Two Novel Cases of Plasma Characterization Through Spectroscopic Modeling

Manolo E. Sherrill

Theoretical Division, Los Alamos National Laboratory

March 28, 2005

Two Cases of Spectroscopic Modeling

Plasmas Formed During Laser Ablation

- Most common laser produced plasma
- $I_{laser} = 1 \times 10^{7} - 1 \times 10^{10}$ W/cm^2
- T_e = 1-3 eV
- Stoichiometrically complex targets
- Time-dep. atomic kinetics
- Opacity and lineshape
- Uniqueness/Topology/Search Tech.

Intense Ultrashort Laser Plasmas

- Non-Maxwellian FEEDF
- Time-dep. atomic and e^- kinetics
- $I_{laser} = 1 \times 10^{15} - 1 \times 10^{21}$ W/cm^2; $\tau = 20 - 300 fs$
- T_e= 1000-1500 eV (Final T.E.)
- Ar cluster targets
- Atomic kinetics and plasma physics?

Spectroscopic Modeling of a Collisionally Confined Laser-ablated Plasma Plume

M. E. Sherrill and R. C. Mancini

Department of Physics, University of Nevada, Reno, Nevada

J. Bailey, A. Filuk, B. Clark, and P. Lake

Sandia National Laboratories, Albuquerque, New Mexico

J. Abdallah, Jr.

Theoretical Division, Los Alamos National Laboratory

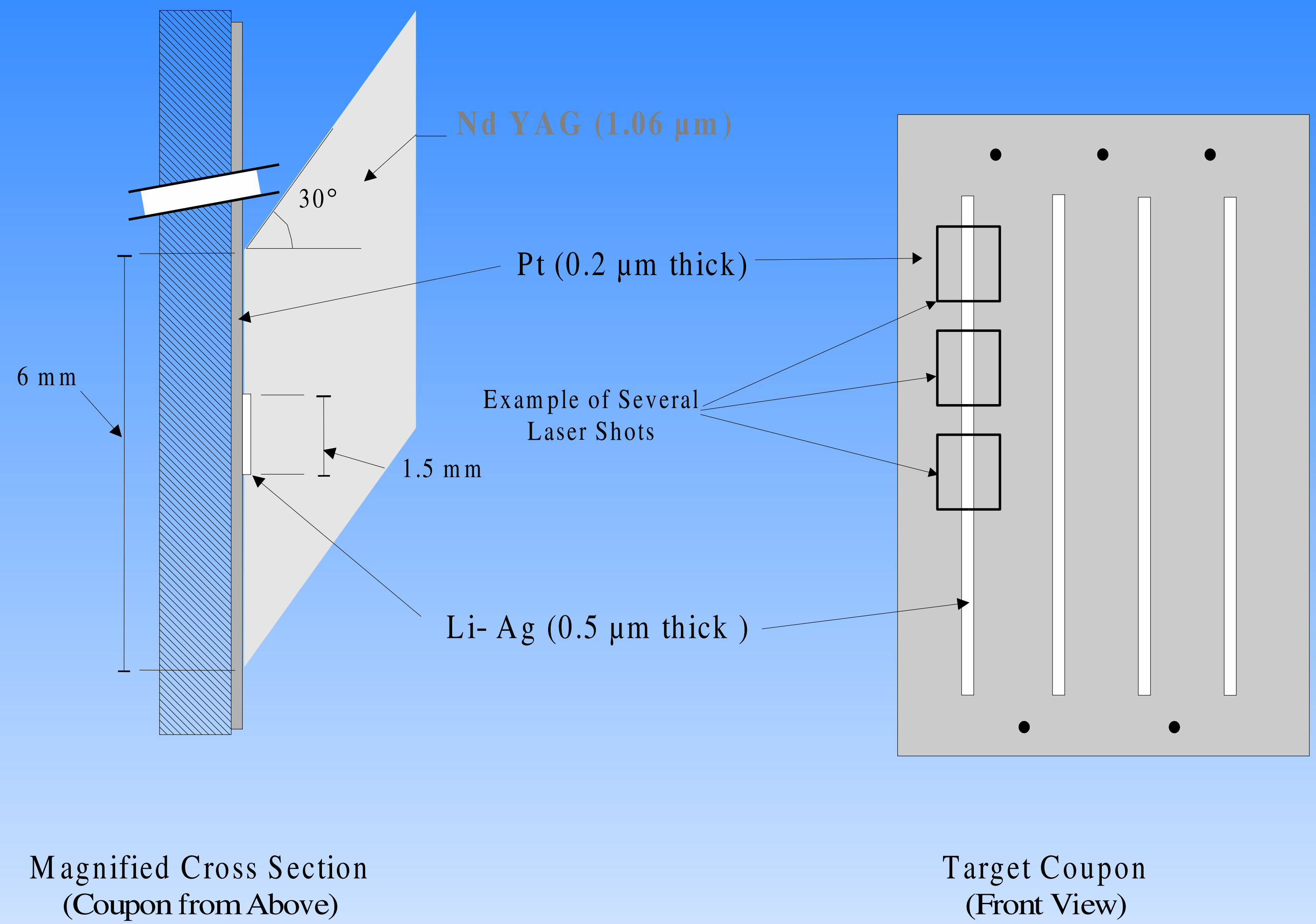


Figure 1: Target Design.

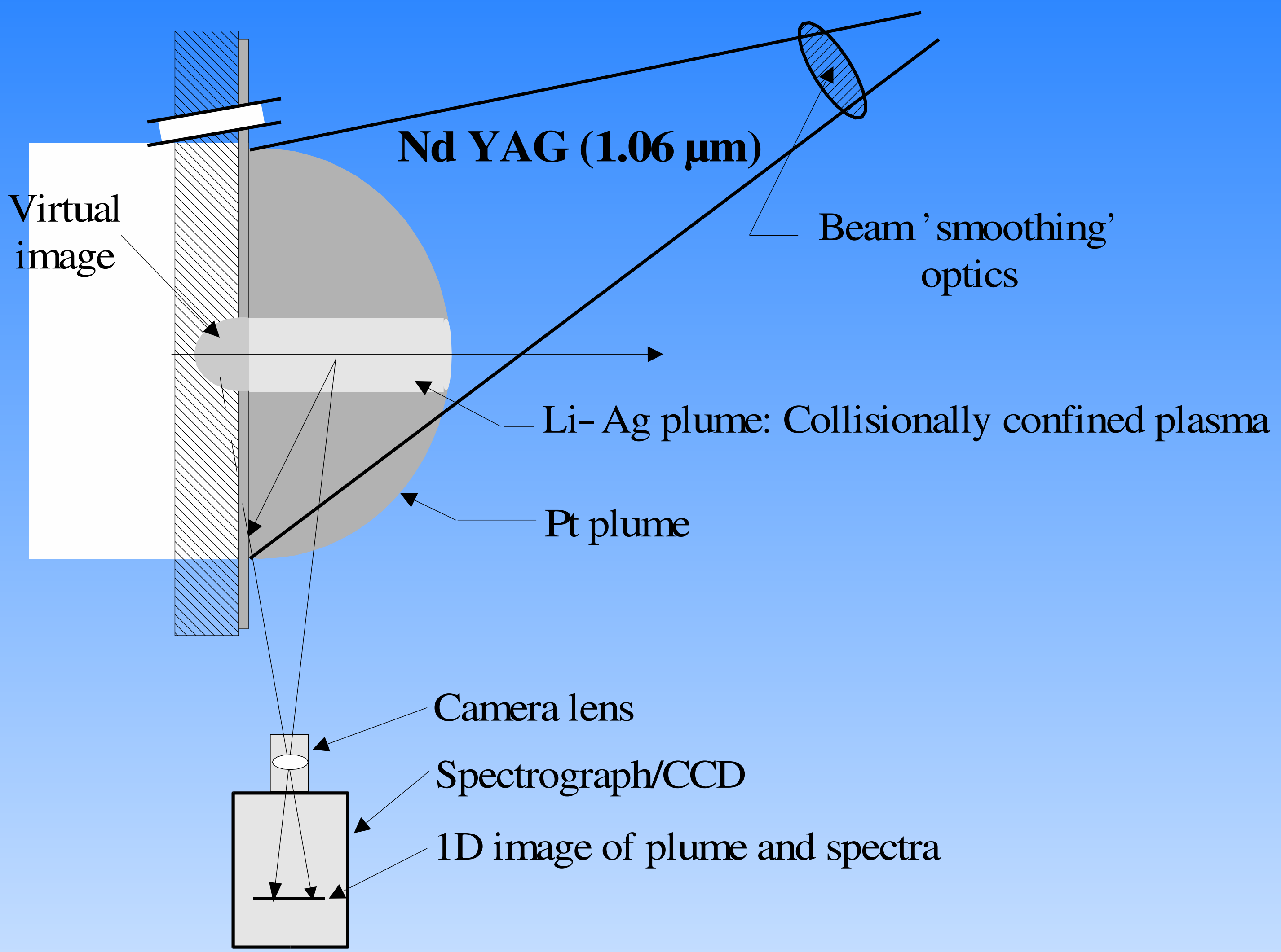


Figure 2: Experimental Configuration.

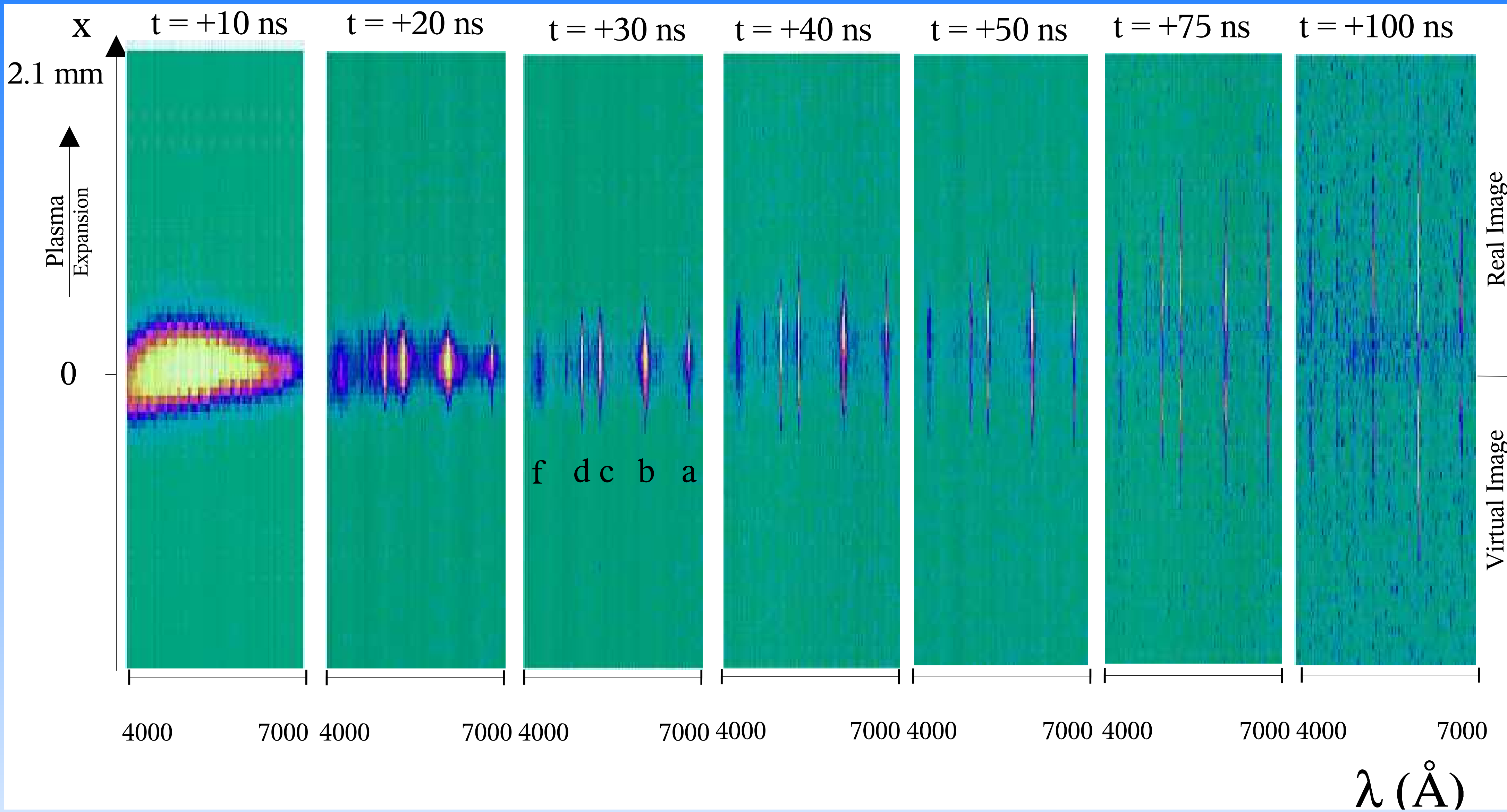


Figure 3: CCD Images.

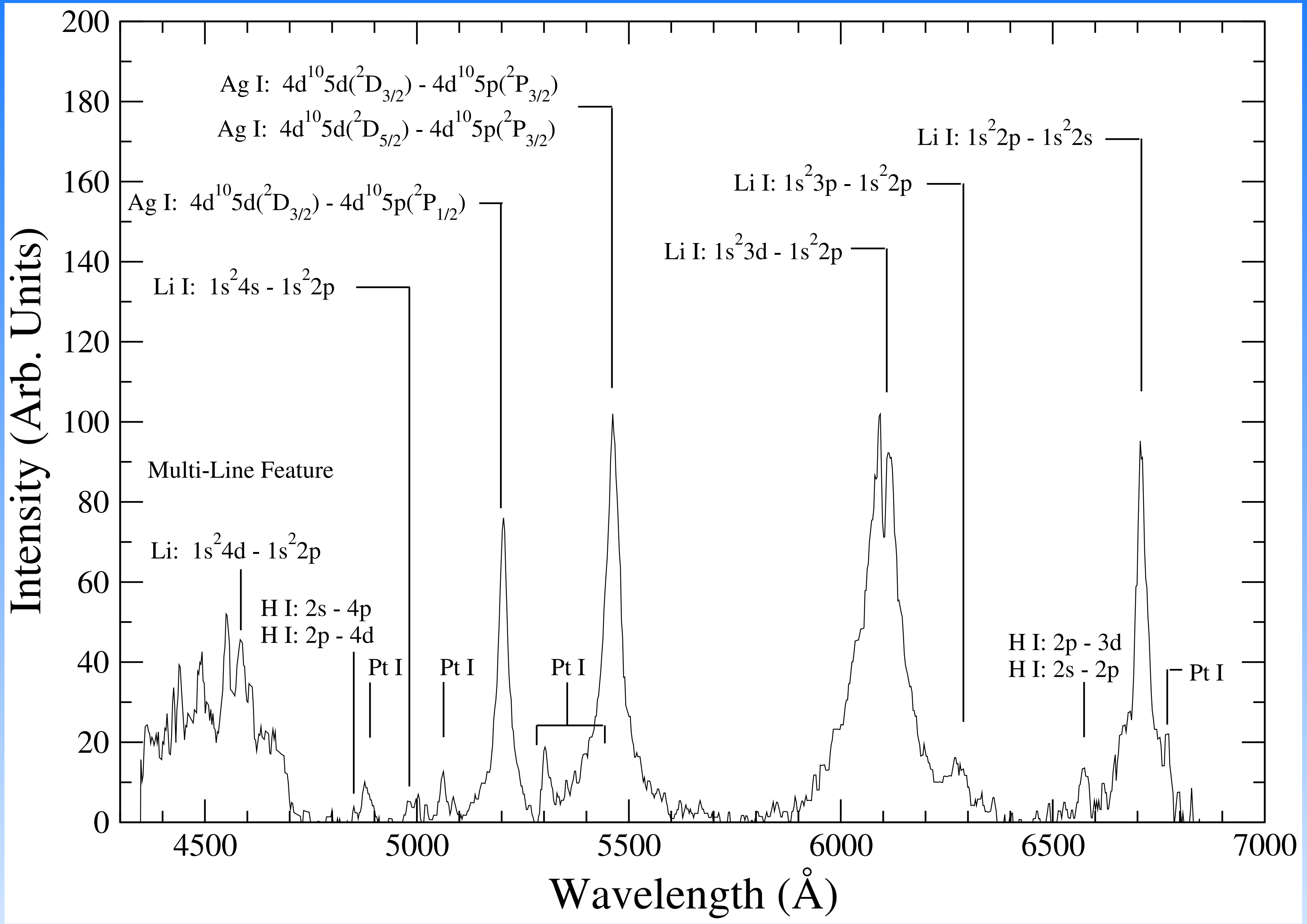


Figure 4: Early in time and close to the target surface, this lineout (x=28.8 μm, t=+20 nsec) displays the high density characteristics of Stark broadened line shapes.

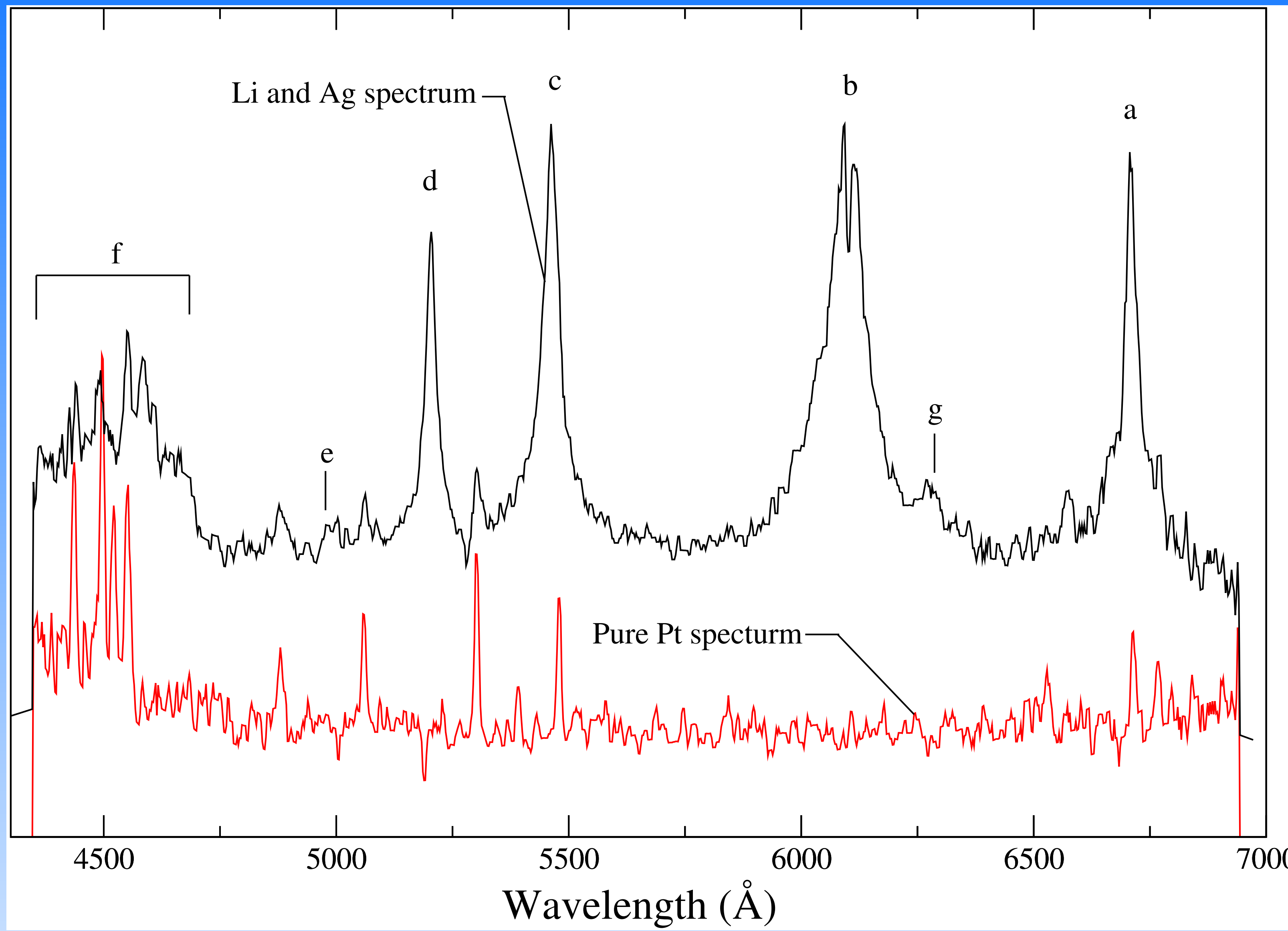


Figure 5: The superposition of two lineouts shows the location of platinum lines in the x=28.8 μm, t=+20 nsec spectrum. The spectrum of a pure platinum target was recorded during the Li-Ag trails at t=+50 nsec, x=56.7 μm from the surface with a 10 ns gate time.

Spectroscopic Model

Best Fit - Synthetic and Experimental Spectra

- Improving atomic structure calculations for high Z neutrals: energy levels and wavefunctions – cross sections and rates
- Reducing the number of calculated cross sections Ag^{+1}, Ag^{+2}, Ag^{+3}. - multi-representational model
- Multi-element collisional radiative model - common free $e-$ pool
- Local plasma effects on the radiator - detailed line shapes
- Non-local effects on the spectra - radiation transport accurate RTE solution
- Plasma gradients – self-reversal feature

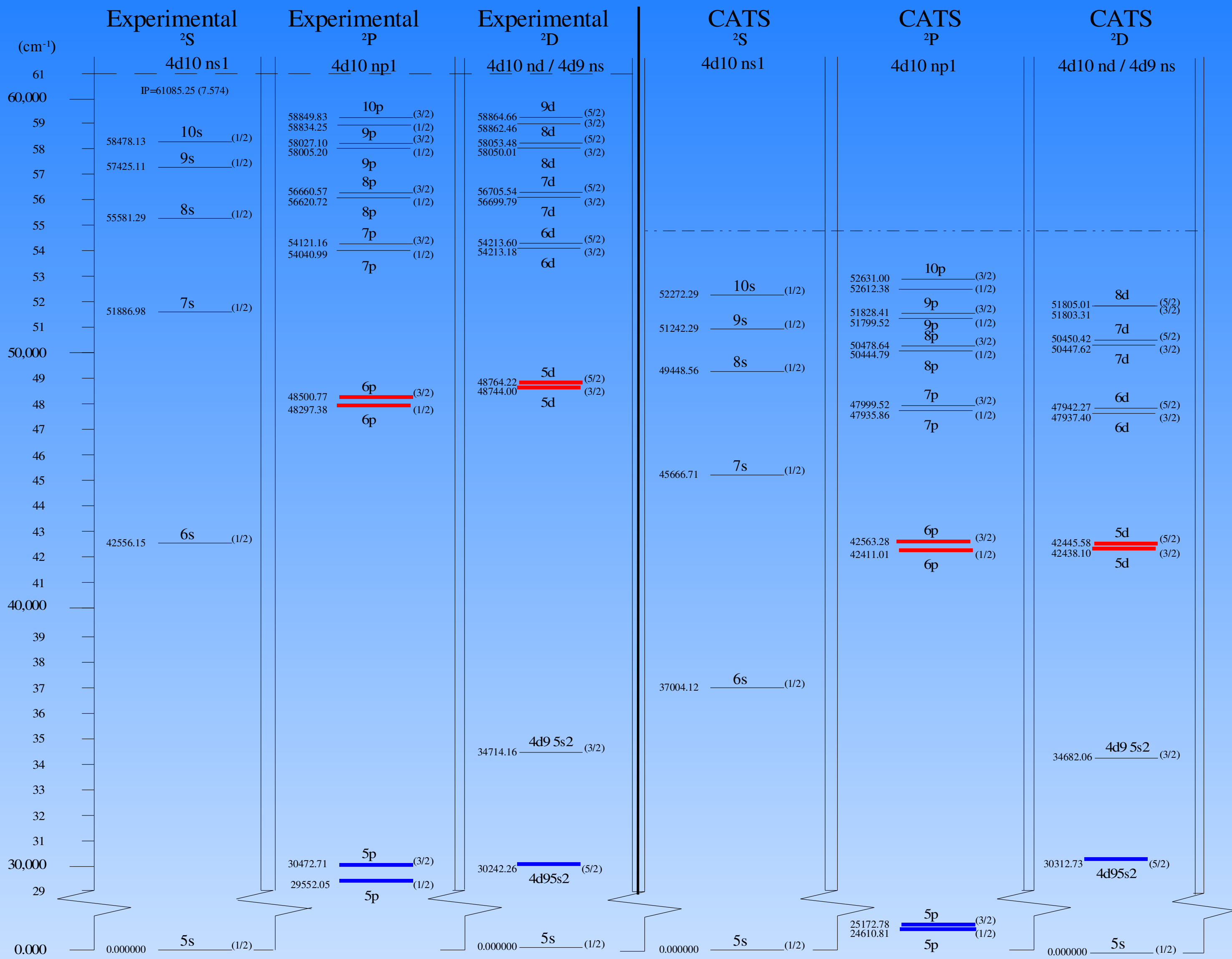


Figure 6: Comparison between experimentally obtained energy level structure (left) and that generated with Cowan's Atomic Structure code (CATS) (right) for neutral Ag.

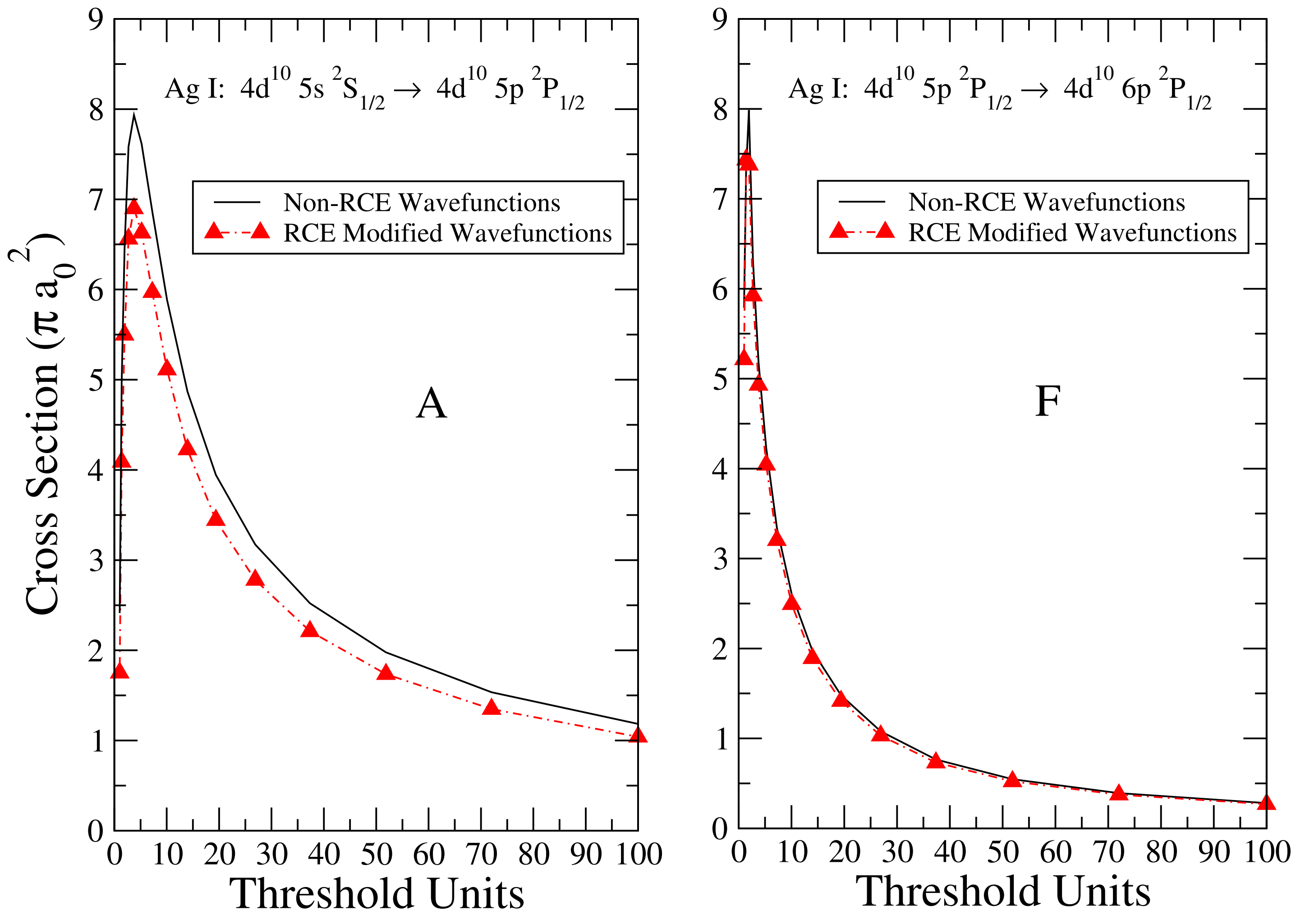


Figure 7: Comparison of electron impact excitation cross sections generated with and with RCE modifications of the wavefunctions. The two figures correspond to an allowed **A** (left) and forbidden **F** (right) transition.

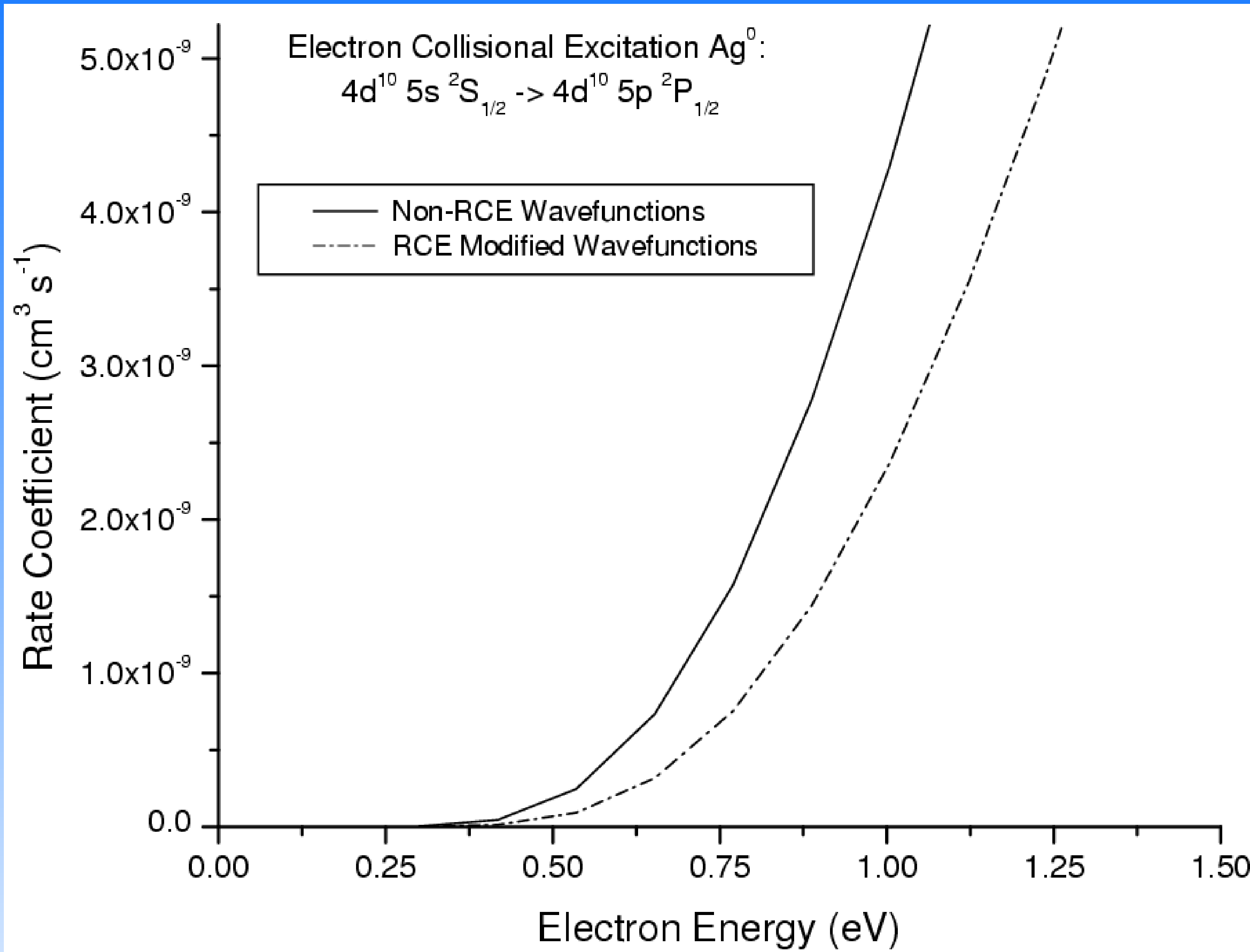


Figure 8: A comparison of an electron impact excitation rate for silver generated with and without wavefunction modifications by the RCE procedure.

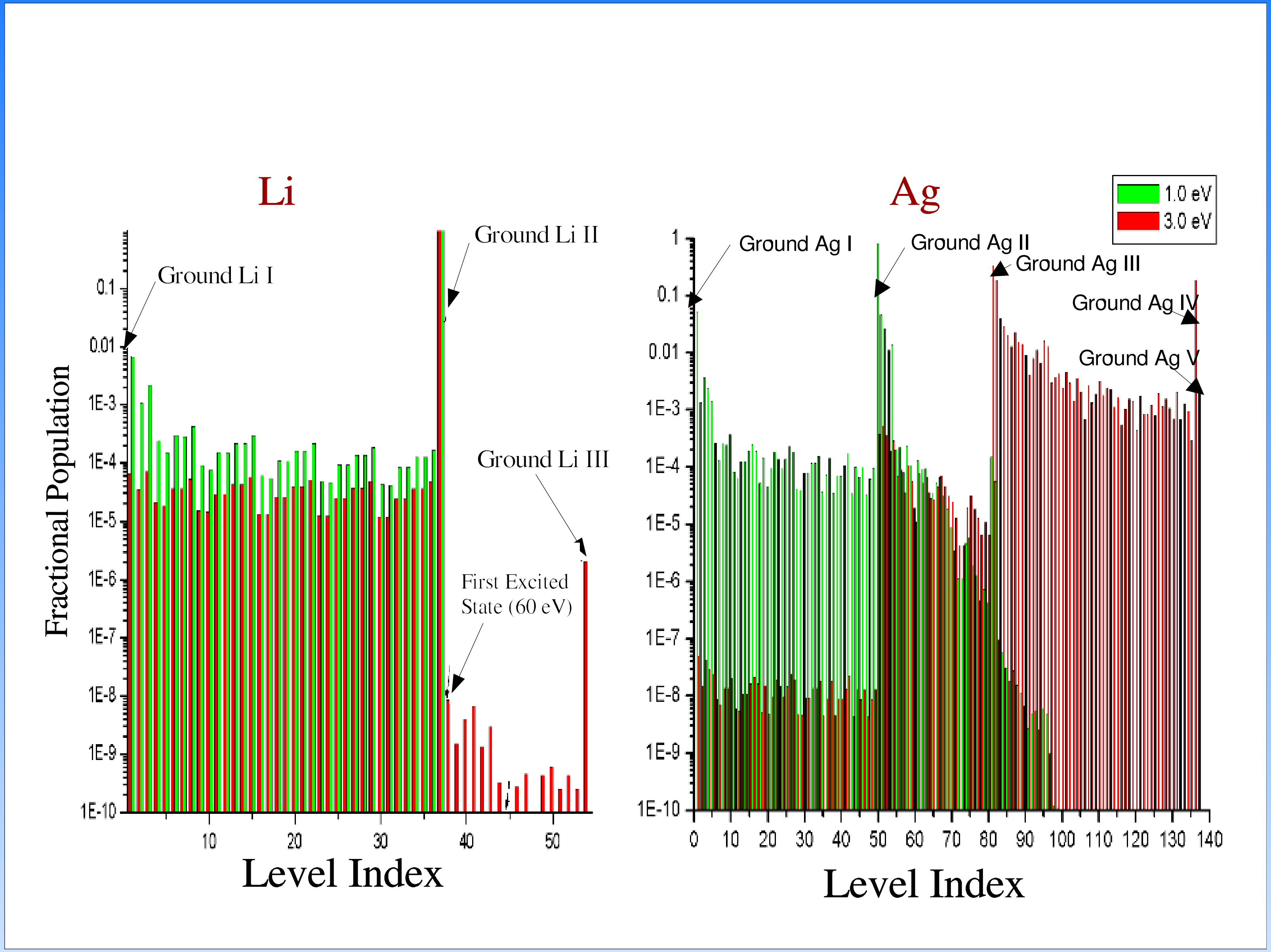


Figure 9: The effect of temperature on fractional level populations for three ionization stages in Li and five in Ag atoms, for an electron density of 1x10^{17} cm^{-3}.

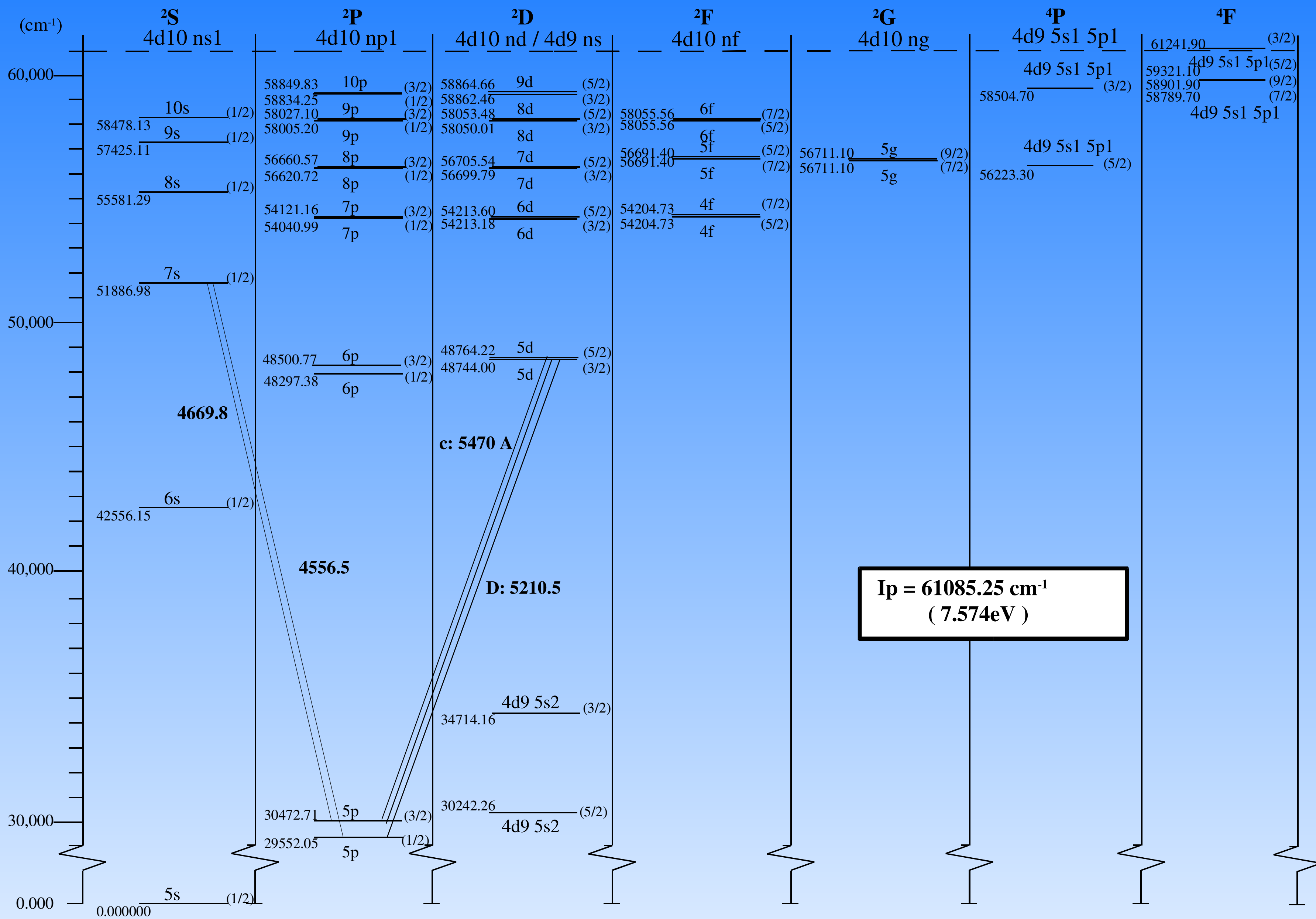


Figure 10: Ag I Grotrian diagram.

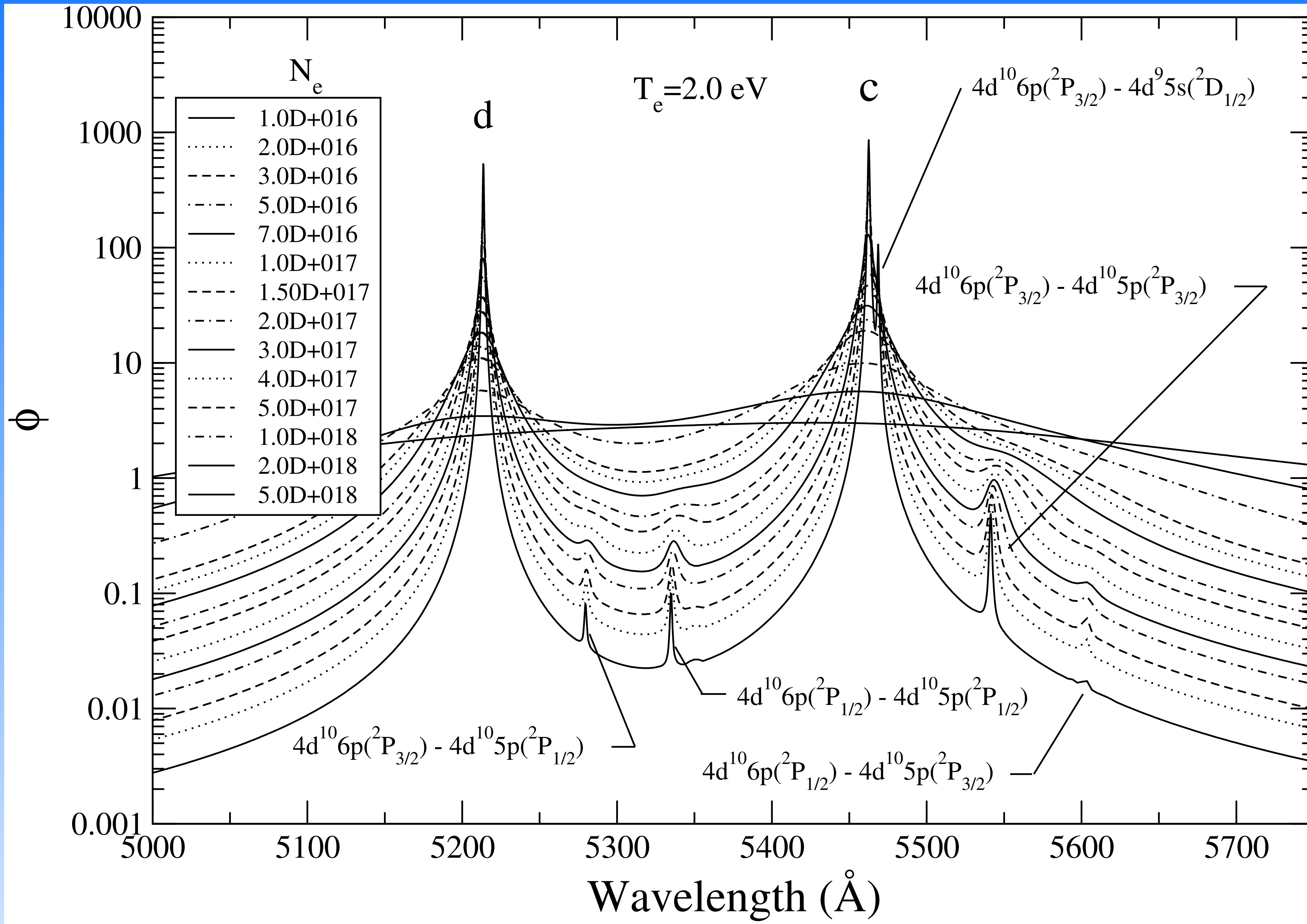


Figure 11: Neutral silver line profiles for various plasma electron densities with the two dominant $5d - 5p$ fine structure transitions.

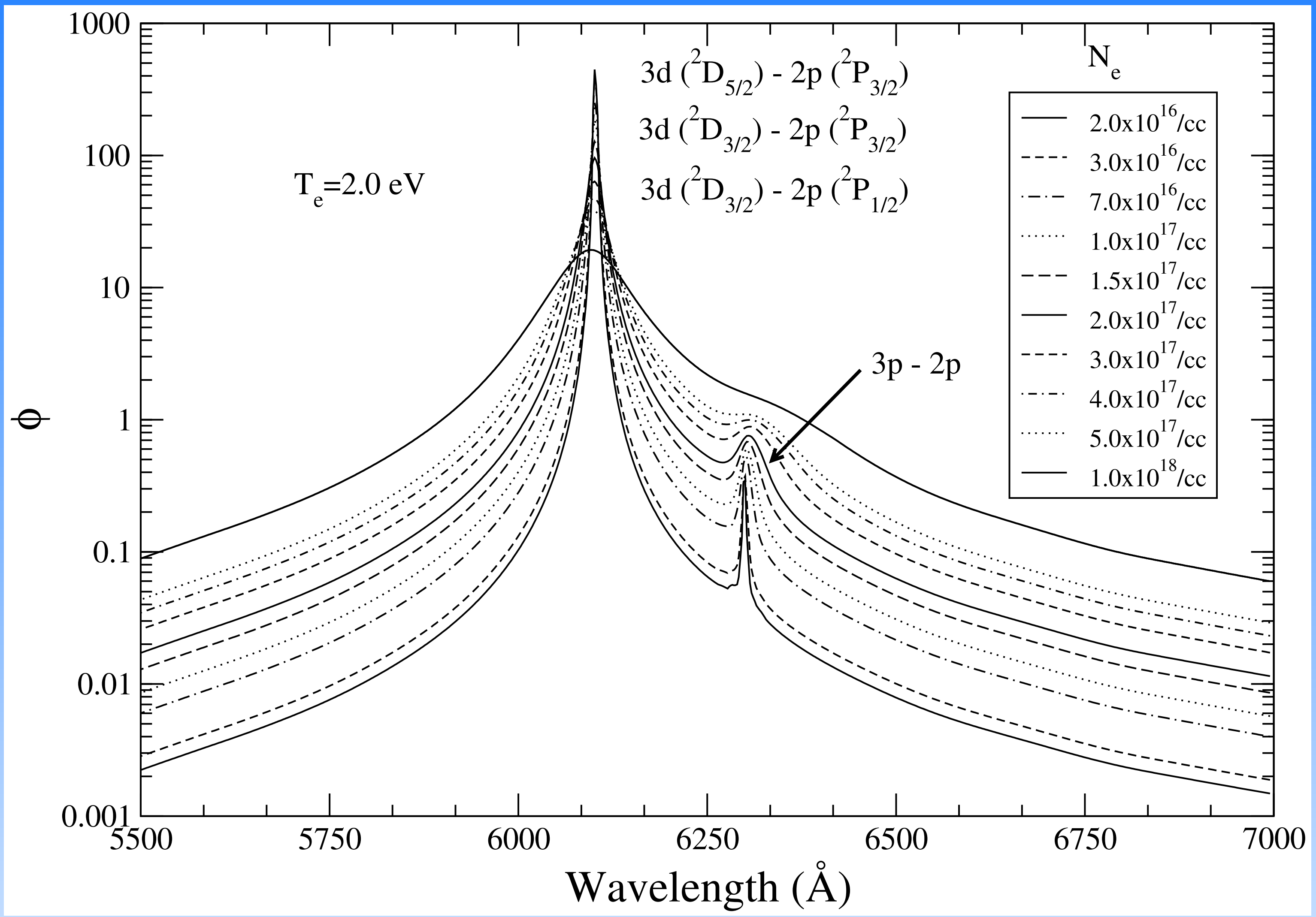


Figure 12: Neutral lithium line profiles at various plasma electron densities for the dominant $3d - 2p$ transition. Note the asymmetry at the peak.

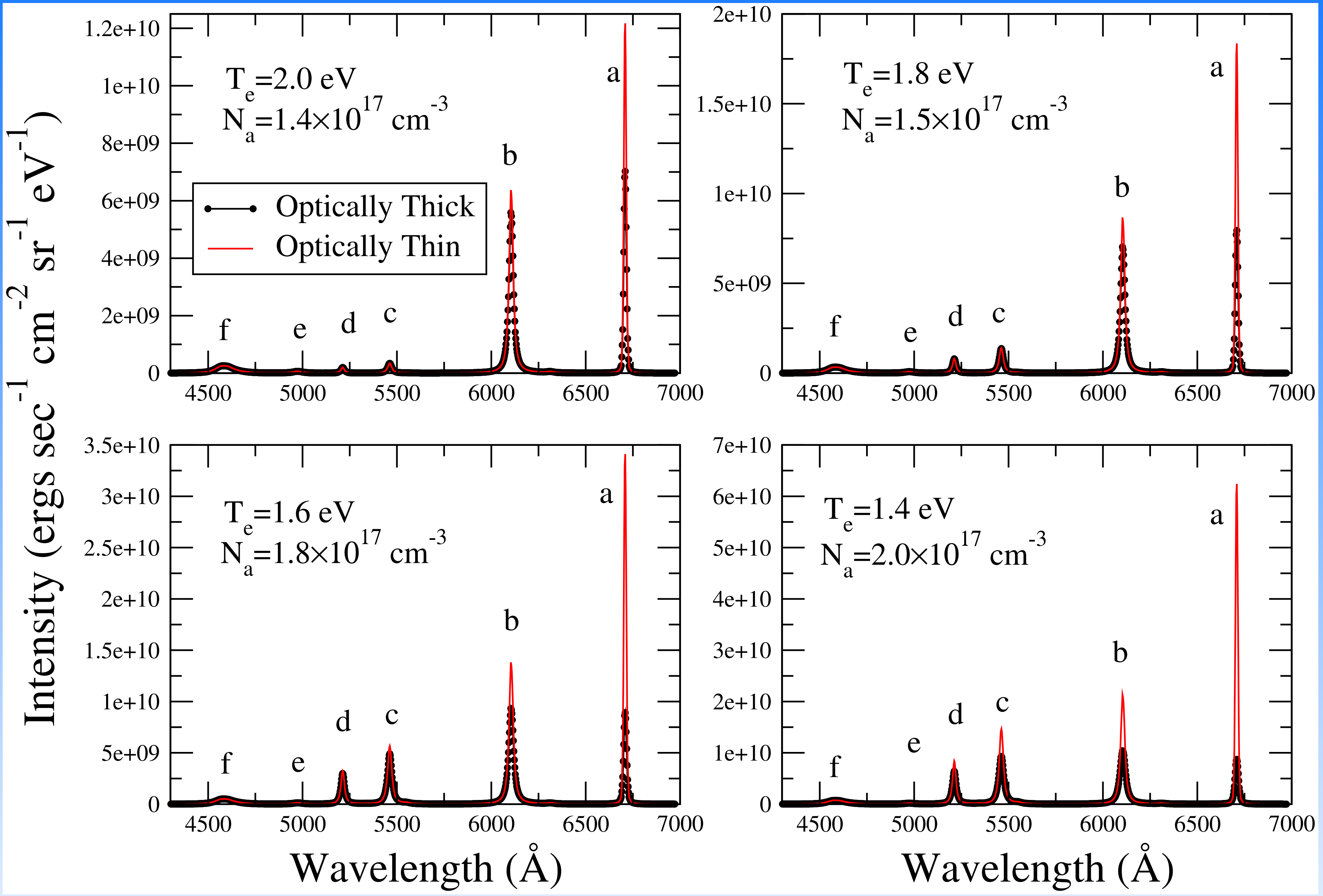


Figure 13: Superposition of optically thin and thick spectra for constant N_e series.

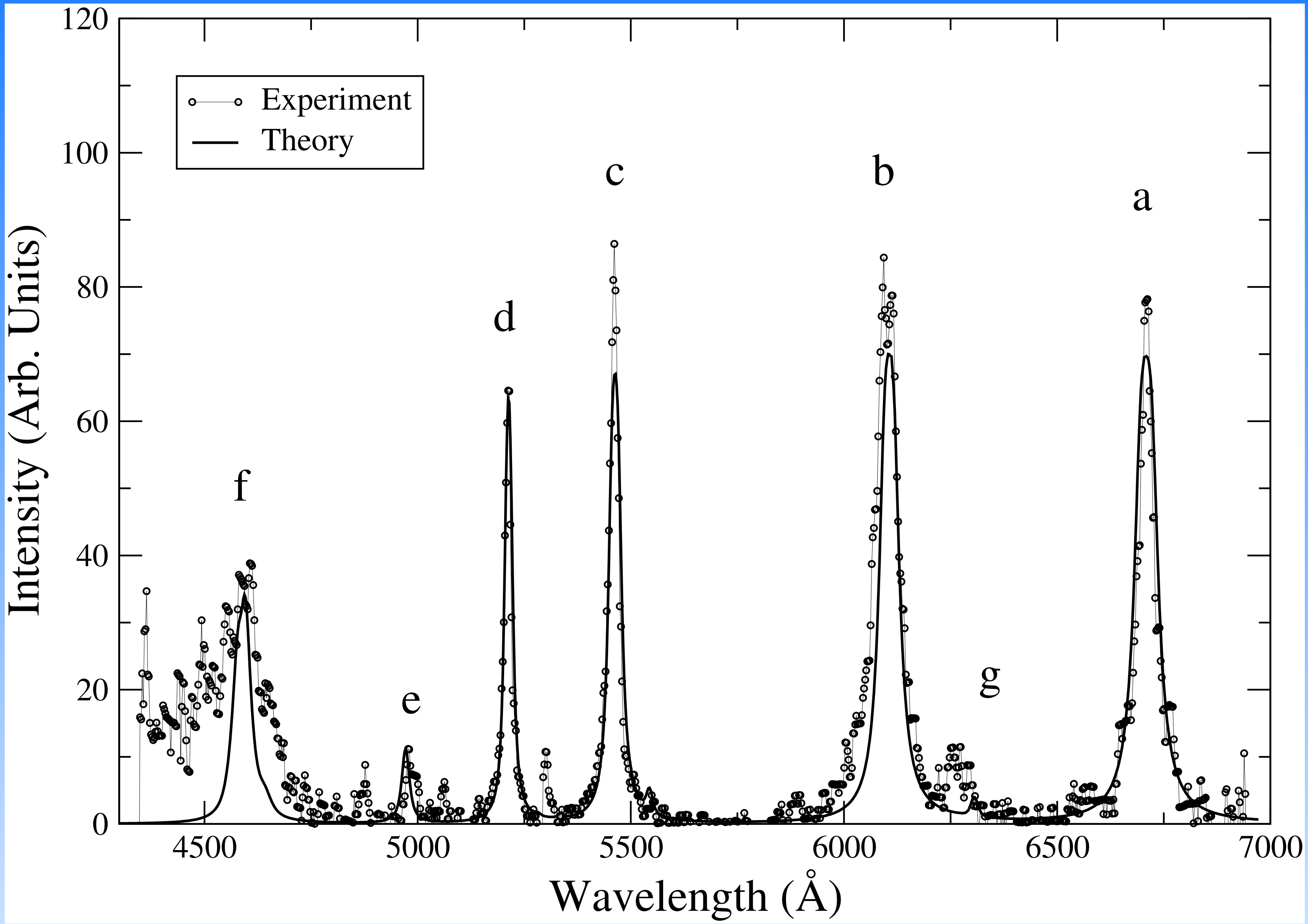


Figure 14: Experimental lineout at x=28.8μm and t=30 nsec with predicted T$_e$=0.8 eV and N$_a$=1.0x10^{17} cm^{-3} uniform synthetic spectra.

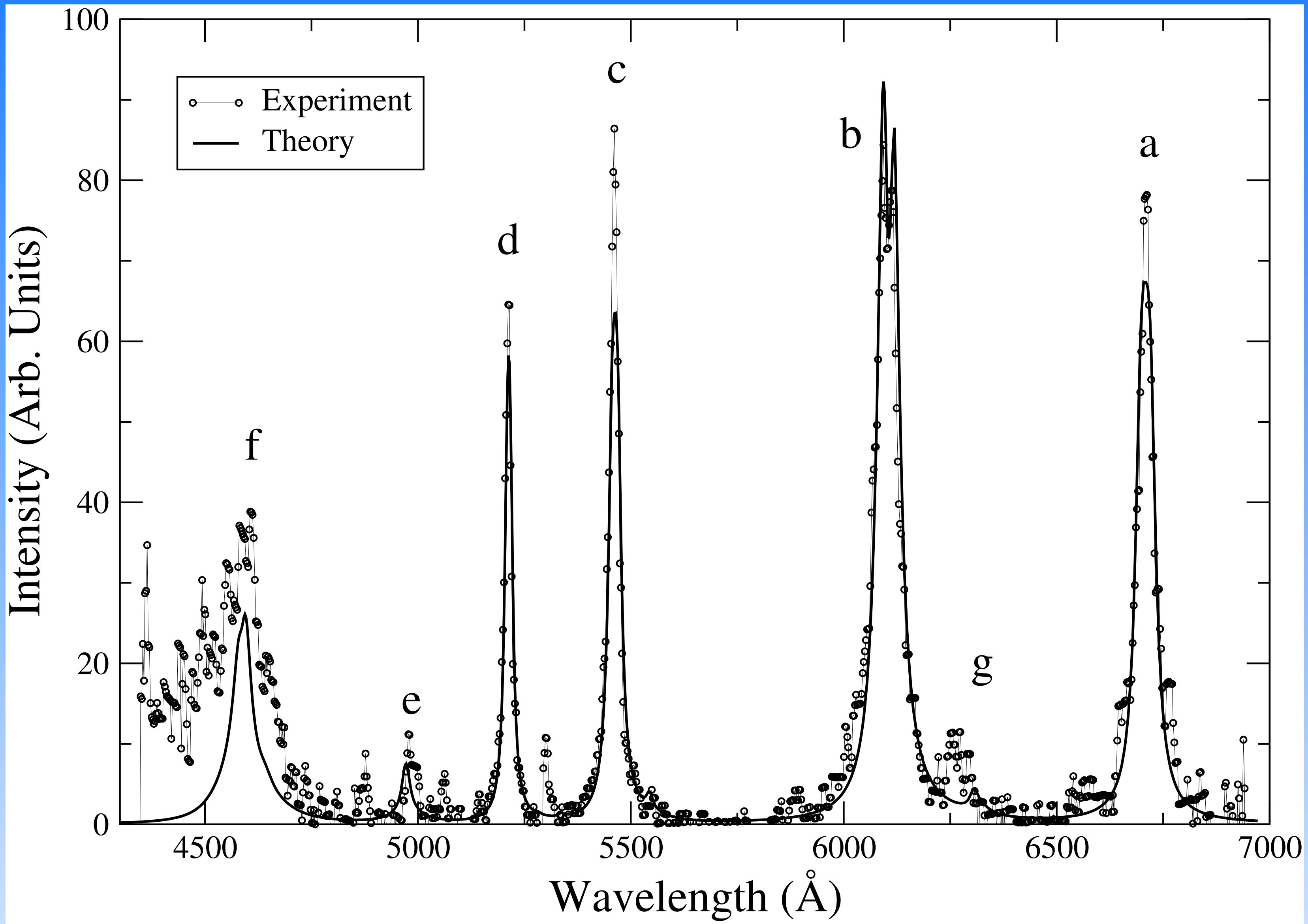


Figure 15: Experimental lineout at x=28.8μm and t=30 nsec with predicted T$_e$=[0.7, 1.8, 1.8, 0.7] eV and N$_a$=[1.0, 2.0, 2.0, 1.0] x10^{17} cm^{-3} 4-zone synthetic spectra.

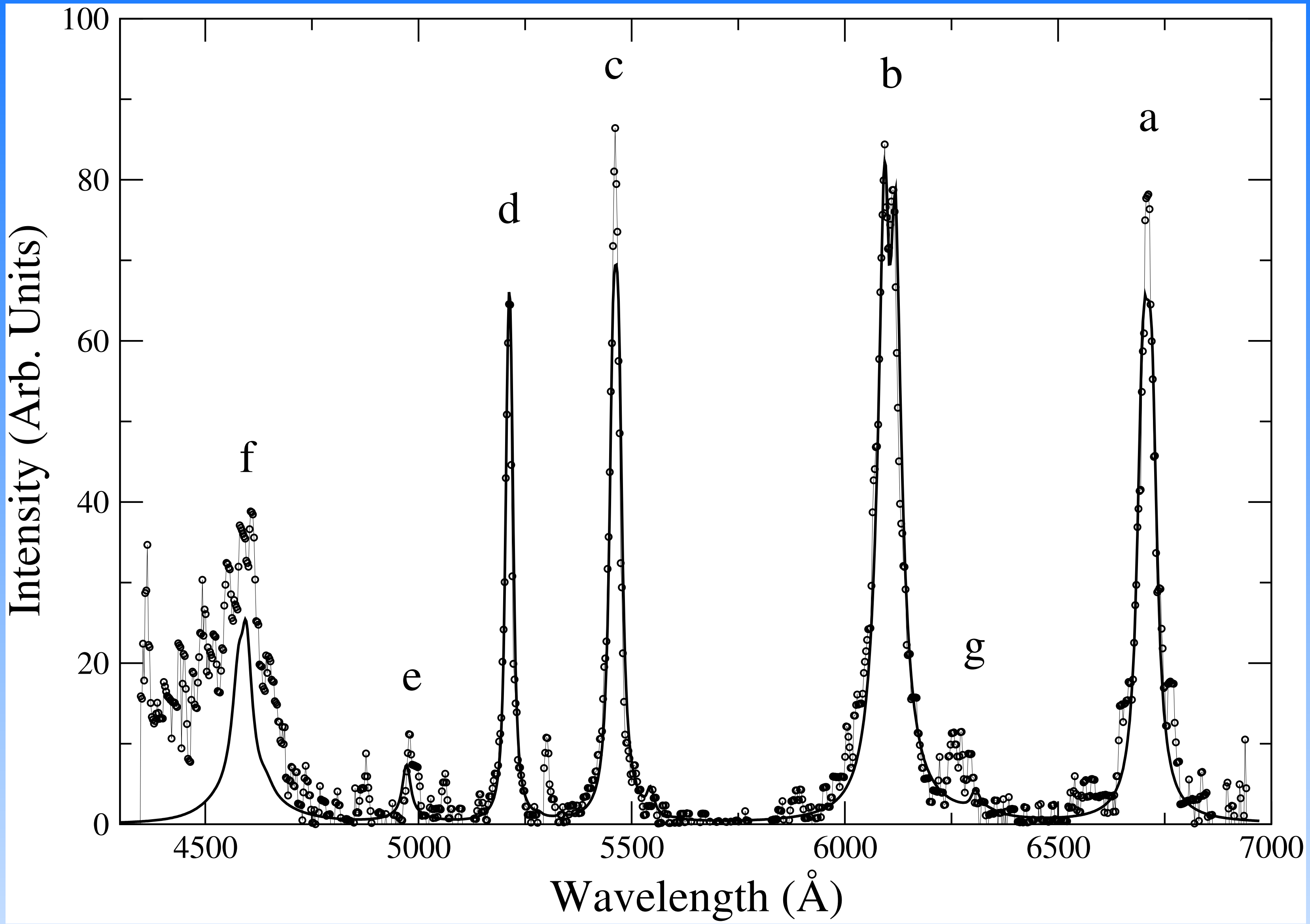


Figure 16: Experimental lineout at x=28.8μm and t=30 nsec with predicted T_e=[0.7, 0.8, 2.3, 2.3, 0.8, 0.7] eV and N_a=[1.0, 1.0, 2.5, 2.5, 1.0, 1.0] x10^{17} cm^{-3} 6-zone synthetic spectra.

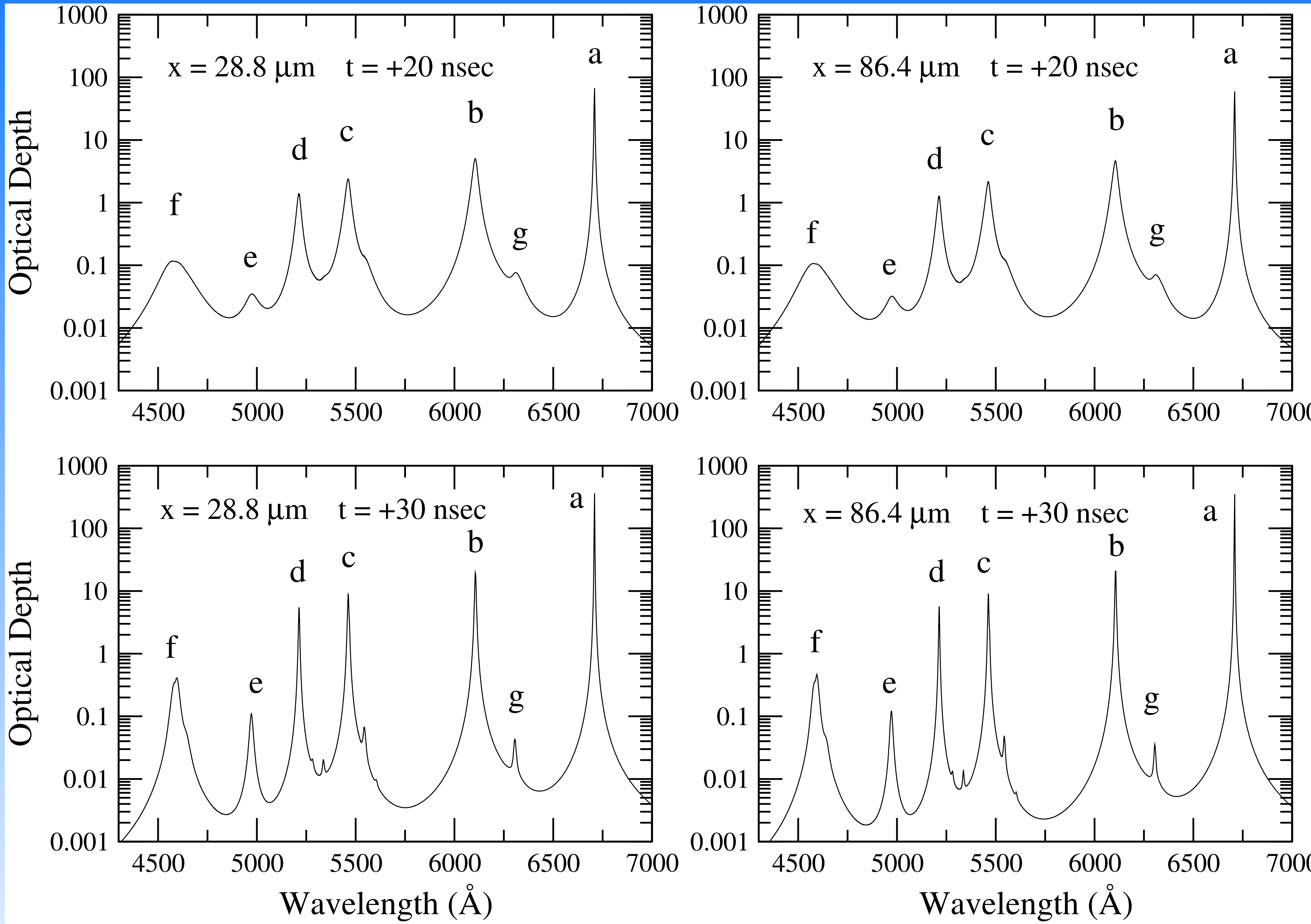


Figure 17: Optical depths for all four 6-zone synthetic spectra.

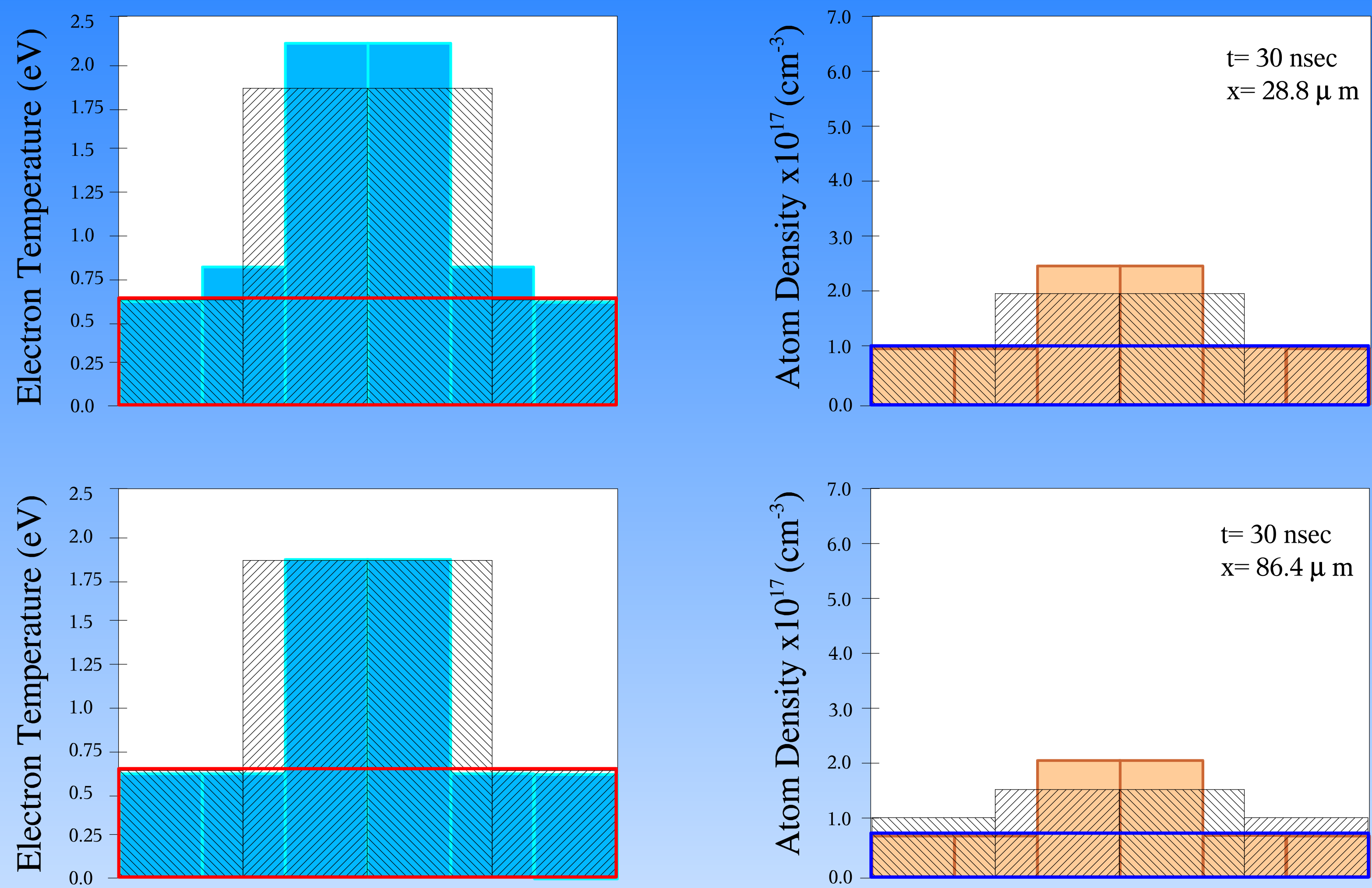
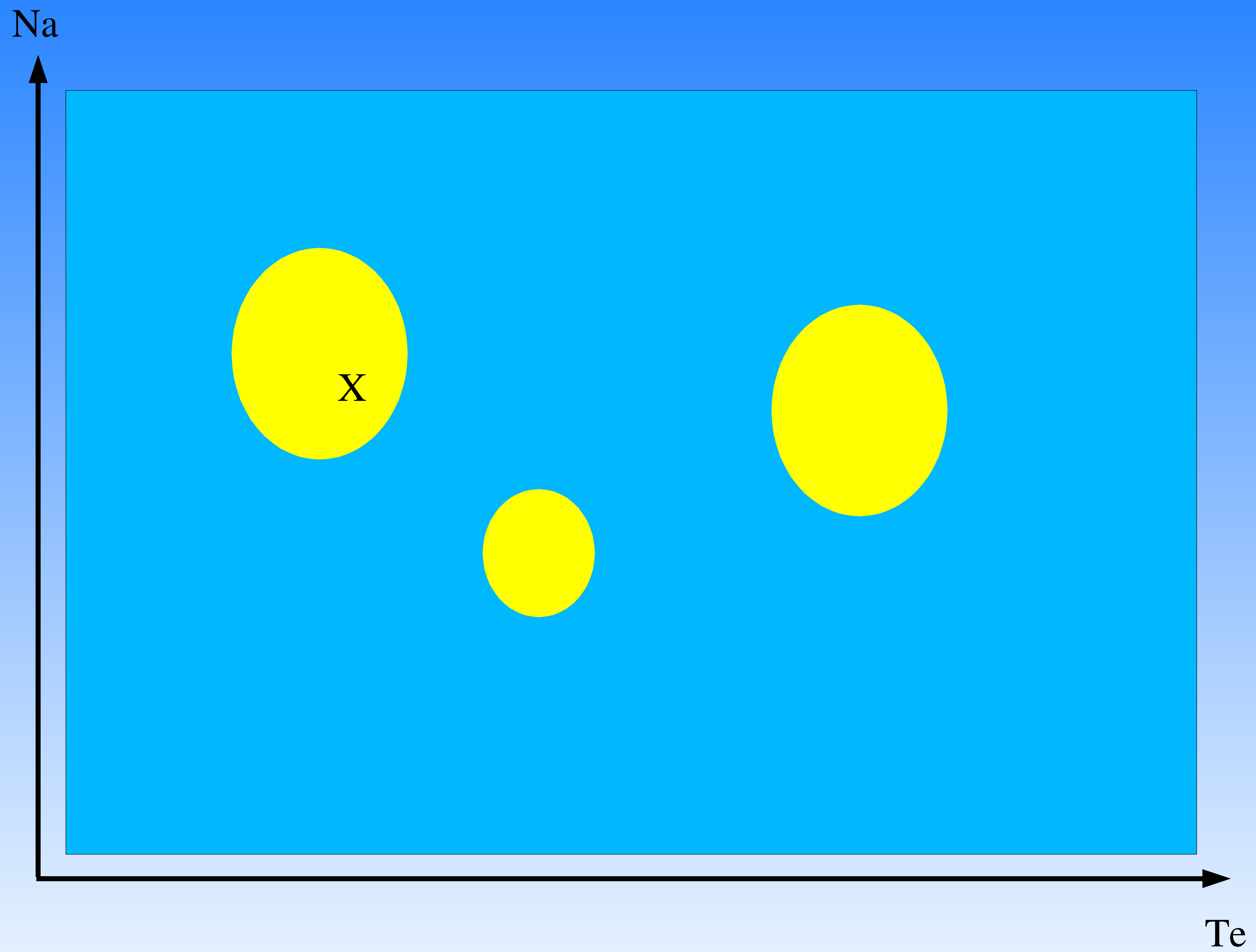


Figure 18: Uniform, 4 and 6 zone T_e and N_a profile results for t= +30 nsec synthetic spectra.

Summary & Insight

Concerning spectra whose features have a larger variation in optical depth

- REVERSE PROBLEM - Large optical depths reduces the sensitivity of the spectra and leads to ambiguity in plasma characterization.
- We see multiple acceptable solution domains.
- Solution space contains many plateaus - standard search techniques begin to fail.
- What does the best fit mean? What do the other plateaus mean?
- Brute force searches to establish this pattern.
- New search approach - mesh refinement: coarse to fine.


Na
X
Te

Coupled electron and atomic kinetics through the solution of the Boltzmann equation for generating time-dependent X-ray spectra

M. E. Sherrill, G. Csanak, J. Abdallah, Jr.

Theoretical Division, Los Alamos National Laboratory

E. S. Dodd

Applied Physics Division, Los Alamos National Laboratory

Y. Fukuda, Y. Akahane, M.Aoyama, N. Inoue, H. Ueda, K. Yamakawa

Advanced Photon Research Center, Japan Atomic Energy Research Institute Kyoto, Japan

A.Ya. Faenov, A. I. Magunov, T.A. Pikuz, I. Yu. Skobelev

Multicharged Ions Spectra Data Center of VNIIFTRI, Mendeleevo, Russia

Introduction

Problem Type

Spectral modeling of highly NON-EQUILIBRIUM PLASMAS (N_a=1.0×10^{20} cm^{-3}).

Time-dependent systems where the electron energy distribution function (EEDF) is NON-MAXWELLIAN.

Implementation

- **Coupled electron and atomic kinetics**

  Stepping: EK solution is propagated to time $t+1$ with level populations from the AK at t. Then the AK are propagated to time $t+1$ with the EEDF from $t+1$. The $t+1$ level populations from the AK model are provided to the EK.

- **Simultaneous?**

  Too slow!

Introduction

- **Common Examples**

  Electron beam driven X-ray lamps for lithography

  Intense short pulse laser driven plasmas

Electron Kinetics

- **Boltzmann Transport Equation**

$$\frac{\partial f(\vec{r},\vec{p},t)}{\partial t} + \frac{\vec{p}}{m} \bullet \nabla_r f(\vec{r},\vec{p},t) + F \bullet \nabla_p f(\vec{r},\vec{p},t) = \left(\frac{\partial f(\vec{r},\vec{p},t)}{\partial t}\right)_{coll} \tag{1}$$

- **0-D Boltzmann Equation** - (Non-relativistic)

$$\frac{\partial f(E,t)}{\partial t} = \left(\frac{\partial f(E,t)}{\partial t}\right)_{coll} \tag{2}$$

Formalism

Following J.Bretagne et al[a] we write the Boltzmann equation for the free-electron distribution function in the following form,

$$\frac{\partial}{\partial t} f(E,t) = \left(\frac{\partial f}{\partial t}\right)_{el(e-e)} + \left(\frac{\partial f}{\partial t}\right)_{in} + A(t)S(E_p, E) \tag{3}$$

where

$$\left(\frac{\partial f}{\partial t}\right)_{in} = K_{ion} + K_{3BR} + K_{exc} + K_{de-exc} \,. \tag{4}$$

---

[a] J. Phys. D, **14** pp. 1225-39

Formalism

Ionization Contribution

$$K_{ion}(E) = \sum_{iljm} K^{ion}_{iljm}(E) \tag{5}$$

where $K^{ion}_{iljm}(E)$ is the contribution to $K_{ion}(E)$ from the

$$e^- + (il) \longrightarrow e^- + e^- + (jm) \tag{6}$$

ionization process where (il) indices refer to the initial ion and its quantum state and (jm) to the final ion and its quantum state, respectively. $K^{ion}_{iljm}(E)$ can be given in the form,

$$\begin{aligned} K^{ion}_{iljm}(E) = N_{il}\Big[& \int_{E+E_0}^{2E+E_0} v'\sigma_{iljm}(E', E'-E_0-E) f(E',t)\, dE' + \\ & + \int_{2E+E_0}^{E_{max}} v'\sigma_{iljm}(E', E) f(E',t)\, dE' + \\ & - v f(E,t) \int_0^{(E-E_0)/2} \sigma_{iljm}(E, E'')\, dE'' \Big] \end{aligned} \tag{7}$$

Formalism

Ionization Contribution: The total ionization cross section for a process will be denoted by $Q_{iljm}(E)$ (in units of cm^2) and thus it can be obtained from $\sigma_{iljm}(E, E'')$ via the formula,

$$Q_{iljm}(E) = \int_0^{(E-E_0)/2} \sigma_{iljm}(E, E'')\, dE'' \tag{8}$$

In the present work we assumed that $\sigma_{iljm}(E', E)$ can be written in the following form,

$$\sigma_{iljm}(E', E_1) = Q_{iljm}(E')\, \Omega(E', E_1) \tag{9}$$

where $\Omega(E, E_1)$ is described as follows (Astrophys. J, 381 pp 597-600 1991)

$$\begin{aligned}\Omega(E, E_1) &= \frac{1}{(E - E_0)(E^2 + aE_0^2)} \\ &\quad \times \left[2(a+1)E_0^2 + \frac{160(E + E_0)\left(E_1 - \frac{1}{2}(E - E_0)^4\right)}{(E - E_0)^3} \right]\end{aligned} \tag{10}$$

and is normalized, that

$$\int_0^{(E-E_0)/2} \Omega(E, E_1)\, dE_1 = 1 \tag{11}$$

Numerical Solution of the Boltzmann Equation

Bin representation

The EEDF can be represented a series of constant values.

$$f(E,t) = f(E_u) \text{ for } E_u - w_u/2 \leq E \leq E_u + w_u/2 \tag{12}$$

where E_u is the midpoint of the uth bin, w_u is the width of the uth bin and N is the total number of bins.

Final Form of the Boltzmann Equation

$$\frac{d}{dt} f(E_u, t) = \sum_{v=1}^{N} M_{u,v}(t) f(E_v, t) \tag{13}$$

where the $M_{u,v}(t)$ matrix contains all the collisional contributions.

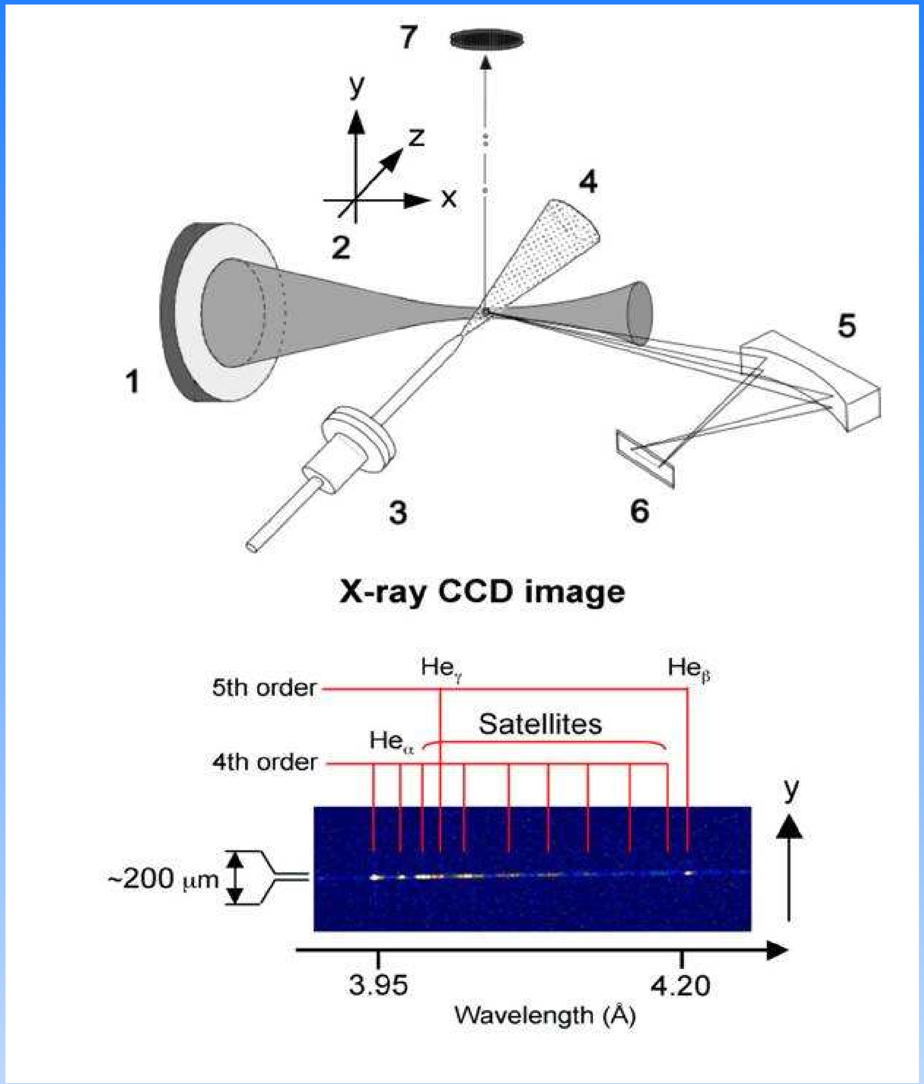


Figure 19: Laser: 200mJ, I=1.0e19 W/cm^2, 30 fs, Ti:sapphire laser. Prepulse: 1 ns duration 10^5 contrast ratio. Target: 1μm cluster target. FSSR:focusing spectrometers with spatial resolution, $\lambda/\delta\lambda=$1667 . Spectrum: 600 shots per spectrum.

Modeling the Experiment

- **Laser-Target Interaction**[a]

  Target 100K clusters in beam volume

  Beam more intense in the center, less on perimeter

- **Laser-Cluster Interaction**

  1 - 10 billion atoms

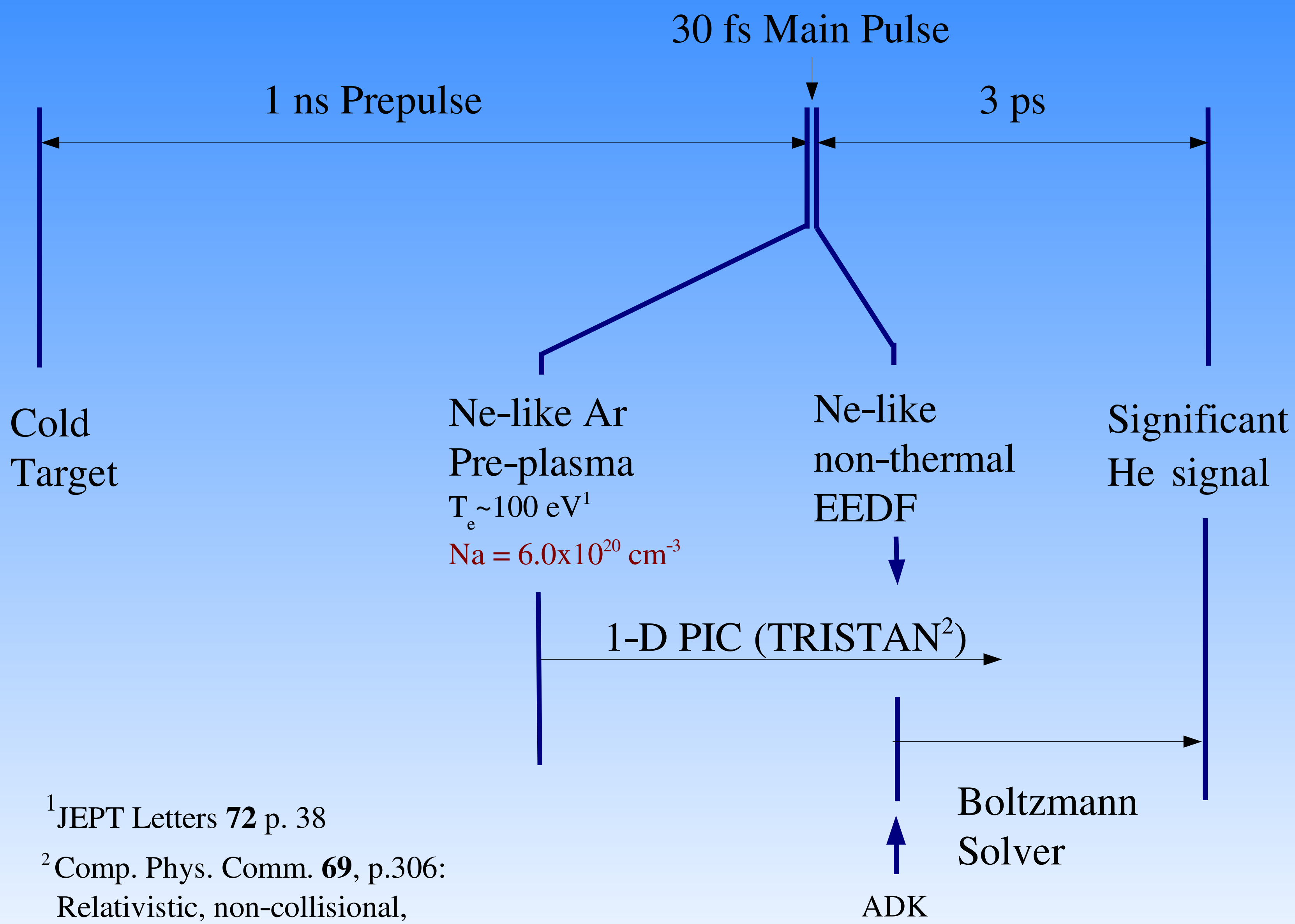
  1 μm cluster vs 10 μm main pulse

  3D - PROBLEM

---

[a] JEPT Letters, **78** pp. 115-118

Plasma Evolution



[1] JEPT Letters **72** p. 38

[2] Comp. Phys. Comm. **69**, p.306: Relativistic, non-collisional, non-ionizing.

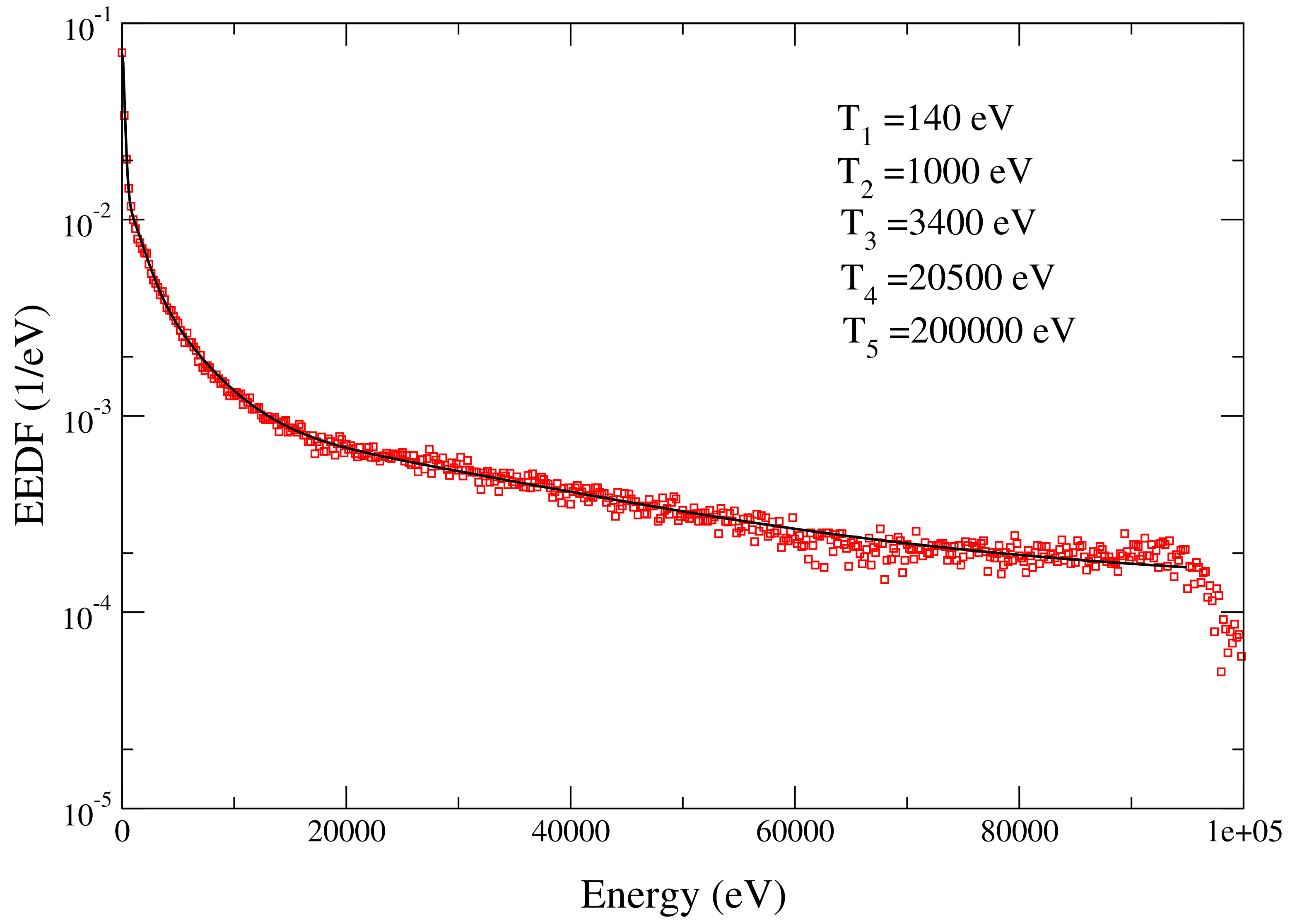
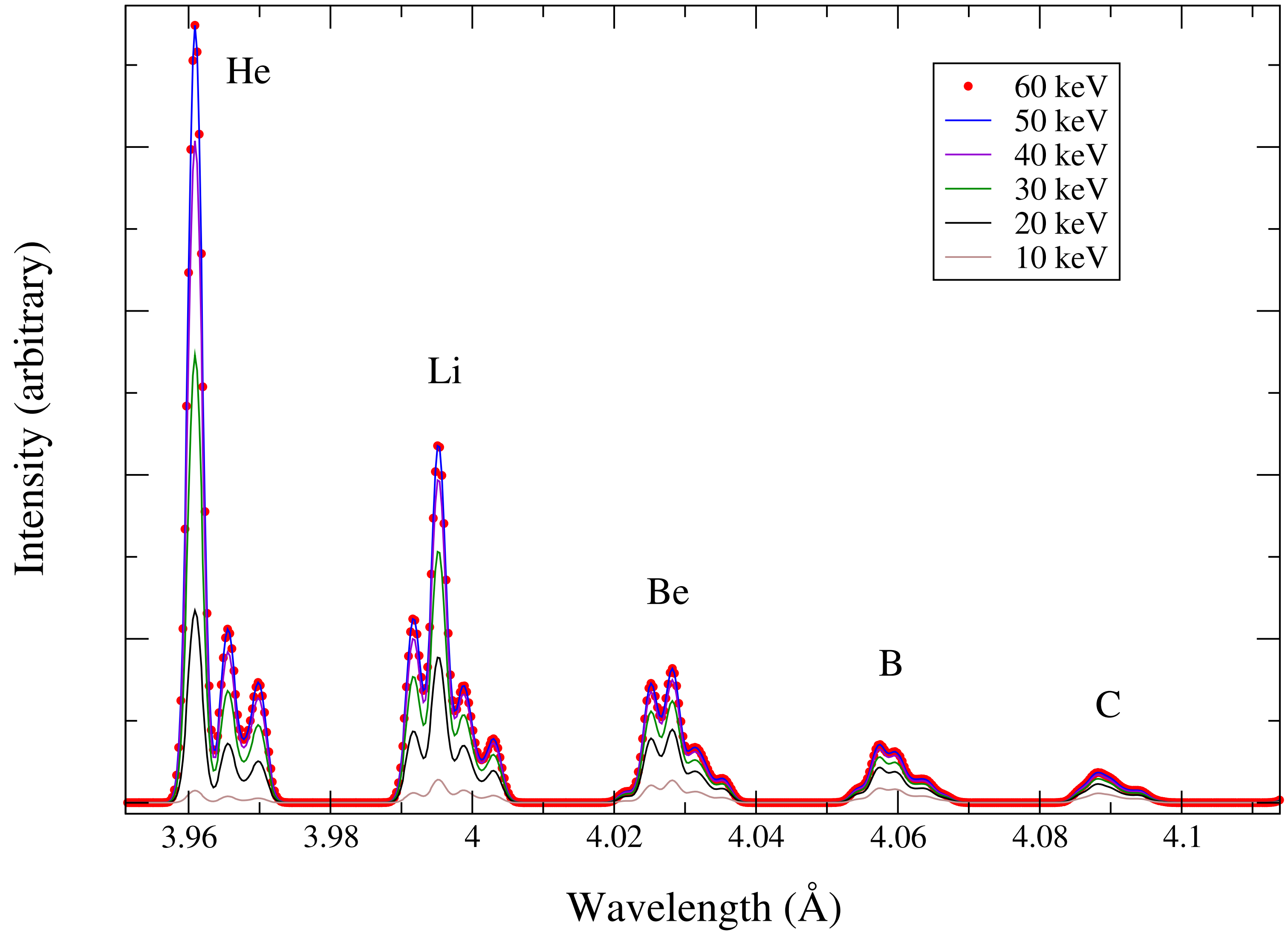
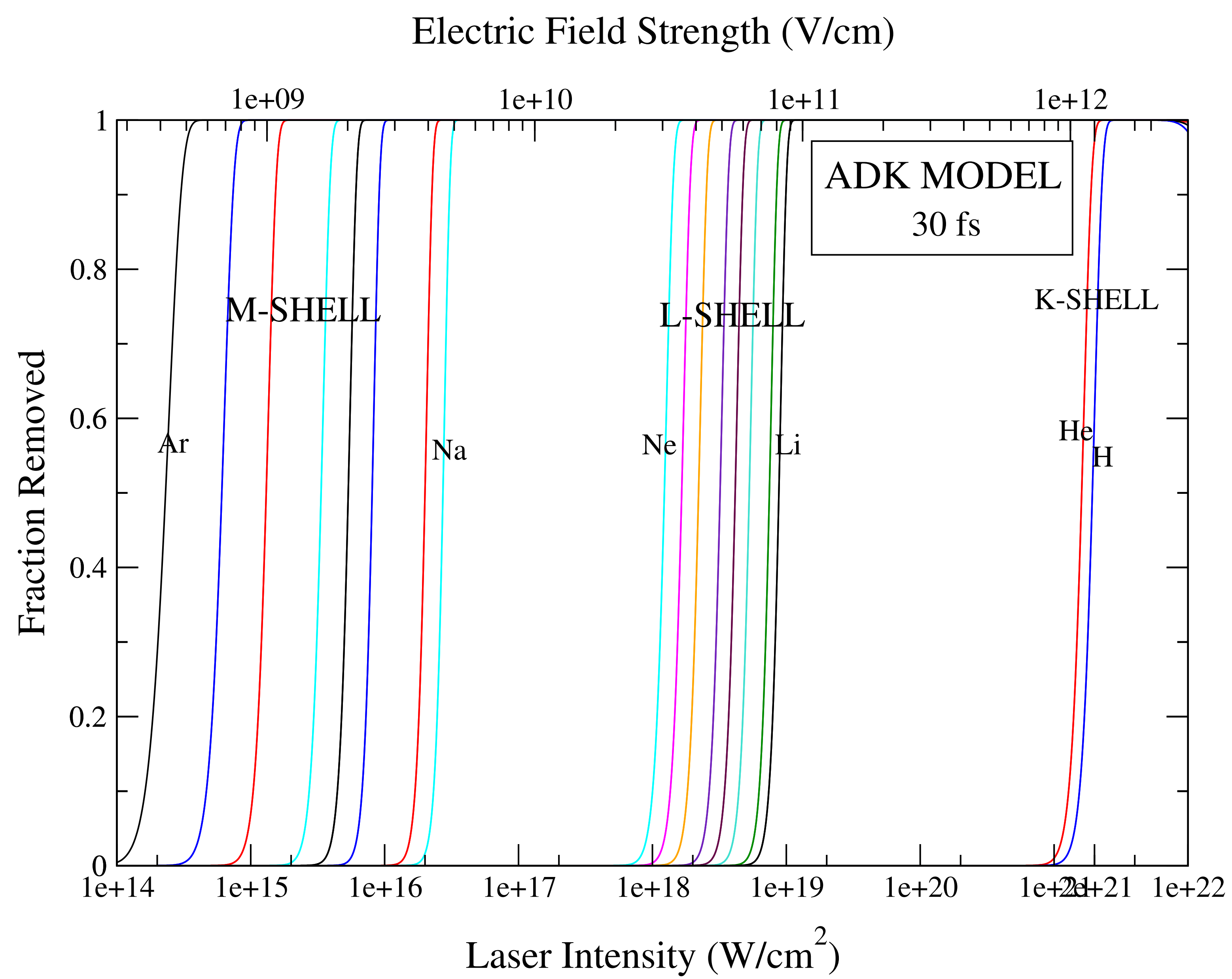


Figure 20: PIC produced EEDF at the end of the laser pulse.

EEDF Effects on Spectra

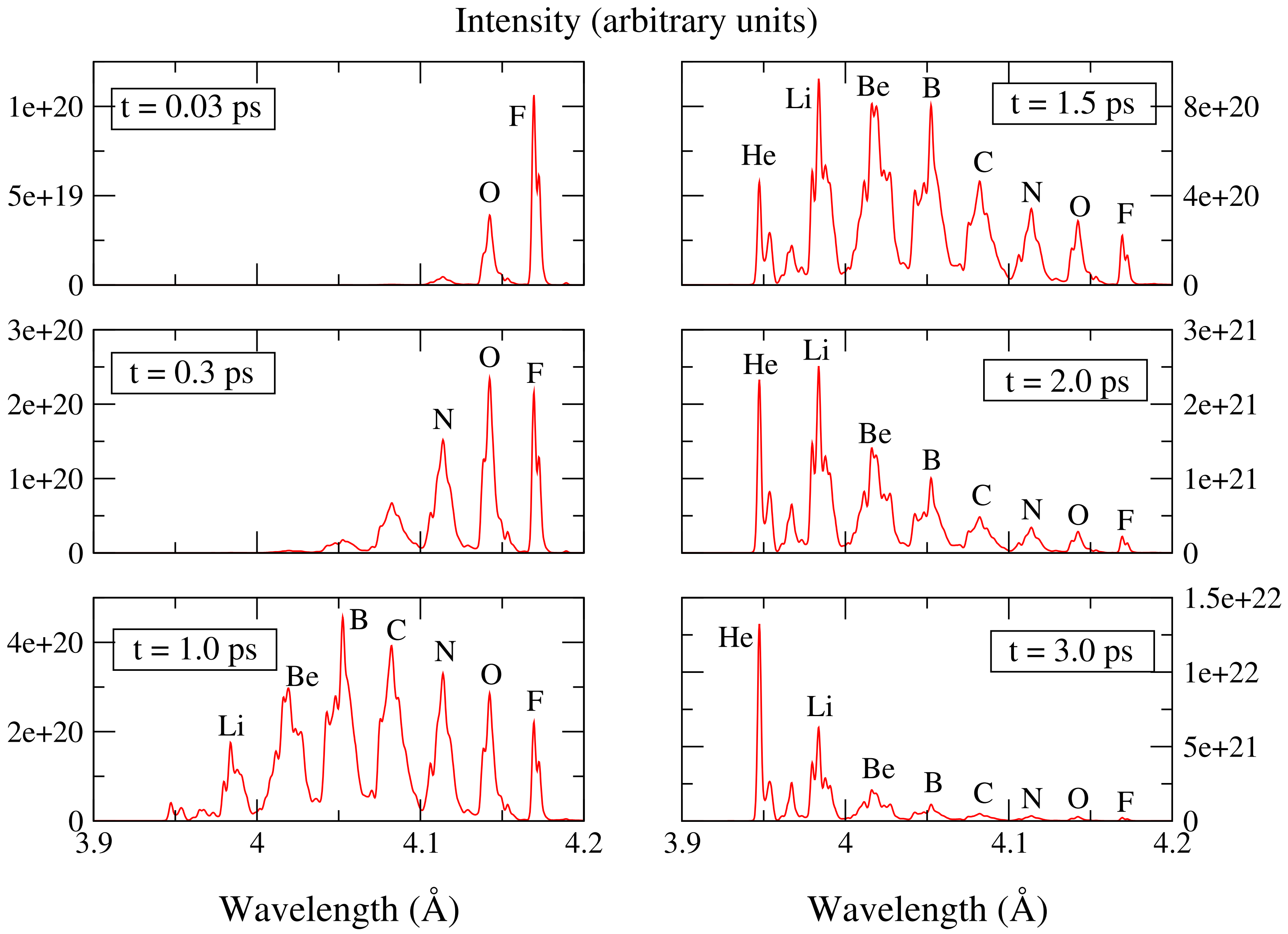



Electric Field Strength (V/cm)
1e+09
1e+10
1e+11
1e+12
ADK MODEL
30 fs
M-SHELL
L-SHELL
K-SHELL
Ar
Na
Ne
Li
He
H
Fraction Removed
0
0.2
0.4
0.6
0.8
1
1e+14
1e+15
1e+16
1e+17
1e+18
1e+19
1e+20
1e+21
1e+22
Laser Intensity (W/cm2)

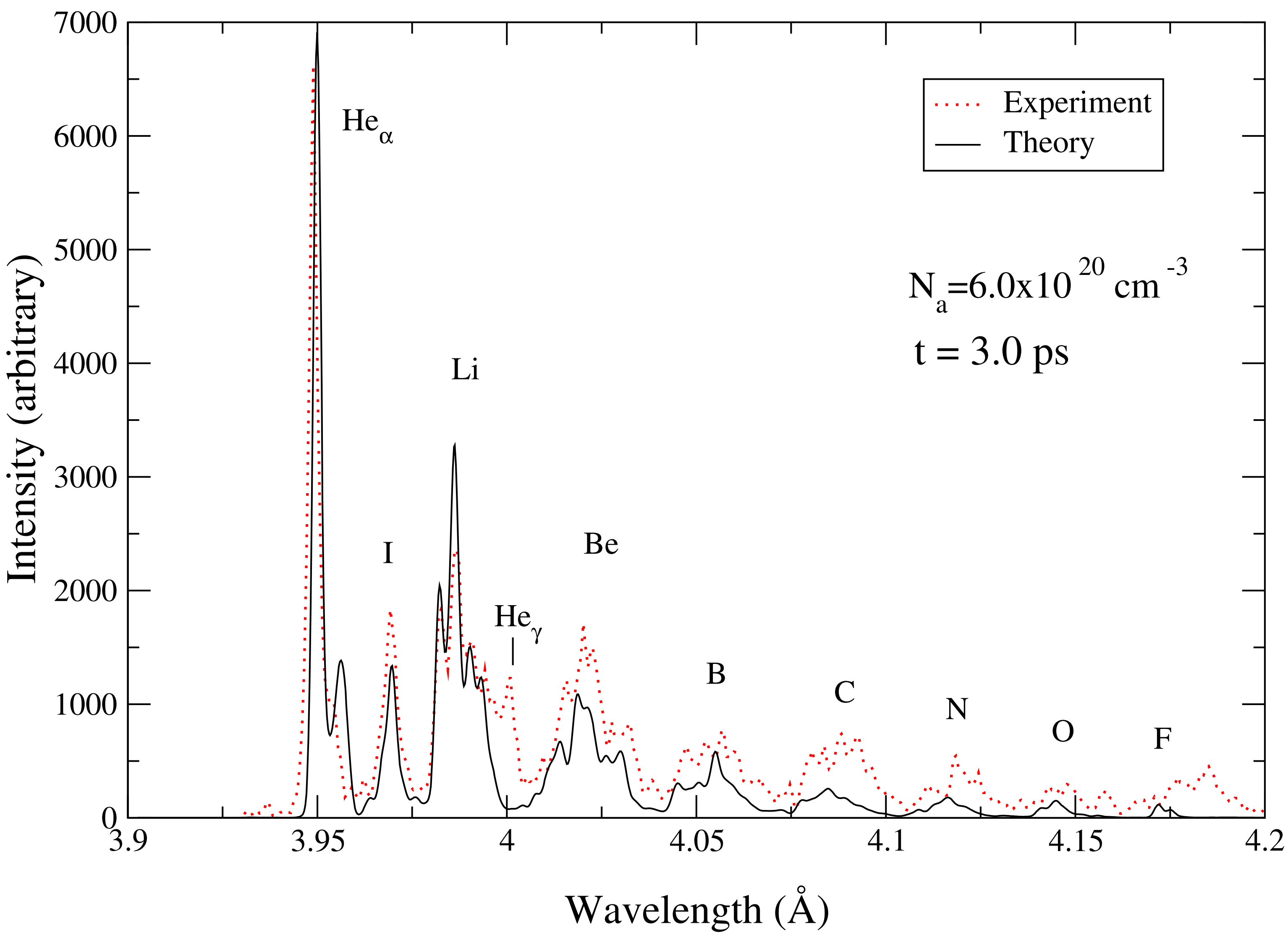
Plasma Evolution

- CATS - Cowan’s Atomic Structure Code
- ACE - Collisional Excitation
- GIPPER - Collisional Ionization
- 3000 fine structure levels, Ne-like - H-like ionization stages.
- Configurations were truncated to principle number n=3.
- All possible n=1,n=2 X-ray transitions within the n=3 manifold.

Time Evolution of Integrated He_α Spectra



Comparison of Time Integrated He_α Spectrum



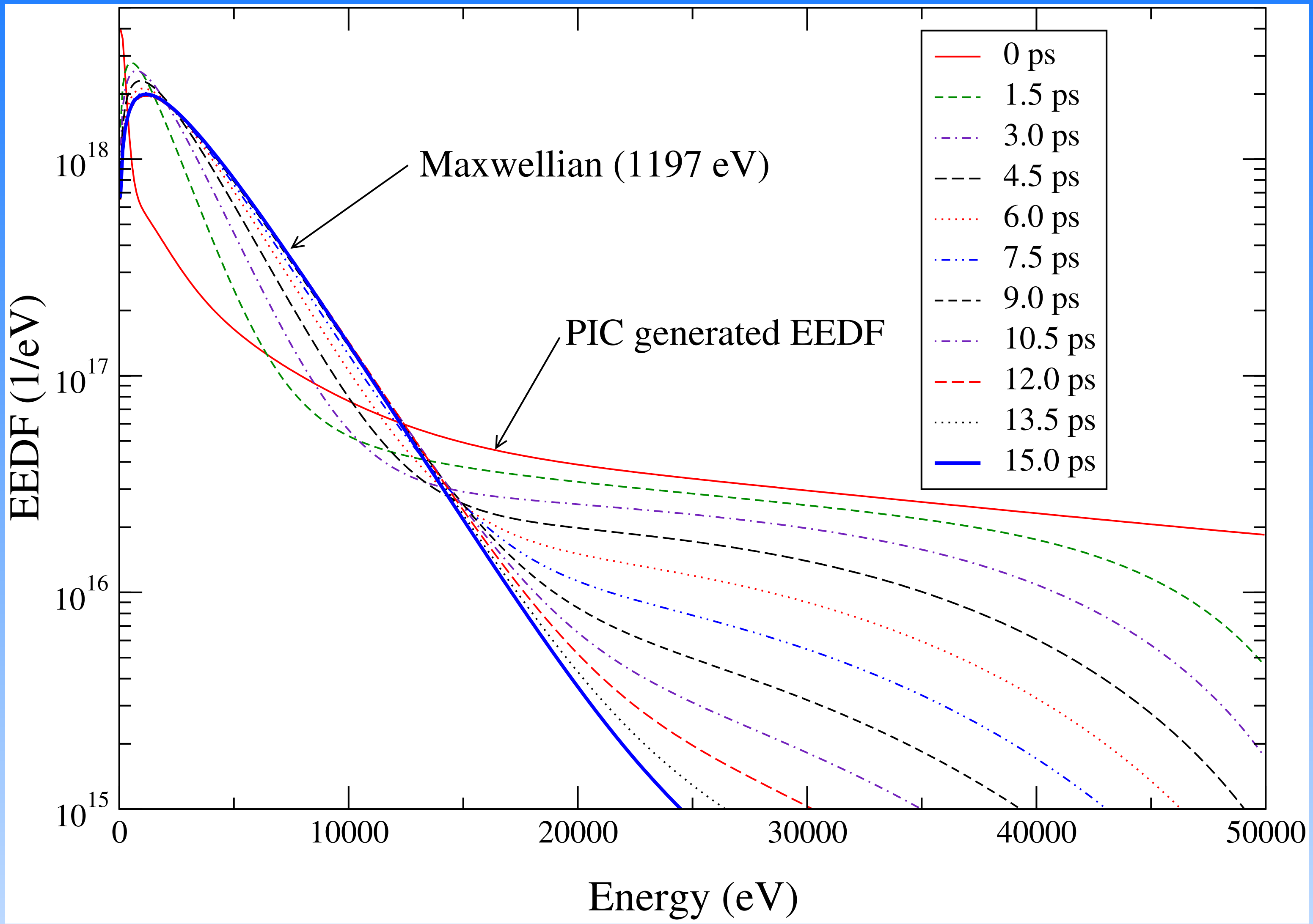


Figure 21: The time history of the EEDF, for times after the end of the laser pulse, computed by the Boltzmann solver for a constant atomic number density (N$_a$=6.0×10^{20} cm^{-3}). The system evolves into a Maxwellian (1197 eV) in approximately t=12.0 ps.

Conclusion/Summary

- Used 1-D particle-in-cell code to provide the initial EEDF of the Boltzmann solver.
- EEDF after 3 ps was far from equilibrated (13 ps)
- 50keV - 60keV of the EEDF is required to obtain a consistent spectra.
- From this model we predicted 6.0x10^{20} cm^{-3} represented an average density for the highly ionized contributions to the He$_\alpha$ spectrum.

Future Work

- Complete 3-body recombination
- Include autoionziation and dielectronic recombination
- Consider a non-constant volume approximation.
- Pursue comparisons between collisional PIC and Boltzmann Solver solutions.

Insight

Plasma and Atomic Physics

- Detailed description of the state of the plasma - The importance of plasma physics?
- Electron and Ion interaction?
- Emission spectra is of the order of the material expansion time - Coulomb explosion?
- Higher Z material and relativistic cross section - Plasma physicist assumption?