Nanoscale Manufacturing - Nonlinear Nanocomposites for Magnetostrictive Actuators and Photonic Devices

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1. Introduction: Our goal is to develop manufacturing techniques for nano-composite and nano-heterogeneous materials and devices that combine the functional advantages obtained from the "size-tunable" properties of nanocomposite materials with the fabrication and direct-write advantages available from nanoparticles (NP) manufactured and delivered in aerosol form. We focus on developing manufacturing techniques utilizing NP generated by Laser Ablation of Microparticles (LAM) from an aerosol source. Two important application areas that benefit from compositional variations on the nanoscale: a) nanostructured giant magnetostrictive films with high magnetization that can be driven at low fields due to a spin reorientation transition, and b) nonlinear photonic devices that have increased optical conversion efficiency due to both the enhancement of the nonlinear effects by nano-heterogeneity and the ability to phase match the interacting optical waves by coherently mixing active and inactive regions in a nano-composite optical media. Both applications require three-dimensional nanocomposite structures. These nanocomposite structures are difficult or impossible to produce in size scales that are practical for many devices using existing manufacturing technologies.

2. The LAM Process: We have been granted a patent for a method of producing NP by LAM.[2] In the LAM apparatus, a high-energy excimer laser pulse (λ= 248 nm, t_p = 12 ns) illuminates an aerosol of microparticles (1 to 20 μm dia.), initiating breakdown and shock wave formation at each microparticle. Nucleation of NP follows in the low-pressure region behind the shock. This traveling wave process is energetically efficient and has high mass-conversion efficiency.[3] We have demonstrated that it is possible to directly write films, micron-scale lines, and other patterned structures onto substrates using supersonic jet deposition of NP of a wide range of inorganic materials. This favors its use as the aerosol source for materials used in supersonic-jet deposition. For direct writing, the LAM process is followed by a virtual impactor to eliminate larger particles so that a uniform, NP aerosol is sent to the impaction chamber as shown in Fig. 1. The NP aerosol (at ~1 atm.) is fed through a nozzle into the impaction chamber that is under vacuum. The pressure differential between the chambers accelerates the

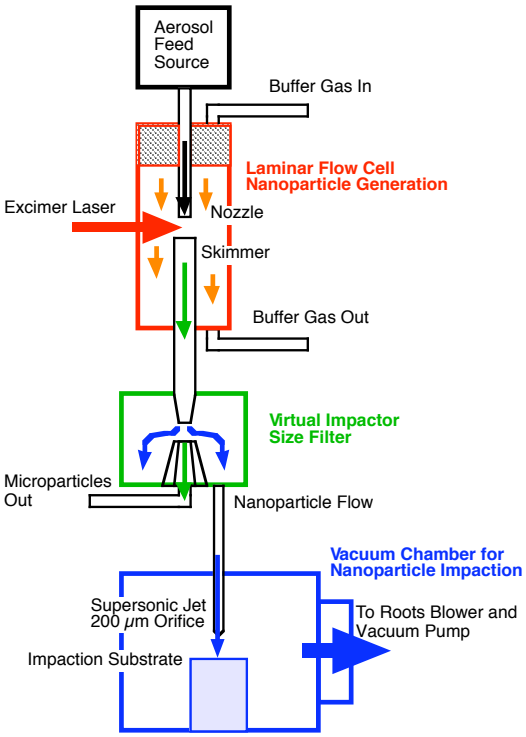

Figure 1. A continuous-flow LAM reactor followed by a virtual impactor that removes unablated microparticles. The formed NP enter a supersonic jet for impaction onto a substrate placed on an x-y table.

NP up to ~800 m/s (in He) before they impact onto a substrate. At these velocities, the NP have sufficient kinetic energy to bond to the substrate. Moving the substrate using a computer-controlled x-y table within the NP impaction chamber produces patterned structures. The NP impaction chamber is connected through a load lock to a thin-film deposition chamber that has both ion-beam sputtering and pulsed laser ablation (PLD) sources. NP films are routinely transferred to this chamber to be coated by PLD with dielectric materials or protective layers.

3. Nonlinear Photonic Nanoparticle Materials: In order to produce optical materials based on the enhanced nonlinear properties of NP, suitable NP and host materials systems need to be identified and developed. For example, a semiconductor material such as GaN might be used to produce NP that are to be embedded in AlN, a material with a larger bandgap that prevents unwanted energy transfer away from the NP. A study was undertaken to produce AlN host films that were either single crystal, or highly oriented polycrystals with minimum optical absorption. Other systems chosen for study are silver NP in SiO_2 or Al_2O_3, and Si NP in SiO_2. Here, we will report results from the GaN – AlN system.

To measure the optical nonlinearity of the GaN NP, tunable Ti:sapphire laser excitation at 800 nm (100 fs pulses) was used to measure the third harmonic generation (THG). The GaN NP samples were made with the LAM process using He buffer gas and GaN microparticle feedstock. A line of NP was written ~1 mm wide and 3 μm thick onto a sapphire substrate. The line's thickness versus transverse position is shown in the profilometer scan in Fig. 2. The third harmonic generation signal was measured using a ~1 μm laser spot size as a function of position across the same sample. Those data are also shown in Fig. 2. In this case, the porous NP film is sufficiently thick in the center to show coherent dephasing or phase mismatch as the film approaches a thickness equal to two coherence lengths. Thinner films do not show this dip, and thicker films would show the return of a peak at the center. This is not due to optical absorption.

The third harmonic generation data were used to estimate the third-order susceptibility of the GaN nano-porous material. The value obtained was equal to that of bulk GaN. We estimate that the mass density of the film is 50%, so that the enhancement due to the nanostructured material is a factor of 2. In scanning the laser wavelength ±50 nm, no variation in third harmonic signal was observed. This indicates that neither one photon, two photons, nor three photons were energy resonant or close to energy resonance with energy states in the GaN NP. Though we have not as yet observed a significant enhancement in third order effects, the NP here are not surrounded by a glass of differing refractive index that causes an enhancement of the field within the NP. We are now developing nanocomposite materials of dielectric/NP mixtures.

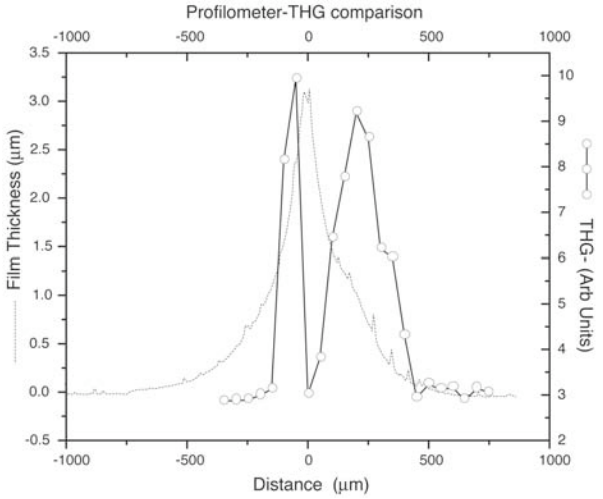

Figure 2. GaN NP film thickness profile and optical third harmonic generation signal as a function of transverse position across the film.

4. Encapsulating Materials: A necessary companion material is needed for every optical NP composite. For the case of GaN NP, we have selected AlN as the larger bandgap host material. The literature is inconclusive and incomplete in describing AlN of film quality as a function of deposition conditions. In particular, the effects of laser fluence and background nitrogen gas pressure are contradictory. To resolve these discrepancies, we are conducting a thorough investigation of film quality as measured by optical properties (complex refractive index corrected for wedged films), XRD analysis including rocking curves to ascertain the grain structure and orientation, EDS, SEM, and profilometry. We have observed that at higher background nitrogen pressure (10^{-3} to 10^{-4} T), low optical absorption films were produced for a wide range of substrate temperatures (100 to 800°C) even though the crystallographic orientation of the films changed changes as a function of temperature.

5. Magetostrictive Actuators: To begin this work we have utilized pre-alloyed Terfenol-D ($Fe_2Tb_{0.27}Dy_{0.73}$) feedstock powder. The first trials at film deposition produced oxidized films with little magnetization. From a series of experiments, we determined that it was most likely that the oxidation was taking place during the ablation process due to oxygen that leaked into or was not purged from the ablation buffer gas. After taking great pains to eliminate all oxygen from the entire process, non-oxidized films of Terfenol-D are regularly obtained. For magnetometer testing and for later actuation testing, the films were deposited on Si wafer substrates cut into cantilevers. The films were 1.5 by 6.5 mm, and covered with ~300 nm of gold. The film volume was determined from profilometer cross-sections. They were in fact quite uniform in profile along the entire 6.5 mm cantilever length. Assuming a 50% packing density, the film weight was determined based on the film volume and bulk Terfenol-D mass density. These measurements were used to convert the raw magnetometer data into an absolute dipole density (emu/g). The M vs. H curves of the NP films showed good agreement in shape with the bulk material. The absolute value of saturation magnetization of the film was 44% of that measured for the bulk material. This value is comparable to films deposited by other techniques such as sputtering (for thinner films). Hysteresis losses were acceptable and the small loop closely resembled that of the bulk material. We find this result very encouraging, and so we are moving on to fabricate films (some deposited in a static magnetic field) for dynamic actuation testing.

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