Angular Correlations of Gamma Rays in Cascade from Levels in Lu^{175} and Hf^{177}

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The directional angular correlations of the 208.36–112.97 kev gamma-gamma cascade in Hf^{177} and the 89.36–343.40 and 282.57–113.81 kev cascades in Lu^{175} have been measured with a coincidence scintillation spectrometer using NaI detectors. The observed correlation functions obtained with dilute aqueous solution sources are $W(\theta)=1-(0.1614\pm0.0015)P_2(\cos\theta)$, $W(\theta)=1+(0.001\pm0.004)P_2(\cos\theta)$, and $W(\theta)=1+(0.221\pm0.004)P_2(\cos\theta)$, respectively.

These results, taken together with conversion coefficient measurements, indicate that δ^2 for the 282.57-kev gamma ray in Lu^{175} is 0.05. The sign of δ, defined below, is positive. The mixing ratio of the 208.36-kev gamma ray in Hf^{177} is $M2/E1=0.001$; in this case the sign of δ is negative. The sign of δ for the 113.81-kev gamma ray in Lu^{175} is positive, and the sign of δ for the 112.97-kev gamma ray in Hf^{177} is negative.

I. INTRODUCTION

SINCE the directional angular correlation of the 208.36–112.97 kev gamma-gamma cascade in Hf^{177} was first measured,[1] the spin of the ground state has been measured[2] to be $\frac{7}{2}$ instead of the $\frac{1}{2}$ or $\frac{3}{2}$ as indicated by previous work. The properties of the energy levels of Hf^{177} have been discussed along with a detailed analysis of the levels of Lu^{175} in terms of the Bohr-Mottelson strong-coupling unified model by Chase and Wilets.[3] In order to obtain as much information about these levels as is possible from gamma-gamma directional angular correlation measurements, the correlation function of the 208.36–112.97 cascade in Hf^{177} was remeasured with the improved apparatus now in use, and those of the 89.36–343.40 and the 282.57–113.81 kev cascades in Lu^{175} were also measured.

II. EXPERIMENTAL RESULTS

The correlation functions were obtained from measurements at 19 angles with a coincidence scintillation spectrometer using NaI detectors, and the data were analyzed as previously described.[4] In each case correlation functions were measured for both dilute aqueous solution sources and solid sources in order to obtain an idea of the perturbation of the intermediate state of the cascade in question by extranuclear fields. Also in each case the window of the differential analyzer associated with each detector was set to accept only the pulses in the full-energy peak of one of the gamma rays of the cascade.

The decay schemes of Hf^{175} and Yb^{175} as given by Mize, Bunker, and Starner[5] are shown in Fig. 1. The decay scheme of Lu^{177} as given by Wiedling[6] is shown in Fig. 2. The angular correlation function of the 89.36–343.40 kev cascade in Lu^{175} was measured in a series of experiments with a total of 2.9×10^5 coincidence counts, using a dilute aqueous solution source. The true coincidence rate was of the order of 0.4 count per second, and the random rate was 12% of this. The correlation function obtained was isotropic, and the experiment was repeated with a dry powder source. Again an isotropic correlation was found.

The correlation function of the 282.57–113.81 kev cascade in Lu^{175} was measured with a dilute aqueous solution source, a total of 3.5×10^5 coincidence counts being obtained. The true coincidence counting rate was of the order of 0.9 count per second, and the random rate was 6% of this. The correlation was measured with a dry source in a set of experiments in which 5.4×10^5 coincidences were observed. Only a very small possible effect of the state of the source on the correlation function was observed, as discussed below.

The correlation function for the 208.36–112.97 kev cascade in Hf^{177} was obtained from a series of experiments with a dilute aqueous solution source in which a total of 2.7×10^6 coincidence counts were measured. In this case the true coincidence counting rate was about 8 counts per second, and the random rate was 15% of this. Again the experiment was repeated with a dry source, this time obtaining a total of 1.8×10^6 coincidence counts; and a small attenuation of the coefficient of the correlation function was observed.

III. ANALYSIS OF DATA

The data obtained for the 89.36–343.40 kev cascade in Lu^{175} were fitted by functions of the form $W(\theta)=1+A_2P_2(\cos\theta)+A_4P_4(\cos\theta)$ and $W(\theta)=1+A_2P_2(\cos\theta)$. A statistical ratio test was carried out, as described in a previous paper,[4] to determine if the dropping of the $P_4(\cos\theta)$ term was statistically significant. It was found that the correlation function best representing the data is given by $W(\theta)=1+(0.001\pm0.004)P_2(\cos\theta)$. As pointed out above, both liquid and solid sources yielded isotropic angular distributions. Since the decay of Hf^{175} is by orbital electron capture and since only an upper limit on the lifetime of the intermediate state

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[3] D. M. Chase and L. Wilets, Phys. Rev. **101**, 1038 (1956).
[4] Ernest D. Klema, Phys. Rev. **102**, 449 (1956).
[5] Mize, Bunker, and Starner, Phys. Rev. **100**, 1390 (1955).
[6] Tor Wiedling, *Directional Correlation Measurements and Some Other Related Investigations of Excited Nuclei* (Almquist and Wiksells Boktryckeri AB, Uppsala, 1956).

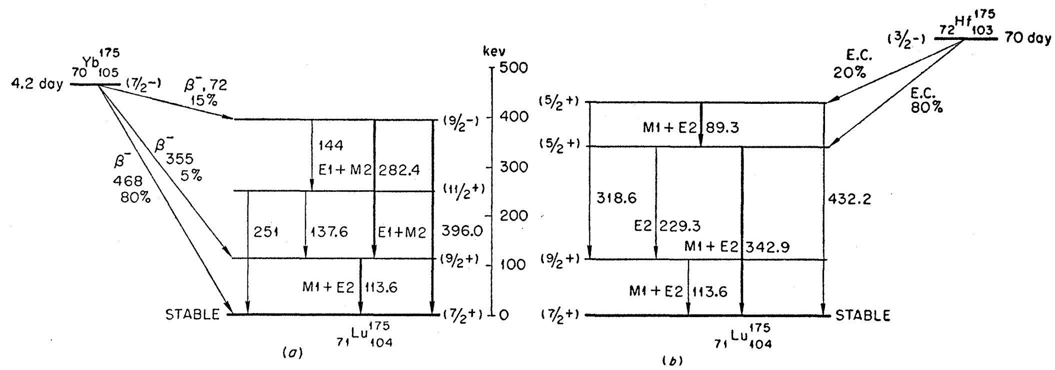

FIG. 1. (a) Disintegration scheme of Yb^{175}. (b) Disintegration scheme of Hf^{175}.

of the cascade has been measured as 10^{-9} second,[7] the function given above may be in doubt because it may represent an attenuation of the true correlation function coefficients due to extranuclear fields.

The correlation function obtained for the 282.57–113.81 kev cascade in Lu^{175} is given by $W(\theta)=1+(0.221\pm0.004)P_2(\cos\theta)$. This measurement is in good agreement with that of Wiedling,[6] who obtained $W(\theta)=1+(0.227\pm0.004)P_2(\cos\theta)$. The correlation function found with a dry source in the present experiments is given by $W(\theta)=1+(0.210\pm0.003)P_2(\cos\theta)$. However, Wiedling[6] measured A_2 as a function of the viscosity of water solutions of $Lu(NO_3)_3$. He varied the viscosity of the solutions by adding different amounts of water-free glycerine. He found that within the experimental errors there was no attenuation; thus one can assume that the coefficient obtained for dilute aqueous solution sources is the true one.

The correlation function measured for the 208.36–112.97 kev cascade in Hf^{177} is given by $W(\theta)=1-(0.1614\pm0.0015)P_2(\cos\theta)$. Again this result is in good agreement with that of Wiedling[6] for dilute solution sources; namely, $W(\theta)=1-0.160P_2(\cos\theta)$. Wiedling has also studied the effect of viscosity on this angular correlation and has arrived at the following unperturbed angular correlation function: $W(\theta)=1-(0.163\pm0.002)P_2(\cos\theta)$. In the present experiments, the correlation function found for a solid source was given by $W(\theta)=1-(0.149\pm0.002)P_2(\cos\theta)$.

The present work is in disagreement with a recent measurement made by Ofer.[8] With a dilute aqueous solution source he found $W(\theta)=1-(0.130\pm0.010)\times P_2(\cos\theta)$ for the 208.36–112.97 kev cascade in Hf^{177}. Thus as a further check on our experimental apparatus, additional calculations were carried out on the data obtained with the dilute solution source. In order to see whether the apparatus introduces statistically significant coefficients of odd Legendre polynomials, which are known from theory not to be present in the physical situations we consider,[9] a fit was made to the function

$$W(\theta)=1+A_1P_1(\cos\theta)+A_2P_2(\cos\theta)+A_3P_3(\cos\theta)+A_4P_4(\cos\theta).$$

Then the odd polynomials were left out one at a time and new least-squares fits were made. Statistical ratio tests were performed, and it was found that neither the coefficient of $P_1(\cos\theta)$ nor that of $P_3(\cos\theta)$ was statistically significant.

IV. INTERPRETATION OF CORRELATION FUNCTIONS

Since the spin of the ground state of Lu^{175} has been measured to be $\frac{7}{2}$,[10] from the correlation function of the 89.36–343.40 kev cascade one may hope to obtain the spins of the initial and intermediate states of the cascade. The spin of the 432.76 kev level from which the cascade originates is of especial interest.

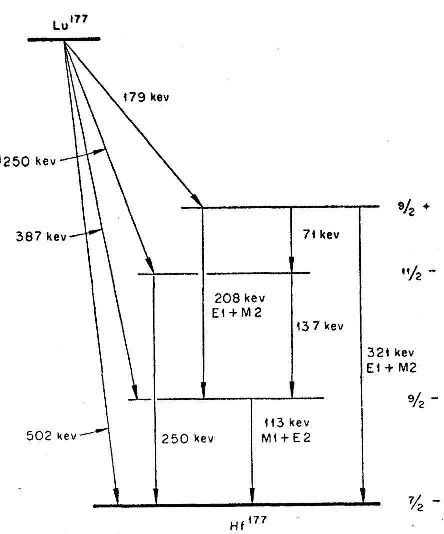

FIG. 2. Disintegration scheme of Lu^{177}.

[7] F. K. McGowan, Oak Ridge National Laboratory Report ORNL-1705 (unpublished), p. 18.
[8] S. Ofer, Nuclear Phys. 3, 479 (1957).
[9] Stuart P. Lloyd, Phys. Rev. 85, 904 (1952).
[10] H. Schüler and T. Schmidt, Naturwiss. 22, 714 (1934).

From scintillation spectrometry, Mize, Bunker, and Starner[5] tentatively assigned it spin $\frac{5}{2}$. However, Burford, Perkins, and Haynes[11] and Hatch, Boehm, Marmier, and DuMond[12] have assigned spin $\frac{7}{2}$ to it. In their discussion of the levels of Lu^{175} in terms of the unified model, Chase and Wilets[3] state that spin $\frac{7}{2}$ is preferred by the model and that it would be difficult to account for a second $\frac{5}{2}+$ state (the 343.40-kev level is $\frac{5}{2}+$).

Both of the gamma rays in the cascade are known to be mixed. The measured K-conversion coefficient[7] of the 343.40-kev gamma ray and both the measured $K/(L+M)$ ratio[11] and absence of observable conversion in the L_{II} and L_{III} subshells[5] indicate that it is a mixture of $M1$ and $E2$ radiation. The measured L-subshell conversion coefficients[5] of the 89.36-kev gamma ray indicate that it is a mixture of $M1$ and $E2$ also. The fact that the observed angular correlation function is isotropic greatly simplifies its analysis.

Assuming that the observed angular correlation function is unperturbed, one can analyze the experimental results as follows: the expression for A_2 in the correlation function $W(\theta)=1+A_2P_2(\cos\theta)+A_4P_4(\cos\theta)$ for the case of a mixed-mixed cascade can be written as the product of two factors, one involving only the first transition and the other involving only the second. For each transition a quantity δ is defined such that δ^2 is the ratio of the intensity of the 2^{L+1}-pole radiation to that of the 2^L-pole radiation in the transition. The sign convention used for δ in the present work is that given by Biedenharn and Rose.[13] Often δ^2 is called the mixing ratio of the transition in question.

Since the mixing ratio of the 343.40-kev gamma ray is known, it can be shown that the factor involving this transition in the expression for A_2 does not vanish. Thus the values of the mixing ratio of the 89.36-kev gamma ray for which A_2 vanishes can easily be solved for, and the laborious calculations of mixed-mixed correlation functions can be avoided.

All nine possible sequences of $\frac{3}{2}$, $\frac{5}{2}$, $\frac{7}{2}$, 9/2, 11/2 $(D+Q)\frac{5}{2}$, $\frac{7}{2}$, $9/2(D+Q)\frac{7}{2}$ with a spin difference of zero or one unit between the states involved in a given transition have been considered. For the sequence $\frac{5}{2}(D+Q)\frac{5}{2}(D+Q)\frac{7}{2}$, the values of δ for the 89.36-kev gamma ray for which A_2 vanishes are 5.71 and -0.392. Only the latter is compatible with the mixing ratio of this gamma ray obtained from the measurement of the L-subshell conversion coefficients, $\delta^2\sim 0.1$.[5] The value of -0.392 leads to an $E2/M1$ intensity ratio of 0.15, and the corresponding A_4 is -0.0004. For the sequence $\frac{7}{2}(D+Q)\frac{5}{2}(D+Q)\frac{7}{2}$, $\delta=-4.18$ and $\delta=-0.100$ make A_2 vanish. The second value of δ leads to an $E2/M1$ intensity ratio of 0.010, which would be in fair agreement with the value[12] of 0.03 obtained by a combination of conversion coefficient measurements, but would be in serious disagreement with the value of 0.1.

For the sequence $\frac{3}{2}(D+Q)\frac{5}{2}(D+Q)\frac{7}{2}$, the condition that A_2 vanish is given by $\delta=-10.1$ or $\delta=0.193$. The latter leads to a mixing ratio of 0.037, in fair agreement with both values obtained from the conversion coefficient measurements. The corresponding A_4 is 0.0002. It is of interest to point out that a spin of $\frac{3}{2}$ for the 432.76-kev level might make this level the analog of the 618.9-kev level in Ta^{181}.[14] There are no other fits of the angular correlation data in the sequences investigated which lead to mixing ratios between 0.03 and 0.2.

The other cascade measured in Lu^{175} also terminates at the ground state. The spin of the intermediate state is known since it is the first member of a rotation band based on the ground state of Lu^{175} and has been formed by Coulomb excitation.[15] The 113.81-kev gamma ray has been extensively studied at a number of laboratories. The L-subshell conversion coefficients[5,12,16] have been measured, the K-conversion coefficient[5,11,12,17] has been measured, and the K/L ratio[11,16,18,19] has been measured. Since these results are in fair agreement, we have taken the mixing ratio of the 113.81-kev gamma as

$$E2/M1=0.18.$$

In the calculations discussed below, δ_1 refers to the 113.81-kev gamma ray and δ_2 refers to the 282.57-kev gamma. The angular distribution function[20] for the mixed-mixed cascade has been programmed for calculation on the ORACLE. The coefficients A_2 and A_4 are calculated for both signs of δ_1 as a function of δ_2 for 72 values of δ_2 from 0.015 to 100. Coefficients for both signs of δ_2 are calculated. In the present case calculations were made for the sequences $\frac{7}{2}$, 9/2, $11/2(D+Q)9/2(D+Q)\frac{7}{2}$.

For both sequences, $\frac{7}{2}(D+Q)9/2(D+Q)\frac{7}{2}$ and $11/2\times(D+Q)9/2(D+Q)\frac{7}{2}$, the values of δ_2 which lead to values of A_2 and A_4 in agreement with experiment, $\delta_2=-0.75$ with $\delta_1=0.42$, $\delta_2=0.048$ with $\delta_1=-0.42$, and $\delta_2=0.034$ with $\delta_1=-0.42$, respectively, are incompatible with other measurements of the mixing ratio of the 282.57-kev gamma ray, as discussed below.

For the sequence $9/2(D+Q)9/2(D+Q)\frac{7}{2}$, with $\delta_1=0.42$ and $\delta_2=0.22$, $A_2=0.221$ and $A_4=-0.0021$. This is the only fit which is compatible with all the experimental data; thus for the case of the present cascade the magnitude of δ_2 and the signs of both δ_1

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and δ_2 have been determined. The above statement depends upon a subjective estimate of the precision of the value of δ_1 as obtained from conversion coefficient measurements. However, it is felt that it is almost certain that the sign of δ_2 is positive. Since it is not possible to assign a well-defined error to the value of δ_1, we have given above only the value of δ_2 corresponding to the value of δ_1 used in the calculations. This value of δ_2 leads to an $M2/E1$ intensity ratio of the 282.57-kev gamma ray of 0.05, in fair agreement with a previous angular correlation measurement[6] (0.04) and with the ratios[5,12] of 0.027 ± 0.016 and 0.02 obtained from conversion coefficient measurements.

Since the spin of the ground state of Hf^{177} is now known to be $\frac{7}{2}$ and since the 112.97-kev gamma ray has been observed by Coulomb excitation,[15] the measurement of the 208.36–112.97 kev cascade is of interest in determining the spin of the 321.33-kev level from which the cascade originates and the mixing ratios of the gamma rays. From the measured K-conversion coefficient[1] of the 112.97-kev gamma ray, one finds that it is a mixture of $E2$ and $M1$ radiation. If one uses Sliv's[21] calculated K-conversion coefficients, he finds that the intensity ratio of $E2/M1$ is 0.945/0.055. This mixing ratio leads to L-subshell ratios calculated from the tables of Rose[22] as follows: $L_{\mathrm{I}}/L_{\mathrm{II}}=0.18$ and $L_{\mathrm{III}}/L_{\mathrm{I}}=4.9$. These are in good agreement with the values measured by Wiedling[6] of 0.18 ± 0.02 and 4.8 ± 0.5, respectively. Thus in the analysis of the angular correlation data, the mixing ratio of the 112.97-kev gamma ray is taken to be known.

The spin of the 321.33-kev level from which the cascade originates is most probably 9/2, as determined both from the data of Wiedling[6] and that of Ofer.[8] According to Wiedling's work, if the spin were $\frac{7}{2}$, one would expect to see the conversion line of the highly-converted 71.64-kev gamma ray, which would be $M2$ or $E3+M2$. It was in fact not seen in the beta spectrometer. Spin 11/2 is ruled out by the information from the continuous beta spectrum. According to Ofer's work, the measurements of the K-conversion coefficients of the 71.64- and the 208.36-kev gamma rays show that they are both predominantly $E1$. From this information he concludes that the spin and parity of the 321.33-kev level is either $9/2^+$ or $11/2^+$. He rules out the $11/2^+$ assignment both on the basis that it would lead to a second-forbidden beta transition to this level, which is not the case ($\log ft=6.3$), and on the basis of his angular correlation measurements.

In order to see if the present angular correlation measurement can give any information about the spin of the 321.33-kev level, we have calculated the correlation functions of the following sequences, 11/2, 9/2, $\frac{7}{2}(D+Q)9/2(D+Q)\frac{7}{2}$, using the values of ±4.6 for δ_1, the square root of the mixing ratio of the 112.97-kev transition to the ground state. A fit of the experimental data was found for each of the above spin assignments.

Thus the present measurement gives no information about the spin of the 321.33-kev level, but it is felt that the other experimental results given above are definite enough that one is justified in analyzing the angular correlation data using spin 9/2 for the level. With this assignment one finds for $\delta_1=-4.6$ and $\delta_2=-0.033$, $A_2=-0.161$ and $A_4=-0.0003$. This result is in agreement with the work of Mann[23] on the angular correlation of the gamma rays in Hf^{177} following the decay of Ta^{177}, in that he finds that the sign of δ_2 is much more likely to be negative than positive. It leads to a mixing ratio of the 208.36-kev gamma ray of $M2/E1=0.001$, much lower than the value of 0.01 given by Wiedling.[6]

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