Atomic processes

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Courtesy to Tibor Kibédi (ANU)

DDEP Training Workshop

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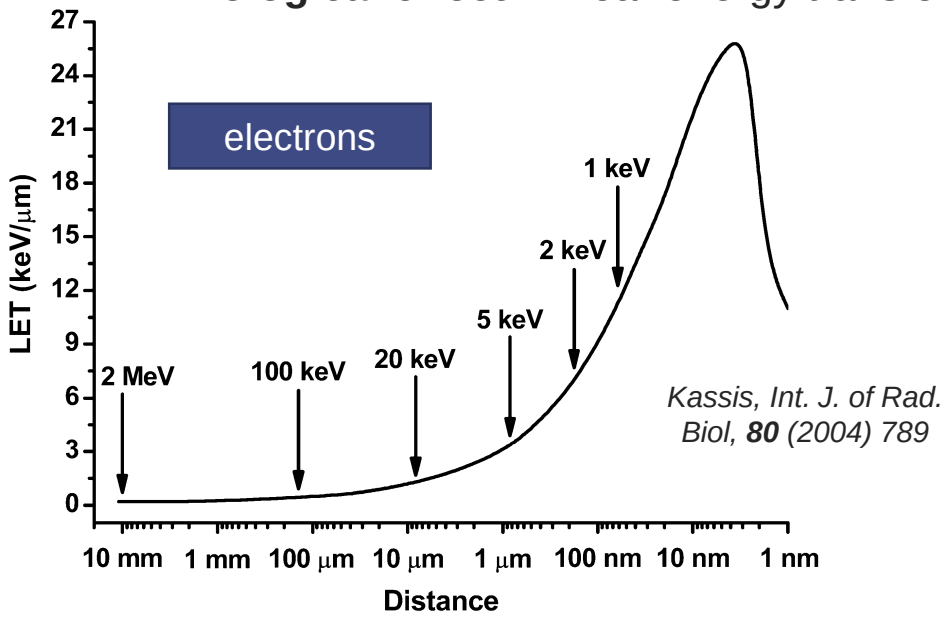
Medical applications

Regaud and Lacassagne (1927)

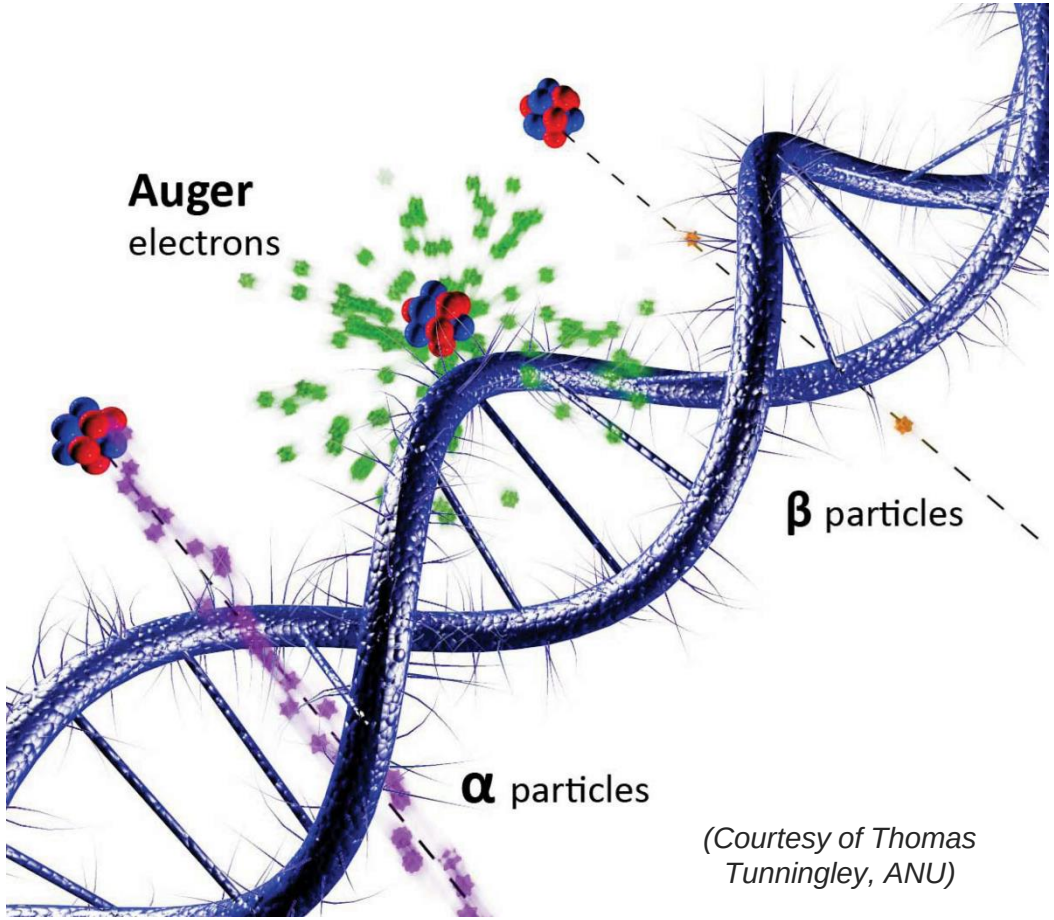
"The ideal agent for cancer therapy would consist of heavy elements capable of emitting radiations of molecular dimensions, which could be administered to the organism and selectively fixed in the protoplasm of cells one seeks to destroy."

Biological effect: Linear energy transfer



*Kassis, Int. J. of Rad. Biol, **80** (2004) 789*

Targeted tumour therapy



(Courtesy of Thomas Tunningley, ANU)

Nucleus decay is a sequential process

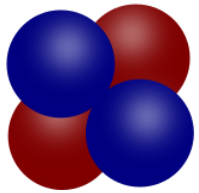
X-ray and Auger electron spectra are an integral part of the radiations emitted in nuclear decay.

Particles emitted

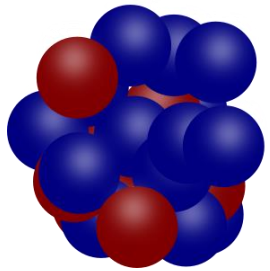
Observable

1. **Initial decay**
   - β-
   - β+ / electron capture
   - Alpha
   - Fission

electron



alpha



ion

$T_{1/2}$, Energy, Intensity

2. **Nucleus de-excitation**
   - γ transition
   - Internal conversion

electron

Photon

$T_{1/2}$, Energy, Intensity

3. **Atomic de-excitation**
   - X-rays
   - Auger electrons

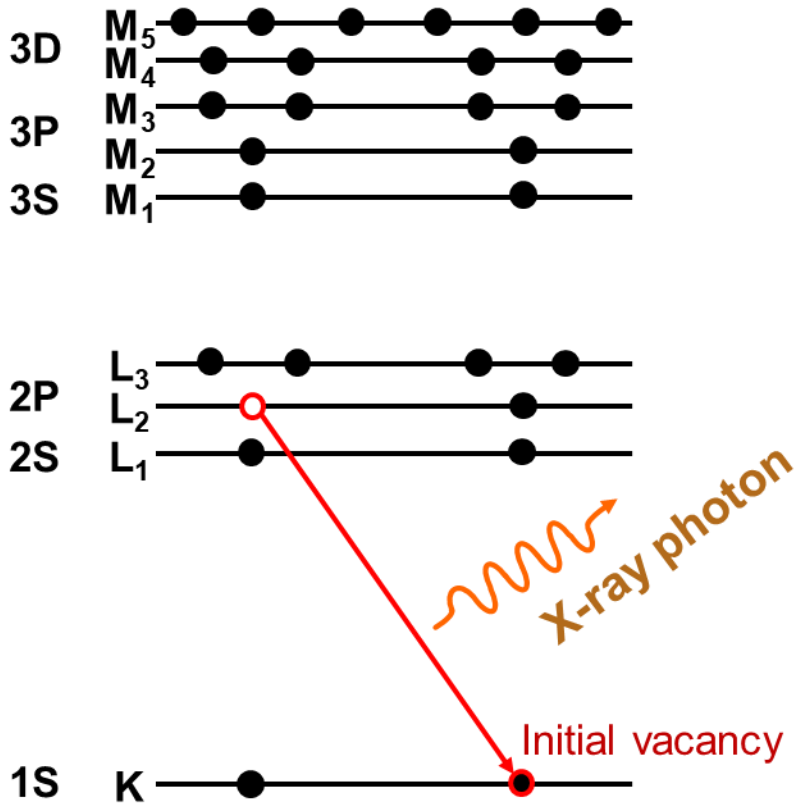
electron

Photon

Energy, Intensity

Atomic radiations

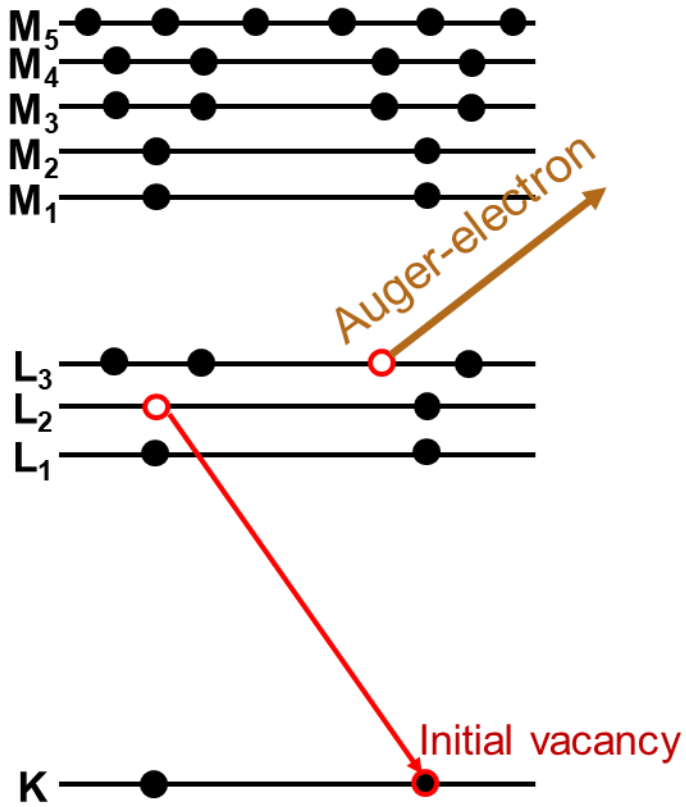
X-ray emission



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

Kα2 X-ray
1 secondary vacancy

Auger-electron

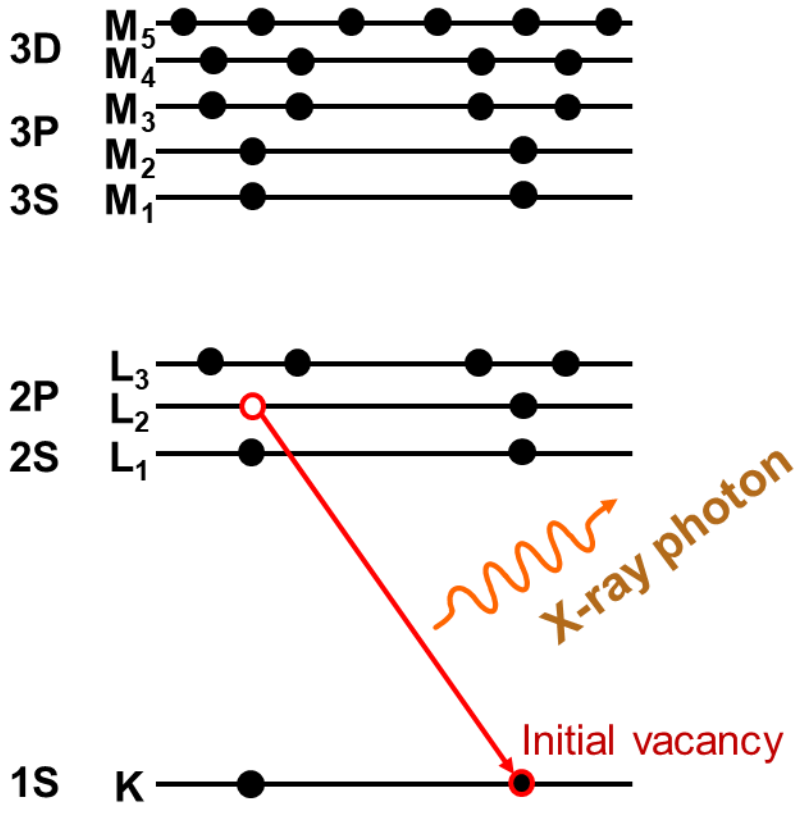


$$E_{KL2L3} = E_K - E_{L2} - E_{L3}^{L2}$$

K L2 L3 Auger-electron
2 new secondary vacancies

Atomic radiations

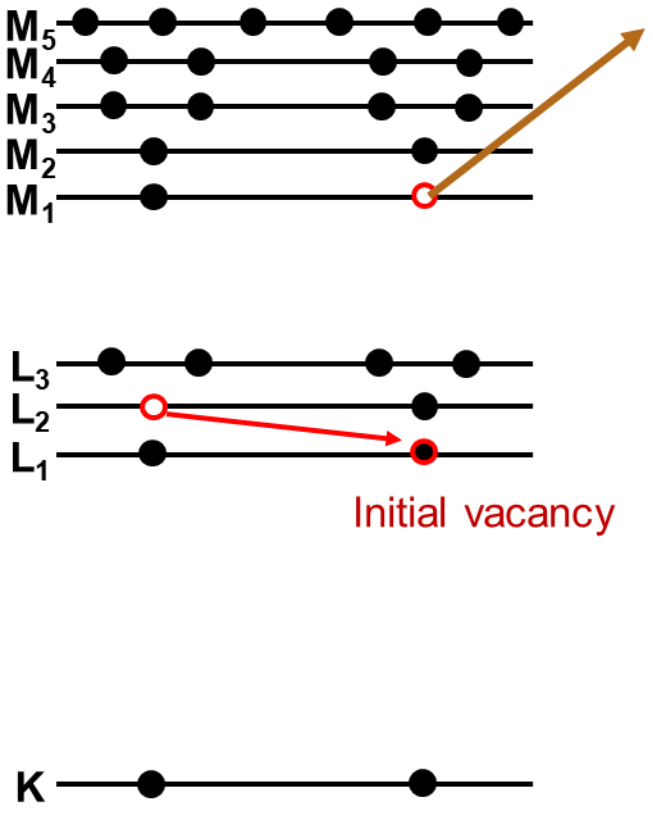
X-ray emission



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

Kα2 X-ray
1 secondary vacancy

Coster-Kronig electron



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

L1 L2 M1 Coster-Kronig transition
2 new secondary vacancies

Relaxation

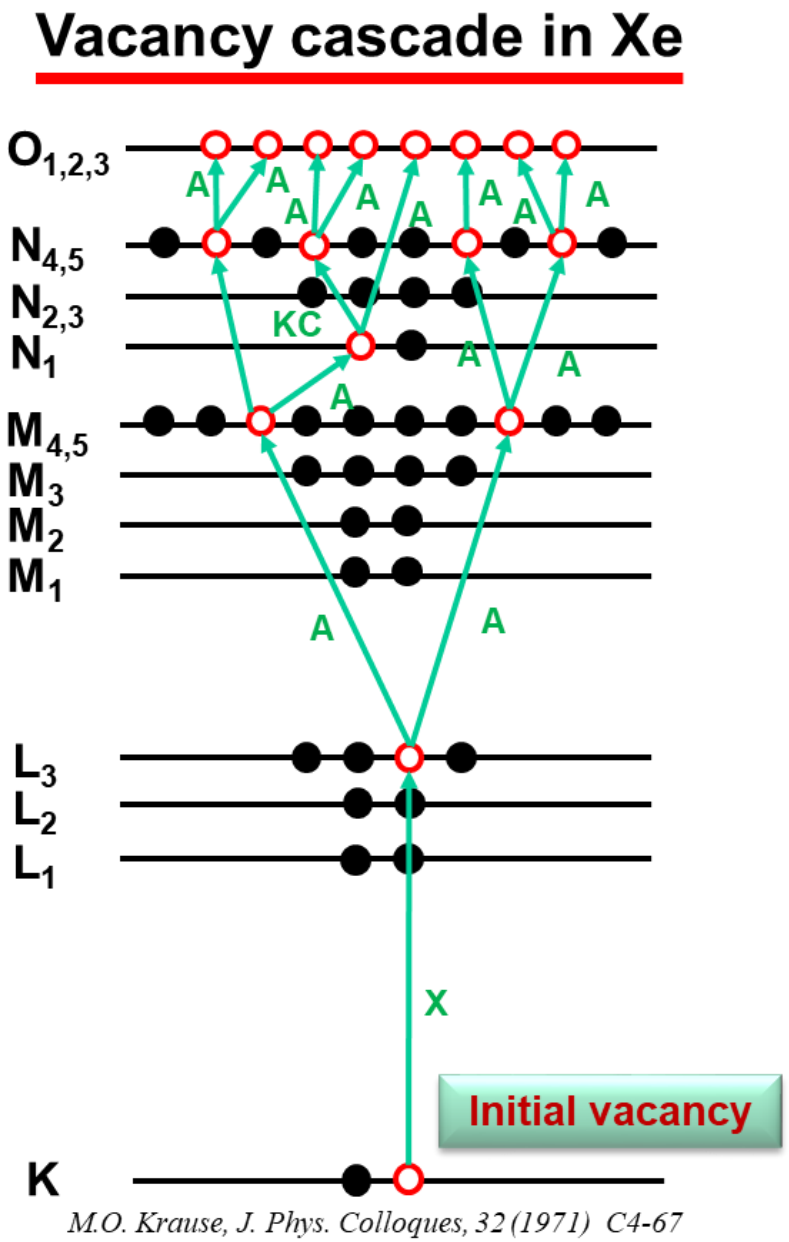
Vacancies can be produced by electron impact, photo-ionisation, ion-atom collision, **internal conversion, electron capture, secondary processes accompanying β-decay or electron capture**.

Full relaxation of an initial vacancy creates vacancy cascade involving **X**-ray (Radiative) and **A**uger as well as **C**oster-**K**ronig (Non-Radiative) transitions

Many possible cascades for a single initial vacancy. Deterministic approach possible for low Z, Monte Carlo otherwise or simplified deterministic.

Typical relaxation time is ~10^{-15} seconds, thus almost independent of nuclear processes.

→ Many vacancy cascades following a single ionisation event!



Siegbahn and IUPAC notations

$$
\begin{array}{lll}
\mathrm{K}\alpha & \mathrm{K}\alpha_2 & \mathrm{K}-\mathrm{L}_2 \\
 & \mathrm{K}\alpha_1 & \mathrm{K}-\mathrm{L}_3
\end{array}
$$

$$
\begin{array}{lll}
\mathrm{K}'\beta_1 & \mathrm{K}\beta_3 & \mathrm{K}-\mathrm{M}_2 \\
 & \mathrm{K}\beta_1 & \mathrm{K}-\mathrm{M}_3 \\
 & \mathrm{K}\beta_5'' & \mathrm{K}-\mathrm{M}_4 \\
 & \mathrm{K}\beta_5' & \mathrm{K}-\mathrm{M}_5
\end{array}
$$

$$
\begin{array}{lll}
\mathrm{K}'\beta_2 & \mathrm{K}\beta_2 & \mathrm{K}\beta_2'' \quad \mathrm{K}-\mathrm{N}_2 \\
 & & \mathrm{K}\beta_1' \quad \mathrm{K}-\mathrm{N}_3 \\
 & \mathrm{K}\beta_4 & \mathrm{K}\beta_4'' \quad \mathrm{K}-\mathrm{N}_4 \\
 & & \mathrm{K}\beta_4' \quad \mathrm{K}-\mathrm{N}_5
\end{array}
$$

$$
\begin{array}{lll}
\mathrm{L}_\ell & \ell & \mathrm{L}_3-\mathrm{M}_1 \\
\mathrm{L}_\alpha & \alpha_2 & \mathrm{L}_3-\mathrm{M}_4 \\
 & \alpha_1 & \mathrm{L}_3-\mathrm{M}_5 \\
\mathrm{L}_\eta & \eta & \mathrm{L}_2-\mathrm{M}_1
\end{array}
$$

$$
\begin{array}{lll}
\mathrm{L}_\gamma & \gamma_2 & \mathrm{L}_1-\mathrm{N}_2 \\
 & \gamma_3 & \mathrm{L}_1-\mathrm{N}_3 \\
 & \gamma_4 & \mathrm{L}_1-\mathrm{O}_{2,\ 3} \\
 & \gamma_5 & \mathrm{L}_2-\mathrm{N}_1 \\
 & \gamma_1 & \mathrm{L}_2-\mathrm{N}_4 \\
 & \gamma_6 & \mathrm{L}_2-\mathrm{O}_4
\end{array}
$$

$$
\begin{array}{lll}
\mathrm{L}_\beta & \beta_6 & \mathrm{L}_3-\mathrm{N}_1 \\
 & \beta_4 & \mathrm{L}_1-\mathrm{M}_2 \\
 & \beta_3 & \mathrm{L}_1-\mathrm{M}_3 \\
 & \beta_{10} & \mathrm{L}_1-\mathrm{M}_4 \\
 & \beta_9 & \mathrm{L}_1-\mathrm{M}_5 \\
 & \beta_1 & \mathrm{L}_2-\mathrm{M}_4 \\
 & \beta_2 & \mathrm{L}_3-\mathrm{N}_5 \\
 & \beta_{15} & \mathrm{L}_3-\mathrm{N}_4 \\
 & \beta_7 & \mathrm{L}_3-\mathrm{O}_1 \\
 & \beta_5 & \mathrm{L}_3-\mathrm{O}_{4,\ 5}
\end{array}
$$

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

Energy $E_{X_{KY}} = E_K - E_Y$

Intensity $I_{X_{KY}} = n_K \times \omega_K$

For L_1 shell $I_{X_{L_1Y}} = n_{L_1} \times \omega_{L_1}$

Auger-electron

$E_{KXY} = E_K - E_X - E_Y^X$ ← in an ion

$I_{KXY} = n_K \times a_K$

$\omega_K + a_K = 1$

$I_{L_1XY} = n_K \times \left(a_{L_1} + f_{L_1L_2} + f_{L_1L_3}\right)$

$\omega_{L_1} + a_{L_1} + f_{L_1L_2} + f_{L_1L_3} = 1$

Koster-Cronig

Atomic recommandations in DDEP

- ✓ Fluorescence yields (ω_K, $\overline{\omega_L}$, $\overline{\omega_M}$, n_{KL}, $\overline{n}_{LM}$)
- ✓ Atomic emission energies

  X-ray

  - K-shell: energy detailed by subshell K_α(K-L_i), $K'\beta_1$ (K-M_i)…
  - L-shell: range of energy by group (L_α, L_β, L_γ)
  - M+ shells: no recommendations

  Auger

  - K-Augers: range of energy by subshell: KLL, KLX, KLY…
  - L-shell: range of energy for the whole shell
  - M+ shells: no recommendations
- ✓ Atomic emission intensities (X-ray and Auger)
  - Absolute values provided for K and L shells
  - Relative values provided for K shell only

→ More detailed information available but not given.

LNE – LNHB/CEA **Table de Radionucléides**

3 Atomic Data

3.1 Mn

ω_K	:	0,321	(5)
$\bar{\omega}_L$	:	0,0047	(7)
$\bar{\omega}_M$	:	0,000027	(2)
n_{KL}	:	1,478	(4)
$\bar{n}_{LM}$	:	1,996	(8)

3.1.1 X Radiations

		Energy (keV)	Relative probability
X_K			
	$K\alpha_2$	5,88772	51
	$K\alpha_1$	5,89881	100
	$K\beta_1$	6,49051	20,5
	$K\beta_5''$	6,5354	
X_L			
	$L\ell$	0,5576	
	$L\alpha$	0,6394 - 0,6404	
	$L\eta$	0,5695	
	$L\beta$	0,64636 - 0,7694	
	$L\gamma$	0,65826 - 0,65826	

3.1.2 Auger Electrons

	Energy (keV)	Relative probability
Auger K		
KLL	4,953 - 5,210	100
KLX	5,671 - 5,895	27,2
KXY	6,370 - 6,532	1,85
Auger L	0,47 - 0,67	

Evaluation of atomic data

Atomic measurements of good quality are scarce. Data are usually estimated from existing tables and codes.

DDEP <u>current</u> evaluation process for atomic data

1. Atomic shell vacancies are calculated based on
   a) Internal conversion coefficients — BrIcc code — regularly updated (2011 version)
   b) Electronic capture — BetaShape code — regularly updated (2024 version)

2. Fluorescence yields are taken from tables
   a) ω_K and $\overline{\omega_L}$ from Schönfeld and Janssen — 1995
   b) $\overline{\omega_M}$ from a fit performed on the data of Hubbell — 1994

3. X-ray energies are taken from tables
   a) Evaluated by Bearden (Z < 10) — 1967
   b) Evaluated by Deslates (Z > 10) — 2003

4. Auger energies are taken from the table evaluated by Larkins — 1977

5. Emission code is used to calculated X-ray an Auger Intensities — not maintained — (2006 version)

Limitations

In ENSDF evaluations, use of the RADLST code (1988).

→ This code does much more than atomic radiations which are poorly treated (K and L_1 shells only).

Common problems / limitations

- In some cases, neutral atom binding energies are used for atoms with vacancies, i.e. for ions.
- Only single initial vacancy is considered. Secondary vacancies are ignored.
- Atomic radiations only from primary vacancies on a limited number of shells.
- Limited information on sub-shell rates:
  - → None with RADLST.
  - → L_1, L_2, L_3 vacancies only, filling from M, N, O sub-shells.
- Auger electrons below ~1 keV are often omitted.

BrIccEmis / NS_Radlist

Developed By T. Kibédi from ANU.

Atomic transition rates from EADL (1991PeZY).

Atomic transition energies calculated using RAINE (2002Ba85), with semi-empirical corrections (2020TEZY).

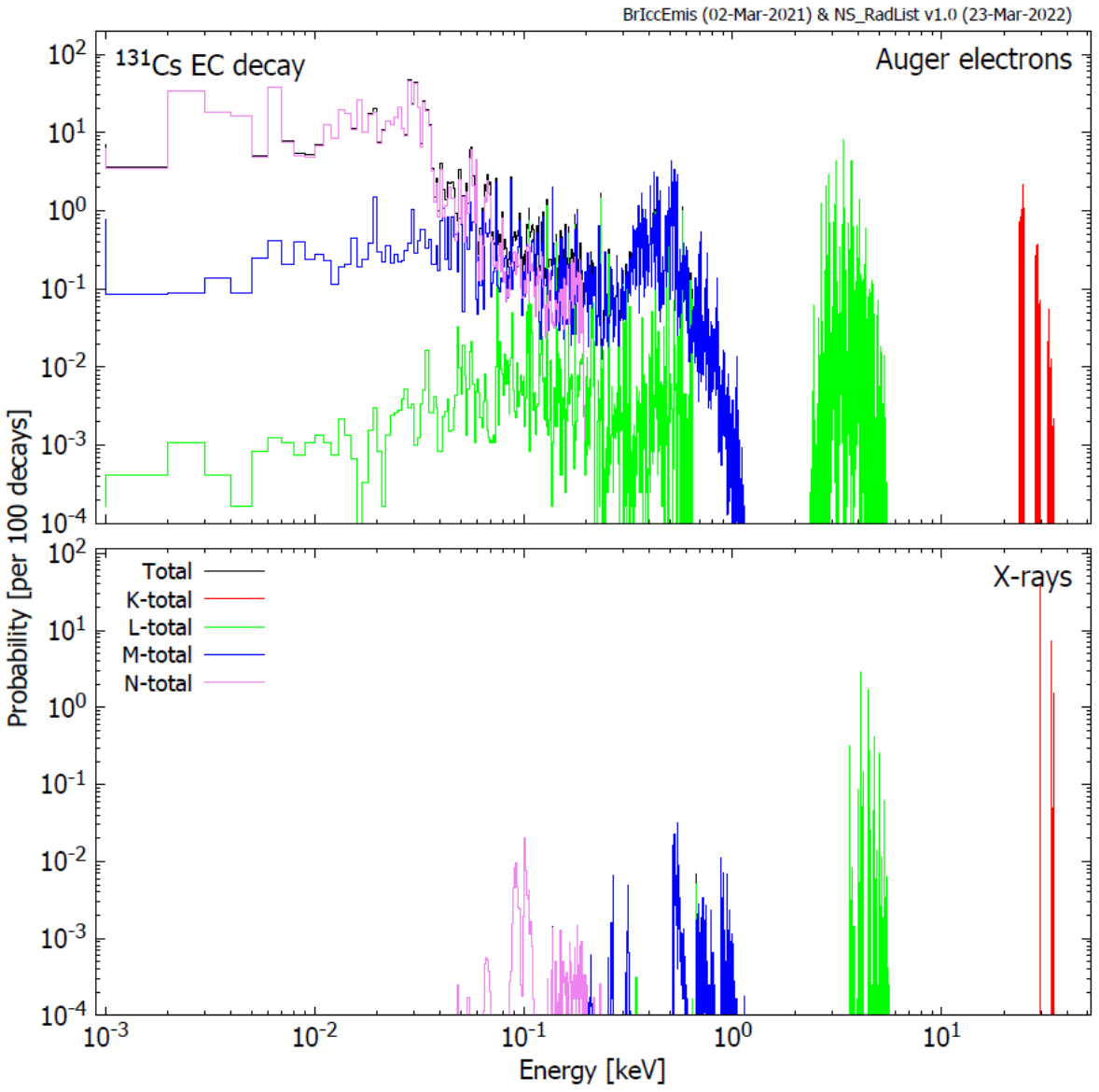
Initial atomic vacancy from EC & IC (nuclear decay mode).

BrIccEmisDB (219 MB) → Monte Carlo approach.

- Precalculated atomic spectra for Z=6 to 100 with initial vacancy on K to O shells; 1 million simulations.
- Binned with 1 eV.
- X-rays and Auger electrons.

✓ New ENSDF format for including atomic information, accepted by the NSDD network.

→ Release of NS_Radlist still pending.



Situation in DDEP evaluations

Old evaluations $\lesssim$ 2008

→ Atomic information from RADLST.

“Usual” evaluations $\gtrsim$ 2006

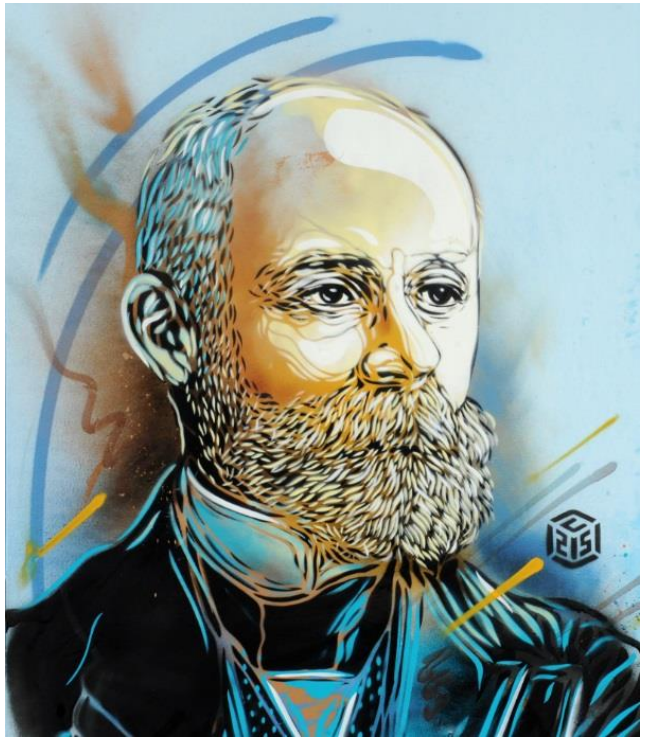
→ Atomic information from EMISSION.

Recent evaluations where X rays and Auger electrons are of high importance (^{131}Cs, ^{103}Pd/^{103m}Rh)

→ Atomic information from NS_Radlist (private communication).

Future: use of NS_Radlist from T. Kibédi

→ Benefit from sub-shell information from BetaShape and BrIcc. Code must be available.

→ Developments are required to adapt Saisinuc, the database and all the output files (pdf Tables; ENSDF, Lara and PenNuc files). Format is different; information is not exactly the same; database must be extended; calculation and checking codes must be revised.

Thank you for attention