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X-RAY SCATTERING FROM LOW Z MATERIALS — EXPERIMENTAL
RESULTS FOR POLYSTYRENE

By

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I. INTRODUCTION

Knowledge of the energy distribution and magnitude of X-rays scattered from low Z materials is of importance to many laboratory and field experiments. We have selected amorphous polystyrene (CH) for detailed experimental investigation and will present results obtained from scattered X-ray measurements using intense high purity incident K_{α} X-ray beams from 4 to 36 keV. Part of our motivation was to test a new Monte Carlo code which was developed by one of us (EML) to calculate single and plural total X-ray scattering for the case of a collimated beam incident on a slab.

In Section II, after discussing general X-ray scattering theory, we will present first order calculations of single scattering from a slab. Using these approximate formulae we then compare total X-ray scattering from various low Z materials. Practical considerations involved in the choice of low Z scatterers are reviewed in Section III.

The details of the polystyrene X-ray scattering measurements are presented in Sections IV, V, and VI; and the Monte Carlo calculation is outlined in Section VII. In Section VIII we compare, for each incident X-ray energy and scattering angle, the measured total scattered X-ray intensity (normalized to one incident photon) and scattered X-ray energy distribution with the corresponding results obtained using the Monte Carlo code. We then discuss the serious disagreement between experiment and calculation which is attributed to diffraction enhancements arising from an average polystyrene intrachain carbon-carbon bond length. The plural scattering calculations of the Monte Carlo code are also presented in Section VIII and a more accurate experimental test is suggested involving further work at higher incident X-ray energies.

II. SCATTERED X-RAY FLUX CALCULATIONS

A) General Theory

The differential cross section for the scattering of X-rays from a free atom⁽¹⁾ is given by

$$d\sigma/d\Omega_T = F^2 d\sigma/d\Omega_{\text{Thompson}} + (Z - f) d\sigma/d\Omega_{\text{KN}} \quad (1)$$

in units of barns/atom.ster

Here the coherent contribution for scattering from a free electron is given by

$$d\sigma/d\Omega_{\text{Thompson}} = \frac{1}{2} r_o^2 (1 + \cos^2\theta) \quad (2)$$

where the square of the classical electron radius $r_o^2 = 7.940 \times 10^{-2}$ barns and θ = the angle between the incident and scattered photon directions.

For the atomic form factor F we have used the numerical Hartree-Fock calculations of Cromer and Mann⁽²⁾. Their results were given in the form of coefficients for their analytical fit to $F(\theta, E)$ calculated as a function of the parameter

$$x = \frac{E \sin \theta/2}{12.39831}$$

where E is the incident photon energy in keV. The differential cross section for incoherent scattering from a free electron is given by the well known Klein Nishina formula⁽³⁾

$$d\sigma/d\Omega_{\text{KN}} = \frac{1}{2} r_o^2 [1 + k(1 - \cos\theta)]^{-2} \left[1 + \cos^2\theta + \frac{k^2(1 - \cos\theta)^2}{1 + k(1 - \cos\theta)} \right] \quad (3)$$

where

$$k \equiv E/511 \quad (E \text{ is in keV})$$

When multiplied by the factor $(Z - f)$ the differential scattering from a free atom is obtained where Z is the atomic number and f is the incoherent scattering form factor. Here we have used the calculations of Cromer and Mann⁽⁴⁾ except that in the case of carbon we have used the results of Brown⁽⁵⁾.

B) First Order Calculations of Single Scattering from a Slab

Consider the case of a beam of photons from a source of intensity N_0 photons/sec a distance R from a slab scatterer of thickness T making an angle ψ with the incident beam as shown in Figure 1. Let A = the beam area at the scatterer. Let s be the distance traveled in the slab by the photon to the volume element $dV = da ds$ and s_r or s_t be the distance traveled out of the slab by the scattered photon in the reflective or transmitted direction respectively. The angle θ between the scattered photons and the incident beam, the exit angle ϕ , and the angle ψ are related by

$$\phi = 90^\circ + \psi - \theta \quad (\text{reflection mode}) \quad (4)$$

The total scattering from the volume element into a solid angle Ω (ster) is given by

$$\frac{d\sigma}{d\Omega_T} \Omega n \exp(-\mu s) \exp(-\mu_2 s_r) dV \quad (5)$$

where the transmission into and out of dV is

$$\exp(-\mu s) \text{ and } \exp(-\mu_2 s_r)$$

μ and μ_2 are the total absorption coefficients (in cm^{-1}) for the incident and scattered photons respectively, and n is the number of atoms per cm^3 in the scatterer, given by

$$n = A_0 \rho / w \quad (6)$$

where A_0 is Avogadro's number, ρ is the density in gms/cm³, and w is the molecular wt. of the material in gms/mole.

Assuming a narrow beam and a small scattering solid angle so that θ (or θ_t) does not depend on s we integrate over the slab, i.e., over the slant thickness $ds = \frac{dt}{\sin\psi}$ to obtain the total number of single scattered photons N_s .

$$N_s = \frac{N_0}{4\pi R^2} \int da \int \frac{d\sigma}{d\Omega_T} \Omega n e^{-\mu t_1/\sin\psi} e^{-\mu_2 t_1/\cos\phi} \frac{dt_1}{\sin\psi} \quad (7)$$

since

$$s = t_1/\sin\psi \text{ and } s_r = t_1/\cos\phi \quad (8)$$

Then

$$N_s = N_0 \frac{A}{4\pi R^2} \frac{d\sigma}{d\Omega_T} \frac{\Omega n}{\sin\psi} \frac{1}{g} (1 - e^{-gT}) \quad (9)$$

where

$$g = \mu/\sin\psi + \mu_2/\cos\phi \text{ and } A = \int da$$

We then define the scattering efficiency ϵ_s , i.e., the number of scattered photons per ster per incident photon by

$$\epsilon_s = \frac{N_s}{N_0 (A/4\pi R^2) \Omega} \quad (10)$$

We have

$$\epsilon_s = \frac{d\sigma}{d\Omega_T} \frac{n}{\sin\psi} \frac{1}{g} (1 - e^{-gT}) \quad (\text{reflection mode}) \quad (11)$$

For coherent scattering and low energy incoherent scattering, $\mu = \mu_2$. For $\theta = 90^\circ$, $\psi = 45^\circ$, and $\mu \sim \mu_2$ Equation (11) reduces to

$$\epsilon_s (90^\circ) = \frac{d\sigma}{d\Omega_T} \frac{n}{2\mu} [1 - \exp(-2.83\mu T)] \quad (12)$$

A useful approximation for small μT is

$$\epsilon_s \approx \frac{d\sigma}{d\Omega_T} n \frac{T}{\sin\psi} \quad (13)$$

For the transmission mode, i.e., for scattered photon emission from the back side of the slab since

$$s_t = t_2/\sin\phi_t \text{ and } \phi_t = 180^\circ - (\theta_t + \psi) \quad (\text{transmission mode}) \quad (14)$$

we similarly obtain

$$\epsilon_s = \frac{d\sigma}{d\Omega_T} \frac{n}{\sin\psi} \exp(-\mu_2 T/\sin\phi_t) \frac{1}{G} (1 - e^{-GT}) \quad (\text{transmission mode}) \quad (15)$$

where

$$G = \mu/\sin\psi - \mu_2/\sin\phi_t$$

The maximum value of ϵ_s is obtained when

$$T = \frac{1}{G} \ln \frac{\mu \sin\phi_t}{\mu_2 \sin\psi} \quad (16)$$

and is given by

$$\epsilon_{s \max} = \frac{d\sigma}{d\Omega_T} \frac{n}{\mu} \exp(-\mu_2 T/\sin\phi_t) \quad (\text{transmission mode}) \quad (17)$$

Note that as the factor G approaches zero

$$\frac{1}{G} (1 - e^{-GT}) \text{ approaches } T$$

Thus when $\phi_t = \psi$ and $\mu = \mu_2$ we have

$$\epsilon_s = \frac{d\sigma}{d\Omega_T} \frac{n T}{\sin\psi} \exp(-\mu T/\sin\psi) \quad (18)$$

It can be readily shown that ϵ_s has a maximum value for $T = \sin\psi/\mu$ and is therefore given by

$$\epsilon_{s \text{ max}} = 0.367 \frac{d\sigma}{d\Omega_T} \frac{n}{\mu} \quad (\text{transmission mode, } \phi_t = \psi, \mu \sim \mu_2) \quad (19)$$

Note that the maximum value of the scattering efficiency is simply related to the ratio of the differential scattering to the total absorption coefficient.

C) Comparison of Total X-ray Scattering from Low Z Materials

In order to compare the low energy X-ray scattering efficiency of various low Z materials we have calculated their maximum scattering efficiencies for the transmission mode. A commonly used scattering geometry was chosen, i.e., $\theta_t = 90^\circ$ and $\psi = 45^\circ$ for which $\phi_t = \psi$. We also have assumed that $\mu = \mu_2$ and have used Equation (19) to obtain the results in Table 1 for lithium, beryllium, boron, and polystyrene (CH) scatterers. Theoretical X-ray differential scattering cross sections were used⁽⁶⁾; and the total absorption cross sections were obtained from the compilation of McMaster et al⁽⁷⁾. Note that for the thickness $T = \sin 45^\circ/\mu$ which maximizes transmission mode scattering, the scattering at $\theta = 90^\circ$ in the reflective case is obtained as calculated from Equation (12) by multiplying the values in the table by a factor of only 1.18. Moreover reflective 90° scattering for a very thick slab is greater by only a factor of 1.36.

For each of the scatterers we note a rapid decrease in the scattering efficiency with a decrease in energy. The increase in the differential scattering cross section with a decrease in energy is counteracted by a more rapid increase in the total cross section.

III. CHOOSING LOW Z SCATTERING MATERIALS

Li metal Lithium metal scatterers were developed for a field experiment requiring high scattering efficiency. Because of its extreme chemical reactivity an encapsulation technique was devised.

Using a pure argon atmosphere lithium metal was melted in a carbon die to obtain a disk of uniform thickness. The disk was placed in a lucite ring and sandwiched between 1 mil Be foils forming an air tight arrangement. Although the encapsulation technique was successful, severe calibration problems were encountered*. The low energy scattered X-ray spectra contained a complex Bragg scattering contribution which varied with each sample and which changed significantly for small changes in the sample angle ψ (Figure 1). Under such conditions an accurate calibration requires duplicating the field geometry as closely as possible in the lab and aligning the scatterer in the field experiment as similarly as possible.

Unfortunately the Bragg contribution to the scattering distribution cannot be calculated. Thus calibration uncertainties of as much as $\pm 15\%$ are to be expected for energies below 7 keV where Bragg effects are the most significant. For field experiments requiring the maximum scattering efficiency we suggest trying encapsulated lithium hydride which is more hygroscopic than lithium metal but which may involve fewer difficulties from Bragg contributions.

We would expect to encounter similar problems using Be metal as a scatterer since scattered X-ray lines due to crystalline diffraction enhancements have been observed⁽⁸⁾. Unlike Be metal beryllium-hydride is not yet available with adequate purity to be used as a low energy X-ray scatterer.

*Lithium calibration measurements were made using bremsstrahlung from the molybdenum target of the R&D group low energy X-ray facility. We wish to thank N. Del Grande for collaborating on these measurements.

The practical problems involved in using boron as a scatterer are not sufficiently compensated by its modest increase in scattering efficiency compared with CH.

Polystyrene was chosen as a scatterer for detailed experimental study because of its following advantages:

- 1) It reacts extremely slowly with air and water vapor.
- 2) It can be obtained with sufficient purity.
- 3) Its thickness and uniformity can be adequately controlled.
- 4) It can be obtained as an amorphous hydrocarbon, i.e. it will not exhibit sharp crystalline diffraction enhancements.

IV. EXPERIMENTAL ARRANGEMENT

The experimental arrangement used for X-ray scattering experiments at the R&D group high-flux, low energy X-ray facility is shown in Figure 2. The source, which provides nearly monoenergetic X-rays from 1.5 to 8 keV, is discussed in Ref. 9. The incident beam was defined by a 1.27 cm diam collimator located in the vacuum chamber only 10 cm from the slab scatterer. The spectrum of scattered X-rays was observed using a 5 mm diam Si(Li) detector of 3 mm thickness and with 195 keV resolution for the Fe-55 line at 5.8 keV. The angular position of this detector was precisely varied to obtain scattering angles from 35 to 145° with two degree source to detector angular resolution. The angle of the slab scatterer ψ was adjusted for maximum scattering efficiency, i.e., such that $2\psi = \theta$. A 4 mm diameter gold collimator inside the Si(Li) detector cryostat defined the scattered beam (minimizing detector edge penetration effects). Additional detector collimation was used so that only X-rays from the scatterer could be observed. A highly collimated Si(Li) detector at the end of the X-ray exit pipe (793 cm from the source) was used to obtain the incident X-ray spectra, and to monitor the incident and, when possible, the transmitted beam flux.

Scattering measurements at 16, 25, and 36 keV were made using a very similar scattering geometry and the filtered K_{α} X-ray source described in Ref. 10. In this case scattered spectra were obtained with the same Si(Li) detector and with nearly the same angular resolution. The beam flux in the higher energy range was monitored using a NaI(Tl) detector.

V. EXPERIMENTAL PROCEDURE

Si(Li) X-ray pulse height analyzer spectra were accumulated at $\theta = 45^\circ$, 90° and 135° for all energies. Finer angular increments were used for $E = 4$ keV after significant deviations from the calculated scattering efficiencies were observed. The total number of photons incident on the scatterer during each run needed to obtain a scattered spectrum was determined using the following information:

- 1) The solid angle subtended by the defining collimator of the monitor detector compared to that of the scatterer.
- 2) The transmitted count rate measured during the run.
- 3) The ratio of monitor counts observed during short runs with and without the scatterer.

The latter runs were used to minimize the effects of slow drift of the incident beam intensity. A highly collimated NaI(Tl) detector with a 5 mil Be entrance window and suitable energy discrimination was used to monitor the incident count rate for measurements of 16 keV and above. At the low energy facility a windowless, highly collimated Si(Li) detector located at the end of the exit vacuum pipe was used for monitoring. In this case an incident beam monitor spectrum was integrated over the same energy range as that of the Si(Li) scatter detector. In obtaining scattered photon spectra for energies below 7 keV the transmission count rate was too low to be of practical use in monitoring the incident count rate. This necessitated correcting scattered X-ray data obtained between various monitor, i.e., sample out, runs for the slow increase in beam intensity which occurred each day. The contribution to the estimated uncertainty in the incident beam intensity in these cases varied from 1 to 2%.

VI. DATA ANALYSIS

A) Scattering Efficiency

Each of the Si(Li) X-ray spectra was integrated over a suitable energy range to obtain the total number of scattered X-rays during the run. A fluorescent peak at 3.7 keV due to calcium impurity (only 0.036% by weight) was subtracted from each spectrum with incident energies below 7 keV and was large enough to limit this preliminary study to energies of 4 keV and above. Each of these scattered count rates, the incident count rate, the detector efficiency, and the detector solid angle were used to determine the scattering efficiency defined in Equation (10).

B) Scattering Efficiency Measurement Uncertainties for $E \leq 8$ keV

Uncertainties in all the geometrical parameters involved in the experiments which were conducted to determine low energy scattering efficiencies were negligible compared to the estimated error of 1.3% in the monitor collimator area. Combining the uncertainties in the geometrical parameters in quadrature yielded a value of 1.5%, a systematic error contribution to all the low energy measurements. Additional measurement errors in each of the scattered photon intensities N_s included that due to counting statistics (varying from 0.2 to 0.6%) and that due to the calcium impurity correction. The latter varied from zero to 1.4% and, of course, increased as the incident energy approached 3.7 keV.

Measurement errors which were incurred in the determination of each of the incident photon intensities included the error due to count rate drift (varying from zero to at most 2%) and the error in the measurement of the monitor deadtime estimated to be $\pm 1\%$ for all energies. (A corresponding deadtime error in the system used to measure scattered flux was negligible due to the low scattered photon count rates.)

In the measurement of the ratio N_s/N_m , errors in the efficiencies of the detectors tend to cancel since they were both similar 3 mm thick Si(Li) systems obtained from Berkeley. The only difference in construction, i.e., a 2 mil Be window in front of the scattering detector, resulted in errors in N_s conservatively estimated to be $\frac{1}{2}\%$ for all energies. The final error in each low energy scattering efficiency value was between 2 to 3% and was obtained by combining in quadrature all of the errors discussed above.

C) Scattering Efficiency Measurement Uncertainties for $E \geq 16$ keV

A 3% systematic error was incurred in each of the higher energy measurements due to the combined uncertainties in the geometry. This was due mainly to an estimated error of 2.5% in the solid angle of the scattered photon detector. In determining scattered photon intensities, errors due to statistics and background subtraction were negligible compared to errors in the detector efficiencies (2, 4, and 8% at 16, 25, and 36 keV respectively). Measurement errors (due to counting statistics, drift, and detector deadtime) incurred in determining the incident photon intensities were estimated to be 1% or less. Combining the above errors in quadrature yielded uncertainties of 4, 5, and 9% for scattering efficiencies measured at 16, 25, and 36 keV respectively.

Before presenting our experimental results we will discuss corresponding scattering efficiencies which were calculated using X-ray scattering factors for neutral carbon and hydrogen atoms (Ref. 3 and 5).

VII. MONTE CARLO CALCULATION

The relations 11 and 15 give a good approximation to the scattering efficiency for the case when the incident and scattered beam have little divergence and multiple scattering is negligible. In order to more accurately calculate the scattered spectrum for the typical slab geometry used in laboratory or field experiments, a Monte Carlo code was developed.

A) Procedure

The experimental geometry is duplicated very accurately in this calculation. An energy spectrum is input, e.g., a spectrum measured with a beam monitor detector, is used for the input source spectrum. X-rays are tracked in the slab to obtain the single, double, and triple scattering contributions to the detector spectrum as follows:

The differential cross sections given in Equations (1), (2), and (3) are used along with their respective scattering factors. The total absorption cross sections were obtained from McMaster et al*.

The procedure is as follows with all decisions made by sampling random numbers:

- 1) A particle is born with an energy determined by the input spectrum and with a weight of $1/N$ where N is the total number of particle histories to be run.
- 2) The direction of the particle is chosen such that it will pass through the collimation and strike the scatterer, the weight of the particle being reduced by a factor equal to the solid angle subtended by the collimation at the source.

*In the case of the CH scattering experiments, we normalized tables of total cross sections (Ref. 7) to that measured for each of the incident photon energies. We wish to thank A. Oliver for measuring these total cross sections using the Hilger facility.

- 3) The particle is forced to interact somewhere along its path through the scatterer. The weight is further reduced by the factor $1 - e^{-\mu s}$ where μ is the total cross section and s is the path length through the target. This factor is just the probability that the particle interacts in the target.
- 4) The distance and direction from the point of interaction to the detector are calculated. The scattered contribution at the detector is then given by:

$$W \frac{1}{\mu} \frac{d\sigma}{d\Omega_x} \Omega \exp(-\mu_2 s_r)$$

Where: W = the particles' weight

μ = the total cross section at the primary energy

$\frac{d\sigma}{d\Omega}$ = the differential cross section, a function of energy and the scattering angle

x = either coherent or incoherent

Ω = the detector solid angle, approximated by the detector area over the square of the distance from the interaction point to the detector

μ_2 = the total cross section at the scattered energy

s_r = the distance out of the scatterer in reflection geometry

These represent the single scattered coherent and incoherent contributions at the detector.

- 5) We now return to the interaction point and determine whether the scattering event is coherent and incoherent. The particle is allowed to scatter into an arbitrary direction, and the weight is reduced by the factor $\frac{4\pi}{\mu} \frac{d\sigma}{d\Omega_x}$ (the probability of scattering coherently or incoherently at that angle).

Steps 3) and 4) are repeated to yield the **doubly scattered** contributions. This process is terminated after the third scatter since the contributions for further scatters are negligible.

B) Uncertainties in the Calculated Value of ϵ_s and Expt/Calc for $E \leq 8$ keV

The statistical uncertainty in the calculated scattering efficiency varied from 0.6 to 0.9%. This was obtained in each case by running 20 problems with 2000 particles and calculating the external error. The calculated results were insensitive to the target thickness but **sensitive to the error in the experimental total cross section. The corresponding components** due to these cross section errors varied from 2 to 4%. Use of the analytical fit to the carbon coherent atomic form factor F resulted in no significant error contribution. The incoherent carbon atomic form factor f was more difficult to fit, however the largest error contribution due to deviations of the fit was 0.8%. Thus the total uncertainty in each calculated value of the scattering efficiency ϵ_s , obtained by combining the above uncertainties in quadrature, was dominated by the error in the total cross section. These uncertainties varied from 2 to 4% and when combined with the measurement errors yield uncertainties between 3 and 5% for the ratio of measured to calculation scattering efficiency ($E \leq 8$ keV).

C) Uncertainties in the Calculated Value of ϵ_s and Expt/Calc for $E \geq 16$ keV

For the higher energy range the uncertainty in each value of the scattering efficiency obtained using the Monte Carlo calculation was similarly due almost entirely to the uncertainty in the total absorption cross section. This was estimated to be 2% for all $E \geq 16$ keV. Compared to this source of error the uncertainties due to statistics and deviations of analytical scattering form factor fits were negligible. The uncertainty in the ratio of experimental to calculated scattering efficiencies was in each case due mainly to the uncertainty in the experimental measurement. As discussed in Section VI-C, these estimated uncertainties are 4, 5, and 9% for measurements at 16, 25, and 36 keV respectively.

VIII. RESULTS AND DISCUSSION

The polystyrene X-ray scattering results are summarized in Table 2 for the low energy range and Table 3 for incident energies from 16 to 36 keV. The number scattering efficiency measured for each incident energy, slab thickness, and scattering angle is presented along with the ratio of measured to calculated values. Ratios of coherent and single scattered photons to the total number of scattered photons are presented in the last two columns. These ratios were obtained from Monte Carlo calculations.

A) Plural Scattering

Data were taken at 8 and 36 keV incident energy for scatterers varying considerably in thickness in order to test the plural scattering contributions determined by the Monte Carlo code. Such contributions were found to be less than 3% at and below 8 keV and were not significant. We were thus justified in obtaining most of the low energy data with one sample of 0.127 cm thickness. Since a larger fraction of plural scattering was expected for the case of 36 keV photons, we obtained scattered spectra for 3 slabs of varying thickness. We note that with a decrease in thickness the calculated plural scattering into the detector at 90° varied from 24 to 11% and the measured scattering efficiency decreased by factor of 3; however, the ratios of experimental to calculated efficiencies for the 3 slabs are in good agreement. The agreement is also fairly good at 45° and 135° and is certainly within experimental error. The results at 36 keV are independent of scatterer thickness indicating that there are no serious problems in the calculation involving the geometry; e.g., handling of plural scattering.

B) Expt/Calc Scattering Efficiency Ratios

The ratio of measured to calculated values of the polystyrene scattering efficiency are shown in Figure 3 plotted as a function of the scattering parameter χ . A large broad enhancement of the scattering efficiency was observed at $E = 4$ keV and $\chi \sim 0.1$. A definite systematic

variation in scattering efficiency with χ (but not necessarily θ) for the 1.27 mm sample can be seen. Note the secondary enhancements at $\chi = 0.22$ and 0.44. This strong Bragg interference enhancement corresponds to an average d-value of $\sim 4.7\text{\AA}$.

This effect has been observed in previous X-ray studies of amorphous polystyrene⁽¹¹⁾ and has been attributed to the average intrachain carbon-carbon bond length. According to these studies a weaker enhancement should also be observed at about twice this d spacing due to an average carbon-carbon bond distance between polymer chains. To observe this experimentally requires energies lower than those used in this preliminary study. Similar broad Bragg enhancements are expected for all CH polymers (Ref. 11). Therefore there is no particular advantage in extending the study to other such materials. One of the amorphous polymers which appears slightly different (polymethyl methacrylate) would yield an enhancement distribution similar to that of Figure 3 but shifted to about 75% lower values of χ . However this polymer contains oxygen which tends to significantly reduce the scattering efficiency.

As shown in Figure 4 the experimental efficiencies for $E \geq 16$ keV tend to be slightly greater than their corresponding calculated values. In general, the difference does not greatly exceed the estimated error in the ratio. No significant variations with angle or scattering parameter χ can be noted.

We conclude that the scattering efficiency of pure CH scatterers can be calculated to better than 25% above $\chi = 0.15$ using the Monte Carlo code and form factors which neglect these interatomic effects. For greater accuracy particularly below $\chi = 0.15$ the curve of Figure 3 should be used as a correction to the calculation. We expect that due to the amorphous nature of polystyrene these results can be applied to most high purity commercial CH samples. However, more work is needed to confirm this. Note that we have obtained very good agreement at 8 keV using two different samples.

C) Comparison of Experimental Observed and Calculated Scattered X-ray Spectra

Shown overlayed in Figure 5 are examples of the incident X-ray spectra obtained with the Si(Li) monitor detector used at the low energy high flux facility. By varying the source high voltage we have obtained additional spectra which indicate that the low energy tail associated with each peak is due mainly to bremsstrahlung background and not to detector response. Shown also with each experimentally obtained spectrum is the source input spectrum generated for use in the Monte Carlo code. The spectra observed for 90° X-ray scattering from the 1.27 mm polystyrene sample are shown overlayed in Figure 6 along with the corresponding Monte Carlo results. Note that the calculated and observed spectra in each case are plotted absolutely and are not normalized to each other. One can see in the expanded plot of Figure 7 that the spectral shape agreement would be much better if the previously discussed interatomic effects were taken into account; i.e., if we multiplied the amplitude of the calculated spectrum by the ratio of observed to calculated total scattering efficiency.

The unresolved K_{α} lines of the calcium impurity are an interesting feature of the 4 and 5 keV spectra of Figure 8. Note the relative increase in the K_{α} production at the incident energy (4.09 keV) closest to the calcium K-edge.

The spectrum observed for 90 degree scattering of 36 keV Pr K_{α} 's from 3.2 mm thick polystyrene is shown in Figure 9 along with the Monte Carlo prediction. Although the spectral shape agreement is not good the predicted distribution of doubly and triply scattered events is reasonable. These data were obtained with a Si(Li) which has poor efficiency in this energy range. In the near future we plan to improve the comparison with the aid of a larger, more efficient intrinsic Ge detector and a cleaner incident spectrum (obtained with a thicker post-filter). It should also be noted that the incident spectrum used for each of the higher energy Monte Carlo calculations was a simple K_{α} and K_{β} distribution not based on an experimentally observed incident spectrum. The goal of further work, of

course, is to more accurately test the plural scattering calculations of the Monte Carlo code and may be best met by extending the measurements to higher incident energies. This is now possible with the new high resolution Ge detector.

REFERENCES

- (1) R. W. James, The Optical Principles of the Diffraction of X-rays, (Cornell Press, 1965).
- (2) D. T. Cromer and J. B. Mann, "X-ray Scattering Factors Computed from Numerical Hartree-Fock Wave Functions". Acta Cryst. A24, 321 (1968).
- (3) Handbuck Der Physik, Vol. XXXIV Corpuscles and Radiation in Matter II, S. Flugge Ed., "Compton Effect" by R. D. Evans.
- (4) D. T. Cromer and J. B. Mann, J. Chem. Phys. 47, 1892 (1967).
- (5) R. T. Brown, EGG-1183-1505 (January, 1971).
- (6) R. J. Stinner, W. H. McMaster and N. Kerr Del Grande, Theoretical Differential Scattering Cross Sections of X-rays for H, He, Li, Li⁺, Be, B, C, and C in the Valence State, Report UCRL-14403 (1965).
- (7) W. H. McMaster, N. Kerr Del Grande, J. H. Mallett and J. H. Hubbell, Compilation of X-ray Cross Sections, Report UCRL-50174, Section II, Rev. I (1969).
- (8) Private Comm. N. Del Grande and A. Oliver, LLL, Livermore (1970).
- (9) J. L. Gaines, Nuclear Inst. and Meth. 102, 7 (1972).
- (10) P. J. Ebert, J. L. Gaines, and G. R. Leipelt, Nucl. Inst. and Meth. 99, 29 (1972).
- (11) Encyclopedia of Polymer Science and Technology, Volume 13, R. F. Boyer Ed., (John Wiley & Sons, 1970), pp. 128-447.

TABLE 1

Comparison of Low Z X-ray Scattering Materials

SAMPLE	E keV	μ cm^{-1}	T_{max} mm	(a) $\frac{d\sigma}{d\Omega_T}$	(b) $\epsilon_{s \text{ max}}$ $\times 10^{-4}$	(c) $\frac{\epsilon_{s \text{ max}}}{\epsilon_{s \text{ max}} (\text{CH})}$
${}^3\text{Li}$	2	13.9	0.51	0.22	2.7	4.2
	4	1.61	4.38	0.17	18.1	5.9
	8	0.26	27.5	0.14	92.9	6.9
	15	0.12	61.7	0.12	172.0	2.3
${}^4\text{Be}$	2	135.0	0.05	0.40	1.3	2.1
	4	15.2	0.46	0.24	7.1	2.3
	8	1.9	3.74	0.20	46.8	3.5
	15	0.54	13.2	0.16	139.0	1.9
${}^5\text{Boron}$	2	387.0	0.02	0.66	0.8	1.30
	4	45.6	0.16	0.36	3.8	1.23
	8	5.3	1.34	0.24	21.8	1.61
	15	1.1	6.52	0.21	93.4	1.25
Polystyrene ${}_{6+1}\text{CH}$	2	296.0	0.02	1.06	0.06	1.0
	4	33.7	0.21	0.59	3.1	1.0
	8	4.4	1.61	0.34	13.5	1.0
	15	0.70	10.1	0.29	74.6	1.0

(a) Total differential scattering cross-section in units of barns/atom·ster ($\theta = 90^\circ$).(b) $\epsilon_{s \text{ max}}$ = maximum scattering efficiency $\left(\frac{\text{scattered photons}}{\text{incident photon} \cdot \text{ster}} \right)$ for $\theta_t = 90^\circ$, $\psi = 45^\circ$, $\mu = \mu_2$, and transmission mode calculated using Equation (19).

(c) The maximum scattering efficiency (t mode) relative to that of CH.

TABLE 2

SUMMARY OF RESULTS
Low Energy Scattering from Polystyrene CH (T = 0.13 cm)

E keV	θ	(a) χ	(b) ϵ_s $\times 10^{-3}$	Expt/Calc	Scattering	
					Coherent %	Single %
4.09	35°	0.099	3.23 $\pm$ 0.10	2.53 $\pm$ 0.10	96	99.6
	45°	0.126	2.17 $\pm$ 0.07	2.16 $\pm$ 0.09	93	99.5
	60°	0.165	0.676 $\pm$ 0.02	1.00 $\pm$ 0.04	88	99.3
	75°	0.200	0.454 $\pm$ 0.013	0.95 $\pm$ 0.04	82	99.1
	90°	0.233	0.472 $\pm$ 0.014	1.24 $\pm$ 0.05	75	98.9
	135°	0.305	0.443 $\pm$ 0.013	0.99 $\pm$ 0.04	60	99.2
4.95	45°	0.152	1.63 $\pm$ 0.04	1.01 $\pm$ 0.04	89	99.2
	90°	0.282	0.556 $\pm$ 0.014	0.95 $\pm$ 0.04	64	98.4
	135°	0.369	0.677 $\pm$ 0.017	0.97 $\pm$ 0.04	49	98.8
5.90	45°	0.182	1.92 $\pm$ 0.06	0.81 $\pm$ 0.04	85	98.8
	90°	0.337	0.79 $\pm$ 0.02	0.93 $\pm$ 0.05	54	97.8
	135°	0.440	1.31 $\pm$ 0.04	1.25 $\pm$ 0.06	41	98.4
6.93	45°	0.214	3.00 $\pm$ 0.06	0.91 $\pm$ 0.04	78	98.3
	90°	0.395	1.29 $\pm$ 0.03	1.09 $\pm$ 0.05	45	97.3
	135°	0.516	1.50 $\pm$ 0.03	1.04 $\pm$ 0.05	35	98.1
8.05	45°	0.248	4.52 $\pm$ 0.09	1.11 $\pm$ 0.03	71	97.8
	90°	0.459	1.68 $\pm$ 0.03	1.15 $\pm$ 0.03	38	97.0
	135°	0.600	1.76 $\pm$ 0.04	1.00 $\pm$ 0.03	30	98.0
8.05 ^(c)	45°	0.248	-	-	71	97.6
	90°	0.459	2.04 $\pm$ 0.04	1.16 $\pm$ 0.03	38	96.2
	135°	0.600	2.36 $\pm$ 0.05	1.03 $\pm$ 0.03	30	97.2

(a) $\chi \equiv \frac{\sin \theta/2}{\lambda} = \frac{E \sin \theta/2}{12.398}$

(b) $\epsilon_s \equiv N_s/N_{in}$ ster [The scattering efficiency defined by Equation (10).]

(c) T = 0.26 cm

TABLE 3

SUMMARY OF RESULTS

X-Ray Scattering from Polystyrene CH

E keV	T cm	θ	(a)	(b)	Expt/Calc	Scattering	
			χ	ϵ_s $\times 10^{-3}$		Coherent %	Single %
15.74	0.64	45°	0.49	18.7 ± 0.5	1.11 ± 0.05	34.9	90.7
		90°	0.90	8.8 ± 0.3	1.08 ± 0.04	18.8	88.1
		135°	1.2	10.2 ± 0.4	1.10 ± 0.05	12.4	91.5
25.19	1.27	45°	0.78	37. ± 2.	1.07 ± 0.06	21.4	84.3
		90°	1.44	17.9 ± 0.9	1.04 ± 0.06	6.2	78.4
		135°	1.88	21.0 ± 1.0	1.11 ± 0.06	2.4	83.9
35.86	1.27	45°	1.11	39. ± 3.	0.92 ± 0.08	12.2	82.8
		90°	2.05	18.6 ± 1.6	0.98 ± 0.09	1.5	76.1
		135°	2.67	24. ± 2.	1.19 ± 0.10	0.6	82.4
35.86	0.64	45°	1.11	25. ± 2.	0.91 ± 0.08	12.5	87.8
		90°	2.05	10.2 ± 0.9	0.97 ± 0.09	1.6	83.1
		135°	2.67	12.2 ± 1.1	1.13 ± 0.10	0.6	88.3
35.86	0.32	45°	1.11	14.7 ± 1.3	0.94 ± 0.08	12.9	92.1
		90°	2.05	5.3 ± 0.5	0.98 ± 0.09	1.7	88.9
		135°	2.67	6.3 ± 0.5	1.13 ± 0.10	0.6	92.7

$$(a) \quad \chi = \frac{\sin \theta/2}{\lambda} = \frac{E \sin \theta/2}{12.398}$$

$$(b) \quad \epsilon_s \equiv N_s/N_{in} \cdot \text{ster} \quad [\text{The scattering efficiency defined by Equation (10).}]$$

Figure 1 Geometry defined for narrow beam X-ray slab scattering.

Figure 2 Experimental arrangement used for low energy scattering experiments.

Figure 3 The ratio of measured to calculated X-ray number scattering from amorphous polystyrene for incident energies from 4 to 8 keV.

Figure 4 The ratio of measured to calculated X-ray number scattering from amorphous polystyrene for $E = 16, 25$, and 36 keV.

Figure 5 Incident X-ray spectra obtained with a Si(Li) monitor detector (labeled according to energy) and corresponding Monte Carlo code input.

Figure 6 Spectra of low energy X-rays scattered at 90° from polystyrene and corresponding calculated spectra.

Figure 7 Spectra of 6, 7, and 8 keV X-rays scattered at 90° from polystyrene and corresponding calculated spectra.

Figure 8 Spectra of 4 and 5 keV X-rays scattered at 90° from polystyrene and corresponding calculated spectra.

Figure 9 Spectra of 36 keV X-rays scattered at 90° from polystyrene.

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