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Photoelectric interaction below the K edge

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Total photon cross sections are measured, using the transmission method in the heavy elements U, Th, Pb, and Au at energies 30.9, 35.9, and 55.4 keV, to study the photoelectric interaction below the K edges of these elements. A krypton-filled proportional counter with a 512-channel analyzer is used as the detector of photons. The photoelectric cross sections, obtained by subtracting the small scattering contributions from the total cross section, are compared with theoretical predictions of Scofield and of Storm and Israel. General agreement is obtained, except for U and Th at 30.9 keV where the present experimental values show a slight preference to the calculations of Storm and Israel rather than the theoretical ones used by Scofield.

INTRODUCTION

Of the possible types of interactions between photons and atoms, the photoelectric effect, scattering, and pair creation are prominent among others over a wide range of energies. The lower the energy and the heavier the atom, the more predominant is the photoelectric effect. Above the K edge the K-shell photoeffect comprises more than 80% of the total interaction, especially in heavy elements. There have been extensive theoretical and experimental studies on this effect,[1] culminating in what may be called a "good harmony" between theory and experiment to a large extent.

The situation below the K edge with regard to the L-shell and higher-shell photoelectric effect is, however, far from satisfactory. While there are relatively few experimental studies on these aspects, until recently the theoretical studies have been unsatisfactory because of the complexity of screening effects for L- and higher-shell electrons. The advent of modern high-speed computers, which made possible mathematical computations heretofore impossible, has stimulated considerable interest in theoretical research in recent years. As a result, there are now available extensive tabulations of accurate L- and higher-shell photoelectric cross sections computed using sophisticated theoretical models. Although this development started a matching reaction on the experimental side, there have been some notable discrepancies among the recent experimental results of different investigators, particularly for heavy elements at such photon energies where the K-shell effect is ruled out and L and higher shells above contribute to the total photoelectric cross section.

The present work is thus motivated by the aim of carrying out a systematic investigation on the photoelectric effect in the region of low photon energies, using heavy elements, to clear up the discrepancies, as well as to contribute to the much needed experimental data on low-energy photoelectric cross sections.

With this end in view, four heavy elements U, Th, Pb, and Au and three photon energies 30.9, 35.9 and 55.4 keV were selected, the latter corresponding to the K x rays of radioactive sources ^{133}Ba, ^{141}Ce, and $^{175+181}$Hf, respectively. In all these cases, the K-shell photoelectric effect is energetically ruled out, while the L-shell photoeffect contributes a major share to the total atomic cross section; the other minor contributions arise from the photoeffect in the M, N, and O shells and the coherent and incoherent scattering at these low energies. The total scattering cross section is, however, only a small fraction of the total atomic cross section, and this allows the photoelectric cross section to be deduced with almost the same accuracy as the total cross section.

Taking advantage of this fact, the total atomic cross sections are first determined for the elements and photons, using the transmission method, a modified narrow beam geometry and a proportional counter system equipped with a Nuclear Data 512 channel analyzer. The coherent and incoherent scattering contributions, which are small and theoretically computed, are then subtracted from the total cross sections to obtain the respective photoelectric cross sections for all the elements and energies studied in the present work. The results are compared with the more recent theoretical and experimental data to draw conclusions on the photoelectric interaction below the K edge.

EXPERIMENTAL DETAILS

Among the various methods for determining the photoelectric cross sections, that of obtaining them from total cross section by subtracting all other contributions is the best suited to low energies where the photoelectric effect dominates the interaction. This so called "subtraction method" is not only simple but also capable of yielding the best accuracy which is essentially decided by the statistics of counting. As it is easy to realize high accuracy using the multichannel analyzer in the cross-section measurement for the elements and energies used in the present work, this method is adopted in preference to others. To detect photons, a krypton-filled proportional counter is employed which has a resolution of about 10% at 14.4 keV. It has a side entrance window of 1 in. diameter, covered by a 0.01-in.-thick beryllium foil. The proportional counter spectrometer consisted of a solid-state charge-sensitive head amplifier, main amplifier, and a Nuclear Data 512 channel analyzer equipped with a Hewlett Packard 1208B display, 7004B X-Y recorder, and an IBM selectric typewriter attached with an autofinger. The usual narrow-beam geometry is modified to include slightly larger solid angles to compensate for the low efficiency of the proportional counter. However, appropriate corrections are applied for the small-angle coherent and incoherent scattering contributions. Uniform circular foils of $\frac{1}{2}$ in. diameter with thicknesses ranging from 10-100 mg/cm^2 are used as absorbers. The foils used in the experiment are stored with a grease layer so as to minimize the high surface oxidization of uranium and thorium. However, the foils are cleaned repeatedly with alcohol before use.

The experimental procedure simply consisted in taking a series of transmission spectra with the multichannel analyzer, progressively increasing the absorber thickness. To analyze the transmitted spectrum a computer program is utilized to correct for a polynomially fitted background and to extract the intensity of the K x-ray peak of interest by appropriate Gaussian fitting. The mass attenuation coefficient of each element at each photon energy is then determined by a least-squares fitting, and the total atomic cross section is extracted with the corresponding standard deviation.

DISCUSSION OF THE ERRORS

The overall experimental accuracy is mainly determined by the statistics of counting and the uniformity of the absorber foils used. At the energy 55.4 keV, however, an additional uncertainty arises owing to the use of a mixed source of $^{175+181}$Hf. In the present measurement the statistical error was limited to 0.5% by collecting sufficient number of counts in the photopeak. The percentage error due to the nonuniformity of the absorber foils was estimated as $\frac{1}{2}(\Delta t)^2$, where (Δt) is the relative fractional variation of the thickness. For thicker foils, this error was negligible. In the case of thinner foils, the relative variation in the thickness was investigated by scanning the foil with a finely collimated beam of 662-keV γ rays and the percentage error was found to lie between (0.5–1)%. Furthermore, only the central portion of the foil, as defined by the defining slits, was irradiated in the experiment, and its thickness was measured by cutting out that portion and carefully weighing it with an electrical balance. Thus the overall experimental error on the cross sections at the energies 30.9 and 35.9 keV is expected to lie between 1 and 1.5%.

However, at the energy 55.4 keV, additional error was anticipated due to the use of a mixed source, $^{175+181}$Hf, which emits a combination of Lu and Ta x-rays at 52.965, 54.070, 56.277, and 57.532 keV, unresolved by the proportional counter. From the relative strengths of the two isotopes in the source (as specified by the suppliers) and using the relative intensities of the $K\alpha$ and $K\beta$ lines of Lu and Ta, an effective energy of 55.4 keV was estimated for the combined peak. However, as a result of the distorted shape of the photopeak, the Gaussian fitting routines could result in a slightly different centroid. Thus the experimental errors on the cross sections at this energy were placed around 3%.

RESULTS AND DISCUSSION

The total atomic cross sections measured in the present work are compared with the results of earlier investigation in Table I. In this connection, it may be pointed out that most of the earlier work, including the recent systematic investigations of McCrary *et al.*,[2] were conducted using bremsstrahlung radiation from x-ray tubes at selected photon energies, not identical with those of the present work. Although Wiedenbeck[3] and Perkin and Douglas[4] have used radioactive x-ray sources, the x-ray energies selected by them hardly coincide with those used in the present work. In this sense, the present experimental results, which consist of 12 data points (the total cross sections of U, Th, Pb, and Au at 30.9, 35.9, and 55.4 keV energy) may be considered as a new contribution to the pool of experimental data badly needed in this low-energy region. For the purpose of comparison with the present results, the experimental results of previous investigators are interpolated

TABLE I. Experimental total cross sections (in barns per atom).

Energy (keV)	Uranium Present results	Uranium Previous results	Thorium Present results	Thorium Previous results	Lead Present results	Lead Previous results	Gold Present results	Gold Previous results
30.9	14 480 ± 280	15 230 ± 100 [a]	13 240 ± 170	13 570 ± 100 [a]	9430 ± 150	9940 ± 70 [a]	8180 ± 150	8410 ± 30 [a]
		14 290 ± 430 [b]		13 050 ± 260 [c]		9220 ± 180 [c]		8180 ± 160 [c]
35.9	10 200 ± 150	10 380 ± 60 [a]	9210 ± 190	9440 ± 50 [a]	6500 ± 130	6800 ± 40 [a]	5660 ± 100	5660 ± 20 [a]
		9890 ± 300 [b]		9240 ± 180 [c]		6450 ± 130 [c]		5710 ± 110 [c]
55.4	3340 ± 80	3370 ± 15 [a]	3030 ± 110	3020 ± 10 [a]	2060 ± 50	2070 ± 10 [a]	1800 ± 40	1760 ± 10 [a]
		3270 ± 100 [b]		3020 ± 60 [c]		2020 ± 40 [c]		1800 ± 40 [c]

[a] McCrary *et al.*, Ref. 2.
[b] Perkin and Douglas, Ref. 4.
[c] Wiedenbeck, Ref. 3.

with respect to energy in order to obtain values corresponding to the present energies for each common element studied. The errors shown on these values are also obtained by suitable interpolation.

It can be observed from Table I that there are notable discrepancies at 30.9 and 35.9 keV between the two sets of earlier measurements. It can be seen that the present results generally follow the trend of the results of Wiedenbeck[3] and Perkin and Douglas,[4] who also employed radioactive x-ray sources as in the present work. In the majority of the cases, the values of McCrary *et al.*[2] are higher than all other results including the present ones. It appears that the cross sections obtained by them using continuous x-ray sources are slightly overestimated.

As mentioned earlier, the coherent and incoherent scattering cross sections (interpolated from the tables of Storm and Israel[5]) are subtracted from the measured total atomic cross sections, to yield the respective photoelectric cross sections for the four elements at the three photon energies. As the subtracted total scattering contribution is limited to values between 4% and 15%, the extracted photoelectric cross sections are nearly as accurate as the measured total cross sections, i.e., to within (1–1.5)%. Since all the photon energies are below the K edges of all the elements used in the present work, the atomic photoeffect comprises that due to L, M, N, and O shells in each element. In Table II, these fairly accurate photoelectric cross sections are compared with the more recent theoretical predictions of Scofield[6] and of Storm and Israel.[5]

It can be noted from Table II that the two theoretical values themselves show some discrepancies up to 2%, particularly for the lowest energies and heaviest elements. Of course a part of the discrepancy, about 0.5%, may be due to errors in the interpolations used in obtaining these values at the respective energies. Owing to the fact that the experimental error on the presently measured cross sections is also of the order of (1.5–3)%, a general agreement can be noticed between present results and both the theoretical values in all cases except for uranium and thorium at the lowest energy, 30.9 keV. In both of these cases, the present values are a few percent smaller than both the theoretical values, but appear to be closer to those of Storm and Israel.[5] There are two significant differences in the theoretical calculations of Scofield[6] on the one hand and of Storm and Israel[5] on the other. Although both employed the relativistic Hartree-Slater potential, Storm and Israel used a Slater Exchange potential which is about $\frac{2}{3}$ that used by Scofield.

TABLE II. Experimental and theoretical photoelectric cross sections.

Energy (keV)	Uranium Expt.	Uranium Theory	Thorium Expt.	Thorium Theory	Lead Expt.	Lead Theory	Gold Expt.	Gold Theory
30.9	13 580 ± 280	14 503 [a] 14 268 [b]	12 820 ± 170	13 307 [a] 13 142 [b]	8940 ± 150	9179 [a] 9045 [b]	7740 ± 150	7890 [a] 7774 [b]
35.9	9680 ± 150	9809 [a] 9648 [b]	8700 ± 190	8982 [a] 8858 [b]	6100 ± 130	6141 [a] 6058 [b]	5290 ± 100	5268 [a] 5180 [b]
55.4	3050 ± 80	3113 [a] 3054 [b]	2750 ± 110	2836 [a] 2783 [b]	1840 ± 50	1897 [a] 1863 [b]	1600 ± 40	1613 [a] 1586 [b]

[a] J. H. Scofield, Ref. 6.
[b] E. Storm and H. I. Israel, Ref. 5.

Furthermore, they introduced experimental binding energies in the place of theoretical ones used by Scofield. The present experimental values show a slight preference towards the predictions of Storm and Israel, at least for heavy elements at low photon energies.

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