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Atomic radiations in nuclear decay

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Outline

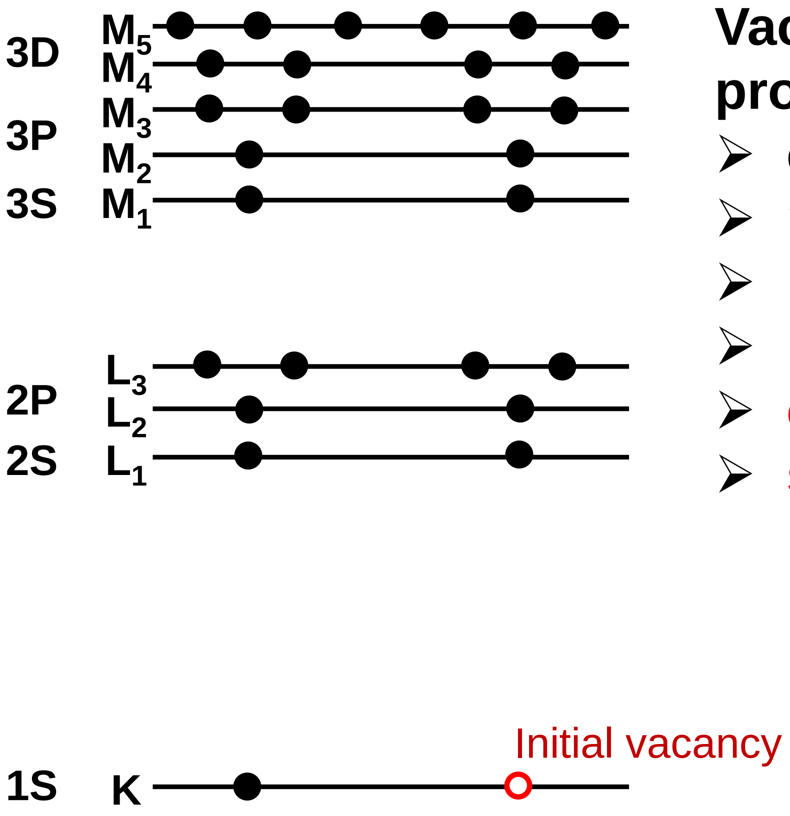
Talk is largely based on

<u>Kålmån Robertson</u> (ANU) , Honours project, October 2010 (literature review, new model of Non-radiative transition rates)

<u>Boon Quan Lee</u> (ANU), special research project (BrIccEmission)

- **Radiative and Non-radiative atomic transitions in nuclear decay**
- **Nuclear and atomic data**
- **Existing programs to evaluate atomic radiations**
- **New model to evaluate Non-radiative transition energies and rates**
- **Future directions**

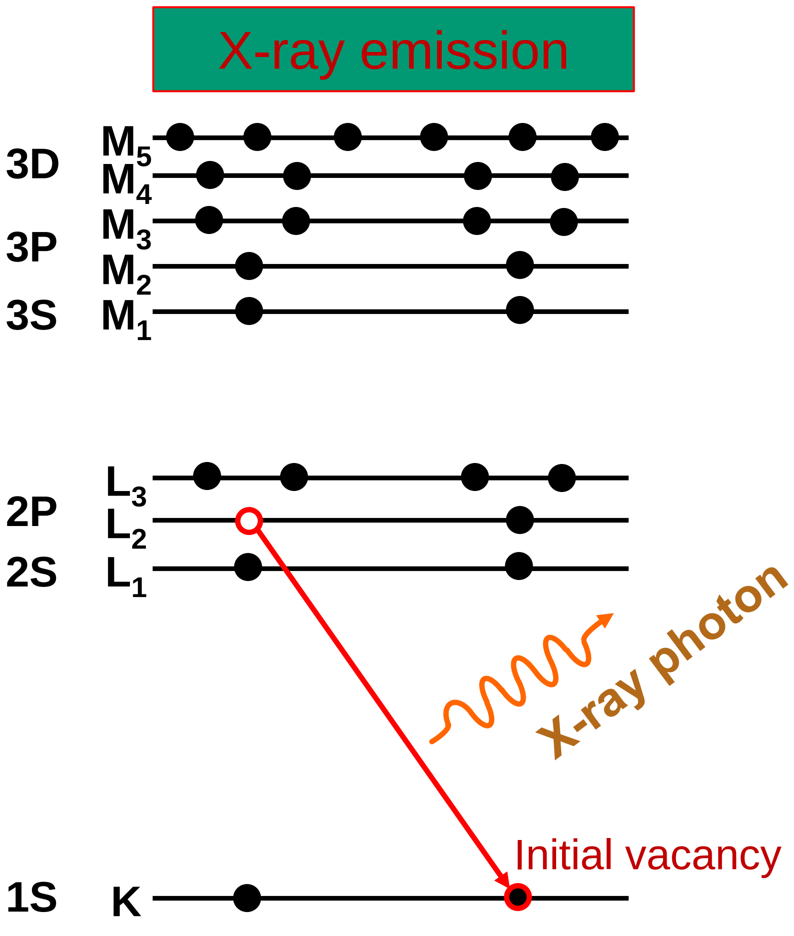
Atomic radiations - Basic concept



Vacancies on the inner-shell can be produced by

- **electron impact**
- **photo ionization**
- **ion-atom collision**
- **internal conversion**
- **electron capture**
- **secondary processes accompanying β-decay or electron capture**

Atomic radiations - Basic concept



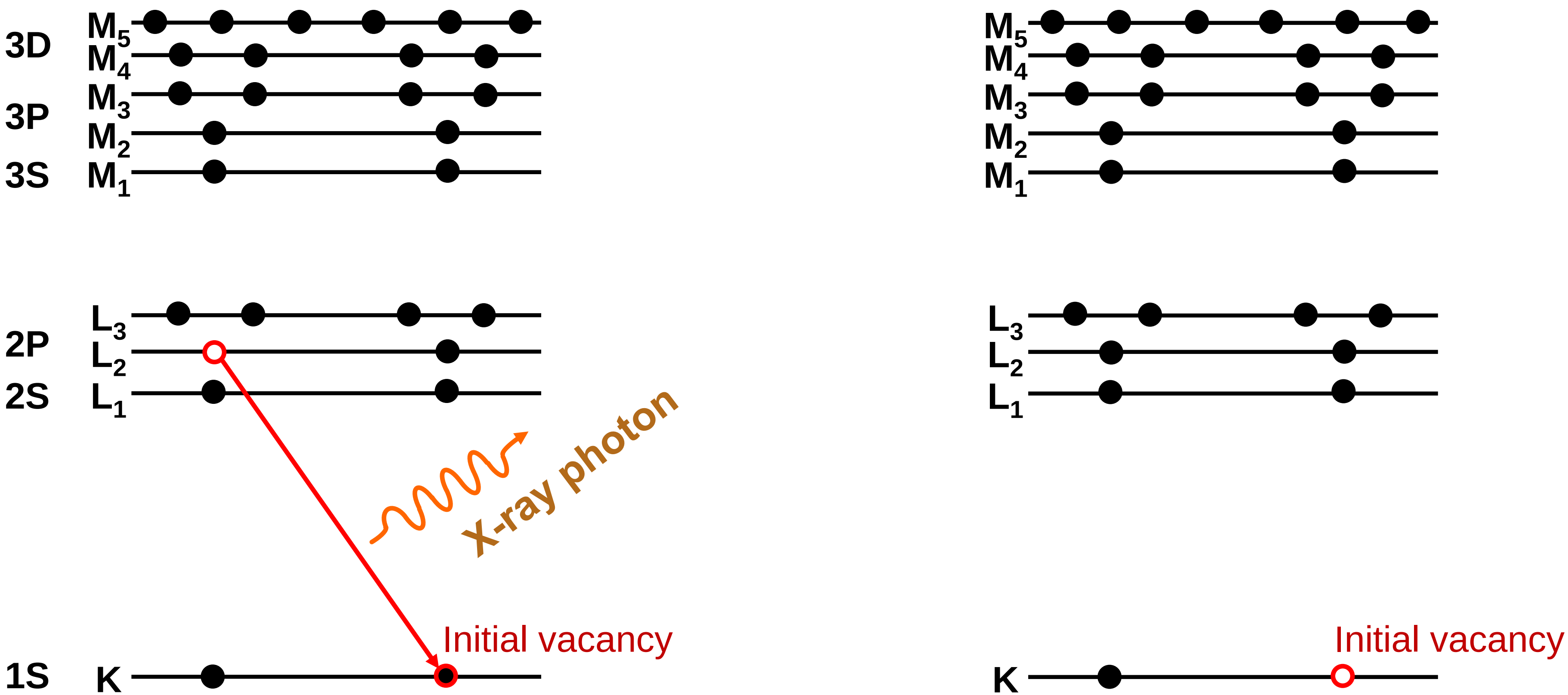
$$E_{X_{K\alpha 2}} = E_K - E_{L2}$$

Kα2 X-ray

1 secondary vacancy

Atomic radiations - Basic concept

X-ray emission



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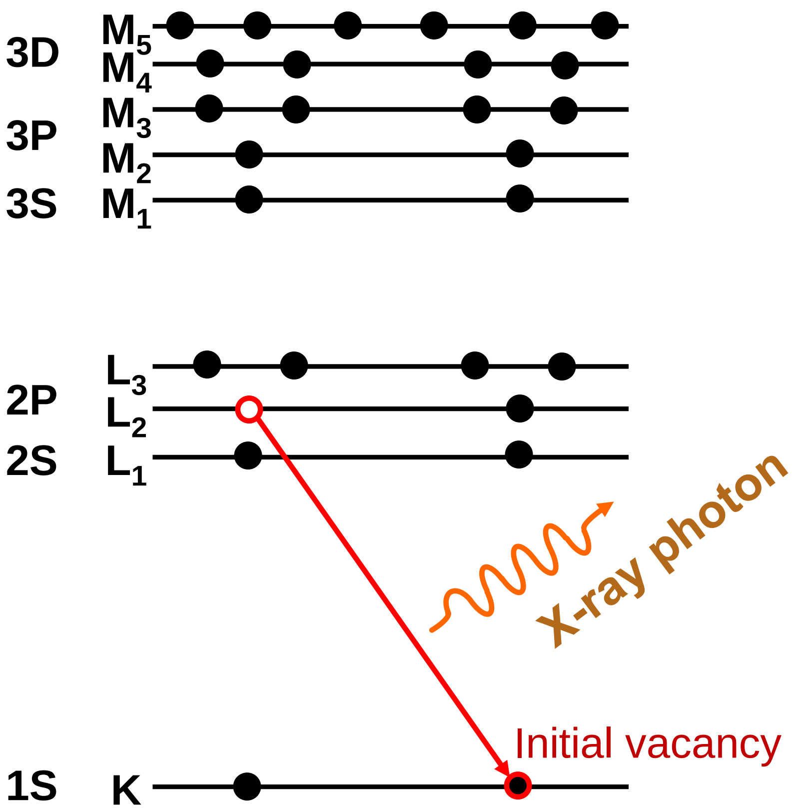
Atomic radiations - Basic concept

X-ray emission

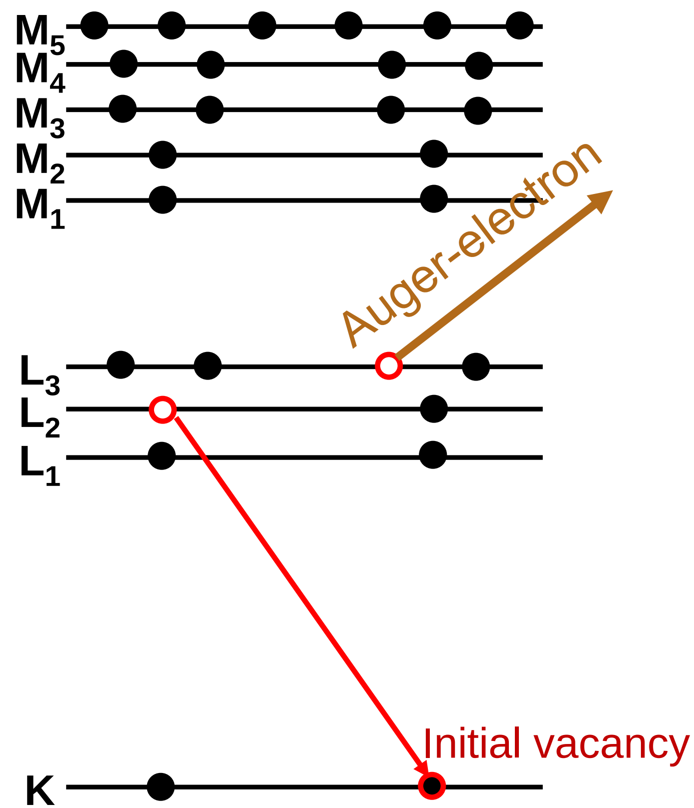


$$E_{X_{K\alpha 2}} = E_K - E_{L2}$$

Kα2 X-ray

1 secondary vacancy

Auger-electron

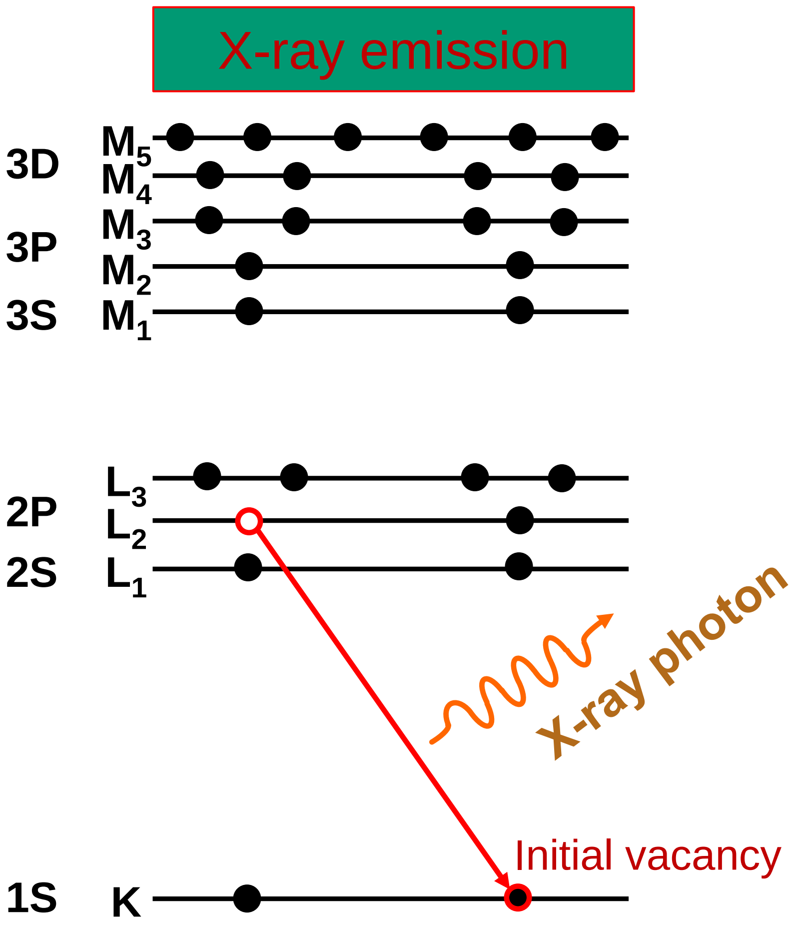


$$E_{K L2 L3} = E_K - E_{L2} - E_{L3}^{L2}$$

K L2 L3 Auger-electron

2 new secondary vacancies

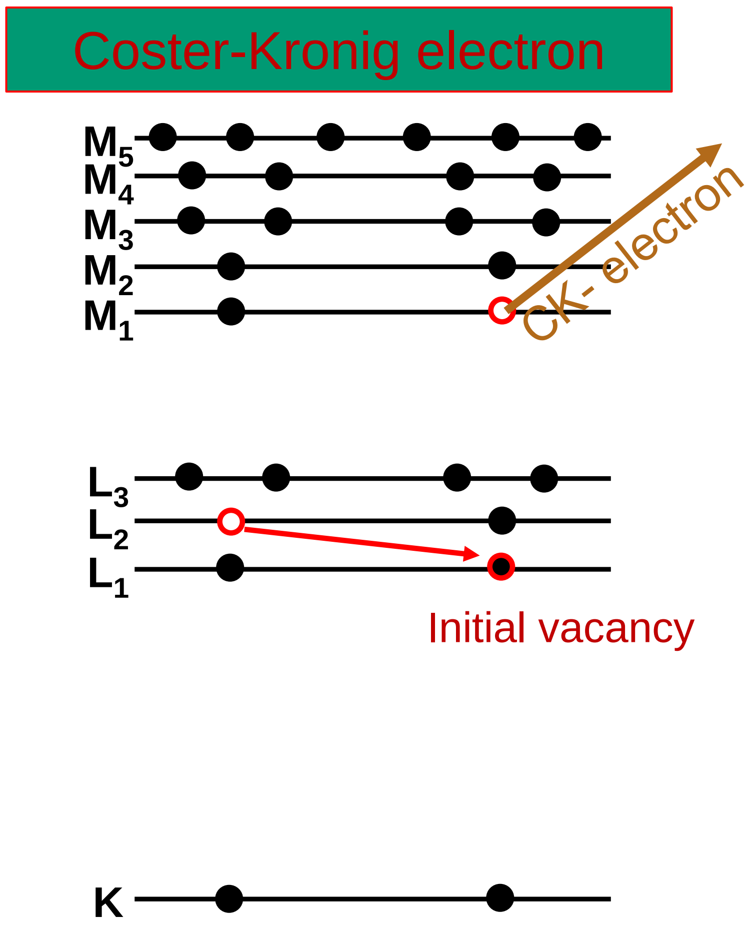
Atomic radiations - Basic concept



$$E_{X_{K\alpha 2}} = E_K - E_{L2}$$

Kα2 X-ray

1 secondary vacancy



$$E_{L1L2M1} = E_{L1} - E_{L2} - E_{M1}^{L2}$$

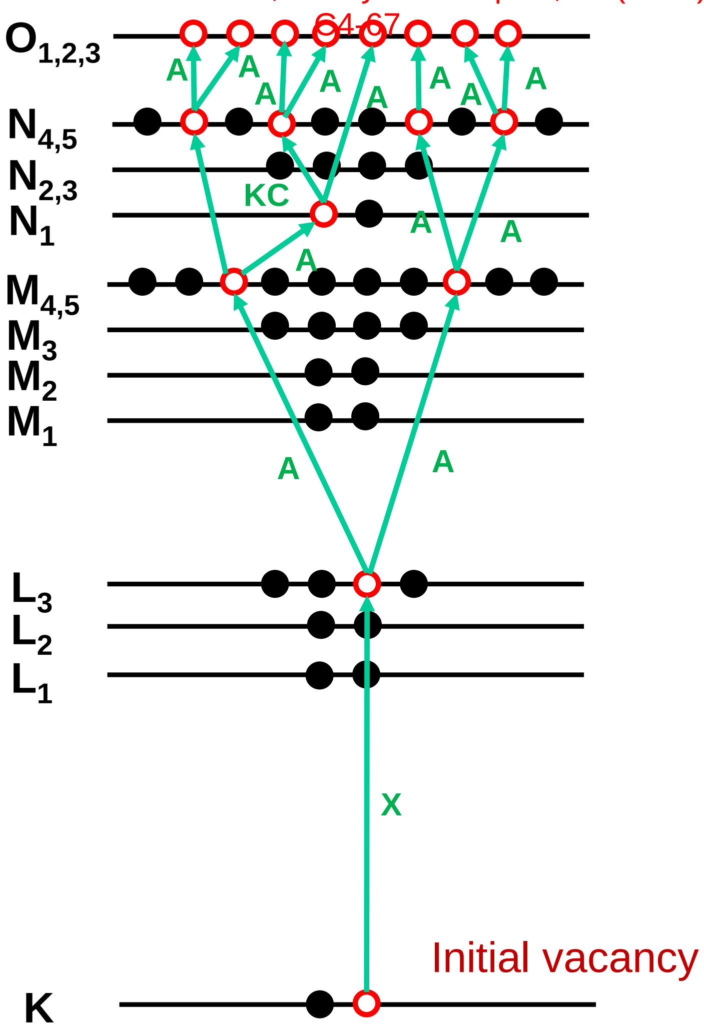
L1 L2 M1 Coster-Kronig transition

2 new secondary vacancies

Atomic relaxation and vacancy transfer

A vacancy cascade in Xe

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



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

Transition energies and Rates

For a single initial vacancy on the K-shell following nuclear decay

Number of primary vacancies

Internal conversion $$n_K = P_\gamma \times \frac{\alpha_K}{1+\alpha_T}$$

Electron capture $$n_K = P_\varepsilon \times P_K$$

	X-ray emission	Auger-electron
Energy	$E_{X_{KY}} = E_K - E_Y$	$E_{KXY} = E_K - E_X - E_Y^X$ (in an ion)
Intensity	$I_{X_{KY}} = n_K \times \omega_K$	$I_{KXY} = n_K \times a_K$
for L1 shell	$I_{X_{L1Y}} = n_{L1} \times \omega_{L1}$	$I_{L1XY} = n_K \times (a_{L1} + f_{L1L2} + f_{L1L3})$

$$\omega_K + a_K \equiv 1$$

$$\omega_{L1} + a_{L1} + f_{L1L2} + f_{L1L3} \equiv 1$$

Existing data bases

Nuclear data

ENSDF continuously updated

2008Ki07, 2002Ba85 (BrIcc): *Conversion coefficients*

1998Sc28, Schönfeld : *Electron Capture Probabilities*

EC program Schönfeld, Chu & Brown

Atomic Data (incomplete)

1972Bb16, Bambynek et al.,: *X-Ray Fluorescence Yields, Auger, and Coster-Kronig Transition Probabilities*

1984 update in an Conf. Proc.

1977La19, Larkins: *Semi-empirical Auger energies (for single initial vacancies only!)*

1979Kr13, Krause, *Radiative and Non-RadiativeYields for K and L Shells*

Existing data bases

Atomic Data (cont)

Evaluated Atomic Data Library (EADL),

[1] D.E. Cullen, et al., "Tables and Graphs of Atomic Subshells and Relaxation Data Derived from the LLNL Evaluated Atomic Data Library (EADL), Z = 1 - 100," Lawrence Livermore National Laboratory, UCRL-50400, Vol. 30, October 1991.

[2] D.E. Cullen "Program RELAX: A Code Designed to Calculate X-Ray and Electron Emission Spectra as Singly Charged Atoms Relax Back to Neutrality," Lawrence Livermore National Laboratory, UCRL-ID-110438, March 1992.

X-ray, Auger and Coster-Kronig energies & transition rates for single vacancies

Neutral atom binding energies, primary source of atomic data need to be confirmed!

Existing programs/data bases

Atomic Data (cont.)

•**1994Hu23, Hubbell et al.:** *Review, Bibliography, and Tabulation of K, L, and higher atomic shell X-Ray Fluorescence Yields*

many errors found, see J. Phys. Chem. Ref. Data, 33 (2004) 621!

• **1995ScZX, 1996Sc06, Schönfeld and Janssen:** *absolute K and L X-ray rates*

- **2007Pu05, Puri:** *rel. L and M X-ray intensities*
- **2009 Campbell (ADNDT 95 p115; not in NSR)**
  L1 Fluorescence and Coster-Kronig Probability
- **2009 Campbell for Group 4:** *review of atomic data X-ray transition and Coster-Kronig transition probabilities*
  - *primary source of data often not clearly stated*
  - *1996Sc06 Scofield no data for Z<30*
  - *Last extensive theoretical DF work by Scofield (1974)*
  - *L-shell: extensive review is long overdue*
  - *M-shell almost no data*
  - *Should theory being used to fill the gaps in systematics?*

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Existing programs/data bases

	Nucl Data	Atomic Data	Atomic Radiations	Comment
EDISTR Dillman (1985)	ENSDF	unknown	unknown	NEA PSR-0191 EDISTR.
RADLST Burrows (1988)	ENSDF, HsIcc	1972Bb16 Bambynek	K,L X-rays and Auger	Used for MIRD
RELAX Cullen (1992)	ENDL	EADL	K,L,M,N X-rays and Auger	Neutral atom BE
IMRDEC Stepanek (1997)	ENSDF, own ICC	EPDL, EEDL	K,L,M,N X-rays and Auger	Bug: decay parent BE used; only Monte-Carlo vacancy propagation program
EMISSIN Schönfeld & Janssen (2000)	Manual DDEP	1995ScZX 1996Sc06 Schönfeld	K,L X-rays and Auger	Inaccurate below Z=25; Extended for L subs-shell 2006Be35 Marie-Martine Be

Existing programs

Common problems / limitations

- Neutral atom binding energies are used for atoms with vacancies; i.e. for ions
- Single initial vacancy is considered. Secondary vacancies are ignored
- Atomic radiations only from K and L shell
- Limited information on sub-shell rates
- Auger spectrum below ~1 keV is often omitted

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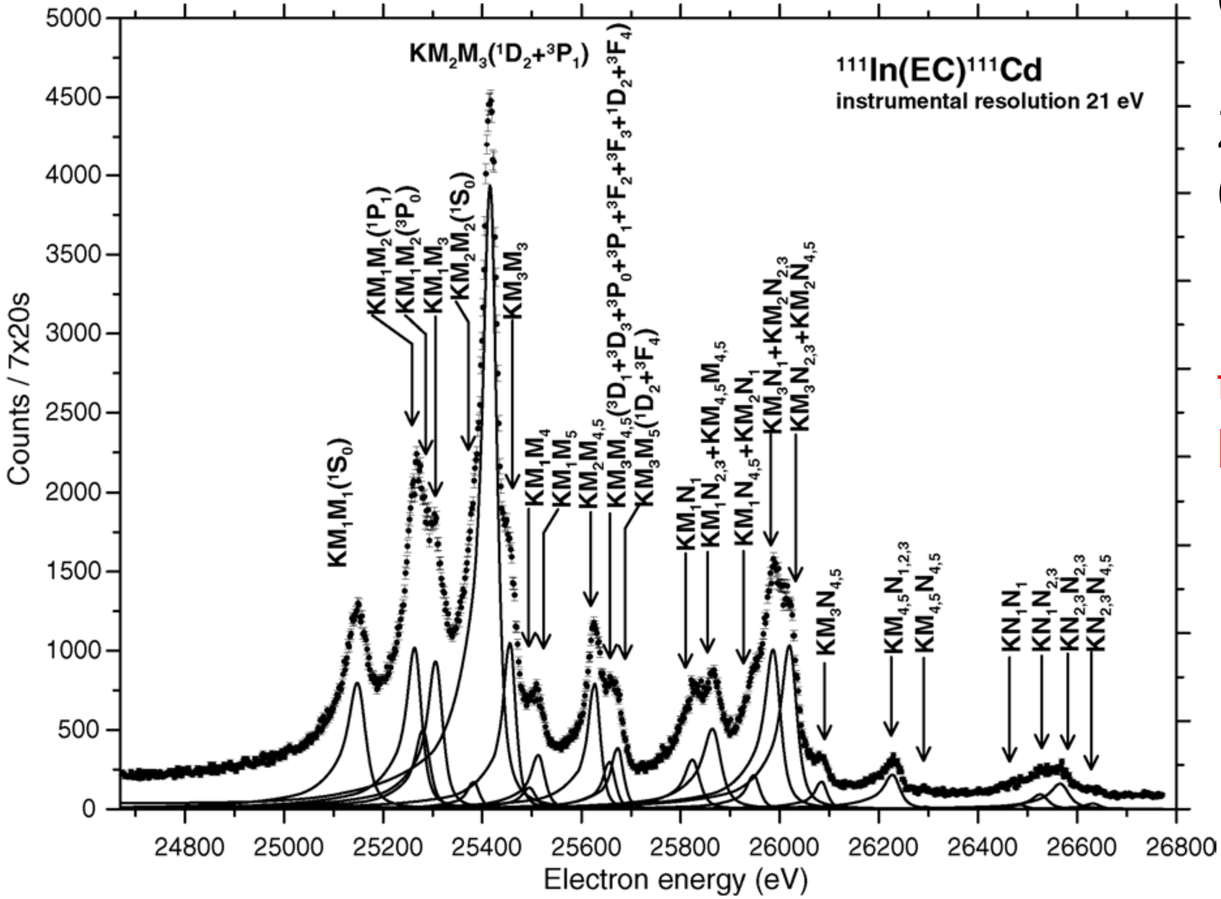
New experimental results

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



KNN Auger spectrum of ^{111}Cd from the EC-decay of ^{111}In measured with 21 eV instrumental resolution and 2 eV s
ycles with an exposition of 20 s per point in each sweep. Results of the spectrum decomposition into components are

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

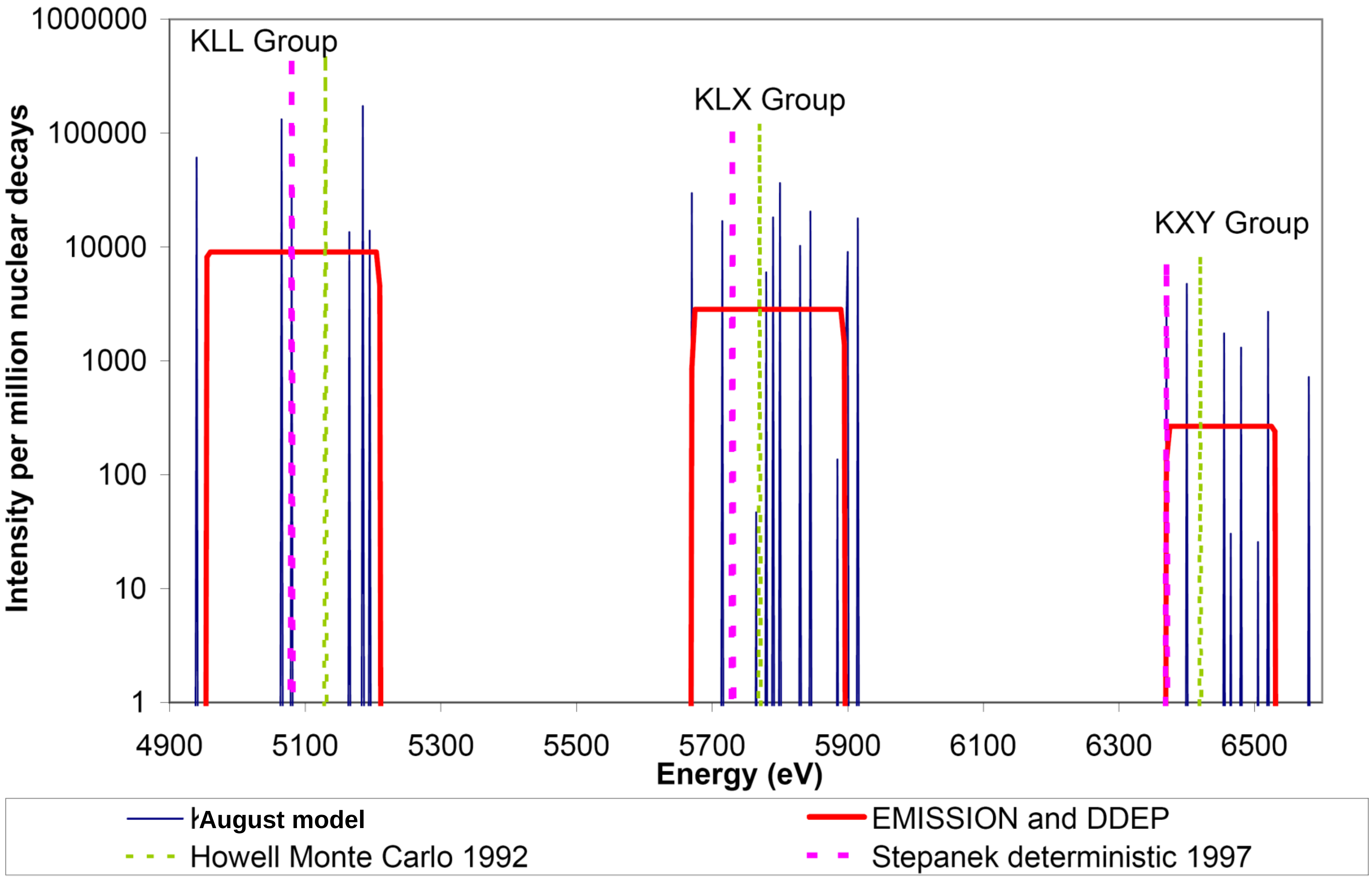
Need to extend theoretical models beyond L-shell

New model to evaluate Non-Radiative transitions (Kalman`s model)

- Primary vacancies from BrIcc (for CE) and EC (for electron capture)
- Binding energies calculated using RAINE for electron configurations of a neutral atom or an ion with single or multiple vacancies
- Auger and CK transition rates calculated using non- relativistic perturbation theory with screened hydrogenic wave functions (Simple general formalism for all shells and transitions)
- Propagate vacancies until they reach valence shells (or the shells immediately below)
- Partially Defrosted Orbitals: calculations for all electronic configurations (complete treatment) but using single-vacancy transition rates

$^{55}Fe + e^- \rightarrow {}^{55}Mn + \nu$:

K-Auger electrons (5 eV bins)

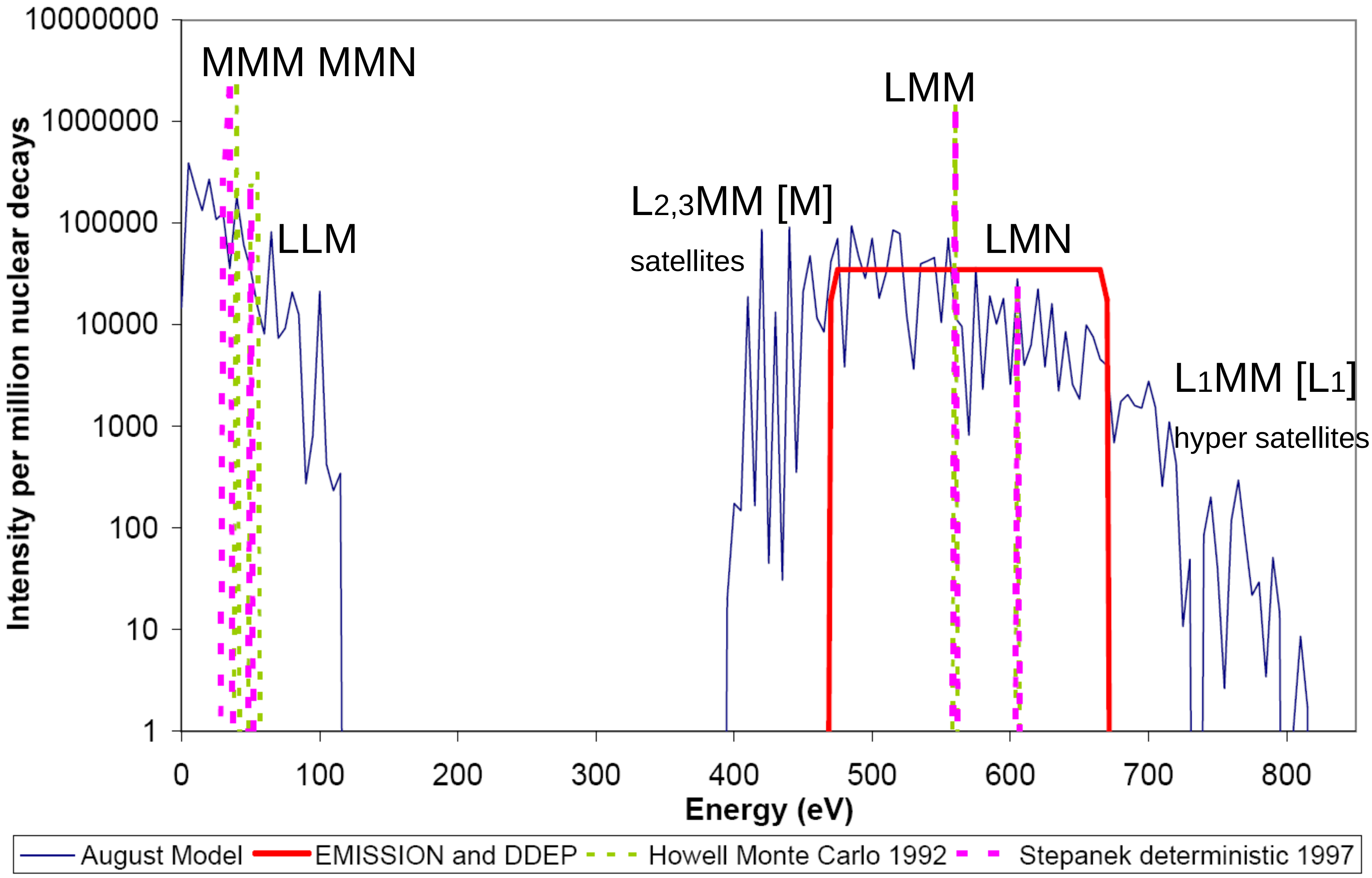


Slide courtesy of Kalman Robertson

$^{55}Fe + e^- \rightarrow {}^{55}Mn + \nu$

3.7×10^6 Auger electrons and 2.8×10^5 X-rays per 10^6 nuclear decays

L- and *M*- Auger electrons (5 eV bins)



Slide courtesy of Kalman Robertson

BrIccEmis

with Boon Quan Lee

- Uses ENSDF file for input nuclear structure data
- Uses atomic data from Schönfeld and Janssen (1995ScZX,1996Sc06)
- Calculates X-ray and Auger-electron spectrum for K and L shell
- Future plans:
  - Calculate theoretical binding energies using the RAINE code
  - Incorporate up-to date atomic data on X-ray emission
  - Incorporate Kalman`s model to calculate detailed energy spectra of Auger and Coster-Kronig transitions

BrIccEmis results for $^{55}Fe + e^- \rightarrow {}^{55}Mn + \nu$

with Boon Quan Lee

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Results:
--------
Vacancy Creation:
-----------------
N(K)...............................: 0.8853(17)
N(L)...............................: 1.407(5)
N(L1)..............................: 0.314(5)
N(L2)..............................: 0.515(6)
N(L3)..............................: 0.576(7)

Auger Yield:
---------------------
a(K)...............................: 0.679(5)
a(L1)..............................: 0.044(16)
a(L2)..............................: 0.99660(20)
a(L3)..............................: 0.99610(20)
```

BrIccEmis results for ^{55}Fe + e- → ^{55}Mn + ν

with Boon Quan Lee

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X-Ray Emission Probabilities:      Intensity                       Energy(keV)
----------------------------
p(XK)..............................: 0.284(5)
p(XL)=N(L)*<OmegaL>................: 0.0066(10)
p(XL1).............................: 0.000104(7)
p(XL2).............................: 0.00204(13)
p(XL3).............................: 0.00309(17)
p(XL)=p(XL1)+p(XL2)+p(XL3).........: 0.00523(21)
p(KAlpha)..........................: 0.250(4)
p(KBeta)...........................: 0.0340(7)
p(KAlpha1)(K-L3)...................: 0.166(3)                          5.899(0)
p(KAlpha2)(K-L2)...................: 0.0845(14)                        5.888(0)
p(K'Beta1)(K-M2,3,4,5).............: 0.0340(7)            6.486(0)          6.536(0)
p(K'Beta2)(K-N2,3,4,5).............: 0.000(0)             6.539(0)          6.539(0)

Partial L-Shell X-Ray Emission Probability:
------------------------------------------
pX(L1-M2).....p(XLBeta4)...........: 3.60E-5(23)                      0.7159(0)
pX(L1-M3).....p(XLBeta3)...........: 6.8E-5(5)                        0.7226(0)
pX(L2-M1).....p(XLEta).............: 0.00204(13)                      0.5675(0)
pX(L3-M1).....p(XLl)...............: 0.00309(17)                      0.5564(0)
```

BrIccEmis results for ^{55}Fe + e- → ^{55}Mn + ν

with Boon Quan Lee

```
Auger Electron Emission Probability: Intensity
-----------------------------------

K-Shell
-------
p(eaK)..............................: 0.601(5)
p(KLL)..............................: 0.466(4)
p(KLX)..............................: 0.1267(18)
p(KXY)..............................: 0.00862(20)

L-Shell
-------
p(eaL)=N(L)*(1-<Omega>)............: 1.400(5)
p(eaL1).............................: 0.014(5)
p(eaL2).............................: 0.597(6)
p(eaL3).............................: 0.789(9)
p(eaL)=p(eaL1)+p(eaL2)+p(eaL3).....: 1.400(12)
```

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Conclusions

- Existing atomic data on X-ray, Auger and Coster-Kronig transitions is incomplete
- Existing programs only cover radiations from single vacancies on the K and L-shell. Sub-shell transition rates are often not known. Neutral binding energies are used.
- New program being developed to treat all atomic shells and multiple vacancies to evaluate detailed energy spectra of atomic radiations using ENSDF file as input for nuclear structure data