Alpha-Gamma Angular Correlation in Ionium (Th^{230})†

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The angular correlation between the alpha particle leading to the first excited state of Ra^{226} at 67.8 kev and the subsequent gamma ray to the ground state leads to a definite assignment of $I=2^+$ for this state. The correlation is weakened by what is believed to be an interaction of the nuclear electric quadrupole moment with crystalline electric field gradients in the source. The lifetime of the excited state is about 1×10^{-8} second.

1. INTRODUCTION

THE study of the fine structure in the alpha radiation of ionium has revealed two major lines at 4.682 Mev (~75 percent) and 4.612 Mev (~25 percent), respectively, separated by about 70 kev.[1] The gamma-ray spectrum following the alpha decay was studied by means of the conversion electrons by several investigators.[2–5] The latest work on the gamma spectrum with a NaI crystal spectrometer and gamma-gamma coincidences[6] leads to the decay scheme shown in Fig. 1. The numbers in parentheses give the intensities in quanta per disintegration. Rosenblum and Valadares,[7] from a comparison of their observed ratios of conversion coefficients in the (unresolved) $L_{I}+L_{II}$ and L_{III} shells with some theoretical values obtained by Gellman *et al.*[8] conclude that the 67.8-kev gamma radiation is of the $E2$ type. Our observations on the alpha-gamma angular correlation establish the sequence $0^+-2^+-0^+$ and thus add another $I=2^+$ case to the growing number of first excited states of even-even nuclei of this type.[9,10]

While these measurements were in progress an explanation was put forward by Abragam and Pound[11] to account for the pronounced departure of the much investigated alpha-gamma angular correlation in radiothorium[12–14] from the predicted shape. Our results in ionium show features very similar to the RdTh case and are believed to be due to similar causes.

2. EXPERIMENTAL DETAILS

An over-all schematic diagram of our experimental arrangement is shown in Fig. 2. An electromagnetically separated source of 90-percent pure Th^{230} (in the form of $ThCl_4$) deposited on a thin rubber hydrochloride film, furnished by the Argonne National Laboratory, was located at the center of a small hemispherical vacuum chamber made of 0.033-in. aluminum. This chamber was mounted directly on the front face of an RCA 5819 photomultiplier tube and was continuously evacuated. The source diameter was 4.7 mm. A thin, single crystal of terphenyl was prepared from a supersaturated solution of terphenyl in xylene and was about 0.001 in. thick, i.e., about equal to the alpha-particle range. The crystal turned out to be completely insensitive to all but alpha radiation. It was mounted in the center of the multiplier face and subtended a half-angle of five degrees at the source. The gamma detector consisted of a 1.40 cm×1.56 cm×0.50 cm NaI(Tl) crystal mounted on another 5819 tube and also subtended a half-angle of about 5 degrees at the source. The small thickness of this crystal reduced the background and response to higher energy gamma rays without affecting the near 100 percent efficiency

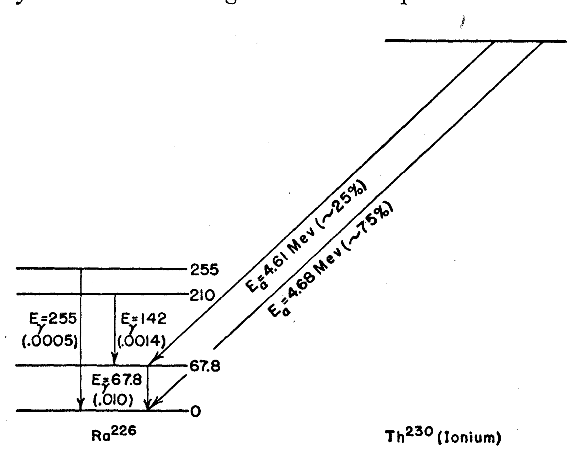

FIG. 1. Decay scheme of Ra^{226} as proposed in reference 6. Gamma-ray energies are given in kev. Numbers in parentheses give intensities in quanta/disintegration.

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[2] J. Teillac, Compt. rend. **227**, 1227 (1948); Albouy, Faraggi, Riou, and Teillac, Compt. rend. **229**, 435 (1949).
[3] S. Rosenblum and M. Valadares, Compt. rend. **232**, 501 (1951).
[4] C. J. D. Jarvis and M. A. S. Ross, Proc. Phys. Soc. (London) **A64**, 535 (1951).
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[10] G. Scharff-Goldhaber, Phys. Rev. **90**, 587 (1953).
[11] A. Abragam and R. V. Pound, Phys. Rev. **89**, 1306 (1953).
[12] Kulchitski, Latyshev, and Bulyginski, Doklady Akad. Nauk. (S.S.S.R.) **64**, 57 (1949).
[13] Beling, Feld, and Halpern, Phys. Rev. **84**, 155 (1951).
[14] Battey, Madansky, and Rasetti, Phys. Rev. **89**, 182 (1953).

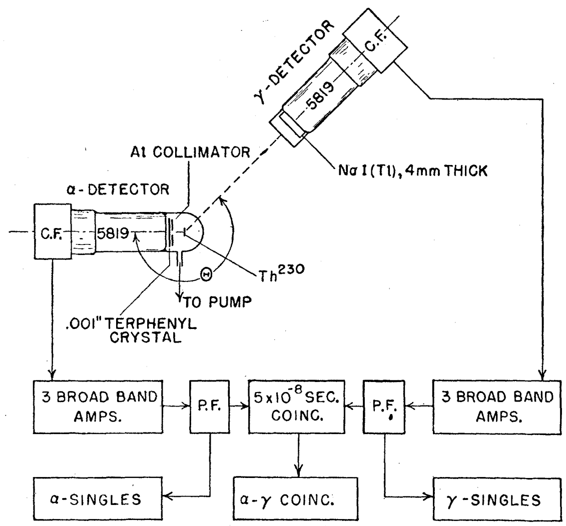

FIG. 2. Schematic diagram of experimental arrangement. Θ is the angle referred to throughout this paper. The entire apparatus is surrounded by a 2-in. lead shield.

for the radiation of interest (67.8 kev). The gamma detector could be set at angles between 84° and 276° with respect to the alpha counter direction. We were thus able to investigate angles on either side, thus having a check on possible systematic left-right differences. The correlation was measured over the hemisphere $\Theta=90°$ to $\Theta=180°$, which is necessarily identical to the hemisphere $\Theta=0°$ to $\Theta=90°$ because all correlation functions involve only even powers of $\cos\Theta$.

The cathode follower outputs from the two multipliers were amplified by three broad band amplifiers (Hewlett-Packard and SKL) in cascade and fed into two pulse-forming stages using Philips EFP-60 secondary emission pentodes. A diode limiter network in the gamma channel prevented overloading of the amplifier chain by large pulses. The standard height outputs of the shaping stages entered a fast coincidence circuit similar to one described by DeBenedetti and Richings.[15] An inherent delay in the gamma channel of about 7×10^{-8} second, probably due to the slower pulse coming from the NaI crystal, was compensated by an appropriate length of cable. The output in turn was stretched, amplified, and recorded by both a scaler and a pen recorder. Both single channels were continuously monitored. The pen recorder made it possible to check the arrival in time of the very small number of coincidences (<8 counts/hour) in this experiment and to distinguish any spurious counts. The resolving time of the circuit was measured to be about 5×10^{-8} second, both by the double-source method and from delay cable measurements (see below). The accidental coincidence rate never exceeded five percent of the total rate at any angle.

[15] S. DeBenedetti and H. J. Richings, Rev. Sci. Instr. 23, 37 (1952).

The gamma-ray pulse-height distribution as obtained with a conventional linear amplifier and single-channel analyzer is shown in Fig. 3. During our coincidence experiments the gamma-ray pulse height was selected by integral bias just below the 68 kev peak. The effect of the higher energy gamma rays at 142 kev and 255 kev will be discussed below. The small peak at about 35 kev is due to the escape of some of the iodine K x-radiation ($\sim$30 kev) following the photoelectric conversion of the gamma rays. We showed this to be so by means of a critical absorber check with tin.

The alpha-ray pulses received from the source presented a rather broad range of pulse heights due to source thickness, although the system itself was capable of good resolution, as may be seen from Fig. 4. It was nevertheless possible to keep the noise contribution to less than 10 percent of the total counting rate in the alpha channel. The entire apparatus was surrounded by two inches of lead to reduce the rather high gamma-ray background from our building.

Our results were obtained during a more or less continuous run of about 1000 hours and consist of six coincidence counting rates at the crucial points of the angular correlation. The angle was changed at least every 24 hours to minimize systematic errors. The photomultiplier voltages were monitored by a precision potentiometer and were kept constant to within 4 volts.

3. RESULTS

The final values of the coincidence rates at six angles together with the various major corrections which were

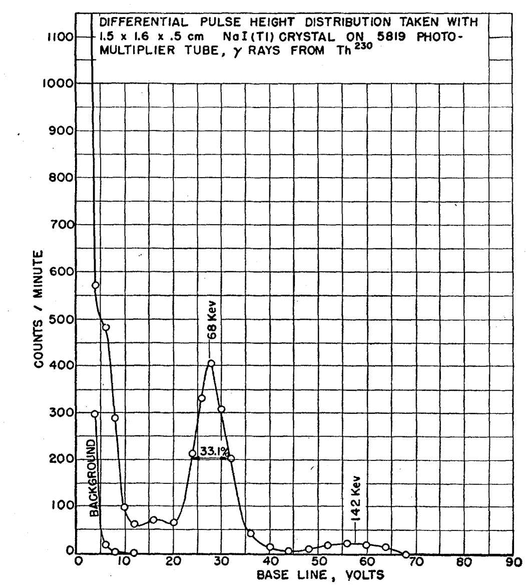

FIG. 3. Pulse-height distribution in the gamma channel. The 255-kev gamma ray is not shown. Bias during experiment set between 68-kev peak and small peak at about 35 kev. The latter is an iodine escape peak (see text).

applied are shown in Table I. All rates were normalized according to the singles rates in both the alpha channel and the gamma channel, the latter by means of a strong ionium source counted before and after each run. This was necessitated by the low gamma rate (~130 counts/minute) and its consequent sensitivity to changes in the background rate (~30 counts/minute). The values at 84° and 276° (not 90° and 270° because of an obstruction) as well as at 135° and 225° are seen to be in fairly good agreement. Those two pairs of points were combined for the final computations. The point at 120° was taken merely to rule out a possible $0^+-3^--0^+$ sequence. We refer to papers by Arnold[16] and Falkoff and Uhlenbeck[17] for the basic theoretical expressions for alpha-gamma angular correlations. Figure 5 shows a plot of the experimental results. Our results agree best with the $\sin^2 2\Theta$ function expected for the transition $0^+-2^+-0^+$ except for considerable deviations believed to be due to extranuclear effects which we shall discuss presently.

4. CORRECTIONS

All corrections applied to the raw results were quite small and can be seen in Table I.

(1) Contribution of Higher Energy Gamma Rays

Because of the integral pulse-height selection in the gamma channel we made a crude check on the angular correlation for energies greater than 68 kev at the low rate points (84° and 180°). This check indicated approximate isotropy, which is reasonable in view of the undoubtedly more complicated (and hence more nearly isotropic) nature of the main higher energy component at 142 kev ending on an $I=2^+$ level. We have therefore subtracted an appropriate constant amount (0.43 count/hour) from all observed points. This agrees with our experimentally observed ratio of gamma-ray intensities, which in turn is in good agreement with the work of Rasetti and Booth.[6]

(2) Effects of Finite Source and Detector Size

The observed angular correlation was approximated by an expression of the form $W(\Theta)=a+b\sin^2\Theta$ and

TABLE I. Summary of experimental results. All numbers in coincidence counts per hour. (1) are original data (normalized according to single rates); (2) corrected for accidentals; (3) corrected for other gamma rays; (4) corrected for finite detectors and source; (5) standard deviations (statistical only).

Θ	(1)	(2)	(3)	(4)	(5)
84°	3.62	3.51	3.08	2.97	±0.17
120°	8.61	8.50	8.07	8.12	±0.56
135°	9.05	8.94	8.51	8.63	±0.43
180°	3.52	3.41	2.98	2.75	±0.15
225°	8.82	8.71	8.28	8.40	±0.36
276°	4.12	4.00	3.57	3.46	±0.27

[16] W. R. Arnold, Phys. Rev. **80**, 34 (1950).
[17] D. L. Falkoff and G. E. Uhlenbeck, Phys. Rev. **79**, 323 (1950).

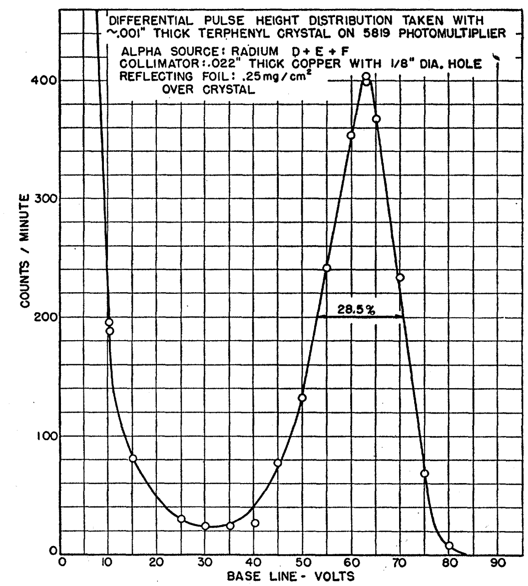

FIG. 4. Pulse-height distribution in the alpha channel, as obtained with a thin RaD+E+F (polonium) alpha source.

integrated over both the extent of the source and the solid angles subtended by the two crystals. This correction turned out to be quite small (~5 percent in the worst case) and is also shown in Table I.

(3) Minor Corrections

The correction due to accidental coincidences was already mentioned. Several long runs with incoherent sources at the usual counting rates confirmed the value (5×10^{-8} sec) of the resolving time obtained regularly with a stronger gamma source. The scattering of gamma rays in the thin spherical shell was found to be negligible.

5. INTERPRETATION

(1) Spin and Parity Assignment

A comparison of the experimental angular correlation with various possible sequences $I_A-I_B-I_C$ leaves $0^+-2^+-0^+$ as the most likely choice even before considering coupling effects: a *less* pronounced anisotropy than the observed one obtains for nonzero values of I_A and I_C (ground states of Th^{230} and Ra^{226}) because of the distribution of nuclei over magnetic substates. Furthermore these two nuclei undoubtedly have $I=0^+$ in their ground states because of their even-even character. This leaves the various $0^+-I_B-0^+$ possibilities. $I_B=1^-$ requires a maximum at 90° and minima at 0° and 180°; $I_B=3^-$ leads to minima at 0°, 60°, 120°, and 180° with maxima at 30°, 90°, and 150°. Furthermore, the lifetime of an $E3$ transition would be too long to have yielded coincidences with our resolving time; this is true *a fortiori* for all $I_B>3$. Hence we are left with $0^+-2^+-0^+$ as the only satisfactory choice. The even parity of the inter-

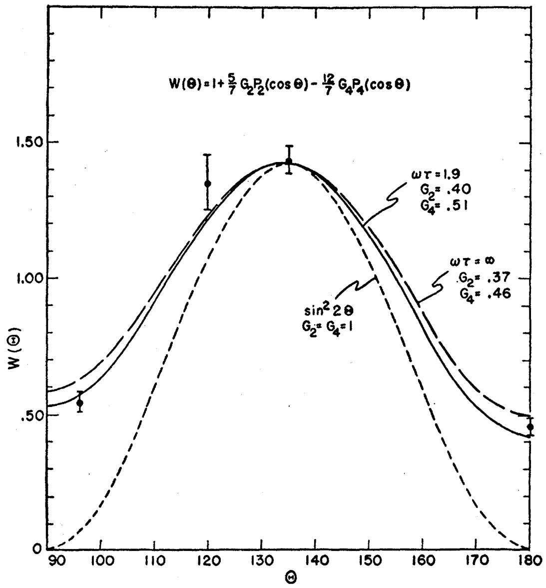

FIG. 5. Plot of experimental results of alpha-gamma angular correlation. Left and right points have been combined to appear in one quadrant. Standard deviations shown are statistical only. Short-dashed curve shows $\sin^2 2\Theta$ correlation for $0^+-2^+-0^+$ case and bare nucleus. Solid curve shows theoretical electric quadrupole attenuation for parameters shown in figure. Long-dashed curve shows maximum theoretical attenuation ($\omega\tau=\infty$). General expression used is given on top of figure [see reference (11)].

mediate state follows from the plausible assumption of even ground states for even-even nuclei.

(2) Extra-nuclear Effects

Even though a $\sin^2 2\Theta$ angular correlation comes closest to the observed results, we note a very definite attenuation of the angular pattern (see Fig. 5) that cannot be due to experimental causes, as we have seen above. In Fig. 6 we have plotted the effect on the $0^+-2^+-0^+$ sequence of both nuclear magnetic dipole coupling with electronic magnetic fields[18] and nuclear electric quadrupole coupling with crystalline electric field gradients.[11] Both effects depend on the product $\omega\tau$; ω is the Larmor precession frequency in the magnetic case and the characteristic splitting frequency between the $m_B=0$ and $m_B=\pm 1$ substates in the electric case, τ is the mean lifetime of the intermediate state. The quantities $G_2(\omega\tau)$ and $G_4(\omega\tau)$ are the "attenuation" coefficients of the second and fourth Legendre polynomials occurring in the angular correlation function. All curves refer to the case of a crystalline powder, i.e., no preferential orientation. The magnetic dipole interaction curves apply to the special but representative case $J=\frac{1}{2}$ for the electronic configuration. Figure 7 illustrates the dependence of the electric quadrupole attenuation coefficients G_2 and G_4 on $\omega\tau$. We see that the coefficients are relatively insensitive to the value of $\omega\tau$ above $\omega\tau\sim 1$. The corresponding plot for the magnetic case can be found in a paper by Heer.[19] Although our data certainly do not permit a definite choice of $\omega\tau$, we can see from Fig. 5 that $\omega\tau$ must be larger than about 1.9. Magnetic interaction to any large extent is ruled out from the relative magnitudes of the 90° and 180° points. We see in Fig. 6 that the 90° point in the magnetic case *always* lies *below* the 0°−180° point. Alghough equality is certainly not excluded by our results, the *strength* of our anisotropy would require values for G_2 and G_4 (magnetic) that would produce a 180°/90° ratio greater than 2, which is certainly ruled out. Our upper limits for G_2 and G_4 are 0.40 and 0.51 respectively, while the lower limits are given by the $\omega\tau=\infty$ (irreducible minimum) values $G_2(\infty)=0.37$ and $G_4(\infty)=0.46$, so that $0.37\leqslant G_2\leqslant 0.40$; $0.46\leqslant G_4\leqslant 0.51$. These rather narrow limits on the coefficients actually correspond to an infinite upper limit on $\omega\tau$.

(3) Lifetime Considerations

Rasetti and Booth[6] estimate that the lifetime of the 67.8-kev state is less than 0.3 microsecond from gamma-gamma coincidence measurements. From the dependence of alpha-gamma coincidence counting rate on delay inserted in either channel we believe that we can set an upper limit of $\sim 1\times 10^{-8}$ second on the mean lifetime of the 67.8-kev state in Ra^{226}. A slight asymmetry in the delay curve seems to indicate that the

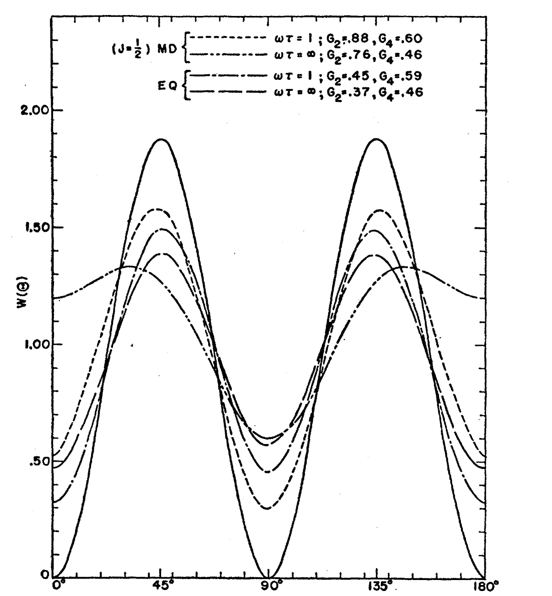

FIG. 6. Electric and magnetic coupling effects on alpha-gamma angular correlation. All curves are for $0^+-2^+-0^+$ sequence, and for isotropic distribution of the extranuclear fields (powder). For an explanation of symbols, refer to text. Magnetic curves are calculated for special case $J=\frac{1}{2}$ according to reference (18); electric curves according to reference (11). Solid curve is undisdistorted $\sin^2 2\Theta$ distribution. Note characteristic difference between electric and magnetic 180°/90° ratios.

[18] K. Alder, Helv. Phys. Acta **25**, 234 (1952).

[19] E. Heer, Physica **18**, 1215 (1952).

lifetime may actually be of that order of magnitude. A further argument in favor of a lifetime considerably shorter than our resolving time is the fact that our experimentally observed coincidence rate agrees almost exactly with the rate to be expected from the known alpha branching ratio,[1] total internal conversion coefficient[5] and our solid angles.

The lifetime calculated on the basis of Weisskopf's single particle expression[20] for an electric quadrupole transition of this energy yields a value

$$\tau = 2.4\times 10^{-6} \text{ second.}$$

The experimental value for the total internal conversion coefficient of the 67.8-kev transition[5] is about 22, so that the corrected lifetime is about 1×10^{-7} second. This is at least ten times the experimental value. Although Weisskopf's formula gives a lower limit on the independent particle picture, it is not too surprising to find a matrix element $\gg 1$ for the transition between low-lying states of an even-even nucleus where cooperative effects are presumably very important.[21,22]

6. CONCLUSIONS

Our measurements seem to establish an $I=2^+$ assignment for the first excited state of Ra^{226}. The deviation of the observed angular correlation from the pure $\sin^2 2\Theta$ distribution predicted for a bare nucleus and a $0^+-2^+-0^+$ transition receive a satisfactory interpretation on the basis of the coupling of the nuclear electric quadrupole moment to a crystalline electric field gradient in the source material.[11] Although the deviation is thus explained, the large recoil energy imparted to the residual nucleus by the first alpha emission ($\sim$100 kev) frustrates attempts to extract the actual value of the quadrupole moment because of the impossibility of knowing the appropriate electric field gradient at the site of the nucleus. Gamma-gamma angular correlation studies seem to offer considerably greater promise in this respect.[23] It is interesting to note that the only other known case of alpha-gamma angular correlation showing these interaction effects is that of radiothorium (Th^{228}), leading to Ra^{224}.[12–14] This nucleus and the one studied by us differ by just two neutrons and could be examined in the same chemical environment. Hence one could in principle obtain a ratio of quadrupole moments of Ra^{224} and Ra^{226} in their first excited states as soon as the lifetimes are pinned down more accurately. This whole argument is of course weakened in practice by the inherent insensitivity of the correlation to the value of $\omega\tau$.

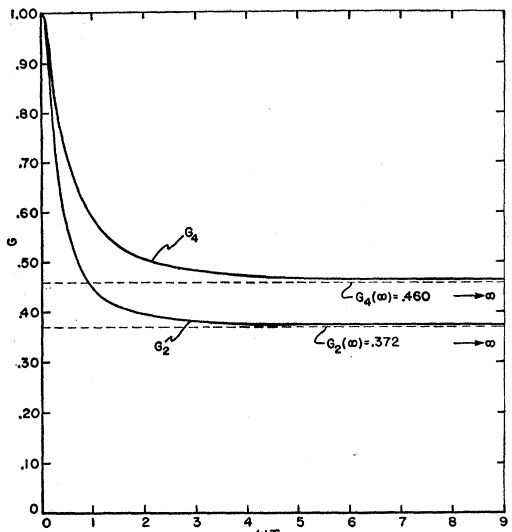

FIG. 7. Theoretical attenuation coefficients for $I=2$ nucleus with electric quadrupole coupling. Case of crystalline powder source (isotropic distribution of extranuclear field gradient). For explanation of symbols refer to text. Note insensitivity of coefficients to value of $\omega\tau>1$. Limiting values are shown by dashed horizontal lines.

We express our thanks to Dr. Paul Fields of the Argonne National Laboratory for supplying us with the electromagnetically separated source of ionium and to Professor Franco Rasetti for sending us his manuscript in advance of publication.

Note added in proof:—The angular correlation between the alpha particles of ionium and *conversion electrons* (92.3-percent L shell, 7.7-percent M shell) corresponding to the same 70-kev transition in Ra^{226} as the gamma ray in our work has recently been determined [R. R. Roy and M. L. Goes, *Nature* **172**, 360 (1953)].

[20] J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley & Sons, Inc., New York, 1952), p. 627.
[21] A. Bohr and B. Mottelson, Phys. Rev. **89**, 316 (1953).
[22] K. W. Ford, Phys. Rev. **90**, 29 (1953).
[23] Albers-Schönberg, Hänni, Heer, Novey, and Scherrer, Phys. Rev. **90**, 322 (1953).