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: Nonlinear optical processes in functional plasmonic hybrid structures

Application Reference N°: 4659

1) Purpose of the visit

The purpose of the visit was to perform series of spectroscopic nonlinear-optical studies, including second harmonic generation and open/closed aperture z-scan measurements in a number of novel materials with high potential for nonlinear-optical sensing. The materials included plasmonic nanostructures (namely gold and silver nanoparticles) which exhibit high local field enhancement contributing to high nonlinear optical response and allow for precise sensing of low qualities of biological samples and topological insulators – novel material insulating in the bulk but possessing topologically protected conducting surface states. Nonlinear optics of topological insulators is mostly unexplored by the time being. Therefore the main goal was study second harmonic generation from the surface of prototypical topological insulators Bi_2Se_3 and Bi_2Te_3.

2) Description of the work carried out during the visit

The main attempt was focused on experiments on optical second harmonic generation in topological insulators. The experimental setup for the time and spectrally resolved second harmonic generation in topological insulators and topological insulators thin films was built and fully automated. For these experiments samples of Bi_2Te_3 thin films down to 7 nm in thickness (with an average being about 20-30 nm) were produced by mechanical exfoliation

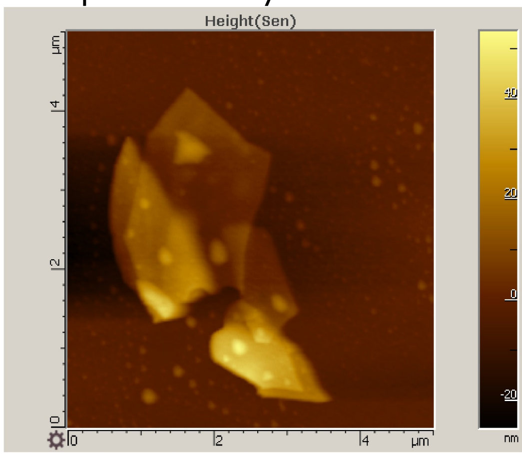


Figure 1a: AFM image of the typical Bi_2Te_3 exfoliated flake

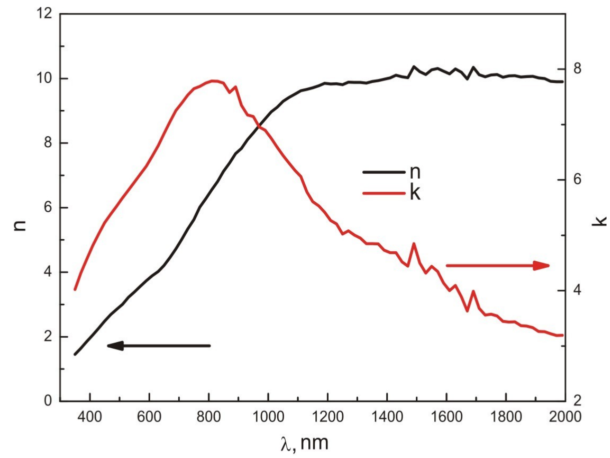


Figure 1b: n,k spectra from spectroscopic ellipsometry

technique and characterized by the means of optical and AFM microscopy (Fig 1a). Bulk crystals of Bi_2Te_3 were characterized with spectroscopic ellipsometry (Fig 1b) which was analyzed with point-to-point interpolation algorithm and allowed for precise determination of the real and imaginary parts of the refractive index in a broad spectral range. Based on the results of the spectroscopic ellipsometry the fundamental wavelength for second harmonic generation was chosen to be 1300 nm in order to produce comparable nonlinear-optical response from the both surfaces of the thin film in transmission. The spectroscopic ellipsometry also explicitly confirmed absence of any oxidized surface layer on the samples of the topological insulators.

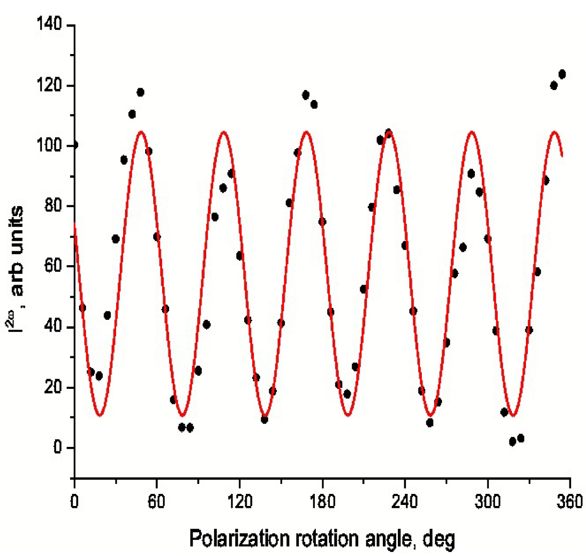
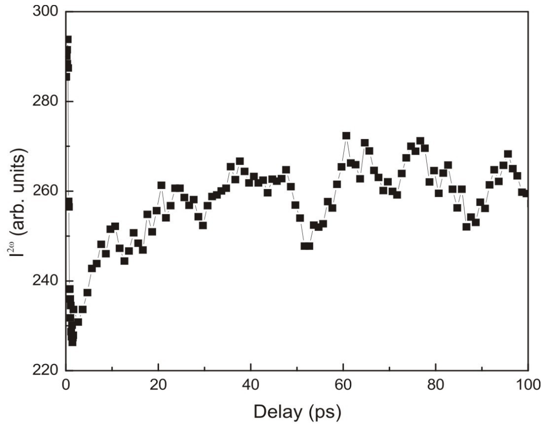


Fig 2: SHG rotational anisotropy from Bi_2Te_3 film

Figure 2 represents results of second harmonic rotational anisotropy measured in transmission from the semitransparent film of Bi_2Te_3. The observed 6-fold symmetry indicates the presence of the (111) crystallographic plane on the surface of the topological insulator film. For the time-resolved measurements the 800 nm beam was used as a pump. Figure 3 represents the results of the time-resolved SHG studies in Bi_2Te_3 thin films. The measured kinetics demonstrate a conventional biexponential decay typical for these kind of materials with the clearly resolved periodic oscillations with two different periods. The fast oscillation, corresponding to the period about 0.6 ps is consistent with the A_{1g}^1 transversal optical phonon mode with the frequency 1.86 THz which has been observed so far with



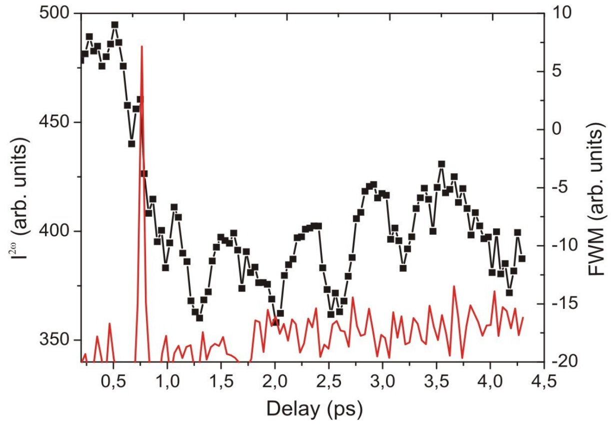
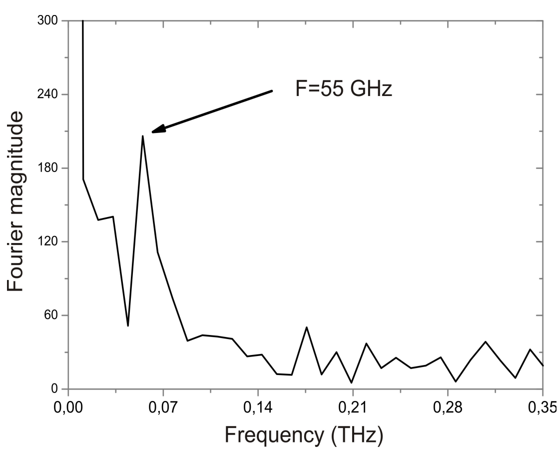


Figure 3a: Long-time scale time-resolved SHG

Figure 3b: Short-time scale time-resolved SHG

Raman scattering [1] and conventional linear pump-probe measurements [2], however for the first time here observed in second harmonic response. The second oscillation has the period about 20-30 ps which is to long to be explained with any sort of optical phonon mode. One of the possible



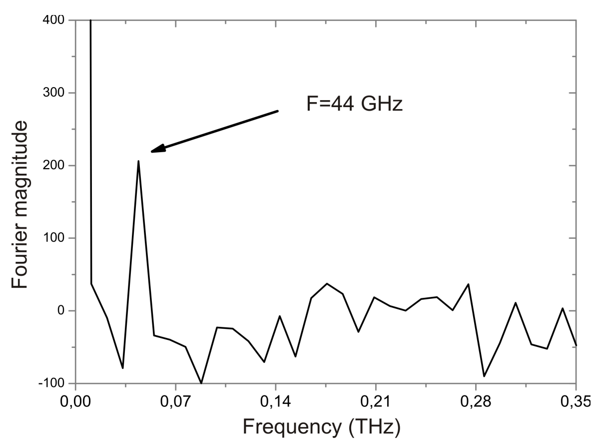


Fig 4a: FFT of the SHG kinetics for the "thin" Bi_2Te_3 film

Fig 4b: FFT of the SHG kinetics for the "thick" Bi_2Te_3 film

explanations of these results in a standing acoustic wave which is formed across the thin film and therefore has the period determined by the thickness of the film. To prove this assumption we performed time-resolved SHG studies for two Bi_2Te_3 flakes with different thickness as determined by difference in their optical transmission. Results of fast fourier transform for these two cases are presented in figures 4 (a and b). It its clearly visible that the frequencies of the observed oscillation depend on the thickness of the topological insulator film. Taking the velocity of sound from [3] to be 2460 m/s one can estimate he thickness of the film to be 22.4 nm for the "thin film" and 28 nm for the "thick film" which is comparable to the values obtained from AFM studies for typical flakes. Physical mechanism that underlies the contribution of a standing acoustic wave to the second harmonic response is believed to be so-called strain-induced second harmonic generation that can be phenomenologically described by the following additional elements of the second order susceptibility tensor:

$$\chi_{ijk}^{(2)} = P_{ijklm}\sigma_{lm},$$

where P_{ijklm} is the nonlinear optoelastic tensor and the σ_{lm} is the tensor of the mechanical stress induced by the standing acoustic wave.

References:
[1] Y. Wang, L. Guo, X, Xu, J, Pierce, R. Venkatasubramanian, Phys. Rev. B 88, 064307 (2013)
[2] A.Q. Wu, X. Xu, R. Venkatasubramanian, Appl. Phys. Lett 92, 011108 (2008)
[3] J.O Jenkins, J.A. Rayne, R.W. Ure, jr., Phys. Rev. B 5, 3171 (1972)

3) Description of the main results obtained

1) Second harmonic generation from Bi_2Te_3 thin flakes demonstrated for the first time. Crystal symmetry confirmed to be (111)
2) Time-resolved SHG studies in thin films of a topological insulator performed for the first time.
3) Signatures of coherent optical phonons in bismuth telluride demonstrated in SHG response for the first time
4) Oscillations with period 20-30 ps observed in long-time scale SHG pump-probe measurements and attributed to formation of the standing acoustic wave across the thin film of topological insulator

4) Future collaboration with host institution (if applicable)

Future collaboration with the group of experimental biophysics and nanotechnology is planned to continue and extend the research on topological insulators as well as other two-dimensional materials such as monolayer MoS_2. The experimental findings on contribution of a standing acoustic wave in thin films of topological insulator to second harmonic response need to be generalized in the form of thorough thickness-dependent studies with independent control over the thickness of the exfoliated films. Other research project on topological insulators involves nonlinear magnetoopics from the surface states of topological insulators coupled via exchange interaction to a thin magnetic layer on the surface. Lastly plasmonic hybridization and studies of the topological insulator surface with deposited biological samples will be performed.

Another research project involves monolayer films of MoS_2 - a material known for its remarkable optoelectronic properties that allow to explore internal valey degree of freedom. Nonlinear optics of MoS_2 and other 2D TMDs is predicted to demonstrate several new phenomena due to the unique electronic properties of the material which have not been explored yet.

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)***

6) **Other comments (if any)**