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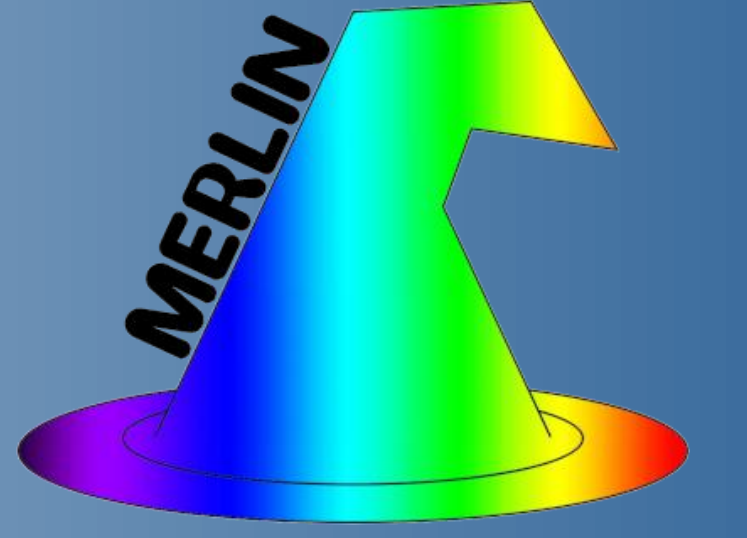
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MERLIN, a RT-LTE software supporting the LIBS diagnostic: application to H-isotopes measurements

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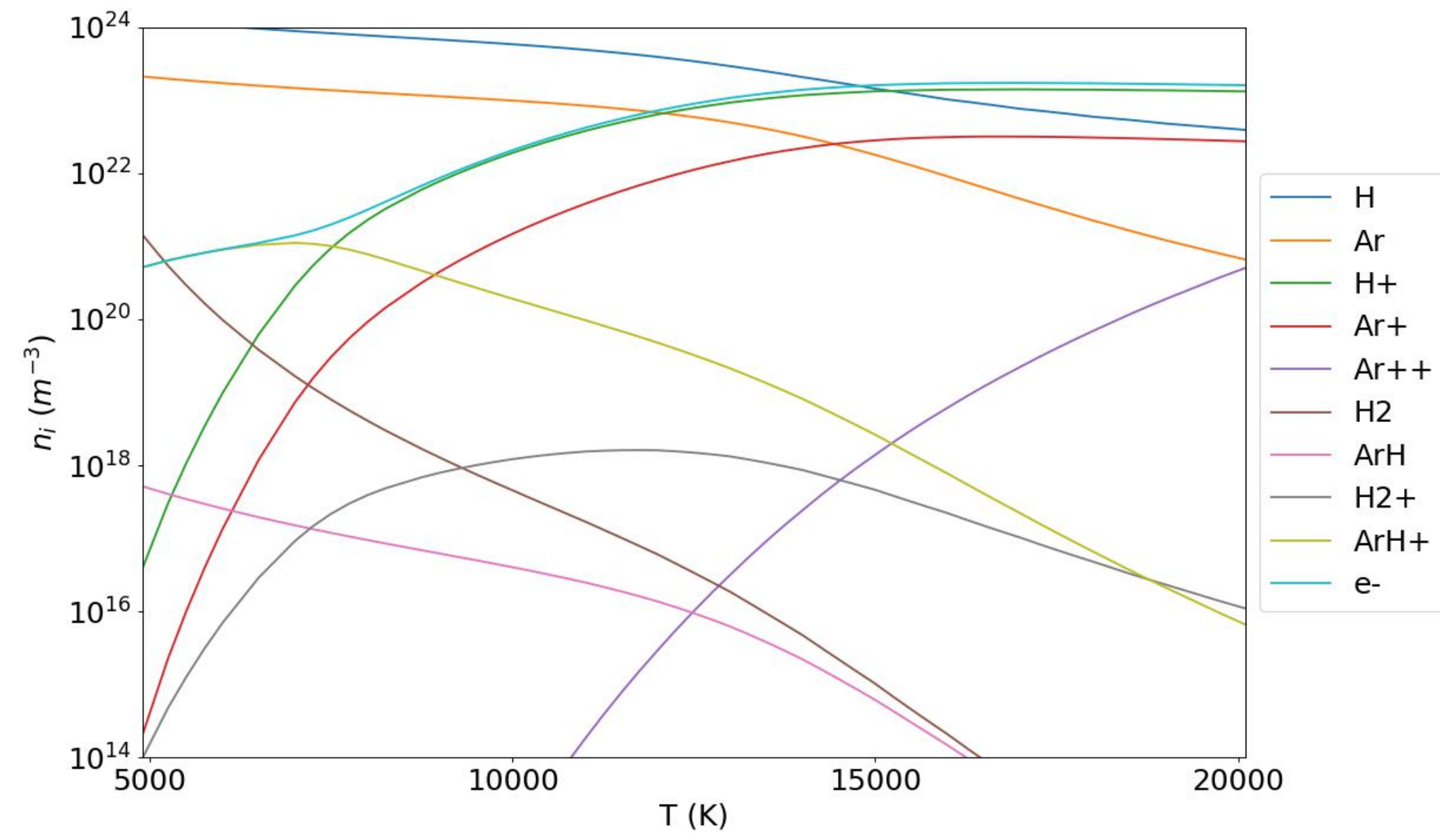
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CONTEXT & OBJECTIVES

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.

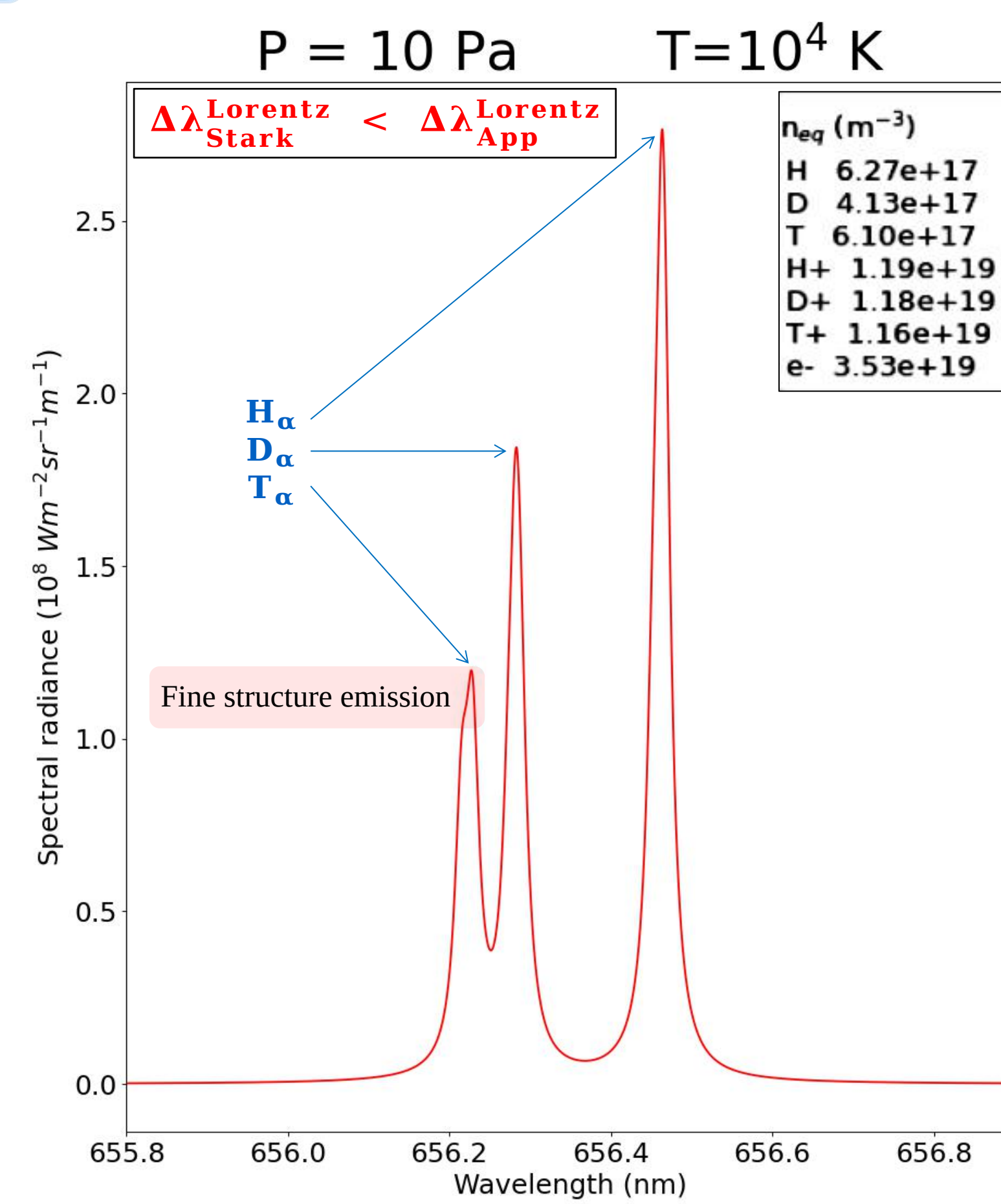
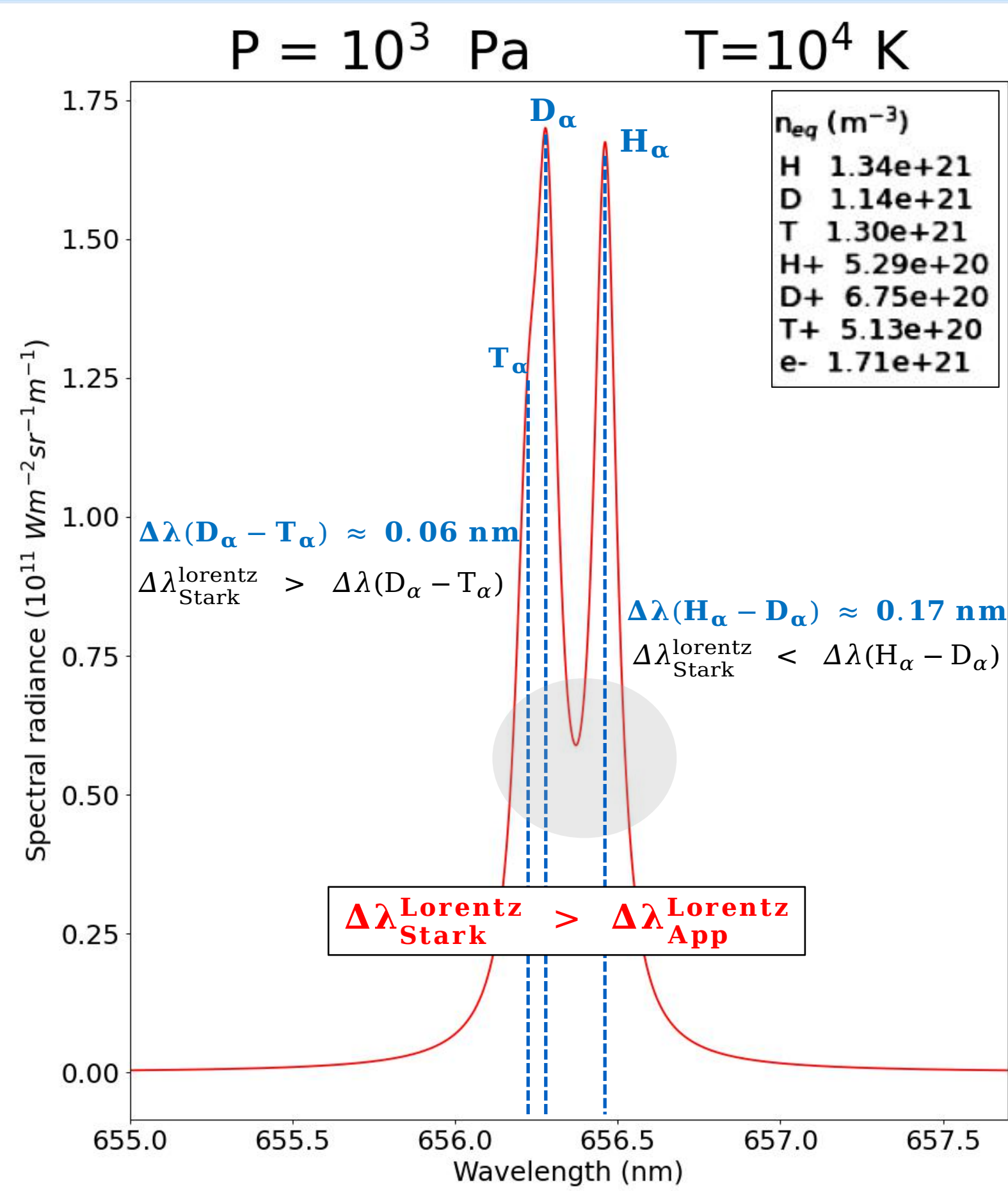
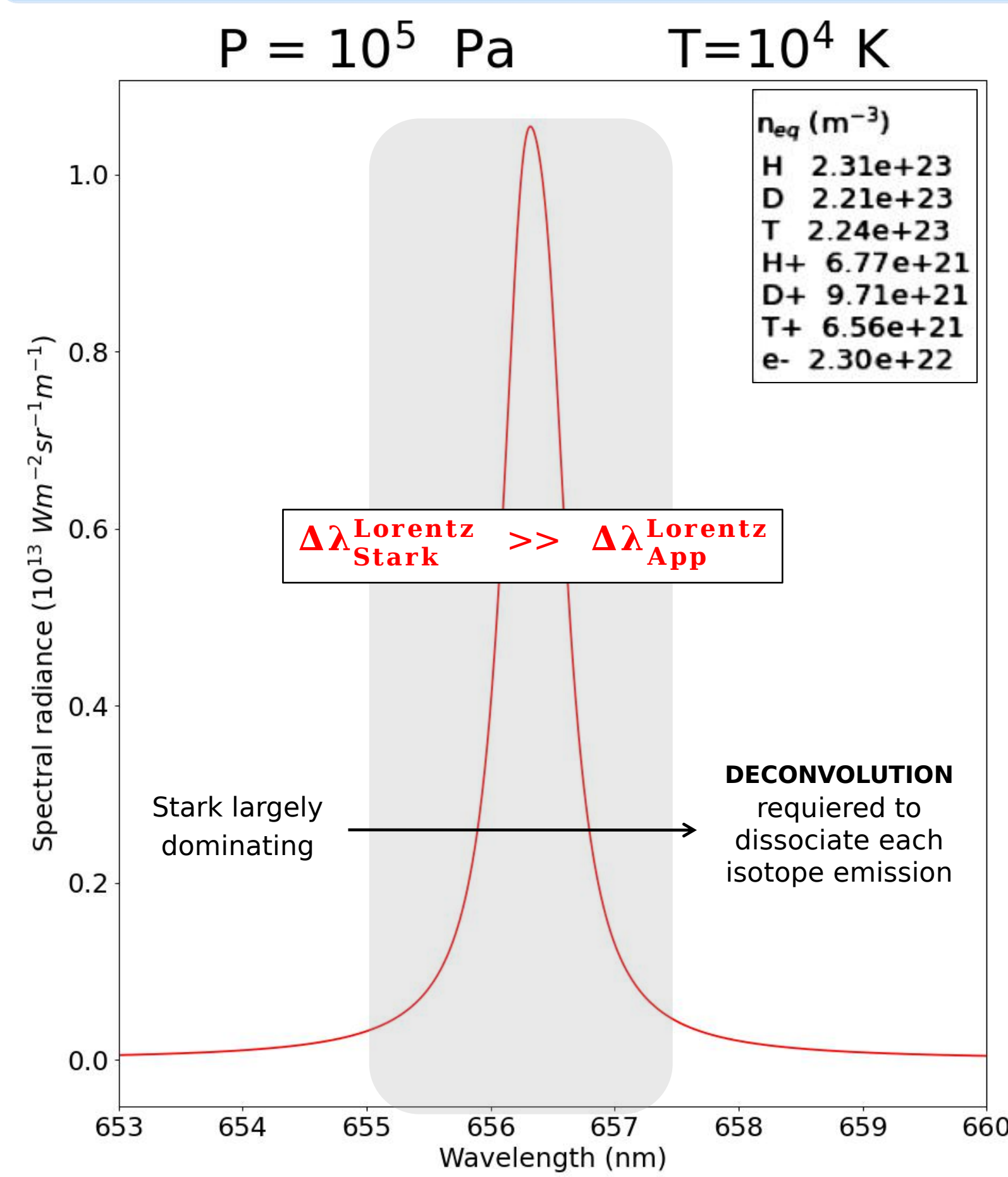
MERLIN (MultiElemental Radiative equiLilibrium emissioN) has been developed to ensure a computational support OES experiments. This suit is based on Radiative Transfer (RT) Local Thermal Equilibrium (LTE) solving for homogeneous emitting media. Ionizations and dissociations equations are dynamically 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 :

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

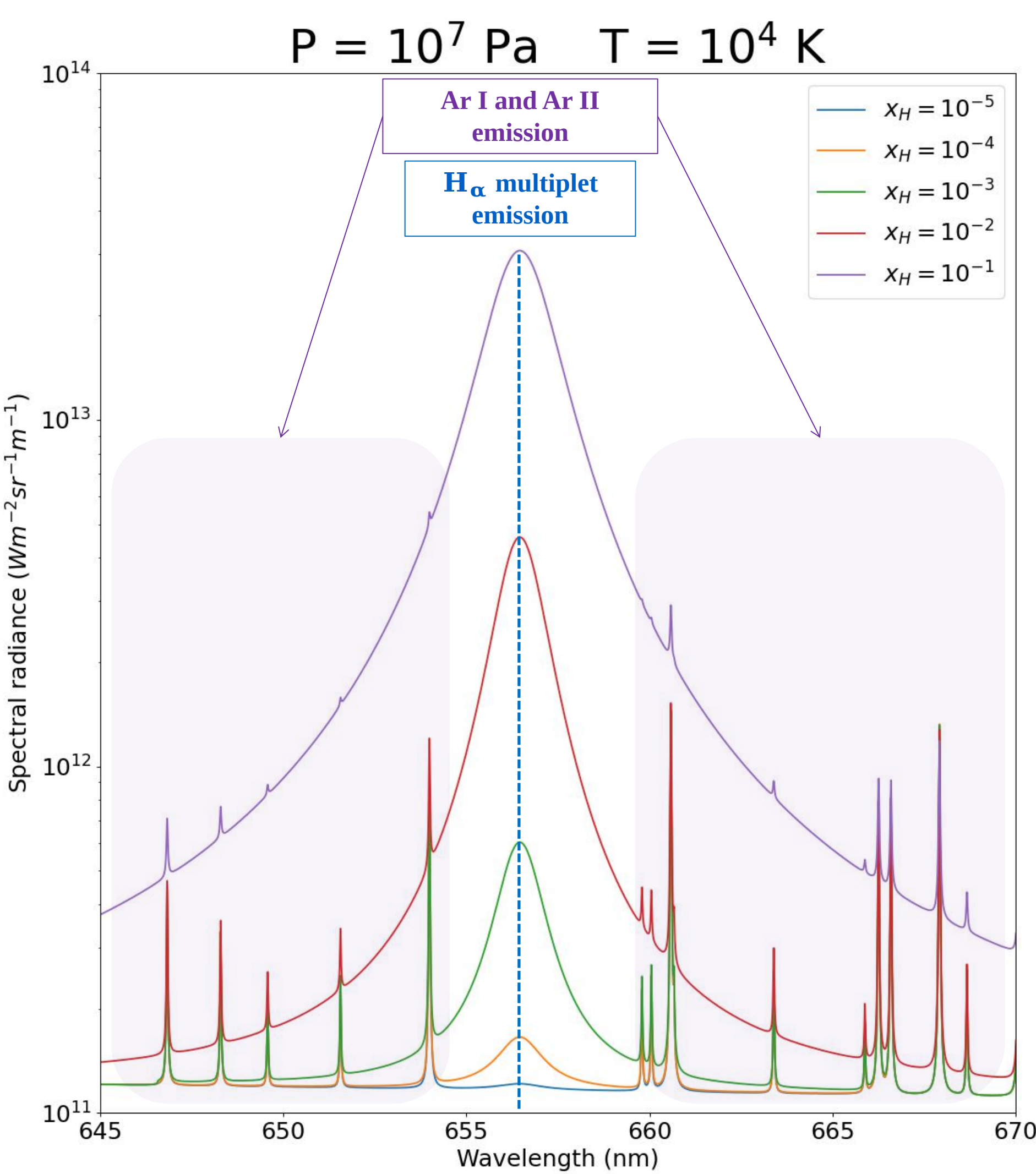
$$\Delta\lambda_{Doppler}^{Gauss} = 0 \text{ nm}$$

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

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

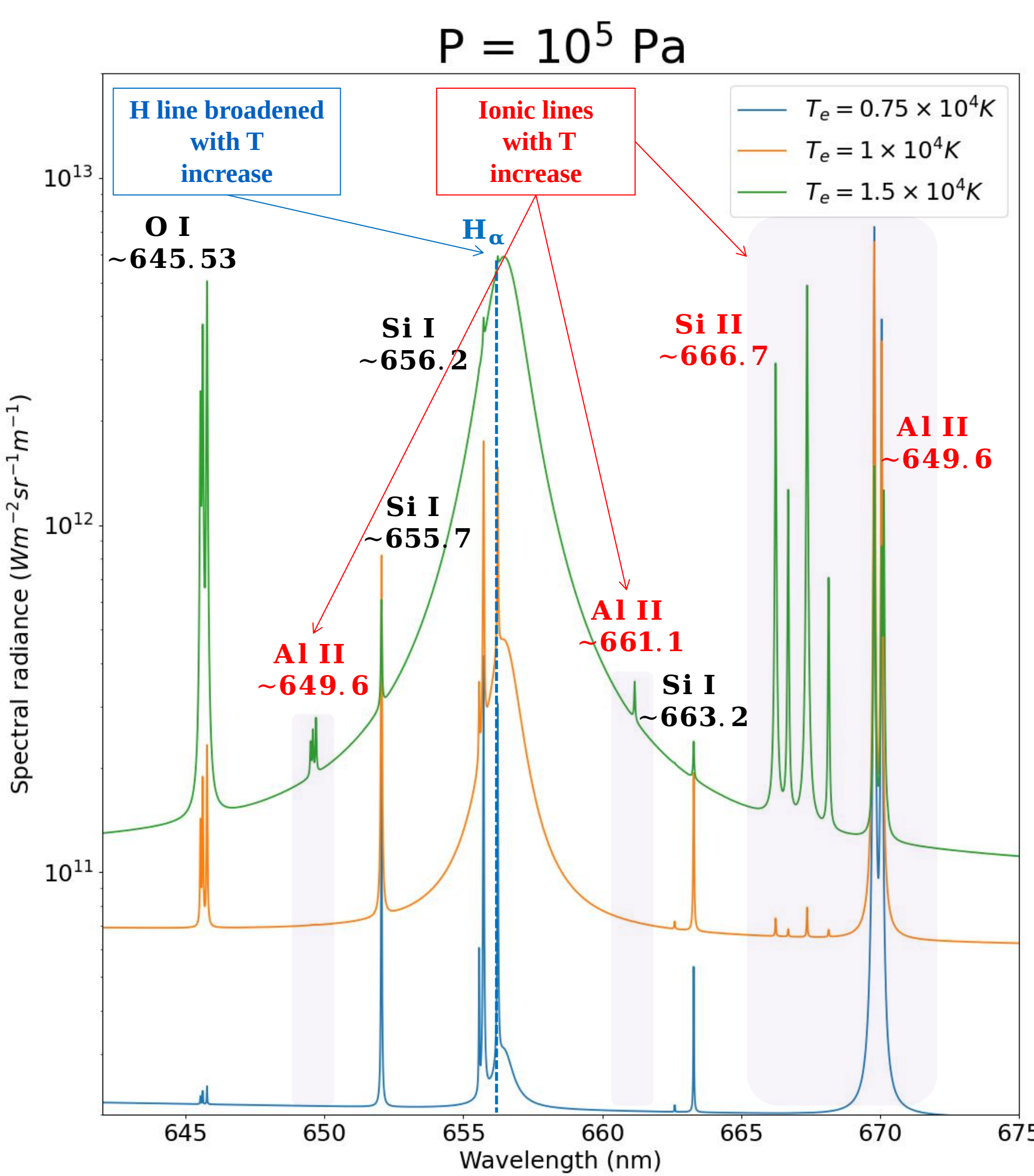
- Higher pressure leads to a significant Stark broadening which is n_e dependant.
- A competition between apparatus and Stark broadenings rules the resolution power.
- Element A_{ki} ratios verifies equilibrium verifies at low densities.

H mole fraction in Ar-H₂ mixture



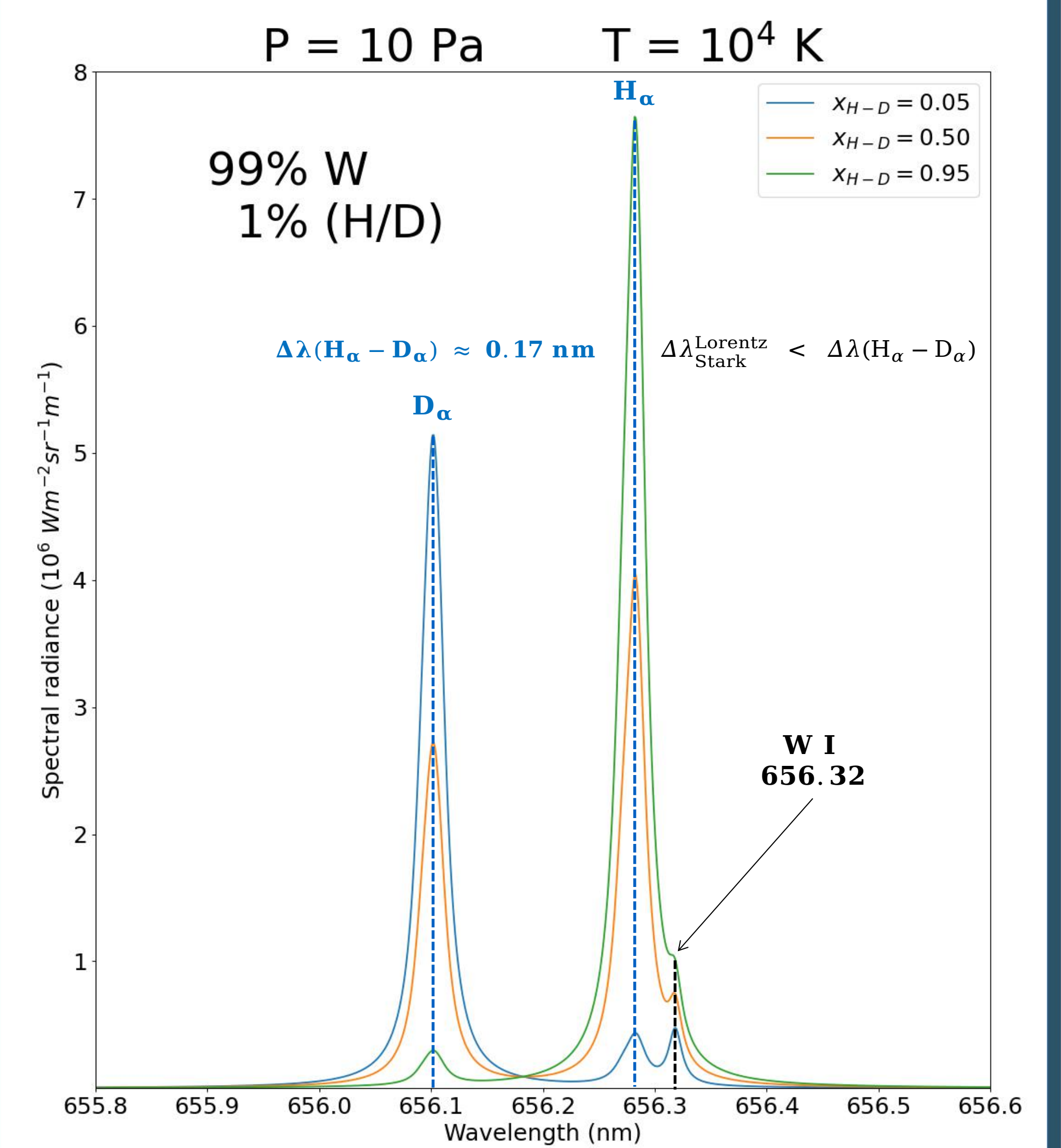
- H₂ dilution leads to significant H α emission
- For every non-H lines : $\Delta\lambda_{Stark}^{Lorentz} = \text{default correlation}$
- Since H ionization potential is lower than the one of Ar, diluting leads to greater n_e values. $(E_H^I = 13.598 \text{ eV}, E_{Ar}^I = 15.759 \text{ eV})$
- Since 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. Gigosos 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