

AD-A042 228

EMORY UNIV ATLANTA GA

L SHELL FLUORESCENCE YIELDS AND COSTER-KRONIG TRANSITION PROBAB--ETC(U)

JAN 75 J M PALMS, P V RAO, R E WOOD

DA-ARO-D-31-124-70-678

F/G 20/8

UNCLASSIFIED

ARO-9119.4-P

NL

| OF |  
AD  
A042228



END

DATE  
FILMED  
8-77

| REPORT DOCUMENTATION PAGE                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        |                                                                                  | READ INSTRUCTIONS<br>BEFORE COMPLETING FORM |
|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|----------------------------------------------------------------------------------|---------------------------------------------|
| 1. REPORT NUMBER<br><b>ARO 949.4-P1</b>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                          | 2. GOVT ACCESSION NO.                                                            | 3. RECIPIENT'S CATALOG NUMBER               |
| 4. TITLE<br><b>L Shell Fluorescence Yields and Coster-Kronig Transition Probabilities Using Radioactive Sources.</b>                                                                                                                                                                                                                                                                                                                                                                                                                             | 5. TYPE OF REPORT & PERIOD COVERED<br><b>Final Report, 6-26-70 - 1-1-75</b>      |                                             |
| 7. AUTHOR(s)<br><b>John M./Palms,<br/>P. Venugopala/Rao<br/>R. E./Wood</b>                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | 6. PERFORMING ORG. REPORT NUMBER<br><b>26 MAY 74 - 1 JAN 75</b>                  |                                             |
| 9. PERFORMING ORGANIZATION NAME AND ADDRESS<br><b>Emory University, Atlanta, Ga. 30322</b>                                                                                                                                                                                                                                                                                                                                                                                                                                                       | 8. CONTRACT NUMBER(s)<br><b>DA-ARO-D31-124-70-G78<br/>DA-ARO-D31-124-73-G129</b> |                                             |
| 11. CONTROLLING OFFICE NAME AND ADDRESS<br><b>U. S. Army Research Office<br/>Box CM, Duke Station<br/>Durham, North Carolina 27706</b>                                                                                                                                                                                                                                                                                                                                                                                                           | 10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS                      |                                             |
| 14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)                                                                                                                                                                                                                                                                                                                                                                                                                                                                      | 12. REPORT DATE<br><b>1 Jan 1975</b>                                             |                                             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                  | 13. NUMBER OF PAGES<br><b>7 (12) 10p.</b>                                        |                                             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                  | 15. SECURITY CLASS. (of this report)<br><b>Unclassified</b>                      |                                             |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                  | 15a. DECLASSIFICATION/DOWNGRADING SCHEDULE<br><b>NA</b>                          |                                             |
| 16. DISTRIBUTION STATEMENT (of this Report)<br><b>Approved for public release; distribution unlimited.</b>                                                                                                                                                                                                                                                                                                                                                                                                                                       |                                                                                  |                                             |
| 17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)<br><b>NA</b>                                                                                                                                                                                                                                                                                                                                                                                                                                          |                                                                                  |                                             |
| 18. SUPPLEMENTARY NOTES<br><b>The findings in this report are not to be construed as an official Department of the Army position, unless so designated by other authorized documents.</b>                                                                                                                                                                                                                                                                                                                                                        |                                                                                  |                                             |
| 19. KEY WORDS (Continue on reverse side if necessary and identify by block number)<br><b>Fluorescence yields, Non-radiative transitions, Decay schemes, x-ray detectors electron detectors, multiple vacancy atomic states</b>                                                                                                                                                                                                                                                                                                                   |                                                                                  |                                             |
| 20. ABSTRACT (Continue on reverse side if necessary and identify by block number)<br><b>Accurate knowledge of x-ray fluorescence yields is an important ingredient in the solution of many practical problems and in the understanding of the decay mechanisms of atomic states characterized by inner-shell vacancies. In the present work, coincidence and singles techniques, employing high resolution semiconductor x-ray detectors and radioactive isotopes, are applied to obtain information on the decay of inner-shell</b> → next page |                                                                                  |                                             |

ADA 042228

DDC FILE COPY

→ vacancy atomic states. Precision measurements of K-shell fluorescence yields at  $Z = 78, 82$  and  $92$  are performed. X ray and Coster-Kronig yields at  $Z > 60$  are measured. It is clearly established that a discrepancy exists in the interpretation of experimentally observed  $L_2$  subshell Coster-Kronig transition probability around  $Z = 80$ . A careful and detailed study of the decay of (LL)-, (LX)-vacancy atomic states at  $Z = 82$  (in the decay of  $^{207}\text{Bi}$ ) is made by (x-ray)-(x-ray) and (x-ray) Auger electron coincidence techniques.

UNCLASSIFIED

L SHELL FLUORESCENCE YIELDS  
AND  
COSTER-KRONIG TRANSITION PROBABILITIES  
USING RADIOACTIVE ISOTOPES

Final Report

P. Venugopala Rao, J. M. Palms, R. E. Wood

January 1975

U. S. Army Research Office

DR-ARO-D31-124-70-G78  
DR-ARO-D31-124-73-G129

Emory University  
Atlanta, Georgia

Approved for Public Release  
Distributions Unlimited

|                                 |                                                   |
|---------------------------------|---------------------------------------------------|
| ACCESSION for                   |                                                   |
| NTIS                            | White Section <input checked="" type="checkbox"/> |
| DDC                             | Buff Section <input type="checkbox"/>             |
| UNANNOUNCED                     | <input type="checkbox"/>                          |
| JUSTIFICATION _____             |                                                   |
| BY _____                        |                                                   |
| DISTRIBUTION/AVAILABILITY CODES |                                                   |
| Dist.                           | 1/ or SP. CIAL                                    |
| A                               |                                                   |



## I. STATEMENT OF THE PROBLEM

Accurate knowledge of x-ray fluorescence yields is an important ingredient in the solution of many practical problems and in the understanding of the decay mechanisms of atomic states characterized by inner-shell vacancies. Before the advent of high resolution semiconductor detectors reliable information existed only on the x-ray yields of single K-shell vacancy atomic states. Late in the 'sixties, application of coincidence techniques using semiconductor x-ray detectors to measure the fluorescence yields of  $L_2$  and  $L_3$  subshells at high  $Z$ , where L shell characteristic x rays' energies are in the range of 10 to 15 keV, was successfully demonstrated at Emory. The problem was to extend these measurements over a wide range of  $Z$  values in order to obtain sufficiently accurate values for L subshell yields. During the four-year period of this grant, this objective was not only completely realized, but also ground work was done for the studies of the decay of complex atomic states. These studies, besides providing a reliable experimental technique, have provided sufficient incentive and encouragement to theoreticians in atomic physics to provide careful theoretical estimates of x-ray emission rates and non-radiative transition rates as evidenced in the work of Crasemann, Bhalla, McGuire and Scofield. This final report is to summarize and put in proper perspective the work performed at Emory.

## II. RESULTS

### a) Measurement of $L_2$ and $L_3$ subshell yields

By measuring coincidences between  $K\alpha$  x rays and L x rays,  $L_2$  subshell Coster-Kronig yields  $f_{23}$  were measured. The Coster-Kronig yields at 7 atomic

numbers in the region  $63 \leq Z \leq 82$  were carefully measured. (Ref. 1,6,2). These were compared with the theoretical estimates. The general trend of the theoretical estimates of  $f_{23}$  is substantiated by these experiments but there exists a substantial discrepancy between theory and experiment in the region around  $Z = 80$ . The experimental values are 25% higher than theoretical estimates in this region. In an experiment in which M x rays (3 keV to 4 keV) were observed in coincidence with  $K_\alpha$  and  $K_\beta$  x rays it was established that the  $f_{23}$  Coster-Kronig transition probability does not have a substantial radiative component. In addition  $\omega_2$  and  $\omega_3$  were measured at  $Z = 73$ <sup>(1)</sup> and L x ray emission rates from  $L_2$  and  $L_3$  subshells were measured in the region of  $65 \leq Z \leq 94$ .<sup>(2)</sup>

#### b) Measurement of $L_1$ subshell yields

Compared to  $L_2$  and  $L_3$  subshells, very little information was available on  $L_1$  subshell decay. An important contribution of this work was the development of methods to obtain information on the x ray emission from  $L_1$  subshells. Three basic techniques were employed: The first technique involved studying a well resolved L x ray spectrum and analyzing it to determine the number of  $L_1$  subshell characteristic rays emitted by the samples and calculating the  $L_1$  subshell characteristic fluorescence yield  $\omega_1$  by using the knowledge of  $L_1$  subshell vacancy states produced in the sample. This method was employed at two Z-values 82 and 83 (Ref: 2,17). This method can be successfully employed at high Z using suitable radioactive isotopes (in which L shell ionization takes place substantially) and high resolution x-ray detectors.

The second technique was to employ L conversion electrons as signals of L vacancy states and observe L x rays in coincidence with them as in the

case of  $Z = 82$ . This method is also potentially capable of being extended to other isotopes by properly choosing the isotope in which L-conversion is taking place predominantly in  $L_1$  subshell.

The third method was to select nuclear transitions in which  $L_1$  conversion is predominant. By observing L x ray emission, by singles and coincidence methods, average  $L_1$  subshell fluorescence yields  $\omega_1$  were measured at  $Z = 63, 73, 77$  and  $83$ . (Ref. 1,17,18).

c) Si(Li) spectrometer for low energy photons and electrons

These measurements just discussed, required a high resolution Si(Li) spectrometer the efficiency of which had to be known. Hence a considerable part of research involved developing methods to measure the efficiency of the detector using routinely available laboratory-standard radioactive isotopes. High resolution windowless Si(Li) spectrometers, which can also simultaneously detect electrons emitted by radioactive isotopes in Auger process and internal conversion process (and with a facility to rapidly change the electron sources mounted inside the vacuum) was constructed. (Ref. 4).

d) Measurement of high-Z K-shell fluorescence yields

By comparing the relative intensities of K Auger electrons and K x rays emitted in the radioactive decay of carrier free  $^{195}\text{Au}$ ,  $^{207}\text{Bi}$ , and  $^{235}\text{Np}$ , the K-fluorescence yield  $\omega_K$  was determined with high accuracy at  $Z = 78, 82$ , and  $92$ . There were no precision measurements available in this region prior to this work. (Ref. 5).

In addition relative intensities of (K-LL), (K-LX) and (K-XY) Auger electrons at  $Z = 82$  and  $92$  were measured. (Ref. 5,15,17).



e) Theoretical studies

As part of this investigation, it was necessary to have clear and definite information on the L-vacancy production in various radioactive isotopes used in this work. Hence a comprehensive and systematic study of the L and M shell vacancy production following the decay of a K vacancy or L vacancy state was made. All available experimental values of Auger transition rates in literature were compiled, initially reviewed and tabulated for use by research workers in this area. Theoretical estimates based on j-j coupling model were computed and compared with experiment data. (Ref. 7).

f) Decay scheme studies

In addition, it was also necessary to obtain more precise information on decay schemes of radioactive isotopes used in this work. Particularly careful study of the decay schemes of  $^{151}\text{Gd}$ ,  $^{159}\text{Dy}$ , and  $^{194}\text{Os}$  was completed. (Ref. 10,18).

g) Study of double vacancy states

The most significant contribution of this work, however, was to extend the above discussed experimental techniques to study the double inner-shell vacancy states. (Ref. 9,13,15,17). The basic principles and details of the experimental technique using the Si(Li) detector and x ray detector were given in Ref. 15. In this work the radioactive decay of LL, LX and MX vacancy states in Pb created during the decay of  $^{207}\text{Bi}$  was studied by observing coincidences between K Auger electrons and LX rays, LX rays and LX rays, and KX rays and MX rays. The angular correlation between the two cascade LX rays emitted during the decay of double L-vacancy states was studied. This study was the first of its kind and can be extended to yield



information on double vacancy states over a large range of high-Z values above  $Z = 70$ .

### III. CONCLUSIONS

The advantage of coincidence techniques in the study of the decay of inner shell vacancy atomic states (popular in nuclear spectroscopy) was clearly demonstrated. This was facilitated by the judicious use of high resolution semiconductor detectors.

A wealth of information was accumulated on the decay of  $L_1$ ,  $L_2$  and  $L_3$  subshell vacancy states at high  $Z$  ( $>60$ ). It was clearly established that a discrepancy exists in the interpretation of experimentally observed  $L_2$  subshell Coster-Kronig transition probability around  $Z = 80$ .

Precision measurements of K-shell fluorescence yields at high  $Z$  were performed, thus extending the use of the semi empirical equation  $[\omega_K/(1-\omega_K)]^{1/4} = B_0 + B_1Z + B_3Z^3$  to predict fluorescence yields.

For the first time careful measurements were performed to study the decay of double inner shell vacancy states and ground work was laid to extend these techniques over a wide range of  $Z$ .

IV. LIST OF PUBLICATIONS

1. Electron capture decay of  $^{181}\text{W}$ : L subshell fluorescence and Coster-Kronig yields in Ta, Z Physik 239, 423-428 (1970). S. Mohan, R. W. Fink, R. E. Wood, J. M. Palms, P. Venugopala Rao.
2. High Z L subshell x-ray emission rates, Phys. Rev. A3, 1568-1575 (1971). P. Venugopala Rao, J. M. Palms and R. E. Wood.
3. Does the Coster-Kronig transition probability have a radiative component? Phys. Rev. A4, 846-849 (1971). M. H. Chen, B. Crasemann, P. Venugopala Rao, J. M. Palms, and R. E. Wood.
4. Si(Li) spectrometers for electrons and low-energy photons, Nucl. Instr. Meth. 94, 245-252 (1971). R. E. Wood, P. Venugopala Rao, O. H. Puckett, and J. M. Palms.
5. Precision determination of high Z subshell fluorescence yields from  $^{195}\text{Au}$ ,  $^{207}\text{Bi}$  and  $^{235}\text{Np}$  decays, Z. Physik 249, 373-385 (1972). J. S. Hausen, J. C. McGeorge, R. W. Fink, R. E. Wood, P. Venugopala Rao, and J. M. Palms.
6.  $L_2$ - $L_3$  Coster-Kronig transition probability at  $Z = 82$ , Phys. Rev. A5, 11-13 (1972). R. E. Wood, J. M. Palms, and P. Venugopala Rao.
7. Atomic vacancy distributions produced by inner-shell ionization. Phys. Rev. A5, 997-1012 (1972). P. Venugopala Rao, M. H. Chen and B. Crasemann.
8. Mean L fluorescence yields at  $Z = 63$  and  $83$ . (1973). Proc. International Conference on Innershell ionization Phenomena and Future Applications, Atlanta, U.S. Atomic Energy Comm. Rep. No. CONF-720404, Oak Ridge, Tenn. R. E. Wood, V. R. Veluri, J. M. Palms, and P. Venugopala Rao.
9. L-shell fluorescence yields of double vacancy states in Pb. (1973). Proc. International Conference on Innershell ionization Phenomena and Future Applications. Atlanta, U.S. Atomic Energy Comm. Rep. No. CONF-720404, Oak Ridge, Tenn. V. R. Veluri, R. E. Wood, J. M. Palms, and P. Venugopala Rao.
10. K-electron capture probabilities in the decays of  $^{151}\text{Gd}$  and  $^{159}\text{Dy}$ . Z. Physik 260, 47-56 (1973). H. Genz, R. E. Wood, J. M. Palms, and P. Venugopala Rao.
11. X-ray fluorescence yields, Auger and Coster-Kronig transition probabilities, Rev. Mod. Phys. 44, 766-813 (1972). W. Bambynek, B. Crasemann, R. W. Fink, H. Freund, Hans Mark, C. D. Swift, R. E. Bice and P. Venugopala Rao.
12.  $L_2$  subshell Coster-Kronig yields at high Z, J. Phys. B: Atom. Molec. Phys. 7, 1486-1493 (1974). V. R. Veluri, R. E. Wood, J. M. Palms, and P. Venugopala Rao.

13. Radiative decay of double inner-shell vacancy states in Pr and Pb, *Physica Fennica* 9, supplement 51, 253-255 (1974).
14. "Inner-shell transition measurements with radioactive atoms," Chapter in *Atomic Inner-shell Processes*, ed. B. Crasemann, Academic Pr. vol. 2, p. 1 (1975). P. Venugopala Rao.
15. Average L-x-ray fluorescence yields of double-vacancy states in Pb, *J. Phys. B: Atom. Molec. Phys.* 10, 399-411, 1977. P. Venugopala Rao, R. E. Wood and V. R. Veluri.

#### Theses

16. H. Genz: Measurements of Electron Capture Probabilities (Ph.D. 1971, Emory University).
17. V. R. Veluri: L X-ray Emission Probabilities from Singly and Doubly-ionized Atoms (Ph.D. 1973, Emory University).
18. P. A. Indira: The Decay of Osmium-194 L-shell Fluorescence Yields at  $Z = 59$  and  $77$  (M.S. 1974, Emory University).

#### Books

Proceedings of the International Conference on Inner-shell Ionization Phenomena and Future Applications, Vols. I, II, III and IV. 2384 pages, U.S. Atomic Energy Commission Rept. No. CONF-720404. Oak Ridge, Tenn. (1973).

### V. PARTICIPATING PERSONNEL

|                   |                                |
|-------------------|--------------------------------|
| J. M. Palms       | Professor of Physics           |
| P. Venugopala Rao | Associate Professor of Physics |
| R. E. Wood        | Associate Professor of Physics |