Terahertz Spectroscopy of 2D Materials

Lyubov V. Titova

Department of Physics, Worcester Polytechnic Institute, Worcester, MA 01609

Two-dimensional, or 2D, materials are attracting considerable attention as a testbed for new physics and as candidates for applications in flexible nanoscale high-speed optoelectronics, solar energy conversion, and chemical sensing. Most unique properties of 2D materials stem from their highly anisotropic optical and electronic properties. Terahertz (THz) spectroscopy provides access to those properties with ultra-high time resolution and without the complications of electrical contacts. I will describe how we apply time-resolved THz spectroscopy to probe ultrafast dynamics of charge carriers in two 2D layered materials with vastly different properties: semiconducting GeS and GeSe and metallic MXene $Ti_3C_2T_{x.}$

We find that above band gap photoexcitation of group-IV monochalcogenides GeS and GeSe with ultrashort laser pulses results in emission of nearly single-cycle THz pulses due to a zero-bias surface shift current as excitation of an electron from the valence to the conduction band produces a spatial shift of the electron charge density [1]. Experimental demonstration of THz emission by the surface shift current puts this layered group-IV monochalcogenides forward as a candidate for next generation shift current photovoltaics, nonlinear photonic devices and THz sources.

Layered 2D $Ti_3C_2T_x$ Mxenes, where T_x is a surface termination groups such as =O, –OH, or –F, are metallic and promising as precious-metal-free flexible transparent electrodes, conductive materials for electrochemical energy storage, and even as candidates for femtosecond laser mode locking and optical isolation applications. Using THz spectroscopy, we have studied intrinsic carrier dynamics and microscopic conductivity in Mxene thin films. We demonstrate that $Ti_3C_2T_x$ conductivity and THz transmission can be manipulated by photoexcitation, as absorption of near-infrared, 800 nm pulses causes transient suppression of the conductivity that recovers over hundreds of picoseconds [2]. The possibility of control over THz transmission and conductivity by photoexcitation suggests the promise for application of $Ti_3C_2T_x$ Mxenes in THz modulation devices and variable electromagnetic shielding.

[1] "Ultrafast Zero-bias Photocurrent in GeS Nanosheets: Promise for Photovoltaics," K. Kushnir, M. Wang, P.D. Fitzgerald, K.J. Koski, L.V. Titova, *ACS Energy Lett*. 2, 1429 (2017).

[2] "Equilibrium and non-equilibrium free carrier dynamics in two-dimensional $Ti_3C_2T_x$ MXenes: THz spectroscopy study," G. Li, K. Kushnir, Y. Dong, S. Chertopalov, A.M. Rao, V.N. Mochalin, R. Podila and L.V. Titova, *2D Materials* 5, 035043 (2018).