Development of a discrete energy-loss model for low-energy electrons in gold using the ELF approach

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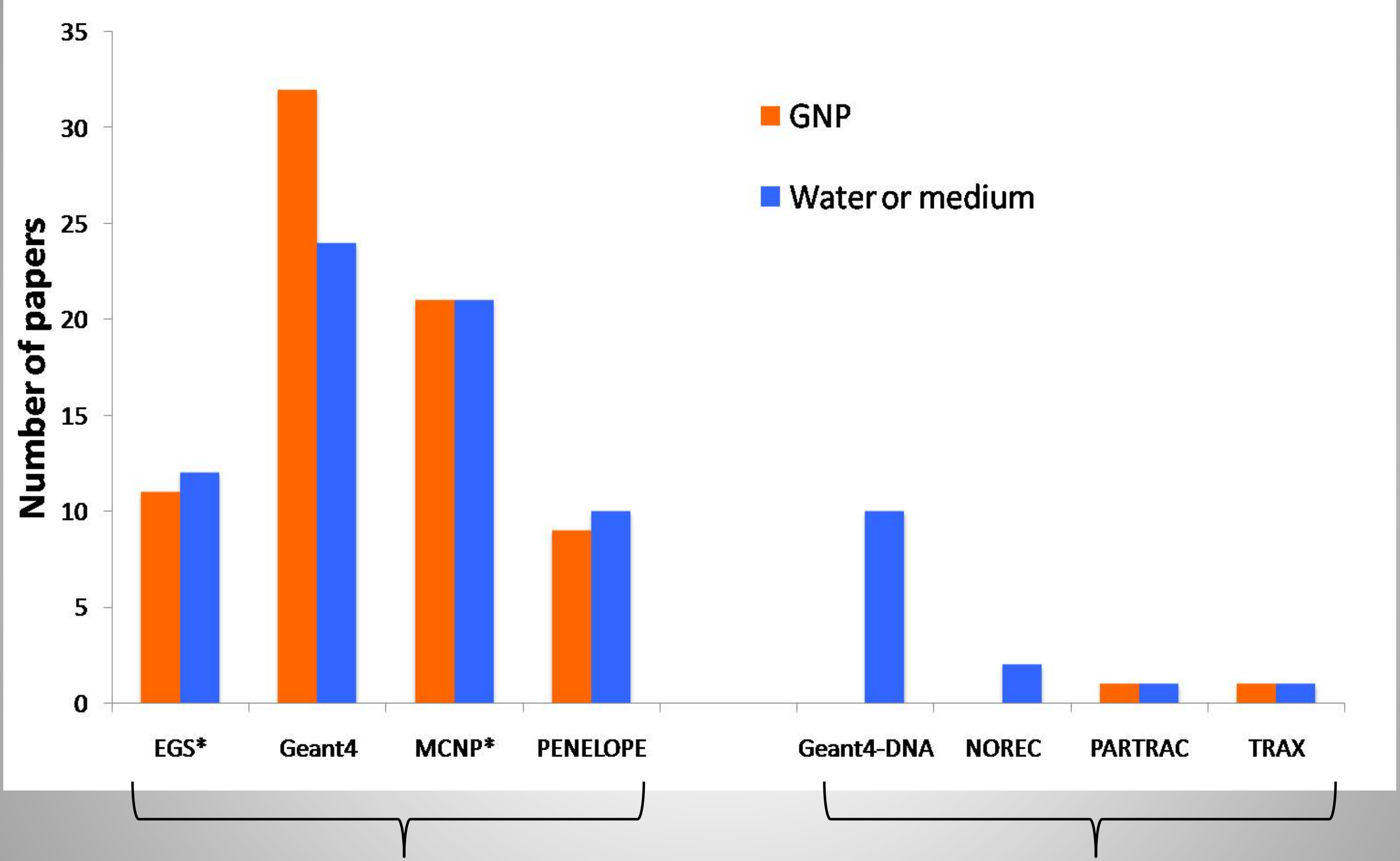
Why discrete electron models for gold are needed?

- Gold nanoparticles (GNPs) are currently being studied as a mean to increase therapeutic efficacy in radiotherapy
- GNP enhance the local dose under keV photon irradiation because of their higher photo-absorption cross section compared to water
- The increase of local dose around the GNP is mainly due to the emission of a large number of low-energy Auger electrons
- Discrete physics models are necessary for studying the energy deposition at the **nanoscale** by these low-energy (sub-keV) electrons

Papers on Monte Carlo GNP radioenhancement as of August 2018



Most studies have used condensed-history or “macroscopic” MC codes which are NOT suitable for the nanoscale

Only a small number of studies have used track-structure MC codes which offer nanometer resolution

The first discrete GNP electron models of Geant4 (GNP2016)

- Developed by Sakata and co-workers in 2016 (J. Appl. Phys.)
- GNP2016 models improve the low-energy EM models of Geant4 (Livermore, Penelope) in two important ways:

✓ Allow event-by-event electron transport in gold medium which is needed for GNP radioenhancement studies

✓ Allow much higher spatial resolution by “safely” extending the tracking & production cutoff energies down to 10 eV

Motivation for a new model

Deficiencies of the GNP2016 energy-loss model:

- ✓ The energy-loss channels (ionizations, excitations, plasmon) are treated by different theories
  - o **Not self-consistent**
- ✓ Ionization cross sections are based on an atomic model (RBEBV)
  - o **Condensed-phase effects are ignored**
  - o **Not justified at low energies (sub-keV)**
- ✓ Plasmon excitations are based on Quinn's model
  - o **Free-electron model**
  - o **Neglects plasmon damping (assumes infinite plasmon lifetime)**
  - o **Not accurate below few 100 eV**

Aim of this work (2018)

To provide an alternative discrete model for GNP based on the ELF (energy-loss-function) approach

- The benefits of the ELF approach:
  - ✓ It is robust
  - ✓ Self-consistency tests are available
  - ✓ It is based on experimental data for solid-Au
  - ✓ It accounts for condensed-phase effects

GNP2016 vs. GNP2018 models

Process	2016 models	2018 models
Elastic	ELSEPA	ELSEPA
Ionization	Rel. Binary Encounter Bethe Vriens	Dielectric
Excitation	Exp.+Dirac B-Spline R Matrix	Dielectric
Plasmon	Quinn	Dielectric
Bremsstrahlung	Seltzer & Berger	Seltzer & Berger

Methodology

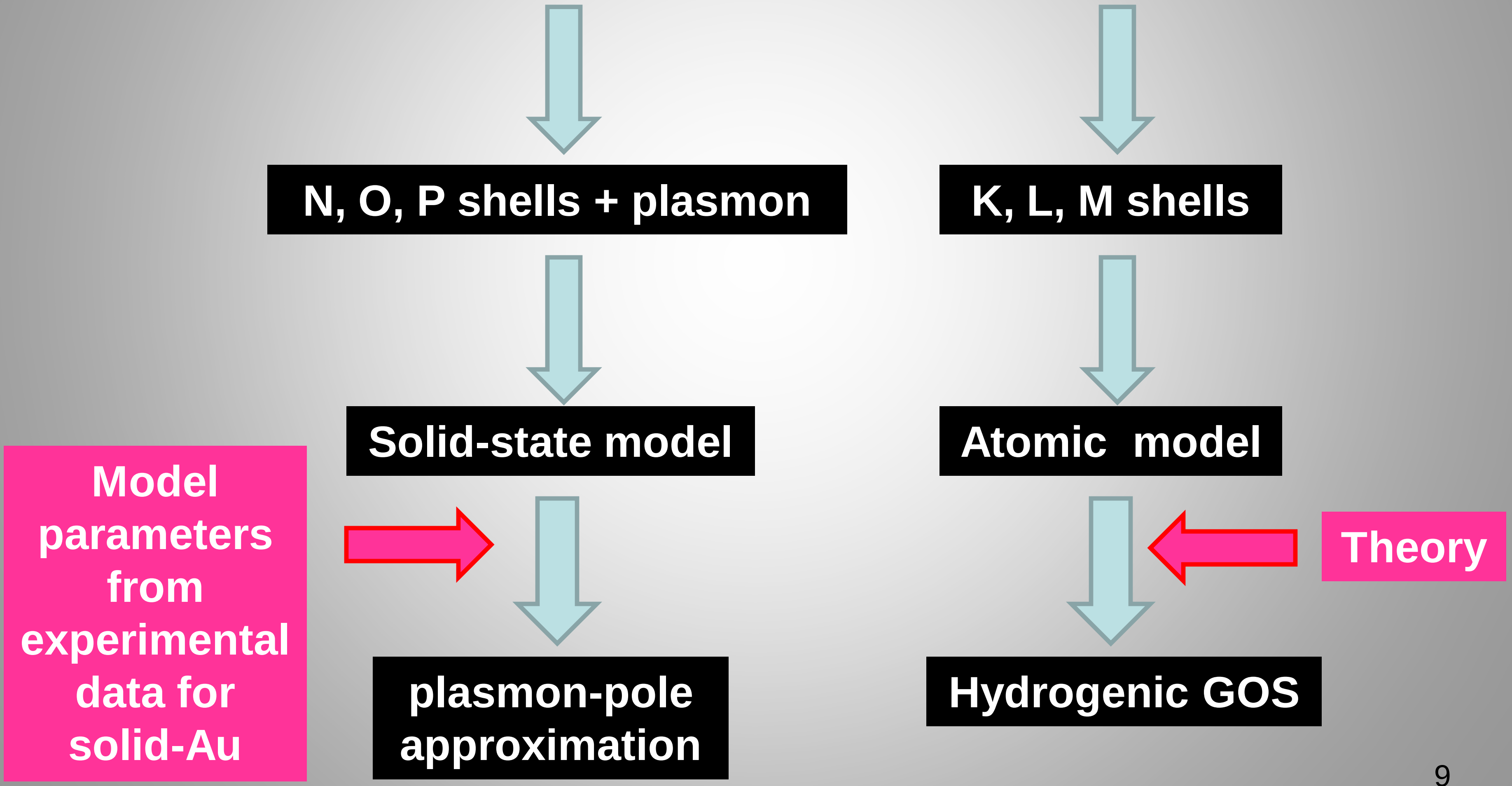
- The ELF approach starts from the double-differential cross section of the **plane-wave Born approximation (PWBA):**

$$\frac{d^2\sigma_{\mathrm{PWBA}}}{dEdq}=\frac{1}{\pi a_0 NZ}\frac{1}{T}\frac{1}{q}\overbrace{\mathrm{Im}\left[-\frac{1}{\underbrace{\varepsilon(E,q)}_{\text{Dielectric-response-function (DRF)}}}\right]}^{\text{Energy-loss-function (ELF)}}$$

In PWBA the ELF is the main material property to calculate energy-loss cross sections

ELF model for gold

$$\mathrm{ELF} = \mathrm{Im}\left[-\frac{1}{\varepsilon(E,q)}\right]_{\text{outer-shells}} + \mathrm{Im}\left[-\frac{1}{\varepsilon(E,q)}\right]_{\text{inner-shells}}$$



N, O, P shells + plasmon

K, L, M shells

Solid-state model

Atomic model

Model parameters from experimental data for solid-Au

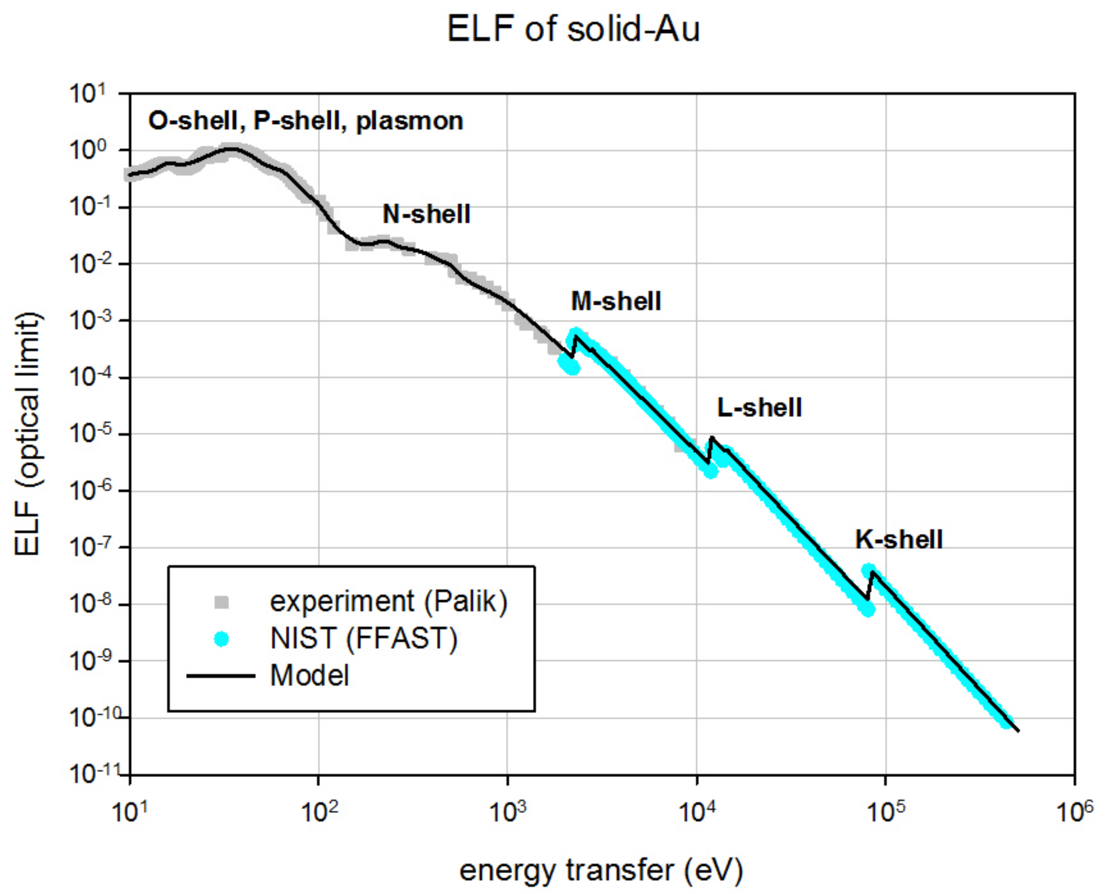
plasmon-pole approximation

Theory

Hydrogenic GOS

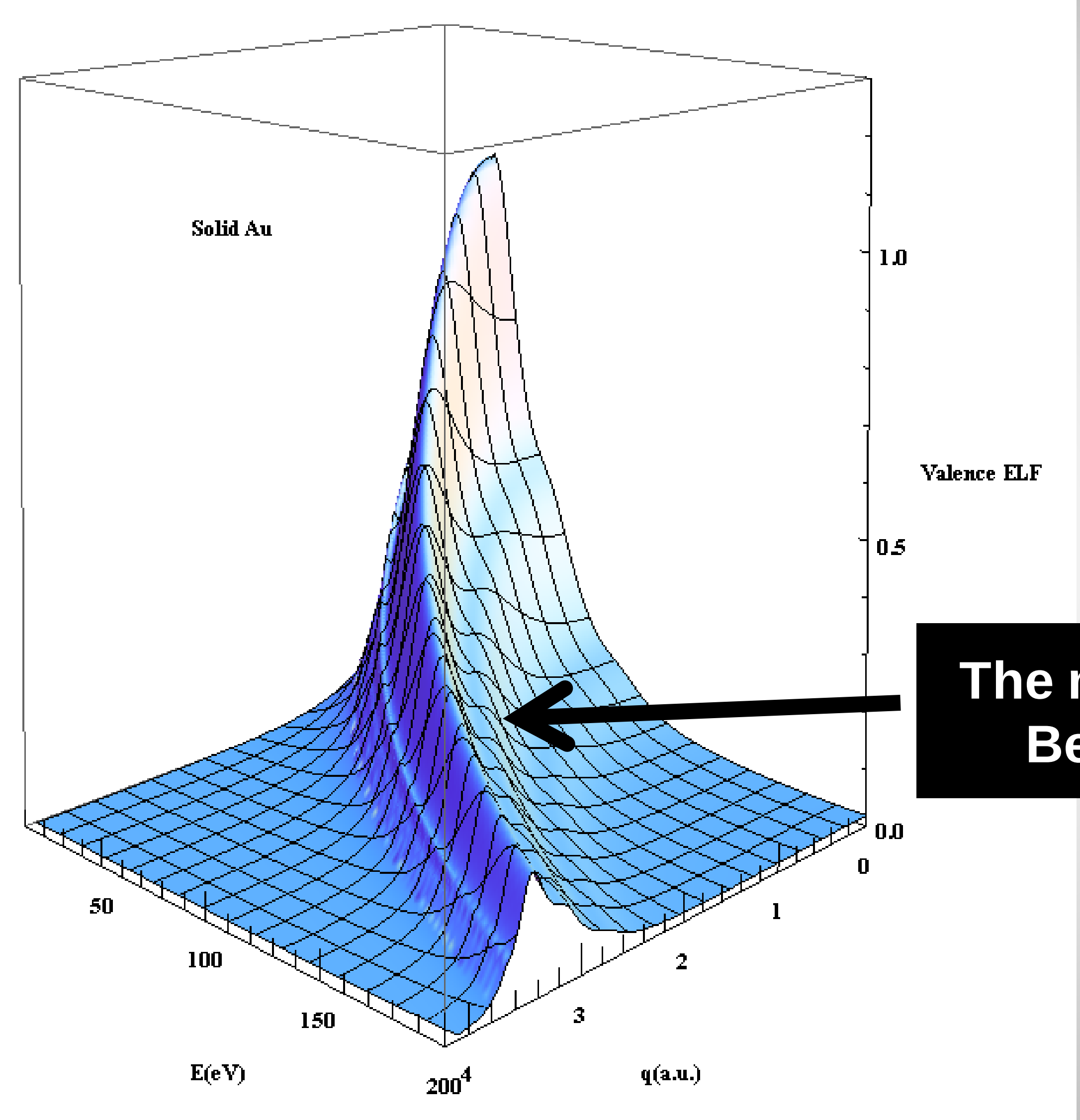
- The ELF model considers the following energy-loss channels above 10 eV:
  - ✓12 outer ionization sub-shells: N_1, N2, N3, N4-5, N6, N7, O1, O2, O3, O4, O5, P1 (binding energies from EADL)
  - ✓6 inner ionization sub-shells: K1, L1, L2-3, M1, M2-3, M4-5 (binding energies from EADL)
  - ✓Plasmon channel (~35 eV)

ELF model at q=0



Overall good representation of experimental & NIST data

ELF model at q>0



The model reproduces the Bethe ridge at high-q

First self-consistency test: The f-sum-rule test

f-sum-rule

$$Z = \frac{1}{2\pi^2 N}\int_0^\infty E\,\mathrm{Im}\underbrace{\left[-\frac{1}{\varepsilon(E,q=0)}\right]}_{\textbf{ELF model}} dE$$

Z(model) versus Z(Au)

difference < 1%

Second self-consistency test: The I-value test

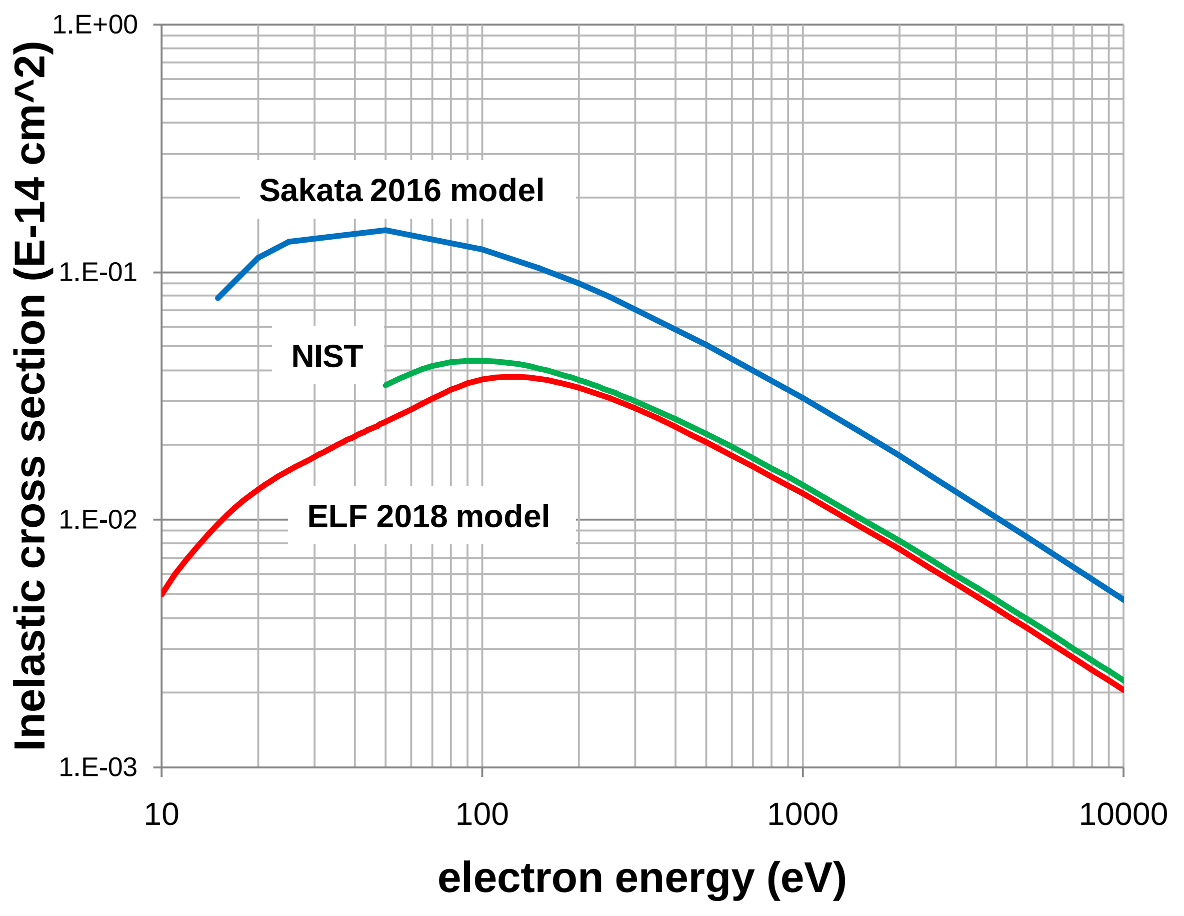
Mean excitation energy (I-value):

$$\ln I = \frac{\int_0^\infty E \ln(E) \, \mathrm{Im}\left[-\frac{1}{\varepsilon(E, q=0)}\right] dE}{\int_0^\infty E \, \mathrm{Im}\left[-\frac{1}{\varepsilon(E, q=0)}\right] dE}$$

I-value (model) vs. I-value (ICRU)

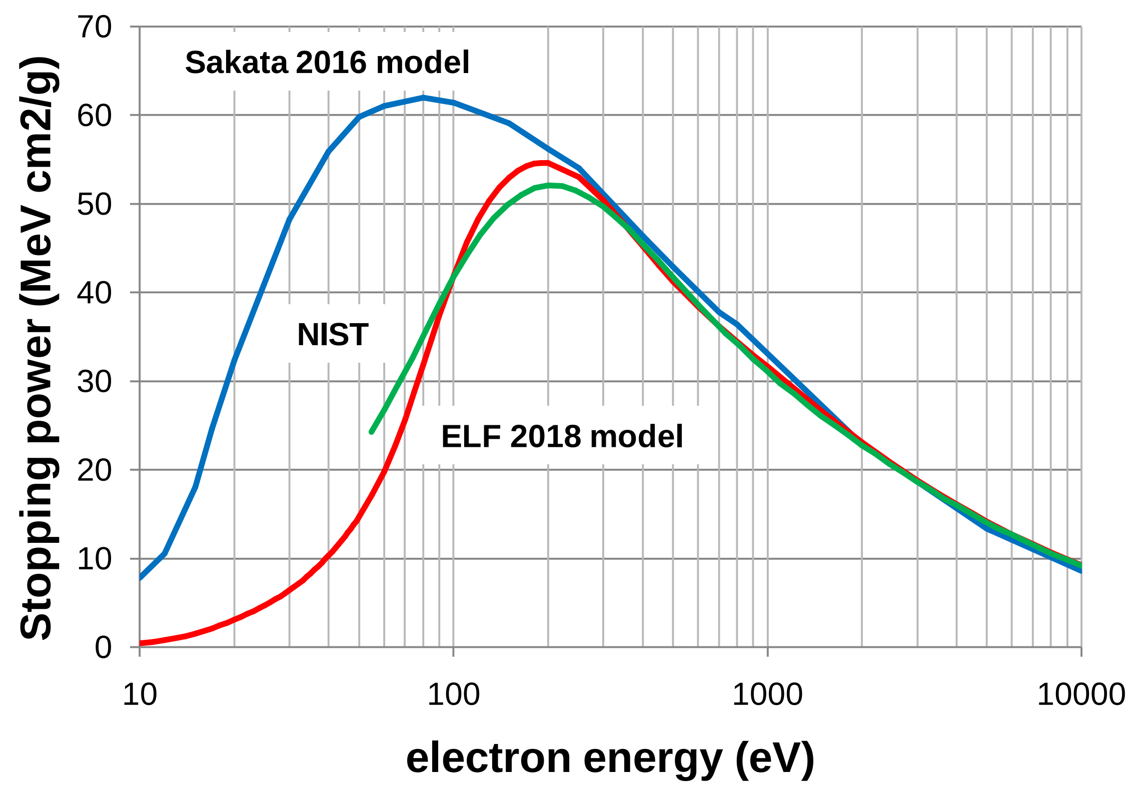
difference < 0.1%

Total inelastic cross section: comparison against NIST (TPP formula)



- ✓ Sakata 2016 is 200-300% higher than NIST
- ✓ ELF 2018 is in good agreement (~10%) with NIST

Stopping power: comparison against NIST (Shinotsuka et al. 2012)



- ✓ **Good agreement above few 100 eV**
- ✓ **Below 300 eV Sakata 2016 departs from both ELF 2018 and NIST**
- ✓ **ELF 2018 is in good agreement with NIST over the whole energy range**

Summary

- An alternative discrete model for gold has been developed based on the ELF approach. The new model uses experimental data for solid-Au and dielectric response theory
- The new model:
  - ✓ is (more) robust
  - ✓ satisfies important self-consistency tests
  - ✓ offers better agreement with available NIST data
- We recommend that the new model should be preferred in the energy range from 100 eV to 10 keV
- From 10 eV to 100 eV it must be considered as qualitative until further refinements are made (e.g., addition of non-Born effects, more elaborate dispersion relations, and discrete excitations)

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Thank you for the attention!