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Properties of amorphous aluminate dielectrics synthesized via photosensitized pulsed laser ablation of luminescent targets

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Abstract

The dielectric properties of (Ce,Tb)$MgAl_{11}O_x$ films deposited by pulsed laser deposition are reported. The optical absorption from the rare earth dopant in the polycrystalline target material serves as an efficient photosensitizer for laser ablation when a laser source operating in the ultraviolet is employed. The deposited films are amorphous with few, if any, particulates evident on the surface. Metal–insulator–metal device structures were fabricated using these films, measuring the dielectric and leakage current properties. The relative dielectric constant of the amorphous aluminate is on the order of 10. Leakage currents as low as 6×10^{-8} A/cm^2 at an applied field of 0.6 MV/cm were realized.
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1. Introduction

In recent years, pulsed laser deposition has emerged as an enabling technique in oxide thin-film research. It has been implemented in materials and device research involving a number of different oxide material classes, including the deposition of oxides as gate dielectrics in wide bandgap device structures [1–5]. However, one area where pulsed laser deposition has limitations is in the formation of ultra-high bandgap insulating thin films. There is significant interest in the formation of large bandgap gate dielectrics for electric field-gated structures involving wide bandgap semiconducting channel materials, such as GaN and ZnO. For the electric field gating of wide bandgap semiconductors, the bandgap of the gate oxide material must necessarily be large as compared to the semiconductor so as to prevent the injection of charge carriers into the channel via transport through the gate oxide valence or conduction bands. As such, these gate dielectrics must have ultra-large bandgaps typically seen in silicates and aluminates. In addition, the gate oxide must be pinhole free and preferably amorphous so as to prevent leakage through grain boundaries. For the deposition of gate dielectrics using pulsed laser deposition, these ultra-wide bandgap dielectrics present a significant challenge. Pulsed laser deposition is dependent on an optical absorption process that yields ablation of the target material. For ultra-wide bandgap materials, band-edge absorption is minimal. Ablation is dependent on non-linear absorption processes (requiring high instantaneous power in a sub-ns pulsed laser source) or absorption via crystalline defects in the ablation target. For conventional pulsed laser deposition that employs an excimer laser source, this ablation process for ultra-wide bandgap dielectrics is quite inefficient and yields significant particulate densities that are detrimental to the performance of a gate dielectric. In addition, the energetics of the pulsed laser deposition process promotes crystallization of the deposition film which is often not desirable.

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In this paper, we describe an approach that circumvents this limitation. In particular, the deposition of a multi-cation $Ce_{0.33}Tb_{0.67}MgAl_{11}O_x$ amorphous films utilizing pulsed laser ablation of luminescent $Ce_{0.33}Tb_{0.67}MgAl_{11}O_{19}$ sintered targets is described. The optical transitions associated with the rare earth ions serve the role of photosensitizer for the optical absorption in the aluminate host yielding efficient ablation. One question to be addressed is whether the ablation dissociates the host sufficiently to minimize particulate formation. Another question is whether the film properties are adversely affects by the photosensitizer dopant. The dielectric properties of the $Ce_{0.33}Tb_{0.67}MgAl_{11}O_x$ amorphous films are discussed primarily within the context of dc dielectric constant, leakage current, and potential utility as a gate dielectric for wide bandgap oxide semiconductor devices.

2. Experimental details

The ablation targets were fabricated from commercial $Ce_{0.33}Tb_{0.67}MgAl_{11}O_{19}$ phosphor powders (Stanford Materials). Crystalline $Ce_{0.33}Tb_{0.67}MgAl_{11}O_{19}$ is an efficient green phosphor with a strong absorption at ~253.7 nm [6–10]. The powders were pressed into 1 in. targets and annealed in air at 1200 °C for 14 h, 1400 °C for 14 h, and at 1500 °C for 24 h. The targets displayed bright green luminescence when exposed to 254 nm ultraviolet light. The $Ce_{0.33}Tb_{0.67}MgAl_{11}O_x$ (CTMA) amorphous films were deposited in a conventional pulsed laser deposition system (base pressure 10^{-4} Pa) using a KrF 254 nm excimer laser as the ablation source. The strong absorption at 254 nm yields an efficient laser ablation process with virtually no particulate ejection from the target. Note that the use of a photosensitizer for achieving efficient ablation has been used in the deposition of polymeric and other organic materials [11]. For these soft materials, the introduction of the photosensitizer is needed to decouple the vaporization process from absorption by the fragile host polymer. In the case of wide bandgap inorganic dielectrics, the rare earth photosensitizer yields highly efficient absorption needed to achieve ablation of the host. In this work, the films for gate dielectric measurements were deposited at 100 °C in 0.1 Pa oxygen. For the gate dielectric measurements, metal–insulator–metal (MIM) diodes were fabricated using Al for the top electrode and indium–tin-oxide (ITO)-coated glass for the bottom electrodes. The thickness of the ITO was 100 nm. The deposited CTMA films had a thickness of 100 nm. Aluminum was deposited by magnetron sputtering at room temperature for the top electrode. The diameter of the diodes was 200 μm using metal shadow masks. Current–voltage (I–V) characteristics were measured over a temperature range of 273 to 573 K in air. The capacitance of the diodes was measured at 1 MHz.

3. Results and discussion

The $Ce_{0.33}Tb_{0.67}MgAl_{11}O_x$ films were optically transparent and amorphous. The films did not display luminescent when exposed to ultraviolet radiation which is the expected result for noncrystalline materials. X-ray diffraction of the films grown at various temperatures showed no evidence of crystallinity. Fig. 1 shows a field-emission scanning electron microscopy image of a $Ce_{0.33}Tb_{0.67}MgAl_{11}O_x$ amorphous film deposited on indium–tin-oxide-coated glass. Note that there is no evidence for particulates on the surface for this field of view. Surveys across other areas of the surface revealed almost no resolvable particulates within the resolution of the scanning electron microscope. The lateral structures seen in the image are related to the underlying grain structure of the ITO polycrystalline film.

Using the CTMA thin-film material, several multi-layer device structures have been investigated. Fig. 2 shows a cross-section scanning electron microscope image of a $Si/SiO_2/Ti/Pt/ZnO/Ce_{0.33}Tb_{0.67}MgAl_{11}O_x$ multi-layer stack in which the CTMA serves as the insulating dielectric for a metal–insulator–semiconductor diode. Note that the grain structure is clearly evident in the (Zn,Mg)O and Pt layers, but no grain structure is seen in the $Ce_{0.33}Tb_{0.67}MgAl_{11}O_x$ layer. Fig. 3 shows the I–V characteristics of Al/CTMA/ITO metal–insulator–metal (MIM) diode measured at 25 °C. The leakage current density for CTMA films was very low, measured at room temperature to be $\sim 10^{-6}$ A/cm^2 at an applied field of 5 MV/cm. From the capacitance measurements, the dielectric constant for the amorphous CTMA was determined to be ~10, which is similar to that of Al_2O_3. In order to explore the mechanism of leakage current in the CTMA films, temperature-dependent I–V measurements were performed. The I–V characteristics on MIM diodes were measured from 298 to 573 K are shown in Fig. 4. Note that log of the leakage current density is linear with respect to the square root of the applied electric field. This holds true for all temperatures, indicating that the

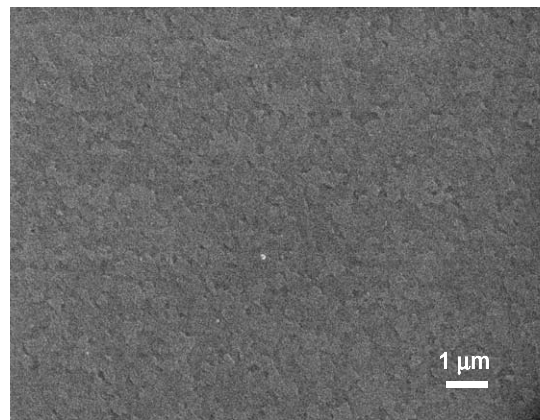

Fig. 1. SEM image of a $Ce_{0.33}Tb_{0.67}MgAl_{11}O_x$ film on ITO-coated glass.

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