Incoherent-scattering cross sections in some lanthanum-group rare-earth elements and Ta, Pb, and Bi derived from the measured total attenuation cross sections in compounds

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The whole-atom integrated incoherent-scattering cross sections have been obtained from the total attenuation cross sections in the elements La, Ce, Pr, Nd, Sm, Gd, Dy, Ho, Er, Ta, Pb, and Bi at photon energies of 279.2, 514, 661.6, and 1115.5 keV. The total attenuation cross sections for the elements were derived with the aid of the mixture rule from the total attenuation cross sections of the compounds measured by performing transmission experiments in a good-geometry setup. The effect of multiple scattering of photons on the measured values has been corrected by using an extrapolation technique. The present values of the incoherent-scattering cross sections are found to agree fairly well with the recent theoretical values of Hubbell *et al.*

INTRODUCTION

A survey of literature on the incoherent-scattering process reveals that though the theoretical calculations have been updated from time to time, the experimental investigations are relatively few.[1] In particular, the experimental data on the whole-atom integral incoherent-scattering cross sections are found to be scarce. The data are found to be scanty, particularly in the case of low-Z, rare-earth elements and also a few high-Z elements like Ta and Bi. Such gaps in the currently available data necessitate a rapid filling up with some reasonably accurate experimental work, particularly in view of the fact that the recent theoretical values of Hubbell *et al.*[2] are to be tested adequately against experimental data. With this end in view and also in view of the various difficulties encountered in obtaining the pure foils of such elements it was felt worthwhile to perform transmission experiments with simple compounds (like oxides) of these elements. The results for some low- and medium-Z elements, at incident photon energies 279.2, 514, 661.6, and 1115.5 keV were reported in our previous paper.[3] Here, we report the experimentally estimated whole-atom integral incoherent-scattering cross sections of some of the lanthanum-group rare-earth elements and a few high-Z elements like Ta, Pb, and Bi for the same energies. These cross sections were extracted from the total attenuation cross sections of the elements which were derived with the aid of mixture rule[4] from the measured total attenuation cross sections of their compounds. The results are compared with the other known experimental data, the Klein-Nishina[5] free-electron values, and the recent self-consistent-field Hartree-Fock theoretical bound-electron values of Hubbell *et al.*[2]

EXPERIMENTAL DETAILS

The geometrical setup, the gamma-ray detector, and the sources used in the transmission experiment are similar to the ones used in our previous measurements.[3,6] Each compound under investigation was confined in a cylindrical plastic container. The sample thus prepared was weighed in an electrical balance exactly to the third decimal place. The weighings were repeated a number of times to get concordant values of the mass. A mean of this set of concordant values was taken to be the mass of the sample. The inner diameter of each container was determined separately with the help of a traveling microscope by the usual method. Using the mean values of the mass as well as the radius, the mass per unit area of the sample was determined. The uncertainty in the value of the mass per unit area was less than 0.05% for all the samples. A list of compounds used along with their

TABLE I. Purities and molecular weights of the compounds.

Sl no.	Compound	Molecular weight	Percentage purity	Manufacturers
1	La_2O_3	325.82	99.0	Reechim PVT. LTD., USSR
2	CeO_2	172.12	99.9	Indian Rare earths Ltd.
3	PrO_2	172.91	99.5	Koch-Light Lab, England
4	Nd_2O_3	336.48	99.9	Leico Industries, U.S.A.
5	Sm_2O_3	348.70	99.9	British Drug houses
6	Gd_2O_3	362.50	99.9	Koch-Light Lab, England
7	Dy_2O_3	373.00	99.9	Leico Industries, U.S.A.
8	Ho_2O_3	377.86	99.5	John Baker Ltd., U.S.A.
9	Er_2O_3	382.56	99.9	Koch-Light Lab, England
10	Ta_2O_5	441.89	99.9	Fluka AG, Beochs SG, Switzerland
11	$(HCOO)_2Pb$	297.25	99.5	British Drug houses
12	Bi_2O_3	495.96	99.0	Reechim PVT. LTD., USSR

purities, molecular weights, and the names of the manufacturers is given in Table I. In the transmission experiment, spectra were recorded by placing the empty container and the sample alternately in the path of the beam. This procedure was repeated at least ten times for each sample at each energy. The recording time was selected such that at least 10^5-10^6 counts were recorded at the photopeak. Thus, the error due to counting statistics was kept below 0.3%. Nonuniformity in the sample material, if any, was checked by exposing different parts of the sample material to the incident beam. It was found that any discrepancy in the intensity was within counting statistics.

From the spectra thus obtained, the total attenuation cross sections of the compounds were obtained using the extrapolation technique of Puttaswamy *et al.*[7] In this technique, two channels symmetrically located on either side of the centroid of the photopeak were selected and their counts added. This procedure was repeated until the entire photopeak was encompassed. In each case, the total attenuation cross sections were calculated using the relation

$$\sigma_{\text{tot}} \mid_i = \frac{A}{0.6025}\left[\ln\frac{(I_0/I)}{t}\right], \tag{1}$$

where $\sigma_{\text{tot}} \mid_i$ is measured in barns per molecule, A is the molecular weight of the compound, t is the sample thickness in g cm^{-2}, I_0 and I are the unattenuated (with empty container) and attenuated (with sample) intensities, respectively. Then a plot of $\sigma_{\text{tot}} \mid_i$ versus the channel number "i" was extrapolated as $i \to 0$. This procedure was repeated for all the samples at the four energies of present interest. With this method, one can correct for the possible effect due to multiple scattering. The multiple scattering of photons results in several photons degraded in energy. These photons contribute to the lower energy side of the photopeak. This results in an overestimation of the intensity I and hence an underestimation of the $\sigma_{\text{tot}} \mid_i$ according to (1). By using the extrapolation technique described above, this effect is eliminated. Thus, we obtain a set of total attenuation cross sections $\sigma_{\text{tot}} \mid_{i=0}$ for each sample at each energy. From this set a mean value of the total attenuation cross sections is obtained for each compound at each energy. These values have been listed in Table II along with the errors.

ERRORS

The errors tabulated in Table II are mainly a result of the counting statistics. The other possible sources of error in this method are (1) small-angle scattering contributions to the measured values, (2) nonuniformity of the sample material, (3) multiple scattered photons reaching the detector, (4) sample impurity, (5) detector resolution, (6) photon-dose buildup effects, (7) dead time of the counting instrument, and (8) pulse-pile-up effects. In the geometrical setup used in the transmission experiment at a distance of 21 cm between the source and the sample, the solid angle subtended was less than a few millisteradians. In this sample position, the maximum angle of scattering from sample to detector was 49 min. Theoretical estimates[8] predict that the contribution of coherent scattering as well as incoherent scattering at such small an-

TABLE II. Total attenuation cross sections in compounds (barns per molecule). Figures in parentheses are the experimental errors.

Sl no.	Compound	279.2 keV	514 keV	661.6 keV	1115.5 keV
1	La_2O_3	110.24(0.47)	52.07(0.33)	42.36(0.32)	30.68(0.26)
2	CeO_2	59.48(0.26)	28.08(0.20)	22.98(0.18)	16.44(0.15)
3	PrO_2	61.85(0.27)	29.02(0.19)	23.42(0.19)	16.66(0.14)
4	Nd_2O_3	127.72(0.58)	57.41(0.35)	45.82(0.32)	32.40(0.26)
5	Sm_2O_3	139.52(0.60)	60.61(0.39)	48.28(0.36)	33.70(0.28)
6	Gd_2O_3	153.92(0.68)	64.49(0.42)	50.48(0.39)	34.92(0.30)
7	Dy_2O_3	167.74(0.68)	68.73(0.43)	53.12(0.42)	36.28(0.32)
8	Ho_2O_3	175.06(0.75)	70.73(0.48)	54.56(0.41)	37.06(0.32)
9	Er_2O_3	183.44(0.80)	72.51(0.47)	55.70(0.42)	37.88(0.33)
10	Ta_2O_5	233.08(0.99)	89.53(0.56)	68.28(0.54)	45.00(0.36)
11	$(HCOO)_2Pb$	179.72(0.75)	65.78(0.40)	49.27(0.37)	31.92(0.27)
12	Bi_2O_3	344.56(1.48)	116.49(0.73)	83.54(0.61)	51.34(0.46)

small angles for photons of energy 1 MeV is less than 0.66% and 0.03%, respectively, of the measured total attenuation cross sections of lead. At lower energies this percentage contribution should still decrease. This indicates that no small-angle scattering correction is required for the measured values. The effects due to the nonuniformity of the sample material and the multiple scattering have been discussed in the earlier part of this paper.

The error due to the sample impurity could be a significant factor only when large percentages of high-Z impurities are present in the sample. However, in all the samples used in the present study the content of high-Z impurities was less than 0.005%. Thus, the error due to sample impurity is expected to be negligible and hence no sample impurity corrections are required for the measurements.

Since monoenergetic photons were used it is obvious that the detector resolution (8% for ^{137}Cs) is sufficient for the purpose of the experiment. However, it may be argued that photon-dose buildup effects as well as complex events such as the liberation of K-, L-, or M-shell electrons followed by bremsstrahlung could interfere.

The photon-dose buildup is a function of target thickness and atomic number, and of primary photon energy which combines to determine the intensity and spectrum of secondary photons seen by the detector in addition to the primary photons. Since our experiment is a good-geometry experiment and the effects of multiple scattering have been corrected, the dose buildup is considered to be negligible.

As far as the complex events leading to bremsstrahlung production are concerned, the secondary photons released in such events are of extremely low energy and intensity compared to the primary beam. The resolution of the detector is sufficient to resolve the energy difference between the primary and the secondary photons, and hence these secondary photons are recorded as background and thus they do not contribute to the measured intensity I. There is an inbuilt provision for dead-time correction in the multichannel analyzer. By keeping an optimal count rate, the pulse-pile-up effects were kept to a minimum.

Thus, the overall errors on the measured values of the total attenuation cross sections of the compounds are less than 2 to 3%.

RESULTS AND DISCUSSIONS

Using the total attenuation cross sections of the compounds listed in Table II, the total attenuation cross sections were derived with the aid of mixture rule, by subtracting the derived cross section of oxygen[3] from the cross sections of Table II, for all elements except lead. In the case of lead, the subtracted cross sections included those of hydrogen, carbon, and oxygen[3] from the cross sections for lead formate. Using 99.9% pure foil, the total attenuation cross sections for lead were determined in the same setup. A good agreement between the derived and the foil values at all energies increased confidence in our measurements. The derived total attenuation cross sections of the elements at the four energies of interest have been listed in Table

III along with the other experimental values,[9–11] and also the cross section interpolated from the Veigele's data[12] for 279.2, 514, and 661.6 keV and the data of Hubbell *et al.*[13] for 1115.5 keV. A good agreement can be noticed among these values within the stated errors. The error on the derived

TABLE III. Total attenuation cross sections in elements (barns per atom). Figures in parentheses are the errors.

Element	279.2 keV	514 keV	661.6 keV	1115.5 keV
La	50.86(0.51)[a]	22.63(0.37)[a]	18.06(0.36)[a]	12.91(0.29)[a]
	49.92(0.18)[b]	22.39[c]	18.08(0.30)[d]	12.50(0.07)[d]
	50.70[c]		17.97(0.05)[b]	12.45(0.06)[b]
			17.78[c]	12.69[h]
Ce	53.80(0.31)[a]	23.54(0.24)[a]	18.82(0.22)[a]	13.20(0.18)[a]
	53.70[c]	23.17[c]	18.56(0.30)[d]	12.79(0.07)[d]
			18.62[c]	13.00[g]
Pr	56.17(0.32)[a]	24.48(0.24)[a]	19.26(0.23)[a]	13.42(0.17)[a]
	56.23[c]	24.27[c]	19.05[c]	13.29[h]
Nd	59.60(0.62)[a]	25.30(0.39)[a]	19.79(0.36)[a]	13.77(0.29)[a]
	59.57[c]	25.12[c]	19.73(0.31)[d]	13.43(0.07)[d]
			19.50[c]	13.62[g]
Sm	65.50(0.63)[a]	26.90(0.43)[a]	21.02(0.39)[a]	14.42(0.31)[a]
	65.31[c]	26.61[c]	20.91(0.32)[d]	14.02(0.07)[d]
			20.89[c]	14.24[g]
Gd	72.70(0.71)[a]	28.84(0.45)[a]	22.12(0.42)[a]	15.03(0.32)[a]
	70.23(0.23)[b]	28.51[c]	22.18(0.34)[d]	14.69(0.08)[d]
	72.44[c]		21.71(0.09)[b]	14.67(0.07)[b]
			21.88[c]	14.93[g]
Dy	79.61(0.71)[a]	30.96(0.46)[a]	23.44(0.45)[a]	15.71(0.34)[a]
	79.43[c]	30.55[c]	23.17[c]	15.57[g]
Ho	83.27(0.78)[a]	31.96(0.51)[a]	24.16(0.44)[a]	16.10(0.34)[a]
	83.18[c]	31.62[c]	23.71[c]	15.92[g]
Er	87.46(0.83)[a]	32.85(0.50)[a]	24.73(0.45)[a]	16.51(0.35)[a]
	87.10[c]	32.36[c]	24.27[c]	16.34[g]
Ta	109.44(1.03)[a]	39.09(0.60)[a]	28.94(0.58)[a]	18.45(0.39)[a]
	108.30[c]	38.46[c]	28.51[c]	18.27[g]
Pb	163.34(0.82)[a]	52.66(0.49)[a]	37.37(0.45)[a]	22.62(0.33)[a]
	158.91(0.83)[b]	51.61(0.44)[e]	36.48(0.31)[e]	22.04(0.19)[e]
	159.24(0.62)[f]	51.29[c]	36.77(0.12)[f]	21.95(0.10)[f]
	162.86(0.70)[e]		36.86(0.03)[d]	22.01(0.03)[d]
	162.18[c]		36.93(0.17)[b]	21.75(0.11)[b]
			36.31[c]	22.39[g]
Bi	168.02(1.49)[a]	54.84(0.75)[a]	38.65(0.63)[a]	23.24(0.48)[a]
	167.88[c]	53.70[c]	37.58[c]	22.99[g]

[a]Present values.
[b]Conner *et al.* (Ref. 11).
[c]Veigele values (Ref. 12).
[d]Sinha (Gowsami) and Chaudhuri (Ref. 10).
[e]Values from measurements made with foils.
[f]Gopal (Ref. 9).
[g]Hubbell *et al.* (Ref. 13).

TABLE IV. Whole-atom integral incoherent-scattering cross sections in elements (barns per atom). Figures in parentheses are the errors.

Element	279.2 keV Experimental	279.2 keV KN (Ref. 5)	279.2 keV Hubbell (Ref. 2)	514 keV Experimental	514 keV KN (Ref. 5)	514 keV Hubbell (Ref. 2)
La	20.19(0.54)[a] 19.99(0.51)[b]	20.69	20.14	15.62(0.41)[a] 15.85(0.37)[b]	16.30	16.08
Ce	20.54(0.36)[a] 20.39(0.31)[b]	21.05	20.49	15.89(0.29)[a] 15.99(0.24)[b]	16.58	16.36
Pr	20.90(0.37)[a] 20.91(0.32)[b]	21.41	20.84	16.17(0.30)[a] 16.45(0.24)[b]	16.87	16.65
Nd	21.25(0.65)[a] 21.17(0.62)[b]	21.77	21.19	16.44(0.43)[a] 16.74(0.39)[b]	17.15	16.92
Sm	21.96(0.66)[a] 21.70(0.63)[b]	22.50	21.89	16.99(0.47)[a] 17.20(0.43)[b]	17.73	17.48
Gd	22.67(0.74)[a] 22.24(0.71)[b]	23.23	22.50	17.53(0.49)[a] 17.71(0.45)[b]	18.30	18.04
Dy	23.38(0.74)[a] 23.12(0.71)[b]	23.95	23.29	18.08(0.50)[a] 18.20(0.46)[b]	18.87	18.60
Ho	23.73(0.81)[a] 23.55(0.78)[b]	24.31	23.64	18.35(0.55)[a] 18.55(0.51)[b]	19.16	18.88
Er	24.08(0.86)[a] 23.63(0.83)[b]	24.68	23.99	18.62(0.54)[a] 18.67(0.50)[b]	19.44	19.16
Ta	25.85(1.06)[a] 24.62(1.03)[b]	26.49	25.75	19.99(0.64)[a] 19.93(0.60)[b]	20.87	20.56
Pb	29.04(0.87)[a] 27.76(0.82)[b] 24.33(0.62)[c]	29.76	28.91	22.45(0.55)[a] 21.73(0.49)[b]	23.44	23.09
Bi	29.39(1.52)[a] 29.12(1.49)[b]	30.12	29.25	22.72(0.79)[a] 22.24(0.75)[b]	23.73	23.37

values for the elements e_{element} is estimated as follows:

$$e_{\text{element}} = \pm \left[e_{\text{comp}}^2 + \sum_i e_i^2 \right]^{1/2}, \quad (2)$$

where e_{comp} is the rms error on the compound cross section (Table II) and e_i is the error on the subtracted cross section of element i.

From the derived total attenuation cross sections of the elements, the whole-atom integral incoherent-scattering cross sections of the elements were extracted by using the graphical method described fully in our previous paper.[3] The incoherent-scattering cross sections were also obtained from the derived total attenuation cross sections of the elements by subtracting the theoretical values of the photoeffect cross sections of Scofield[14] and the coherent scattering[15] as well as pair-production cross sections (for 1115.5 keV only) of Hubbell *et al.*[13] The graphical values and the subtraction values of the whole-atom integral incoherent-scattering cross sections thus obtained have been listed in Table IV along with the other available experimental data,[9] the bound-electron values of Hubbell *et al.*,[2] and the free-electron Klein-Nishina values.[5] It can be noticed from Table IV that while the present values agree fairly well with the bound-electron values of Hubbell *et al.*[2] within the stated errors, they differ systematically from the free-electron values. The stated errors e_{inc} on the cross sections of Table IV are estimated as follows:

TABLE IV. (*Continued.*)

Element	661.6 keV Experimental	661.6 keV KN (Ref. 5)	661.6 keV Hubbell (Ref. 2)	1115.5 keV Experimental	1115.5 keV KN (Ref. 5)	1115.5 keV Hubbell (Ref. 2)
La	14.71(0.37)[a] 14.24(0.36)[b]	14.60	14.51	11.45(0.29)[a] 11.63(0.29)[b]	11.41	11.41
Ce	14.96(0.24)[a] 14.57(0.22)[b]	14.86	14.76	11.65(0.18)[a] 11.81(0.18)[b]	11.61	11.61
Pr	15.22(0.25)[a] 14.81(0.23)[b]	15.12	15.01	11.85(0.17)[a] 11.95(0.17)[b]	11.81	11.82
Nd	15.48(0.37)[a] 14.98(0.36)[b]	15.37	15.27	12.05(0.29)[a] 12.17(0.29)[b]	12.01	12.02
Sm	16.00(0.40)[a] 15.65(0.39)[b]	15.88	15.77	12.45(0.31)[a] 12.60(0.31)[b]	12.41	12.42
Gd	16.51(0.43)[a] 16.02(0.42)[b]	16.40	16.27	12.86(0.32)[a] 12.92(0.32)[b]	12.81	12.82
Dy	17.03(0.46)[a] 16.50(0.45)[b]	16.91	16.77	13.26(0.34)[a] 13.36(0.34)[b]	13.21	13.22
Ho	17.29(0.45)[a] 16.58(0.44)[b]	17.17	17.02	13.46(0.34)[a] 13.61(0.34)[b]	13.41	13.43
Er	17.54(0.46)[a] 16.83(0.45)[b]	17.42	17.28	13.66(0.35)[a] 13.80(0.35)[b]	13.61	13.63
Ta	18.33(0.59)[a] 18.11(0.58)[b]	18.70	18.54	14.66(0.39)[a] 14.82(0.39)[b]	14.61	14.63
Pb	21.16(0.47)[a] 19.76(0.45)[b] 19.97(0.12)[c]	21.01	20.81	16.47(0.33)[a] 16.69(0.33)[b]	16.41	16.45
Bi	21.41(0.65)[a] 19.99(0.63)[b]	21.26	21.06	16.67(0.48)[a] 16.92(0.48)[b]	16.61	16.66

[a]Present values obtained by the graphical method.
[b]Present values obtained by the subtraction method.
[c]Gopal (Ref. 9).

$$e_{\text{inc}} = \pm(e^2_{\text{element}} + e^2_{\text{gr}})^{1/2} , \quad (3)$$

where e_{gr} is the rms error on the fitted value, e_{element} is the error on the derived total attenuation cross sections of the element as obtained from (2).

CONCLUSIONS

A consistent set of total attenuation cross sections and whole-atom integral incoherent-scattering cross sections of the elements La, Ce, Pr, Nd, Sm, Gd, Dy, Ho, Er, Ta, Pb, and Bi at energies 279.2, 514, 661.6, and 1115.5 keV has been obtained. These values are in good agreement with the theoretical values.[2,12,13] To our knowledge, the experimental data presented here (except perhaps that of lead, and the Conner *et al.*[11] La and Gd 279.2-keV values as cited) are the first of their kind at energies 279.2 and 514 keV.

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