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 two PhD thesis, we optimized Sb_2Te_3 and Bi_2Se_3 layers to obtain significant nonlinear absorption. More specifically, the saturable absorption behavior obtained was the highest ever reported in the field of nonlinear optics by similar experimental techniques.[2-5]
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 is still not sufficiently explored. The target of the thesis is to shed light on the origins of the nonlinear optical properties of 2D topological insulators (like Sb_2Te_3, Bi_2Te_3 and 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 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. 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.

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. An optical parametric amplifier has been very recently installed at the exit of the femtosecond laser, which will allow tuning the laser wavelength at the UV, visible and IR parts of the spectrum (200 nm to 2.5 μm). This is an ideal laser system for the thesis, as it will allow the investigation of the impact of the repetition rate (tunable between 1 Hz and 40 MHz), the wavelength and the pulse duration (the latter can be adjusted from 80 fs up to 20 ps) on the nonlinear optical responses.

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 will allow to generate photo-excited carriers, while less intense probe pulses 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.

The combination of these two approaches will allow retrieving the full spectral dependence of the investigated topological insulators and understand the underlying photon/ electron interactions.

Apart from the nonlinear optical investigations the student will participate at all the experimental steps required for obtaining giant optical nonlinearities. This procedure includes thin film deposition, annealing, X-Ray Diffraction studies (XRD), Scanning Electron Microscopy (SEM) studies and Atomic Force Microscopy (AFM) investigations.

Bibliography:

1) M. Kowalczyk et al. Optical Materials Express 6, 2273-2282 (2016).
2) R.-N. Verrone et al. ACS Applied Nano Materials 3, 7963-7972 (2020).
3) C. Moisset et al. Nanoscale Adv. 2, 1427-1430 (2020)
4) A. Karimbana-Kandy et al. Optical Materials 143, 114211 (2023).
5) A. Karimbana-Kandy et al. « Pulse Duration Dependent Optical Nonlinearities of Bi_2Se_3 Thin Films » accepted in Optics Express (2023).