Small-angle elastic scattering of ^{152}Eu and ^{154}Eu γ rays

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Differential cross sections for the elastic scattering of 344-, 411-, 444-, 723-, 779-, 868-, 964-, 1005-, 1086-, 1112-, 1275-, and 1408-keV γ rays by $_{29}$Cu, $_{42}$Mo, $_{50}$Sn, $_{73}$Ta, $_{82}$Pb, and $_{92}$U have been measured for scattering angles of 2°, 3°, 5°, 7°, and 10°. These cross sections generally agree to better than 15% with theoretical cross sections for Rayleigh plus nuclear Thomson scattering, where the Rayleigh scattering amplitudes have been represented by relativistic Hartree-Fock-Slater modified form-factor amplitudes.

I. INTRODUCTION

The elastic scattering of γ rays by atoms is the coherent superposition of Rayleigh, nuclear Thomson, Delbrück, and nuclear resonance scattering processes. The advent of high-resolution Ge(Li) detectors has stimulated several detailed experimental studies of elastic scattering of 0.1–2.0-MeV γ rays[1–12] and has been accompanied by more accurate theoretical calculations of Rayleigh scattering[13–16] and Delbrück scattering.[17,18]

For energies below a few MeV the dominant process is Rayleigh scattering and any detailed interpretation of experimental results for elastic scattering depends critically on the accuracy of the theoretical calculations for Rayleigh scattering. The most accurate calculations of Rayleigh scattering[13,15,16] are based upon the second-order S matrix in the bound-interaction picture, which allows for atomic binding effects on the initial, intermediate, and final electron states, and involves multipole expansions of the initial and final photon fields and numerical solution of inhomogeneous radial Dirac equations for each atomic subshell. These numerical partial-wave calculations require vast amounts of computer time and, consequently, have been restricted primarily to K- and L-shell scattering. These accurate calculations have made possible[19] a careful examination of the validity of the widely used form factor and modified form-factor approximations of Franz,[20] which are believed to be accurate for small momentum transfers when the photon energy is large compared to the binding energies of the atomic electrons. Such form factors can be used[15] to obtain outer-shell contributions which, when combined with the accurate inner-shell S-matrix calculations, yield total-atom Rayleigh amplitudes claimed to be accurate to $O(1\%)$.

Small-angle elastic scattering of γ rays below 2 MeV is of particular interest since in this region Rayleigh scattering is the only significant contributing process to the cross section. Furthermore, in this region there will be major contributions to Rayleigh scattering from outer atomic subshells for which only form-factor amplitudes are available, and therefore experiment can provide a direct check on the adequacy of these approximate amplitudes.

In this paper we report experimental cross sections for elastic scattering of 344-, 411-, 444-, 723-, 779-, 868-, 964-, 1005-, 1086-, 1112-, 1275-, and 1408-keV γ rays through scattering angles of 2°, 3°, 5°, 7°, and 10° by targets of $_{29}$Cu, $_{40}$Mo, $_{50}$Sn, $_{73}$Ta, $_{82}$Pb, and $_{92}$U. The results are compared with theoretical cross sections for Rayleigh plus nuclear Thomson scattering where the Rayleigh amplitudes have been obtained from existing tabulations of nonrelativistic Hartree-Fock form factors,[21] relativistic Hartree-Fock form factors,[22] and relativistic Hartree-Fock-Slater modified form factors.[23] For several elements comparisons have also been made with the recent unpublished tabulations[16] for Rayleigh plus nuclear Thomson plus Delbrück scattering where the Rayleigh amplitudes used are the numerical partial-wave S-matrix amplitudes of Kissel and Pratt for the inner-shell electrons, and, for the outer electrons, modified form-factor amplitudes for the real parts of the scattering amplitudes and inner-shell amplitudes scaled by ratios of photoeffect cross sections for the imaginary parts of the amplitudes.

II. THEORY

The differential cross section for elastic scattering of unpolarized γ rays of energy $\hbar\omega$ through an angle θ into the element of solid angle $d\Omega$ is

$$\frac{d\sigma}{d\Omega}=r_e^2\,|f(\omega,\theta)|^2\,, \tag{1}$$

where $f(\omega,\theta)$ is the total elastic scattering amplitude in units of the classical electron radius r_e. For γ-ray energies up to a few MeV, $f(\omega,\theta)$ is a coherent superposition of amplitudes for Rayleigh R, nuclear Thomson T, Delbrück D, and nuclear resonance N scattering, i.e.,

$$f=R+T+D+N\,. \tag{2}$$

If each scattering process is described in terms of circularly polarized photons, then

$$\frac{d\sigma}{d\Omega}=r_e^2[\,|f^+(\omega,\theta)|^2+|f^-(\omega,\theta)|^2]\,, \tag{3}$$

where f^+ and f^- are the amplitudes for no spin flip and spin flip, respectively, that is, for no change and change, respectively, in the state of circular polarization.

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In the form-factor and modified form-factor approximations the Rayleigh amplitudes for the nth atomic subshell are

$$R_n^{\pm} = -\tfrac{1}{2}(1 \pm \cos\theta) F_n(q) , \tag{4}$$

where F_n is either the form factor

$$F_n(q) \rightarrow f_n(q) = \int d^3x\, \psi_n^{\dagger}(\mathbf{x}) \exp(i\mathbf{q}\cdot\mathbf{x}) \psi_n(\mathbf{x}) , \tag{5}$$

or the modified form factor

$$F_n(q) \rightarrow g_n(q) = \int d^3x\, \psi_n^{\dagger}(\mathbf{x}) \times \exp(i\mathbf{q}\cdot\mathbf{x}) m_e c^2 [E_n + V(\mathbf{x})]^{-1} \psi_n(\mathbf{x}) . \tag{6}$$

E_n is the energy of the atomic electron in state $\psi_n(\mathbf{x})$, V is its potential energy, and

$$\hbar q = (2\hbar\omega/c)\sin(\theta/2) \tag{7}$$

is the momentum transfer from the photon to the atom.

Nonrelativistic Hartree-Fock total-atom f form factors have been tabulated by Hubbell *et al.*[21] For the relativistic case, analytical results for f_n and g_n for a point Coulomb potential have been obtained by Smend and Schumacher,[24] whereas numerical tabulations are available for Dirac-Hartree-Fock total-atom f form factors[22] and for Dirac-Hartree-Fock-Slater (DHFS) total-atom g form factors.[23] A computer program[25] is now available to compute DHFS individual subshell and total-atom f and g form factors. The nonrelativistic form-factor and relativistic modified form-factor results agree well with numerical second-order S-matrix results[15,19] for small momentum transfers $x < 10$ Å^{-1}, where

$$x = \lambda^{-1}\sin(\theta/2) = \frac{1}{12.3}\hbar\omega(\text{keV})\sin(\theta/2)\ \text{Å}^{-1} . \tag{8}$$

In contrast, there is poorer agreement for the relativistic form-factor results.

The amplitudes for nuclear Thomson scattering from a nucleus of mass M and charge Z are independent of photon energy and are given by

$$T^{\pm} = -\left[\frac{Z^2 m_e}{2M}\right](1 \pm \cos\theta) , \tag{9}$$

and therefore have the same angular dependence as the form-factor Rayleigh amplitudes. The maximum (zero-angle) nuclear Thomson contribution T^+ ranges from -0.0034 for Al to -0.0196 for U and is therefore relatively insignificant in comparison with the zero-angle Rayleigh amplitude $R^+(\omega,0) \approx -Z$.

For γ-ray energies below $2m_ec^2$, the Delbrück amplitudes are real and small and, for small scattering angles, are given accurately by[17]

$$D^{\pm}(\omega,\theta) = \frac{(\alpha Z)^2}{2}\left[\frac{\hbar\omega}{m_e c^2}\right]^2 \frac{a^{\pm}}{32\times 72}(1 \pm \cos\theta) ,$$
$$a^+ = 73, \quad a^- = 45 . \tag{10}$$

These amplitudes are much smaller than the nuclear Thomson amplitudes, ranging from 0.001 for Al to 0.055 for U in the case of 1005-keV γ rays. The exact calculations of Papatzacos and Mork[17] decrease even more strongly with θ than (10) and, consequently, we can ignore Delbrück scattering for γ-ray energies below $2m_ec^2$. For energies above $2m_ec^2$, the largest contribution from Delbrück scattering in our investigation will occur for the 1408-keV γ ray. The significant amplitude $D^+(\omega,0)$ can be estimated from the 1.33-MeV amplitudes of Papatzacos and Mork[17] and is

$$D^+(1.33\ \text{MeV},0) = 0.241(\alpha Z)^2 . \tag{11}$$

Hence D^+ is of opposite sign to T^+ and ranges from $-0.6T^+$ for Al to $-5.6T^+$ for U. Amplitudes at 2°, 5°, and 10° were interpolated from the tables of Bar-Noy and Kahane[18] and were found to be significant at 10° and for high Z.

Nuclear resonance scattering arises from excitation of the giant-dipole resonance which can be represented[26] by a Lorentzian, or a superposition of two Lorentzians, centered around $\approx 80A^{-1/3}$ MeV and with width(s) of ≈ 3–5 MeV. This nuclear resonance scattering is negligible for γ-ray energies considered here and will be ignored.

The total differential cross section for Rayleigh plus nuclear Thomson scattering, when the form-factor amplitudes (4) are used for Rayleigh scattering, is

$$\frac{d\sigma}{d\Omega} = \tfrac{1}{2}r_e^2(1+\cos^2\theta)\left[\sum_n F_n + \left[\frac{Z^2 m_e}{M}\right]\right]^2 . \tag{12}$$

These theoretical cross sections (12) have been computed for the various form-factor models using cubic spline interpolation of the tabulated Rayleigh amplitudes.[21–23] The ratio of cross sections using relativistic form factors[22] to those using relativistic modified form factors (RMFF) (Ref. 23) increased smoothly with x, the ratio increasing from near unity at small x to values ranging from 1.1 ($Z=29$) to 1.5 ($Z=92$) at $x=10$ Å^{-1}. Cross sections obtained using nonrelativistic form factors[21] generally differed by less than 10% from the RMFF cross sections, but the variation with x was uneven, a result of unreliable interpolation based upon the coarse grid of the Hubbel *et al.*[21] tabulations and anomalous oscillations.[19]

For the case of 1408-keV γ rays, inclusion of a contribution from Delbrück scattering reduces the total cross sections, the decrease ranging from less than 1% for the lighter elements at 2° to a maximum of $\sim$7% for $_{82}$Pb and $_{92}$U at 10°.

The cross sections (12) computed using RMFF Rayleigh amplitudes have been compared with the new tabulations[16] of cross sections for Rayleigh plus nuclear Thomson plus Delbrück scattering based upon more extensive numerical partial-wave calculations of Rayleigh amplitudes. The agreement between the two sets of cross sections is always better than 5% for the small values of x ($x \lesssim 10$Å^{-1}) considered in the present investigations. The difference is therefore only of the order of the errors arising from the use of cubic splines to interpolate these tabulations to the ^{152}Eu and ^{154}Eu energies.

III. EXPERIMENTAL DETAILS

The γ rays from a 0.5-Ci source containing ^{152}Eu and ^{154}Eu were scattered into a Ge(Li) detector which had a resolution of 2.5 keV full width at half maximum (FWHM) at 1330 keV. At each scattering angle, experimental runs were done with thick and thin targets of each element studied in order to check for multiple-scattering effects, with a carbon target to obtain a Compton line shape, and with a 0.1-mCi source containing ^{152}Eu and ^{154}Eu placed at the target position to obtain the elastic line shape. These runs were interspersed with no-target runs to obtain background spectra. All spectra were recorded by a 4096-channel plus-height analyzer.

At the small scattering angles used in this investigation the elastic scattering peak and Compton peak overlap and the relative intensities $I_{\text{el}}(\omega,Z,\theta,t)$ and $I_{\text{Compton}}(\omega,Z,\theta,t)$ for scattering of a photon of incident energy $\hbar\omega$ through an angle θ from a target of atomic number Z and thickness t were extracted using a least-squares method based upon the experimentally measured Compton and elastic line shapes. The differential cross sections are then given by

$$\frac{d\sigma(\omega,Z,\theta)}{d\Omega}=\frac{I(\omega,Z,\theta,t)}{G(S\text{-}T)G(T\text{-}D)N(\omega)\epsilon(\omega')n_Z T(\omega,Z,\theta,t)}\,, \tag{13}$$

where $G(S\text{-}T)$ and $G(T\text{-}D)$ are source-target and target-detector geometrical factors, $N(\omega)$ is the source strength of γ rays of energy $\hbar\omega$ after correction for radioactive decay, $\epsilon(\omega')$ is the detector photopeak efficiency at the scattered photon energy $\hbar\omega'$, n_Z is the atomic density of the target, the $T(\omega,Z,\theta,t)$ is the target transmission factor. The ratio of elastic cross sections at two different scattering angles is therefore

$$\frac{d\sigma_{\text{el}}(\omega,Z,\theta_2)}{d\sigma_{\text{el}}(\omega,Z,\theta_1)}=\frac{T(\omega,Z,\theta_1,t)}{T(\omega,Z,\theta_2,t)}\frac{I_{\text{el}}(\omega,Z,\theta_2,t)}{I_{\text{el}}(\omega,Z,\theta_1,t)}\,. \tag{14}$$

Equation (14) must be multiplied by various geometrical factors if different source-target-detector geometries are used at the two scattering angles. From Eq. (13) the ratio of elastic to Compton cross sections is[3,12]

$$\frac{d\sigma_{\text{el}}(\omega,Z,\theta)}{d\sigma_{\text{Compton}}(\omega,Z',\theta)}=\frac{I_{\text{el}}(\omega,Z,\theta,t)}{I_{\text{Compton}}(\omega,Z',\theta,t)}\times\frac{T(\omega,Z',\theta,t)}{T(\omega,Z,\theta,t)}\frac{n_{Z'}}{n_Z}\frac{\epsilon(\omega^{\text{Compton}})}{\epsilon(\omega)}\,, \tag{15}$$

where $\hbar\omega^{\text{Compton}}$ is the energy of the γ ray after Compton scattering.

Absolute elastic cross sections were obtained from (15) by using theoretical Compton cross sections for a light element Z' (in our case carbon) where the relationship

$$\frac{d\sigma_{\text{Compton}}(\omega,Z',\theta)}{d\Omega}=S(\omega,Z',\theta)\frac{d\sigma_{\text{KN}}(\omega,\theta)}{d\Omega} \tag{16}$$

between the cross section for Compton scattering by bound electrons and the Klein-Nishina cross section $d\sigma_{\text{KN}}/d\Omega$ for Compton scattering from a free electron is believed to be valid. The incoherent scattering function $S(\omega,Z',\theta)$ is a measure of the probability that the atomic electrons can undergo transitions after absorbing the change in photon energy $\hbar(\omega-\omega^{\text{Compton}})$ resulting from the photon scattering through an angle θ as if the electrons were free. Extensive tabulations of $S(\omega,Z',\theta)$ are given in Hubbell *et al.*[21] For $Z'=6$ the minimum value of S was $0.82Z$ for 3° scattering of 344-keV γ rays and hence the approximation (16) should be accurate. Experimental values for relative Compton cross sections for the light element Z' at different energies for a given scattering angle were obtained from

$$\frac{d\sigma_{\text{Compton}}(\omega_1,Z',\theta)}{d\sigma_{\text{Compton}}(\omega_2,Z',\theta)}=\frac{I_{\text{Compton}}(\omega_1,Z',\theta,t)}{I_{\text{Compton}}(\omega_2,Z',\theta,t)}\times\frac{T(\omega_2,Z',\theta,t)}{T(\omega_1,Z',\theta,t)}\times\frac{N(\omega_2)\epsilon(\omega_2^{\text{Compton}})}{N(\omega_1)\epsilon(\omega_1^{\text{Compton}})}\,. \tag{17}$$

The efficiencies $\epsilon(\omega^{\text{Compton}})$ for the various energies were interpolated from the efficiencies at the source energies which were obtained by comparing the intensities in the direct source spectrum from the 0.1-mCi source with a given source strengths.[27] The energy dependence obtained agreed very closely with the theoretical dependence predicted by (16), thus permitting absolute elastic cross sections to be calculated from (15) by either normalizing to a carbon Compton cross section at each energy or at one particular energy.

The absolute elastic cross sections reported in this paper were obtained by normalizing to the theoretical carbon Compton cross section at 7°. Absolute cross sections obtained by normalizing to carbon Compton cross sections at 3°, 5°, and 10° differed by a maximum of 9% from the cross sections based upon 7°. As a further check the procedure was repeated using aluminum Compton cross sections at 3°, 5°, and 7° which yielded elastic cross sections differing from those based upon carbon by about 10%.

The rapid decrease in the elastic scattering cross sections at small scattering angles requires small angular acceptances and accurate determination of the scattering angle if accurate cross sections are to be obtained. At 2° the horizontal and vertical angular acceptances were 0.46° and 0.05°, respectively, and the average theoretical cross section over the detector face corresponded to a scattering angle of 2.025°. Thus no application of angular acceptance corrections was necessary. The zero angle, and hence each scattering angle, was determined to ±0.1° by finding the position of maximum count rate for a no-target run and then checking this setting by measuring the peak positions of the strong γ lines Compton-scattered through 15° and comparing these energies with the Compton formula for $\hbar\omega^{\text{Compton}}$.

IV. RESULTS AND DISCUSSION

The measured cross sections for elastic scattering of γ rays with energies ranging from 344 to 1408 keV through scattering angles of 2°, 3°, 5°, 7°, and 10° by targets of $_{29}$Cu, $_{40}$Mo, $_{50}$Sn, $_{73}$Ta, $_{82}$Pb, and $_{92}$U are shown in Fig. 1

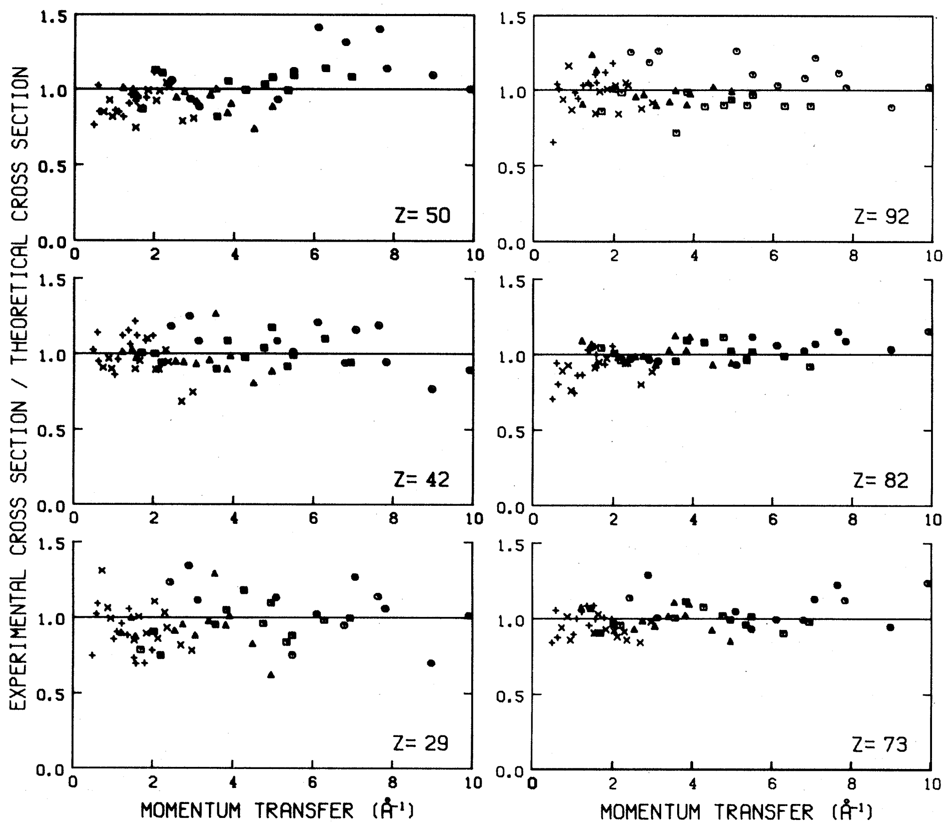

FIG. 1. Ratio of measured differential cross section to theoretical RMFF cross sections for different atomic targets as a function of momentum transfer x. The various scattering angles are denoted as follows: $+$, 2°; $\times$, 3°; $\triangle$, 5°; $\square$, 7°; and $\circ$, 10°.

as the ratios of the experimental cross sections to the theoretical cross sections (12) for Rayleigh plus nuclear Thomson scattering in which RMFF Rayleigh amplitudes have been used. The experimental results are in agreement with the theoretical calculations, although there is significant scatter of $\lesssim 15\%$, the scatter being the greatest for the lightest element (Cu). This scatter arises from the combined effects of pure statistical error, which is $\lesssim 5\%$ for an isolated peak, and the least-squares curve-fitting errors which, for overlapping elastic and Compton peaks, are sensitive to channel shifts introduced to compensate for amplifier drifts during each run and which can be as large as 15%. Other sources of error are associated with angle determination, attenuation coefficients used in the evaluation of target transmission factors, geometry factors resulting from different source-target-detector geometries, and normalization using carbon theoretical Compton cross sections, but all these errors are expected to be systematic more than statistical.

Several workers[4,6–9,12] have investigated the small-angle elastic scattering of γ rays, but only Ramanathan *et al.*[4] have used the same γ-ray energies. The ratios of the present experimental results to those of Ramanathan *et al.* for a given target material vary with γ-ray energy and scattering angle, the variation being greatest for Cu. The ratio averaged over energy and angle has the values 0.99 ($_{29}$Cu), 1.06 ($_{73}$Ta), and 1.13 ($_{82}$Pb), indicating that, in general, the present results lie above those of Ramanathan *et al.* Interpolation of the present results to the energies of 1173 and 1333 keV studied by Kane *et al.*[12] and 468 keV studied by de Barros *et al.*[7] indicated agreement in nearly all cases to within 10%, the exceptions being 10° scattering of 468-keV γ rays in $_{82}$Pb and $_{50}$Sn, where the present results are $\lesssim 30\%$ lower than those of de Barros *et al.*, and for 5° and 10° scattering of 1332-keV γ rays by Cu, where the present results are 20–25% below those of Kane *et al.* Finally, a qualitative comparison with the 317-keV results of de Barros *et al.*[8] indicates that the present 344-keV results lie smoothly below the 317-keV results at each angle as expected.

The present results for Pb are significantly lower than the previously published results of Chitwattanagorn *et al.*[10] for many energies and angles, but in all cases are in much better agreement with the theoretical cross sections than the previous results. Most of the discrepancies have been traced back to the lower resolution (4.5 keV) of the Ge(Li) detector used in that investigation which prevented separation of the elastic and Compton components at small angles for the $_{11}$Al target and hence affected the normalization procedure used to obtain cross sections.

We conclude that there is no clear disagreement between the present experimental results and theoretical cross sections computed using either RMFF or numerical partial-wave Rayleigh amplitudes for elastic scattering of γ rays in which the momentum transfer x is less than 10 Å^{-1}. Further reduction in the scatter of the experimental results is necessary before any inadequacies in the theoretical calculations will be apparent.

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