PHOTODISINTEGRATION OF ^{16}O AND ^{63}Cu BY PHOTONS OF VARIABLE ENERGY

By J. G. CAMPBELL*

[*Manuscript received August* 31, 1955]

Summary

Proton capture reactions in light nuclei provide γ-radiation with energy controllable by variation of proton energy. The radiation from proton capture by ^{7}Li has been used to investigate the (γ,n) reactions in ^{16}O and ^{63}Cu over an energy range of 300 keV around 17·7 MeV. No resonant behaviour was observed in ^{16}O. A variation in the ^{63}Cu photoneutron cross section is interpreted as the combined effect of many unresolved levels.

I. INTRODUCTION

The possibility that nuclei might absorb γ-radiation through excitation of discrete energy levels, by a process inverse to monochromatic γ-emission, has been recognized for many years. Kuhn (1929) reported an unsuccessful attempt to detect selective scattering of ThC″ radiation by ^{208}Pb, which was based on the expectation that the radiation from the 2·62 MeV excited state of ^{208}Pb (resulting from β-emission by ThC″) would excite that same state in other identical ^{208}Pb nuclei. It is only recently, however, that the effect has been successfully detected.

Kuhn's experiment failed because the emission and absorption of the radiation must involve loss of energy to nuclear recoil, so that the energy absorbed is less than the excitation energy of the original level by an amount sufficient to take it off resonance. In recent experiments along the same lines this energy has been restored by causing the emitting nuclei to be moving towards the scattering nuclei so that the photon energy is increased by the Doppler effect. This motion has been achieved by direct mechanical means (Moon 1951 ; Davey and Moon 1953), by heating (Malmfors 1952), and by recoil from a previous emission (Ilakovac 1954). The resonant scattering so obtained has enabled the lifetimes of the states concerned to be determined.

Recent work on the activation curves resulting from disintegration of light nuclei by betatron radiation has disclosed abrupt changes in slope, or " breaks ", which are interpreted as the effect of resonant absorption by discrete nuclear levels. The positions of several of the breaks for the reaction $^{16}O(\gamma, n)^{15}O$ found at Saskatchewan (Katz *et al.* 1954) have been confirmed by a group at Illinois (Penfold and Spicer, personal communication). Similar effects in the activation curve for $^{63}Cu(\gamma, n)^{62}Cu$ were reported by Phillips (1953), but neither of the groups at Saskatchewan and Illinois has been able to confirm this.

This paper reports the results of experiments carried out on these two reactions using the γ-radiation from the bombardment of lithium by protons.

* Physics Department, University of Melbourne.

II. Principle of Experiments

Consider a stationary nucleus of mass $(A-1)M$ being struck by, and capturing, a proton of mass M and kinetic energy E_p. The compound nucleus will recoil with velocity $\sqrt{(2E_p/M)}/A$. It is assumed to be travelling with this full recoil velocity when it is de-excited to its ground state by the emission of a photon which emerges at angle θ to the recoil direction. The energy of the photon in the laboratory is

$$E=\left(Q+E_p\frac{A-1}{A}\right)\left(1-\left[Q+E_p\frac{A-1}{A}\right]\Big/2MAc^2\right)\left(1+\frac{\cos\theta}{Ac}\sqrt{\frac{2E_p}{M}}\right), \quad \ldots (1)$$

where Q is the binding energy of the proton in the compound nucleus and c the velocity of light. $E_p(A-1)/A$ is the fraction of the proton kinetic energy which is available for excitation in the centre-of-mass system. The second factor is the correction due to the small amount of energy taken by the compound nucleus in recoiling from the emission of the photon, and is actually an expansion terminated at the first order, the approximation being valid for $[Q+E_p(A-1)/A]\ll 2MAc^2$. The third factor is the Doppler correction due to motion of the centre of mass.

The photon energy can therefore be controlled by varying either the proton energy E_p or the angle θ. In these experiments, θ was held constant and E_p varied. Assuming the ground state of the final nucleus to be sharp, the homogeneity in any direction is limited by the energy spread of the proton beam and the thickness of the target, and so can be of the order of 1 keV, although in the present work thicker targets were necessary to obtain sufficient intensity.

The ^{7}Li$(p, \gamma)^8$Be reaction has a Q-value of 17·242 MeV (Ajzenberg and Lauritsen 1955). The ground state of ^{8}Be has a width of a few electron-volts, which is small enough to be ignored, and so the photons from transitions to the ground state have energy given by (**1**). Transitions also occur to a broad ($\sim$2 MeV) state at excitation 3 MeV, but since the spread in the resulting photon energies is large compared with the range of variation available they could not produce fine structure effects. The cross section of the ^{7}Li$(p, \gamma)^8$Be reaction exhibits the well-known resonance at 441 keV proton energy, above which it drops to a low but nearly constant level.

In these experiments, the samples of oxygen and copper were exposed to the γ-radiation from the bombardment of thin lithium targets by the proton beam from a 700 keV electrostatic generator. The angle θ was held constant, and the proton energy E_p varied over a range of about 300 keV. The activation induced by photodisintegration was measured at each proton energy.

III. ^{16}O$(\gamma, n)^{15}$O Experiment

Because of the small cross section of this reaction and the low intensity of γ-radiation available, it was necessary to detect the ^{15}O produced with the highest possible efficiency. This was done by counting the 1·68 MeV positrons emitted by ^{15}O, the half-life of which is 118 sec (Ajzenberg and Lauritsen 1955). 4π geometry was obtained by making a scintillation counter in which the scintillator itself was rich in oxygen. The scintillator used was a saturated solution of *p*-terphenyl in *p*-dioxane, which contains 36 per cent. oxygen by weight, with a trace of diphenylhexatriene to improve the pulse height. It was contained in a small Pyrex vessel in the form of a truncated cone, of semi-angle 30°, with a volume of about 30 cm^3. The vessel was wrapped in aluminium foil and coupled optically to a photomultiplier (E.M.I. type 6260) with mineral oil.

The pulses from the photomultiplier were fed through a unity gain pre-amplifier and a 1008 amplifier to two 1009 scaling units switched together with connected inputs. The discriminator levels were set to correspond to ionization in the scintillator of 0·6 and 2·2 MeV respectively, so that the difference between their readings gave the counts in a channel bounded by these values.

The thin lithium targets were prepared by evaporation of lithium metal *in vacuo* on to copper disks, and as this was done in a separate vacuum system it was necessary to transfer the targets without exposure to air. This transfer was carried out under an atmosphere of dry nitrogen, so that an unknown amount of lithium would have combined to form Li_3N, but this is still a satisfactory target material.

The proton beam currents used were between 30 and 50 μA. The dioxane scintillator was placed with its axis along the line of the proton beam, and the apex of the truncated cone at the target spot. Thus the γ-radiation entering the dioxane had values of θ ranging from 0 to 30°.

After each bombardment, it was necessary to remove the scintillator from the vicinity of the target, to avoid counts due to stray activities. The counter unit, consisting of scintillator, photomultiplier, preamplifier, and light-tight box, was placed during irradiation in a kinematic mounting with assured reproducibility of position and orientation. When each bombardment was completed, the unit was lifted off the mounting and slid into a " burrow " in a pile of lead bricks. The scintillator was there shielded from stray radiation by at least 10 cm of lead in all directions except at the mouth of the burrow. In order to eliminate counts due to energetic cosmic ray particles, a tray of 17 Geiger counters was inserted in the lead pile just above the scintillator, with a sensitive area about 10 in. square. The dioxane pulses, after passing through a 3 μsec delay line, were put through a gate which was closed for 8 μsec after a pulse from any of the Geiger tubes. Thus the background in the channel was reduced to 4 counts per minute.

The photon flux was monitored by a sodium iodide scintillation counter. The cylindrical NaI(Tl) crystal, $1\frac{1}{2}$ in. long and $1\frac{1}{2}$ in. in diameter, in conjunction with a 6260 photomultiplier, gave pulses which were amplified and fed to a single-channel pulse height analyser. In order that the counter should respond only to the γ-radiation from transitions to the ground state of ^8Be, the channel was set at a level corresponding to an ionization in the crystal of 16 MeV (Campbell and Boyle 1954). The crystal was placed on a line from the target 20° from the forward direction, which was about the mean of the angular range covered by the dioxane.

The proton beam falling on the target was monitored by two circuits in series, a conventional pulse integrator and a " leaky " integrator of the type first discussed by Snowdon (1950). The purpose of the latter was to measure the apparent integrated current as registered by a system with a decaying memory, the decay time of this being equal to that of the activation being studied.

The procedure in each bombardment was to set the proton beam on the target for about 5 min, with the scintillator in position on its kinematic mounting. During the bombardment, the γ-monitor and the pulse integrator were switched on together for a period sufficient to determine the ratio (integrator count)/(γ-count) to sufficient statistical accuracy. At the end of the bombardment, the leaky integrator was read, and simultaneously the stopclock was started which timed the activation counting. After a 30-sec delay, during which the dioxane counter was transferred to the burrow, the counts were recorded during 4 min, followed by a 4-min delay and a further 4-min count. The difference between these two counts was taken as the measure of the activation and the final measure of the photodisintegration cross section at the energy of the bombardment was then

$$\frac{\text{(activation count) (integrator count)}}{\text{(leaky integrator reading) }(\gamma\text{-count})}.$$

No attempt was made to measure the cross section absolutely.

To determine the mean energy of the photons causing the disintegration, equation (**1**) was used. The mean of E_p was taken to be the incident proton energy less half the target thickness. This is not quite true, because of the variation of proton yield with energy, and if necessary a more accurate mean could be calculated in each case. The mean of $\cos\theta$, over the material of a cone of semi-angle 30°, is $0\cdot 933$.

Figure 1 shows one of the sets of results obtained. Also shown are points taken in a control experiment when the dioxane scintillator was replaced in the vessel by a solution of *p*-terphenyl in toluene, in which case the only activation detected would be that due to the oxygen in the glass, which would be expected to give between 15 and 20 per cent. of the counts. The results are quite consistent with this estimate.

The vertical lines through the points in Figure 1 denote the 67 per cent. statistical confidence limits. These are shortest in the region of the resonance in the lithium proton capture cross section, where the γ-flux is most intense.

In the case of the results shown in Figure 1, the lithium target used was about 80 keV thick. Several other sets of results were taken, with thinner lithium targets and hence poorer statistical accuracy. In all of these, the points were consistent with a constant photodisintegration cross section* in the region available.

Using the result of Wäffler and Younis (1949) that this cross section has the value $0\cdot 54\pm 0\cdot 14$ mbarn for the lithium resonance radiation, it is possible to place an upper limit on any (γ, n) resonance in this region. It can be concluded

* In a note on a preliminary experiment (Campbell 1954) it was claimed that a resonance had been detected in this range. This was erroneous, and resulted from stray target activations, mainly ^{13}N.

that a sharp resonance between 17·6 and 17·9 MeV would certainly have been observed if its integrated cross section were as great as 0·05 MeV-mbarn.

This result is to be compared with the results of the betatron work mentioned above. Katz *et al.* (1954) covered this range of photon energies and detected no resonance. Penfold and Spicer (personal communication), however, report a break corresponding to an energy level in ^{16}O at 17·71 MeV with integrated cross section 0·18 MeV-mbarn.

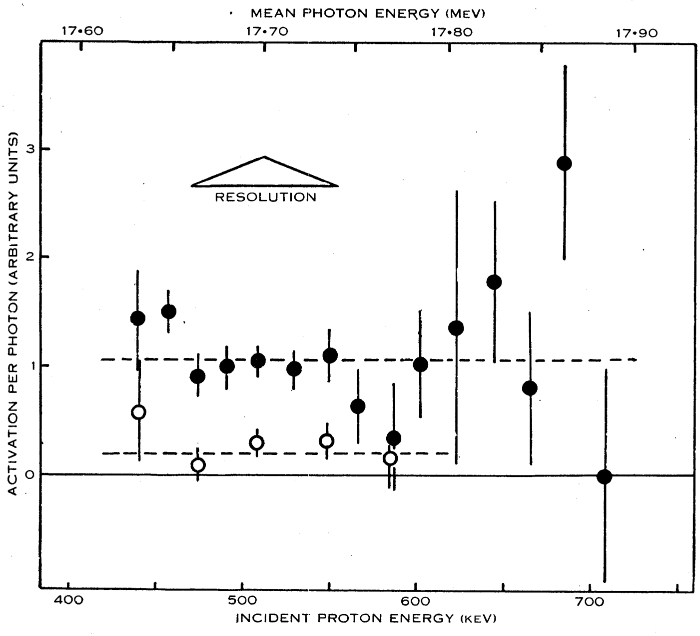

Fig. 1.—A set of results for $^{16}O(\gamma,n)^{15}O$. Closed circles : dioxane scintillator ; open circles : toluene scintillator.

IV. $^{63}Cu(\gamma, n)^{62}Cu$ Experiment

In this experiment, the copper metal sample, which was in the form of an annulus of the exterior of a cone, 2 in. in diameter with a central hole $\frac{3}{8}$ in. in diameter, was placed around the target so that it intercepted radiation emerging at 90°. The Doppler spread in the radiation entering the sample depended on the angle it subtended at the target. Two samples were used, one subtending $82° \leqslant \theta \leqslant 98°$ and the other $60° \leqslant \theta \leqslant 98°$.

After irradiation, the sample was removed to between a pair of sodium iodide scintillation counters in coincidence, which detected the positron annihilation radiation from ^{62}Cu. Two flat aluminium cones were sprung together to form a sandwich around the sample, ensuring that all positrons were absorbed. The counting system was in a pile of lead bricks to reduce background.

The procedure was similar to that in the oxygen experiment, the leaky integrator now being set to a half-life of $10 \cdot 0 \pm 0 \cdot 1$ min, the mean of recent determinations of the half-life of ^{62}Cu (U.S. National Bureau of Standards 1950). The sodium iodide counter measuring the flux of ground-state γ-radiation was placed below the target, at the mean angle subtended by the sample.

The activation was counted for 20 min, followed by a 20-min delay and a further 20-min count. The relative photodisintegration cross section was found as before.

The results with the thinner sample indicate a dip in the cross section at about 550 keV incident proton energy, but it is of the same magnitude as the statistical uncertainties. The results for the thicker sample are shown in Figure 2, and a similar dip appears here too.

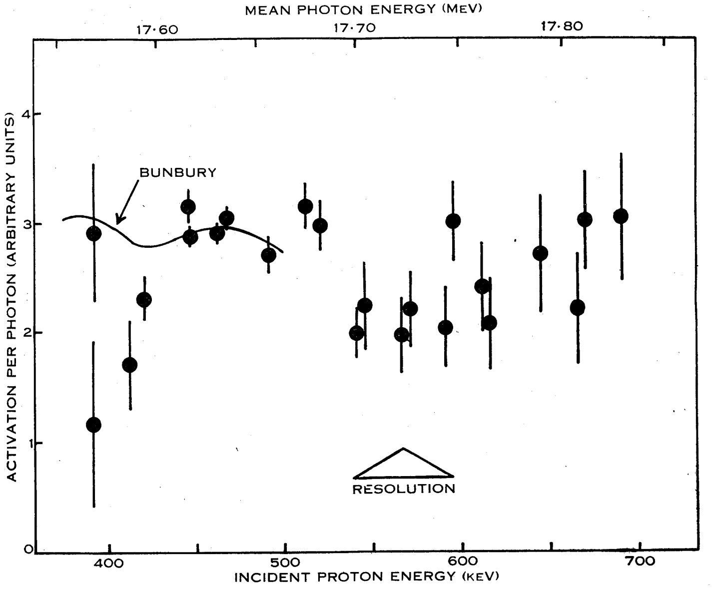

Fig. 2.—A set of results for $^{63}Cu(\gamma,n)^{62}Cu$. Also shown are results of Bunbury (1954).

While this experiment was being planned, Bunbury (1954) reported an experiment on this reaction using the same lithium γ-radiation. In that work the radiation from the 441 keV proton capture resonance was used, the energy being varied by alteration of angle. This has the advantage of high γ-intensity, but the range of energy variation is more limited. The curve obtained by Bunbury is shown on Figure 2, with ordinate adjusted to fit. The statistical uncertainties in the present experiment would have obscured the fine structure observed by Bunbury, and the dip at 550 keV is beyond the range of photon energy obtainable by Doppler shift of the resonance radiation.

It is necessary to interpret the copper results in terms of known theory. There are little data available about medium-weight nuclei at high excitation, but Blatt and Weisskopf (1952, pp. 371–2) have given a rough, semi-empirical formula based on a statistical nuclear model. With the constants given for mass number 63, this formula gives the mean spacing of all levels to be about

24 eV at excitation 17·6 MeV. An experiment such as the present one could not resolve these levels individually, but the average integrated cross section within the finite interval of resolution would be expected to show some variation, just as the needle of a count-rate meter fluctuates even when its integrating time is long compared with the mean pulse spacing.

Since each point in Figure 2 has a total resolution width of about 50 keV, then each would include the effects of about 2000 levels. If these levels are spaced randomly along the energy scale, then the number of levels affecting each point is statistically distributed about 2000 according to the Poisson distribution, with standard deviation $\sqrt{2000}$. Assuming in the first instance that all levels have the same integrated cross section, then the apparent cross section of independent points will fluctuate with proportional standard deviation 2 per cent. To take account of the differences in level cross sections, it might be assumed that one-quarter of them are large and equal (these would be the ones giving electric dipole transitions) and the remainder small enough to be ignored. The result is now an expected variation in apparent cross section with standard deviation 4 per cent. This is to be compared with the experimental standard deviation of about 15 per cent., which would be the fluctuation of a smooth line of best fit through the points of Figure 2. These figures, while not being in close agreement, are within the same order of magnitude, and taking account of the rudimentary nature of the level density formula it can be concluded that the results are not inconsistent with this model.

V. Acknowledgments

The author has appreciated the continued encouragement of Professor L. H. Martin, and acknowledges the assistance of Mr. B. M. Spicer in communication of results prior to publication and in discussion.

VI. References

Ajzenberg, F., and Lauritsen, T. (1955).—*Rev. Mod. Phys.* **27** : 77.

Blatt, J. M., and Weisskopf, V. F. (1952).—" Theoretical Nuclear Physics." (John Wiley & Sons : New York.)

Bunbury, D. St. P. (1954).—*Proc. Phys. Soc. Lond.* A **67** : 1106.

Campbell, J. G. (1954).—*Phys. Rev.* **95** : 1357.

Campbell, J. G., and Boyle, A. J. F. (1954).—*Aust. J. Phys.* **7** : 284.

Davey, W. G., and Moon, P. B. (1953).—*Proc. Phys. Soc. Lond.* A **66** : 956.

Ilakovac, K. (1954).—*Proc. Phys. Soc. Lond.* A **67** : 601.

Katz, L., Haslam, R. N. H., Horsley, R. J., Cameron, A. G. W., and Montalbetti, R. (1954).—*Phys. Rev.* **95** : 464.

Kuhn, W. (1929).—*Phil. Mag.* **8** : 812.

Malmfors, K. G. (1952).—*Ark. Fys.* **6** : 49.

Moon, P. B. (1951).—*Proc. Phys. Soc. Lond.* A **64** : 76.

Phillips, K. (1953).—*Phil. Mag.* **44** : 533.

Snowdon, S. C. (1950).—*Phys. Rev.* **78** : 299.

U.S. National Bureau of Standards (1950).—Nuclear data. Circ. U.S. Nat. Bur. Stand. No. 499.

Wäffler, H., and Younis, S. (1949).—*Helv. Phys. Acta* **22** : 614.