Modelling of nonlinear light up-conversion from intense femtosecond laser pulses

DESY Summer Student Programme 2023
Project Presentation

Group: CFEL-ATTO
Supervisor: Josina Hahne

David Amorim (University of Glasgow)

1. Motivation

- The CFEL-ATTO group uses THG of few-femtosecond NIR laser pulses in a gas cell to generate ultrashort UV pulses
- The aim of this project was to produce simulations of the THG process to:
  a. Reproduce the experimental conditions in the gas
  b. Study the effects and parameters shaping UV generation
  c. Compare the new gas chip to the old (2019) gas cell

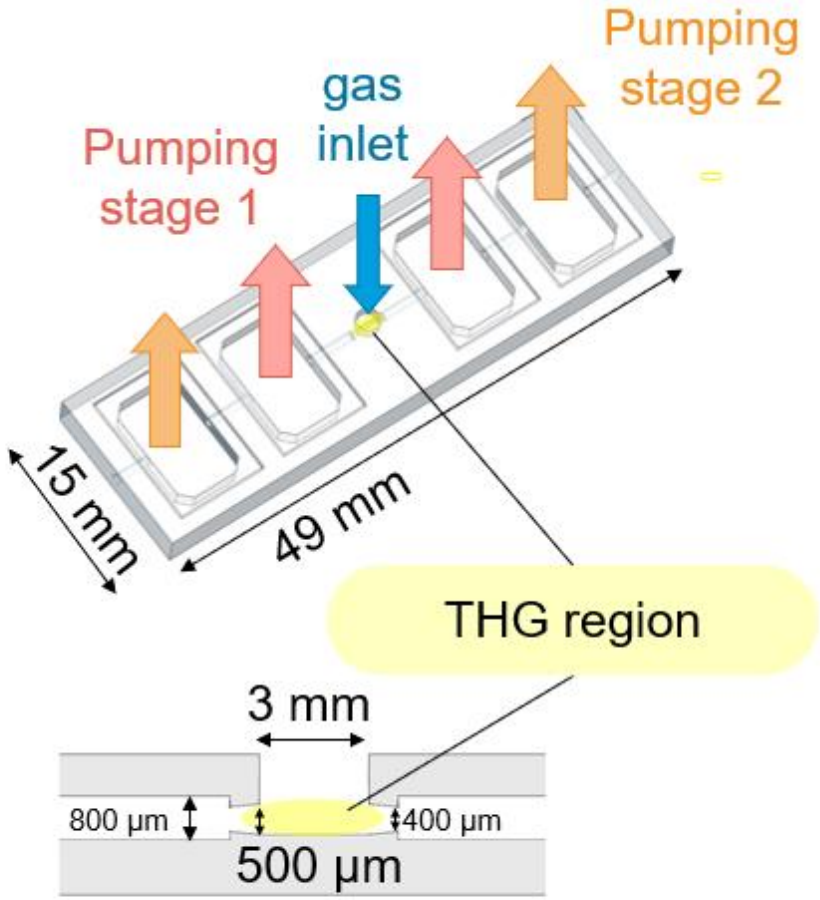

Fig. 1: the fused silica chip used for gas confinement.
Reproduced from Josina Hahne's master's thesis defence

2. Background

- The simulations were produced using the *Luna.jl* propagation solver [1], which formed the basis of a software package produced for the project
- *Luna.jl* numerically solves the unidirectional pulse propagation equation, which is derived without a slowly-varying envelope approximation [2]
- The nonlinear polarisation response in *Luna.jl* considers the Kerr effect as well as photo-ionisation: $\boldsymbol{P}^{NL} = \boldsymbol{P}^{Kerr} + \boldsymbol{P}^{ion}$, with

$$\boldsymbol{P}^{Kerr}(t,\boldsymbol{r}) = \chi^{(3)}[\boldsymbol{E}(t,\boldsymbol{r})]^3 \qquad (1)$$

$$\frac{\partial}{\partial t}\boldsymbol{P}^{ion}(t,\boldsymbol{r}) = \frac{I_p}{\boldsymbol{E}(t,\boldsymbol{r})}\frac{\partial n_e}{\partial t} + \frac{e^2}{m_e^2}\int_{-\infty}^{t} n_e(t')\boldsymbol{E}(t',\boldsymbol{r})dt' \qquad (2)$$

where I_p is the ionisation potential, n_e is the number of free electrons and all other symbols have their usual meaning

3. Simulation inputs

- The simulation input was based on measured data, as far as possible:
  - Time-intensity data of the NIR input beam is read in
  - The spatial profile of the input beam is assumed to be a Gaussian
  - *The gas density profile in the central interaction region is approximated using a gradient model:*

$$p(z_0 < z < z_1)^2 = p(z_0)^2 + (z - z_0)\frac{p(z_1)^2 - p(z_0)^2}{z_1 - z_0} \quad (3)$$

- While a variety of gases, pressures, and input powers can be simulated, this presentation focusses on Argon and Neon

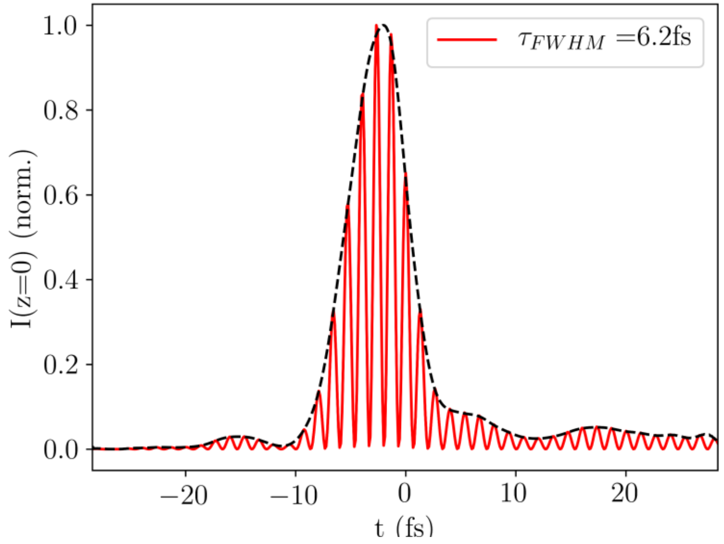

Fig. 2: NIR input pulse in the time domain

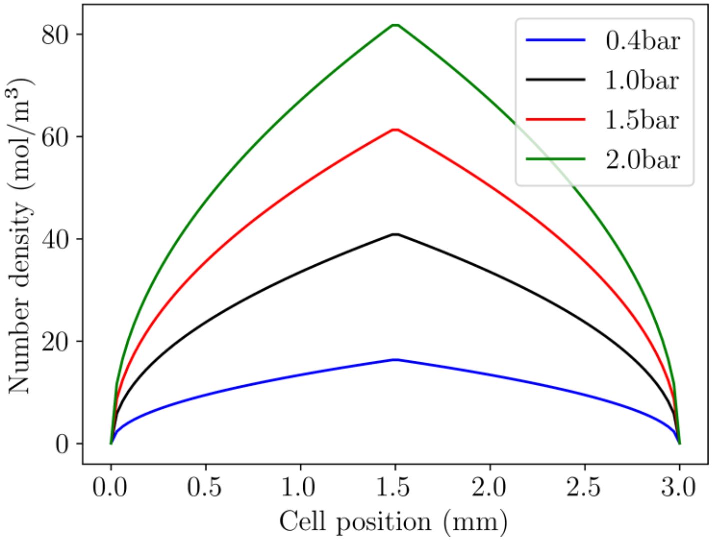

Fig. 3: gradient model of the gas distribution

4.I Comparison with experiment

The simulated UV energies qualitatively reproduce experimental trends, although energy values are overestimated by a factor of 10 and pressures must be rescaled (factor 2.5):

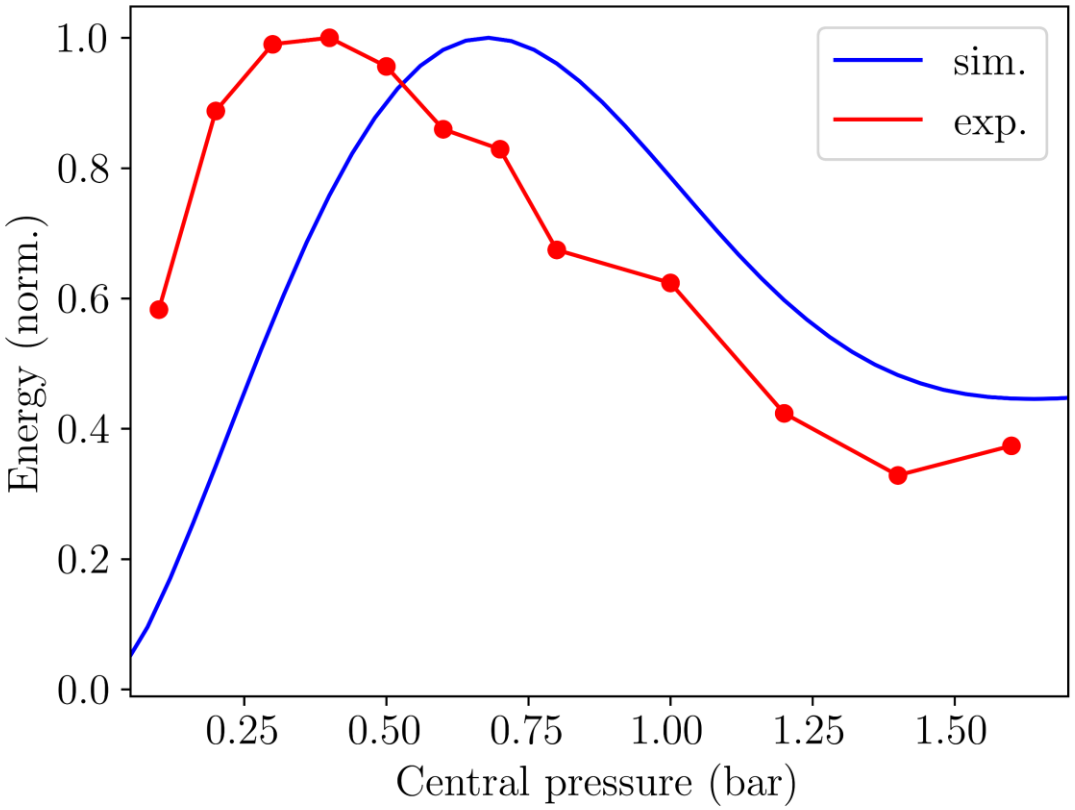

Fig. 4: Normalised UV output energies for Argon with 150mW input beam power. Simulation pressures are rescaled.

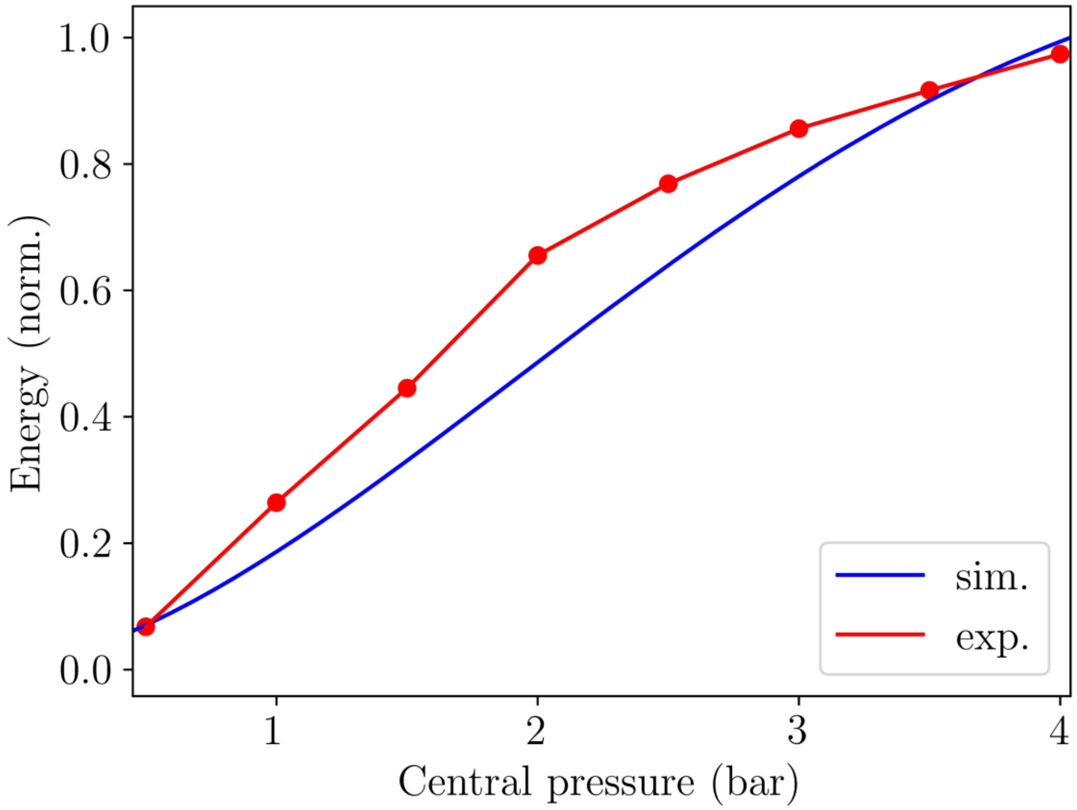

Fig. 5: Normalised UV output energies for Neon with 400mW input beam power. Simulation pressures are rescaled.

4.I Comparison with experiment

Using rescaled pressures, the simulated UV spectra agree reasonably well with experiment:

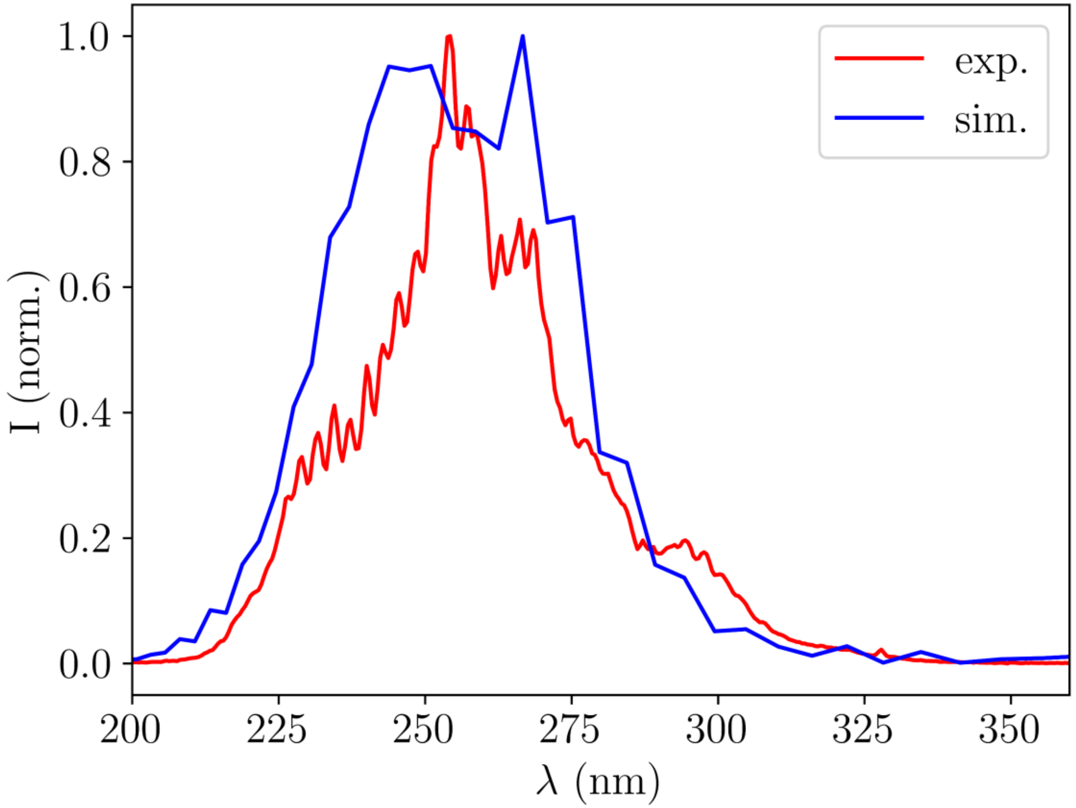

Fig. 6: UV output spectra for Argon with 150mW input beam power and 0.4bar rescaled central pressure

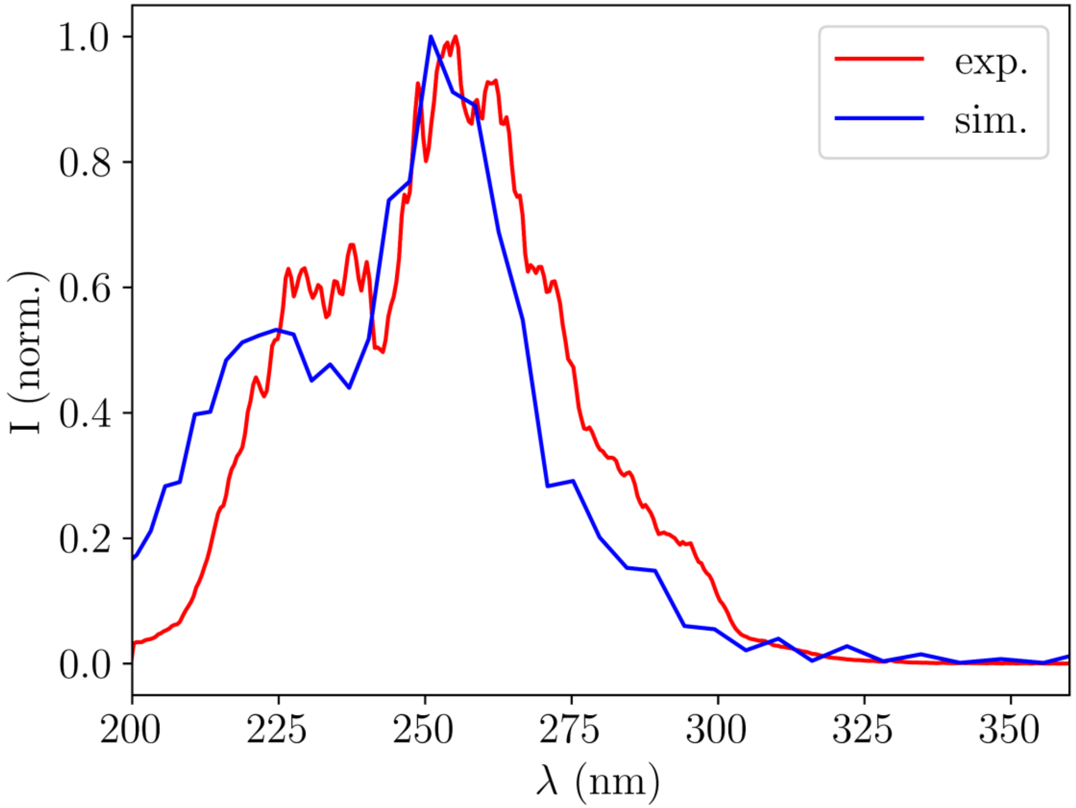

Fig. 7: UV output spectra for Neon with 400mW input beam power and 2.0bar rescaled central pressure

4.I Comparison with experiment

Across different (rescaled) pressures, general spectral trends can be reproduced:

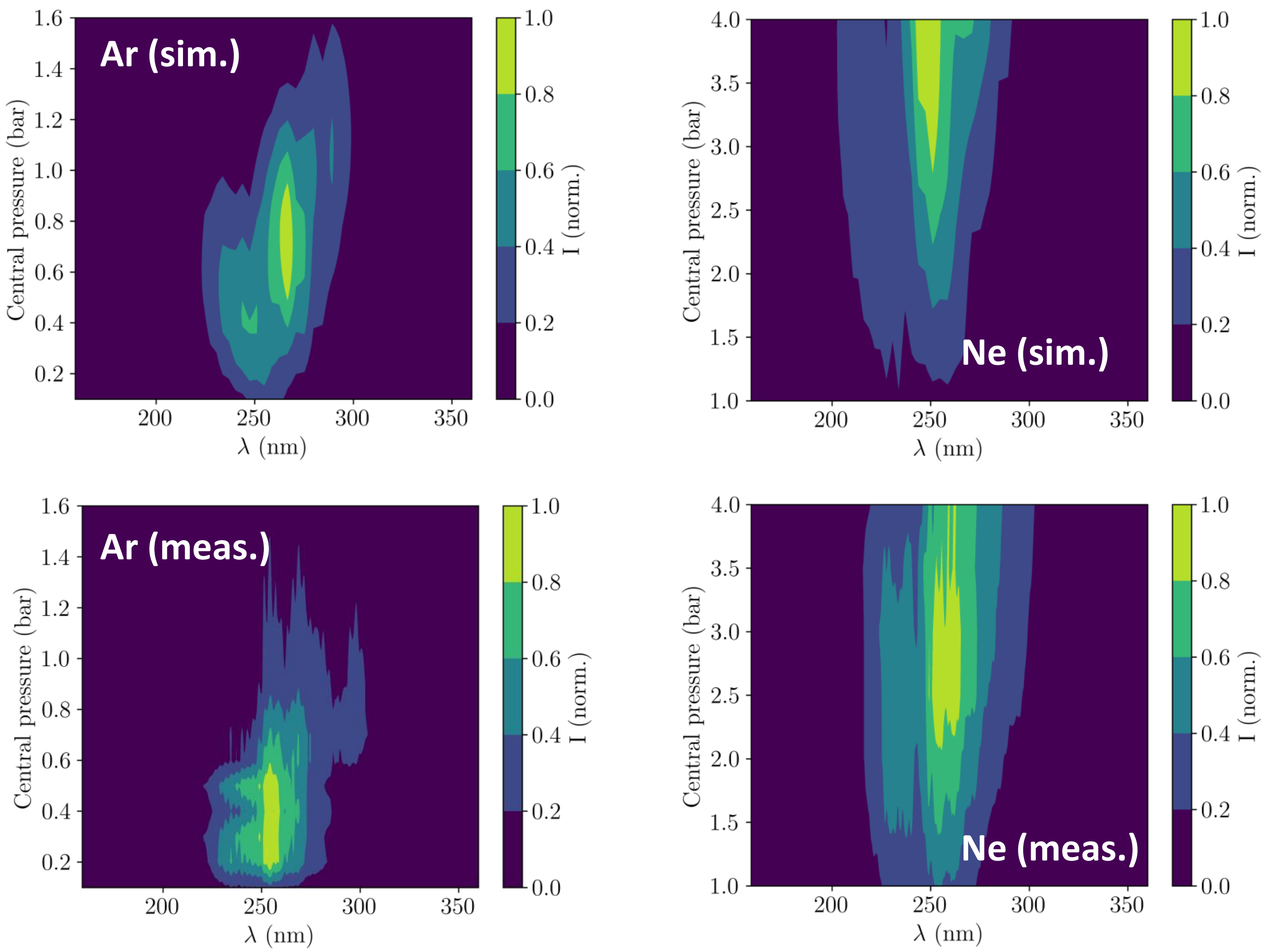

Fig. 8: UV output spectra at different rescaled central pressures. Left column: Argon (150mW), right column: Neon (400mW). Upper row: simulations, bottom row: measurements

4.II Self-defocusing

The simulations show that ionisation-induced self-defocusing at the trailing edge contributes to pulse compression, in qualitative agreement with literature [3]:

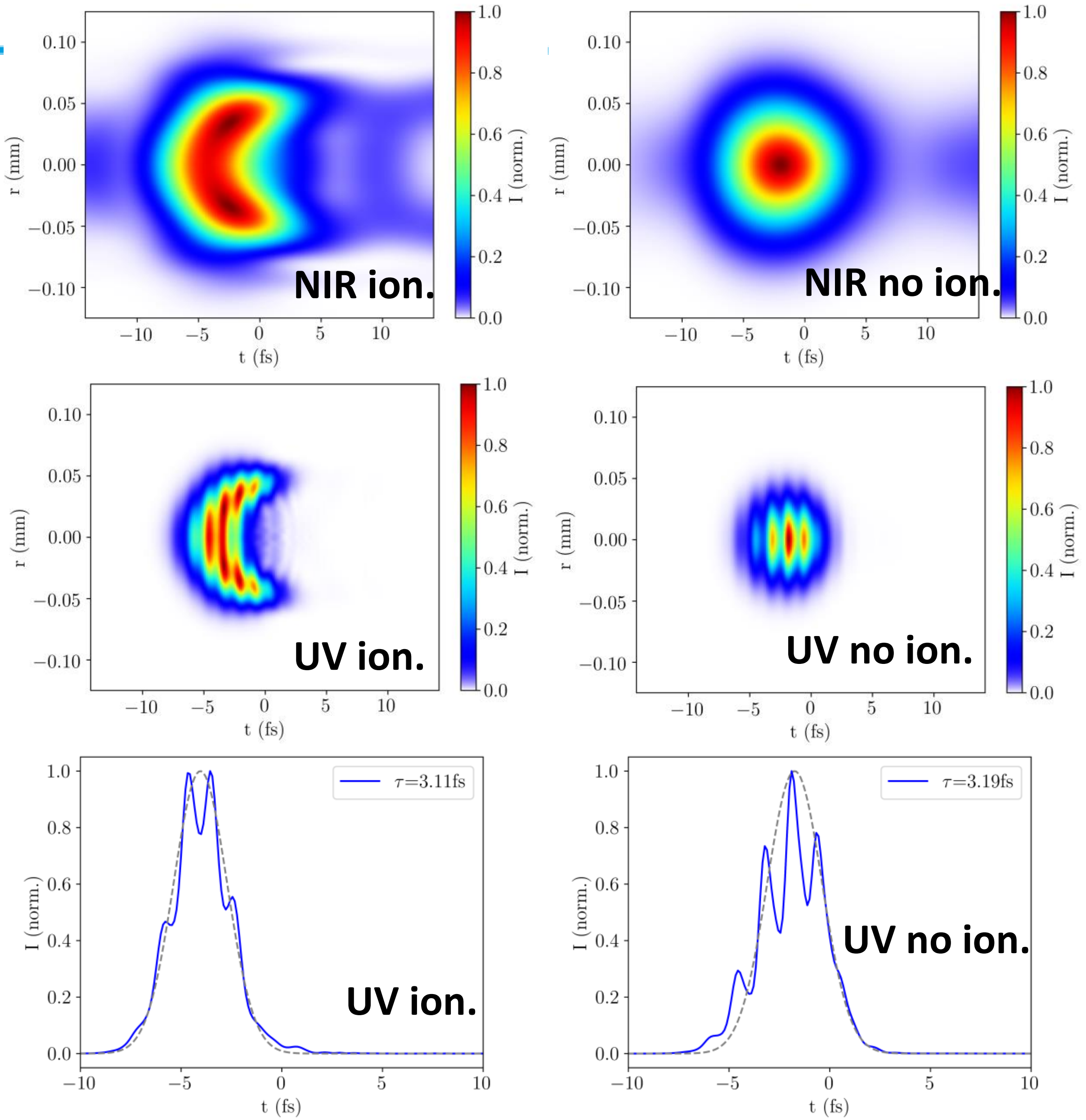

Fig. 9: NIR (top row) and UV (middle and bottom rows) output beams with (left column) and without (right column) ionisation using Argon at 150mW NIR power and 0.4bar rescaled central pressure

4.III Effects of chirp

The simulations indicate that using chirped input pulses could potentially improve UV pulse properties:

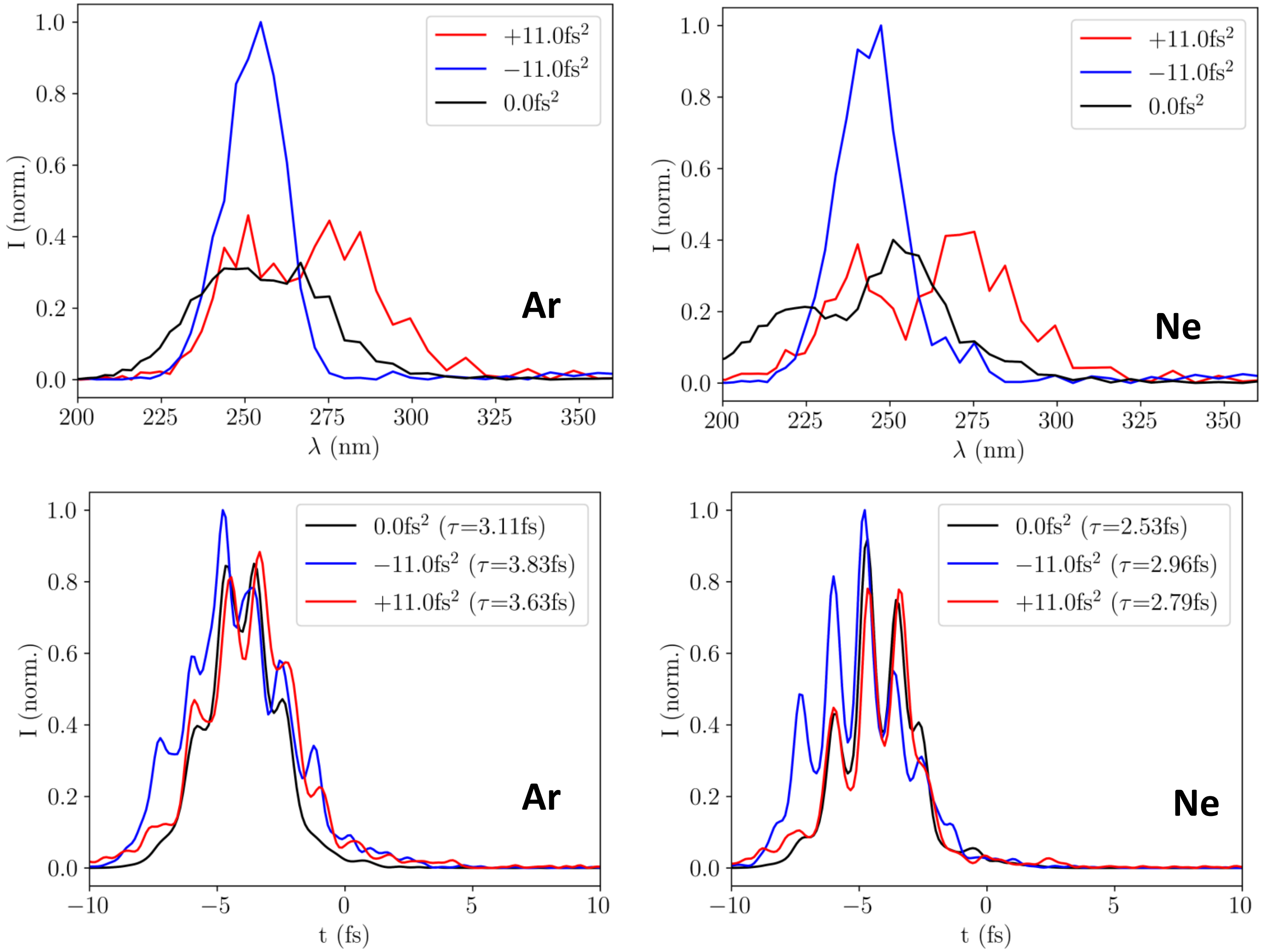

Fig. 10: UV output spectra and temporal profiles for different GVD values of the input pulse. Left column: Argon (150mW, 0.4bar), right column: Neon (400mW, 2.0bar); pressures rescaled

4.IV Comparing the gas cells

- *COMSOL* simulations for the gas density distribution of the new and old cell were produced (by Josina Hahne)
- Currently, these models do not sufficiently consider interactions between gas molecules at high pressures
- They do allow for a qualitative comparison of the new and the old cell, however

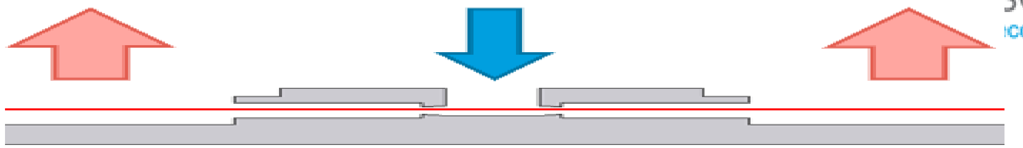

Fig. 11: side view of the new chip Reproduced from Josina Hahne's master's thesis defence

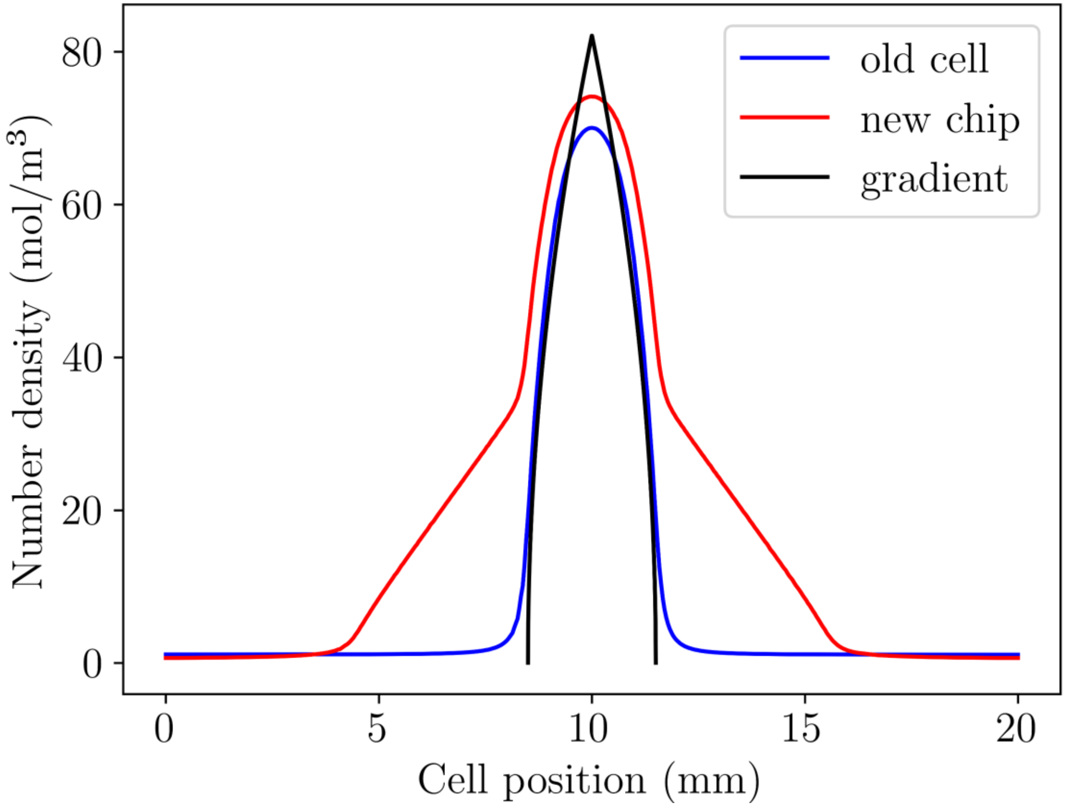

Fig. 12: Gas density distribution models for the old cell, the new chip and the gradient model, all at 2.0bar

4.IV Comparing the gas cells

The simulations indicate that the new chip to delivers more intense UV pulses with broader spectra, in agreement with experiments:

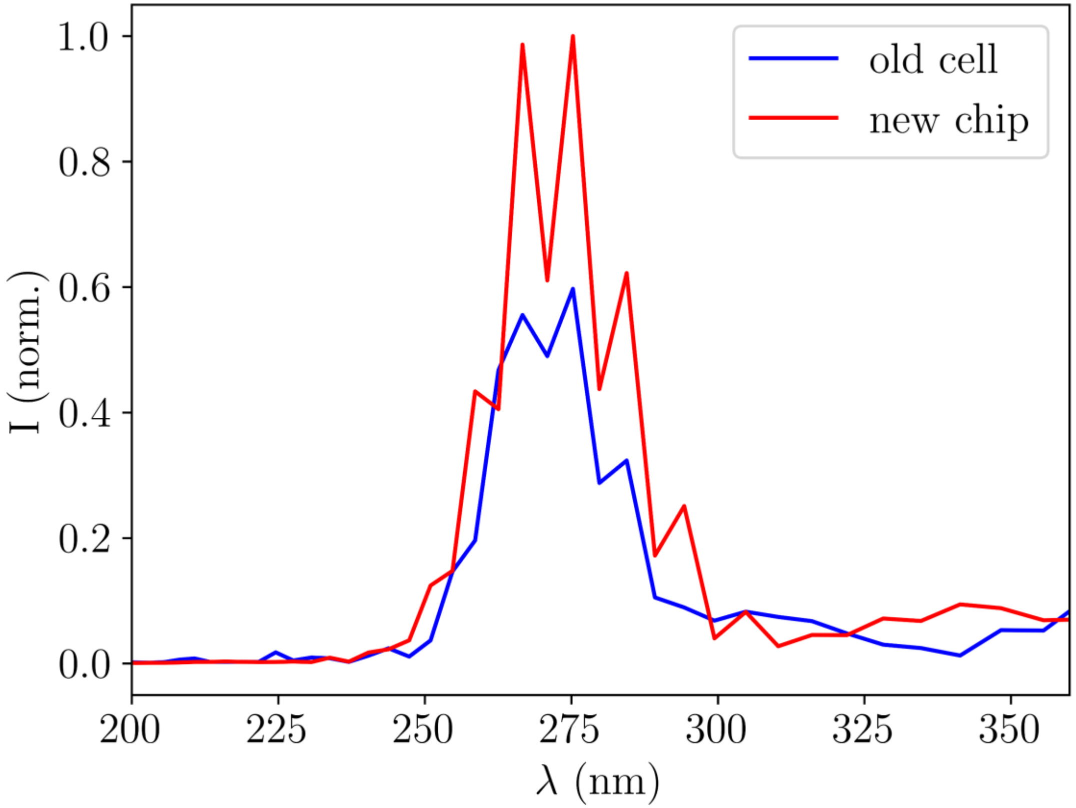

Fig. 13: UV output spectra with new and old gas cell for Argon at 150mW NIR power and 0.4bar rescaled central pressure

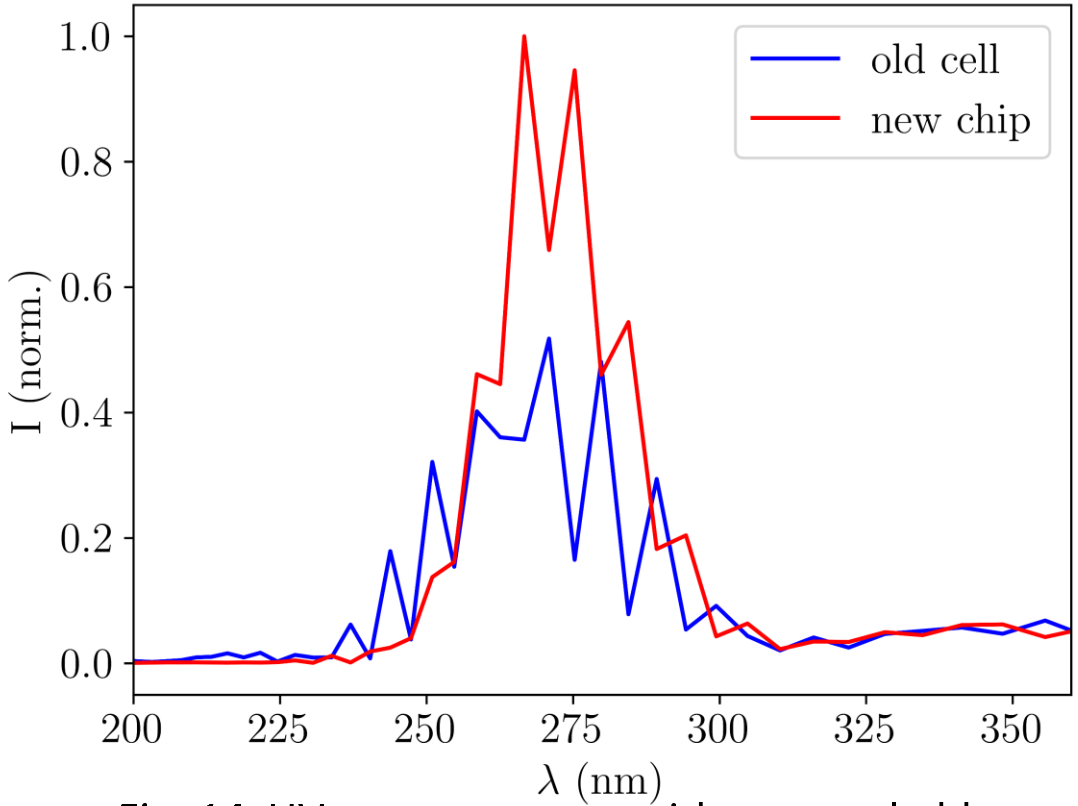

Fig. 14: UV output spectra with new and old gas cell for Neon at 400mW NIR power and 2.0bar rescaled central pressure

4.IV Comparing the gas cells

- Interestingly, the gradient distribution results in significantly more intense UV output despite being nearly identical to the old cell in the central interaction region
- This seems to indicate a large absorptive effect of the low-density regions outside the THG region

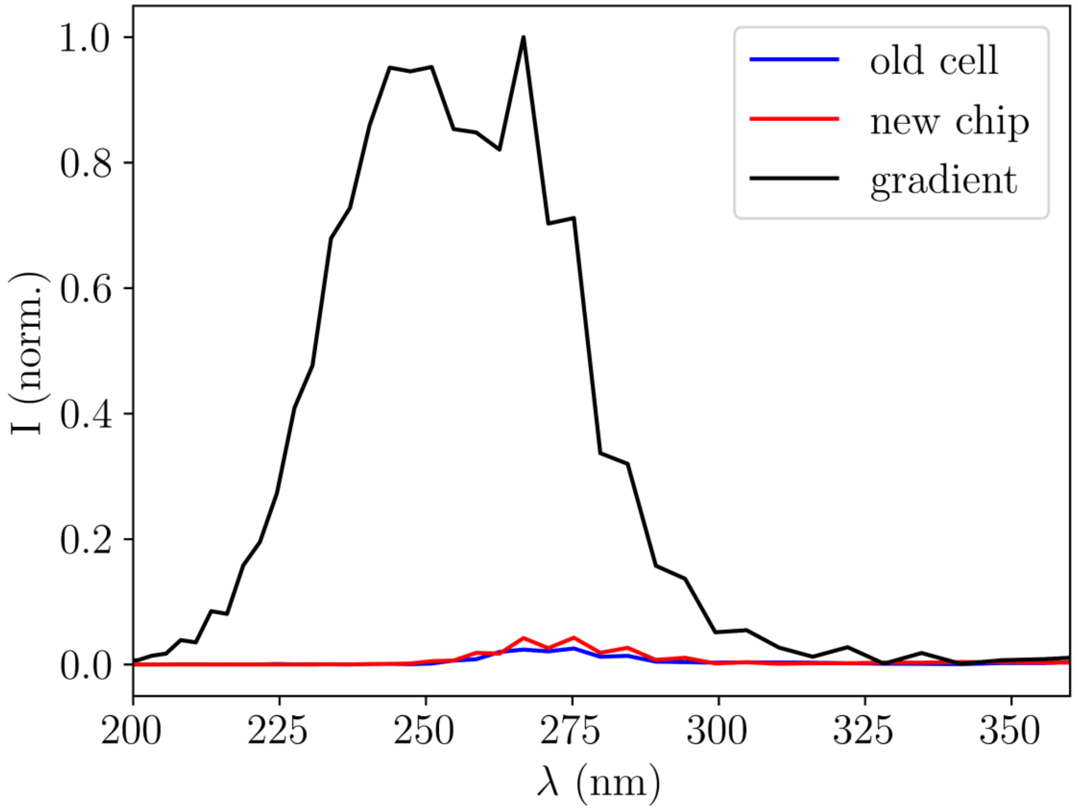

Fig. 15: UV output spectra with new and old gas cell and the gradient model for Argon at 150mW NIR power and 0.4bar rescaled central pressure

5. Discussion & conclusion

- The THG simulations at their current stage resulted in some promising outputs, showing qualitative agreement with both experiment and literature
  - Some of the results shown will form part of a submission to *JPhys Photonics*

- There is significant room for further improvements:
  - Base the spatial profile of the input beam on measured data
  - Use *COMSOL* simulations to accurately model the gas distribution inside and outside the central interaction region

- With additional expansions, especially with regards to gas density modelling, the simulation code could in the future serve as the basis of a theoretical framework to complement experiments

6. Acknowledgments & references

CFEL SCIENCE
Attosecond Science

Acknowledgments

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References

[1] C. Brahms and J. C. Travers, “*Luna.jl*,” (2023); https://doi.org/10.5281/zenodo.8242646

[2] M. Kolesik and J. V. Moloney, “*Nonlinear optical pulse propagation simulation: From Maxwell’s to unidirectional equations*,” Phys. Rev. E **70**, 036 604 (2004)

[3] F. Reiter, U. Graf, et al. “*Route to Attosecond Nonlinear Spectroscopy*,” Phys. Rev. Lett. **105**, 243 902 (2010)