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Angular Dependence of Fluorescence X-Rays and Alignment of Vacancy State Induced by Radioisotopes

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1. Introduction

This chapter concerns angular distribution measurements for fluorescence X-ray and the alignments of atoms with inner-shells vacancy resulting from ionization by radioisotope sources. The discussion on this topic is done by evaluating measurements of X-ray fluorescence parameters (such as cross-section, alignment parameter, polarization degree) from sample in various emission angles.

When an atom is ionized in one of its inner shells, the electrons rearrange themselves to fill the vacancy, with the transition energy released as a photon or transferred to another electron. The following X-ray or Auger electron may have an isotropic or non-isotropic angular distribution. The study of alignment of the inner-shell vacancy in ions can provide information about ionization process and the wave functions of inner-shell electrons, and calculations showed that the alignment was a sensitive testing parameter for theoretical models. For the last five decades there have been both theoretically and experimentally renewed efforts towards better understanding of the physics concerned with alignment of atoms with inner-shells vacancy and/or angular dependence of fluorescent X-rays emitted atoms induced photons or charged particle (electrons, protons, heavy ions). Generally, the alignments of atoms with inner-shells vacancy resulting from ionization by photons are investigated by measuring the anisotropic emission of X-ray lines using a detector (such as Si(Li) or Ge(Li)) and radioisotope photon source in various emission angles.

2. Historical background and current status of topic

The aim of paper interested in this topic is to determine the relationship between the angular distributions of X-rays with respect to total angular momentum values (J) of vacancy states. It is well-known that when radioisotope source, X-ray tube or charged particles produce vacancies in atoms at energy levels with $J>1/2$, the resulting ions will be aligned. The signature of this alignment is the anisotropic angular distribution of the emitted characteristic X-ray radiation, or the degree of polarization of the X-ray radiation. Total angular momentum (J) of vacancy states after photoionization is greater than 1/2, the population of its magnetic sub-states is non-statistical by the ionized atoms and this is reason of this anisotropic behavior. A lot of theoretical studies have been reported so far along this topic (Mehlhorn 1968; Mc Farlene, 1972; Berezhko and Kabachnik 1977; Sizov and Kabachnik, 1980, 1983) and the predictions of these researchers have been experimentally supported by some researchers (Schöler and Bell, 1978; Pálinkás, 1979, 1982; Wigger et al., 1984; Jesus et al., 1989; Mitra et al., 1996). The experimental study of alignment generally involves measurements of the angular distribution or polarization of the induced X-rays (Hardy et al., 1970; Döbelin et al., 1974; Jamison and Richard, 1977; Jistchin et al., 1979, 1983; Pálinkás, et al., 1981; Stachura et al., 1984; Bhalla, 1990; Mehlhorn, 1994; Papp 1999). In 1969, Cooper and Zare, (1969) first suggested a theoretical model relevant to aligned photon induced atoms. According to calculation by Cooper and Zare, (1969), after photoionization the inner-shell vacancy states have statistical population of magnetic substates. The vacancies produced after photoionizationin sub-shells are not be aligned at all and so the angular distribution of the fluorescent X-rays subsequent to photoionization will be isotropic. In 1972, 3 years after Cooper and Zare, the predictions of Flügge et al., (1972) showed that when vacancies are created in states with J>1 2, the population of its magnetic sub-states are non-statistical and therefore the resulting ions will be aligned. Mc Farlane (1972) calculated the polarization of X-rays from the decay of a vacancy in the $2p_{2/3}$ sub-shell using hydrogenic wave- functions in the Bethe approximation and the first Born approximation. After Caldwell and Zare (1977) first made an experimental investigation of the photon-induced alignment of Cd and they measured the degree of polarization of the emitted radiation from Cd. Since then, many experiments and calculations have been done to study the alignment of atoms and angular dependence characteristic X-rays by measuring either the angular distribution or the degree of polarization of the emitted X-rays. All these studies confirmed either alignment or not-alignment of the atoms after photoionization. The angular correlation between ionizing and fluorescent X-rays has been calculated relativistically, including all the radiation multipoles using single particle wavefunctions calculated in the Hartree–Slater model, by (Scofield, 1976). More recently, Scofield, (1989) used a relativistic model to study the angular distribution of the photoelectrons produced from photo- ionization by linear polarize photons and its inverse process (radiative recombination) in the energy region of 1–100keV. Scofield, (1989) found that the cross-section has a maximum at 90^{o} compared to the direction of the incoming photons in the x–z plane (polarization plane) while the cross-section is independent of the angle between the incoming photon and the ejected electron in the y–z plane (normal to the polarization plane). Kamiya et al., (1979) measured L X-rays of Ho and Sm produced by protons and ^{3}He impacts with Si(Li) detector over the incident energy ranges E_p = 0.75–4.75MeV and $E_{3_{He}}$ = 1,5–9,4 MeV in the direction of 90° to the projectile. Kamiya, et al., (1979) reported that the ratios of X-ray production cross-sections for the La and Ll lines depend clearly on projectile energy, but are independent of the projectile charge. Theoretical values of the alignment parameter for different states of various atoms calculated using the Herman-Skillman wave functions, have been reported by Berezhko and Kabachnik, (1977). The very strong anisotropy was reported for the emission of L lines for various elements by several scientists (Kahlon, et al., 1990a,b, 1991a,b; Ertuğrul, et al., 1995, 1996; Ertuğrul, 1996, 2002; Kumar, et al., 1999 Sharma and Allawadhi, 1999; Seven and Koçak, 2001,2002; Seven, 2004; Demir, et al., 2003). However, in all these investigations, the observed anisotropy is much higher than the predicted theoretical values of Scofield, (1976) and Berezhko and Kabachnik, (1977). On the other hand, anisotropic emission for L X-rays of Pb, Th and U was reported by some scientists (Mehta, et al., 1999; Kumar, et al 1999, 2001). Recently, Yamaoka et al., (2002, 2003) performed experiments using synchrotron radiation to determine the angular distribution of

L X-ray photons of Pb and Au. Although they found an isotropic distribution of the Pb L_3 lines within the experimental errors, non-isotropic angular distribution of the Au L_3 lines have been obtained. Papp and Campbell, (1992) reported the magnitude of the anisotropy and the alignment parameter for the L lines of Er. The alignment parameter of the ions of Xe was obtained by Küst, et al., (2003).

Kahlon et al., (1990a) reported experimental investigation of the alignment of the L_3 subshell vacancy state produced after photoionization in lead by 59.57 keV photons. The values of differential cross sections for the emission of the Ll, $L\alpha$, $L\beta$ and $L\gamma$ X-ray lines were determined at different emission angles varying from 40° to 120°. It was seen from the results that the Ll, and $L\alpha$ peaks show anisotropic emission, while the $L\beta$ and $L\gamma$ peaks are emitted isotropically. The angular dependence of emission intensity of L shell X rays induced by 59.57 keV photons in Pb and U was investigated by Kahlon et al., (1990b) measuring the normalized intensities of the resolved L X-ray peaks at different angles varying from 40° to 140°. It was observed that while the Ll and $L\alpha$ peaks (originating from J=3/2 state) show some anisotropic angular distribution, the emission of the $L\beta$ and $L\gamma$ peaks are emitted isotropically. Kahlon et al., (1991a) measured the angular distribution and polarization of the L shell fluorescent X-rays excited by 59.54 keV photons in Th and U. It was found that the $L\gamma$ group of L X-rays is isotropic in spatial distribution and unpolarized but, the Ll and $L\alpha$ groups are anisotropically distributed and polarized. Although no anisotropy of the $L\beta$ group is detected, it was slightly polarized. Kahlon et al., (1991b) investigated the differential cross sections for emission of Ll, $L\alpha_2$, $L\alpha$, $L\beta$ and $L\gamma$ groups of L X-ray lines induced in Au by 59.54 keV photons at different angles varying from 40° to 120°. The L X-rays represented byLl, $L\alpha_2$ and $L\alpha$ peaks were found to be anisotropic in the spatial distribution while those in $L\beta$ and $L\gamma$ peaks were isotropic. Papp and Campbell, (1992) measured angular distributions of the L_l, $L\alpha_{1,2}$ and $L\beta_{2,15}$ transitions of erbium in the angular range of 70°–150° following photoionization by 8.904 keV photons. A Johansson-type monochromatic was used to select the Cu $K\beta_1$ line for ionization. Anisotropy parameters for Ll, $L\alpha_{1,2}$ and $L\beta_{2,15}$ were found as 0.052±0.016, 0.16±0.022 and 0.012±0.015, respectively. Ertugrul et al., (1995, 1996a, 1996b) measured differential cross-sections for the emission of Ll, $L\alpha$, $L\beta$ and $L\gamma$ X-rays of Au, Hg, Tl, Pb, Bi, Tb and U at different emission angles varying from 45° to 135°. They found that Ll and $L\alpha$ peaks are emitted isotropically, while $L\beta$ and $L\gamma$ peaks show anisotropic emission. Sharma and Allawadhi, (1999) measured values of Ll, $L\alpha$ and $L\beta$ differential X-ray production cross sections in Th and U at 16.896 and 17.781 keV at emission angles 60°, 70°, 80° and 90°. From the results of the measurements it was evident that, in the present case, all the three Ll, $L\alpha$ and $L\beta$ differential X-ray production cross sections depend on the emission angle and thus, the emission is anisotropic. Demir et al., (2000) indicated differential cross-sections for the emission of M shell fluorescence X-rays from Pt, Au and Hg by 5.96 keV photons at seven angles ranging from 50° to 110° at. The differential cross-sections were found to decrease with increase in the emission angle, showing an anisotropic spatial distribution of M shell fluorescence X-rays. Seven and Koçak (2001, 2002) measured the Ll, $L\alpha$, $L\beta$ and $L\gamma$ X-ray production cross-sections in U, Th, Bi, Pb, Tl, Hg, Au, Pt, Re,W, Ta, Hf, Lu, and Yb using 59.5 keV incident photon energies in the angular range 40°-130°. Although differential cross sections for Lβ and Lγ X-rays were found to be angle independent within experimental error, those for the Ll and La X-rays were found to be angle dependent. Ertugrul et al., (2002) measured the alignment parameter the $I_{L\alpha}/\ I_{Ll}$ intensity ratio. The Ll and $L\alpha$ X-rays of the elements were measured with a Si (Li) detector at a direction of 90° to the projectile. The L3 edges of Nd, Gd, Tb, Dy, Ho, Er, Yb, Hf, Ta, W, Au, Hg, Tl, Pb, Bi, Th and U the elements were excited with the K X-ray energy of 17.781($MoK_{\alpha,\beta}$), 16.896($NbK_{\alpha,\beta}$), 14.980(RbK_{β}), 13.300(BrK_{β}), 12.503(SeK_{β}), 12.158($BrK_{\alpha,\beta}$), 10.983(GeK_{β}), 10.073($GeK\alpha_{1},_{\beta}$), 9,572(ZnK_{β}), 8.976($CuK_{\beta2}$), 8.907(CuK_{β}), 8.265(NiK_{β}), 7.649($CoK_{\beta1}$), 6.490($MnK_{\beta1}$) keV from the selected elements, respectively. They noticed that the L_3 X-rays show large anisotropy, the measured alignment parameter varying from -0.115 to +0.355. Demir et al., (2003) reported Ll, $L\alpha$, $L\beta$ and $L\gamma$ X-ray differential cross-sections, fluorescence cross-sections and σ_{L1}, σ_{L2} and σ_{L3} subshell fluorescence cross-sections for Er, Ta, W, Au, Hg and Tl at an excitation energy of 59.6 keV. The differential cross-sections for these elements have been measured at different angles varying from 54° to 153°. The Ll and $L\alpha$ groups in the L X-ray lines were found to be spatially anisotropic, while those in the $L\beta$ and $L\gamma$ peaks are isotropic. The Ll, $L\alpha$, $L\beta_{2,4}$, $L\beta_{1,3}$ and $L\gamma$ X-ray production cross-sections and L-subshell fluorescence yields ω_1 and ω_2 in Th and U have been determined by Seven (2004) at an incident photon energy of 59.54 keV by measuring differential cross-sections with angles changing from 40° to 130°. The Ll, $L\alpha$ and $L\beta_{2,4}$ X-rays have an anisotropic spatial distribution while $L\beta_{1,3}$ and $L\gamma$ X-rays have isotropic spatial distributions. Özdemir et al., (2005) measured the angular dependence of L_3 subshell to M-shell vacancy transfer probabilities for the elements Lu, Hf, Ta, W, Os and Pt at the excitation energies of 5.96 keV and K X-rays of Zn, Ga, Ge, and As, respectively, at seven angles varying from 120° to 150°. It was observed that angular dependence from L_3 subshell to M-shell vacancy transfer probabilities increase with increasing cosθ. The angular dependence of M X-ray production differential cross-sections for selected heavy elements between Lu and Pt have been measured by Durak (2006) at 5.59 keV of incident photon energy and at seven emission angles in the range of 120°-150°. Angular dependence of M X-ray production differential cross sections has been derived, using the M-shell fluorescence yields, experimental total M X-ray production cross sections and theoretical M-shell photoionization cross sections. M X-ray production differential cross-sections were found to decrease with increase in the emission angle, showing an anisotropic spatial distribution of M X-rays. Angular dependence from L_3 subshell to M-shell vacancy transfer probabilities for selected heavy elements from Au to U were measured by Özdemir and Durak (2008) at different angles varying from 120° to 150°. It was observed that angular dependence from L_3-subshell to M-shell vacancy transfer probabilities increase with increasing cosθ. Apaydın et al., (2008) measured Mi (i = a + β) X-ray production differential cross sections for Re, Bi and U elements at the 5.96 keV incident photon energy in an angular range 135°-155°. They found that the angular dependence M X-rays production cross sections decrease with increase in the emission angle, showing anisotropic spatial distribution

Kumar et al., (1999) investigated the angular dependence of emission of L x-rays following photoionization at 22.6 and 59.5 keV in 82Pb by measuring the intensity ratios $I_{Ll}/I_{L\gamma}$, $I_{L\alpha}/I_{L\gamma}$ and $I_{L\beta}/I_{L\gamma}$ at different angles varying from 50° to 140°. The measured intensity ratios for various L x-rays were found to be angle independent within experimental error. Mehta et al., (1999) measured the L_l, L_α, L_η, $L\beta_6$, $L\beta_{2,4}$, $L\beta_{1,3}$, $L\beta_{9,10}$ and L_γ x-ray production differential cross sections in 92U using the 22.6- and 59.5-keV incident photon energies in an angular range 43°–140°. Differential cross sections for various L x rays were found to be angle independent within experimental error. Puri et al., (1999) measured The Ll, $L\alpha$, $L\beta_{2,4}$, $L\beta_{1,3}$ and $L\gamma_{1,5}$ X-ray production differential cross sections in 90Th have at 22.6 keV incident photon energy in an angular range 50° -130° The measured differential cross sections for various L X-rays were found to be angle-independent within experimental error. Kumar et al., (2001a) measured the the Ll, $L\alpha$ and $L\beta_{2,5,6,715}$ X-ray fluorescence (XRF)

differential cross-sections in Pb at the 13.6 keV incident photon energy ($E_{L3} < E_{inc} < E_{L2}$, E_{Li} being the Li sub-shell binding energy) and in the angular range 90-160°. At this incident photon energy, the L_3 sub-shell vacancies ($J = 3/2$) are produced only due to the direct ionization and the reduction in the observed anisotropy in the emission of the Ll, $L\alpha$ and $L\beta_{2,5,6,715}$ X-rays due to the transfer of unaligned L_1 and L_2 subshell vacancies ($J = 1/2$) to the L_3 sub-shell through Coster–Kronig transitions was eliminated. The differential cross-sections for various x-rays were found to be angle-independent within experimental error. The L X-ray production (XRP) differential cross sections in Th and U have been measured by Kumar et al., (2001b) at the 17.8 keV incident photon energy ($E_{L3} < E_{inc} < E_{L2}$, E_{Li} is the Li subshell ionization threshold) in an angular range 90°-160°and at the 25.8 and 46.9 keV incident photon energies ($E_{L1} < E_{inc} < E_K$) at an angle of 130°. The present measurements rule out the possibility of a strong angular dependence of differential cross sections for various L_3 subshell X-rays following selective photoionization of the L_3 subshell. Tartari et al., (2003) investigated the anisotropy of L X-ray fluorescence induced by 59.54 keV unpolarized photons by means of an experimental procedure which allows the relative L X-ray production cross section to be evaluated without taking account of the angular set-up and the instrumental efficiency. Thick targets of Yb, Hf, Ta, W and Pb are considered, and the angular trend of the relative experimental ratios, $I_{L\alpha}/I_{L\beta}$, is calculated by simple evaluations of the peak area alone. Within the experimental uncertainties, which were found to be of the order of 1.6% in the worst cases, the results do not show any significant angular dependence of the L*a* emission lines. Santra et al., (2007) measured the angular distribution of the *L* X-ray fluorescent lines from Au and U induced by 22.6-keV X-rays in the angular range of 70°–150°. No strong anisotropy was observed as mentioned by some groups. In the case of Au, a maximum anisotropy of 5% was observed while for U it was within experimental errors 2%. From the angular distribution of the L_l line of Au, the alignment parameter was obtained and its value was found to be 0.10±0.14. Kumar et al., (2008) investigated alignment of the M_3(J= 3/2), M_4(J= 3/2) and M_5(J= 5/2) subshell vacancy states produced following photoionization in the M_i (i=1-5) subshells of Au, Bi, Th and U through angular distribution of the subsequently emitted *M* X-rays. The unpolarized Mn *K* X rays (E_{KX}=5.97 keV) from the 55Fe radioisotope were used to ionize the *Mi* subshells in an angular range 90°-160°and the emitted *M* X-rays were measured under vacuum using a low energy Ge detector. The *M* X-ray spectra taken at different emission angles were normalized using the isotropically emitted *K* shell (J= 1/2) X-rays measured simultaneously from a 23V thin target placed adjoining the *M* X-ray target. The present precision measurements infer that anisotropy in the $M_{\alpha\beta\gamma}$ X-ray emission shows trends and order of magnitude predicted by theoretical calculations, i.e., anisotropy parameter (β_2)~0.01.
In the recent experimental study (Han et al., 2008), the angular distribution of characteristic K and L X-rays, emitted from Sm, Eu, Gd Tb, Dy, Ho, and Er as a result of K and L shell vacancies produced by 59.54 keV photon impact was investigated. Thus, K and L X-rays emitted from these elements were simultaneously measured in the same experimental geometry. In this study, Sm, Eu, Gd, Tb, Dy, Ho, and Er lanthanides were chosen since both K shell and L shell electrons of these elements can be excited simultaneously by an Am-241 point source. Also, K and L peaks of the chosen elements are well resolved. Earlier experimental investigations have been only performed on the K X-ray cross sections or on the angular distribution of L X-rays. This is the first report of the angular distributions of L*i* X-ray and K*i* X-ray ($i = a, \beta$) cross sections for Sm, Eu, Gd, Tb, Dy, Ho, and Er at different angles. It is well known that K X-ray cross sections have no angular dependency (J = 1/2). The experimental investigation on K X-ray cross sections at different angles was made to

check the validity of the angular dependency of experimental L X-ray cross sections. The experimental K X-ray cross sections were compared with theoretically calculated values and fairly good correspondence was observed. This means that the present measurements regarding angular dependency of L X-rays are reliable.

In following the work of us (Han et al., 2009) experimental results of the angular distribution of characteristic X-rays were introduced. We preferred to use of $I_{La} / I_{Ll}(\theta)$ intensity ratios to obtain the values of alignment parameters (A_2). In that case, the background subtraction problem is considerably reduced and statistical errors are significantly less. It was observed from measured intensities that L*a* and L*l* X-ray intensities for the L_3 sub-state depended on the emission angle, meaning that L*a* and L*l* X-rays had an anisotropic spatial distribution. Thus, the L*a* to L*l* intensity ratios for a set of elements was determined and alignment parameters for each element were obtained using these ratios. In this study, three L sub-shells electrons were excited. Therefore, alignment parameter values are influenced by Coster–Kronig transitions from vacancies induced in the L_1 or L_2 sub-shells. L_1 and L_2 sub-shells have the same J= 1/2 value therefore the transferred vacancies are not-aligned and the observed anisotropy of the X-rays is attenuated. For this reason, corrected value of the alignment parameter was calculated using attenuation factor F. If photon energies exciting only L_3 sub-shell electrons are chosen, the alignment parameter will be independent from Coster–Kronig transitions

In more recently study (Han and Demir, 2011a), we investigated the angular distribution of characteristic L X-rays emitted from heavy elements (Pt, Au, Pb, Bi, Th and U) as a result of L shell vacancy production by 59.54 keV photon impact and angular distribution of Compton scattering photons from the same elements. Thus, emitted fluorescent L X-rays and Compton scattering photons from elements were simultaneously measured in the same experimental geometry. Earlier experimental investigations have been only performed on the angular distribution of L X-rays or Compton scattering photons. This is the first report of the angular distribution of L*i* (*i*= *l*, *a*, β and γ) X-rays fluorescent and Compton scattering differential cross sections for Pt, Au, Pb, Bi, Th and U at different angles in the same experimental geometry. It is well known that Compton scattering differential cross sections have angular distribution. The experimental investigation on Compton scattering differential cross sections at different angles was made to check the validity of angular distribution of experimental L X-rays fluorescent differential cross sections. The experimental Compton scattering differential cross sections were compared with theoretically calculated values and fairly good correspondence was observed. This means that the present measurements regarding angular distribution of L X-rays are reliable. In the meantime, L_3-subshell alignment of Th and U ionized by 59.5 keV photons has been investigated by evaluating the angular dependence of L*i* (*i*=*l*, *a*, η, β and γ) X-ray lines. The angular dependence measurements were performed by measuring the fluorescence cross section, σ_{Li} (*i*= *l*, *a*, η, β and γ) and $\sigma_{Ll}/\sigma_{L\gamma}$, $\sigma_{L\eta}/\sigma_{L\gamma}$, $\sigma_{La}/\sigma_{L\gamma}$ and $\sigma_{L\beta}/\sigma_{L\gamma}$ ratios at different angles. It was observed from the measurements that L*i* (i=*l* and *a*) X rays for the L_3-subshell depended on the emission angle and had an anisotropic spatial distribution. On the other hand, there was no dependence of emission angle and any significant anisotropy for other L X rays. The both L*l* and L*a* X-rays originate from the filling of vacancies in states L_3-subshell with J = 3/2. The results of measurements indicate that the L_3-subshell vacancy states with J =3/2 are aligned, whereas L_1, and L_2 vacancy states with J =1/2 are non-aligned. Integral cross-sections for the L*i* (*i*= *l*, *a*, η, β and γ) X-rays and L subshell fluorescence yields ω_i (*i*= 1, 2 and 3) were also determined and results were compared with theoretically calculated values and results of others and fairly good correspondence was observed. The Lγ X-rays, originating purely from the L_1 and L_2 subshells, having isotropic emission were used to normalize the intensities of the anisotropic L*l* and the L*a* X-rays originating from the L_3 subshell. It was observed from measurements that L*l* and L*a* X-ray for the L_3 sub-state depended on the emission angle, meaning that L*l* and L *a* X-rays had an anisotropic spatial distribution. On the other hand, the Lβ and Lγ X-rays don't show any significant anisotropy. The fluorescence cross sections for L*l* and L*a* X-rays are decreased with increased emission angles (Han and Demir, 2011b).

3. Conclusion

In the light of all these, above; data from different researchers show contradictory and the existing results on the angular dependence of fluorescence X-ray and the alignment of atoms with inner-shells vacancy following ionization are still controversial and quite confusing. Therefore, more experimental and theoretical investigations should be required to settle the present discrepancies

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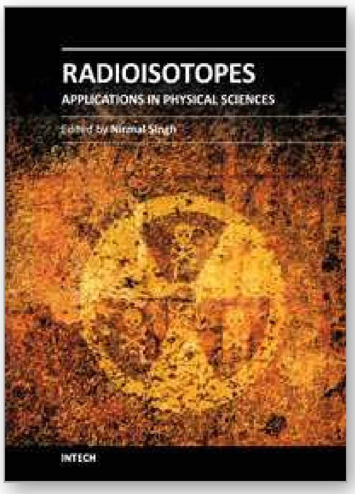

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The book Radioisotopes - Applications in Physical Sciences is divided into three sections namely: Radioisotopes and Some Physical Aspects, Radioisotopes in Environment and Radioisotopes in Power System Space Applications. Section I contains nine chapters on radioisotopes and production and their various applications in some physical and chemical processes. In Section II, ten chapters on the applications of radioisotopes in environment have been added. The interesting articles related to soil, water, environmental dosimetry/tracer and composition analyzer etc. are worth reading. Section III has three chapters on the use of radioisotopes in power systems which generate electrical power by converting heat released from the nuclear decay of radioactive isotopes. The system has to be flown in space for space exploration and radioisotopes can be a good alternative for heat-to-electrical energy conversion. The reader will very much benefit from the chapters presented in this section.