

Whole-atom integral incoherent scattering cross sections of elements Na, Cl, K, Cu, Br, Rb, Ag, Cd, I, and Hg at 661.6 keV

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The whole-atom integral incoherent-scattering cross sections of elements Na, Cl, K, Cu, Br, Rb, Ag, Cd, I, and Hg have been derived with the aid of the mixture rule from the incoherent-scattering cross sections of their simple halides. The cross sections of the halides are extracted from their total attenuation cross sections measured by performing transmission experiments in a modified narrow-beam setup. The results are found to be in good agreement with the values assembled by Hubbell *et al.* [J. Phys. Chem. Ref. Data **4**, 471 (1975)] that are interpolated to 661.6 keV.

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INTRODUCTION

Incoherent scattering is an important process of γ -ray interaction in the energy region 200 keV to about 2000 keV. A literature survey on incoherent-scattering suggests that there have been a number of theoretical as well as experimental investigations on the process.

Theoretically, the scattering of γ rays by bound electrons is envisaged as a second-order process in which there are initial, intermediate, and final states of the electron. Exact calculations of the incoherent-scattering cross sections are very tedious in view of the fact that the final state of the electron can be any one of the discrete states in the continuum. The complexity of this problem is understood by the point that the exact calculation of coherent-scattering cross sections (in which the initial and final states are the same) is itself very tedious. The aforesaid complexities have led many investigators to adopt a simpler approach to the calculation of the incoherent-scattering cross sections. In this approach, the free-electron cross section is assumed to be reduced as a result of electron binding to the atom by a certain factor $S(q, z)$ which is a measure of the electron binding. This factor $S(q, z)$ is called the incoherent-scattering function. Most of the theoretical investigations on the incoherent scattering have been centered around the calculation of $S(q, z)$, by assuming different models for the atomic charge distribution. The Thomas-Fermi model [1] and the Hartree-Fock model [2] have been widely used. Hubbell *et al.* [3] have tabulated the differential as well as the integral incoherent-scattering cross sections of elements in the atomic number range $Z=1-100$ for the energy region 100 eV to 100 MeV, calculated using the self-consistent-field Hartree-Fock bound-electron wave functions.

Recently, Bergstrom *et al.* [4] have reported theoretical work on incoherent scattering that goes beyond the incoherent-scattering-function approximation. In this

work, Compton scattering from bound electrons is studied within external-field quantum electrodynamics and the independent-particle approximation but without making use of any additional approximations, such as impulse or incoherent-scattering-factor approximations. The calculation done by them is of the doubly differential cross sections for scattering of unpolarized and polarized photons from bound atomic electrons as a function of scattered photon energy and angle, based on a numerical evaluation of the second-order S matrix in self-consistent screened atomic potentials. They also discuss the cross section singly differential in scattered-photon angle, emphasizing the contribution of terms neglected when making the incoherent-scattering-factor approximation as well as implication for total cross sections and for attenuation coefficients.

On the experimental side, there has been comparatively less work. The earlier experimental work was motivated to verify the Klein-Nishina formula. The later work can be broadly classified into (1) The study of differential incoherent-scattering cross sections (2) The study of the integral incoherent-scattering cross sections. Although measurements of differential incoherent-scattering cross sections abound in literature, the experiments primarily motivated to measure the whole-atom integral incoherent-scattering cross sections are very scarce. In general, we can observe that two types of methods are widely used, viz. (1) the subtraction method (2) the extrapolation method. In the first method [5], the total attenuation cross sections are experimentally determined using a narrow-beam good-geometry setup. From the total attenuation cross sections, the whole-atom integral incoherent-scattering cross sections are derived by subtracting the interpolated theoretical values of the other competing processes like coherent scattering and photo effect. Obviously, the main drawback in the subtraction technique stems from the fact that its accuracy is seriously dependent on the theoretical values and is good enough only in a region where incoherent scattering is a very significant mode of interaction. The second method [6] although it is not dependent on theory, however, depends on how accurately the low- Z total attenuation

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cross sections are determined.

In light of the above, it was felt essential to develop an alternative method for the determination of whole-atom integral incoherent-scattering cross sections of elements. In this paper, we report the results of such a method for the elements of atomic number in the range $Z = 11-80$ for ^{137}Cs γ rays. The elemental cross sections are found to be in good agreement with the theoretical data of Hubbell *et al.* [3] within the range of experimental errors.

EXPERIMENTAL DETAILS

The radioactive source ^{137}Cs used in the present experiment was in the form of a radiographic capsule of strength 10 mCi. It was procured from the Bhabha Atomic Research Centre, Bombay, India. The detector was a well type NaI (TI) crystal of dimension $4.5 \times 5.0 \text{ cm}^2$ with a well size of diameter 1.7 cm and depth 3.9 cm, optically coupled to an RCA 8053 photomultiplier tube. The detector signal was suitably amplified by a linear amplifier and the spectral analysis including the data processing was performed in a personal computer based multichannel analyzer.

Each compound in fine powder form was filled in cylindrical plastic containers. The mass of the compound was determined in an electrical balance correct to the third decimal place. The inner diameter of the plastic containers was determined using a traveling microscope. From this, the mass per unit area was calculated. In the case of Cu, 99.9% pure foils were used. The thickness of Cu foils was determined and from this the density thickness (mass per unit area) was calculated by multiplying it by its density (8.9 g cm^{-3}).

The experimental setup was as shown in Fig. 1. It was similar to the usual narrow beam good geometry setup, except for the fact that the sample was kept exactly inside the well of the detector. The method involves the determination of the transmission spectra corresponding to this sample position.

While the sample is kept exactly inside the well there will be a maximum contribution of scattering (single as

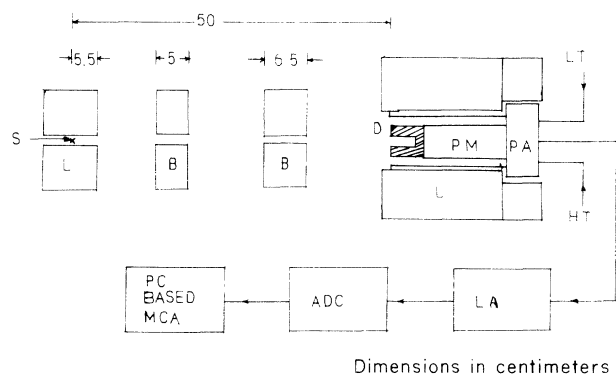


FIG. 1. Experimental setup: S—source position, L—lead shielding, B—lead collimator, D—detector, PM—photomultiplier, Pa—preamplifier, LA—linear amplifier, ADC—analogue to digital converter, HT—high voltage supply, and LT—voltage supply.

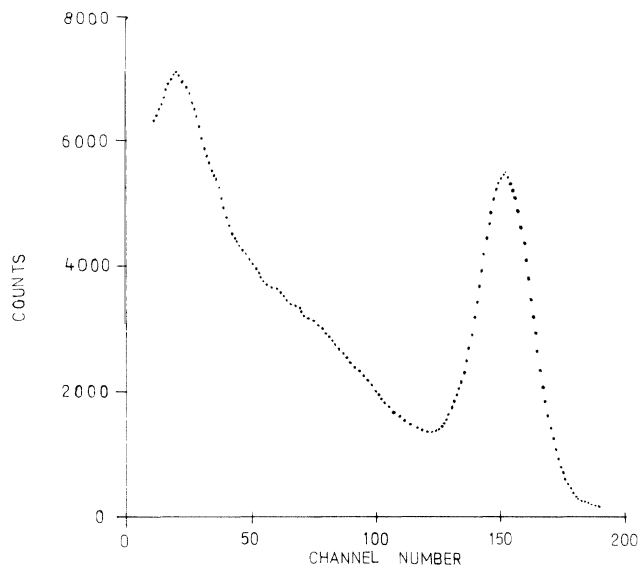


FIG. 2. Typical spectrum recorded for a sample HgCl_2 inside the NaI(Tl) detector well.

well as plural) in all directions (due to 4π geometry) to the resulting transmitted spectra (Fig. 2). Since these scattered photons contribute significantly to the lower-energy side of the photopeak of these spectra, it was noticed that there was a considerable increase in the transmitted intensity relative to the sample position outside the well. This was attributed to the fact that the sample position outside the well represents a typical narrow beam good geometry situation in which almost no energy-degraded (scattered) photons can reach the detector. This fact is made use of in the present study to evaluate the whole-atom integral incoherent-scattering cross sections of elements.

EVALUATION OF THE WHOLE-ATOM INTEGRAL INCOHERENT-SCATTERING CROSS SECTIONS OF ELEMENTS

To evaluate the incoherent-scattering cross sections, the attenuated (with sample) and the unattenuated (with empty container) spectra are used. For this purpose, the sample and the empty container were placed alternately in the path of the beam inside the well and the corresponding transmitted intensities I and I_0 were determined as the area under the photopeaks of the respective recorded spectra. Then, these values were used in the formula

$$\sigma_{\text{tot}} = A \ln(I_0/I) / 0.60225t \quad (1)$$

to calculate the total attenuation cross sections for the sample in units of barn/mole, where A is the molecular weight of the sample and t is the mass per unit area.

Next, to the transmitted intensities I and I_0 the counts corresponding to ten channels towards the lower-energy side of the photopeak in the respective spectra were added and, again, the σ_{tot} was calculated using Eq. (1). This procedure was repeated by adding the counts corresponding to ten channels to the left of the earlier case and every

time the σ_{tot} was calculated using Eq. (1). This was done till the entire region of the transmitted spectra up to the backscattered peak was encompassed.

RESULTS AND DISCUSSION

The values of σ_{tot} calculated in the case of each sample were plotted as a function of the number of channels used. A typical plot for HgCl_2 sample is shown in the Fig. 3. It can be noticed from Fig. 3 that σ_{tot} progressively decreases up to a certain value beyond which it remains constant irrespective of the number of channels considered. This saturation value of σ_{tot} is expected to be a result of the maximum scattering contribution to the transmitted intensity. As such, the saturation value σ_s would represent that value of σ_{tot} which is obtained when all the incoherently scattered photons would have reached the detector. Similarly, when the plot is extrapolated to zero channel number, this extremum value of σ_{tot} should obviously correspond to a value σ_0 which would have been obtained when no incoherently scattered (or scattered) photons could reach the detector. It is, therefore, evident that while σ_0 represents a value of σ_{tot} obtained in an ideal narrow beam good geometry setup, σ_s is that value of the total attenuation cross section obtained in a typical broad beam setup in which the angle of acceptance is such that all scattered photons reach the detector. The difference ($\sigma_0 - \sigma_s$), therefore, should yield the whole-atom integral incoherent-scattering cross section σ_{inc} of the sample.

The values of the σ_{inc} were evaluated using the above method for all the samples used in the present study. These have been listed in Table I along with the experimental errors.

The experimental errors in the present method are mainly a result of (1) counting statistics, (2) small-angle scattering contributions, (3) nonuniformity of the sample material, (4) sample impurity, (5) photon dose buildup effects, and (6) dead time of the counting instrument.

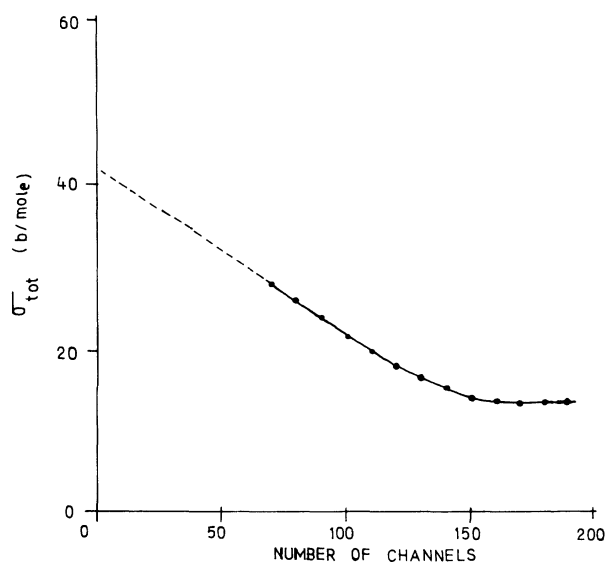


FIG. 3. Plot of σ_{tot} vs the number of channels for HgCl_2 .

TABLE I. The whole-atom integral incoherent-scattering cross section for compounds (b/mol).

| Compound | $(\sigma_{\text{incoh}})_{\text{expt}}$ | $(\sigma_{\text{incoh}})_{\text{theor}}$ |
|-----------------|---|--|
| NaCl | 7.28 | 7.16 |
| KCl | 9.33 | 9.21 |
| CuCl | 11.89 | 11.71 |
| KBr | 13.84 | 13.73 |
| RbCl | 13.93 | 13.72 |
| AgCl | 16.48 | 16.30 |
| KI | 18.38 | 18.32 |
| HgCl_2 | 29.08 | 28.88 |
| CdI_2 | 39.41 | 39.12 |

In the transmission experiment, the counting time was chosen such that at least 10^5 – 10^6 counts were recorded within the photopeak area. Thus, the error due to counting statistics was less than 0.3% in all cases.

In the present experimental setup, a distance of 50 cm was maintained between the source and the detector. This corresponds to a maximum angle of scattering from the sample to the detector of 31 min. According to the theoretical estimates, the contribution of coherent as well as incoherent scattering at such small angles to the measured cross section at 661.6 keV is negligibly small.

Nonuniformity of the sample was checked by exposing different portions of the sample material to the incident beam. It was found that any discrepancy in the attenuated intensity in each case was within counting statistics.

The error due to sample impurity can be high only when a large percentage of high Z impurities are present in the sample. In all the compounds used in the present study, the content of high Z impurities was less than 0.005%. Hence, sample impurity corrections were not applied to the measured data.

The pulse pile-up effects and the photon dose buildup effects were kept to a minimum by choosing an optimal count rate and optimal counting time. The photon dose buildup is a function of the sample thickness, its atomic number and the incident energy. In the present study, the effect of dose buildup is kept to a minimum by choosing a sample thickness such that the transmission ratio is in the range 0.1–0.4.

There was a built-in provision for dead-time correction in the multichannel analyzer.

Thus, the overall error on the measured cross sections was less than 2%. From the cross sections for the samples, the incoherent-scattering cross sections for the individual elements were derived with the aid of the mixture rule [7]. Since, most of the samples were halides, the cross sections of chlorine, bromine and iodine were first determined. The cross section for chlorine was calculated as the difference between the cross sections for Cu and CuCl. The integral incoherent-scattering cross sections for the elements Na, K, Rb, Ag, and Hg were derived by suitably subtracting the cross section for chlorine from the cross section of their chlorides. The cross section for bromine and iodine was obtained by subtracting the cross section for potassium from KBr and KI cross sections. The iodine cross section was subtracted from the cross

TABLE II. The whole-atom integral incoherent-scattering cross section for elements (b/atom).

| Elements | $(\sigma_{\text{incoh}})_{\text{expt}}$ | $(\sigma_{\text{incoh}})_{\text{theor}}$ |
|----------|---|--|
| Na | 2.83±0.32 | 2.81 |
| Cl | 4.45±0.28 | 4.35 |
| K | 4.88±0.34 | 4.86 |
| Cu | 7.44±0.15 | 7.36 |
| Br | 8.96±0.44 | 8.87 |
| Rb | 9.48±0.39 | 9.37 |
| Ag | 12.03±0.43 | 11.95 |
| Cd | 12.40±0.93 | 12.20 |
| I | 13.50±0.50 | 13.46 |
| Hg | 20.18±0.64 | 20.18 |

sections for cadmium iodine to get the cross section for cadmium. Thus, using this method, the whole-atom integral incoherent-scattering cross sections for the elements Na, Cl, K, Br, Rb, Ag, Cd, I, and Hg have been determined. These values have been listed in Table II along with the errors and compared with the interpolated values of Hubbell *et al.* The error on the elemental cross section " e_{element} " was calculated as

$$e_{\text{element}}^2 = e_{\text{compound}}^2 + e_{\text{halogen}}^2 \dots,$$

$$\begin{aligned} \text{where } e_{\text{halogen}}^2 &= e_{\text{Cu}}^2 + e_{\text{CuCl}}^2 \text{ for chlorine} \\ &= e_{\text{K}}^2 + e_{\text{KBr}}^2 \text{ for bromine} \\ &= e_{\text{K}}^2 + e_{\text{KI}}^2 \text{ for iodine.} \end{aligned} \quad (2)$$

Thus, the overall error on elemental cross sections were calculated to be about 3–4%. A good agreement can be observed between theory and experiment within the range of experimental errors.

CONCLUSIONS

It is felt that this method embarks upon an alternative technique for the determination of the whole-atom integral incoherent-scattering cross sections of elements. Since the method makes use of merely the transmitted spectra, it appears to be much superior to the earlier methods like subtraction technique of extrapolation method, as it is not seriously dependent on subtracted theoretical contributions. However, the suitability of the method at lower energies needs to be thoroughly investigated, because if found suitable, this will be one of the more accurate methods for the determination of whole-atom integral incoherent-scattering cross sections of elements.

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