Rayleigh scattering of 661.6-keV γ rays in Cu, Zn, Sn, and Pb at forward angles

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A method for measuring the Rayleigh scattering cross sections of γ rays at forward angles is described. The measured integral Rayleigh scattering cross sections below 7° for 661.6-keV γ rays in copper, zinc, tin, and lead are presented and compared with form-factor calculations. The present results show a good agreement with the nonrelativistic form-factor values of Hubbell *et al.* (1975) as pointed out by Kissel *et al.* (1980) and Roy *et al.* (1983). In the case of high-Z elements, the relativistic form-factor values appear to be slightly too large.

I. INTRODUCTION

The elastic scattering of γ rays by atoms is in general a coherent sum of Rayleigh scattering, nuclear Thomson scattering, nuclear resonance scattering, and Delbruck scattering.[1] The Rayleigh scattering is dominant at all scattering angles for photon energies less than 1 MeV. At higher energies the Rayleigh scattering is more and more forward peaked. Below 700 keV the other competing coherent processes, viz., nuclear Thomson, nuclear resonance, and Delbruck scattering, are negligible.[2]

Recently great progress has been made in the understanding of Rayleigh scattering, both theoretically and experimentally. However, the present knowledge of Rayleigh scattering is not completely satisfactory and efforts are being continued with more and more realistic wave functions and realistic treatments of the scattering process.[3–13] Rayleigh scattering amplitudes have been calculated on the basis of Hartree-Fock-Slater (HFS) wave functions following the computational method of (i) form factor (FF), (ii) modified form factor (MFF), and (iii) the second-order S matrix of QED. Of these, only (iii) may be considered exact; the others are approximations with limited applicability. Exact calculations of Rayleigh scattering amplitudes are extremely difficult to make.[9,10] Therefore, most of the tabulations of Rayleigh scattering amplitudes make use of approximate methods such as the FF approximation, as the integration involved is relatively simple. Hubbell *et al.*[3] have tabulated the atomic form factors $F(x,Z)$ based on the nonrelativistic Hartree-Fock (HF) wave functions following the method of Cromer and Mann,[4] for all elements from $Z=1$ to 100 over a wide range of momentum-transfer values. At higher energies and for heavy atoms, relativistic effects must be taken into account. Recently Hubbell and Overbo[5] have tabulated $F(x,Z)$ values based on the relativistic HF wave functions. The relativistic form-factor approximation neglects the binding effects of intermediate states. A modified form-factor approximation has been suggested wherein the binding of the intermediate states is taken into account in an approximate way.

Exact numerical calculations of Rayleigh scattering amplitudes have been attempted since 1954 by various workers[6–8] by developing a relativistic formalism in the second-order perturbation theory. More recently Johnson and Cheng[9] and Kissel and Pratt[10,11] have made exact numerical calculations of Rayleigh scattering amplitudes for various energies and atomic subshells of a series of elements based on the second-order S matrix and screened relativistic Dirac-Hartree-Fock-Slater (DHFS) wave functions. There are in general many subshells of the atom contributing to the scattering amplitude, and exact S-matrix calculations are extremely difficult for the higher shells. However, as has been noted by Brown and Mayers[12] and discussed in detail by Kessel *et al.*,[11] the Rayleigh amplitudes for small scattering angles may well be approximated by the MFF. And as pointed out by Kissel and Pratt[10] and Roy *et al.*,[13] the Rayleigh scattering amplitude obtained using the nonrelativistic FF approximation are also adequate for describing Rayleigh scattering at forward angles.

A number of systematic experimental studies have been carried out to study the small-angle Rayleigh scattering of γ rays by several investigators[14–26] using radioactive isotopes and (n,γ) reaction facilities at various reactor centers. Most of the studies (e.g., Refs. 17–24) at small angles have been performed using the shadow cone method. This method, although widely employed, suffers from a number of difficulties. Because of inherent difficulties in measuring absolute values of the scattering cross sections, only cross sections relative to some low-Z elements such as carbon or aluminum target were made. These approximate methods used to estimate the absolute cross sections are very likely to introduce large errors in the measured values.

Very few investigators[14,15] have employed techniques other than the shadow cone method at small angles. Hauser and Mussgnug[15] used a method employing the annihilation radiation and the coincidence technique to make measurments of differential Rayleigh scattering cross sections at small angles from 0.5 to 10°, which is feasible only in the case of annihilation radiation. Belskii and Starodubtsev,[14] on the other hand, have measured the integral Rayleigh scattering cross sections of ^{60}Co γ rays at foward angles ($<3°$) using a technique which permitted them to measure the Rayleigh scattered radiation as a small addition to the direct transmitted intensity. Their experimental setup was similar to that used in the total-attenuation cross-section measurements.

The above survey of the experimental data reveals inconsistencies and contradictory results from one study to another.[14–26] Many workers[21–25] have reported fairly good agreement with FF calculations of Rayleigh scattering cross sections at forward angles. However, it is observed by some of the investigators[14–16] that the form-factor approximation is inadequate to represent the Rayleigh scattering even at forward angles where it is expected to be adequate.[11,13] Under these circumstances the authors felt the need for reinvestigation of the Rayleigh scattering at forward angles, adopting a different experimental technique which involves minimum corrections. In this work, we describe a method for measuring the integral Rayleigh scattering cross sections of 661.6-keV γ rays in Cu, Zn, Sn, and Pb at angles less than 7°. The experimental method is somewhat similar to that of Belskii and Starodubtsev,[14] but an altogether different technique and procedure have been used for varying the scattering angle and determining the integral scattering cross sections. The method is simple and straightforward, as it involves just the measurement of the scattered intensity as a small addition to the direct transmitted beam. It is free from most of the approximations[23] used in the earlier methods in estimating the absolute cross sections. The only correction used here is the subtraction of small but inseparable Compton scattering contributions similar to other earlier methods at forward angles. The experimental results obtained are compared with the theoretical cross sections computed using the tabulated form-factor values of Hubbell *et al.*[3] and Hubbell and Overbo.[5]

II. PRINCIPLE OF THE METHOD

In the widely employed shadow cone method for measuring the Rayleigh scattering cross sections at small angles, the direct γ beam is prevented from reaching the detector by using a direct beam absorber (DBA) kept in between the source and the detector. Only the photons scattered from the ring scatterer placed around the DBA are allowed to impinge on the detector. In the present study the experimental setup used in the total-attenuation cross-section measurements is slightly modified in order to measure, for a given scatterer, (i) the transmitted intensity by eliminating as far as possible all the scattered photons and (ii) the intensity comprising the transmitted photons and the photons scattered within a cone of half-angle θ, by suitably positioning the scatterer and the collimators.

If I_1 represents the scattering-free transmitted intensity, then from the well-known attenuation relation we have

$$\mu t = \ln(I_0/I_1) , \tag{1}$$

where I_0 is the direct beam intensity, μ is the total-attenuation coefficient, and t is the thickness of the scatterer. On the other hand, for the same scatterer, if I_2 represents the transmitted plus the scattered intensity within a forward cone of half-angle θ, then the total-attenuation coefficient μ will be decreased by a small amount $\Delta\mu$ such that

$$(\mu - \Delta\mu)t = \ln(I_0/I_2) . \tag{2}$$

Here $\Delta\mu$ represents the scattered contribution within a forward cone of half-angle θ. From the above relations one can deduce an expression for $\Delta\mu$:

$$\Delta\mu = \ln(I_2/I_1)/t \tag{3}$$

in cm^{-1}. Expressing $\Delta\mu$ in cm^2/atom and writing it as $\Delta\sigma_{\text{sca}}$, we get

$$\Delta\sigma_{\text{sca}} = (A/N\rho t)\ln(I_2/I_1) , \tag{4}$$

in cm^2/atom, where A is the atomic weight, N is Avogadro's number, and ρ is the density of the scatterer. $\Delta\sigma_{\text{sca}}$ consists of both coherent and incoherent (Compton) scattering cross sections from 0 to θ as they are not separable at forward angles, i.e.,

$$\Delta\sigma_{\text{sca}} = \int_0^\theta d\sigma_{\text{coh}} + \int_0^\theta d\sigma_{\text{incoh}} . \tag{5}$$

Equation (4) is used to extract the integral scattering cross sections by measuring I_1 and I_2. Further, the integral coherent scattering cross sections are obtained by subtracting the relatively small and theoretically calculated accurate integral incoherent scattering cross sections at forward angles using the values of incoherent scattering functions.[3]

The differential coherent scattering cross sections $(d\sigma_{\text{coh}}/d\Omega)$ can also be obtained from the above study by taking the difference of any two consecutive values of $\Delta\sigma_{\text{sca}}$ and dividing it by an appropriate solid-angle factor. However, the experimental accuracy will suffer, and it is not advisable for better and meaningful comparison with the theory.

III. EXPERIMENT

A. Experimental details

The experimental arrangement used in the present study is depicted in Fig. 1. The 661.6-keV γ beam was obtained from a 10-mCi ^{137}Cs source S. It was housed in a lead collimator C. The source, in the form of a steel-welded capsule, was procured from the Isotope Division, Bhabha Atomic Research Centre, Bombay. C, C_1, C_2, and C_3 are lead collimators, each of which was 7 cm in thickness. The collimators C_2 and C_3 had collimating holes of less than 0.4 cm in diameter and were kept separated by a distance of 73 cm. Hence, when the scatterer was placed at position P_1, the angle of acceptance (determined entirely

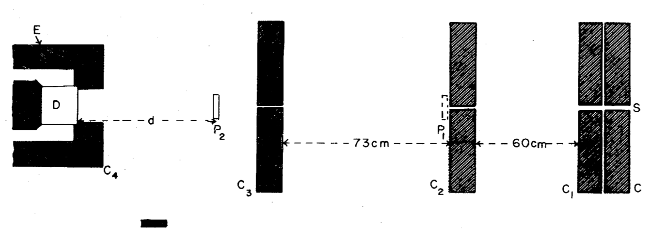

FIG. 1. Diagram of the experimental arrangement (not to scale). S is the Cs-137 source; D is the NaI(Tl) detector; C, C_1, C_2, C_3, and C_4 are lead collimators; E is the detector lead shielding; P_1 and P_2 are the scatterer positions.

by C_3) was about 0.1°, which leads to a small scattering contribution less than 0.01 b/atom in the case of lead.

The detector D, a $2''\times 1.75''$ NaI (Tl) crystal supplied by Electronics Corporation of India Ltd., Hyderabad, India, was kept in a lead housing E. It was placed at a distance of 79 cm from C_3. The collimator C_4, kept in front of the detector, had a step, 0.5 cm in depth and a diameter slightly greater than the diameter of the NaI(Tl) crystal can, cut symmetrically around its collimating hole of diameter 4.18 cm, so that the crystal can be fitted into the step groove correctly with its center coinciding with the centroid of the crystal. Copper, zinc, tin, and lead of high purity (99.9%) in the form of circular foils of uniform thickness were used as scatterers. Their thicknesses were chosen such that μt is of the order 1 to 2. The alignment of the experimental setup was done optically using a good telescope. The output of the detector was fed to a 1024-channel analyzer after suitable amplification and shaping from a linear amplifier. The experiment was performed in an air-conditioned room using a stabilized power supply. The drift in the electronic system was negligible.

B. Scattering angle

In order to define the scattering angle, it is very necessary to see that the beam is made as narrow as possible so that the area of the scatterer exposed to the beam can be treated as a point scatterer for all practical purposes. The experiment was therefore so designed using collimators C_2 and C_3 having very narrow collimating holes, to obtain a highly collimated sharp beam of photons. The angles of scattering θ can be varied by varying the angle of acceptance of the detector at the position P_2. This can be done by changing the distance d between the scatterer and the detector. In this experiment, the angle θ was varied by keeping the position of the detector fixed at a distance of 79 cm from C_3 and by moving the scatterer (position P_2) axially between C_3 and D. The separation distances between C_2, C_3, and D were so chosen such that the γ beam emerging out of C_3 was practically the same diameter up to the detector.

IV. MEASUREMENTS

The spectra were recorded by placing each of the scatterers at P_1 and at different P_2 positions. The P_2 positions were chosen to cover the angular range up to 7°. The distance from the middle of the scatterer to the front surface of the NaI(Tl) crystal was used for calculating the angle θ. The time for each spectrum was so chosen to get counts under the photopeak of the order of 10^5 or more in order to minimize the statistical error. The counts under the entire photopeak were taken to be the intensity. The spectrum obtained by placing a lead absorber of 20 cm in thickness in the path of the γ beam between C_2 and C_3, which would completely absorb the 661.6-keV γ rays, was taken as the background spectrum. This was checked by recording the spectrum upon removing the source, and it was found to be in good agreement within the experimental error. The experiment was repeated at least five times for each position and the scatterers. The background subtracted intensities I_1 and I_2 were used to obtain the integral scattering cross sections using the relation given in Sec. II for each of the P_2 positions and for the scatterers used.

V. RESULTS AND DISCUSSION

The measured cross sections are the sum of the contributions from the Rayleigh and Compton scattering because they cannot be separated in the angular region considered in this study. However, the Compton contribution is negligibly small at these forward angles. And the Compton cross sections calculated on the basis of the nonrelativistic HF incoherent scattering function approximation are found to be quite adequate in describing the experimental values.[23–25] Hence, the experimental integral Rayleigh scattering cross sections are obtained by subtracting the small and considered to be accurate theoretical integral incoherent scattering cross sections from the measured total integral scattering cross sections for each of the elements, viz., Cu, Zn, Sn, and Pb, as they will not impair the overall accuracy any further. The experimental Rayleigh cross sections thus obtained are given in Tables I and II. The experimental errors shown include the statistics of counting and that of the measurement of the thickness t of the scatterer. In addition to this, there is about a 5% error in the measurement of the angle, mainly due to the spread in the γ beam incident on the scatterer.

There is only one similar experimental study available in the literature, viz., the work of Belskii and Starodubtsev.[14] They have made measurements of the integral Rayleigh scattering cross sections of the ^{60}Co γ rays at two angles below 3°. Their experimental results are greater than the values predicted by the Debye-Franz theory. Comparison of their values with our results in the suitable momentum-transfer range shows that their values are considerably lower. Even the Debye-Franz theoretical values considered by them for comparison are very much lower than the presently available form-factor theoretical cross sections.

The integral Rayleigh cross sections calculated using

TABLE I. Comparison of the measured and calculated integral Rayleigh scattering cross sections of 661.6-keV γ rays for copper and zinc (b/atom).

Distance d (cm)	Angle θ (deg)	Copper Theor.	Copper Expt.	Zinc Theor.	Zinc Expt.
69.6	1.72	0.048	0.05±0.01	0.053	0.05±0.01
50.7	2.36	0.060	0.06±0.01	0.065	0.06±0.01
36.6	3.27	0.073	0.07±0.01	0.079	0.08±0.01

TABLE II. Comparison of the measured and calculated integral Rayleigh scattering cross sections of 661.6-keV γ rays for tin and lead (b/atom).

Distance d (cm)	Angle θ (deg)	Tin Nonrelativistic	Tin Relativistic	Tin Expt.	Lead Nonrelativistic	Lead Relativistic	Lead Expt.
69.6	1.72	0.167	0.168	0.15±0.03	0.552	0.561	0.52±0.04
50.7	2.36	0.223	0.225	0.22±0.03	0.745	0.760	0.79±0.04
36.6	3.27	0.285	0.290	0.27±0.03	0.944	0.970	0.94±0.04
26.3	4.54	0.339	0.344	0.33±0.03	1.143	1.180	1.13±0.04
19.2	6.21	0.383	0.389	0.40±0.03	1.338	1.399	1.33±0.04

the nonrelativistic coherent form factors $F(x,Z)$ of Hubbell *et al.*[4] and the relativistic $F(x,Z)$ values of Hubbell and Overbo[5] were used for comparison with the experimental Rayleigh scattering cross sections. The integral Rayleigh scattering cross section was theoretically calculated for each of the elements, viz., Cu, Zn, Sn, and Pb, by integrating numerically the product of the Thomson expression for the free electrons and the square of the $F(x,Z)$ value over the range from 0 to θ. Similarly, the theoretical integral incoherent (Compton) scattering cross section used for subtraction was calculated in each case by integrating numerically the product of the Klein-Nishina expression for free electrons and the incoherent scattering function over the range from 0 to θ. These computations were performed in a TDC 316 computer. The theoretical integral Rayleigh scattering cross sections thus obtained are given in Tables I and II along with the experimental values.

The variation of the calculated and measured integral Rayleigh scattering cross sections with the scattering angle θ is shown in Figs. 2 and 3. The solid curve represents the theory and the circles with the error bars denote the experimental Rayleigh cross sections. For the lighter elements such as Cu and Zn, both the nonrelativistic and the relativistic form-factor theory gives the same values. Therefore, only the theoretical integral Rayleigh scattering cross sections based on the nonrelativistic HF wave functions were used for comparison in the case of Cu and Zn. On the other hand, in the case of heavier elements such as Pb and Sn, the integral cross sections obtained using the nonrelativistic HF wave functions are slightly lower than those obtained using relativistic HF wave functions. Curve *a* in Fig. 3 represents the nonrelativistic form-factor theory, whereas curve *b* denotes the relativistic theory. The difference in the theoretical integral cross sections based on nonrelativistic HF and relativistic HF wave functions is appreciable only in the case of lead.

As can be observed from Tables I and II and Figs. 2

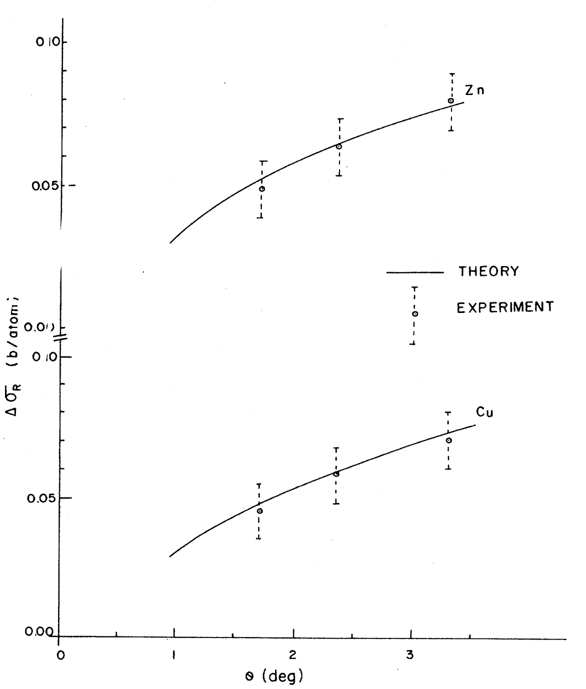

FIG. 2. Comparison of the calculated and measured integral Rayleigh scattering cross sections of 661.6-keV γ rays for copper and zinc. The solid curve represents the nonrelativistic form-factor values of Hubbell *et al.* (1975).

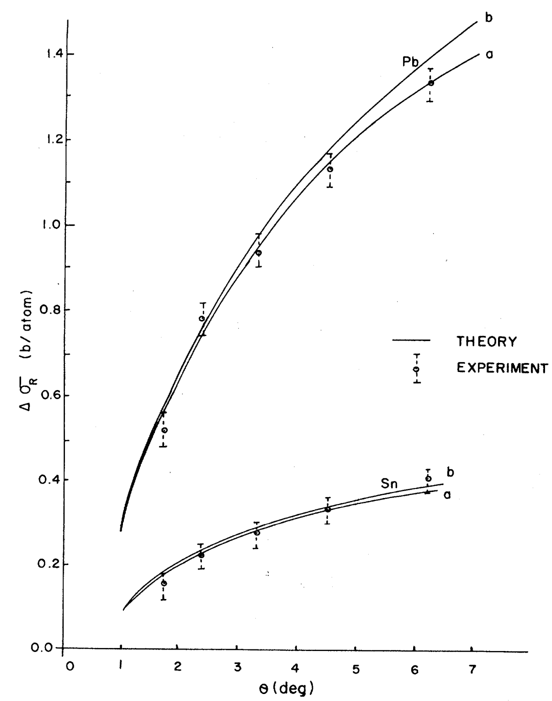

FIG. 3. Comparison of the calculated and measured integral Rayleigh scattering cross sections of 661.6-keV γ rays for tin and lead. Curve *a* represents the nonrelativistic form-factor values of Hubbell *et al.* (1975). Curve *b* represents the relativistic form-factor values of Hubbell and Overbo (1979).

and 3, the experimental integral Rayleigh scattering cross sections are in good agreement with the form-factor theory within experimental errors over the angular region considered and for all elements studied. However, in the case of lead, the experimental values are slightly lower than theoretical cross sections based on the relativistic HF wave functions. But they are in very good agreement with the nonrelativistic form-factor theory. Our observation is in agreement with the conclusions drawn by other investigators in the momentum-transfer region considered here.[20–25] We have reported the Rayleigh scattering cross sections only up to 3.27° in the case of Cu and Zn because the Compton cross sections become appreciable at angles greater than about 4° for these lighter elements.

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