Research Networking Programmes

Short Visit Grant ☒ or Exchange Visit Grant ☐

(please tick the relevant box)

Scientific Report

The scientific report (WORD or PDF file – maximum of eight A4 pages) should be submitted online within one month of the event. It will be published on the ESF website.

Proposal Title: Broadband saturable absorption in graphene and its application to femtosecond mode-locked lasers

Application Reference N°: 6658

1) Purpose of the visit

The purpose of the visit was to design and assemble a pump-probe setup with few-femtosecond resolution and use it to study ultrafast carrier dynamics in graphene samples used at the femtolab at Porto and at Madrid. The setup was designed to be compact and portable to serve as a moveable diagnostics system for future use in Madrid. Other complementary characterization techniques, such as z-scan, were also used to study the graphene samples.
This project also included a computational part. Using the experience in intracavitary dynamics simulation of the host group in Madrid, we aimed to understand the novel dynamical behaviour observed in the graphene-based Ti:Sapphire system at Porto, shown in Fig. 1 [1].

2) Description of the work carried out during the visit

Pump-probe systems require very fine alignment, with lengthy fine-tuning and several stages of setup diagnostic measurements, which is why this visit was planned for 3-4 weeks instead of just 2 weeks, as it further included numerical simulation work.
Over the first weeks, work was focused on building and testing the new pump-probe system. After the setup was assembled, alignment and pulse synchronization were verified by viewing the interference pattern in the overlapping region of the pump and probe spots, by observing spectral interference and using second harmonic generation.

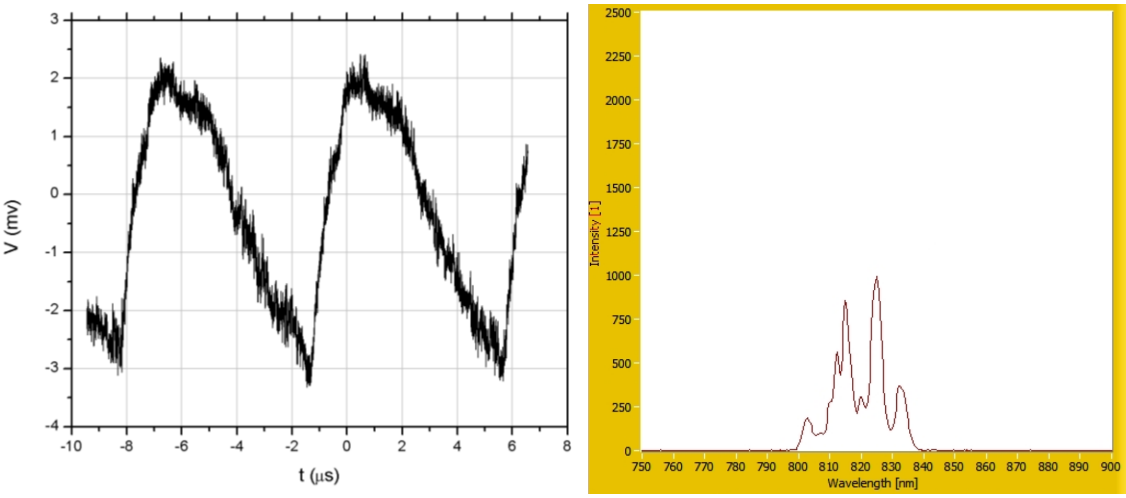

Figure 1 – Left: Temporal output of the graphene-based Ti:sapphire system at Porto. Right: Corresponding emission spectrum. To the best of our knowledge, this is the widest emission spectrum ever recorded in a graphene-based system. Nevertheless, no ultrashort pulses are produced, and as such, mode-locking operation is not occurring as expected.

Over the latter part of the visit, we focused on numerical simulation of Q-switching in Ti:Sapphire cavities. We started by focusing on understanding the regime observed at Porto, by using similar cavity parameters.

Afterwards, we shifted towards a broader range of simulation parameters, to study Q-switching in Ti:Sapphire systems in general, using graphene films with a varying number of layers as saturable absorbers. We expect to submit this work to a scientific journal in the coming weeks.

Detailed description

May 5th-8th: Work began with system design and alignment. Pump-probe systems require very fine alignment, and as such the first days were exclusively dedicated to system design, alignment and optimization. Fig. 2 shows the setup configuration as it was changed throughout this work.

May 9th-13th: During this time, due to technical issues we were unable to carry on with work in the lab and literature research was done on pump-probe experiments in graphene.

May 14th: We performed z-scans for the available graphene samples but we did not observe any evidence of saturable absorption in any of them. The noise level was higher than the expected change of 1% in transmittance, and no results were obtained even with prolonged averaging.

May 15th: We used the reflectivity change in Silicon to try to determine the zero delay point. Around zero delay, the reflectivity should rise to a sharp peak the abruptly decline [2]. However, we did not observe this behaviour and were unable to find zero delay with this measurement.

May 16th: We removed the final probe arm mirror and placed a 50/50 beam splitter to couple the output beams. This resulted in a small lateral separation between the two output beams and thus a smaller interaction angle in the focal point of the lens/parabolic

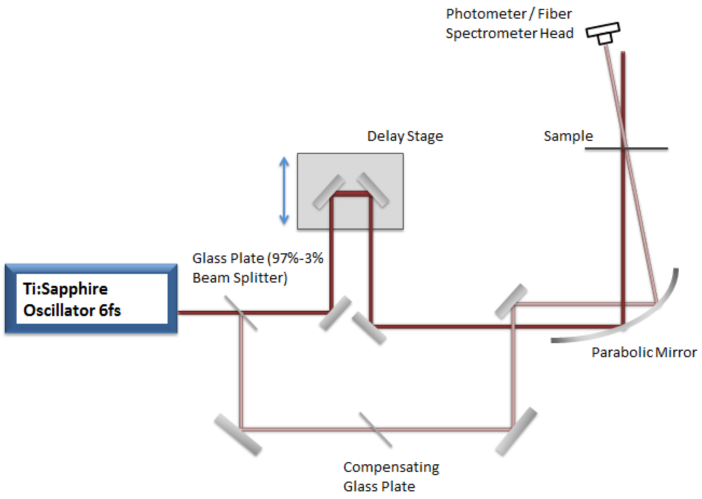

Figure 2a – Initial pump-probe setup.

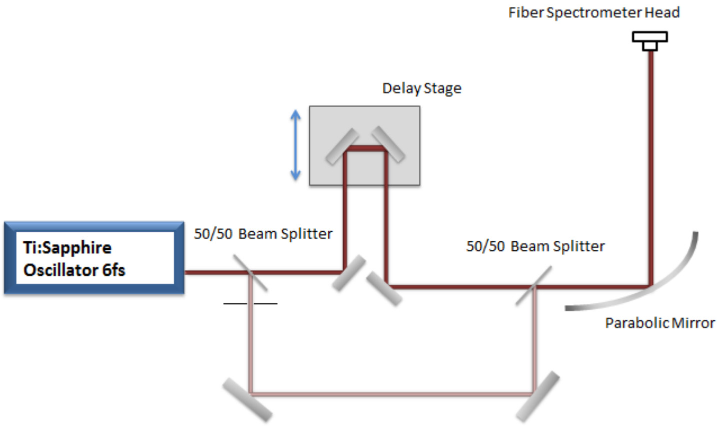

Figure 2b – Using a beam splitter as coupler allows for smaller beam separation and small interaction angles in the focal point. The input beam was also replaced for a compensated 50/50 beam splitter, allowing us to remove the compensating glass plate. The probe beam is attenuated with a small aperture. The beams were made collinear, for easier occurrence of spectral interference.

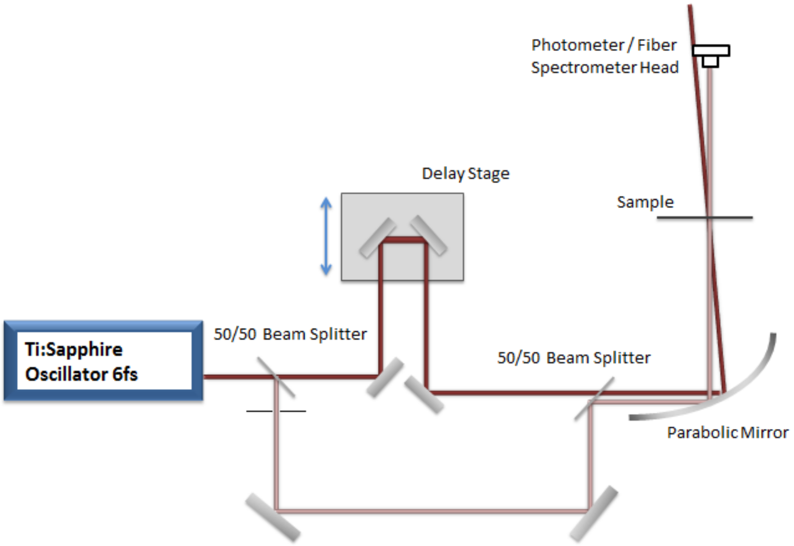

Figure 2c – Final pump-probe setup.

mirror placed afterwards. We also replaced the glass plate serving as the input beam splitter for another 50/50 beam splitter.

Since these 50/50 beam splitters are designed to have equal transmission and reflection optical paths, they allowed us to remove the compensating glass plate in the probe path, which hampered SHG with the probe beam. At this point the output beams were made collinear for easier synchronization detection, which would later be changed to allow detection of the probe alone.

May 18th: In the continuous wave regime, if the system is properly aligned, interference will occur for all delay values but the pattern is only visible to the naked eye when the system is close to zero delay, when there are few rings with high contrast. We observed the interference pattern with a CW input and, by adjusting the delay to minimize the number of rings, we determined a range in which to perform a very slow and thorough scan with the laser in mode-locked operation.

We observed an interference pattern on the overlapping region of the two laser spots while the system was in mode-locking operation, which ensures a close proximity to zero delay. To verify this, we measured the output in a fiber spectrometer and observed spectral interference. The pattern changes when the delay is varied and is only visible in a very small delay range around zero delay.

May 19th-20th: To optimize the available delay range, the delay stage was moved and finely realigned. We again observed spectral interference with a pulsed input. Furthermore, when generating second-harmonic, we observed an 8:1 intensity ratio between the overlapping beams and the pump or probe alone, which only occurs when the pulses are synchronized, shown in Fig. 3.

The output beams were made parallel but noncollinear to enable us to detect just the probe. As the beams were no longer collinear, we placed a thin glass plate which transmitted the probe beam while reflecting the pump beam in such a way that they become collinear. With this configuration we were able to measure spectral interference with noncollinear output beams. Results are shown in Fig. 4.

May 21st-22nd: With the noncollinear setup aligned and with synchronized pulses, we studied the available graphene samples using a z-scan setup again to select the most promising ones. This technique is faster and less demanding but allows us to observe saturable absorption to evaluate the intensity of the effect in each sample. As before, no clear saturation behaviour was observed and the noise level was still higher than the expected changes in transmittance. We performed a pump-probe measurement in a CdS (Cadmium Sulfide), which should show a higher increase in transmission than graphene [3]. But again, laser power fluctuations made it impossible to draw any conclusions from the measurements.

May 23rd-30th: The system remained too unstable for pump-probe measurements to be performed. During this time, we focused on the computational part of this project. The objective was to study possible Q-switching behaviour in graphene-based Ti:Sapphire oscillator at Porto. The goal here was to learn, not just run an already written program with the system data from Porto. The first goal was to reproduce the results published in [4], using 4th order Runge-Kutta method in Python. After that, we ran simulations for the oscillator at Porto.

Using the cavity specifications from Porto, Q-switching did not occur in our simulations. The parameters changed were the layer number (1 to 4 layers) and the carrier relaxation time of graphene. This allowed us to study the possibility of a low quality sample, with impurities or surface defects.

After this, we studied Q-switching in cavities containing Ti:sapphire and graphene acting as a saturable absorber. This work studied the possibility of passive Q-switching occurring with various cavity lengths, number of layers in the graphene absorber and pumping intensity. This work is still in development, and will be continued by Dr. Rosa Weigand and Tiago Pinto.

3) Description of the main results obtained

The first part of this work consisted in designing and building a compact and portable pump-probe system to future use in Madrid. Alignment and pulse synchronization were measured using spectral interferometry and second-harmonic generation. These measurements, described in the section above, are shown below.

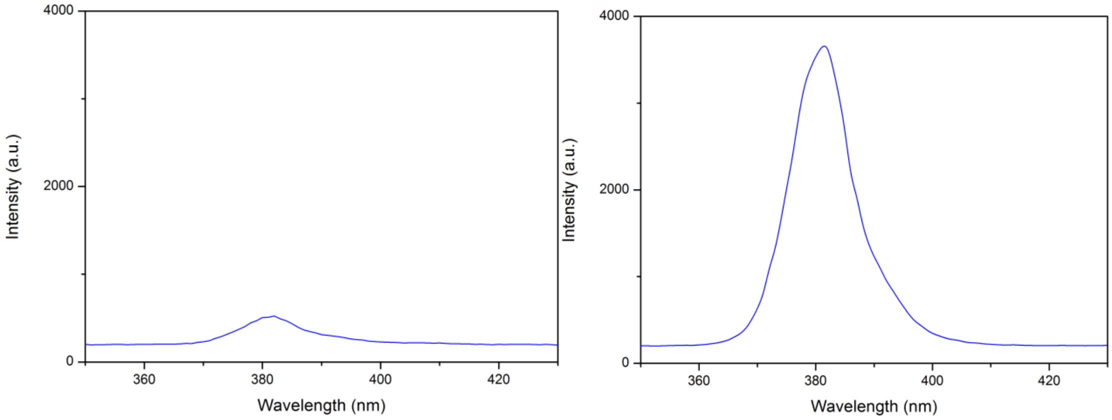

Figure 3 –SHG intensity for collinear output beams close to the zero delay. Left: probe; Right: pump+probe. These signals show an 8:1 intensity ratio only when the beams overlap in the crystal and the pulses are synchronized.

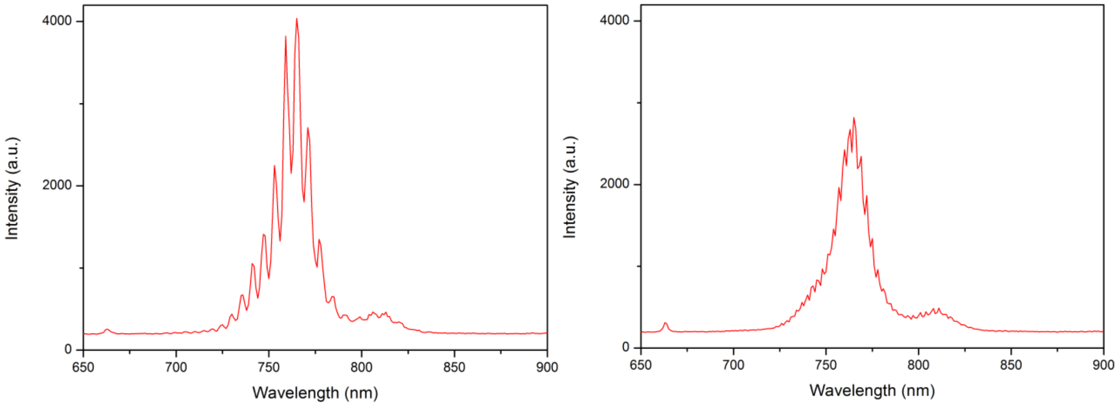

Figure 4 – Noncollinear spectral interference for two different delay values. Both the contrast and fringe width change when the delay is varied.

As previously mentioned, due to laser instability and unknown sample quality, we were unable to measure saturable absorption with the pump-probe or the z-scan setup. The expected changes are very small and require exceedingly stable systems, while still producing sufficiently high output powers in order to produce saturation.

We will now discuss the numerical results obtained. Firstly, we successfully reproduced the results obtained in [4], to ensure the proper functioning of the program. Afterwards, using the cavity parameters from Porto, the simulations yielded no Q-switching operation, only CW operations, after relaxation oscillations, as shown in Fig. 5.

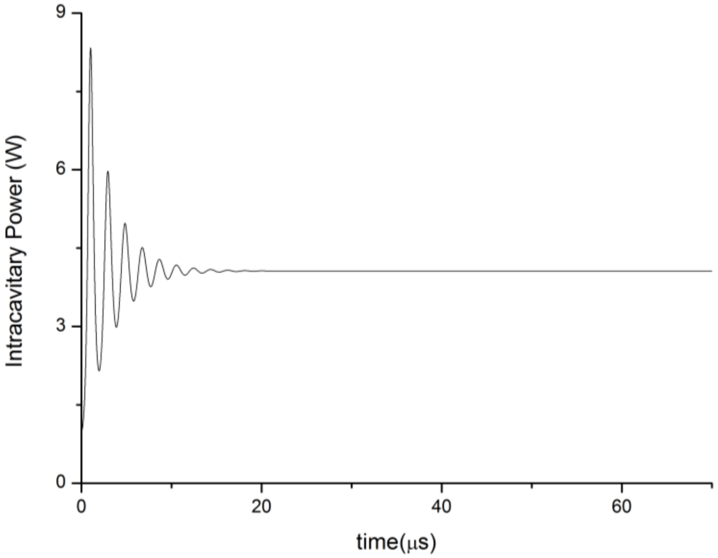

Figure 5 – Q-switching dynamics simulation for the cavity from Porto. After initial relaxation oscillations, the intracavitary power quickly becomes constant, initiating stable CW regime.

Afterwards, we started conducting studies of more general Q-switching dynamics in Ti:sapphire and graphene systems. This work is still being developed but below we show some preliminary results, for a 10 cm long linear laser cavity, with a single-layer graphene sample. Q-switching behaviour is clearly observed. Peak intensity quickly rises until a stable value is achieved. Our work's foci include the changes in pulse energy, duration, repetition rate and other parameters with changes in cavity specifications. We expect this work to be completed in a near future.

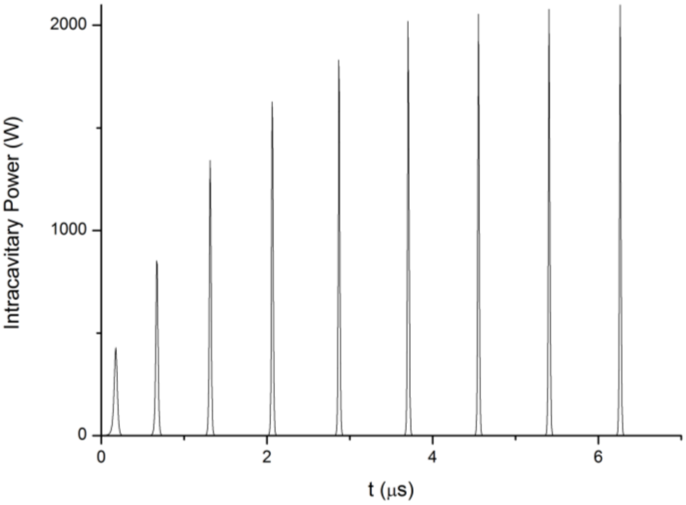

Figure 6 – Q-switching simulation for a linear 10cm long Ti:sapphire laser cavity with a single-layer graphene saturable absorber

4) Future collaboration with host institution (if applicable)

The numerical work started in the latter part of the visit is still being continued by Dr. Rosa Weigand and Tiago Pinto. Furthermore, collaborations between the Optics Department at Madrid and femtolab at Porto already exist in other projects. We wish to continue this trend with future collaborations involving graphene and ultrashort laser pulses.

5) Projected publications / articles resulting or to result from the grant *(ESF must be acknowledged in publications resulting from the grantee's work in relation with the grant)*

We expect to submit the numerical work being done on Q-switching in Ti:Sapphire systems with graphene to a scientific journal in the forthcoming weeks.

6) Other comments (if any)

We would like to thank ESF for its financial support in project. We will acknowledge and thank ESF for its support in publications and presentations regarding the work done during this visit.

References

[1] Ultrafast Saturable Absorption in Single-Layer Graphene with Application to Broadband Mode-Locking, Tiago Pinto, MSc Thesis submitted October 2013.
[2] A. J. Sabbah and D. M. Riffe, Femtosecond pump-probe reflectivity study of silicon carrier dynamics, Physical Review B 66, 165217, 2002.
[3] H. P. Li, C. H. Kam, Y. L. Lam, W. Ji, Optical nonlinearities and photo-excited carrier lifetime in CdS at 532nm, Optics Communications, 190 (2001) 351-356, 2001.
[4] An Nd:YLF laser Q-switched by a monolayer-graphene saturable-absorber mirror, Paloma Matía-Hernando, José Manuel Guerra and Rosa Weigand, 10.1088/1054-660X/23/2/025003, 2013.