Method of determining the value of the Coster-Kronig parameter (f_{23})

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The present work demonstrates the utility and efficacy of a technique to measure the Coster-Kronig parameter (f_{23}) of elemental targets. For this purpose a pure gold foil was irradiated in a beam of 88-keV γ rays from a radioactive ^{109}Cd source. The photoinduced K and L x rays emitted from the gold foil were then measured in coincidence, with the data collected in the list mode. On analysis, the f_{23} value of gold was found to be 0.119 ± 0.003. The technique demonstrated here is different in the sense that, in contrast to the earlier coincidence methods, where the x rays emitted directly from a radioactive source are detected in coincidence, in the present method the x rays of the elements under investigation are obtained by photoionization.

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I. INTRODUCTION

Precise determination of all the atomic parameters such as fluorescence yields (ω_i), Coster-Kronig rates (f_{ij}), and Auger yields (a_i) is very important because these are used in (1) converting x-ray production cross sections to ionization cross sections and to compare the same with the predictions of different theoretical models, (2) to compare the measured values with the Dirac-Hartree-Slater (DHS) calculations for the same, and (3) in the elemental analysis such as proton-induced x-ray emission, and x-ray fluorescence, etc.

All the measurements performed so far to determine the values of the different atomic parameters for the L subshells can be broadly classified into two categories: (1) Single spectrum method and (2) coincidence method. In the single spectrum method characteristic x -rays of the target elements are detected when target atoms are ionized by x rays, γ rays, electrons, or protons or when processes like electron capture or internal conversion occur in a radioactive atom. In order to evaluate the different atomic parameters, one has to use the theoretical values of the ionization or photoelectric cross sections of the different subshells where, a built-in uncertainty of a few percent always exists. Compounded with the other errors present in a particular measurement, the overall uncertainty of the values of the atomic parameters obtained in this method cannot be brought down below a few percent. In the coincidence experiments done to date, a long-lived radioactive isotope was used directly as the source and the L x rays coming from it were measured in coincidence with the K x rays or internal conversion electrons. With prior knowledge of the geometry and the efficiency of the detector, the coincidence x-ray yields were used to obtain the values of the different atomic parameters. It is a very powerful technique where, in favorable cases, the atomic parameters, especially the values of f_{23}, can be measured with an overall uncertainty of 1% or less [1].

The limitations of both, the single spectrum and coincidence methods are that in the first method, the uncertainty of the measured values cannot be brought down below a few percent while in the second method, a long-lived radioactive source is required which is not possible for all elements. An alternative approach to such measurements would be to excite an elemental target with an x ray or γ ray source to ionize its K shell and subsequently measure the K and L x rays of the target element in coincidence. As we have explained later, this method would be almost entirely free of the limitations and uncertainties mentioned above. So far as our knowledge goes, no such effort has ever been made earlier. Here we would like to demonstrate that with modern x-ray detectors and data acquired in the list mode, elemental targets excited with a suitable radioactive source can be used to obtain the values of f_{23} with high precision. The radioactive source is used here only to excite the K shell of the element being investigated. It should be emphasized that the method demonstrated here is quite a general one, in that, high-precision values of f_{23} of almost all elements can be obtained.

It should be noted that the values of the atomic parameters measured before 1970 had shown large errors and also large scatter among themselves [2]. Precision measurements using high-resolution solid-state detectors and list-mode data acquisition started in the mid 1970s only. It was observed that the data for different atomic parameters, especially those for the Coster-Kronig transition probabilities show large deviations (in the range of $70 \leqslant Z \leqslant 90$) from the theoretical as well as the semiempirical predictions [1]. Moreover, the values obtained by the synchrotron radiation method were always found to lie below the values determined by the coincidence technique [1]. One possible reason for this could be that in all previous coincidence experiments radioisotopes had been used as the source of x rays and the f_{23} values measured were for the daughter atom. It is possible that the nuclear cascades present in such sources would give rise to some unwanted coincidence events. To remove all such ambiguities *a more general, cleaner, and high-precision* measurement of Coster-Kronig transition probabilities has long

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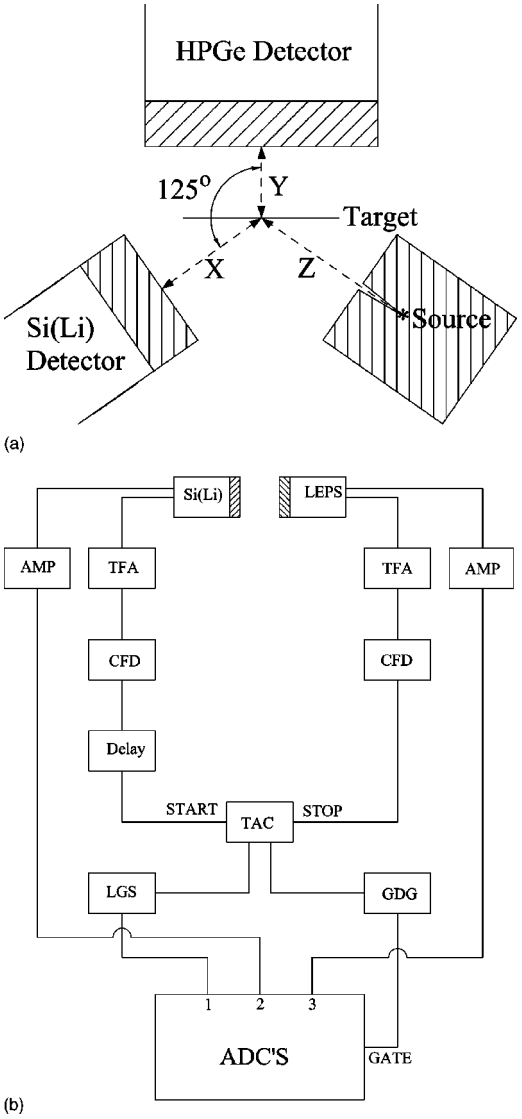

FIG. 1. (a) A schematic diagram of the experimental setup. $X=20$ mm, $Y=40$ mm, $Z=3$ mm; distances are not to the scale. (b) Electronic circuit used in the experiment.

been warranted. Our present measurement is an endeavor in that direction.

The main point where the present approach radically differs from the coincidence techniques used earlier [3–6] lies in the way the primary vacancy distribution in the target is created. Previously it was determined by the deacay scheme of the radioactive source used for the measurement. In the present method the primary vacancies are created by photoionization. This procedure is different in that by choosing suitable excitation sources we can control the primary vacancy distribution. The rest of the procedure of detection and analysis is very similar to what was done in the earlier coincidence techniques [3–6]. The expression that is used to determine the value of f_{23} in the K x-ray L x-ray coincidence is given by

$$f_{23}=\frac{C(L\alpha,K\alpha_2)n(K\alpha_1)}{C(L\alpha,K\alpha_1)n(K\alpha_2)}W(\theta), \tag{1}$$

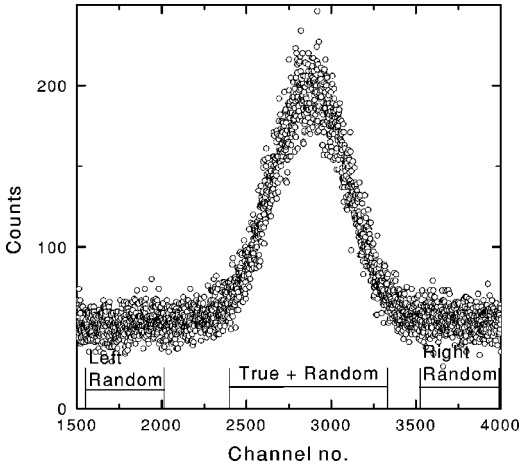

FIG. 2. Positions of the three windows set on the TAC spectrum.

where $C(L\alpha,K\alpha_2)$, $C(L\alpha,K\alpha_1)$ are the $L\alpha$ x rays measured in coincidence with $K\alpha_2$ and $K\alpha_1$, respectively, $n(K\alpha_2)$ and $n(K\alpha_1)$ are the number of $K\alpha_2$ and $K\alpha_1$ x rays measured in coincidence with the $L\alpha$ x rays, and $W(\theta)$ is the angular correlation coefficient between $K\alpha_1$ and $L\alpha$ lines. When the two detectors are placed at an angle of 125° the value of $W(\theta)$ becomes 1.

II. EXPERIMENTAL DETAILS

A 2-mCi ^{109}Cd source, shielded in a lead container provided the 88-keV γ rays which excited the target sample. A Au foil of thickness 53.7 mg/cm^2 mounted on an Al frame was used as the target. The 88-keV γ rays intereacted with the Au atom to create K shell vacancies, thus initiating the K-L x-ray cascades in the Au atom.

The K x rays ($K\alpha_1$ and $K\alpha_2$) were detected with an ORTEC LEPS high-purity Ge (HPGe) detector of energy resolution 160 eV at 5.9 keV. A DSG Si(Li) detector of resolution 140 eV at 5.9 keV was used to detect the L x rays. The relative angle between the two detectors was kept at 125° to eliminate the angular correlation effect between $K\alpha_1$ and $L\alpha$ x rays. A schematic diagram of the experimental setup is shown in Figs. 1(a) and 1(b).

The data were collected for 60 days and were stored as separate files every 10–24 h intervals. All the intermediate data sets were then added up to get the final spectrum. During the 60 days run, no peak shift was noticed. The TAC, L x-ray, and K x-ray events were registered in ADC1, ADC2, and ADC3, respectively.

III. DATA ANALYSIS

The data were analyzed event by event by using the software INGASORT developed at the Nuclear Science Center, New Delhi, India. Relevant details of the analysis are given below.

In order to find the value of $C(L\alpha,K\alpha_1)$, first of all, three windows were set on the TAC spectrum (obtained from ADC 1) as shown in Fig. 2. The first window was set on the central region (prompt) of the spectrum which included true as well

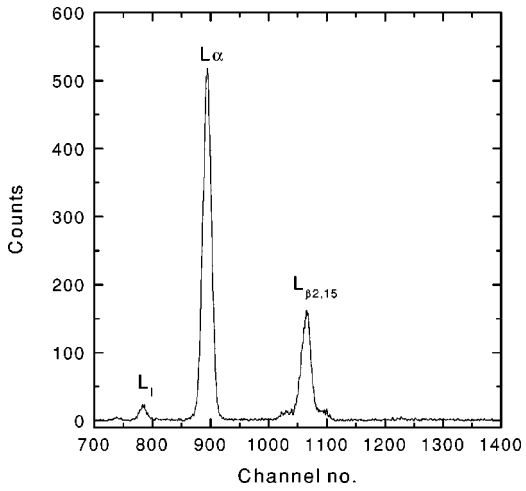

FIG. 3. True coincidence spectrum of L x rays gated by $K\alpha_1$.

as random events. With the half of the width of the first window, the second and the third windows were set on the continuum of the left and right side of the central peak as shown in the figure. These two windows essentially include the random events only. From all the L x-ray events, two L x-ray spectra, $S_{Lx}^{L}(K\alpha_1)$ and $S_{Lx}^{R}(K\alpha_1)$, were then generated by setting a common gate on $K\alpha_1$ and simultaneously by setting another gate either on the second (left) or third (right) window of the TAC spectrum. Another L x-ray spectrum $S_{Lx}^{T+R}(K\alpha_1)$ was generated in a similar manner by setting one gate on $K\alpha_1$ and the other on the prompt of the TAC spectrum. The true coincidence spectrum of L x rays with $K\alpha_1$ can now simply be obtained by subtracting $S_{Lx}^{L}(K\alpha_1)$ and $S_{Lx}^{R}(K\alpha_1)$ from $S_{Lx}^{T+R}(K\alpha_1)$. The subtracted spectrum is shown in Fig. 3. The true coincidence spectrum of L x rays with $K\alpha_2$ was obtained in a similar fashion and is shown in Fig. 4. Areas under the $L\alpha$ peak in Figs. 3 and 4 give us the values of $C(L\alpha,K\alpha_1)$ and $C(L\alpha,K\alpha_2)$, respectively. For the true coincidence events of $n(K\alpha_1)$ and $n(K\alpha_2)$, the process of gating was the same, i.e., two random spectra were

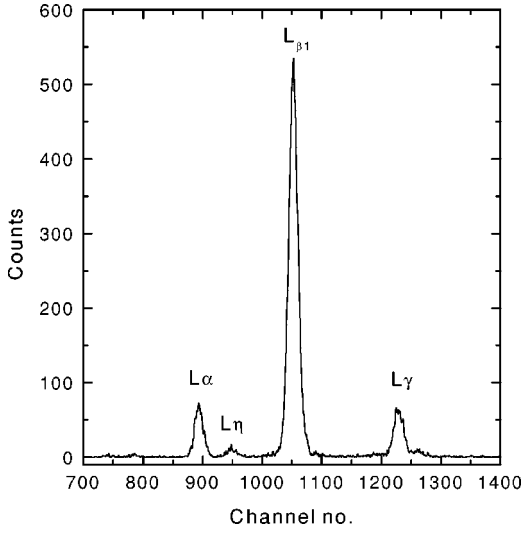

FIG. 4. True coincidence spectrum of L x rays gated by $K\alpha_2$.

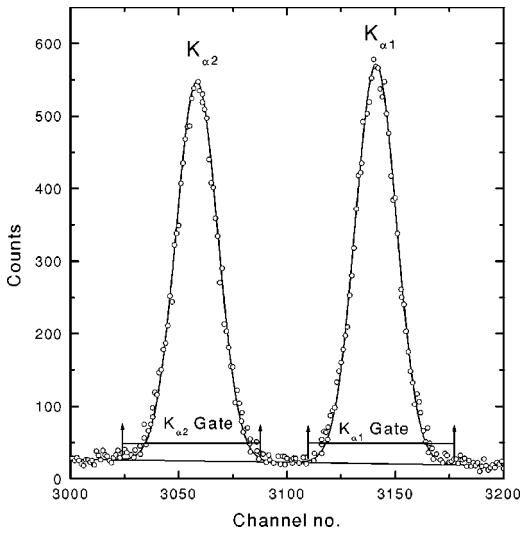

FIG. 5. Positions of the $K\alpha_1$ and $K\alpha_2$ gates.

subtracted from the true and random spectra. Having obtained all these quantities, the value of f_{23} for Au was found from Eq. (1) to be 0.119 ± 0.003.

Putting the gates on $K\alpha_1$ and $K\alpha_2$ is the most tricky part of the data evaluation especially when these two peaks are not completely resolved. In such cases, the contribution of $K\alpha_2$ in $K\alpha_1$ (and vice versa) should be carefully estimated and the prescription for this has been elaborated in several papers [4,6]. In our case, as can be seen from Fig. 5, the $K\alpha_1$ and $K\alpha_2$ peaks are well separated. So errors arising out of putting gates on them are minimal.

IV. DISCUSSION

So far as other measured values of f_{23} for Au are concerned, there are only three values available in the literature. Jitschin *et al.* [7] and Werner and Jitschin [8] used synchrotron radiation to selectively ionize different L subshells of the target and by observing the jump in the intensity of a particular L line at the L edges, determined the values of the different Coster-Kronig transition probabilities. Their measured values of f_{23} for Au are 0.100 ± 0.009 and 0.101 ± 0.010, respectively. Öz *et al.* [9] used an ^{241}Am source with different sets of secondary targets to selectively ionize different L subshells and measured all the Coster-Kronig transition probabilities of elements in the range $59 \leqslant Z \leqslant 90$ with an error of 3–12 %. They have not mentioned which sets of secondaries were used for which elements and also did not include corrections due to the scattered γ rays. Their measured value of f_{23} for Au is 0.125 ± 0.013. McGuire [10], using nonrelativistic Hatree-Slater calculations predicted a value of 0.132. Chen *et al.* [11] using nonrelativistic screened hydrogenic wave functions calculated the atomic parameters for 14 elements. An interpolation through these points gives a value of 0.100 for Au. Later, Chen *et al.* [12] made an *ab initio* relativistic calculation for 18 elements using the independent particle model with Dirac-Hartree-Slater wave functions. The interpolated value for Au from this set of data is 0.129. Puri *et al.* [13] calculated different atomic

parameters for the elements $Z=25$–95 using the radiative emission rates of Scofield [14] and nonradiative emission rates from Chen *et al.* [15]. Their calculated value of f_{23} for Au is 0.129. The most widely used value of f_{23} of Au is the semiempirical value of Krause [16] which is 0.122.

It is clear that our measured value of 0.119 ± 0.003 is lying above the values determined by the synchrotron method, but very close to the semiempirical value of Krause [16], and slightly less than the value predicted by the relativistic formalism of Chen *et al.* [12]. It has been pointed out by Jitschin [1] that atomic parameters like f_{12}, f_{13}, and f_{23} determined by the synchrotron radiation method always lie below the values obtained using the x-ray–x-ray coincidence technique. This trend has also been observed by McGhee and Campbell [17] who used coincidence technique with radioactive isotopes as sources of primary L vacancies. One of the main differences between these two techniques is that in the synchrotron radiation method, vacancies in the L subshells are produced by photoionization whereas in the coincidence technique, the same are produced either via electron capture or by internal conversion. McGhee and Campbell [17] speculated that the different values of Coster-Kronig transition probabilities obtained by these two methods might be due to the difference in the physical mechanism creating the initial L vacancies.

For Au, there are no coincidence data available using a radioactive isotope. McGhee and Campbell [17] have measured f_{23} values for neighboring elements ($Z=80$, 81, and 82) using the coincidence method with radioactive sources. If one compares these data with the theoretical predictions and the semiempirical values of Krause [16], it is found that the data give excellent agreement with Krause's values. Our present result which was based on an x-ray–x-ray coincidence method, where an elemental target of Au was excited with γ rays (photons) agrees, within experimental errors, with the value of Krause [16]. It can perhaps be conjectured that the values of the Coster-Kronig transition probability, f_{23}, determined by the x-ray–x-ray coincidence technique remain very close to one another irrespective of the mechanism of creating the primary vacancies, e.g., the vacancies created in a daughter atom by radioactive decay or vacancies created in an elemental target by direct photon excitation. So our present result provides experimental evidence against the speculation of McGhee and Campbell [17]. The reason why the values of Coster-Kronig transition probabilities determined by synchrotron radiation method always lie below the values determined by the coincidence technique is still not clear and further investigations in this line are needed.

While writing this report we came across an old work of Douglas [18] who had excited Ho and Tm targets with an ^{241}Am source and measured their f_{23} values. Partly due to the poor resolution of the detectors and partly due to the targets of medium Z elements used by him, he could not completely resolve the $K\alpha_1$ and $K\alpha_2$ components, and had to use edge filters of Sm and Gd for selective absorption of one of these components. Use of filters produced K x-ray lines of Sm and Gd which were very close to the $K\alpha_1$ and $K\alpha_2$ x rays of the targets. Consequently the mode of gating was quite imprecise. At times, the contribution of the filter x-ray lines was estimated to be more than 10%. He also had to make a correction for the unresolved $L\eta$ line that was lying buried under the intense $L\alpha$ lines. In their work, the proper choice of filters was also fortuitous, in that it would be very difficult to find such filters in a general case. Besides, Douglas prepared all gates with hardware and presumably had to continue data collection over longer time periods.

Our present method as reported here is quite a general one. Any element can be excited by a suitable radioactive x ray or γ source or in an x-ray tube and the present technique can be applied to measure its Coster-Kronig probabilities. Through such an excitation, no cascading is produced like the same occurring while using a radioactive source. Moreover, if the data are acquired over a long enough period, the accuracy of the measurement can be brought down to $\sim 1\%$.

ACKNOWLEDGMENTS

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