A Titanium Nitride based Metamaterial for Applications in the Visible

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Abstract: Epitaxially grown TiN/$Al_{0.6}Sc_{0.4}N$ superlattice behaves as a hyperbolic metamaterial (HMM) in the visible range. Since HMMs enhance photonic-density-of-states and reduce lifetime of an emitter, we observed nine times decrease in lifetime of a dye molecule placed close to this HMM.

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1. Introduction

New materials as substitutes for noble metals could greatly boost the performance of many plasmonic and metamaterial devices [1]. The application horizon of these devices expands significantly by the incorporation of new plasmonic materials. For example, TiN as an alternative plasmonic material could make the devices CMOS compatible. Also, the high melting temperature and hardness of TiN are very useful for applications such as thermal radiation engineering and energy harvesting [2]. New plasmonic materials such as TiN also enable better devices and exploration of new physics associated with them. For example, hyperbolic metamaterials (HMMs) are shown to produce broadband enhancement in the photonic-density-of-states (PDOS). Plasmonic materials such as TiN are found to make high performance HMMs in the visible [2]. Hence, we perform an experiment to observe the enhancement in PDOS associated with TiN-based HMMs. In this report, we summarize the findings of an experiment designed to probe the PDOS of a TiN/$Al_{0.6}Sc_{0.4}N$ HMM.

2. TiN/$Al_{1-x}Sc_xN$ superlattice as HMM

The fabrication of a good quality HMM would require ultrathin and smooth alternating layers of metal and dielectric with sharp interfaces. This is possible if the metal/dielectric superlattice can be epitaxially grown. TiN grows as an epitaxial film on MgO (100). AlN when alloyed with 40 at% of ScN can be stabilized in the cubic rocksalt phase and matches the lattice constant of TiN. We performed optimization of the growth of $Al_{1-x}Sc_xN$ thin films sandwiched between TiN layers, and found that $Al_{0.6}Sc_{0.4}N$ can be stabilized in the cubic phase and grows lattice matched on TiN. Thus, we prepared HMMs comprising TiN as the plasmonic material and $Al_{0.6}Sc_{0.4}N$ as the dielectric material. A Transmission Electron Microscopy (TEM) image of the interface of TiN and $Al_{0.6}Sc_{0.4}N$ layers is shown in Fig 1a. The figure also shows electron diffraction patterns recorded from the TiN and $Al_{0.6}Sc_{0.4}N$ layers. Sharp interfaces and perfect lattice matching are evident.

The optical properties of the superlattice were characterized using a spectroscopic ellipsometer (J.A. Woollam Co.). The superlattice was modeled with an effective medium model with uniaxial anisotropy. The dielectric functions are extracted for the two orthogonal directions, viz. parallel to layers (||) and perpendicular to layers ($\perp$). Figure 1b shows the dielectric functions of a superlattice consisting of 5 nm thin TiN and 5 nm thin $Al_{0.6}Sc_{0.4}N$. The superlattice shows transverse positive (type-1) hyperbolic dispersion around 550 nm and transverse negative (type-2) hyperbolic dispersion for longer wavelengths. Thus, TiN/$Al_{0.6}Sc_{0.4}N$ superlattice behaves as a HMM for visible and longer wavelengths.

In order to probe the PDOS of TiN/$Al_{0.6}Sc_{0.4}N$ HMM, we conduct an experiment similar to those reported in references [4-6]. A 300 nm thick HMM comprising of 5 nm each TiN and $Al_{0.6}Sc_{0.4}N$ layers (TiN top layer) is grown. On the top of this sample is grown a 5 nm thin spacer of $Al_{0.6}Sc_{0.4}N$. A 100 µM solution of LD800 dye in SU-8 (1:5 in thinner) is spin coated onto the HMM. The lifetime of the dye is measured using fluorescence lifetime imaging system (MicroTime 200 Picoquant). The pump wavelength was 635 nm (88 ps pulses) and the detection wavelength range spanned from 650 nm to 720 nm. The average lifetime of dye on HMM was recorded to be 0.26 ns and that on bare MgO was 2.1 ns (see Fig. 1c). A control sample was prepared consisting of 5 nm thick

$Al_{0.6}Sc_{0.4}N$ on top of 5 nm thick TiN film (on MgO substrate). The average lifetime of dye recorded on the control sample was 0.7 ns. Overall, we observe nearly 9x reduction in lifetime of dye molecules on TiN/$Al_{0.6}Sc_{0.4}N$ HMM compared to the bare substrate and nearly 3x reduction in lifetime compared to the control sample.

3. Conclusion

The reduction in lifetime of dye molecules close to the TiN/$Al_{0.6}Sc_{0.4}N$ HMM is larger than that obtained with the control sample. This could indicate that the radiative decay rate of the emitter is higher in the case of the HMM. Nearly nine times enhancement of decay rate of emitters close to the HMM indicates a large enhancement in PDOS as predicted by the theory. Such high performance metamaterials enabled by a better plasmonic material such as TiN opens a wider horizon for the applications of HMMs and new avenues in exploring exciting physics with metamaterials.

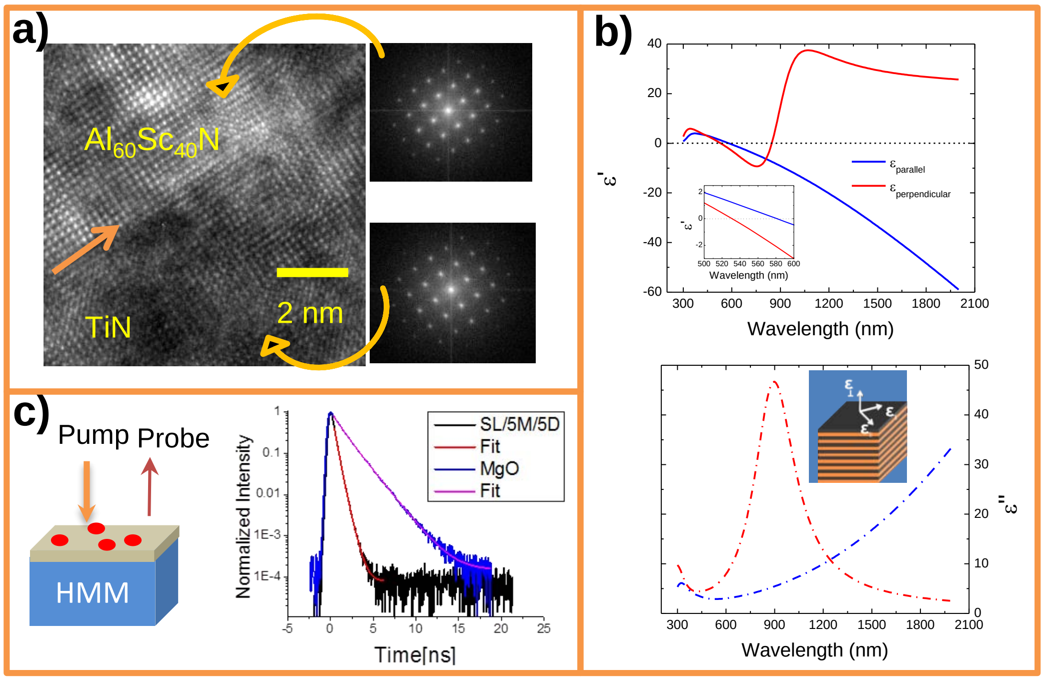

Fig 1. a) TEM image of the TiN/$Al_{0.6}Sc_{0.4}N$ superlattices. Clear and distinct interface of the constituent layers are observed in this high resolution phase contrast micrograph of the interface. The side panels are diffractograms from the image, demonstrating the cube on cube epitaxial relationship between the layers. b) Dielectric functions of the uniaxial effective medium formed by the superlattice. The superlattice shows hyperbolic dispersion in the visible and longer wavelengths. c) The experimental set-up (left panel) consisting of LD800 dye molecules placed on a TiN/ $Al_{0.6}Sc_{0.4}N$ (5 nm each) HMM with a $Al_{0.6}Sc_{0.4}N$ spacer of 5 nm. The lifetime of the dye molecules is measured in reflectance mode as shown. The right panel shows the result of lifetime measurement on the superlattice and bare MgO substrate. The fitted curves extract the lifetimes of dyes on HMM and bare MgO substrate to be 0.26 ns and 2.1 ns, respectively.

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