Abstract

With a revolutionary technological improvement in the field of ultrafast lasers, shorter time domain laser pulses with broader bandwidth have become available for high resolution ultrafast spectroscopic techniques. Not only the high peak power from ultrafast laser pulse is suitable for inducing electronic nonlinearity resulting in nonlinear absorption and nonlinear refraction from non centrosymmetric materials, but also the shorter pulse width enables it to excite and then probe the characteristic life time of the charge carriers, excitons and phonons constitute the system. Sufficient spectral bandwidth of an ultrafast laser pulse can also be used to generate new frequency such as terahertz radiation using nonlinear crystals by frequency mixing processes. In the present thesis we have exploited these advantages and have investigated the ultrafast nonlinear optics and time resolved spectroscopic behavior of carbon based 2D materials such as graphene oxide (GO), reduced graphene oxide and semiconductor system such as Gallium Selenide (GaSe), Indium (In) doped GaSe and Cromium (Cr) doped GaSe.

Pristine graphene sheet consisting entirely of sp^2- hybridized carbon atoms has already been identified having large optical nonlinearity. However GO, a precursor of graphene having both sp^2 and sp^3 - hybridized carbon atom is increasingly attracting cross-discipline researchers for its controllable properties by reduction of oxygen containing groups that reside in GO. Thus GO is chosen for investigation not only due to it's ease of fabrication, solvent and substrate compatibility but also due to achievable band gap engineering using various reduction techniques (photo thermal and chemical) which establish the control over oxygen functionality in GO sheets. Both real and imaginary part of third order optical nonlinearity is probed in GO and gradually reduced GO via single beam open aperture and close aperture z-scan at variable wavelength and pulse energies which conclude that reduction of oxygen functional groups on GO plane can be used to tune it's nonlinear optical properties. Variation of pump wavelength and pump power using broadband transient absorption spectroscopy (TAS) in these materials further reveal unique features such as delay dependent optical switching, non-degenerate nonlinear absorption property as well as different carrier relaxation path ways. Also, terahertz time-domain spectroscopy (THz-TDS) has been carried out in GO, partially reduced GO to achieve tunability of optical transmission and electrical properties in THz regime. Lastly, doped and un-doped GaSe crystals grown by modified vertical Bridgman method are investigated for phonon

mode absorption by THz-TDS at room temperature. The effect of doping on plasma frequency and momentum relaxation time are explained under the framework of modified Drude-Smith model. The resonance peaks of THz absorption spectra are attributed to the different combinations of phonon modes obtained by Raman spectroscopy.

Keywords: Femtosecond laser, Z-Scan, Transient absorption spectroscopy, THz time domain spectroscopy, Graphene Oxide, Reduced Graphene Oxide, GaSe, In-GaSe, Cr-GaSe