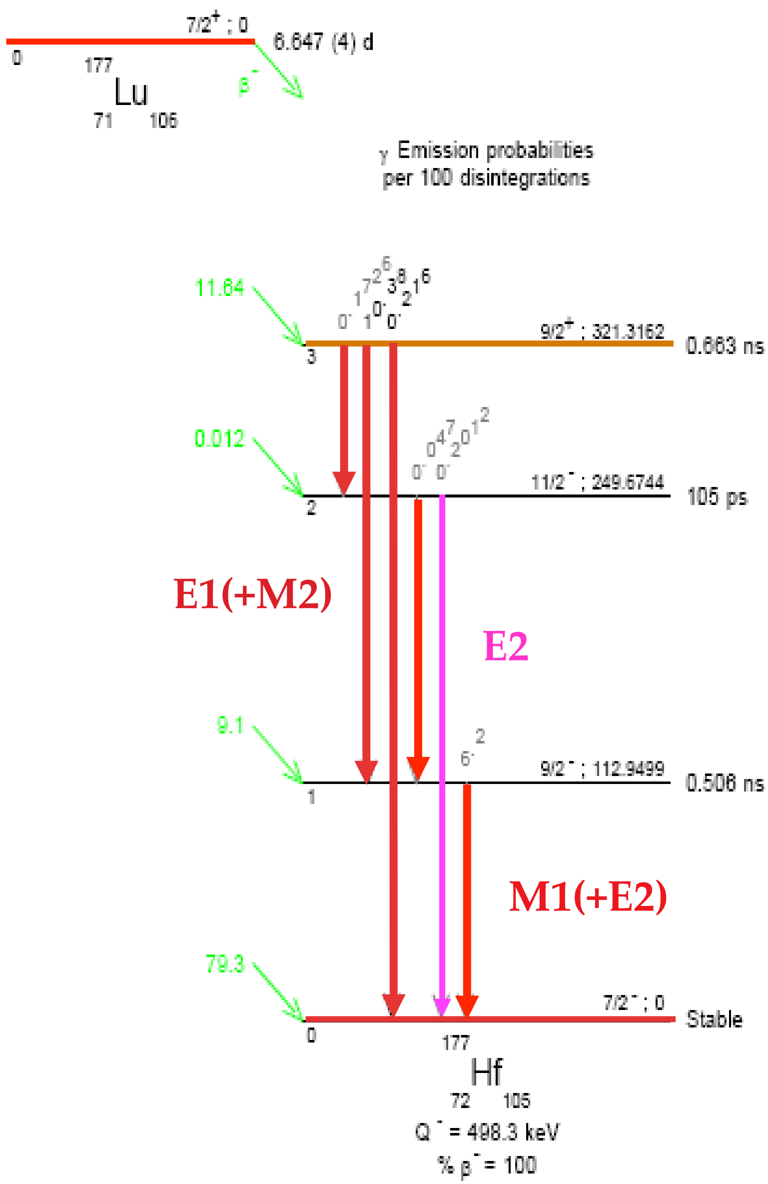


detailed decay scheme is required

- ✓ parent – T1/2, BR
- ✓ daughter – signature radiation energies and emission probabilities – both nuclear & atomic

Reaction cross sections:

- ✓ one or two strongest gamma rays
- ✓ absolute emission probabilities – Iγ/decay
- ✓ mostly nuclear, but ICC and X-rays are needed is some cases

Decay data for medical applications:

- ✓ both the nuclear and atomic data (Auger, Coster-Kronig, etc)

MIRD, TOI, LiveChart, NuDat, ENDF, **JEFF**, JENDL, etc.
ENSDF, DDEP, NUBASE (no spectral data)

There is no magic bullet, both ENSDF & DDEP have Pros and Cons

which one is better – it is difficult to answer – depends on a particular evaluator – **it is a NIGHTMARE to the end-users**

in many of those evaluations no information is provided on what are the data problems and what would be useful to be measured in order to improve them (perhaps with a few exceptions – depend on evaluator's experiences and knowledge)

Nowadays, essentially each applied project that needs reliable and up to date decay data (e.g. our CRP for example) needs to make its own decay data assessment

erosion of expertise in decay data evaluation worldwide, including at IAEA

Decay Data – What is evaluated?

- **Q values -** G. Audi et al, AME2012 – new tables in press – surprises driven by new measurements – don't use end-point energies!

- **Level Properties:** E (ΔE), J^{π}, $T_{1/2}$ ($\Delta T_{1/2}$), BR(Decay mode(s))
  - ✓ E (ΔE) – least-squares fit procedure to ALL available data (not only decay – high-precision reaction data) -> should be used to determine signature radiations, e.g. E_{γ}, E_{β}, E_{α}, …
  - ✓ J^{π} – important when dealing with large decay data schemes -> defines transition multipolarities and ICC
  - ✓ $T_{1/2}$ ($\Delta T_{1/2}$) - in most cases under control (except ^{186m}Re for example), but there is no consistency (recipe) between different evaluations
  - ✓ BR – in many cases only one mode measured, but the second inferred from 100-%BR1; lack of separating EC from β+: %EC+%B=100 in ENSDF or β+=100 in NUBASE -> what is measured and what is deduced

Decay Data – What is evaluated-cont.?

- **Gamma Radiation Properties:** E_γ (ΔE_γ), I_γ (ΔI_γ), Mult., δ ($\Delta\delta$)
  - ✓ E_γ (ΔE_γ) – need to be evaluated in a relation to a particular nuclear level (not only decay – high-precision reaction data, e.g. bent-curve spectrometers); the recommended ones determined from lsq-fit level energies
  - ✓ I_γ (ΔI_γ) – MUST be evaluated. One must consider BR from reactions for weakly populated levels in β/α decay
  - ✓ Mult. – sometime inferred from the decay scheme and from reactions data – important to deduce ICC
  - ✓ δ ($\Delta\delta$) – Must be evaluated. Frequently reactions data must be consulted
  - ✓ careful when dealing with E0 or mixed E0+M1+E2 transitions: simplified approaches use experimental ICC and $I\gamma$(tot); or penetration effect for ICC (mostly for heavy nuclei)

Decay Data – What is evaluated-cont.?

- **Beta Radiation Properties:** E_β (ΔE_β), I_β (ΔI_β)
  - ✓ E_β (ΔE_β) – it is not a discrete, usually maximum and mean energies are deduced from the known decay scheme and decay Q value -> would be useful to provide the full beta spectrum
  - ✓ I_β (ΔI_β) – deduced from intensity balances - > need to look carefully if $I_{\beta+}$ has been measured, usually deduced from the (calculated) $I_{\beta+}$/EC ratio
- **Alpha Radiation Properties:** E_α (ΔE_α), I_α (ΔI_α)
  - ✓ E_α (ΔE_α) – from level energy differences & Qα values; directly measured ones are usually with low uncertainties
  - ✓ I_α (ΔI_α) – both directly and indirectly (from Iγ)
- **Atomic Radiation:**
  - ✓ CE, X-rays, Auger and Coster-Kronig are derived quantities, except ICC for mixed E0+M1+E2 transitions and those affected by penetration

Auger, Coster-Kronig and super-Coster-Kronig

- ✓ low-energy (10 eV–10 keV) electrons – short range (a few nm to 1 μm)
- ✓ commonly emitted by radionuclides that decay by EC, CE or IT

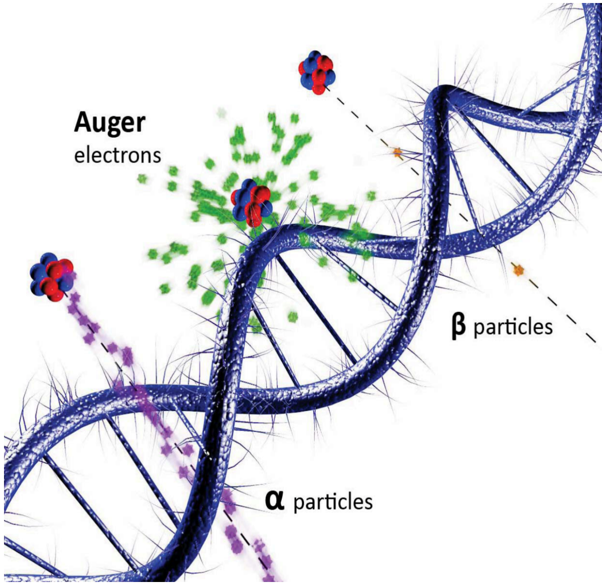
INVITED COMMENTARY

Cancer Therapy with Auger Electrons: Are We Almost There?

Amin I. Kassis, PhD
Harvard Medical School
Boston, Massachusetts

J. Nucl. Med. 44 (2003)

- ✓ high toxicity – highly localized energy deposition in a small volume
- ✓ availability of many radionuclides with variable physical half-life
- ✓ emission of gamma rays - useful for imaging

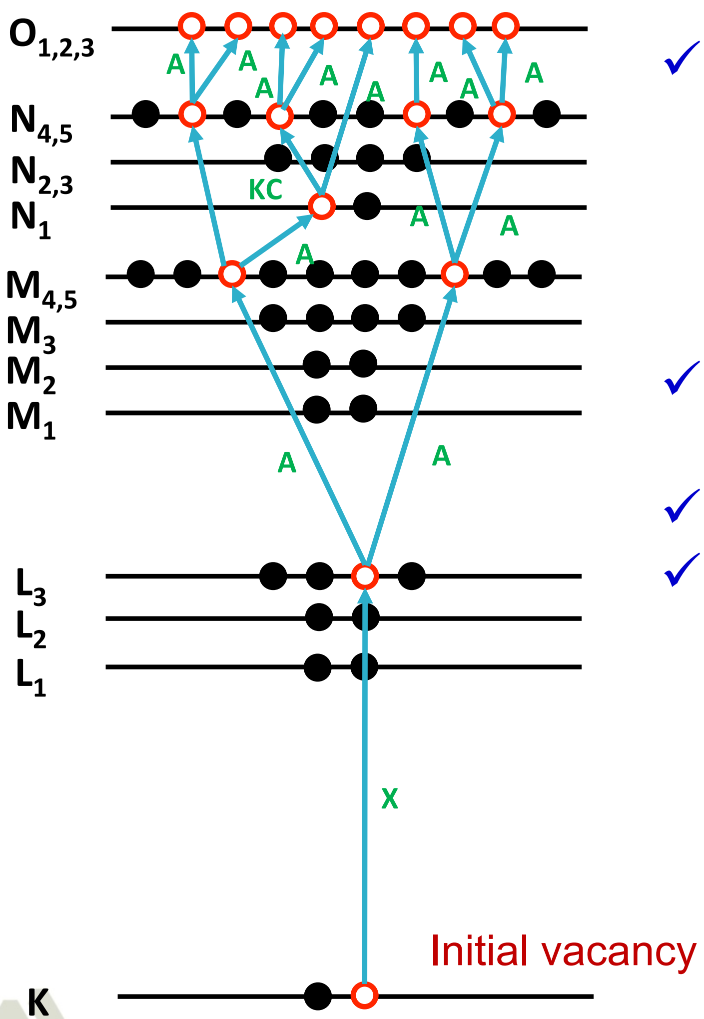


more than 100 Auger emitters
popular ones: ^{99m}Tc, ^{111}In, ^{123,125}I, ^{67}Ga, ^{193m}Pt
new emerging ones: ^{117,119}Sb, ^{165}Er

Atomic relaxation and vacancy transfer

A vacancy cascade in Xe

From M.O. Krause, J. Phys. Colloques, 32 (1971) C4-67



- ✓ **Full relaxation of an initial inner shell vacancy creates vacancy cascade involving X-ray (<u>Radiative</u>) and Auger as well as Coster-Kronig (<u>Non-Radiative</u>) transitions**
- ✓ **Many possible cascades for a single initial vacancy**
- ✓ **Typical relaxation time ~10^{-15} seconds**
- ✓ **Many vacancy cascades following a single ionisation event!**

ENSDF (RADLIST) & DDEP (EMISSION) only initial vacancy in the K shell

courtesy to T. Kibedi, ANU

Existing calculations & approaches

	RADAR	DDEP	Eckerman & Endo (2007)	Howell (1992)	Stepanek (2000)	Pomplun (2012)
Nuclear decay data	ENSDF	DDEP	ENSDF	ENSDF	ENSDF	ICRP38
Conversion coefficients	HsIcc	RpIcc/BrIcc	RpIcc, 1978 Band	RpIcc	2000 Stepanek	HsIcc, 1971 Dragoun, 1976 Band
Electron Capture Ratios	1971 Gove & Martin	1995 Schönfeld	1977 Bambynek	1971 Gove & Martin, 1970Martin	1971 Gove & Martin, 1970Martin	1971 Gove & Martin
Atomic transition rates	1972 Bambynek, RADLST	1974 Scofield, 1995 Schönfeld & Janßen, 2006 Be et al., EMISSION	1991 Perkins, EDISTR04	1979 Chen, 1972/1975 McGuire, 1983 Kassis, 1974 Scofield, 1974 Manson & Kenedy	1991 Perkins	1979 Chen, 1972/1975 McGuire, 1970 Storm & Israel, 1979 Krause
Atomic transition energies	1970 Bearden & Burr, Neutral atom	1977 Larkins, Semi-empirical	1991 Perkins, Neutral atom	Z/Z+1 (Auger), Neutral atom (X-ray)	Dirack-Fock calculation	1991 Desclaux, Dirack-Fock calculation
Vacancy propagation	**Deterministic**	**Deterministic**	**Deterministic/MC** (+++)	**Monte Carlo** with charge neutralization	**Monte Carlo**	**Monte Carlo**

courtesy to T. Kibedi, ANU

Existing calculations
Auger electron yield per nuclear decay

	RADAR	DDEP	Eckerman & Endo (2007)	Howell (1992)	Stepanek (2000)	Pomplun (2012)
^{99m}Tc (6.007 h)	0.122	0.13	4.363	4.0		2.5
^{111}In (2.805 d)	1.136	1.16	7.215	14.7	6.05	
^{123}I (13.22 h)	1.064	1.08	13.71	14.9		6.4
^{125}I (59.4 d)	1.77	1.78	23.0	24.9	15.3	12.2
^{201}Tl (3.04 d)	0.773	0.614	20.9	36.9		
Vacancy propagation	**Deterministic**	**Deterministic**	**Deterministic/MC** (+++)	**Monte Carlo** with charge neutralization	**Monte Carlo**	**Monte Carlo**

courtesy to T. Kibedi, ANU

BrIccEmis – Monte Carlo approach for vacancy creation and propagation

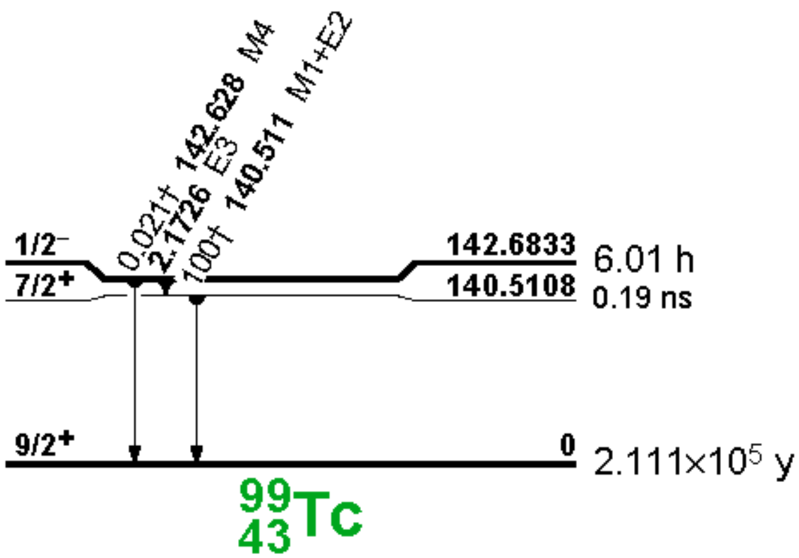
- Initial state: neutral isolated atom
- Nuclear structure data from ENSDF
- Electron capture (EC) rates: Schönfeld (1998Sc28)
- Internal conversion (IC) coefficients: BrIcc (2008Ki07)
- Auger and X-ray transition rates: EADL (1991 Perkins)
  Calculated for single vacancies!
- Auger and X-ray transition energies: RAINE (2002Ba85)
  Calculated for actual electronic configuration!
- Vacancy creation and relaxation from EC and IC are treated independently
- *Ab initio* treatment of the vacancy propagation:
  - Transition energies and rates evaluated on the spot
  - Propagation terminated once the vacancy reached the valence shell

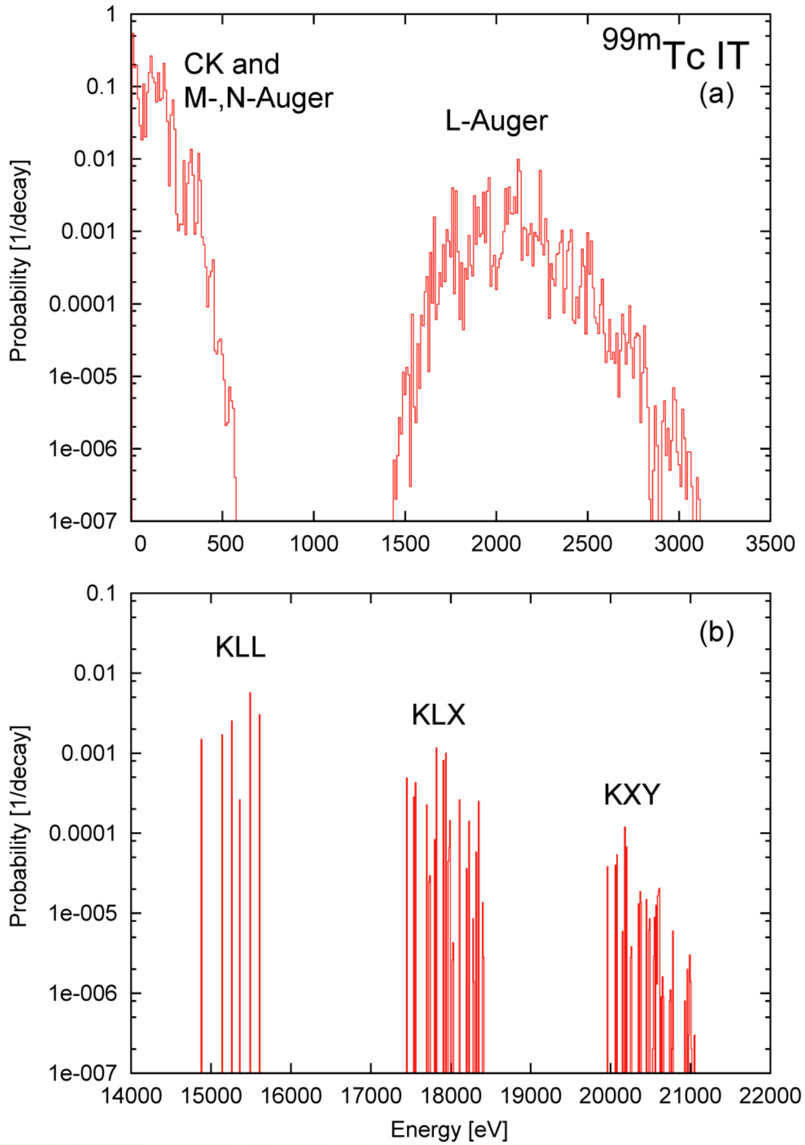
- integration of nuclear and atomic data using an ENSDF formatted file (platform independent) – essentially completed

^{99m}Tc atomic radiations

BrIccEmis: spectrum from 10 M simulated decay events

2.1726 keV below L-shell BE





No experimental spectrum to compare with

courtesy to T. Kibedi, ANU

^{99m}Tc atomic radiations – Auger electrons

	DDEP	BrIccEmis
KLL	[14.86:15.58] ***1.49E-2***	15.37 ***1.48E-2***
KLX	[17.43:18.33] ***2.79E-3***	17.85 ***5.58E-3***
KXY	[19.93:21.00] ***2.8E-4***	20.27 ***5.07E-4***
K-total	***2.15E-2***	16.15 ***2.08E-2***
CK LLM		***2.08E-2*** 0.054
CK LLX		0.144 ***9.48E-3***
LMM		2.016 ***9.02E-2***
LMX		2.328 ***1.41E-2***
LXY		2.654 ***6.07E-4***
L-total	[1.6:2.9] ***1.089E-1***	1.765 ***1.24E-1***

courtesy to T. Kibedi, ANU

^{99m}Tc atomic radiations – X-rays

	DDEP	BrIccEmis
$K\alpha_1$	18.3672 ***4.21E-2***	18.421 ***4.05E-2***
$K\alpha_2$	18.251 ***2.22E-2***	18.302 ***2.13E-2***
$K\beta$	20.677 ***1.30E-2***	20.729 ***1.18E-2***
L	[2.134:3.002] ***4.82E-3***	2.466 ***4.72E-3***
M		0.263 ***7.83E-4***
N		0.047 ***8.73E-1***

courtesy to T. Kibedi, ANU

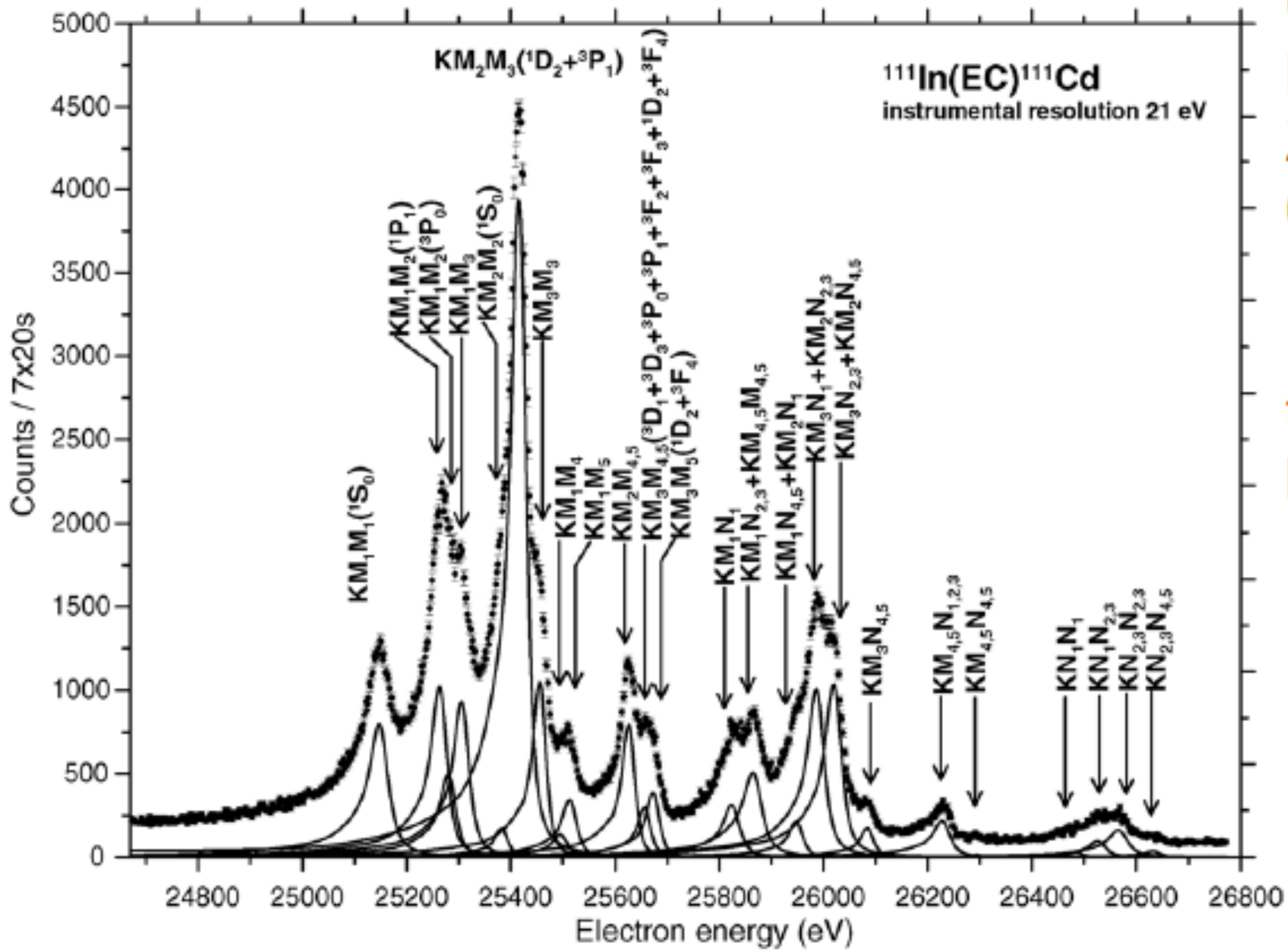
^{99m}Tc atomic radiations – Auger electrons

	DDEP	BrIccEmis
CK MMX		0.104 ***7.10E-1***
MXY		0.170 ***1.10E+0***
Super CK NNN		0.014 ***5.36E-1***
CK NNX		0.012 ***8.45E-1***
Total yield Auger electron per nuclear decay	***0.13***	***3.37***

courtesy to T. Kibedi, ANU

Available Experimental Data

A. Inoyatov et al. / Journal of Electron Spectroscopy and Related Phenomena 151 (2006) 193–198



Only a handful data on KMM Auger-electrons: Z=25, 26, 36, 46, 54, 62, 69, 78, 84

Need to extend theoretical models beyond L-shell

light- source facilities could be very useful – they have a big user community, but often lacking expertize in NP

courtesy to T. Kibedi, ANU

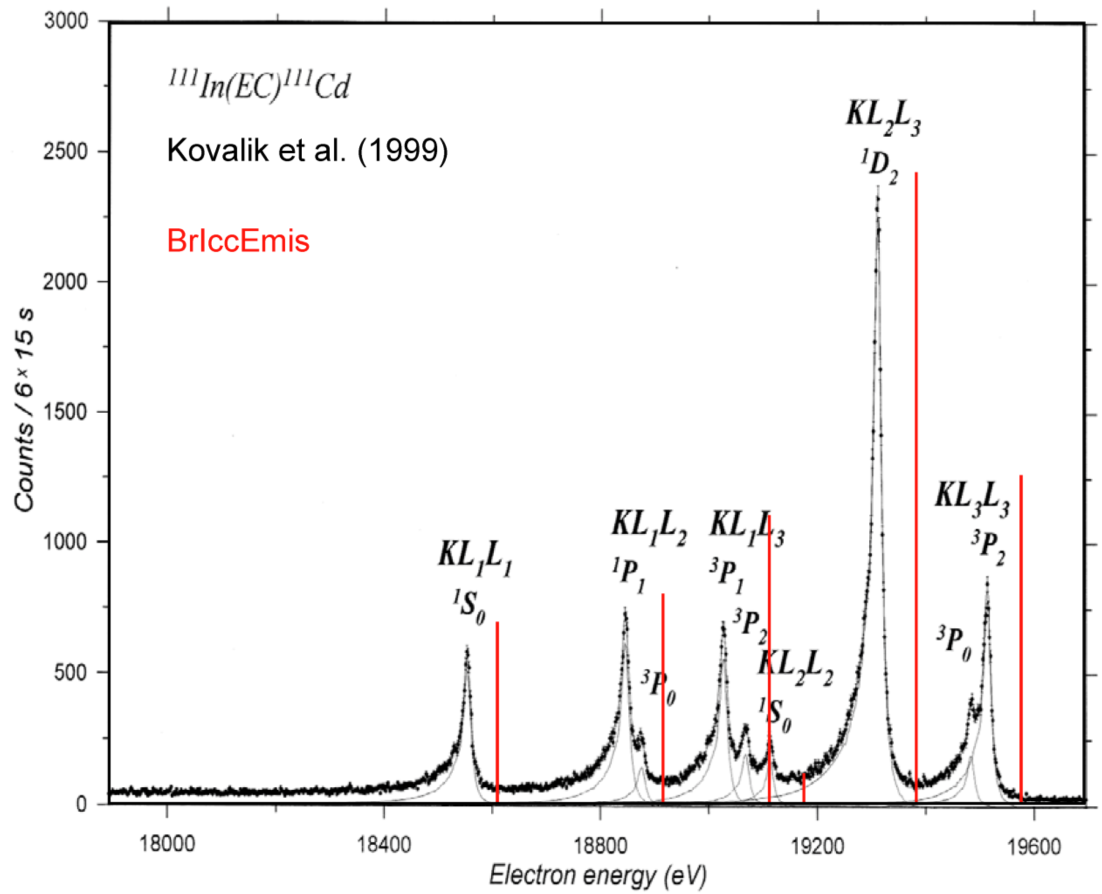
^{111}In – experiment vs calculation

A. Kovalik, et al., J. of Electron Spect. and Rel. Phen. **105** (1999) 219

- ESCA; FWHM = 7 eV
- Calculated energies are higher
- $KL_2L_3(^1D_2)$ energy (eV):

19319.2(14)	Experiment Kovalik (1999)
19308.1	Semi-empirical Larkins (1979La19)
19381	RAINE (2002Ba85)

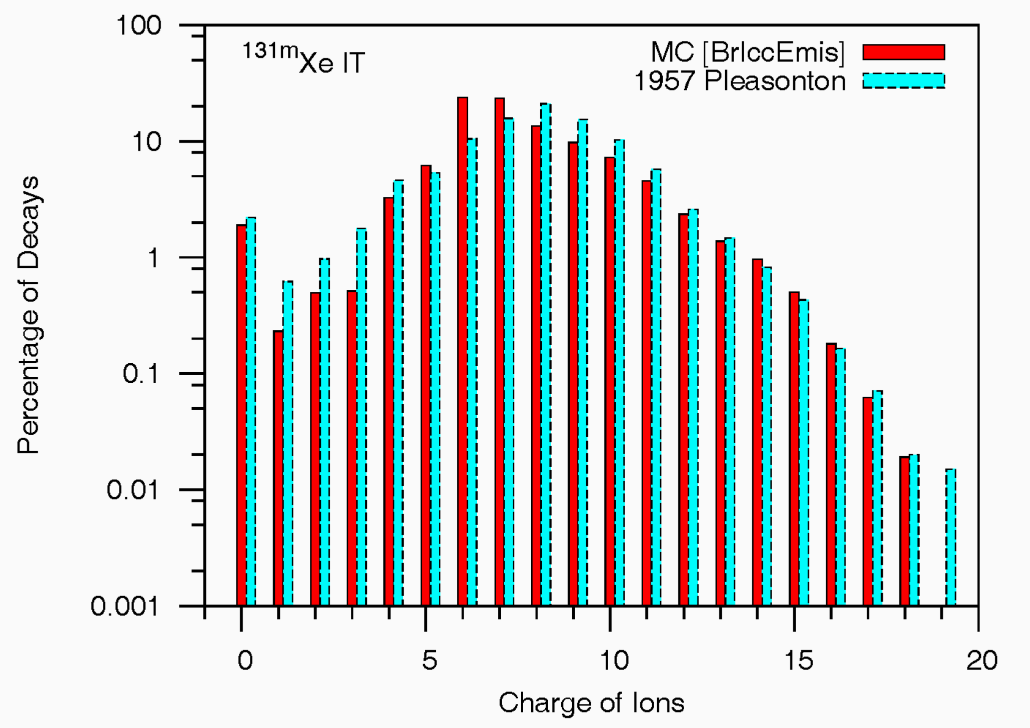
- Multiplet splitting could not be reproduced in JJ coupling scheme
- Similar discrepancies have been seen in other elements (Z=47, Kawakami, Phys. Lett **A121** (1987) 414)



courtesy to T. Kibedi, ANU

^{131m}Xe IT – charge state at the end of atomic relaxation

- Only a handful of measurements exist for ionization by nuclear decay
- ^{131m}Xe: F. Pleasonton, A.H. Snell, Proc. Royal Soc. (London) **241** (1957) 141
- ^{37}Ar: A.H. Snell, F. Pleasonton, Phys. Rev. **100** (1955) 1396
- Good tool to asses the completeness of the vacancy propagation
- BrIccEmis: mean value is lower by ~0.7-1.0 charge



courtesy to T. Kibedi, ANU

BrIccEmis

- Reads the ENSDF file, evaluates absolute decay intensities of EC, GAMMA, CE and PAIR transitions
- Simulates a number (100k-10M) radioactive decays followed by atomic relaxation
- Electron configurations and binding energies stored in memory (and saved on disk). New configurations only calculated if needed. (^{55}Fe: 15 k, ^{201}Tl: 1300k)
- Emitted atomic radiations together with shells involved stored like histories in large files (several Gb)
- Separate files for X-rays and Auger electrons
- Smaller programs to sort/project energy spectra, produce detailed reports

Near-future Plans

- BrIccEmis: calculation intensive approach (hours to days)
- RelaxData (under development):
  - ✓ Nuclear decay event (EC or CE) produces a SINGLE INITIAL vacancy
  - ✓ Considering a single atomic vacancy the relaxation process independent what produced the vacancy
  - ✓ Compile a database of atomic radiation spectra for
    - produced by a single initial vacancy on an atomic shell
    - Carry out calculations of all elements and shells
  - ✓ Example: ^{55}Fe EC, 7 shells for Z=25 and 26, calculated in a couple of hours (1 M each shell)
  - ✓ Replace EADL fixed rates and binding energies from RAINE with GRASP2k/RATIP calculations
- BrIccRelax (under development): Evaluate primary vacancy distribution and construct atomic spectra from the data base (20 seconds for ^{55}Fe EC)

Need to be under the CRP umbrella – invite T. Kibedi to join the CRP
Developed tools at the IAEA medical portal – must be the focal point for data
Beneficial to future medical isotopes CRP's

courtesy to T. Kibedi, ANU

Proposal

IAEA-NDS Medical Portal
www.iaea-nds.ord/

repository for recommended both reactions (already there) & decay data
DD: have to be in the MIRD for the end users

Repository

- ✓ ENSDF-like file with the evaluated data
- ✓ DDEP-like file with the description of the evaluation
- ✓ Atomic radiations (X-rays & Auger) added on in house (development needed – need to bring T. Kibedi (ANU) onboard as suggested by ALN)
- ✓ Final tables etc. are easy to be created, similar to the safeguard data development

courtesy to T. Kibedi, ANU