



HAL
open science

MERLIN, a RT-LTE software supporting the LIBS diagnostic: application to H-isotopes measurements

Aurélien Favre, Morgan Lesage, Vincent Morel, Arnaud Bultel, Pascal Boubert

► To cite this version:

Aurélien Favre, Morgan Lesage, Vincent Morel, Arnaud Bultel, Pascal Boubert. MERLIN, a RT-LTE software supporting the LIBS diagnostic: application to H-isotopes measurements. International Workshop on LIBS, Dec 2020, Szeged, Hungary. hal-03011810

HAL Id: hal-03011810

<https://hal.science/hal-03011810>

Submitted on 18 Nov 2020

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

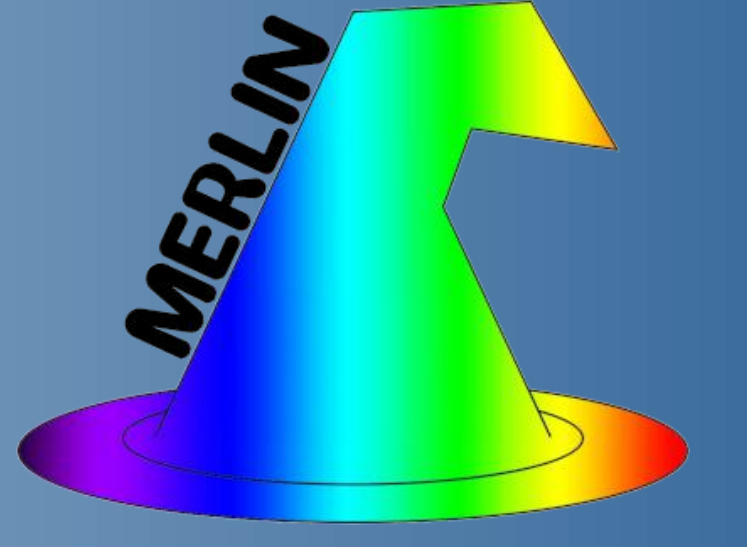


MERLIN, a RT-LTE software supporting the LIBS diagnostic: application to H-isotopes measurements

AURÉLIEN FAVRE[‡], Morgan LESAGE, VINCENT MOREL, ARNAUD BULTEL, PASCAL BOUBERT

CORIA, UMR CNRS 6614, Normandie Université, 76801 Saint-Étienne du Rouvray, FRANCE

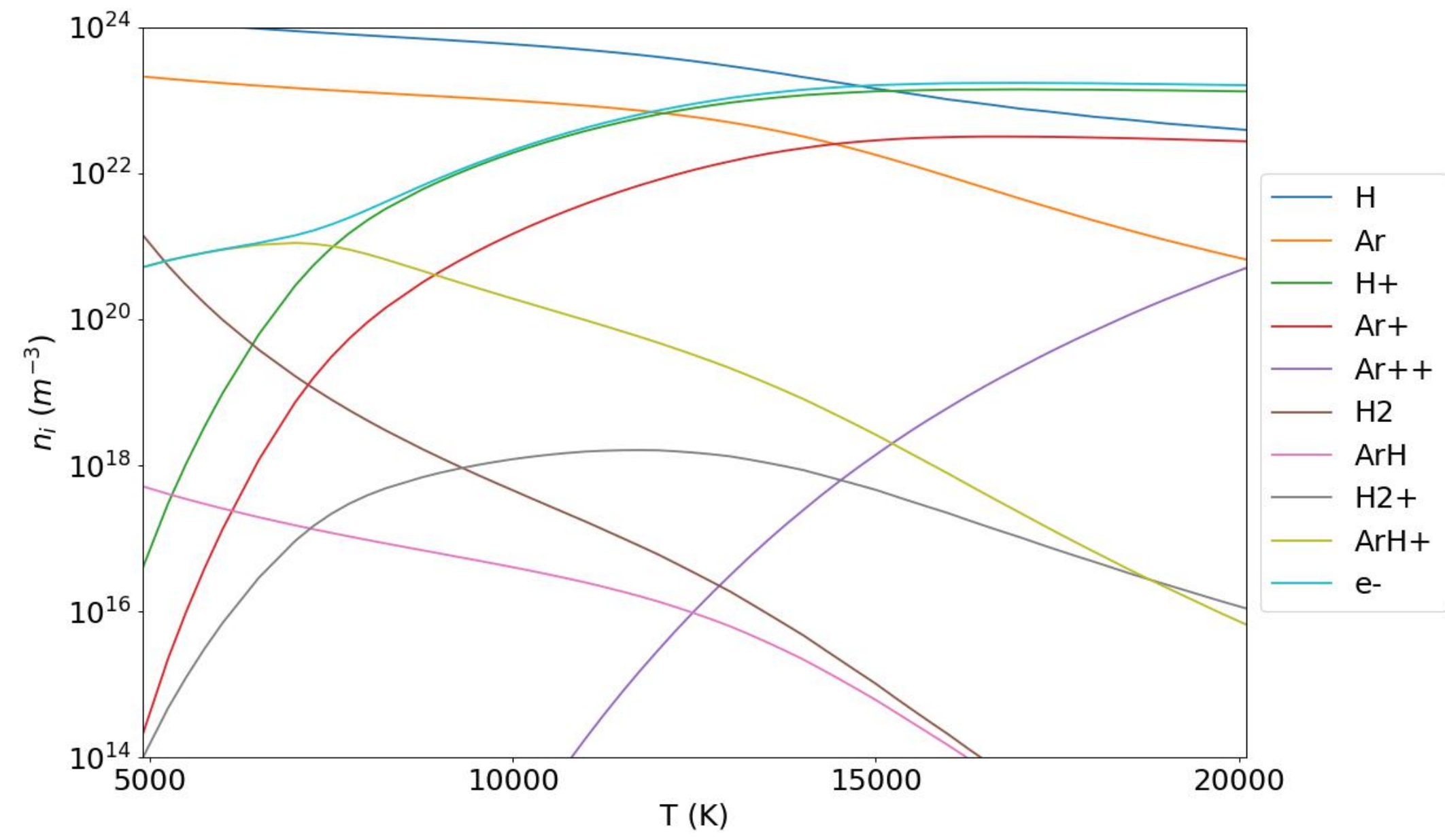
[‡] aurelien.favre@coria.fr



CONTEXT & OBJECTIVES

LIBS (Laser-Induced Breakdown Spectroscopy) enables the determination of targets composition using Optical Emission Spectroscopy (OES). An efficient way to derive an accurate composition is to iterate the computation of the emission spectrum with respect to the best fit of experimental observations.

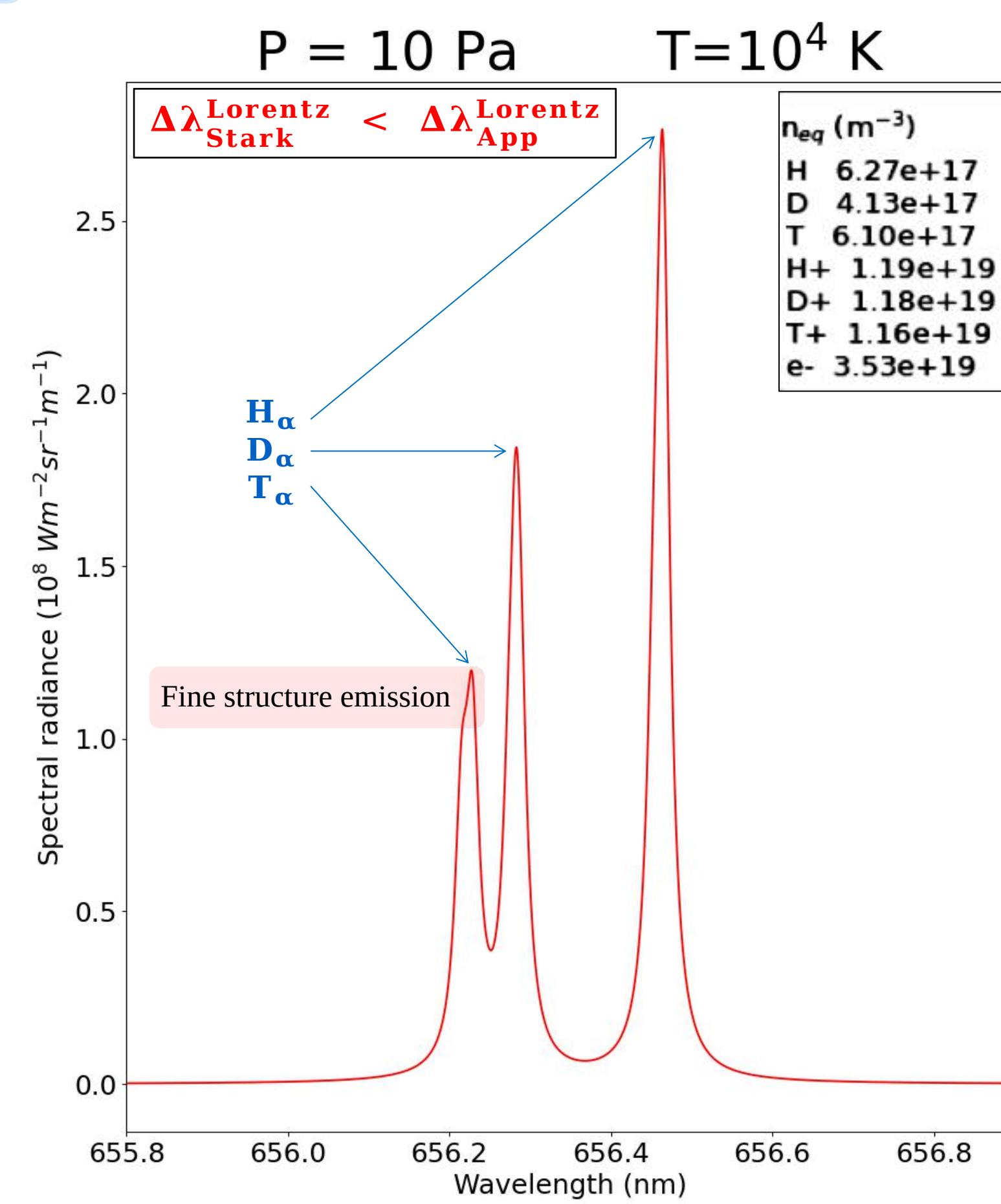
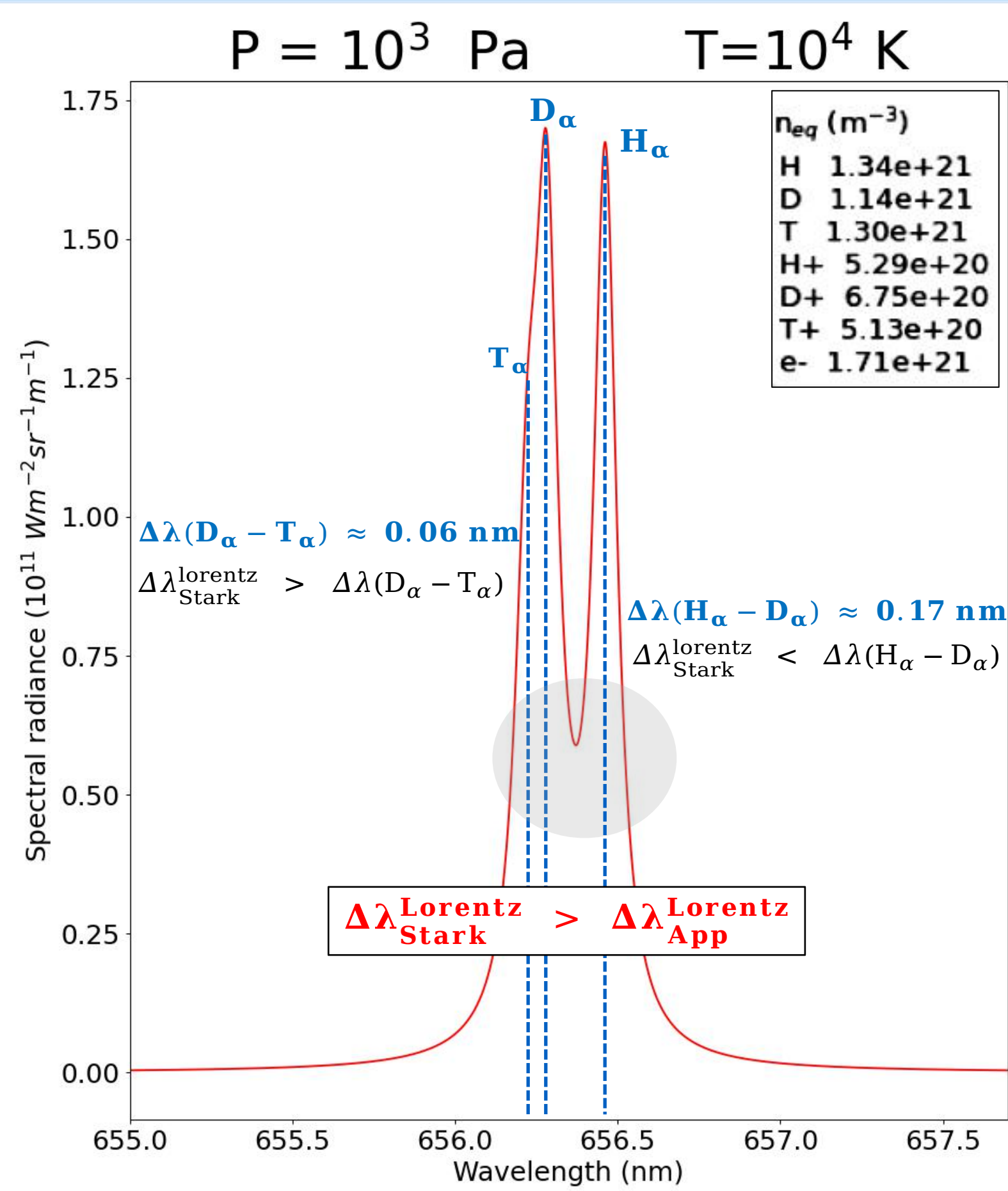
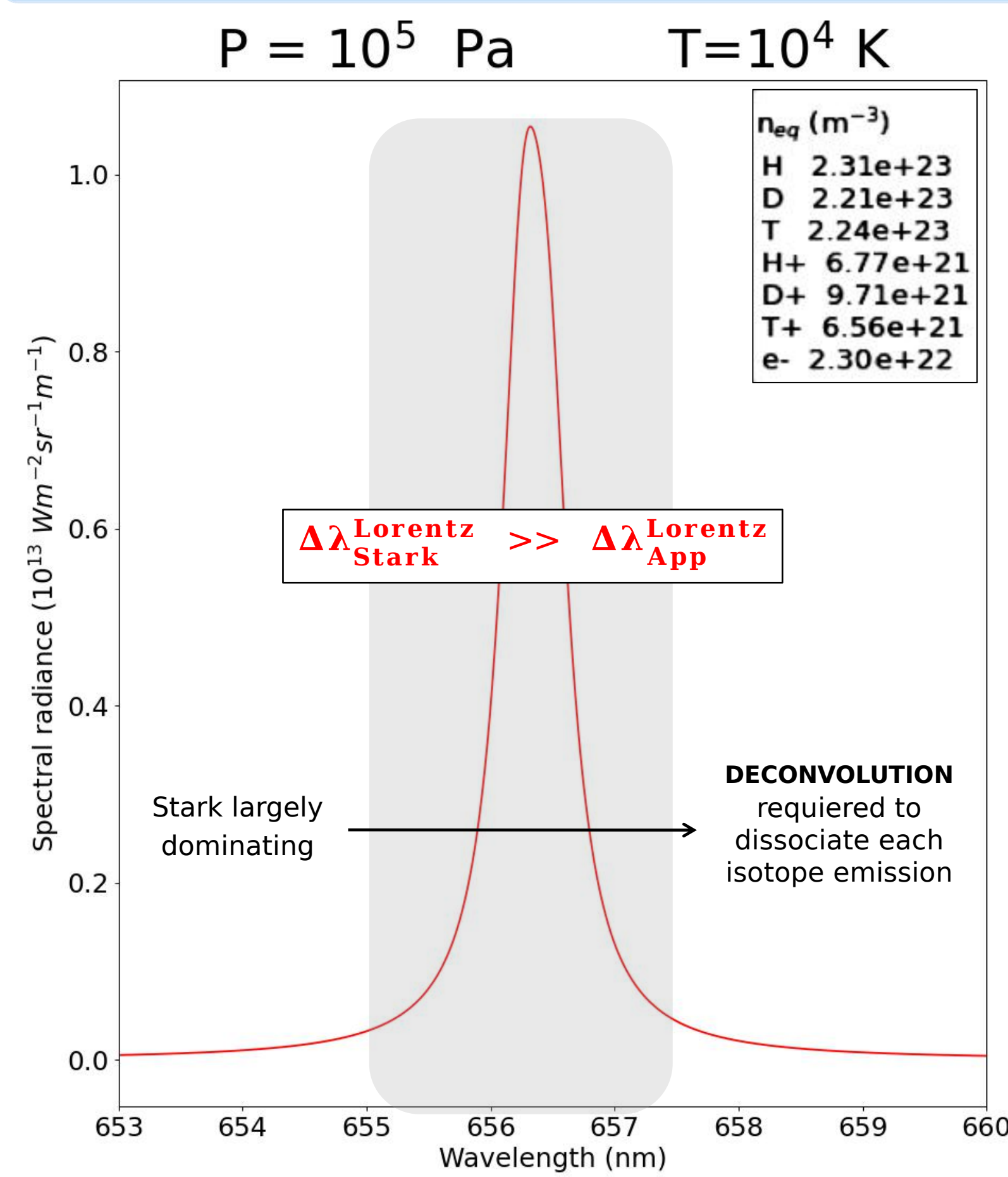
MERLIN (MultiElemental Radiative equiLilibrium emissioN) has been developed to ensure a computational support OES experiments. This suit is based on Radiative Transfer (RT) Local Thermal Equilibrium (LTE) solving for homogeneous emitting media. Ionizations and dissociations equations are dynamically generated and solved from elemental mole fractions. Spectral data are online requested to generate the emission spectrum of the mixture.



Example of equilibrium composition of a (0.25 Ar - 0.75 H₂) mixture versus temperature. MERLIN shows a satisfactory agreement with published results

Upper levels population density driven by a Boltzmann statistic then allow a computation of emission spectrum.

H, D & T stoichiometric mixture: isotopic resolution power



Balmer- α HDT LTE emission spectra for a 1 mm wide emitting plasma with the following broadening parameters :

$$\Delta\lambda_{VDW}^{Lorentz} = 0 \text{ nm}$$

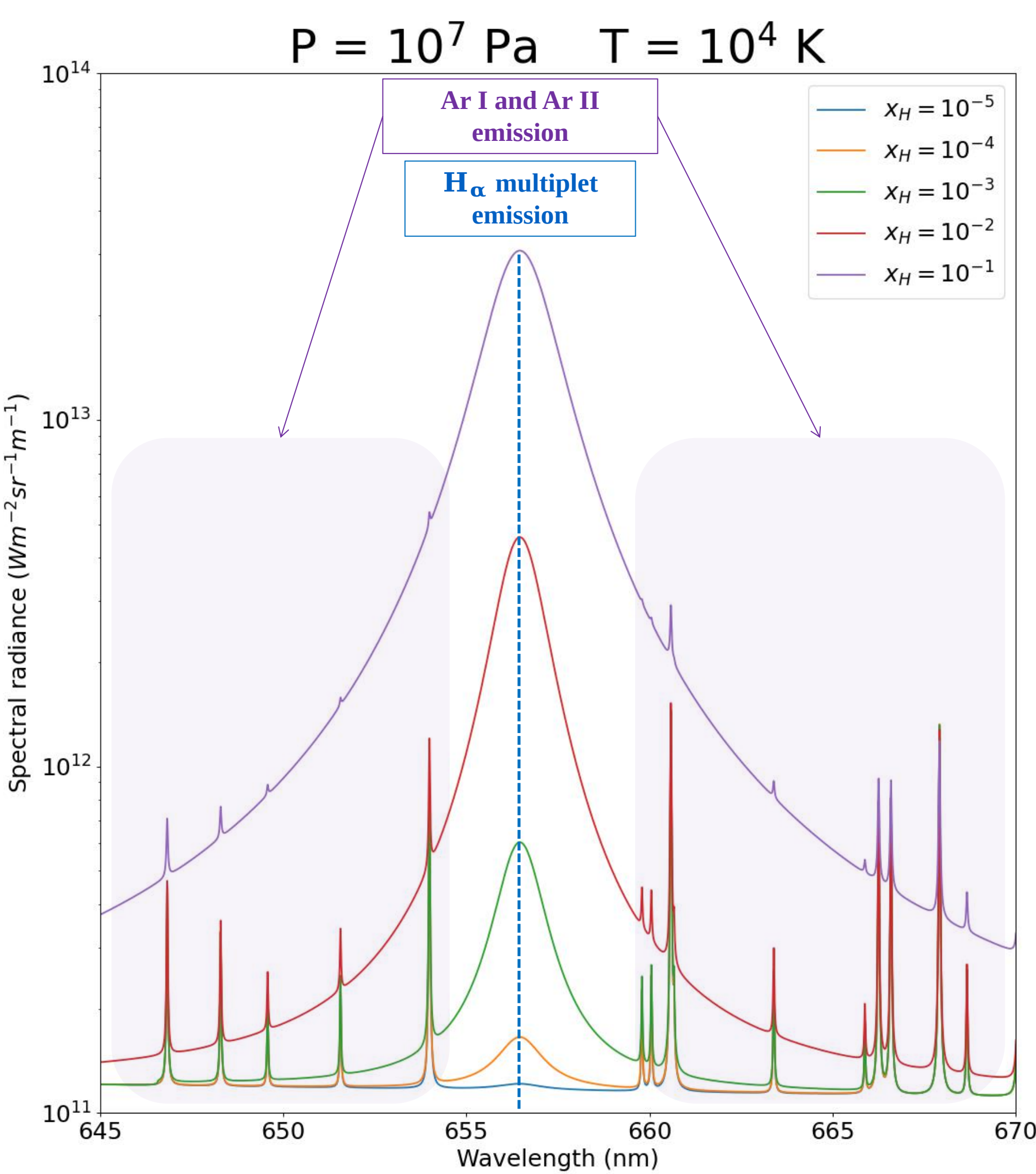
$$\Delta\lambda_{Doppler}^{Gauss} = 0 \text{ nm}$$

$$\Delta\lambda_{App}^{Lorentz} = 0.0145 \text{ nm}$$

$$\Delta\lambda_{H-Stark}^{Lorentz} = 1.098 \left(\frac{n_e}{10^{23}}\right)^{0.67965} \text{ nm}$$

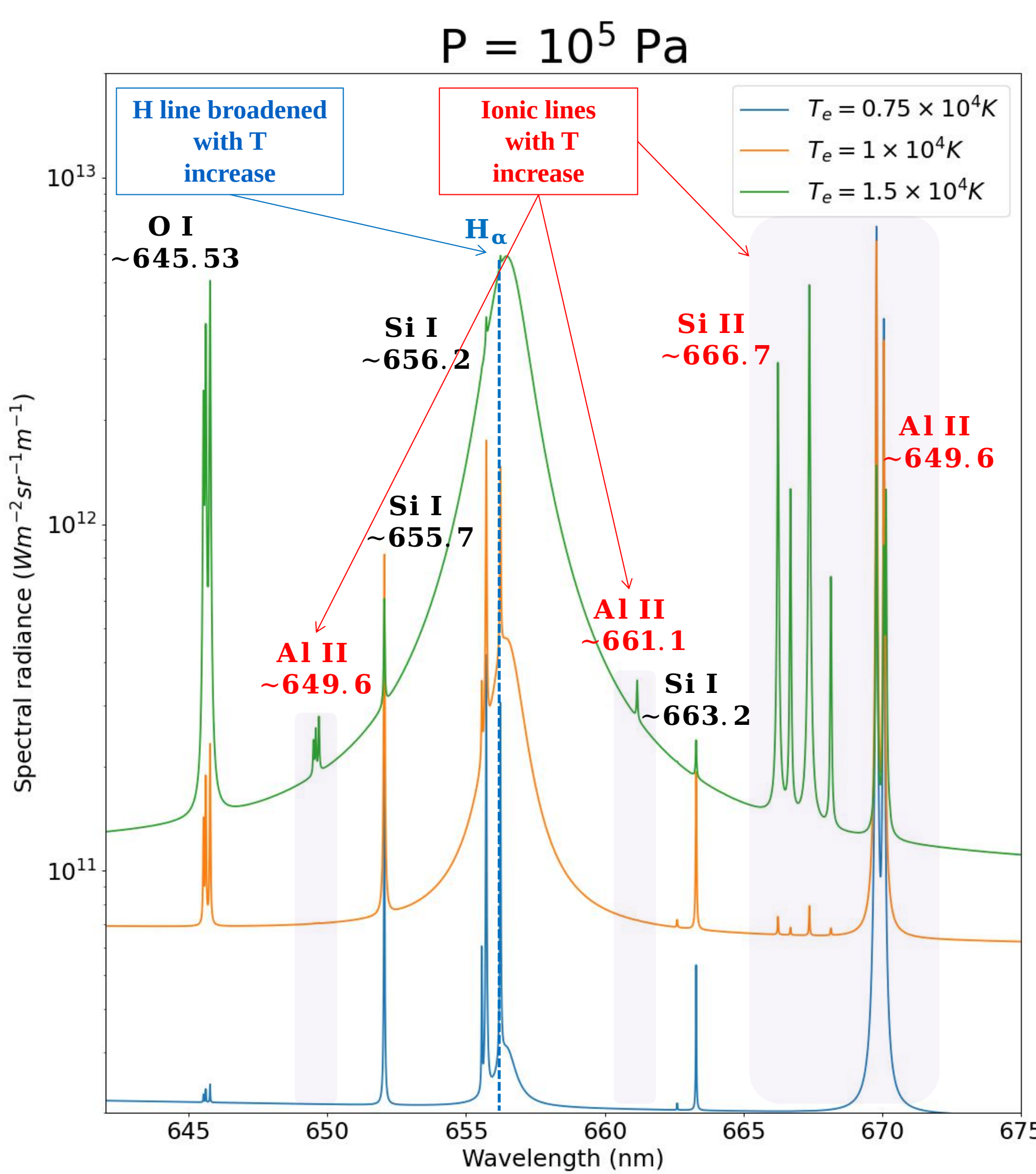
- Higher pressure leads to a significant Stark broadening which is n_e dependant.
- A competition between apparatus and Stark broadenings rules the resolution power.
- Element A_{ki} ratios verifies equilibrium verifies at low densities.

H mole fraction in Ar-H₂ mixture



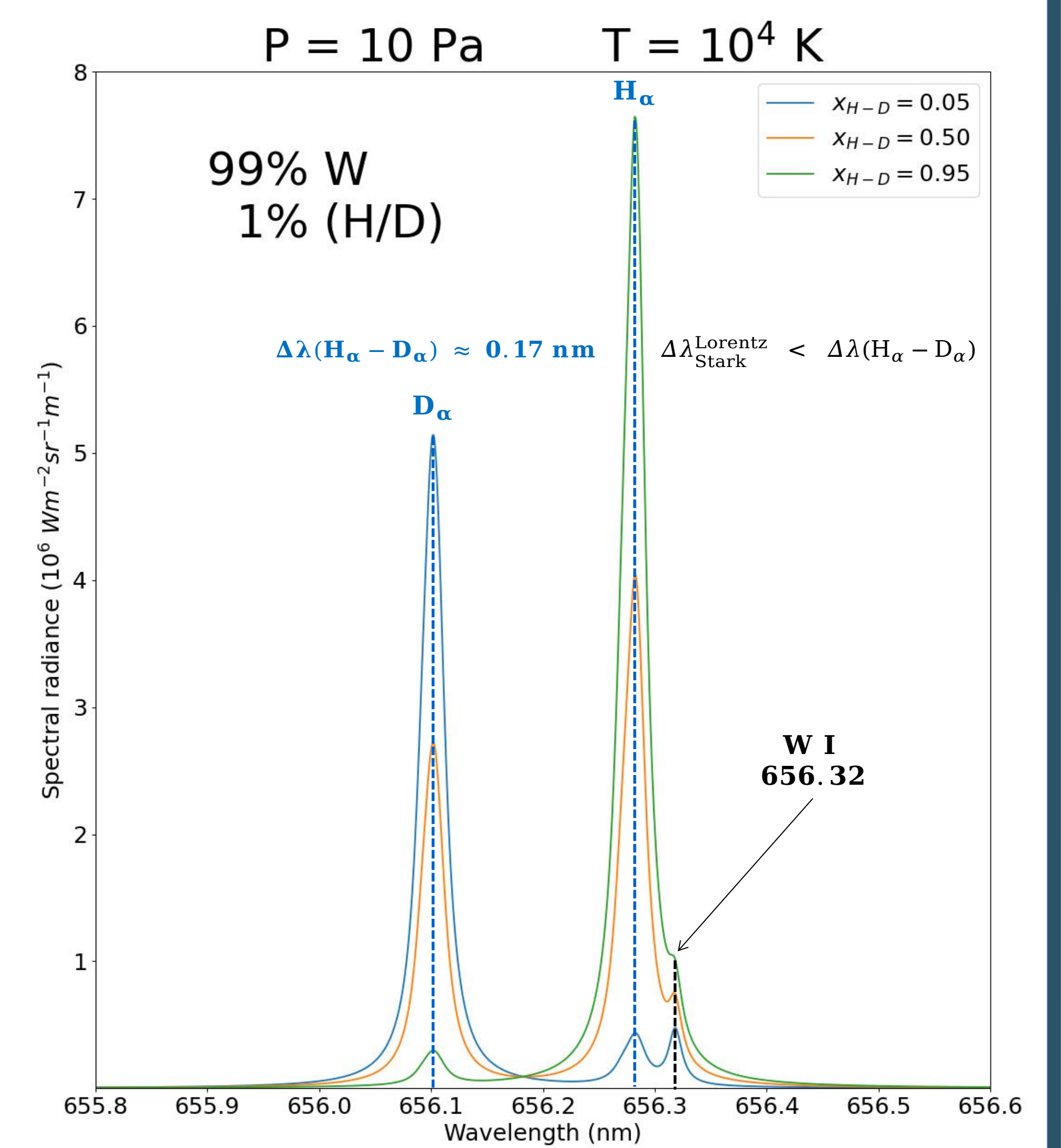
- H₂ dilution leads to significant H α emission
- For every non-H lines : $\Delta\lambda_{Stark}^{Lorentz} = \text{default correlation}$
- Since H ionization potential is lower than the one of Ar, diluting leads to greater n_e values. $(E_H^I = 13.598 \text{ eV}, E_{Ar}^I = 15.759 \text{ eV})$
- Since the ArH dissociation potential is very weak, dilution does not induce strong n_{Ar} / n_{Ar+} variations at T and P constant.
- Argon and Hydrogen diagnostic appears to be apart one from the other with dilution.

Temperature in zeolite-like structure



- Zeolite = porous structure with high hydrogen retention potential. COMPOSITION : 0.087H / 0.087Al / 0.217Si / 0.609O
- For every non-H lines : $\Delta\lambda_{Stark}^{Lorentz} = \text{default correlation}$
- Temperature increase heads to ionization degree growth (ionic lines, strong increase in n_e and significant n_H decrease).
- H α and S I lines interfere : direct diagnostic appears to be tricky: necessity to probe other Balmer lines.

H/D mole ratio in bulk W



- Light element probing in bulk heavy metallic matrix such as W. COMPOSITION : Constant 0.99W / 0.01(H-D) with variable x_{H-D} ratio
- For every non-H lines : $\Delta\lambda_{Stark}^{Lorentz} = \text{default correlation}$
- W I 656.32 nm is $\Delta\lambda(W I - H\alpha) \approx 0.03 \text{ nm}$ from H α which perturbs direct H probing : Al matrix alternative.
- Weak emission due to very low (H-D) charge : necessity to probe in low pressure plasma to reduce Stark as possible.

CONCLUSION & PERSPECTIVES

NECESSARY TOOL TO ANALYZE LIBS EXPERIMENTS



Many upgrades are planned: potential lowering, resonant broadening, Zeeman broadening, non-homogenous media, multi-T support, etc...

REFERENCES

J. Liu et al. J. Chem. Phys 130 (2009) 174306
 A. W. Mizolek et al., Fund. and Appl., Cambridge Univ. Press (2006)
 R. Fantoni et al., Spectrochimica Acta Part B 129 (2017) 8-13
 Z. Mijatovic et al., Spectrochimica Acta Part B 166 (2020) 105821
 A. Gigoso et al., Spectrochimica Acta Part B 58 (2003) 1489-1504

P. Stancil et al. The Astrophysical Journal 430 (1994) 360-370
 R. W. Patch et al., J. Chem. Phys 49 (1968) 961
 M. A. Maltsev et al., TVT 53 (2019) 367-370
 M. Capitelli et al., Springer (2012) ISBN: 978-1-4419-8181-3
 P. Fauchais et al., Springer (1994) ISBN: 978-1-4899-1339-5