26 September 2024

Summer Student Programme Report: Rare Isotopes in Diamond for Fundamental Symmetry Tests

Zeno Maesen

Supervisor: Dr. Shandirai Malven Tunhuma (KU Leuven)

Summary

This document is a report on the work I performed in the Quantum Photonics Lab at ISOLDE, CERN during a 13 week period in the context of the CERN Summer Student Programme. First, the need for a photoluminescence setup is motivated in the context of implanting rare isotopes in diamond to perform fundamental symmetry test using permanent electric dipole moments. Then the existing setup and its issues are outlined. Lastly, a short overview of the software that I developed for improved control of the setup is given. A more detailed account of the software and its capabilities can be found in the manual I wrote, which is added in appendix.

Contents

1 Introduction

In modern day physics there is a great interest for any interaction that violates charge conjugation parity (CP) symmetry, as these are crucial in explaining the matter-antimatter asymmetry we observe in our universe. Importantly, the Standard Model (SM) has some amount of CP violation, but much too little to explain the observed asymmetry. Different extensions to the SM have been proposed that introduce new sources of CP violation. To test these extensions, a better understanding of the amount of CP violation in our universe is crucial. Various ways to measure CP violation are currently being tried. One of these ways involves measuring the Electric Dipole Moment (EDM) of various systems: neutrons, electrons, nuclei and so on. EDMs provide a nice way to measure CP violation as a permanent EDM of a fundamental particle necessitates CP violating interactions, as is illustrated on Figure 1, and is free of SM backgrounds.

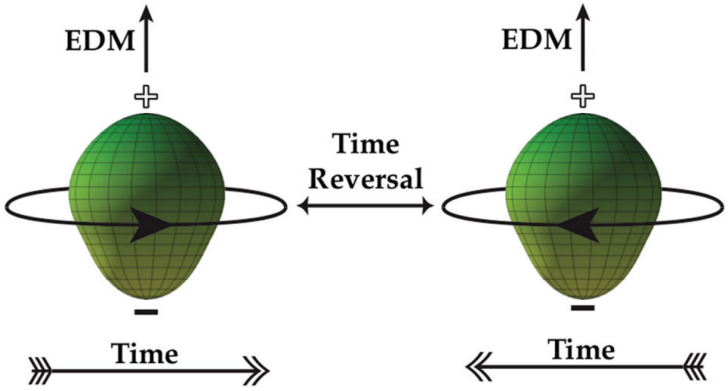


Figure 1: Illustration of how an EDM violates CP-symmetry. The only axis of quantization of a fundamental particle is provided by the spin. After a time reversal transformation the direction of the spin is flipped, but the EDM remains unchanged. Effectively, the sign of the EDM has changed - because it is defined relative to the spin - and thus breaks time reversal symmetry. Due to the CPT theorem, time reversal (T) symmetry violation, also implies CP violation. Figure adapted from [4].

In this report we focus on the EDM of paramagnetic atoms and molecules. In any system, measuring the EDM is hard, as any EDM will be extremely small. However, in atoms and molecules the situation is even worse due to the Schiff theorem. The Schiff theorem states that the EDM of a nucleus is exactly cancelled by a dipole moment formed by the surrounding electron cloud [2]. Luckily, the Schiff theorem only holds for point-like nuclei and in the non-relativistic limit. For larger nuclei, the atom or molecule can still have a moment, called the Schiff moment. But, these Schiff moments are again extremely small and hard to measure. One way to improve the sensitivity of the Schiff moment to the EDM is to use octupole-deformed nuclei [1], such as ^{229}Pa. In octupole-deformed nuclei there is a ladder of nearly degenerate parity doublet states. If there is a parity violating interaction, the parity doublets can mix and the Schiff moment is strongly enhanced. For example, ^{229}Pa is predicted to be 100000 more sensitive than the current limits set by ^{199}Hg [4].

However, octupole-deformed isotopes are very hard to produce and relatively short-lived. Consequently, to study them we need an efficient way to capture, detect and manipulate them.

Implanting octupole-deformed isotopes into defects in diamond has garnered some attention in recent times as a promising measurement scheme [4, 3]. There are two main advantages specific to diamond

1. If the defect is optically active, well established and advanced techniques for their manipulation are commonplace as optical defects in diamond have been studied extensively.

2. If the defect is non-centrosymmetric, large internal electric fields occur that eliminate the need for external electric fields, while simultaneously being larger and more stable. Such fields make measuring the EDM significantly easier.

As the reader may have noticed, there are two conditions for these advantages to actually hold: the defect must be optically active and have a non-centrosymmetric structure [3]. However, it is not known if these types of defects actually form for the isotopes of interest, such as ^{229}Pa. The goal for the work at ISOLDE is to implant ^{231}Pa, as a longer lived proxy to ^{229}Pa, in diamond and see if the formed defects are optically active using photoluminescence (PL), and non-centrosymmetric using emission channelling. In case they do form, the implantation and growth conditions can then be optimised and hopefully someday be used to measure the EDM of ^{229}Pa.

2 Photoluminescence setup at ISOLDE

When photons with a certain energy hit a diamond, some of the electrons are excited to a higher energy state. After some time these electrons decay back down to the ground state, potentially via a series of steps. The spectrum of light emitted in this process is called the emission spectrum and contains much information about the defects. For example, Figure 2 shows the spectrum of the stereotypical NV-defect - a substitutional N atom bound to a C vacancy - in diamond. Lines characteristic of the neutral NV^0 and singly charged NV^- are obviously visible. This process is called PL.

Figure 3 shows the PL setup at ISOLDE (508-R-001). There are two lasers: a green ($\lambda = 532\,\text{nm}$) and blue ($\lambda = 457\,\text{nm}$) laser. The lasers are guided by a set of mirrors onto the sample. The sample stage can be moved around using a set of stepper motors. The emitted light is guided to a monochromator and CCD combination that allows for the measurement of the spectrum. Originally, the different components of the setup were controlled using separate sets of - rather unpleasant to use - software. My project was to create one programme that is able to control all of the setup, in a, hopefully, more user-friendly way. This software is shortly described in the next section.

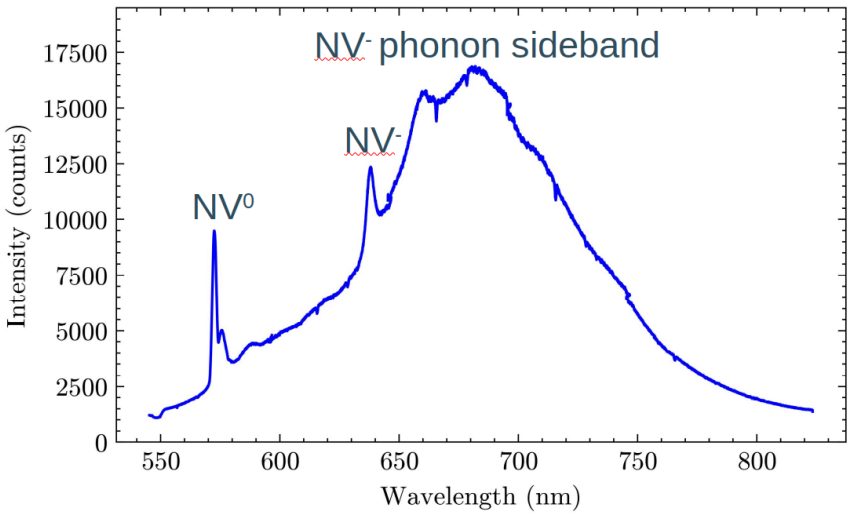


Figure 2: PL spectrum of NV defects in diamond measured in the PL lab at ISOLDE.

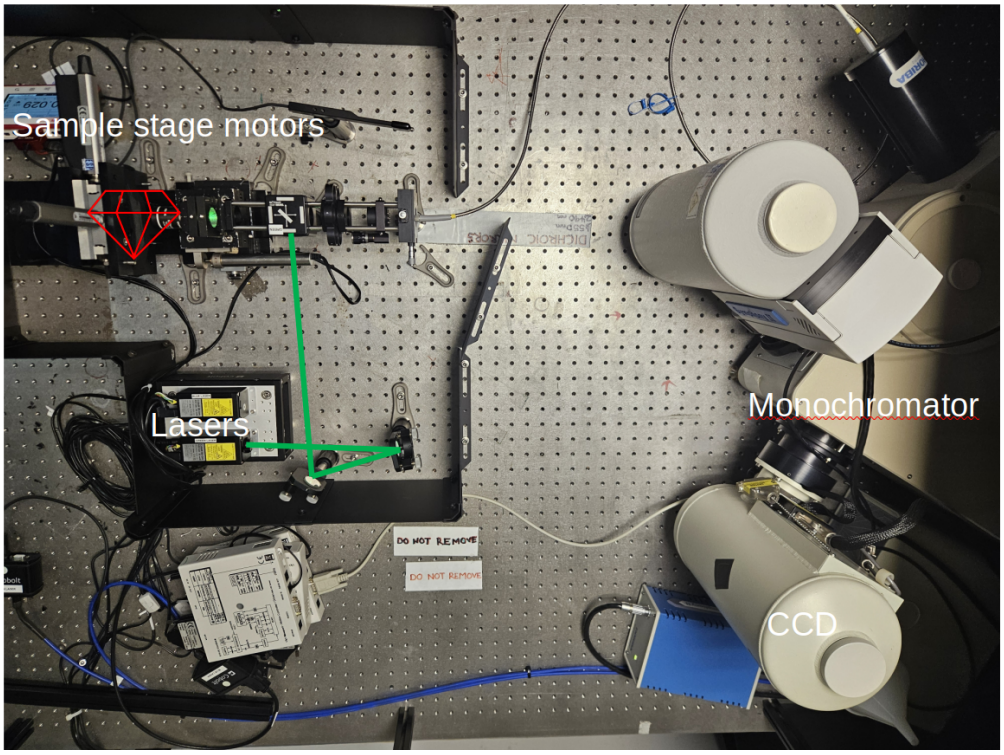


Figure 3: Image of the PL setup at ISOLDE.

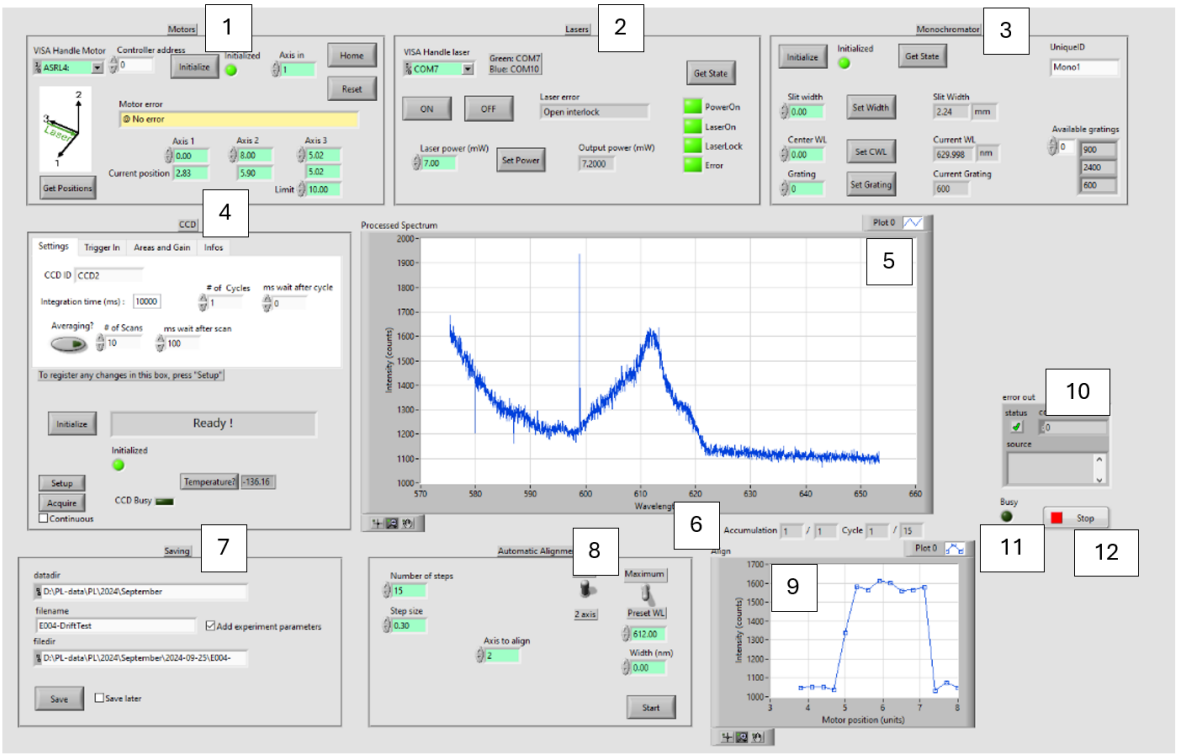


Figure 4: Screenshot of the new PL control software.

3 New PL control software

Figure 4 shows a screenshot of the LabVIEW software I created. The program is based on a consumer-producer architecture which provides a much smoother UI experience than simpler architectures. The main components are

1. Motor control
2. Laser control
3. Monochromator control
4. CCD control and acquisition
5. Spectrum display
6. Accumulation and cycle counter
7. Data saving configuration
8. Automatic alignment
9. Alignment display
10. Error display
11. Busy indicator
12. Stop button.

To summarise, the program can control all components of the setup as desired. Furthermore, because all components are in the same programme, they can talk to each other. Consequently, previously impossible features such as automatic sample alignment - using the motors and CCD at the same time - and improvements to safety by turning off the lasers when the program is terminated, are now implemented. A more detailed overview of all of the features can be found in the manual in appendix.

References

[1] P. A. Butler. Pear-shaped atomic nuclei. *Proceedings of the Royal Society A: Mathematical, Physical and Engineering Sciences*, 476(2239):20200202, 2020.

[2] Eugene D. Commins, J. D. Jackson, and David P. DeMille. The electric dipole moment of the electron: An intuitive explanation for the evasion of Schiff's theorem. *American Journal of Physics*, 75(6):532–536, 06 2007.

[3] Ian M. Morris, Kai Klink, Jaideep T. Singh, Jose L. Mendoza-Cortes, Shannon S. Nicley, and Jonas N. Becker. Rare isotope-containing diamond color centers for fundamental symmetry tests, 2023.

[4] Jaideep Taggart Singh. A new concept for searching for time-reversal symmetry violation using pa-229 ions trapped in optical crystals. *Hyperfine Interactions*, 240(1), March 2019.

Appendix: manual

1 Introduction

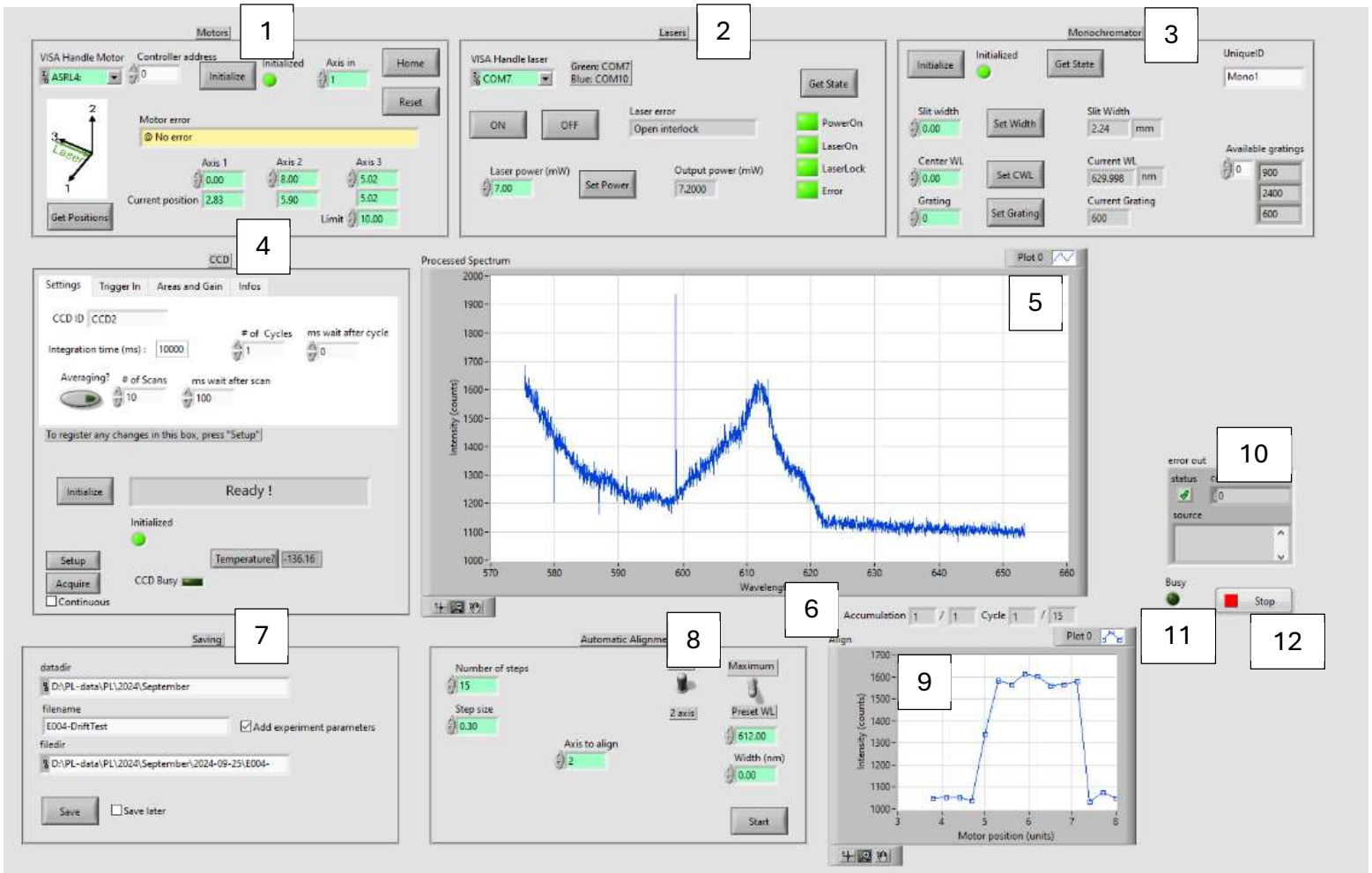
This LabView software was developed by Zeno Maesen in the context of the CERN Summer Student Programme of 2024, for the control of the PL setup at ISOLDE (508-R-001).

The software is implemented in LabVIEW using a consumer-producer architecture, which provides a smoother UI experience than other architectures. One key consequence is that the UI remains responsive even when it is busy in the background, e.g., when a measurement is ongoing. This also means that **when a button is pressed many times, all these presses are queued up and executed later**.

Neither the programme nor this manual is complete, so the user is invited to make modifications to both if necessary.

2 Overview

The screenshot below shows an overview of the programme and its different components.

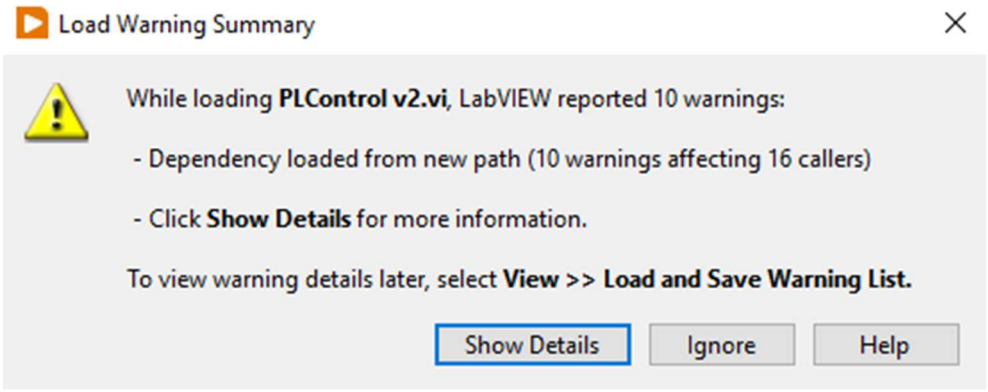


1. Motor control
2. Laser control
3. Monochromator control
4. CCD control and acquisition
5. Spectrum display
6. Accumulation and cycle counter
7. Saving configuration
8. Automatic alignment
9. Alignment display
10. Error display
11. Busy indicator
12. Stop button

3 QuickStart guides

3.1 Taking a single measurement

1. Open PL Control v2
2. After all drivers have been loaded and compiled, the following pop-up is shown. Press 'Ignore' to continue



3. Run the program by clicking the arrow on the top left

4. Select the correct laser and press 'ON'. If the interlock is closed, the laser can then be turned on by toggling the safety key. The power can be set by entering it in 'Laser power (mW)' and clicking 'Set Power'.
5. Initialize the monochromator by pressing 'Initialize'. Set the desired slit width and centre wavelength by entering the value in the corresponding field and clicking the set button.
6. Initialize the CCD by pressing 'Initialize'. Set the desired exposure time in 'Integration time (ms)' and register it by pressing 'Setup' (this might take a while). Press 'Acquire' to make the acquisition and the result will be displayed on the central graph.
7. Stop the program by pressing 'Stop' (this also turns off the laser).
8. Close the program in the standard way and choose 'Don't Save – All' in the pop-up.

3.2 Multi-acquisition, saving and plotting

1. Perform steps 1 through 5 from guide 2.1
2. Initialize the CCD and set the exposure time. Turn on ‘Averaging?’ and fill in the desired number of accumulations in ‘# of Scans’ and the wait between accumulations in ‘ms wait after scan’. Set the number of cycles and wait between cycles. To register all of these values, press ‘Setup’.
3. In case you want to save all cycles: put the directory where data should be saved in ‘datadir’, set the filename (for example to the sample name), check ‘Add experiment parameters’ in case you want to automatically add these and lastly mark ‘Save later’.
4. Press ‘Acquire’ in CCD to start the acquisition. Spectra are plotted and saved after every cycle is finished.

For plotting the data there are two options:

Terminal operation

1. Open command prompt
2. Navigate to the data directory using ‘cd data/dir/’ (if the data is located on the D drive, you first must execute ‘D:’ without cd).
3. Open python by typing ‘python3.12’
4. Import the PL plotting module ‘from plplot import *’
5. Run ‘plotCycles(‘filename’,number of cycles)’. ‘filename’ is the common part of the files for all cycles: filename_i_experiment_parameters.
6. If you want to see the decay at a certain wavelength, use ‘plotCycles(‘filename’,number of cycles,wavelength)’.

Notebook operation

1. Open command prompt
2. If the data is located on the D drive, navigate there using ‘D:’
3. Open jupyter notebook using ‘python3.12 -m notebook’
4. Navigate to the data directory
5. Either create a new notebook or a copy of the ‘PL plot.ipynb’ notebook that can be found on the desktop.
6. Modify existing cells to suit your needs and press ‘Ctrl+Enter’ to execute a cell.

3.3 Aligning

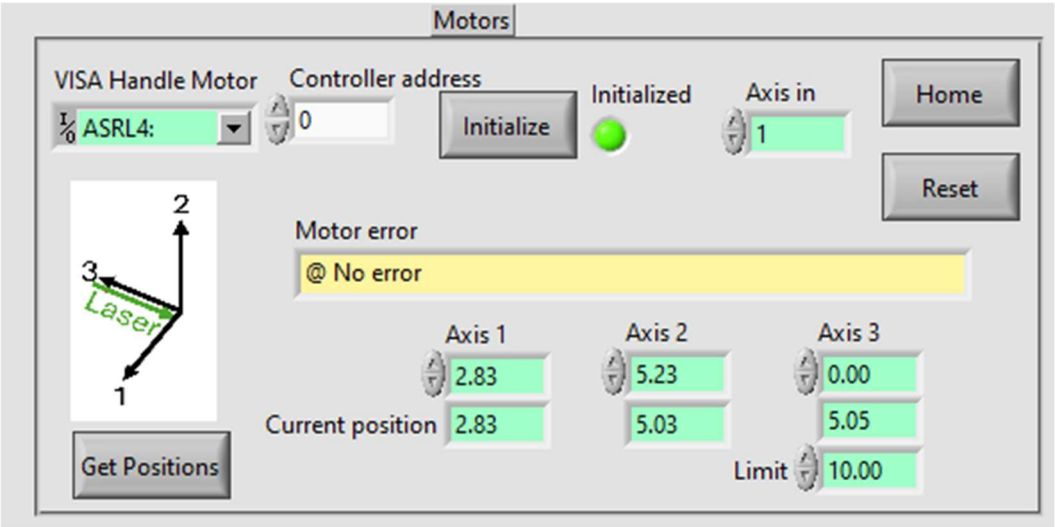
Here we describe a process to align the sample and scan over it completely to find any interesting spots.

1. Perform steps 1 through 6 from guide 2.1
2. Make sure the laser is hitting the sample and manually move the sample stage to a small distance away from the objective lens.
3. Initialize the motors.
4. Do an initial acquisition to make sure you are seeing the sample. If you don't see anything, one thing you can try is to play around with the objective, controlled by axis 3, but make sure not to crash into the sample.
5. Once the sample has been found, alignment can start. First, move the objective as close as possible to the sample by moving on axis 3. Set the current axis 3 motor position in 'Limit' so it is impossible to go even closer. Switch automatic alignment to '1 axis' mode and 'Axis to align' to 3. Enter the desired 'Number of steps' and 'Step size'. A good starting point is 10 steps of 0.05 units. After the scan finishes, the motor automatically moves to the spot of highest intensity. Keep an eye on the spectra during the scan as the one with highest intensity often is not the optimal one.
6. Now we find the centre of the sample. Move axis 1 – to higher values – such that the laser is just off the sample. Do another single axis alignment. Here the number of steps can be smaller and the step size depends on the sample size. Determine the centre position and sample size from the graph.
7. Repeat for axis 2 and move to the centre of the sample.
8. Switch to 2 axis alignment. Enter the number of steps and step size such that their product is about the sample size. The full scan takes the number of steps squared scans, so choose the step size carefully as to not make the scan too long (the exposure time from the CCD tab is used). Finally, press start and a 2D scan of the sample is made and plotted.

4 Detailed overview

Not every UI element is explained as many are self-explanatory.

4.1 Motors

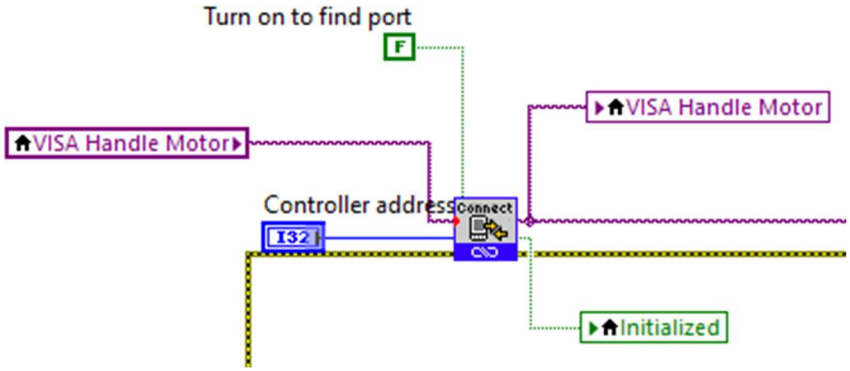


- ***VISA Handle Motor***: doesn't need to be changed for regular operation.
- ***Controller address***: doesn't need to be changed for regular operation.
- ***Axis in***: controls the axis on which the 'Home' and 'Reset' buttons are executed
- ***Motor error***: one common error is 'NOT REF smartstage error' (usually preceeded by a popup saying 'Error: the controller is not in ready state') which is fixed by homing the relevant axis.
- ***Get positions***: updates the positions of all axis
- ***Axis i***: the top control sets the new position (updated after any change) and the bottom shows the readout position. Everything in 'units' defined by the motors.
- ***Limit***: sets the upper limit for movement of 'Axis 3' to avoid the sample stage crashing into the objective. Should be set by the user at every start up.

Remarks

- The drawing shows how the different axis of the stage move (the arrows point in the direction of larger numbers).

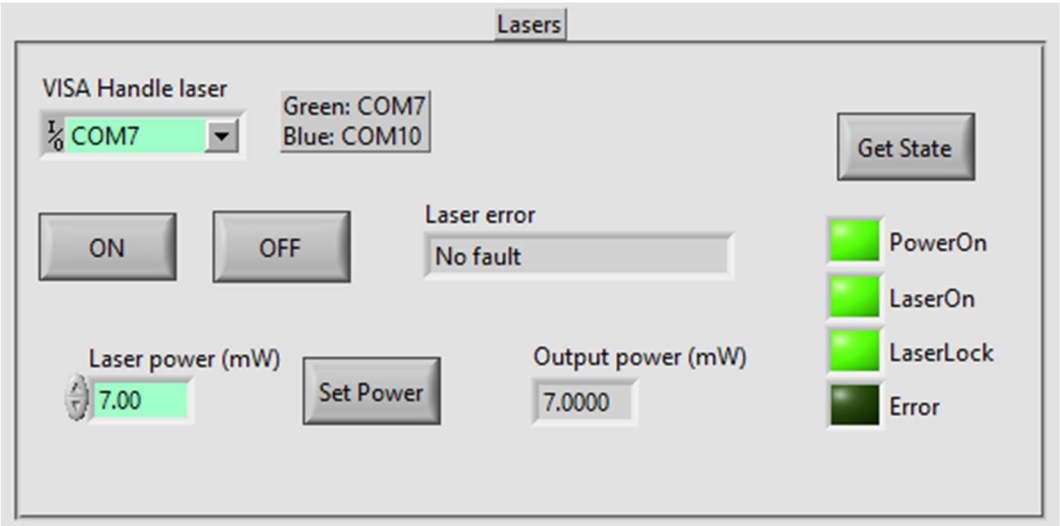
- If the port for the motor ever changes, changing it in the software is actually a little hard (as opposed to for the lasers, where it is trivial). In “InitMotor” in the



consumer loop, change the ‘Turn on to find port’ to ‘T’. If ‘Initialize’ is then pressed, a pop-up will then appear where you can select the correct port. By modifying the default value to the new port, ‘Turn on to find port’ can be turned back off for easier initialization. However, note that there is a difference between the default value of ‘VISA Handle Motor’ and the one it has during operation.

- Sometimes, the communication with the motors has a problem, which shows up in the ‘Error out’ display as ‘RS232 communication timed out’. One method that can be tried to fix this is unplugging the motors.

4.2 Lasers

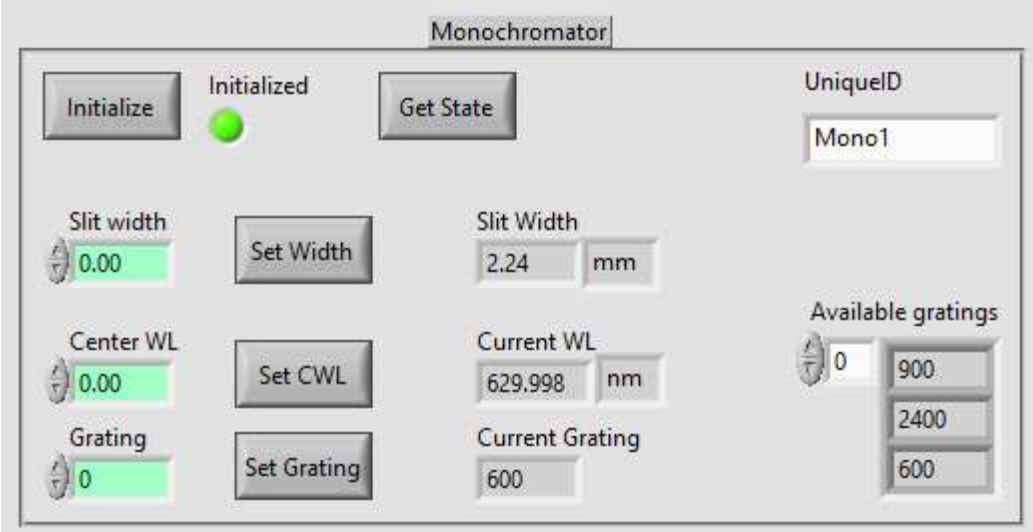


- ***Get State***: updates ‘Output power (mW)’ and status indicators.
- ***Laser error***: does not work properly. The main problems are that it says the interlock is open even though it isn’t (still turns the laser on) and it doesn’t display the need for the ‘Key ON/OFF’ toggle.
- ***Indicator lights***: similar problem to ‘Laser error’.

Remarks:

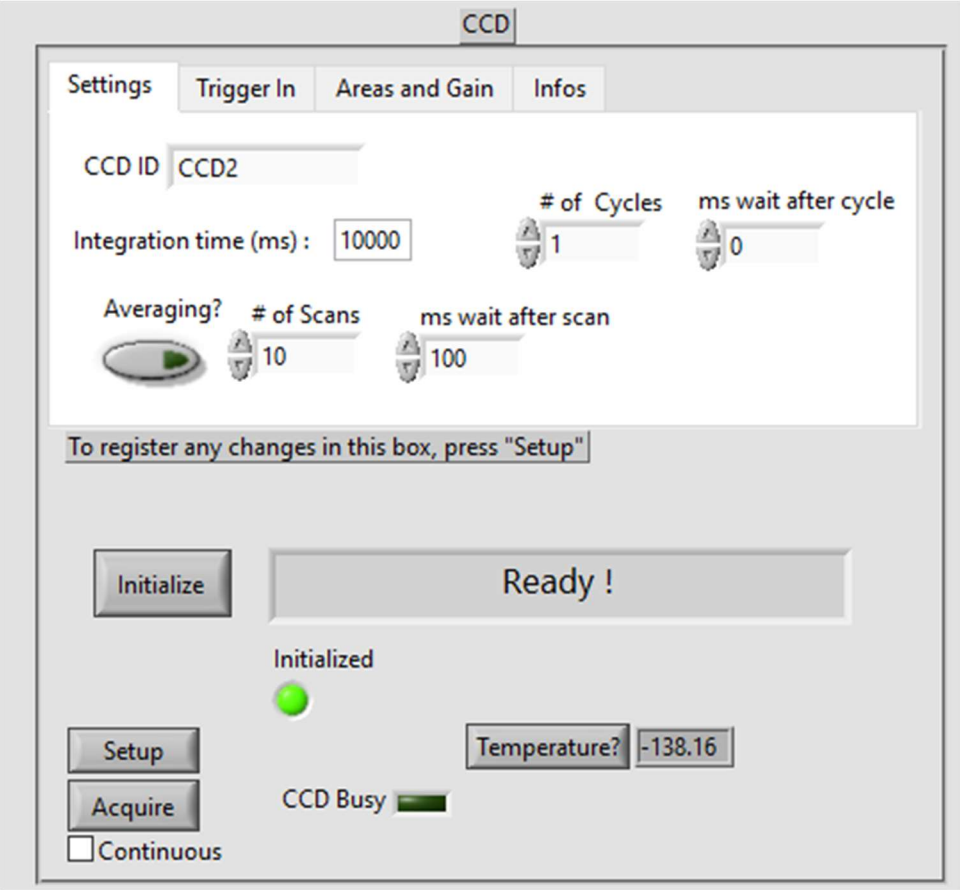
- **When the program is properly stopped**, i.e., through the ‘Stop’ bottom at the bottom right, **the laser is automatically turned off**.

4.3 Monochromator



- ***UniqueID***: doesn’t need to be changed for regular operation.
- ***Grating***: sets the grating based on the list shown in ‘Available gratings’. For example, the 600 lines/mm grating would be set by inputting ‘2’.

4.4 CCD



- ***CCD ID***: doesn't need to be changed for regular operation. If anyone ever wants to add the IGA, it should have ID 'CCD1'.
- ***Averaging?***: needs to be turned on to do multiple acquisitions. If it is turned off, will always do a single scan, even if '# of Scans' is larger than 1.
- ***# of Scans***: number of accumulations that are averaged.
- ***# of Cycles***: number of times the accumulations are repeated, used for RPL. Works even if 'Averaging?' is turned off.
- ***Setup***: takes the values in the multi-tab above and writes them to the CCD. Thus, to register any change in the multi-tab box, 'Setup' needs to be pressed. For some reason this takes some time, so don't be surprised.
- ***Continuous***: if checked and 'Acquire' is pressed, the measurement is repeated until 'Continuous' is unchecked.

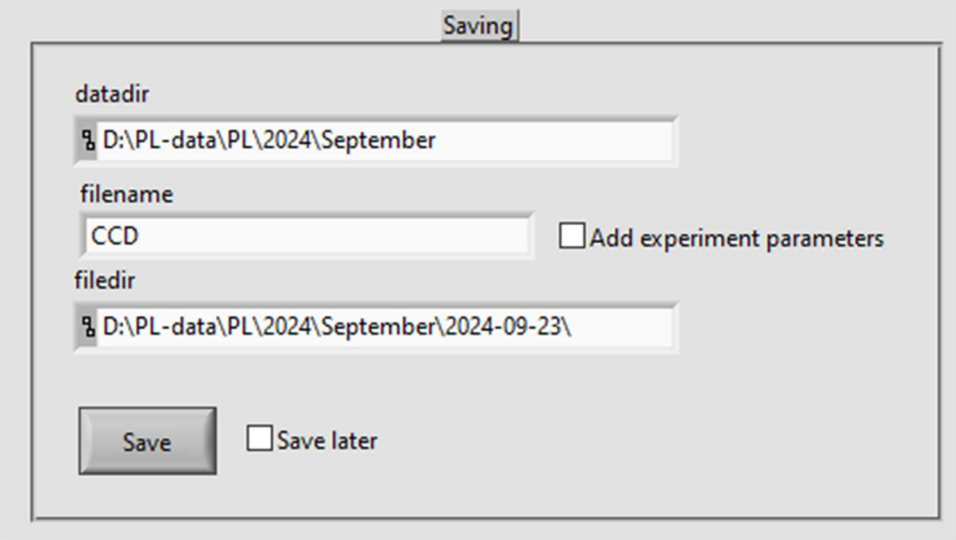
Remarks:

- The progress of an acquisition is displayed on the bottom right

- Everything in the 'Trigger In' and 'Areas and Gain' tabs is untested.

- A measurement is only plotted after all acquisitions have been made, which also removes the previous plot.
- No dark current or cosmic ray removal is implemented. A basic cosmic ray remover can be found in the added Python software.

4.5 Saving



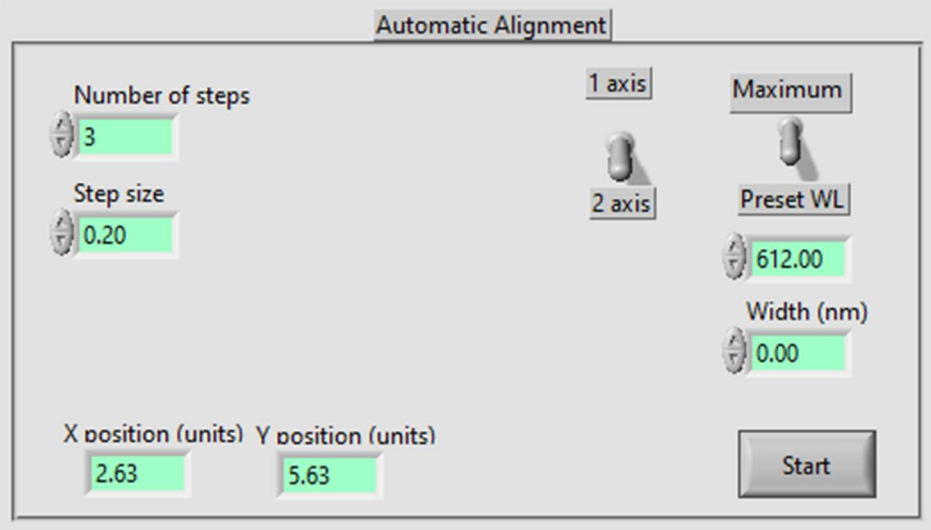
- ***datadir***: directory where the data is to be saved.
- ***filename***: basic part of the filename, e.g., the sample name.
- ***Add experiment parameters***: if checked, the centre wavelength, slit width, number of cycles, number of accumulations and exposure time are appended to the filename.
- ***filedir***: final directory+filename, does not need to be modified by the user in normal operation.
- ***Save***: saves the spectrum that is currently plotted.
- ***Save later***: if it is checked, all subsequent spectra are saved. **Must be turned on to do multiple cycles.**

Remarks:

- Data is saved in a standard csv file but with a tab (‘\t’) as separator (if python pandas is used, this needs to be input). The first column of the csv is the wavelength. If there is no multi-accumulation the second column is the CCD counts. If there is multi-accumulation, the second column is the average counts over all accumulations and the subsequent columns are counts of individual accumulations. The time of acquisition is not explicitly saved in the data file but can be extracted from the file creation time.
- The full filename is ‘filename_i_experiment_parameters_j.csv’ where i is a unique identifier that makes sure that **old data is never overwritten,** and j is the cycle

count. Both i and j are always present. It's best not to use underscores in 'filename' as this might cause problems with the identifier i.

4.6 Auto-alignment



- ***1 axis – 2 axis***: controls the alignment mode (see more details below). The exposure time set in the CCD box is used, but no support for multi-acquisition is implemented. The progress of the alignment is displayed in the cycle counter.
- ***Maximum – Preset WL***: controls the way the intensity at a certain spot is quantified. If it is 'Maximum' the maximum intensity is used. Unless you are working with a very intense signal it is probably not a good idea to work with this option. 'Preset WL' allows you to enter a wavelength that is used to determine the intensity. If 'Width (nm)' is zero, the count at the given wavelength is interpolated. Otherwise, an integral over the range WL-Width to WL+Width is used.

Alignment modes:

- ***1 axis:*** measures a spectrum, quantifies the intensity with the chosen method, moves 'Step size' units on 'Axis to align' (towards 0) and repeats this process 'Number of steps' times. After all steps are completed, the motor automatically moves to the location with the highest intensity. When aligning the focus (axis 3), it is important to note that **the motor moves the sample stage away from the objective**. This is done so the user can manually bring the sample as close as possible to the objective, and then have the alignment happen without risk of crashes.
- ***2 axis***: measures a square of spots with the start spot as centre. Only implemented for alignment in the axis 1 and 2 plane. First, the motor moves to the corner of the square with the lowest axis 1 and 2 positions, which (I think) corresponds to the top right corner of the sample, seen from the perspective of the laser. After the measurement in the corner is done, 'Step size' units are moved (away from 0) on axis 1. This is repeated 'Number of

steps' times, after which a single step is made in the axis 2 direction and axis 1 moves back to the starting value. When the whole square has been measured, the motors automatically move to the position of highest intensity. Note that **the total number of measurements is the number of steps squared**. During the scan, the intensities are displayed on the 'Intensity Graph', where **the bottom left corner of a pixel corresponds to its actual coordinates**.

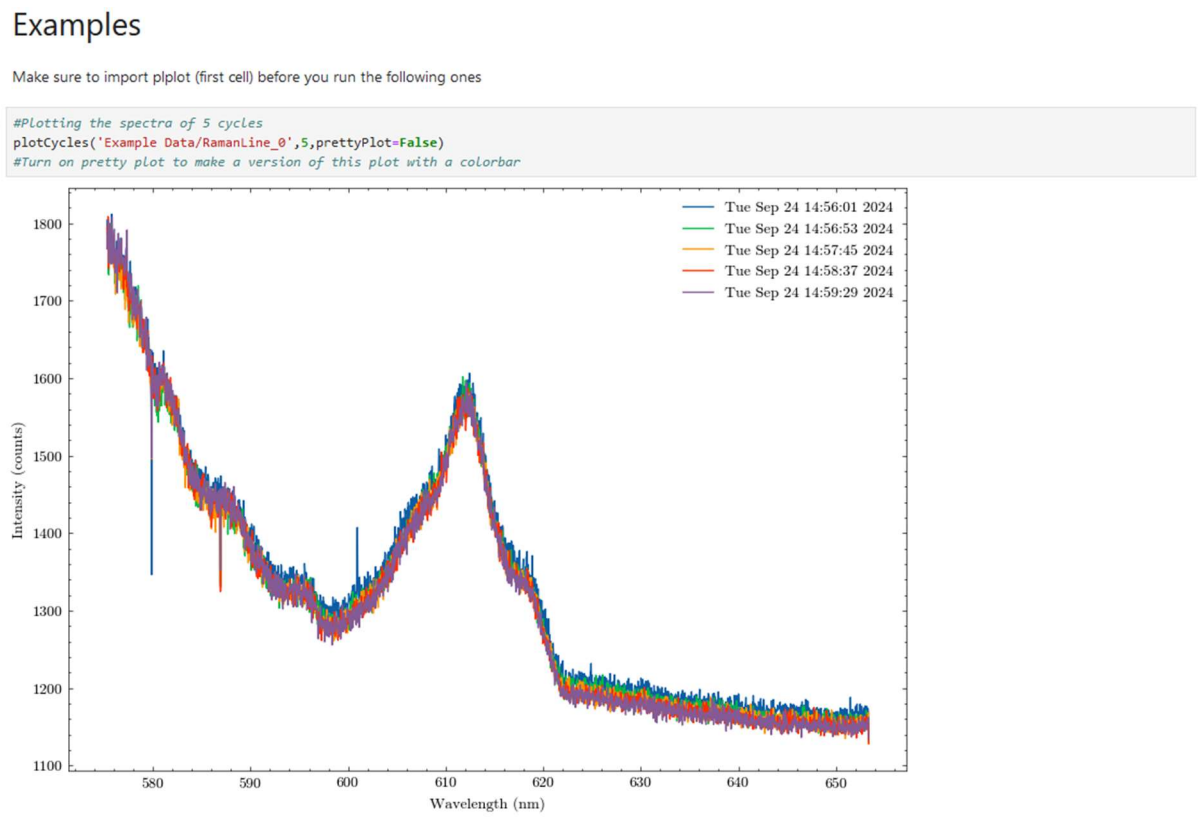
Remarks:

- Weird things often happen when scanning over the sample, so it is always recommended to keep an eye on the spectra in case the spot with the highest intensity is not actually the optimal one.
- In case the user would like to export the intensity graph, this can be done by right-clicking the graph and clicking 'Export'.

4.7 Python plotting

All the python functions for plotting are located at 'C:\Users\QPL508\plmodule\plplot\plplot.py'. They can be imported anywhere on the PL PC by using 'from plplot import *'.

All functionality is illustrated in a jupyter notebook ('PL Plot.ipynb', located on the desktop) that can also serve as a template.



Remarks:

- The time at which a cycle was measured is determined from the last modification date of the data file. Consequently, if raw data files are modified after creation, this will be wrong. So, as is always the case, **don't modify raw data files**.
- The cosmic ray removal is rather rudimentary and could definitely use improvements

4.8 Common errors and troubleshooting tips

- In case of problems, it is always a good idea to try the software of the individual components again. However, for these to function properly, LabVIEW needs to be shut down completely, i.e., not just stopping PL Control, but closing everything LabVIEW (even the annoying pop-up that shows up after closing the program itself).
- If the programme needs to be stopped while it is busy, be it to abort a measurement or because it has frozen, the 'Stop' button cannot be used. Instead, the LabVIEW termination button needs to be pressed. When this is done, the laser is not turned off.