

X-Ray interactions With Superheavy Atoms

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August 18th, 2016

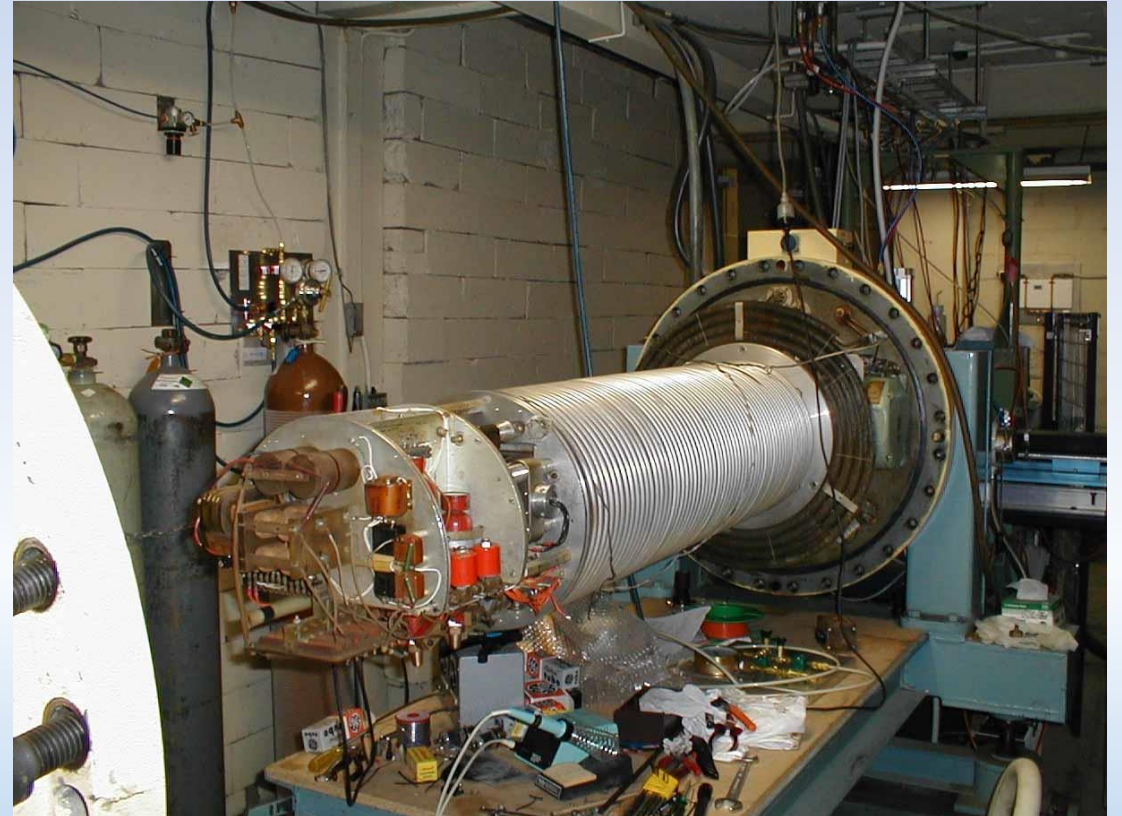
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Introduction

Superheavy Elements

- ❖ Start from $Z = 104$
- ❖ Have very short half-lives
- ❖ Made artificially
- ❖ Created in a particle accelerator



Particle Accelerator, https://upload.wikimedia.org/wikipedia/commons/5/5f/2mv_accelerator-MJC01.jpg

Extended Periodic Table

(Superheavy elements may not exist, and may not follow the order of this table even if they do)

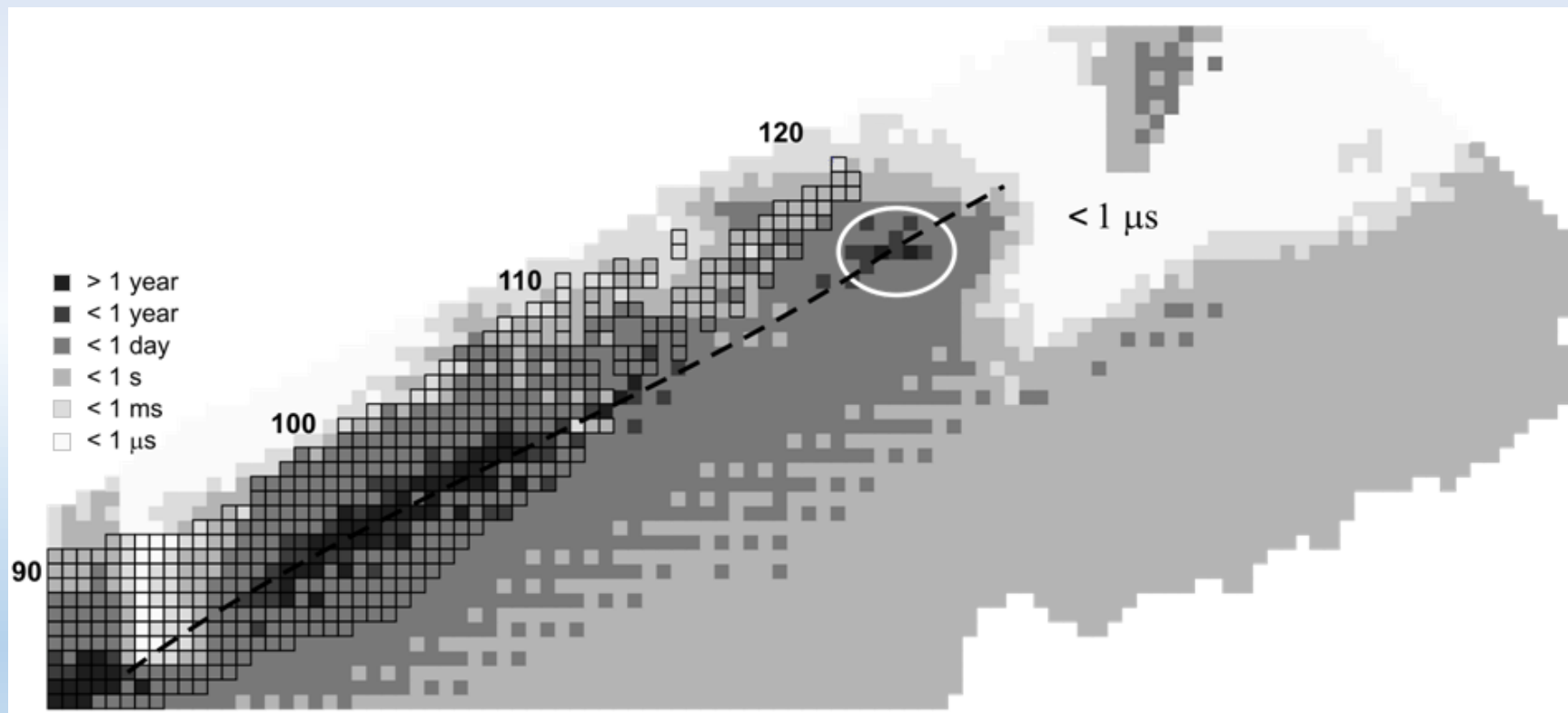
1	1																	2																																
	H																	He																																
2	3	4													5	6	7	8	9	10																														
	Li	Be													B	C	N	O	F	Ne																														
3	11	12													13	14	15	16	17	18																														
	Na	Mg													Al	Si	P	S	Cl	Ar																														
4	19	20													21	22	23	24	25	26	27	28	29	30	31	32	33	34	35	36																				
	K	Ca													Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr																				
5	37	38													39	40	41	42	43	44	45	46	47	48	49	50	51	52	53	54																				
	Rb	Sr													Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe																				
6	55	56													57	58	59	60	61	62	63	64	65	66	67	68	69	70	71	72	73	74	75	76	77	78	79	80	81	82	83	84	85	86						
	Cs	Ba													La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn						
7	87	88													89	90	91	92	93	94	95	96	97	98	99	100	101	102	103	104	105	106	107	108	109	110	111	112	113	114	115	116	117	118						
	Fr	Ra													Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr	Rf	Db	Sg	Bh	Hs	Mt	Ds	Rg	Cn	Uut	Uuq	Uup	Uuh	Uus	Uuo						
8	119	120	121	122	123	124	125	126	127	128	129	130	131	132	133	134	135	136	137	138	139	140	141	142	143	144	145	146	147	148	149	150	151	152	153	154	155	156	157	158	159	160	161	162	163	164	165	166	167	168
	Uue	Ubn	Ubu	Ubb	Ubt	Ubq	Ubp	Ubh	Ubs	Ubo	Ube	Utn	Utu	Utb	Utt	Utq	Utp	Uth	Uts	Uto	Ute	Uqn	Uqu	Uqb	Uqt	Uqq	Uqp	Uqh	Uqs	Uqo	Uqe	Upn	Upu	Upb	Upt	Upq	Upp	Uph	Ups	Upo	Upe	Uhn	Uhu	Uhb	Uht	Uhq	Uhp	Uhh	Uhs	Uho
9	169	170	171	172	173																																													
	Uhe	Usn	Usu	Usb	Ust																																													

Extended Periodic Table, <https://sciencepicks.files.wordpress.com/2011/12/extended-periodic-table.png>

Why study them???

Island of Stability

- The idea that heavy elements near the isotope ^{300}Ubn (element 120) with near the magic number of protons and neutrons will be much more stable.



Island of Stability, https://upload.wikimedia.org/wikipedia/commons/7/7b/Island_of_Stability_derived_from_Zagrebaev.png

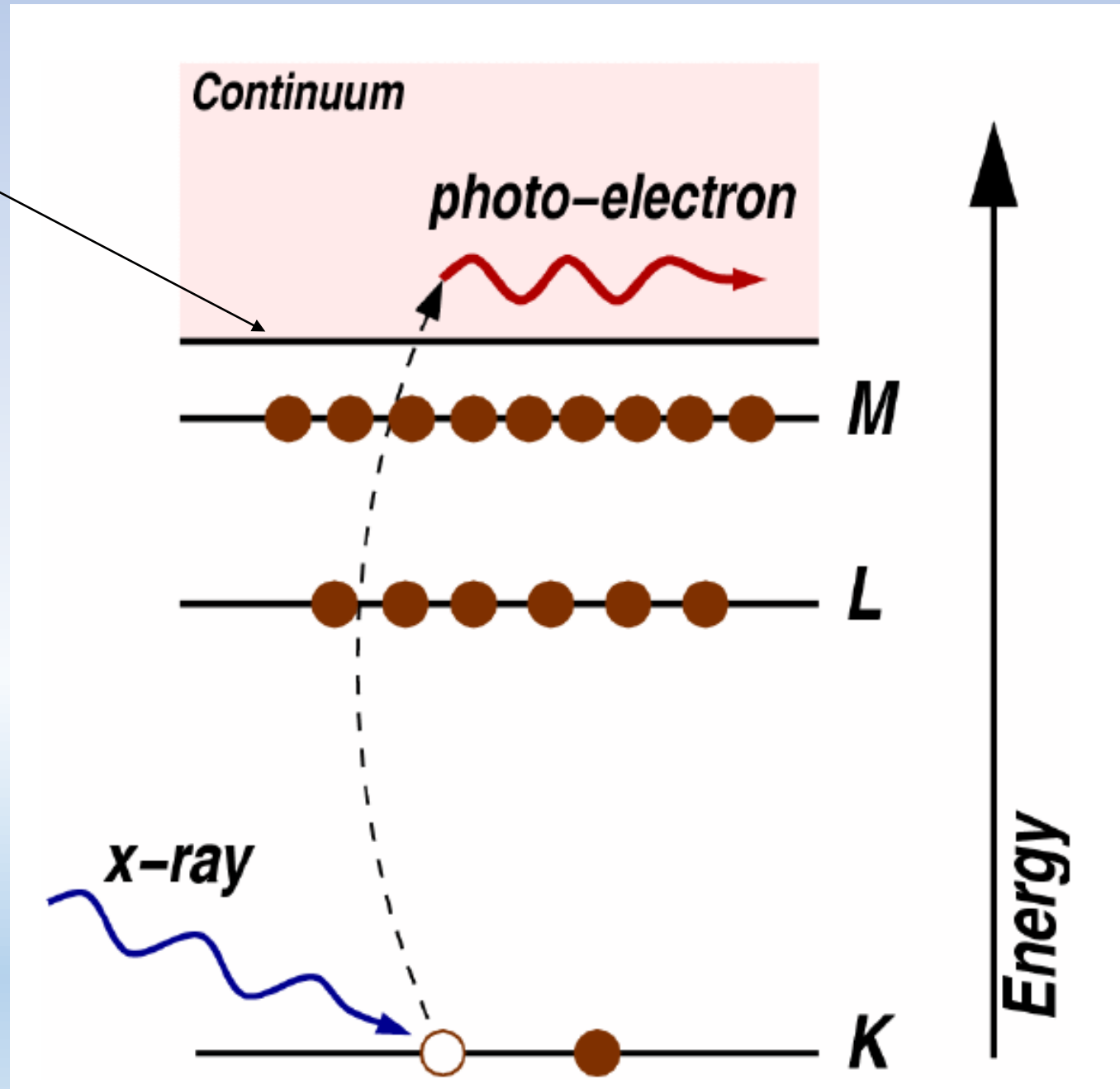
XAFS Theory

XAFS – X-Ray Absorption Fine Structure

- XAFS tells us how x-rays are absorbed by a system of atoms, including free atoms, molecules and solids at energies around the core-level binding energies of the atom.
- XAFS depends on the atomic structure and electronic and vibrational properties of the material; therefore, XAFS can be calculated for every element on the periodic table.
- XAFS can be used to characterize materials, including atomic and electronic structure.
- Absorption coefficient μ , $I = I_0 e^{-\mu t}$ Beer's Law.
- $\mu \propto \sum_f |\langle i|d|f \rangle|^2 \delta(E_f - E_i - \hbar\omega)$ - Fermi's Golden Rule.

Photoelectric Effect

Fermi Level



EXAFS – Extended X-Ray Absorption Fine Structure

- Edges are the sharp rises in absorption at certain energies. They occur at roughly the binding energy of each core-level electron.
- EXAFS is what is happening well above the edges.
- EXAFS Equation

$$\chi(k) = \sum_j \frac{N_j f_j(k) e^{-2k^2 \sigma_j^2}}{k R_j^2} \sin[2k R_j + \sigma_j(k)]$$

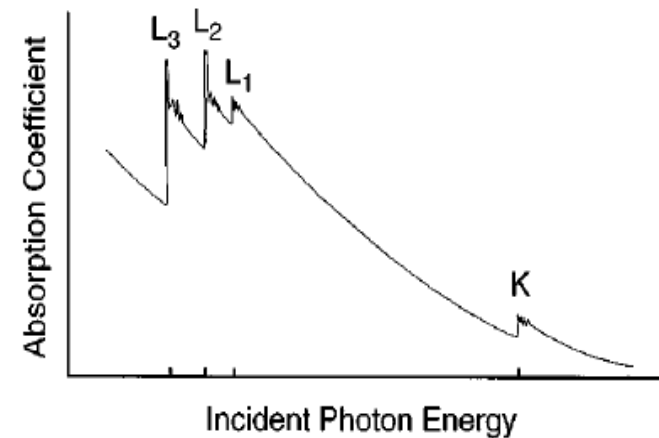
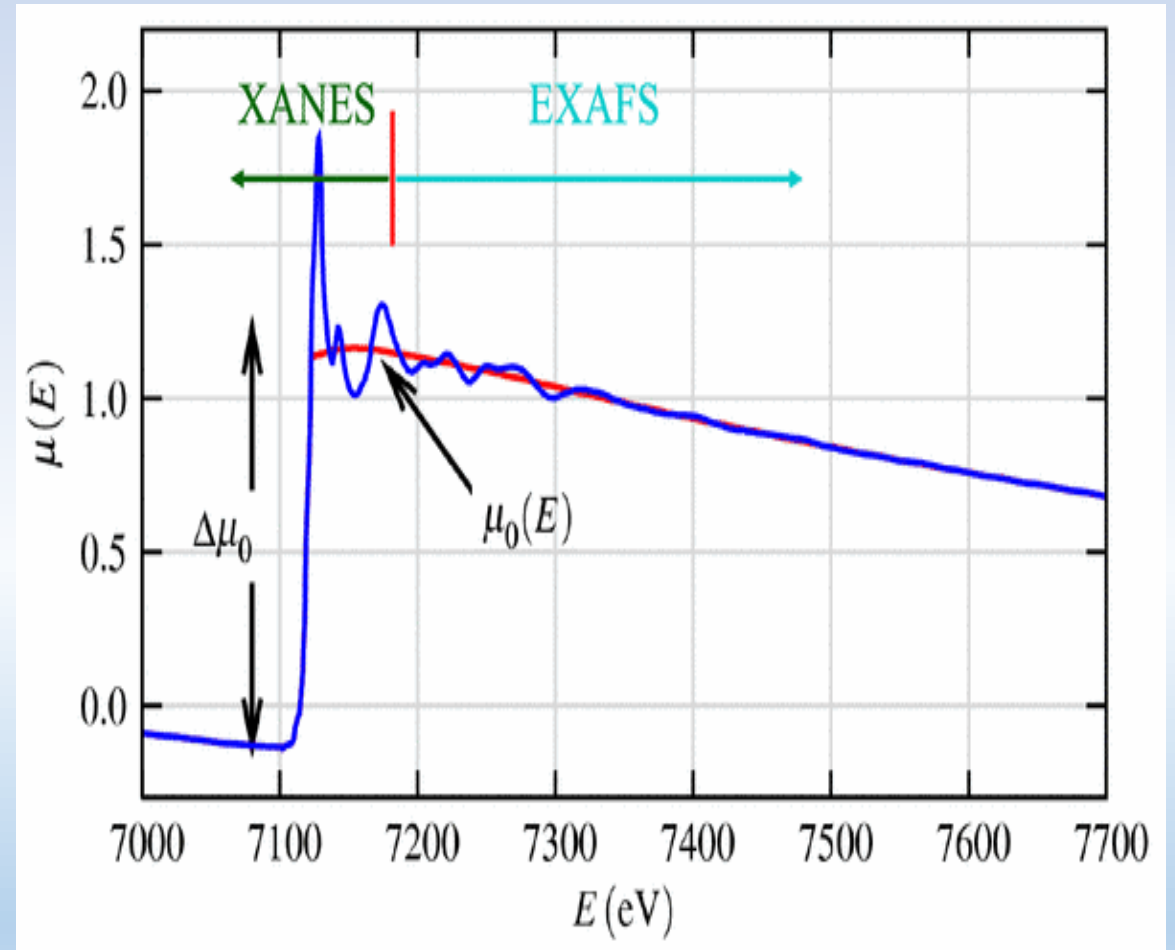


FIG. 2. Schematic view of x-ray absorption coefficient as a function of incident photon energy. Four x-ray edges are shown: K , L_1 , L_2 , and L_3 . Note that the overall decrease in absorption as a function of energy is punctuated by four sharp, step-function-like increases at each edge. Above each edge are the oscillatory wiggles known as the EXAFS. This figure was redrawn; it is based on Fig. I.2 of Müller (1980).

XANES – X-Ray Absorption Near Edge Structure

- XANES is typically within 30 eV of the main absorption edge.
- XANES can be used as a fingerprint to identify the presence of a particular chemical species.



FEFF9

The screenshot displays the FEFF9 software interface on a Linux desktop. The desktop background is a starry space scene. Several windows are open:

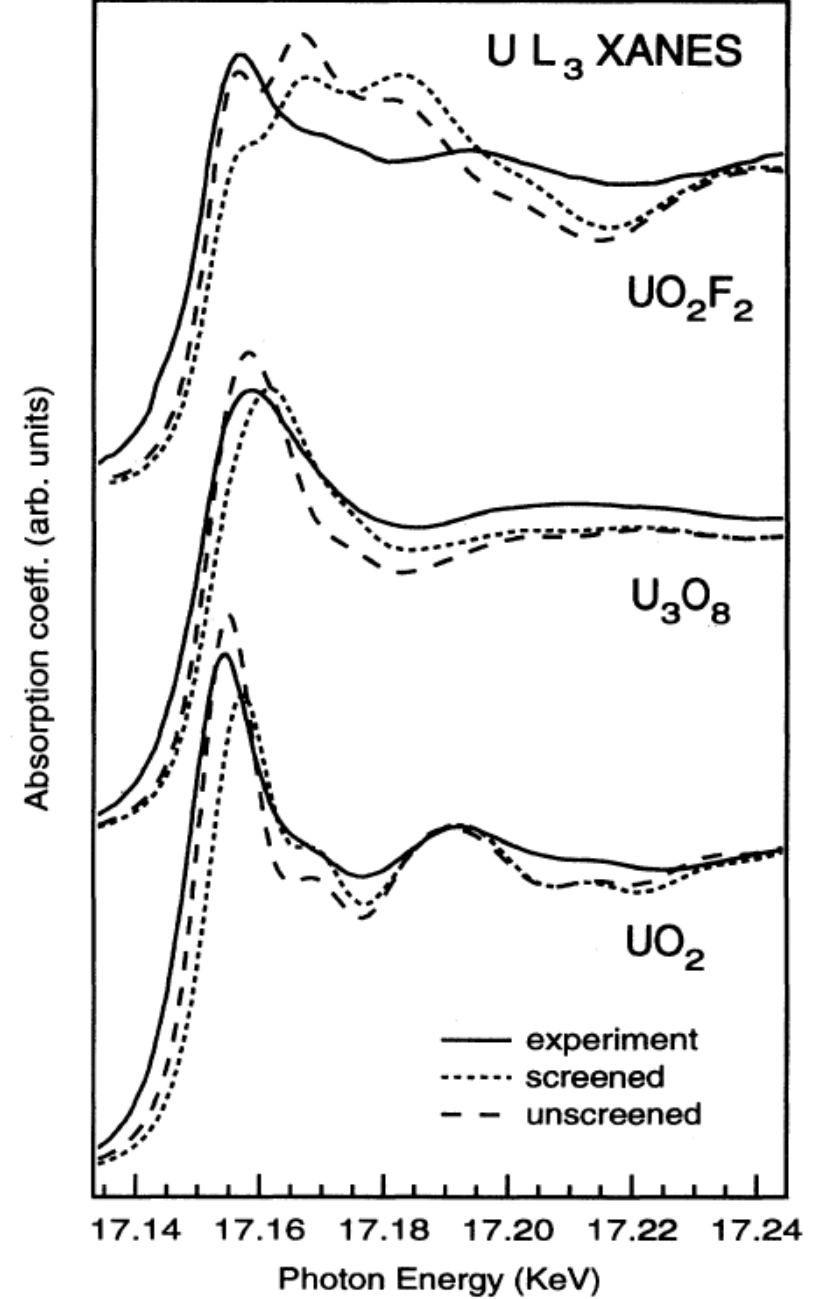
- Terminal Window (FEFF 9.5.1):** Shows the execution of the FEFF9 program. The output includes:


```

      FEFF 9.5.1
      Core hole lifetime set to: 7.57179181115714 4V.
      Thorium(IV) Bisoxalato(II) Bisquap(II) Dihydrate
      Th IC2 O42 O42 O42 O42
      Calculating potentials ...
      Electron configuration set to feff7 recipe.
      free atom potential and density for atom type 1
      free atom potential and density for atom type 2
      free atom potential and density for atom type 3
      initial state energy
      overlapped potential and density for unique potential 0
      overlapped potential and density for unique potential 1
      overlapped potential and density for unique potential 2
      overlapped potential and density for unique potential 3
      Calculating potentials ...
      muffin tin radii and interstitial parameters
      iph, romipho'boho, romipho'boho, fapipho
      0 1.63341E+00 1.68668E+00 1.15000E+00
      
```
- Plot Window:** Shows a graph of potential (y-axis, 0.00E+00 to 1.00E+01) versus distance (x-axis, 0 to 1.754). The plot shows a sharp initial peak followed by a series of smaller oscillations that decay towards a constant value.
- Main Control Window:** Contains various settings for the calculation. Key options include:
 - Special Settings:** EXAFS (checked), EDGE (checked), LDOS (unchecked).
 - Module Options:** SCF (unchecked), SO2 (checked), EXCHANGE (checked), COREHOLE (checked).
 - Run Module:** potentials (checked), fdfs (checked), phase shifts (checked), path expansion (checked), cross-section (checked).
- 3D Molecular Model Window:** Displays a ball-and-stick model of the Thorium(IV) Bisoxalato(II) Bisquap(II) Dihydrate complex. The thorium atom is represented by a large blue sphere, oxygen atoms by red spheres, and carbon atoms by grey spheres. The structure is highly symmetric and complex.
- Atoms Data Editor Window:** Shows two tables of data:

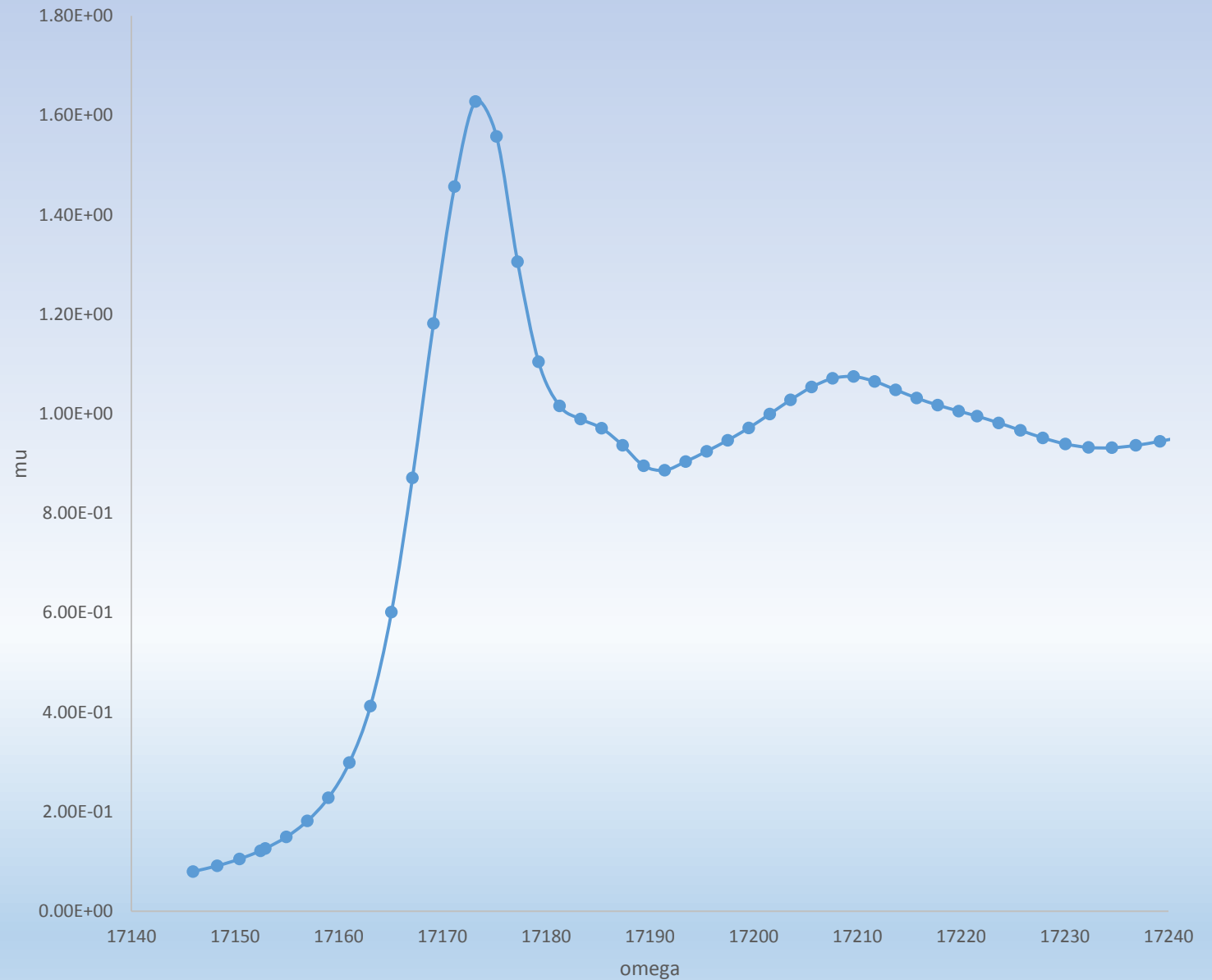
Potentials						
atom	l	mag	lmax-1	lmax-2	smooth	smooth
0	90 Th	1	3	3	0.001	
1	90 Th	3	3	4		
2	8 O	1	1	48		
3	6 C	1	1	38		

Atoms						
atom	z	l	lmax	lmax	lmax	comment
0	90	0	0	0	Th	0.00000 0
1	90	1.967	1.077	2.02	Th	2.41101 1
2	90	1.967	-1.077	2.02	Th	2.41101 2
3	8	-2.09	-1.357	2.04	O	2.50279 3
4	8	-2.09	-1.357	2.04	O	2.50279 4
5	6	0.92	1.836	2.08	C	2.50496 5
6	6	0.92	-1.856	2.08	C	2.50496 6
7	6	-0.841	0.357	2.05	C	2.51821 7
8	6	-0.841	-0.357	2.05	C	2.51821 8
9	8	-0.333	2.431	2.02	O	2.58815 9
10	8	-0.333	-2.431	2.02	O	2.58815 10
11	6	0.484	0.484	3.01	C	3.32841 11
12	6	-0.484	-0.484	3.01	C	3.32841 12
13	6	0.548	1.859	3.01	C	3.33163 13
14	6	-0.548	-1.859	3.01	C	3.33163 14
15	6	-2.337	2.337	3.02	C	3.38900 15
16	6	-2.337	-2.337	3.02	C	3.38900 16
17	6	-1.312	0.881	3.02	C	3.37417 17
18	6	-1.312	-0.881	3.02	C	3.37417 18
19	8	-1.877	0.308	2.06	O	4.33494 19
20	8	-1.877	-0.308	2.06	O	4.33494 20
21	8	0.816	2.501	2.05	O	4.54819 21
22	8	0.816	-2.501	2.05	O	4.54819 22
- Help Window (COREHOLE):** Provides information about the COREHOLE card, explaining that it specifies which edge to calculate and how the core state is treated. It lists options: `FEFF` (equivalent to the old `NORHOLE` card), `RPA` (screened core hole), and `FSR` (Final State Rule core hole). It also includes instructions on how to use these options.



Uranium Dioxide UO_2

UO₂ L3 Edge XANES



Erbium Oxide Er_2O_3

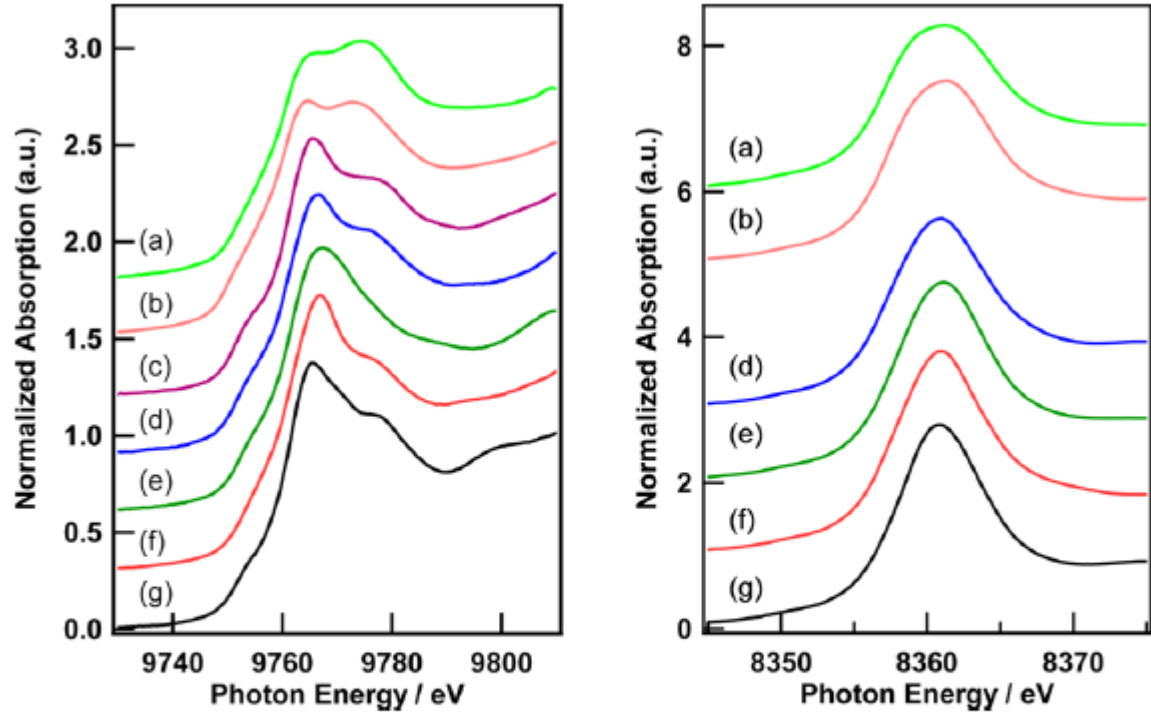
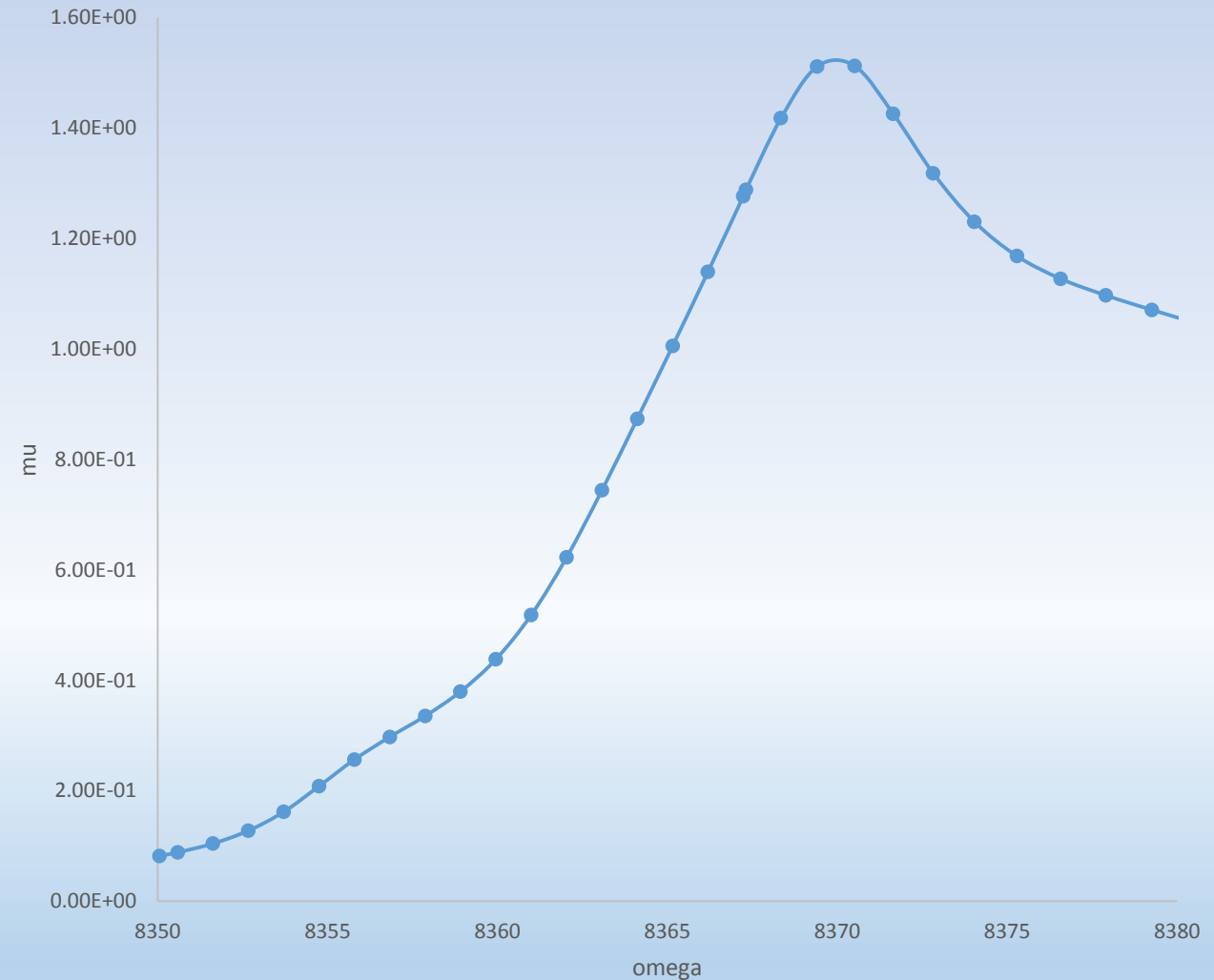


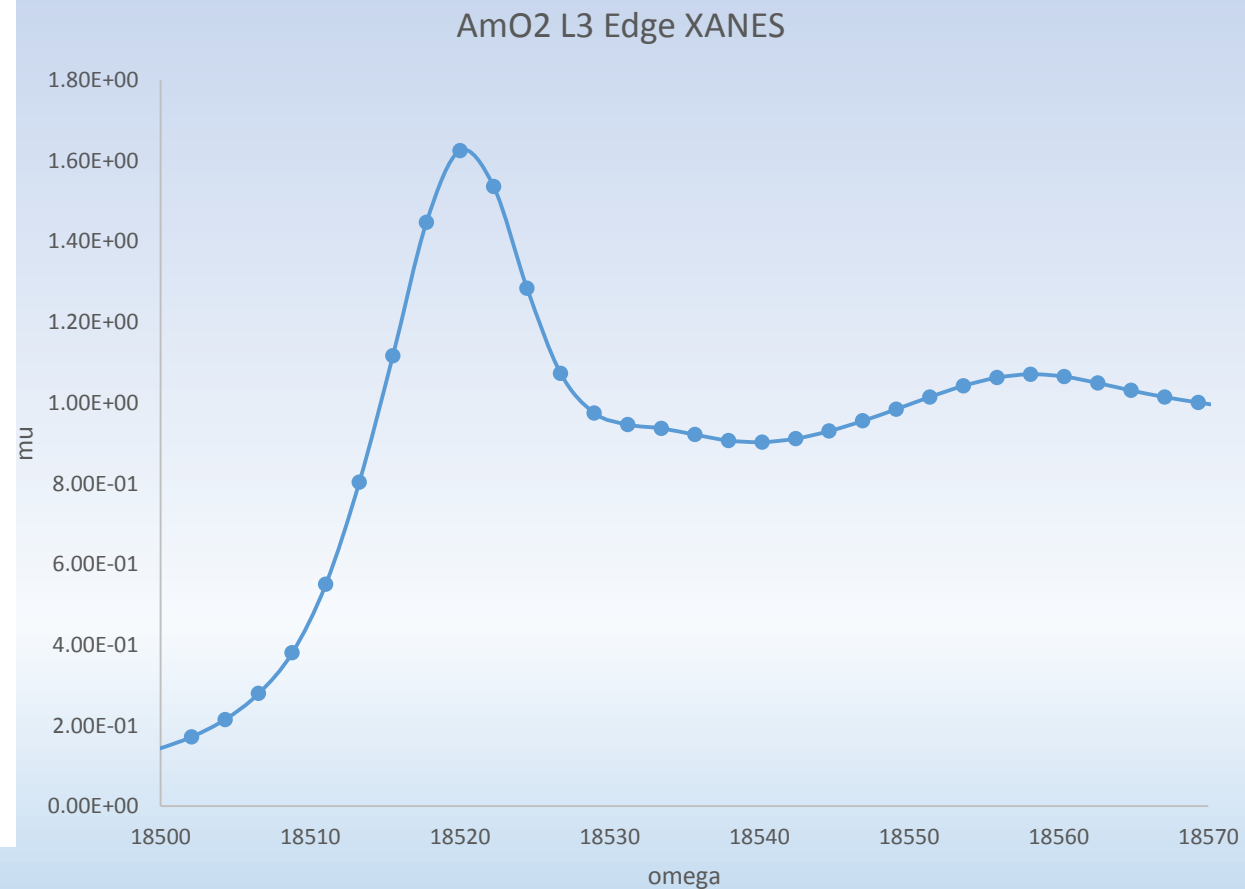
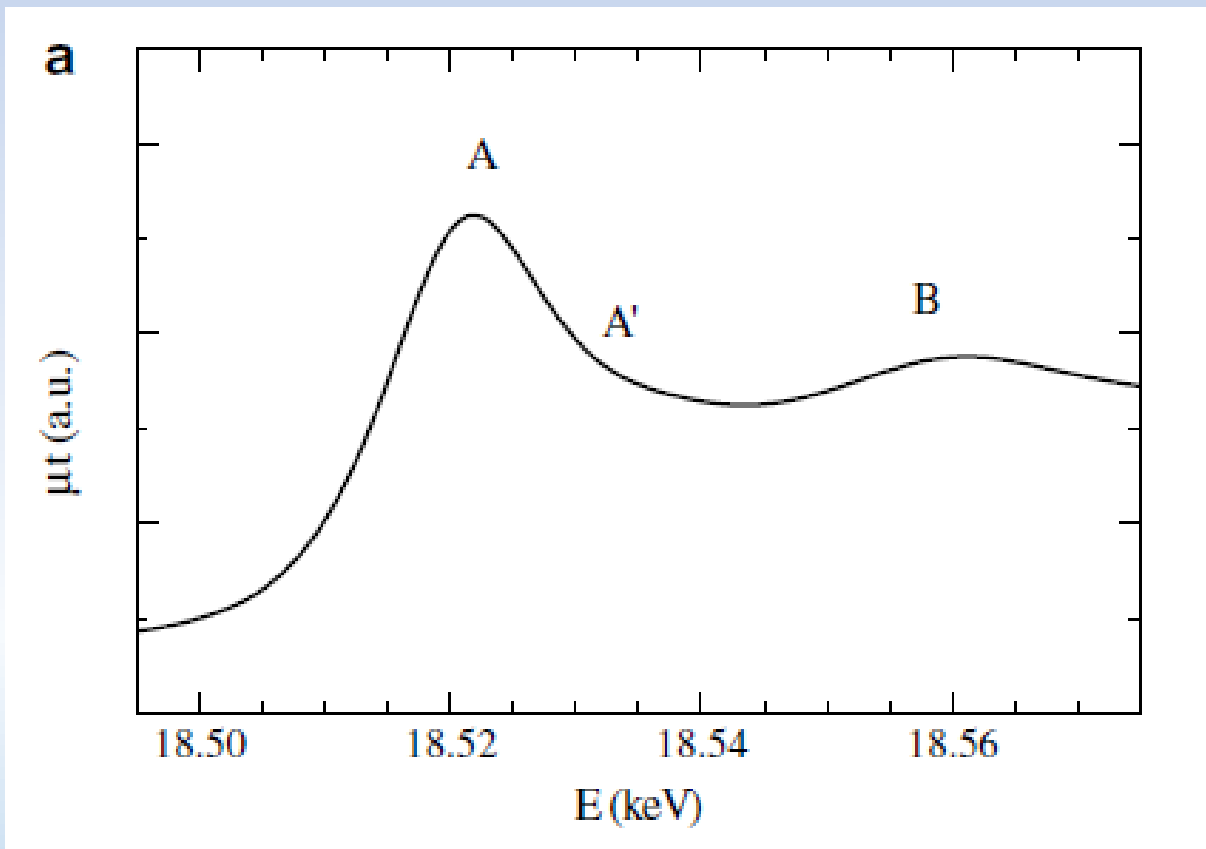
Figure 5. Er L_1 - (left) and L_3 -edge (right) XANES spectra of Er oxides: (a) $Er_2Cu_2O_5$, (b) Er_2O_3 , (c) $BaNiEr_2O_5$, (d) $BaCuEr_2O_5$, (e) $ErNbO_4$, (f) $Er_3Al_5O_{12}$, and (g) $ErVO_4$.

Hiroyuki Asakura, Tetsuya Shishido, Kentaro Teramura, and Tsunehiro Tanaka, *Local Structure and L_1 - and L_3 -Edge X-ray Absorption Near Edge Structure of Late Lanthanide Elements (Ho, Er, Yb) in Their Complex Oxides*, J. Phys. Chem. C 2015, 119, 8070–8077

Er2O3 L3 Edge XANES



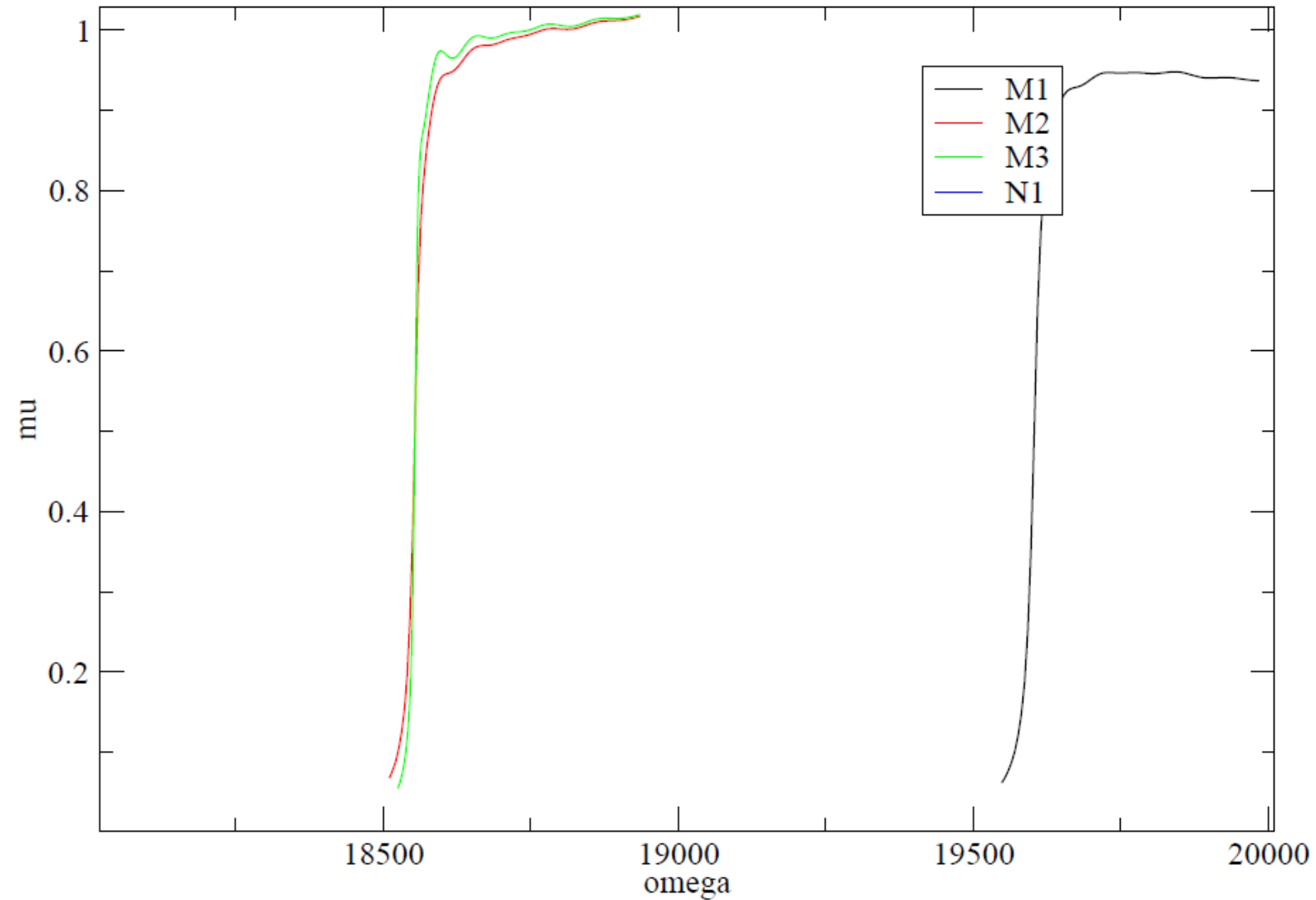
Americium Dioxide AmO_2



Tsuyoshi Nishi, Masami Nakada, Akinori Itoh, Chikashi Suzuki, Masaru Hirata, Mitsuo Akabori, *EXAFS and XANES Studies of Americium Dioxide with fluorite structure*, J. Of Nuclear Materials, 2007

XANES of Untrinium (Utn2)

Z = 130 XANES M1, M2, M3, N1



Thomson Scattering Cross Section & Thomas-Fermi Model

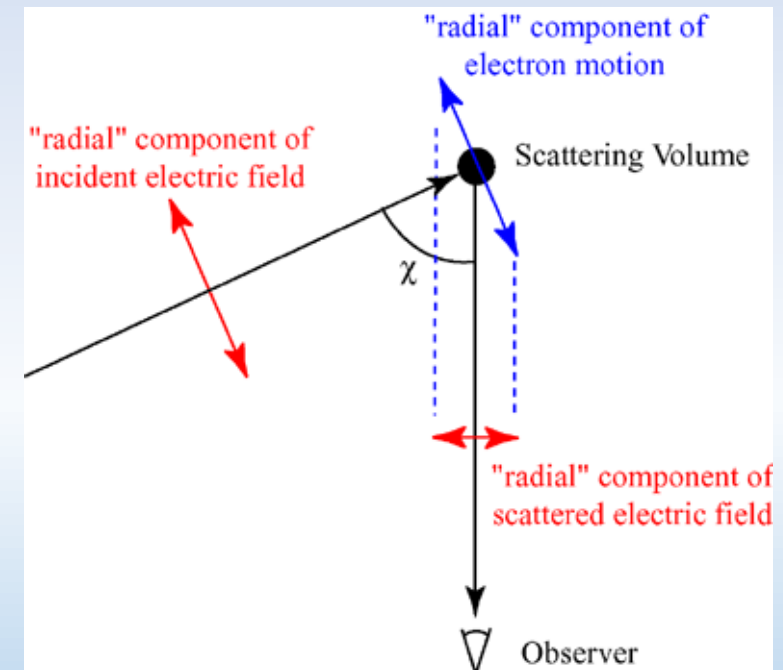
Thomson Scattering

- The elastic scattering of X-rays from free electrons.
- The atomic scattering factor

$$f_0 = \sum_{n=1}^Z \int_0^{\infty} 4\pi r^2 \rho_n(r) \frac{\sin(qr)}{qr} dr$$

- Momentum transfer

$$q = \frac{4\pi \sin(\theta_B)}{\lambda}$$



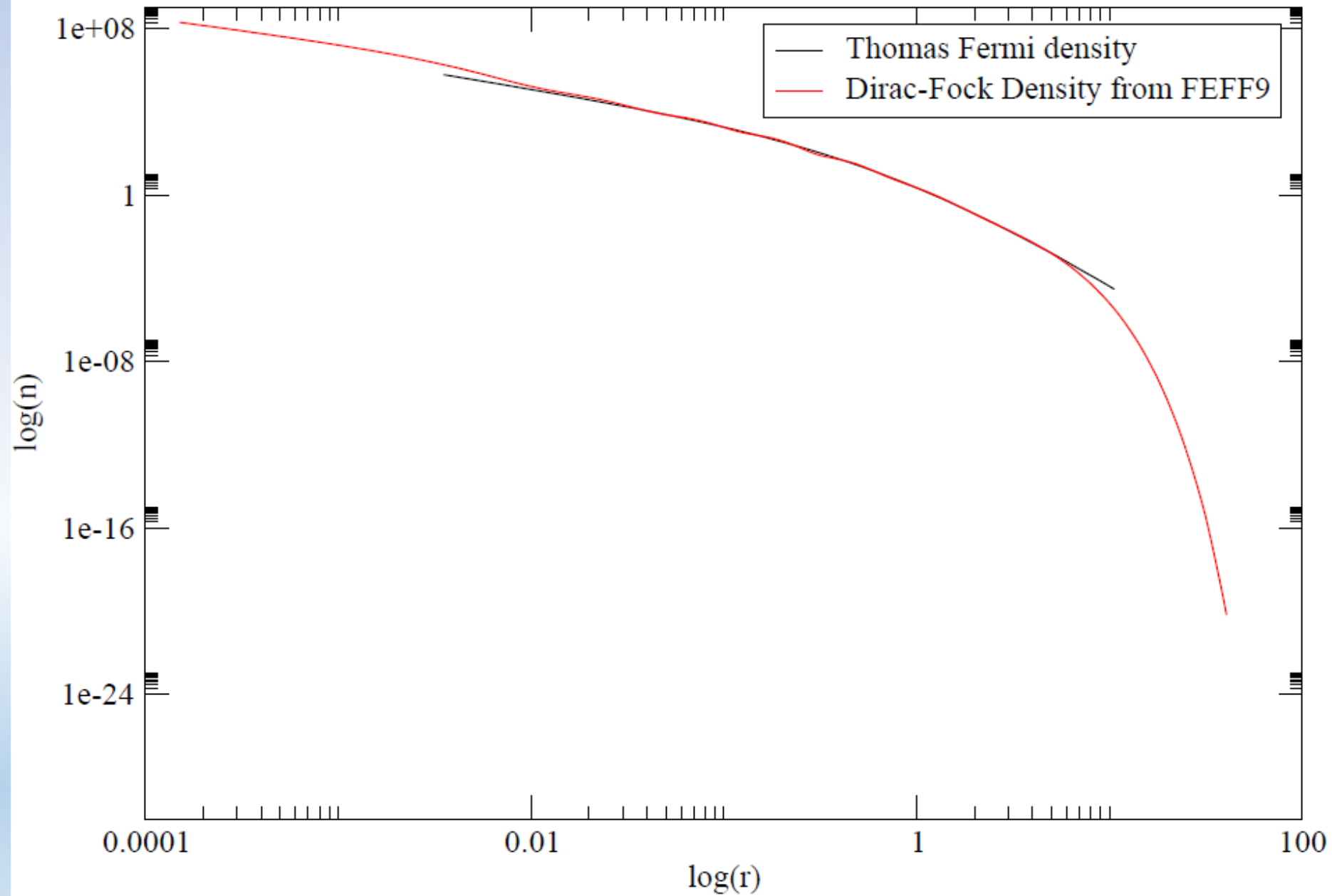
Thomson Scattering,
https://upload.wikimedia.org/wikipedia/commons/b/bf/Thomson_scattering_geometry.png

Thomas Fermi model

- $n = \frac{[2(\varphi - \varphi_0)]^{3/2}}{3\pi^2}$
- $r = xbZ^{-1/3}, b = 0.885$
- $\varphi(r) = \frac{Z^{4/3}}{b} \frac{\chi(x)}{x}$
- Thomas Fermi equation $x^{1/2} \frac{d^2\chi}{dx^2} = \chi^{3/2}$
- TF density $n(r) = Z^2 f\left(\frac{rZ^{1/3}}{b}\right), f(x) = \frac{32}{9\pi^3} \left(\frac{\chi}{x}\right)^{3/2}$

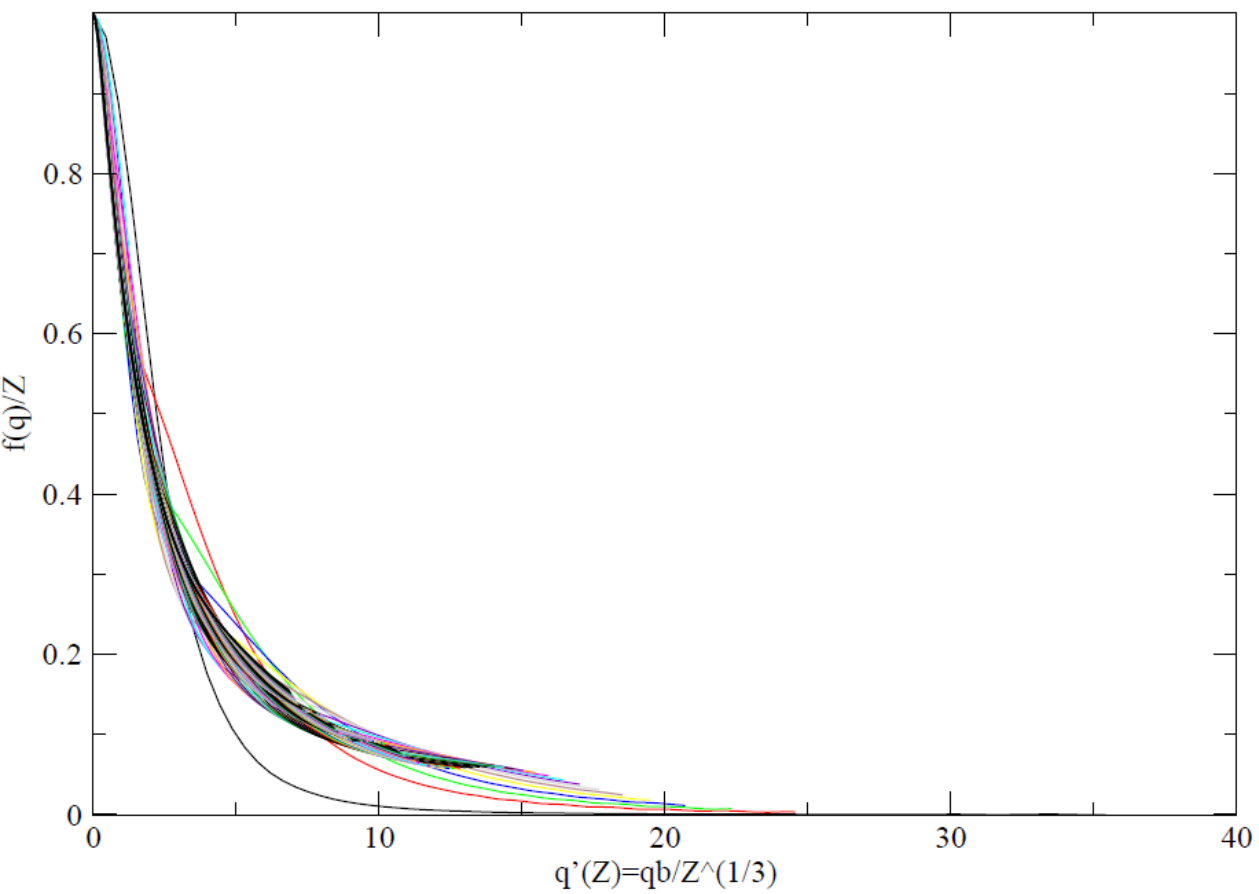
Density vs. radius for Thomas Fermi and FEFF9

$Z = 126$



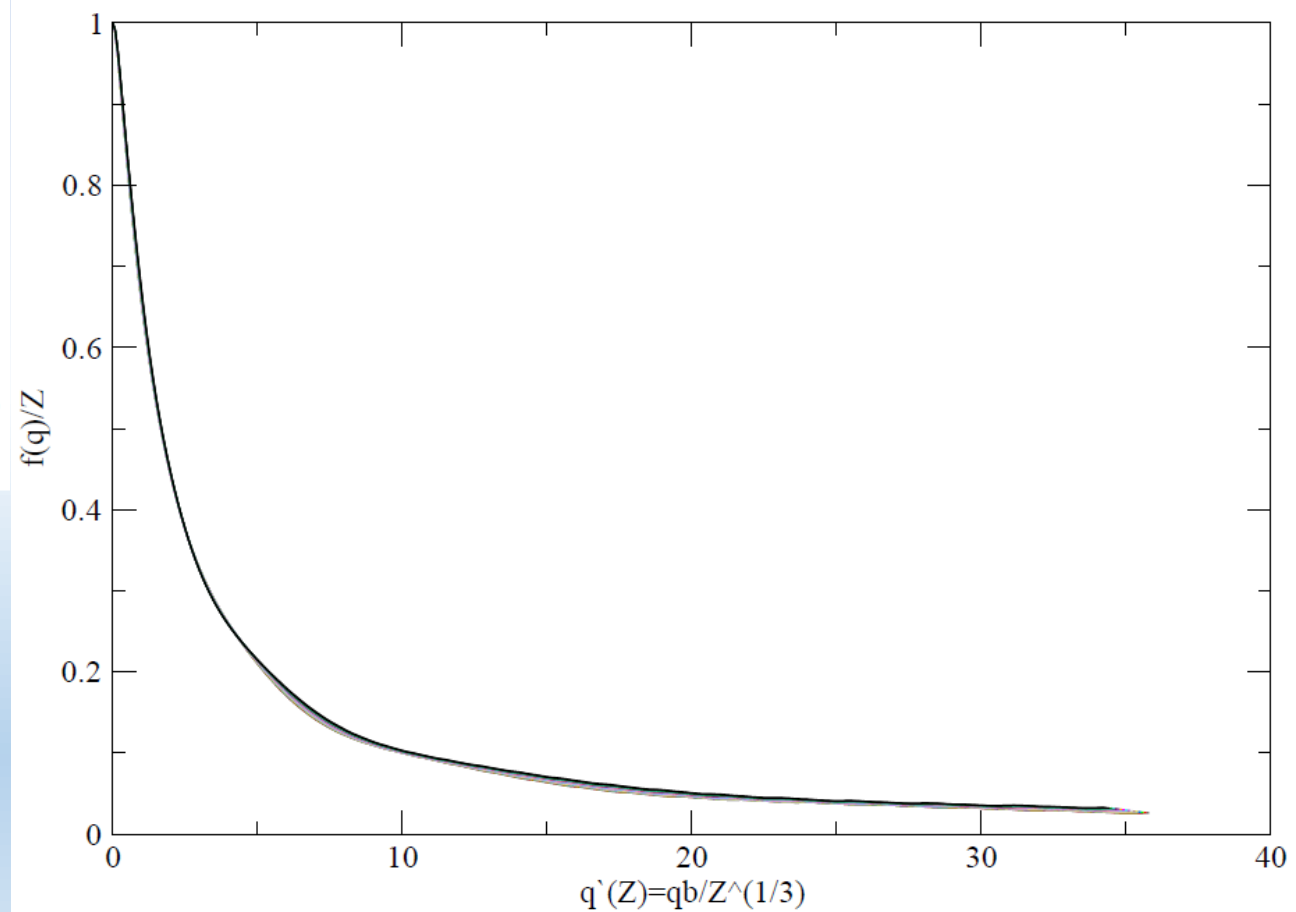
Thomson Scattering

$Z = 1-138$



Thomson Scattering

$Z = 121-138, 0 < q < 201$



Future Work

- Calculation of phase shifts using FEFF9
- Comparison to WKB approximation
- Developing a model to calculate radii for superheavy molecules.

Acknowledgments

Thank you to professor John Rehr, Joshua Kas, and Fernando Vila for their guidance through my projects.

Thank you to Ron Musgrave for the machine shop lessons.

Thank you to REU coordinators and the NSF.

References

J. J. Rehr, R. C. Albers, *Theoretical Approaches to X-ray Absorption Fine Structure*, Reviews of Modern Physics, Vol. 72, No. 3, 2000

Pekka Pyykko, *A suggested Periodic Table up to $Z \leq 172$, based on Dirac-Fock calculations on atoms and ions*, Phys. Chem. Chem. Phys., 2011,13, 161-168

Hiroyuki Asakura, Tetsuya Shishido, Kentaro Teramura, and Tsunehiro Tanaka, *Local Structure and L1- and L3-Edge X-ray Absorption Near Edge Structure of Late Lanthanide Elements (Ho, Er, Yb) in Their Complex Oxides*, J. Phys. Chem. C 2015, 119, 8070–8077

Tsuyoshi Nishi, Masami Nakada, Akinori Itoh, Chikashi Suzuki, Masaru Hirata, Mitsuo Akabori, *EXAFS and XANES Studies of Americium Dioxide with fluorite structure*, J. Of Nuclear Materials, 2007

E. A. Hudson, J. J. Rehr, J. J. Bucher, *Multiple-scattering calculations of the uranium L3-edge x-ray-absorption near-edge structure*, Physical Review B, 1995

Matthew Newville, *Fundamentals of XAFS*, Consortium for Advanced Radiation Sources, 2004

J. Kas, *Theory and Calculation of X-Ray Absorption*, <https://www.bnl.gov/ps/nsls/workshops/2006/exafs/talks/Kas.pdf>, 2006

V. L. Eletsii, V.S. Popov, *The Thomas-Fermi method for $Z > 137$* , Zh. Eksp. Teor. Fiz. 73, 2046-2059, 1977

A. Messiah, *Quantum Mechanics*, Dover Publications Inc., Mineola, New York, 1999

L. D. Landau, E. M. Lifshitz, *Quantum Mechanics, non-relativistic theory*, Pergamon Press, Oxford, New York, Beijing, Frankfurt, Sao Paulo, Sydney, Tokyo, Toronto, 1977

Gwyndaf Evans, *Classical X-ray scattering*, <http://www.gwyndafevans.co.uk/thesis-html/node11.html>, 1994

Ray Wong, Josh Alamillo, *EXAFS: Theory*, http://chem.libretexts.org/Core/Physical_Chemistry/Spectroscopy/X-ray_Spectroscopy/EXAFS%3A_Theory, 2016