\gamma$ cascade has been measured in chemical compounds and in a liquid Pb—Tl alloy. The values for the anisotropy of the Pb^{204} cascade in the chemical compounds vary between 0.08 ± 0.04 and 0.46 ± 0.04, thus showing a pronounced influence of extranuclear fields. The anisotropy in the liquid Pb—Tl alloy, 0.49 ± 0.05, excludes the assignment 7^- for the second excited state of Pb^{204}. The only decay scheme compatible with all experimental data requires the assignments 2^+ and 6^+ to the first and second state, respectively. The first γ transition in Pb^{204} must be a mixture of approximately 90 percent $E4$ and 10 percent $M5$.

I. INTRODUCTION

RECENT experimental[1–3] and theoretical[4–6] evidence indicates that the influence of extranuclear fields in solids and liquids on the directional correlation of nuclear γ rays is largely (or entirely) due to the interaction of the nuclear quadrupole moment with inhomogeneous electric fields. Most of the experimental work up to now has been done with the $\gamma-\gamma$ cascade of Cd^{111}. However, the fact that this cascade follows K capture in In^{111} introduces some uncertainty in the interpretation of the experiments: the electronic shell, excited after the K capture, may have properties different than those of its ground state. This difficulty does not exist in the case of isomeric transitions. Therefore, we decided to investigate the directional correlation of the $\gamma-\gamma$ cascade in Pb^{204}.

Pb^{204} has been carefully studied by the Brookhaven group.[7] They found that the isomeric transition takes place in two steps (γ_1: $E=0.905$ Mev, $T_{\frac{1}{2}}=65$ min; γ_2: $E=0.374$ Mev, $T_{\frac{1}{2}}=3\times10^{-7}$ sec), and measured the conversion coefficients of the two successive γ rays. Moreover, they determined a value $A=0.22\pm0.05$ for the anisotropy $A\equiv[W(180°)/W(90°)]-1$ of the directional correlation function $W(\theta)$. Comparison of the conversion coefficients and the half-lives of the two γ rays with theory led to the identification of γ_1 as $E5$ and γ_2 as $E2$. The first two excited states of Pb^{204} therefore were given the assignments 2^+ and 7^-.[8] The theoretical anisotropy for the resulting cascade $7(E5)2(E2)0$, $A_{\text{theor}}=0.407$, need not be in disagreement with the experimental value 0.22, because extranuclear fields can attenuate the theoretical correlation.

The data of the Brookhaven group show that Pb^{204} has many excellent properties for an investigation of the effect of extranuclear fields on the nuclear directional correlation. The decay scheme is simple, the γ-ray energies are suitable for work with scintillation counters, and the half-life of the intermediate state (3×10^{-7} sec) is long enough to expect strong perturbations. The only experimental difficulty is the short half-life (65 min) of the initial isomeric state. All measurements must be made in a relatively short time, and no involved source preparation is possible.

Despite the fact that no K capture or beta decay complicates the problems, we found that the influence of the extranuclear fields on the correlation of Pb^{204} is not as simple as we expected. The present paper is only a preliminary report and contains no conclusions as to the nature of the interactions between the nucleus and its surroundings. We shall show, however, that the directional correlation in some of the present measurements is really undisturbed, and we provide consistent spin and parity assignments for the Pb^{204} levels. Further investigations of the influence of extranuclear fields can therefore be based on a unique decay scheme and a knowledge of the highest possible anisotropy.

II. APPARATUS AND CORRECTIONS

The isomeric state in Pb^{204} was produced by irradiating metallic thallium with 12-Mev deuterons. Unfortunately, such a bombardment produces simultaneously, and with much greater intensity, the 52-hr Pb^{203}. A counter system for the determination of the directional correlation of Pb^{204} has therefore to be so designed that it discriminates effectively against coincidences from Pb^{203}.

We measured the directional correlation of Pb^{204} with NaI scintillation counters.[9] The desired discrimination against the Pb^{203} radiation was achieved by using suitable front absorbers and discriminator levels, and by inserting a delay in channel I of the circuit. The relevant counter data are given in Table I. All coincidences due to scattered radiation and to Pb^{203} were reduced by a factor of more than 10^3.

* Assisted by the joint program of the U. S. Office of Naval Research and the U. S. Atomic Energy Commission.

[1] Albers-Schönberg, Hänni, Heer, Novey, and Scherrer, Phys. Rev. **90**, 322 (1953).

[2] Albers-Schönberg, Heer, Novey, and Rüetschi, Phys. Rev. **91**, 199 (1953).

[3] R. M. Steffen, Phys. Rev. **90**, 1119 (1953).

[4] A. Abragam and R. V. Pound, Phys. Rev. **89**, 1306 (1953).

[5] R. V. Pound and A. Abragam, Phys. Rev. **90**, 993 (1953).

[6] A. Abragam and R. V. Pound, Phys. Rev. **92**, 943 (1953).

[7] Sunyar, Alburger, Friedlander, Goldhaber, and Scharff-Goldhaber, Phys. Rev. **79**, 181 (1950).

[8] M. Goldhaber and R. D. Hill, Revs. Modern Phys. **24**, 230 (1952).

[9] J. S. Lawson, Jr., and H. Frauenfelder, Phys. Rev. **91**, 649 (1953).

TABLE I. Characteristics of the counting system.[a]

Counter	α_0	Lead shielding lateral	Lead shielding front	Discriminator level	Delay	Resolving time
I	25°	1.5 cm	0.5 cm	0.55 Mev	0.432 μsec	
II	25°	1.5 cm	0.2 cm	0.22 Mev	none	0.310 μsec

[a] α_0 denotes the width of the counter efficiency curve at half-maximum.[9]

The measured anisotropy was corrected for finite angular resolution of the counters and for scattering in the source. The first correction was straightforward.[9] The correction for scattering of the γ rays in the source, however, presented a serious difficulty. Sources thin enough to make the scattering correction small have two disadvantages: the activity is extremely low, and possible surface effects (e.g., in solids: oxidation and field gradients at the boundaries; in liquids: adsorption at the wall) are relatively more important.

We therefore measured the anisotropy in sources of different diameters and extrapolated to zero thickness by the following semiempirical method: We calculated for each source the probability w_1w_2 that a genetically related γ-ray pair, $\gamma_1-\gamma_2$, escape from the source without scattering. The measured anisotropy A_m for each source was then plotted against the corresponding value of w_1w_2. A simple calculation shows that for "thin" sources, A_m should vary linearly with w_1w_2. (A "thin" source means that the probability for single scattering of the γ rays is much larger than that for multiple scattering.) Therefore a straight line was fitted to the points and extrapolated to $w_1w_2=1$ to obtain the anisotropy for zero-source thickness.

In order to test the method experimentally, and to find its limits of applicability, we measured the anisotropy of a very small Ni^{60} source (linear dimensions <0.05 cm) surrounded by cylindrical lead absorbers of different thicknesses. These results are shown in the lower part of Fig. 1. In the region $w_1w_2\sim0.4$ to 0.8, the measured values *can* be fitted by a straight line. The extrapolation to $w_1w_2=1$ then gives a result $A=0.175$ which is only slightly higher than the correct value 0.167. An overestimation of the correction is reasonable: the multiple scattering of the γ rays, which increases with decreasing w_1w_2, tends to enhance the "smearing-out" effect, and bends the measured curve towards the w_1w_2-axis. We expect our correction method to work better for cascades with lower γ-ray energies than Ni^{60}, e.g., Pb^{204}: at high γ-ray energies, the scattering occurs predominantly in the forward direction, and the multiple scattering therefore is more important in averaging out the correlation than is the single scattering.

TABLE II. Anisotropy A of Pb^{204} and Cd^{111} in various compounds.

Compound	Solvent	Anisotropy $A=\frac{W(180°)}{W(90°)}-1$ $T=-185$°C	$T=+25$°C	Number of independent measurements
$Pb(NO_3)_2$	HNO_3 (1*N* − 16*N*)	0.08 ± 0.04	0.30 ± 0.03	5
		0.22 ± 0.03	0.42 ± 0.03	3
$Pb(CH_3CO_2)_2$	NH_4 Acetate, sat.	0.10 ± 0.05	0.38 ± 0.03	2
$Pb(HSO_4)_2$	H_2SO_4 36*N*		0.23 ± 0.05	1
			0.44 ± 0.03	1
H_2PbCl_6	HCl 12*N*		0.46 ± 0.04	4
$In_2(SO_4)_3$	H_2SO_4 36*N*	−0.04 ± 0.02	−0.10 ± 0.01	2
	32*N*	−0.04 ± 0.02	−0.15 ± 0.01	1
	18*N*	−0.03 ± 0.01	−0.21 ± 0.01	2
	1*N*	−0.03 ± 0.01	−0.20 ± 0.01	3
	aqueous	−0.02 ± 0.02	−0.20 ± 0.01	1
$InCl_3$	HCl 12*N*	−0.04 ± 0.01	−0.21 ± 0.01	1
	1*N*	−0.02 ± 0.01	−0.19 ± 0.01	1

III. MEASUREMENTS WITH CHEMICAL COMPOUNDS

The object of the first set of measurements was to observe the directional correlation of Pb^{204} in various liquid sources at several temperatures. We hoped to get, in analogy with the case of Cd^{111}, the highest anisotropy in the liquid state.[3,10,11] Furthermore, we expected to find direct evidence for the influence of extranuclear fields by cooling the specimens down to liquid nitrogen temperature and thereby solidifying them. These experiments were carried out with several Pb compounds in various solvents.

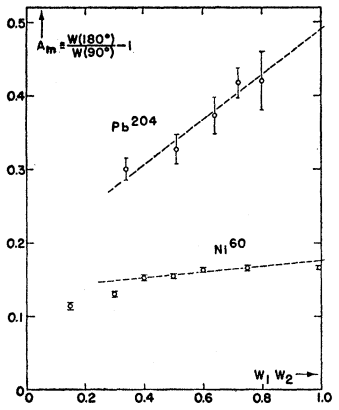

FIG. 1. Measured anisotropy A_m as a function of source thickness (w_1w_2) for Pb^{204} and Ni^{60}.

The Pb^{204} sources were produced by irradiating metallic thallium with deuterons in the cyclotron. The radioactive Pb^{204} and the simultaneously produced 52-hr Pb^{203} were chemically separated from the Tl by extracting the Tl as $TlCl_3$ with ether from a 4*N* HCl solution. The desired Pb compound was then synthesized and sealed in a thin-walled Pyrex tube. We also investigated Cd^{111}. Typical results are summarized in Table II.

Cd^{111} shows the expected behavior: the solid (frozen) state displays a markedly reduced correlation as compared with that in the liquid state. All measurements at −185°C are consistent with a value $A=-0.031\pm0.005$. The liquid sources yield, in accordance with earlier investigations,[3,10,11] a value of the anisotropy close to the value $A=-0.23$.[1] Only $In_2(SO_4)_3$ in con-

[10] J. C. Kluyver and M. Deutsch, Phys. Rev. **87**, 203 (1952).
[11] R. M. Steffen, Phys. Rev. **89**, 903 (1953).

centrated H_2SO_4 deviates strongly from the maximum value.

Pb^{204} also exhibits a large anisotropy in the liquid state and considerably reduced values in the solid state. However, sources which are expected to give the same results have widely differing anisotropies. This fact is especially visible in the cases of $Pb(NO_3)_2$ and $Pb(HSO_4)_2$. Only H_2PbCl_6, where the Pb is tetravalent, shows consistently high values. This may be due to the fact that the Pb in $(PbCl_6)^=$ has coordination number 6, and is octahedrally surrounded by the 6 Cl atoms. In this case the electric field gradient at the side of the Pb nucleus should be small.

In both series of measurements (Pb^{204} and Cd^{111}), heating the liquids to 90° did not increase the anisotropy above its value at 25°C.

IV. MEASUREMENTS WITH A LIQUID LEAD-THALLIUM ALLOY

Table II shows that the behavior of the Pb^{204} correlation function in liquids is rather erratic. We therefore

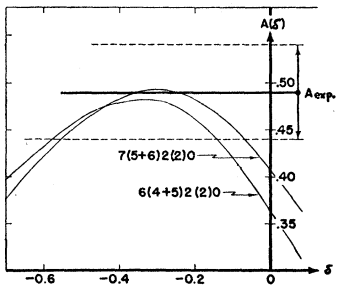

FIG. 2. Theoretical anisotropy $A(\delta)$ as a function of the mixing ratio between competing multipoles, and experimental anisotropy A_{exp}.

sought sources in which the influence of crystalline fields might be controlled, or made vanishingly small.

Lead single crystals (face-centered cubic) or lead melt[2] would constitute a good choice, but experimental difficulties prevent their use: after production in the cyclotron, the Pb^{204} is dispersed in metallic thallium. The chemical separation of the Pb from Tl and the transformation back into its metallic state then invariably yields very weak sources.

Our sources for this experiment therefore were prepared directly from the target material. The irradiated thallium was carefully melted and cleaned in vacuum and sealed into a Pyrex glass tube. Such a source can be regarded as a Pb-Tl mixed crystal[12,13] with vanishingly small Pb content. Because Pb-Tl mixed crystals with high Tl concentration possess, below the melting point, a body-centered cubic phase,[14] we do not expect appreciable electric field gradients at the lattice sites. However, we considered liquid Tl to be even better suited for the present work: in the molten state, metals retain their structure in small regions.[15] The Pb^{204} atoms therefore, as in the cubic phase, would experience only small field gradients in most sites. Moreover, the rapid Brownian motion in the melt would probably average out any residual local field gradients, which could arise even in the cubic phase from lattice imperfections and strains.

TABLE III. Lifetime and conversion data for the γ rays of Pb^{204}.

γ	Assignment	K-shell conversion coefficient exptl[a]	K-shell conversion coefficient theor	γ half-life (sec) exptl[a]	γ half-life (sec) theor[b]
γ_1	$E5$	0.06	0.06	4.3×10^{3}	1×10^{4}
γ_2	$E2$	0.034	0.040	3×10^{-7}	8×10^{-10}

[a] See reference 7.
[b] See reference 16.

All measurements were therefore made with the source at 390°C. The final results include only those sources which showed a mirrorlike surface and which did not change in appearance during the experiment. (In two cases, the Pyrex tube leaked during the measurement. The process of oxidation caused a continuous decrease in anisotropy, which was observed over a period of two hours, as well as a visible change in the appearance of the material.)

In order to apply our correction method for scattering of the γ rays in the source, we measured the anisotropy of sources with different diameters. The results are shown in the upper part of Fig. 1. The extrapolated anisotropy for the Pb^{204} $\gamma-\gamma$ cascade is $A=0.49\pm0.05$. This value is in good agreement with the anisotropy $A=0.46\pm0.04$, reported for H_2PbCl_6 in Table II.

V. THE DECAY OF Pb^{204}

As pointed out in the introduction, the first and second excited state of Pb^{204} have been assigned spin and parity 2^+ and 7^-, respectively.[8] These assignments have been made on the basis of lifetime and conversion coefficient considerations for the two transitions $7^-(\gamma_1)2^+$ and $2^+(\gamma_2)0^+$. The relevant experimental[7] and theoretical[16] data are given in Table III. They agree especially well for γ_1.

However, our experimentally determined anisotropy 0.49 ± 0.05 for the directional correlation between γ_1 and γ_2 is higher than the theoretical value 0.407 for the cascade 7(5)2(2)0. We therefore looked for a possible modification in the decay scheme of Pb^{204} which could explain the measured anisotropy.

The assignment 2^+ for the first excited state cannot be altered: the measured conversion coefficient and the K/L conversion ratio of γ_2 (which, as a transition to the ground state 0, must be pure), exclude any transition except $E2$. Moreover, a state 2^+ agrees with the Gold-

[12] W. Hückel, *Structural Chemistry of Inorganic Compounds* (Elsevier Publishing Company, Inc., New York, 1950), p. 703.
[13] Y. C. Tang and L. Pauling, Acta Cryst. **5**, 39 (1952).
[14] H. Lipson and A. R. Stokes, Nature **148**, 437 (1941).
[15] Reference 12, p. 851.
[16] J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952), p. 627.

TABLE IV. Mixing ratios and conversion coefficients.

Second excited state Spin	Parity	First transition mixture	Mixing ratios δ_{min}	δ_{max}	K-shell conversion coefficient theoretical for δ_{min}	δ_{max}	exptl
6	+	$E4+M5$	−0.15	−0.58	0.031	0.096	0.06
	−	$M4+E5$			0.17	0.14	
7	+	$M5+E6$	−0.07	−0.55	0.32	0.27	
	−	$E5+M6$			0.06	0.17	

haber-Sunyar rule.[17,18] The fact that the experimental half-life is longer than the theoretical one is also not unexpected for an electric transition.[16]

If we restrict ourselves to pure multipole radiations for the first transition also, then no possible spin assignment for the second excited state yields a large enough anisotropy. This forces us to assume that the first transition γ_1 is mixed.

Even if we admit a mixed transition, the spin of the second excited state is restricted to 6 or 7. A spin value of 5 (or less) is excluded by the fact that the half-life of γ_1 is $\sim 10^7$ times longer than what one expects for a transition with spin change 3.[17] A spin of 8 (or more) would require a transition at least 10^6 times faster than a single proton transition,[16] and a conversion coefficient larger than the experimentally determined value.

We therefore assume a spin of 6 or 7 for the second excited state. The mixed transition to the first excited state 2^+ then can proceed with or without parity change, and we are left with the four possibilities listed in Table IV. In order to decide among these four cascades and to determine the mixing ratio δ of the competing multipoles, we have calculated the anisotropy A as a function of δ.[19] [δ^2 is defined as the intensity ratio of the $(L+1)$-pole radiation to the L-pole radiation.] The resulting curves $A(\delta)$ and the measured anisotropy A_{exp} are plotted in Fig. 2.

We have stated above that the spins 6 and 7 for the second excited state are the only values compatible with lifetime and conversion considerations. Hence, the anisotropy of the Pb^{204} $\gamma-\gamma$ cascade cannot assume values larger than those given by the two theoretical curves in Fig. 2. However, our experimental value of the anisotropy in a Pb-Tl melt lies approximately on the maximum of the theoretical curves. Therefore the directional correlation is not appreciably disturbed by extranuclear fields.

The lower limit of the experimental anisotropy determines the extreme mixing ratios δ_{min} and δ_{max} (see Fig. 2). These mixing ratios and the K conversion coefficients calculated from them are given in Table IV.

A comparison of the experimental with the theoretical conversion coefficients rules out the mixtures $M4+E5$ and $M5+E6$.

The decision between the two remaining possibilities has to be made on the basis of lifetime estimates. In Table V we compare the experimental partial half-lives for an intermediate value of δ ($\delta=0.3$) with the corresponding theoretical values for a single proton transition.

These data induce us to abandon the $E5+M6$ cascade for the following reason. Single-particle estimates constitute lower limits for the half-lives. In the case $E5+M6$, however, the observed magnetic transition would be faster than the theoretical estimate by a factor of $\sim 10^7$, whereas the corresponding factor in the first alternative, $E4+M5$, would be only ~ 20. Ratios of the latter order of magnitude have been found for $M4$ transitions[17,20] but no factor of the order of 10^7 has ever been observed. A mixture of $E4$ and $M5$ explains also the measured conversion coefficient. (The excellent agreement in Table III between theoretical and experimental data for the first transition seems to have been only an accident.)

Taking all information together, we conclude that the second excited level in Pb^{204} is a 6^+ state, and the first transition γ_1 is a mixture of approximately 90 percent $E4$ and 10 percent $M5$. (A similar mixture between $E3$ and $M4$ has recently been found by Hill and Mihelich.[20]) The transition probabilities for the $E4$ in the first, and the $E2$ in the second transition are reduced, compared with those for a single proton, by factors of 1×10^{-5} and 2×10^{-3}, respectively. This reduction is very probably due to the fact that the protons in Pb^{204} form a closed shell. It is also interesting to note that Moszkowski's calculation[21] for the transition probability of a single neutron yields values of this order of magnitude.

TABLE V. Experimental and theoretical half-lives for a mixed transition.

Mixture	Exptl partial half-life (sec) E	M	Theor half-life[a] (sec) E	M
$E4+M5$	5×10^3	5×10^4	7×10^{-2}	1×10^6
$E5+M6$	5×10^3	5×10^4	1×10^4	4×10^{11}

[a] See reference 16.

VI. ACKNOWLEDGMENTS

We thank Professors J. M. Blatt, F. Coester, M. Gell-Mann, H. S. Gutowsky, F. E. Low, D. Pines, C. P. Slichter, and Dr. R. Norberg for valuable discussions, Mr. D. H. Fraembs for his continuous help during measurements and evaluation, and Mr. W. H. Rosenfeld for help with the calculations.

[17] M. Goldhaber and A. W. Sunyar, Phys. Rev. **83**, 906 (1951).
[18] G. Scharff-Goldhaber, Phys. Rev. **90**, 587 (1953).
[19] L. C. Biedenharn and M. E. Rose, Revs. Modern Phys. **25**, 729 (1953).
[20] R. D. Hill and J. W. Mihelich, Phys. Rev. **89**, 323 (1953).
[21] S. A. Moszkowski, Phys. Rev. **89**, 474 (1953).