

The Disintegration of Cu⁶¹ *

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By the use of very thin and uniform sources that were prepared by thermal evaporation, the positron spectrum of Cu⁶¹ continues to show an excess of particles in the low energy region. The presence of nuclear gamma-rays and internal conversion electrons indicates that the disintegration of Cu⁶¹ is complex. Magnetic spectrometer studies of the radiations from Cu⁶¹ have led to a tentative disintegration scheme.

I. INTRODUCTION

ACCORDING to the *ft* values,¹ the beta-disintegrations of both Cu⁶¹ and Cu⁶⁴ are of the allowed type. The Fermi theory of beta-decay predicts that such an allowed transition should produce a straight-line Fermi-Kurie (F-K) plot. However, experimental results have been obtained^{2,3} which seemed to indicate that the F-K plots for these two isotopes are non-linear in the low energy region. This non-linearity appeared as an excess of low energy particles, and in the case of the positrons from Cu⁶¹ this excess was present for all energies below 500 kev.

For the positron and negatron spectra of Cu⁶⁴ it has been shown⁴ that the deviations from the Fermi theory could be caused by an instrumental distortion. When thin sources were used in a study⁵ of S³⁵ and when gaseous sources were used in a study of tritium,⁶ the resulting spectra were found to be in good agreement with the Fermi theory to quite low energies. Further investigations of instrumental distortions⁷ have led to the conclusion that, with a properly designed chamber, the major portion of such distortions could be attributed to source thickness.

To test the dependence of the distortions on variation in source thickness, the positron distribution of Cu⁶¹ was selected. The fact that deviations in this spectrum occur at higher energies than in most other known allowed positron and negatron spectra seemed to favor a large dependence on source thickness.

In order that very thin and uniform sources could be prepared, an evaporator was constructed. The sources were prepared by thermal evaporation of copper from a hot tungsten filament onto a source backing.

Studies of the positron spectrum of Cu⁶¹ using sources prepared in this way showed the same type of deviation⁸ that had previously been observed.³ It follows

that this deviation is most probably real rather than instrumental and is caused by either a complex disintegration or by some departure from the Fermi theory.

Previous coincidence studies⁹ failed to disclose any nuclear gamma-rays of intensity comparable to the intensity of the annihilation quanta. This, however, does not eliminate the possibility of a complex disintegration.

A later investigation¹⁰ of both the positrons and negatrons of Cu⁶⁴ using sources prepared by evaporation gave spectra consistent with the Fermi theory as low as 50 kev; therefore it appears likely that the disintegration of Cu⁶¹ is complex. This conclusion is supported by the detection of gamma-rays in the photoelectron spectrum and internal conversion electron lines in the negatron radiations from Cu⁶¹.

II. EXPERIMENTAL DETAILS

A. The Small 180° Magnetic Spectrometer (5.7 Cm Radius of Curvature)

The investigations of the continuous beta-spectra were made using a small 180° spectrometer of 5.7 cm radius.⁷ Baffles are used at 30°, 90°, and 150°. The width of an internal conversion line at one-half of the maximum intensity is 1.2 percent; this was checked against the internal conversion lines of I¹³¹, using sources of 42 micrograms per square centimeter average thickness.

Measurements of the magnetic field are made with a flip coil and a ballistic galvanometer. The galvanometer is calibrated against the internal conversion lines of Th(B+C) and I¹³¹. To control the magnetic field current, a constant voltage d.c. generator and storage battery combination is employed. This regulator restricts variations in current to less than ±0.5 percent. An end-window Geiger-Mueller counter is used having a zapon window for which electron transmission begins at 3 kev.

B. The Large 180° Magnetic Spectrometer (13.8 Cm Radius of Curvature)

Because of the high intensity sources which are needed for measurements of photoelectrons, a large 180°

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¹ E. J. Konopinski, *Rev. Mod. Phys.* **15**, 209 (1943).

² C. S. Cook and L. M. Langer, *Phys. Rev.* **73**, 601 (1948); J. Backus, *Phys. Rev.* **68**, 59 (1945); C. S. Wu and R. D. Albert, *Phys. Rev.* **75**, 1107 (1949).

³ C. S. Cook and L. M. Langer, *Phys. Rev.* **74**, 227 (1948).

⁴ G. E. Owen and H. Primakoff, *Phys. Rev.* **74**, 1406 (1948).

⁵ R. D. Albert and C. S. Wu, *Phys. Rev.* **74**, 847 (1948); Price, Motz, and Langer, *Phys. Rev.* **77**, 744 (1950).

⁶ Curran, Angus, and Cockroft, *Phil. Mag.* **40**, 53 (1949).

⁷ G. E. Owen and C. S. Cook, *Rev. Sci. Inst.* **20**, 768 (1949).

⁸ G. E. Owen and C. S. Cook, *Phys. Rev.* **76**, 1536 (1949).

⁹ W. Gentner and E. Segrè, *Phys. Rev.* **55**, 814 (1939); Bradt, Gugelot, Huber, Medicus, Preiswerk, and Scherrer, *Helv. Phys. Acta*, **18**, 252 (1945).

¹⁰ Langer, Moffat, and Price, *Phys. Rev.* **76**, 1725 (1949); G. E. Owen and C. S. Cook, *Phys. Rev.* **76**, 1726 (1949).

magnetic spectrometer,¹¹ having more lead shielding between source and counter than the smaller spectrometer, was employed for investigating the gamma-rays from Cu^{61} .

This instrument has a resolution of 0.6 percent for internal conversion lines, and 1.5 percent for photoelectron lines, when a 50 mg/cm^2 uranium radiator is used. The field current, which is regulated electronically,¹² has a variation of less than 0.05 percent.

The Geiger-Mueller counter is of the end-window type employing a zapon window whose cut-off for

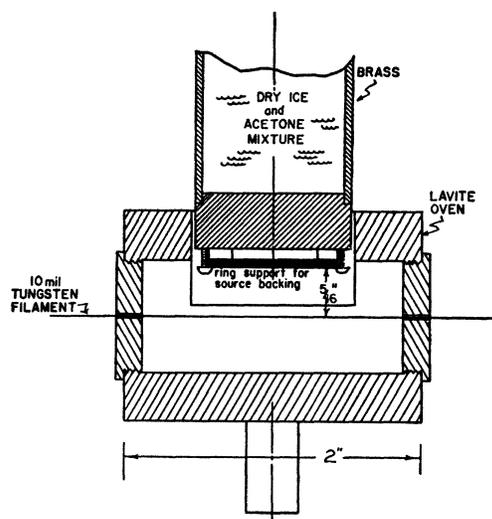


FIG. 1. Detail of the evaporator oven.

electrons is about 7 kev. To maintain a constant pressure in this counter, which has a large aperture, a pressure regulator is inserted in the filling system.¹³

C. The Photographic Spectrometer

The photographic spectrometer is of the usual 180° type and has a maximum radius of curvature of 10 cm. In order that the photographic plates and sources can be loaded and placed in position in a short time, the sources and photographic detection plates are mounted on a removable plug. This plate holder is provided with a light-tight cover which can be removed after the plug assembly is under vacuum.

The magnetic field is provided by a permanent magnet. This magnet employs two Alnico sections in order to maintain a large and stable field. The core is magnetized by means of a series of low resistance coils placed in position about the Alnico sections.

D. The Evaporator

In order to prepare sources which would be thin and uniform, an evaporator was constructed. The vacuum

¹¹ F. N. D. Kurie and M. Ter-Pogossian, *Phys. Rev.* **74**, 677 (1948); Ter-Pogossian, Cook, Goddard, and Robinson, *Phys. Rev.* **76**, 909 (1949).

¹² Elmore and Sands, *Electronics, National Nuclear Energy Series* (McGraw-Hill Book Company, Inc., New York, 1949), Vol. V-1, pp. 390-393.

¹³ Ter-Pogossian, Robinson, and Townsend, *Rev. Sci. Inst.* **20**, 289 (1949).

chamber consists of a large bell jar open at the top. A brass plate placed over this opening carries a plunger that can be removed from the chamber through an "O" ring seal.¹⁴ The end of this plunger can be placed in a Lavite oven located on the bell jar base, and provides a means of supporting the source holder during the evaporation process. A detail of the oven is shown in Fig. 1.

The active copper is first electroplated onto a 0.01-inch diameter tungsten wire, and the wire is then placed in position in the Lavite oven. A source backing of the 0.00025-inch aluminum foil is mounted on an aluminum supporting ring and is covered by an aluminum mask. An opening in this mask allowed an exposed area of $1.0 \text{ cm} \times 0.1 \text{ cm}$ at the center of the backing. The foil and mask were then held $5/16$ inch from the filament. A current of 5 amperes flowing for 10 seconds was observed to evaporate practically all of the copper which had been plated.

It was found advantageous to cool the backing with a dry-ice and acetone mixture which was located in the brass plunger as shown in Fig. 1. Later this procedure was modified by inserting a hollow brass sleeve, which carried the cooling mixture, into the plunger; this permitted the removal of the cold reservoir before the plunger was exposed to air and prevented the formation of ice on the source backing.

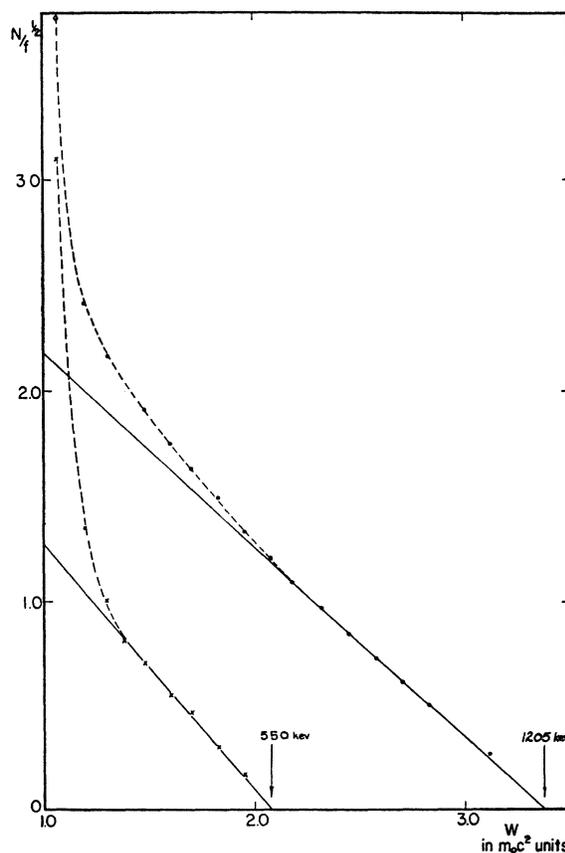
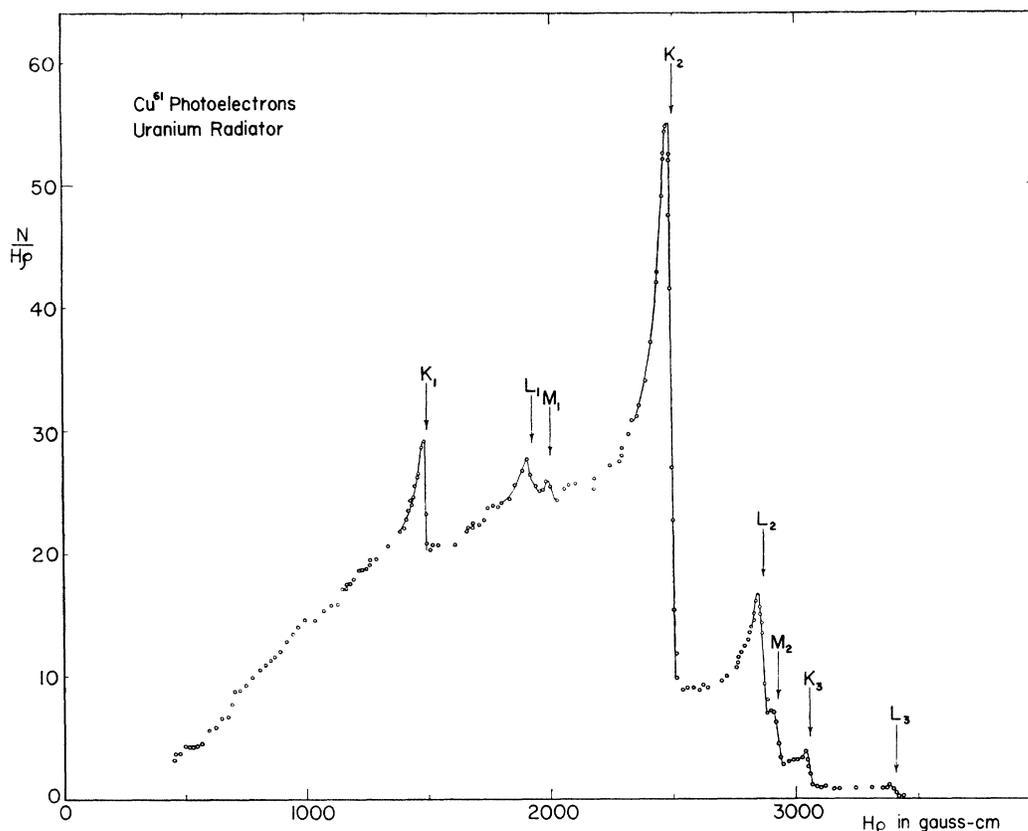


FIG. 2. F-K plot of the positrons of Cu^{61} showing the resulting F-K plot of one low energy group.

¹⁴ F. N. D. Kurie, *Rev. Sci. Inst.* **19**, 485 (1948).

FIG. 3. Photoelectron spectrum of Cu^{61} .

E. Production and Separation of Sources

The Cu^{61} isotope was prepared carrier free by (d, n) and ($d, 2n$) reactions on nickel. The copper was separated from the nickel by an electroplating process described by Steigman.¹⁵

A calculation of the equilibrium concentrations from the Nernst equation indicated that no nickel would be carried with the copper in a given electroplating process.

Since there were other short-lived copper activities formed in the bombardment, a time of approximately

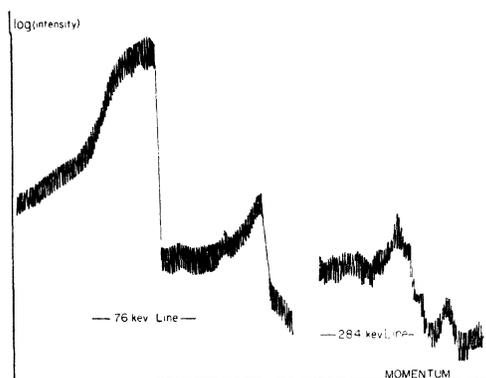
2 hours was allowed for these to die out. Approximately one half-life after the initial determination of the spectrum, the positron radiations from the source were re-studied, and after correction for loss of intensity because of the decay of the source, the results from both measurements were found to agree within one percent of each other.

A study¹⁶ of the extent to which Cu^{64} will be present as an impurity in this source indicates its positron activity as being considerably less than one percent of the total positron activity of the sample.

The sources prepared by evaporation could neither be observed visually nor weighed, and were detectable only by their activity on the exposed region of the backing. Rough orders of magnitude of the thickness could be calculated from the activity present. Such calculations give a thickness of 10^{-4} microgram per sq. cm. This figure represents a lower limit to the actual thickness, because of the probable presence of small amounts of stable copper originating as an impurity in the original chemically pure nickel target. The actual thickness is probably not greater than 10^{-2} microgram per sq. cm.

III. RESULTS

The F-K plot of the positrons from Cu^{61} , using a source prepared by evaporation, is shown in Fig. 2. The excess of particles below 500 kev remains. Because

FIG. 4. Microphotometer tracing of the internal conversion lines of Cu^{61} as recorded by the photographic spectrometer.

¹⁵ J. Steigman, Phys. Rev. **53**, 771 (1938).

¹⁶ C. S. Cook and C. H. Chang, Phys. Rev. **78**, 171 (1950).

it was found that sources of Cu^{64} prepared in the same manner gave agreement with the Fermi theory to energies as low as 50 keV, the evidence suggests that Cu^{61} is complex. If a second lower energy spectrum is derived by subtracting the Fermi allowed shape of the high energy spectrum from the low energy excess, an F-K plot is obtained which has a maximum energy of 550 ± 25 keV, with a corresponding momentum distribution having an intensity of 4 percent of the total positron intensity. This second F-K plot is also shown in Fig. 2.

From consideration of the positron spectrum (and F-K plot) alone, the low energy excess in the secondary spectrum may or may not be real. It may represent a tertiary spectrum but most likely a considerable part of it represents a distortion caused by scattering from the aluminum backing.

The photoelectron spectrum resulting from the gamma-rays and annihilation radiation of Cu^{61} incident on a 50 mg/cm² uranium radiator is shown in Fig. 3. The *K*, *L* and *M* photoelectron lines resulting from the annihilation quanta are designated by K_2 , L_2 and M_2 respectively. Additional photoelectron lines, designated by K_1 , L_1 , K_3 and L_3 correspond respectively to the *K* and *L* lines for two nuclear gamma-rays having energies 284 ± 3 keV and 655 ± 3 keV. These have also been found recently by a Swiss group.¹⁷

In addition, the Zürich group observed¹⁷ an internal conversion line corresponding to a gamma-transition at 70 keV. If this transition appeared as a gamma-ray its *L* photoelectron line should have appeared at 49 keV

(760 gauss-cm). However, no trace of such a line could be found.

A search of the negatron radiations from Cu^{61} by means of the photographic spectrometer revealed the existence of *K* and *L* internal conversion peaks corresponding to an energy approximately that reported above.¹⁷ In addition, a very weak internal conversion line corresponding to the *K* radiation of the 284 keV transition was observed. Microphotometer tracings of the photographic plates which revealed these internal conversion lines are shown in Fig. 4. Intensities are on an arbitrary logarithmic scale. In order to obtain more accurate information concerning the relative intensities of these lines, they were also studied in the 6-cm radius of curvature spectrometer. The results are shown in Fig. 5. The usual background plus that caused by the negatron radiation from a small amount of Cu^{64} present in the source¹⁶ was subtracted. The energies of the transitions in Ni^{61} as observed from the internal conversion lines are 76 ± 2 keV and 284 ± 3 keV.

IV. DISCUSSION AND TENTATIVE DISINTEGRATION SCHEME

In order to arrive at a disintegration scheme the comparison of the relative intensities of the several radiations from Cu^{61} is necessary. The intensity of the photoelectron lines may be compared to the total positron intensity through the annihilation radiation, whereas the internal conversion peak intensities may be compared directly with the intensity of the beta-distribution.

The comparison of areas of the photoelectron lines in

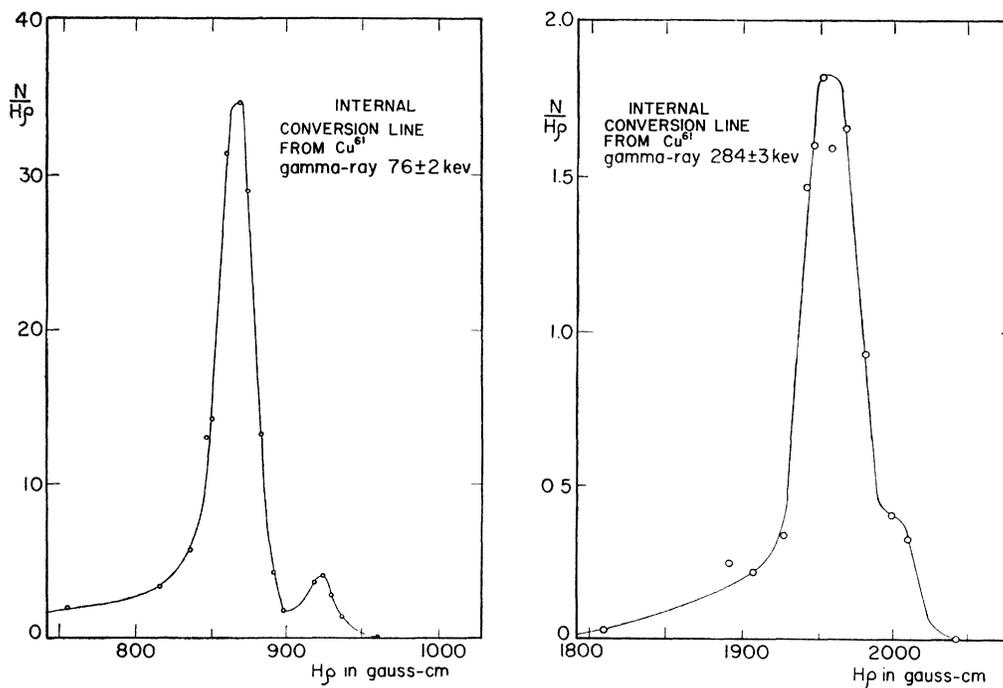


FIG. 5. Internal conversion lines of Cu^{61} as recorded by the constant radius spectrometer.

¹⁷ Boehm, Blaser, Marmier, and Preiswerk, Phys. Rev. **77**, 295 (1950).

TABLE I. Disintegration data for Cu⁶¹.

Radiation	Energy in keV	Observed intensity in % of total β^+ intensity	Calculated K/β^+ ratio	Internal conversion coefficient	Percent of total no. of disintegrations	Possible type of radiation
β_{1205}^+	1205±5	96±1	—	—	63±0.6	Allowed
K_{1205}	1205±5	—	0.29	—	18±0.15	Allowed
β_{550}^+	550±25	4.1±0.8	—	—	2.5±0.12	Allowed
K_{550}	550±25	—	~4.8	—	12.5±4	Allowed
β_{266}^+	266±13	—	—	—	0.03±0.007	Assumed
K_{266}	266±13	—	~100	—	2.3±0.8	Assumed
β_{190}^+	190±20	—	—	—	(1.3±0.6)10 ⁻⁵	Assumed
K_{190}	190±20	—	~500	—	0.65±0.3	Assumed
γ_{284}	284±3	4.5±1	—	0.015	2.9±0.7	Allowed
γ_{76}	76±2	0.93±0.3	—	very high	0.65±0.3	Electric
γ_{655}	655±3	25±9	—	0	18±6	Quadripole ?

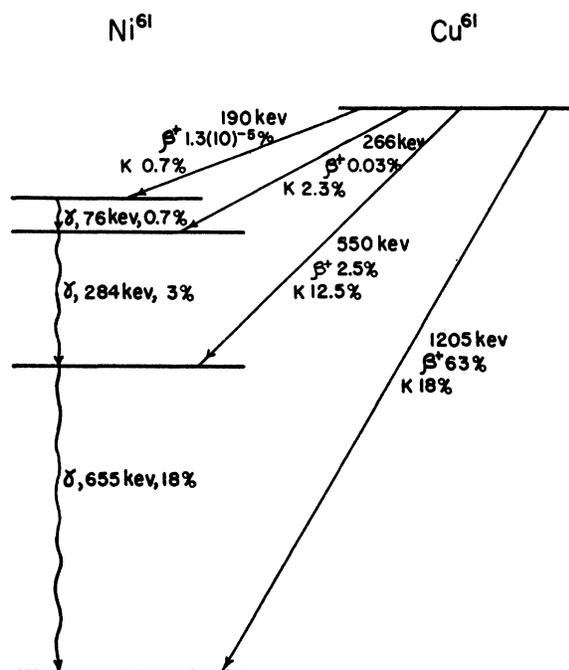
FIG. 6. Proposed decay scheme for Cu⁶¹.

Fig. 3 with appropriate corrections for change in intensity as a function of energy for the photoelectric effect¹⁸ indicates that there are approximately 25 percent as many 655 keV quanta observed as positrons. The relative percentage of 284 keV quanta with respect to positrons is even smaller, being about 4.5 percent. The internal conversion line at 284 keV is only about 0.068 percent as intense as the positron radiations. This leads to an internal conversion coefficient for the 284 keV gamma-transition of $0.068/4.5=0.015$. Since the monochromatic radiation from the 76-keV transition was found in the internal conversion spectrum but not in the photoelectron spectrum, it has been assumed to be very highly internally converted. It is approximately one percent as intense as the positrons.

¹⁸ W. Heitler, *The Quantum Theory of Radiation* (Oxford University Press, London, 1944), second edition, p. 124.

In addition to the positron disintegration of Cu⁶¹, the K-capture transitions must also be considered. The theoretically expected (K -capture/positron) ratio for an allowed Cu⁶¹ transition having a maximum energy 1.205 Mev is¹⁹ $K/\beta^+=0.29$. The corresponding expected ratio for the 550-keV transition is $K/\beta^+=4.8$.

Using this information and the intensities of the 75-keV and 284-keV gamma-transitions, one can construct the tentative disintegration scheme for Cu⁶¹ illustrated in Fig. 6. The experimental evidence offered does not conclusively prove the validity of this disintegration scheme. The proposed scheme is, however, consistent with all experimental evidence and is the only one which to date we have been able to construct which is consistent with this evidence.

The results of this set of experiments are summarized in Table I. It should be noted at this time that, using the evidence given in Table I as arranged in accordance with the disintegration scheme of Fig. 6, the resulting total (K -capture/positron) ratio now expected for Cu⁶¹ is $K_{61}/\beta_{61}^+=0.55$. This is the value found experimentally by Bouchez and Kayas²⁰ in their original experiments and is somewhat higher than the more recent values which have been reported.^{21,22}

A radioactive isotope, Co⁶¹, has been reported as decaying to Ni⁶¹ by negatron emission with a maximum energy of approximately 1.1 Mev.²³ Since sufficient energy is available it is possible that the disintegration scheme proposed here can be further checked through a study of the radiations from Co⁶¹.

The writers would like to express their appreciation to Dr. H. Primakoff and Dr. P. S. Jastram for their many suggestions, and to Mr. M. Ter-Pogossian for assistance in the photoelectron study.

¹⁹ E. Feenberg and G. Trigg, submitted to Rev. Mod. Phys.

²⁰ R. Bouchez and G. Kayas, *J. de phys. et rad.* **10**, 110 (1949).

²¹ Huber, Ruetsche and Scherrer, *Helv. Phys. Acta* **22**, 375 (1949).

²² Bouchez, de Groot, Nataf, and Tolhoek, *Physica* **15**, 863 (1949).

²³ G. T. Seaborg and I. Perlman, *Rev. Mod. Phys.* **20**, 585 (1948).