

the existence of a superstructure having a unit cell of  $4a \times 4a \times c$ . Our analysis, which is near completion, seems at present to give the atomic arrangement shown schematically in Fig. 11, in which an antiparallel set of dipolar orientations due to opposite shifts of heavy ions (probably Pb ions) may be noted. Details of the x-ray analysis will be published in the near future.

In conclusion, we wish to express our sincere thanks to Dr. S. Roberts for his many helpful comments on our work.

*Note added in proof.*—Recently, C. Kittel [Phys. Rev. **82**, 729 (1951)] discussed the possibility of realization of antiferroelectricity, showed that antiferroelectric crystals may be expected to

occur in nature, and gave some criteria for identifying them. Very recently, S. Roberts [Phys. Rev. **83**, 1078 (1951)] has observed a very small but definite piezoelectric effect in polarized PbZrO<sub>3</sub> ceramic, in contrast to the negative result of our pyroelectric test. But the observed effect is very small, being approximately one-thousandth that of BaTiO<sub>3</sub>, and this suggests that the spontaneous polarization of this crystal may also be so small that its detection by a pyroelectric test is beyond the limit of our apparatus. Our experiments have shown that the spontaneous polarization of PbZrO<sub>3</sub> may be, if it exists, far less than one-hundredth that of BaTiO<sub>3</sub>. So it must be concluded, in reference to the recent result of x-ray analysis of PbZrO<sub>3</sub> single crystal by Sawaguchi, Maniwa, and Hoshino [Phys. Rev. **83**, 1078 (1951)], that PbZrO<sub>3</sub> is strongly antiferroelectric in the direction perpendicular to the *c*-axis and very weakly ferroelectric in the *c*-direction.

## Bombardment of Copper with Tritons

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(Received July 13, 1951)

Tritons have been accelerated to 6.5 Mev in a cyclotron and made to bombard stacks of copper foils. Among the triton induced reactions, the following have been identified: Cu<sup>63</sup>(*t,d*)Cu<sup>64</sup> (12.9 hours), Cu<sup>63</sup>(*t,n*)-Zn<sup>65</sup> (250 days), Cu<sup>65</sup>(*t,He*<sup>3</sup>)Ni<sup>66</sup> (2.6 hours), and Cu<sup>65</sup>(*t,p*)Cu<sup>67</sup> (2.44 days) with relative cross sections for the various reactions as 11:64:6:10, respectively. The Oppenheimer-Phillips process appears to be operative in case of tritons of this order of energy.

### I. INTRODUCTION

TRITONS have already been used as bombarding particles for effecting nuclear reactions. In the earlier experiments,<sup>1</sup> the tritons had been produced by some suitable primary reaction, like Be+*d*, and the tritons thus obtained were then made to bombard various target nuclei. The maximum energy of the tritons was about 10.5 Mev. Later workers<sup>2-5</sup> used low-energy tritons and obtained reactions on tritium and lithium.

In the present investigations, tritium gas obtained in small amounts through the courtesy of the Isotopes Division of the Oak Ridge National Laboratory have been accelerated to 6.5 Mev in the cyclotron.<sup>6</sup> Various triton reactions with copper have been made. Relative cross sections and certain other reaction characteristics have been studied.

### II. EXPERIMENTAL

A few cubic centimeters of tritium gas were sealed in glass capsules which were contained in a special housing

connected to the cyclotron vacuum chamber through suitable capillary tubes and stop cocks. Arrangements were made so that from the outside the seal of a capsule could be crushed open and the gas fed between the dees in controlled amounts. The ion source was a single tungsten filament at the center of the vacuum chamber with the accelerating plates placed above and below the dee surfaces. The frequency of the oscillator was reduced from its normal 10.4 to 6.7 megacycles by the use of a bank of vacuum condensers placed in parallel both with the dee and the grid lines. The maximum current rating of the magnet could then be utilized. The natural leakage of the vacuum system was reduced to a practical minimum of 2 to 3 cc per hour. The gas from the exhaust of the backing pump was reinjected in controlled amounts through a closed recirculation system. The gaseous impurities introduced into the vacuum system through natural leakage consisted of nitrogen and oxygen. A stage of purification was incorporated in the circulation system so that the tritium could be freed from these gases, if and when necessary, with the use of a liquid hydrogen trap.

A stack of copper foils each 0.001 inch thick was mounted in a special target holder. The stack was thus shielded on all sides except for a small slot through which the triton beam bombarded the foils. The bombardment was continued, through the recirculation system, for two hours. The activities produced in

<sup>1</sup> D. N. Kundu and M. L. Pool, Phys. Rev. **72**, 101 (1947); **73**, 22 (1948).

<sup>2</sup> Sanders, Allen, Almquist, Dewan, and Pepper, Phys. Rev. **79**, 238 (1950).

<sup>3</sup> Los Alamos Group, Phys. Rev. **79**, 238 (1950).

<sup>4</sup> Allen, Almquist, Dewan, and Pepper, Phys. Rev. **81**, 315 (1951).

<sup>5</sup> E. Almquist, Can. J. Research **28A**, 433 (1950).

<sup>6</sup> Pool, Kundu, Weiler, and Donaven, **82**, 305 (1951).

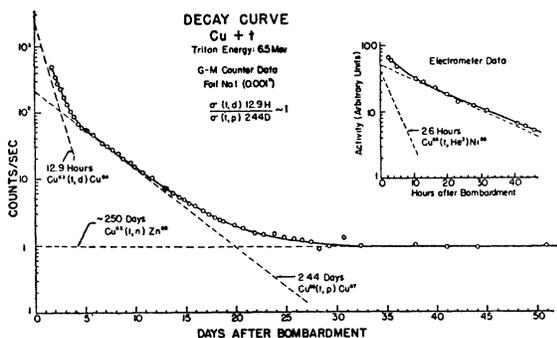


FIG. 1. Decay curve of the first copper foil after bombardment with 6.5-Mev tritons showing the half-lives 2.6 hours, 12.9 hours, 2.44 days, and 250 days.

individual foils were followed in the customary way with G-M counters and electrometers under the same geometry for each apparatus. Frequent absorption measurements were made at different stages of the decay to confirm the identification of the activities. From the extrapolated activities at the time of the termination of the bombardment, the saturation intensities corresponding to an infinite duration of bombardment for the various periods were calculated. Each saturation intensity was then converted into the number of disintegrations per second,  $I_\infty$ , by taking into account the known disintegration scheme of the activity in question and also the counting characteristics of the instruments. For two activities produced from the same target isotope in the same bombardment, the relative cross section was estimated as  $\sigma(1)/\sigma(2) = I_\infty(1)/I_\infty(2)$ , where the numbers stand for the activities compared.

### III. RESULTS

The decay curve of the first copper foil is given in Fig. 1. The insert shows the initial part of the decay made by use of an ionization chamber and electrometer. Each curve has been analyzed into its various component periods as indicated and these latter are separately examined in the following paragraphs. A similar study of the second foil is shown in Fig. 2. Here the activities are much lower in magnitude. The third and the rest of the deeper foils were also measured but no activity attributable to the triton bombardment was found. A few typical aluminum absorption measurements on the first copper foil are reproduced in Fig. 3, and are discussed below in conjunction with the corresponding periods.

#### 2.44-Day $\text{Cu}^{67}$

This activity is found to be produced with considerable clearness in the first foil with an intensity at the end of the bombardment of 218 counts per second as seen in Fig. 1. The same activity was also produced in the second foil (Fig. 2) but the corresponding initial intensity was only 0.87 count per second. The absorp-

tion curve made 10 days after bombardment, shown in Fig. 3, was at a time when the activity was predominately of the 2.44-day half-life. This half-life is thus found to be associated with negatrons of maximum energy 0.54 Mev. The activity results from the reaction  $\text{Cu}^{65}(t,p)\text{Cu}^{67}$  in agreement with the assignment made<sup>7</sup> on the basis of bombardments on enriched isotopes of Ni and on Cu and Zn.

If the relative stopping power of copper be taken as 2.56 mg/cm<sup>2</sup> as equivalent to 1 cm of air at 760 mm pressure and 15°C from the data of Mano,<sup>8</sup> the 6.5-Mev tritons will have a range of 2.16 mils in copper and will, therefore, penetrate into the third foil. The maximum energy of the tritons entering the first three foils are 6.5, 4.5, and 1.5 Mev, respectively. The  $(t,p)$  reactions thus proceeds with measurable yield of  $\text{Cu}^{67}$  with tritons of energy less than 4.5 Mev.

#### 12.9-Hour $\text{Cu}^{64}$

This activity is produced only in the first and the second foils with initial intensities 2400 and 23 counts per second respectively as found from Figs. 1 and 2.  $\text{Cu}^{64}$  is known<sup>9</sup> to decay by the emission of 0.57-Mev negatrons (43 percent), 0.66-Mev positrons (21 percent) and  $K$ -capture (36 percent). The beta-end point measured at 0.61 Mev, and shown in Fig. 3, is approximately in agreement as a mean value. The reaction is  $\text{Cu}^{63}(t,d)\text{Cu}^{64}$  or  $\text{Cu}^{63}(t,np)\text{Cu}^{64}$ .

#### 2.6-Hour $\text{Ni}^{65}$

This half-life is observed only in the first foil. Part of the decay curve followed with the electrometer is shown in the insert in Fig. 1 from which it is seen that the ionization produced by this activity is comparable with that by the 12.9-hour  $\text{Cu}^{64}$ . The reaction is  $\text{Cu}^{65}(t,\text{He}^3)\text{Ni}^{65}$ . Here, as in the previous  $(t,d)$  case, the out-going

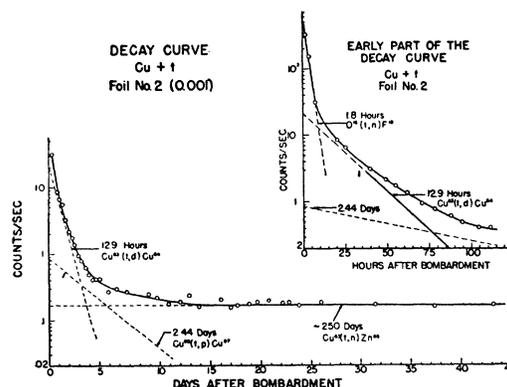


FIG. 2. Decay curve of the second copper foil after bombardment with 6.5-Mev tritons. The 1.8-hour  $\text{F}^{18}$  activity from traces of oxygen in the foil is seen in the initial part of the curve.

<sup>7</sup> D. N. Kundu and M. L. Pool, Phys. Rev. 78, 488 (1950).

<sup>8</sup> G. Mano, Ann. phys. 1, 407 (1934); J. phys. et radium 5, 628 (1934).

<sup>9</sup> "Nuclear data," Circular of the National Bureau of Standards 499, p. 59, Sept. 1, 1950.

particle is probably a single  $\text{He}^3$  nucleus rather than some other suitable number of separate particles. The ratio of the cross sections  $\sigma^{65}(t, \text{He}^3)/\sigma^{65}(t, p) = 0.6$ .

### 250-Day $\text{Zn}^{65}$

An activity with a long half-life of approximately this duration was produced in the first and second foil only. The initial activity in the first foil was 1 count per second (Fig. 1) whereas in the second foil (Fig. 2), it is only 0.17 count per second. The reaction is  $\text{Cu}^{63}(t, n)\text{Zn}^{65}$ . The apparent low activity is understandable in the light of the successive capture characteristics<sup>9</sup> of  $\text{Zn}^{65}$ . This radioactive nucleus decays largely by  $K$ -capture (96.7 percent). However, the relative cross section on calculation is found to be  $\sigma^{63}(t, d)/\sigma^{63}(t, n) = 0.17$ .

### IV. DISCUSSION

The production of the 2.44-day activity definitely established that the bombarding agent had been tritons; for, this activity is not produced from copper by protons, deuterons, or alpha-particles. Also, because this activity was not produced beyond the triton range, the successive capture of two neutrons, available from spurious sources, is ruled out as a mechanism for the production of  $\text{Cu}^{67}$ .

When the isotopic abundances of  $\text{Cu}^{63}$  (69.09 percent) and  $\text{Cu}^{65}$  (39.91 percent) are taken into account along with the decay scheme of the radioactive product nuclei, the relative cross sections of the different reactions for 6.5-Mev tritons become  $\sigma^{63}(t, n) : \sigma^{63}(t, d) : \sigma^{65}(t, p) : \sigma^{65}(t, \text{He}^3) = 64 : 11 : 10 : 6$ . Therefore, it is evident that with tritons as bombarding particles, it is as easy to introduce a single neutron into a  $\text{Cu}^{63}$  nucleus and form  $\text{Cu}^{64}$  as it is to introduce two neutrons or a dineutron into  $\text{Cu}^{65}$  and form  $\text{Cu}^{67}$ .

The explanation as to how the 2.6-hour  $\text{Ni}^{65}$  activity could be formed appears to be interesting. The reaction involved is necessarily of the  $(t, \text{He}^3)$  type. The question arises, even though the tritons of 6.5 Mev could still form a compound nucleus, the out-coming  $\text{He}^3$  will have to overcome a very steep potential barrier by virtue of its double charge. One way of explaining the phenomenon would be to suppose that the  $\text{He}^3$  overcame the potential barrier on "borrowed energy" obtained from the building of the three added nucleons to the target  $\text{Cu}^{65}$  nucleus and returned the amount necessary to maintain energy conservation as it left the barrier. An alternative view may be to postulate that the triton never entered the target nucleus at all but thus was only a charge exchange between a neutron within the triton and a proton within the target nucleus when the triton was in the close vicinity of the nucleus. The phe-

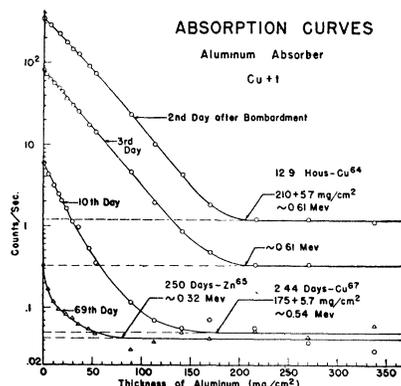


Fig. 3. Absorption curves on the first copper foil at different stages during the decay.

nomenon of charge exchange, however, is known so far only in the domain of very high energy collisions between nucleons.

With regard to the  $(t, p)$  reaction, it may be asked whether a part at least of the observed yield was through a stripping process analogous to the Oppenheimer-Phillips<sup>10</sup> mechanism in deuteron reactions. Compared to the deuteron, the triton is a more tightly bound nucleus. However, the extent of the stripping in Oppenheimer-Phillips mechanism depends on the ratio of the kinetic energy of the bombarding particle to its binding energy. From the curves calculated by Bethe<sup>11</sup> in the case of the deuteron, one finds that even when the ratio is  $\frac{1}{2}$  the penetration probability by the Oppenheimer-Phillips process for copper is 10 times larger than that by the compound nucleus formation process although this ratio is hard to evaluate quantitatively due to the difficulties involved in the wave-mechanical treatment with the triton, it is still expected that for values of  $\frac{1}{2}$  or even less, the Oppenheimer-Phillips process would contribute substantially to the total yield in the  $(t, p)$  and  $(t, d)$  reactions.

### V. ACKNOWLEDGMENTS

It is a pleasure to acknowledge the help received from the Development Fund of the Ohio State University. Our indebtedness to Mr. F. G. Boyle and Col. E. C. Mallery for the valuable help in constructional details, and to Mr. Paul E. Wiler at the operational stage is gratefully acknowledged. Thanks are also due the Ghosh Board of the Calcutta University for providing travel facilities for one of the authors.

<sup>10</sup> J. R. Oppenheimer and M. Phillips, Phys. Rev. 48, 500 (1935).

<sup>11</sup> H. A. Bethe, Phys. Rev. 53, 39 (1938).