PHYSICAL REVIEW VOLUME 143, NUMBER 3 MARCH 1966

Praseodymium-141 Photoneutron Cross Section to 65 MeV*

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(Received 20 May 1965; revised manuscript received 9 August 1965)

The cross section for the reaction $Pr^{141}(\gamma,n)Pr^{140}$ has been measured through 65 MeV. Yield curves of Pr^{140} radioactivity were analyzed by least-structure analysis. Eleven distinct resonances are resolved below 25 MeV with a total integrated cross section to 30 MeV of 1790±100 MeV mb. The cross section falls to zero in the region of 30 MeV and rises slightly with no resolved structure above 35 MeV. The integrated cross section from 30 to 65 MeV is 440 MeV mb.

I. INTRODUCTION

The electric-dipole resonance, or "giant resonance," in the interaction cross section of photons with nuclei has been the subject of intensive investigation for many years. The existence of appreciable cross section (or oscillator strength) for photonuclear interactions at energies well above the giant resonance has been inferred in many instances because of failure of the integrated cross section in the giant-resonance region to exhaust the dipole-oscillator-strength sum rule.[1–4] Danos[5] has suggested the existence of electric-dipole overtones based on a hydrodynamical model of the nucleus. The idea of $E1$ overtones has been considered within the framework of a nuclear shell model by Carver, Peaslee, and Taylor.[4] Gillet[6] and others have calculated transition probabilities for nuclear electric-quadrupole transitions based on a shell model with particle-hole interactions. The energies of such transitions would, in most instances, fall above the energy of the giant resonance.

More recently exhaustive calculations have been done regarding the structure of the giant resonance,[7,8] and the effect of $E2$ photon absorption on the resonance shape.[9–11] Again the effect is predicted to be a broadening or splitting of the resonance (even for nuclei with little or no static deformation) and oscillator strength at higher energies. These calculations have been applied specifically to odd-A nuclei[12] with the tentative conclusion that oscillator strength should be distributed in the giant-resonance region in a manner not dramatically different from that of even-A nuclei.

Until recently high-resolution data regarding structure in the giant-resonance region, and more particularly, above the giant-resonance region has been sparse. This has been due primarily to difficulty associated with extraction of nuclear cross sections from yield-curve measurements made with electron accelerators providing only bremsstrahlung spectra of photons. Various methods for resolution of this problem have been proposed in the past.[13,14] Such methods do not provide stable solutions for the cross sections above the giant resonance. The use of positron annihilation in flight or (p,t) reactions to provide a flux of fairly monochromatic gamma rays has been successful,[15–17] but such methods have not been extended to energies above about 30 MeV.

A procedure for extraction of nuclear cross sections from bremsstrahlung-induced yield curves over large energy ranges has been developed in this laboratory by Cook.[18,19] The method is based on a smoothing technique for solution of Volterra equations of the first kind.[20]

During the past several years repeated measurements of the $O^{16}(\gamma,n)O^{15}$ reaction cross section from threshold energy to 65 MeV have been made using bremsstrahlung photons from the Iowa State University synchrotron.[21–24] Each measurement has revealed appreciable structured cross section above the giant resonance and improved measurement techniques have made possible the resolution of essentially all known structure within the giant resonance.[24] (Reference 22 contains details of the data extraction technique in addition to early results.) The O^{16} measurements were made by counting

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* Work was performed at the Ames Laboratory of the U. S. Atomic Energy Commission.

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the annihilation radiation from the $\beta+$ activity of O^{15} nuclei resulting from the photoneutron reaction. During this sequence of measurements, yield curves for the reaction $Pr^{141}(\gamma,n)Pr^{140}$ were also made from threshold energy to 65 MeV. Since Pr^{140} is also a $\beta+$ emitter with mean lifetime 3.4 min, these yield curves were measured in precisely the same manner as were the O^{16} yield curves, with only samples, irradiation times, and counting times changed. (This lifetime is very close to the 2.0-min lifetime of O^{15}.) In these measurements 120 yield points were measured for each yield curve in increments of 0.5 MeV.

In addition to the wide energy-range measurements, data were taken on Pr^{141} in 0.125-MeV increments, the yield curves just spanning the giant-resonance region. These measurements were made in an effort to resolve structure in the giant-resonance region. Such structure was thought to be present because of apparent asymmetry in the giant-resonance curve both in the abovementioned measurements and in measurements reported by Rice, Bolen, and Whitehead.[25]

II. EXPERIMENTAL PROCEDURE

With exception of samples and irradiation times the experimental procedures used in measurement of the yield curves are described extensively in Ref. 24. A summary of the techniques will be presented here.

Samples were disks of pure Pr^{141} 1.9 cm in diameter and 1.59 mm thick. Thin samples were used to minimize degradation of the bremsstrahlung spectrum in passing through the samples. The sample thickness represented less than one-tenth of one attenuation length over the entire energy spectrum above threshold energy. The samples were prepared shortly before each experiment and carefully weighed before and after each experiment. Oxidation during the measurement of a set of yield curves, a period of about two weaks, was found to be negligible. Fifteen such samples were available for the extended energy range experiment and thirty samples were used in the experiment covering the giant-resonance region. Every effort was made to fabricate uniform samples. In order to eliminate errors due to sample variation, each sample was irradiated and counted at 30 MeV a minimum of three times before the yield curves were measured. On the basis of these measurements a table of sample normalization factors was prepared and used during the experiment. Corrections due to sample nonuniformity were less than 4%.

The samples were irradiated in eclipsing geometry with bremsstrahlung radiation from electrons striking an internal synchrotron target. The photon beam was, in this instance, collimated to a half-angle of 0.021 rad. After passing through the sample, the photon beam was monitored by a National Bureau of Standards type P2 ionization chamber and an activity computer was used in conjunction with the ionization-current electrometer.

Samples were irradiated for 5 min, one minute was allowed to elapse during which the sample was removed to the radioactivity counting system, then the annihilation photons from Pr^{140} were counted for 5 min. The counting system was identical to that used in the O^{16} experiments.[24] At bombarding energies greater than 19 MeV more than 3×10^5 counts were recorded in each detector so that small count rate, or dead time, corrections were necessary.

Since data were taken at energies above the $(\gamma,2n)$ and $(\gamma,3n)$ thresholds of Pr^{141}, it was necessary to correct the radioactivity count for the presence of these longer lived activities (4.5 and 2.0 h, respectively). Independent measurements were made on these activities and a set of correction factors established. Maximum correction was 1.5% at 65 MeV. Because of the presence of these activities it was found necessary to precount each sample before actual irradiation and counting. For data points at very low energies, where small count rates were anticipated, certain samples were set aside and never used above the $(\gamma,2n)$ threshold. This minimized the residual activity correction necessary at these points.

Previous to each experiment careful checks were made of the photon beam position as a function of energy. Machine adjustments were made such that beam movements were reduced to a negligible amount.[24]

Absolute Cross-Section Measurement

Absolute-cross-section measurements were performed by irradiating carefully measured samples at 21 MeV and counting the radioactivity in a 4×4-in. well-type NaI(Tl) crystal. The resulting activity spectrum was recorded on a multichannel analyzer. The efficiency of this detection system has been calculated by a computer technique developed in this laboratory by Dingus and Stewart.[26] The efficiency is calculated to be $(93\pm2)\%$. Since the decay of Pr^{140} to Ce^{140} goes partly by electron capture, it was necessary to correct the data accordingly. Less than 0.35% of the electron-capture transitions decay to excited states of Ce^{140}, so essentially the only recorded counts for these transitions were the fluorescence x rays.[27] The recorded spectrum was compared to that of a pure $\beta+$ emitter (O^{15}) so that the low-energy x-ray events could be accounted for. The comparison showed that about 11% of the x-ray events were recorded and this number, in conjunction with the branching ratio given in Ref. 27, (50%), was used to calculate the true number of Pr^{140} decays during the counting live time. A correction was made to account for decay of the sample during the extended count period resulting from counting for a fixed analyzer live time. Corrections were also made to account for the

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existence of longer lived components of activity resulting from (γ,$2n$) and other reactions. The number of (γ,n) events during the irradiation period was then calculated from the total count.

The collected ionization chamber charge was converted to energy incident upon the sample by the factor listed in NBS Monograph 48, and the result normalized by energy contained within a Schiff integrated-over-angle thin-target bremsstrahlung spectrum.

The total number of (γ,n) events per unit of incident energy per target nucleus was then calculated directly and all measured yield curves were normalized accordingly.

The combined errors of the factors contributing to the absolute cross section were about 5.5%. Since the reproducibility of the yield curves is better than $\frac{1}{2}$% the integrated cross sections should have values accurate to about ±6%. This figure should not be applied to the value of the cross section at any particular energy since such values depend strongly on experimental resolution. Integrated cross-section values are much less dependent upon resolution than are individual values.

III. TREATMENT OF DATA

Essentially two separate experiments were performed. In the first experiment data were accumulated from threshold energy through 65 MeV in 0.5-MeV increments. In the second, data were accumulated from threshold energy through 24 MeV in 0.125-MeV increments. Three complete yield curves of 120 points each were measured in each experiment. Standard deviations were calculated for each energy using the three data points available for that energy. In this manner average standard deviations were calculated for each experiment. Since each experiment contains a significant number of points at energies where nuclear counting statistics are less than 0.15%, it is possible to separate counting statistics from other random errors in the yield points. The average random errors with counting statistics removed for the two experiments were 0.41% for the 0.5-MeV and 0.25% for the 0.125-MeV experiment.

Cross sections were extracted from each individual yield curve and from the averaged curves for each experiment by the least-structure method.[18,19,22] In essence the method accepts as an approximation to the cross section the smoothest sequence of points which is consistent with very carefully evaluated statistical errors on the yield data points. Artificial yield curves are calculated from the smoothed cross section and compared to actual yield curves. Deviations are allowed which are consistent with data random errors.

In some instances the smoothing process causes the calculated yield to deviate from the measured yield excessively in some regions of the yield curve and very little in others. Since the deviations are averaged over the entire yield curve such a solution might appear mathematically acceptable, but it would result in excessive smoothing of the cross section in some regions and insufficient smoothing in others. When this is observed, a sequence of weighting factors is applied to the analysis procedure so that the smoothing process will be equally effective everywhere.[19]

In addition to resolution functions, the analysis method provides a highly correlated sequence of error bars, one for each cross-section point. Since this is so, error bars are provided only at intervals in the plotted curves demonstrate the error trend.

Since smoothing establishes a high degree of successive point correlation in the cross sections, cross-section curves should be drawn through the point sequence rather than just within the error bars. A new measurement might provide new point sequence near the limits of the error bars but all the points in a localized region would still be correlated.

The validity of structure observed in cross sections by this method is strikingly demonstrated in the case of $O^{16}(\gamma,n)O^{15}$ and $C^{12}(\gamma,n)C^{11}$ by comparison with many other experimental techniques.[24,28] The agreement is of course limited to the giant-resonance region since other techniques have not been extended to higher energies.

EXPERIMENTAL RESULTS

The threshold energy to 65-MeV cross section resulting from the yield curves taken in 500-keV increments is shown in Fig. 1. The cross section rises to a peak at 15.2 MeV, then drops sharply to zero at 25 MeV. The cross section is nearly zero between 25 and 36 MeV, at which point it begins to rise slowly and reaches a maximum of approximately 20 mb at 65 MeV. The horizontal error bar at the peak of the cross section indicates that the experimental resolution at that point was about 1.25 MeV. The very small oscillations in the cross section above 36 MeV did not appear to be correlated in the individual curves and there is no evidence for any resonance structure in this region.

Integrated Cross Section

The integrated cross section to 30 MeV is found to be (1790±100) MeV mb. This cross section is for the (γ,n) reaction only and it is somewhat higher than that published by Rice, Bolen, and Whitehead,[25] although it agrees well with their cross section not corrected for neutron multiplicity.

Above 36 MeV the rising cross section contributes another 440 MeV mb so that the cross section integrated to 65 MeV becomes (2230±135) MeV mb.

In the NBS Monograph 48 describing the response of the P2-type ionization chamber two curves are presented representing the extreme limits of the chamber response (charge per incident energy as a function of

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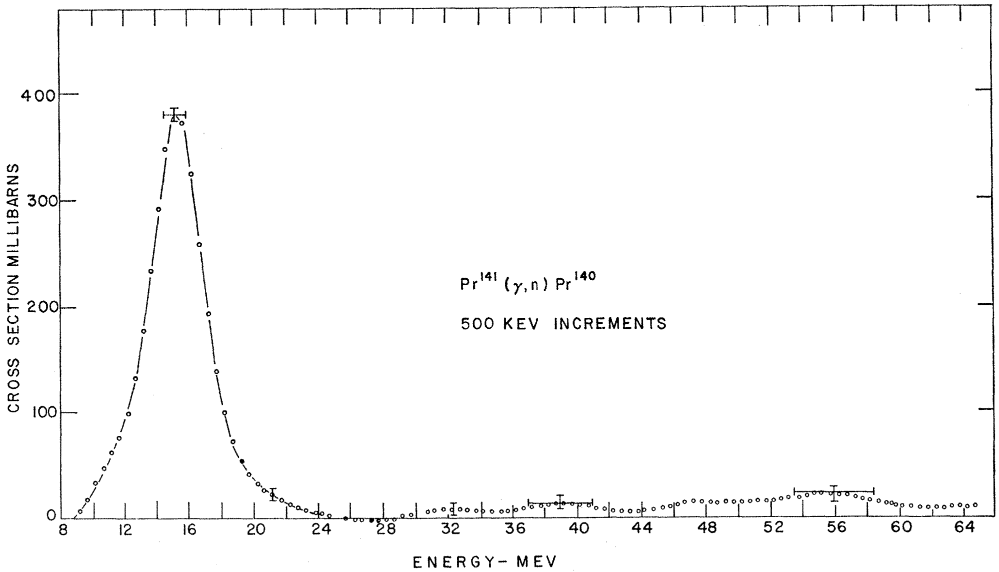

FIG. 1. Cross section for $Pr^{141}(\gamma,n)Pr^{140}$ from threshold energy to 65 MeV. The cross-section curve was calculated from yield data taken at 0.500-MeV increments. Horizontal error bars represent full width at half-maximum (FWHM) of experiment resolution and not uncertainty in energy.

energy). Since each point on the activation yield curve is normalized to dosimeter response at that energy, these limiting response values will appear as variations upward or downward on the cross sections. Cross sections to 65 MeV were calculated using each of the limiting response curves. The results of these modifications were negligible at energies below 25 MeV. Above 25 MeV one limit increased the integrated cross section to 32% of that below 25 MeV (573 MeV mb), while the other limit reduced the integrated cross section to 13% (232 MeV mb). In each case the cross section above 25 MeV looks very much the same as that shown in Fig. 1 with the entire curve raised or lowered slightly. The lowered curve remains very close to zero over the entire range with only a few regions of slight positive cross section.

While there is no reason to expect that the entire cross section to 65 MeV results from electric-dipole transitions, it is of interest to compare the integrated cross section with known $E1$ sum-rule calculations. The sum rule of Levinger and Bethe[29] with 0.5 inserted as the fraction of exchange force in the two-body interaction, yields 2882 MeV mb. This appears to be consistent with the measured result since one can expect small additions to the (γ,n) cross section from (γ,p) events and events resulting in the emission of more than one nucleon.

Giant-Resonance Region

The results of higher resolution cross-section measurement in the giant-resonance region are shown in Fig. 2. Eleven distinct resonances have been resolved in the region below 20 MeV. With the exception of the resonances at 15.45 and 15.85 MeV, each of the resonances shown was resolved in each of the individual cross-section determinations. These resonances appear to have widths of 100 to 200 keV and spacings about 800 keV. It appears very likely that many of these rather narrow resonances actually consist of further unresolved structure. The horizontal error flags indicate the half-width of the experimental resolution. Since the resolution has become approximately equal to the level spacing at 15.5 MeV, the two above-mentioned resonances were not resolved in two of the three individual curves, but were resolved in the remaining curve and in the average curve. Inspection of slightly undersmoothed data indicates splitting into two resonances in all curves. In addition there is evidence in slightly undersmoothed cross sections that the resonance at 14.60 MeV actually consists of two sharper but unresolved peaks.

Table I lists each of the resonances observed and the standard deviation of energy obtained by comparing the resonance position in each of the individual cross-section curves. In addition, the half-width of the resolution function is presented at each resonance energy. In general the experimental errors in the energies are less than the resolution-function half-width.

The peak value of the cross section is very close to 400 mb, somewhat higher than that obtained from the 0.5-MeV data. This simply reflects the higher resolution of these data and the peaks would be extended even higher if better resolution were obtainable. The integrated cross sections to 21 MeV of the two sets of data agree within 1.5%.

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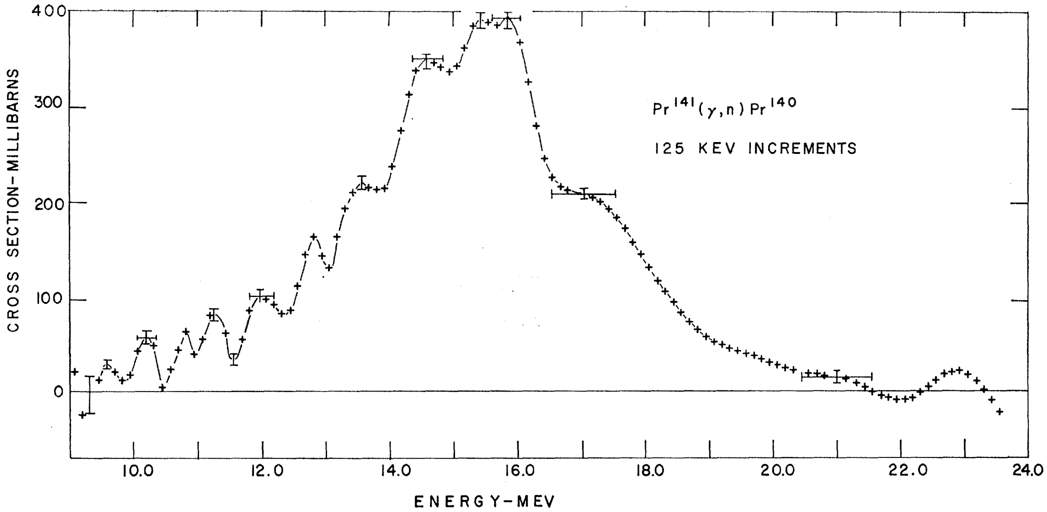

FIG. 2. Cross section for $Pr^{141}(\gamma,n)Pr^{140}$ from threshold energy to 24 MeV. The cross-section curve was calculated from yield data taken at 0.125-MeV increments. Horizontal error bars represent FWHM of experiment resolution and not uncertainty in energy.

CONCLUSIONS

In many recent reports of the (γ,n) cross section of heavy elements it has been observed that it was difficult to fit the result to a single Lorentz line.[25,30,31] In each case careful examination of the cross section reveals evidence for oscillator strength or structure above and below the main cross-section peak. This experiment demonstrates clearly that such structure exists and in addition, that the region of great oscillator strength is split into several resonances even in nuclei with little static deformation.

TABLE I. Resonance energies below 25 MeV and integrated cross sections.

Mean resonance energy (MeV)	Standard deviation (MeV)	Resolution half-width (MeV)
9.58	0.090	0.270
10.17	0.080	0.280
10.81	0.190	0.300
11.23	0.070	0.310
11.95	0.130	0.330
12.78	0.070	0.350
13.58	0.210	0.370
14.64	0.030	0.400
15.45 15.70	···	0.420
15.87	···	0.450
17.20	···	0.810

$$\int_{E_{th}}^{30} \sigma\, dE = 1794 \pm 100 \text{ MeV mb}; \quad \int_{E_{th}}^{65} \sigma\, dE = 2235 \pm 135 \text{ MeV mb}^{a}.$$

[a] The integral to 65 MeV is based on 0.500-MeV increment data. The integrated cross sections to 30 MeV for the 0.125- and the 0.500-MeV data differ by less than 1.5%.

It is evident from Fig. 1 that no structure or resonances exist in the (γ,n) cross section above 25 MeV. This is the region in which oscillator strength representing $E1$ overtones or electric-quadrupole transitions might be expected to appear according to the simple hydrodynamic model. In this respect the results differ drastically from those of the $O^{16}(\gamma,n)O^{15}$ experiment in which extensive structure appears to about 60 MeV.

The general appearance of the cross section at high energies is very similar to the recently presented result of Moffatt and Reitmann[32] for the photoneutron cross section of thallium 203 to 105 MeV. Their result included all neutron multiplicities and showed no high-energy structure, although their smooth cross section may have been the result of the method of analysis.

It is still possible that a high-resolution experiment in the high-energy region may reveal structure in the $(\gamma,2n)$ cross section which would represent $E1$ overtone absorption strength.

In Fig. 2, the resonance appearing near 17.25 MeV appears to be too low in energy to qualify in a simple way as an $E1$ overtone or as an "electric-quadrupole giant resonance," although the latter may not be completely unreasonable. Much attention has been given recently to the effect of quadrupole collective motion on the giant dipole resonance shape.[9,33]

The strong resonances in the region of 14.6 and 15.5 MeV, each of which is suspected of being doublet, may be candidates for the "upper" and "lower" resonances described in the report by Danos *et al.* regarding collective oscillations in odd-A nuclei. They suggest that the lower of the peaks should be split into two roughly equal components separated by about 100 keV. The

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results of this experiment indicate the persistence of rather sharp resonances through the giant-resonance region.

Recently, Izumo[34,35] has presented a theory of "partial equilibrium" for nuclear reactions in which a few (3–7) nucleons share excitation energy. The model is an attempt to explain the existence of "intermediate resonances" or clumps of oscillator strength ranging in width from 100 to 400 keV. It may be that the resonances observed in this experiment below 14 MeV are examples of such intermediate resonances.

One prediction of the Izumo theory is that the resonance structure of different nuclei in which the same number of nucleons share in the excitation should look the same (although displaced slightly in energy). The theory of Danos *et al.*, on the other hand, suggests that the photon absorption spectrum of odd-A nuclei should be somewhat different than even-A nuclei, even though the presence of the odd nucleon does not make a dramatic difference.

It seems clear that the relative merits of the various collective oscillation theories for heavy nuclei can only be tested by a succession of high-resolution experiments on a variety of such nuclei in the near future. The results of this experiment suggest that such experiments are possible and will be carried out.

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Number-Conserving Approximation for the Theory of the Pairing Interaction in Nuclei

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(Received 25 October 1965)

The pairing interaction is studied by means of the equations of motion. An approximation is derived which adheres strictly to particle-number conservation, but is otherwise quite close to the formalism of the Bardeen-Cooper-Schrieffer (BCS) theory. It yields ground-state energies and single-particle properties considerably improved over those of the latter theory, as evidenced by calculations for (i) the model of Pawlikowski and Rybarska, (ii) the model of the Ni isotopes solved exactly by Kerman, Lawson, and MacFarlane. Though it does not yet surpass in accuracy several alternative improvements of the BCS theory, it is capable of systematic further development, including the treatment of more realistic potentials.

I. INTRODUCTION

The present systematic theory of medium and heavy nuclei[1,2] is based upon ideas and methods borrowed from the theory of superconductivity[3] (the BCS theory). During the past few years there have been numerous efforts, detailed below, to improve on the basic approximation of this theory—the treatment of the properties of a given nucleus as the average of the properties of an ensemble of muclei. If we restrict our attention momentarily to the pairing interaction, as has been the case in almost all work of this sort, there now exist exactly soluble models, both of spherical[4] and of deformed[5] nuclei against which to measure the accuracy of the BCS theory and its proposed extensions.

One new approach has been related to Lipkin's[6] idea of allowing for the "curvature" of the separation energy as a function of particle number, i.e., of replacing the operator used in the BCS theory,

$$\mathcal{H}=H-\lambda A\,, \tag{1.1}$$

where H is the Hamiltonian, λ the separation energy or chemical potential, and A the number of particles, by a more general operator

$$\mathcal{H}=H-\lambda f(A)\,. \tag{1.2}$$

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