Thesis subject:

Name of the laboratory: Institut Fresnel, Laser-Matter Interaction team (ILM)

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Subject's title: Ultrafast nonlinear optical response and dynamics of 2D thin films

Subject description:

Two-dimensional thin films are well-known for their high optical nonlinearities. For this reason, they are currently the best candidates for mode-locking of laser systems.[1] Recently, during a PhD thesis (Richard Verrone, thesis in progress), we optimized the thickness of Sb_2Te_3 layers to obtain significant nonlinear absorption. More specifically, the saturable absorption behavior obtained by thin films having thicknesses of about 10 nm was the highest ever reported in the field of nonlinear optics by similar experimental techniques and excitation conditions.[2]

These high optical nonlinearities are emanating from the topological insulator character of the layers which can be observed in 2D structures. However, the relation between the structural characteristics of topological insulators and their optical nonlinearities are still not clear. The target of the thesis is to shed light into the origins of the nonlinear optical properties of 2D topological insulators (like Sb_2Te_3, Bi_2Te_3, Bi_2Se_3). This will allow a better understanding of the physical mechanisms that give rise to the nonlinear refraction and absorption of the thin films. The objectives of the thesis are the following:

Objective 1: Thin film deposition and preparation. The thin films will be deposited by the electron beam deposition technique available in a modern facility established in the Fresnel Institute (Espace Photonique). An optimal crystallization of the thin film layers is necessary in order to enhance the optical nonlinearities. This is currently done by our group by heating the thin films in an oven and requires approximately one hour of annealing to achieve the highest nonlinearities. During this thesis a new experimental setup will be built, which will allow a higher precision annealing by using a high repetition rate femtosecond laser. Preliminary measurements carried out by our group have shown that annealing durations lower than 1 minute can be expected. Another advantage of this procedure is that the optical nonlinearities will be detected in-situ, during the optical annealing, which will allow a monitoring of the procedure and the possibility to stop it when an optimal nonlinearity is obtained. Finally, electron microscopy, optical microscopy and X-Ray diffraction studies will be performed in order to characterize the obtained crystalline structures.

Objective 2: Nonlinear optical studies. The deposited and optimized 2D layers will be studied by means of the Z-scan technique, already existing in Institute Fresnel. For these studies a femtosecond laser system will be employed. This is a hybrid (crystal/fiber), passively mode-locked laser delivering 400 fs duration pulses at 1030 nm. The oscillator provides pulses at a 40 MHz repetition rate. By means of a pulse picker and an acousto-optic modulator, both integrated in the laser system, the repetition rate will be adjusted. The next months an optical parametric amplifier will be installed, which will allow tuning the laser wavelength at the UV, visible and IR parts of the spectrum. This is an ideal laser system for the thesis, as it will allow the investigation of the impact of the repetition rate, the wavelength and the pulse duration (the latter can be adjusted from 80 fs up to 20 ps) on the nonlinear optical responses. These studies have never been previously done and will provide valuable information concerning the origin of the nonlinearities. Indeed, many physical contributions can arise during the laser-matter interaction that takes place in these complex materials, such as excitation of the systems to higher states, thermal effects, bleaching of the absorption due to Pauli blocking and contributions related with the topological-insulator character of the thin films.

Objective 3: Ultrafast dynamics of the 2D layers. A deeper understanding of the laser-matter interaction will be achieved through a thorough study of the carrier dynamics of the topological insulators during their excitation with light. For this reason, a pump-probe optical spectroscopy setup will be built by the PhD student. Briefly, the higher energy pump pulses (typically few nJ) will allow to generate photo-excited carriers, while less intense probe pulses (typically about 100 pJ) will detect the transmittance change of the sample. These studies will allow a precise study for several different delays between the pump and the probe pulses, which will be adjusted by an appropriate delay line. During the first part of the thesis single-wavelength studies will be performed by means of the aforementioned setup.

In the second part of the thesis, a more complex approach, will be employed in order to obtain a simultaneous detection of the ultrafast dynamics for several different wavelengths. In this case fs supercontinuum pulses will be employed, which will be generated by focusing the femtosecond laser beam on an appropriate transparent material (such as BaF_2) and using the self-phase modulation attribute of the latter.

The combination of these two approaches will allow retrieving the full spectral dependence of the investigated topological insulators, determine the relaxation times of the carriers and understand the underlying photo-physics.

The candidate shall have a solid background related with nonlinear optics.

Bibliography:
1) M. Kowalczyk *et al.* Opt. Mater. Express 6, 2273-2282 (2016).
2) R.-N. Verrone *et al.* ACS Appl. Nano Mater. 3, 7963-7972 (2020).
3) K. Iliopoulos *et al.* Appl. Phys. Lett. 101, 261105 (2012).
4) C. Moisset *et al.* Nanoscale Adv. 2, 1427-1430 (2020)