



Electronics kinetics Induced by XUV Excitation in Warm Dense Gold

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Abstract

This report, in the context of the DESY Summer Student Program 2016, presents the experiment concerning the study of Electronics kinetics Induced by XUV Excitation in Warm Dense Gold, performed at FLASH between August 29th and September 8th 2016. The experiment consisted in the excitation of 4f electrons in gold using 50fs X-Ray laser pulses at 5.06nm. By hitting a gold foil with this intense and short pulse, one can manage to observe a state of matter called Warm Dense Matter, which is characterized by densities too high to be considered as a weakly-coupled plasma, and temperatures too important to correspond to condensed matter theory. This is somehow a regime between solid state and plasma, which is present in many fields such as astrophysics or fusion science. The goal of this experiment is to measure the dependence of the dielectric function on pump-probe delay, probe frequency and excitation energy density. The datas will be used to understand some kinetics processes, such as three-body recombination, Auger decay and electron-electron scattering. Through these datas, one can find out the effect of these processes on electron thermalization dynamics in WDM.

Group : FS-FL

Supervisor : Sven Toleikis

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1 Introduction

My work at DESY was mainly to assist my supervisor and the team working on the experiment (people from SLAC, CA, USA and from the University of Alberta, Canada). I actually helped them to prepare the whole experiment, beginning from nothing to finally get all the equipment ready. I was also present during the measurements, but not for data analysis.

The experiment consisted of shooting with soft X-Ray FEL femtosecond pulses on a thin gold foil. The result of such an intense pulse on matter is vaporization of matter almost instantly at our scale. But one can manage to probe what happens in an atomic scale with femtosecond pulses, which are way smaller in time than the typical vaporization time (few picoseconds). We are then capable to study a very strange regime of matter : the Warm Dense Matter (WDM). This regime is a transition between solid state and plasma, characterized by high densities and temperatures, which do not correspond to any theoretical model so far. The main problem is we are not able anymore to make some common assumptions that simplify the equations of state, and wave equations. This is why it is very important to study this regime, to get some benchmark data in order to develop models.

Warm Dense Matter is a very important regime in physics. Actually it is present in a lot of fields such as astrophysics - we can find WDM in inner cores of some planets or on the surface of neutron stars -, or fusion science - WDM regime is reached at the very beginning of inertial confinement fusion. By understanding the properties of matter in this regime, we can progress in many fields involved with WDM.

The goal of the experiment performed at FLASH was to measure the dielectric function of warm dense gold as a function of time, by measuring its reflectance and transmittance at different wavelengths. The expected data will be used as benchmarks for future experiments and models.

In this report I will explain first what Warm Dense Matter is, then I will describe the experiment, its goal and technical aspects, and finally I will resume my work at FLASH and what I learned from my stay here at DESY. In the appendix one can find some additional pictures, photos and graphs.

2 Context and overview

2.1 Warm Dense Matter (WDM)

WDM is a typical regime of matter characterized by temperatures near Fermi level - approximately between 1eV and 100eV ($1\text{eV} \Rightarrow 11,605\text{K}$) -, and densities between a tenth and ten times density of solid state. WDM is an important topic because it is present in astrophysics (inner core of some planets, etc.) and in fusion applications. This is a very complex regime for theoretical modelisation, because neither condensed matter theory, nor simplifying assumptions of plasma physics are valid. For example state equations of WDM, which link pressure, density and temperature are not well known yet. On Figure 1, one can see the typical phase diagram where the WDM regime zone has been highlighted.

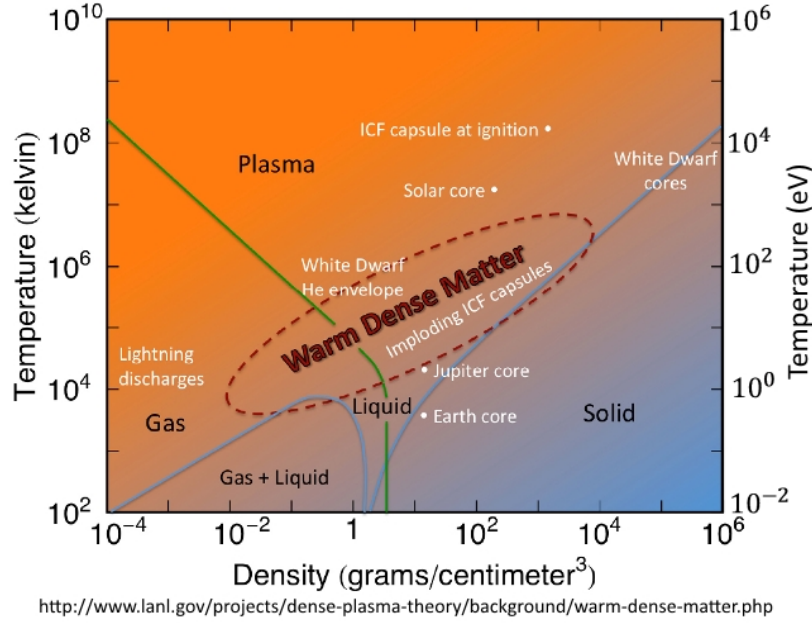


Figure 1: Warm Dense Matter approximated phase diagram

In WDM, the temperatures can be sufficient to ionise atoms, and for matter to be partially coupled and degenerated. For this point we need to introduced two parameters :

- **The ion-ion coupling parameter** Γ_i is given by potential energy V over kinetic energy E_k of ions in equation 1 :

$$\Gamma_i = \frac{V}{E_k} \quad (1)$$

Assuming that ions have the charge Ze and 3 degrees of freedom, V and E_k are given by equations 2 and 3 respectively :

$$V = \frac{(Ze)^2}{4\pi\epsilon d} \quad (2)$$

$$E_k = \frac{3}{2}k_B T_i \quad (3)$$

where ϵ is the dielectric function, k_B is the Boltzmann constant, T_i is the ions temperature and d is the mean distance between ions, approximated to the ionic density n to the power $-1/3$. We can now rewrite the equation 1 as :

$$\Gamma_i \simeq \frac{(Ze)^2 n^{1/3}}{6\pi\epsilon k_B T_i} \quad (4)$$

This parameter Γ_i gives us a clue on how coupled is the matter. Typically for $\Gamma_i \ll 1$, ions are weakly coupled and potential terms can be neglected, such as in a standard plasma.

- **The electrons degeneration parameter** Λ_e consider the wave nature of the electrons, and is given by the de Broglie wavelength of the electrons over the mean distance d approximated to the ionic density n to the power $-1/3$. The de Broglie wavelength of the electrons λ_{dB} is given by the equation 5 :

$$\lambda_{dB} = \frac{h}{\sqrt{2m_e k_B T_e}} \quad (5)$$

where m_e is the electron's mass. The equation of the degeneration parameter is then :

$$\Lambda_e = \frac{\lambda_{dB}}{n^{-1/3}} = \frac{h n^{1/3}}{\sqrt{2m_e k_B T_e}} \quad (6)$$

This parameter gives us a clue on how electrons must be described. For high densities and/or low temperatures, $\Lambda_e \ll 1$, the interactions between electrons are then dominated by the coulombian interaction, and are described by the Maxwell-Boltzmann distribution. For low densities and/or high temperatures, $\Lambda_e \geq 1$, then the electrons must be described with quantum equations, as they are degenerated and Pauli principle applies. They are then described by the Fermi-Dirac distribution, given by equation 7 :

$$f(E) = \frac{1}{1 + \exp\left(\frac{E - E_F}{k_B T_i}\right)} \quad (7)$$

where E is the energy level and E_F is the chemical potential or the Fermi energy.

For WDM, Γ_i and Λ_e are close to 1, so this is why this is a very difficult regime to describe, because one must consider the electrons in a quantum way and cannot use the assumptions of weakly-coupled plasma physics.

2.2 Experiment at FLASH

In this section I will present the processes of the experiment, its goal, and the technical aspects involved.

The experiment relies on the excitation of 4f electrons in gold ($Au : [Xe]4f^{14}5d^{10}6s^1$) using a 50fs laser at 5.1nm (i.e. 245eV). The main goal here is to measure the dependence of the dielectric function ε on pump-probe delay t , probe frequency ω and excitation energy density $\Delta\epsilon$. The datas will be used as benchmarks to find out the role of basic kinetic processes including three-body recombination, Auger decay and electron-electron scattering on the electrons thermalization in Warm Dense Matter.

The experiment took place at the Beam Line 3 (BL3) at FLASH. We used a 20nm-thick gold foil as sample, resting on a 3cm × 5cm, 1.1mm thick stainless steel plate with 105 550μm diameter holes as targets. The whole experiment took place in a vacuum chamber at pressure around 6×10^{-8} mbar. The total shot time was 80 hours, with 2 minutes/shot, so for a total of 2400 shots, or potential data points. The Figure 2 is a scheme of the CUBE setup (area of the vacuum chamber where the sample was set). On this figure are represented the sample (Gold foil), the FEL beam path, the probe path, the Fourier Domain Interferometry (FDI) beam path and the optical devices (objectives and band-pass filter).

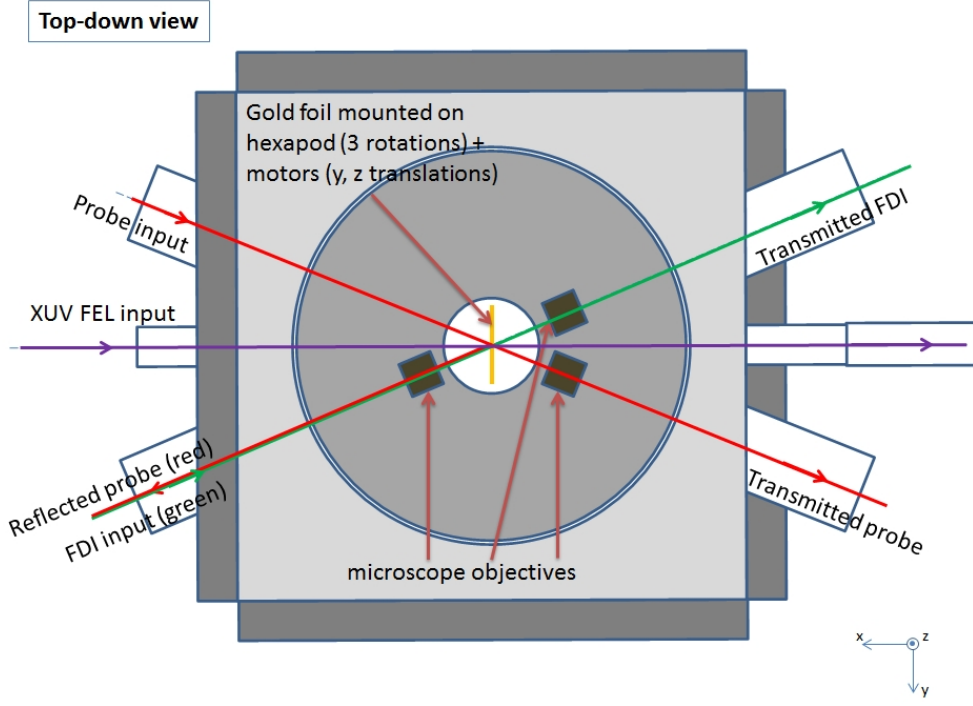


Figure 2: Scheme of the CUBE setup

To probe the sample, we used an optical laser at 800nm. This optical laser went first by a grating compressor, which is able to reduce the pulse duration up to 50fs via a specific optical path. This pulse was split in two beams. One went by an Optical Parametric Amplifier (O.P.A.) which can amplify the beam and change it's wavelength to a specific one. We used 10 different wavelengths using this method to probe the sample, between 400nm and 1000nm. This beam was then injected in the cube as shown in the Figure 2. The probes have been used to measure the reflectance $R(t, \omega, \Delta\epsilon)$ and the transmittance $T(t, \omega, \Delta\epsilon)$ of the shot sample at these wavelengths, using CCD cameras. The other one was used as the Fourier Domain Interferometry (F.D.I.) beam. This 800nm 50fs beam was used to detect the expansion of the heated gold sample. We also used a Ce:YAG crystal screen coupled with a CCD camera to measure XUV fluorescence and know radiation loss in the sample.

The results were then used as inputs to solve numerically Helmholtz equations - which describe electromagnetic wave propagation in a uniform dielectric slab - to finally yield the dielectric function :

$$\varepsilon(t, \omega, \Delta\epsilon) = \varepsilon_r(t, \omega, \Delta\epsilon) + i\varepsilon_i(t, \omega, \Delta\epsilon) \quad (8)$$

The real part $\varepsilon_r(t, \omega, \Delta\epsilon)$ represent the "energy storage" and the imaginary part $\varepsilon_i(t, \omega, \Delta\epsilon)$ represent the "energy loss" in the dielectric material.

The Figure 3 is a sketch of the experimental setup. One can notice the grating compressor, the Optical Parametric Amplifier (O.P.A), the CUBE, CCD cameras, YAG crystal and paths of the differents laser beams.

3 My work at FLASH

During my stay at DESY, I took part in a large-scale experiment, which permitted me to be aware of what is the daily work of a researcher. Indeed we have had to set up the entire experiment, from nothing to the final stage. My job was to assist my supervisor and the team during the set up and the beam-time.

3.1 Before the beam-time

Before the beam-time (from July 19th to August 29th), the work was to set up the whole experiment, beginning from nothing to finally have everything done as good as possible. The different steps of the work were the following (in chronological order) :

- Assembly of the different external parts of the vacuum chamber - vacuum pumps, pressure sensors, flanges - first vacuum test without internal parts : $6 \times 10^{-8} \text{ mbar}$
- Alignment of the center of the cube and the FEL beam using the alignment laser of the BL3 and motors of the chamber (these motors allow precise motions along x, y and z axis).
- Setup of the laser tables, where the pulse compressor, the OPA, mirrors and focusing lenses were installed (see Figure 3)
- Setup of the internal parts of the chamber - hexapod, motors for linear translations along y and z axis, sample plate, microscope objectives (see Figure 2)
- Triggering the CCD cameras to the DAQ (data acquisitions system)
- Connecting the vacuum interlock (pressure control) to the control system. The vacuum interlock controls the pressure inside the vacuum chamber, and allows the fast shutter opening only if this pressure is below a specific threshold, set by users. This process is a 3-times process (3 different thresholds control). This vacuum interlock system is an additional safety system which keeps the fast shutter closed if a vacuum problem happens.
- Connecting the FLASH2 fast shutter control to FLASH1 computers.
- Optimization of the different parts (O.P.A., cameras positions, etc.)

See in the appendix part 5 on figures 8 to 15 are some photos of the experimental setup we prepared before the beam-time.

3.2 During the beam-time

During the beam-time (from August 29th to September 6th), the work was to acquire data as mentioned in part 2.2. The beam-time was divided in $3 \times 48h$ shifts.

The beginning of the beam-time was somehow disappointing, because the FEL beam has problems (missing gas attenuator, vacuum problems with the electron gun). We had to wait until the operators fixed the problems and gave us the beam back. But then another problem was that the beam energy was neither high nor stable enough ($[15 \pm 10] \mu J$). Therefore this was enough to make some calibration shots. The real data acquisitions began on the 3rd day shift at 22^h49, with a pulse duration of 58 fs, a pulse energy of $[30 \pm 9] \mu J$, at $[5.07 \pm 0.03] \text{ nm}$ and $[0.25 \pm 0.02] \text{ nC}$ charge.

To acquire datas, the team made a LabView program that proceed the following :

- Take pictures of the sample transmission, reflection and of the YAG screen view before the shot (plus record all other datas such as energy beam, wavelength and charge)
- Open the fast shutter to do a one-shot on the sample with the FEL
- Take pictures on-shot of the sample transmission, reflection and of the YAG screen view (plus record all other datas such as energy beam, wavelength and charge)
- Move the plate to the next sample (among the y axis)

With this program, the user can set up the amount of shots he wants to proceed and the starting position (y,z coordinates setup). He can also change the pump-probe delay on then delay control system of FLASH. As only pictures of before the shot and on-shot were taken and displayed to the user, we also added a real time camera display of the sample (10 fps, mono-8 display) to have a live see of what happened in the CUBE.

During the datas acquisitions, two persons were needed at least. One was managing the Lab-View program while the other was setting the pump-probe delay and writting the acquisition parameters - date and time of acquisition, shots numbers, pump-probe delay and starting position (y,z coordinates) - and any other comment on the acquisition on a logsheet. Both were also looking at the pictures, parameters display and live images to notice any interesting event.

On the Manual Control window figure 4, X, Y, and Z controls are the controls of sample rotation around X, Y and Z axis respectively. U, V and W are parameters to set the hexapod reference position. A and B are respectively the controls to translate the sample among Z and Y axis. Units are in mrad for rotations and mm for translations. We can also see displays of each cameras view.

On the Scan window figure 5, we can see at the top images of the sample's transmission and reflection, and YAG image (FEL transmission) before the shot. Under these images are the same views taken on the FEL shot. On this window the user can also enter parameters for the automatic shot process (number of shots, starting position, direction of sample motion, etc.).

On the figure 6 is the SASE energy display in real time. On the left graph is the long term SASE energy display and on the right graph is the short term one. This window was used to check if the beam energy was stable and high enough to start a shot sequence.

On the figure 7 is the logsheet and delay control window. On the left side we can see the logsheet. The user had to collect the shot sequence datas such as time, shift number, run number, shots numbers, number of good shots in the sequence, starting positions and any other comment. On the right side we can see the delay control panel with which we can set the pump-probe delay (in picoseconds). Also at the top is the LOLA bunch profile monitor, that gives us the beam profile and pulse duration.

During the beam-time we had to fix many problems, such as program bugs, cameras malfunctions or decalibration problems. Most of the time, problems were not on our side but were FEL problems (unstable and/or wrong beam energy, wrong wavelength, no beam at all).

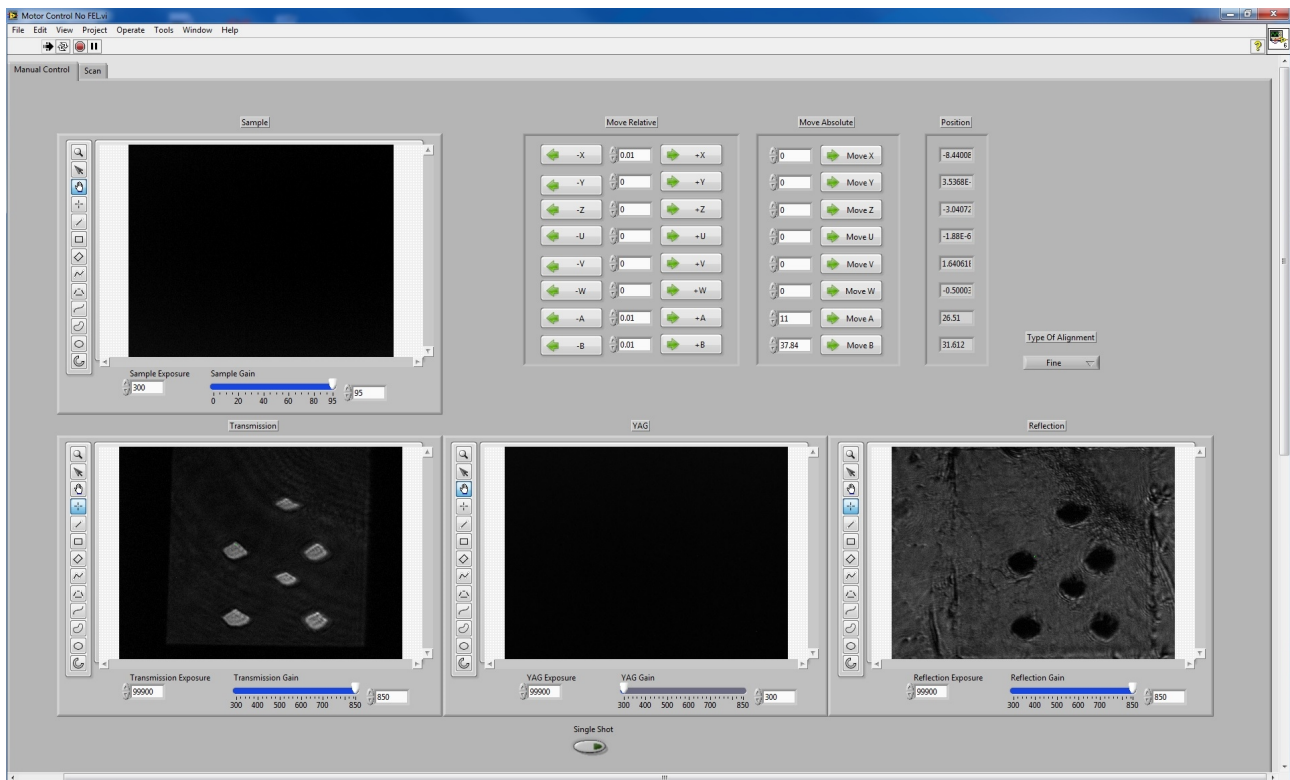


Figure 4: Screenshot of the Manual Control window

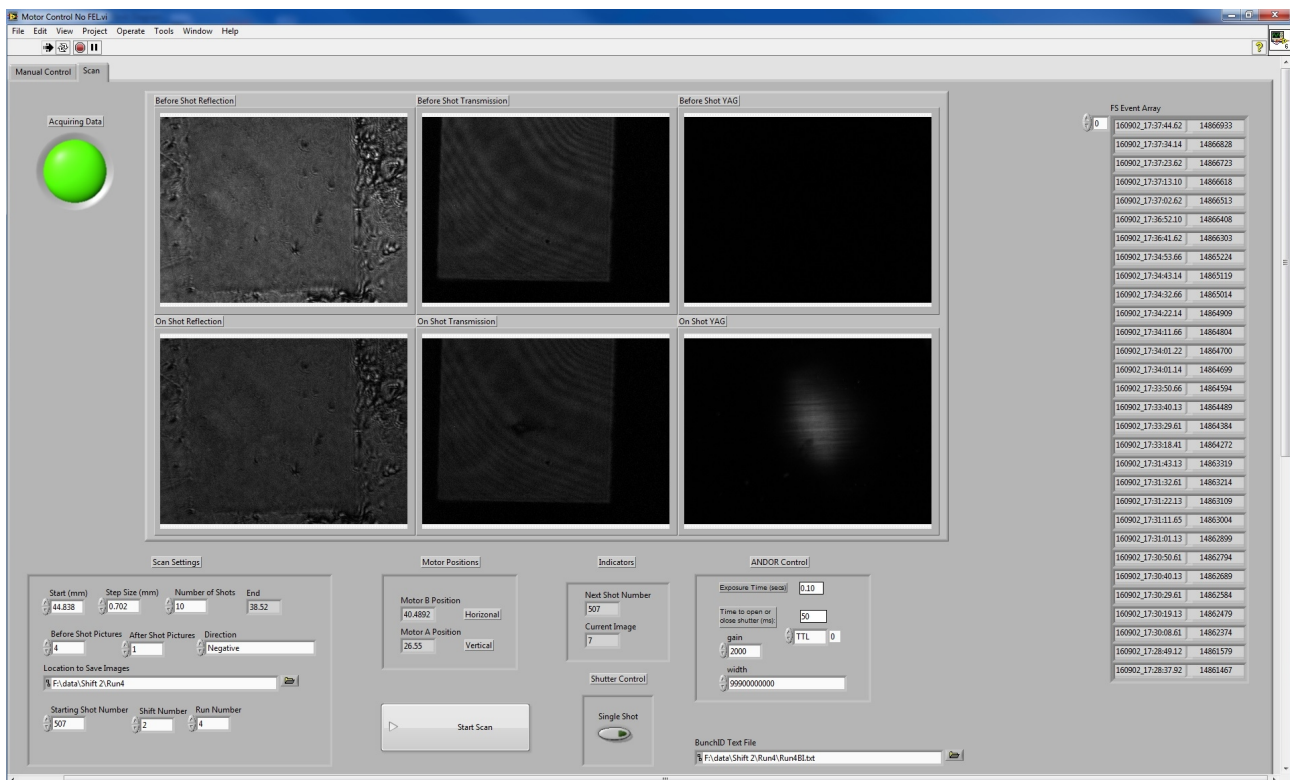


Figure 5: Screenshot of the Scan window

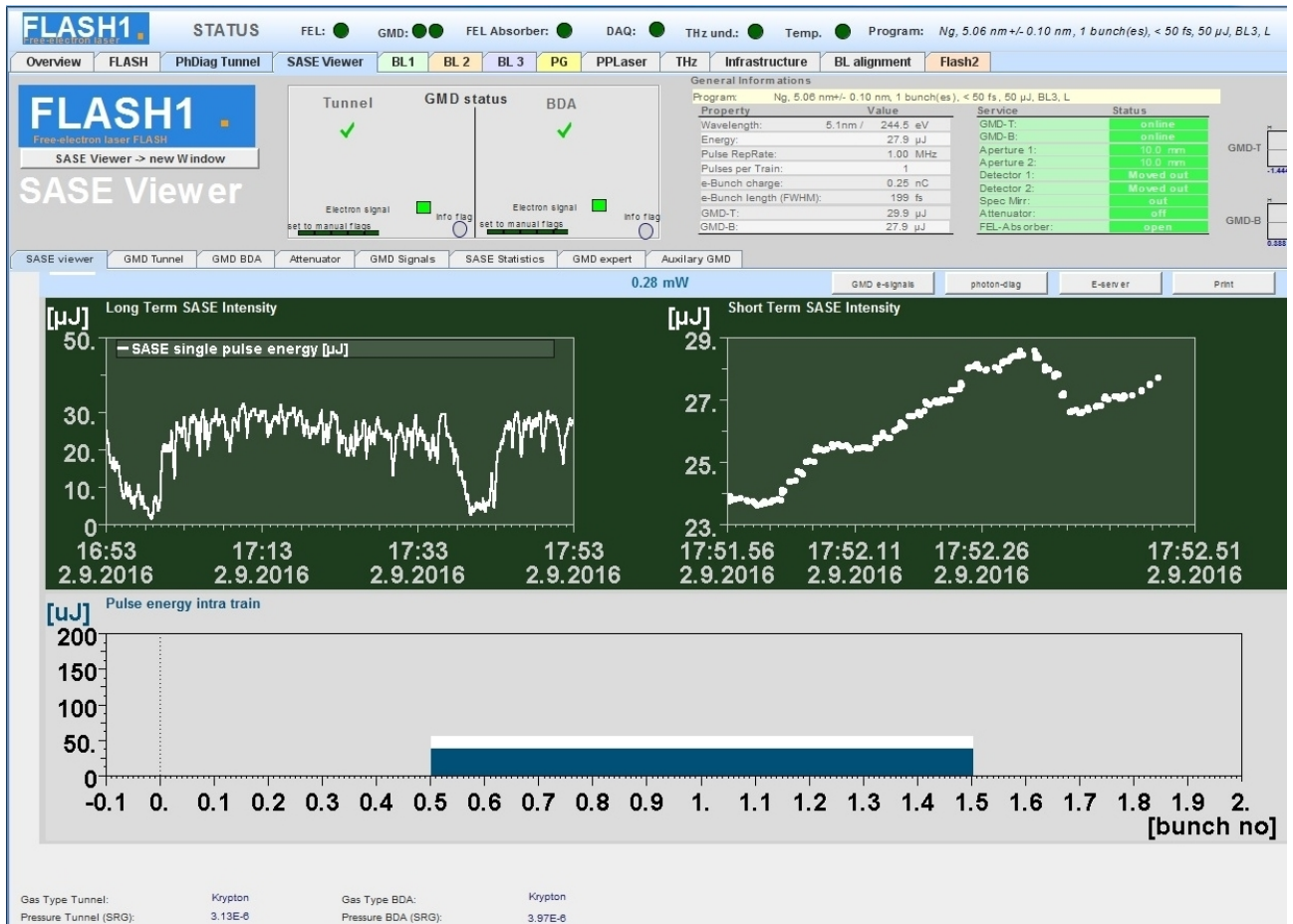


Figure 6: Screenshot of the SASE energy graph display

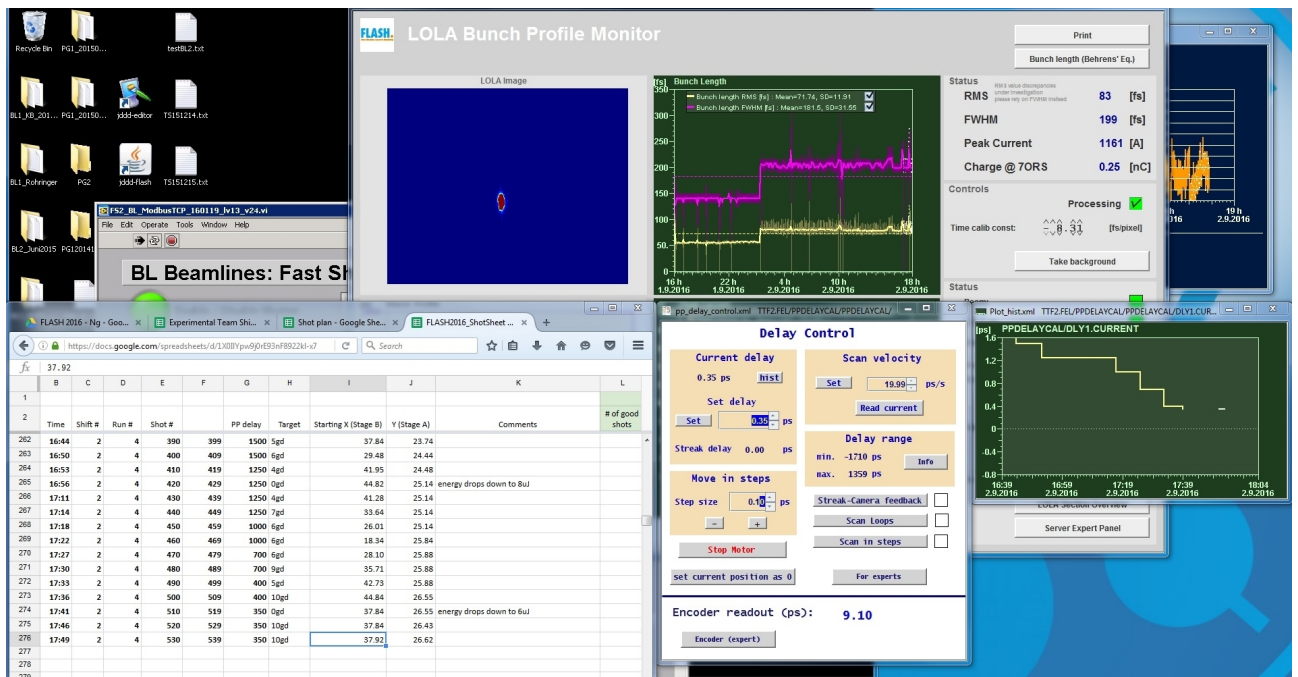


Figure 7: Screenshot of the delay control, pulse duration display and logsheet window

4 Acknowledgements

My Summer Student program at DESY was a unique experience for me. Indeed I have learned a lot on what is the daily work of a researcher, and I also took part in a very large scale experiment. The many problems we had to fix added to the on-time data analysis enriched a lot my experience of real scale experiments, because we always have to expect the unexpected and we have to react appropriately to the events we encounter.

That is why I would like to thank my supervisor Sven Toleikis for having me in his team during my stay. I really liked to work with him on this experiment, because I was able to appreciate how much work such an experiment requires. It was very useful for me because I want to work as a researcher after my Master and PhD.

I would also like to thank the Summer Students Program organizers for allowing me to join this very interesting program.

I want to thank also the team I worked with for several weeks. The talks we had permitted me to understand and to learn some processes and methods, and many vocabulary.

I also thank all people who were working around me at FLASH (Mark, Torsten, Nikola, Andreja, Cédric, Erlund, Sören, Stefan, and many others) for their help or sometimes just talking. I really appreciated working among them.

And finally I would like to thank all the summeries, who have made this summer a unique one among all. Thanks Sandra for enduring me during these two months. Thanks Orestis, my co-Muser of the summer. Thanks Emma, Jasmine, Ivette, Mi, Elsa, John, Magnus, Joe, Sean, Hans, Guillermo, Gergo, Colm, Dominik, Max and all the others for these great moments we shared and for the weekly barbecues.

Thank you all.

Adrian

5 Appendix

In this part are some photos of the experimental setup and camera views during the beam-time.

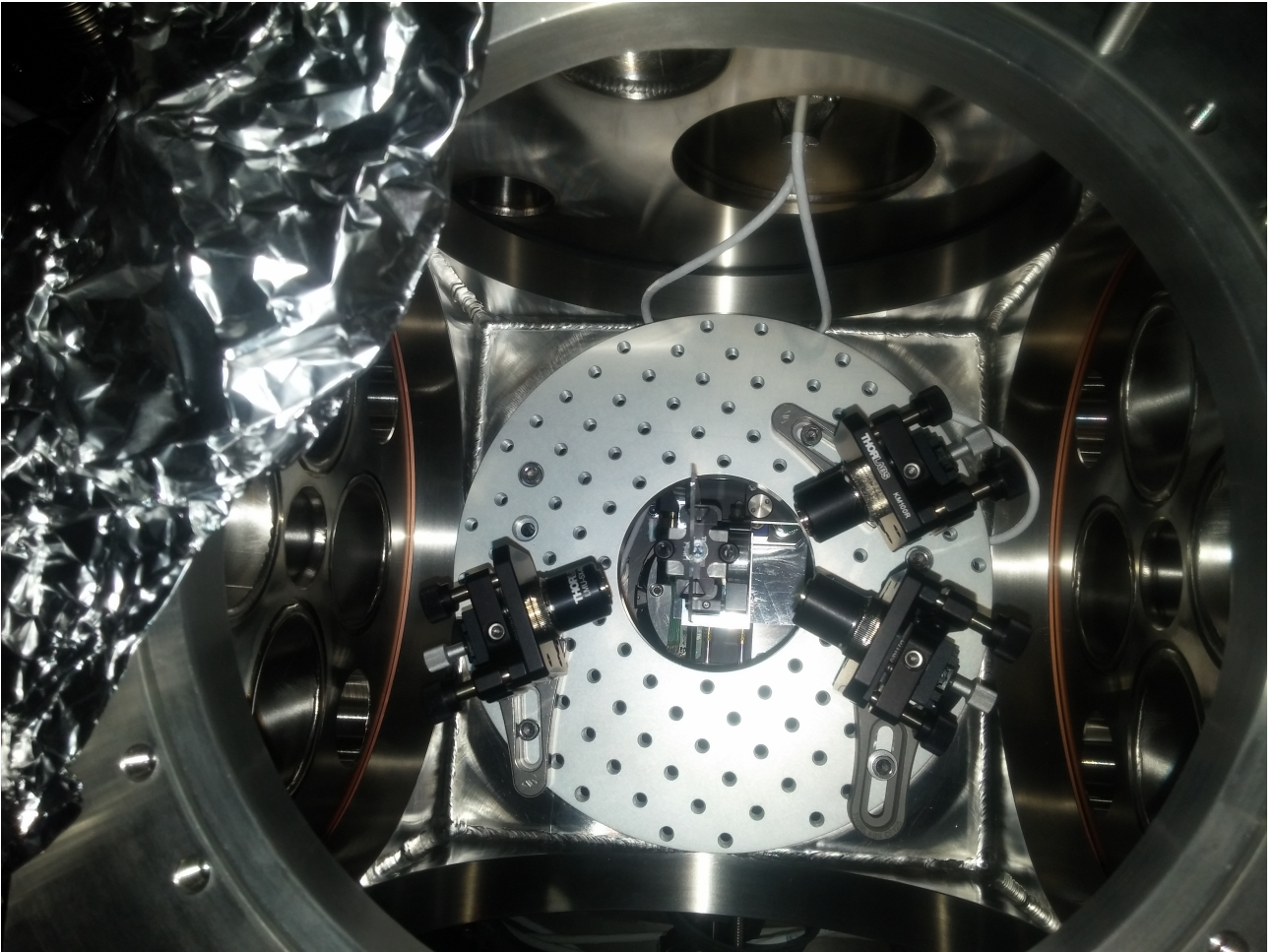


Figure 8: Top down view photo of the inside of the CUBE



Figure 9: Photo of the inside of the CUBE - ready to experiment



Figure 10: Photo of the inside of the O.P.A.

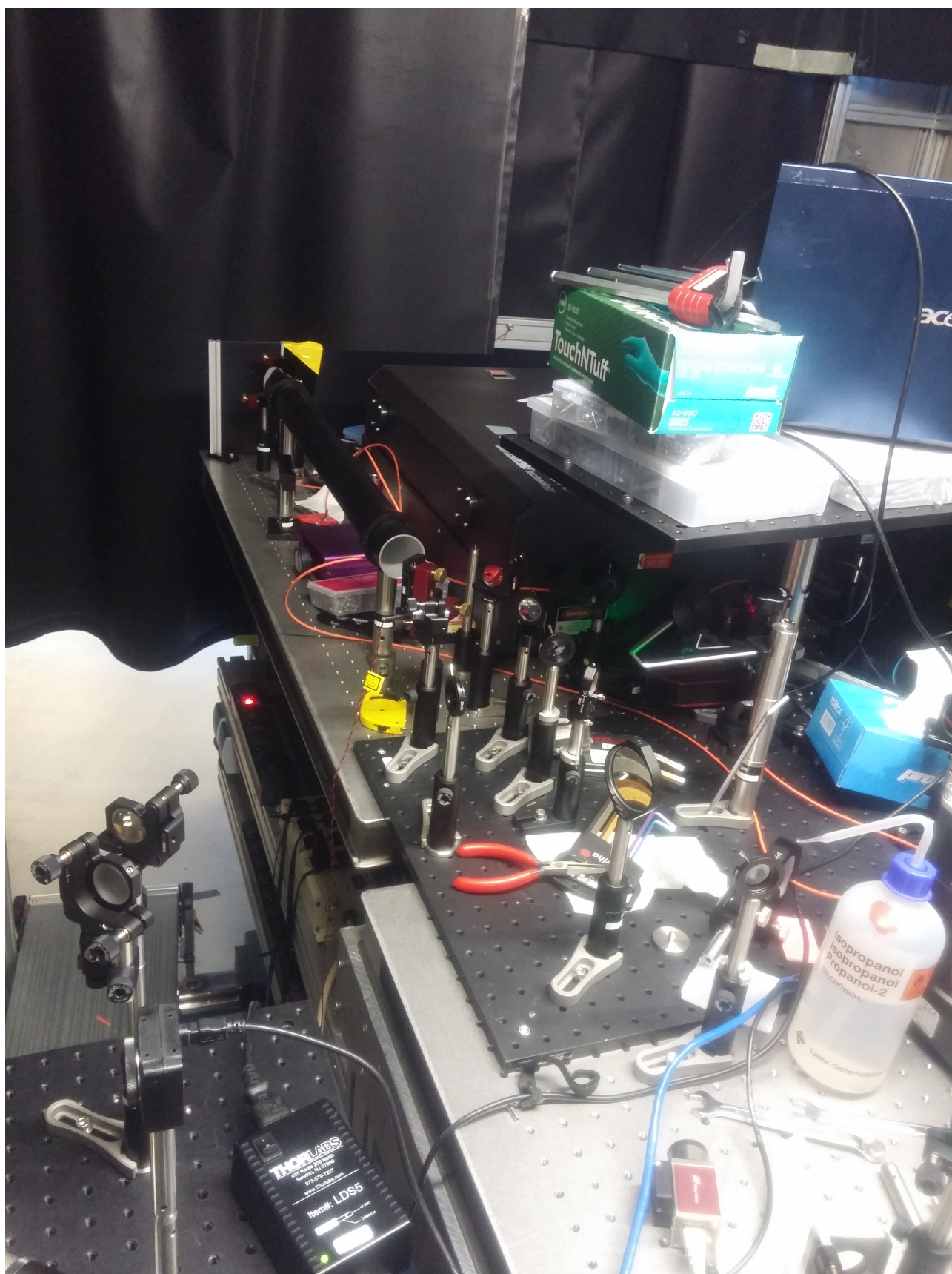


Figure 11: Photo of the OPA optical table and breadboard



Figure 12: Delay line and FDI beam path

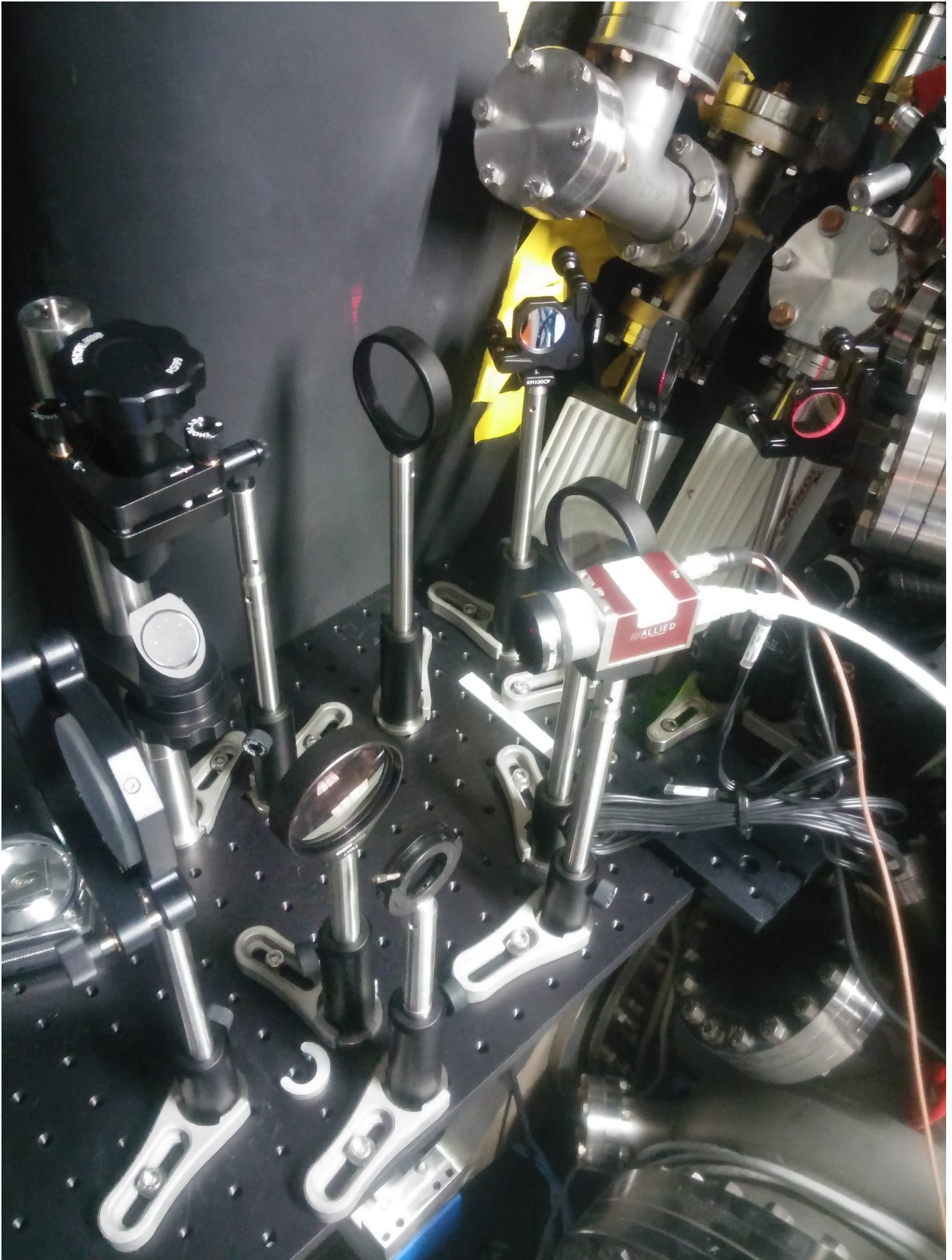


Figure 13: Reflection CCD camera (front) and FDI input (back-right)

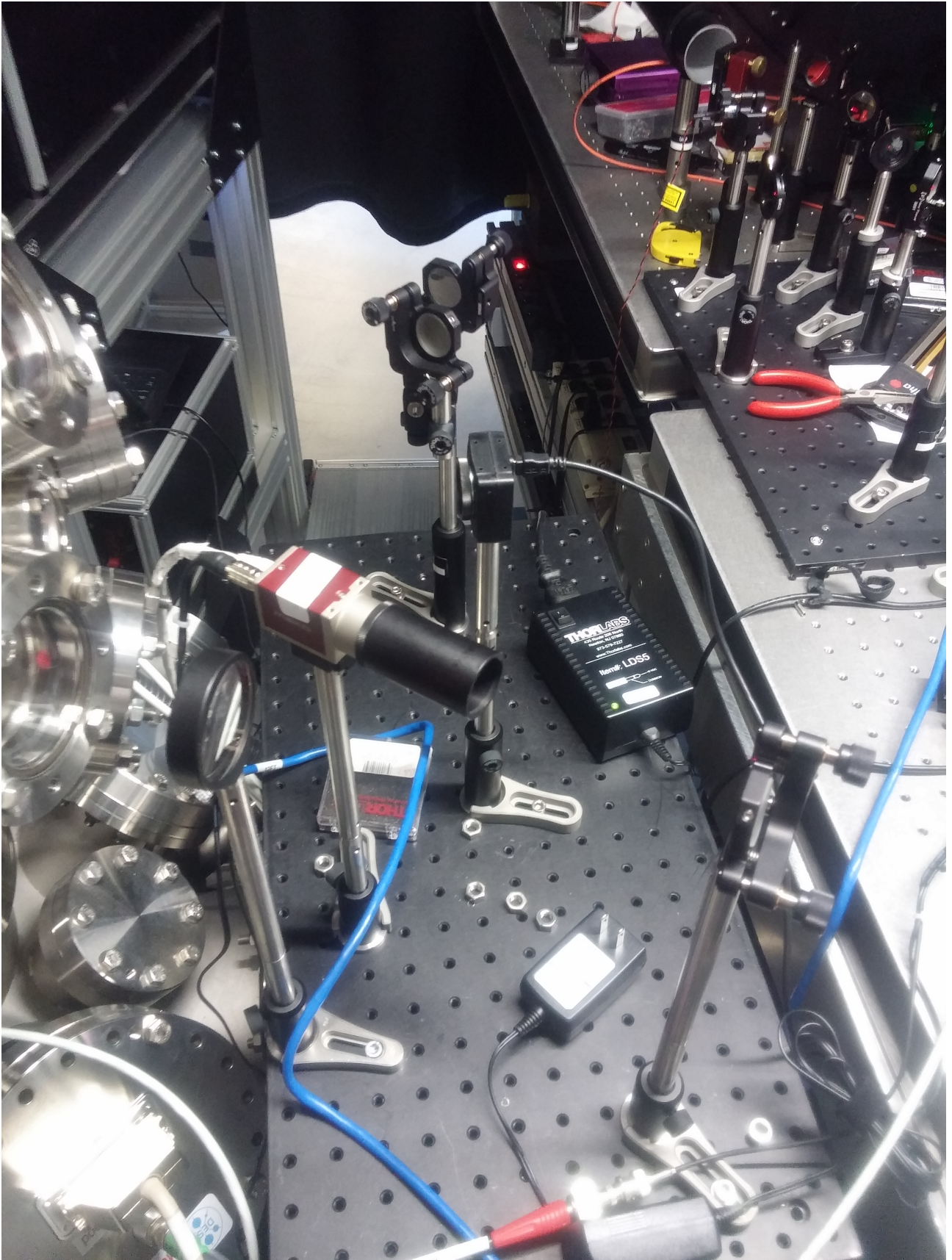


Figure 14: Transmission CCD camera (front) and FDI output (back)

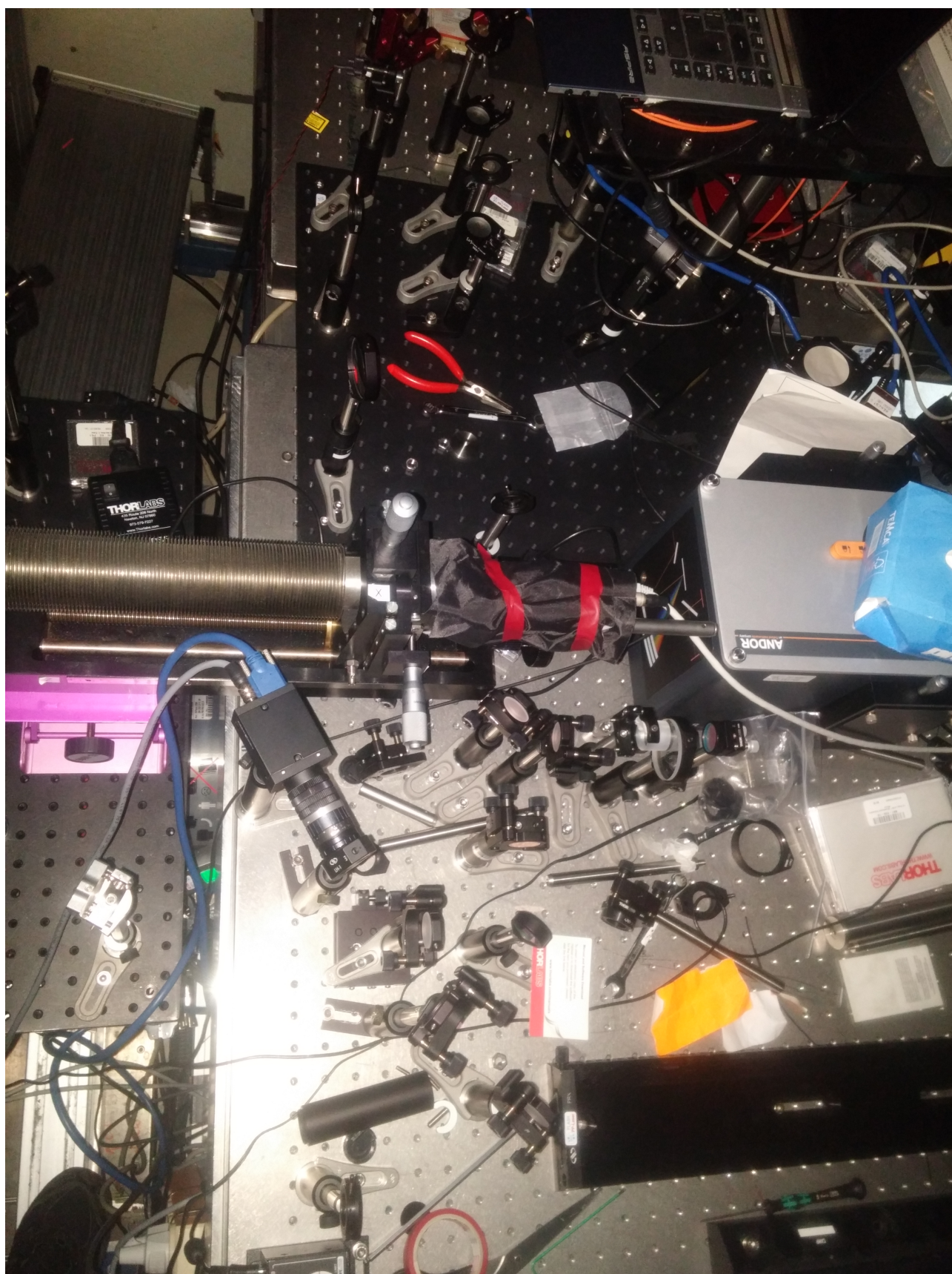


Figure 15: Ce:YAG (left-center) and spectrometer (right)

6 Bibliography and references

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