IRSN
INSTITUT
DE RADIOPROTECTION
ET DE SÛRETÉ NUCLÉAIRE

Faire avancer la sûreté nucléaire

MODELLING THE PRODUCTION OF ELECTRONS AND X-RAYS FOR MONTE CARLO DOSIMETRY CALCULATIONS

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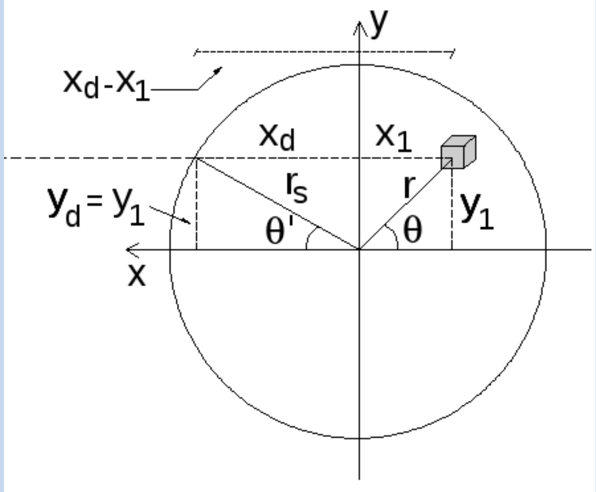


31èmes Journées L.A.R.D. – 13 et 14 Octobre 2014

CONTEXT

PRIMARY AND SECONDARY EMISSION OF PARTICLES

⇒ **Environment (aerosols)**

⇒ **Accurate elemental characterization using PIXE analytical technique**

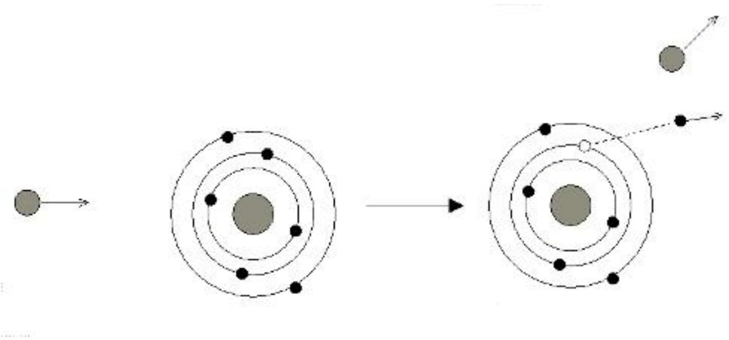
⇒ **Biomedical applications**

⇒ **Precise determination of the deposited energy in cells - Microdosimetry (∝ number of emitted X-rays, Auger electrons, etc)**

⇒ **Number of detected X-rays:**

$$N_{ij} = \frac{\Omega}{4\pi}\epsilon_j N T_j C_i \int_{E_{out}}^{E_{in}} \frac{\tau_j(x(E))}{S(E)} \boxed{\sigma_{ij}^{X}(E)} dE$$

ATOMIC EXCITATION - DEEXCITATION



⇒ H and He ion beams - PIXE (Particle induced X-ray Emission) analysis

⇒ β, α (He ions), γ particles, etc - nuclear decay

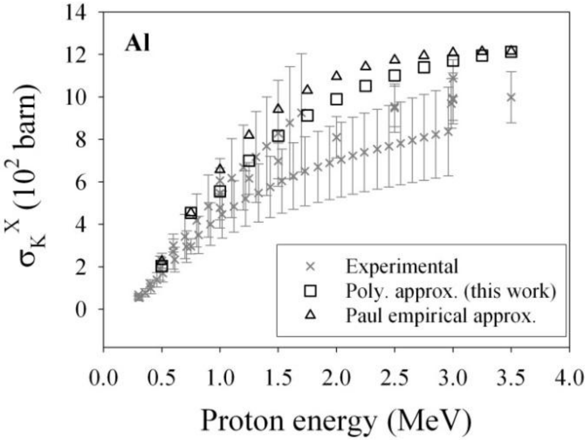
⇒ Atomic ionisation

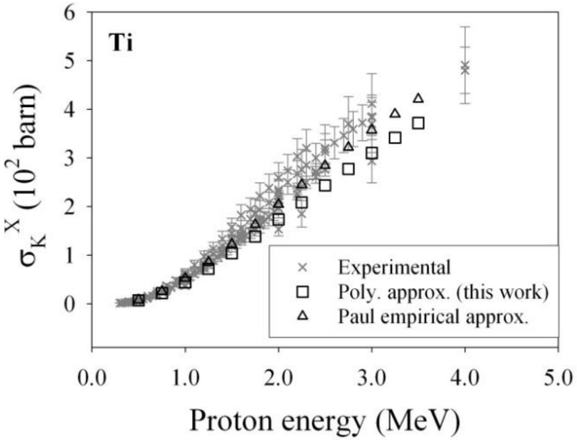
⇒ Ionisation cross-sections

⇒ X-ray production cross-sections - ionisation cross-sections

⇒ Experimental values - large discrepancies between data sets

⇒ Theoretical values - experimental data dependency?





UNIVERSAL IONISATION CROSS-SECTIONS – H AND HE IONS

⇒ **Theoretical approach - universal ionisation cross-sections**

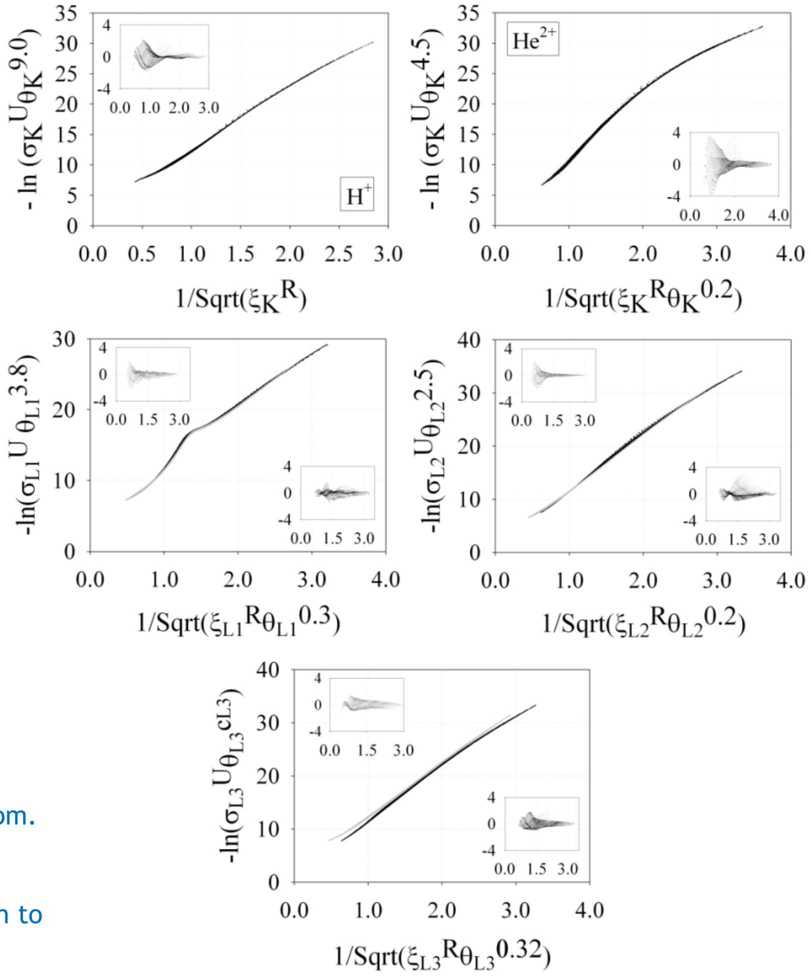
⇒ **7th order polynomial approximation**

⇒ **0.1 MeV to 10 MeV (100 MeV)**

$$\sigma^U_{n,o}(\xi^R_{n,o},\zeta_{n,o},\theta_{n,o}) = \frac{\sigma^{ECPSSR}_{n,o}(\xi^R_{n,o},\zeta_{n,o},\theta_{n,o})}{8\pi a_0^2 Z^2_{proj} C_{n,o}(x_q)}\eta_{n,o} Z^2_{n,o}\theta_{n,o}^{1+c^U_{n,o}}$$

⇒ **Allows exclusion of experimental data dependency**

⇒ **Partially implemented in Geant4 since 2012**



References:

Taborda, A.; Chaves, P. C.; Reis, M. A., « Polynomial approximation to universal ionisation cross-sections of K and L shells induced by H and He ions », X-Ray Spectrom. 40:127, 2011

Taborda, A.; Chaves, P. C.; Carvalho, M. L.; Reis, M. A., « Polynomial approximation to universal M-shell ionisation cross-sections induced by H+ and He2+ ions », X-Ray Spectrom. 42:177, 2013

UNIVERSAL IONISATION CROSS-SECTIONS – HE IONS

⇒ **Implemented in Geant4 → can be used in dosimetry calculations**

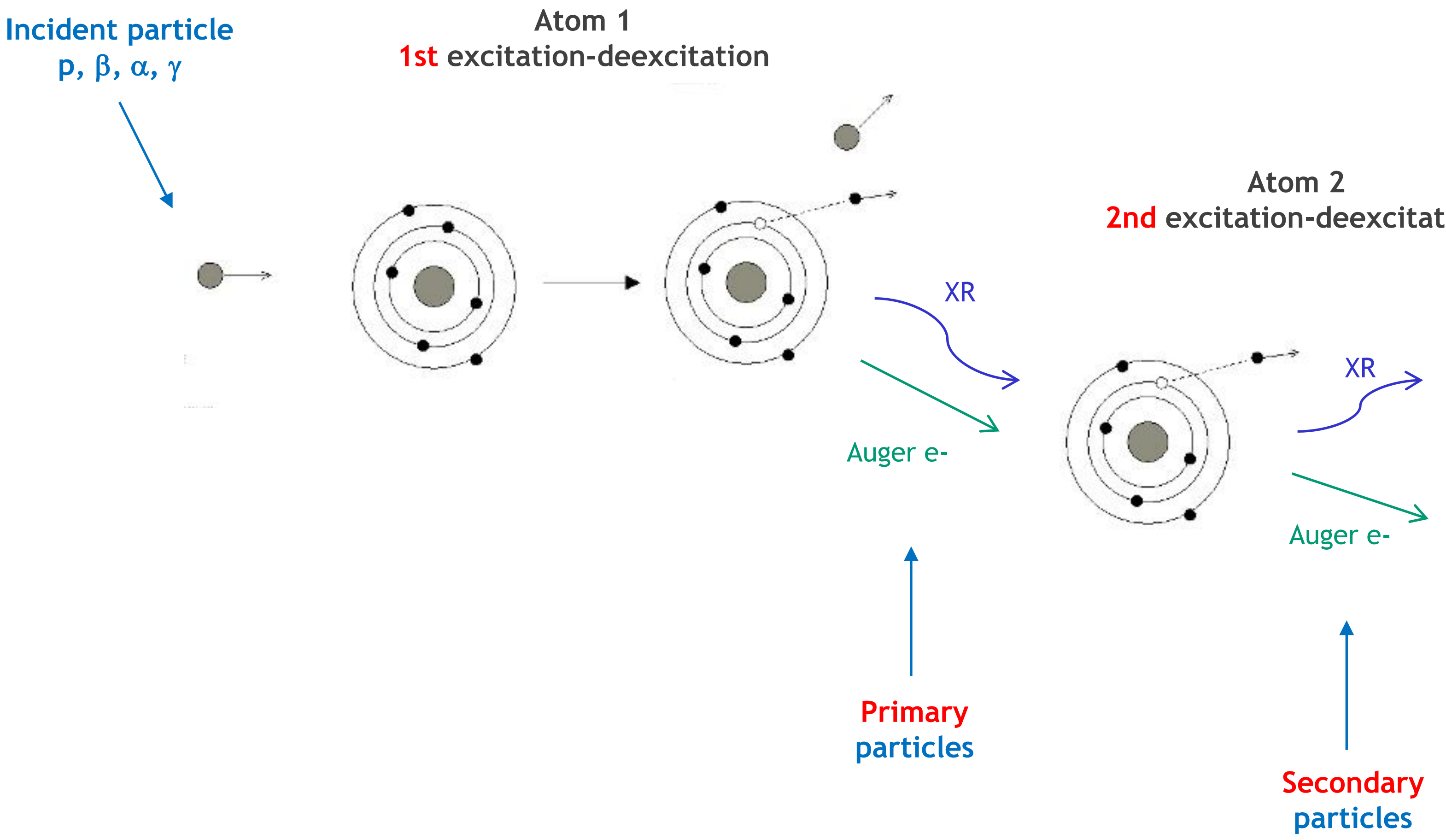
⇒ **Alpha particles emitting radionuclides, e.g.**

⇒ **Energies < 10 MeV**

Radionuclide	**α particle energy (MeV)**
211At	5.8
213Bi	5.9
223Ra	5.7
225Ac	5.8
227Th	6.0
…	…

Energies values taken from RADAR – the radiation dose assessment resource

PRIMARY AND SECONDARY PARTICLES



Incident particle
p, β, α, γ

Atom 1
1st excitation-deexcitation

Atom 2
2nd excitation-deexcitation

XR

Auger e-

XR

Auger e-

Primary particles

Secondary particles

SECONDARY X-RAYS

PENETRATION FUNCTION MODEL (REIS1996)

⇒ **Number of X-rays *B* produced in a volume *dV* induced in *dV* by X-rays *A***

$$d\chi_{B,A} = P_A(x) R_{B,A} Q_A(r) T_B(x_1, r, \theta) dV$$

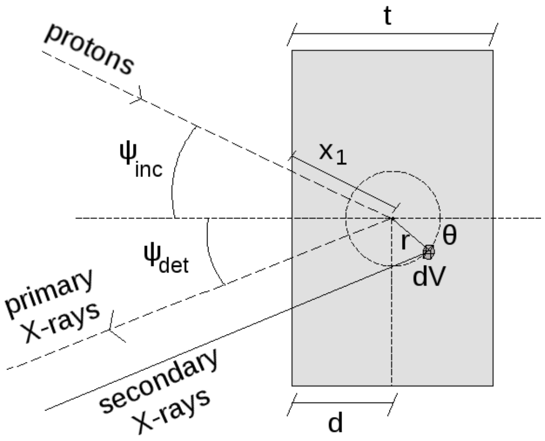
Probability of X-rays *A* produced by impact of particles at depth x

Probability that an absorbed X-ray *A* produces a secondary X-ray *B*

Density of probability of a X-ray *A* being absorbed at a distance *r* from the emission point

Transmission factor of the X-rays *B* from the volume *dV* up to the target surface, calculated in the detector direction

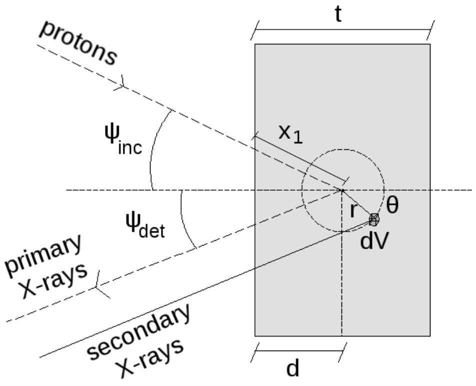
2D infinite sample



Secondary fluorescence emitted by the target is referred to the point where the emission of the primary radiation takes place

Reis, M. A., Alves, L. C., Jesus, A. P., Nucl. Instrum. Meth. B, 109/110: 134-138, 1996

SECONDARY X-RAYS



PENETRATION FUNCTION MODEL (REIS1996)

$$d\chi_{B,A} = P_A(x) R_{B,A} Q_A(r) T_B(x_1, r, \theta) dV$$

⇒ Probability of primary X-rays: $P_A(x) = \sigma_A^X(E_p),$

⇒ Probability that XR$_A$ produces XR$_B$: $R_{B,A} = \frac{\sigma_B^{photo}(E_A)\omega_B k_B}{\mu(A)} f_B$

⇒ Absorption of XR$_A$ at r from x: $Q_A(r) = \frac{\mu(A)}{4\pi r^2} e^{-\mu(A)r}$

⇒ Transmission of XR$_B$ to surface: $T_B(r_1, r, \theta) = e^{-\frac{\mu(B)(x_1 \cos\Psi_{inc} + r\cos\theta)}{\cos\Psi_{det}}}$

$$Q_{B,A}(x_1) = \int_{V_{target}} Q_A(r) T_B(x_1, r, \theta) dV$$

⇒ Total number of secondary X-rays in thick sample:

$$\chi_{B,A} = P_A(x) R_{B,A} A(x_1) \left[I_1 + e^{\frac{\mu(B)}{\cos\Psi_{det}}} E_1[\mu(A) x \cos\Psi_{inc}] - E_1[g^+ x \cos\Psi_{inc}] \right]$$

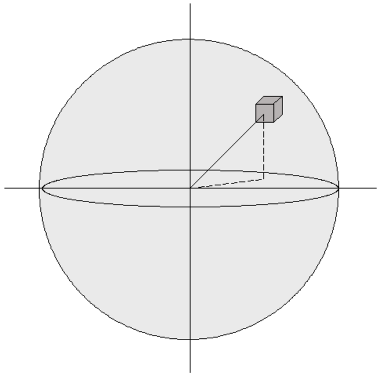
GENERAL MODEL

PENETRATION FUNCTION MODEL - advantages

⇒ **Secondary fluorescence emitted by the target is referred to the point where the emission of the primary radiation takes place**

⇒ **For 2D infinite samples: analytical expressions**

⇒ **Especially useful for spherical samples**



⇒ **Number of particles *B* produced in a volume *dV* induced in *dV* by particles *A* (particles: X-rays, Auger e$^-$)**

$$d\chi_{B,A} = P_A(x) R_{B,A} Q_A(r) T_B(x_1, r, \theta) dV$$

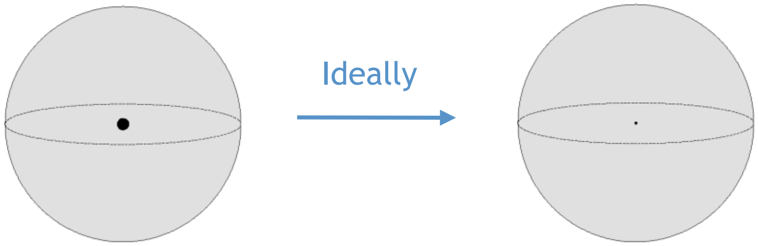
Determination of $P_A(x)$, R_{BA}, $Q_A(r)$ and $T_B(x_1,r,\theta)$!

GEOMETRY CONSIDERATIONS

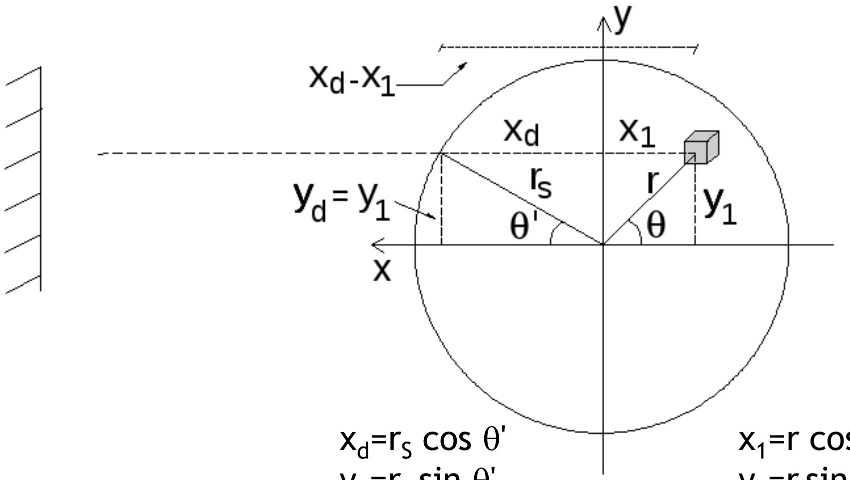
CENTRAL POINT EMISSION

⇒ **Nanoparticle described by ideal geometrical model**

Fe core: ∅ 5 nm, Si coating: ∅ 200 nm



Detector >> nanoparticle



$x_d = r_s \cos\theta'$
$y_d = r_s \sin\theta'$

$x_1 = r \cos\theta$
$y_1 = r \sin\theta$

Distance inside sphere: $x_d - x_1$

$y_d = y_1$

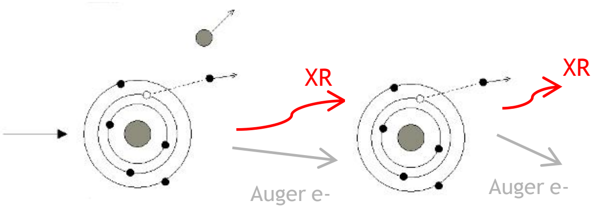
$\theta' = \arcsin(r \sin\theta / r_s)$

First step:

primary X-ray -> secondary X-ray

for homogeneous sphere

PRIMARY X-RAY



SECONDARY X-RAY

$$d\chi^{X_p,X_s} = P^{X_p}(E_H)R^{X_p,X_s}Q^{X_p}(r)T^{X_s}(r_S,r,\theta)dV$$

- $T^{X_s}(r_S,r,\theta) = e^{-\mu(E_{X_s})[x_d - x_1]} = e^{-\mu(E_{X_s})\left[\sqrt{r_S^2 - r^2\sin^2\theta} - r\cos\theta\right]}$
- $Q^{X_p,X_s} = \int_{V_{sphere}} Q^{X_p}(r)T^{X_s}(r_S,r,\theta)dV$
- $Q^{X_p,X_s} = \int_0^{r_S}\int_{-1}^{1}\frac{\mu(E_{X_p})}{2}e^{-\mu(E_{X_p})r}e^{-\mu(E_{X_s})\left[\sqrt{r_S^2 - r^2(1-y^2)} + ry\right]}dydr$

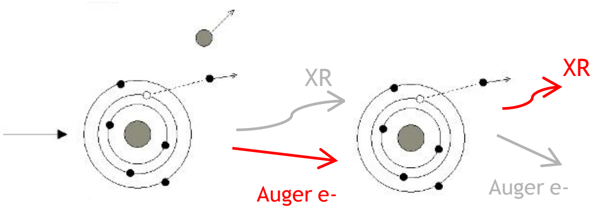
$y = -cos\theta$

⇒ **Number of secondary X-rays:**

$$\chi^{X_p,X_s} = \sigma^{X_p}(E_H)\frac{\sigma_s^{photo}(E_{X_p})\omega_s k_{X_s}}{\mu(E_{X_p})}f_s.$$

$$.\quad\int_0^{r_S}\int_{-1}^{1}\frac{\mu(E_{X_p})}{2}e^{-\mu(E_{X_p})r}e^{-\mu(E_{X_s})\left[\sqrt{r_S^2 - r^2(1-y^2)} + ry\right]}dydr$$

PRIMARY AUGER E$^-$



SECONDARY X-RAY

⇒ **Fluorescence yield and Auger yield**

⇒ Fe(Z=26): ω_K=0.340, a_K=(1-ω_K)=0.660; Si(Z=14): ω_K= 0.050, a_K=0.950

⇒ **Energy loss of primary particles**

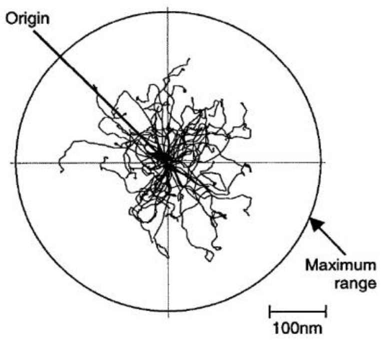


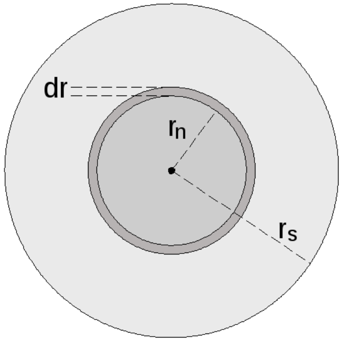
Figure: Electron trajectories in silicon at initial energy 3.5 keV (Fig 3 in Kataoka1999)

$$E(r) = E_0 - \int_0^{rn} \frac{dT}{dr} dr$$

E_0 : initial energy of primary electrons

$-dT/dr$: total energy loss

r_n : distance to dV

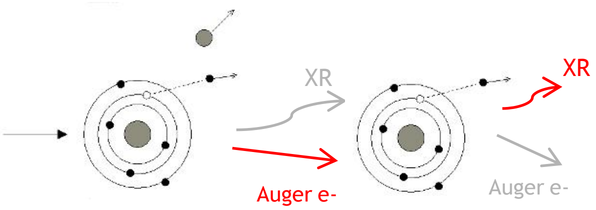


⇒ **Number of electrons at position r:**

$$N(r) = N_0 - \int_0^{r_n} N(r)\sigma^{I,e}(E_r) dr$$

M. O. Krause, J. Phys. Chem. Ref. Data, 8: 307-327, 1979
Y. Kataoka, N. Kawahara, T. Arai, M. Uda, JCPDS-International Centre for Diffraction Data, 1999, 76-83

PRIMARY AUGER E$^-$



SECONDARY X-RAY

$$\Rightarrow \quad d\chi^{e_p,X_s} = P^{e_p}(E_H,r)R^{e_p,X_s}Q^{e_p}(r)T^{X_s}(r_S,r,\theta)dV$$

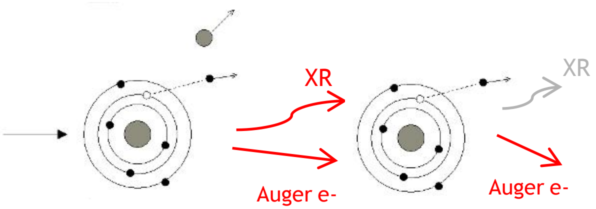
⇒ **Absorption of primary electrons**

- $R^{e_p,X_s} \equiv 1$
- $Q^{e_p,X_s}(r)dV = \dfrac{\sigma_s^{I,e}(E_r)\omega_s k_{X_s} f_s}{4\pi r^2}dV$

⇒ **Number of secondary X-rays:**

- $$\chi^{e_p,X_s} = \int_{V_{sphere}} P^{e_p}(E_H,r)Q^{e_p,X_s}(r)T^{X_s}(r_S,r,\theta)dV$$
- $$\chi^{e_p,X_s} = \frac{\omega_s k_{X_s} f_s}{2}\int_0^{r_S}\int_{-1}^{1} P^{e_p}(E_H,r)\sigma_s^{I,e}(E_r)e^{-\mu(E_{X_s})\left[\sqrt{r_S^2-r^2(1-y^2)}+ry\right]}dydr$$
- $$P^{e_p}(E_H,r) = N(0) - \int_0^{r_n} N(r)\sigma_s^{I,e}(E_r)dr$$

SECONDARY AUGER E$^-$



PRIMARY X-RAY OR PRIMARY AUGER E$^-$

⇒ **Transmission of secondary electrons**

$$T^{e_s}(r_S, r, \theta) = e^{-\mu_m(E_r)\left[\sqrt{r_S^2 - r^2\sin^2\theta} - r\cos\theta\right]}$$

⇒ **Primary X-ray - secondary Auger e$^-$**

$$\chi^{X_p,e_s} = \sigma^{X_p}(E_H)\frac{\sigma_s^{photo}(E_{X_p})(1-\omega_s)k_{e_s}}{\mu(E_{X_p})} f_s . \quad . \int_0^{r_S}\int_{-1}^{1} \frac{\mu(E_{X_p})}{2} e^{-\mu(E_{X_p})r} e^{-\mu_m(E_r)\left[\sqrt{r_S^2-r^2(1-y^2)}+ry\right]} dydr$$

⇒ **Primary Auger e$^-$ - secondary Auger e$^-$**

$$\chi^{e_p,e_s} = \frac{(1-\omega_s)k_{e_s}f_s}{2}\int_0^{r_S}\int_{-1}^{1} P^{e_p}(E_H,r)\sigma_s^{I,e}(E_r) e^{-\mu_m(E_{X_s})\left[\sqrt{r_S^2-r^2(1-y^2)}+ry\right]} dydr$$

APPLICATIONS

X-RAY EMITTING NANOPARTICLE

⇒ Fe core coated with Si

Central point: Fe(Z=26)
Sphere: Si (Z=14)
Sphere radius (r_s): 100 nm

Ep=3.0 MeV
E(Si-KL23)=1.740 keV

X-ray transition: KL23
Auger transition: K-L23L23

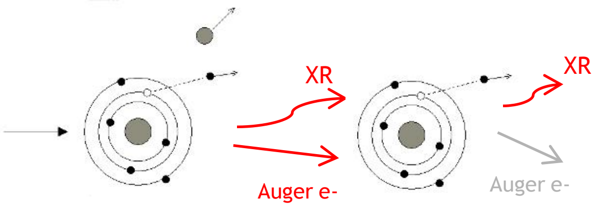
Fe X-ray production cross-section: 145.67 b
Fe Auger e^- production cross-section: 213.99 b

Fe fluorescence yield: 0.34
Si fluorescence yield: 0.05

⇒ How many Si KL23 X-rays exit the nanoparticle?

$$\chi^{X,X}_{Si,Fe} = 0.0097$$

$$\chi^{e,X}_{Si,Fe} = 0.0140$$



M. O. Krause, J. Phys. Chem. Ref. Data, 8: 307-327, 1979
J. H. Scofield, At. Data Nucl. Data Tables, 14: 121-137, 1974
F. P. Larkins, At. Data Nucl. Data Tables, 20: 311-387, 1977
D. L. Walters, C. P. Bhalla, Atomic Data, 3: 301-315, 1971

APPLICATIONS

AUGER E⁻ EMITTING NANOPARTICLES

⇒ **Fe core coated with Si**

Central point: Fe(Z=26)
Sphere: Si (Z=14)
Sphere radius (r_s): 100 nm

Ep=3.0 MeV
E(Si-K-L23KL23)=1.846 keV

X-ray transition: KL23
Auger transition: K-L23L23

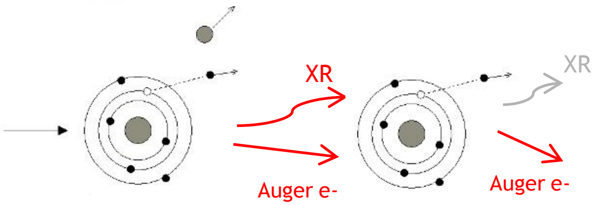
Fe X-ray production cross-section: 145.67 b
Fe Auger e^- production cross-section: 213.99 b

Fe fluorescence yield: 0.34
Si fluorescence yield: 0.05

⇒ **How many Si K-L23L23 Auger electrons exit the nanoparticle?**

$$\chi_{Si,Fe}^{X,e} = 0.1239$$

$$\chi_{Si,Fe}^{e,e} = 0.1778$$



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SUMMARY AND CONCLUSIONS

⇒ Secondary X-ray and electron emission expressions have been obtained for primary X-rays and Auger electrons for an ideal spherical model

⇒ Can be used for PIXE analysis (impact of particles inducing the primary particles emission) or consider an emitting radionuclide

⇒ As a simple model it can be used for the validation and interconparison with results from Monte Carlo codes

⇒ Number of electrons or X-rays leaving the nanoparticle fundamental for the accurate account of deposited energy (and possible damage to cells)

⇒ To be further developed to become a more realistic model: one direction for exiting the nanoparticle and only one step of electron cascade is not realistic

THANK YOU

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