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 Ni⁶⁰ 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 Ni⁶⁰ and Ti⁴⁶, 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 Pb²⁰⁷ was also measured to be $(1.60 \pm 0.1) \times 10^{-10}$ seconds by reading the slope of the delayed time distribution. The upper limit for Ni⁶⁰ and

Ti⁴⁶ is consistent with the predictions of the vibrational model, dispelling the suspicion aroused by an earlier measurement.^{9,10} It has been shown¹⁶ 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 betagamma (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.

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Mean Lives of the First Two Excited States of P³¹

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Nuclear resonance fluorescence techniques have been used to measure the mean lives of the first and second excited states of P³¹. 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 $\tau = (7.16_{-1.0}^{+1.3})$ $\times 10^{-13}$ sec. Similarly, $\tau = (4.2_{-1.0} + 1.5) \times 10^{-13}$ 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, $\delta = -0.20 \pm 0.03$, -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 theoretical and experimental interest in nuclei in the *2s-ld* 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.

Experimentally¹ 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 Wright² have measured these lifetimes by resonant scattering of bremsstrahlung. Their quoted errors of $\pm 35\%$ are

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

¹ P. M. Endt and C. Van der Leun, Nucl. Phys. 34, 1 (1962). 2 E. C. Booth and Kenneth A. Wright, Nucl. Phys. 35, 472 (1962).

larger than our usual errors, so that a continuation of our measurements seemed to be justified. Indeed, we seem to be in considerable disagreement with one of their values, as is discussed later.

EXPERIMENTAL DETAILS

The experimental procedures for measuring a lifetime by nuclear resonance fluorescence when the γ rays are produced in a nuclear reaction have been discussed in several publications.^{3,4} We mention the basic principle, that the resonance scattering cross section is proportional to the level width, and then confine the following description of experimental details to points of particular importance for the present measurements.

A. The γ -Ray Source

The high (p,n) threshold for P^{31} $(Q=-6.2 \text{ MeV})$ suggests the reaction $P^{31}(p,p')P^{31*}$ as a suitable source of γ radiation. Thick-target yield curves, obtained with the Bartol-ONR Van de Graaff accelerator, showed that the intensity of the 1.26-MeV γ ray from the decay of the first excited state was suitable at proton energies around 4.0 MeV. The 2.23-MeV radiation from the second excited state decay was still negligible at that energy, and reached a useable intensity only at around 4.95 MeV. It was necessary that targets be made from 99.999% pure phosphorous⁵ to keep the neutron background down to a tolerable level. Targets were prepared by pressing this material into the shallow gold cup of Fig. 1 with pressures of the order of 1500 kg/cm² . Durable targets resulted if the initial bombardment was with only a fraction of a microampere of beam and several hours were taken to reach the final value of $5 \mu A$. By visual inspection, the thin platinum foil

³ F. R. Metzger, in *Progress in Nuclear Physics*, edited by O. R. Frisch (Pergamon Press Inc., New York, 1959), Vol. 7, Chap. 2.
⁴ V. K. Rasmussen, F. R. Metzger, and C. P. Swann, Phys. Rev. 123, 1386 (1961), and Refs

over the phosphorous had then disappeared, but no change in yield, such as would be expected if $\approx 150 \text{ keV}$ of platinum were removed from the beam path, was observed. It is at least plausible that the top layer of the target was then some Pt—P compound.

A typical pulse-height spectrum of the γ radiation from this source for $E_p = 3.9$ MeV is shown in Fig. 2. The 1.78-MeV γ ray is from $P^{31}(p,\alpha)$ Si^{28*}. It is clear that silicon comparison scatterers should not be used. The intensity of the 2.23 γ observed would require a correction to a scattering measurement for the 1.26-MeV line, but, since the self-absorptions of the two lines are small and not too different, there is no appreciable correction for a self-absorption measurement. Any effect of weaker, higher energy, lines (these were observed, but not in detail) was estimated to be negligible. At 4.95 MeV, the intensity of the 2.23-MeV line had increased by about a factor of 10.

FIG. 2. Gamma-ray spectrum resulting from the inelastic scattering of 3.90-MeV protons by phosphorous. The 661-keV calibration source gives the line marked "Cs¹³⁷". The 1.28- and 2.23-MeV lines are from inelastic scattering, and the 1.78-MeV line from $P^{31}(p,\alpha)$ Si²⁸. The linear amplifier used limited at a value corresponding to channel 98, so that higher energy γ rays from background, $P^{31}(p,\gamma)$ S³², etc., appear in these top channels.

B. Scatterers, Absorbers, and γ **-Ray Detectors**

Figures 3 and 4 give typical experimental arrangements, and show one of the scatterers and two of the absorbers used. These were made by packing powdered phosphorous into thin walled (0.03 in.) aluminum containers. A second scatterer, not shown, had the same diameters as the one illustrated but was 1 in, long rather than 4 in. Comparison scatterers and absorbers were made from magnesium, with dimensions suitably modified to give the same number of electrons per cm², i.e., they were matched for Compton scattering.⁶

Since the ordinary electronic attenuation of the γ rays in the absorbers was quite considerable, and since the

⁵ Obtained from A. D. Mackay, Inc., 198 Broadway, New York, New York,

⁶ Nonresonant elastic scattering is negligible for these cases, as is emphasized by the observation that the counting rate with a magnesium scatterer was the same as that with no scatterer, within the 2% statistical error of the latter rate.

phosphorous was somewhat difficult to pack uniformly (the densities attained varied from 1.3 to 1.4), the absorbers were carefully checked for matching and uniformity with a Co⁶⁰ source. Differences between the phosphorous and magnesium absorber were found to be less than 1% .

Two NaI(Tl) crystals were used, the $1\frac{3}{4}$ -in.-diam by 2-in.-long crystal shown in Fig. 3 and the 3 -in. \times 3 -in. crystal shown in Fig. 4. The 3-in. crystal was first used in an arrangement similar to Fig. 4, except that only the 3-in.-diam tungsten attenuator of Fig. 3 was available. For the 1.26-MeV γ rays and a Mg scatterer (or no scatterer), a definite P absorber—Mg absorber difference was observed and was attributed to inadequate shielding of the crystal. Although in principle this could be subtracted out, the data were discarded and new data obtained when the larger attenuator of

FIG. 3. One of the experimental arrangements used with the 1.75-in.-diamX2-in. crystal.

Fig. 4 became available. No such undesirable effects were noted with the 2.23-MeV γ rays. The higher energy of the γ rays of interest and the small intensity of any still higher probably offer an adequate explanation, so that these measurements were not repeated.

Angular distributions for resonance scattering were measured with a modification of the arrangement of Fig. 3. The absorbers were dispensed with, the attenuator was moved somewhat further from the target, and the scatterer was moved quite a bit further from the target, so that the scattering angle could be changed by changing only the crystal-target distance. For the 1.26-MeV γ ray, the 1-in.-long scatterer and scattering angles of 90°, 126°, and 146° were used. For intensity reasons, the 4-in. scatterer was used for the 2.23-MeV *y* ray. The nominal scattering angles were 90°, 110°, 126°, and 149°.

FIG. 4. One of the experimental arrangements used with the 3-in. crystal.

C. Auxiliary Measurements

Certain incidental measurements concerned with detailed properties of the γ -ray source and detector were performed as described previously.^{3,4} A determination that the polarization of resonant-energy γ radiation from the source was negligible was made for the 1.26- MeV γ ray only, the method used being that described in Ref. 4, p. 1389.

The result of one of these auxiliary measurements is shown in Fig. 5, in which the relative cross section for resonant scattering of the 1.26-MeV γ ray is plotted against the mean angle with the proton beam of the γ rays striking the scatterer. The rectangular outline indicates the angular region (for forward angles) over

FIG. 5. Relative resonant cross section as a function of the angle between the proton beam and 1.26-MeV photons incident on the scatterer. As explained in the text, the kinematics of the (ρ, ρ') reaction and of the resonant scattering requires that there be no photons of resonant energy forward of $\approx 34^{\circ}$. The nonzero effect for 25° indicates that the velocity of the P^{31*} nuclei has been appreciably changed by scattering or slowing down within the radiative lifetime,

which resonant scattering should be observed.⁷ The occurrence of resonant scattering outside of this region is a clear indication that there has been scattering and/or large slowing down of the P^{31*} nuclei between the time they recoiled from the inelastic proton scattering and their radiative decay. The calculation of the scattering cross section from the scattered intensity is then more involved, since the incident photon intensity per eV will depend on details of the P³¹* energy loss.

FIG. 6. (a) Scattered spectra for the 1.26-MeV γ ray with scatterers and absorbers as labeled and the arrangement of Fig. 4. (b) P-Mg scatterer differences for P and Mg absorbers. The solid curves are derived from the spectrum obtained by exposing the same NaI crystal to direct 1.26 -MeV γ rays by subtracting a small, constant background and normalizing for a self-absorption of 15.7%.

⁷ In the laboratory system, velocities for the P^{31*} recoils will lie in a forward cone. Photons of the resonant energy will be emitted at $\approx 89^\circ$ to these velocities, and thus should *not* be observed in a related forward (or backward) cone.

A self-absorption measurement of the cross section, if feasible, avoids this difficulty.

Similar results were obtained for the 2.23-MeV level. We note at this point that a measure of the amount of slowing down is obtained by comparing the apparent lifetime from a scattering measurement with the selfabsorption lifetime. In this way, one finds that the number of photons per eV is increased by a factor of 2 for the 1.26-MeV line and a factor of 1.5 for the 2.23- MeV line.

RESULTS—1.26-MeV LEVEL

A. Self-Absorption

Data taken with various absorber and scatterer combinations and a comparison of the appropriate difference spectra with the line shape observed in direct exposure of the crystal to the 1.26-MeV line are shown in Fig. 6. Spectra showing a much larger range of γ -ray

FIG. 7. Angular distribution for resonant scattering of 1.26-MeV photons. The solid curve is the function $A_0+A_2P_2(\cos\theta)$ with $A_2/A_0=0.638$.

energies were taken in 100-channel subgroups of a 400 channel analyzer in some cases, mainly to ascertain that nothing unexpected or unfavorable was happening elsewhere in the spectrum. The 20-channel analyzer data, as in Fig. 6, however, was used in determining counting rates. The scattered intensity was taken as the sum of channels 8-12 of the Mg absorber, P-Mg scatterer difference. The resonant attenuation or selfabsorption is then the Mg-P absorber difference expressed as a percentage of the scattered intensity.

The experimental absorptions with the absorbers of Figs. 3 and 4 were $(10.7 \pm 1.9)\%$ and $(15.7 \pm 2.8)\%$. Taking the effective temperature of the absorber and scatterer as 300° K, the corresponding level widths are $(1.1\pm0.2)\times10^{-3}$ eV and $(0.77\pm0.17)\times10^{-3}$ eV. The weighted average of $(0.92 \pm 0.13) \times 10^{-3}$ eV corresponds to a mean life

$$
\tau\!=\!(7.16_{-1.0}^{+1.3})\!\times\!10^{-13}\;\mathrm{sec},
$$

The errors quoted for the widths are purely statistical but those on the mean life include a rough allowance for other uncertainties.

B. Angular Distribution and Mixing Ratio

The experimental points for the 1.26-MeV angular distribution and a least-squares fit of $A_0+A_2P_2$ (cose) to them are shown in Fig. 7. We found $A_2/A_0=0.638$. Figure 8 is a plot of A_2/A_0 versus δ , the $E2/M1$ amplitude ratio for the transition. The region of values allowed by the statistical errors on the angular distribution is shaded, and corresponds to

FIG. 8. Plot of A_2/A_0 versus δ , the *E2/M1* mixing ratio, for the transition $\frac{1}{2} - \frac{3}{2} - \frac{1}{2}$. The shaded area indicates the values allowed by the data of Fig. 7.

RESULTS—2.23 MeV LEVEL

A statistically meaningful self-absorption for the 2.23-MeV γ ray could be readily obtained only with the larger absorber as in Fig. 4. The data of Fig. 9 give $(8.6\pm2.1)\%$, corresponding to a width of (1.57 ± 0.40) X 10~³ eV, and a mean life

$$
\tau = (4.2_{-1.0}^{+1.5}) \times 10^{-13} \text{ sec},
$$

where the error on the lifetime incudes an allowance for other experimental uncertainties.

The angular distribution was consistent with that expected for a pure quadrupole, $\frac{1}{2} - \frac{5}{2} - \frac{1}{2}$ transition. It should be noted, however, that if any question arises as to these spin assignments, further work would be required on the angular distribution, and possible polarization effects looked into, before our work could be regarded as an unambiguous verification of the spin.

DISCUSSION

The values given by Booth and Wright² for the mean lives of these states are 2.2×10^{-13} sec for the first excited state and 4.5×10^{-13} for the second, with errors

FIG. 9. (a) Scattered spectra for the 2.23-MeV γ ray for various absorbers and scatterers. (b) P-Mg scatterer differences. The P and Mg absorber data have been added together. The points designated by an \times are from direct exposure of the NaI crystal to the 2.23-MeV γ ray.

of $\pm 35\%$. We agree for the second excited state, but disagree for the first by a factor of more than 2, which is completely inconsistent with our self-absorption measurement. Indeed, it may be quickly verified that the first excited state is longer lived than the second, rather than the contrary. For "thin" absorbers and scatterers (small self-absorption), the self-absorption SA can be written⁸

$$
SA = nl \frac{g_2}{g_1} \frac{\lambda^2}{4(\pi)^{1/2}} \frac{\Gamma}{\Delta},
$$

where nl is the absorber thickness in resonant nuclei/ cm^2 , g_2/g_1 are statistical weights for the excited and ground states, and Δ is the thermal Doppler width of the absorption line. Since the experimental arrangements for the two measurements were almost identical, one can easily find an approximate ratio of the two lifetimes as $\tau_2/\tau_1=0.5$, which is reasonably close to the result 0.59 for the more complete calculation, but in contrast with Booth and Wright's value 2.1.8a

Broude et al.⁹ and McCallum¹⁰ have used angular distribution and linear polarization measurements to determine values of the *E2/M1* mixing ratio for the first excited state. Broude *et al.* give $\delta = -0.20 \pm 0.06$ or -1.18 ± 0.24 , both of which are consistent with our values. McCallum gives $\delta = -0.25 \pm 0.15$ or $2.5 < \delta < 10$, strongly favoring $\delta \approx -0.2$. On the other hand, Andreev *et al.*¹¹ find, by Coulomb excitation, $\tau(E2) = 4.8 \times 10^{-12}$ sec. Using our value for the *Ml* component, this gives $|\delta|$ =0.40, which is hard to reconcile with our angular distribution or with the work of Broude *et al.* The bulk of the evidence seems to favor $\delta = -0.20$.

This value of *8* is used to obtain the partial widths of Table I which summarizes what is now known about these transitions. The Weisskopf single-particle widths Γ_W are calculated according to Wilkinson's article,¹²

TABLE I. Partial widths (in units of MeV, or 10^{-3} eV) and ratios to single-particle widths for the first two excited states of P 31 . The *E2/M1* intensity ratio for the 1.26-MeV transition is taken as 0.04. The Weisskopf single-particle values are according to Ref. 12, the $B(E2)/B(E2)$ ^{SP} values to Ref. 13. See also Ref. 14. The Coulomb excitation value is from Ref. 11.

Transition	$\Gamma(M1)$ (meV)		$\Gamma(E2)$ (meV) $\Gamma(E2)$ Coulomb	(meV) excit. $\Gamma(M1)/\Gamma_W B(E2)_{\rm SP}$	$B(E2)$ /
$1.26 \rightarrow 0$	0.88	0.035	0.137	0.02	0.9
$2.23 \rightarrow 0$ $2.23 \rightarrow 1.26$	< 0.024	1.57		< 0.0014	$\binom{(3.5)}{3.6}$

the $B(E2)/B(E2)_{SP}$ values from Eqs. (IV.3) and (V.1) of the review article of Alder et al.^{13,14} The qualitative features of Table I, i.e., that the *E2* transition is rather strong and the *Ml* transitions are relatively weak, have been known for some time and have been referred to in two theoretical discussions of the low-lying levels of P 31 . Broude *et al.¹⁵* applied the Nilsson model, and Thankappan and Pandya¹⁶ considered the coupling of the odd proton in shell-model states to vibrational excitation of the core. Both of these approaches were essentially successful in locating low-lying levels, but neither could claim too much success in predicting electromagnetic properties. In particular, the qualitative features of Table I are accounted for, although not too closely. Quantitative predictions are not given.

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¹³ K. Alder, A. Bohr, T. Huus, B. Mottelson, and A. Winther, Rev. Mod. Phys. 28, 432 (1956).

14 That both these transition strength units, which differ only by a model-dependent statistical factor, should be used is a consequence of the fact that for most lifetime measurements the natural reference seems to be to Weisskopf units, while those who measure the inverse transition by Coulomb excitation prefer $B(E\lambda)_{SP}$ especially when speaking of quadrupole enhancements. Note that in this second case, the transition-up and transitiondown values are related by the actual level spins involved, and that $B(E2)/B(E2)_{\text{SP}}$ is independent of this consideration, providing the equations of Ref. 13 are used properly.

15 C. Broude, L. L. Green, and J. C. Willmott, Proc. Phys. Soc. (London) 72, 1122 (1958).

¹⁶ V. K. Thankappan and S. P. Pandya, Nucl. Phys. 19 , 303 (1960).

⁸ F . R. Metzger, Phys. Rev. **103,** 983 (1956), Eq. (4).

^{8ª} *Note added in proof.* E. C. Booth has informed us that a recent, to be published, repetition of his measurements is in agreement with our results.

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¹² D. H. Wilkinson in *Nuclear Spectroscopy*, edited by

^{1960),} Part B, Chap. 5.