Optically Induced Absorption Changes in a Polycrystalline ZnO/ZnMgO Thin-Film Heterostructure

Samuel Haeuser, Trenten Smith

Faculty Sponsors: Eric Gansen, Seth King, Department of Physics

ABSTRACT

Zinc oxide (ZnO) is a semiconductor material exhibiting a wide bandgap in the ultraviolet (UV) spectral region, and is a promising material for use in short-wave optoelectronic devices such as optical switches. In this work, we investigate the all-optical switching properties of a polycrystalline ZnO/ZnMgO heterostructure grown by DC sputter deposition. A120-ps control pulse tuned to the band edge of the film modifies the transmission of a weaker signal pulse. In the absence of the control pulse, the signal light is heavily absorbed by the ZnO film. When incident, the control pulse resonantly excites electron-hole pairs in the film that decrease the material's absorption by filling energy states and screening the electric field built into the ZnO. Consequently, more signal light is transmitted by the film. Here, we present the results of differential-absorption measurements that show how the absorption changes vary with polarization, control power, and the time delay between the control and signal pulses.

INTRODUCTION / BACKGROUND

Optical communication and processing systems require switches that can be used to encode information on a beam of light or stream of light pulses. An optical switch functions similarly to an electrical switch. Electrical switches are commonly used to control the passage of electric current in both household and commercial circuits. When the electrical switch is turned off the current is blocked, and when it is turned on the current is allowed to pass. By comparison, an optical switch is used to control the passage of light; when it is turned off the light is blocked, and when it is turned on the light is allowed to pass. Thus, digital information can be encoded on the optical signal, allowing the information to be written to a disk for storage or transmitted from one place to another.

One way optical switching can occur is by a simple mechanical shutter; however, mechanical processes are far too slow to be useful in today's communication and computation systems, motivating interest in optical switches that use electronic processes in semiconductor materials. In these devices, the signal light is sent through the semiconductor material where the amount of light that is transmitted through the material is modulated by an electrical or optical control.

Electrically controlled switches use voltage to modulate the transmission of semiconductor material. These devices, commonly referred to as electro-absorption modulators (EAMs), often use multiple-quantum-well (MQW) structures for the switching medium that consist of alternating layers of semiconductor materials with differing bandgap energies. Modulation is achieved by applying a voltage across the MQW which shifts the absorption resonances near the band edge of the wells to lower energies through the quantum-confined Stark effect (QCSE) [1]. This effect can be used to modulate the transmission of signal light that is tuned to the absorption resonance. When no voltage is applied, the signal light is highly absorbed by the MQW (the switch is off). Conversely, when the voltage is applied, the MQW becomes more transparent to the signal (the switch is on). EAMs have been shown to operate at speeds much faster than that of mechanical shutters [2]; however, their speeds are still limited by that of conventional electronics.

Alternatively, in an all-optical switch (AOS), an optical control pulse is used to modulate the signal light. Such devices are amongst the fastest switches known, exhibiting speeds on sub-ps timescales, and are well suited for specific applications, such as serial-to-parallel conversion and header recognition [3]. The QCSE [4] as well as other coherent and incoherent processes in semiconductor MQW structures have been investigated for all-optical switching applications. For example, devices based on coherent many-body effects [5] and spin-polarized virtual excitons [6] have been shown to exhibit switching times on the order of 100 fs.

The operating wavelength of an optical switch is determined by the material composition used for the switching medium. Many of the AOSs demonstrated to date use GaAs films and MQW structures and operate in the infrared (IR) spectral region [4-6]. There is also demand for switches and modulators that operate in the ultraviolet (UV) spectral region. Such devices can be integrated into Blu-ray optics/electronics and non-line-of-sight (NLOS) optical communication systems. Switches based on GaN have been shown to provide efficient modulation in the ultraviolet (UV) spectral region [7]; however, the materials used can produce toxic side effects known for affecting lungs, reproductive systems, and kidneys [8]. Alternatively, ZnO is non-toxic, exhibits a UV band edge of 375 nm, and is lattice matched to GaN, so it can be directly integrated with GaN lasers and light-emitting diodes (LEDs). For these reasons, ZnO is a promising switching material. In fact, an optically controlled switch based on ZnO has been demonstrated [9]; however, little has been done in studying the optical nonlinearities necessary for producing ZnO-based AOSs.

In this work, we investigate the all-optical switching properties of a polycrystalline ZnO/ZnMgO heterostructure grown by DC sputter deposition. In this investigation, we performed differential-absorption measurements using 120-ps control and signal pulses tuned to the band edge of the ZnO thin film. We show how the absorption changes vary with polarization, control power, and the time delay between the control and signal pulses, and discuss how these measurements provide evidence for the underlying switching mechanism.

METHODS

In an AOS, the control and signal pulses are spatially overlapped in the switching material, as shown in Figure 1. In the absence of the control light, the signal pulse is largely absorbed by the material, representing the off state of the switch. When incident, the control pulse turns the switch on by changing the absorptive properties of the material, thus changing the amount of signal light passed by the switch. This switching action requires a nonlinear interaction between the signal light and the control light in the material. The strength of the nonlinearity dictates the contrast between on the on and off states, while the temporal dynamics of the nonlinearity determines the response time of the switch.

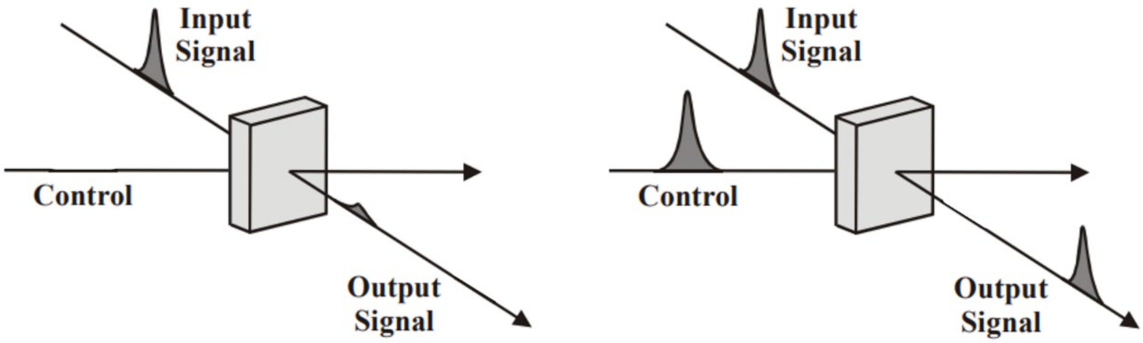


Figure 1. Basic geometry of an all-optical switch (AOS). (Left) Off-state: In the absence of the control pulse, the signal light is highly absorbed. (Right) On-state: The control pulse changes the absorption of the switching material so that the transmission of the signal light is increased.

Switching Material: ZnO Thin Films. In order to create a UV AOS, we need a material that exhibits strong optical nonlinearities in the UV spectral region. ZnO is a semiconductor material that is well suited for this purpose. Semicondutors are a classification of materials that derive their name from their conductive properties that fall between that of conductors and insulators. They are staples of the electronics industry due to the fact that their conductive properties can be easily controlled through external stimuli. For example, because their conductivity is highly affected by light, they are often used as optical detectors. Similarly, their absorptive properties are also affected by illumination, making them ideally suited for AOSs.

The basic absorptive properties of ZnO is shown in Figure 2 and can be understood in terms of its band structure. As illustrate in the figure, the absorption spectrum is characterized by an abrupt increase in absorption for wavelengths less than ~375 nm along with the presence of absorption resonances. In all semiconductor materials, the discrete energy levels in which electrons can reside, are closely grouped together to form bands separated by forbidden gaps. The most important bands of ZnO, as they relate to the optical nonlinearities of the material in the UV spectral region, are the valence band (VB) and the conduction band (CB). The VB is the highest energy band that is mostly filled with electrons, and the CB is the lowest energy band that is mostly void of electrons. In ZnO, these bands are separated by a bandgap energy of ~3.37 eV at room temperature. A photon with energy greater than 3.37 eV (i.e. wavelength less than 375 nm) is absorbed by the material when it excites an electron from the VB into the CB. This excited electron leaves behind a "hole" (an empty state in the VB). The bound electron-hole pair is known as an exciton and exhibits its own discrete energy resonances near the band edge called exciton resonances. The excition binding energy is particularly large in ZnO, in comparison to other semiconductor materials (e.g. GaAs), resulting in the pronounced absorption resonances apparent in its absorption spectrum. When the material is cooled, the band edge shifts to a lower wavelength, as shown in Figure 2.

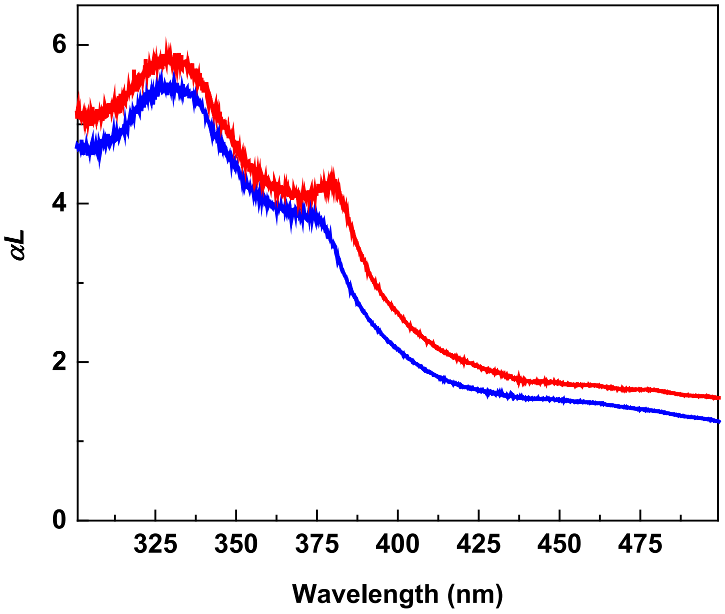
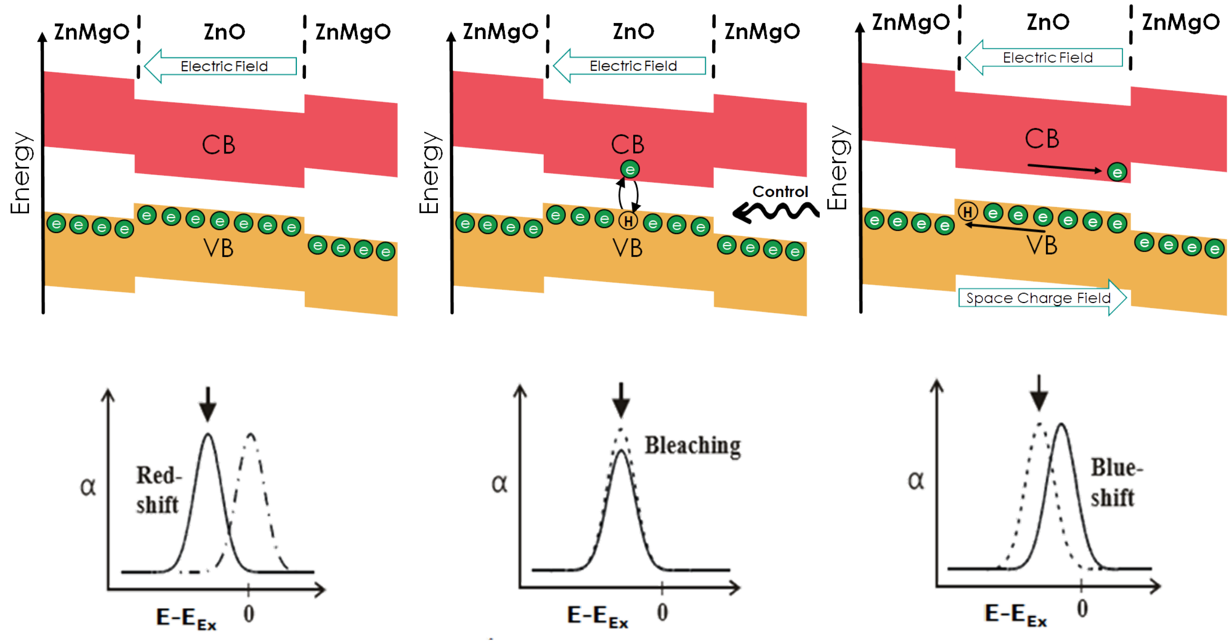


Figure 2. Absorption spectra of a120-nm-thck layer of ZnO (red curve) at room temperature and (blue curve) cooled to 78 K. Here, α is the absorption coefficient, and L is the layer thickness. The ZnO film is combined with other materials in the heterostructure described in Figure 4.

All-Optical Switching Mechanism: QCSE. Exciton resonances exhibit particularly strong optical nonlinearities, which can be exploited for all-optical switching. As shown in Figure 3, when a thin layer of ZnO is sandwiched between two layers of ZnMgO (which have a wider bandgap than ZnO), a quantum well is formed and the optical nonlinearities associated with the QCSE can be utilized for all-optical switching. Here, the control and signal pulses are tuned to the fundamental exciton resonance near the band edge and therefor are highly absorbed by the ZnO thin film. ZnO/ZnMgO quantum wells are known to possess a built-in electric field, which tilts the bands and causes the exciton resonance to be initially shifted to lower energy (known as a red shift), as shown in Figure 3(a). When incident, the control pulse is absorbed when it excites electron-hole pairs, filling some of the exciton states, as illustrated in Figure 3(b). This state filling in the ZnO reduces the number of available states (Pauli blocking) and therefore reduces the amount of signal light absorbed by bleaching the exciton resonance. The excitons are eventually pulled apart by the static electric field, creating a space charge field that opposes the built-in field, as shown in Figure 3(c). This results in a correction of the bands, shifting the exciton resonances to higher energies (a blue shift). Since the laser is tuned along the band edge, blue shifting the exciton resonance again results in a decrease in the amount of signal light absorbed. This effect persists until the electron-hole pairs recombine or are swept out of the material. Although recombination times of picoseconds-nanoseconds are common for many direct-gap semiconductor materials, the recombination time in our ZnO/ZnMgO heterostructure is expected to be extended due to the spatial separation of the electron-hole pairs.



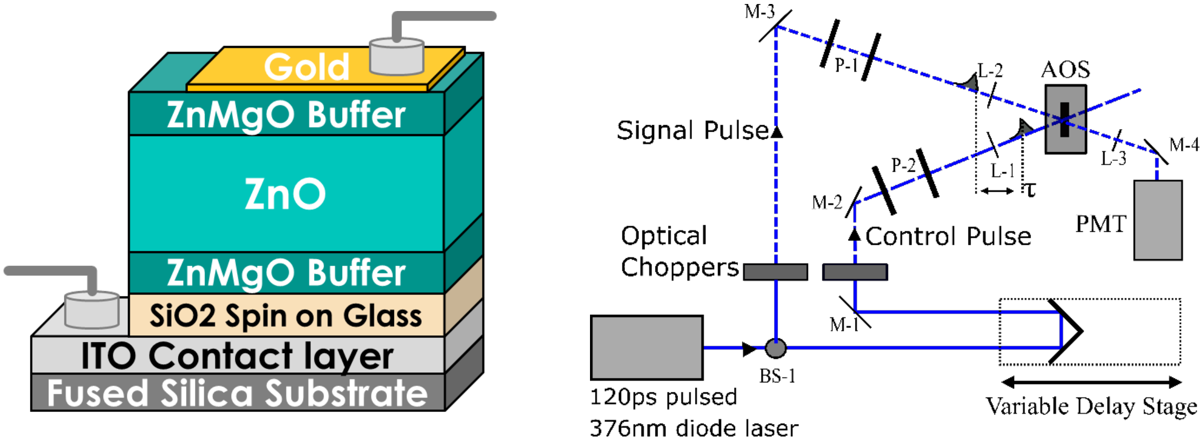
(a) (b) (c)

Figure 3. (a) The unexcited ZnO/ZnMgO single-well heterostructure, showing the bent energy bands and the redshifted exciton resonance (solid line) caused by the static electric field. The absorption without an electric field is represented by the dash-dotted curve centered at the exciton energy, Ex. (b) The control pulse excites electron-hole pairs. This initially bleaches the exciton resonance due to state filling (solid line). (c) The electrons and holes separate due to the static electric field producing a space charge field that screens the built-in field, thus blue shifting the resonance. The arrows over the absorption spectra indicate tuning of the control and signal.

EXPERIMENTAL RESULTS

Experiential Setup. The ZnO/ZnMgO single-well heterostructure that is the subject of this study is shown in Figure 4(a) and was grown, fabricated, and tested in house at the University of Wisconsin-La Crosse. The device is constructed of polycrystalline ZnO and ZnMgO thin films that are grown by DC sputter deposition on a commercially available ITO-coated fused silica substrate. The active ZnO layer is 120-nm thick and is bookended by two ZnMgO buffer layers. The ZnMgO layers function as barriers to keep the electron-hole pairs confined to the ZnO well and to buffer the lattice mismatch between the ZnO and the substrate materials. Before the ZnO and ZnMgO layers are grown, the ITO-coated fused silica substrate is coated with a layer of SiO_2 spin-on glass. The SiO_2 layer is added as a diffusion block to keep the indium from diffusing from the ITO layer into the ZnO-based layers during the grown process. We chose to study a single-well structure in order to simplify the carrier dynamics involved with the optical switching. ZnO exhibits a strong exciton binding energy, allowing us to use a single layer that is relatively thick by quantum-well standards. However, further studies aimed at device optimization will employ multiple layers that are considerably thinner than 120 nm. Since ZnO/ZnMgQ quantum wells exhibit a built-in electric field, no external applied field is needed; however, for the purposes of characterizing the electrical properties of the heterostructure, a semitransparent top layer of Au and electrical contacts are added to the device. Contact to the conductive layers is accomplished by using silver epoxy to attach leads so that voltage can be applied to the layers. The added Au layer is largely responsible for the background absorption observed in Figure 2.

The switching action is tested using the experimental apparatus shown in Figure 4(b). The light source used in the experiment is a UV pulsed diode laser that emits 120-ps pulses, with a center wavelength of 376 nm, at a repetition rate of 40MHz. Using a beam splitter (BS), the laser output is separated into two paths. The control path is 93% of the pulse intensity, whereas the signal path is 7%. This separation of power is done to create a large state filling effect due to the presence of a control pulses while minimizing the number of carriers excited by the signal pulses. The control path includes a retroreflector that is mounted on a movable stage so that its optical path length can be varied. This allows us to alter the time delay (τ) between the control and signal pulses. Here, positive τ refers to the control pulse reaching the heterostructure before the signal pulse, and negative τ refers to the control pulse reaching the structure after the signal pulse. To increase the carrier density excited by the control light, lenses are used to focus the control and signal pulses onto the heterostructure surface. The control pulses are focused to a spot diameter of 16 μm, while the signal pulses are focused to a slightly smaller spot diameter of 13 μm to ensure that they encounter a high density of carriers. Polarization optics are used to independently control the power and polarization of the control and signal light. The heterostructure is housed in a Joules-Thompson refrigerator, allowing the material to be cooled to temperatures ranging from 78 K to room temperature during the measurements.



(a) **(b)**

Figure 4. (a) Depiction of ZnO/ZnMgO single-well heterostructure. The layers are not to scale. The active layer for switching is a120-nm-thick layer of ZnO surrounded by two ZnMgO buffer layers. (b) The experimental setup for differential-absorption measurements consisting of mirrors (M), linear polarizers (P), Lenses (L), a beam splitter (BS), and a photo multiplier tube (PMT).

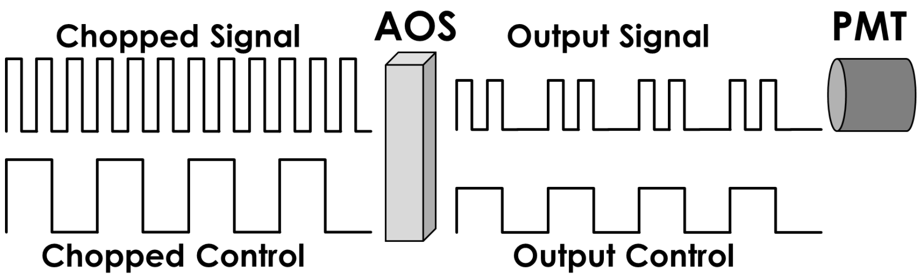


Figure 5. Schematic of the differential-absorption measurements. The signal and control are mechanically chopped at two distinct frequencies. Consequently, the change in the output signal caused by the control, Δ*S*, is modulated at both frequencies. These signal changes are passed by the cascaded lock-in amplifiers, while the scattered control output is blocked. Both chopping frequencies are low in comparison to the laser repetition rate (40 MHz) and thus each chop carries many laser pulses.

To determine the control's impact on the signal, we perform differential absorption measurements using a photo-multiplier tube (PMT) and a cascaded lock-in amplifier technique, shown schematically in Figure 5. Here, mechanical choppers are used to modulate the signal and control beams at two distinct frequencies and are used to reference two cascaded lock-in amplifiers. The first amplifier in the cascade is referenced to the higher chopping frequency of the signal and has a time constant small enough to pass the lower modulation frequency of the control. The second amplifier is referenced to the control frequency and can have a longer time constant in order to combat noise. Since only the control-induced changes in the signal light, ΔS, contains both frequencies, it is the only signal that is passed by the amplifier pair. By normalizing this value by the signal passed by the heterostructure in the absence of the control, S, the control-induced change in the aborption of the ZnO heterostructure [$-\Delta\alpha L = \ln(\Delta S/S)$] is determined. This technique alows very small changes in the absorption to be measured, while at the same time keeping scattered control light from contaminating the measurements.

Experiential Results. In Figure 6, $-\Delta\alpha L$ is plotted as a function of τ for co-linearly polarized control and signal pulses at two different samples temperatures. In both cases, the control induces a reduction in the amount of signal light absorbed by the ZnO. The fact that absorption changes are observed even when the control and signal pulses are not overlapped in time is consistent with the changes being due to control-induced carriers, as opposed to an interference effect between the control and signal fields. The largest absorption changes are observed at a slightly positive delay time (τ~200 ps), coinciding with when the carrier density excited by the control pulse reaches its maximum value. The absorption changes vary little with τ, indicating that the material is not recovering between pulses. The data indicate that the carrier lifetimes are much greater than the 25-ns time interval of the laser pulses, providing strong evidence that the electron-hole pairs are spatially separating in the heterostructure. Much larger changes in absorption are observed when the ZnO structure is cooled to 78 K than those observed when the structure is held at room temperature. At the cooler temperature, the laser pulses are tuned lower on the absorption band where the signal pulses are particularly sensitive to shifts in the band edge. This observation is again consistent with screening of the built-in field caused by spatially separated electron-hole pairs.

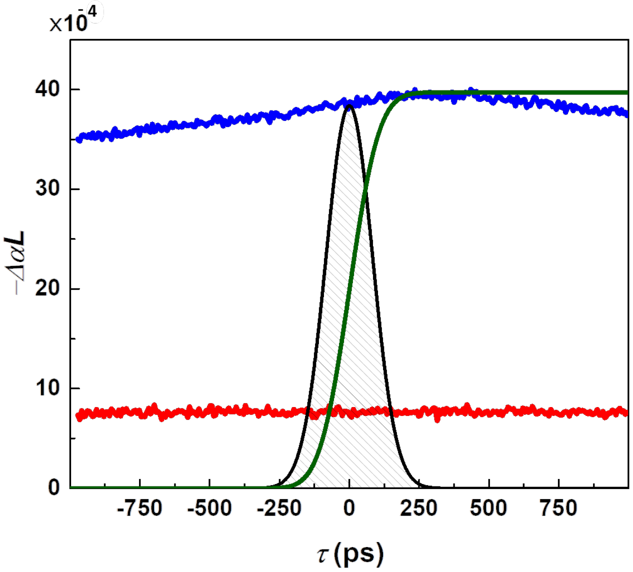
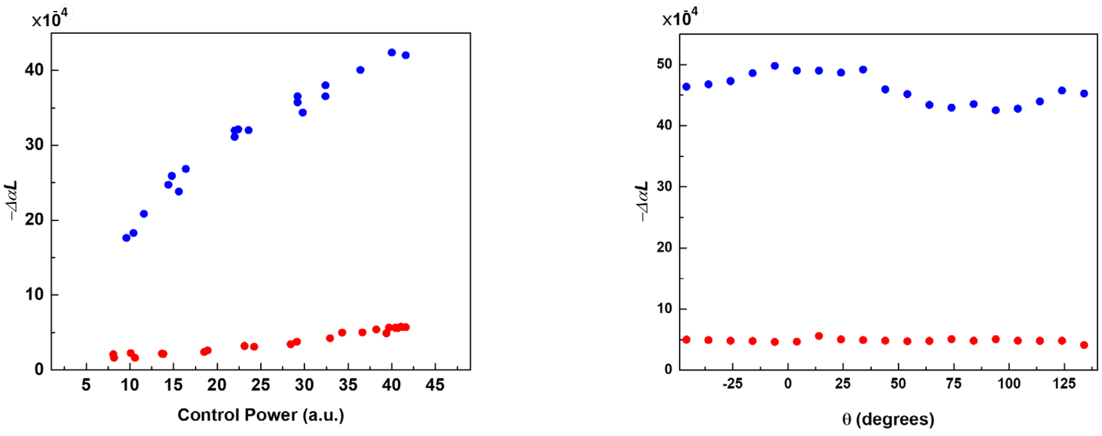


Figure 6. Absorption changes as a function of τ for co-linearly polarized control and signal pulses for an operating temperature of (red) 300 K and 78 K (blue). The black curve represents the intensity profile of the control pulse, and the green curve shows the integrated control pulse as it excites the sample.

In Figure 7, we show how $-\Delta\alpha L$ varies with control power and polarization. As shown in in Figure 7(a), the magnitude of the absorption changes increase with increasing control power. These changes are not perfectly linear with control power, but instead show signs of saturation. Figure 7(b) shows how the absorption changes depend on the polarization of the control pulses. Here, θ represents the azimuthal angle of the linearly polarized control pulses, where $\theta = 0^{\circ}$ represents control and signal pulses with the same linear polarization, and $\theta = 90^{\circ}$ represents control and signal pulse with orthogonal polarizations. There is very little dependence on the polarization state of the pulses, providing further evidence that the absorption changes are caused by a population of control-excited carriers.



(a) (b)

Figure 7. Absorption changes as a function of (a) control power and (b) azimuthal angle of the control polarization for an operating temperature of (red) 300 K and 78 K (blue).

CONCLUSIONS

Optically induced absorption changes of polycrystalline ZnO/ZnMgO heterostructure grown by DC sputter deposition have been demonstrated using 120-ps control and signal pulses. The presence of a control pulse is shown to alter the absorption properties of the material as seen by a weaker signal pulse. These effects persist long after the control pulse has exited the heterostructure, increase with increasing control power, and show little dependence on the azimuthal angle of the control field, consistent with the excitation of electron-hole pairs rather than interference effects. The absorption changes do not recover between laser pulses and enhanced changes are observed when the device is cooled so that the control and signal pulses are tuned closer to the band edge, consistent with the creation of a space charge field associated with spatially separated electrons and holes. Altering the absorption properties of ZnO using an optical control is a key element in creating AOSs that operate in the UV spectral region. Future work on the project will be aimed at increasing the contrast ratio of the device and decreasing the recovery time of the switch. Both of these objectives may be accomplished by constructing and testing ZnO/ZnMgO MQW devices with thinner well thickness.

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REFERENCES / LITERATURE CITED

[1] Miller, D. A. B., Chemla, D. S., Damen, T. C., Wood, T. H., Burrus, C. A., Gossard, A. C., and Wiegmann, W. (1984) "Band-edge electroabsorption in quantum well structures: The quantum-confined Stark effect," Phys. Rev. Lett. 53, 2173.

[2] Wood, T. H. (1988) "Multiple quantum well (MQW) waveguide modulators." *Journal of Lightwave Technology* 6, 743.

[3] Nishikawa, Y., Tackeuchi, A., Yamaguchi, M., Muto, S., and Wada, O. (1996) "Ultrafast all-optical spin polarization switch using quantum-well etalon." *IEEE Journal of Selected Topics in Quantum Electronics* 2, 661.

[4] Miller, D. A. B., Chemla, D. S., Damen, T. C., Gossard, A. C., Wiegmann, W., Wood, T. H., and Burrus, C. A. (1985) "Electric-field dependence of optical-absorption near the band-gap of quantum-well structures." *Physical Review B* 32, 1043.

[5] Gansen, E. J., Jarasiunas, K., Hawkins, S. A., Stevens, M. J., and Smirl, A. L. (2001) "Coherent all-optical polarization switching based on exciton-exciton interactions in quantum wells." *Journal of Applied Physics* 90, 1077.

[6] Gansen, E. J., Jarasiunas, K., and Smirl, A. L. (2002) "Femtosecond all-optical polarization switching based on the virtual excitation of spin-polarized excitons in quantum wells." *Applied Physics Letters* **80**, 971.

[7] Kao, C.-K. Bhattacharyya, A. Thomidis, C. Paiella, R. Moustakas, T. (2011). Electroabsorption Modulators Based on Bulk GaN films and GaN/AlGaN Multiple Quantum Wells. Journal of Applied Physics 109.

[8] Tanaka, A. (2004). Toxicity of indium arsenide, gallium arsenide, and aluminium gallium arsenide. Department of Hygiene, Graduate School of Medical Sciences, Kyushu University.

[9] Zhang, X. Y., Dhawan, A., Wellenius, P., Suresh, A., and Muth, J. F. (2007) "Planar ZnO ultraviolet modulator," Appl. Phys. Lett. 91, 071107.