PHYSICAL REVIEW C VOLUME 2, NUMBER 2 AUGUST 1970

Search for Double Gamma Emission from the First Excited States of Ca^{40} and Zr^{90}†

P. Harihar,*‡ J. D. Ullman, and C. S. Wu
Physics Department, Columbia University, New York, New York 10027

(Received 9 January 1970)

A search for double γ emission from the first excited 0^+ states in Ca^{40} and Zr^{90} has been carried out using a NaI(Tl) scintillation spectrometer and a sum-coincidence technique. No definite evidence for double γ emission is found in either case. In the 3.35-MeV, 0^+-to-ground-state transition in Ca^{40}, an upper limit of 4×10^{-4} has been established for the branching ratio of double γ emission with respect to internal pair emission. For the 1.75-MeV 0^+-to-ground-state transition in Zr^{90}, an upper limit of 1.8×10^{-4} has been obtained for the branching ratio of double γ emission with respect to the sum of internal-conversion electrons and internal pairs. The significance of these results in the light of existing theories is discussed.

I. INTRODUCTION

Electromagnetic transitions of higher order involving the simultaneous emission and absorbtion of more than one quantum are usually so much slower than competing processes that it is difficult to observe them. They can best be studied when the first-order radiative transition probability vanishes, as in spin-zero–to–spin-zero transitions, where the emission of a single quantum is forbidden by angular momentum conservation.

Two-quantum emission was first considered theoretically by Göppert-Mayer[1] for atoms and later by Sachs[2] and others[3-5] for nuclei. Nuclear zero-to-zero transitions between the first excited state and the ground state, with no parity change, occur in O^{16}, Ca^{40}, Ge^{72}, and Zr^{90}. Since cascade transitions cannot occur, these excited states normally decay by emission of internal-conversion electrons and internal pairs. The lowest-order radiative transition permissible is the second order, which should occur principally by the simultaneous emission of two quanta. Thus, unless there is a parity change, in which case the nonradiative processes would also be inhibited, two-quanta emission should occur in competition with internal conversion and internal pair creation.

The simultaneous emission of two γ rays in the second order is assumed to take place via intermediate states which lie above the 0^+ first excited state. As in all three-body decays, the spectrum is expected to be continuous, with the sum of the energies of the two quanta equal to the total zero-to-zero transition energy. The theoretical spectrum shape depends both on the multipolarity of the γ rays and the location of the intermediate states above the first excited state. However, since the transition probability is greatest for the lowest multipoles radiated, dipole emission only may be considered. The intermediate dipole states may lie just above the 0^+ first excited state or higher up in the region of the giant resonance, where the density of dipole states is large, as observed experimentally. The angular correlation between the two γ rays is the same as for dipole cascades.

Experimental searches for double γ emission in zero-to-zero transitions have been conducted by several investigators. The results of Ryde[6] for the 1.75-MeV 0^+-to-0^+ transition in Zr^{90} yields an upper limit of 4.2×10^{-4} for the branching ratio with respect to all other decay modes. Vanderleeden and Jastram[7] found an upper limit of 8×10^{-5} for the same branching ratio. In the 3.35-MeV 0^+-to-0^+ transition in Ca^{40}, Sutter[8] has established an upper limit of 2×10^{-3} for the branching ratio with respect to internal pair emission. Finally, Alburger and Parker[9] have studied the 6.05-MeV 0^+–to-ground-state transition in O^{16} and have obtained an upper limit of 1.1×10^{-4} for the branching ratio with respect to internal pair emission.

The value of this branching ratio gives useful information on the nature of the 0^+ ground and excited states. It is one more piece of data to fit, along with transition rates for single γ emission from higher states to the states in question and the rate of e^+-e^- emission. In fact, the unusual amount of information available on these states makes the prediction of their properties a severe test of any nuclear model.

The upper limits quoted above already tell a great deal. Single-particle estimates of the branching ratio[4,5] come out in the region of 10^{-2} -10^{-3}, which is 1 to 2 orders of magnitude greater than the upper limits quoted. Since single-particle estimates of the e^+-e^- decay rate are of the right order of magnitude, double γ emission seems to be strongly hindered in the cases discussed above. Any proposed structure of these 0^+ states must explain this hindrance.

In view of the theoretical significance of this work, it is necessary to pursue the search for double γ emission with improved experimental technique capable of attaining better limits on the branching ratio. The nuclei Ca^{40} and Zr^{90} were chosen in order to verify the then existing results.[10,11]

II. EXPERIMENTAL PROCEDURE

The first excited 0^+ state was either produced by inelastic resonance scattering of protons from a target or by the decay of a long-lived radioactive parent nucleus. The γ rays were detected by two NaI(Tl) detectors mounted on photomultiplier tubes, placed 180° with respect to each other, with the target or source between them. The simultaneous emission of two γ rays was ascertained by a fast-coincidence circuit with a resolving time in the neighborhood of 30 nsec. Coincidences due to Compton backscattering were reduced by lead collimators. The pulses from one of the inner dynodes of the photomultipliers were summed by a linear adder, and the added output was recorded in a multichannel pulse-height analyzer by a gating pulse supplied by a conventional fast-slow coincidence circuit. Since the sum of the energies of the two γ rays is always equal to the total zero-to-zero transition energy, a peak corresponding to this energy would be expected in the gated sum spectrum when the calibration is identical for the two channels. The summing technique is thus particularly suited to the study of double γ emission.

A Na^{22} source was placed between the two detectors for energy calibration. The γ-ray singles spectrum of Na^{22} in one of the detectors shows peaks at 0.511 and 1.28 MeV. The gains of the two photomultipliers were adjusted so that the 0.511- and 1.28-MeV peaks from both the detectors occurred in the same channels in the multichannel pulse-height analyzer. The ungated (without coincidence requirement) sum spectrum from the adder then gives five calibration points at 0.511, 1.02, 1.28, 1.79, and 2.3 MeV. The gated sum spectrum, on the other hand, gives calibration points at 1.02, 1.79, and 2.3 MeV. The 1.79-MeV peak was particularly useful in the search for double γ emission from Zr^{90}, where a sum peak was expected at 1.75 MeV. Measurement of the single and coincidence rates also gave the photopeak efficiencies of the counters for 0.511- and 1.28-MeV γ rays.

III. MEASUREMENTS AND RESULTS

A. Ca^{40}

The first excited 0^+ state at 3.35 MeV in Ca^{40} (Fig. 1) was populated by inelastic resonance scattering of protons from a calcium target. The 6.5-MeV Columbia Van de Graaff accelerator was used for this purpose. The background due to proton-induced activity in the brass beam pipes was considerably reduced by collimating the beam to a fine spot, using tantalum collimators. The target chamber and Faraday cup were also lined with 3-mil tantalum for the same reason. The detectors were two 5-in.×5-in. NaI(Tl) detectors. The general setup is shown in Fig. 2.

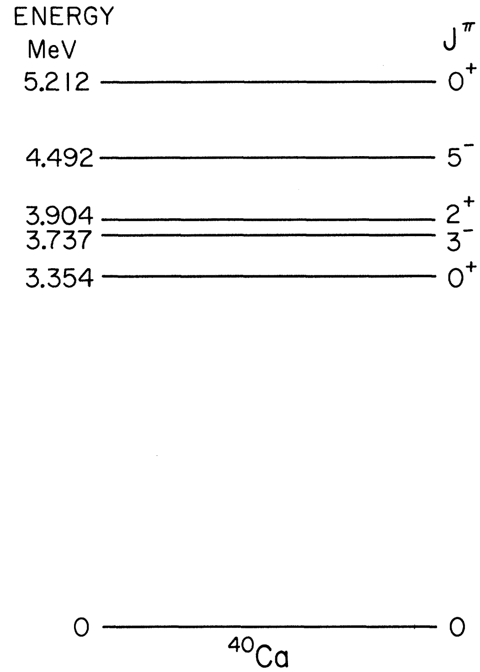

FIG. 1. Energy levels of Ca^{40}.

A natural metallic calcium target was used in our preliminary measurements. The target was prepared by evaporating natural calcium metal on a 5-mil tantalum backing. The target thickness was 0.2 mg/cm^2. On bombarding the target with 5-MeV protons, it was found that an induced activity produced in the reaction $Ca^{44}(p, n)Sc^{44}$ gave a sum peak in the 3.35-MeV region when the proton beam was turned off. The final measurements were therefore carried out using a target enriched to greater than 99.98% in Ca^{40}, in the form of $Ca^{40}O$. The target was 0.25 mg/cm^2 thick, on a 1.5-mil tantalum backing. A check for beam-induced activity in the target showed that no activity could be detected in the 3.35-MeV region of the sum spectrum. Furthermore, a γ-ray singles spectrum in one of the side channels showed that no γ rays from Sc^{44} were present.

With the target in the proton beam, the excitation function for the production of the first excited state was measured. Two large resonances were observed at proton energies 5.02 and 5.08 MeV, in agreement with the work of Bent and Kruse.[12] The

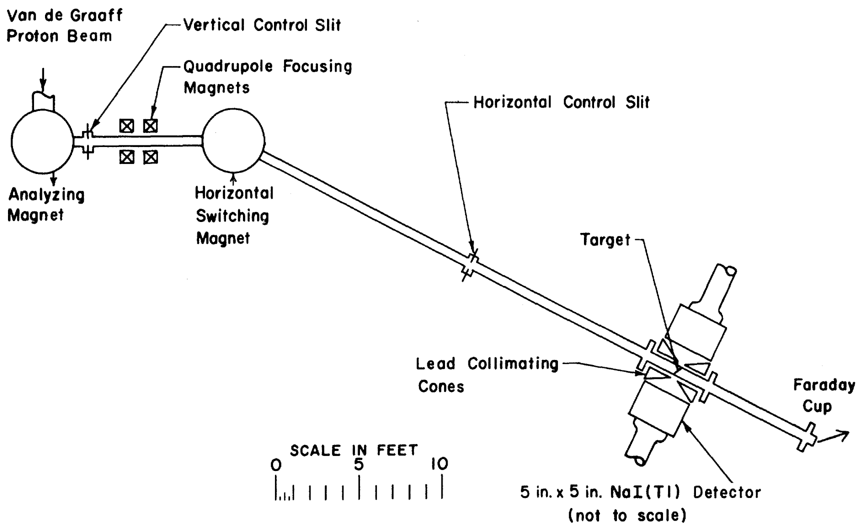

FIG. 2. Experimental layout for the investigation of double γ emission in Ca^{40}.

beam energy was fixed on the 5.08-MeV resonance, and the gated sum spectrum was recorded in a multichannel pulse-height analyzer for a preset integrator charge. The beam energy was then varied to a value slightly *below* or *above* the 5.08-MeV resonance, and the measurements were repeated. A sum peak at 3.35 MeV, which shows up more prominently when the beam energy is *on* the 5.08-MeV resonance than when it is either *below* or *above* it, would indicate the decay of the excited 0^+ state by double γ emission. The gated sum spectra with the beam energy *on*, *above*, and *below* the 5.08-MeV resonance are shown in Fig. 3. The number of 0.511-MeV coincidences was measured for a given integrator charge with the beam energy *on* and *below* the 5.08-MeV resonance to obtain the number of internal pairs emitted by the 0^+ excited state.

Figure 3 shows no evidence of double γ emission. An upper limit on the counting rate due to double γ emission was estimated, taking into account the resolution of the apparatus. The branching ratio was then calculated from

$$\frac{P_{\gamma\gamma}}{P_\pi + P_e} \leq \frac{N_{\gamma\gamma}}{N_{\pi 0}(1+Ne/N_{\pi 0})}\,\frac{(\Omega\epsilon_{p\,0.511})^2}{(\Omega\epsilon_{p\,E})^2}, \qquad (1)$$

where $P_{\gamma\gamma}$ is the probability for the decay of the 0^+ excited state by double γ emission, P_π is the probability for the decay of the 0^+ excited state by internal pairs, P_e is the probability for the decay of 0^+ excited state by conversion electrons, $N_{\gamma\gamma}$ is the upper limit on the number of double γ counts for a given integrator charge, $N_{\pi 0}$ is the number of observed 0.511-MeV coincidences for the same integrator charge, $(N_e/N_{\pi 0})$ is the ratio of conversion electrons to internal pairs, and $(\Omega\epsilon_{pE})$ is the photopeak efficiency[13] for the energy E, which corresponds to half the energy of the 0^+ excited state, and solid angle Ω.

Corrections were made to take into account the angular correlation between the two simultaneous γ rays – assuming an $(E1, E1)$ transition, the energy selection in the two side channels, which limits the portion of the double γ spectrum that would be observed, and absorption of the γ rays in the material between the target and the detector. Finally, a correction was also made to take into account the positrons that are not annihilated between the detectors.

The upper limit on the number of counts that might have been present in the 3.35-MeV region without being detected is estimated to be 450 for an integrator charge of 920 μC. The number of 0.511-MeV coincidences for the same integrator charge is 1.4×10^7. The upper limit on the branching ratio with respect to internal pairs – $N_e/N_{\pi 0}$ being negligible – was computed to be 4×10^{-4}.

B. Zr^{90}

The first excited 0^+ state at 1.75 MeV in Zr^{90} is produced in the β^- decay of Sr^{90} and Y^{90}. Since only a small fraction (<0.02%) of the β^- decay of Y^{90} feeds the first excited state in Zr^{90} (Fig. 4), it is necessary to use a strong source. However, the β^- decay of Sr^{90} and Y^{90} produce intense bremsstrahlung in the container and hence limit the source strength that can be used. To reduce the bremsstrahlung background, a beryllium container was used. The source used in this experiment was a fission product and was at least 7 yr old at the time of the experiment. The source strength was 240 μC. The detectors were two 3in.×3in. NaI(Tl) crystals. Without shielding the detectors, the gated background sum spectrum showed a peak in the 1.75-MeV region. The detectors were therefore shielded with 4 in. of lead (Fig. 5). With the

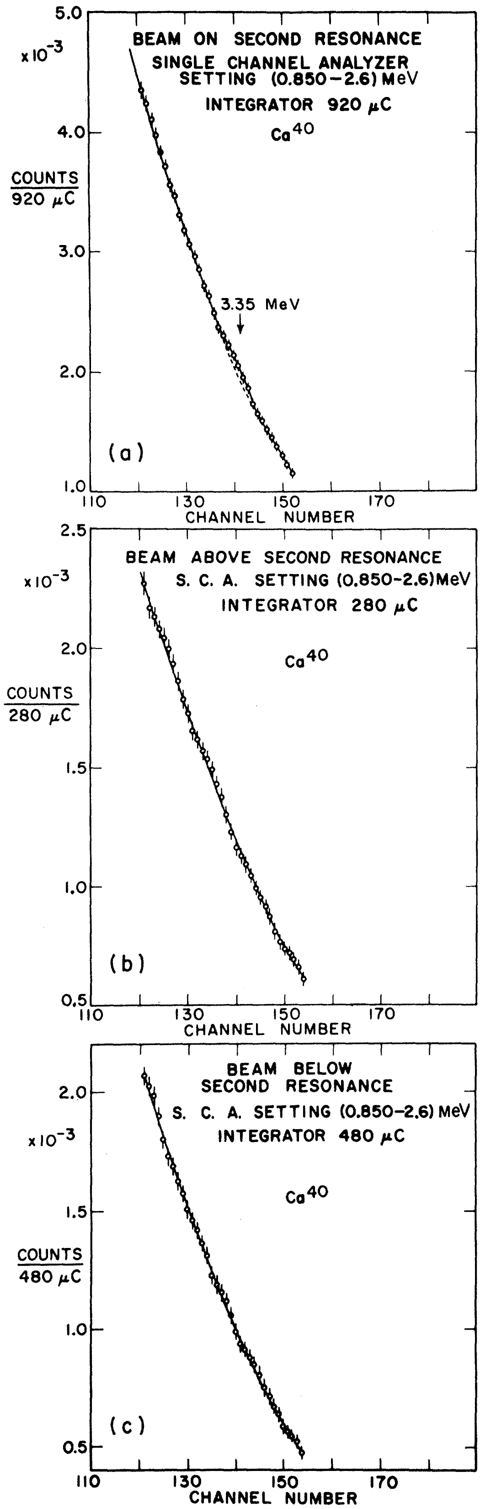

FIG. 3. (a) Gated total coincidence sum spectrum with beam energy *on* second resonance, and side channels open from 0.850 to 2.6 MeV. (b) Gated total coincidence sum spectrum with beam energy *above* second resonance. (c) Gated total coincidence sum spectrum with beam energy *below* second resonance.

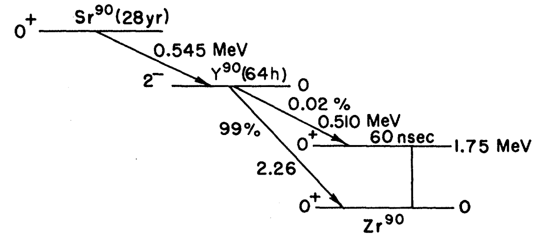

FIG. 4. Decay scheme of Sr^{90}.

shielding, the gated background sum spectrum was reasonably flat in the region of interest. To reduce gain shifts due to temperature variations, the detectors and the photomultipliers were housed in a constant-temperature box. Because of the small counting rate, data were accumulated for several hundred hours. The results are shown in Figs. 6 and 7. The gated accidental sum spectrum was recorded with a 110-nsec delay in one of the channels of the fast coincidence circuit. The number of pairs emitted by the 1.75-MeV 0^+ state was obtained from the number of 0.511-MeV coincidences in a given time interval.

Figure 7 shows a rather smooth spectrum decreasing with increasing energy and dropping to zero just below the 2.26-MeV endpoint energy of the Y^{90} β spectrum. A large fraction of this is probably due to double internal bremsstrahlung (simultaneous emissions of two photons in the electric field of the nucleus during β decay[7]). At the 1.75-MeV energy of the $0^+ \rightarrow 0^+$ transition in Zr^{90}, there is a suggestion of a peak. However, in view of the poor statistics and the absence of supporting evidence, such as measurements of the energy distribution and the angular correlation of the pairs of γ's in that sum-energy region, we cannot accept this as a definite observation of double γ emission. The counting rate under the peak was therefore used to set an upper limit on the branch-

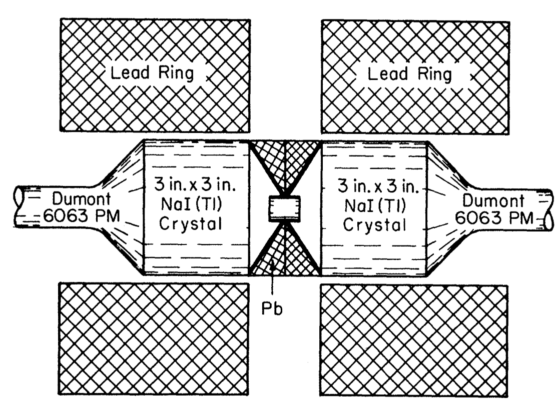

FIG. 5. Geometry of detecting apparatus. Sr^{90} is sandwiched between two Be blocks.

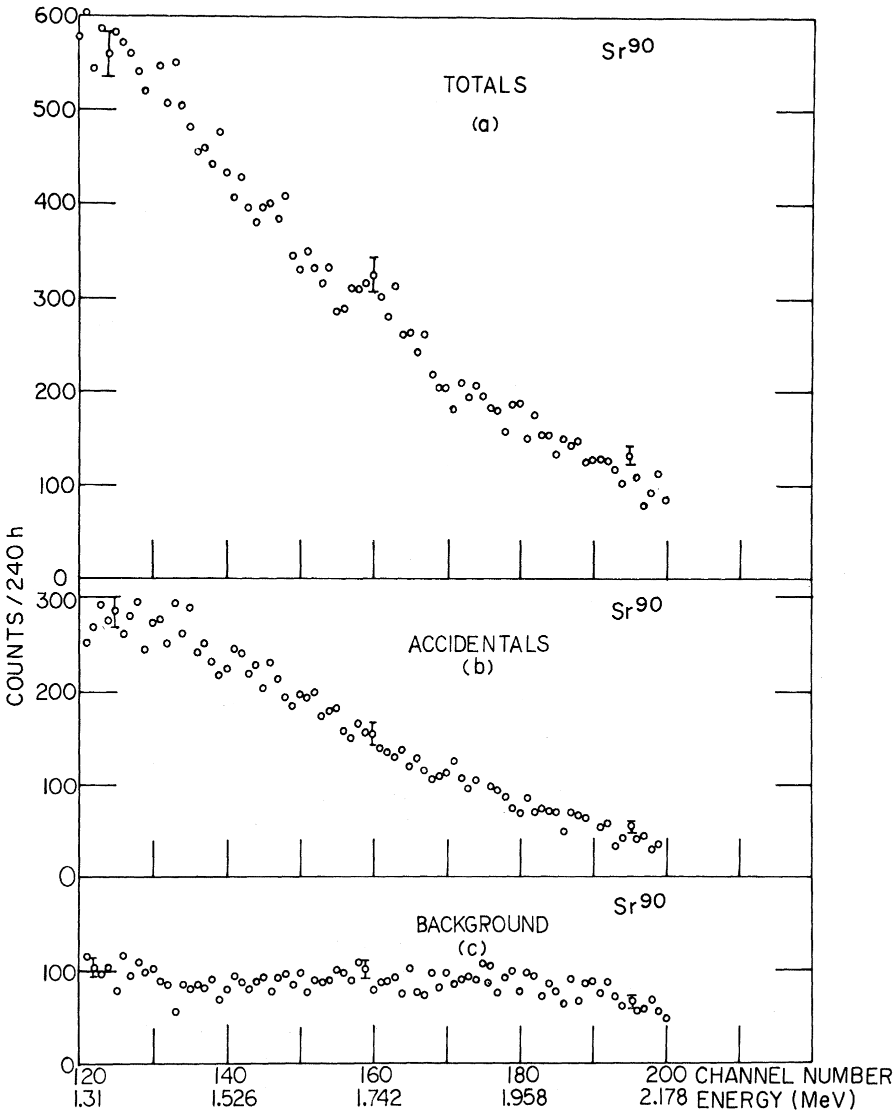

FIG. 6. (a) Gated total coincidence sum spectrum of Sr^{90} with side channels open from 0.600 to 1.5 MeV. (b) Gated accidental coincidence sum spectrum of Sr^{90}. (c) Gated background coincidence sum spectrum without source.

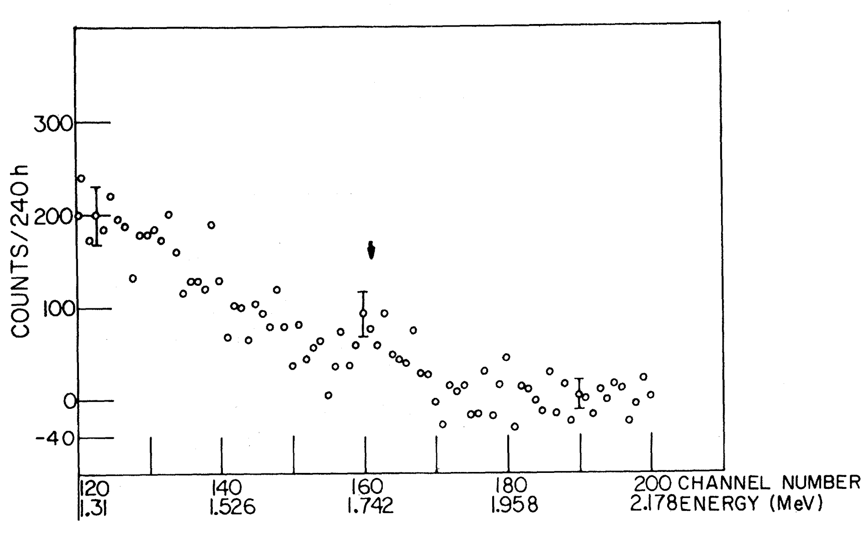

FIG. 7. Gated coincidence sum spectrum of Sr^{90}, accidentals and background subtracted. Expected peak position is indicated by arrow.

ing ratio for double γ emission with respect to the sum of internal conversion and internal pair creation. The branching ratio was computed from Eq. (1). The angular correlation between the two γ rays [assuming an $(E1, E1)$ transition], energy selection in the side channels, and absorption of the γ rays in the material between the source and the detectors were considered in computing the upper limit. The counting rate under the sum peak is 1.9 counts/h. The number of 0.511-MeV coincidences was 5.1×10^4 per hour. The ratio $N_e/N_{\pi 0}$ was taken as 2.38.[10] The upper limit on the branching ratio was computed to be 1.8×10^{-4}.

DISCUSSION

This work has confirmed other work,[6,7] which shows that the branching ratio to double γ emission in Zr^{90} is small. Our result is consistent with the estimate of Yoccoz,[14] who used spherical shell-model wave functions proposed by Talmi and Unna.[15] He obtains a value of 1.6×10^{-4}, consistent with our limit but greater than the limit obtained by Vanderleeden and Jastram.[7]

In the case of Ca^{40}, this work has considerably improved on previous upper limits[8] on the branching ratio to double γ emission and has shown that in this case, too, double γ emission is strongly hindered in comparison with single-particle estimates. It is now clear that in three cases discussed in the Introduction – O^{16}, Ca^{40}, and Zr^{90} – the upper limit to the branching ratio to double γ emission is of the order of 10^{-4}, or nearly 2 orders of magnitude smaller than single-particle estimates.

In the case of O^{16}, there is strong evidence[16,17] that the 0^+ first excited state is a member of a deformed rotational band. This identification depends not only on the spacing of states of increasingly high angular momentum, but on enhanced $B(E2)$ values and scattering experiments as well. Brown and Green[18] have incorporated this into a theory in which they regard the 6.05-MeV 0^+ state and the ground state in O^{16} as a linear combination of 0p0h, 2p2h, and 4p4h states, the latter two components being deformed. The enhanced $B(E2)$ rates[18] and the monopole matrix element leading to e^+-e^+ emission[19] are reasonably well explained by this model. Bertsch,[20] using the theory of Brown and Green, has calculated the branching ratio to double γ emission and has obtained a value of 9×10^{-5}, just lower than the current experimental limit.

No comparable theoretical calculations exist for Ca^{40}, Ge^{72}, and Zr^{90}, although Bertsch states that a similar theory should apply to the hindrance of the 2γ mode in Zr^{90}. However, in all these cases, one can identify the deformed rotational nature of the first excited state on the basis of level spacings. It seems reasonable to hope that a similar theory might hold for all four cases. More theoretical work on the cases other than O^{16} seems to be worthwhile, as well as further experimental work to determine values, not only upper limits, for the double γ branching ratio.

ACKNOWLEDGMENTS

We sincerely thank Dr. R. Chasman, who participated in this investigation and contributed invaluably to it at the very beginning. We are grateful to Dr. Y. K. Lee, Dr. L. W. Mo, and Dr. G. Sutter for criticism, discussions, and assistance rendered in various stages of the work.

---

† Work supported in part by the U. S. Atomic Energy Commission.

*Now at Lowell Technological Institute, Lowell, Massachusetts.

‡ Submitted in partial fulfillment of the Ph.D. degree, Columbia University.

[1] M. Göppert-Mayer, Ann. Phys. (Paris) 9, 273 (1931).
[2] R. G. Sachs, Phys. Rev. 57, 194 (1940).
[3] J. R. Oppenheimer and J. S. Schwinger, Phys. Rev. 56, 1066 (1939).
[4] J. Eichler and G. Jacob, Z. Physik 157, 286 (1959).
[5] D. P. Grechukhin, Zh. Eksperm. i Teor. Fiz. 32, 1036 (1957) [transl.: Soviet Phys. – JETP 5, 846 (1957)]; Nucl. Phys. 62, 273 (1965).
[6] H. Ryde, Arkiv Fysik 23, 247 (1963).
[7] J. V. Vanderleeden and P. S. Jastram, Phys. Letters 19, 27 (1965).
[8] G. Sutter, Ann. Phys. (Paris) 8, 323 (1963).
[9] D. E. Alburger and P. D. Parker, Phys. Rev. 135, B294 (1964).
[10] M. Nessin, T. H. Kruse, and K. E. Ecklund, Phys. Rev. 125, 639 (1962).
[11] H. Ryde, P. Thieberger, and T. Alvager, Phys. Rev. Letters 6, 475 (1961).
[12] R. D. Bent and T. H. Kruse, Phys. Rev. 109, 1240 (1958).
[13] S. H. Vegors, Jr., L. L. Marsden, and R. L. Heath, Phillips Petroleum Co. Report No. IDO-16370 (unpublished).
[14] J. Yoccoz, J. Phys. Radium 22, 685 (1961).
[15] I. Talmi and I. Unna, Nucl. Phys. 19, 225 (1960).
[16] E. B. Carter, G. E. Mitchell, and R. H. Davis, Phys. Rev. 133, B1421 (1964).
[17] R. W. Bauer, A. M. Bernstein, G. Heymann, E. P. Lippincott, and N. S. Wall, Phys. Letters 14, 129 (1965).
[18] G. E. Brown and A. M. Green, Nucl. Phys. 75, 401 (1966).
[19] E. Boeker, Phys. Letters 21, 69 (1966).
[20] G. F. Bertsch, Phys. Letters 21, 70 (1966).