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Optically induced modulation of insulator-metal transition materials in thin-films and plasmonic nanostructures

All-optical information processing is superior to conventional electronic devices because light can achieve faster modulation speeds and can propagate without loss (no Ohmic loss). The realization of all-optical information processing relies on the ability to modulate light using light, that is, to develop an integrated device that switches light with light in the same way as the transistor switches an electron current with a current. Such all-optical modulators would consume less energy and have a small footprint amenable for on-chip integration so that advanced optical communication systems (guiding and modulating optical signals in their optical form without electrical conversion) could be realized.

All-optical modulators based on highly correlated oxides such as VO_2 and NbO_2 have been proposed. As these materials undergo an insulator-metal transition, their optical properties change drastically enabling the strongmodulation of the light. The insulator-metal transition can be induced optically. The current understanding of the mechanism for the ultra-fast modulation follows the Mott-Hubbard description of the insulator-metal transition. The transition occurs when the concentration of the optically induced carriers reaches a threshold above which the bandgap of the metal oxide material collapses causing an increase in the carrier concentration that results in a drastic change in the material reflection. This effect appears as a threshold in the magnitude of the reflectance as a function of the pump intensity, and is a nonlinear behavior. At a higher threshold (high fluence regime), the relaxation of photo-excited carriers induces a crystal phase change that results in a long-lived metallic state. Recently, a photo-induced transition described as a transition from the monoclinic semiconducting phase to a metastable monoclinic metallic phase has been reported for VO_2 under a mid fluence regime. However, the observation of the full-time dynamics (response and recovery) of this photo-induced transition has not been reported and fully characterized.

CLARK reports the first observation of the full-time dynamics (response and recovery) of the optically induced VO_2 metastable monoclinic phase under a mid fluence regime, clarifies the modulation of VO_2 and NbO_2 under the low fluence regime and designs an all-optical waveguide-integrated modulator using VO_2. The thesis manuscript consists of seven Chapters.

Chapter One reviews the field of all-optical modulators and introduces insulator-metal transition materials. The importance of pump-probe ultrafast time-resolved spectroscopy to characterize the time-dependence of the photoinduced response of the transition materials is emphasized.

In Chapter Two, the pump-probe setup with a 200 fs resolution developed for this project is described. Emphasis is placed on the reduction of the noise that allows for the accurate determination of the recovery times and the re-pump analysis (analysis of two consecutive pump pulses separated by a delay of 100 ps) that helps to clarify the response of the material.

The fabrication and characterization of the insulator-metal transition thin films are explained in Chapter Three. Accurate determination of the thin-film thickness required for the analysis of the results is achieved using X-ray reflectance.

In Chapters Four and Five, the results and interpretation of the ultrafast spectroscopy on the VO_2 and NbO_2 thin films and nanostructures are reported. For VO_2, the transition from the semiconducting monoclinic phase to the metastable metallic monoclinic phase is observed for the first time under a mid-fluence regime. The metal monoclinic phase is found to fully recover on an ultrafast sub-nanosecond scale (full recovery in 600 ps), thus making GHz all-optical modulation possible. Within this recovery time, a minimum lifetime or persistence of the order of tens of picoseconds is also observed. The length of this persistence can be controlled by the pump fluence and thus offers a unique means to control the duration of the induced modulation with possible applications in neuromorphic computing. The mechanism for such a transition is discussed by recording the optically induced modulation for different film thicknesses, and a thermal origin (phonon temperature) of this transition mechanism is suggested. Under a low fluence regime, VO_2 and NbO_2 exhibit similar behavior with a full recovery time of 100 and 200 picoseconds, respectively. The slightly faster recovery of VO_2 is attributed to its high thermal conductivity. The observed relation between the modulation and the pump intensity is reported to be linear in a first approximation. Indeed, no threshold for a transition in the modulation is observed, and the re-pump experiment confirms the linearity of the modulation response (two delayed pump pulses of the same intensity give the same modulation). The mechanism for light modulation under the low regime fluence is attributed to the known effects of photoinduced carriers in semiconductors with a dominant effect of the bandgap shrinkage for VO_2 and NbO_2. The subsequent relaxations of the photo-induced carriers and also the phonon relaxation explain the observed relatively slow recovery times.

In Chapter Six, the control of the insulator-metal transition by the excitation of plasmonic modes in a plasmonic nanostructure that includes the highly correlated metal oxide materials is investigated. An optical modulator is realized by integrating the phase-transition materials into a resonant plasmonic nanostructure. Upon triggering a phase transition by plasmonic excitation, the proposed device changes from being transparent to the signal light to absorbing to the signal light. This change in the optical properties is made possible by pumping the device with a control light that causes the resonance of the plasmonic nanostructures. The resonance of the plasmonic nanostructures enhances the electric field which should help to trigger a reversible and ultrafast phase transition of the phase-transition material. An Au/VO_2 plasmonic nanohole array is used to demonstrate this concept and a reduced threshold for the insulator-metal monoclinic transition as well as a strong modulation are reported.

In Chapter Seven, the design of an all-optical waveguide-integrated modulator using VO_2 is proposed and its performance analyzed theoretically. The modulator consists of a waveguide modified by a sub-wavelength plasmonic grating with the VO_2 filled nanogaps. In the proposed modulator, the control light excites gap plasmons within the nanogaps, resulting in an enhancement of photoabsorption. The photoexcited electrons trigger a photo-induced insulator-to-metal phase transition. Upon the phase change, the waveguide cannot transmit the signal light, which is instead absorbed in the hybrid nanostructure. An estimated extinction rate of 26.8 dB/µm is obtained. The signal light and control light are both transmitted through the same waveguide, allowing a fully integrated design with on-chip data processing.

よって本論文は博士（工学）の学位請求論文として合格と認められる。