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Exploring Nanostructured Complex Oxides Produced via Cluster-Assembling for Enhanced Dielectrics Applications

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Extended Abstract

Nanomaterials showing enhanced dielectric properties are of great interest for the next generation of transparent capacitive and electronic device applications. Zinc oxide (ZnO), with its wide bandgap (3.37 eV) and high-binding energy exciton (60 meV) is a promising multifunctional semiconductor material for several optical and electrical applications due to its physical and chemical properties [1]. In our study, we focus on the possible synthesis of complex nanostructured ZnO-based oxides with temperature and frequency independent colossal dielectric constants by employing Supersonic Cluster Beam Deposition (SCBD) [2] and subsequent doping. Doping with gadolinium and iron was performed to modify the reorganization of ZnO nanostructured material after thermal treatments and assess their effect on the dielectric properties.

SCBD is a Physical Vapor Deposition technique that is used to fabricate nanostructures after gas-phase synthesis of nanoparticles with control over their size, structure, and physicochemical properties [2]. ZnO thin films were fabricated on silicon substrates via SCBD and then doped by soaking the as-deposited films in highly diluted $GdCl_3$ and $FeCl_3$ isopropanol solutions. A time-controlled doping method enabled the incorporation of Gd and Fe ions at variable levels. The films were then annealed at 400–650°C to study temperature effects on morphology and structure.

X-Ray Photon Spectroscopy (XPS) was used to study the surface chemical composition, effectiveness of doping, and oxidation states of ZnO nanoparticles. The XPS spectra at the corresponding peaks of Zn *2p*, O *1s*, Gd *3d*, and Fe *2p*, photoemissions are a confirmation of the presence of doping elements on the surface of the film, and we have not observed peaks corresponding to impurity of any kind: we thus consider it as a sufficiently high-purity doping process.

Raman spectroscopy was used to study the vibrational and structural characteristics of the prepared samples. Raman features include the LO phonon mode and a weak band near 275 cm^{-1}, which may arise from the activation of a normally Raman-inactive (silent) mode and are in accord with the ultrafine character of the deposited nanoparticles [3-4]. Distinct Raman peaks attributed to the presence of Gd or Fe were not observed in the Raman spectra, in accord with their incorporation as dopants at low concentration. The search for a signature of their presence and effect on the ZnO local bonding environment to be observed as spectrum alterations is underway but limited by the need for an extremely accurate background subtraction.

Surface morphology and particle shape characterization were conducted by using scanning electron microscopy (SEM) and atomic force microscopy (AFM). SEM images were collected at a magnification of 150,000x resolution and 100 nm. SEM images show a complex morphology of the film, featuring micro-cracks, porosity, and agglomerated clusters. AFM provided high-resolution information on surface topography as a

function of annealing temperature. When the annealing temperature is raised to 650°C, we observe grain size increase, and the larger particles start showing some polyhedral facets as a signature of crystallization.

This study demonstrates the potential of our method, which uses SCBD to deposit a low-density amorphous phase with an ultrafine nanostructure followed by time-controlled doping via exposure to a diluted solution and an eventual thermal annealing, to tune the nanoscale-enhanced features of ZnO nanoparticles.

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