

# XUV Spectra from Plasmas of Second Transition Row Elements Generated by fs Laser Pulses

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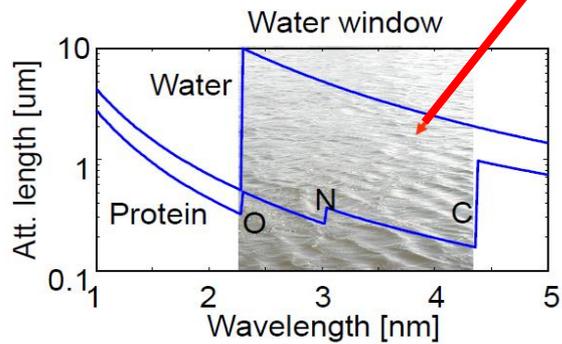
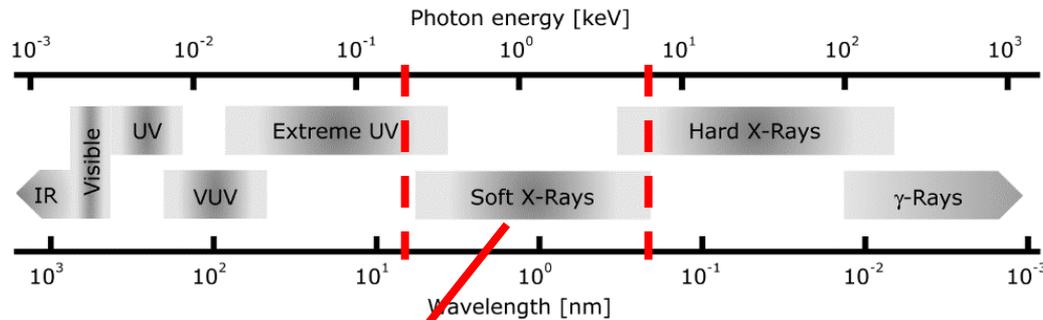
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- Motivation
- Introduction
- Results and Discussion:
  - Section 1: XUV spectra of 2<sup>nd</sup> transition row elements:  
identification of 3d-4p and 3d-4f transition arrays
  - Section 2: Femtosecond LPPs from 2<sup>nd</sup> transition row elements
- Conclusion
- Acknowledgments

Feasibility of using 2<sup>nd</sup> transition row elements as possible candidates for

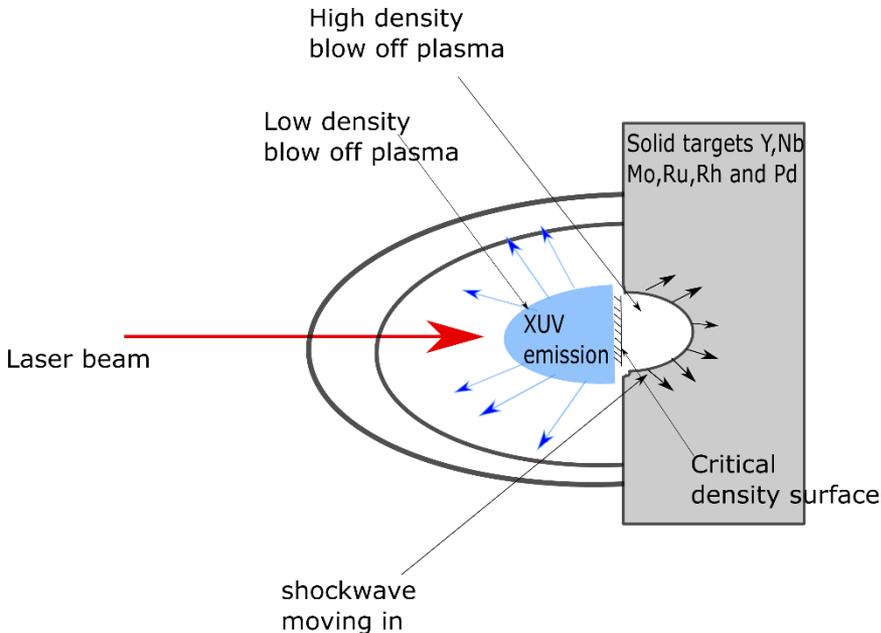
- **Water window** sources
- **Next generation lithography** (6.X nm).
- **Lower laser intensities** (and electron temperatures of 150 to 300 eV)
- Optimum matching of spectral output - **multilayer mirrors**.



- Water window absorption contrast 2.3-4.4 nm (284-543 eV)
- Relative transparency of water allows investigation of biomolecules, cells and proteins
- Possibility to study thick ( $\sim 10 \mu\text{m}$ ) objects

High power laser intensity focused onto a solid targets in vacuum forms a

- Short lived high temperature,
- High density plasma



Plasma temperature depends on laser intensity ( $\Phi$ ) and wavelength( $\lambda$ ),  $T_e(\text{eV}) \propto (\lambda^2 \Phi)^{3/5}$

Laser produced plasma (LPPs) expansion velocity  $\approx 10^6\text{-}10^7 \text{ cms}^{-1}$

Critical electron density, depends on laser wavelength  $n_{ec} (\text{cm}^{-3}) = 10^{21} / \lambda^2 [\text{micron}]$

Spectroscopy of LPPs provide detail information on

- The transitions and electronic structure of highly ionized atoms,
- Allow source optimization.

- Results and Discussion:

Section 1: XUV spectra of 2<sup>nd</sup> transition row elements: identification of 3d-4p and 3d-4f transition arrays.

The positions of the reflectance peaks of currently available MLMs are compared with the present experimental data.

### Periodic Table of the Elements

|                        |                |
|------------------------|----------------|
| Atomic<br>Number       | Atomic<br>Mass |
| <b>Symbol</b>          |                |
| <b>Name</b>            |                |
| Electron Shells        |                |
| Electron Configuration |                |

Element symbol represents state at room temperature.  
**Solid, Liquid or Gas**

| Period | 1<br>IA<br>1A                   | 2<br>IIA<br>2A                  |                                |                                     |                                 |                                     |                                  |                                   |                                  |                                    |                                   |                                   | 13<br>IIIA<br>3A                   | 14<br>IVA<br>4A                  | 15<br>VA<br>5A                     | 16<br>VIA<br>6A                   | 17<br>VIIA<br>7A                   | 18<br>VIIIA<br>8A                 |
|--------|---------------------------------|---------------------------------|--------------------------------|-------------------------------------|---------------------------------|-------------------------------------|----------------------------------|-----------------------------------|----------------------------------|------------------------------------|-----------------------------------|-----------------------------------|------------------------------------|----------------------------------|------------------------------------|-----------------------------------|------------------------------------|-----------------------------------|
| 1      | 1<br>H<br>Hydrogen<br>1.008     |                                 |                                |                                     |                                 |                                     |                                  |                                   |                                  |                                    |                                   |                                   |                                    |                                  |                                    |                                   |                                    | 2<br>He<br>Helium<br>4.003        |
| 2      | 3<br>Li<br>Lithium<br>6.941     | 4<br>Be<br>Beryllium<br>9.012   |                                |                                     |                                 |                                     |                                  |                                   |                                  |                                    |                                   |                                   | 5<br>B<br>Boron<br>10.811          | 6<br>C<br>Carbon<br>12.011       | 7<br>N<br>Nitrogen<br>14.007       | 8<br>O<br>Oxygen<br>15.999        | 9<br>F<br>Fluorine<br>18.998       | 10<br>Ne<br>Neon<br>20.180        |
| 3      | 11<br>Na<br>Sodium<br>22.990    | 12<br>Mg<br>Magnesium<br>24.305 |                                |                                     |                                 |                                     |                                  |                                   |                                  |                                    |                                   |                                   | 13<br>Al<br>Aluminum<br>26.982     | 14<br>Si<br>Silicon<br>28.086    | 15<br>P<br>Phosphorus<br>30.974    | 16<br>S<br>Sulfur<br>32.066       | 17<br>Cl<br>Chlorine<br>35.453     | 18<br>Ar<br>Argon<br>39.948       |
| 4      | 19<br>K<br>Potassium<br>39.098  | 20<br>Ca<br>Calcium<br>40.078   | 21<br>Sc<br>Scandium<br>44.956 | 22<br>Ti<br>Titanium<br>47.88       | 23<br>V<br>Vanadium<br>50.942   | 24<br>Cr<br>Chromium<br>51.996      | 25<br>Mn<br>Manganese<br>54.938  | 26<br>Fe<br>Iron<br>55.845        | 27<br>Co<br>Cobalt<br>58.933     | 28<br>Ni<br>Nickel<br>58.693       | 29<br>Cu<br>Copper<br>63.546      | 30<br>Zn<br>Zinc<br>65.38         | 31<br>Ga<br>Gallium<br>69.723      | 32<br>Ge<br>Germanium<br>72.631  | 33<br>As<br>Arsenic<br>74.922      | 34<br>Se<br>Selenium<br>78.971    | 35<br>Br<br>Bromine<br>79.904      | 36<br>Kr<br>Krypton<br>84.798     |
| 5      | 37<br>Rb<br>Rubidium<br>84.468  | 38<br>Sr<br>Strontium<br>87.62  | 39<br>Y<br>Yttrium<br>88.906   | 40<br>Zr<br>Zirconium<br>91.224     | 41<br>Nb<br>Niobium<br>92.906   | 42<br>Mo<br>Molybdenum<br>95.9      | 43<br>Tc<br>Technetium<br>98.907 | 44<br>Ru<br>Ruthenium<br>101.07   | 45<br>Rh<br>Rhodium<br>102.906   | 46<br>Pd<br>Palladium<br>106.4     | 47<br>Ag<br>Silver<br>107.868     | 48<br>Cd<br>Cadmium<br>112.414    | 49<br>In<br>Indium<br>114.818      | 50<br>Sn<br>Tin<br>118.711       | 51<br>Sb<br>Antimony<br>121.760    | 52<br>Te<br>Tellurium<br>127.6    | 53<br>I<br>Iodine<br>126.905       | 54<br>Xe<br>Xenon<br>131.29       |
| 6      | 55<br>Cs<br>Cesium<br>132.905   | 56<br>Ba<br>Barium<br>137.328   | 57-71<br>Lanthanide Series     | 72<br>Hf<br>Hafnium<br>178.49       | 73<br>Ta<br>Tantalum<br>180.948 | 74<br>W<br>Tungsten<br>183.84       | 75<br>Re<br>Rhenium<br>186.207   | 76<br>Os<br>Osmium<br>190.23      | 77<br>Ir<br>Iridium<br>192.22    | 78<br>Pt<br>Platinum<br>195.085    | 79<br>Au<br>Gold<br>196.967       | 80<br>Hg<br>Mercury<br>200.592    | 81<br>Tl<br>Thallium<br>204.384    | 82<br>Pb<br>Lead<br>207.2        | 83<br>Bi<br>Bismuth<br>208.980     | 84<br>Po<br>Polonium<br>[209]     | 85<br>At<br>Astatine<br>[210]      | 86<br>Rn<br>Radon<br>[222]        |
| 7      | 87<br>Fr<br>Francium<br>223.020 | 88<br>Ra<br>Radium<br>226.025   | 89-103<br>Actinide Series      | 104<br>Rf<br>Rutherfordium<br>[261] | 105<br>Db<br>Dubnium<br>[262]   | 106<br>Sg<br>Seaborgium<br>[266]    | 107<br>Bh<br>Bohrium<br>[264]    | 108<br>Hs<br>Hassium<br>[269]     | 109<br>Mt<br>Meitnerium<br>[268] | 110<br>Ds<br>Darmstadtium<br>[271] | 111<br>Rg<br>Roentgenium<br>[272] | 112<br>Cn<br>Copernicium<br>[285] | 113<br>Nh<br>Nihonium<br>[284]     | 114<br>Fl<br>Flerovium<br>[289]  | 115<br>Uup<br>Ununpentium<br>[288] | 116<br>Lv<br>Livermorium<br>[293] | 117<br>Uus<br>Ununseptium<br>[294] | 118<br>Uuo<br>Ununoctium<br>[294] |
|        |                                 |                                 |                                | 57<br>La<br>Lanthanum<br>138.905    | 58<br>Ce<br>Cerium<br>140.116   | 59<br>Pr<br>Praseodymium<br>140.908 | 60<br>Nd<br>Neodymium<br>144.242 | 61<br>Pm<br>Promethium<br>144.913 | 62<br>Sm<br>Samarium<br>150.36   | 63<br>Eu<br>Europium<br>151.964    | 64<br>Gd<br>Gadolinium<br>157.25  | 65<br>Tb<br>Terbium<br>158.925    | 66<br>Dy<br>Dysprosium<br>162.500  | 67<br>Ho<br>Holmium<br>164.930   | 68<br>Er<br>Erbium<br>167.259      | 69<br>Tm<br>Thulium<br>168.934    | 70<br>Yb<br>Ytterbium<br>173.055   | 71<br>Lu<br>Lutetium<br>174.967   |
|        |                                 |                                 |                                | 89<br>Ac<br>Actinium<br>227.028     | 90<br>Th<br>Thorium<br>232.038  | 91<br>Pa<br>Protactinium<br>231.036 | 92<br>U<br>Uranium<br>238.029    | 93<br>Np<br>Neptunium<br>237.048  | 94<br>Pu<br>Plutonium<br>244.064 | 95<br>Am<br>Americium<br>243.061   | 96<br>Cm<br>Curium<br>247.070     | 97<br>Bk<br>Berkelium<br>251.080  | 98<br>Cf<br>Californium<br>251.080 | 99<br>Es<br>Einsteinium<br>[254] | 100<br>Fm<br>Fermium<br>257.095    | 101<br>Md<br>Mendelevium<br>258.1 | 102<br>No<br>Nobelium<br>259.101   | 103<br>Lr<br>Lawrencium<br>[262]  |

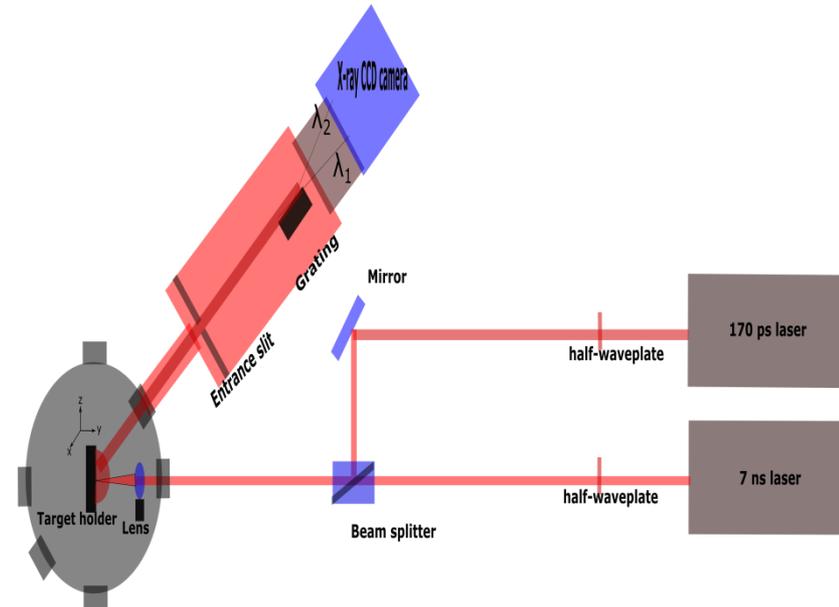
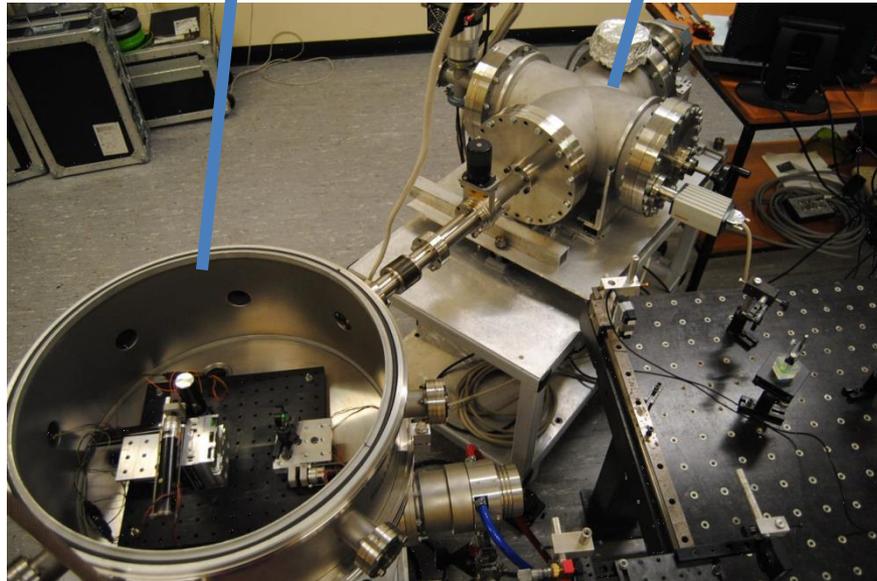
- Alkali Metal
- Alkaline Earth
- Transition Metal
- Basic Metal
- Metalloid
- Nonmetal
- Halogen
- Noble Gas
- Lanthanide
- Actinide

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## Experimental setup at University College Dublin

Vacuum chamber

Spectrometer

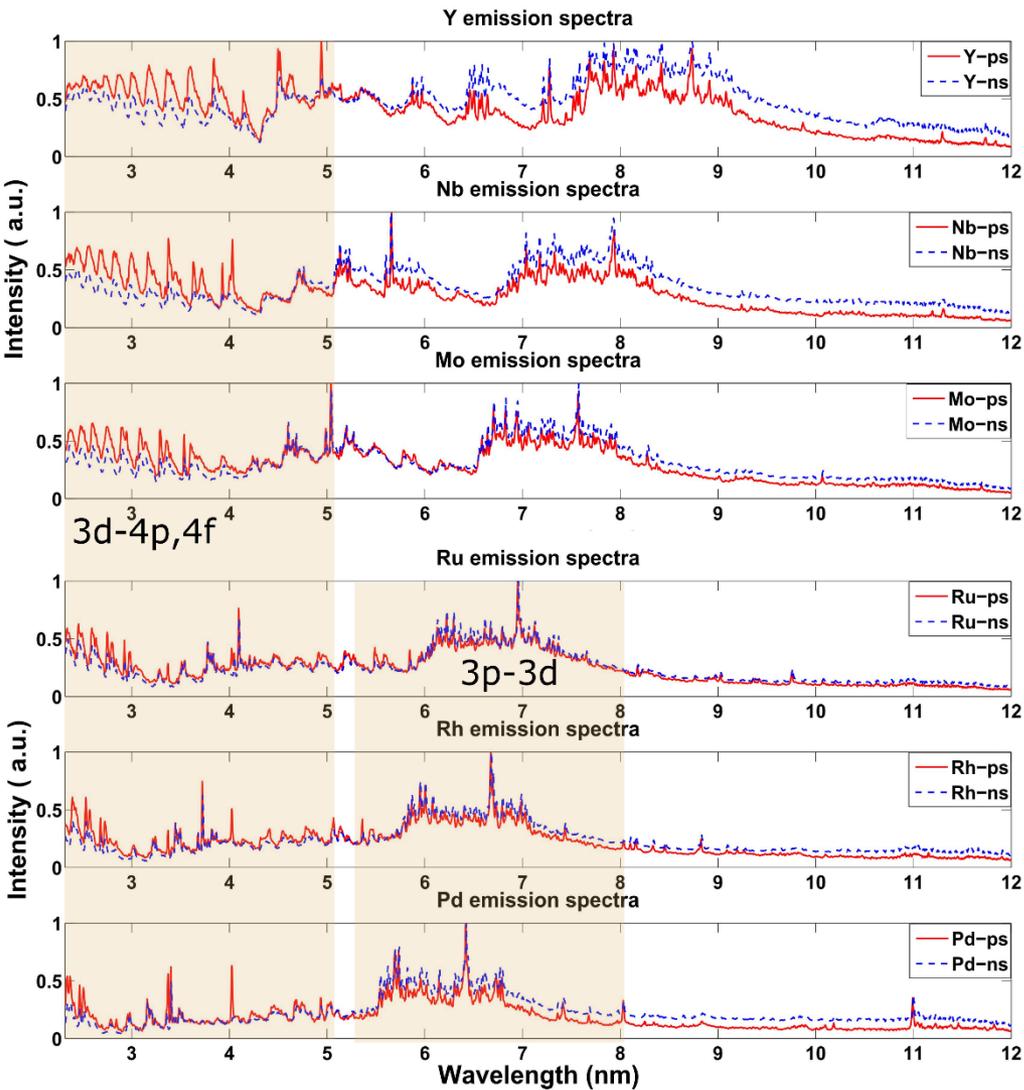


| Parameter                                  | ns laser                     | ps laser                     |
|--|------------------------------|------------------------------|
| Model                                      | Contium surelite             | EKSPLA                       |
| Maximum pulse energy (mJ)                  | $\approx 600$                | $\approx 227$                |
| Pulse length (ns)                          | $\approx 7$                  | $\approx 0.17$               |
| Maximum power density (W/cm <sup>2</sup> ) | $\approx 2.2 \times 10^{12}$ | $\approx 3.4 \times 10^{13}$ |

## Spectrometer

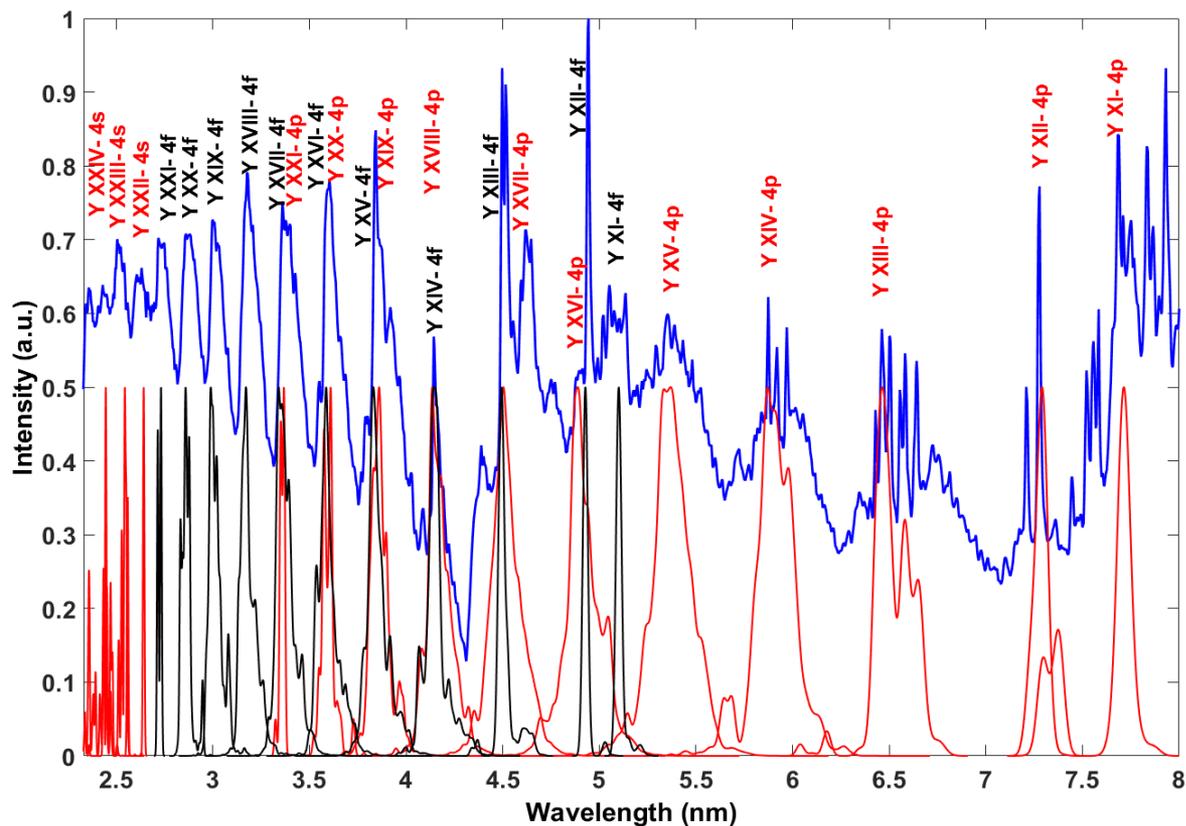
Parameter

|                        |   |
|------------------------|---|
| Spectrometer           | flat-field grazing-incidence            |
| Grooves                | 1200 per mm/variable line space grating |
| Spectral resolution    | $\approx 0.02$ nm                       |
| Wavelength uncertainty | $\approx 0.005$ nm                      |



- Emission spectra of 6 elements from 170 ps and 7 ns Nd:YAG laser pulses.
- Spectra are normalized to the highest intensity.

- The Cowan code models atomic spectra based on the superposition of configurations method, developed by Robert D. Cowan.
- The code numerically calculates radial wave functions in order to determine the transitions set by the user.
- The Schrodinger equation is then solved using the calculated wave functions, which outputs a set of oscillator strengths as a function of wavelength.
- The term energies, electrostatic, spin-orbit and exchange parameters can be scaled as an aid to interpreting the experimental spectrum.
- Calculated spectra using the Cowan code - Y, Nb, Mo, Ru, Rh and Pd : Water window source & BEUV source.



The experimental spectrum of yttrium (blue) with synthetic spectra obtained from Cowan code calculations.

(R. Lokasani et al, *J. Phys. B* 48 (2015), 245009)

- UTA has too many lines to identify individual transitions.
- Energy levels and spectral distributions can be parameterized statistically in terms of moments of the array (*Bauche, and Bauche-Arnoult, Phys. Scripta T40, (1992), 58*)
- The general  $n^{\text{th}}$  moment for transitions between configurations  $a$  and  $b$  is given by

$$\mu_n(a - b) = \frac{\sum_{m,m'} [|\langle m' | H | m \rangle - \langle m | H | m \rangle|^n |\langle m | D | m' \rangle|^2]}{\sum_{mm'} |\langle m | D | m' \rangle|^2}$$

where  $D$  is the electric dipole operator and sum runs over all states  $m, m'$  of configurations  $a$  and  $b$  respectively

- First moment  $\mu_1$  gives the average value of the weighted mean wavelength of the UTA. Width is  $\sigma = [\mu_2 - (\mu_1)^2]^{1/2}$

| Ion Stage | Transition                 | $\mu_1$ (nm) | Width (nm) | UTA Peak (nm) | Observed UTA range (nm) | Previously identified spectral range (nm) |
|-----------|----------------------------|--------------|------------|---------------|-------------------------|---|
| Nb XIII   | $3d^{10} 4s - 3d^9 4s 4p$  | 5.86         | 0.119      | 5.93          | 5.5-6.4                 | 5.7-6.5 <sup>[24,22,25,23]</sup>          |
|           | $3d^{10} 4s - 3d^9 4s 4f$  | 4.02         | 0.019      | 4.03          | 4-4.2                   |   |
| Nb XIV    | $3d^{10} - 3d^9 4p$        | 5.6          | 0.02       | 5.65          | 5.5-5.72                | 5.5-5.75 <sup>[26,24,21,22,25]</sup>      |
|           | $3d^{10} - 3d^9 4f$        | 3.91         | 0.017      | 3.92          | 3.92-4.1                | 3.9-4.5 <sup>[26,24,21]</sup>             |
| Nb XV     | $3p^6 3d^9 - 3d^8 4p$      | 5.14         | 0.069      | 5.13          | 4.8-5.4                 | 4.8-5.4 <sup>[24,28,27,21]</sup>          |
|           | $3p^6 3d^9 - 3d^8 4f$      | 3.62         | 0.037      | 3.63          | 3.5-3.84                |   |
|           | $3p^6 3d^9 - 3p^5 3d^{10}$ | 7.58         | 0.289      |               |                         | 7.3-8.1 <sup>[27,29,30,21,24]</sup>       |
| Nb XVI    | $3p^6 3d^8 - 3d^7 4p$      | 4.71         | 0.077      | 4.7           | 4.5-5                   | 4.49-4.9 <sup>[33,24]</sup>               |
|           | $3p^6 3d^8 - 3d^7 4f$      | 3.21         | 0.752      | 3.37          | 3.25-3.6                | 3.2-3.5 <sup>[24]</sup>                   |
|           | $3p^6 3d^8 - 3p^5 3d^9$    | 7.5          | 0.445      |               |                         | 6.8-8.7 <sup>[31]</sup>                   |
| Nb XVII   | $3p^6 3d^7 - 3d^6 4p$      | 4.33         | 0.076      | 4.35          | 4.1-4.6                 | 4.1-4.5 <sup>[24]</sup>                   |
|           | $3p^6 3d^7 - 3d^6 4f$      | 3.1          | 0.042      | 3.16          | 3-3.3                   |   |
|           | $3p^6 3d^7 - 3p^5 3d^8$    | 7.5          | 0.539      |               |                         | 6.9-8.75 <sup>[34]</sup>                  |
| Nb XVIII  | $3p^6 3d^6 - 3d^5 4p$      | 4            | 0.07       | 4             | 3.9-4.2                 |   |
|           | $3p^6 3d^6 - 3d^5 4f$      | 2.98         | 0.04       | 2.98          | 2.9-3.15                |   |
|           | $3p^6 3d^6 - 3p^5 3d^7$    | 7.5          | 0.599      |               |                         |   |
| Nb XIX    | $3p^6 3d^5 - 3d^4 4p$      | 3.71         | 0.062      | 3.72          | 3.55-3.91               |   |
|           | $3p^6 3d^5 - 3d^4 4f$      | 2.82         | 0.037      | 2.82          | 2.7-2.91                |   |
|           | $3p^6 3d^5 - 3p^5 3d^6$    | 7.47         | 0.636      |               |                         |   |
| Nb XX     | $3p^6 3d^4 - 3d^3 4p$      | 3.46         | 0.052      | 3.44          | 3.3-3.6                 |   |
|           | $3p^6 3d^4 - 3d^3 4f$      | 2.68         | 0.033      | 2.68          | 2.6-2.75                |   |
|           | $3p^6 3d^4 - 3p^5 3d^5$    | 7.47         | 0.651      |               |                         |   |
| Nb XXI    | $3p^6 3d^3 - 3d^2 4p$      | 3.23         | 0.042      | 3.22          | 3.12-3.33               |   |
|           | $3p^6 3d^3 - 3d^2 4f$      | 2.55         | 0.028      | 2.56          | 2.5-2.64                |   |
|           | $3p^6 3d^3 - 3p^5 3d^4$    | 7.49         | 0.643      |               |                         |   |
| Nb XXII   | $3p^6 3d^2 - 3d^1 4p$      | 3.02         | 0.029      | 3.03          | 2.9-3.13                |   |
|           | $3p^6 3d^2 - 3d^1 4f$      | 2.44         | 0.022      | 2.44          | 2.4-2.6                 |   |
|           | $3p^6 3d^2 - 3p^5 3d^3$    | 7.52         | 0.61       |               |                         |   |
| Nb XXIII  | $3p^6 3d - 4p$             | 2.84         | 0.01       | 2.84          | 2.7-2.9                 |   |
|           | $3p^6 3d - 4f$             | 2.33         | 0.007      | 2.34          | 2.2-2.3                 |   |
|           | $3p^6 3d - 3p^5 3d^2$      | 7.57         | 0.54       |               |                         | 7.2-8.12 <sup>[35]</sup>                  |

Weighted mean wavelengths, widths UTA observed range, observed wavelength peak from experimental spectra  
(R. Lokasani et al. J. Phys. B 48 (2015), 245009)

| Material               | Wavelength(nm) | Reflectivity (%) | Observed UTA Peak                  |
|------------------------|----------------|------------------|------------------------------------|
| Cr/V                   | 2.42           | 9                |                                    |
| Cr/Ti                  | 2.73           | 17               |                                    |
| TiO <sub>2</sub> /ZnO  | 2.74           | 29               | Mo XIX (3d-4f)<br>Ru XVIII (3d-4f) |
| Cr/Sc                  | 3.12           | 32               |                                    |
| Cr/Sc                  | 3.14           | 21               |                                    |
| Cr/Sc B <sub>4</sub> C | 3.15           | 32.1             | Mo XXI (3d-4p)<br>Pd XX (3d-4p)    |
| Cr/Sc                  | 3.35           | 10               | Y XVII (3d-4f)                     |
| Cr/Sc                  | 3.37           | 5.5              | Y XXI (3d-4p)<br>Nb XVI (3d-4f)    |

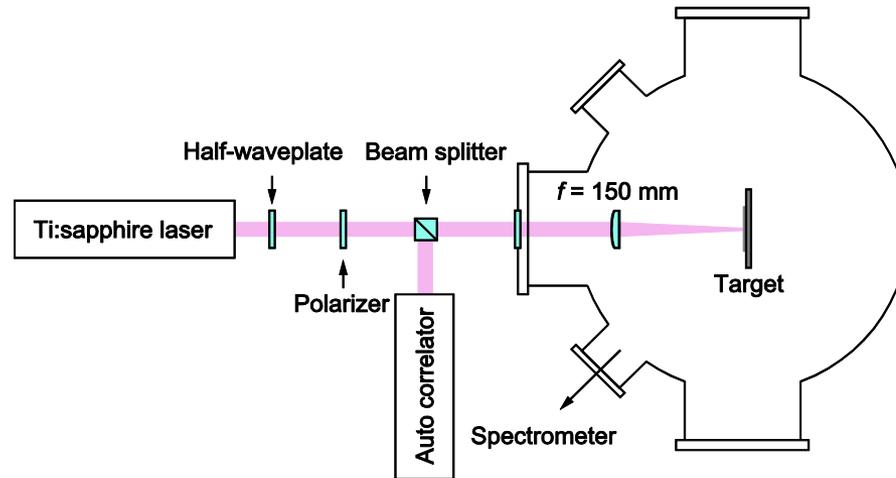
Peak wavelength and percentage reflectivity of different multilayer mirrors matched to UTA peaks from the present experimental data. (R. Lokasani et al, *J. Phys. B* **48** (2015), 245009)

## Section 2: femtosecond LPPs from 2<sup>nd</sup> row transition elements

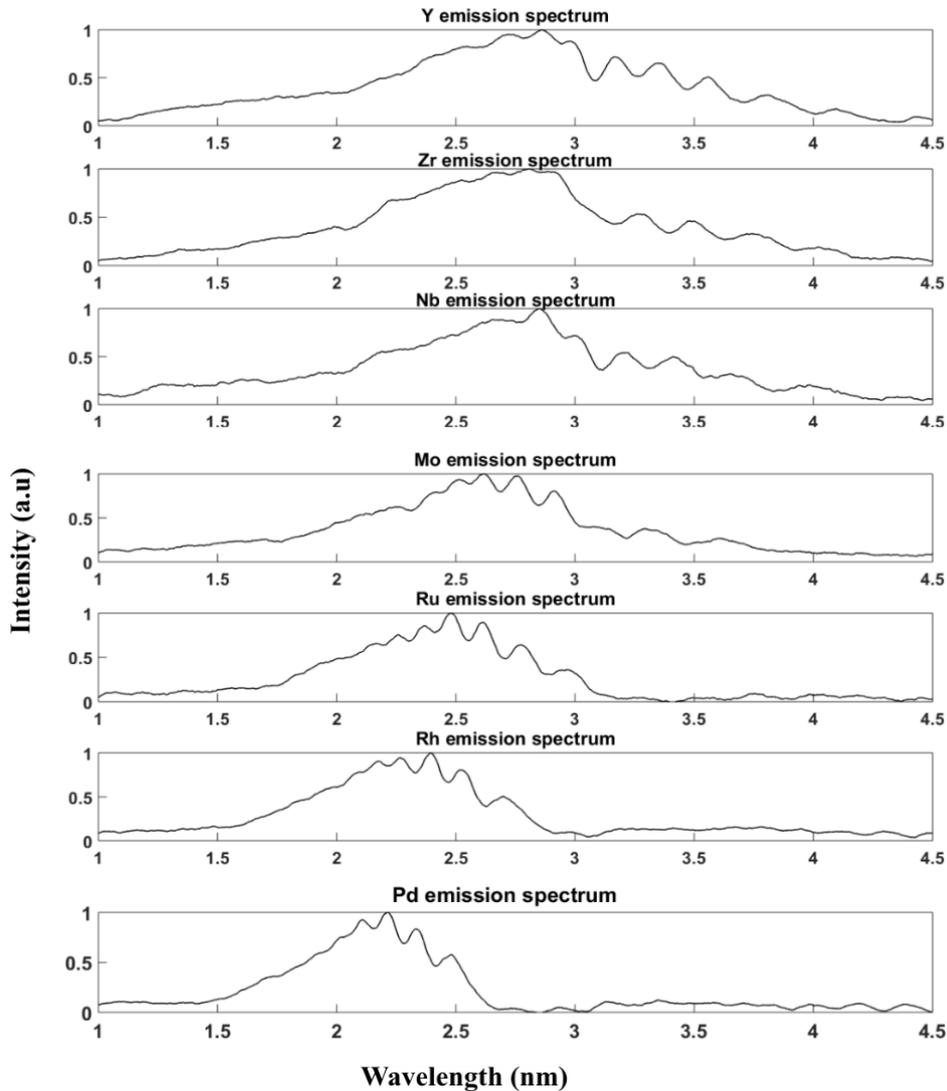
## Laser parameters

Czech Technical University in Prague

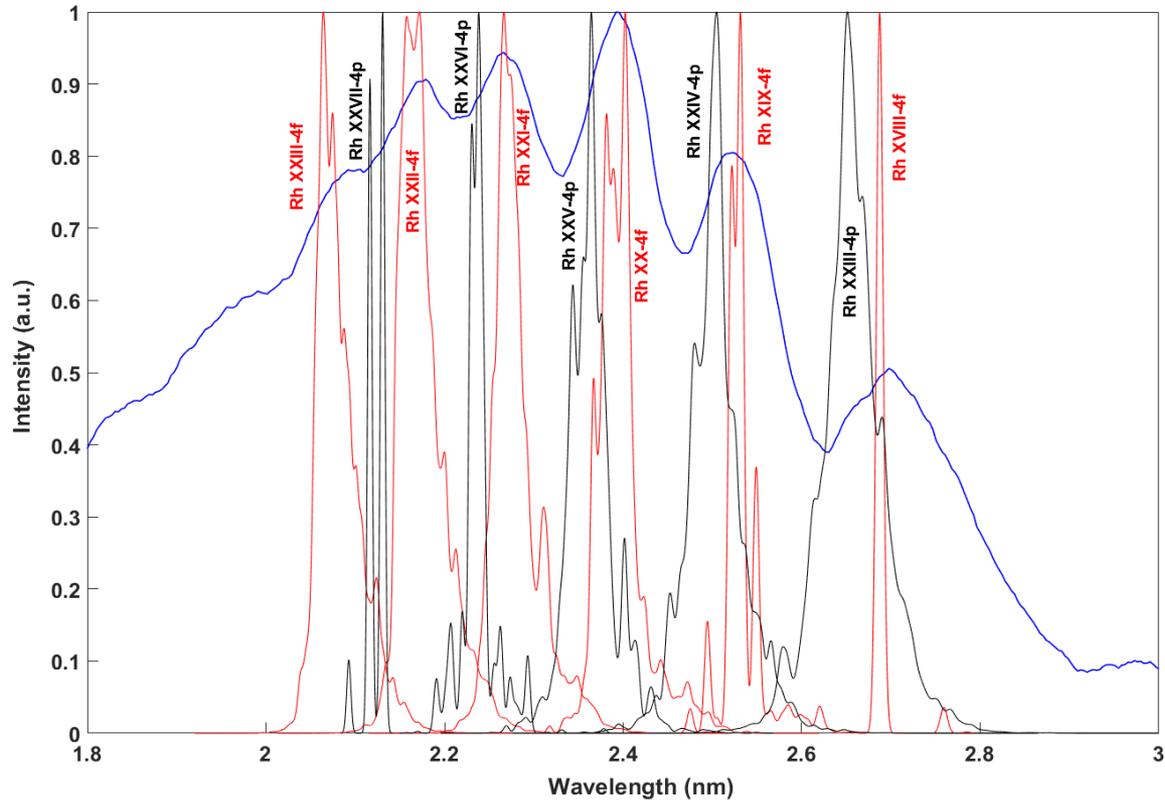
|                                    |                         |
|------------------------------------|-------------------------|
| Parameter                          | fs laser                |
| Maximum pulse energy (mJ)          | ≈ 10                    |
| Pulse length (fs)                  | ≈ 65                    |
| Laser                              | Titanium-Sapphire laser |
| Wavelength                         | 805 nm                  |
| Energy used in the experiment (mJ) | 4.5                     |



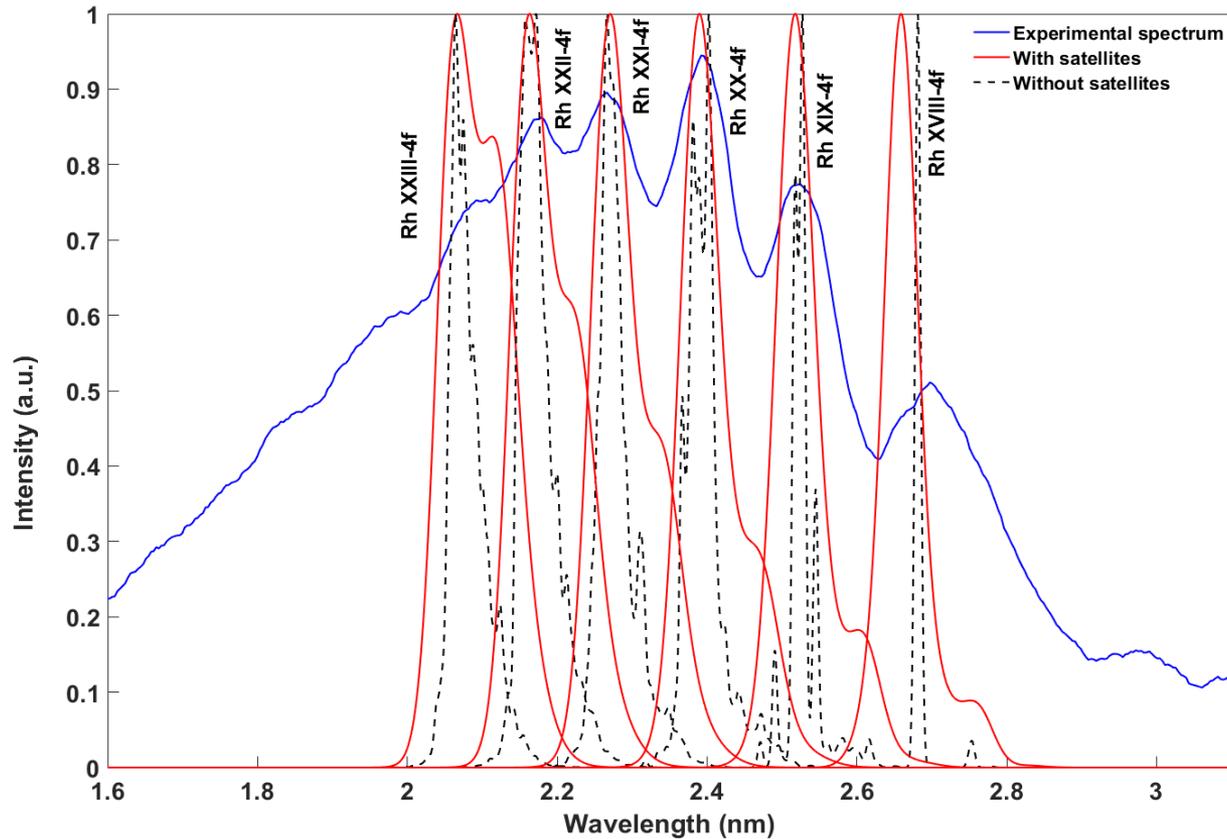
Schematic diagram of the experimental apparatus.



- Emission spectra of Y, Zr, Nb, Mo, Ru, Rh and Pd from plasmas produced by a femtosecond Titanium-Sapphire laser with a pulse width 65fs. (*R. Lokasani et al, J. Phys. B 50 (2017),*)



The experimental spectrum of Rh (blue) with synthetic spectra obtained from Cowan code calculations.



- The measured emission spectrum (blue) from a Rh target with spectra calculated by the Cowan code with satellites (red solid lines) and without satellites (black dashed lines).
- Included satellites were  $3d^{n-1}4s-3d^{n-2}4s4f$ .

(R. Lokasani et al. *J. Phys. B* 50 (2017), )

| Ion stage                   | Calculated mean wavelength (FAC) | Calculated UTA width (FAC) | Calculated mean wavelength (Cowan) | Calculated UTA width (Cowan) | Measured in present experiment | Measured in experiment [1] |
|-----------------------------|----------------------------------|----------------------------|------------------------------------|------------------------------|--------------------------------|----------------------------|
| Ruthenium 3d-4f transitions |                                  |                            |                                    |                              |                                |                            |
| Ru XVII                     | 2.94                             | 0.015                      | 2.91[1]                            | 0.01                         | 2.92                           | 2.92                       |
| Ru XVIII                    | 2.76                             | 0.025                      | 2.74[1]                            | 0.028                        | 2.74                           | 2.74                       |
| Ru XIX                      | 2.60                             | 0.029                      | 2.59[1]                            | 0.029                        | 2.59                           | 2.59                       |
| Ru XX                       | 2.47                             | 0.029                      | 2.46[1]                            | 0.029                        | 2.46                           | 2.46                       |
| Ru XXI                      | 2.34                             | 0.028                      | 2.34                               | 0.028                        | 2.35                           | -                          |
| Ru XXII                     | 2.24                             | 0.026                      | 2.23                               | 0.026                        | 2.24                           | -                          |
| Ru XXIII                    | 2.14                             | 0.023                      | 2.13                               | 0.024                        | 2.14                           | -                          |
| Rhodium 3d-4f transitions   |                                  |                            |                                    |                              |                                |                            |
| Rh XVIII                    | 2.69                             | 0.014                      | 2.67[1]                            | 0.013                        | 2.69                           | 2.68                       |
| Rh XIX                      | 2.53                             | 0.023                      | 2.52[1]                            | 0.025                        | 2.52                           | 2.53                       |
| Rh XX                       | 2.40                             | 0.026                      | 2.39[1]                            | 0.026                        | 2.39                           | 2.39                       |
| Rh XXI                      | 2.29                             | 0.026                      | 2.27                               | 0.026                        | 2.26                           | -                          |
| Rh XXII                     | 2.18                             | 0.025                      | 2.17                               | 0.026                        | 2.17                           | -                          |
| Rh XXIII                    | 2.08                             | 0.024                      | 2.07                               | 0.024                        | 2.08                           | -                          |
| Palladium 3d-4f transitions |                                  |                            |                                    |                              |                                |                            |
| Pd XIX                      | 2.48                             | 0.013                      | 2.46[1]                            | 0.013                        | 2.46                           | 2.47                       |
| Pd XX                       | 2.35                             | 0.021                      | 2.33[1]                            | 0.022                        | 2.34                           | 2.34                       |
| Pd XXI                      | 2.23                             | 0.023                      | 2.21                               | 0.023                        | 2.21                           | -                          |
| Pd XXII                     | 2.12                             | 0.024                      | 2.11                               | 0.024                        | 2.1                            | -                          |
| Pd XXIII                    | 2.03                             | 0.023                      | 2.02                               | 0.023                        | 2.01                           | -                          |

- Mean wavelengths and UTA widths (in nm) of 3d-4f transitions in Ru, Rh and Pd ions calculated with the FAC and Cowan codes.

(1) (R. Lokasani et al. *J. Phys. B* **48** (2015), 245009)

- Identified 3d-4p, 3d-4f and 3p-3d transitions in 6 elements from LPP spectra.
- UTA statistical approach was applied for isoelectronic series of all elements.
- The focus was on  $\Delta n=1$  3d-4p and 3d-4f transitions, which are more intense in the LPPs created with ps pulses and appear at shorter wavelengths.
- Transitions in Mo indicate that it might be particularly suitable for use with  $\text{TiO}_2/\text{ZnO}$  and Cr/Sc  $\text{B}_4\text{C}$  MLMs with reflectance peaks at 2.74 and 3.15 nm, respectively.
- Transitions from higher ionization states are clearly demonstrated in the spectra emitted from Ru, Rh and Pd targets heated by the femtosecond laser.
- The use of low to moderate energy fs lasers as potential high brightness sources for XUV metrology, is a topic worthy of further study.



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