論 文 の 内 容 の 要 旨

論文題目　Optically induced modulation of insulator-metal transition materials in thin-films and plasmonic nanostructures（金属・絶縁体転移マテリアルの薄膜及びプラズモニック構造の光誘起変調）

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The demand for higher speed and larger volume telecommunications networks and higher speed computing devices is expected to increase significantly in coming years. The reliance of modern devices on electrical signals for all data processing and manipulation, however, represents a significant hurdle that must be overcome to meet this demand. Electrical signals suffer from resistive losses that make electrically based devices inefficient and demand large amounts of power. Furthermore, the operating frequency of electrical devices is limited by the excessive heat generated from the resistive losses, along with other fundamental phenomena affecting electrical signals, such as inductance and capacitance.

The replacement of electrical signals with optical signals, which have much lower propagation losses than electrical signals and don’t suffer from inductance and capacitance related signal distortion, is the most promising way to meet the demands for more efficient and higher speed telecommunications and computing devices. Optical signals, transmitted by fibre optic cables, are already widely used for high-efficiency and high-speed data transmission in large scale telecommunications networks and server interconnects in data centres; however, the modulation and processing of these signals requires electro-optic devices, such as electro-optic modulators, photodetectors and photo-diodes. This adds significant inefficiency due to the frequent need to convert optical signals to electrical signals and back, and limits the maximum modulation speed that can be achieved.

For these reasons, the development of all-optical modulators that modulate optical signals using other optical signals is of great interest. All-optical modulators have been widely studied for many years, with silicon photonic based all-optic modulators with modulation frequencies nearing THz speeds having been demonstrated; however, the vast majority of devices studied thus far have relied on weak non-linear phenomena. This reliance on weak non-linear phenomena has necessitated large microscale sizes and has resulted in modulation depths that are insufficient for use as all-optical switches in all-optical computers. For all-optical computing and all-optical telecommunications devices to be a viable alternative to electrical computing and electro-optical devices, all-optical modulators with nanoscale sizes and strong modulation depths must be developed.

Insulator-metal transition materials, such as VO_2 and NbO_2, are of particular interest for the development of all-optical modulators. Insulator-metal transition materials undergo a transition from an insulating state to a metallic state upon reaching a specific insulator-metal transition temperature, and as a result of the transition their optical properties change drastically. In the case of VO_2 this has been used to develop nanoscale thermo-optic and electro-optic modulators with large modulation depths. When it was demonstrated that the insulator-metal transition of VO_2 can be induced through optical excitation, the potential for VO_2 to be used in an all-optical modulator was realized; however, thus far, all-optical modulators using VO_2 have been limited to low MHz speeds and the insulator-metal transition was induced photo-thermally. Although the initial dynamics of the photo-induced insulator-metal transition of VO_2 has been widely studied, the recovery of VO_2 from this photo-induced state and whether insulator-metal transition materials can recover at sub-ns speeds to achieve ultrafast all-optical modulation has yet to be studied.

In this thesis, the optically induced modulation of two insulator-metal transition materials, VO_2 and NbO_2 is studied, with a focus on the dynamics of the recovery from the photo-induced state after excitation by a femtosecond laser pulse. The effect of nano-structuring on the recovery dynamics is then investigated, followed by a demonstration of strong modulation and sub-ns recovery using a plasmonic nanohole array structure. Finally, a waveguide-integrated all-optical modulator using VO_2 as an active material with a nanoscale size, extremely high modulation extinction ratio and low switching threshold is designed to demonstrate the potential of these insulator-metal transition materials for all-optical modulation in integrated photonic devices.

In the initial study of the recovery dynamics of the optically induced modulation of VO_2 and NbO_2 thin-films, three main conclusions are drawn. First, the optically induced modulations of VO_2 and NbO_2 at low pump fluences are found, contrary to the consensus in the literature, to be a purely linear process, unrelated to the insulator-metal transition and with recovery dynamics determined by the thermal dissipation of the thin-films. In a mid-fluence region, evidence of the recently discovered monoclinic metal state of VO_2 is found, and for the first time, it is demonstrated that VO_2 can recover from this monoclinic metal on an ultrafast, sub-ns timescale. This represents the first demonstration of the full recovery of VO_2 on a sub-ns timescale after optical excitation, and the first evidence of a non-thermal recovery process for the monoclinic metal state of VO_2.

In order to determine the maximum potential modulation speed of a VO_2 based all-optical modulator, the effect of film thickness and nano-structuring on the recovery dynamics of VO_2 and NbO_2 are studied. It is found that the monoclinic metal state of VO_2 has a minimum lifetime of 10s of ps and that thermal dissipation does not represent a bottleneck in the recovery of VO_2 to its equilibrium optical properties after undergoing an optically induced insulator-metal transition. Through the use of ultra-thin thin-films, full recovery of VO_2 in as little as 100 ps and NbO_2 in as little as 200 ps after optical excitation was demonstrated and full recovery of VO_2 after a photo-induced insulator-metal transition to the monoclinic metal phase in as little as 600 ps was demonstrated.

Strong modulation with a low pump fluence and fast recovery is demonstrated using a Au/VO_2 plasmonic nanohole array. The plasmonic nanohole array allows for a small volume of VO_2 to enable ultrafast recovery of the VO_2 for high-speed modulation while maintaining a strong modulation. The periodic plasmonic structure is also designed to concentrate the pump light that induces the insulator-metal transition in VO_2. The structure is fabricated using nanofabrication techniques and then the optically induced modulation of the structure is characterized. A reduced threshold, compared to a VO_2 thin-film, for the monoclinic metal state is found and a strong modulation intensity is obtained, despite the low volume fraction of VO_2

in the structure. In the mid-fluence region, the structure was found to have a strong modulation after undergoing an insulator-metal transition to the monoclinic metal state while recovering on a sub-ns timescale, representing the first demonstration of a VO_2 based all-optical modulator with sub-ns recovery.

To demonstrate the potential of all-optical modulators using insulator-metal transition materials, a waveguide-integrated all-optical modulator incorporating VO_2 with a nanoscale size is designed. A sub-wavelength Au/VO_2 nanostructure is used to take advantage of the concentrating effects of the plasmonic modes supported by the Au in order to enable low threshold switching of the all-optical modulator. The Au/VO_2 nanostructure is further found to behave as an effective medium, whose dimensions can be optimized to enhance the transmittance of the device when VO_2 is in its insulating state and minimize the transmittance of the device when VO_2 is in its metallic state, thus achieving strong modulation while maintaining a nanoscale size. A 550 nm long, 320 nm wide and 300 nm tall all-optical modulator with an extinction ratio of 26.85 dB/µm, the highest yet reported for a VO_2 based optical modulator, is designed.

The demonstration of the optically induced modulation of insulator-metal transition materials with full recovery of the modulation in as little as 600 ps, and the VO_2 based nanoscale all-optical modulator with extremely strong modulations detailed in this thesis shows the potential for insulator-metal transition materials to enable high-speed integrated all-optical modulators. The discovery of a ps-scale bottleneck in the recovery of the optically induced monoclinic metal state precludes the possibility of THz speed all-optical modulators as had been hoped for all-optical computing; however, GHz speeds all-optical modulators have the potential to revolutionize optical signal processing in telecommunications networks. Furthermore, the ability to control the length of the induced modulation using pump fluence is a unique feature unseen in electrical modulator devices that may see practical use in new burgeoning fields such as neuromorphic computing and memristors.