**2327-4**

## Joint ICTP-IAEA Workshop on Fusion Plasma Modelling using Atomic and Molecular Data

*23 - 27 January 2012*

# Atomic Processes Modeling in Plasmas
# Modeling Spectroscopic Observables from Plasmas

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# Atomic processes modeling in plasmas

## modeling spectroscopic observables from plasmas

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Joint ICTP-IAEA Workshop on Fusion Plasma Modelling Using Atomic and Molecular Data
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Modeling atoms and photons present in plasmas

# THEORETICAL PLASMA SPECTROSCOPY

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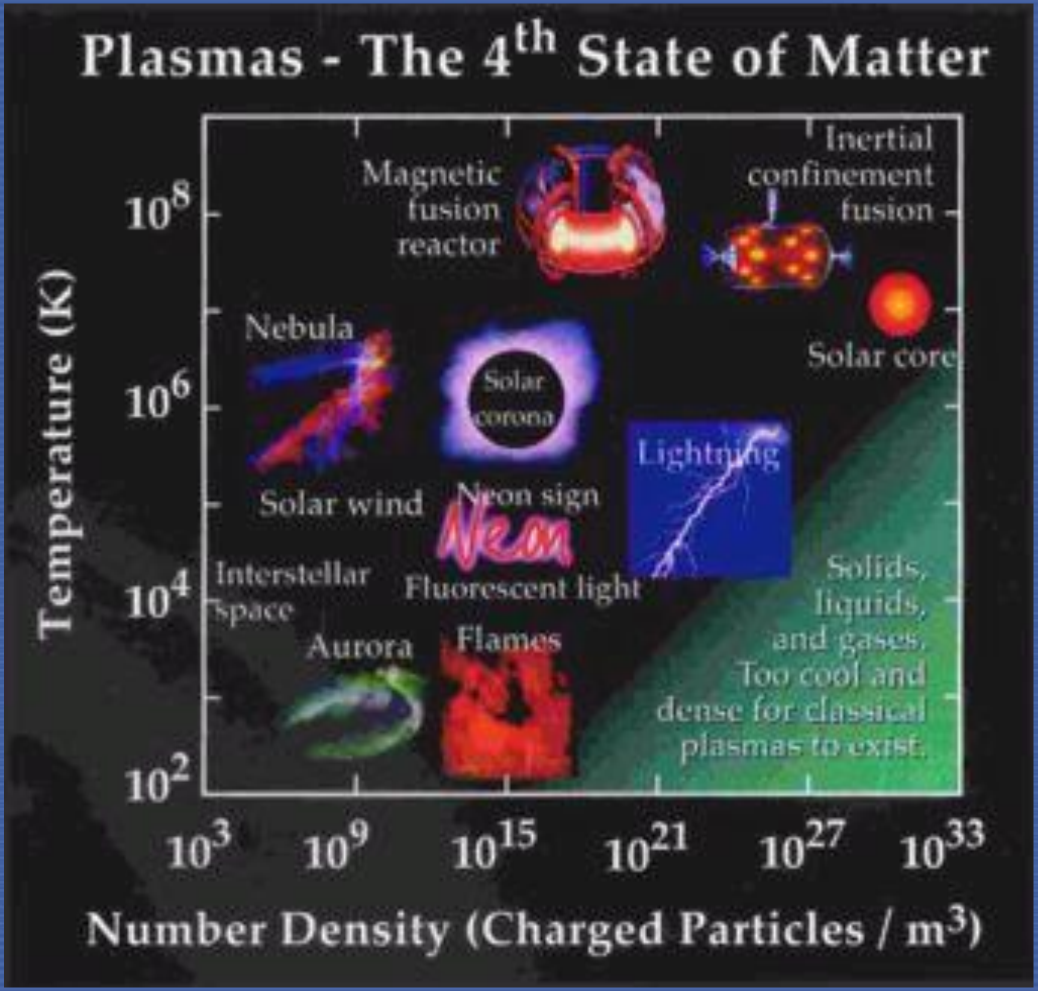
# Plasmas occur over a vast range of conditions



Temperature

$10^{-6}$K – 100 keV ($10^9$K)

Density

$10^5 – 10^{23}$ $cm^{-3}$

Astrophysical observations

- CHANDRA/XMM

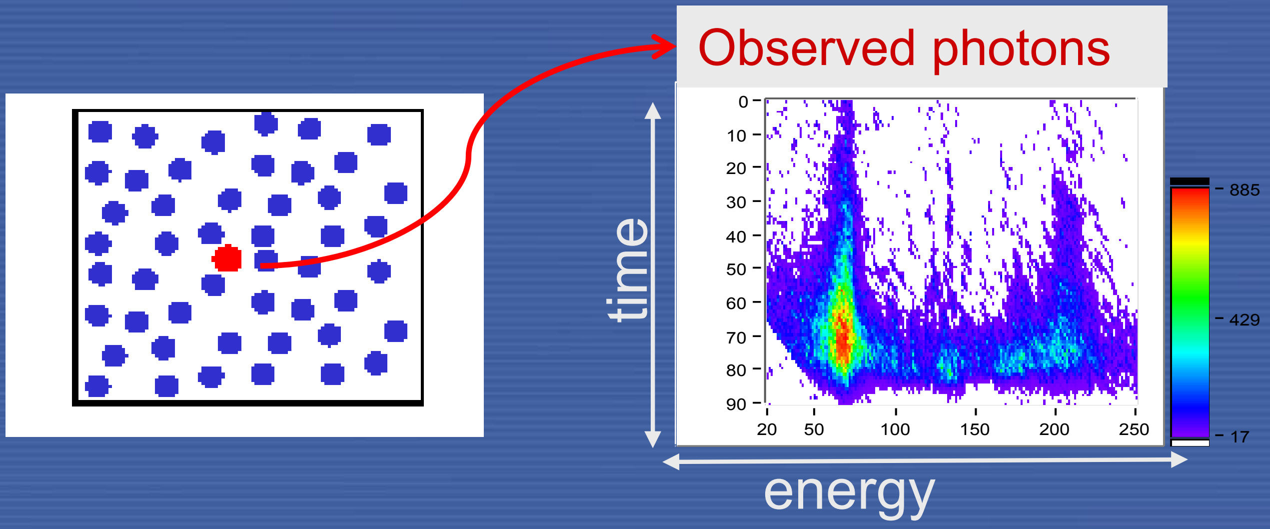
Plasma processing

Fusion research

- ICF (Inertial confinement fusion)
  - Laser-produced plasma
  - Beam-produced plasma
  - Z-pinch plasmas
- MCF (Magnetic confinement fusion)
  - Tokamak plasma

Ultracold plasmas

# Spectroscopic Observables:
# Photons carry information of plasma



Theoretical Plasma spectroscopy

Atoms respond to perturbations of the ensemble of surrounding particles and/or external fields

- Electronic transitions
- Photon-atom interactions
- Atomic level shifts and broadens

The microscopic response of the atom carried by photons provides information on the macroscopic environment

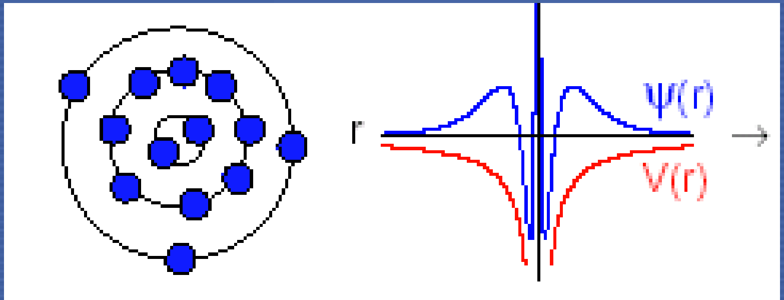
- $T_e$ and $n_e$
- Electromagnetic fields
- Plasma wave modes

One can obtain plasma thermodynamic properties

One can understand plasma collective behavior

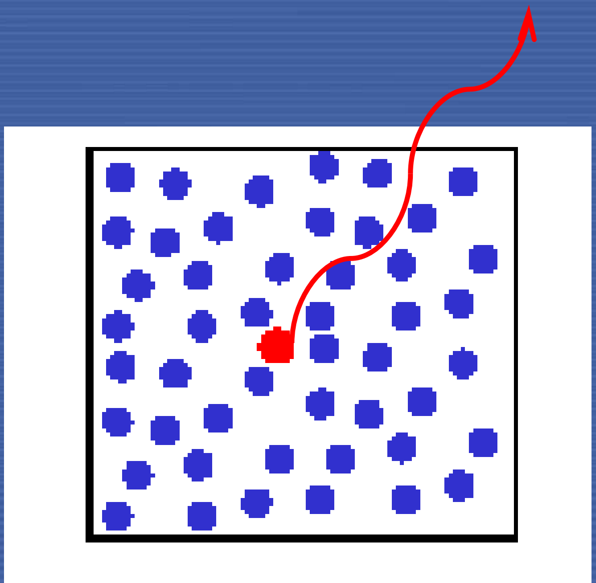
One can verify macroscopic fluid descriptions, e.g., hydrodynamics and particle theories, e.g., kinetic theory

# Theoretical plasma spectroscopy is a highly interdisciplinary field



Quantum physics

- Atomic physics
- Collision physics
- Line shape theory

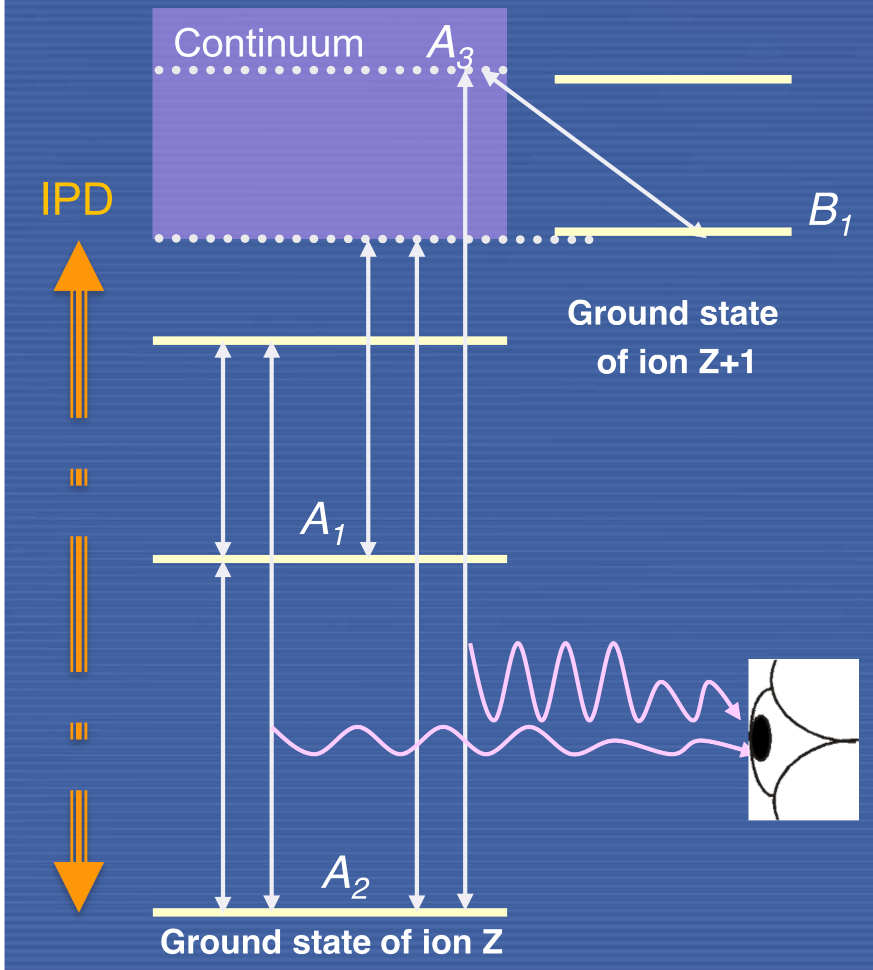


Classical physics

- Radiation transport
- Hydrodynamics
- Plasma kinetic theory

# 1) Atomic Data required

**Energy levels of an atom**



BOUND-BOUND TRANSITIONS

$A_1 \rightarrow A_2 + h\nu_2$ Spontaneous emission

$A_1 + h\nu_1 \leftrightarrow A_2 + h\nu_1 + h\nu_2$ Photo-absorption or emission

$A_1 + e_1 \leftrightarrow A_2 + e_2$ Collisional excitation or deexcitation

BOUND-FREE TRANSITIONS

$B_1 + e \rightarrow A_2 + h\nu_3$ Radiative recombination

$B_1 + e \leftrightarrow A_2 + h\nu_3$ Photoionization / stimulated recombination

$B_1 + e_1 \leftrightarrow A_2 + e_2$ Collisional ionization / recombination

$B_1 + e_1 \leftrightarrow A_3 \leftrightarrow A_2 + h\nu_3$ Dielectronic recombination

(autoionization + electron capture)

# 2) Population Kinetics modeling required

$$\frac{dn_i}{dt} = -n_i \sum_{j \neq i}^{N\max} W_{ij} + \sum_{j \neq i}^{N\max} n_j W_{ji}$$

$$W_{ij} = B_{ij} \overline{J_{ij}} + n_e C_{ij} + \beta_{ij} + n_e \gamma_{ij}$$

$$W_{ji} = A_{ij} + B_{ji} \overline{J_{ji}} + n_e D_{ji} + n_e(\alpha_{ji}^{RR} + \alpha_{ji}^{DR}) + n_e^2 \delta_{ij}$$

| | |
|---|---|
| $A_{ij}$ | Spontaneous emission |
| $B_{ij}$ | Stimulated absorption or emission (I > j) |
| $C_{ij}$ | Collisional excitation |
| $D_{ij}$ | Collisional deexcitation |
| $\alpha_{ij}^{DR}$ | Dielectronic recombination |
| $\alpha_{ij}^{RR}$ | Radiative recombination |
| $\beta_{ij}^{RR}$ | Photoionization+stimulated recombination |
| $\gamma_{ij}$ | Collisional ionization |
| $\delta_{ij}$ | Collisional recombination |

The key is to figure out how to manage the infinite set of levels and transitions of atoms and ions into a model with a tractable set of levels and transitions that represents a physical reality!

# 3) Radiation transport required

- Radiation intensity *I(r,n,ν,t)* is determined self-consistently from the coupled integro-differential radiation transport and population kinetic equations

$$[c^{-1}(\partial/\partial t)+(\mathrm{n}\cdot\nabla)]I(\mathrm{r},\mathrm{n},\nu,t)=\eta(\mathrm{r},\mathrm{n},\nu,t)-\chi(\mathrm{r},\mathrm{n},\nu,t)\,I(\mathrm{r},\mathrm{n},\nu,t)$$

- Opacity *χ(**r**,**n**,ν,t)* and emissivity *η(**r**,**n**,ν,t)* are obtained with population densities and radiative transition probabilities

$$\chi_\nu=\sum_i\sum_{j>i}[n_i-(g_i/g_j)n_j]\alpha_{ij}(\nu)+\sum_i(n_i-n_i^{*}e^{-h\nu/kT})\alpha_{i\kappa}(\nu)+\sum_\kappa n_e n_\kappa\alpha_{\kappa\kappa}(\nu,T)(1-e^{-h\nu/kT})$$

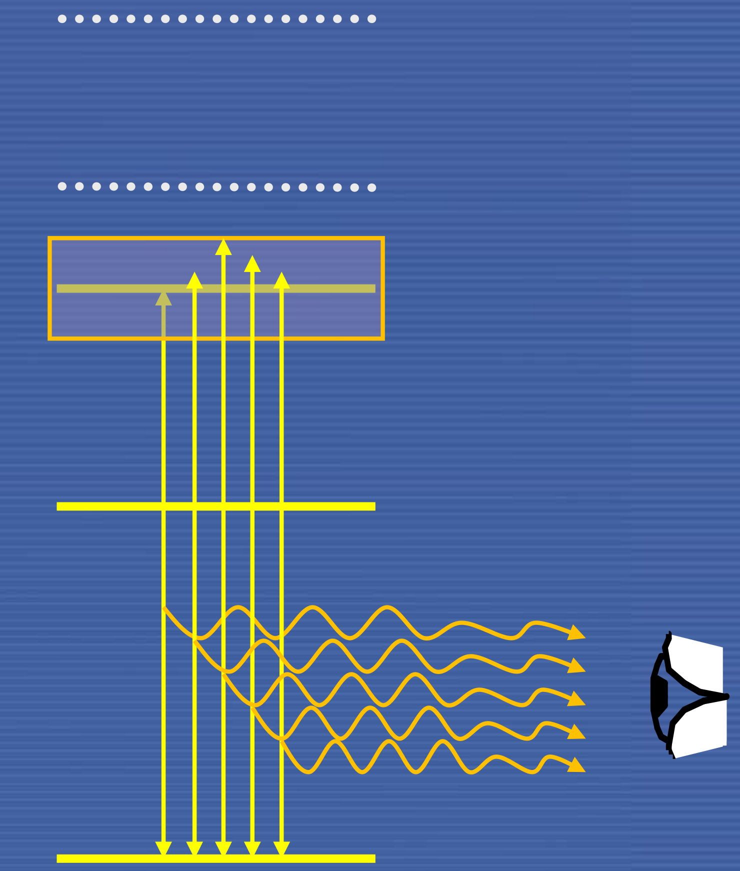
$$\eta_\nu=\left(2h\nu^3/c^2\right)\left[\sum_i\sum_{j>i}(g_i/g_j)n_j\alpha_{ij}(\nu)+\sum_i n_i^{*}e^{-h\nu/kT}\alpha_{i\kappa}(\nu)+\sum_\kappa n_e n_\kappa\alpha_{\kappa\kappa}(\nu,T)e^{-h\nu/kT}\right]$$

Radiation field carries the information on atoms embedded in plasmas through population distributions

# 4) Line shape models required

- Line shape theory is a theoretically rich field incorporating quantum-mechanics and statistical mechanics

- Line shapes have provided successful diagnostics for a vast range of plasma conditions
  - Natural broadening (intrinsic)
  - Doppler broadening ($T_i$)
  - Stark broadening ($N_e$)
  - Opacity broadening
  - Resonance broadening (neutrals)



Ground state of ion Z

# 5) Hydrodynamics required

- Time scales are very different between atomic processes and classical particle motions : separation between QM processes and particle mechanics

→ Is this a valid assumption?

- Hydrodynamics simulations
  - Fluid treatment of plasma physics
    - Mass, momentum and energy equations solved
  - Plasma thermodynamic properties
  - LTE (Local Thermodynamic Equilibrium) (assumed)

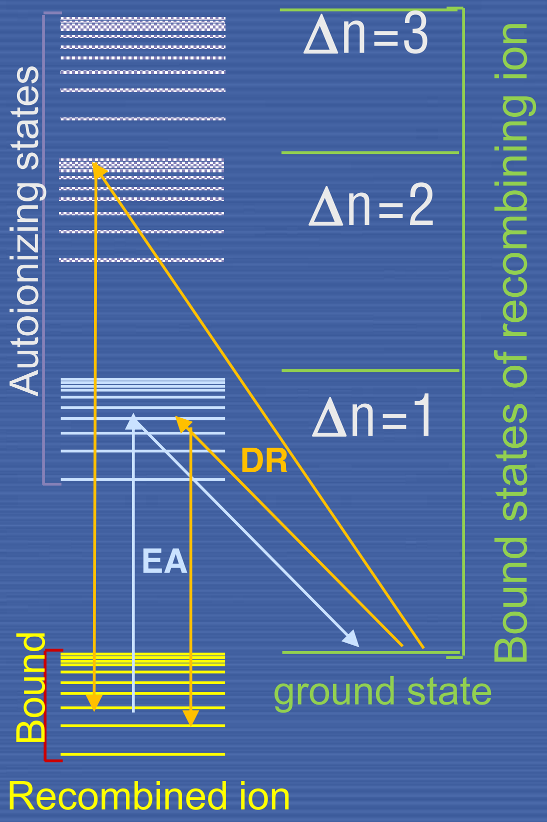
- PIC (Particle-In-Cell) simulations
  - Particle treatment of plasma physics
    - Boltzmann transport and Maxwell equations solved
  - Electron energy distribution function
  - Simple ionization model (assumed)

Level population distribution determined by rate equations for non-local thermodynamic equilibrium (NLTE) state plasmas

# COLLISIONAL-RADIATIVE POPULATION KINETIC MODELS

# Population Kinetics Models

Mean ionization states, Charge state distributions, Spectral intensity, Emissivity, Opacity, Equation of state, Electrical conductivity require population distributions of ions in the plasma.



*Coronal plasmas (low $N_e$) ⇒ Rate formalism*

Charage state distributions are determined by rates of *collisional ionization and excitation-autoionization (EA)* & *radiative recombination and dielectronic recombination (DR)* *originating from the ground states*

*LTE plasmas (high $N_e$) ⇒ Statistical distributions*

Collisional processes are dominant and the population distribution is governed by *Boltzmann relations and Saha equation*.

.

*Collisional-radiative plasmas (intermediate $N_e$) ⇒ Rate equation model*

A population distribution is determined by *rate equations considering collisional and radiative processes* at a given temperature and density. The results should converge to coronal or the LTE limit at low and high $N_e$ limits, respectively.

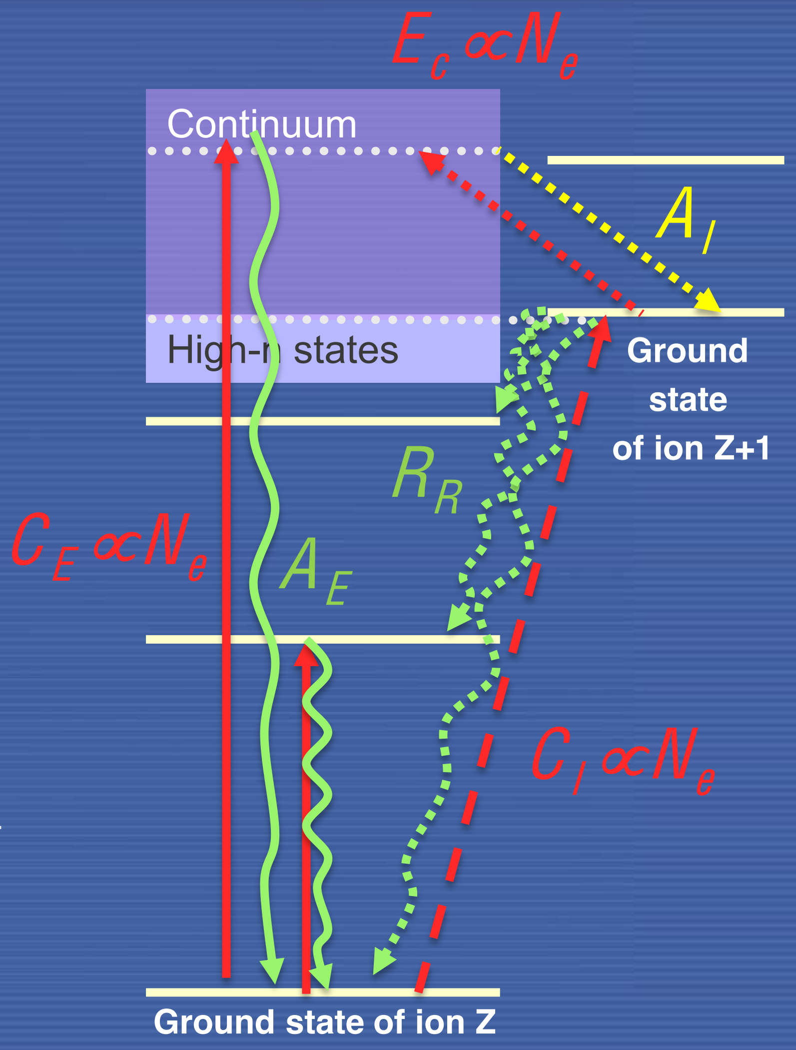
# Coronal Limits (low densities)

- At low densities, excited state population densities are so low that they are assumed to be populated from the ground state by collisional excitation **$C_E$** and depopulated by spontaneous emission **$A_E$**. Therefore the excited population density is *proportional to $N_e$*
- Charge state distribution is *independent of $N_e$*

$$n_Z(C_I + E_A) = n_{Z+1}(R_R + D_R)$$

$$E_A = \sum_i^{\infty} C_E^i B_{Z+1}^{i,a} \quad B_{Z+1}^{i,a} = \frac{\sum_{k>Z} A_E^{ik} B_{Z+1}^{i,a} + \sum_{l\in Z+1} A_I^{il}}{\sum_{j\in Z}^{\infty} A_E^{ij} + \sum_{k>Z} A_E^{ik} + \sum_{l\in Z+1} A_I^{il}}$$

$$D_R = \sum_i^{\infty} E_C^i B_Z^{i,d} \quad B_Z^{i,d} = \frac{\sum_{j\in Z}^{\infty} A_E^{ij} + \sum_{k>Z} A_E^{ik} B_Z^{i,d}}{\sum_{j\in Z}^{\infty} A_E^{ij} + \sum_{k>Z} A_E^{ik} + \sum_{l\in Z+1} A_I^{il}}$$



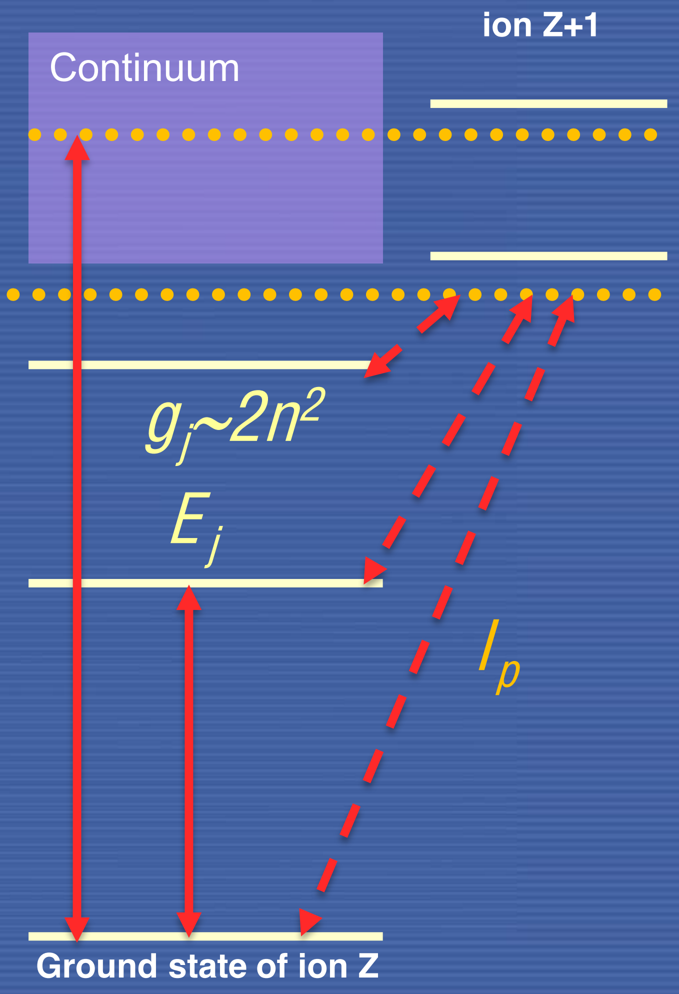
Coronal Model

# LTE Limits (high densities)

- Population/depopulation processes are totally dominated by collisions and therefore rates should be in detailed balance.
- The 3-body recombination process dominates and hence the high-lying states are important for charge state balance.
- A population distribution is an explicit function of "local" plasma conditions, governed by Boltzmann statistics and Saha equations.

$$n_j = \frac{N_i}{Z_i(T)} g_j e^{-E_j/kT} \qquad Z_i(T) = \sum_{j\in i}^{j_{max}} g_j e^{-E_j/kT}$$

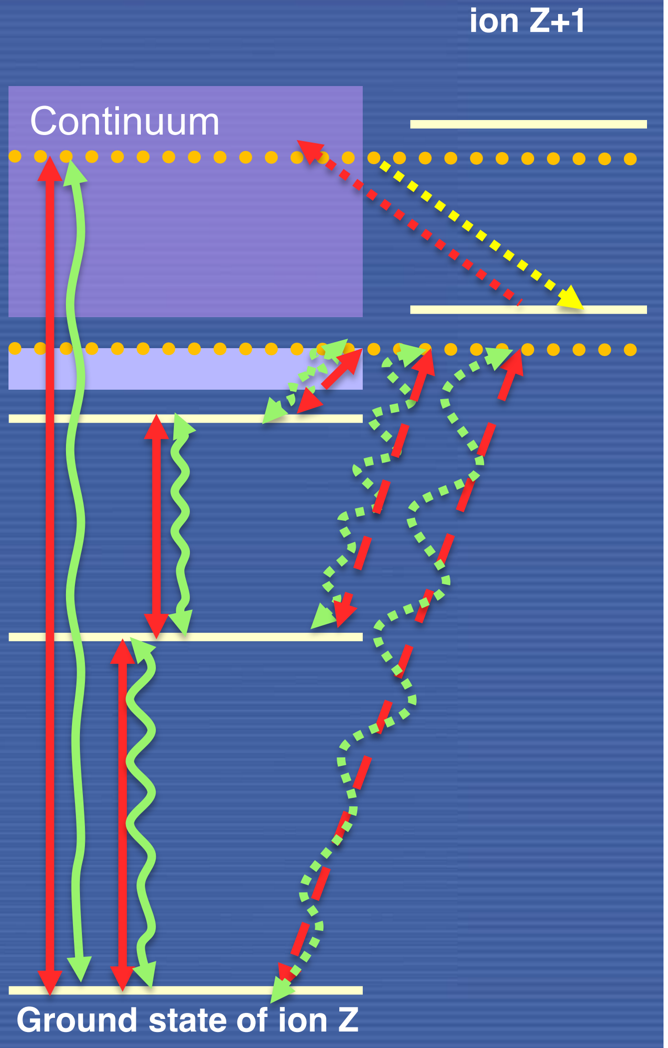
$$\frac{N_e n_{i+1}}{n_i} = \frac{2(2\pi m k_B T)^{3/2}}{h^3} \frac{Z_{i+1}(T)}{Z_i(T)} e^{-I_p/kT}$$



**LTE Model**

# Collisional-Radiative Regime

- Population distribution is obtained by rate equations considering collisional and radiative processes, along with plasma effects
- *Excited states* are substantially populated and *increase the total ionization* by step-wise ionization processes while the 3-body recombination to these states is proportional to $n^3$ and hence they significantly enhance the total recombination.
- Plasma effects such as non-local radiation transport, fast particle collisions and density effects should be included in the model.
- For optically thick lines, the self-absorption should be included to reduce the radiative processes.

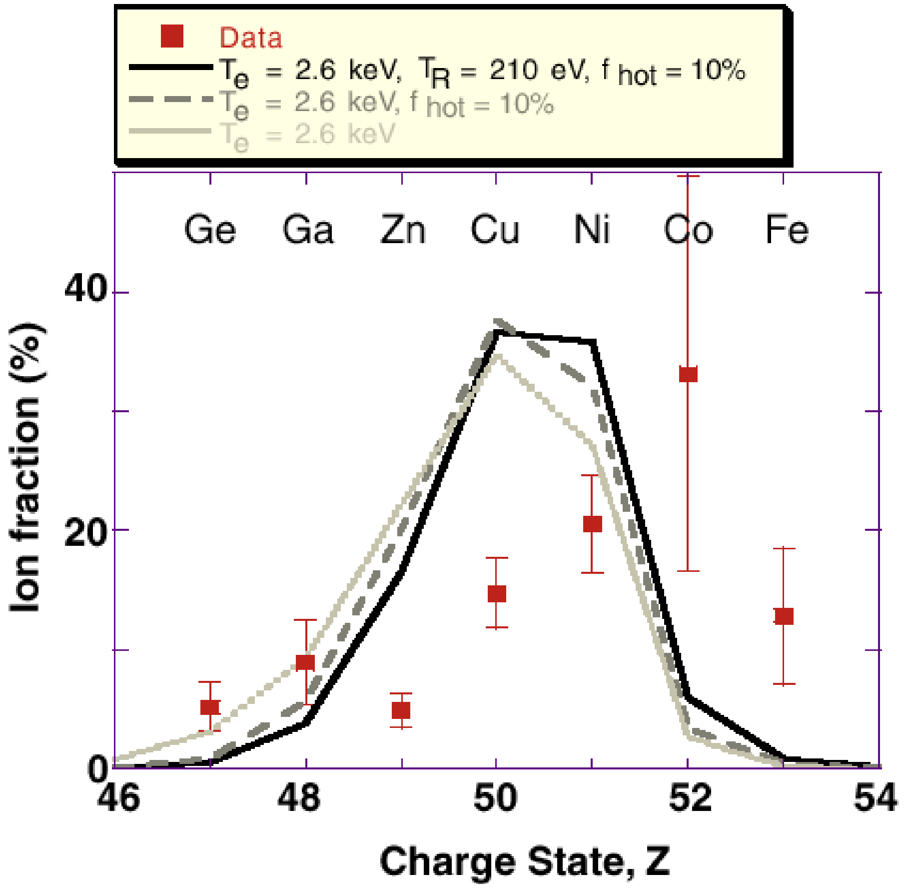


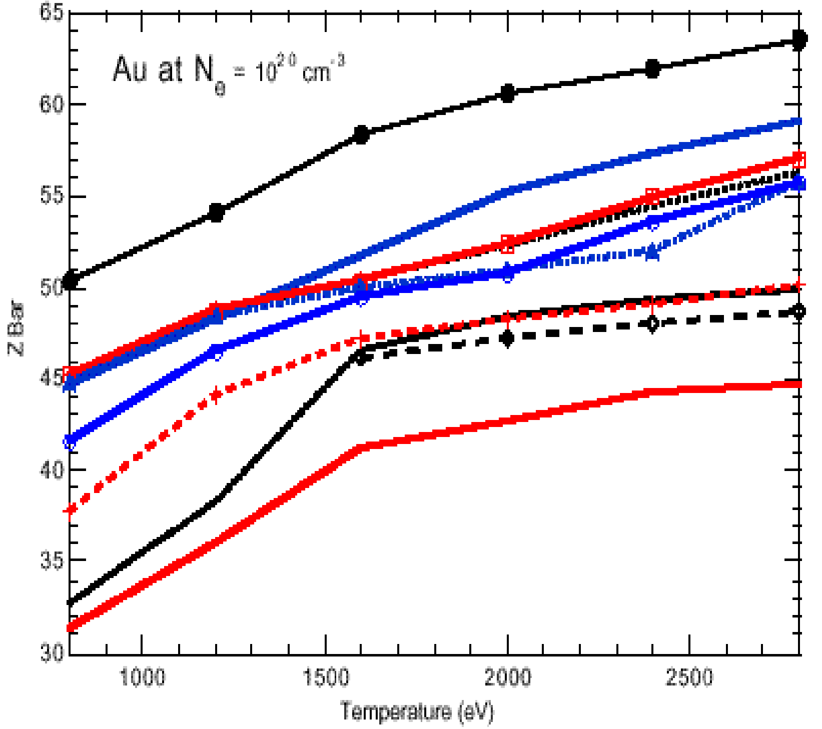
Collisional-Radiative Model

# Non-LTE plasmas have well documented problems for experiment and theory

Au M-shell emission
Glenzer et al. PRL (2001)

$1^{st}$ Non-LTE workshop (1996) documented large differences between codes for Au





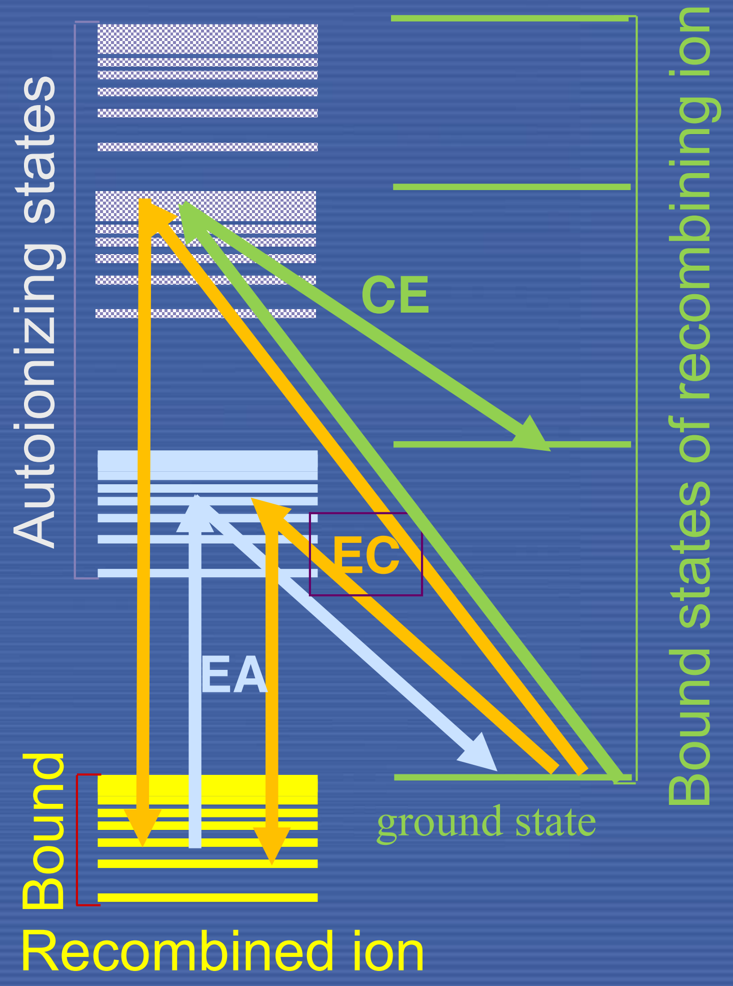
- Note the wide range of predicted average ionization state

DR and EA Processes

# LOW DENSITY LIMITS

# Processes of dielectronic recombination (DR) and excitation autoionization (EA)

- DR is a two-step process where an electron is captured leaving the recombined ion at **autoionizing (Ai)** states.
- Subsequently, the states stabilize either to the bound states making the recombination process complete or back to the ion by Auger process
- The **electron capture (EC)** can enhance **resonant collisional excitation** (CE) when the state autoionizes to the excited state of the recombining ion
- **EA** process is a reverse process to DR and an electron is excited to Ai states which subsequently ionize by Auger processes



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# Detailed atomic data are obtained using the **HULLAC** code

- Used to compute energy levels, oscillator strengths, collisional excitation and ionization cross-sections, photoionization cross-sections and autoionization rates
  - For atomic structure, Dirac equations are solved for multi-configuration, intermediate-coupling energy eigenvalues for fine structure levels using a parametric potential
  - Autoionization rates and radiative rates are computed using the multi-configuration wave functions from Dirac equations
  - Collisional cross-sections are computed in the distorted wave approximation

# Many levels and configurations are required for DR/EA modeling

| Iso | HULLAC | model | DR channels |
|---|---|---|---|
| H-like | 58 | 58 | NA |
| He-like | 636 | 320 | 2lnl’ 3lnl’ |
| Li-like | 1676 | 454 | 1s2lnl’ 1s3lnl’ 1s4lnl’ |
| Be-like | 2425 | 837 | 3lnl’ 1s2l$^{2}$nl’ |
| B-like | 4710 | 1510 | 2l3l’nl” 1s2s$^{2}$2pnl’ 1s2p$^{3}$nl’ |
| C-like | 8070 | 2544 | 2s2p3lnl’ 2s$^{2}$3lnl’ 2p$^{2}$3lnl’ 1s2s$^{2}$2p$^{2}$nl’ |
| N-like | 16437 | 3775 | 2s$^{2}$2p3lnl’ 2s2p$^{2}$3lnl’ 2p$^{3}$3lnl’ 1s2s$^{2}$2p$^{3}$nl’ |
| O-like | 14680 | 3881 | 2s$^{2}$2p$^{2}$3lnl’ 2s2p$^{3}$3lnl’ 2p$^{4}$3lnl’ 1s2s$^{2}$2p$^{4}$nl’ |
| F-like | 12559 | 2773 | 2s$^{2}$2p$^{3}$3lnl’ 2s2p$^{4}$3lnl’ 2p$^{5}$3lnl’ 1s2s$^{2}$2p$^{5}$nl’ |
| Ne-like | 7021 | 1549 | 2s$^{2}$2p$^{4}$3lnl’ 2s2p$^{5}$3lnl’ 2p$^{6}$3lnl’ 1s2s$^{2}$2p$^{6}$nl’ |
| Na-like | 3600 | 693 | 2s$^{2}$2p$^{5}$3lnl’ 2s2p$^{6}$3lnl’ |
| Mg-like | 13521 | 1878 | 2s$^{2}$2p$^{5}$3l$^{2}$nl’ 2s2p$^{6}$3l3l’nl” 2s2p$^{6}$(3l)$^{2}$nl’ |
| Al-like | 4087 | 2457 | 2s$^{2}$2p$^{5}$3s$^{2}$3lnl’ 2s2p$^{6}$3s$^{2}$3lnl’ |
| Si-like | 9052 | 2119 | 2s$^{2}$2p$^{5}$3s$^{2}$3l$^{2}$nl’ 2s2p$^{6}$3s$^{2}$3l$^{2}$nl’ |

# Model assumptions

- Charge states from bare nuclei to Si-like Kr are considered when their abundances are greater than 0.01%
- The maximum principal quantum numbers for levels included are 8 and 6 for bound and autoionizing levels, respectively, and the maximum angular momentum quantum number of up to 5 is used
- The high-n state of autoionizing levels included in the model are limited to those which contribute more than 70% of the full-scale DR calculations at the coronal limit
- Ionization potential depression (IPD) model of Stewart and Pyatt is used to suppress bound states due to continuum lowering
- Partial LTE approximation is applied to use configuration-average model for high-lying configurations which brings about max. 6% uncertainty in charge state balance at the coronal limit

# Selecting DR channels affect charge state distribution: *big time*

A. Complete model with *LLn/LMn/KLn* DR channels with n <7

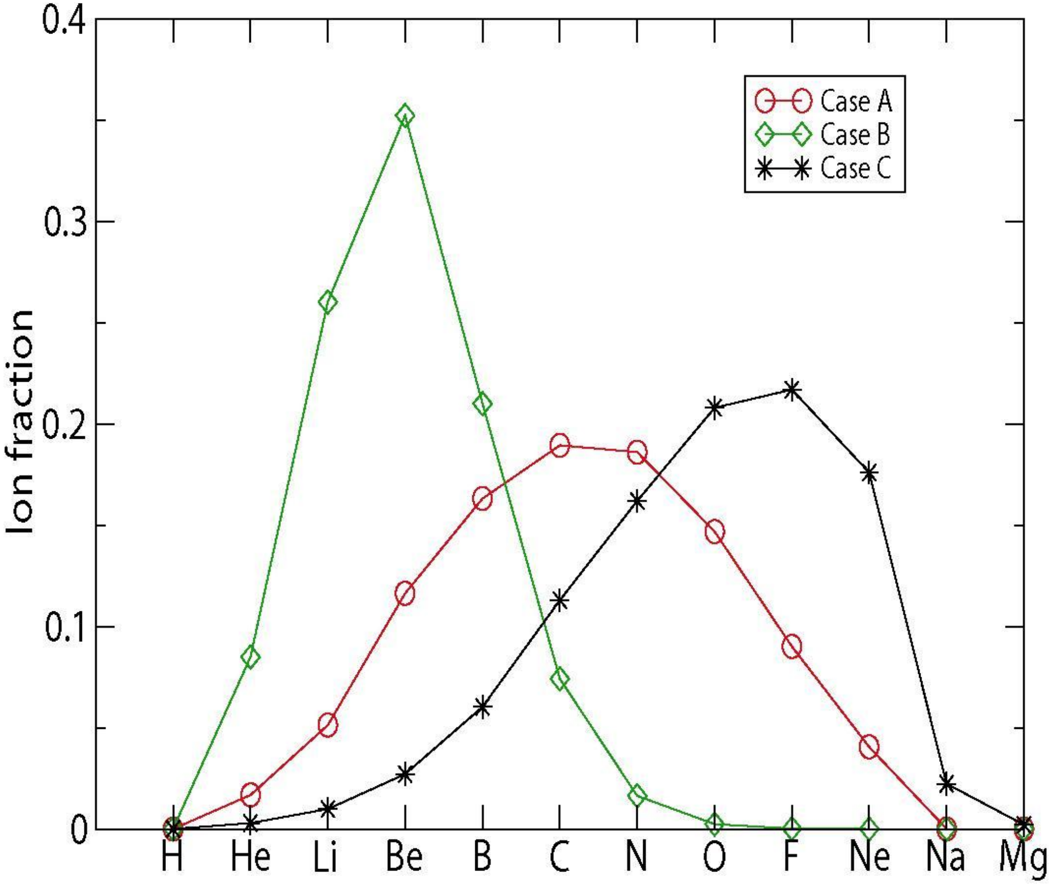
B. Complete model with *KLn* DR channels

C. Coronal model with *LLn/LMn/KLn* DR channels with n ~ ∞

⇒Our model will be valid at Ne~$10^{20}$ $cm^{-3}$

Krypton Charge states

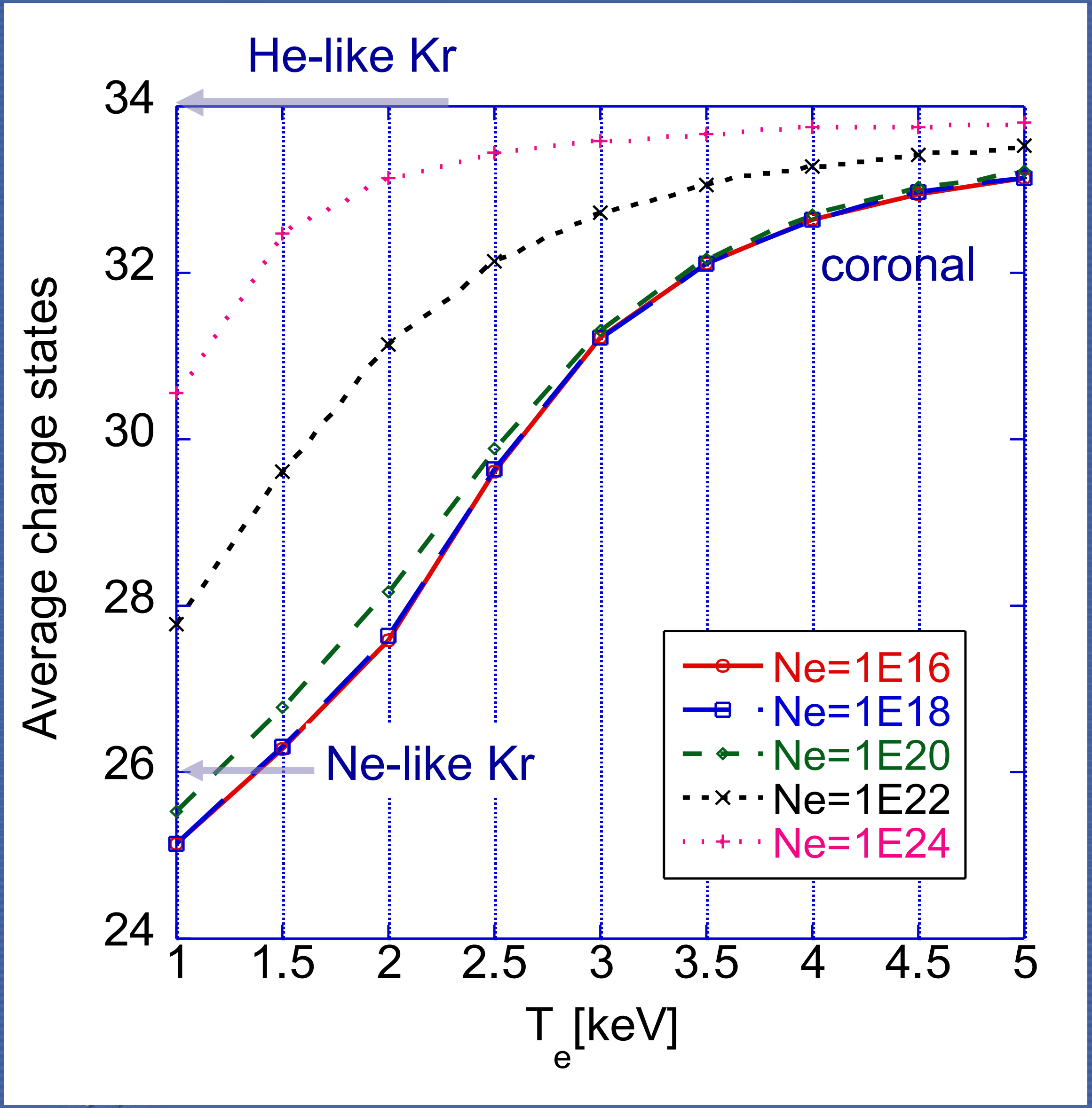
$T_e$=2500 eV/$N_e$=$10^{14}$$cm^{-3}$

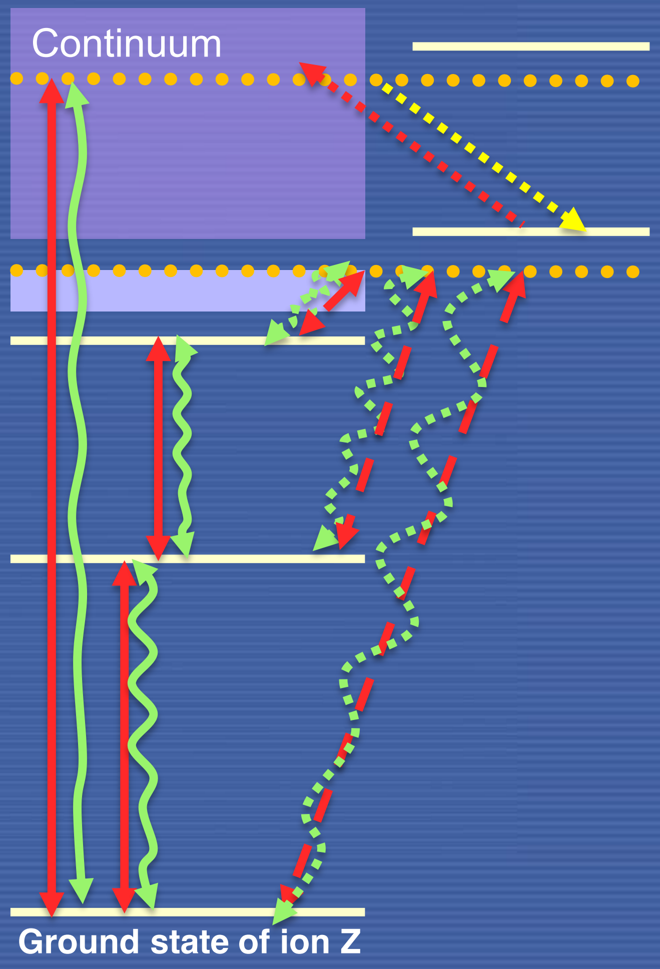


Step-wise excitation

# INTERMEDIATE DENSITIES

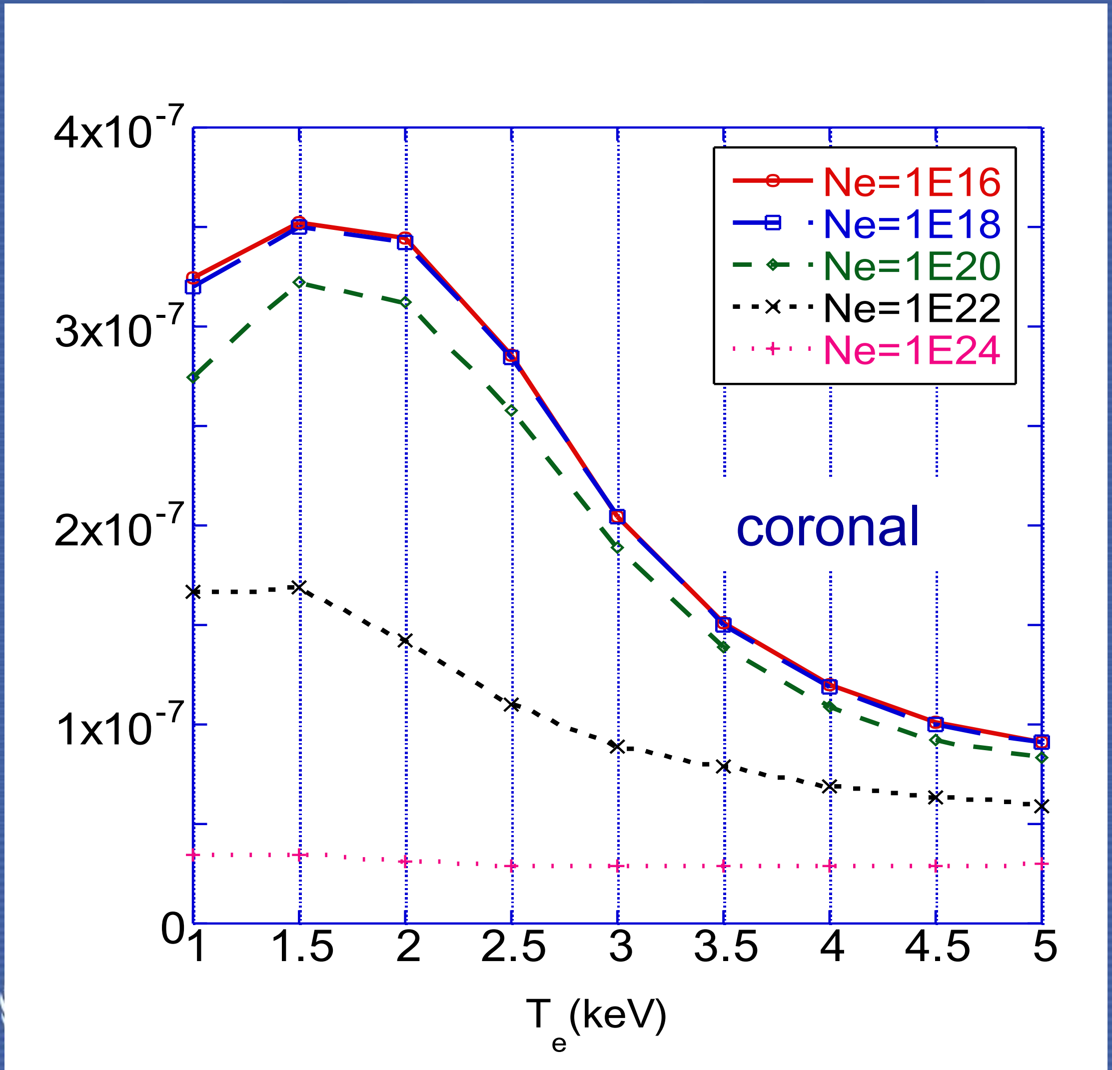
# Excited states contribute to ionization and recombination processes





# Excited level population densities and radiative loss rates are *not linear* with $N_e$

Calculated Kr radiative cooling rates per $N_e$ [eV/s/atom/cm$^{-3}$]



\# of radiative transitions using HULK code

| Ion | HULLAC+DHS |
|---|---|
| 1 | 3049 |
| 2 | 27095 |
| 3 | 30078 |
| 4 | 404328 |
| 5 | 3058002 |
| 6 | 5882192 |
| 7 | 7808014 |
| 8 | 6202123 |
| 9 | 5544814 |
| 10 | 1050919 |
| 11 | 841094 |
| Sum | 30,851,708 |

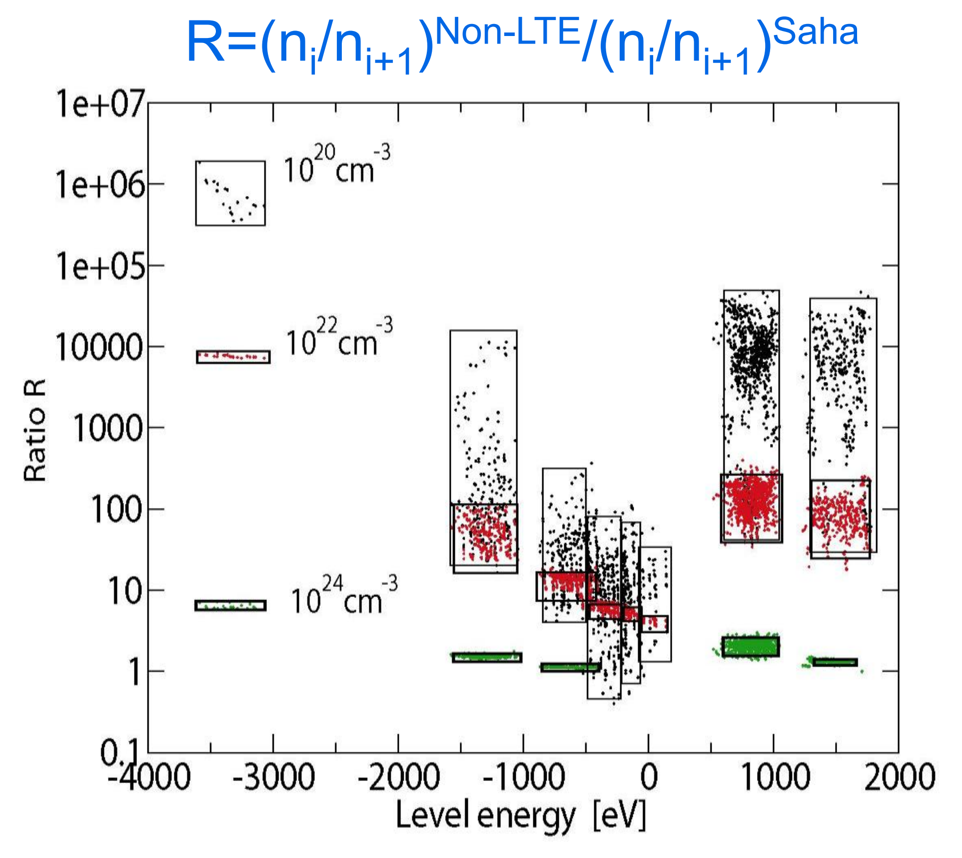
Ionization potential depression

# HIGH DENSITY LIMITS

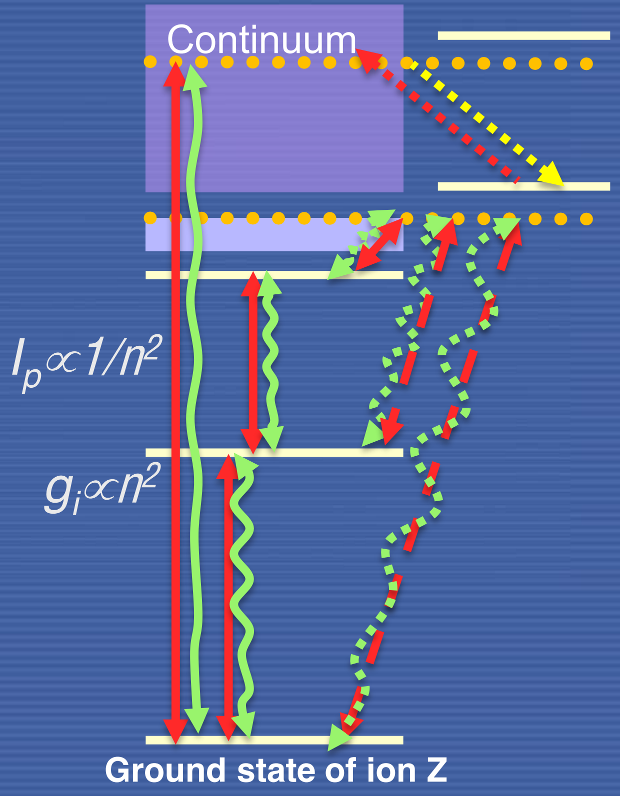
# Highly excited states are in Saha equilibrium with the continuum at high densities

- For Maxwellian electrons, the collisional recombination rate coefficient is given by the detailed balance with collisional ionization rate coefficient
- Recombination rates to high lying hydrogenic levels are <u>proportional to *n*$^4$</u> and *<u>inversely proportional to T$^2$</u>*.

$$R_{colrec}$$
$$= N_e \frac{h^3}{2(2\pi m k_B T)^{3/2}} \frac{g_i}{g_{i+1}} e^{I_p/k_B T} R_{colionz}$$
$$\propto N_e^2 \frac{g_i}{g_{i+1}} e^{I_p/k_B T} \frac{1}{I_p (k_B T)^2}$$

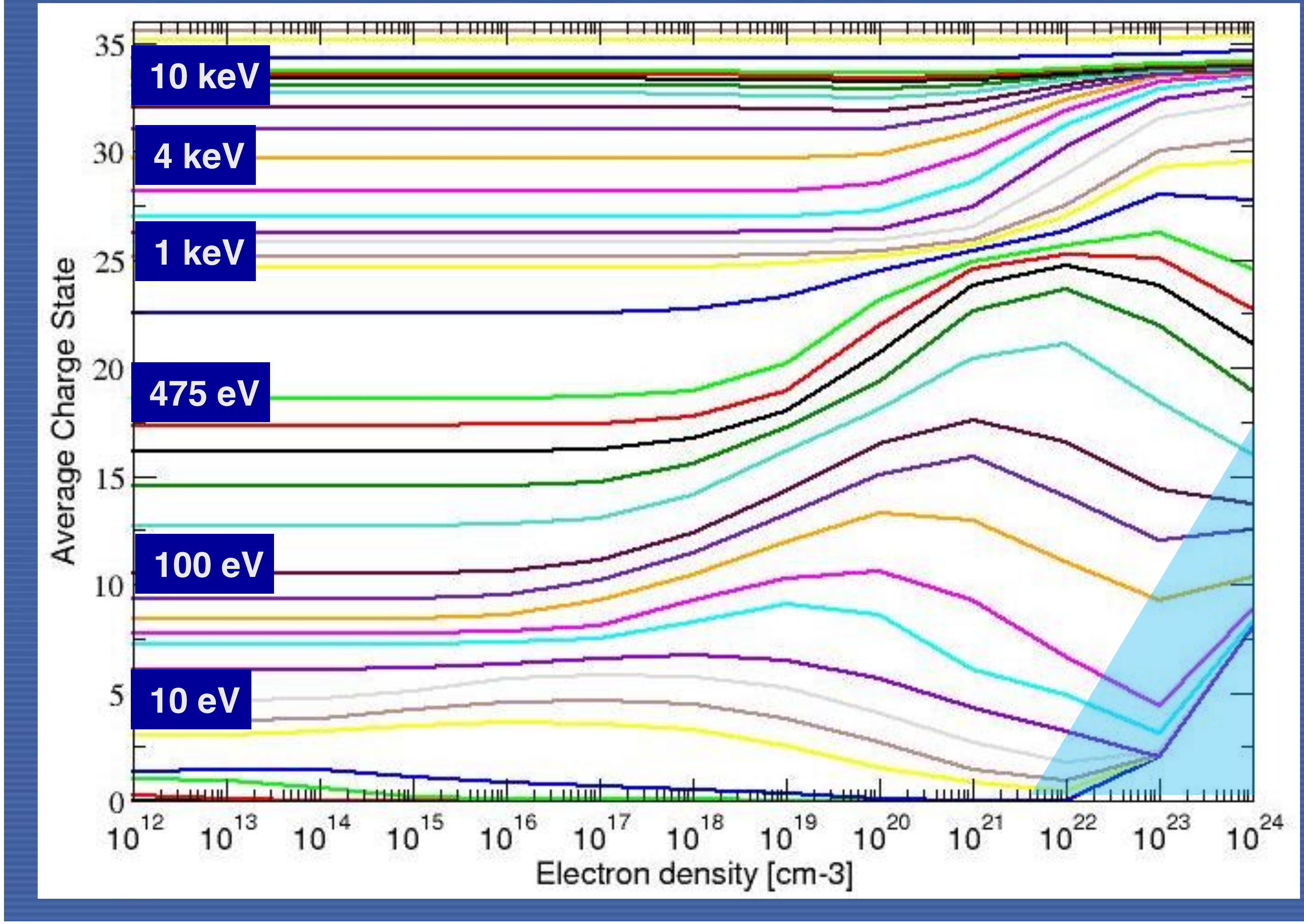


$R=(n_i/n_{i+1})^{Non\text{-}LTE}/(n_i/n_{i+1})^{Saha}$



# Average charge states are sensitive to the treatment of high-lying states

- Due to the 3-body recombination processes, the average charge state *decreases after* reaching the maximum value as $N_e$ increases, then *increases again* due to the ionization potential depression.



**Krypton**

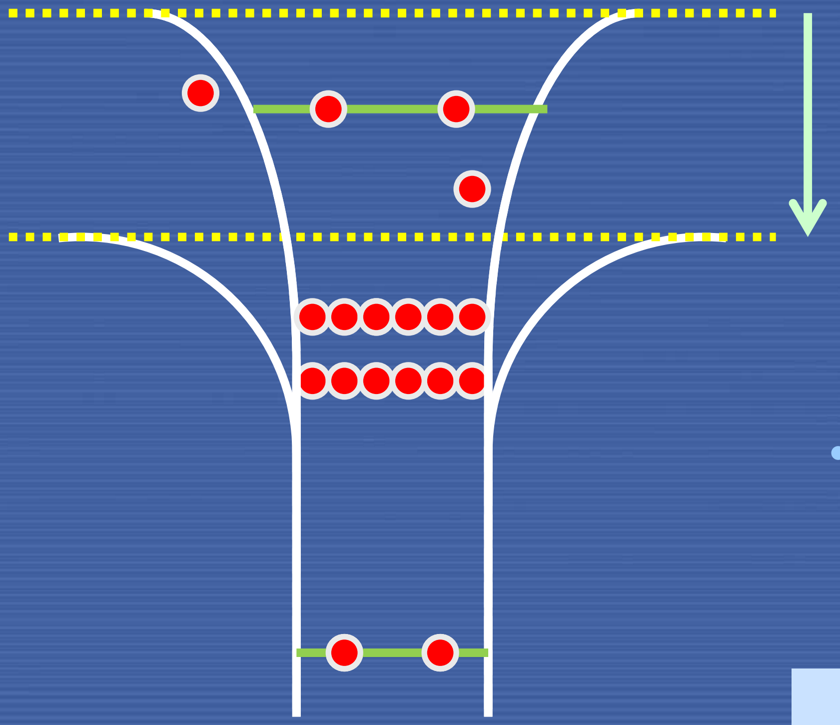
**FLYCHK**

**$T_e$=0.5 eV-100 keV**

**$N_e=10^{12}-10^{24}$ cm$^{-3}$**

# Plasma effects: Ionization potential depression & Rate changes

- High-lying states are no longer bound due to interactions with neighbouring atoms and ions. (pressure ionization)
- Ionization potentials are a function of plasma conditions



Isolated atom continuum

*Ionization potential depression (IPD)*

Embedded atom continuum

- For a given $n_e, T_e$, the IPD $\Delta\chi$, is calculated using a *Stewart-Pyatt* type model

Where $r_i$ and $r_d$ are the ion sphere and Debye radii

$$\Delta\chi = 2.16\text{x}10^{-7}\ \frac{z}{r_i}\left(\left(1+\left(\frac{r_d}{r_i}\right)^3\right)^{2/3} - \left(\frac{r_d}{r_i}\right)^2\right)\quad (eV)$$

Opacity effects and opacity broadening

# DENSITY EFFECTS ON RADIATIVE PROCESSES

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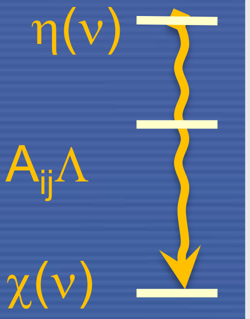
# Escape probability: Modified radiative rates due to optical depth effects

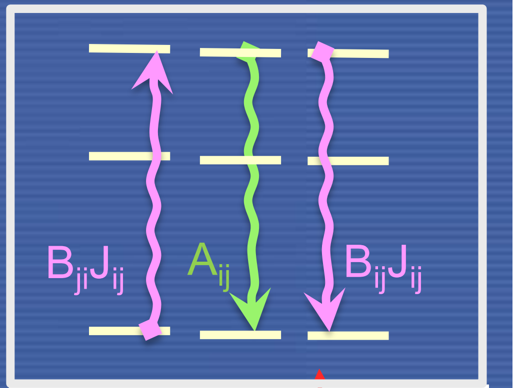
## Escape probability Λ reduces the total A rate

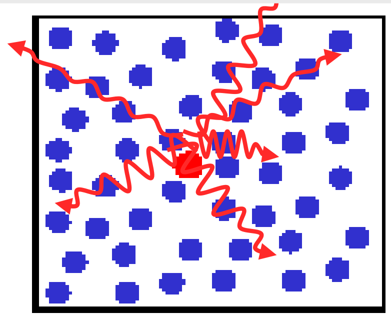
$$R_{ij} = n_i(A_{ij} + B_{ij}\bar{J}_{ij}) - n_j B_{ji}\bar{J}_{ij} \equiv n_i A_{ij}\Lambda(n_i, n_j)$$

$$\bar{J}_{ij} = \int \phi(\nu)d\nu \frac{1}{4\pi}\oint I(\nu)\, d\omega$$

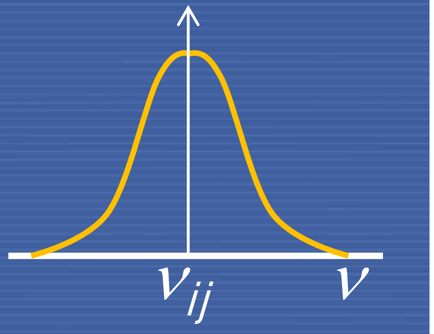
$$\mu \frac{\partial I(\nu)}{\partial \tau(\nu)} = I(\nu) - \frac{\eta(\nu)}{\chi(\nu)}$$







$\phi(\nu)$ : line profile



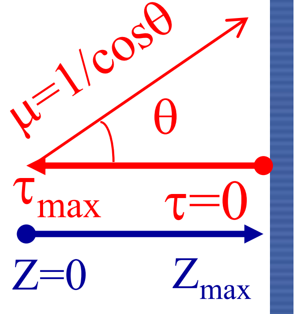
## Frequency-averaged escape probabilities

$$P_e(\tau) = \int_0^\infty d\nu\, \phi(\nu) e^{-\tau\phi(\nu)/\mu\phi(\nu=0)}$$

$$\overline{P_e(\tau)} = \int_0^1 P_e\left(\frac{\tau}{\mu}\right) d\mu$$

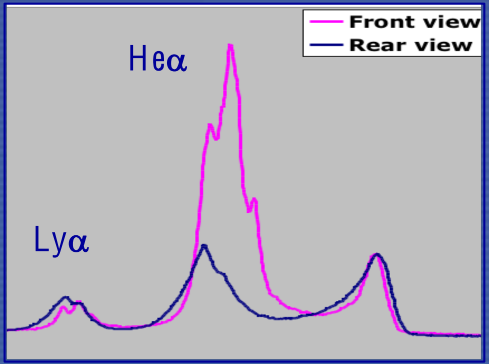
$$\tau(z,\nu) = \int_z^{z_{max}} \chi(z',\nu)dz'$$

Planar geometry



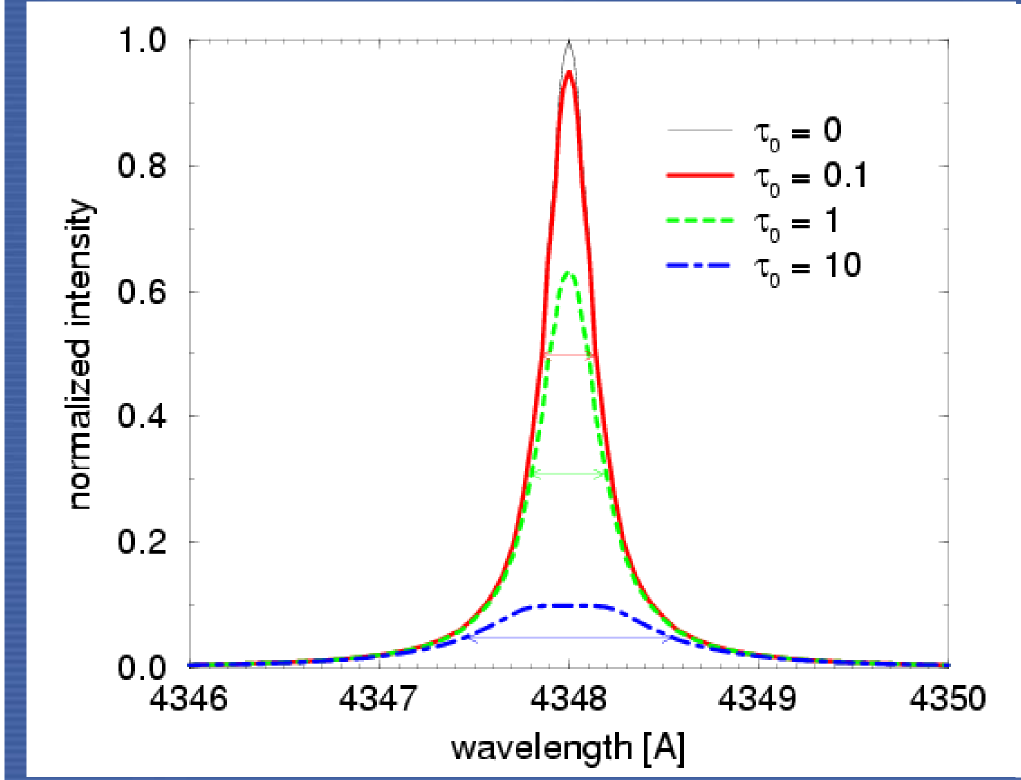
# Modified line intensities by opacity broadening

Measured ratios between Ly- α and He- α lines are only possible for $T_e > 1$ keV for optically thin case

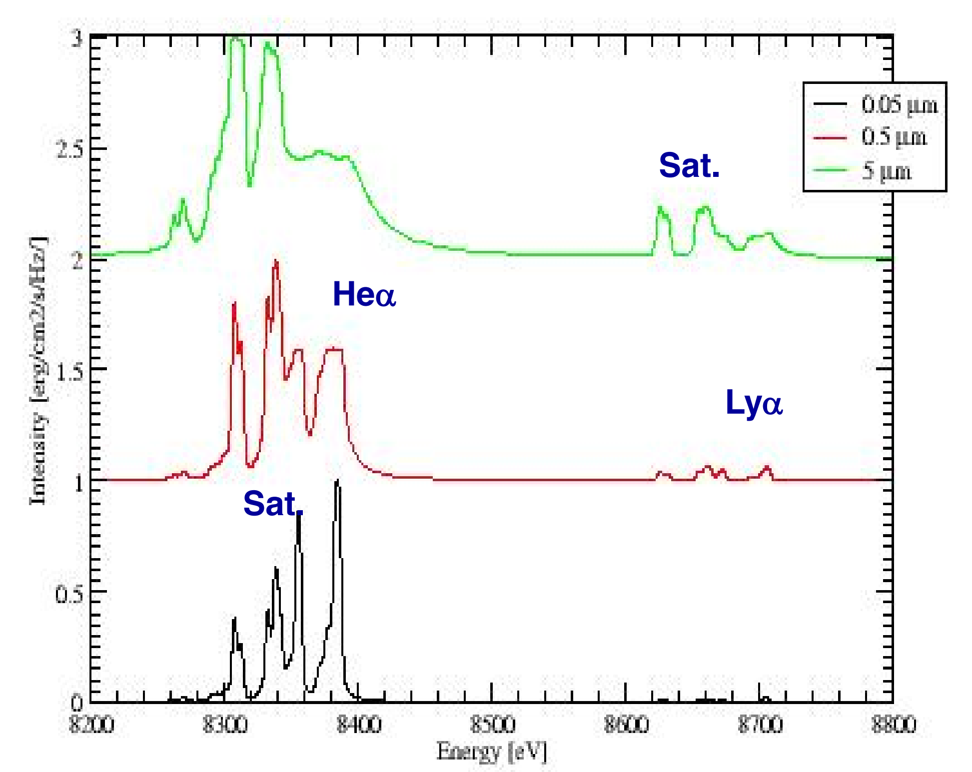


Measured ratios between Ly- α and He-α lines increase with plasma size/opacity effects

Opacity broadening due to line-core absorption



Cu Solid, $T_e$=2 keV, $N^{hot}$=2% 200keV



Reality Check

# NON-EQUILIBRIUM EFFECTS

# Consideration of the non-equilibrium effects for spectral analysis

- Laboratory plasmas are complex
  - Time-dependence → time-dependent NLTE model
  - Spatial non-uniformity →NLTE radiation transport code
  - External radiation field
  - Non-thermal particles

- Non-equilibrium states due to other sources should be carefully considered before applying the NLTE model for measured spectra

FLYCHK

# GENERALIZED POPULATION KINETICS MODEL

IAEA

# FLYCHK Model : *simple, but complete*

FLYCHK → HULLAC / FAC / MCDF

*(n)* *(nl)* *(nlj)* *(detailed-term)*

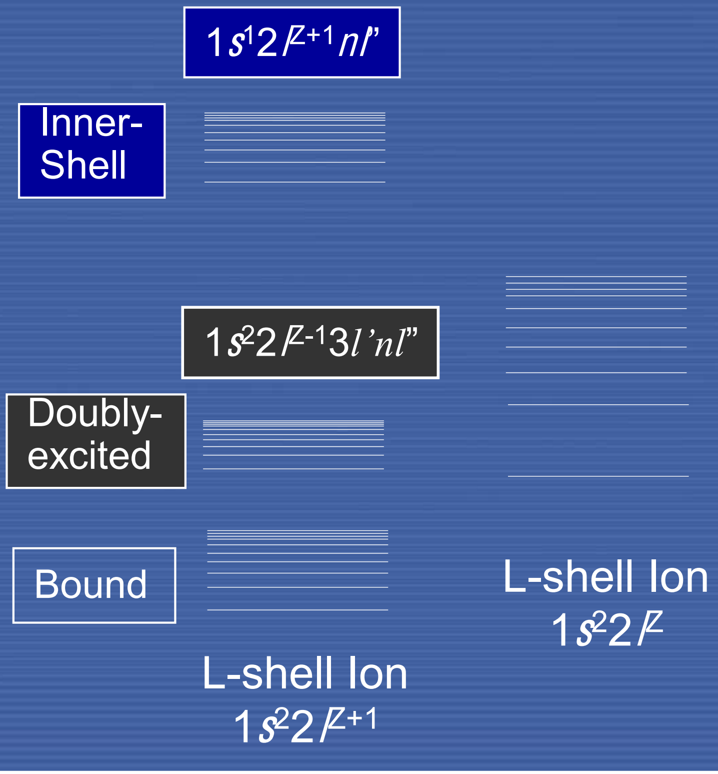
- Screened hydrogenic energy levels with relativistic corrections
- Dirac Hartree-Slater oscillator strengths and photoionization cross-sections
- Fitted collisional cross-section to PWB approximation
- Semi-empirical cross-sections for collisional ionization
- Detailed counting of autoionization and electron capture processes
- Continuum lowering (Stewart-Pyatt)

# Application to a wide range of Z and experiments:

## *Excitation autoionization (EA) /Dielectronic recombinationa (DR) processes are modeled with doubly-excited and inner-shell (IS) excited states*

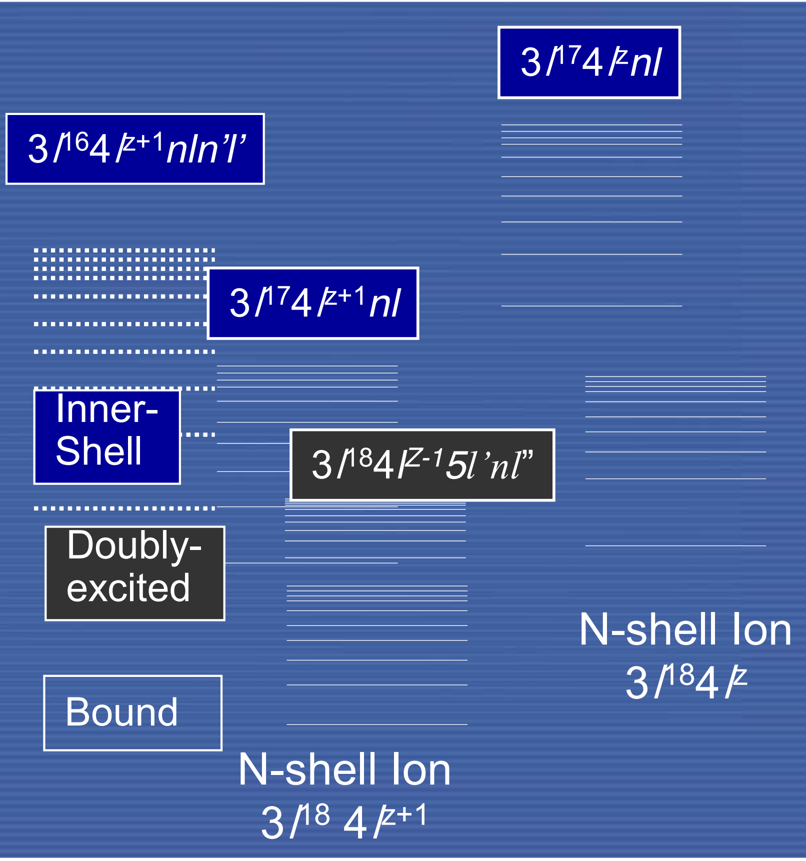
### Low Z atom

Promotion of IS electrons leads to states far from continuum limit and *rarely matters in CSD*



### High Z atom

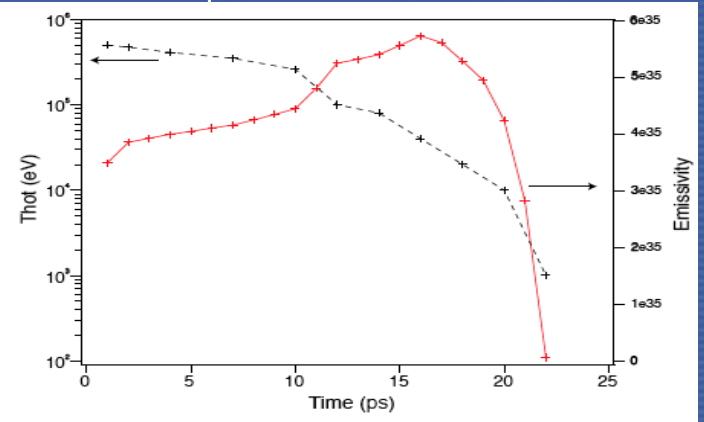
Promotion of **IS** electrons can lead to states near the continuum limit and EA/DR process of **IS** is ***critical***

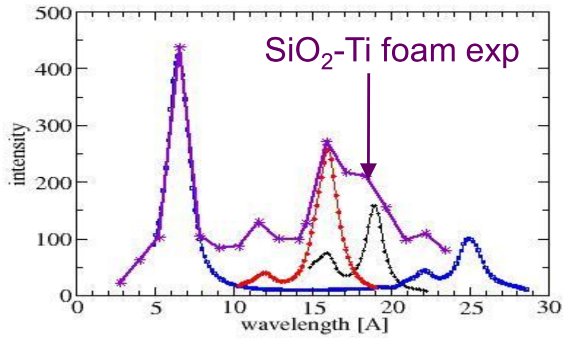


# Applications to Plasma Research

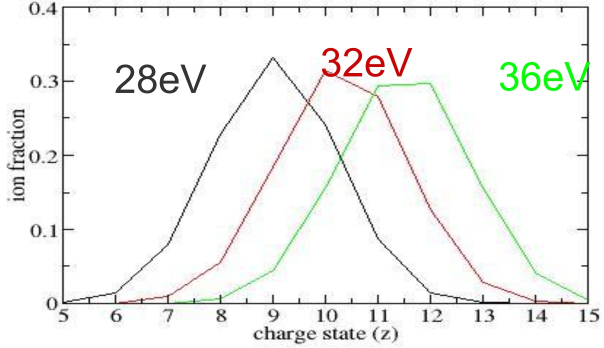
- Short-pulse laser-produced plasmas
  - Arbitrary electron energy distribution function
  - Time-dependent ionization processes
  - K-α shifts and broadening: diagnostics
- Long-pulse laser-produced plasmas
  - Average charge states
  - Spectra from a uniform plasma
  - Gas bag, Hohlraum (H0), Underdense foam
- Z-pinch plasmas: photoionizing plasmas
- Proton-heated plasmas: warm dense matter
- EBIT: electron beam-produced plasmas
- EUVL: Sn plasma ionization distributions
- TOKAMAK: High-Z impurities

Time-dependent Ti Kα emissivities





Tin charge state distributions



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# Available to the community at password-protected NIST website: http://nlte.nist.gov/FLY

**Advantages**: simplicity and versatility→ applicability

- <Z> for fixed any densities: electron, ion or mass
- Mixture-supplied electrons (eg: Argon-doped hydrogen plasmas)
- External ionizing sources : a radiation field or an electron beam.
- Multiple electron temperatures or arbitrary electron energy distributions
- Optical depth effects

**Outputs:** population kinetics code and spectral synthesis

- <Z> and charge state distribution
- Radiative Power Loss rates under optically thin assumption
- Energy-dependent spectral intensity of uniform plasma with a size

**Caveats**: simple atomic structures and uniform plasma approximation

- Less accurate spectral intensities for non-K-shell lines
- Less accurate for low electron densities and for LTE plasmas
- When spatial gradients and the radiation transport affect population significantly