$Ca^{40}(p,\gamma)Sc^{41}$ Reaction

J. W. BUTLER
Nucleonics Division, U. S. Naval Research Laboratory, Washington, D. C.
(Received March 24, 1961)

Proton capture by Ca^{40} has been studied by means of observations of both the prompt gamma rays from resonance states in the compound nucleus and the delayed positrons from the decay of the ground state. Targets of CaO were prepared by the electrodeposition of Ca onto a Pt backing followed by the oxidation of the Ca and the purging of impurities by heat. These targets were bombarded by protons from a 2-Mv Van de Graaff accelerator, producing the reaction $Ca^{40}(p,\gamma)Sc^{41}$. The gamma rays from this reaction were observed with the use of a 3-in. diam by 3-in. NaI(Tl) crystal and a 256-channel pulse-height analyzer. Positrons from the decay of Sc^{41} were detected with the use of a thin plastic phosphor, 1.5 in. diam by 0.012 in. thick. Two resonances in the reaction were observed at bombarding energies of 650±5 kev and 1850±10 kev. Two other possible resonances were observed at 1550±15 kev and 1630±15 kev. The 650-kev resonance corresponds to an excited state in Sc^{41} at 1.723±0.011 Mev, has an integrated cross section of 0.02 ev barn (factor of 2 uncertainty either way), has a width of less than 5 kev, and involves a gamma ray whose energy was measured to be 1.71±0.03 Mev. The 1850-kev resonance corresponds to an excited state in Sc^{41} at 2.883±0.014 Mev, has an integrated cross section of 0.3 ev barn (factor of 2 uncertainty either way), has a width of less than 10 kev, and involves a gamma ray whose energy was measured to be 2.89±0.02 Mev.

INTRODUCTION

SINCE the nuclide Ca^{40} has closed shells for both protons and neutrons, observations of low-lying excited states for the system Ca^{40}+nucleon have particular importance for the independent particle model of the nucleus. The energies of gamma rays following thermal neutron capture by Ca^{40} have been measured by Adyasevich *et al.*[1] and others, and further information on energy levels in Ca^{41} has been obtained by Braams,[2] Bockelman and Buechner,[3] and others using magnetic analysis of proton groups from the $Ca^{40}(d,p)Ca^{41}$ reaction.

At the time the present experiment was undertaken, very little was known about the mirror reactions to those given above, involving proton capture. The $Ca^{40}(p,\gamma)Sc^{41}$ reaction had not previously been observed, and only one report had been published on the $Ca^{40}(d,n)Sc^{41}$ reaction. The present experiment concerns the detection of resonances in the $Ca^{40}(p,\gamma)Sc^{41}$ reaction, and the measurement of their parameters: width, cross section, and energies of emitted gamma rays.

The only other stable nuclides with double closed shells of equal numbers of protons and neutrons are He^4 and O^{16}. The $He^4(p,\gamma)Li^5$ reaction has never been observed, and no resonance has been observed in the $O^{16}(p,\gamma)F^{17}$ reaction below a bombarding energy of 2 Mev. The nonresonant capture for this reaction has an extremely low cross section and is attributed to a direct capture process.

EXPERIMENTAL PROCEDURE

The protons were supplied by the NRL Nucleonics Division 2-Mv Van de Graaff accelerator. A 90° magnetic beam analyzer, calibrated with the $Al^{27}(p,\gamma)Si^{28}$ resonance at 992 kev and controlled by proton magnetic resonance equipment, was used to define and control the proton energy. The gamma rays following proton capture were detected by the use of a 3-in. diam by 3-in. NaI(Tl) crystal mounted on a type 6363 multiplier phototube. The crystal was shielded from background radiation by approximately 2 in. of lead and was mounted at 0° with respect to the proton beam as shown in Fig. 1. The phototube pulses were analyzed with an Argonne-type 256-channel pulse-height analyzer.

Since the residual nuclide Sc^{41} is unstable, having a half-life of less than 1 sec, emitting positrons as it decays to Ca^{41}, excitation curves were determined with respect to delayed positron emission as well as prompt gamma-ray emission. The observation of delayed positrons has certain advantages over the observation of the prompt gamma rays. For example, two of the most prolific gamma-ray emitting reactions under proton bombardment are $N^{15}(p,\alpha\gamma)C^{12}$ and $F^{19}(p,\alpha\gamma)O^{16}$, both involving stable final nuclides. Since both these reactions involve the emission of energetic alpha particles, resonances arising from these reactions are much wider, on the average, than those involving (p,γ) reactions. Ex-

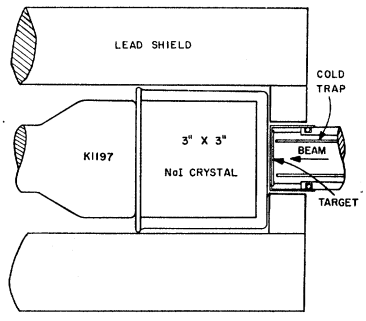

FIG. 1. Geometrical arrangement of the target holder (including the cold tube extension of the liquid-nitrogen cold trap) and the NaI(Tl) crystal for making the gamma-ray measurements. For the positron measurements, the NaI(Tl) crystal was replaced with a thin plastic phosphor. The arrangement possessed axial symmetry.

[1] V. P. Adyasevich, L. V. Groshev, A. M. Demidov, and B. N. Lutsenko, Atomnaya Energ. **1**, 28 (1956); J. Nuclear Energy **3**, 325 (1956).
[2] C. M. Braams, Phys. Rev. **103**, 1310 (1956).
[3] C. K. Bockelman and W. W. Buechner, Phys. Rev. **107**, 1366 (1957).

tremely small amounts of these contaminants, N^{15} and F^{19}, therefore can obscure the gamma-ray yield from (p,γ) reactions having relatively low cross sections. But since the product nuclides of the $(p,\alpha\gamma)$ reactions on N^{15} and F^{19} are stable, a delayed positron detector is completely insensitive to these contaminant $(p,\alpha\gamma)$ reactions.

For positron detection a thin plastic phosphor, Pilot-B, was mounted on a type-6292 multiplier phototube and placed in the same position as the NaI(Tl) crystal in Fig. 1. The phosphor had a diameter of 1.5 in. and a thickness of 0.012 in.

Because of the high beta background when the beam was on target, the following bombard-count cycling procedure was used. The beam was allowed to strike the target for 1 sec, then a shutter stopped the beam about 10 ft from the target, and for the next second the delayed positrons were counted. An automatic cycling device repeated this cycle many times for each bombarding energy used.

Target preparation was a difficult problem because of the chemical affinity of Ca for many elements and compounds, and also because of the very small cross section of the $Ca^{40}(p,\gamma)Sc^{41}$ reaction. Evaporated targets proved inadequate because of the large amount of "air" and other materials in the "vacuum" space carried onto the target by the evaporating Ca atoms. The amount of nitrogen and oxygen carried onto the target, for example, was several times greater than the total amount calculated to exist in the "vacuum" volume of the evaporator at any one time. The reason for this is not clear, but presumably there is a rapid interchange between "vacuum gas" and "wall gas," and the gettering action of the Ca forms a "sink" which "soaks up" the gas over a period equivalent to several volume interchanges. Nitrogen on the target presented a particular problem for gamma-ray measurements because of the relatively large cross section for the $N^{15}(p,\alpha\gamma)C^{12}$ reaction.

In previous experiments (e.g., the bombardment of nickel isotopes[4] by protons) electrodeposition techniques were developed and proved to be particularly suitable for producing extremely clean targets. Therefore procedures for the electrodeposition of Ca were investigated. Of course, normal aqueous solutions were unsuitable because of the chemical activity of Ca; so various organic solvents and Ca salts were tried. Ethyl alcohol and $CaCl_2$ gave the best results. $Ca(NO_3)_2$ produced a satisfactory Ca plating, but left too much occluded N^{15} on the target, leading to the $N^{15}(p,\alpha\gamma)C^{12}$ reaction mentioned above.

Prior to the electrodeposition of Ca for the target, the alcohol solution was purged of water content by an extra large electric current (the order of 100 ma) plating Ca onto a cathode different from the target blank. Then the 0.75-in. diam×0.010-in. thick Pt target blank was substituted for the previous cathode and a current the order of 10 ma was allowed to flow for the order of 1 min, the target thickness being approximately proportional to the product of electric current and time.

Special handling techniques were used for transferring the metallic Ca from the electroplating solution to a quartz spring balance for weighing and thence to the target holder. The electroplating system and the spring balance were inside a standard "dry box" with a dry helium atmosphere. Also inside the dry box was the target holder, including a valve; this valve was closed after insertion of the weighed target and kept closed until the assembly was placed in position on the Van de Graaff accelerator and the system on the other side of the valve evacuated.

The target was protected by a cold trap similar to the one previously described[4] except that the cold tube was retractable during the time the target-holder valve was closed and could be repositioned under vacuum near the target as shown in Fig. 1 when the target-holder valve was reopened.

Even when these metallic Ca targets were initially clean and protected by the surface at liquid-nitrogen temperature, they would, after a few hours of bombardment, pick up minute traces of fluorine, which would give rise to gamma rays through the $F^{19}(p,\alpha\gamma)O^{16}$ reaction. These gamma rays tended to mask the yield from the $Ca^{40}(p,\gamma)Sc^{41}$ reaction because the $F^{19}(p,\alpha\gamma)O^{16}$ reaction, like the $N^{15}(p,\alpha\gamma)C^{12}$ reaction, has resonances with relatively very large cross sections.

The most successful targets resulted from the deliberate oxidation of the electrodeposited Ca, forming the nonvolatile CaO. This property of CaO permitted the purging of impurities by high temperatures; and the relatively less chemical activity of CaO reduced the probability of recontamination. The targets used for the final data of the present experiment were purged of impurities by being "ignited" to white heat with an oxygen-natural-gas torch over a period of 5 to 15 min. Targets, which after long periods of bombardment picked up small amounts of fluorine or other contaminants, could be repurged by the reapplication of the igniting process. The oxygen, primarily O^{16}, contributed relatively little to the total observed gamma-ray yield because O^{16} is also doubly magic and has small, and well-known, proton-capture cross sections.

It was necessary to prevent the inner cone of the torch flame from touching the Pt disk, because if the inner cone did touch the Pt disk, it would carbonize the Pt, and the yield from the $C^{12}(p,\gamma)N^{13}$ reaction would tend to mask that from the $Ca^{40}(p,\gamma)Sc^{41}$ reaction.

The thickness of the targets could be controlled by the regulation of the amount of current and the time allowed for the deposition. Several different targets were used with several different thicknesses, the range of thicknesses for the CaO layer being 50 to 500 $\mu g/cm^2$ corresponding to energy losses of about 10 to 100 kev for incident protons of 1 Mev in penetrating the layer.

[4] J. W. Butler and C. R. Gossett, Phys. Rev. **108**, 1473 (1957).

RESULTS

Excitation curves for both gamma rays and positrons were first obtained with a relatively thick target (about 75 kev thick to 1-Mev protons) and with relatively wide bombarding energy intervals (about 50 kev). The bombarding energy range was from 500 to 1900 kev. There were only a few regions where the yield was appreciably above the background, and these regions were then reinvestigated with thinner targets and smaller bombarding energy intervals. The lowest energy region of interest (the region near 650 kev) was also investigated with a thicker target (about 100 kev) because it was difficult to establish with certainty whether or not a resonance actually existed in this region. (If the observed resonance peak changed its width in proportion to the target thickness, it was necessarily due to a nuclide uniformly distributed throughout the volume of the target and not due to a surface contaminant.) The net positron yield (total counts minus background counts) for this thicker target is illustrated in Fig. 2. The vertical bars on the datum points give the standard statistical deviations for the net counts. The sharp rise at 650±5 kev is due to the presence of a narrow resonance at that energy. The relatively long slope on the high-energy side is due primarily to energy-loss straggling of the protons in penetrating the target.

The gamma-ray yield at the 650-kev resonance was somewhat obscured by the background, but there was one peak in the net gamma-ray spectrum, and this peak was at 1.71±0.03 Mev. This energy is consistent with the Q value for the reaction, discussed later, and the assignment of this resonance to the $Ca^{40}(p,\gamma)Sc^{41}$ reaction is reasonably certain.

The next region of interest is the interval from 1500 to 1900 kev. The positron counts for this region at 15-kev intervals for a 15-kev thick target are illustrated in Fig. 3. The two peaks at 1550±15 kev and 1630±15 kev could be resonances in the $Ca^{40}(p,\gamma)Sc^{41}$ reaction, but their assignment is uncertain. The net gamma-ray yield from these resonances was not sufficiently large to enable the identification of the gamma-ray energies involved because of the higher gamma-ray background in this energy region compared with the region about 650 kev.

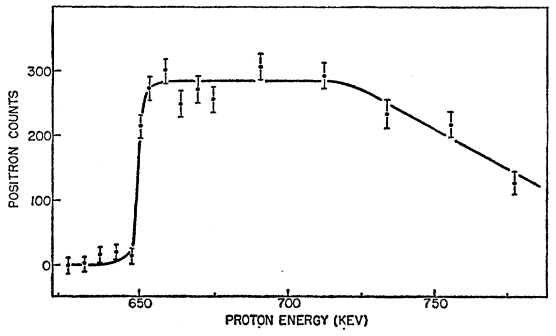

FIG. 2. Positron excitation curve for the $Ca^{40}(p,\gamma)Sc^{41}$ reaction in the vicinity of the lowest energy resonance observed. The target for these particular data was about 100 kev thick. Each datum point involved 1000 μcoul of protons in a repeated cycle arrangement described in the text. Background has been subtracted.

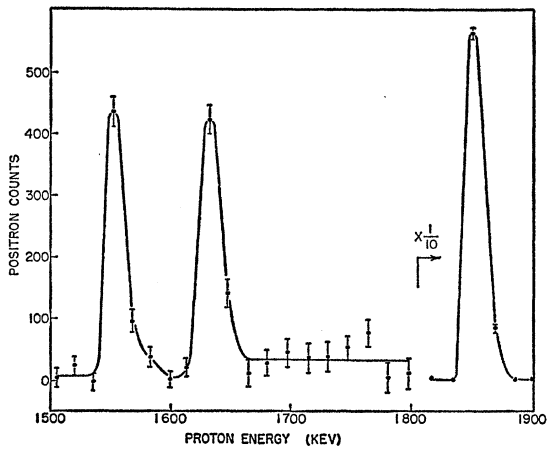

FIG. 3. Positron excitation curve for the $Ca^{40}(p,\gamma)Sc^{41}$ reaction in the upper energy region. The target for these particular data was about 15 kev thick. Each datum point involved 1000 μcoul of protons in a repeated cycle procedure described in the text. Background has been subtracted.

Even though great care was exercised in the preparation of the targets, apparently the "chemically pure" calcium used in the electroplating bath contained minute traces of magnesium because most of the known resonances in the $Mg^{24}(p,\gamma)Al^{25}$ reaction were evident in the yield curve. Al^{25} has a half-life of approximately 7 sec and is also a positron emitter. Even though these resonances were weak, they could conceivably have obscured some resonances in the $Ca^{40}(p,\gamma)Sc^{41}$ reaction.

Since the two possible resonances at 1550 and 1630 kev do not correspond to any resonances listed in a comprehensive table[5] of (p,γ) resonances, they are tentatively assigned to the $Ca^{40}(p,\gamma)Sc^{41}$ reaction. [There is a resonance listed at 1620 kev for the $Mg^{24}(p,\gamma)Al^{25}$ reaction, but its width, 36 kev, is too great to account for the resonance shown in Fig. 3 at 1630 kev, and the $Mg^{24}(p,\gamma)Al^{25}$ reaction has another resonance[6] at 1660 kev of even greater intensity than the one at 1620.]

The relatively strong resonance at 1850±10 kev is assigned with reasonable certainty to the $Ca^{40}(p,\gamma)Sc^{41}$ reaction. The gamma-ray yield at this resonance was sufficiently large to enable the measurement of the spectrum, displayed in Fig. 4. The three highest energy peaks correspond to the "total-absorption," "single-escape," and "double-escape" peaks of a 2.89±0.02-Mev gamma ray.

The peak at 0.88±0.01 Mev was present in both the

[5] J. W. Butler, U. S. Naval Research Laboratory Report 5282, April 9, 1959 (unpublished); *1959 Nuclear Data Tables*, edited by K. Way, National Academy of Sciences, National Research Council (U. S. Government Printing Office, Washington, D. C.), p. 134.

[6] A. E. Litherland, E. B. Paul, G. A. Bartholomew, and H. E. Gove, Phys. Rev. **102**, 208 (1956).

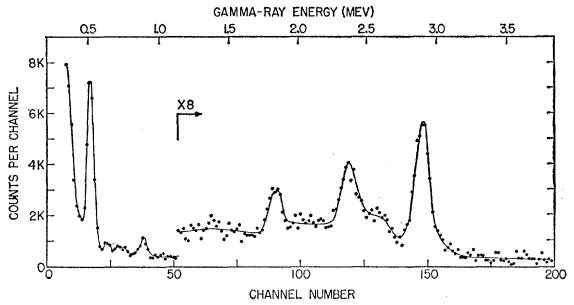

FIG. 4. Gamma-ray spectrum for the 1850-kev resonance. The spectrum was accumulated for 1000 μcoul of protons. The spectrum just below resonance was subtracted from the spectrum at resonance to obtain the net resonance spectrum shown here.

background spectrum and the resonance spectrum, but did not completely subtract out because of its great relative intensity. Its origin is not quite clear, but it probably does not arise from the $Ca^{40}(p,\gamma)Sc^{41}$ reaction. A possible source for this gamma ray is the $O^{17}(p,p'\gamma)O^{17}$ reaction. (There is a known energy level in O^{17} at 0.871 Mev, and the target consisted of natural CaO.)

The peak at 0.511 Mev is the annihilation radiation itself, resulting principally from the positron decay of the isotopes formed upon proton capture.

Since there is no evidence of a gamma-ray cascade (i.e., there appears to be only a single gamma-ray energy associated with the 1850-kev resonance), it appears likely that the 2.89-Mev gamma ray is a ground-state transition and thus corresponds to the energy of the excited state in Sc^{41}. If we make this assumption, the Q value for the $Ca^{40}(p,\gamma)Sc^{41}$ reaction is calculated to be 1.09±0.02 Mev.

Hinds and Middleton[7] have recently measured the $Ca^{40}(He^3,d)Sc^{41}$ reaction Q value to be −4.414±0.010 Mev from which we calculate, using the mass tables of Everling *et al.*,[8] the $Ca^{40}(p,\gamma)Sc^{41}$ reaction Q value to be 1.079±0.010 Mev in excellent agreement with the present results. Because of the smaller uncertainty in the measurement of Hinds and Middleton, we use their value in computing the energies of the excited states in the compound nucleus corresponding to the observed resonances. These results are summarized in Table I. The two possible resonances at 1550 and 1630 kev are included in Table I although their assignment to the $Ca^{40}(p,\gamma)Sc^{41}$ reaction is somewhat dubious.

The integrated cross section $\int\sigma dE$ for the 1850-kev resonance was obtained from the gamma-ray counts in the total absorption peak of the 2.89-Mev gamma ray. Various related factors, such as the geometry, attenuation, intrinsic crystal peak efficiency, number of bombarding protons, and proton-stopping power of the target, were taken into account. The angular distribution of the gamma ray was not measured because of the low yield, and the value of $\int\sigma dE$ was therefore calculated assuming isotropy. Since the crystal subtended about 40% of the total solid angle, and since symmetry about 90° is expected, about 80% of the total solid angle is accounted for. Therefore it is unlikely that the assumption of isotropy introduces an error greater than a factor of 2. The formula for calculation of $\int\sigma dE$ has been previously discussed.[4]

TABLE I. Summary of resonances in the $Ca^{40}(p,\gamma)Sc^{41}$ reaction and related information. A question mark indicates that the assignment of that resonance to Ca^{40} is doubtful. The value of the energy of the excited state in Sc^{41} was calculated from the Q value of the $Ca^{40}(He^3,d)Sc^{41}$ reaction of Hinds and Middleton and from the proton bombarding energy at resonance. The uncertainty in the integrated cross sections is a factor of 2 either way.

Proton bombarding energy at resonance (kev)	Energy of excited state in Sc^{41} (Mev)	Gamma-ray energy observed (Mev)	Integrated cross section $\int\sigma dE$ (ev barn)	Resonance width Γ (kev)
650±5	1.723±0.011	1.71±0.03	0.02	<5
1550±15?	2.590±0.018?	···	0.03?	<10?
1630±15?	2.669±0.018?	···	0.03?	<10?
1850±10	2.883±0.014	2.89±0.02	0.3	<10

The values of $\int\sigma dE$ for the other resonances were determined with respect to that for the 1850-kev resonance by the ratios of relative positron counts. For the 650-kev resonance (Fig. 2) the amount of material between the target and positron detecting plastic phosphor was the minimum—the amount shown in Fig. 1. For this resonance, background was low and the resonance counting rate was low, so a minimum of positron attenuation was desirable. But for the data of Fig. 3, the background and resonance counting rates were both higher; and a higher "signal-to-noise" ratio was achieved by the use of an extra absorber, the extra amount used being 0.050 in. of Al. The relative positron counting rate for the 650-kev resonance was corrected for the smaller amount of attenuation when $\int\sigma dE$ was calculated.

The resonance widths of Table I are simply upper limits to the conventional "full width at half-maximum." The actual values of Γ are probably much smaller, perhaps less than 1 kev for the 650-kev resonance.

DISCUSSION

Although the $Ca^{40}(p,\gamma)Sc^{41}$ reaction has not been previously observed, the expected Q value may be calculated from measurements on other reactions. Plendl and Steigert[9] measured the Q value of the $Ca^{40}(d,n)Sc^{41}$ reaction to be −0.57±0.05 Mev from which the Q value of the $Ca^{40}(p,\gamma)Sc^{41}$ reaction is readily calculated to be 1.65±0.05 Mev in disagreement with the value determined above from the data of the present experiment.

However, some very recent results at several different laboratories are in agreement with the Q value from

[7] S. Hinds and R. Middleton (private communication).
[8] F. Everling, L. A. König, J. H. E. Mattauch, and A. H. Wapstra, Nuclear Phys. **15**, 342 (1960).
[9] H. S. Plendl and F. E. Steigert, Phys. Rev. **116**, 1534 (1959).

the present experiment. Wegner and Hall[10] have measured the Q values of both the $Ca^{40}(d,n)Sc^{41}$ and the $Ca^{40}(He^3,d)Sc^{41}$ reactions. From their measurement of the $Ca^{40}(He^3,d)Sc^{41}$ reaction Q value, and the mass tables of Everling *et al.*,[8] we calculate that the $Ca^{40}(p,\gamma)Sc^{41}$ reaction Q value should be 1.02 ± 0.10 Mev; and from their measurement of the $Ca^{40}(d,n)Sc^{41}$ reaction Q value, we calculate that the $Ca^{40}(p,\gamma)Sc^{41}$ reaction Q value should be 0.90 ± 0.07 Mev. The former value is in good agreement with the present experiment, while the latter value is only marginally in agreement. Macefield *et al.*[11] have also recently measured the Q value of the $Ca^{40}(d,n)Sc^{41}$ reaction, and from their results we calculate that the $Ca^{40}(p,\gamma)Sc^{41}$ reaction Q value should be 1.080 ± 0.015 Mev in excellent agreement with the present results. The most recent measurement of the $Ca^{40}(He^3,d)Sc^{41}$ reaction Q value is that of Hinds and Middleton,[7] who obtained the value -4.414 ± 0.010 Mev from which we calculate the $Ca^{40}(p,\gamma)Sc^{41}$ reaction Q value to be 1.079 ± 0.010 Mev, also in excellent agreement with the present results.

Comparisons of excited states in Sc^{41} with other experiments are also possible. Wegner and Hall,[10] using the $Ca^{40}(He^3,d)Sc^{41}$ reaction, found excited states in Sc^{41} at 1.69, 3.35, 5.10, and 6.01 Mev. The lowest of these states corresponds to the resonance at 650 kev ($E_x=1.723$ Mev) in the present experiment while the others are beyond the energy range of the present experiment.

Macefield *et al.*,[11] using the $Ca^{40}(d,n)Sc^{41}$ reaction, found excited states at 1.709 ± 0.030 Mev and 2.476 ± 0.030 Mev. Again the lower state corresponds to the lowest energy resonance of the present experiment. The upper state would correspond to a resonance at 1431 ± 32 kev. No resonance was observed in the present experiment at this energy, but the background was relatively high in this energy region, so a resonance could easily have been missed. Since the present experiment was completed before the results of these other experiments were available, no particular effort was made to look for a resonance at about 1431 kev. Since the Van de Graaff accelerator is now being used to accelerate tritium, it is not feasible to repeat the experiment in the near future.

It is interesting to compare the density and intensities of observed resonances in this reaction with those observed in other reactions.[4] With the $Ca^{40}(p,\gamma)Sc^{41}$ reaction, two definite resonances were observed, and there is reason to believe a third one should have been observed as discussed above. These two observed resonances have $\int\sigma dE$ values of 0.02 and 0.3 ev barn. With the $Ni^{58}(p,\gamma)Cu^{59}$ reaction over the same energy range, 17 resonances were observed with an average $\int\sigma dE$ value of 0.30 ev barn, the maximum individual resonance value being 2.1 ev barns. With the $Ni^{60}(p,\gamma)Cu^{61}$ reaction, 54 resonances were observed with an average $\int\sigma dE$ value of 0.42 ev barn, the maximum individual resonance value being 2.3 ev barns. For the $Co^{59}(p,\gamma)Ni^{60}$ reaction, about 150 resonances were observed with an average $\int\sigma dE$ value of 1.9 ev barns. [The values of $\int\sigma dE$ for the $Co^{59}(p,\gamma)Ni^{60}$ reaction could not be obtained for individual resonances because of their close spacing; so the maximum individual resonance value is unavailable.]

The order of considering the above reactions has been in the direction of increasing numbers of nucleons outside closed shells or subshells for the target nuclides. The resonance density and average $\int\sigma dE$ values tend to increase as the number of nucleons outside the closed shells or subshells increases.

ACKNOWLEDGMENT

The author wishes to express his gratitude to Dr. C. R. Gossett for his assistance in performing the experiment.

[10] H. E. Wegner and W. S. Hall, Phys. Rev. **119**, 1654 (1960).
[11] B. E. F. Macefield, J. H. Towle, and W. G. Gilboy (to be published).