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Measured and simulated high-energy x-ray photoelectron spectra for validation of radiation transport physics models

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presented by Tim Flanagan, Sandia National Labs
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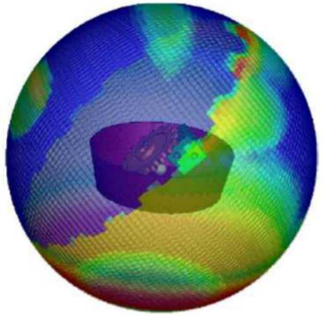
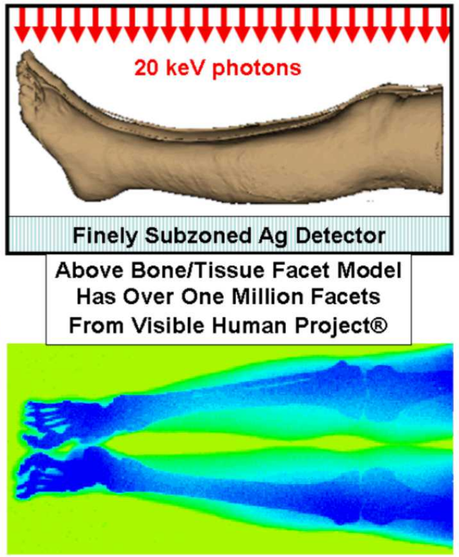
Radiation effects community must accurately predict how ionizing radiation interacts with matter. *(Created, absorbed, in, out, and through)*

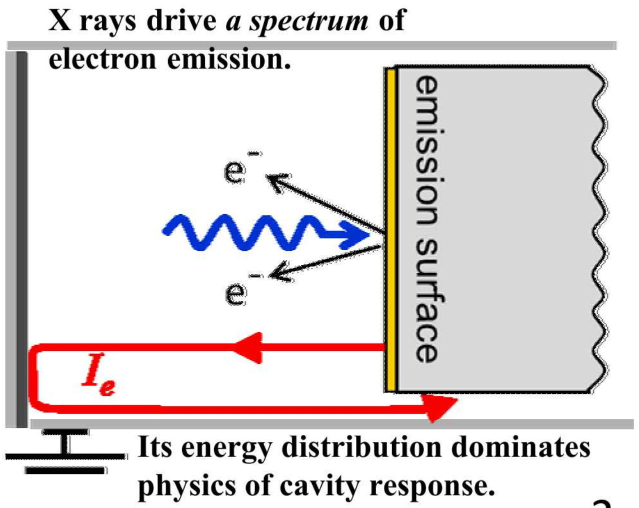
- Integrated Tiger Series (ITS) is a Monte-Carlo code that transports radiation in arbitrary geometries.
- SCEPTRE solves the Boltzmann-transport equation on a fine mesh with symmetry.
- Forward transport (one specified source → many separate responses)
- Backward transport (one detector response → from many sources [directions, energies])
- Fully-coupled photon and electron transport.
- pair-production, bremsstrahlung, Compton scattering, photoelectric absorption and relaxation, elastic/inelastic scattering.
- Do not have equal confidence in all energy ranges (vast).





ITS-Simulated Dose(Ag) Distribution

While utility is broad, many regimes lack validation. Lower energy photoemission is one such area.



We are addressing validation with tools used at synchrotrons by the materials science community.

Materials research / atomic-molecular physics:

- Scan small kinetic energy ranges.
- In high resolution (sub-eV) for a long time.
- *chemically-induced binding energy shifts; atomic concentration; electronic properties of buried interfaces; specific resonant interactions; solid state effects*
- interested in *specific* elastic photoelectron and Auger peaks, often at low E_B's; remove inelastic

These experiments:

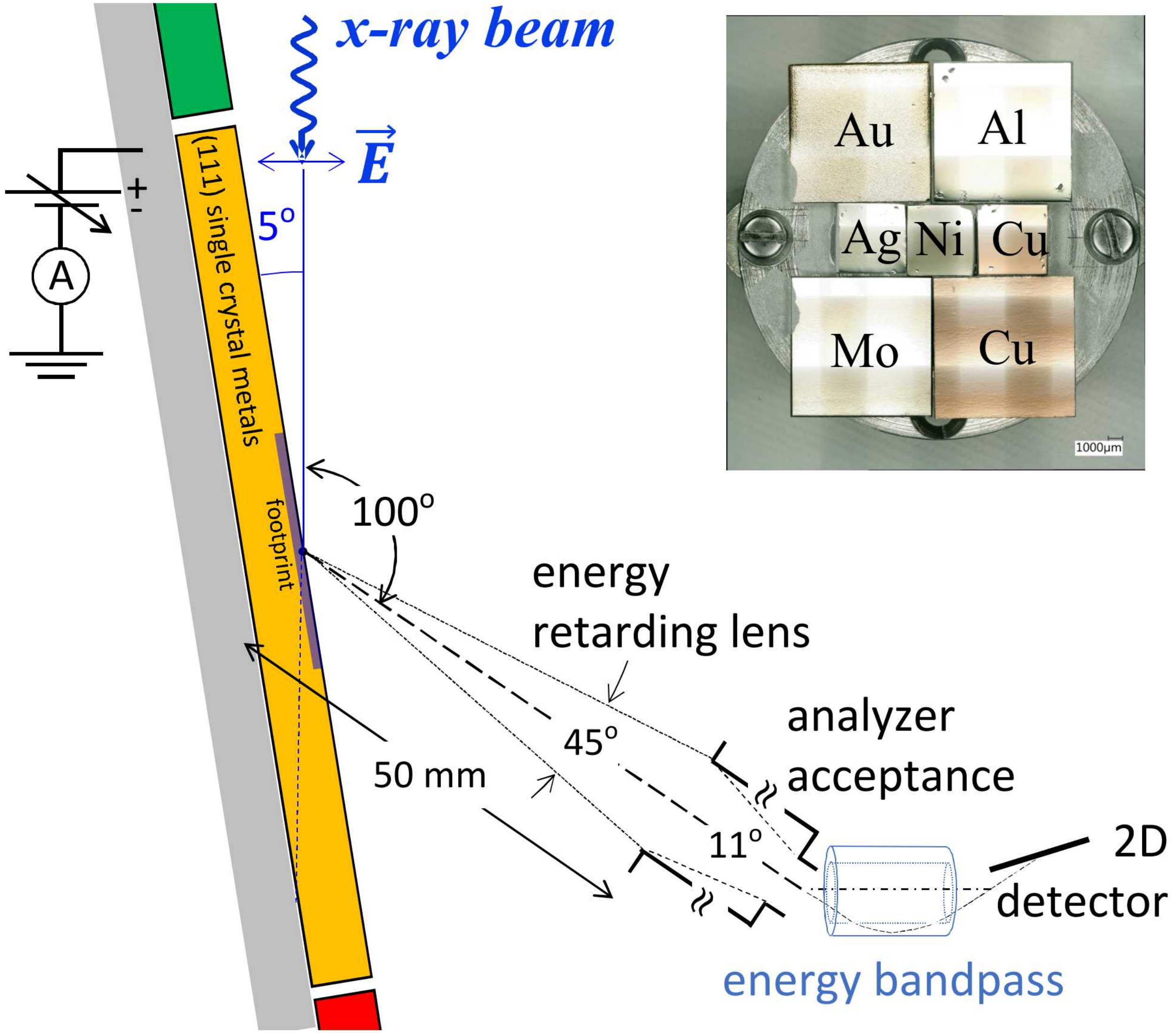
- Scan large range (up to photon energy)
- Lower resolution, reduced statistics.
- Test radiation transport physics models.
- Inelastic "background" just as important as elastic information.
- ***Measurements are a probability distribution for surface photoemission*.**

Need as large a kinetic energy range as possible, so we went to SpLine at BM25 at the ESRF.

SpLine

Spanish CRG Beamline

Geometry of experiment at SpLine.



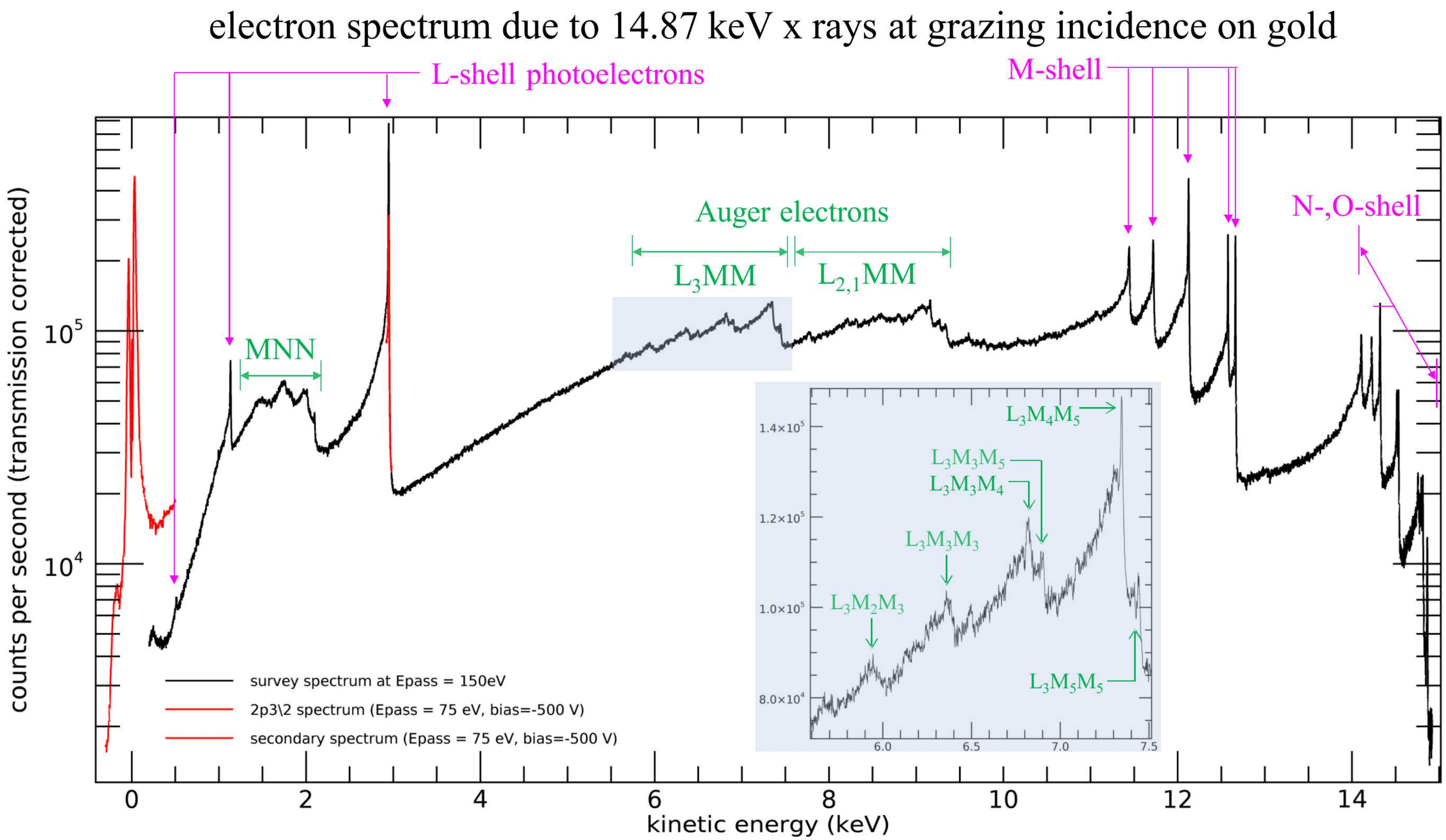
Electron kinetic energy analyzer.

- Custom cylindrical sector electrostatic analyzer measures kinetic energies from 10’s to 15,000 eV
- Full survey scan is several hours duration.
- Retarding electrostatic lens operates has energy-dependent transmission (fixed spatial magnification)
- For further details see literature references from J. Rubio-Zuazo and G.R. Castro (last slide).

X-ray beam tunable 8 to 26 keV.

- 8.0, 11.0, 12.99, 14.87, 18.0, 21.554 keV
- beam intensity varies with energy, but 10^{11} -10^{12} ph/sec.
- Post-monochromator realitive beam intensity monitored with a proportional counter
- Intensity dependence removed in all scans (small).

Measured gold survey spectrum driven by 14.87 keV- x rays.

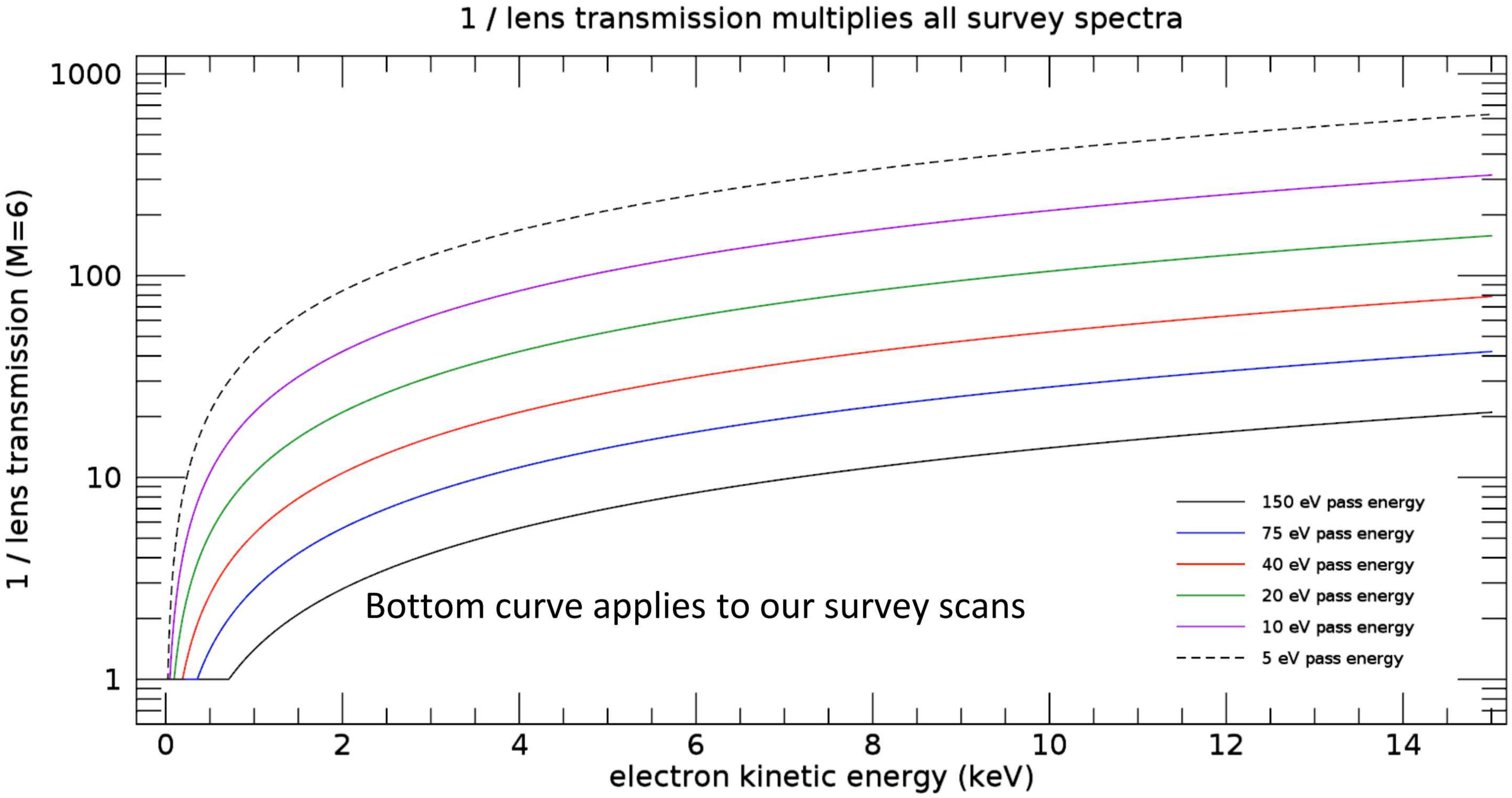


- 5 hour scan time
- 150 eV pass energy; 1 eV step size from 200 to 14,920 eV; 0.9 s dwell (90 images)
- Main scan: Sample unbiased, but grounded through a current meter.
- Low energy scan: sample bias = -500 V. Influence of field lines near sample need to be explored.

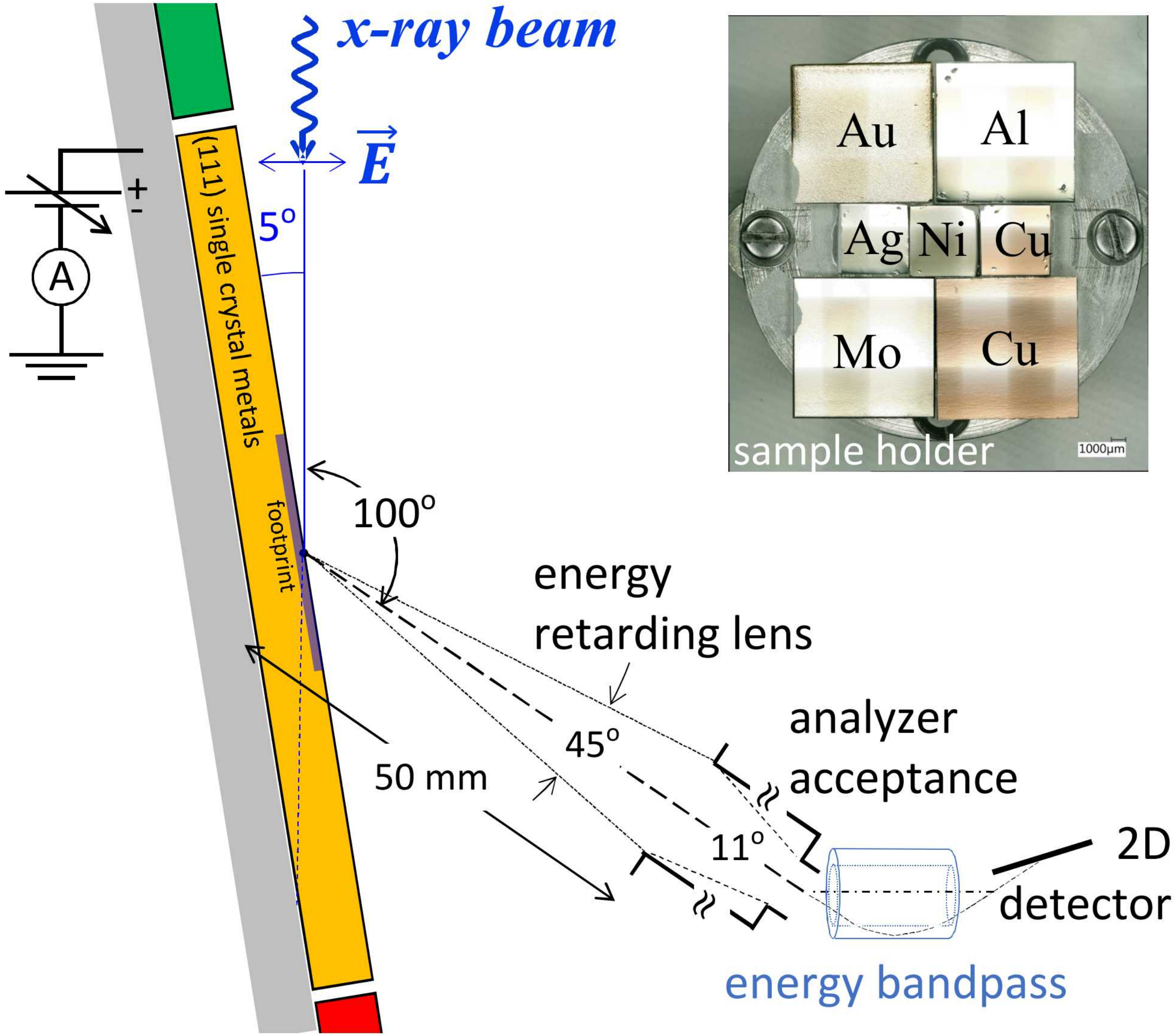
Transmission of the retarding lens is a critical quantity when scanning over such large kinetic energy ranges.

Relative to raw CPS at 2D detector, it does lift the high-energy portion of spectrum.

- Energy dependence of transmission correction is as plotted for E_{pass} = 150 eV.
- Correction linearly varies from 1 to 20 between 0.7 and 15 keV.
- We apply the kinetic energy dependence detailed in "Rubio-Zuazo *et al.*, *Review of Scientific Instruments* **81** 043304 (2010)," which is vetted with measurements and simulations.

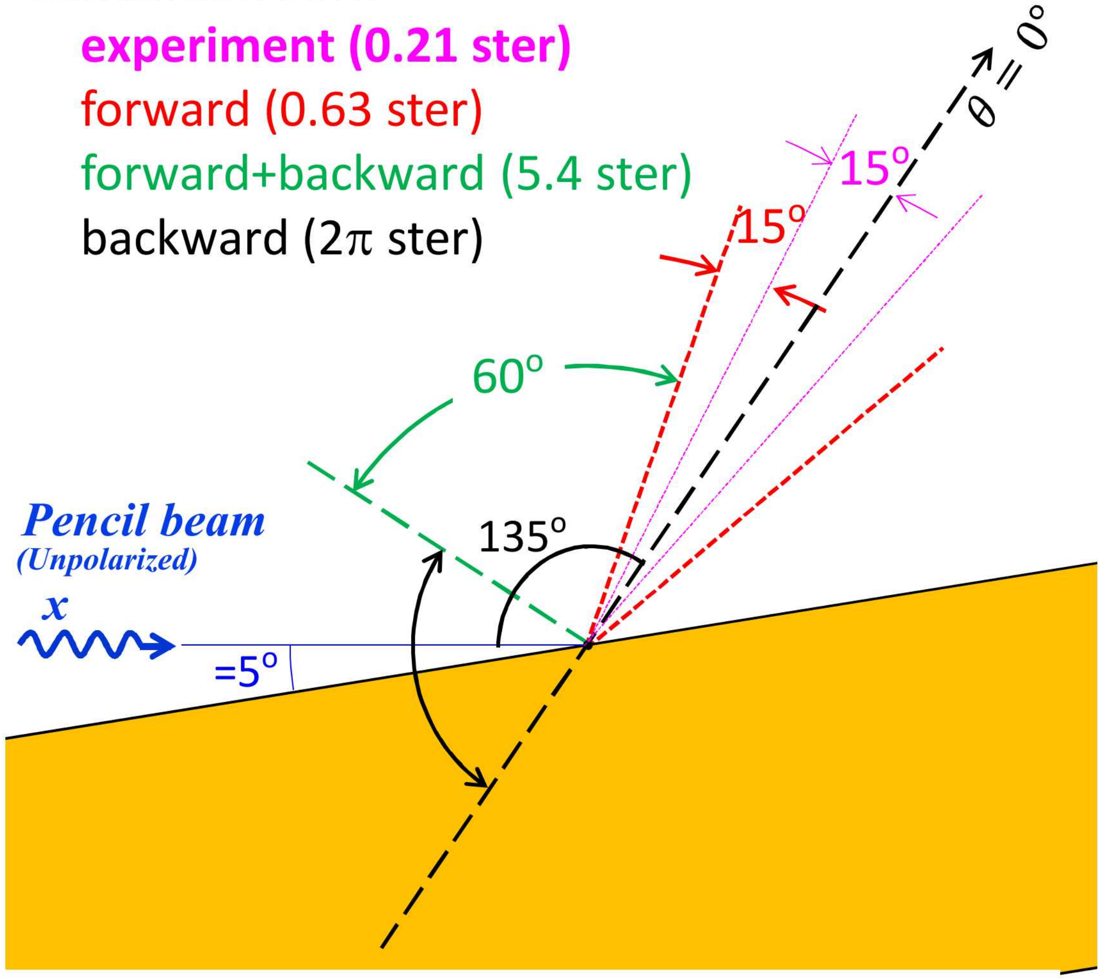


Geometry of experiment at SpLine.

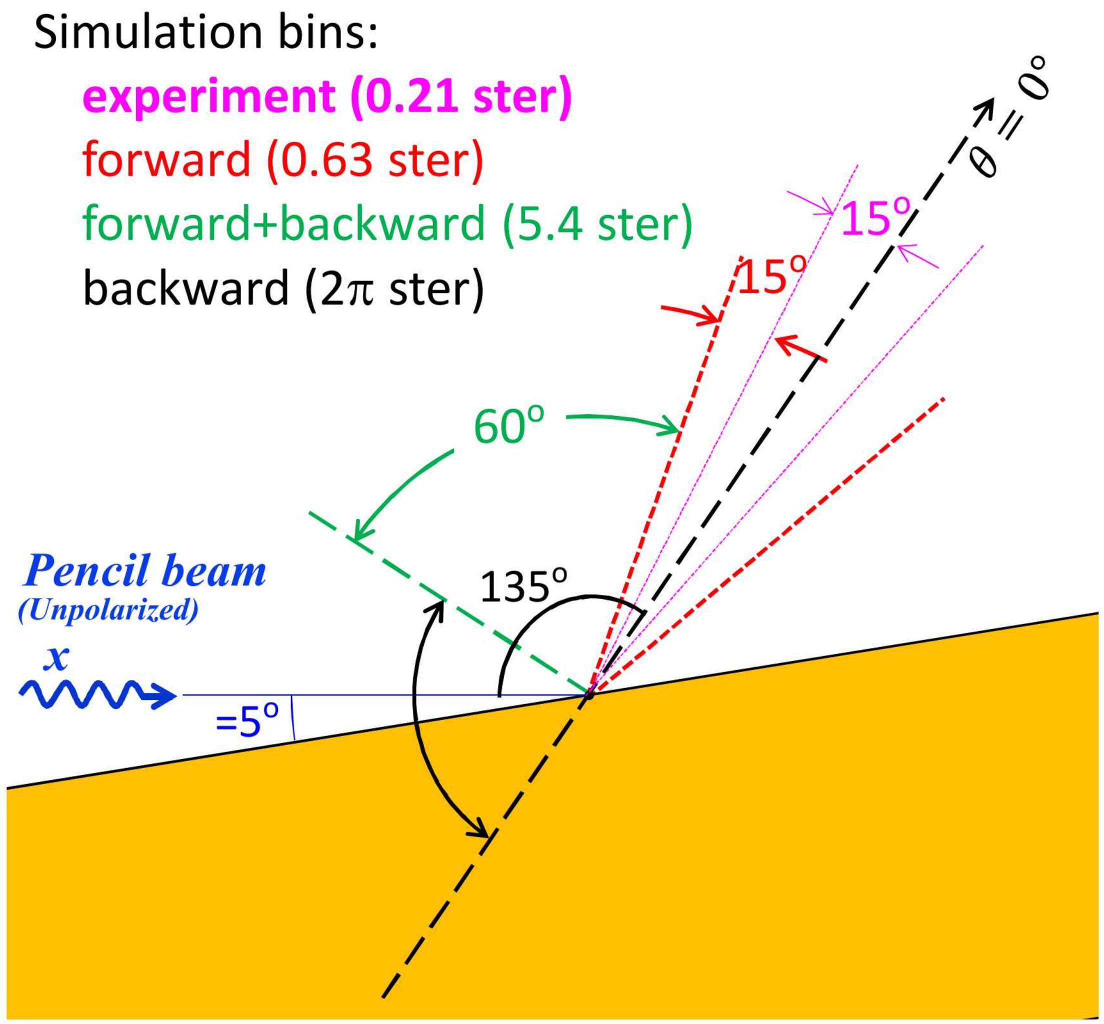


Geometry of Monte-Carlo simulation.

Simulation bins:
- experiment (0.21 ster)
- forward (0.63 ster)
- forward+backward (5.4 ster)
- backward (2π ster)

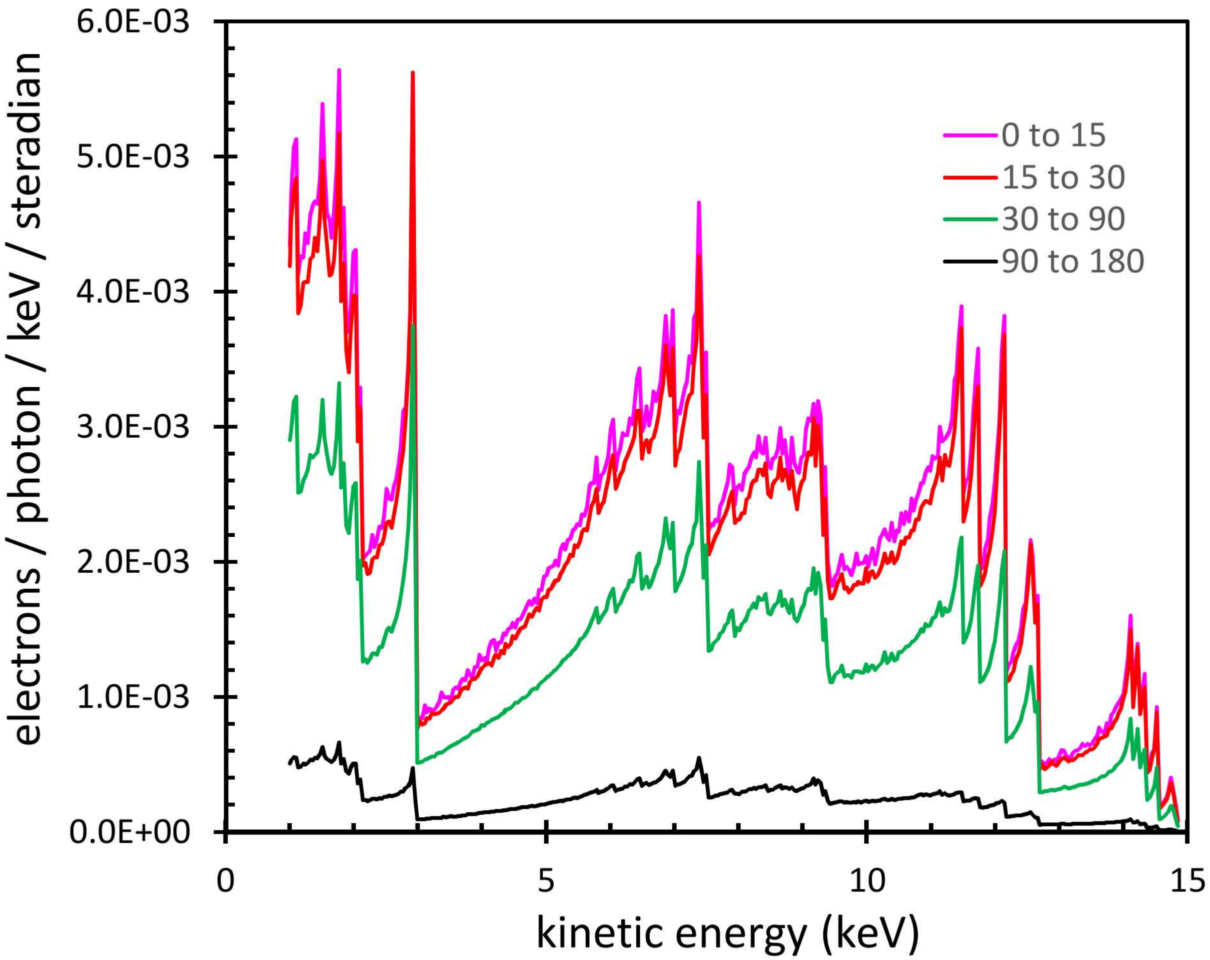


Geometry of Monte-Carlo simulation.



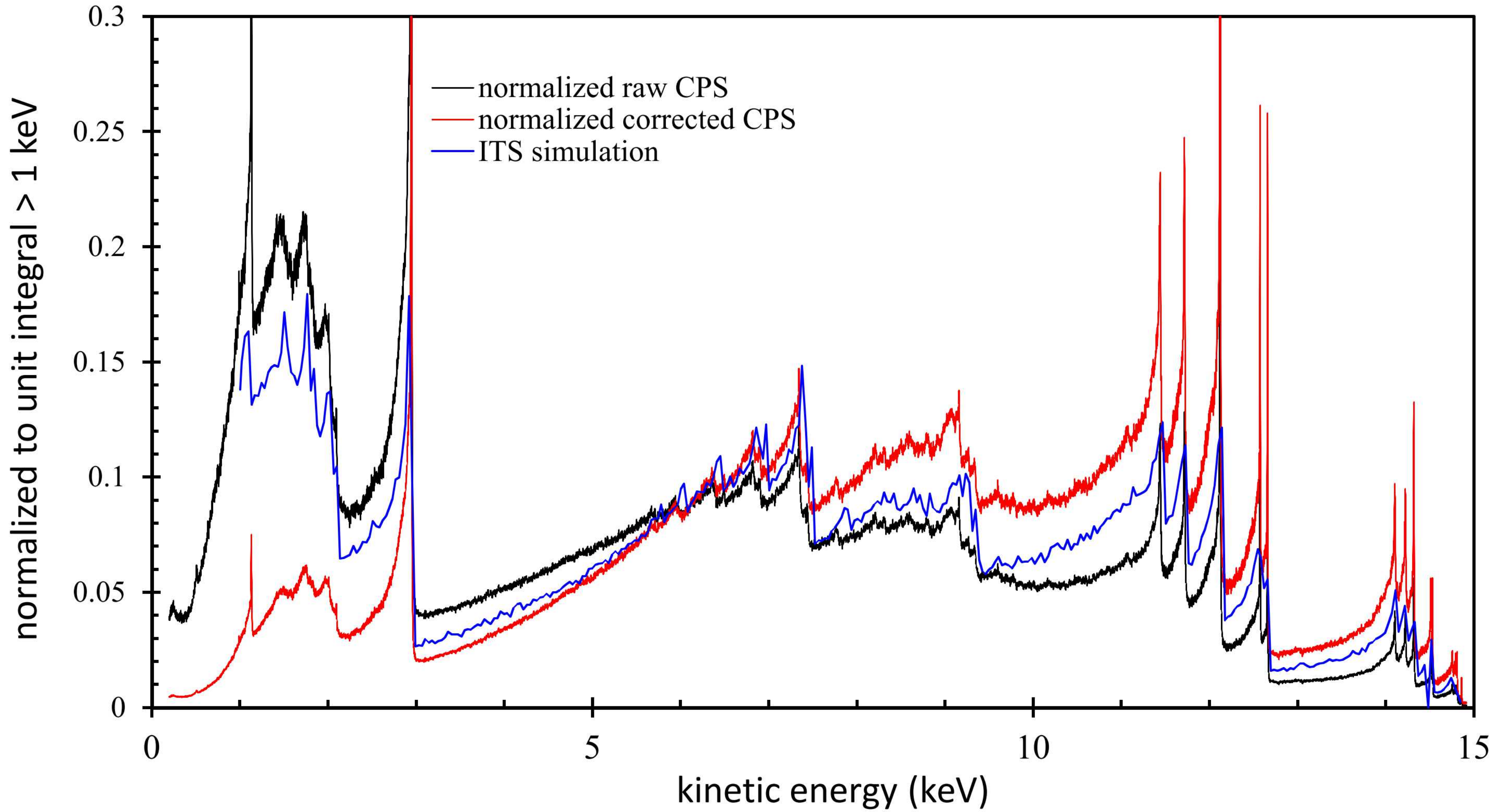
Simulation of 14.87 keV on gold

- Integrates to 0.148 electrons / photon
- dN/dE weakly angle dependent
- Angular bin spectra similar when normalized



Need a simple (and fair) way to compare measurements with simulations.

- Ordinate of CPS measurements are similar to arbitrary units. Abscissae known with great precision.
- Monte Carlo predicts absolute physical units. Matching HAXPES resolution computationally expensive or impractical.
- Normalize spectra so they integrate to unity > 1 keV.
- Raw analyzer counts-per-second (black) are plotted, as well as transmission-corrected CPS (red).



Caveats

- Elastic peaks are not nearly as prominent in simulation.
- Due to 40 eV energy bins.
- 1 eV in measurement.
- Plot cuts off measured $2p_{3/2}$ elastic peak for clarity.

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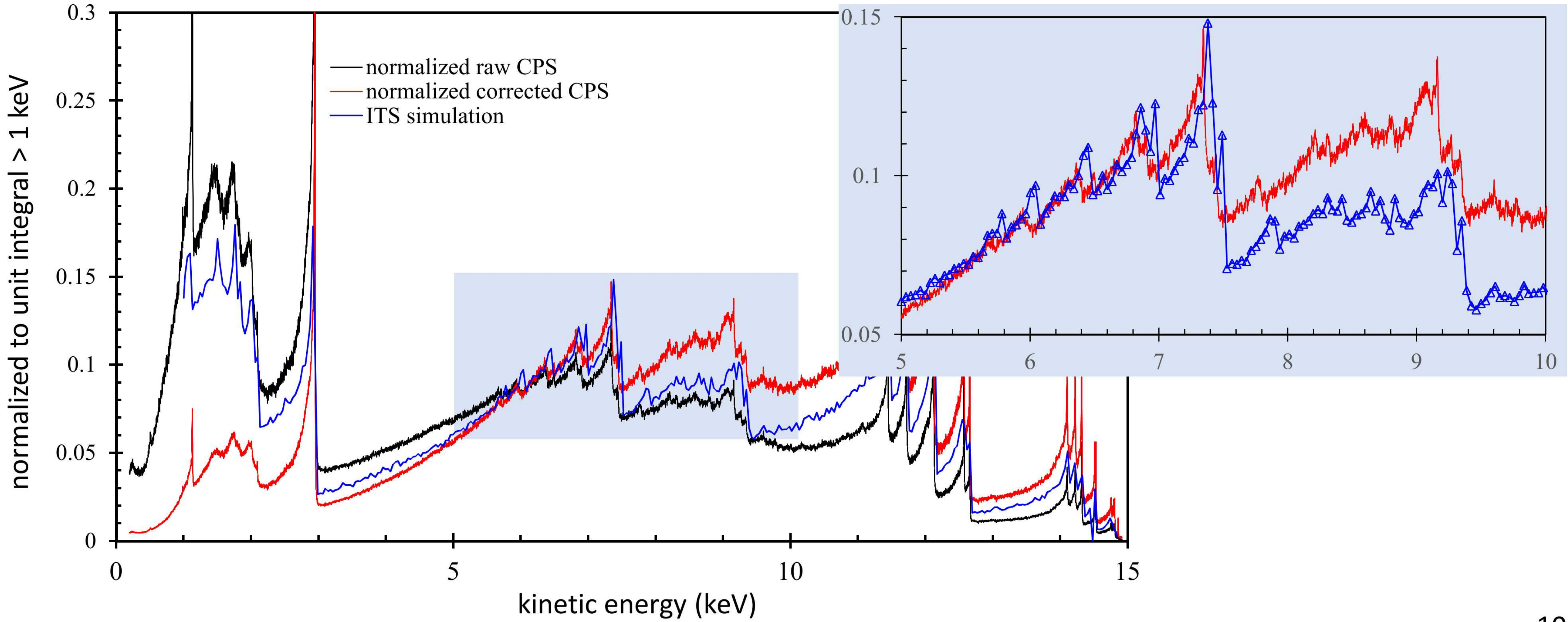
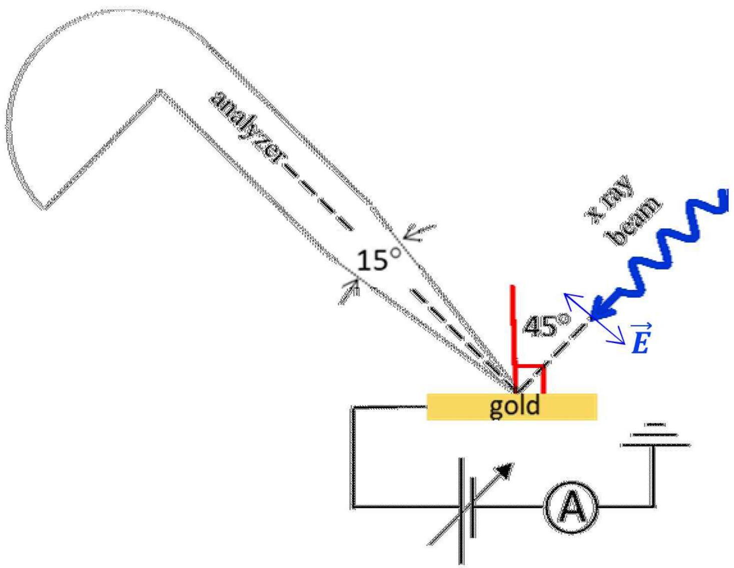


Need HAXPES spectra in units of electrons/ photon



Multiple analyzer's measure the spectral shape (counts)

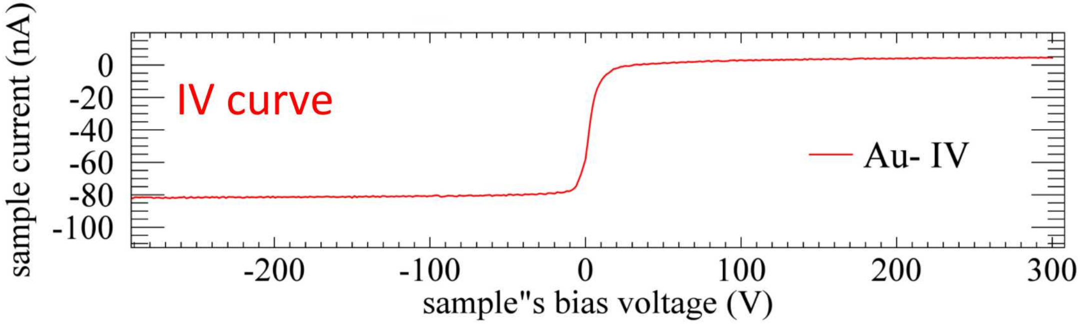
- transmission and throughput differ across electron analyzers
- x-ray flux and angular access to sample differ across beamlines
- Cannot use analyzers in absolute sense [1 count ≠1 electron ??]

Characterize x ray beam:

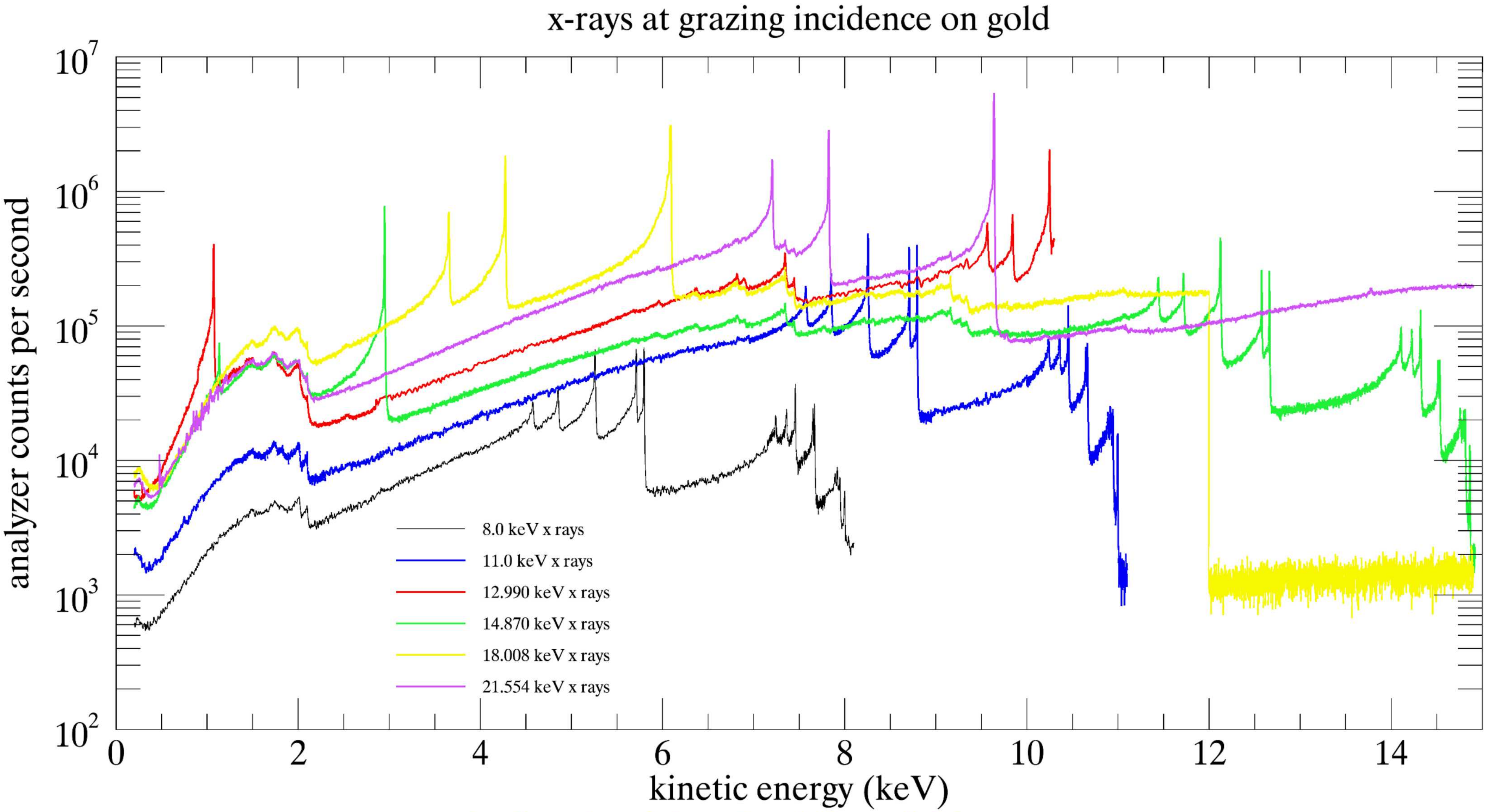
- Measure flux using calorimetry in a sample and/or PIN diodes [recent experiments at NSLS-II (Weiland, Woicik)]
- spectral width is well-known at any beamline

Characterize total electron emission:

- measure sample current from grounded sample
- discriminate true secondaries from high-energy
- [I-V characteristics, at NSLS-2 SST]

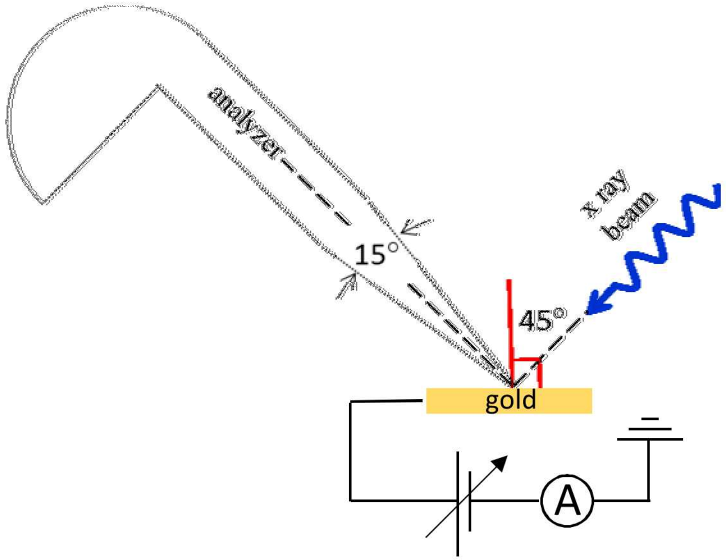


THANK YOU FOR YOUR ATTENTION



Similar measurements were made for Ag, Mo, Cu, Ni and Al

BACKUPS



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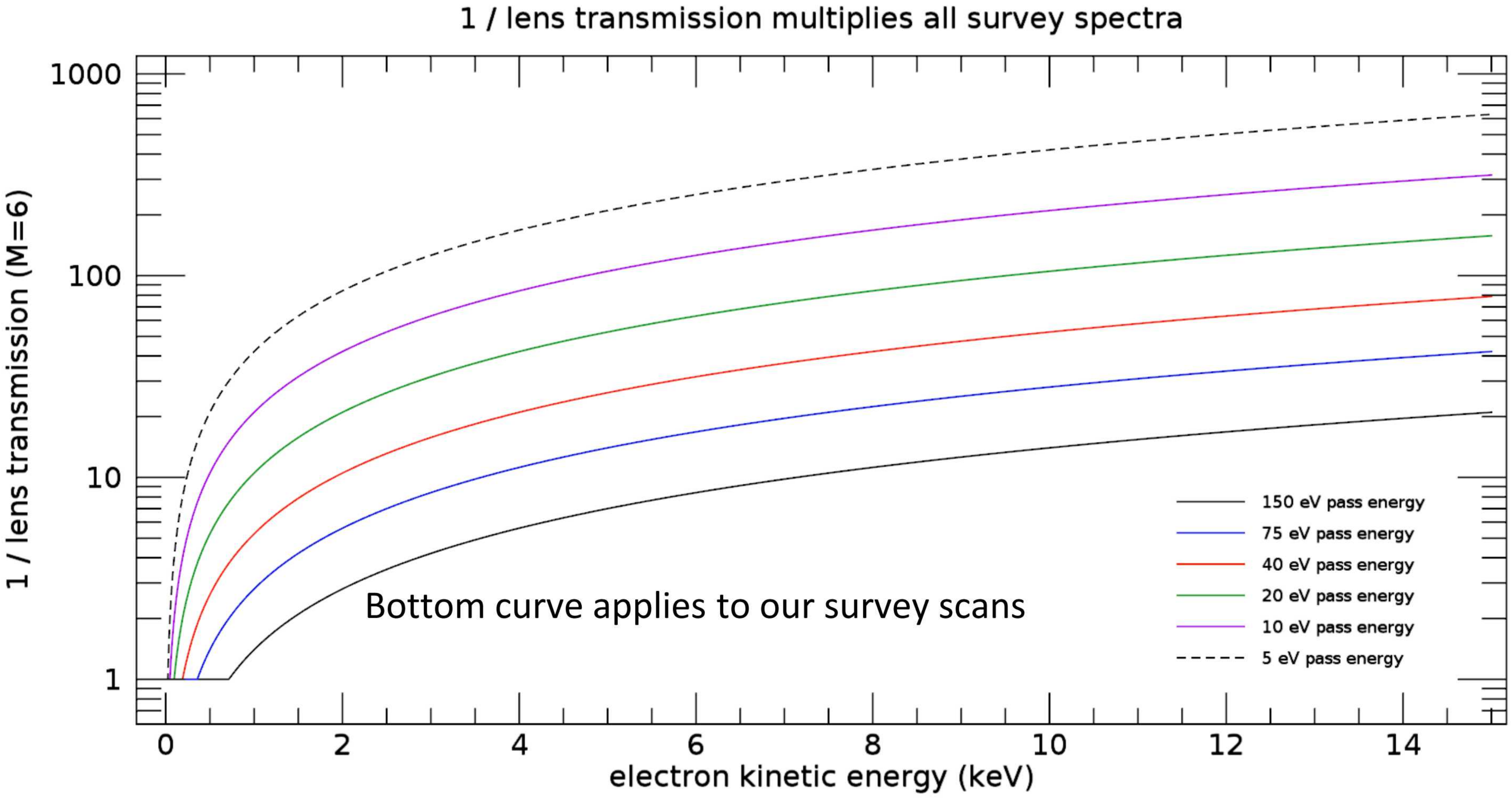
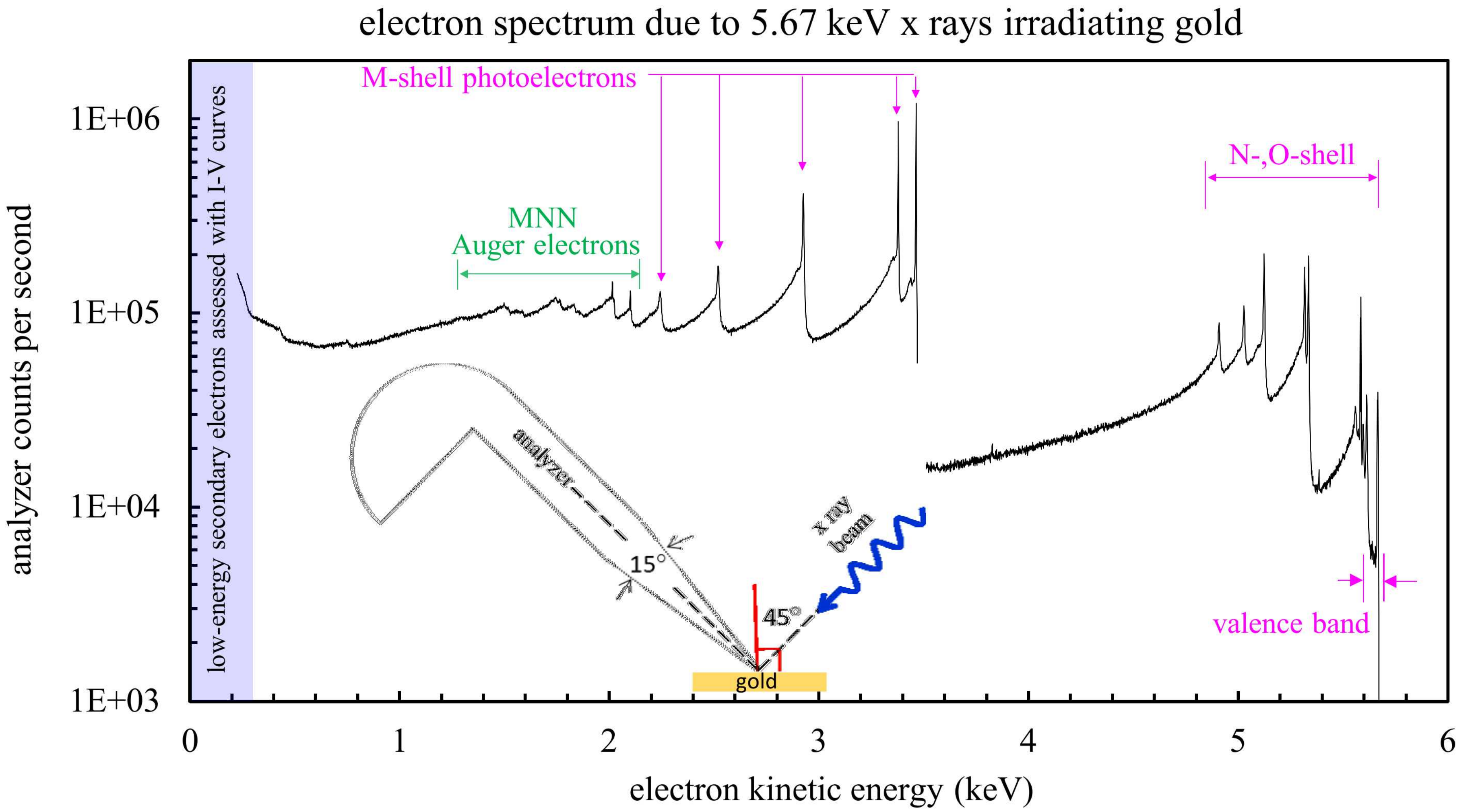
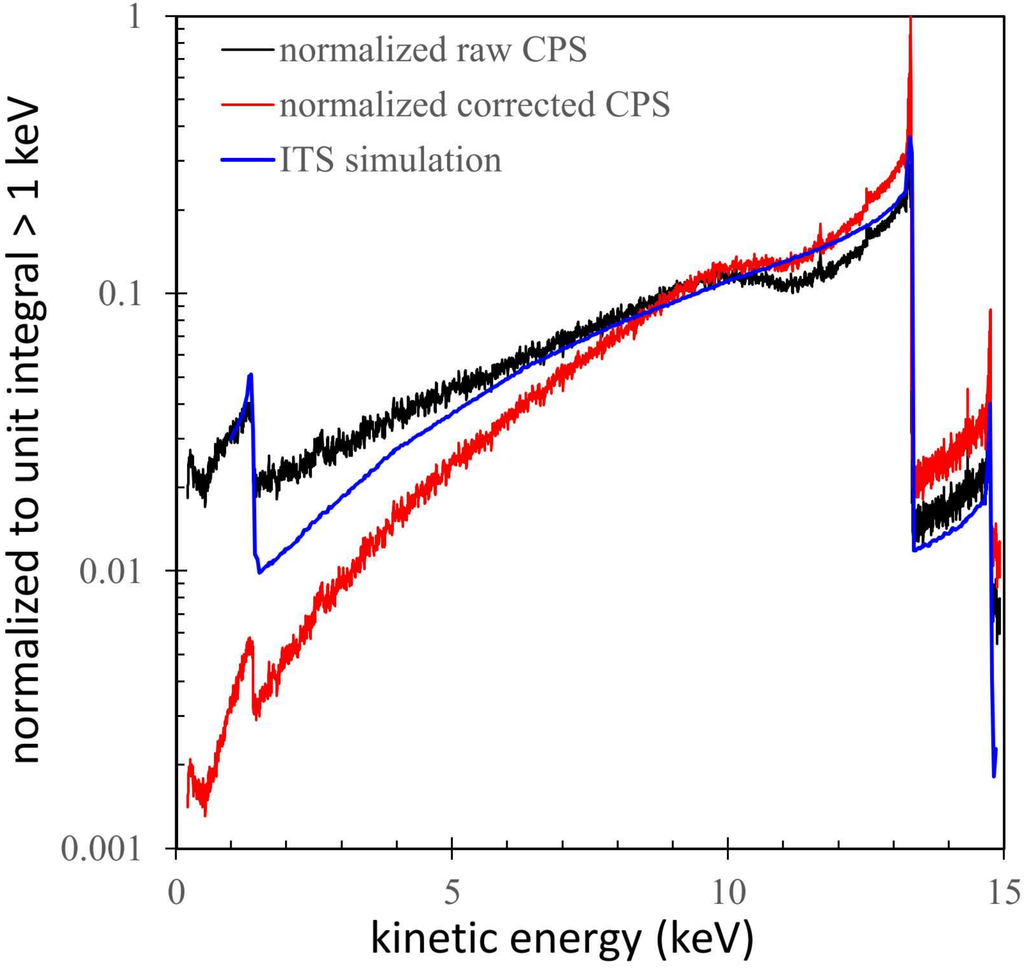




electron spectrum due to 5.67 keV x rays irradiating gold
M-shell photoelectrons
N-,O-shell
MNN
Auger electrons
low-energy secondary electrons assessed with I-V curves
analyzer
15°
x ray
beam
45°
gold
valence band
analyzer counts per second
1E+06
1E+05
1E+04
1E+03
0
1
2
3
4
5
6
electron kinetic energy (keV)

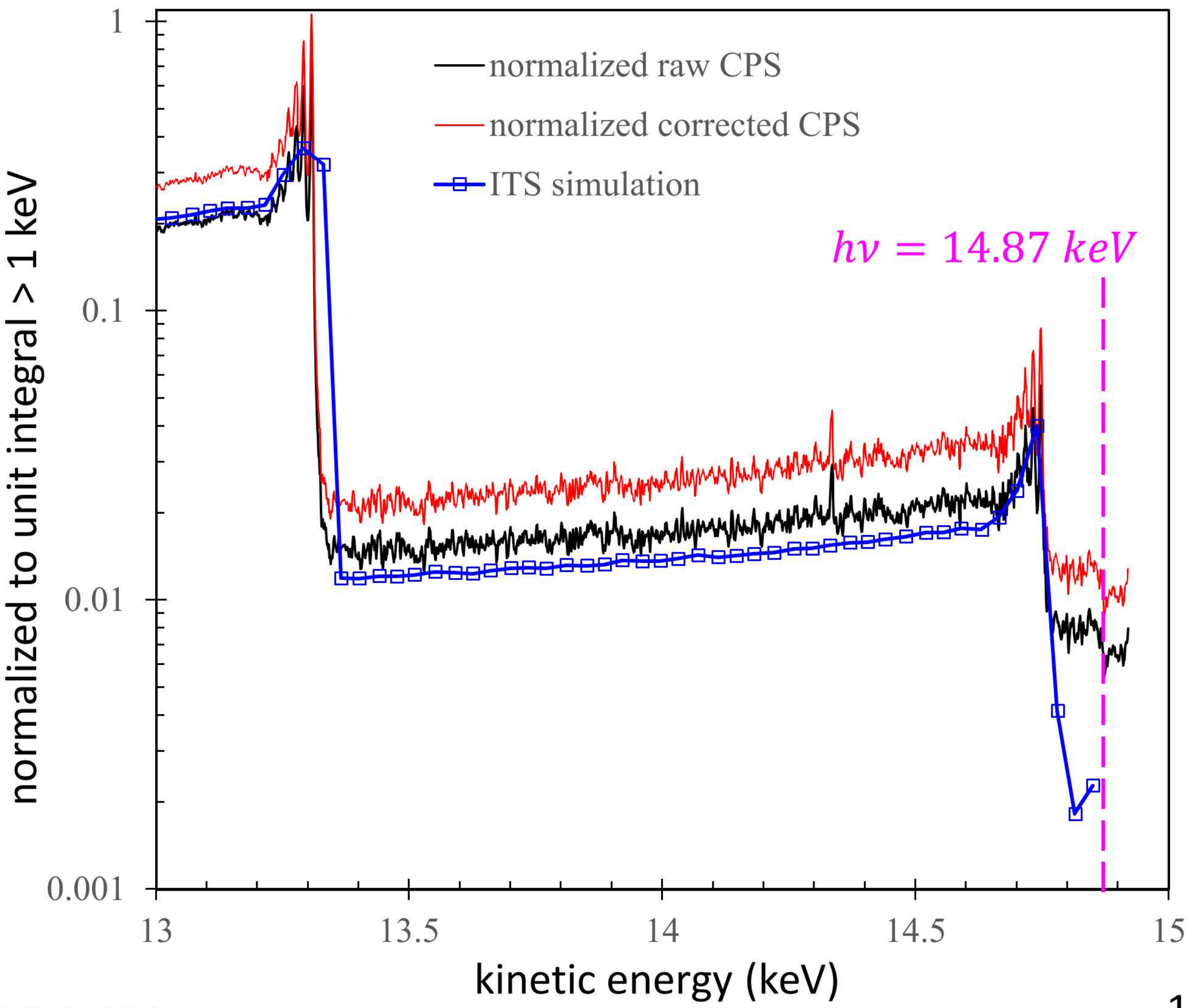
Aluminum full surveys feature almost entirely inelastic processes.

Simulations are integrated over full 2π steradians, due to geometry error. Will need to be redone.



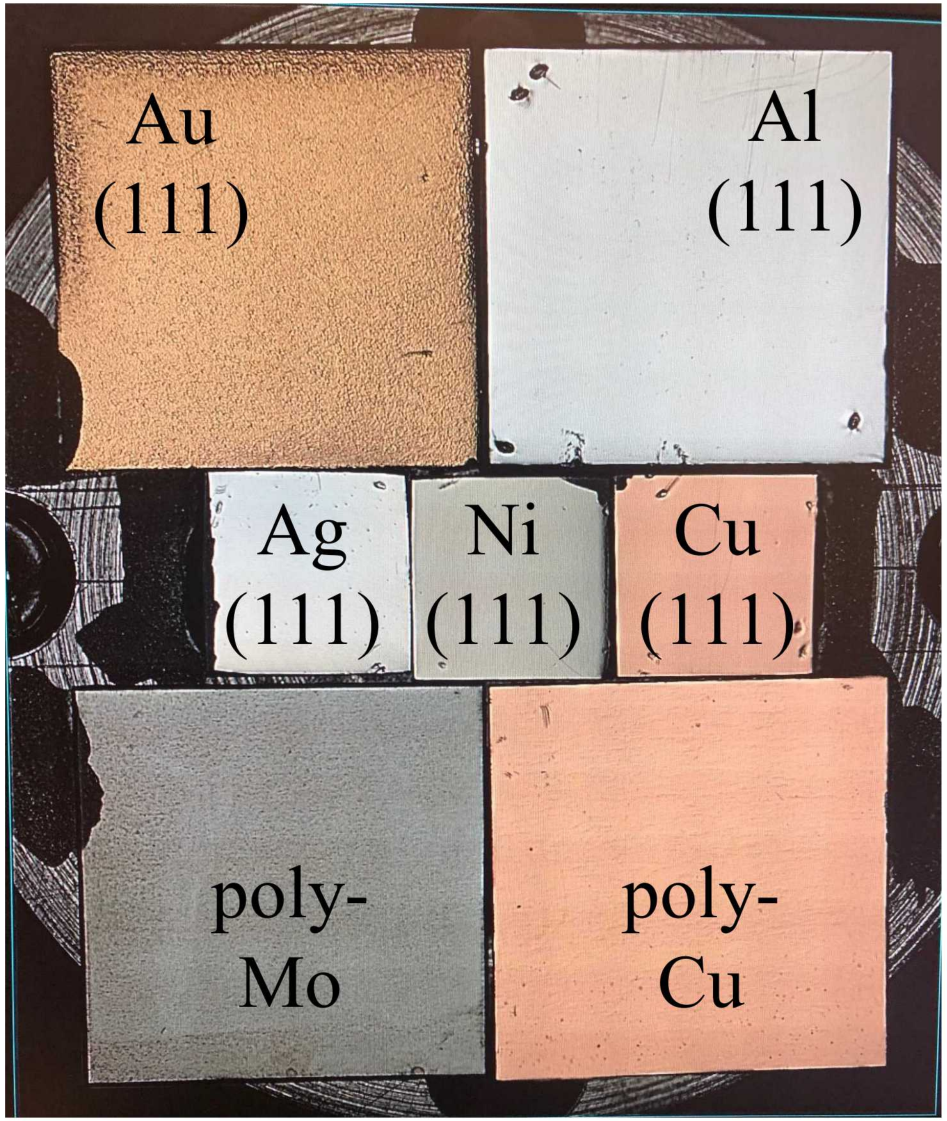
But obvious things like plasmon losses in aluminum are not modeled here.

Simulations use atomistic, not a solid-state model.



SAMPLE HOLDER

before sputtering



after sputtering