PHYSICAL REVIEW A VOLUME 39, NUMBER 3 FEBRUARY 1, 1989

Measurement of the $L\beta$ to $L\alpha$ intensity ratio through a selective excitation of L_{III} subshell in elements of $Z \geq 70$

G. Sree Krishna Murty, M. V. S. Chandrasekhar Rao, C. V. Raghavaiah, S. Bhuloka Reddy, G. Satyanarayana, and D. L. Sastry

Swamy Jnanananda Laboratories for Nuclear Research, Andhra University, Visakhapatnam 530 003, Andhra Pradesh, India

(Received 22 March 1988; revised manuscript received 29 September 1988)

The intensity ratio of $L\beta_2+L\beta_5+L\beta_6$ to $L\alpha_1+L\alpha_2$ was measured in seven elements ($Z \geq 70$) through a selective excitation of the L_{III} subshell using an x-ray tube. A high-resolution Si(Li) detector system coupled to a computer controlled multichannel analyzer was used in these measurements. The results show that the relative transition probabilities $L\beta$ and $L\alpha$ agree with the relativistic Hartree-Fock-Slater calculations in the case of Yb and U, while a discrepancy of 3.1–8.7 % between experiment and theory is found for the other elements.

INTRODUCTION

In recent years the availability of high-resolution solid-state detectors and crystal spectrometers and sophisticated data-acquisition systems has made it possible to obtain rich information on K and L x-ray-component ratios. These results are found to be useful in checking theories as well as in nuclear physics applications. We have been carrying out measurements on $I(K\beta)/I(K\alpha)$ (Refs. 1 and 2) and L-component ratios[3] with the help of a high-resolution [full width at half maximum (FWHM)=160 eV at 5.9 keV of the Mn $K\alpha$ line] Si(Li) detector and using x-ray tube and radio isotopes for the purpose of exciting the sample. The resolution of the Si(Li) detector does not permit the splitting of different K and L components. As such, many of the authors have reported[4–12] only the intensity ratios of the $K\beta$, $K\alpha$, Ll, $L\alpha$, $L\beta$, and $L\gamma$ lines. It is to be noted that each component represents the sum of different $K\beta$ and $K\alpha$ components. However, a crystal spectrometer helps resolve all the individual components like $K\alpha_1, K\alpha_2, K\beta_1, K\beta_2$, etc. The disadvantage of the crystal spectrometer is poor transmission, which results in large uncertainties in the measurements.

Salem *et al.*[13] have measured the K- and L-component ratios in elements with $Z \geq 57$ using an electron source of excitation and a crystal spectrometer. Their results show good agreement between experiment and theory based on the relativistic Hartree-Fock-Slater (RHFS) potential for the $I(L\alpha_2)/I(L\alpha_1)$ ratio but a discrepancy of about 22% in the $I(L\beta_{2,15})/I(L\alpha_1)$ ratio is reported for elements of $Z \approx 82$. The earlier measurements,[4–11] by different authors concerned with the relative intensities of L x-ray lines who measured the $I(L\alpha)/I(L\beta)$ and $I(L\alpha)/I(L\gamma)$ ratios using a Si(Li) detector, show wide discrepancies.

In the present studies we have used x-ray-tube excitation for exciting the samples in seven elements with $Z \geq 70$. As the x-ray tube voltage can be varied from 5 to 50 kV there is a flexibility of selectively exciting the L subshells. For instance, if the excitation energy is fixed at 15 kV, which is less than the L_{I} and L_{II} subshell binding energy of Bi, only the L_{III} subshell vacancies can be created via photoionization. Similarly, the excitation energy is fixed in the case of other elements also for L_{III} subshell excitation. In this situation only transitions to L_{III} subshell take place from the higher subshells resulting in the emission of Ll, $L\alpha$, $L\beta_{2,15}$, $L\beta_5$, and $L\beta_6$ characteristic x-ray lines. Thus, the transitions to L_{I} and L_{II} subshells are suppressed. This nomenclature is that followed by Siegbahn. In our earlier study,[14] we have reported L-component intensity ratios corresponding to sum of all the L components arising out of L_{I}, L_{II}, and L_{III} subshell excitation. If one is confined to only one subshell excitation, there is an added advantage of eliminating corrections due to Auger and Coster-Kronig transitions. This method of exciting the L_{III} subshell in the seven elements referred to earlier is followed in the present study. Such an attempt appears to have not been made so far. The detection of L x rays was accomplished using a Si(Li) detector. In these measurements we expect information on the relative intensities of $L\alpha_2+L\alpha_1$ sum and $L\beta_{2,15}+L\beta_5+L\beta_6$ sum because of limited resolution of the detector. The results thus obtained are compared with those due to Salem *et al.* and RHFS calculations.

EXPERIMENTAL DETAILS

Sample preparation. The targets were used in powdered form, with lead in the form of the acetate and thorium as the sulfate. The other elements were taken in the form of oxides. They were uniformly spread on a cellophane tape backing fixed to a Perspex ring. The thickness of the sample is of the order of 1 mg/cm^2.

The experimental arrangement was described in several of our earlier papers.[1,2] It essentially comprises of an x-ray generator fitted to a rhodium anode, whose voltage can be varied from 5 to 50 kV and the current up to 500 μA, the maximum power being 25 W. The Si(Li) detector has a Be window of thickness 7.5 μm and an active area of 30 mm^2. The detector is coupled to a cooled field-effect transistor (FET) preamplifier and maintained at liquid-nitrogen temperature. The pulses were amplified,

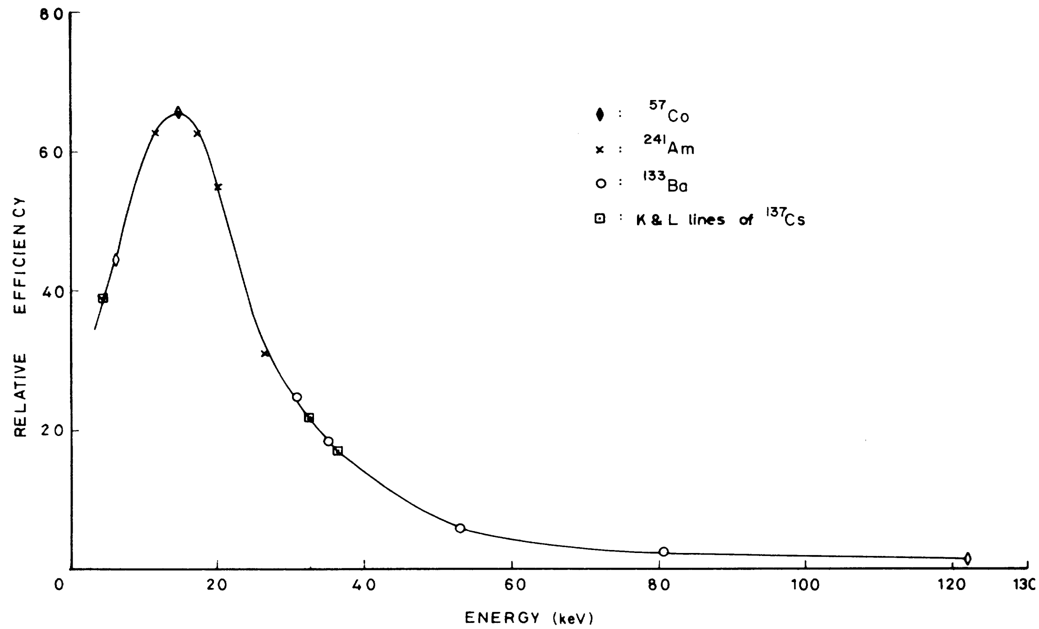

FIG. 1. Efficiency calibration of the Si(Li) detector system in units of keV.

processed, and analyzed by the computer-controlled 2000 multichannel analyzer. The system has peripherals including a video monitor, a plotter, and a printer. The keyboard facilitates different computer operations. The system was calibrated for energy and efficiency using standard lines of reference, which would fall in the present region of interest. The efficiency calibration is shown in Fig. 1. The error in efficiency calibration is essentially due to (1) error in the reported intensities of standard lines,[15,16] and (2) counting statistics. For the present efficiency calibration one could expect a maximum error of 2% depending on the energy of interest. The validity of this calibration was checked for the $I(K)/I(L)$ ratio of the 662-keV transition in the decay of ^{137}Cs ($E_{K\alpha}=32.06$ keV, $E_{K\beta}=36.37$ keV, and $E_{L\alpha}=4.47$ keV). It is found to be 5.31 ± 0.3, which is in good agreement with the previous result[17] [$I(K)/I(L)=5.25\pm0.3$], thus establishing the validity of the efficiency calibration down to 4.47 keV. The efficiency at 4.47 keV was taken from the straight line extrapolated to this energy in Fig. 1. Two typical x-ray fluorescence (XRF) spectra from bismuth were excited at 15 and 20 kV and a current of 100 μA. The same are shown in Figs. 2 and 3, respectively. Figure 3 shows the $L\gamma$ lines due to the excitation of L_{I} and L_{II} subshells, while the same completely disappear in Fig. 2, which confirms the nonexcitation of the L_{I} and L_{II} subshells at 15 kV. The data in each case were collected for sufficient amount of time so as to ensure good statistics of about 1% even in the case of weak Ll line. The $L\alpha$ and $L\beta$ intensities due to different components were computed graphically after subtracting background. The intensities thus obtained were corrected for efficiency using Fig. 1 and self-absorption in the sample. Instead of applying the usual sum rule to determine the absorption coefficient of a compound at a particular energy, an auxiliary experiment was performed in each case to determine μ. This μ was used to determine the self-absorption in each sample using the relationship,

$$\frac{I}{I_0}=\frac{1}{\mu t}(1-e^{-\mu t}) ,$$

where t is the thickness of the sample and μ is the mass absorption coefficient. This expression for self-absorption

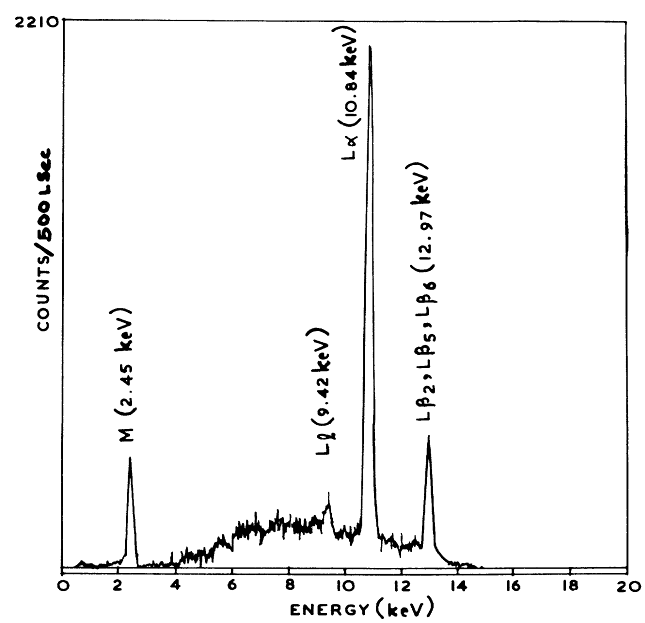

FIG. 2. L x-ray-fluorescence spectrum from Bi at an exciting voltage of 15 kV.

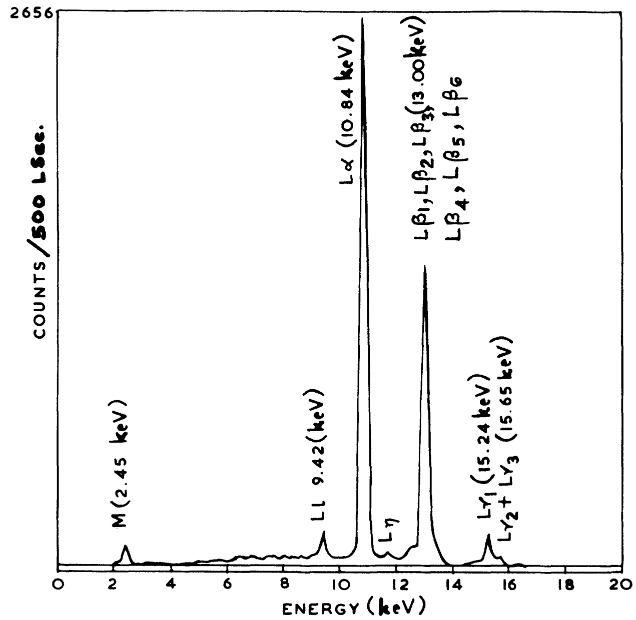

FIG. 3. L x-ray-fluorescence spectrum from Bi at an exciting voltage of 20 kV.

follows the integration over the thickness t of the sample. The absorption correction for $L\alpha$ ($E_{L\alpha}$=10.83 keV) was evaluated to be 19%, while for $L\beta$ ($E_{L\beta}$=13.01 keV) it is 9% in bismuth. The error in the absorption correction depends on counting statistics in the transmission experiment. In the case of $L\alpha$ line it is of the order of 0.7% while the same for the $L\beta$ line is 2%. The large error in the $L\beta$ line correction is due to its poor intensity. After applying the absorption correction, the resulting intensities were corrected for absorption in an air column of 5.4 cm and Be window of thickness 7.5 μm. However, the last two corrections were found to be small relative to the efficiency correction. The final relative intensities of $L\alpha$ and $L\beta_{2,15}$, $L\beta_5$ and $L\beta_6$, thus obtained, are summarized in Table I. Table I includes the theoretical values due to Scofield based on RHFS calculations and the most probable value reported by Salem *et al.* for the same lines. The values of $L\alpha_1$ and $L\alpha_2$ reported by Salem *et al.* were added to get the $L\alpha$ values given in Table I, while $L\beta_{2,15}$, $L\beta_5$, and $L\beta_6$ were added to get $L\beta$ values in Table I. The sources of errors are essentially due to (1) counting statistics in gross and background counts which contributes 1.3%, (2) instrumental instability which contributes 0.25%, (3) error due to efficiency correction which contributes 2%, (4) error due to absorption correction which is of the order of 2.1%. The compounded error ranges from 3.2 to 3.8% and the same are given in Table I for each ratio.

DISCUSSION

From an examination of Table I, it may be noted that the present values of $I(L\beta)/I(L\alpha)$ in Yb and U agree with the theory as well as with the values of Salem *et al.*, within experimental uncertainties. The same for the other elements differ from theory by 3.1 to 8.7%. Taking into consideration the experimental uncertainties, Salem *et al.* have reported that their experimental values differ from theory by about 20%.

CONCLUSION

By exciting only the L_{III} subshell, the ratio of

$$I(L\beta_{2,15}+L\beta_5+L\beta_6)/I(L\alpha_1+L\alpha_2)$$

is measured in seven elements ($Z \geq 70$) using an energy-dispersive x-ray-fluorescence (EDXRF) spectrometer. The results show good agreement with theory in Yb and U. About 3.1–8.7 % discrepancy between theory and experiment is noted for the other five elements.

ACKNOWLEDGMENTS

The authors acknowledge the financial support given by the research and development wing of the Ministry of Defence, Government of India to carry out the present studies.

TABLE I. Experimental and theoretical $I(L\beta_{2,15+5+6})/I(L\alpha_{1+2})$ ratio.

Element	Atomic number Z	$I(L\beta_{2,15+5+8})/I(L\alpha_{1+2})$ Ratio Present work	Salem *et al.*[a]	theory (Scofield)[b]
Yb	70	0.182±0.006	0.185	0.181
Hg	80	0.243±0.008	0.257	0.220
Tl	81	0.245±0.009	0.263	0.224
Pb	82	0.256±0.008	0.277	0.228
Bi	83	0.256±0.009	0.270	0.232
Th	90	0.276±0.010	0.298	0.258
U	92	0.290±0.011	0.301	0.301

[a]Reference 13.
[b]Reference 18.

[1]N. Venkateswara rao, S. Bhuloka Reddy, G. Satyanarayana, and D. L. Sastry, Physica **138C**, 215 (1986).
[2]N. Venkateswara rao, Bh. Sankara Rao, Ch. Suryanarayana, S. Bhuloka Reddy, G. Satyanarayana, and D. L. Sastry, Portugal Phys. **17**, 35 (1986).
[3]C. V. Raghavaiah, N. Venkateswara rao, S. Bhuloka Reddy, G. Satyanarayana, and D. L. Sastry, J. Phys. **20B**, 5647 (1987).
[4]A. Johnson, Z. Phys. **36**, 426 (1926).
[5]S. K. Allison, Phys. Rev. **30**, 245 (1927); **32**, 1 (1928).
[6]V. Hicks, Phys. Rev. **36**, 1273 (1930); **38**, 572 (1931).
[7]V. J. Andrew, Phys. Rev. **42**, 591 (1932).
[8]A. H. Compton and S. K. Allison, *X-Rays in Theory and Experiment* (Van Nostrand, New York, 1935), p. 642.
[9]R. W. G. Wyckoff and F. D. Davidson, Phys. Rev. **36**, 1883 (1965).
[10]C. Victor, Ann. Phys. (Paris) **6**, 183 (1961).
[11]M. Goldberg, Ann. Phys. (Paris) **7**, 329 (1962).
[12]P. V. Rao, J. M. Palms, and R. E. Wood, Phys. Rev. A **3**, 1568 (1971).
[13]S. I. Salem, R. T. Tsutsui, and B. A. Rabbani, Phys. Rev. A **4**, 1728 (1971).
[14]C. V. Raghavaiah, Ph.D. thesis, Andhra University, India, 1988.
[15]Mauro S. Dias and Cleide Renner, Nucl. Instrum. Methods **193**, 91 (1982).
[16]C. Michael Lederer and Virginia S. Shirley, *Table of Isotopes,* 7th ed. (Wiley and Sons, New York, 1978).
[17]E. Baldinger, E. Haller, Helv. Phys. Acta. **42**, 949 (1969).
[18]J. H. Scofield, At. Data Nucl. Data Tables **14**, 121 (1974).