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L_{III} SUBSHELL INTENSITY RATIOS OF Au

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ABSTRACT

In this study, L_{III} subshell intensity ratios were measured using an energy dispersive x-ray spectroscopy method. Only the L_{III} subshell of gold was excited by the characteristic x-rays of the element used as the secondary source (Br- K_{α,β} rays from NaBr). The emitted x-rays were measured with a Si(Li) detector system coupled to a multi-channel analyser and computer system. L₁/L_{α1,2}, L_{2,15}/L_{α1,2}, L_{β6}/L_{α1,2} intensity ratios of Au were determined by spectral analysis.

The experimental results are compared with those of other experimenters and with the Scofield calculations.

Key Words: EDXRF; L-subshell; Intensity ratios

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INTRODUCTION

Accurate determination of intensity ratios for medium—and high— Z elements are important because of their wide use in many areas of basic and applied science. For example, these data are used in the fields of atomic, molecular and radiation physics and nondestructive testing of materials and elemental analysis using x-ray fluorescence techniques, and they are also important to verify result obtained from the theoretically calculated methods.

In earlier investigations^{1–10}, all L_I , L_{II} and L_{III} subshells were observed together and these authors have reported only the intensity ratios of the L_I , L_{α} , L_{β} , L_{γ} lines because of difficulties arising from the overlapping of L subshell in the L_{β} region.

The method which was used to obtain the L_{III} subshell spectrum of the elements Pb, Th, U in a previous study¹¹, had been developed in this study, to obtain only Au L_{III} subshell spectra and $L_I/L_{\alpha 1,2}$, $L_{2,15}/L_{\alpha 1,2}$, $L_{\beta 6}/L_{\alpha 1,2}$ intensity ratios were determined.

EXPERIMENTAL

A gold foil with thickness $2.5\mu\text{m}$ and purity better than 99.9%, was obtained from Goodfellow company. To obtain the L_{III} x-ray spectra of Au a secondary source was used. The secondary source was irradiated by the Ag-K x-rays from a Cd-109 radioisotope source of about 370 MBq and the Au foil was irradiated by the characteristic K-x rays of this secondary source as shown in the experimental set up in Fig. 1. The secondary source was chosen such that its characteristic x-ray will excite the L_{III} subshell of the element concerned but will not excite the L_I and L_{II} subshell of Au. Br K x-rays ($K_{\alpha} = 11.921\text{ keV}$, $K_{\beta} = 13.378\text{ keV}$) was used to excite the L_{III} subshell of the Au. The binding energy of the Au L_{III} subshell is 11.919 keV.

The data were collected by a Si(Li) detector coupled to a Canberra-85 multi-channel analyser. The efficiency calibration of the Si(Li) detector was effected with a calibrated radioisotope variable source (Cu-K, Rb-K, Mo-K, Ag-K, Ba-K, Tb-K x-rays) from Amersham International.

Spectral Analysis

To give an idea of the complexity of the L_{III} subshell spectra, a gold L x-ray spectrum obtained by direct excitation of gold foil with the Ag K-x rays of Cd-109 is shown in Fig. 2.



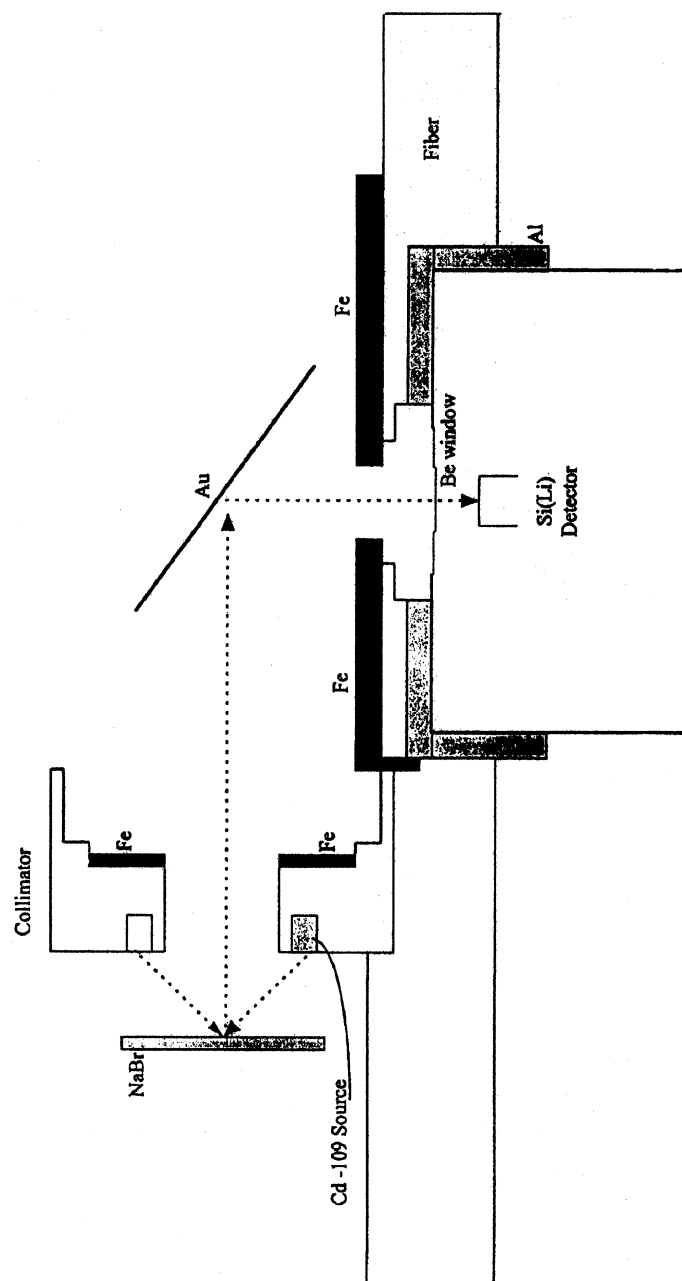


Figure 1. Experimental set up used to measure L_{III} X-rays.

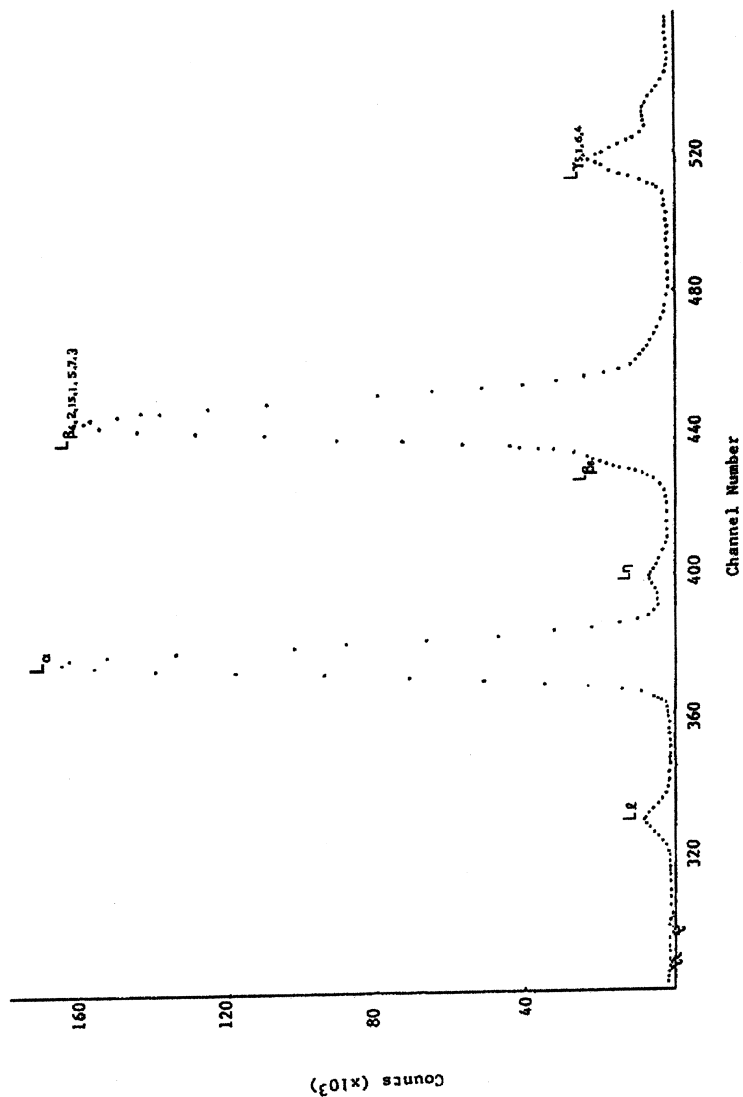


Figure 2. Gold L X-ray spectrum obtained by excitation of gold with Cd-109 source.



The Cd-109 photons, which are scattered Rayleigh (R) and Compton (C) from the secondary source are unfortunately capable of exciting the gold in the L_I and L_{II} subshell. To eliminate this effect a new scatterer source, which has the same I_R/I_C scattering ratio for the photon of the main source (Cd-109) as that of the secondary source, was prepared by mixing Co₂O₃ and cellulose samples in different ratios. All Compton and Rayleigh intensities (I_C, I_R) were calculated by the AXIL software program and I_R/I_C values against the amount of cellulose in Co₂O₃ are shown in Fig. 3. Also these values vs. amount of cellulose in Co₂O₃ were fitted to the following analytical form;

$$(I_R/I_C) = 0.632 + 0.240 \cdot \exp(-1.237 \cdot S) \quad (1)$$

with $r = 0.997$ and S is amount of cellulose in Co₂O₃.

The I_R/I_C ratio of the scatterer sample used for determination of the L_{III} subshell intensity ratio of gold was 0.687 and on the other hand the amount of the added cellulose (S) in Co₂O₃ is calculated by using this value and Eq. (1). The scatterer sample prepared was used instead of a secondary source and a spectrum of L x-rays caused by scattered Cd - 109 photons obtained (Shown in Fig. 4). This spectrum was time normalised and subtracted from of the L_{III} subshell spectrum. The stripped spectrum of the only L_{III} subshell of gold obtained in this way is shown in Fig. 5.

RESULTS AND DISCUSSION

The data collection time was chosen to ensure good statistics for each peak. The L_I, L_{α1,2}, L_{β6} and L_{β2,15} intensities were computed graphically after background subtraction and efficiency corrections were done. The efficiency correction was determined by using the absorption in the air path and the Be-window of the detector.

The error in efficiency calibrator is due to the errors in the reported activities of the standard sources and intensities of standard lines. In this work the quoted error limits on L_I/L_{α1,2}, L_{2,15}/L_{α1,2} and L_{β6}/L_{α1,2} values include an additional 8% uncertainty in the photo - peak detection efficiency.

L_{III} subshell intensity ratio values of Au are given in Table 1. The present experiment values of L_I/L_{α1,2}, L_{2,15}/L_{α1,2} agree with experimental values of Rao et al.¹² Rao et al.¹³, Jesus et al.³, Salem et al.¹ and Scofield theoretical values.¹⁴ But, the L_{β6}/L_{α1,2} value was about three time different from the values obtained by other worker and from the theoretical value.



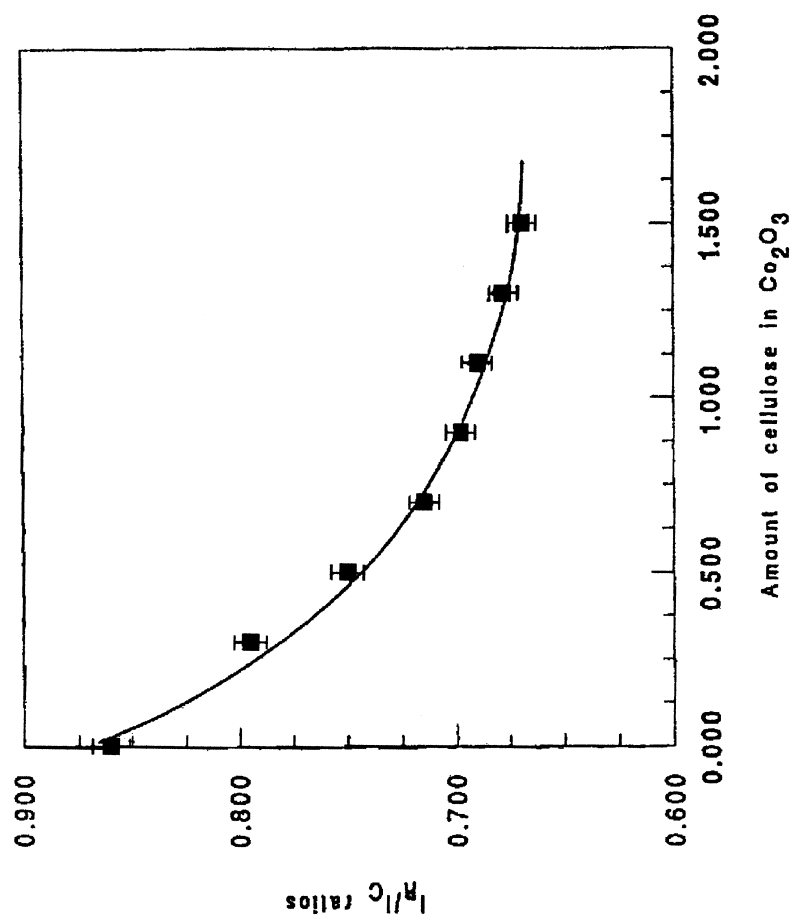


Figure 3. I_R/I_C ratios vs. amount of cellulose in Co_2O_3 .



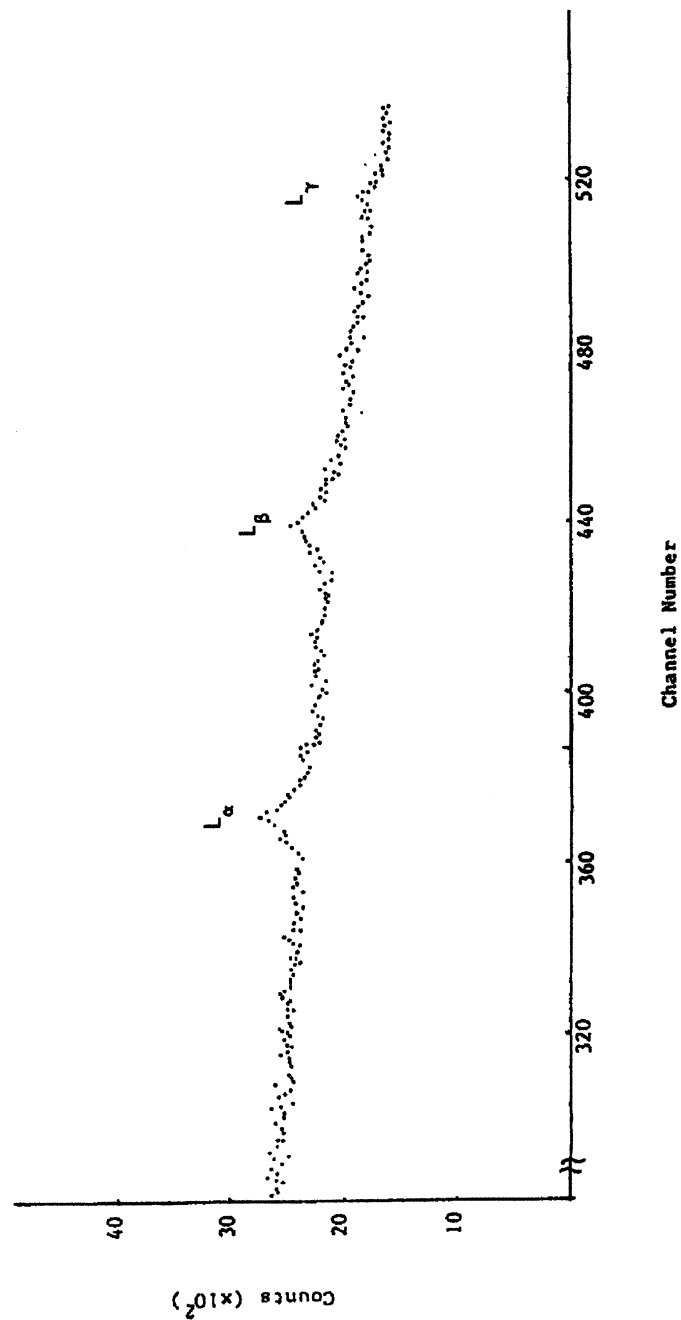


Figure 4. Gold L X-ray spectrum obtained by using scatterer sample.



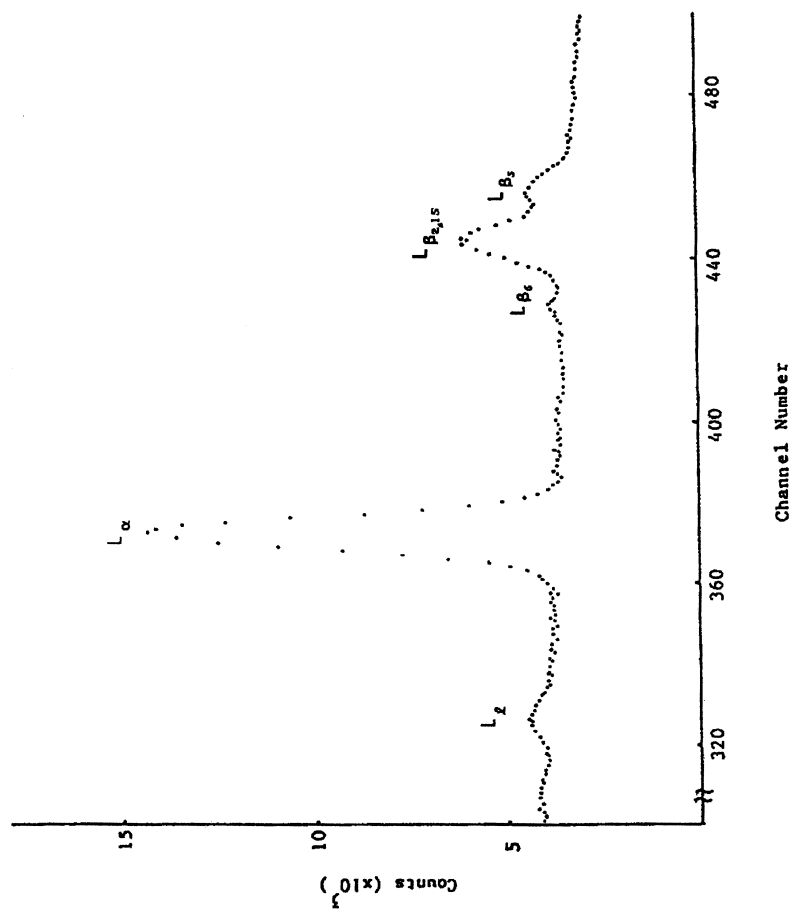


Figure 5. Gold L_{III} subshell spectrum.



Table 1. L_{III} Subshell Intensity Ratios

Intensity Ratio	Present Exp.	Ref. 12	Ref. 13	Ref. 1	Ref. 3	Theoretical Calculation
$L_I/L_{\alpha 1,2}$	0.0464 ± 0.0025	0.052	0.048 ± 0.004	—	—	0.05027
$L_{\alpha 1,2}/L_I$	—	—	—	—	21 ± 2	19.8926
$L_{2,15}/L_{\alpha 1,2}$	0.2316 ± 0.0118	—	—	0.2510	—	0.17513
$L_{\beta 6}/L_{\alpha 1,2}$	0.0045 ± 0.0003	—	—	0.012	—	0.01227

Possible sources of errors include; counting statistics, background determinations, spectrometer efficiency and sample and air absorption. Of these, the first is the most important. And the other hand, in some studies,^{15–16} in the transition of L_{III} subshell at high Z- elements it is observed that the transition of electrical and magnetic dipol and magnetic quadrupol are mixing. Because of that, especially, for the intensity ratio of $L_{\beta 6}/L_{\alpha 1,2}$ it is necessary to carry out theoretical calculations by considering the mixtures in these transitions.

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