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### Deep subwavelength photonic multilayers fabricated by atomic layer deposition

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Photonic dielectric multilayers are one of the most studied systems in optics [1]. Applications include Bragg mirrors, antireflection coatings, band-pass filters etc. Research traditionally focused on layered structures with thicknesses d compared to the wavelength of light  $\lambda$  and omitted much thinner layers. It was commonly assumed that the description of such deep  $(d\ll\lambda)$  subwavelength multilayers is governed by effective media approximation (EMA) theory [2]. However recent theoretical publications [3], [4] suggest that EMA assumption may fail if the angle of incidence is close to the angle of total internal reflection. The transmission spectrum in this case becomes different than predicted by EMA. The spectrum becomes sensitive to layers thicknesses on nanometer scale and their order in multilayer. Moreover the EMA breakdown effect can be enhanced by placing the multilayer in photonic resonator set-up [4] as it shown in Fig 1. In this work we present the fabrication approach to the described structure.

 $TiO_2$  and  $Al_2O_3$  metal oxides were selected as the multilayer components and silicon nitride (Si<sub>3</sub>N<sub>4</sub>) as photonic resonator. The fabrication of the multilayers was performed in a commercial hot-wall ALD system (Picosun R200, Finland). The precursors used for Al<sub>2</sub>O<sub>3</sub> and TiO<sub>2</sub> deposition were trimethylaluminum Al( $CH_3$ )<sub>3</sub> and titanium tetrachloride (TiCl<sub>4</sub>), respectively. An oxidant source in both processes was deionized water. The deposition temperature was chosen at 120C in order to prevent the crystal anatase phase transition of TiO<sub>2</sub> known to occur at temperatures above 150C [5]. Such transition would increase the films roughness. To establish the deposition rate,  $Al_2O_3$  and  $TiO_2$  films with thickness ranging between 10 and 50 nm were deposited on Si substrates, with subsequent ellipsometric characterization of the films thicknesses and refractive indices. It was determined that  $TiO_2$  and  $Al_2O_3$ have a constant growth rate of 0.047 and 0.089 nm/cycle respectively.

The substrates for the samples were fabricated by depositing 1  $\mu$ m of Si<sub>3</sub>N<sub>4</sub> (the resonator layer) on 100 mm silicon < 100 > wafers using low-pressure chemical vapor deposition. The process was carried out at 780C with ammonia (NH<sub>3</sub>) and dichlorosilane (SiH<sub>2</sub>Cl<sub>2</sub>) as reactive gases. Thickness and refractive index of the deposited silicon nitride was measured and confirmed using spectroscopic ellipsometry. The deposited Si<sub>3</sub>N<sub>4</sub> film was carefully analyzed for existence of cracks, particles and other defects using dark field optical microscopy. The best-quality wafer with Si<sub>3</sub>N<sub>4</sub> was selected and cleaved in pieces, which were used as substrates for the deposition of  $Al_2O_3/TiO_2$  multilayers. Before inserting each substrate into the ALD reactor, it was placed on a Si carrier wafer. Therefore the Al<sub>2</sub>O<sub>3</sub>/TiO<sub>2</sub> multilayers were grown not only on the  $Si_3N_4$  layer but also on the dummy carrier wafer. After the ALD process was completed, the dummy was cleaved and its cross-section was characterized using scanning electron microscopy (SEM). The SEM images reveal high-quality homogeneous, conformal coatings, as seen in the examples in Figs 2 and 3. Such a method of deposited multilayers characterization turned out to be more feasible than the direct SEM characterization of multilayers on  $Si_3N_4$ , since the latter suffers from issues related to charge accumulation on the silicon nitride.

We believe this fabrication flow opens possibility for experimental demonstration of EMA theory breakdown in deep subwavelength multilayers.

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of each layer is 10 nm.

Figure 2. SEM image of fabricated 200 nm Figure 3. SEM image of fabricated 200 nm TiO<sub>2</sub> TiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> multilayer on Si substrate. Thickness /Al<sub>2</sub>O<sub>3</sub> multilayer on Si substrate. Thickness of each layer is 20nm.