Lifetime of the Second Excited Levels in Ni^{60} and Ti^{46}†

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The lifetime of the second excited levels in Ni^{60} and Ti^{46} was measured by beta-gamma delayed coincidence method employing a time-to-amplitude converter. The centroid shift of the beta-gamma delayed time distribution was measured with respect to the prompt time distribution due to a prompt gamma-gamma cascade which is present in each isotope. Simultaneous accumulation of the delayed and prompt time distribution on a split memory of a multichannel analyzer was made possible by the use of a third sorting detector. An upper limit of 5×10^{-12} sec was established for the mean life of both Ni^{60} and Ti^{46}. The mean life of the first excited state of Pb^{207} was also measured to be $(1.60\pm0.1)\times10^{-10}$ sec from the slope of the delayed time distribution.

I. INTRODUCTION

THE range of applicability of the delayed coincidence method of measurement of lifetime was greatly extended in the past few years mainly due to the advent of scintillators with faster decay constant and photomultipliers with smaller transit time spread. During this period the statistical nature of the processes in the combination of a scintillator and a photomultiplier became better understood.[1] Another important development was the new prompt comparison technique which eliminates the undesirable process of interchange of sources.[2] When one takes advantage of these new developments in the measurement of short lifetime, it is fair to say that an upper limit measurement can be extended to the region of 5×10^{-12} sec.

One immediate application of this new range would be to the levels which one describes as vibrational, such as the low-lying levels in Ni^{60} and Ti^{46}. The first excited states (2+) of these nuclei are accessible by Coulomb excitation[3] and nuclear resonance fluorescence measurement.[4] The present delayed coincidence investigation attempts to measure the lifetime of the second excited states (4+) in Ni^{60} and Ti^{46}, respectively.

The lifetime measurement of the second excited state (4+) in Ti^{46} is also significant in connection with the recent conflicting results of beta-gamma (circularly polarized) correlation in Sc^{46}. A large asymmetry in the correlation was reported by several groups,[5] but the absence of such an asymmetry was also reported by several other laboratories.[6] One proposed explanation of the disappearance of the asymmetry is due to attenuation of the beta-gamma correlation by the extranuclear effects which depend on the chemical structure of the sample.[7] The extranuclear effects could be important if the lifetime of the level were sufficiently long. If one can establish experimentally an upper limit of the order of 5×10^{-12} sec for the lifetime of this level, as the vibrational model predicts, then the possibility of attenuation due to ordinary extranuclear effects which have been known to be present in some of the gamma-gamma cascades becomes very small.

The previous best value for the mean lifetime of the second excited 4+state of Ti^{46} was an upper limit of 3×10^{-11} sec by delayed coincidence method.[8] It is highly desirable that this limit could be improved by at least an order of magnitude. However, for comparison reasons, it is advisable to apply this method of investigation to a nuclide whose decay scheme is similar to that of Sc^{46} but vibrational properties of its first excited level are well understood. The nuclide which was chosen for this comparison purpose is Co^{60}. It should be remarked here that the lifetime of the second excited state for Ni^{60} had been evaluated from a direct $E4$ excitation of the level in a high-energy electron scattering experiment by Crannell *et al.*,[9] together with the measurement of the small branching ratio of the cross-over transition from the 4+state to the ground state by Morinaga *et al.*[10] A mean life of 3×10^{-11} sec is obtained for the partial transition to the first excited level. This value corresponds to an enhancement factor less than unity, which is far too small to be understood on the basis of the vibrational model. The previous measurement of the mean lifetime of this level (4+) in Co^{60} by delayed coincidence method gave an upper limit of

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[10] H. Morinaga and K. Takahashi, J. Phys. Soc. Japan, 14, 1460 (1959).

5×10^{-11} sec,[8] which was not precise enough to resolve the problem.

The present investigation is also desirable from the point of view that it may provide a prompt comparison source for beta-gamma coincidence measurements. In many cases, it is desirable to perform delayed coincidence measurement between the beta particles and the succeeding gamma rays, or between the conversion electrons and gamma rays. The method of utilizing internally converted electrons has proven advantageous in many examples compared to gamma-gamma coincidence, because the conversion electrons require an extremely small dimension of scintillator where small dimension is important in improving the time resolution. Besides a very high detection efficiency is possible with conversion electrons. In all these instances it is desirable to have a convenient source which exhibits prompt beta-gamma coincidences for comparison with delayed beta-gamma coincidences being studied. Co^{60} is possibly such a candidate.

The present setup was also used to determine the lifetime of 570-keV level of Pb^{207} by measuring the logarithmic decay of the delayed gamma-gamma coincidences. The result is in agreement with the latest published data.[11] This provides an additional check on the performance of the present equipments.

II. PRINCIPLE OF THE METHOD

The lifetime of the second excited state of the daughter nucleus was determined by measuring the centroid shift of the time distribution of the delayed coincidence with respect to that of a prompt coincidence. A time-to-amplitude converter with a 6BN6 type coincidence circuit[12] was used. The usual procedure of measuring the centroid shift involves the use of a second source which gives the centroid of the time distribution of a prompt coincidence event. Since the two sources must be interchanged frequently, systematic errors due to changes in counting rate, energy, and position of the sources during the comparison are unavoidable. A new prompt comparison technique without the interchange of the sources was developed in our laboratory[2]; this technique is applicable to nuclei where a single source exhibits an interesting, delayed event as well as a prompt event for comparison, and the events can be distinguished from each other by detecting a third coincident particle. In this technique, first, a pair of fast scintillation detectors and a time-to-amplitude converter measure the prompt and delayed coincidences without discriminating one type of event from the other. Then a third detector, operated in coincidence with one of the main detectors, detects and identifies the third coincident particle. According to the sorting signal from the third detector, the delayed and prompt time distributions are separately stored in a split memory of a multichannel analyzer. The possibility of simultaneous accumulation of delayed and prompt coincidences eliminates many systematic errors including the one due to drift of electronics.

Under certain circumstances it is not necessary to distinguish both prompt and delayed events completely. In the present experiment, for example, the coincidence events which are observed in the time-to-amplitude converter are overwhelmingly due to the delayed events, so that the few prompt coincidence events which are being stored without an identifying signal from the third detector do not appreciably distort the delayed coincidence distribution. The third detector will sort out the prompt events only.

III. EXPERIMENTAL ARRANGEMENTS

The decay scheme of Ni^{60} and Ti^{46} has a gamma-gamma cascade. The mean life of the first 2^+ state is known by Coulomb excitation experiment to be 1.0×10^{-12} sec and 5.5×10^{-12} sec, respectively.[3] If the detectors do not discriminate the two gamma rays in cascade, which is possible when the energies of the two gamma rays are not very different from each other, then the centroid of the resulting time distribution will mark the "time-zero" with great accuracy.

The arrangement of the three scintillators is shown in Fig. 1, and the block diagram of the electronic system is given in Fig. 2. Detector 1 (D1) with a plastic scintillator 2 mm thick and 3 mm in average diameter for beta particles and detector 2 (D2) with a plastic scintillator 3.8 cm thick and 2.5 cm in diameter for gamma rays provide pulses to the time-to-amplitude

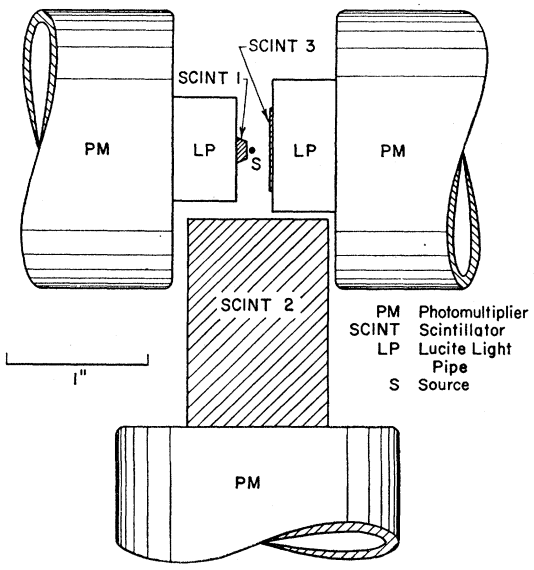

FIG. 1. Experimental arrangement of the detectors. All scintillators are plastic Naton 136. Photomultipliers 1 and 2 are 56 AVP, and 3 RCA 6810.

[11] S. Gorodetzky, R. Manquenouille, R. Richert, and A. C. Knipper, *Nuclear Science Series Report* (Printing and Publishing Office, National Academy of Sciences–National Research Council, Washington 25, D. C., 1962), Vol. 974, p. 79.

[12] R. E. Green and R. E. Bell, Nucl. Instr. 3, 127 (1958).

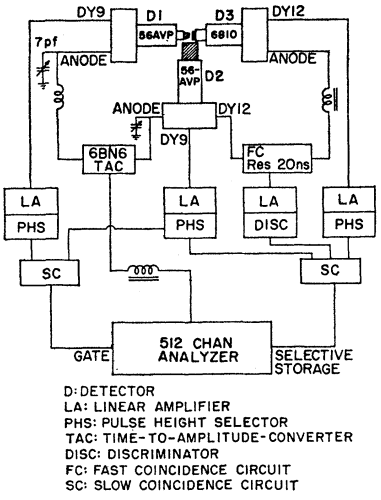

FIG. 2. Block diagram of the electronic system. The additional fast-slow coincidence system to provide selective storage routing signal is also shown.

converter (TAC). The output of TAC is predominantly beta-gamma delayed coincidence events with a small fraction of gamma-gamma prompt coincidences. The fraction depends directly on the ratio of the efficiency of D1 for gamma rays to that for beta rays. Detector 3 (D3) has a plastic scintillator which is only a fraction of 1 mm thick and 1.7 cm in diameter. The D3 and D2 provide pulses to a conventional fast-slow coincidence system with a resolving time of 20 nsec. The sorting signal indicates that a beta particle has been detected in D3 so that the associated coincidence between D1 and D2 must be a gamma-gamma event. In the absence of the sorting signal, the output of TAC is stored in one half of the split memory of a multichannel analyzer. The output of TAC which is accompanied by a sorting signal is selectively stored in the other half.

All possible coincidence processes in the present experimental arrangements are illustrated in Fig. 3. Figures 3(a) and (b) correspond to the double-coincidence processes which are stored without the sorting signal. Of the two processes, beta-gamma coincidence (a) is predominant while gamma-gamma coincidence (b) forms a small background due to the low efficiency of D1 as a gamma-ray detector. Figures 3(c) and (d) correspond to the triple coincidence processes which are selectively stored by means of the sorting signal from D3. Of the two processes, the gamma-gamma coincidence (C) identified by a beta particle in D3 is the main process, while the other process (d) forms the background. The background (d) is low because of the small efficiency of D3 as a gamma detector. In order to obtain a sufficient counting rate of the process (c), the efficiency of D1 as a gamma detector should not be unduly lowered.

Naton 136 plastic scintillators[13] in conjunction with Phillips 56AVP photomultipliers were used for the main detectors, D1 and D2. The output of anodes was shaped by limiters using 404A pentodes and 5-ft stubs of RG63U immediately preceding 6BN6 coincidence circuits of Bell and Green type.[12] One important feature of the present system was the use of a variable capacitor of the order of a few pF inserted between the anode of the 56AVP photomultipliers and the ground so that the size of the anode output could be controlled before it triggered the limiters. This way one can adjust the fraction of the anode pulse that is used to trigger the limiters. The recent analysis of the scintillator statistics combined with the multiplier time dispersion[1] showed that a relatively large fraction of the total anode charge should be used to define time in order to minimize the time jitter. In the present experiment, somewhere around 30% of the photoelectrons used to trigger the limiter gave optimum results. The high voltage for the multipliers, the quiescent current for 404A limiter, and the bias of the 6BN6 grids were optimized to give the best prompt resolution and linearity. The high voltage was set near 1800 V, with the first dynode voltage near 500 V. The scintillators and the photomultipliers were kept in a chamber which held the temperature constant within 1°C. The side channels for slow coincidences were of conventional designs. The time scale was calibrated by use of an air-core trombone delay line which was inserted between D1 and TAC.

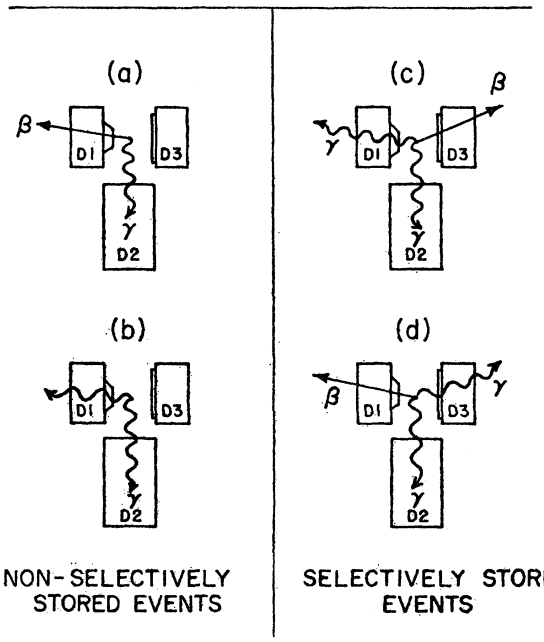

FIG. 3. Schematic diagrams of all possible double- and triple-coincidence events. Events in the left-hand column are stored in the multichannel analyzer without sorting signals, and therefore, correspond to the part which is used as delayed-time distribution. Events in the right-hand column are selectively stored by the routing signal derived from D3, and therefore, correspond to the part which is used as prompt time distribution. In each column, the top diagram represents the predominant main events while the bottom diagram represents the background.

[13] Supplied by Nash-Thompson Ltd., Hookrise South, Tolworth, Surrey, England.

IV. INVESTIGATION OF Ni^{60} AND Ti^{46}

Due to the very close similarity between the decay schemes of the parent nuclei, Co^{60} and Sc^{46}, almost identical procedures apply to the investigation of lifetime of Ni^{60} and Ti^{46} levels. The source was prepared by depositing a small fraction of a microcurie of activity to a circular area of 1 mm in diameter on a Mylar foil. The foil was mounted on an aluminum ring which can be accurately positioned with respect to D1 by means of a calibrated drum. In the case of the present arrangement with a 2×3 mm detector (see Fig. 2) and a point source which is located almost next to the detector, the mean time of flight of electrons with a velocity approaching C is 3×10^{-12} seconds, with the spread in time of flight smaller than this, assuming that most electrons are stopped in the first 1 mm of the path in the scintillator.

The measurement was carried out in a succession of 4-h runs. The delayed and prompt time distribution of a typical 4-h run for Ni^{60} and an 8-h run for Ti^{46} are shown in Figs. 4 and 5. The full width at half-maximum was typically 4.3×10^{-10} sec for Ni^{60} and somewhat larger for Ti^{46}. This number is to be compared with 3.8×10^{-10} sec which was the best value obtained with a higher and narrower energy window setting, disregarding counting rate requirement and the background consideration. With Ni^{60} the typical single counting rates were 160 and 20 counts/sec for D1 and D2, respectively. The rate of routing signal which was obtained from coincidence between D2 and D3 was less than 5/sec. The typical triple coincidence counting rate for prompt time distribution was 180 counts/each 4-h run, with approximately 100 times more double coincidences for the delayed time distribution. With Ti^{46} the counting rate was about 30% smaller. A total of 3000 triple coincidences were accumulated for each

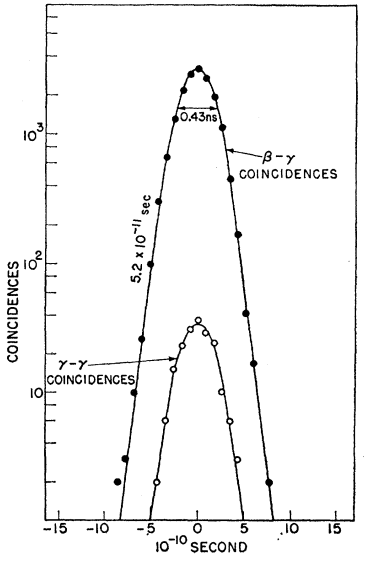

FIG. 4. Result of a typical four-hour run for beta-gamma delayed time distribution and gamma-gamma prompt time distribution obtained for Ni^{60}.

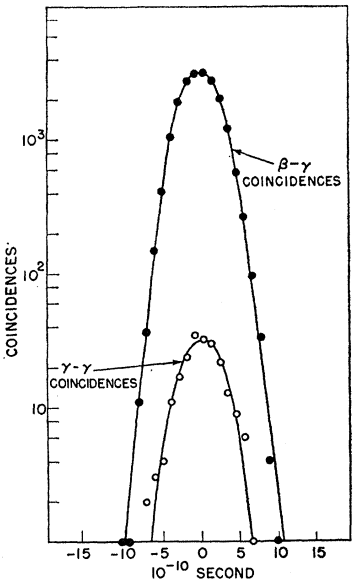

FIG. 5. The beta-gamma delayed time distribution and gamma-gamma prompt time distribution obtained by summing two sets of typical 4-h runs with Sc^{46}.

isotope. The effect of Compton scattering by D2 and D1 is negligible because the source was very close to D1 and D1 is very small. An experimental check was made, however, by placing a 3.5 mm lead absorber in front of D2 which should absorb 90% of the Compton scattered gamma rays, but allow 80% of the primary gamma rays to pass through. No appreciable effect was seen in the normalized triple-coincidence counting rate.

In order to determine the effect of the difference in profile of the beta and gamma spectra which are accepted by the energy selection window of D1, the centroid shift of the delayed time distribution with Ni^{60} was measured again with a much narrower window width. To compensate for the severe reduction in triple coincidence counting rate due to the narrowed window width, the scintillator in D2 was replaced by one 50 mm thick and 38 mm in diameter. This resulted in the increase of counting rate roughly by a factor of three, and also the deterioration of resolving time by 10% giving 4.8×10^{-10} sec. The experiment was repeated with the new arrangement for D2 and a narrower window width for D1 side channel which now accepted 5% of the top of beta spectrum compared to 25% for the previous runs. The observed difference in centroid shift is smaller than the experimental uncertainty; the detail of which is described in Sec. VI.

With the increased counting rate it was also possible to measure the difference in time-of-flight between electrons and photons when the source was moved away from D1 by a known small distance. The measurement of such a definite time interval provides a direct test that the arrangement described was sensitive enough to measure the time interval of the order of magnitude involved in the present experiment. This test was performed with the source-to-D1 distance of <0.1 cm and of 1 cm and with 25% energy selection

window for D1. The observed time shift is in good agreement with that predicted. (See Sec. VI).

V. INVESTIGATION OF Pb^{207}

The lifetime of the first excited level (570 keV) of Pb^{207} was measured with the present setup as an additional check of the performance of the delayed coincidence system and its calibration. The more reliable method of reading the slope of the delayed time distribution was employed. Plastic scintillator of 3.8 cm in height and 2.5 cm in diameter was used in D1 and D2 for this experiment. Ten microcuries of high-specific activity Bi^{207} was encapsuled in an aluminum cylinder 3 mm in outer diameter, 1 mm in wall thickness, and 5 mm in length. The delayed time distribution due to coincidences between 1060 keV and 570-keV gamma rays is shown in Fig. 6 together with the prompt time distribution obtained with Co^{60} gamma rays. The slope of the prompt time distribution was 4.8×10^{-11} sec. The half-life of the 570 keV level in Pb^{207} is thus determined to be $(1.10\pm0.11)\times10^{-10}$ sec, where the 10% uncertainty is a conservative estimate of the systematic error. This value is in good agreement with the latest value obtained by Gorodetzky *et al.*[11]

VI. RESULTS AND DISCUSSION

The new results of the measurement of centroid shift with Ni^{60} are $(-4\pm5)\times10^{-12}$ sec[14] and $(-0.3\pm5)\times10^{-12}$ sec for 25 and 5% window width of energy selection, respectively. The agreement between the two values with different window width indicates that the effect due to the difference in profile of the beta and gamma spectra accepted by the energy selection window is smaller than the experimental uncertainty. The uncertainty of 5×10^{-12} sec is a conservative estimate of the systematic errors, and this value corresponds to one tenth of the slope of the prompt time distribution which we assume as a reasonable index of the accuracy of the measurement by centroid shift. The effects due to the small mixture of beta-gamma coincidence in gamma-gamma coincidence and vice versa is understood to be absorbed in the above uncertainty. The statistical error alone is smaller than this number. The correction due to the small delay in the gamma-gamma cascade is less than 0.5×10^{-12} sec and is in the direction of reducing the lifetime and therefore is not applied. Recently, another measurement of the lifetime of this level in Ni^{60} was reported which gave 5×10^{-12} sec as the upper limit.[15] The result of the measurement of centroid shift with Ti^{46} is $(-4\pm5)\times10^{-12}$ sec. In this case the mean life of the first excited state is of the order of magnitude of the time interval we are interested in,[3] and there is an appreciable correction of 2.5×10^{-12} sec due to the indiscriminate use of the two gamma rays in the delayed beta-gamma coincidences. This correction is as mentioned above in the direction to reduce the upper limit; therefore, it is not applied. Although the magnitude of the measured centroid shift is within the experimental uncertainty, the minus sign of the data is used to indicate that the centroid is shifted as if the electrons are the delayed particles. The occurrence of minus sign, if we can attach any meaning to it, may be connected to the flight-time difference between gamma and beta rays.

The centroid shift for the case of Ni^{60} source which was positioned 1 cm away from the beta detector, D1, was $(2.2\pm0.5)\times10^{-11}$ sec. This value can be interpreted as due to the difference in flight time between photons and electrons of mean momentum equivalent

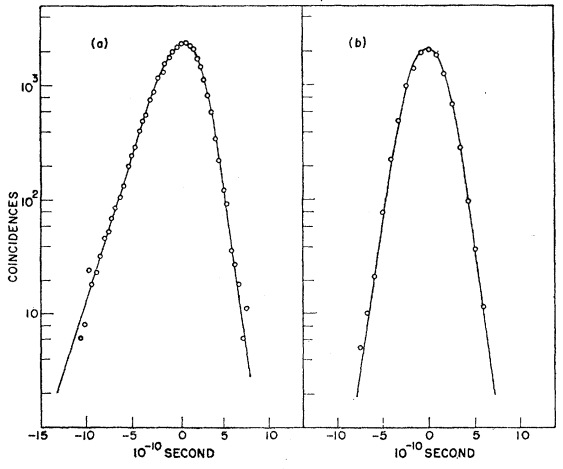

FIG. 6. (a) Delayed time distribution due to coincidences between gamma rays of 1060 and 570 keV in energy from Bi^{207} exhibiting the half-life of excited state at 570 keV in Pb^{207}. (b) The prompt time distribution due to Co^{60} gamma rays.

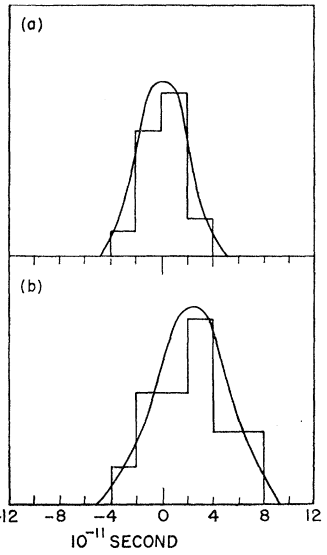

FIG. 7. Histogram of centroid shift measurements of the beta-gamma delayed time distribution for the case of Ni^{60}. The "time-zero" corresponds to the centroid of the gamma-gamma prompt time distribution. The top diagram was obtained when the source was in contact with the beta detector, and the bottom diagram was obtained when the source was 1 cm away from the beta detector. The change in centroid shift is due to the flight-time difference between electrons and photons over a 1 cm path.

[14] Y. K. Lee and C. S. Wu, Bull. Am. Phys. Soc. **7**, 340 (1962).

[15] A. Li and A. Schwarzschild, Phys. Rev. **129**, 2664 (1963).

to 0.6 V/c over a flight path of 1 cm. Histograms of the centroid shift measurements of the beta-gamma delayed time distribution with Ni^{60} are shown in Fig. 7 for both cases of the source immediately next to the beta detector and 1 cm away.

VII. CONCLUSIONS

No detectable centroid shift of the beta-gamma delayed time distribution was observed within experimental uncertainty for both cases of Ni^{60} and Ti^{46}, establishing an upper limit of 5×10^{-12} sec for the mean life of the second excited levels in these nuclei. An auxiliary check by measuring the time-of-flight of electrons over a definite path interval which yielded a results with the correct order of magnitude definitely demonstrates the capability and reliability of our experimental arrangement. The mean life of the first excited state of Pb^{207} was also measured to be $(1.60\pm0.1)\times10^{-10}$ seconds by reading the slope of the delayed time distribution. The upper limit for Ni^{60} and Ti^{46} is consistent with the predictions of the vibrational model, dispelling the suspicion aroused by an earlier measurement.[9,10] It has been shown[16] by angular correlation investigations that the attenuation of the nuclear alignment of the intermediate state by the extra nuclear effect is usually negligible if its lifetime is less than 10^{-11} seconds. Therefore, the upper limit of 5×10^{-12} sec obtained in this investigation suggests that if the beta-gamma (circularly polarized) correlation does attenuate with the change of the chemical composition,[7] then the origin of the perturbation effect must be quite different from those known in gamma-gamma angular correlations.

ACKNOWLEDGMENT

We wish to thank Dr. P. C. Simms for his constant interest. We are also most grateful to Dr. A. Schwarzschild for many helpful conversations.

[16] R. M. Steffen, Phil. Mag. 4, 293 (1955).

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Mean Lives of the First Two Excited States of P^{31}

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Nuclear resonance fluorescence techniques have been used to measure the mean lives of the first and second excited states of P^{31}. The exciting γ radiation was obtained by bombarding 99.999% pure phosphorous with 4.0- and 4.95-MeV protons. Self-absorption of the 1.26-MeV resonant radiation gives $\tau=(7.16_{-1.0}{}^{+1.3})\times10^{-13}$ sec. Similarly, $\tau=(4.2_{-1.0}{}^{+1.5})\times10^{-13}$ sec is found for the 2.23-MeV level. The angular distribution for resonant scattering of the 2.23-MeV radiation is consistent with the expected pure quadrupole nature of this $\frac{1}{2}-\frac{5}{2}-\frac{1}{2}$ transition. The angular distribution for resonant scattering by the 1.26-MeV level allows 4 values of the $E2/M1$ amplitude ratio, $\delta=-0.20\pm0.03$, -1.12 ± 0.08, 0.90 ± 0.06, and 5 ± 0.8. Only $\delta=-0.20$ is consistent with the principal work of others on angular distributions and linear polarization. Our lifetime combined with Coulomb-excitation results gives $|\delta|=0.40$, which is not compatible with our angular distribution.

INTRODUCTION

IN the past few years there has been considerable theoretical and experimental interest in nuclei in the 2s-1d shell, and some rather striking successes in correlating theory and experiment have been attained. One case where there has been only partial success is P^{31}. This paper is a contribution to the experimental knowledge of this nucleus, and reports values for the $E2/M1$ mixing ratio and mean life of the first excited state, and for the mean life of the second excited state.

Experimentally[1] it has been found that the ground and first two excited states have spins $\frac{1}{2}$, $\frac{3}{2}$, and $\frac{5}{2}$, and even parity, and that the second excited state decays directly (>97%) to the ground state. Their excitation energies are 1.265 and 2.232 MeV.

Since this work was begun, Booth and Wright[2] have measured these lifetimes by resonant scattering of bremsstrahlung. Their quoted errors of ±35% are

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† This research was supported by the U. S. Office of Naval Research.

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