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MEASURED 1 TO 40 keV PHOTOABSORPTION CROSS
SECTIONS FOR: Fe, Ni, Sn, Ta, Pt, Au, Pb, U

Nancy Kerr Del Grande

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Measured 1 to 40 keV photoabsorption cross sections for: Fe, Ni, Sn, Ta, Pt, Au, Pb, U

Nancy Kerr Del Grande

University of California, Lawrence Livermore National Laboratory
P. O. Box 808, Mail Stop L-45, Livermore, California, 94550

Abstract

Total absolute photoabsorption cross sections were measured at 1 keV to 40 keV for Fe, Ni, Sn, Ta, Pt, Au, Pb and U. Overall measurement uncertainties were 3%, less than 2% of which were statistical. The measurements tested widely-used theoretical and experimental tables. They favored relativistic Hartree-Slater atomic calculations. Agreement with theory was typically 5% or better, except near the absorption edge energy region with EXAFS peaks. The measured peak and valley magnitudes for the solid samples ranged from 0% to 20% (with an average of 10%) above the atomic calculations for Fe and Ni. Discrepancies with experimental tables, at energies below 6 keV, were 10% to 40%. Authors of widely-used tables have been fraught with problems where there were few or no prior measurements, only interpolations and theory. The present measurements used samples from a few hundred to a few thousand $\mu\text{g}/\text{cm}^2$, such that the mean free path thicknesses, μpx , were 1 and 3 at 1.5 keV. Samples were vapor-deposited on, and floated off of, optically flat glass slides. Their weights per unit area, $\mu\text{g}/\text{cm}^2$, were determined within 2% or 3%. Thicker samples, of known weights per unit area, were used for the higher energy measurements. The ratio of μpx for thick and thin samples tested μx for the thin samples. Back-scattered protons and alpha particles, as well as ion microanalyses, were used to test the samples' purity. Uranium deposits were sandwiched between two beryllium layers to impede surface oxidation. The transmission measurements were taken with a demountable x-ray tube, vacuum crystal monochromator and flow proportional counter, using slits that provided a narrow-beam geometry.

Introduction

X-ray optics instrumental design is largely dependent on the use of optical constants. Optical constants are derived from photoabsorption cross sections. In the soft x-ray region (below 10 keV) photoabsorption is the dominant fundamental process by which photons interact with matter. Our knowledge of soft x-ray photoabsorption is extremely poor. This puts a crack in the foundation of x-ray optics.

Designers of x-ray optics instrumentation depend largely on several widely used tabulations of soft x-ray photoabsorption cross sections (e.g., McMaster et al.,¹ Henke et al.,² Veigele,³ Scofield,⁴ Storm and Israel⁵). There are notable differences in the values given by these tables, particularly at energies between 1 and 10 keV. For elements considered in this study, these differences range from 20 to 60 percent. It is important to clear this up. The problems are both experimental and theoretical.

On the experimental side, we have the unpleasant situation that there exist significant discrepancies between experiments from different laboratories.^{6,7} On the theoretical front, we are finding that the interplay of relativistic and many-body effects can give cross sections which differ from the central-field model results.⁷⁻¹² The goal of this investigation was to measure total absolute photoabsorption cross sections at 1 keV to 40 keV for Fe, Ni, Sn, Ta, Pt, Au, Pb and U. The measurements could then be used to test the widely-used theoretical and experimental tables.

X-ray absorption cross sections have a host of important applications besides the design of x-ray optics instrumentation. Among the defense applications are radiation protection and radiation damage for men and materials, modeling the propagation and effects of an x-ray laser beam in the atmosphere, modeling the effects of a nuclear explosion and for possible new isotope separation methods. In addition, there is interest in x-ray diagnostics of fusion plasmas,¹³ ion-atom collisions and astronomical observations. Finally, it must be emphasized that photoabsorption is one of the most basic physical processes and, thus, an understanding of photoabsorption (which requires experimental photoabsorption data) is a key to understanding the behavior of atoms, ions, molecules and solids.

Discussion of experiments

The instrumentation used to measure the total absolute photoabsorption cross sections (shown in Table 1 as total attenuation coefficients) was a demountable x-ray tube, a vacuum

single crystal monochromator, and a flow-proportional counter.⁶ Statistical fluctuations in x-ray attenuation coefficients ranged from 0.5 to 2%. The overall accuracy was 3% because of the uncertainty in determining the sample weight-per-unit area, ρx (g/cm²), for the thinnest samples.

Table I. 3% Measured Attenuation Coefficients, μ (cm²/g)

E (keV)		Fe	Ni	Sn	Ta	Pt	Au	Pb	U
1.000		8674	L	8610		4634	4663		N
1.4867	Al K α	3677	4686	3677	1836	2152	2208	2592	3639
1.580		3802	4436	3586	1748	2110	2147	2517	3459
2.000		1715	2187	1812	M	1069	1110	1326	1902
3.000		643.3	736.0	644.7	1791	M	M	M	132.9
4.000		258.3	331.6	L	948.0	1119	1164	1272	M
5.000		136.1	177.6	986.1	562.0	644.9	672.6	749.0	M
5.4147	Cr K α_1	111.7	143.6	718.4	471.3	540.9	557.3	622.1	M
5.9467	Cr K β_1	87.72	111.2	558.5	369.8	430.2	440.8	493.2	617.3
6.000		84.87	108.9	540.9	348.3	410.0	427.7	479.5	606.7
6.3986	Fe K α	86.75		468.0					
7.0580	Fe K β_1	62.99		361.5					
7.112	Fe K-edge	(456.6)							
7.4724	Ni K α	386.0	58.12						
8.000		313.6	48.38	257.0	166.3	197.7	208.0	231.0	302.8
8.2647	Ni K β_1		44.68						
8.333	Ni K-edge		(361.9)						
8.3876	W L α_1	278.0		226.1	148.8	173.3	181.6	203.4	
9.000				185.2	120.9	146.9	152.6	170.2	
9.500				161.0	104.4	127.1	133.1	148.4	
9.8615	W L β_2	172.9		141.6		113.1	118.4	131.3	182.5
10.00		172.2	213.6	140.2		111.3	116.6	129.6	180.8
10.50						92.76	102.7	113.2	
11.00						86.16	90.86	100.4	
11.50					L		80.61	89.40	
12.00								79.96	
12.50								71.78	
13.50					167.8	L			
14.00					153.3		L		
14.50					141.4	166.8			
15.00		57.21	71.63	46.86	134.0	156.3	161.2	L	63.87
17.00					92.89	107.2	111.3	123.3	45.31
20.00		25.66	32.51	21.40	62.62	72.22	75.13	84.92	L
24.9424	Ag K α_1	13.67		11.59	35.58	41.72	43.94	48.36	65.17
30.00		8.166		42.60	21.72	25.85	27.33	29.83	40.80
35.00					14.79	17.07	18.06	20.07	27.08
40.00		3.604		19.60	10.21	11.94	12.72	14.20	19.45
Multiplier*		92.72	97.48	197.1	300.4	323.9	327.0	344.0	396.2

(L) implies the average value through EXAFS region

*The multiplier is used to convert from μ (cm²/g) to σ (barns/atom).

Samples were free-standing, ultrapure, vacuum-evaporated target foils. The samples were a few hundred to a few thousand $\mu\text{g}/\text{cm}^2$ such that the mean free path thicknesses, $\mu\rho x$, were 1 and 3 at 1.5 keV. Samples were vapor-deposited on, and floated off of, optically flat glass slides. Their weights per unit area, ρx g/cm², were determined within 2% or 3%. Thicker samples, of known weights per unit area, were used for the higher energy measurements. The ratio of $\mu\rho x$ for thick and thin samples tested ρx for the thin samples.

Back-scattered protons and alpha particles, as well as ion microanalyses, were used to determine the samples' purity. The uranium samples had protective layers of beryllium. Samples were vacuum evaporated Be-U-Be sandwiches and Be-Be target foil samples. There were 16 samples prepared at a time using a precision mask. The protective layers of Be were each about 42 $\mu\text{g}/\text{cm}^2$, whereas the sandwich foils contained about 125 and 340 $\mu\text{g}/\text{cm}^2$ of uranium.^{6,14} The bulk materials used for our measurements had less than 100 parts per million by weight of contaminants.

A double-Be-window demountable-tube x-ray generator by Hilger and Watts, in conjunction with two crystal spectrometers was used for x-ray transmission measurements. An Ag target was used to provide a continuous radiation source for measurements at energies above 5 keV. The impurities in the target provided calibration lines (e.g., Fe, Ni, Cu and W) for aligning the spectrometer.¹⁵ The crystal used was LiF and the gas which filled the proportional counter as p-10.

Both a Mo target and a multi-element target of Al vapor-deposited on Cr (which had Nd and W impurities) were used for the 1 to 5 keV measurements. The flow proportional counter gas was methane at 10 cm H₂O above atmospheric pressure. The absolute measurement energies of continuum radiation were known to 3 or 4 eV. Uncertainties were 2 eV relative to tabulated emission line energies for W(M), Nd(L), Mo(L), Cr(K) and Al(K). The alignment was done in first or second order. Multiple orders were separated with discriminators on a multichannel analyzer. Higher order radiations were eliminated by reducing the target voltage appropriately.

The attenuation coefficients, $\mu(\text{cm}^2/\text{g})$, were derived from transmission measurements, i.e., $\exp(-\mu x)$. Usually two or more measurements, consisting of ten pairs (but occasionally up to twenty) of sample-in, followed by sample-out measurement periods, provided standard deviations of the mean error for μx ranging from 0.2 to 2%. These measurements were followed by background measurements. The backgrounds were determined using exit slits 2° 20' on both sides of the diffracted beam. Corrections for backgrounds were typically 1 or 2%. Dead time corrections were less than 1%. Continuum radiation spectra were used in addition to emission line spectra to obtain the measurements given in Table 1. The uranium data were reported previously.^{6,7}

Critical comparison of experiment with tabulations

For convenience, an analytical expression was derived to smoothly fit the empirical data of Table 1. A logarithmic quadratic regression fit, with parameters a_0 , a_1 , and a_2 , reproduces the measured data between absorption-edge energies to within 2%. Four or more data (per shell) were smoothly fit between the K and L_1 , the L_2 and M_1 , and the M_2 and N_1 shell absorption-edge energies. The quadratic regression fit parameters shown in Table 2 apply to Equation 1:

$$\log_e \mu(\text{cm}^2/\text{g}) = a_0 + a_1 \log_e E(\text{keV}) + a_2 [\log_e E(\text{keV})]^2 \quad (1)$$

Table 2. Quadratic regression fit parameters for Equation 1.

	Above K-edge			Above L ₁ -edge			Above M ₁ -edge			Above N ₁ -edge		
	a_0	a_1	a_2	a_0	a_1	a_2	a_0	a_1	a_2	a_0	a_1	a_2
²⁶ Fe	11.340	-2.620	-0.028	9.135	-2.334	0.164						
²⁸ Ni	12.120	-3.082	0.067	9.448	-2.522	-0.075						
⁵⁰ Sn				10.806	-2.408	-0.059	9.051	-2.007	-0.314			
⁷² Ta				11.150	-2.124	-0.072	9.530	-1.552	-0.274			
⁷⁸ Pt				11.054	-2.346	-0.030	10.127	-2.068	-0.124	8.418	-1.606	-0.691
⁷⁹ Au				11.513	-2.257	-0.047	10.222	-2.142	-0.100	8.446	-1.665	-0.586
⁸² Pb				10.306	-1.506	-0.150	10.133	-1.830	-0.162			
⁹² U				11.560	-2.063	-0.072	9.842	-1.506	-0.197	8.983	-1.707	-0.347

† Fit parameters derived from 4 or more input data (avoiding structure regions) allowed cross sections from widely-used compilations to be compared with 3% experimental data by Del Grande and Oliver, 1982.

Using the fit parameters in Table 2, a critical test of four widely-used tables was possible. The tabulations included relativistic Hartree-Slater theory,^{4,12} experimental data and non-relativistic theory (in the absence of experimental data),^{1,2} and experiment with interpolations or extrapolations where there were no experimental data, at energies above 1 keV.³ The percent difference between the present measurements and the tabulated values is shown in Figure 1.

Figure 1. The percent difference: $100 (\mu_{\text{meas}} - \mu_{\text{tab}}) / \mu_{\text{meas}}$.

A summary of the average effect for the percent differences shown in Figure 1 is given in Table 3. Table 3 gives the average percent difference, standard deviation and number of data values, n , used to critically test the four widely-used tables.

Table 3. Summary of average percent difference: $100(\mu_{\text{meas}} - \mu_{\text{tab}}) / \mu_{\text{meas}}$.

Average % difference, standard deviation, and number of data, comparing new data with compilations¹

	McMaster et al., 1989				Henke et al., 1982				Voigto, 1973				Scofield, 1973			
	K Shell	L Shell	M Shell	N Shell	K Shell	L Shell	M Shell	N Shell	K Shell	L Shell	M Shell	N Shell	K Shell	L Shell	M Shell	N Shell
Fe	2 ± 3	0 ± 2			4 ± 2	5 ± 2			0 ± 1	-3 ± 7			2 ± 4	2 ± 3		
	n = 8	n = 12			n = 4	n = 18			n = 7	n = 8			n = 7	n = 8		
Ni	2 ± 3	0 ± 2			5 ± 2	5 ± 3			2 ± 2	-2 ± 4			3 ± 4	1 ± 3		
	n = 4	n = 13			n = 2	n = 20			n = 4	n = 9			n = 4	n = 8		
Sn	3 ± 2	0 ± 1	-7 ± 11		6 ± 6	2 ± 3			1 ± 2	-3 ± 4	-8 ± 9		2 ± 1	1 ± 3	5 ± 6	
	n = 3	n = 13	n = 5		n = 10	n = 12			n = 3	n = 8	n = 5		n = 3	n = 8	n = 5	
Ta		-1 ± 2	-6 ± 11			-2 ± 3			-1 ± 4	-4 ± 7			1 ± 2	1 ± 3		
		n = 6	n = 9			n = 11			n = 5	n = 6			n = 5	n = 6		
Pt	4 ± 1	-7 ± 7	-6 ± 10			-5 ± 1	-4 ± 3		-5 ± 4	-5 ± 4	-13 ± 17		-3 ± 0	0 ± 2	1 ± 4	
	n = 6	n = 9	n = 4			n = 11	n = 7		n = 5	n = 7	n = 4		n = 5	n = 7	n = 4	
Au	-2 ± 2	-4 ± 2	-8 ± 9			-6 ± 1	-8 ± 4		-3 ± 4	-5 ± 2	-12 ± 17		-3 ± 1	0 ± 2	1 ± 4	
	n = 6	n = 9	n = 4			n = 11	n = 8		n = 5	n = 7	n = 4		n = 5	n = 7	n = 4	
Pb	-2 ± 3	-5 ± 4	0 ± 0			-5 ± 1	-3 ± 4		-5 ± 6	-5 ± 3	-5 ± 4		-3 ± 2	1 ± 3	5 ± 2	
	n = 5	n = 9	n = 2			n = 10	n = 2		n = 4	n = 7	n = 2		n = 4	n = 7	n = 2	
U	0 ± 1	-2 ± 4	5 ± 5			-14 ± 3	-12 ± 5		-1 ± 2	-5 ± 3	-28 ± 13		-2 ± 1	-2 ± 2	0 ± 4	
	n = 4	n = 5	n = 3			n = 6	n = 3		n = 3	n = 4	n = 3		n = 3	n = 4	n = 3	

¹ Energy regions where differences between fits to 2% measurements disagree with previous compilations more than 5% are boxed. Experimental input data were sparse or nonexistent over these energy regions.

Discussion of theory

Agreement between total attenuation coefficients and theory⁴ is typically better than 5% as shown in Table 3 and in Figure 2. The Hartree-Slater calculation is a central-field

Figure 2. Photoionization cross sections derived from two different theoretical models and experiment for Fe, Ni, Sn, Ta, Pt, Au, Pb and U.

model.¹² It does not account for the effects of the induced electric field due to cooperative oscillations of the atom. These many-body effects are approximated in more sophisticated calculations.

A recent study of uranium (metal) experimental data and many-body theory for the uranium atom showed that relativistic one-electron theory is not applicable for extremely soft x-rays (i.e., at 100 to 200 eV).⁷ The giant dipole resonance region for $5d + 5f$ transitions in uranium has photoabsorption cross sections ten times larger than calculations based on the one-electron models. This major difference is associated with excitation of the valence electrons and the $6p$ - and $5d$ -subshells.

Following a recent workshop on "New Directions in Soft X-ray Photoabsorption",¹⁶ Dr. Gary Dooleen at the Los Alamos National Laboratory set up his program to compare many-body calculations with our experimental data.¹⁰ The results of this effort are shown in Figure 2 and Figure 3. The relativistic time-dependent local density approximation (RTDLDA) and the independent particle approximation (IPA) agree within about 5%, at most energies above 1 keV, and agree with the present experimental data.

Figure 3. Photoionization cross sections derived from two different theoretical models and experiment for uranium in the region of M and N absorption-edge energies.

The near-edge structure of uranium was considered in greater detail in a previous paper.⁶ Oscillator strengths for $3d_{5/2} + 5f$ and $3d_{3/2} + 5f$ transitions derived from theory were about twice those derived from our measurements. This structure represents the measured photoexcitation of $3d$ electrons to vacancies in the partially filled $5f$ states. Further refinement of theory is needed to explain near-threshold effects at a level better than 10 or 15%.

An earlier study confirmed the 10 or 15% differences between Hartree-Slater central field calculations and measurements for transition elements: Ti, Fe, Co, Ni, Cu, Zn.¹⁷ This effect is shown in Figure 4. It applies to the energy region within 1 keV of threshold on the high energy side of the K-absorption edge energy.

Figure 4. Precise attenuation coefficients compared with theory.¹⁷

Figure 5. The measured K-absorption edge (jump) ratio of K- plus L- plus outer-shell photoabsorption to L- plus outer-shell photoabsorption.

The Hartree-Slater central field calculations were not corrected for shake excitations which appear to be important at near threshold energies for transition elements Ti-Zn. Measurements at near-edge energies for elements with higher atomic numbers: Zr-U, show that the effect is less than 5%.¹⁹ This is shown in Figure 5.¹⁸

Summary of Results

The results of our measurements supply theorists with the means to assess the predictive capabilities of their theoretical models. They provide an impetus for the development of new theoretical methodologies. They imply that the scarcity of prior 1 to 10 keV measurements has seriously affected the accuracy of widely used soft x-ray cross section tables. They suggest that several theoretical models provide predictive capabilities at the 5% level, except near threshold energies. X-ray optics requires knowledge of soft x-ray photoabsorption cross sections for a variety of neutral, ionized and excited states of a number of elements. It is not feasible to perform laboratory measurements of all the photoabsorption cross sections of all the important states and stages of ionization. It is crucial to employ a theoretical model with known predictive capability. This study has shown that at 1 to 10 keV, several theoretical models have a 5% or better predictive capability at most energies. Thus, theory is able to mend the crack in the foundation of x-ray optics at energies above 1 keV. At lower energies, the theories differ near threshold energies as much as a factor of ten. More measurements, which use synchrotron radiation sources, are needed to test the predictive capability of the more sophisticated many-body theoretical models.

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