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Multi-photon absorption and subband photodetection in monolayer transition-metal di-chalcogenides

Semiconducting materials capable of simultaneously absorbing multi-photons and subsequently generating photoluminescence or photocurrents have great potential for applications in multi-photon microscopy and light frequency up-conversion. Two-Photon Absorption (2PA) and Three-Photon Absorption (3PA) in semiconductors can also be applied to autocorrelation, optical signal processing, sub-band photodetectors, quantum detectors, coherent control and others. Therefore, understanding 2PA and 3PA mechanisms are of direct relevance to the above-mentioned applications. However, there are a lack of theoretical understanding of 2PA and 3PA in recently-emerged, two-dimensional (2D) semiconductors. 2PA phenomena have been utilized for probing dark excitons in monolayer Transition-Metal Di-Chalcogenides (TMDCs) [1]. Large yet varied 2PA coefficients (103~105 cm/GW at 800 or 1040 nm) have been observed experimentally for monolayer MoS2 [2] and WS2 [3, 4] made by Chemical Vapor Deposition (CVD) methods. Notably, a high amount of defects/impurities could present in these CVD-made samples and hence, the observed 2PA might not directly associate with the intrinsic nature of monolayer TMDCs. To the best of our knowledge, there is no report on the magnitude and spectrum of 2PA in highly-crystalline monolayer TMDCs. Furthermore, there is no report on 3PA in TMDCs Here, for the first time, we present our theoretical models [5, 6] to quantitatively predict the 2PA and 3PA magnitude and spectrum in monolayer MoS2. Our models are based on quantum mechanical perturbation theories. To verify our models and to demonstrate multi-photon absorption of TMDCs for applications, we fabricate an exfoliated monolayer MoS2 sub-band photodetector for multiphoton-induced photocurrent measurements. From the photocurrent measurements, we experimentally determine both 2PA and 3PA spectra of monolayer MoS2. We find that our models are in agreement with the measurements, within one order of magnitude. Our theoretical and experimental findings pave the way to develop sensitive infrared MoS2-based multi-photon detectors.

Biography

Wei Ji obtained his B.Sc. in Physics from Fudan University, Shanghai, China (1982) and was awarded M.Sc. in Optoelectronics and Laser Devices and PhD in Physics by Heriot-Watt University, Edinburgh, the UK in 1984 and 1986, respectively. Currently, he is a professor in the Physics Department, National University of Singapore. He is also a visiting professor at Shenzhen University, Shenzhen, China. His research interests cover the nonlinear optical and emission properties of nano-scale materials and their applications for photonics and optoelectronics. He has authored 200 research papers or more published in scientific journals. Total citation of his papers is over 8000 with the h-index of 52 or higher.

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