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FIELD-SENSITIVE MATERIALS FOR OPTICAL APPLICATIONS

Sang H. Choi and Mark Little†
AIAA Senior Member, Sr. Research Scientist
NASA Langley Research Center
Hampton, Virginia 23681-2199

ABSTRACT

The purpose of investigation is to develop the fundamental materials and fabrication technology for field-controlled spectrally active optics that are essential for industry, NASA, and DOD applications such as: membrane optics, filters for LIDARs, windows for sensors and probes, telescopes, spectroscopes, cameras, light valves, light switches, flat-panel displays, etc. The proposed idea is based on the quantum-dots (QD) array or thin-film of field-sensitive Stark and Zeeman materials and the bound excitonic state of organic crystals that will offer optical adaptability and reconfigurability. Major tasks are the development of concept demonstration article and test data of field-controlled spectrally smart active optics (FCSAO) for optical multi-functional capabilities on a selected spectral range.

GENERAL DESCRIPTION OF PRINCIPLES

The field-controlled spectrally smart active optics (FCSAO) is the dynamic optical components and its concept is based on the constraint of quantum electronic behavior (weakly) and dipole moment change (strongly) in a thin-film or quantum-dot domain of materials that are effectively controlled by externally applied field. The principles that the current research activities are footed on are listed in the followings:

Quantum Electronic Constraint – Stark Effects: The effects on atomic and molecular energy level and spectra by externally imposed field give rise to radiative transitions as a consequence of higher order perturbation. Hence, the effect by higher order perturbation results in a weak interaction mainly to atomic level. This effect is generally called as the Stark effect [1]. When an electric field is used, it appears like the electrochromic effect but fundamentally different from the fact that electrochromism is based on the ionic state changes that give rise to the colored or bleached state from either anodic or cathodic charge. The constraint to quantum electronic behavior of atoms and molecules under an applied field is represented by the Stark effect as stated above and can be effectively used for selective spectral transmission in a filtering mechanism by state control through an externally applied field.

Quantum Electronic Constraint – Zeeman Effects: The effect on dipole moment of atoms or molecules by an externally applied magnetic field is often called as the Zeeman effect [2] and appears as level multiplicities and nuclear spins. Because of dipole interaction, the angular momentum vector (L) precesses around the field vector as a coupling with spin-orbit momentum vector (S) when Zeeman energy is much less than spin-orbit interaction energy. However, as the field effect increases (or the Zeeman energy increases), the angular and spin-orbit momenta individually are decoupled, thus resulting in destroying the spherical symmetry of the total Hamiltonian and producing a splitting linear with the field strength. In such a case, the magnetic (M) or Zeeman momentum strongly dictates degeneracy and controls level multiplicities (or level splittings) and nuclear spins, resulting in spectral variations. The Zeeman effect is more vivid for polyatomic molecules due largely to asymmetry nature of molecular structures and complex state levels. In short, the constraint on quantum state imposed by externally applied (magnetic) field gives rise to radiative transitions as a consequence of dipole interaction (magnetic). The Zeeman effect is much more strong and a wide range unlike the Stark effect. The optical transitions by the Zeeman effect encompass emission, absorption, and transmission spectra within atomic or molecular structures. Hence, the impinging quanta of spectra on or through Zeeman materials can be selectively gated for the color options (spectrally variable) rather than bleach (flux density control) by modulating externally applied fields.

Effects of Quantum-Dot Domain: In quantum-dots [3] that contain a small and controllable number (1~1000) of electrons, the electronic states are completely quantized due largely to the quantum confinement, the resonant structure associated with confinement, and granular nature of electric charge. And the energy spectrum is discrete (or eigen-

† ICASE, NASA Langley Research Center

energies). The density of (quantum) states is uniquely populated on discrete energy levels (Dirac δ-function) in quantum-dots unlike quantum-wells or quantum-lines. Therefore, the behavior of electrons in quantum-dots can be controlled by either electric or magnetic field, but only within discrete levels of energy spectrum that allows optical transitions even without Zeeman effect (spin splitting). There are many reported works [4] that proved optical transitions of II-VI and III-V materials with or without the applied field.

Organic Materials with Ouantum Effects: The organic crystals, similarly with II-VI and III-V materials [5] considered for research activity, show very interesting optical and electronic properties that are dictated by excitons. An exciton is a bound electron-and-hole pair that transports energy through a solid. The bound excitonic state of ordered organic crystalline structure (i.e. aluminum tris[8-hydroxyquinoline] or Alq_3) exhibit electroluminescence (EL) and photoluminescence (PL) under the injection of electron-hole pair

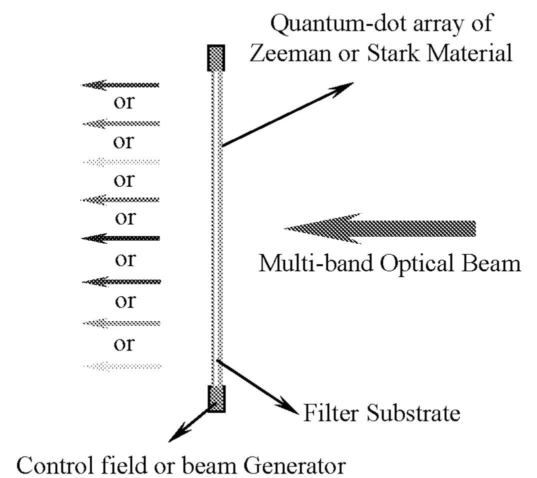

Fig. 1 Zeeman or Stark Effects

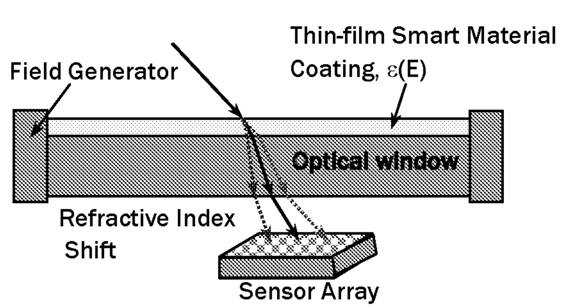

Fig. 2 Lattice Structure Change Effects

injection [6]. The FY02 research also include the organic crystals for FCSAO development.

Figures 1 and 2 show the concept of spectrally active smart optics. The quantum-dot array or thin-film of Zeeman or Stark materials or thin-film organic crystal is developed on an optical window substrate for smart active optical filtering. The thickness of quantum-dots is of tens of nanometers. A material to material for quantum-dots will offer dual filtering functions of spectral variability and intensity on a selected spectral range. From the performance aspect, the FCSAO is superior to other filter concepts. For instance, the response time of FCSAF is of quantum mechanical order since the field-to-dipole interaction is instantaneous. The thermal agitation and other noise sources are easily suppressed by manipulating an applied field. The fabrication of FCSAF requires a thin film technology.

SELECTION OF CANDIDATE MATERIALS

Many of the rare earth (RE) elements in lanthanide series with atomic numbers 58 through 70, is categorized as phosphor materials and therefore, considered for the smart active filter applications. The RE elements form a group of chemically similar

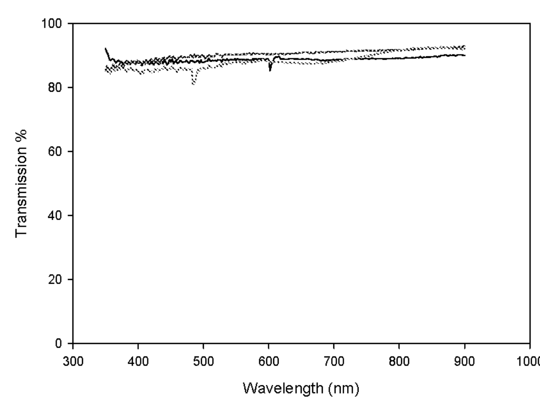

Fig. 3 Typical transmission spectra for $BaTiO_3$ infilled films

elements that have a partially filled 4*f* shell and usually take on a 3+ ionic state (RE^{3+}) of which energy levels are predominantly independent of their surroundings. In general, the 4*f* electronic energy levels of RE ion are shielded from external fields by $5s^2$ and $5p^2$ outer-shell electrons. Other distinctive nature that some RE elements have is the unusually high magnetic moments by the unpaired 4*f* electrons of which angular and spin-orbit momenta individually can be decoupled by applied fields and may result in radiative transition. The wide band-gap of semiconductor hosts, such as GaN, SiN and AlN, doped with RE offers a broad photonic transition at visible and near IR wavelengths.

THIN-FILM DEVELOPMENT AND TESTS OF CANDIDATE MATERIALS

Sol-Gel Films: A SiO_2 film infilled with $BaTi0_3$ is known to have a spectral shift by thermal agitation.

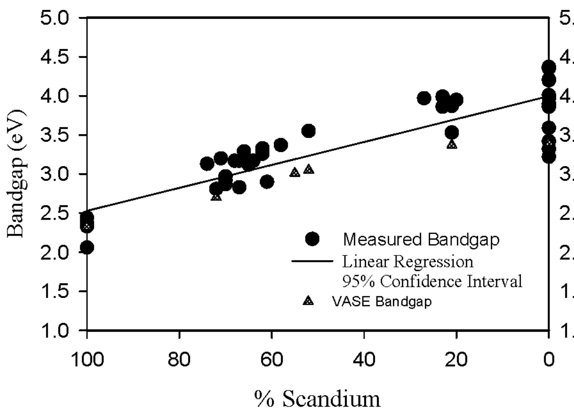

Fig. 4 Optical bandgap versus Scandium concentration.

To date many attempts were made to create a SiO_2 film infilled with a $BaTi0_3$ using the sol-gel method. However, $Si0_2$ adhesion to the substrate is a major problem. The films crack (like a dry river bed) and then flake off with very slight mechanical stimulus. A low concentration of 0.02 μm $Si0_2$ spheres was used to obtain a relatively uniform coating, although cracking was still present to a lesser degree. The films were then infilled with a $BaTi0_3$ precursor solution and sintered at 400-600 °C. However, the characteristic absorption seen by Zhou [7] at 600 nm is not present. This suggests that an insufficient amount of $BaTi0_3$ is entering the material matrix to produce the effect. Typical transmission spectra of a lab sample are shown in Figure 3.

Sputtered Films: Alloys of amorphous GaN/ScN and alloys of GaSiN have been investigated with the variable angle spectroscopic ellipsometer (VASE), test the optical equipment, and establish their usefulness as future optical materials. Concentration of scandium has been given to $Sc_xGa_{1-x}N$ systems. These exhibit band-gap engineering, as shown in Figure 4, which could lead to their use as an electronic material. (Minimal electronic properties measurements have been established in the lab with equipment present to test new materials). In addition, it was found with VASE that the optical properties show a dependence on the film composition. This will be crucial in producing Fabry Perot type filters with controllable properties. The index of refraction *n* was found to vary with film composition as shown in Figure 5 while the extinction coefficient *k* remained almost constant for the same range of wavelength (see Figure 6). Further measurements and, more importantly, additional growth is needed to establish the repeatability of these measurements. However, the initial findings are encouraging.

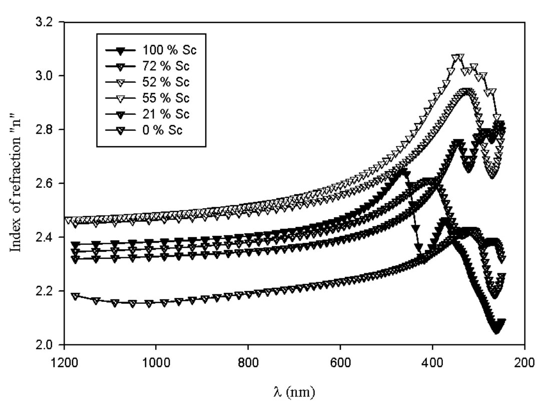

Fig. 5 Index of refraction in the region below optical absorption. Data generated from variable angle spectroscopic ellipsometry (VASE).

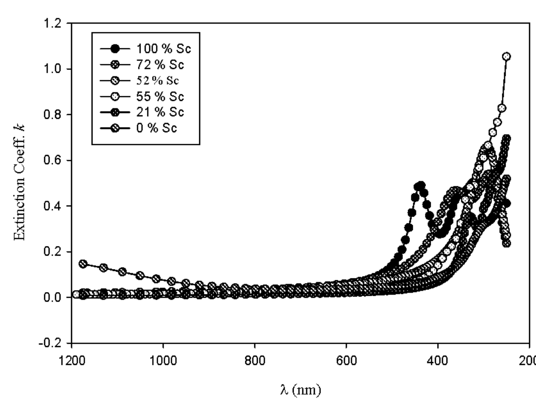

Fig. 6 Extinction coefficient κ from VASE

A system was adapted to the VASE machine to allow for applying a potential to the films while they are being measured. A film of amorphous/microcrystalline GaN shows a slight decrease in index of refraction and extinction coefficient over the range 4-5 eV with increasing applied voltage as shown in Figures 7 and 8. Interestingly, a small amount of spectral shifts appears over the range of 4.5 ~ 5 eV as shown in Figure 8. These shifts correspond to the shifts of refractive indices as appeared in Figure 7. There is also a decrease in the extinction coefficient over this range. The film was measure with zero, 10, and 15 volts applied to contacts on the film edge. With high applied voltages, the change in index of refraction and the spectral shifts will appear much more distinctively than in the low applied voltages. This experiment was done with only electrical fields and resulted in very promising phenomena that will allow the development of smart active optics. In the next phase of experiments, the effects of magnetic field (Zeeman effects) on $Sc_xGa_{1-x}N$ systems will be interrogated to demonstrate both the changes in index of refraction and spectral shifts. The data shown so far is from one sample, one test run, and is above band-gap, and thus highly preliminary and should be reported with caution. However, if it is reliable, it would be a major step forward to the research.

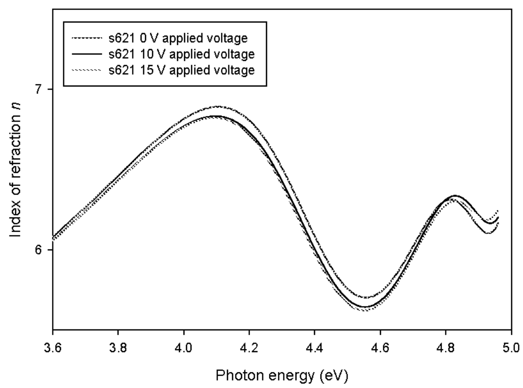

Fig. 7 Change in refractive index with applied voltages

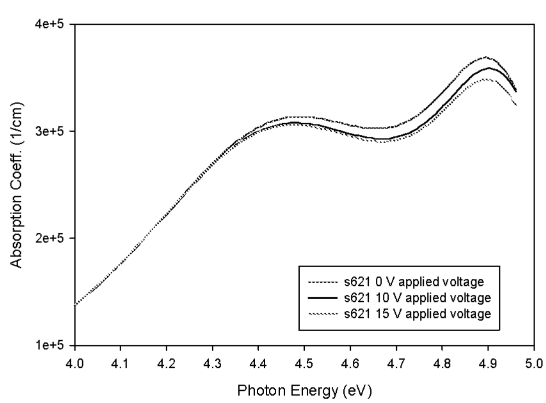

Fig. 8 Change in absorption coefficient with applied voltages.

CONCLUSION

Various thin-film structures of several optical materials were developed to characterize the unique optical properties that are influenced by Zeeman or Stark effects. By the Zeeman and Stark effects, the quantum level transition within a material can be modified with the applied magnetic or electric field. The changes in quantum constraint of a material are normally appeared with a pattern of spectral shift and/or refractive index shift. The first indication of these effects can be verified with the refractive index shift. Some of the tested materials indicate that the shift in refractive index exists. The promising results were obtained from a film of amorphous/microcrystalline GaN which shows a slight decrease in index of refraction and extinction coefficient over the range 4-5 eV with increasing applied voltage up to 15 volts. Interestingly, a small amount of spectral shifts appears over the range of 4.5 ~ 5 eV. These shifts correspond to the shifts of refractive indices. There is also a decrease in the extinction coefficient over this range. With high applied voltages, the change in index of refraction and the spectral shifts will appear much more distinctively than in the low applied voltages. Co-sputtered Al and Er in nitrogen/argon atmosphere producing AlN:ErN alloy has the concentration ratio of 80:20. This sample shows the strongest evidence of possible field induced optical constant shifts. The non-point-to-point (PTP) fit shows strong evidence of a change in the index of refraction *n* and extinction coeff. *k* with applied film. The PTP fit also shows a characteristic change in *n* and *k (to a lesser degree)* with increasing field.

The research on Zeeman and Stark effects with new materials is still underway in our laboratory. The results obtained so far appear promising and indicate that the originally indented idea is valid as expected.

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