AUG 25 1964

UCRL-7813

MASTER

University of California
Ernest O. Lawrence
Radiation Laboratory

COPPER L_{III} SHELL X-RAY
PRODUCTION BY 25-100 keV PROTONS

~~NOT FOR PUBLIC RELEASE - OFFICIAL
DISTRIBUTION MAY BE MADE. OFFICIAL
REQUESTS MAY BE FILED. REPORT
CONTAINS NOTHING OF PATENT INTEREST.
PROCEDURES ON FILE IN RECEIVING
SECTION.~~

Livermore, California

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

This paper was submitted for publication in the open literature at least 6 months prior to the issuance date of this Microcard. Since the U.S.A.E.C. has no evidence that it has been published, the paper is being distributed in Microcard form as a preprint.

UCRL-7813

UNIVERSITY OF CALIFORNIA
Lawrence Radiation Laboratory
Livermore, California

Contract No. W-7405-eng-48

COPPER L_{III} SHELL X-RAY PRODUCTION BY 25-100 keV
PROTONS.

D. L. Potter

J. M. Khan

R. D. Worley

June 16, 1964

Copper L_{III} Shell X-ray Production by 25-100 keV Protons*

D. L. Potter, J. M. Khan, and R. D. Worley

Lawrence Radiation Laboratory, University of California

Livermore, California

June 16, 1964

ABSTRACT

Recent x-ray production studies of L-shell x rays from copper by proton bombardment (100-500 keV) have been extended to 25-keV energy. In the region of the periodic table near copper the higher intensity (L_{β_1} , $L_{\beta_{3,4}}$) characteristic x-ray lines from the L_I and L_{II} shells are absorbed in the L_{III} shell. By using thin copper absorber foils the characteristic x rays from the L_{III} shell of copper have been isolated from the L_I and L_{II} characteristic x rays. The thick target yields of both groups are presented as well as the x-ray production cross section for the L_{III} shell. With a fluorescence yield of 0.05 the L_{III} shell ionization cross section decreases from $3.6 \times 10^{-22} \text{ cm}^2$ at 100 keV to $2.3 \times 10^{-24} \text{ cm}^2$ at 25 keV. The $L_{III}/L_I + L_{II}$ x-ray intensity ratio decreases from 1.4 at 100 keV to an extrapolated value of approximately 1 at zero proton energy.

INTRODUCTION

Characteristic x rays produced when targets are bombarded by heavy ions have been studied since 1913.¹ The study of proton-excited characteristic x rays began in 1933² and continued until 1937.^{3,4} The work was again revived in 1953⁵ and thereafter followed many experiments employing protons of high energy

(> 500 keV).⁶ Recently measurements have been made using protons of energy less than 500 keV⁷⁻⁹ and observing x rays of increasingly longer wavelength.¹⁰ By using a thin window proportional counter and a low energy proton accelerator (0-140 kV) measurements have been extended to 25 keV energy.¹¹

The present work involves the measurement of the thick target yield of characteristic x rays produced in the L shell of copper. With the aid of additional data (the stopping power of copper for protons and the mass absorption coefficient of copper for its own L-shell x rays) one can obtain the x ray production cross section. The ionization cross section obtained is compared with one based on Born approximation considerations and is found to be over two orders of magnitude lower at 25-keV proton energy.

In the region of the periodic table near copper a number of the elements have an L_{β_1} ($L_{II}-M_{IV}$) x-ray line which is greater in energy than the L_{III} absorption edge. Copper L-shell x-ray energies extend from 814 eV (L_2) to 1024 and 1027 eV ($L_{\beta_{3,4}}$) with the L_{III} absorption edge at 935 eV. By placing a copper foil between the thick target sample and the detector, the L_{III} absorption edge of copper was used to attenuate the L_I and L_{II} originated x rays (mass absorption jump ratio at L_{III} edge ≈ 5).¹² Thus the thick target yield, with and without the appropriate copper foil, enables one to separately measure the L_{III} subshell x-ray yield and the combined $L_I + L_{II}$ x-ray yield. The L_{III} yield thus obtained can be employed to calculate the x-ray production cross section for the L_{III} shell. Additionally, by knowing the probability of radiationless transitions from the L_{III} shell the ionization cross section can be determined.

APPARATUS AND METHOD

The apparatus consisted of a proton source, 140-kV accelerating column, analyzing magnet, target chamber, proportional counter, and associated pulse

counting equipment. This accelerator has been described previously¹¹; the current available on the target at 25 kV is $\approx 5 \mu\text{A}$.

The x rays were produced in a thick target sample mounted at 45° to the incident proton beam. The proportional counter, mounted at 90° to the incident proton beam, was maintained at a pressure of 120 mm of Hg. The counter was operated in flow mode, $\approx 30 \text{ cc/min}$, with P-10 gas (90% argon, 10% methane). A 1.8-mg/cm^2 Al window was mounted between the counter volume and the target chamber vacuum. The counter voltage was 1100 V. The counter (2 in. i. d. $\times$ 12 in. length with a 0.003-in. stainless steel center wire) represented ≈ 5 absorption lengths for the copper L_{a_1, a_2} ($L_{III}-M_{IV, V}$) x rays at 120 mm of Hg.

Interposing copper and aluminum foils between the target and proportional counter isolated the L_{III} x-ray lines and allowed measurement of the counter window absorption. The Al absorber (window) was 1.8 mg/cm^2 and the copper absorber was 1.06 mg/cm^2 . The relationship of the observed counts with copper and aluminum foils has been described in detail in ref. 10.

EXPERIMENT

To calculate the ionization cross section from the thick target yield the following expressions are employed:⁶

$$\sigma_I = \frac{1}{\omega} \sigma_x \quad (1)$$

$$\sigma_x = \frac{1}{n} \frac{dI}{dE} \frac{\mu}{d(\rho x)} + \frac{\mu}{np} I_\mu \quad (2)$$

where σ_I and σ_x are in cm^2 , n in atoms/g, $\frac{dE}{d(\rho x)}$ in $\text{keV-cm}^2/\text{g}$, and μ/ρ in cm^2/g .

Since the fluorescence yield ω is a function of the particular shell, the above expressions assume that the radiation observed, (I_μ) , comes from the filling of a single shell in the atom. Additionally, the radiation must be approximately monoenergetic to allow for reliable self-absorption determinations.

The present experiment closely achieves the above conditions in the following manner. Copper foils are inserted between the target and the detector. By virtue of the L_{III} absorption edge the predominant radiations transmitted by the foil are L_{a_1, a_2} , L_η , and L_ζ . Figure 2 from ref. 10 shows an x-ray intensity spectra of copper produced by electron bombardment. It can be seen that the L_η and L_ζ radiations are only a few percent of the total radiations transmitted below the L_{III} absorption edge; therefore the observed radiation is approximately monoenergetic and consisting principally of L_{a_1, a_2} . Assuming the ratio of L_η and L_ζ to L_{a_1, a_2} radiations to be of the same order of magnitude for proton-excited spectra, measurements of the mass absorption coefficient in copper for the radiations transmitted by the copper absorber foil yield a value of $1635(60) \text{ cm}^2/\text{g}$ (constant for proton energies between 40-500 keV).

Table I gives the thick target yields for the L_{III} and $L_I + L_{II}$ radiations and the ionization cross section for the L_{III} radiations. The error in the thick target yield is estimated to be $\pm 15\%$ for all energies except 25 and 30 keV where low signal-to-noise values necessitate an error of $\pm 30\%$. The uncertainty in the ionization cross section is dependent upon fluorescence yield (ω) and self-absorption (μ/ρ) estimates and stopping power $dE/d(px)$ extrapolations. The total uncertainty in the ionization cross section is felt to be less than a factor of 5.

DISCUSSION

A comparison of the theoretical ionization cross section (for the total L shell) using Born approximation calculations with the experimental L_{III} ionization cross section is given in Fig. 1. The experimental cross sections deviate from the theoretical at 25 keV by a factor of 650. This large deviation is of interest if only to support the fact that an adequate theory for L-shell ionization cross sections for low energy protons has yet to be developed. A program has been initiated to study the possible application to L-shell cross sections of a deflected trajectory theory which assumes the Coulomb repulsion of the low energy proton by the nucleus causes a proton trajectory which deviates from the conditions necessary for applicability of the Born approximation. This theory has been successfully applied to K-shell cross sections.¹³

An interesting result of this experiment is the ratio of $L_{III}/L_I + L_{II}$ x-ray intensities. As was noted in ref. 10 and is further shown in Fig. 2 the $L_{III}/L_I + L_{II}$ x-ray intensity ratio appears to extrapolate to unity at zero proton energy.

This ratio has not been observed for electron-excited spectra. Skinner reported that Bullen has found a ratio of 10 for copper L_{III}/L_{II} emission band intensity.¹⁴ Holliday recently reported a ratio of 4 for Cr, Co, and Ni L_{III}/L_{II} emission band intensity.¹⁵ In ref. 10 the authors undertook an examination of the electron-excited spectra of copper and found an $L_{III}/L_I + L_{II}$ x-ray intensity ratio of 5, (negligible L_I contribution observed).

From a statistical approach to x-ray spectra intensities (intensity of x rays originating from a single level given by $2j + 1$) the $L_{III}/L_I + L_{II}$ x-ray intensity ratio should equal unity but as noted above is not observed in electron-excited spectra. (See, however, the spectra of Cu L observed by Cauchois in 1953.¹⁶)

A possible explanation for the difference between the proton and electron data is given by the effect of self-absorption of the target material for its own x rays. Self-absorption plays a role of some prominence in electron-excited x-ray spectra¹⁷ and may be even more important in L x-ray spectra.

The authors are presently investigating the apparent intensity anomalies between proton excited and electron excited x-ray spectra.

REFERENCES

*Work performed under the auspices of the U. S. Atomic Energy Commission.

- ¹J. Chadwick, Phil. Mag. 25, 193 (1913).
- ²C. Gerthsen and W. Reusse, Z. Physik 34, 478 (1933).
- ³O. Peter, Ann. der Physik 27, 299 (1936).
- ⁴M. S. Livingston, F. Genevieve, and E. Konopinski, Phys. Rev. 51, 835 (1937).
- ⁵H. W. Lewis, B. E. Simmons, and E. Merzbacher, Phys. Rev. 91, 943 (1953).
- ⁶E. Merzbacher and H. W. Lewis, Encyclopedia of Physics (Springer-Verlag, Berlin, 1958), vol. 34, p. 166. Contains a general review of the field up to 1957.
- ⁷S. Messelt, Nucl. Phys. 5, 435 (1958).
- ⁸R. C. Jopson, Hans Mark, and C. D. Swift, Phys. Rev. 127, 1612 (1962).
- ⁹J. M. Khan and D. L. Potter, Phys. Rev. 130, A890 (1964).
- ¹⁰J. M. Khan, D. L. Potter, and R. D. Worley, Phys. Rev. 133, A316 (1964).
- ¹¹R. D. Worley, D. L. Potter, and J. M. Khan, "Characteristic x-ray production in the K-shell of magnesium and aluminum by 25 keV to 100 keV protons"(submitted for publication in Phys. Rev.).
- ¹²A. H. Compton and S. K. Allison, X-Rays in Theory and Practice (D. Van Nostrand Co., Princeton, N. J., 1960), 2nd ed.
- ¹³J. Bang and J. M. Hansteen, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. 31, No. 13 (1959).
- ¹⁴H. W. B. Skinner, Phil. Trans. Roy. Soc. (London) Ser. A239, 95 (1940).

- ¹⁵ J. E. Holliday, J. Appl. Phys. 33, 3259 (1962).
- ¹⁶ Y. Cauchois, Phil. Mag. 44, 173 (1953).
- ¹⁷ H. P. Hanson and J. Herrera, Phys. Rev. 105, 1483 (1957).

Table I. Thick-target yield and x-ray production cross sections.^a

E_p (keV)	25	30	40	50	60	70	80	90	100
<u>L_{III} Shell</u>									
I_μ	2.5×10^{-8}	1.05×10^{-7}	5.5×10^{-7}	1.58×10^{-6}	3.3×10^{-6}	6.1×10^{-6}	9.8×10^{-6}	1.47×10^{-5}	2.0×10^{-5}
$\frac{d(I_\mu)}{dE}$	8.2×10^{-9}	2.4×10^{-8}	7.5×10^{-8}	1.4×10^{-7}	2.3×10^{-7}	3.2×10^{-7}	4.3×10^{-7}	5.7×10^{-7}	6.1×10^{-7}
$\frac{dE}{d(\rho x)}^b$	1.3×10^5	1.5×10^5	1.7×10^5	1.9×10^5	2.0×10^5	2.1×10^5	2.1×10^5	2.2×10^5	2.2×10^5
$\frac{1}{n} \frac{d(I_\mu)}{dE} \cdot \frac{dE}{d(\rho x)}$	1.1×10^{-25}	3.8×10^{-25}	1.3×10^{-24}	2.8×10^{-24}	4.9×10^{-24}	7.1×10^{-24}	9.8×10^{-24}	1.3×10^{-23}	1.5×10^{-23}
$\frac{1}{n} \frac{\mu}{\rho} (I_\mu)$	4.3×10^{-27}	1.8×10^{-26}	9.5×10^{-26}	2.7×10^{-25}	5.7×10^{-25}	1.05×10^{-24}	1.7×10^{-24}	2.5×10^{-24}	3.4×10^{-24}
$\omega_{L_{III}} \sigma_I = \sigma_x$	1.1×10^{-25}	4.0×10^{-25}	1.4×10^{-24}	3.1×10^{-24}	5.4×10^{-24}	8.1×10^{-24}	1.15×10^{-23}	1.6×10^{-23}	1.8×10^{-23}
$\omega_{L_{III}} = 0.05$									
$\sigma_I(\text{exp})$	2.3×10^{-24}	8.0×10^{-24}	2.9×10^{-23}	6.1×10^{-23}	1.08×10^{-22}	1.6×10^{-22}	2.3×10^{-22}	3.0×10^{-22}	3.6×10^{-22}
$\sigma_I(\text{theor})^c$	1.5×10^{-21}	2.4×10^{-21}	4.7×10^{-21}	7.8×10^{-21}	1.14×10^{-20}	1.6×10^{-20}	2.0×10^{-20}	2.5×10^{-20}	3.0×10^{-20}
$\sigma_I(\text{theor})/\sigma_I(\text{exp})$	650	300	160	138	105	100	90	83	83
<u>L_I + L_{II} Shells</u>									
I_μ	2.5×10^{-8}	1.05×10^{-7}	4.7×10^{-7}	1.35×10^{-6}	2.6×10^{-6}	5.1×10^{-6}	7.4×10^{-6}	1.05×10^{-5}	1.5×10^{-5}

^aUnits: I_μ - x rays per proton; dI_μ/dE - x rays per proton per keV; $dE/d(\rho x)$ - keV-cm²/g; $\frac{1}{n} = 1.056 \times 10^{-22}$ g/atom; σ_x - cm².

^bS. D. Warshaw and S. K. Allison, Rev. Mod. Phys. 25, 779 (1953).

^cE. Merzbacher and H. W. Lewis, Handbook of Physics (Springer-Verlag, Berlin, 1958), vol. 34, p. 166. Ionization cross section calculated for total L shell.

Fig. 1. Cu(L_{III}) ionization cross section.

GLL-646-1700

Fig. 2. $\text{Cu } L_{III}/\text{Cu } L_I + L_{II}$ x-ray intensity ratio.

FIGURE CAPTIONS

Fig. 1. $\text{Cu}(L_{\text{III}})$ ionization cross section.

Fig. 2. $\text{Cu } L_{\text{III}}/\text{Cu } L_{\text{I}} + L_{\text{II}}$ x-ray intensity ratio.

— LEGAL NOTICE —

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission " includes any employee or contractor of the commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.