Dirección de Formación Académica

CENTRO DE INVESTIGACIONES EN OPTICA, A.C.

Advisor: Dr. Bernardo Mendoza Santoyo

Committee Members:

Dr. Eden Morales Narváez
(Secretary)

Dr. Norberto Arzate Plata
(Evaluator)

Dr. Bernardo Mendoza Santoyo
(Chairperson)

Thesis: "AB-INITIO CALCULATION OF TWO-PHOTON ABSORPTION FOR SEMICONDUCTORS"

Summary:

A theoretical derivation of two-photon absorption (2PA) from bulk, surface and 2 dimensional (2D) semiconductors, based on the length gauge analysis and the electron density operator, is formulated. Within the independent particle approximation, the nonlinear third order susceptibility tensor and the two-photon absorption coefficient are calculated, including the scissors correction needed to correct the well-known underestimation of the local-density-approximation band gap. Using time-reversal symmetry, it is shown that the expression for the susceptibility tensor is finite at omega=0, avoiding nonphysical divergences presented in previous calculations when omega goes to zero. Ab initio band structure calculations using different pseudopotential schemes that include spin-orbit coupling are used to calculate the 2PA for several semiconductors, and the calculations are compared with available experimental results for photon energies that are below the optical band gap. The TPA results for the chosen bulk semiconductors cover a wide range of wavelengths as well as a wide intensity range, in such a way that depending on the sought application one would be able to chose among the studied materials. For instance, the semiconductors with the biggest absorption could be used as components in flourophores for two-photon excitation microscopy, also these materials could be used in multiphoton microfabrication and lithography technologies. Moreover, the materials with wide band gap could be used for optical power limiting, optical data storage, or two-photon photodynamic therapy technologies. In addition, the 2PA susceptibilities for the following surface structures GaAs-1x1(110):Sb, Si-1x1(111):H and clean GaAs-1x1-110 were calculated, obtaining a spectral range that shows a remarkable widening of bandwidth for GaAs-1x1(110):Sb and Si-1x1(111):H structures compared with the corresponding bulk response, and for clean GaAs-1x1-110 surface the spectral shape shows a resonance in the terahertz region. Finally, we calculated the 2PA susceptibility for the following 2D monochalcogenides materials GeSe, SnSe, SnS and GeS. These materials are commonly used in optoelectronic devices and our study motivates the experimental investigation of 2PA in 2D structures. The 2PA susceptibility of above surfaces and 2D materials is as large as the standard bulk semiconductors, thus opening a myriad of application for 2PA, such as non-invasive terahertz medical applications. In the future, we are going to calculate 2PA for other crystalline structures besides FCC, extending the calculation to more types of semiconductors. In addition to our numerical results presented in this work, the theoretical derivation of 2PA presented in this thesis opens the possibility of the study of other third-order nonlinear optical phenomena. For example, four-wave mixing, Raman scattering, third harmonic generation, and the calculation of the nonlinear third order susceptibility in order to obtain the nonlinear refractive index.