ALD Applications
Room Baekeland - Session AA1-WeM2

ALD for Optical Applications

Moderators: Parag Banerjee, University of Central Florida, Matti Putkonen, University of Helsinki

10:45am **AA1-WeM2-1 Atomic Layer Deposition of Perovskite K(Ta_x,Nb_{1-x})O_3 films on Silicon for Integrated Photonics via KOtBu and H_2O**, ***Eric Martin,*** Ohio State University*; J. Bickford,* Army Research Laboratory*; H. Sønsteby,* University of Oslo, Norway*; R. Hoffman,* Army Research Laboratory*; R. Reano,* Ohio State University

Photonic integrated circuits (PICs) have advanced significantly over the last decade with a view towards applications in communications, sensing, and computing in the classical and quantum domains.The fabrication of silicon PICs in particular has transitioned from university cleanrooms to foundry supported process design kits by leveraging mature fabrication techniques from the microelectronics industry.The silicon material system exhibits optical properties that are enabling for compact PICs, such as high index contrast and low optical propagation loss at telecommunications wavelengths, allowing for the demonstration of low power nonlinear optical devices in waveguides based on third order susceptibility.An approach to advance the state-of-the-art performance of nonlinear optics in silicon PICs is to pursue heterogeneous integration. A promising material for this purpose is potassium tantalate niobate (KTN).KTN is a solid solution of $KTaO_3$ and $KNbO_3$ perovskites with a Curie temperature (T_c) that is defined by the Ta:Nb ratio.Bulk KTN crystals show record high Kerr nonlinearity when thermally tuned near T_c.KTN thin films can be grown with controllable Ta:Nb ratios via atomic layer deposition (ALD) using potassium tert-butoxide (KOtBu), tantalum ethoxide ($Ta(OEt)_5$), niobium ethoxide ($Nb(OEt)_5$) and H_2O precursors.To date, deposition of KTN films on silicon substrates has not been reported.In this work, we explore several approaches for obtaining perovskite $KTaO_3$ and KTN thin films on silicon using KOtBu, $Ta(OEt)_5$, $Nb(OEt)_5$, and H_2O precursors. Our approaches include varying the ALD process parameters for the metalorganic and H_2O co-precursors, chemically modifying the substrate surface, deposition of interfacial layers, and implementing ALD equipment modifications.We report the K uptake as a function of ALD process parameters using both refractive index obtained from spectroscopic ellipsometry and quantification from X-ray photoelectron spectroscopy (XPS).For a subset of samples, we report the crystal structure of our RTA annealed ALD films using X-ray diffraction (XRD). For the first time, we demonstrate perovskite $KTaO_3$ and KTN films on silicon by ALD.

11:00am **AA1-WeM2-2 Low-temperature ALD Sb_2Te_3 for Self-powered Broad-band Photodetector**, ***Jun Yang****, A. Bahrami, S. Mukherjee, S. He, S. Lehmann, K. Nielsch,* Institute for Metallic Materials, Leibniz Institute of Solid State and Materials Science Dresden, Germany

P-type Sb_2Te_3 thin film was deposited by atomic layer deposition (ALD) at 80 °C using $(Et_3Si)_2Te$ and $SbCl_3$ as precursors. The good crystal quality, low defect, and excellent uniformity of low-temperature ALD processed Sb_2Te_3 was supported by Raman, transmission electron microscopy, and XPS. Furthermore, high performance self-powered broad-band photodetectors based on Sb_2Te_3/Si heterostructure were fabricated. The photodetector has a wide detection range of 405 to 1550 nm. Meanwhile, a high responsivity of 4287 mA/W at 405 nm, and a quick response speed of 98 µs (t_{rise}) were obtained under 0 bias voltage, which could be ascribed to the strong built in electric field between p-type Sb_2Te_3 thin film and n-type Si. The temperature dependent performance of Sb_2Te_3/Si photodetector was thoroughly examined. Resistivity, conductivity, and carrier concentration of Sb_2Te_3 were carried out at temperature ranging from 273 K to 473 K to reveal intrinsic mechanism. The Sb_2Te_3/Si heterostructure self-powered photodetector with excellent performance based on ALD process shows the great potential application in optoelectronic devices.

11:15am **AA1-WeM2-3 Preparation of High Mobility Indium Hydroxide Doped by Atomic Layer Deposition and Study on Photoelectric Properties**, ***Liangge Xu,*** Harbin Institute of Technology, China

At present, the industrialization of metal mesh and tin doped indium oxide (ITO) film cannot meet the high transmittance requirements of infrared band. Therefore, how to achieve high carrier mobility has become a hot topic. In order to solve the above problems, In_2O_3:H film is selected, which is a special transparent conductive oxide film. On the one hand, hydrogen has a very small volume and mass, and will not produce impurity scattering in the process of electron transport; On the other hand, hydrogen is distributed in the gap of the indium oxide lattice, resulting in almost no lattice distortion, so it does not increase the number of grain boundaries and reduce the grain boundary scattering. The combined effect of these two factors makes the hydrogen doped indium oxide (In_2O_3:H) thin films have high carrier mobility, which can achieve high infrared transmittance by reducing the carrier concentration on the basis of ensuring the electrical properties.

In this paper, the growth rate, microstructure and photoelectric properties of In2O3:H films were investigated by changing the deposition process parameters. When the deposition temperature is 100 ℃, incomplete reaction occurs during deposition, and the growth rate is slow. The film is amorphous. At this time, the carrier mobility is low, which is only 30.18 cm^2/Vs, and the optical properties are good, the transmittance can reach 80% at 4 µm; With the increase of deposition temperature, the growth rate gradually stabilized at 0.1nm/cycle, the amorphous state gradually changed to crystalline state, the carrier mobility increased sharply, and the highest was 64.05 cm^2/Vs, and the carrier concentration and the optical properties decreased. The ratio of deposition time of InCp / (O_2 + H_2O) and the reaction time of InCp have little effect on the electrical and optical properties. With the increase of deposition period, the carrier mobility can reach up to 80 cm^2/Vs, but the optical performance is very poor, the transmission rate at 4 µm is only 39.5%.

11:30am **AA1-WeM2-4 ALD MgF2 Using (EtCp) $_2$Mg and SF_6 Remote Plasma Source**, ***Hoon Kim****, . Huang, . Allen, E. Pierce, , J. Wang,* Corning Inc.

Magnesium fluoride (MgF2) is a key material for Deep Ultra-Violet (DUV) and Far UV optics due to its wide bandgap and high resistance of oxidation. ALD process enables conformal coatings and precise thickness control on 3D shape optics. ALD MgF2 has been demonstrated using anhydrous hydrogen fluoride (HF) as fluorine source. However high concentration HF has safety concern in handling and residue in the reactor. Thus, safer fluorine source should be considered for industrial use. Hhfac has also been evaluated, but it requires O3 to activate the fluorine which results in oxygen and carbon incorporation in the film. The impurities are an absorption source for the UV range. SF6 is evaluated as fluorine source for AlF3, but no report for MgF2. In this study, we demonstrated the MgF2 ALD using SF6 remote plasma. The growth rate, impurity level and conformality were evaluated. Optical properties such as dispersion and transmittance were obtained. Hot wall batch reactor with remote plasma source was employed in this study. Fluorine radical was generated by using SF6 flow through Inducted coupled plasma (ICP) source. (EtCp) 2Mg was used as precursor. A prism shape surrogate was used to evaluate coverage on 3D shape. Thickness and reflectance were measured by an ellipsometer. Impurity of the film was measured by XPS and SIMS. Direct reduction of the precursor using SF6 plasma caused high carbon incorporation in the film due to the reaction of Cp ligand and fluorine. XPS results showed that CFx bonds are detected in the film. To address this issue, reducing agent to remove the Cp ligand from adsorbed precursor was introduced before SF6 exposure. This approach effectively removes the carbon incorporation and forms high purity MgF2 film. This ALD film has lower impurity level than that of in-house PVD MgF2. The growth rate of ALD MgF2 is 0.4Å/cycle at 150oC. The refractive index at 550nm is 1.33 which is same as inhouse PVD MgF2. The deposited film is polycrystalline. Other optical properties will be reported.

11:45am **AA1-WeM2-5 Moisture Sensitivity of ALD Metal Fluorides for Far UV Optical Coatings**, ***Robin Rodríguez****, J. Hennessy, A. Jewell, S. Nikzad,* Jet Propulsion Laboratory (NASA/JPL)

We report on the use of atomic layer deposition (ALD) for the development of metal fluoride thin films relevant to optical coatings operating at far ultraviolet wavelengths (FUV, 90-200 nm). The use of metal fluoride materials like MgF_2, AlF_3, LiF, CaF_2, and LaF_3 is relevant to all FUV optical systems for applications like anti-reflection coatings, dichroic beam splitters, bandpass filters, and reflective mirror coatings. However, many metal fluorides have associated concerns with environmental stability, primarily related to water vapor exposure. For example, LiF-protected aluminum mirror coatings can experience reflectance-losses if stored in modest relative humidity environments (RH ~40%). Initial space-based demonstrations of ALD coatings have shown improved stability by depositing a thin 1–2 nm capping layer of MgF_2 or AlF_3 onto the LiF-protected mirrors. In this work, we have conducted a broader study of the moisture sensitivity of ALD materials. This includes physical vapor deposition (PVD) of AlF_3 mirror coatings capped with ALD MgF_2 layers on Si, as well as stand-alone ALD coatings of MgF_2, AlF_3, CaF_2, and LiF on Si subjected to both long-duration and accelerated aging tests and then characterized by FUV reflectance measurements, spectroscopic ellipsometry, atomic force microscopy, x-ray photoelectron spectroscopy, x-ray diffraction, and secondary-ion mass spectrometry. To the best of our knowledge, this work also reports the first instance of CaF_2 and LaF_3 coatings deposited by ALD using anhydrous HF.

The observed changes in optical properties, surface morphology, and film composition can provide guidelines on storage conditions for these materials for future space instrumentation, and serves as a baseline for the direct comparison of material produced by PVD versus ALD.Finally, we also describe a new test reactor used in the work above that has enabled the deposition of PVD aluminum mirror coatings and ALD metal fluorides within the same vacuum chamber, to prevent oxidation of the aluminum surface and minimizing the time between metal evaporation and ALD encapsulation.

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