OPACITIES: LTE & NON-LTE

Christopher Fontes
Computational Physics Division
Los Alamos National Laboratory

ICTP-IAEA School on Atomic and Molecular Spectroscopy in Plasmas

Trieste, May 6-10, 2019

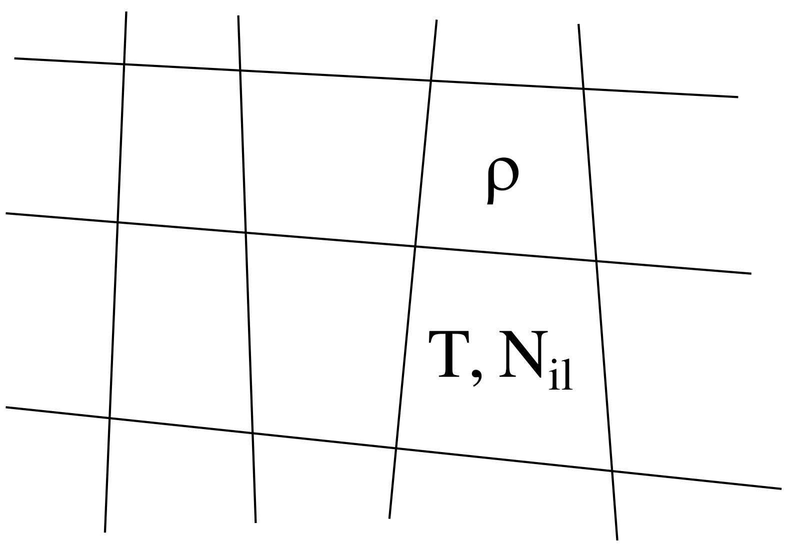
Acknowledgements

- This presentation contains information obtained from many conversations with members of the Los Alamos Atomic Data Team:

  Joe Abdallah
  Bob Clark
  James Colgan
  Peter Hakel
  Dave Kilcrease
  Norm Magee
  Manolo Sherrill
  Honglin Zhang

Some helpful illustrations

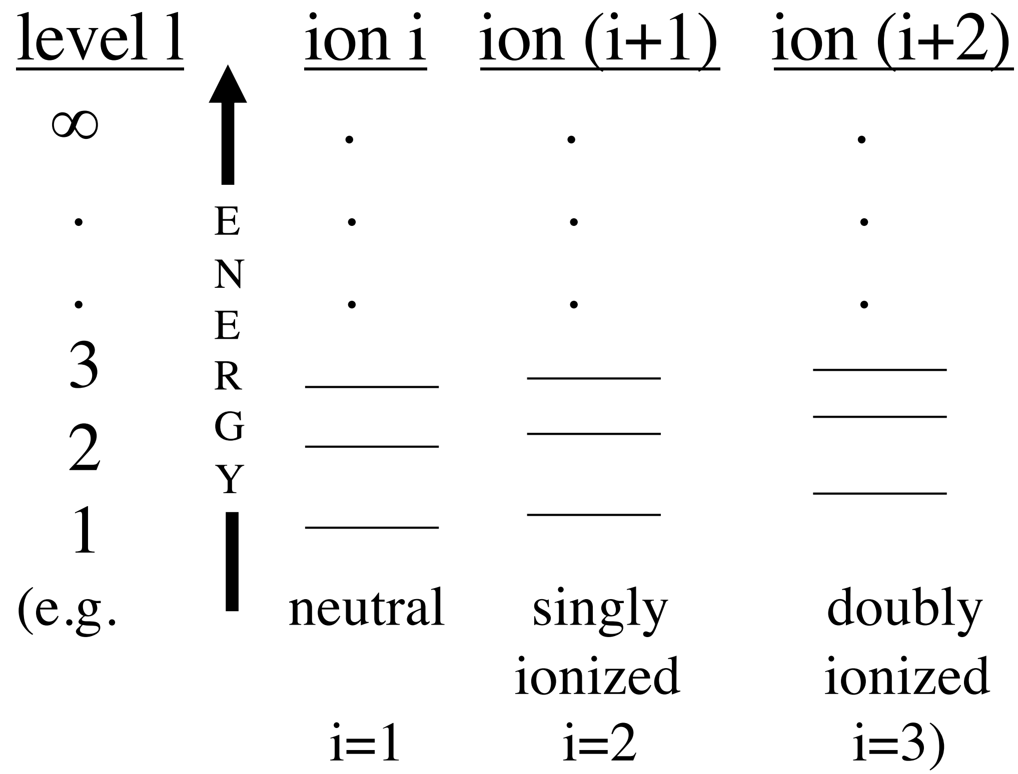
Our sample plasma made up of cells described by temperatures, densities, atomic populations, etc.



N_{il} = number density for level l, ion stage i

$[N_{il}] = 1/cm^3$

Our sample ions/atoms inhabiting each cell



The main players

- Photons (radiation field)
- Bound electrons (orbiting around the nucleus)
- Free electrons (formerly bound electrons that have been ionized by free electrons or photons)

- The photons and electrons interact via fundamental atomic processes, which can be used to determine the atomic level populations, N_{il}
- These populations can then be used to compute an opacity, κ_ν, which is used in radiation transport calculations

Explanatory definitions/symbols

- ρ - ion (or material) mass density: $[\rho]$ = grams/cm^3
- $N_I = \sum_{il} N_{il}$ - ion number density: $[N_I] = 1/\text{cm}^3$; $\{N_I = \rho(A_0/A)\}$
- N_e - free electron number density: $[N_e] = 1/\text{cm}^3$
- T or kT = temperature (ion, electron, radiation): [T] = eV
- $\bar{Z}(\rho,T)$ ("Z bar") or $<Z>$ - average charge state ($N_e = \bar{Z} N_I$)

radiation quantities

- $h\nu$ - photon energy: $[h\nu]$ = eV
- $\kappa_\nu(\rho,T)$ - opacity: $[\kappa_\nu]$ = cm^2/gram
- $\varepsilon_\nu(\rho,T)$ - emissivity: $[\varepsilon_\nu]$ = ergs/(gram sec Hz)
- I_ν - (isotropic) radiation intensity: $[I_\nu]$ = ergs/(cm^2 sec Hz)

Atomic kinetics modeling is an *ab-initio* effort

- There are far too many atomic processes to be measured experimentally
- Furthermore, there are not many experimental measurements of atomic physics data
- Nuclear data are obtained through evaluations which rely on both experimental data and theoretical calculations
- Atomic data (e.g. opacities) are obtained almost exclusively from first-principle calculations (quantum mechanics, wavefunctions, cross sections, etc.)

Road map to opacity

wavefunctions, level energies (Ψ_l, E_l)

$\downarrow$

fundamental cross sections ($\sigma_{l \to l'}$)

$\downarrow$

rate coefficients, rate equations

$\downarrow$

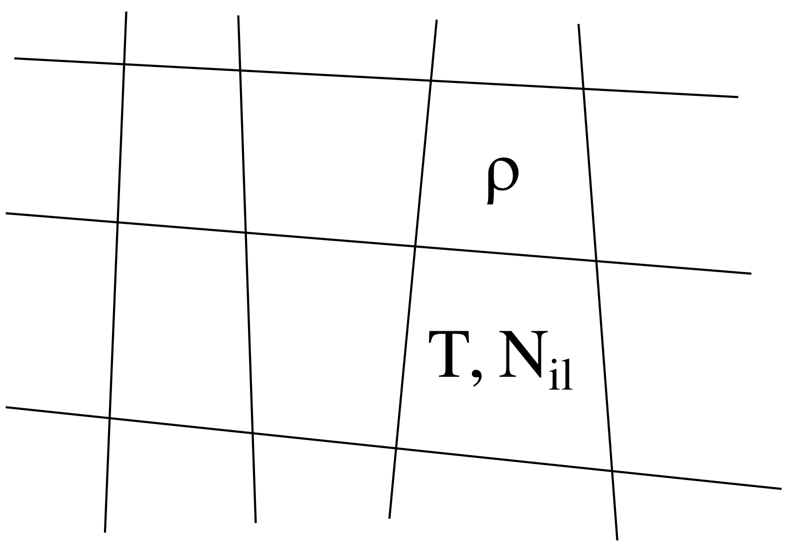
atomic level populations (N_{il})

$\downarrow$

opacity ($\kappa_\nu \sim N_{il} \times \sigma_{l \to l'}^{\text{photo}}$)

Excitation and de-excitation processes

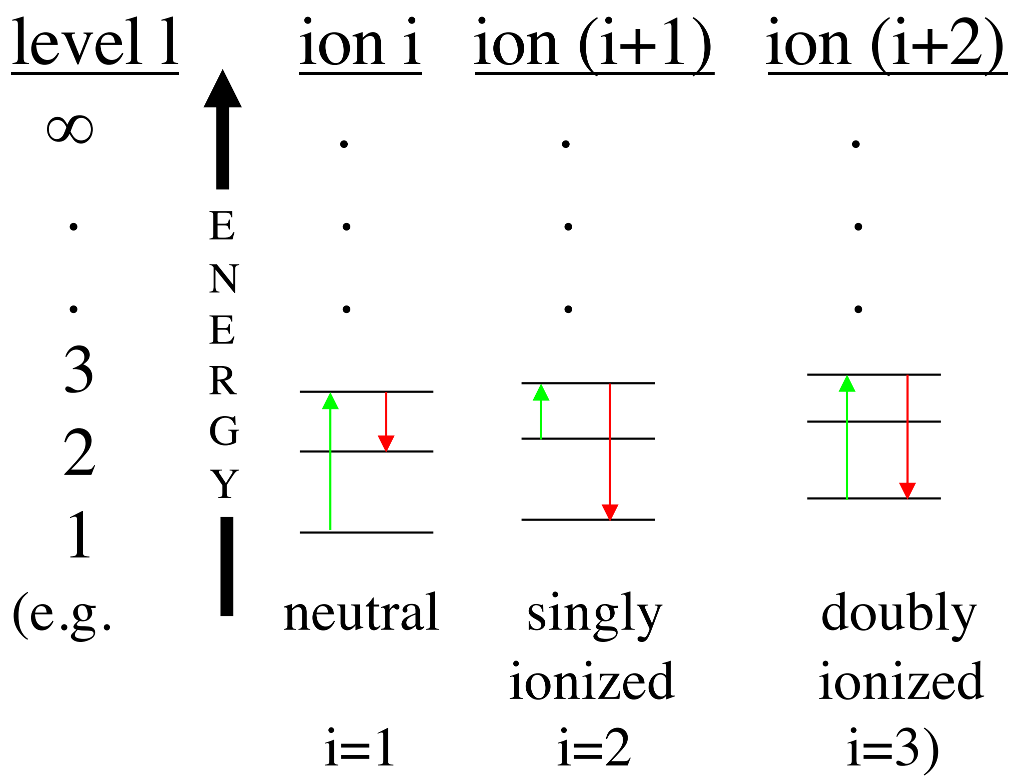
Our sample plasma made up of cells described by temperatures, densities, atomic populations, etc.



N_{il} = number density for level l, ion stage i

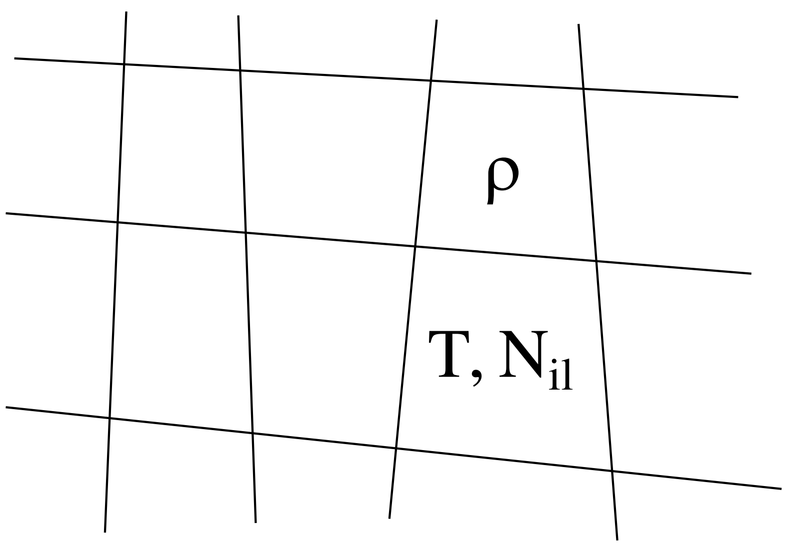
$[N_{il}]$ = 1/cm^3

Our sample ions/atoms inhabiting each cell



Ionization and recombination processes

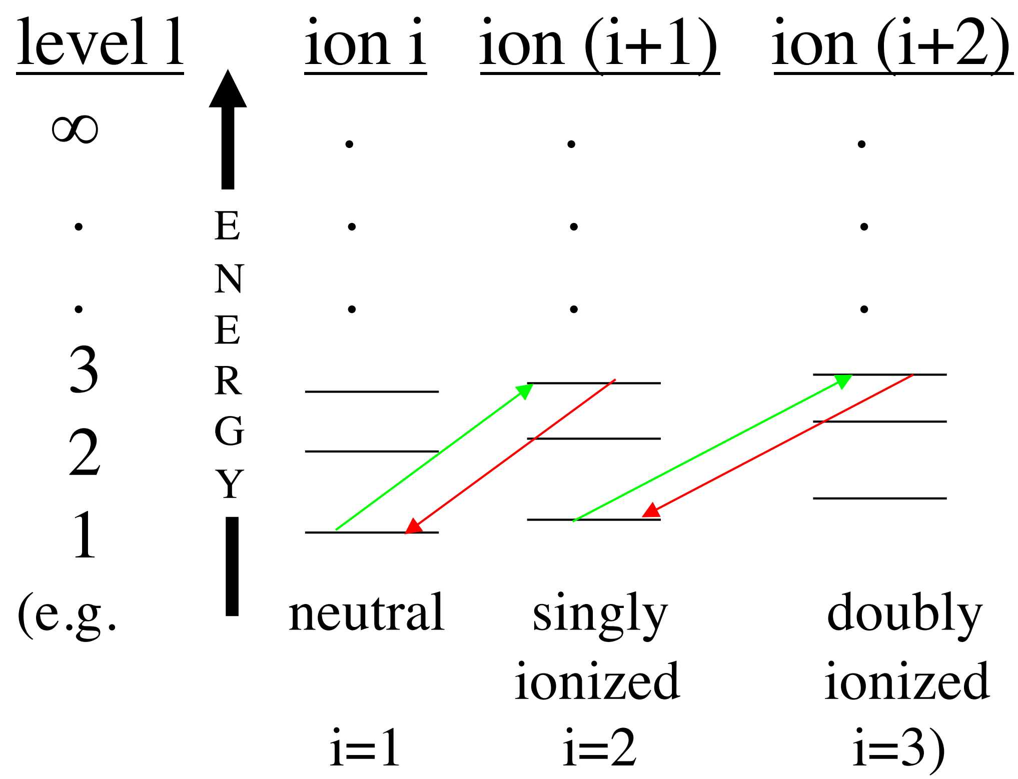
Our sample plasma made up of cells described by temperatures, densities, atomic populations, etc.



N_{il} = number density for level l, ion stage i

$[N_{il}] = 1/cm^3$

Our sample ions/atoms inhabiting each cell



Solving for the atomic level populations, N_{il}

- To obtain an opacity at each point in our sample plasma, we require the fundamental cross sections and the level populations, N_{il}
- The level populations are determined by the following basic atomic processes and their inverses:

process	inverse process
photoexcitation	photo de-excitation
photoionization	radiative recombination
electron collisional excitation	electron collisional de-excitation
electron collisional ionization	three-body recombination
autoionization	dielectronic recombination

- The cross sections for these processes are used in coupled, differential equations, known as "rate equations", which determine the populations N_{il}

The rate equations

- In general, the level populations vary as a function of time
- One must consider all possible processes that can populate and depopulate each level
- The result is a set of non-linear, first-order differential equations
- $\frac{dN_{il}}{dt} =$ (Formation rates) - (Destruction rates)
- In matrix form

$$\begin{pmatrix} dN_{11}/dt \\ \ldots \\ dN_{il}/dt \\ \ldots \\ dN_{nn}/dt \end{pmatrix} = \begin{pmatrix} R_{11} & . & R_{1l} & . & R_{1n} \\ . & . & . & . & . \\ R_{i1} & . & R_{il} & . & R_{in} \\ . & . & . & . & . \\ R_{n1} & . & R_{nl} & . & R_{nn} \end{pmatrix} \begin{pmatrix} N_{11} \\ \ldots \\ N_{il} \\ \ldots \\ N_{nn} \end{pmatrix}$$

The rate equations (continued)

- The order of the rate matrix can vary greatly depending on the complexity of the atomic model
- Average-atom: order ~10, very crude, very fast to compute
- Configuration-average: order ~100-10^7, good compromise, some spectral detail, but maybe not enough to produce high-resolution spectra
- Fine-structure: order ~100-10^{10}, spectrally resolved features, very accurate if complete model can be considered, but can be impractical to solve numerically

A specific example: collisional excitation/de-excitation

- Each element of the rate matrix is computed from fundamental cross sections associated with each process
- Consider collisional excitation/de-excitation as a specific example:

$$\frac{dN_{il}}{dt} = (\text{“rate” of excitations into il}) - (\text{“rate” of de-excitations out of il}) + \ldots$$

$$= \sum_{im} [s(im,il;T)N_e N_{im} - t(im,il;T)N_e N_{il}] + \ldots$$

- s(im,il;T) is the “rate coefficient” for electron collisional excitation from level m to level l in ion stage i, symbolically written as

$$e^- + (\mathrm{im}) \rightarrow (e^-)' + (\mathrm{il})$$

- Similarly, t(im,il;T) represents the rate coefficient for all possible collisional de-excitations into level l of ion stage i

A specific example: collisional excitation/de-excitation (continued)

- The result looks like

$$\mathrm{s}(\mathrm{im},\mathrm{il};T_e) = \int_{E_0}^{\infty} F(E,T_e)\,\mathrm{v}(E)\boldsymbol{\sigma}_{\mathrm{iml}}(E)\,dE$$

- $F(E,T_e)$ is the free-electron energy distribution function
- $\mathrm{v}(E)$ is the velocity of a free electron $[\mathrm{v}(E) = \sqrt{(2E)/m_e}]$
- $\boldsymbol{\sigma}_{\mathrm{iml}}(E)$ is the excitation cross section
- E_0 is the threshold energy, above which excitation can occur

---

- The ***rate*** at which excitations occur from level m to level l is s(im,il;T)N_e and the ***rate per unit volume*** is s(im,il;T)N_eN_{im}

A specific example: collisional excitation/de-excitation (continued)

- The rate coefficients for collisional de-excitation are determined from the principle of detailed balance and can also be expressed in terms of the same excitation cross section
- The rate coefficients for the remaining collisional and photo processes are determined in a similar fashion
- Just as electron-collision processes require a knowledge of the free-electron energy distribution function, $F(E,T_e)$, photo processes require that the photon energy distribution function also be specified
- These concepts lead naturally to a discussion of LTE vs. non-LTE (NLTE) atomic physics

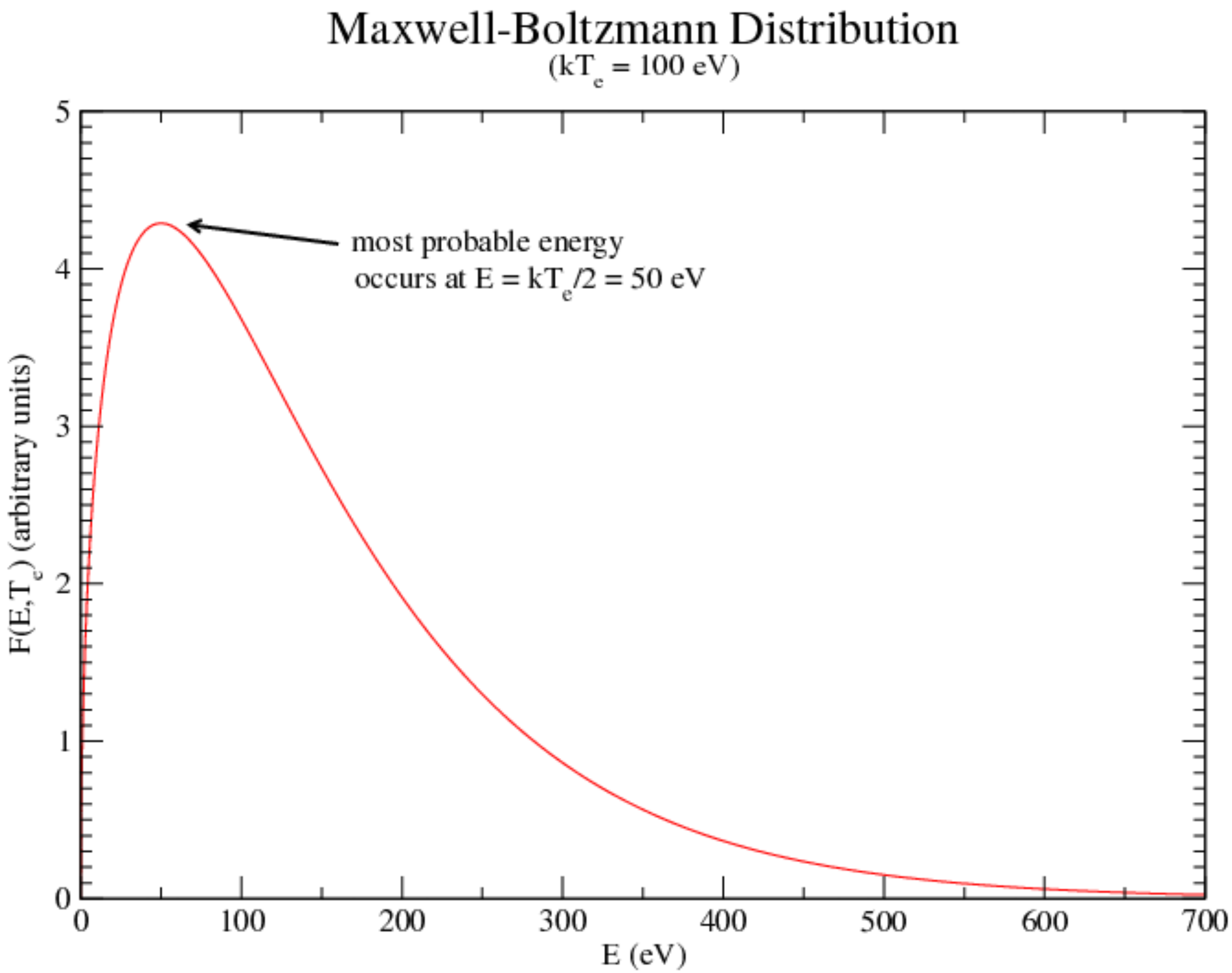
Free electrons in thermodynamic equilibrium

- If the free electrons are in thermodynamic equilibrium (TE) with themselves, then the energy distribution is given by the Maxwell-Boltzmann distribution at an electron temperature T_e

$$F(E,T_e) = \frac{2}{\sqrt{\pi}} \frac{\sqrt{E}}{(kT_e)^{3/2}} e^{-E/kT_e}$$

- This distribution represents the fraction of electrons per unit energy interval that have energies between E and E+dE

Maxwellian distribution at kT_e=100 eV



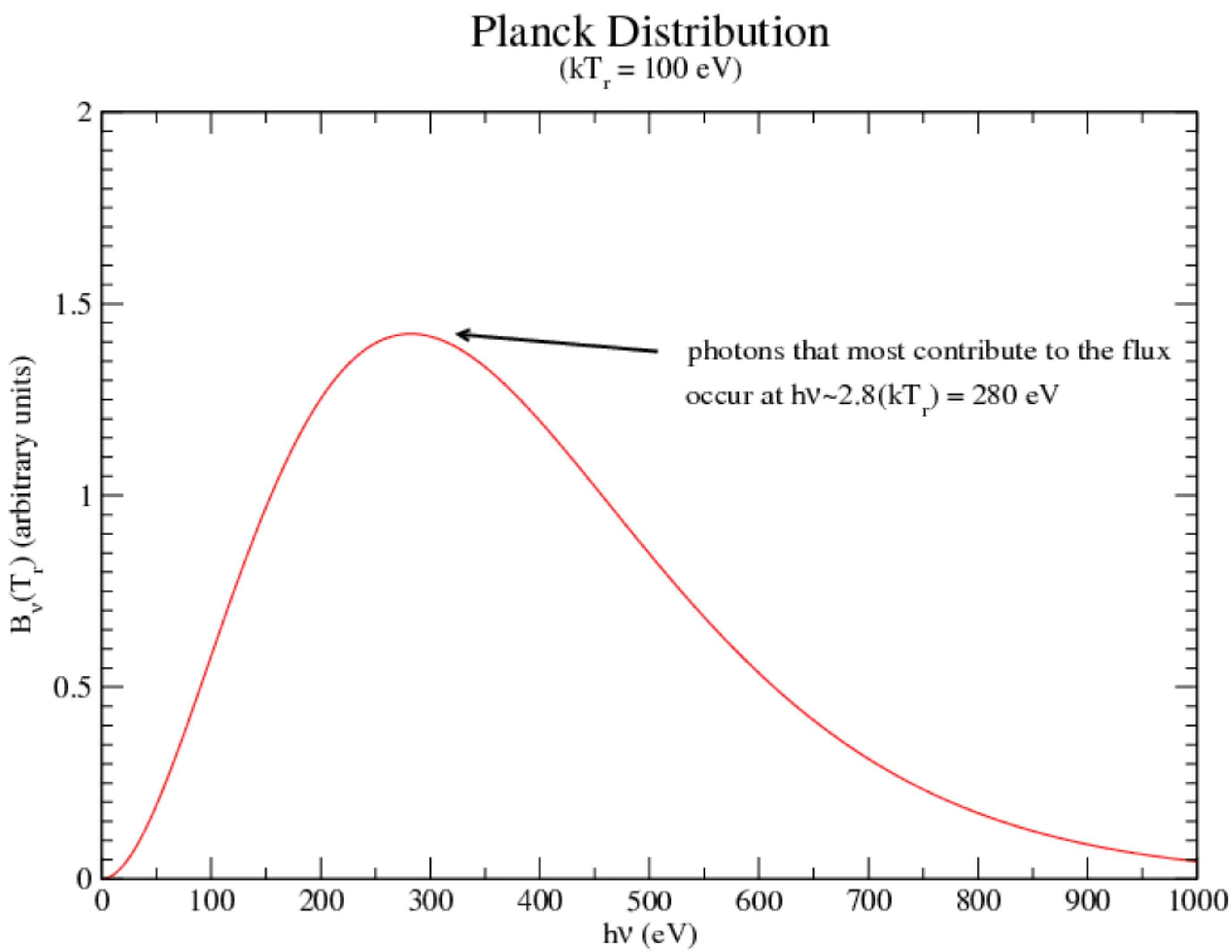
Photons in thermodynamic equilibrium

- Similarly, if the photons are in thermodynamic equilibrium (TE) with themselves, then the energy density distribution is given by the Planck distribution at a radiation temperature T_r

$$B_\nu(T_r) = \frac{2}{(hc)^2} \frac{(h\nu)^3}{e^{h\nu/kT_r} - 1}$$

- This is a flux distribution that represents the amount of radiation energy per unit frequency interval per unit area per unit time per unit solid angle

Planckian distribution at kT_r=100 eV



Local Thermodynamic Equilibrium

- LTE = Local Thermodynamic Equilibrium
- LTE is valid at a particular point in the plasma if the electron and photon distributions are in equilibrium ***with each other*** : $T_e = T_r = T_I = T$. This is one of the "textbook" definitions of LTE.
- There are other descriptions of LTE...

LTE applies if any of the following are true:

- At a given point in the plasma, the (atomic) conditions can be described by a single temperature ($T=T_e=T_r=T_I$)
- The rate at which any atomic process occurs is exactly balanced by the rate of its inverse process (this condition makes the physics much simpler to deal with than NLTE)
- The energy distribution of the free electrons in the plasma is described by a Maxwellian distribution and the radiation field is described by a Planck function (all at the same temperature)
- The FREE ELECTRON density is so high that electron collisions dominate the various atomic processes ("collision-dominated plasma"). In this case, there is not a true balance between all processes, but the following, and perhaps most important, bullet is still true:

LTE from a practical (computational) perspective

- From a computational perspective, LTE means that the atomic level populations, N_{il}, can be solved from the (relatively) simple Saha equation and the Boltzmann relationship

$$\boxed{N_{\mathrm{il}} \propto (N_{\mathrm{i}})e^{-E_{\mathrm{il}}/kT}}$$

- In this case, the N_{il} can be determined from a simple analytic formula that depends on the energy and temperature; there is ***no need to consider the fundamental cross sections***.

- Solving the detailed rate equations with a Maxwellian electron distribution and a Planckian radiation distribution results in a steady-state solution $(dN_{\mathrm{il}}/dt = 0)$ which could have been found by solving the much simpler Botzmann relationship above

Non-LTE

- Non-LTE applies if:
  - LTE conditions are not satisfied (obviously!)
  - System is changing so rapidly that electron and/or photon energy distributions do not reach thermal equilibrium (i.e. Maxwellian or Planckian is not valid, lasers, $T_r \neq T_e$, etc.)
  - Optically thin plasma: radiation escapes and is not available to provide LTE balance among the fundamental atomic processes
- For the NLTE case, the detailed rate equations must be solved to obtain the atomic level populations, N_{il}
- In practice, this solution requires the use of large-scale computing
- NLTE calculations can take as much as 3-4 ***orders of magnitude*** more computing time than LTE calculations

Photon scattering

- One additional, fundamental process must be discussed before an opacity can be constructed: Compton scattering of photons

$$\gamma + e^- \rightarrow \gamma' + (e^-)'$$

- This process differs from free-free absorption in that the incident photon loses only a small portion of its energy when interacting with a free electron, then continues on with a slightly smaller energy

$$\sigma^{\mathrm{THOMSON}} = \frac{8\pi}{3}\left(\frac{\mathrm{e}^2}{m_\mathrm{e}c^2}\right)^2 = 6.66 \times 10^{-25}\,(\mathrm{cm}^2) \quad (h\nu << mc^2)$$

$$\sigma^{\mathrm{COMPTON}}(\nu) = G(\nu)\,\sigma^{\mathrm{THOMSON}}$$

- $G(\nu)$ is a relativistic correction factor that accounts for the case when the photon energy becomes comparable to the electron rest mass and the electron's kinetic energy is treated in a fully relativistic manner

What is an opacity?

- An opacity, κ_ν, describes the coupling between matter and radiation via electron-photon interactions
- Opacity gives a measure of how much radiation a certain material will absorb/scatter (i.e. how "opaque" is the material)
- An opacity can be thought of as a macroscopic quantity that is built up from fundamental atomic cross sections
- The amount of radiation that is absorbed/scattered (i.e. removed) from the ambient radiation field, I_ν, in each cell of our sample plasma is given by:

$$\kappa_\nu I_\nu$$

What is an emissivity?

- An emissivity, ε_ν, gives the amount of radiation that will be emitted by the material in a plasma via electron-photon interactions
- As with the opacity, an emissivity is calculated from fundamental atomic cross sections
- The amount of radiation that is emitted (i.e. added to) the ambient radiation field, I_ν, in each cell of our sample plasma is given by:

$$\varepsilon_\nu/(4\pi) \quad \text{(isotropic emitter)}$$

Why are opacities/emissivities important?

- These quantities are necessary to solve the radiation transport equation
- Assuming problem is time-independent and one-dimensional with isotropic radiation, the transport equation can be written:

emissivity

radiation intensity

$$\frac{1}{\rho}\frac{dI_\nu}{dx} = \frac{\varepsilon_\nu}{4\pi} - \kappa_\nu I_\nu$$

material density

opacity

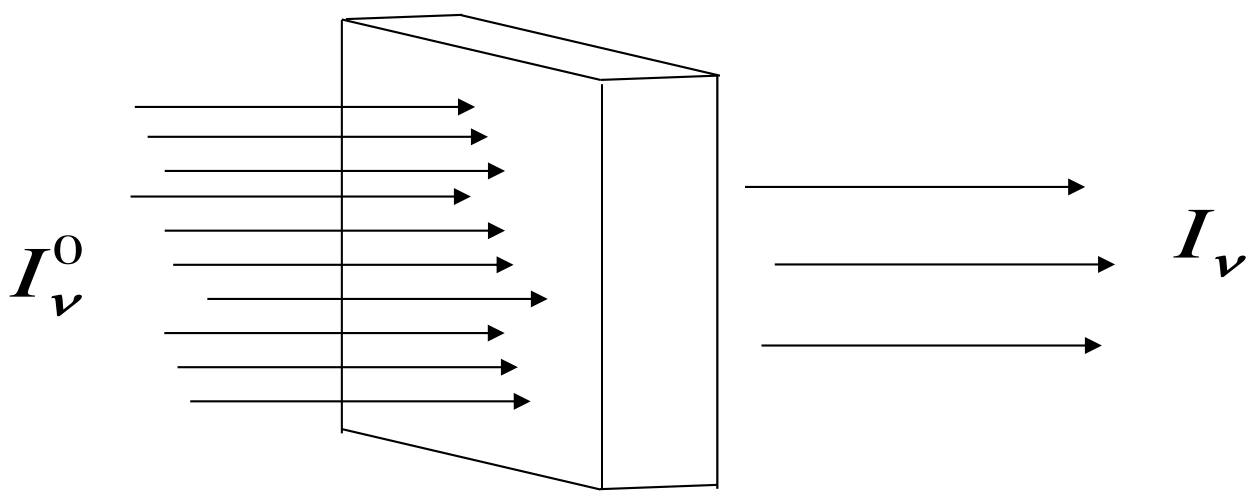
radiation frequency

The classic opacity (transmission) experiment: Optically thin plasma example

- If the plasma is "optically thin", then the emitted radiation will escape and need not be considered in the radiation transport equation:

$$\frac{1}{\rho}\frac{dI_\nu}{dx} = \cancel{\frac{\varepsilon_\nu}{4\pi}} - \kappa_\nu I_\nu$$

- This situation can be illustrated by the following diagram:



A beam of photons passing through a slab of thickness t.

Optically thin plasma example (continued)

- The previous differential equation has a well-known solution:

$$I_\nu = I_\nu^0 e^{-(\rho\kappa_\nu t)}$$

- This sort of "transmission experiment" is the typical way in which opacities are measured
- The quantity λ_ν^{mfp} = $(1/\rho\kappa_\nu)$ has the dimensions of length and is called the **optical mean free path**. The mean free path is a useful physical quantity and is defined as the average distance a photon can travel through a material without being absorbed or scattered. Optically thin plasmas have physical dimensions << λ_ν^{mfp}.

Computing an opacity from fundamental atomic cross sections

- Basically,

  opacity = (atomic population)(cross section)/(mass density)
  (NB: we are only interested in ***photo*** cross sections now)

- When interacting with electrons, a photon can be absorbed (most/all energy given to electrons) or scattered (some energy given to electrons, but photon survives with slightly decreased energy)

$$\kappa_\nu^{\mathrm{TOT}}(\rho,T_e,T_r) = \kappa_\nu^{\mathrm{ABS}}(\rho,T_e,T_r) + \kappa_\nu^{\mathrm{SCAT}}(\rho,T_e,T_r)$$

Compton scattering

$$\kappa_\nu^{\mathrm{ABS}} = \frac{1}{\rho}\sum_{\mathrm{il}} N_{\mathrm{il}}(\rho,T_e,T_r)[\sigma_{\mathrm{il}}^{(\mathrm{bound-bound})}(\nu) + \sigma_{\mathrm{il}}^{(\mathrm{bound-free})}(\nu)] + \kappa_\nu^{(\mathrm{free-free})}$$

material density

atomic level populations

photoexcitation cross sections

photoionization cross sections

inverse Bremsstrahlung contribution

How to compute an opacity

- Compton scattering uses a straightforward formula:

$$\kappa_\nu^{\mathrm{SCAT}} = N_e \sigma^{\mathrm{SCAT}}(\nu)/\rho \;\; [\approx 0.4\bar{Z}/A\ (\mathrm{cm}^2/\mathrm{g}) \text{ for Thomson scattering }]$$

- The free-free contribution is straightforward (Kramers' formula)
- The bound-bound and bound-free contributions are obtained by summing over ALL bound levels of ALL important ion stages
- This sum requires the populations, N_{il}, as well as the relevant photo cross sections, $\sigma_{il}^{\mathrm{photo}}$
- The previous opacity equations are valid for both LTE and NLTE conditions
- The LTE/NLTE difference ***is in how one calculates the atomic populations***, N_{il}

The LANL Suite of Atomic Modeling Codes

Atomic Physics Codes → Atomic Models → ATOMIC

CATS: Cowan Code

RATS: relativistic

ACE: e^- excitation

GIPPER: ionization

http://aphysics2.lanl.gov/tempweb

fine-structure
config-average
UTAs
MUTAs
energy levels
gf-values
e^- excitation
e^- ionization
photoionization
autoionization

LTE or NLTE
atomic level
populations

spectral modeling
emission
absorption
transmission
power loss

LTE OPLIB tables
↓
TOPS

To calculate LTE opacities, you need only:

Atomic Physics Codes → Atomic Models → ATOMIC

CATS: Cowan Code

RATS: relativistic

ACE: e^- excitation

GIPPER: ionization

http://aphysics2.lanl.gov/tempweb

fine-structure
config-average
UTAs
MUTAs
energy levels
gf-values
e^- excitation
e^- ionization
photoionization
autoionization

LTE or NLTE
atomic level
populations

spectral modeling
emission
absorption
transmission
power loss

LTE OPLIB tables → TOPS

What about emissivities?

- Simple relationship for LTE conditions:

emissivity Planck function

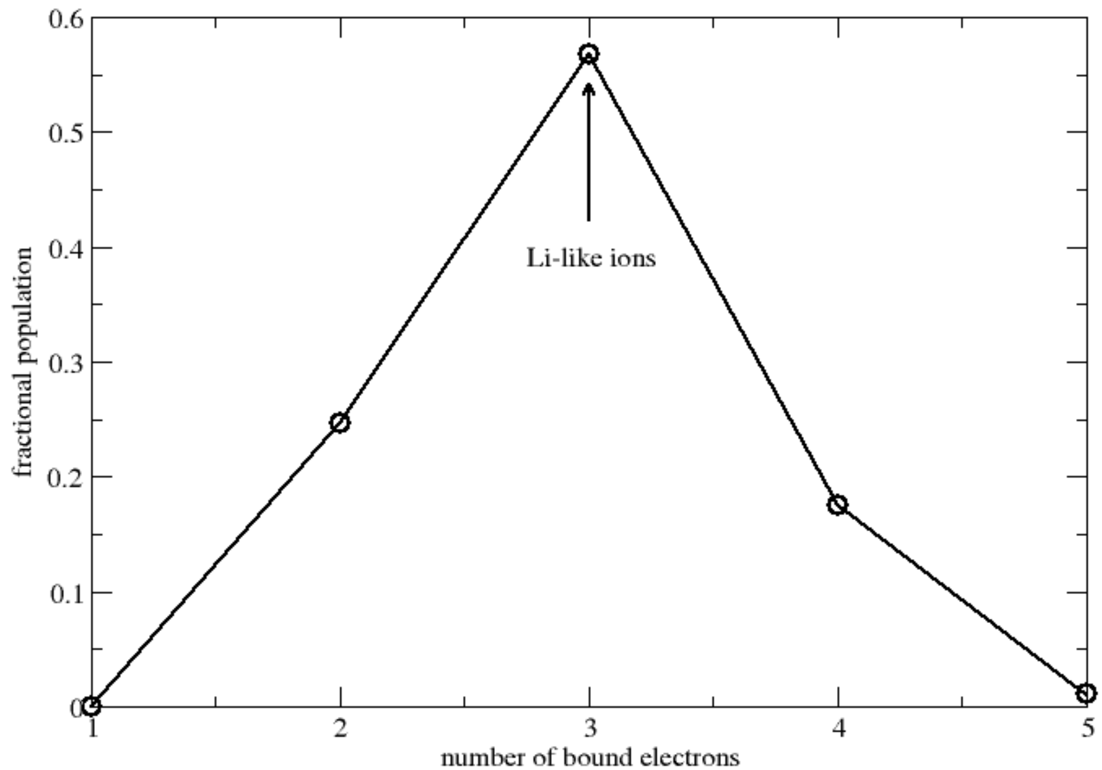
$$\varepsilon_\nu = (4\pi)\kappa_\nu^{\text{ABS}}(\rho, T)B_\nu(T)$$

opacity

- One only needs the opacity to obtain the emissivity when doing LTE calculations
- Non-LTE emissivities require the level populations, N_{il}, along with the cross sections for the ***inverse*** of the photo-absorption processes that were considered for opacities

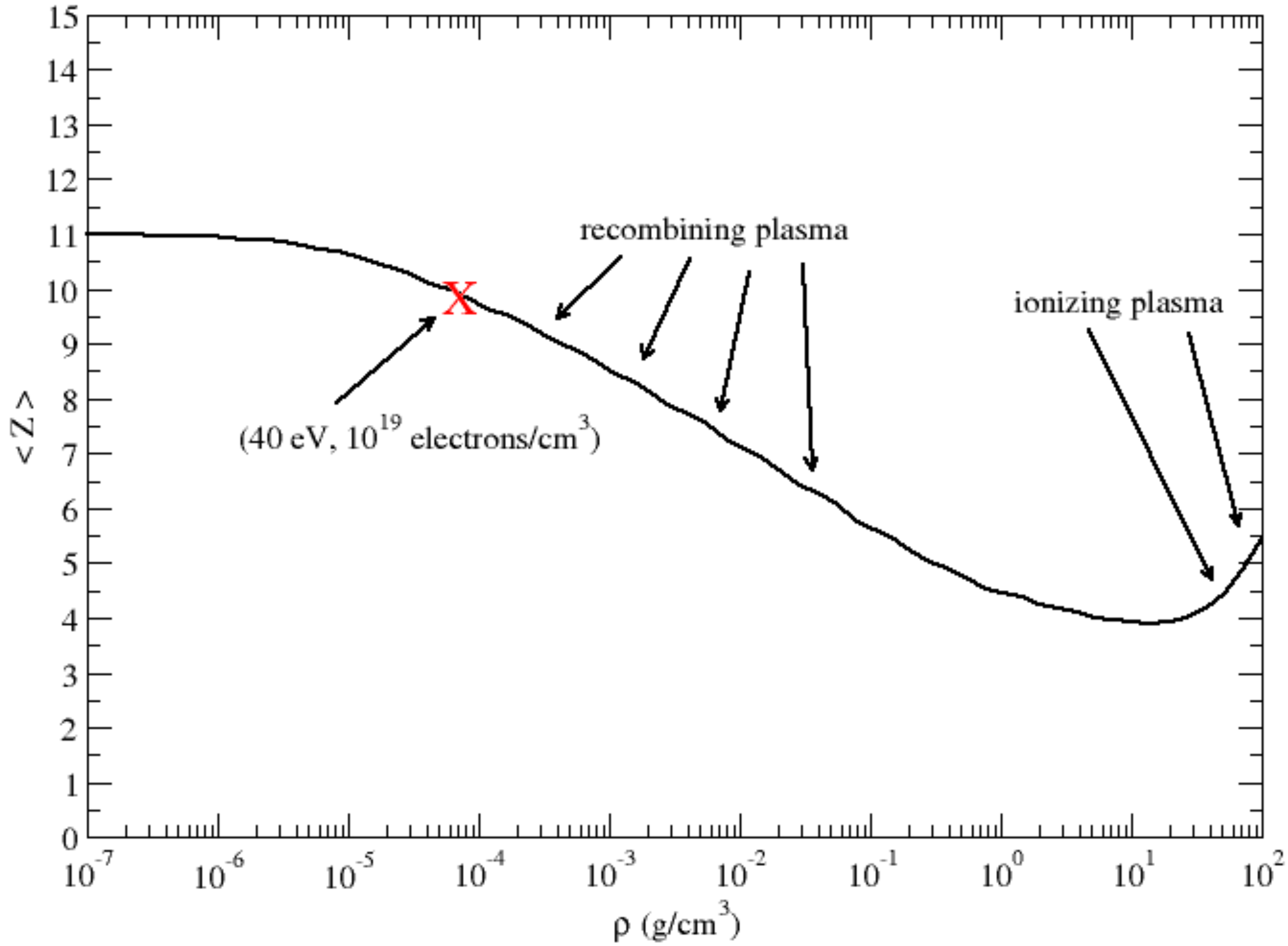
Numerical example of an LTE opacity: Aluminum plasma at kT = 40 eV, $N_e = 10^{19} cm^{-3}$

- For these conditions, <Z>=10.05 $\Rightarrow$ there is an average of ~2.95 bound electrons/ion (Li-like ions are dominant)
- Here is the charge state distribution:



Another useful plot to consider: <Z> vs. ρ

- Here is a plot of <Z> vs mass density for a fixed temperature of 40 eV:



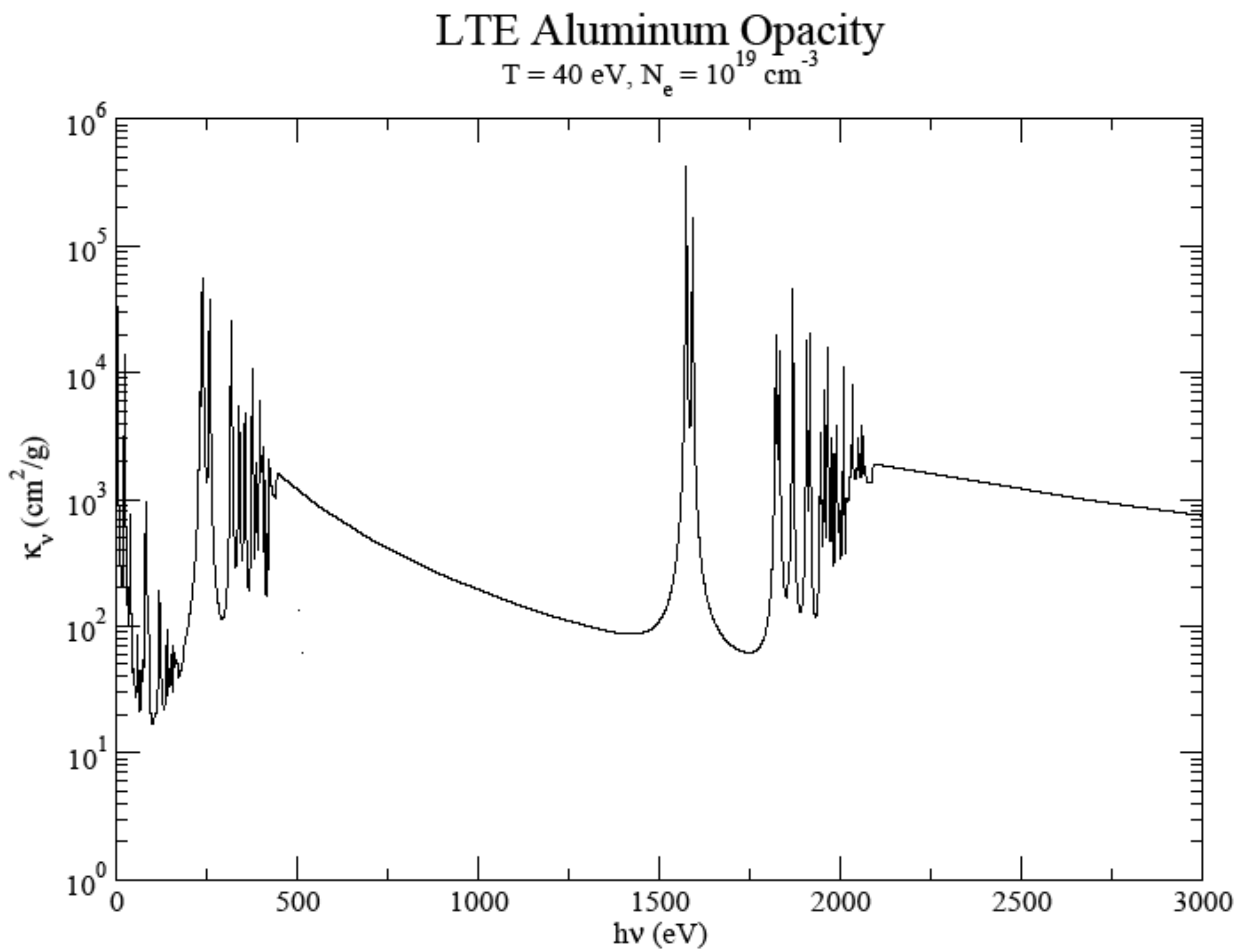
Numerical example of an LTE opacity: Aluminum plasma at $kT = 40$ eV, $N_e = 10^{19}\text{cm}^{-3}$

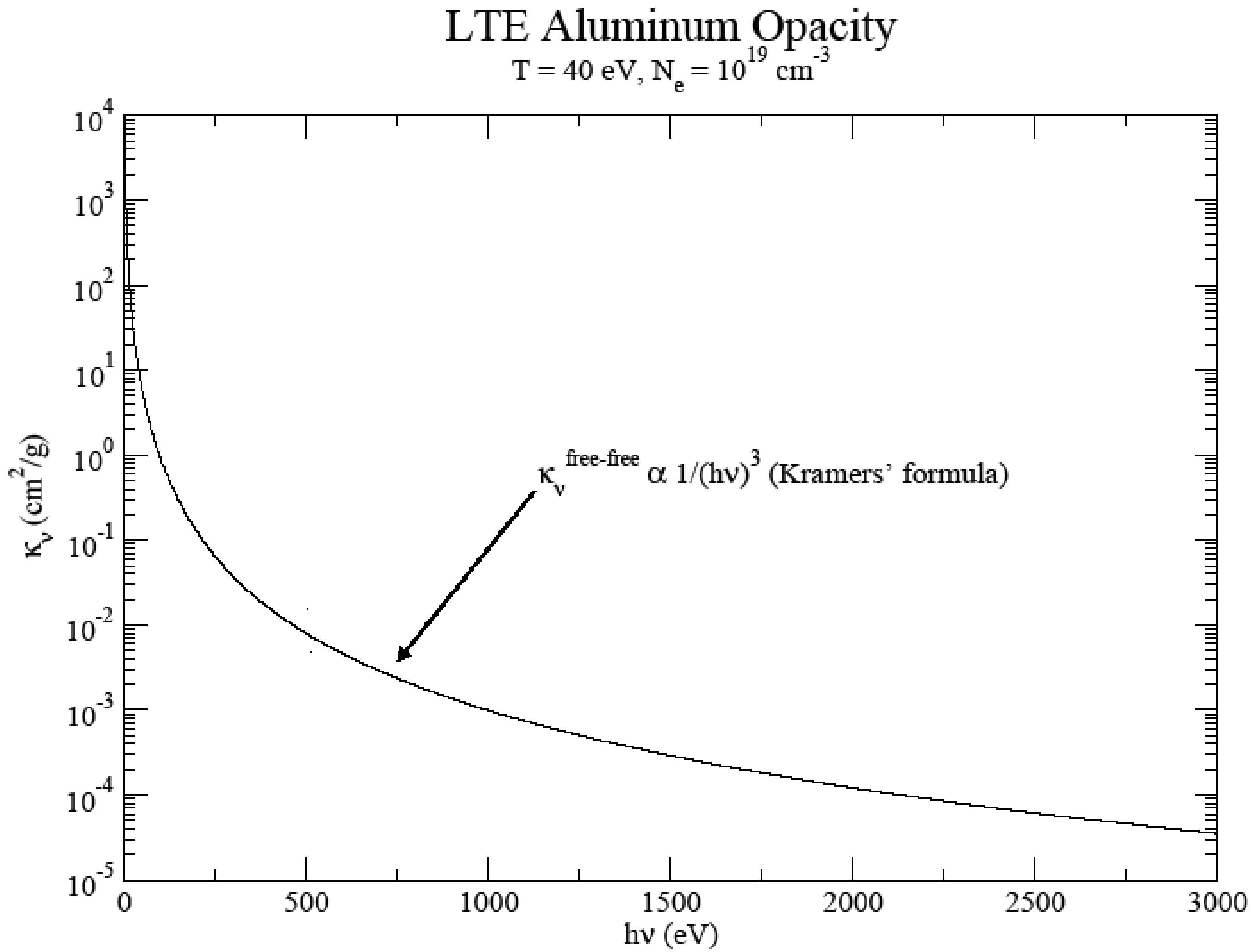
- For these conditions, $\langle Z \rangle = 10.05 \Rightarrow$ there is an average of ~2.95 bound electrons/ion (Li-like ions are dominant)
- The following plots show the contribution to the total opacity from each of the three photo-absorption processes as well as the contribution from Compton scattering

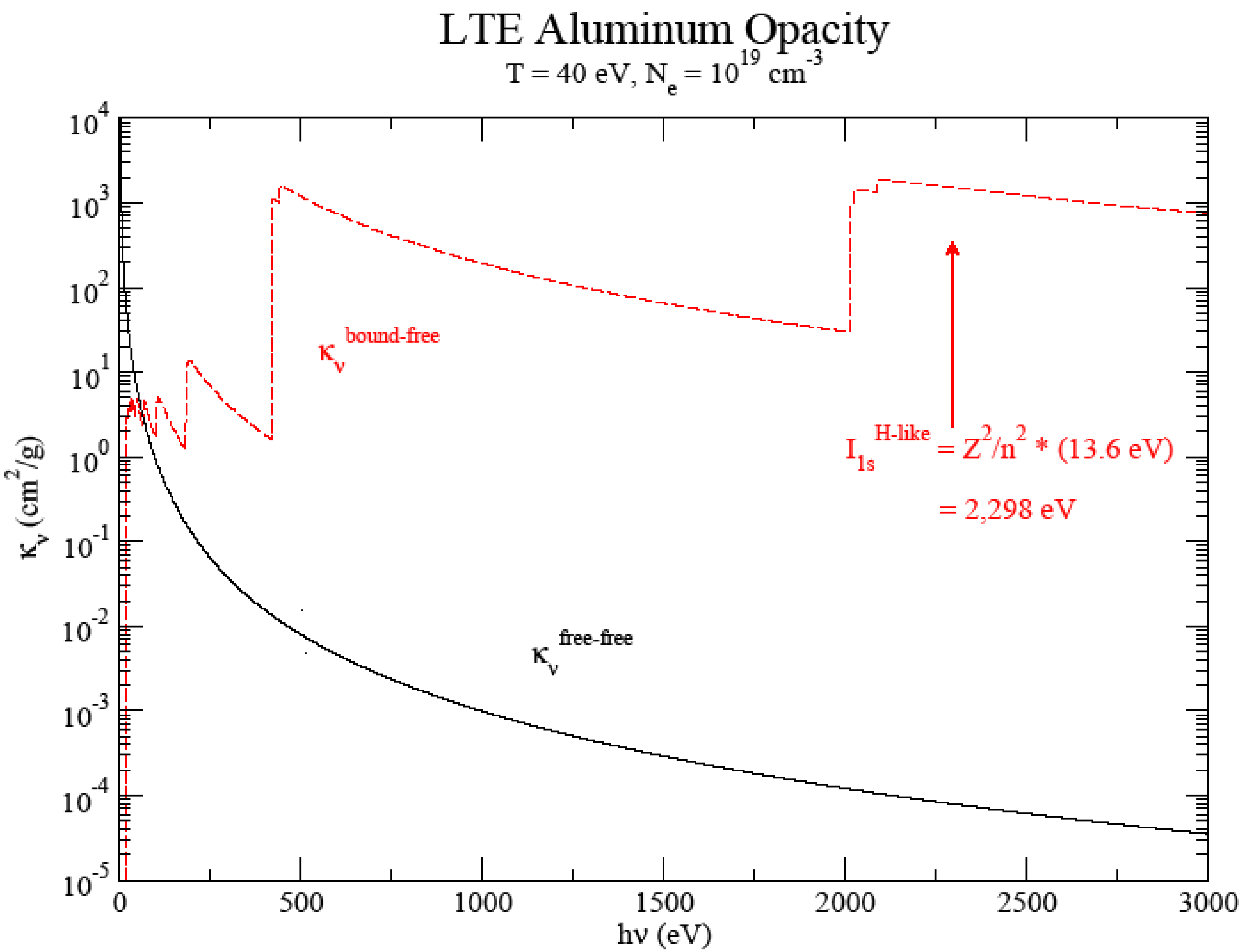
---

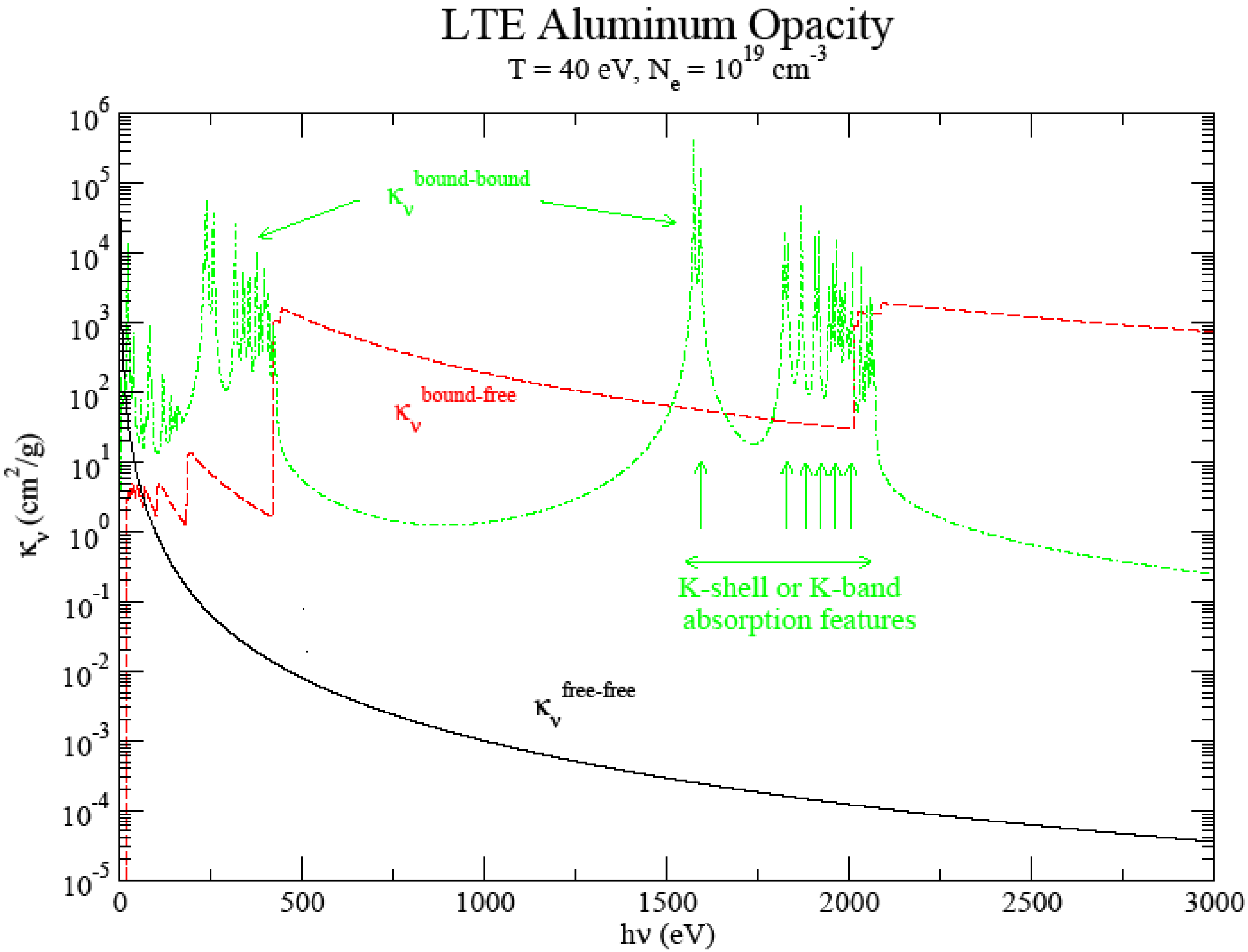
- You will see some arcane spectroscopic notation: bound electrons with the same principal quantum number n are said to inhabit the same "shell". Each shell is identified by a capital letter: n=1, K-shell n=2, L-shell   n=3, M-shell ….
- Bound-bound absorption involving an active bound electron that initiates from the K-shell is referred to as "K-shell" absorption, etc. Bound-bound emission that terminates with a bound electron ending up in the K-shell is referred to as "K-band" emission, etc.

First, a snapshot of the total LTE opacity for this aluminum plasma

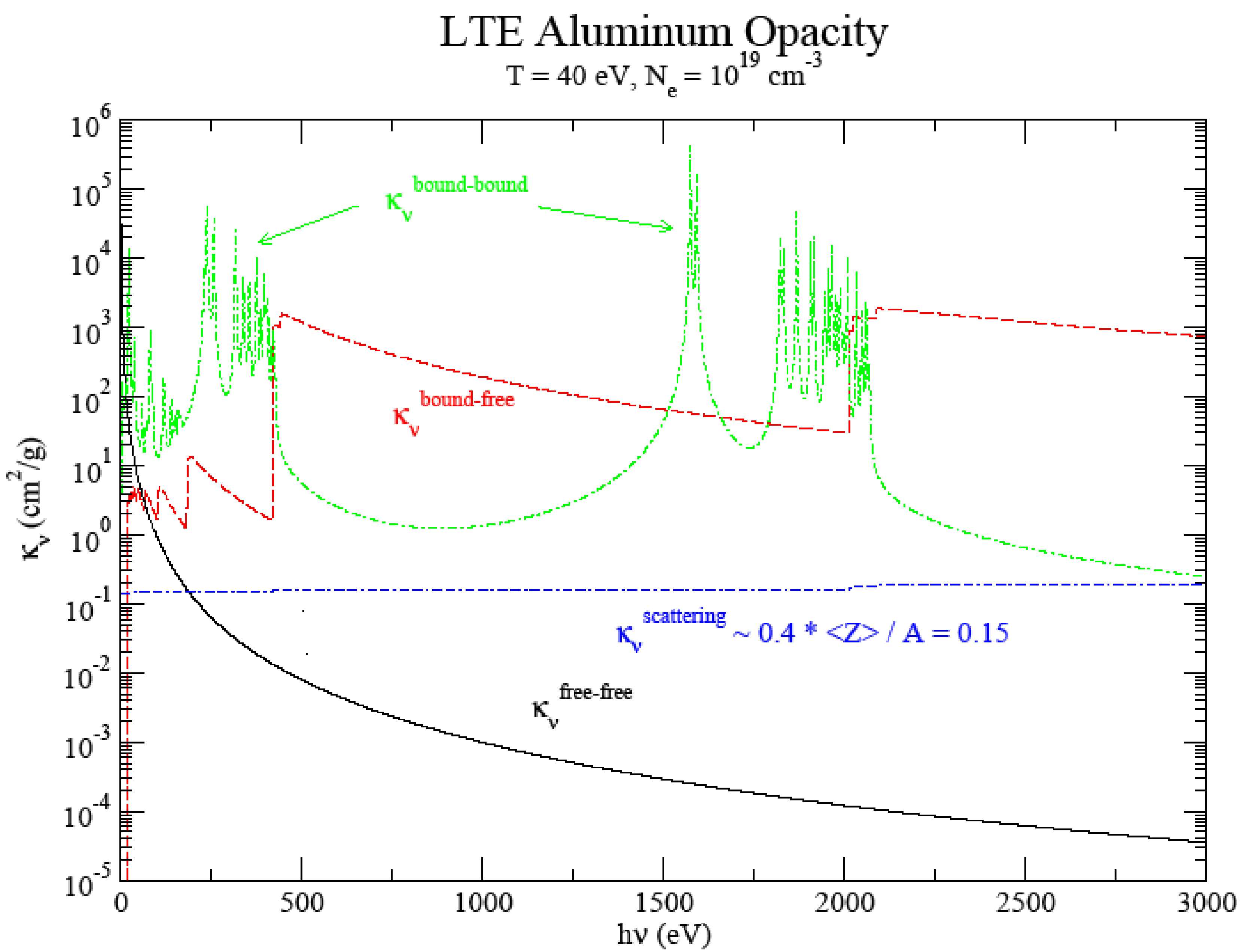



LTE Aluminum Opacity
T = 40 eV, N_e = 10^19 cm^-3
κ_ν (cm^2/g)
hν (eV)
κ_ν^free-free α 1/(hν)^3 (Kramers' formula)
10^4
10^3
10^2
10^1
10^0
10^-1
10^-2
10^-3
10^-4
10^-5
0
500
1000
1500
2000
2500
3000

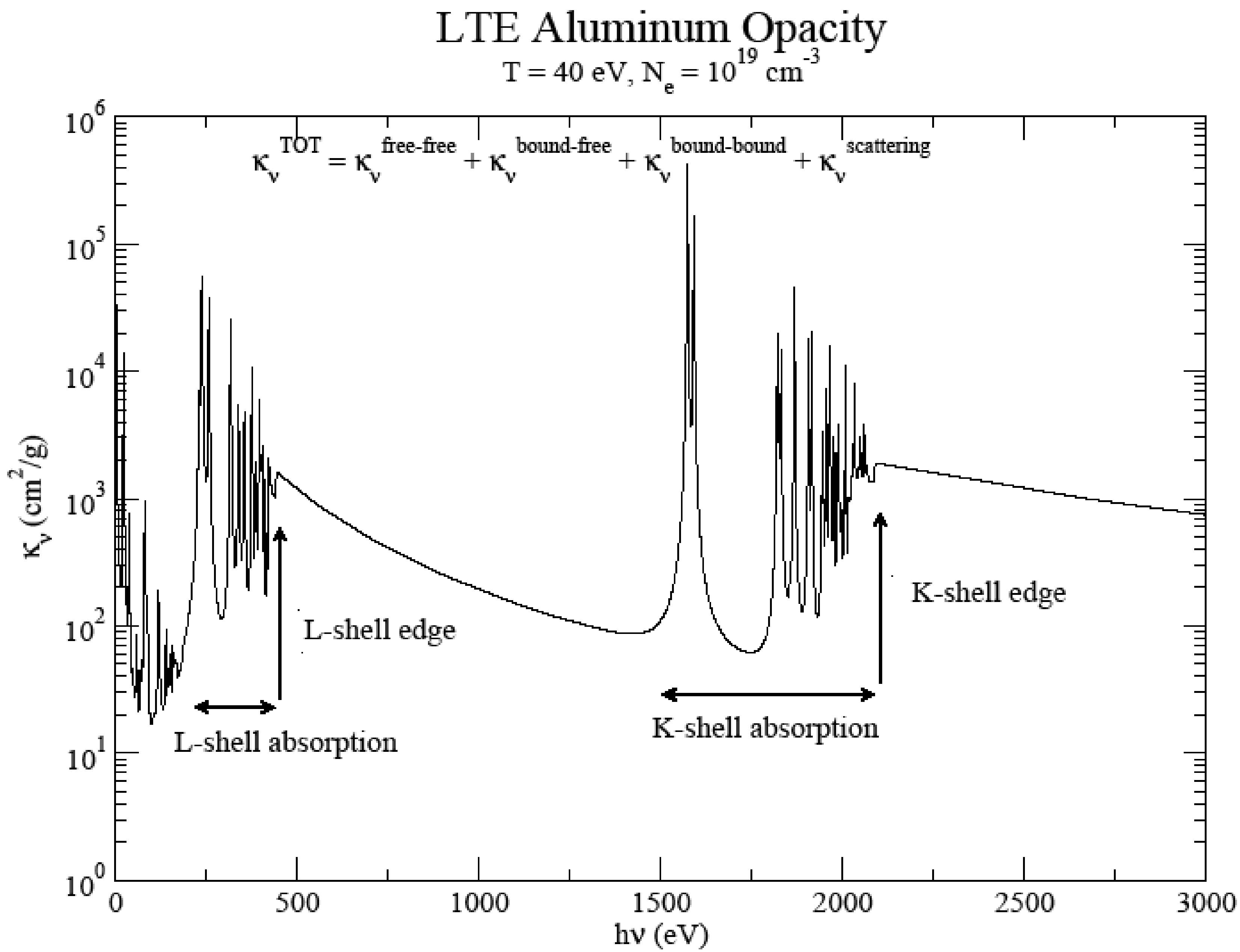

LTE Aluminum Opacity
T = 40 eV, N_e = 10^19 cm^-3
κ_ν (cm^2/g)
hν (eV)
κ_ν^bound-free
κ_ν^free-free
I_1s^H-like = Z^2/n^2 * (13.6 eV)
= 2,298 eV
10^4
10^3
10^2
10^1
10^0
10^-1
10^-2
10^-3
10^-4
10^-5
0
500
1000
1500
2000
2500
3000



LTE Aluminum Opacity
T = 40 eV, $N_e = 10^{19}$ cm^{-3}
$\kappa_\nu^{\text{bound-bound}}$
$\kappa_\nu^{\text{bound-free}}$
$\kappa_\nu^{\text{free-free}}$
K-shell or K-band absorption features
κ_ν (cm^2/g)
hν (eV)



LTE Aluminum Opacity
T = 40 eV, N_e = 10^19 cm^-3
κ_ν bound-bound
κ_ν bound-free
κ_ν scattering ~ 0.4 * <Z> / A = 0.15
κ_ν free-free
κ_ν (cm^2/g)
hν (eV)
10^6
10^5
10^4
10^3
10^2
10^1
10^0
10^-1
10^-2
10^-3
10^-4
10^-5
0
500
1000
1500
2000
2500
3000


LTE Aluminum Opacity
T = 40 eV, N$_e$ = 10^{19} cm^{-3}
$\kappa_\nu^{TOT} = \kappa_\nu^{free-free} + \kappa_\nu^{bound-free} + \kappa_\nu^{bound-bound} + \kappa_\nu^{scattering}$
L-shell edge
K-shell edge
L-shell absorption
K-shell absorption
κ_ν (cm^2/g)
hν (eV)
10^0
10^1
10^2
10^3
10^4
10^5
10^6
0
500
1000
1500
2000
2500
3000