Hard Coatings and Vapor Deposition Technologies
Room Town & Country D - Session B8-2-ThM

HiPIMS, Pulsed Plasmas and Energetic Deposition II

Moderators: Prof. Jon Tomas Gudmundsson, University of Iceland, **Dr. Tiberiu Minea**, Université Paris-Saclay, France

9:00am **B8-2-ThM-4 Hipims Deposition of Ultrathick Au-Ta Coatings: Effects of Deposition Rate and Substrate Tilt**, ***Leonardus Bimo Bayu Aji***, *E. Kim, J. Merlo, S. Shin, G. Taylor, L. Sohngen, A. Engwall, A. Baker, D. Strozzi, B. Bocklund, E. Moore, A. Perron, S. Kucheyev,* Lawrence Livermore National Laboratory, USA

Targets for indirect-drive inertial confinement fusion require hohlraums. These are centimeter-scale sphero-cylindrical heavy-metal canswith wall thicknesses of ~10 microns or larger. The fabrication of hohlraums by physical vapor deposition is challenging as it involves ultrathick coatings on non-planar substrates in the oblique angle deposition regime. Here, we study Au-Ta alloy films deposited on rotating non-planar substrates by high power impulse magnetron sputtering (HiPIMS) at different deposition rates and substrate tilt geometries. We use mass-resolved ion energy spectrometry and optical emission spectroscopy to monitor plasma discharge characteristics. Results show that the deposition rate can be effectively used to control the major film properties relevant to ICF applications, including residual stress and electrical resistivity.

This work was performed under the auspices of the U.S. DOE by LLNL under Contract DE-AC52-07NA27344.

9:20am **B8-2-ThM-5 Self-Sputtering Identification in Helium HiPIMS Discharge with Molybdenum Target**, ***Abderzak el-Farsy***, *E. Mmorel,* Laboratoire de Physique des Gaz et des Plasmas, France; *Y. Yoann Rozier,* SuperGrid Institute, Villeurbanne, France; *T. Minea,* Laboratoire de Physique des Gaz et des Plasmas, France

Magnetron sputtering is a worldwide used process for thin film deposition covering a large panel of applications. In the last two decades, High Power Impulse Magnetron Sputtering (HiPIMS) has been developed and intensively studied. It is an ionized vapor deposition technique exploiting the high plasma density generated during a short time when a huge power (> 10^7 W/m^2) boosts the plasma.

In this contribution, we analyzed the current-voltage waveforms during HiPIMS discharge period working with helium as background gas and a 1-inch molybdenum target. The gas pressure was fixed at 5 Pa, and the pulse duration was 90 ms with a repetition frequency of 50 Hz. From the current waveform of the discharge, two phases were identified: (i) a current peak (up to 90 A) reached at the pulse start (t<30 µs) followed by (ii) a decay transition between 30 to 90 µs towards a steady-like state (I ~ 35 A).

To highlight the physical process behind this behavior, a study was carried out by increasing the applied voltage of the target until 1000 V. Optical emission spectroscopy (OES) qualitatively tracks the neutral and ionized species of He and Mo present in the ionization region. In addition, the plasma electron density can be evaluated from the line profiles of $H_\alpha$and H_β emissions *via* theStark effect by high-resolution OES. Combining all this information, it comes out that the self-sputtering process sustains the discharge in the second part of the pulse (30-90 µs) when the high voltage is applied, while the gas itself (He) plays the dominant role in the beginning, being responsible for the peak current.

9:40am **B8-2-ThM-6 On Working Gas Rarefaction in High Power Impulse Magnetron Sputtering**, ***Kateryna Barynova***, *S. Suresh Babu,* University of Iceland; *M. Rudolph,* Leibniz Institute of Surface Engineering (IOM), Germany; *J. Gudmundsson,* University of Iceland

The ionization region model (IRM) is applied to explore the working gas rarefaction in high power impulse magnetron sputtering discharges operated with graphite, aluminum, copper, titanium and tungsten targets. The various contributions to working gas rarefaction including electron impact ionization, kick-out by the sputtered species, and diffusion, are evaluated and compared for the different target materials, over a range of discharge current densities. For all cases the working gas rarefaction is found to be significant, and to be caused by several processes, and that their relative importance varies between different target materials. In the case of a graphite target, electron impact ionization is the dominating contributor to the working gas rarefaction, with 55 - 64 % contribution, while kick-out, or sputter wind, has negligible influence, whereas in the case of tungsten target, kick-out dominates, with 39 - 48 % contribution. The relative role of kick-out by the sputtered species increases and the relative role of electron impact ionization decreases with increased mass of the target atoms. The main factor influencing how much each process contributes to working gas rareafaction is the mass of the target species, while it is not clear how the ionization potential and the cohesive energy, that determines the most probable velocity with which the sputtered particles leave the target, influence the relative contribution of the various terms.

10:00am **B8-2-ThM-7 Spokes in HiPIMS: Help or Hindrance?**, ***Julian Held***, University of Minnesota, USA; *P. Maaß, M. George, W. Breilmann, S. Thiemann-Monjé, V. Schulz-von der Gathen, A. von Keudell,* Ruhr University Bochum, Germany **INVITED**

Spokes are long wavelength oscillations observed in the magnetized region of high power impulse magnetron sputtering (HiPIMS), as well as other ExB discharges. Initially, spokes in HiPIMS were observed as regions of intense light emission, moving along the E x B direction of the discharge, just above the cathode surface. By now, it has become clear that these bright structures are accompanied by intense fluctuations in electron density, temperature, and plasma potential. These variations in plasma potential might lead to electron and ion heating, as well as increased transport of charged species across the magnetic field lines. As such, spokes have long been considered a possible pathway in increasing the deposition rate of HiPIMS discharges.
In this contribution, we will review our current understanding of the physical mechanisms creating and sustaining the spokes. Based on this, we will try to answer whether spokes have a positive influence on either the deposition rate or the energy of ions and whether or not they can be manipulated to improve these advantages.

10:40am **B8-2-ThM-9 Effect of Plasma Nitriding Pretreatment on the Mechanical and Wear Properties of Tungsten Carbide Substrate, and AlCrN Coating Deposited by High-Power Impulse Magnetron Sputtering**, *F. Yang,* Department of Mechanical Engineering, National Taiwan University of Science and Technology, and Center for Plasma and Thin Film Technologies, Ming Chi University of Technology, Taiwan; *T. Liu,* ***Guan-Lun Shen***, *I. Chen,* Department of Materials Engineering, Ming Chi University of Technology, Taiwan; *Y. Kuo,* Department of Mechanical Engineering, National Taiwan University of Science and Technology, Taiwan; *C. Chang,* Department of Materials Engineering, Ming Chi University of Technology, and Center for Plasma and Thin Film Technologies, Ming Chi University of Technology, Taiwan

This paper examines the mechanical and wear properties of Tungsten Carbide substrates subjected to plasma-nitriding (PN) before, and after being coated with AlCrN by high-power impulse magnetron sputtering (HiPIMS). Low-temperature (∼300 °C) plasma nitriding treatment was applied for various durations (0 hr, 0.5 hr, 1 hr, and 1.5 hr) with the aim of maximizing the adhesion strength and wear resistance by improved the mechanical properties of the tungsten carbide substrate for after AlCrN coating. XPS and TEM revealed the effect of plasma nitriding on the tungsten carbide substrate, the diffusion of nitrogen into the tungsten carbide substrates to form new nitrides as such W-N and C-N bonding. Result in the hardness is enhanced from 1534 to 2034 Hv. After that, the AlCrN deposited on nitride tungsten carbide substrate by HiPIMS process. The measurement results indicate that the adhesion strength was improved from 70 to above 150 N, and the hardness was enhanced from 2257 to 2568 Hv with increasing plasma nitriding durations. Due to substrate hardening effect led to the wear rate can be decreased from 14.5 to 3.4 (10^{-8} mm^3/Nm). Therefore, the AlCrN coating deposited on tungsten carbide substrate with plasma nitriding pretreatment is proved can enhance the mechanical and wear properties of the AlCrN thin film.

Keywords: plasma nitriding, high-power impulse magnetron sputtering, AlCrN coating

11:00am **B8-2-ThM-10 Highly Ionized Pulse Sputtering of Seed Layers for Through Silicon Vias**, ***Juergen Weichart***, Evatec AG, Switzerland

Directional deposition based on HIPIMS has been developed for barrier and copper seed layers in Through Silicon Vias (TSV) with very high aspect ratios up to 20:1. Due to demanding particle and uniformity requirements stationary sputtering with a large target diameter up to 450mm is needed. To avoid redeposition rotating magnets must be designed carefully, which is even more important due to the return effect of metal ions to the target in HIPIMS discharges. A concept of adjustable magnets has been developed to enable a stable peak current with progressive target erosion. High frequency synchronized bias is applied on the pedestal for insulating substrates with an appropriate matching for the highly dynamic impedance conditions at low duty cycles. The challenges to achieve good uniformities on 200mm or on 300mm substrates for the deposition rate, the deposition profile in the vias, and the specific resistivity are demonstrated. With increasing ion impact the average grain size increases from 30 to 70nm, accompanied by an increase of the (002) over the (111) orientation, as measured by Ө-2Ө measurements. Typically, in deep Silicon etched vertical vias of 5µm diameter and 25µm depth, hence with an aspect ratio (AR) of 5:1, the minimal side wall coverage is 4% for Ti or Ta and 2% for Cu, while in vias of 10µm diameter and 100µm depth (AR 10:1) the minimal side wall coverage is 2% for Ti or Ta and 1% for Cu. The difference between Cu and Ti or Ta is explained by the high sputter yield of Cu, which reduces the effective ionization degree for Cu in the plasma. For the subsequent enforcement by well-developed electroplating processes of Cu (ECP) for heterogeneous integration it has been found that 20nm of the seed layer is sufficient.

11:20am **B8-2-ThM-11 Deposition Environment and Microstructure of Transition Metal Nitride Thin Films Deposited at CMOS-Compatible Temperatures for Tunable Optoelectronic and Plasmonic Devices***, **Arutiun P. Ehiasarian,*** Sheffield Hallam University, UK*; R. Bower,* Imperial College London, UK*; D. Loch,* Sheffield Hallam University, UK*; A. Berenov, B. Zou,* Imperial College London, UK*; P. Hovsepian,* Sheffield Hallam University, UK*; P. Petrov,* Imperial College London, UK

Plasmonic metamaterials based on transition metal nitrides have received significant interest to enable environmentally stable and tunable optoelectronic devices.However, the deposition conditions for achieving low optical losses and synthesis temperatures which are compatible with semiconductor processing are unclear. We have deposited binary, ternary and layered transition metal nitride thin films based on titanium and niobium nitride using constant-current High Power Impulse Magnetron Sputtering (HIPIMS) without substrate biasing.Enhanced plasma density, dissociation of nitrogen molecules and metal-to-nitrogen ratio are investigated using optical emission spectroscopy and energy-resolved mass spectroscopy as factors influencing the microstructure and optical properties of the materials.N^+: N_2^+ ratios exceeding 1 were achieved at high peak power density in a stable deposition process due to constant current control mode of operation.The temporal evolution of the discharge operating in a constant current regime is discussed.Epitaxial films on MgO substrates and polycrystalline films on glass and steel both showed low optical losses at a deposition temperature of less than 300°C.A random texture was observed at film thicknesses of 50 nm with Nb-containing films exhibiting an enhanced (200) texture attributed to bombardment by a heavier ion.Continuous well-defined layers with thickness of 5 nm were obtained as observed by TEM.Tunability of the plasma frequency in the ultraviolet to visible spectral ranges was achieved through the Ti:Nb ratio.The dense grain boundaries obtained in the HIPMS deposition environment may contribute to efficient plasmon dispersion in the material.The thin film quality combined with the scalability of the deposition process indicates that HIPIMS can pave the way towards the industrial fabrication of next generation plasmonic devices.

11:40am **B8-2-ThM-12 On the Connection between the Self-Sputter Yield and Deposition Rate in High Power Impulse Magnetron Sputtering Operation***, **Jon Tomas Gudmundsson,*** University of Iceland*; M. Rudolph,* Leibniz Institute of Surface Engineering (IOM), Germany*; K. Barynova,* University of Iceland*; J. Fischer,* Linköping University, Sweden*; S. Suresh Babu,* University of Iceland*; N. Brenning, M. Raadu,* KTH Royal Institute of Technology, Sweden*; D. Lundin,* Linköping University, Sweden*; H. Hajihoseini,* University of Twente, Netherlands

The magnetron sputtering discharge is a plasma discharge-driven physical vapor deposition technique, that is utilized in a range of industries. When the magnetron sputtering discharge is driven by high power unipolar pulses of low repetition frequency, and low duty cycle, it is referred to as high power impulse magnetron sputtering (HiPIMS). HiPIMS operation results in increased ionization of the sputtered species and lower deposition rate than the dc magnetron sputtering discharge, when operated at the same average power. The HiPIMS discharge can contain a large fraction of ionized sputtered material. This means that, at least some fraction, often a significant fraction, of the ions involved in the sputter process are ions of the target material. This also implies that a large fraction of the ions of the sputtered species can be attracted back to the target and is not deposited on the substrate to form a film or coating. Self-sputtering and the self-sputter yield are therefore expected to play a significant role in HiPIMS operation, and have a decisive impact on the film deposition rate, at least for metal targets. We have applied the ionization region model (IRM) [1] to model HiPIMS discharges in argon with a number of different targets [2,3], to study various processes, such as working gas rarefaction and refill processes, the electron heating mechanisms, ionization probability and back-attraction of the sputtered species, and recycling mechanisms. It will be discussed how these processes depend on the mass and ionization potential of the target atom, the discharge current density, and self-sputter yield of the target.

[1] Huo *et al.,* Journal of Physics D: Applied Physics **50**, 354003 (2017)

[2] Gudmundsson *et al.*, Surface and Coatings Technology **442**, 128189 (2022).

[3] Babu *et al.,* Plasma Sources Science and Technology **31**, 065009 (2022)

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