Transmutation of the Separated Isotopes of Neon by Deuterons

ERNEST POLLARD AND WILLIAM W. WATSON
Sloane Physics Laboratory, Yale University, New Haven, Connecticut

(Received May 15, 1940)

Under bombardment by 2.6-Mev deuterons, neon is observed to emit a large yield of protons which fall into at least four groups of energy change values 1.02, 2.15, 3.13, and 4.88 Mev. To assign these to the appropriate isotope, samples of neon enriched in Ne^{22} and Ne^{20} prepared by the thermal diffusion method were bombarded with the result that all groups are found to correspond to excited states of Ne^{21}. The reaction involved is $Ne^{20}(dp)Ne^{21}$. The maximum energy of the protons leads to the value 21.00017 for the mass of Ne^{21}. The bombarded gas was found to be radioactive and the presence of Ne^{23} due to $Ne^{22}(dp)Ne^{23}$ and Na^{21} due to $Ne^{20}(dn)Na^{21}$ was established. By absorption the beta-rays from Ne^{23} were found to consist of a single group of maximum energy 4.1 Mev which leads to a probable value of 23.0010 for the mass of Ne^{23}.

INTRODUCTION

APART from the discovery, by Snell,[1] of the formation of F^{18} from neon and of Na^{22} by Laslett,[2] the deuteron bombardment of neon has not been closely studied. In particular, no observations of proton groups have been made. Such observation is complicated by the fact that neon consists of three isotopes with percentage composition Ne^{20} 90 percent, Ne^{21} 0.27 percent, and Ne^{22} 9.73 percent. While the yield from Ne^{21} can be neglected in comparison with that from the two more abundant isotopes, there is no justification for assuming that Ne^{22} will not contribute appreciably to the protons observed, and therefore unless observations are made with some separation of the isotopes the groups observed cannot be assigned to the appropriate nucleus. Fortunately, the thermal diffusion coefficient of neon is relatively large so that separation by the thermal diffusion method is easy and gives large samples. Such samples have been made and bombarded in this work.

The Oppenheimer-Phillips reaction should give rise to Ne^{21}, Ne^{22}, and Ne^{23}, of which the last is unstable, decaying into Na^{23} with a half-life of 43 seconds.[3] The (d, n) type reaction yields Na^{21}, Na^{22}, both positron radioactive nuclei, and stable Na^{23}. The (d, α) reaction yields F^{18}, F^{19}, F^{20}, well-known nuclei. The work described here has been of two types: the first a direct study of the protons emitted in the Oppenheimer-Phillips reactions, and the second an observation of the induced radioactivity in the gas. The first part showed the existence of a considerable yield of protons, divided into several groups, all of which could be assigned to the formation of Ne^{21} in excited states. The protons due to the reaction $Ne^{22}(dp)Ne^{23}$ were masked by the more prolific yields from $Ne^{20}(dp)Ne^{21}$. The second part showed the formation of Ne^{23} and Na^{21}. The former emits an electron whose maximum energy was determined while the latter, already studied by Creutz, Fox, and Sutton[4] who produced it from Ne^{21} by the reaction $Ne^{21}(pn)Na^{21}$, was found to emit gamma-rays, a result to be expected since Ne^{21}, the resulting nucleus, is found to be rich in excitation levels.

EXPERIMENTAL PROCEDURE

Two features of the experimental work are of interest: the first the production of separated isotopes of neon, the second the bombardment and detection arrangement.

The production of rather large samples of neon gas with good separation of the isotopes by the method of thermal diffusion has already been briefly described by one of us.[5] Since, for one thing, neon atoms are about as close to elastic spheres as any available molecules, the thermal separation of their isotopes would be expected to proceed well. The gas circulated in a single thermal diffusion column two meters long made

[1] A. H. Snell, Phys. Rev. **51**, 143 (1937).
[2] L. J. Laslett, Phys. Rev. **52**, 529 (1937).
[3] E. Amaldi, O. D'Agostino, E. Fermi, B. Pontecorvo, F. Rasetti, and E. Segrè, Proc. Roy. Soc. **149**, 522 (1935); T. Bjerge, Nature **139**, 757 (1937); M. E. Nahmias and R. J. Walen, Comptes rendus **203**, 71 (1936).
[4] E. C. Creutz, J. G. Fox, and R. Sutton, Phys. Rev. **57**, 567 (1940).
[5] W. W. Watson, Phys. Rev. **57**, 899 (1940).

entirely of copper. As the inner hot surface a G.E. Calrod heater $\frac{1}{2}''$ in diameter was used. This heater was mounted concentrically in a vertical, water-cooled copper pipe of 1" I.D. Between the two surfaces a temperature difference of 400°C was maintained, the differential expansion being taken up by a sylphon bellows at the lower end.

At the lower end of the column a glass volume of about 300 cc was connected by means of two glass tubes, one of which was heated electrically. During the run the gas thus circulated convectively between this volume and the "heavy" end of the column. A metal leak at the upper end led to a glass manifold to which three break-seal bulbs each of about 125 cc capacity were attached. Two liters of pure neon gas were pumped into the column with a Toepler pump to a pressure of 1.5 atmospheres. Since the speed of separation should vary[6] as the square root of the pressure, with this fairly high gas pressure approximate equilibrium in the column should have been attained in somewhat less than a day. However, the gas was circulated for two days, after which it was bled off at the top at the rate of about 20 cc an hour into the three break-seal bulbs until the pressure in the column had dropped nearly to atmospheric. The lower end-volume was next isolated from the column by means of two stopcocks in each convector tube, and was then cut off. Finally the contents of the column were pumped back into the original flask for use in some of the bombardment experiments.

Mass-spectrographic analyses showed that the percentages of Ne^{20}, Ne^{21}, and Ne^{22} in this "heavy" gas were 84, 0.43, and 15.7, respectively, while in one of the "light" samples the corresponding percentages were 97.6, 0.163, and 2.28. The ratio of Ne^{22} in the heavy gas to that in the light was thus 6.88, a sufficiently large ratio to produce very appreciable difference in the activities in the two samples when subjected to deuteron bombardment. The Ne^{20} ratio in the two was still so nearly one, however, that in our experiments no differences in the intensity of the resulting proton groups could be detected. And in all samples the Ne^{21} content was so low that no reactions from the bombardment of this isotope could be assigned with any certainty. A subsequent separation of a like amount of gas was carried out in exactly the same manner. Although these second samples of separated neon were not analyzed with a mass spectrometer, we feel certain from our measurement of the relative amounts of Ne^{23} β-activity in the heavy and light samples that the difference in the Ne^{22} content of the two was at least as great as that of the first separation. To test the separation factor in this way a standard volume of neon was bombarded for two minutes with a known beam and the amounts of 43-second activities compared. A factor of between four and eight was estimated in this way, the inaccuracy resulting from difficulties in keeping the beam constant, a certain amount of O^{15} contamination due to residual air in the bombardment chamber and some short-lived effects due to Na^{21}. We estimate the most probable ratio of the Ne^{22} in the two samples to be six.

The arrangement for bombardment and proton detection is shown in Fig. 1. The beam is deflected from the dees of a cyclotron by the usual deflector arrangement and passes through an aluminum foil of 2.7 cm air equivalent[7] into a bombardment chamber as indicated. The foil, which is liable to have to stand pressure in both directions was held in place between rubber gaskets and gave no trouble at all. A system of baffles as indicated confined the effective part of the gas to a volume situated, on the average, 2.5 cm from the entrance foil and 8.5 cm from the exit foil (air equivalent 4.4 cm). The beam continued past the effective bombardment volume into a plate insulated by hard rubber which was used for measurement of the beam current. It is not perfectly clear from the diagram, but care was taken that no proton produced by bombardment of metal surfaces could "see" the exit hole so that no corrections need be made for surface contaminations. On pumping the gas out the yield dropped to the background value. The yield from the neon exceeds that from the same amount of air so that small amounts of air contamination do not affect the results. Such air contamination is liable to be present since the Hyvac pump used to evacuate the bombardment chamber was not run long enough to reduce the

[6] W. H. Furry, R. C. Jones and L. Onsager, Phys. Rev. **55**, 1083 (1939).

[7] 1 cm air equivalent = 1.52 mg/cm^2.

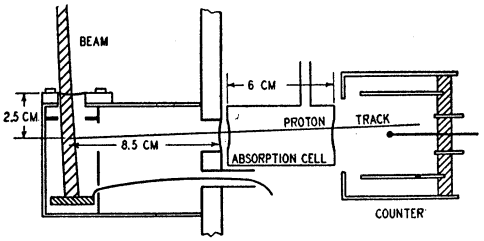

FIG. 1. Arrangement of bombardment chamber and detection apparatus. The beam enters through an aluminum foil and bombards neon gas in the enclosure. Protons emerging at 90° to the beam are detected after passing through various thicknesses of absorption.

air pressure below $\frac{1}{10}$ mm mercury. The protons from the bombarded gas pass into an absorption cell and through a foil changer (not shown) into a proportional counter attached to a conventional amplifier and either a direct or a scale-often recorder. We used pressures varying between five and twenty centimeters of mercury in the chamber and in each case made correction for the absorption introduced both in the incident beam and the emerging protons by the gas itself.

The question of the energy of the incident beam requires some attention. This can be measured in a variety of direct ways but the results are not wholly satisfactory. We found, by varying the pressure of air in the chamber that the maximum energy of the beam *emerging from the deflection plate* was 3.5 Mev, the calculated value. However, there is considerable spread (0.5 Mev) in the energy as measured in this way and it is not easy to decide on the appropriate value to use. We nevertheless find we can determine proton ranges accurately to 1 cm air or less in some cases, and so we used the proton groups from a layer of boron as a means of standardizing the beam energy, with the result that the value 3.2 Mev was decided on. Using this value we have measured the Q values for $H^2(dp)H^3$ and $O^{16}(dp)O^{17}$ with this apparatus and found agreement to within 0.1 Mev, the experimental error. The same figure has been used in many solid target experiments (e.g., $C^{12}(dp)C^{13}$ and $Na^{23}(dp)Na^{24}$) with accurate agreement between our work and that of previous workers. The effective energy in our gas bombardment cell varies between 2.45 and 2.75 Mev according to the pressure of gas used.

Absorbing foils were of aluminum, used in conjunction with the variable air pressure in the air absorption cell. The effective depth of the counter was measured with ThC′ alpha-particles, making allowance for the counting levels and the difference in ionization between protons and alpha-particles.

An account of the use of the cyclotron in observing proton groups is given in a previous paper from this laboratory.[8]

EXPERIMENTAL RESULTS

Proton groups

The first results of bombarding heavy and light samples are shown in Fig. 2. The counting level was set so as to include small "kicks"as well as large, and the two absorption curves are shown together. They have been adjusted as regards yield figures to lie together: the fact that the heavy curve lies at shorter ranges than the light is due to the greater gas pressure in the absorption cell which diminishes the energy of the beam. It can be seen at once that the two curves lie together indicating that the majority of the protons in both cases are due to the overwhelming excess of Ne^{20} in both samples. Two well marked groups appear at 24 cm and 58 cm range while a third is indicated at 35 cm range. To examine the structure of these protons in more detail we changed the counting level to include only large kicks and obtained the results of Fig. 3 where the 35-cm group now shows clearly and in addition a fourth group of still shorter range at 15 cm. The different samples again show no appreciable difference with the possible exception that a group at 40 cm is enhanced in the heavy sample. It should be emphasized that the shape of an absorption curve is strongly dependent on the counting level chosen and that erroneous conclusions can easily be drawn from a limited sample of data. We have taken many runs with the conditions maintained the same throughout as near as we can and in no case have the ratios of the yields of the four groups changed by as much as a factor of two, if we except the doubtful group at 40-cm range. Since our separation factor cannot be less than four we can say definitely that all four clearly marked

[8] E. Pollard, W. L. Davidson, Jr., and H. L. Schultz, Phys. Rev. **57**, 1117 (1940).

groups are due to one neon isotope. That the Ne^{20} isotope is responsible can be deduced in three ways. First, no great change in absolute yield is observed, which means that an isotope whose amount hardly changes is responsible. This is Ne^{20} which is never present to less than 80 percent of the total. Second, the maximum proton range fits well with the expected value from the accurately known masses of Ne^{20} and Ne^{21}. Third, the yield of radioactive Ne^{23} is much less than would result from the observed proton yield. Hence we conclude that the reaction $Ne^{20}(dp)Ne^{21}$ is responsible for the four groups. On this basis we can calculate the Q values for the various groups and determine the excited states of Ne^{21}. In Table I we give various values, obtained from different runs, for the various energy change values. The data are reduced by the procedure given in Livingston and Bethe's article. The values are not of equal weight but are included to give an idea of the amount of scatter possible. We consider our best values to be correct to 0.1 Mev in each case. The largest Q value leads to the value 21.00017 ± 0.00020 for the mass of Ne^{21}, which is to be compared with 20.99968 ± 0.00023 obtained by Bainbridge. The two limits of error nearly overlap. It seems likely that there is a small but significant difference. Such differences between transmutation and mass spectrograph values have recently been reported by Allison.

The group of greatest energy change is rather wider than the others. It may, perhaps, be multiple. We have examined this group in some

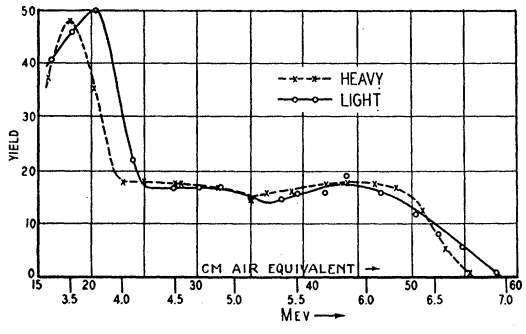

FIG. 2. Absorption curve for protons from heavy and light samples of neon without attempting great resolution. The two curves are almost the same, the difference being due to a slight increase in the bombarding energy for the light sample. All three of the groups are thus due to excited states of Ne^{21}.

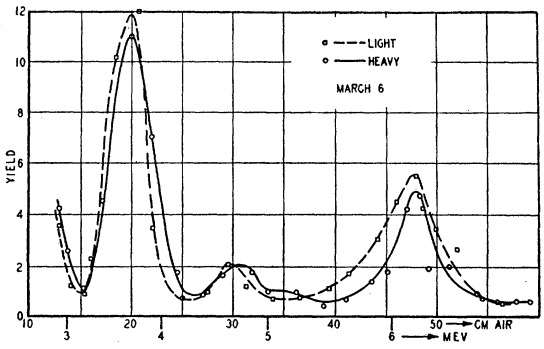

FIG. 3. Differential absorption curve for protons in which only particles near the end of their range are recorded. Four groups are present with an uncertain fifth. The similarity between the curves for heavy and light neon means that all four clearly marked groups are due to excited states of Ne^{21}. The slight hump ending between 5 and 6 Mev could possibly be due to protons from Ne^{22}.

detail in several runs but can find no consistent evidence for structure although we cannot deny the possibility. The difficulty encountered when the counting level is increased still more to increase resolution is the very high background due to neutron recoils, which produce large kicks also. If these were reduced by coincidence counting the resolution might be attempted.

Induced radioactivity

On withdrawing the gas after bombardment and testing it with a Geiger counter it was found to be radioactive decaying with a half-life of 43 ± 3 seconds. This agrees with the period found for Ne^{23} by Amaldi, *et al.*, and by Nahmias and Walen,[3] and so we ascribe it to the new reaction $Ne^{22}(dp)Ne^{23}$, a fact which is proved by the enhancement of the activity in the heavy sample. Our first experiments aimed at plotting an absorption curve for the beta-rays from Ne^{23}. About fifteen samples of ordinary neon were bombarded and quickly transferred to a perforated brass cell with a thin aluminum foil over the top. The brass was thick enough to stop any possible beta-ray. These fifteen samples were placed under a Geiger counter whose wall thickness was calibrated with radiophosphorus, and aluminum absorption foils placed over the cell. The decay of the sample was followed by taking several counts at zero absorption and in this way an absorption curve was gradually built up. This curve is shown in Fig. 4 where two sets of data are shown separately,

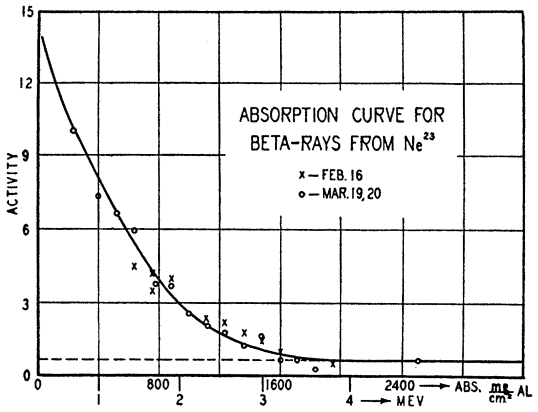

FIG. 4. Reduction of the number of counts in a Geiger-Müller counter as foils of aluminum are placed over a cell containing gas which had been in the bombardment chamber. An end point at 4 Mev is indicated.

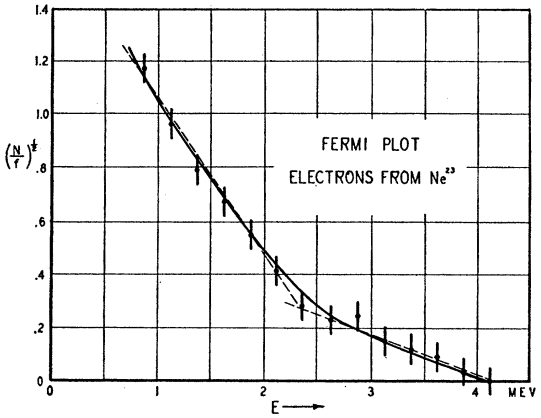

FIG. 5. The data for Fig. 4 reduced to give a Fermi plot. The curvature is probably due to back scattering and variable absorption in the counter. The possibility of two groups as indicated by the dotted lines is discounted by the absence of gamma-rays from this reaction. The end point is 4.1 Mev.

those marked with circles being taken with more counts. The curve appears to reach a maximum value around 4 Mev as determined by Feather's empirical relation

$$\text{Max g/cm}^2 = 0.543E - 0.16.$$

The maximum so obtained uses only the rather poor data where the counts are lowest. We therefore constructed a rough Fermi plot, by marking off the absorption into Mev intervals using the above rule (which checks quite well with magnetic bending work) and reducing the resulting distribution curve in the manner of Kurie, Richardson and Paxton[9] to give a Fermi plot.

TABLE I. *Various energy change values for the reaction* Ne^{20} (dp) Ne^{21}.

EXCITATION	ENERGY CHANGE VALUES (MEV)	BEST VALUE
Ground	4.74, 5.08, 4.80, 4.52, 4.92, 4.92	4.88
First	3.19, 3.17, 3.17, 3.00	3.13
Second	2.20, 1.99, 2.01, 2.23, 1.97, 1.91, 2.01	2.15
Third	1.01, 1.05, 0.98, 0.98	1.02

This is shown in Fig. 5. It will be seen to be either two straight lines or a curve. A calibration Fermi plot using V^{52} prepared by Mr. W. L. Davidson showed the same type of curvature which is to be expected in view of back scattering by the brass and also the variable basic absorption of the cylindrical counter. The Fermi plot however shows a definite intercept with the energy axis at 4.1 Mev which is therefore the appropriate upper limit.

Since the possibility that Na^{23} has an excited state to which Ne^{23} could decay is partly suggested by the shape of the Fermi plot, we looked for the resulting gamma-radiation. A rather weak gamma-ray was found but this did not decay with the 43-second half-life expected. In Fig. 6 the decay curve is shown. It has a half-life of 26 ± 3 seconds and is almost certainly to be identified with Na^{21} discovered by Creutz, Fox, and Sutton[4] and here produced by the reaction

$$Ne^{20}+H^2 \rightarrow Na^{21}+n.$$

As Na^{21} emits positrons this gamma-radiation might easily be due to annihilation radiation, but in view of the large number of excited states of Ne^{21} found we think it more likely that decay to an excited state takes place. An attempt was made to determine the absorption coefficient and values ranging between 2 and 3×10^{-25} for the absorption coefficient per electron in aluminum and brass absorbers were obtained. These indicate that gamma-rays of higher energy than annihilation radiation are present but the low yield and the short half-life made the measurements very difficult. It is probable that our technique of pumping off the bombarded gas operated selectively against sodium which would be deposited on the walls. We feel sure this also accounts for the absence of effects due to fluorine.

[9] F. N. D. Kurie, J. R. Richardson and H. C. Paxton, Phys. Rev. **49**, 368 (1936).

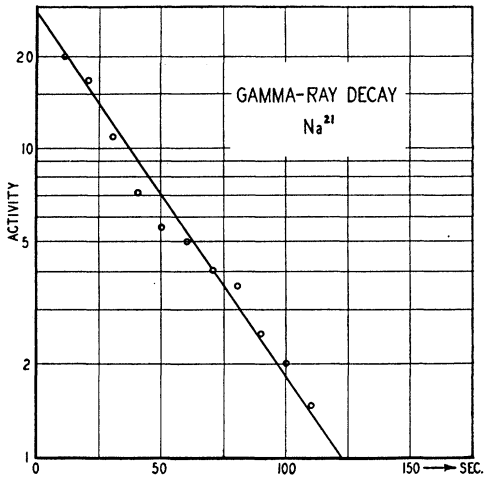

FIG. 6. Decay curve for the gamma-rays found. These are due to Na^{21} and probably consist of a mixture of annihilation radiation and 1.75-Mev gamma-rays from a transition in Ne^{21}.

It would be better to collect the active deposit on a quickly removable solid collector and study the radiations from the deposit.

DISCUSSION

In Fig. 7 we show energy level diagrams for Ne^{21} from our work and comparison values for Ne^{20} taken from Bonner's work on the $F^{19}(dn)Ne^{20}$ reaction.[10] The two are not particularly similar. The possible gamma-ray transitions are indicated. The first excited state for Ne^{21} has recently been observed by Murrell and Smith[11] who studied the $Na^{23}(d\alpha)Ne^{21}$ reaction. They obtain a short range group of alpha-particles corresponding to an excitation energy of 1.6 Mev to be compared with our value of 1.75 Mev. It is very interesting that in our reaction the yield in the first excited state is about one-fourth of that in the ground state, whereas in the mode of formation from $Na^{23}(d\alpha)Ne^{21}$ Murrell and Smith observe roughly equal yields in these two groups. This may be connected with the fact that in our experiments the Oppenheimer-Phillips process is taking place so that there is no compound nucleus in the usual sense.

The Na^{21} gamma-rays we observe are probably a composite of annihilation radiation and gamma-rays from an Ne^{21} nucleus formed in the first excited state.

The mass of Ne^{23} could be found from our measurement of the maximum energy of the beta-particles from Ne^{23} and the mass of Na^{23}. This latter is not definitely known. The work of Murrell and Smith above, together with Bainbridge's mass for Ne^{21} gives a value 22.9961 $\pm$0.0003. Experiments by Pollard and Brasefield[12] on the reaction $Ne^{20}(\alpha p)Na^{23}$ led to a published value of 22.9972$\pm$0.0002, which disagrees with them. Murrell and Smith suggest that a group of greater energy protons was missed. This is unlikely, as careful tests were made by Pollard and Brasefield, and also because no excited state of Na^{23} is apparent in the beta-ray spectrum of Ne^{23}. If the data on the reaction $Ne^{20}(\alpha p)Na^{23}$ are recalculated according to the procedure of Livingston and Bethe a lower value 22.9970 for Na^{23} is found. If now we take our figure for the mass of Ne^{21}, also derived from Ne^{20}, we can deduce from the data of Murrell and Smith the value 22.9966 which gives agreement within the limits of error. Using this we get for Ne^{23} 23.0010 $\pm$0.0005. A check on this is obtained by assuming that the extremely weak group at 41-cm

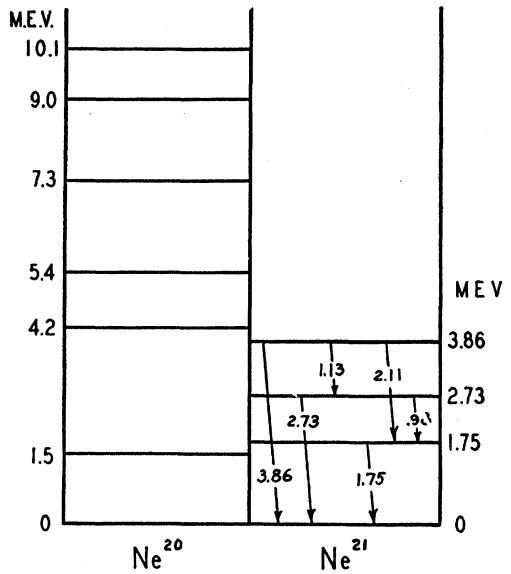

FIG. 7. Energy levels of Ne^{21} compared with those of Ne^{20}. The possible gamma-rays are indicated. The level at 1.75 Mev is to be compared with the figure 1.6 Mev given by Murrell and Smith.

[10] T. W. Bonner, Proc. Roy. Soc. **174**, 339 (1940).
[11] E. B. M. Murrell and C. L. Smith, Proc. Roy. Soc. **173**, 410 (1939).
[12] E. Pollard and C. J. Brasefield, Phys. Rev. **51**, 8 (1937).

range is due to protons from $Ne^{22}(dp)Ne^{23}$ and corresponds to the transition to the ground state. The Q value calculated is 3.6 Mev which gives a mass of 23.0012 for the mass of Ne^{23}. The agreement is reasonable but such data should not be pushed too far.

The proton yield for the $Ne^{22}(dp)Ne^{23}$ reaction can be calculated from the radioactivity observed. With our solid angle for counting we should obtain between ten and thirty per minute. Since when set for great resolution the counter only records about one-twentieth of the total protons passing through it, the size of group to be expected is about that of the indicated group at 41 cm so that it is not surprising that the heavy yields due to Ne^{20} mask the Ne^{22} group. It might be pointed out that without separated isotopes the temptation to ascribe the 33-cm group to Ne^{22} would have been irresistible and an incorrect level scheme for Ne^{21} thus deduced.

In conclusion we wish to thank Mr. W. L. Davidson, Jr., for help in general operations, Messrs. Harry Schultz and A. R. Tobey for keeping the beam in tune during many long hours, and Professor E. O. Lawrence for the gift of the vacuum chamber. The cyclotron construction has been greatly helped by a grant from the George Sheffield Fund.

JULY 1, 1940 PHYSICAL REVIEW VOLUME 58

On the Resonance Levels of Rhodium and Indium

J. HORNBOSTEL, *New York University, Washington Square College, New York, New York*
H. H. GOLDSMITH, *Columbia University, New York, New York*

AND

J. H. MANLEY, *University of Illinois, Urbana, Illinois*

(Received April 15, 1940)

Self-absorption curves as well as mutual absorption curves of the In and the Rh neutron capture levels have been measured. From the experiments, the following values were obtained for the absorption coefficients for self-indication K_s, the resonance cross sections σ_0, the natural widths Γ, the neutron width Γ_N and the spacing $|E_{0\text{In}}-E_{0\text{Rh}}|$ of the two levels: $K_s=11.5$ cm²/g, $\sigma_0=4100\times10^{-24}$ cm², $\Gamma=0.13$ ev, $\Gamma_N=3.5\times10^{-4}$ ev for Rh; $K_s=52$ cm²/g, $\sigma_0=23{,}000\times10^{-24}$ cm², $\Gamma=0.07$ ev, $\Gamma_N=1\times10^{-3}$ ev for In; $|E_{0\text{In}}-E_{0\text{Rh}}|=0.15$ ev.

THE total and the neutron width of a resonance level for slow neutron capture are quantities important in nuclear theory, since from them the probabilities of γ-ray emission and neutron re-emission of compound nuclei can be calculated. On the assumption that only one resonance level is responsible for the neutron capture, the total width Γ of the level may be computed from

$$\Gamma=2E_0^{\frac{3}{2}}E_{th}^{\frac{1}{2}}(K_{th}/K_0)^{\frac{1}{2}}. \tag{1}$$

Here, E_{th} is the thermal and E_0 the resonance energy, K_{th} the thermal absorption coefficient and K_0 the absorption coefficient at exact resonance, and it is assumed that $E_0\gg E_{th}$ and $E_0\gg\Gamma$. The neutron width Γ_N may be found from the relation[1]

$$\sigma_0=\frac{1.30\times10^6}{E_0}\left(1\pm\frac{1}{2j+1}\right)\frac{\Gamma_N}{\Gamma}, \tag{2}$$

if the cross section at exact resonance σ_0 is measured in units of 10^{-24} cm² and E_0 in ev. The factor $(1\pm1/(2j+1))$, with j designating the spin of the capturing nucleus, is unknown and shall, therefore, be included in Γ_N.

In an earlier work,[2] self-absorption measurements to determine σ_0 for the one-volt level of rhodium were performed with thick detectors.

[1] H. A. Bethe, Rev. Mod. Phys. **9**, 69 (1937) (Henceforth referred to as B.), Eq. (548).
[2] Manley, Goldsmith and Schwinger, Phys. Rev. **55**, 39 (1939).