Abstract

Nonlinear optical properties are of paramount importance in diverse directions in scientific and technological contexts, as they enable the manifestation of advanced optical phenomena that transcend linear interactions. After the discovery of the laser, the nonlinear light-matter interaction plays an important role in a wide range of applications such as optoelectronic devices, and ultrashort lasers. Second-order nonlinear optical processes are fundamental phenomena that arise when non-centrosymmetric materials interact with intense light, resulting in the generation of new frequencies through nonlinear polarization effects. On the other hand, when materials encounter intense light, third-order nonlinear optical processes, encompassing nonlinear refraction, nonlinear absorption, the generation of new frequencies, and other distinct responses, significantly influence the realm of modern optics. Nonlinear refraction helps with optical switching, signal processing, and creating solitons, while nonlinear absorption is used to design optical limiters for protecting against strong light and controlling interactions in photonic devices. This thesis explores the investigation of the broadband third-order optical nonlinearity of various opto-electronic materials, including zero-dimensional (0D) carbon dots, two-dimensional (2D) metal monochalcogenide Gallium Telluride (GaTe), and microstructural (three-dimensional network) citraconic acid-directed cadmium-based metallogel (Cd-CA), studied within the femtosecond regime. Using the Z-scan technique, this study investigates the characteristics of light traveling through carbon dots across a photon energy spectrum of 2.07 eV to 2.58 eV. Positive nonlinear refraction coefficients, denoted as n_2', manifest prominently at 2.38 eV and 2.48 eV. Conversely, other excitation energies exhibit negative n_2' values. The dispersion of n_2' values guides the utilization of carbon dots for optical switching across a broad spectral range. Exploring the nonlinear optical (NLO) characteristics of atomically thin 2D layered Gallium Telluride, open aperture (OA), and closed aperture (CA) Z-scan measurements are conducted in the spectral range of 520-700 nm. Notably, exfoliated layered GaTe exhibits robust saturable absorption and significant negative Kerr nonlinearity. The observed nonlinear refraction coefficient, n_2', reached $-(7.61 \pm 0.07) \times 10^{-1}$ cm^2/GW under near band edge excitation at 700 nm. Additionally, the material exhibits a noteworthy nonlinear absorption coefficient, α_{NL}, of $-(18.02 \pm 0.20) \times 10^4$ cm/GW at an intensity of 40 GW/cm^2. Transient absorption spectroscopy provides insights into carrier dynamics and underlying mechanisms, including Pauli blocking. The nonlinear optical data from exfoliated GaTe, discussed in this

study, holds promise for potential applications in ultrafast photonic devices, including optical switching, passive Q-switching, and mode-locking. The citraconic acid-directed supramolecular metallogel of Cd(II), referred to as Cd-CA, showcases thixotropic behavior and self-healing properties. The study extensively explores the charge transport characteristics of Cd-CA metallogel, particularly by fabricating a Schottky barrier diode using an Al/Cd-CA/ITO configuration, unveiling remarkable semiconducting attributes. Additionally, the broadband nonlinear optical (NLO) properties of Cd-CA metallogel are investigated through the femtosecond Z-scan technique at excitation photon energies below its bandgap (E_g= 4.81 eV) at 2.98 eV, 2.34 eV, 1.65 eV, and 1.24 eV. Intriguingly, the gel displays positive nonlinear refraction and multiphoton absorption (MPA), encompassing two-, three-, and four-photon processes. Specifically, at 2.98 eV, the material demonstrates two-photon absorption, at 2.34 eV, it exhibits simultaneous two- and three-photon absorption, and at 1.65 eV excitation, it shows enhanced three-photon absorption. The material also manifests four-photon absorption with a low optical limiting threshold at 1.24 eV. This study elucidates the factors contributing to self-healing, semiconducting, and nonlinear optical properties, offering a potential avenue for its application in optical switches and limiters.

Keywords: Nonlinear optical properties, Carbon-dots, Metal monochalcogenide, Supramolecular metallogel, Schottky diode, Nonlinear refraction, Multiphoton absorption, Saturable absorption, Optical limiting