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On a low intensity ^{241}Am Compton spectrometer for measurement of electron momentum density

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Abstract. In this paper, a new design and construction of a low intensity (100 mCi) ^{241}Am γ-ray Compton spectrometer is presented. The planar spectrometer is based on a small disc source with the shortest geometry. Measurement of the momentum density of polycrystalline Al is used to evaluate the performance of the new design. The measured profile is in good agreement with the existing theoretical data and our density functional calculations.

Keywords. Compton scattering; electron momentum density; band structure calculations, γ-ray scattering.

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1. Introduction

During the last three decades, γ-ray Compton scattering has been used extensively to obtain momentum densities of a variety of materials [1]. The Compton profile, $J(p_z)$, is the projection of the electron momentum density $\rho(\vec{p})$ along the scattering vector (usually z-axis of a Cartesian coordinate system). It is defined as

$$J(p_z) = \int_{p_x} \int_{p_y} \rho(\vec{p}) \mathrm{d}p_x \, \mathrm{d}p_y. \tag{1}$$

An electron with momentum component p_z along the scattering vector shifts the scattered photon energy from E_1 to E_2, where

$$\frac{p_z}{m_0 c} = \frac{\left\{E_2 - E_1 + E_1 E_2 (1 - \cos\phi)/m_0 c^2\right\}}{\left(E_1^2 + E_2^2 - 2E_1 E_2 \cos\phi\right)^{1/2}}. \tag{2}$$

Here ϕ is the photon scattering angle. In crystalline materials, Compton data are mainly interpreted in terms of the difference between the pairs of directional profiles, i.e.

$$\Delta J(p_z) = J_{h\,k\,l}(p_z) - J_{h'\,k'\,l'}(p_z), \tag{3}$$

where $h\,k\,l$ and $h'\,k'\,l'$ denote planes perpendicular to the scattering vector. This approach eliminates the residual systematic errors such as failure of impulse approximation, bremsstrahlung contribution, multiple scattering etc. and also removes the isotropic core contribution from $J(p_z)$. Therefore, the anisotropy in the Compton profiles provides a very useful test of *ab-intio* electronic structure theories.

In spite of the instrumentation related to synchrotron radiations, there is still a need for laboratory sources. In addition to the basic requirement related to measurement of momentum densities, such sources are also useful in X-ray fluorescence, determination of the Fermi momentum, measurements of elastic and inelastic X-ray scattering cross-section etc. ^{241}Am with a principal γ-ray energy of 59.54 keV is a very convenient source because of its commercial availability, a long half-life (432.2 yr), freedom from source contamination and a clean space in the Compton profile region. Besides ^{241}Am isotope, high energy sources such as ^{198}Au (412 keV, $T_{1/2} \sim 2.7$ days) and ^{137}Cs (661.65 keV, $T_{1/2} \sim 30.1$ yr) are also in use for the study of high Z materials (see, for example, [2–4]). Due to the problems related to lower efficiency of solid state detectors at high energies, a short half-life and radiation shielding, only a limited number of high energy Compton spectrometers have been fabricated so far. The most common choice has been 1–5 Ci annular ^{241}Am sources (see, for example, refs [1,5]). The use of annular sources in the measurement of directional Compton profile from single crystals is complicated by the fact that for such an arrangement, the scattering vector ($\vec{k}$) is not a constant. The $\vec{k}$ lies anywhere on the surface of a cone of semi-angle of the order of $\frac{1}{2}(180^\circ - \phi)$. Moreover, the angular beam divergences cause the ϕ to be distributed on a family of cones with varying conic angles. The high degree of imprecision in $\vec{k}$ may be acceptable in the study of isotropic momentum densities but it imposes a serious problem in the directional measurements. To minimise the uncertainty in $\vec{k}$ which is the primary requirement for the study of anisotropy in the momentum densities, a collimated planar geometry with a small disc-type source is very helpful. In this paper, a first ever new design for planar geometry consisting of the lowest activity (100 mCi) ^{241}Am γ-ray source and drastically reduced distances with suitable collimation is presented. To establish the viability of planar geometry, test measurements have been made on a standard sample, namely polycrystalline Al. The experimental data on Al are compared with the available augmented plane wave (APW) calculations of Papanicolaou *et al* [6] and our density functional theory (DFT) based calculations which are undertaken using CRYSTAL03 code [7].

2. Experimental set-up

The layout of our planar ^{241}Am Compton spectrometer, which employs a disc source, is shown in figure 1. The 100 mCi point source ^{241}Am procured from M/s Chemotrade, Germany has the active dimension of 0.4 cm diameter and 0.12 cm length. The sample chamber made of brass (outer diameter 10.0 cm, length 30 cm, wall thickness 0.5 cm) was extended in the direction away from the source so that the walls remain out of the scattering volume. The resolution of γ-ray Compton

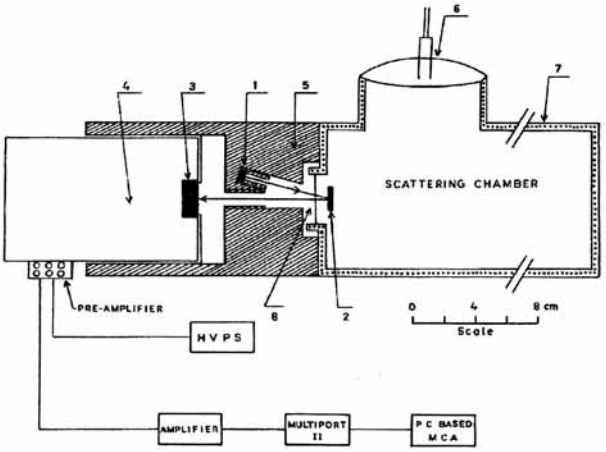

Figure 1. Layout of the first ever 100 mCi ^{241}Am Compton spectrometer. The lined region shows the lead shielding. Shown here are: 100 mCi ^{241}Am disk source (1), sample to be studied (2), HPGe detector crystal (3), HPGe detector capsule (4), lead shielding around the source and the detector (5), evacuation port (6), scattering chamber made of brass (7) and mylar foil to evacuate scattering chamber (8). The standard associated electronics (procured from Canberra, USA) is also shown at the bottom of the figure.

spectrometer consists of two parts: (a) the resolution of the detector and (b) the geometrical resolution due to beam divergences. It is defined as

$$\Delta p_z = \sqrt{\left(\left(\frac{\mathrm{d}p_z}{\mathrm{d}E_2}\right)\Delta E_2\right)^2 + \left(\left(\frac{\mathrm{d}p_z}{\mathrm{d}\phi}\right)\Delta\phi\right)^2}. \tag{4}$$

Here ΔE_2 is the energy resolution of the detector. For the present experimental set-up and other possibilities we have estimated the spread $\Delta\phi$ in the scattering angle using a Monte Carlo (MC) simulation [8] which simulates the path of the incident and scattered photons through the spectrometer. We have calculated the geometrical broadening by simulating the history of 99,999 photons for the sample size 15 mm (diameter), source size 4 mm (diameter) and different distances between the source and the sample as given in table 1. The geometrical broadening at different possible distances (table 1) is shown in figure 2. It is worth mentioning here that for the present MC calculations, we have used a computer program called GEOM developed by the University of Warwick [1, 8]. While taking the output in the program, a Gaussian distribution is approximated by chopping-off the tail and fitting a polynomial in the data points. Using eq. (4), we have computed both the differentiation $\mathrm{d}p_z/\mathrm{d}E_2$ and $\mathrm{d}p_z/\mathrm{d}\phi$ involved in the detector and the geometrical contributions to the resolution for a fixed incident energy of 60 keV and angles typical for Compton scattering. These differentiations are shown in figure 3. From figures 2 and 3, it is evident that to get the best possible resolution, the scattering angle should have the maximum possible value within the experimental geometry. From table 1 and inset of figure 3, it is evident that in the scattering range

Table 1. Distances between source and sample (s–s), sample and detector (s–d) and detector and source (d–s). The scattering angle is based on the intensity distribution of Monte Carlo simulations. Total resolution (Gaussian, full-width at half-maximum) accounts the detector and geometrical resolution as discussed in the text. In atomic units, 1 a.u. of momentum is $1.99289{\cdot}10^{-24}$ kg m s^{-1}.

Distance (cm)			Scattering angle (degree)	Resolution (a.u.)
s–s	s–d	d–s		
3.7	6.8	3.6	158.0	0.57
4.7	7.8	3.6	162.6	0.55
5.7	8.8	3.6	165.2	0.55
6.7	10.6	4.2	169.4	0.54
7.7	11.6	4.2	170.5	0.54
8.7	12.6	4.2	171.5	0.54
9.7	13.6	4.2	172.2	0.54
10.7	14.6	4.2	172.8	0.54

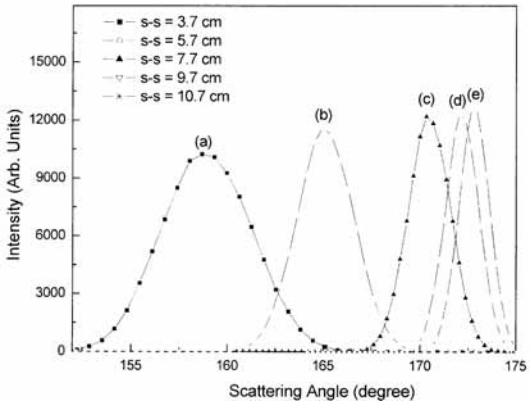

Figure 2. Monte Carlo-based geometrical divergence for typical distances between source and sample (s–s) as detailed in table 1. The present spectrometer is based on the geometrical divergence produced by the peak (b).

163–173° the overall resolution is ultimately limited by the energy resolution of detector which is set by the current state of detector technology.

The count rate C under the Compton profile is proportional to the collimator factor $\Delta\Omega_{\text{eff}}$, which is equal to $\Delta\Omega_{\text{s}} \cdot \Delta\Omega_{\text{d}}$ ($\Delta\Omega_{\text{s}}$ and $\Delta\Omega_{\text{d}}$ being solid angles subtended by the source and detector, respectively, at the sample). As shown in table 1, by increasing source–sample (s–s) and sample–detector (s–d) distances the scattering angle can be increased up to 173° or so. It may improve instrumental resolution by 0.01 a.u., in the scattering range 162–173°, which is of course negligible in such measurements. The drop in intensity while increasing the scattering angle can be calculated from the ratio of intensities as estimated using MC simulations. From

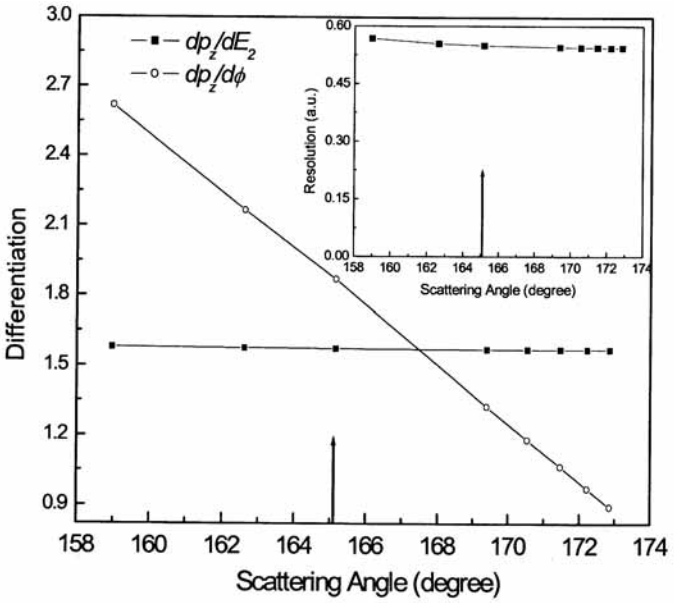

Figure 3. Plot of $\mathrm{d}p_z/\mathrm{d}E_2$ (a.u./keV) and $\mathrm{d}p_z/\mathrm{d}\phi$ (a.u./rad.) for different angles. In the inset, overall momentum resolution (computed using eq. (4)) is also shown. Detector resolution, ΔE_2, in the present set-up is 346 eV at 48.4 keV. The vertical arrow shows the scattering angle chosen in the present set-up.

the present computations (shown in figure 2), it is found that the count rate will reduce by a factor of about 2 when the scattering angle is 173° as compared to 165°. Therefore, at scattering angle 173° the count rate with the 100 mCi source may be insufficient for completion of Compton measurements in a reasonable period of time. The smallest possible distances between source and sample and source and detector will further reduce the background due to air scattering inside the collimators and also the amplitude of low-energy tail in the response function of the detector. Considering the requirement of a better resolution, higher Compton count rate, tolerance of larger divergence at the higher angles, fulfillment of impulse approximation criteria and requirements of interpretation theories, which hold more accurately at higher angles, a scattering angle of 165° was chosen. This led us to a choice of distances between source and sample and source and detector as 5.7 and 8.8 cm, respectively. In earlier conventional planar Compton spectrometers mainly employing 200–300 mCi ^{241}Am sources, the distances were almost double of our distances [1]. In the present set-up, the sample chamber has a small 25 μm thick Mylar window and can be evacuated. The resolution function of the high purity Ge detector (Canberra Model GL0510P) used in the set-up is measured with a weak 1 μCi ^{241}Am source, which could be conveniently placed in front of the detector capsule.

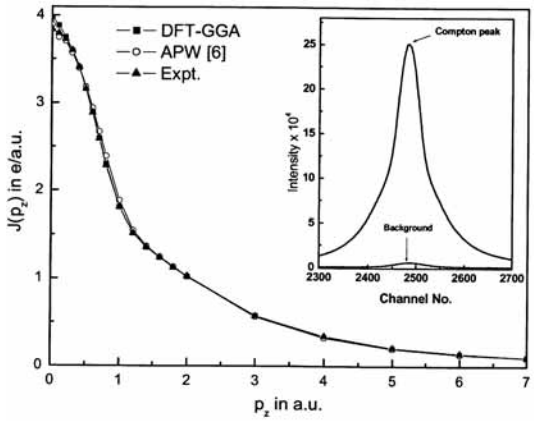

Figure 4. Duly corrected isotropic experimental Compton profile of test sample Al along with convoluted APW theory [6] and the present DFT–GGA profiles. The experimental errors are within the size of symbols used. In the inset, the raw data of Al along with the background are shown. Each data is for a period of 21.22 h.

3. Performance of the spectrometer

To check the performance of the new set-up, measurements have been made on the Compton profile of a standard sample namely Al. In case of Al, it is known that even a free electron profile provides a fairly accurate representation of the Compton profile. Therefore, the Compton profile of Al can be used as a figure-of-merit of any Compton spectrometer. For 2 mm thick Al, a reasonable count rate of 3.3 counts/s at Compton peak (20 eV or 0.019 a.u. channel width) was recorded. The raw data for the Al sample accumulated for 21.22 h along with background are shown in the inset of figure 4. Due to small distances between source and sample; and source and detector, the background contribution due to air scattering was small. To extract out the true Compton profile, first of all, the raw data were corrected for background correction. Thereafter, the profile was corrected for detector resolution (limited to stripping off the low energy tail), energy-dependent corrections like absorption in the sample and Compton scattering cross-section following the approach of Warwick group [1, 8]. The data were than converted into momentum scale and normalized to free atom profile area. To remove the effect of multiple scattering events, the Monte Carlo program of Felsteiner *et al* [9] was used. To compare the experimental Compton profile of Al with DFT calculations, we have also computed the Compton profiles of Al using CRYSTAL03 code [7]. In the present computations, the one-electron Schrödinger equations are solved self-consistently for the one-electron wave function. We have incorporated the generalised gradient approximation (GGA) to the DFT. Using the notations of CRYSTAL03 code, in the DFT–GGA scheme the one-electron equation is given as

$$\left\{T + V + C[\rho(\vec{r})] + V^{\mathrm{XC}}[\rho(\vec{r}), \nabla\rho(\vec{r})]\right\} \Psi_i = \varepsilon_i \Psi_i, \tag{5}$$

where T, V and C are operators of the kinetic energy, external potential energy (in our case potential energy of the nuclei) and classical interaction Coulomb repulsion,

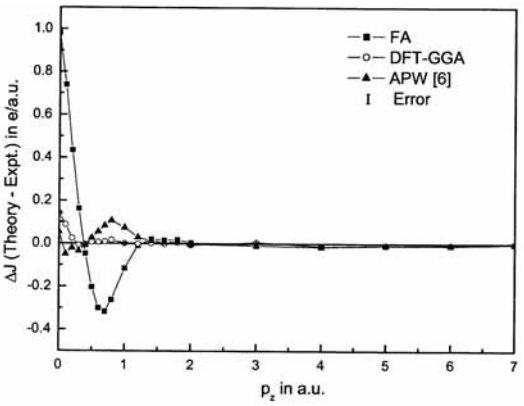

Figure 5. Difference profiles between the convoluted theories and the experiment. The experimental error ($\pm\sigma$) is also shown.

respectively. The exchange correlation potential V^{XC} is defined as a functional derivative of the exchange and correlational energy E_{XC} as

$$V^{\mathrm{XC}} = \frac{\partial E_{\mathrm{XC}}[\rho(\vec{r}), \nabla\rho(\vec{r})]}{\partial\rho(\vec{r})}. \tag{6}$$

In the present DFT–GGA scheme, the exchange of Becke [10] and correlations of Perdew–Wang [11] are considered. All-electron Gaussian basis sets of Al is used from ref. [12]. The space group and lattice parameter of Al are taken as $Fm\bar{3}m$ and 4.0496 Å [13], respectively. In the present computations, the self-consistent field calculations have been performed using 120 $\vec{k}$ points in the irreducible Brillouin zone. To compare our experimental data, we have also considered the APW calculations [6] and the free-atom Compton profiles [14]. To smear the theoretical profile with the instrumental resolution, all the profiles were convoluted with a Gaussian of FWHM 0.55 a.u. Figure 4 shows an excellent agreement between experimental and theoretical values particularly in high momentum side. In figure 5 the difference profiles (convoluted theory – experiment) are plotted using free-atom [14], our DFT–GGA and APW [6] calculations. It is seen that our DFT–GGA theory gives the best agreement with the experiment. The present comparison of experimental data with the band structure calculations on Al shows the correctness of the design of our Compton spectrometer, data analysis and its possible application in the measurement of isotropic and directional profiles.

4. Conclusions

The present work on 100 mCi Compton scattering set-up shows that by adopting a careful design it is possible to increase the effective intensity of scattered γ-ray by a factor of 2 or so. The count rate and the resolution (0.55 a.u. FWHM, Gaussian) of the Compton spectrometer are sufficient to highlight the main features of the band structure calculations of low-Z materials. The real advantage of the design

discussed here depends crucially on keeping the size of the spectrometer as small as possible and can be regarded as a novel guideline for a low-intensity planar Compton spectrometer.

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