The absolute values of Δ' and $\bar{w}/w_\alpha$ for argon plus CO_2 (3%) given in Table II agree with the earlier determination[5] based on the ionization measurements of Brunton and Hanna.

Note added in proof.—Note from Table II that for each gas, the defects Δ and Δ' are approximately the same for light fragments. Consequently, the value of w for these high-velocity particles is very nearly the same as that for alpha particles. This is in accord with the expectation[1] that w is insensitive to the mass and charge of fast particles. In the case of heavy fragments, however, the results show that Δ' is greater than Δ for each gas; thus for heavy fragments we have $w>w_\alpha$. It therefor appears[8] that at the velocities used in the light-fragment measurements no appreciable energy is lost in nuclear recoils; while at the generally lower velocities of the heavy fragments, the energy lost in nuclear recoils is significant and increases with decreasing velocity. The importance of nuclear collisions even at these velocities, $v \gtrsim 2v_0$, where $v_0=e^2/\hbar$, is a consequence[9] of the large nuclear charge of fission fragments.

[8] Suggested by J. Lindhard (private communication, 1955).
[9] N. Bohr, Kgl. Danske Videnskab. Selskab, Mat. fys. Medd **18**, No. 8 (1948).

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Time-Dependent Directional Correlation of 1.1-hr Pb^{204}†

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A study is made of an effect attributable to the interaction of the electric quadrupole moment of the 0.27 μsec level of Pb^{204} with the crystalline electric field in metallic, polycrystalline, thallium in which 68-min Pb^{204} is produced by $(p,2n)$ reaction. The directional anisotropy in the rate of delayed coincidences between the γ ray preceding the 0.27 μsec state and the γ rays following the state is observed as a function of the delay time. Although some discrepancy between the observed function and theory is found, the time dependence suggests a coupling frequency $eQ(\partial^2V/\partial z^2)/h$ of 12 Mc/sec. Measurements of the directional correlation between the two gamma rays in prompt coincidence following the 0.27 μsec state supports the assignment, by Krohn and Raboy, of spin 4 to that state and of some mixing of $M3$ with the dominantly $E2$, 371-kev gamma ray. Evidence of prompt γ rays of several energies, following K-capture in 12-hr Bi^{204}, is also found.

INTRODUCTION

IT has been shown by Abragam and Pound[1] that the interaction of the nuclear quadrupole moment with the gradient of a static electric field results in a periodic attenuation of the coefficients of the directional correlation function of two successively emitted nuclear radiations, provided that the interacting field has an axis of at least threefold symmetry. The result is particularly simple in a source composed of a large number of randomly oriented microcrystals, where it takes the form

$$W(\theta,t)=1+\sum_\nu G_\nu(\omega_0 t)A_\nu P_\nu(\cos\theta). \quad (1)$$

The coefficients A_ν are those appropriate to the decay scheme in the absence of a disturbance and the forms of the time-dependent attenuation functions $G_\nu(\omega_0 t)$ are determined by the spin of the intermediate state. The dimensionless parameter $\omega_0 t$ is proportional to the product of the time t, measured from the first decay, and the strength ω_0 of the quadrupole interaction.

Such an effect is observable if the period characteristic of the interaction is not much longer than the half-life of the disturbed intermediate state. On the other hand, the period of the interaction must be long compared with the minimum resolving time attainable in order that the structure of the attenuation function be well resolved. Present instrumental techniques do not allow a resolving time, with energy selection, less than about 5 millimicroseconds, so that a half-life of at least 50 millimicroseconds is essential. The magnitude of ω_0 determining the period of the attenuation function, $G(\omega_0 t)$, is not known *a priori*, but may be estimated from values of electric quadrupole interactions of stable nuclei in various compounds observed by magnetic and pure quadrupole resonance.

The known nuclei suited to such studies are few. Lead-204 has a level of half-life of 0.27 microsecond, that could allow good resolution of an attenuation function. However, having a closed proton shell and an even number of neutrons, its quadrupole moment is expected to be small and, therefore, the interaction with an extranuclear field gradient might be too weak to exhibit a complete period of the attenuation function, even in the relatively long half-life.

An experiment of this kind in 48-min Cd^{111} has

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* Now at Bell Telephone Laboratories, Murray Hill, New Jersey.
[1] A. Abragam and R. V. Pound, Phys. Rev. **92**, 943 (1953).

recently been reported by Lehman and Miller.[2] Three periods of the attenuation function were observed within two half-lives. The resolution was insufficient to show very detailed structure of the function, since only two or three points were obtained per period.

Measurement of the attenuation function, in principle, allows determination of the spin of the disturbed state since the function has a characteristic detailed shape for each spin value. Its full period is $\tau=2\pi/\omega_0$. Because of a desire to have the nuclei studied in a chemically stable environment, we first worked with 12-hr Bi^{204} produced by proton bombardment of lead. The bismuth was expected to decay by electron capture into the 68-minute isomeric state of Pb^{204}, the 68-minute half-life allowing sufficient time for the lead to come to equilibrium following the disturbance of electron capture. In this way the radiating lead would have been situated in a lead lattice. Unfortunately metallic lead crystallizes in a cubic lattice and some chemical processing would have been required to obtain a crystalline field gradient. Work with sources so produced was abandoned, however, for additional reasons related to the decay scheme, discussed in a later section.

INSTRUMENTATION

The equipment used[3] follows the conventional slow-fast coincidence scheme in which the timing and energy selection functions are separated at the photomultipliers and eventually recombined in a triple coincidence circuit.

Thallium activated sodium iodide crystals ($1\frac{1}{2}$ in. by 1 in.) mounted on RCA 6342 photomultipliers are used. The fast, or timing, channels consist of three Hewlett-Packard distributed amplifiers each, followed by an EFP 60 secondary emission pentode limiter feeding a 6BN6 coincidence detector. Pulse lengths are determined by shorted stub lines, and variable delay may be introduced as cable between amplifier stages, before the limiters. The energy selection channels consist of commercial linear amplifiers followed by single channel, pulse-height analyzers modified for fixed time delay. The modification uses a 6BN6 as an anticoincidence tube and a delay of 0.40 microsecond determined by cable.

To accommodate high counting rates, the triple coincidence resolving time is also 0.40 microsecond, generally requiring that delay be introduced in both slow and fast channels.

Data recording and repositioning of the movable scintillation detector are carried out automatically at the end of a preset interval. The number of triple coincidences, fast coincidences, and the counts in the two analyzers are presented on glow transfer tube registers and photographed at the end of each timing cycle. The fast coincidences, although not needed in the treatment of data, offer an important monitor of the stability of the resolving time of the fast coincidence circuit.

PROCEDURE

Lead-204*m* was prepared by bombardment of thallium metal with protons in the internal beam of the Harvard synchrocyclotron. Alternatively Bi^{204} could be prepared by proton bombardment of lead, the Pb^{204m} daughter separated periodically by chemical means and subsequently reduced to metallic lead. The difficulties in the latter method are numerous, the total time required for separation, purification, and reduction being at least one hour. Electrolytic reduction was found to be slower and generally yielded a spongy deposit which oxidized readily.

The isotopic abundance in natural thallium is 70% Tl^{205} and 30% Tl^{203}. At 21 Mev the favored $(p,2n)$ reaction produces 68-min Pb^{204} and 3.5-hour Pb^{202}. Some Pb^{203} is also produced. The decay schemes of the contaminants are such as to produce no interference with delayed coincidence measurements. Examination of the spectrum of 0.003-inch thick targets bombarded for 10 minutes and aged for 10 half-lives of Pb^{204m} allowed identification of the gamma rays of Pb^{202}, both by their energy and their coincidences with respect to the 963-kev line. A further check was made to show that the half-lives of the intermediate levels are less than 1 millimicrosecond. The 4^+ level, which is perhaps analogous to the 0.27-microsecond level in Pb^{204}, was examined with particular care. Pb^{203} decays by electron capture into 279-kev (95%) and 679-kev (5%) levels of Tl^{203}, according to Wapstra.[4] The 2.3-day half-life makes the contribution of Tl^{203} to the spectrum negligible.

Considering the isotopic abundance and the half-lives, the expected ratio of activity of Pb^{204} to Pb^{202} is 7 to 1 immediately following bombardment. Scintillation spectra obtained during the decay of a target show that the actual ratio is considerably higher, perhaps 15 to 1, indicative of a difference in the cross sections of Tl^{203} and Tl^{205} for a $(p,2n)$ reaction at 21 Mev.

Neither of these isotopes, therefore, interferes with measurements of the delayed cascades in Pb^{204}. Measurements at zero delay, however, are possible only for the 910–895 kev cascade. Even here, the 778–963 kev cascade in Pb^{202} may provide some interference. The 371–895 kev prompt cascade in Pb^{204} interferes with measurement of its own delayed 910–371 kev cascade.

The anisotropy function was measured with a resolving time $\tau=16$ millimicroseconds and a source-to-counter distance of 4.5 cm. The strong attenuation of the coefficient A_4 resulting from such close spacing is not entirely undesirable since it allows a better determination of G_2 to be made directly from the anisotropy. The ratio of true to accidental coincidence counts was as low as 0.2. The source in the form of a crumpled

[2] P. Lehman and J. Miller, Compt. rend. **240**, 298 (1955).
[3] G. K. Wertheim, thesis, Harvard (unpublished).
[4] A. H. Wapstra *et al.*, Physica **20**, 169 (1954).

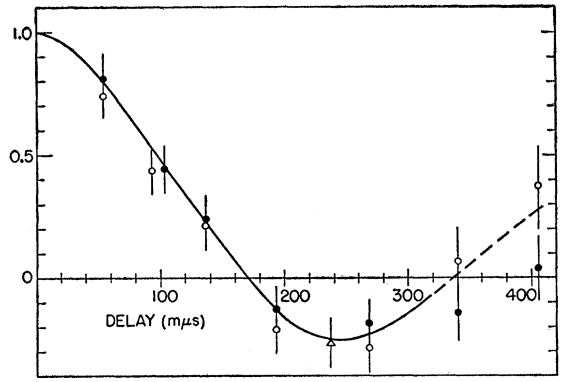

FIG. 1. The attenuation function $G_2(t)$ *vs* coincidence delay time in millimicroseconds. The solid points refer to the 910–895 kev coincidences, the open circles to 910–371 kev and the triangular point to the recrystallized source.

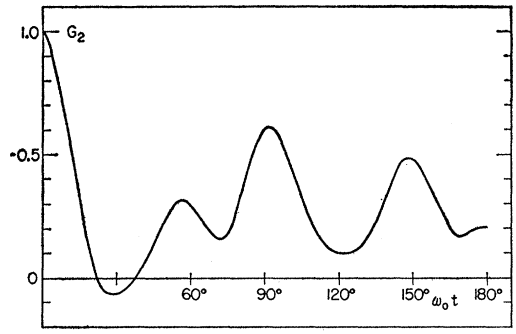

FIG. 2. The theoretical function $G_2(\omega_0 t)$ for $I=4$.

metallic foil was contained in a thin-walled glass tube centered to within 0.1 cm with respect to the activity of the sample. Nonuniform distribution of activity in the target foil makes more accurate positioning difficult. Only counts of pulse heights corresponding to the photopeaks of the two desired gamma rays, as detected in the scintillation spectrometers, were accepted.

DISCUSSION

The time-dependent attenuation function $G_2(t)$ found for the 0.27-microsecond level of Pb^{204} using both the 910–895 and the 910–371 kev cascades is given in Fig. 1. In both cases the function has been determined from the anisotropy $[W(180°)/W(90°)]-1$ under the assumption that A_4 is negligible. The value unity at zero delay corresponds to the relevant anisotropy using the correlation functions reported by Krohn and Raboy[5] for liquid sources. The points from the two cascades agree, within the experimental error, with a single curve. Theory indicates that the attenuations should be the same for the two coincidence schemes except for the effect of internal conversion of the unobserved 371-kev γ ray in the former case and provided that there is no disturbance of the spin 2 level. The fraction of coincidences for which internal conversion occurs is small ($\sim 5\%$). The life of the spin 2 level is certainly less than 6×10^{-10} sec,[6] making the disturbance negligible, if the electric quadrupole moment is not much larger than in the 0.27 μsec level.

Comparison of the experimental curve with the theoretical function for spin 4 (Fig. 2) indicates one discrepancy. The experimental curve goes strongly negative for delays in the interval from 170 to 340 millimicroseconds, while the theoretical curve is relatively less negative for a time less than the initial positive interval. This is also true of the theoretical functions for other possible integral spin assignments. The assignment of this cascade to Pb^{204} is considered to be too well established to justify comparison with the attenuation functions for half odd-integral spin.

The discrepancy cannot be attributed to the after-effects of proton bombardment, even though the curves shown were obtained with material counted without further treatment following irradiation. One target was melted in vacuum and suddenly chilled to prevent the formation of a single crystal, deformed slightly and cut into small pieces as further precautions. It was counted at a delay of 0.236 microsecond corresponding to the minimum of the time-dependent attenuation function previously obtained for the untreated material, and yielded an anisotropy in good agreement with the other data, Fig. 1. This result is not unusual from the point of view of the known properties of metals, which suggest that at temperatures above one-half of the absolute melting point, lattice defects such as those produced by bombardment tend to anneal rapidly.

Although an additive negative anisotropy caused by source absorption could explain the effect, it is considered unlikely that the relatively large negative anisotropy could be so produced in view of the fact that the sources were so small compared to a scattering length. A satisfactory explanation for the discrepancy has not been found.

The frequency characteristic of the interaction may be estimated on the basis of the measured coefficients, ignoring the discrepancy in the negative excursion. If one assumes that the interaction is otherwise similar in nature to that predicted for spin 4, only a small part of the period is accessible and a full period of approximately 3 microseconds is indicated, corresponding to a frequency of 12 Mc/sec for $eQ(\partial^2 V/\partial z^2)/h$. This value may be compared to a frequency of 669 Mc/sec for Bi^{209} in the form of $Bi(C_6H_5)_3$[7] and 708 Mc/sec for Hg^{201} in the form of $HgCl_2$.[8] The low frequency obtained for Pb^{204} compared to those for adjacent isotopes, although not directly comparable because the lead is measured in a metallic state, seems to support the expected smallness of its quadrupole moment, in general agreement

[5] V. E. Krohn and S. Raboy, Phys. Rev. **97**, 1017 (1955).
[6] A. W. Sunyar, Phys. Rev. **98**, 653 (1955).
[7] Robinson, Dehmelt, and Gordy, Phys. Rev. **89**, 1305 (1953).
[8] Dehmelt, Robinson, and Gordy, Phys. Rev. **93**, 480 (1954).

TABLE I. Coefficients and mixing for the (371–895)-kev cascade in Pb^{204}.

4(E2)2(E2)0	A_2	A_4	δ
Basic, pure	0.102	0.009	0
Krohn and Raboy (computed)	0.142	0.000	-0.07 ± 0.02
Measured	0.129 ± 0.006	0.016 ± 0.016	-0.05 ± 0.01

with the systematics of nuclear quadrupole moments near closed shells.

SPIN OF THE 0.27-MICROSECOND LEVEL OF Pb^{204}

In view of the discrepancy between the theoretical and the measured time-dependent attenuation coefficient, $G_2(t)$, an independent check of the spin assignment was made by measuring the directional correlation of the 371–895 kev prompt cascade. The correlations of the two delayed cascades starting from the 68-minute level have been used by Krohn and Raboy[5] to assign multipole mixing to both the $E2$ and $E5$ transitions. The mixing parameter for the $E2$ transition may then be used to compute the coefficients of the $4(E2)2(E2)0$ cascade.

Direct measurement of the 371–895 kev cascade is complicated by the prompt 963–421-kev cascade in Pb^{202}, present in our targets to one part in 15. Correction for the contribution of the delayed 910–371 kev cascade in Pb^{204} is readily kept small by using a 6 millimicrosecond resolving time. The anisotropy of the interfering cascade in Pb^{202} has been independently measured in targets prepared in the same manner but allowed to age for 10 half-lives of Pb^{204} and was found to be 0.21 by Rebka.[9] Since this anisotropy is similar in magnitude to that obtained for Pb^{204} the error produced by the inclusion of some coincidences from it in the measured correlation is greatly reduced. Narrow window energy selection was used to minimize this source of error, and the data were corrected only for the contribution from the delayed cascade in Pb^{204}.

The results are compared to the computed values of Krohn and Raboy in Table I, and indicate agreement within the experimental error. The errors shown for our measurements are those of statistical origin only. The mixing parameter obtained from the fast cascade is -0.05 ± 0.01 which compares with -0.07 ± 0.02 obtained by Krohn and Raboy from the delayed cascade. The directional correlation function obtained is not compatible with an unmixed, basic 4, 2, 0 cascade, nor with 3, 2, 0 which has a negative A_2. We conclude that measurements on the prompt cascade support the assignment of spin 4 as well as mixing of the 371-kev gamma ray.

SOME REMARKS ON THE DECAY OF BISMUTH-204

In connection with the above experiment, the 68-minute isomer of Pb^{204} was prepared in equilibrium with its 12-hour Bi^{204} parent, yielding a more convenient source for extended coincidence counting. It was found, however, that the decay of Bi^{204} is followed by a complicated cascade in lead which, to a large extent, bypasses the 68-minute level but not the 0.27-microsecond level. The possibility of disturbance of the directional correlation of the prompt cascades following electron capture and the inability to select the desired γ ray uniquely confuse the use of this source for the delayed coincidence experiments. This is especially true because a dominant gamma ray of 981 kev leading into the 0.27-microsecond level is found.

Sources of this type were prepared by proton bombardment of lead and radio lead. A limited exploration of the energy dependence of the $(p,3n)$ reaction in lead was undertaken in the vicinity of the energy suggested by similar results for Bi^{209}.[10] The radio lead had a composition of 0.06% Pb^{204}, 88.2% Pb^{206}, 8.8% Pb^{207}, and 2.9% Pb^{208} as well as traces of other members of the decay chain.[11] Traces of normal lead are included in this analysis. The targets were 0.003-inch lead foils, corresponding to a proton energy loss of approximately 1 Mev in penetrating the target. Optimum production of Bi^{204} was obtained at 31 Mev.

Estimates indicate that Bi^{204} activity must dominate immediately following bombardment of either the lead or the radio lead, while the radio-lead targets should remain relatively free of other activity for 4 or 5 half-lives of Bi^{204}. These conclusions are in essential agreement with the experiment, provided that a large number of gamma rays not previously reported but observed here are correctly attributable to the decay of Pb^{204}.

The production of 68-minute Pb^{204m} via Bi^{204} was confirmed by a chemical separation. Lead separated from the target exhibited the known three-gamma-ray spectrum of Pb^{204m}. A second separation after two hours yielded Pb^{204m} comparable to the first in intensity, confirming that Pb^{204m} was produced from a longer-lived substance whose half-life was established to be 11.6 ± 0.02 hours. The lead fraction decayed with a 1.1-hour half-life, and exhibited the known directional correlation of the 910–895 kev cascade. Coincidence spectra confirmed the existence of a prompt (371–895)-kev cascade as well as delayed cascades of 910–371 and 910–895 kev within the limits of energy resolution of the scintillation spectrometers. The half-life of the intermediate level was also confirmed.

The presence of other gamma rays in this decay scheme was initially suspected from the interference which they produced in the directional correlations of delayed coincidences. Using photopeak energy selection, the full anisotropy could be obtained from the separated lead fraction in H_2PbCl_6 solution, while un-

[9] G. Rebka (private communication).

[10] P. Morrison, *Experimental Nuclear Physics*, edited by E. Segrè (John Wiley and Sons, Inc., New York, 1953), Vol. 2, p. 167.

[11] *Radio Elements and Accessories*, Catalogue C, Atomic Energy of Canada, Ltd., Ottawa, Canada, p. 119.

separated sources in this form gave no reproducible results. Molten metal sources, which had been shown by Frauenfelder *et al.*[12] to give the full anisotropy using 68-min Pb^{204} only, gave similar inconclusive results. The interference was subsequently shown to be caused by a gamma ray of 981 kev in a prompt cascade leading into the 0.27-microsecond level.

Energies of several dominant gamma rays have been obtained[13] from internal as well as external conversion spectra. The spectra obtained by internal conversion, but with source thickness great enough to allow some photoelectric effect in the lead, gave energies of 371, 668, 895, 910, and 982 kev. Use of a uranium converter produced energies of 372, 895, and 981 kev. The conversion spectra of other lines have not been thoroughly investigated but evidence of lines at 1730 and 2100 kev was found in the form of Compton edges in the double-focusing spectrometer. Gamma rays of approximate energies 1200, 1730, and 2100 kev have been found in the scintillation spectra in the range above 1 Mev. Delayed coincidence scintillation spectra have indicated that the 668-, 910-, 981-, 1200-, and 1730-kev gamma rays lead into the 0.27-microsecond level. Prompt coincidences have been observed between 981 and 668, 981 and 1200, and 371 and 895 kev. The half-lives of the gamma rays in the foregoing were found to be 11.6 ± 0.2 hours by observation of the whole scintillation spectrum over several half lives. The observed similarity of the scintillation spectra of bombarded lead and radio lead is consistent with the interpretation ascribing these gamma rays to Pb^{204}.

A second 12-hour bismuth isotope, Bi^{203}, does not appear to provide any interference. This presumably decays by electron capture into Pb^{203} which in turn decays with a 2.3-day half-life by electron capture into Tl^{203}, which emits 95% 279-kev and 5% 400- and 679-kev radiation. Thus there is an initial growth of the dominant 279-kev line, which has been observed after five Pb^{204} half-lives. Bombardment of radio-lead at higher energy, so as to favor the production of Bi^{203}, did not produce any additional gamma rays.

A chemical separation has shown that no more than 25% of the gamma rays going into the 0.27-microsecond level come from the 68-minute level in unseparated targets. This is in reasonable agreement with an earlier study by Templeton *et al.*[14] in which it was concluded that only 4% of the decays of Bi^{204} produce 1.1-hr Pb^{204m}.

We tentatively assumed a decay scheme as in Fig. 3, and counted the combined delayed coincidences of 981–895 kev and 910–895 kev. If the fraction of decays coming through the 981-kev gamma ray is α, it may be shown that the observed coincidence rate N_c, following a chemical separation of lead is given by

$$N_c \propto \frac{1-\alpha}{T_1-T}\exp(-t/T_1)-\frac{1-\alpha T_1 T^{-1}}{T_1-T}\exp(-t/T), \quad (3)$$

where T is the mean life of Bi^{204} and T_1 the mean life of Pb^{204m}. Comparison of the observed time-dependent coincidence rate with this equation indicates that $\alpha=0.75$.

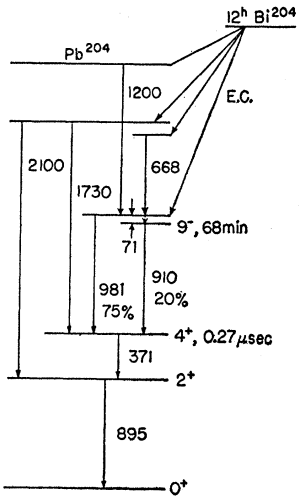

FIG. 3. A very tentative partial decay scheme for Bi^{204}—Pb^{204}, based on the observed γ rays and the coincidence data.

The data presented are designed primarily to demonstrate the existence of a specific interference which makes directional correlation measurement of the cascades of 910–371 and 910–895 kev extremely difficult if not impossible in unseparated targets. The 981-kev gamma ray provides this interference. Since it follows promptly upon electron capture, the correlation between it and either of the two gamma rays following the 0.27-microsecond level cannot be used safely in a determination of the delayed coincidence attenuation coefficients characteristic of the static electric quadrupole effect.

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[12] Frauenfelder, Lawson, Jentschke, and DePasquali, Phys. Rev. **92**, 513 (1953).

[13] The authors are indebted to Professor K. T. Bainbridge and R. Narcisi for these measurements.

[14] Templeton, Howland, and Perlman, Phys. Rev. **72**, 766 (1947).