Development of a setup for time resolved MOKE measurements of ultrafast magnetization phenomena Internship Synopsis

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1 Introduction

Exploring and understanding spin-related phenomena in materials using light to probe, modify and control magnetic properties are of great interest from both fundamental scientific and technological points of view. On one hand, it is of scientific interest to understand transient phenomena of collective spin systems that have been driven out of equilibrium. On the other hand, technological demands for ever-increasing writing and reading speed for magnetic storage and data processing have fueled efforts to find ever-faster ways to control magnetic states. One of the most exciting, yet puzzling challenges in condensed matter physics today is whether one can detect, understand and control macroscopic spin ordering in highly non-equilibrium, non-thermal states at femtosecond time scales. The development of sub $100fs$ laser pulses introduced the possibility of probing and controlling magnetism at ultrafast time scales. Indeed, it has been discovered that such ultrafast pulses can induce sub-picosecond demagnetization in nickel [1], stimulating studies on a wide range of magnetic materials. Exciting magnetic material with ultrafast laser pulses induce processes that are orders of magnitude faster than those of the traditional thermal-magnetic processes [2].
An intense femtosecond optical pulse induces a non-equilibrium electron gas which subsequently thermalizes back to a Fermi-Dirac distribution. This thermalization is due to electron-electron interactions, and occurs typically within 100s of femtoseconds, e.g., $\sim 500fs$ has been measured for nobel metals [3], [4]. In parallel, the hot electron gas relaxes its energy to the lattice via electron-phonon scattering, with a typical time constant of $1-10ps$ for transition metals [1]. During this time, the temperature exchange between the electron (T_e) and the lattice (T_l) can be studied and the characteristic times of the microscopic interactions can correspondingly be deduced from such measurements.
The challenge is to understand how the initial hot electron distribution can induce spin dynamics. This question is non-trivial, since the optically induced electronic excitation preserves the electron's spin. Currently, there is no consensus about which physical phenomenon is responsible for this ultrafast demagnetization [5]. Or the more recently discovered all-optical magnetization switching [6]. In order to study these phenomena and find new information, time resolved experiments have to be performed. A large fraction of these experiments is based on Magneto-Optical (MO) effects.

1.1 Ultrafast Magneto-Optical Effect

To investigate spin-related phenomena in condensed matter, one can use optical light in the visible to Infra Red (IR) wavelength range. Indeed, when light is interacting with a magnetic material, it can get modified - this is what we call a magneto-optical (MO) effect. A variety of linear MO effects such as Kerr Effect (MOKE) and Faraday spectroscopy, have provided detailed information and insight into collective excitations and other phenomena, providing direct time-domain information about the magnetic properties of excited states with high temporal resolution [7]. The photo-excitation of a magnetic system with ultrashort laser pulses can strongly alter the thermodynamic equilibrium of the different degrees of freedom (charge carrier, spin and lattice), triggering a variety of dynamical processes. Subsequently, MO techniques are capable of directly revealing the spin and charge fluctuations associated to these processes. Since the MO effect alters the electromagnetic wave as it passes through a medium that has a magnetic field, it is widely used in both static and time resolved experiments. In the latter case, to access ultrafast magnetization dynamics, femtosecond pulses are used to give information about the transient magnetization state. Time resolved MOKE spectroscopy is extremely relevant for accessing genuine spin dynamics at femtosecond scales: it can detect the magnetization of a fraction of an atomic layer of a magnetic material [8]. In addition, it can be employed to observe the perpendicular magnetic anisotropy of ultra-thin films [9]. And it can be combined with a good lateral resolution down to $0.2\mu m$, allowing observations of magnetic domains [10].

1.2 Goal

The proposed internship project aims to optimize the laboratory's current static MOKE setup in order to use it for time-resolved experiments. Hysteresis loop measurements taken with the current MOKE setup present some drift after saturation as well as significant noise. Also, time resolved experiments require a particularly stable set-up because the measurement has to be repeated for each pump-probe delay. Therefore, a new setup will be implemented (See Section 4, Figure 8). The goal is to eliminate any drift and noise while increasing the signal. Once this setup is mounted, measurements will be taken and compared with those obtained using the previous setup. Finally, the setup will be applied in time-resolved experiments using a femtosecond laser. The scientific motivation for this development is that a characterization of samples is needed, before they are studied at large scale x-ray free electron laser user facilities. Thus, a functional, reliable time resolved MOKE setup is crucial for the selection of the best samples to be tested.

2 LCPMR

The "Laboratoire Chimie Physique-Matière et Rayonnement" (LCPMR) is a French research laboratory attached to the Université Pierre and Marie Curie (Paris 6) and the Centre National de la Recherche Scientique (CNRS). It has a variety of teams composed of chemists and physicists, experimentalists and theoreticians that are specialized in the study of interactions of radiation with matter. The systems studied range from condensed matter (complex materials, interfaces) to atoms and molecules isolated or adsorbed on surfaces. It is also involved in nanoscience and attoscience (study of the dynamics of systems on the attosecond time scale). The laboratory is nationally and internationally recognized for its expertise in the development and use of XUV and X-ray spectroscopy techniques. There are many

advanced equipments such as the X-ray photo-electron and ultraviolet photo-electron spectrometer. The laboratory has a successful partnership with the French synchrotron radiation facility SOLEIL. Several instruments developed at the LCPMR are permanently installed at SOLEIL: a high-resolution instrument for resonant inelastic X-ray scattering studies, a near-ambient pressure X-ray photo-electron spectrometer and a high energy photo-electron spectrometer for experiments on solids and gases. With these equipments, the matter can be studied from condensed state to atoms, molecules and clusters, isolated or absorbed on surfaces. Based on the research area, there are three research sections in the laboratory, they focus on electronic structure of complex materials, reactive surfaces and interfaces, and reactivity under radiation of species in the gas phase, respectively. The theoretical component of the laboratory comprises two teams in the modeling of solids and molecular systems excited in an internal layer, as well as in the description of the response to ultra-short times of atomic and molecular systems subjected to intense fields.
I am doing my internship in the section "structure électronique de matériaux complexes" and more precisely in the team "systèmes fortements corrélés - Matériaux magnétiques". This team is using XUV and soft X- rays (photons from 10 to 2000 eV) to probe strongly correlated and magnetic materials. They are studying interactions between electrons, spins and lattices and particularly, ultrafast magnetism dynamics after excitation by short laser pulses. For that reason, the team has a close collaboration with SOLEIL where a resonant inelastic x-ray scattering end-station has been developed conjointly and where the team is involved in the development of the femtoslicing facility. For the ultrafast study of magnetic dynamics, the team is working in close collaboration with the Laboratoire d'Optique Appliqué where a high harmonic generation source is used and with the XUV free electron lasers of FERMI in Italy and FLASH in Germany. To perform experiments in such infrastructures, they are growing their samples with a magnetron sputtering chamber and characterizing their magnetic properties at room temperature and low temperature thanks to magneto-optical experiments.

3 Magneto Optical Kerr Effect (MOKE)

MO effects occur due to the modification of light by the magnetization of the material with which the light is interacting. One of these effects, known as the Magneto-Optical Kerr Effect (MOKE) was first observed in 1887 by John Kerr[11]. The MOKE effect is directly proportional to the magnetization M, which renders it useful for the study of surface magnetism. It gives specific magnetization information such as orientation, uniformity and strength of the magnetization in thin films. One can probe the magnetization in very small regions of a material [12] or in real device applications. Thus, MOKE has emerged as an important technique in the study of surface magnetism and extensively used to characterize magnetic materials. The MOKE effect is also the basis of commercially available magneto optical devices.

3.1 Principles of MOKE

MOKE can be described macroscopically by the dielectric tensor theory [13] or microscopically, where the coupling between the electric field of the light and the magnetization occurs through the spin-orbit interaction [14]. In this article the effects will be described using the idea of the Lorentz force [15] .
In general, MOKE is the change of polarization and/or intensity of a light beam reflected by a ferromagnetic surface. The measured quantity, e.g the rotation of polarization, is a linear function of the magnetization of the ferromagnetic material. The simplest model of MOKE

is to consider a Lorentz-Drude model of a metallic film [16].

3.1.1 Polarization of light

In order to understand MOKE, one needs to understand how the state of polarization of the reflected light is dependent on the initial polarization and the MO geometry in which it is being used.
Light is an electromagnetic wave where the electric and magnetic fields oscillate perpendicularly to each other and to the direction of propagation. Light is called unpolarized if the direction of the electric field fluctuates randomly in time, whereas if it is well defined, it is called polarized. The plane which contains the electric field and the direction of propagation is called the plane of polarization. Depending on how the electric field is oriented, one can classify polarized light into three types (See Figure 1):

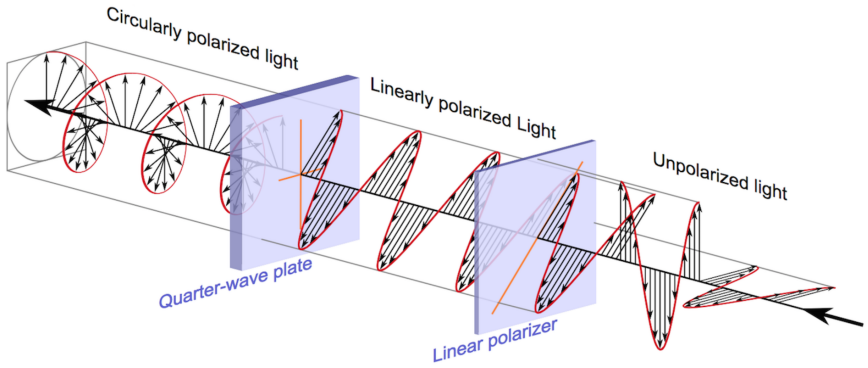

Figure 1: Types of polarization of light.

Linear polarization: the electric field of light is confined to a single plane along the direction of propagation. *Circular polarization:* the electric field of light has two linear components that are perpendicular to each other, equal in amplitude, but have a phase difference of $\pi/2$. The resulting electric field rotates in a circle around the direction of propagation and, depending on the rotation direction, is called left-or right-hand circularly polarized light. *Elliptical polarization:* the electric field of light describes an ellipse, as a result of the combination of two linear components with differing amplitudes and/or a phase difference that is not $\pi/2$. In our experiment, we will only use linearly polarized incoming light.

The two orthogonal linear polarization states that are most important for reflection and transmission are referred to as p- and s-polarization. P-polarized (from the German parallel) light has an electric field polarized parallel to the plane of incidence, while s-polarized (from the German senkrecht) light is perpendicular to this plane. The plane of incidence is also known as the scattering plane - the plane which contains the incident and reflected light beam.

3.2 The MOKE effect

Linearly polarized light which is reflected off a metallic surface becomes generally elliptically polarized. However, if the incident light is either p- or s- polarized, then the reflected light

will still be linearly polarized upon reflection because the reflecting surface is a plane of symmetry of the system. When the linearly polarized light is reflected off a magnetic surface, the magnetism destroys this symmetry. The polarization changes during the reflection, since the reflected light not only has a p-component (as in the ordinary metallic reflection), but an additional small s-component which is generally out of phase with the p-component.
This makes the light elliptically polarized with its major axis rotated from its initial incident polarization plane (See Figure 2). A similar effect occurs for s-polarized light. The two effects are known as the Kerr ellipticity and Kerr rotation.

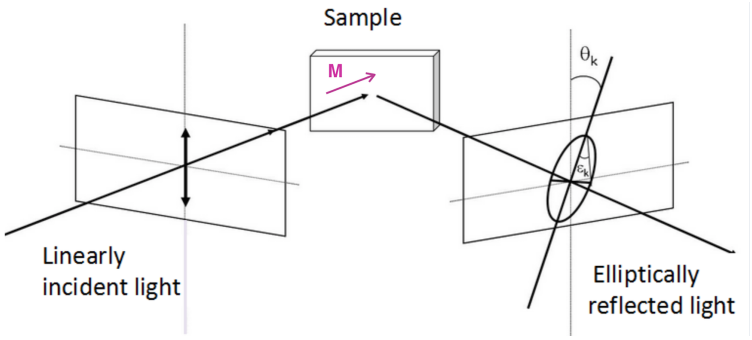

Figure 2: Scheme of the reflection of a linearly polarized incident light from a magnetic sample.

Essentially, what MOKE measures directly is the magneto optic response of the medium, i.e the change in the incident polarization of light.
This response consists of two parts:
a)The change in the polarization of the phase component of the reflected light which is responsible for the *rotation* (θ).
b)The change in the polarization of the out-of-phase component of the reflected light which is responsible for the *ellipticity* (ε).
There are principally three MOKE geometries which are classified depending upon the magneto optic geometry being employed (See Figure 3). The effects are dependent on the orientation of the magnetization with respect to the scattering and sample planes.

* Longitudinal Kerr effect: The magnetization is in the plane of the sample and parallel to the scattering plane.

* Transverse Kerr effect: The magnetization is in the plane of the sample and perpendicular to the scattering plane.

* Polar Kerr effect: The magnetization is perpendicular to the sample plane and parallel to the scattering plane.

The longitudinal and transverse effects are generally used to study the in-plane magnetic anisotropy, whereas the polar configuration is used to study thin films which exhibit perpendicular, magnetic out-of-plane anisotropy. In the transverse effect there is no change in the polarization of the incident light. This can be seen in the Figure 4, where a vector representation using the idea of Lorentz force indicates how p and s-polarized light interact in the three MO geometries. The electric field of the linearly polarized light incident upon the material excites the electrons in it so that they oscillate parallel to the incident polarization which

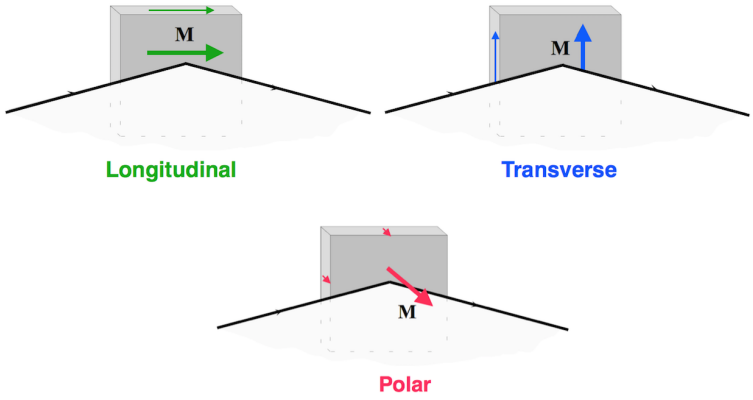

Figure 3: Scheme of **longitudinal**, **transverse** and **polar** MOKE geometries. The color arrows refer to the direction of the magnetization M.

makes a normal component E_N in the reflected light. On the other hand, the Lorentz force induces a relatively *small* component perpendicular to both the primary motion (E_N) and the direction of magnetization and it's referred to the Kerr component E_K.
Generally, the two components are not in phase so that the superposition of both components is responsible for the magnetization-dependent rotation of the polarization. In the longitudinal and polar effects (Figure 4:a,b) p- or s-polarized light will become elliptically polarized with its major axis rotated (Kerr rotation) because there is an additional orthogonal electric field component being induced due to the Lorentz force. The transverse effect (Figure 4:c) involves no change in polarization since there is either no Lorentz force present (s-polarized case) or the induced component has the same polarization as the incident polarization (p-polarized case), instead it involves a change in the intensity of the light (Kerr reflectivity). The intensity changes depend on the component of magnetization perpendicular to the plane of incidence.
These effects are described (using the dielectric theory [13] ,[17]) by assuming that the linearly polarized light is made up of the superposition of two circular components, L- and R- circularly polarized light. For each polarized mode, the magnetic medium has a different refractive index. Therefore, the two modes travel with different velocities and attenuate differently in the material. Thus after reflection, the two modes recombine to produce Kerr rotation and ellipticity. The general form of the dielectric tensor (in the x, y, z basis) which represents the effects of a magnetic medium is given by [13]:

$$M = \varepsilon_0 \begin{bmatrix} 1 & -iQ_z & iQ_y \\ iQ_z & 1 & -iQ_x \\ -iQ_y & iQ_x & 1 \end{bmatrix}$$

where $Q_{x,y,z}$ are the Voigt magneto optic constants that describe the magneto optical effect and are at first order proportional to the magnetization of the material and ε_0 is the dielectric constant.
The magnetic sample has a refractive complex index n which can equivalently be represented using the dielectric tensor (in the s, p basis), where a reflection matrix is produced by solving

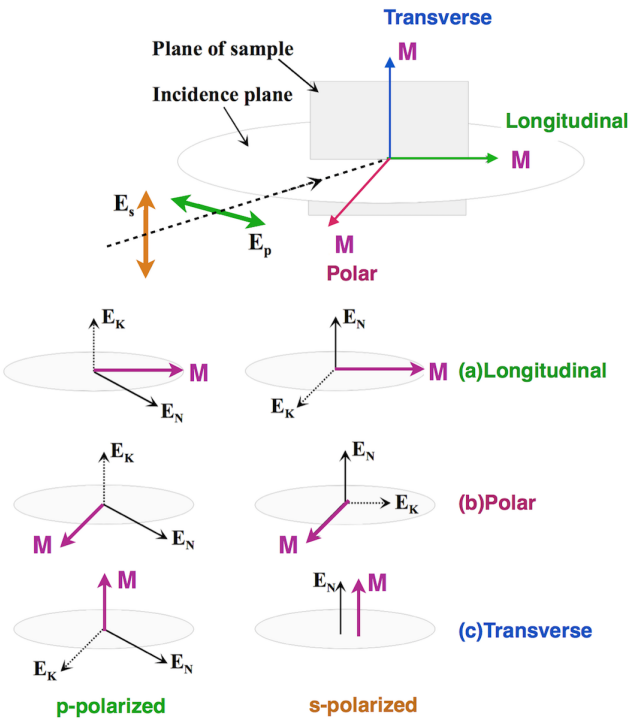

Figure 4: Schematic illustration of the Magneto Optic interactions (**Longitudinal**, **transverse** and **polar**) using the idea of Lorentz force.

the Maxwell equations [17] and is shown as:

$$R = \begin{bmatrix} \tilde{r}_{pp} & \tilde{r}_{ps} \\ \tilde{r}_{sp} & \tilde{r}_{ss} \end{bmatrix} \tag{1}$$

Where $\tilde{r}_{ij} = r_{ij}e^{i\delta_{ij}}$. The matrix R (equation 1) associates the p-polarized and s-polarized components of both incident and reflected light. The coefficients r_{ij} are the ratio of the incident j polarized electric field and the reflected i polarized electric field [16], δij are the corresponding phase angles.
Thus, the complex Kerr angles can be written as follows:

$$\Theta_K^p = \theta_K^p + i\varepsilon_K^p = \frac{\tilde{r}_{sp}}{\tilde{r}_{pp}} \tag{2}$$

$$\Theta_K^s = \theta_K^s + i\varepsilon_K^s = \frac{\tilde{r}_{ps}}{\tilde{r}_{ss}} \tag{3}$$

Where θ_K and ε_K are Kerr rotation and Kerr ellipticity respectively.
Using equations 2, 3 and the coefficients of the matrix R (equation 1), we can obtain:

$$\theta_K^p = \frac{r_{sp}}{r_{pp}} cos(\delta_{sp} - \delta_{pp}); \varepsilon_K^p = \frac{r_{sp}}{r_{pp}} sin(\delta_{sp} - \delta_{pp}) \tag{4}$$

$$\theta_K^s = \frac{r_{ps}}{r_{ss}} cos(\delta_{ps} - \delta_{ss}); \varepsilon_K^s = \frac{r_{ps}}{r_{ss}} sin(\delta_{ps} - \delta_{ss}) \tag{5}$$

4 MOKE Results

As mentioned in the introduction, a large number of MO studies have been performed to investigate how a laser pulse can effectively change the magnetic moment at femtosecond time scales. In particular, ultrafast demagnetization in a thin metallic ferromagnetic Ni film was first seen using ultrafast MOKE [1]. The authors observed an ultrafast reduction (within $2ps$) in MOKE signals. In this way they made a measurement of time-dependent remanent magnetization, which was interpreted as "photoinduced demagnetization" (See Figure 5 a), [1]). In a more extreme case, intense laser pulses were shown to increase the electron and spin temperature even above the Curie temperature, driving a ferromagnetic to paramagnetic phase transition on the femtosecond time scale. Figure 5 b) shows an example of complete quenching of ferromagnetism induced by laser pulses, it occurred within $500fs$ in $CoPt_3$ [18] .

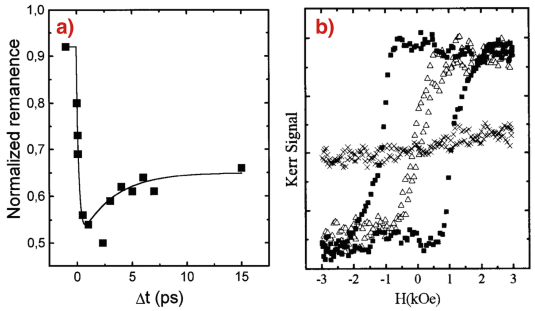

Figure 5: Femtosecond demagnetization in a) Ni and b)$CoPt_3$. a) Normalized remnant magnetization for a Ni film decreased within the first piscosecond after excitation. b) Ferromagnetic hysteresis loops of $CoPt_3$ at different time delays (squares: no pump; triangles: $-1ps$; cross: $630ps$). It shows that a ferromagnet was converted to its paramagnetic state after $\sim 500fs$. Figures a) and b) were taken from [1]and [18] respectively.

The goal of this internship is to optimize the static MOKE set-up of the LCPMR to perform TR-MOKE experiment. This will allow us to have results similar as the ones just shown on different magnetic thin films that are later on used at bigger user facilities.

4.1 MOKE at LCPMR

In the laboratory, the current MOKE experimental setup consists of a HeNe laser passing through a polarizer (to make the light linearly polarized). Then, the light passes through a focusing lens which focuses it on to the sample surface. The sample is magnetized via an electromagnet, and mounted on a rotatable sample holder. This allows the sample to rotate so that the setup can do polar or longitudinal MOKE geometries. The reflected light is modulated periodically by a photoelastic modulator, which sets its frequency. Then, the light goes through an analyzer (polarizer) and is detected by a photo-diode. The voltage from the detector is recorded by the computer through a lock-in amplifier set at the same frequency as

the modulator. Figure 6 shows the general scheme of the MOKE setup.

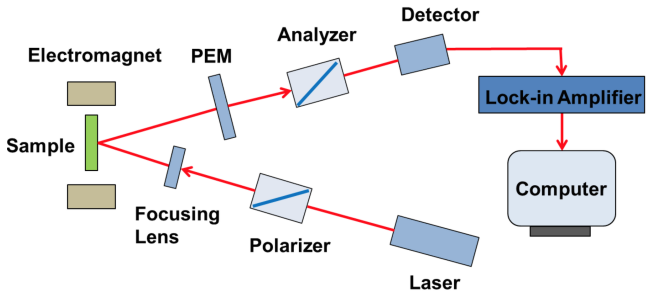

Figure 6: Current experimental setup for MOKE. Figure taken from [19].

Figure 7 shows typical hysteresis loops taken with the current setup in the longitudinal and polar MOKE geometries. Figures a) and c) show "good" measurements in both MOKE gemetries, whereas in Figure b) one can clearly observe a "drift" after saturation of the material which has no physical explanation and is therefore attributed to the experimental setup. In Figure d) one can see the problem of too much noise in the signal.

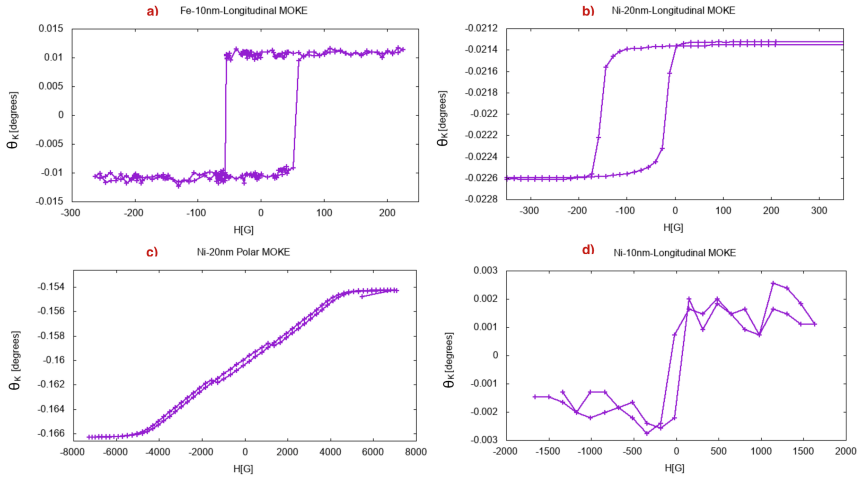

Figure 7: a) Typical hysteresis loop measurement obtained with the longitudinal MOKE geometry. b) Example of a "shift" in the hysteresis loop after saturation of the sample. c) Typical hysteresis loop measurement obtained with the polar MOKE geometry. d) Example of noise presented in the measurements.

As an attempt to optimize the current static MOKE setup in order to reduce signal noise and eliminate the unwanted "drift", a new setup will be implemented. This will differ from the initial setup in that the reflected light will be passed through a Wollaston prism, which will

separate the two components of the elliptically polarized reflected light. After the components are split, each light beam is detected by a photo-diode. The voltage from each detector will be recorded by the computer. Figure 8 shows schematically how the new experimental setup will be done. Both setups will be mounted during the internship and measurements will be taken

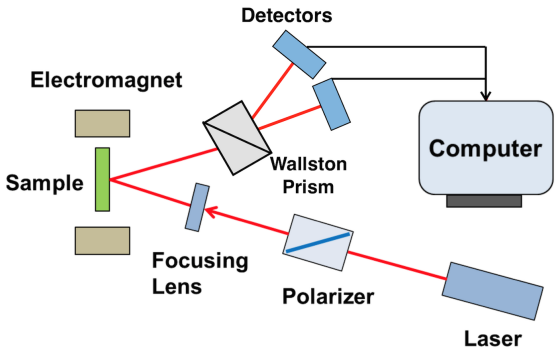

Figure 8: New proposed experimental setup for MOKE. The Walltson prism separates the reflected light into two perpendicular components.

to compare them. Finally, the best setup will be implemented for time resolved measurements at the Laboratoire d'Optique Appliquée, where there is a IR laser providing $60fs$ short pulses and the pump-probe technique will be used.

5 Proposed Internship Schedule

- Until start of April: Bibliography review, introduction to the experiment and writing of the synopsis.
- Until start of May: Mount static MOKE experiment in 2 different configurations to reduce the noise and drift.
- Until mid May: Optimization of the prefered configuration in order to apply it for time resolved measurements.
- Until mid June: Work at Laboratoire d'Optique Appliquée (LOA) to setup the TR-MOKE experiment.
- Until mid July: Use the experimental TR-MOKE setup to measure different samples with different chemical composition and with different pump fluence. Analysis of the experimental results and preparation of the final talk.

6 Conclusion

This document reviews the most relevant aspects of the internship and is intended to serve as a starting point for the work to be realized in the following months. The scientific context and motivation of the project is presented, as well as a review of the current status of the

static MOKE setup and some results obtained with it. It also presents a general view of the optimized MOKE experiment to be mounted, the goals to be achieved and the corresponding schedule.

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