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Near-infrared electroluminescence from atomic layer doped Al_2O_3:Yb nanolaminate films on silicon

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ABSTRACT

Nanolaminate Al_2O_3:Yb films are fabricated by atomic layer deposition on silicon, from which intense 977 nm electroluminescence is obtained. By precise controlling the Yb agglomerate and the distance among Yb^{3+} ions, the power density up to 2.67 mW cm^{-2} and external quantum efficiency of 1.8% are estimated from the nominal 1.27 at.% doped device. The charge transport is ascribed to the Poole-Frenkel conduction mechanism and the electroluminescence originates from the impact-excitation of Yb^{3+} ions by hot electrons. The distance for the presence of non-radiative interaction of Yb^{3+} ions in Al_2O_3 matrix is concluded to be ~3 nm.

Integrated optoelectronics is expected to provide high performance and sustainable solution to the current limitations of Si microelectronics industry. The electrically-driven light sources integrated on silicon are indispensable and have triggered the scientific attention for years [1–3]. Silicon compatibility remains challenging for semiconductor-based approaches due to the inefficient light emission of silicon and the complexity and high cost of heterogeneous integration techniques [4]. Rare-earth (RE) doped materials offer certain unique advantages and can be important alternatives, the emissions from which show much less sensitivity to the temperature together with low fluorescence background [5,6]. Ytterbium (Yb) plays an important role in fiber amplifiers and lasers at ~1 μm wavelength region, which is of exceptional interest for various optical applications [7–9]. The intense and broad Yb^{3+} absorption lines are well suited for near-infrared (NIR) InGaAs diode pumping, and the small Stokes shift between absorption and emission spectra is in favor of efficient laser. The kW-scale Yb-doped fibers have been the leading contenders because of the excellent efficiency [10]. Moreover, Yb^{3+} ion serves as the structural probe and the sensitizer for other RE ions [11,12].

Al_2O_3 is a promising host material for Yb^{3+} ions due to its high stability and a wide range of transparency. In addition, the good adhesion to Si surface makes Al_2O_3 attractive in the microelectronics and optoelectronics [13,14]. Although a good platform for optical excitation, the insulating property impedes effective electric injection. So far there are few reports on the electroluminescence (EL) from Yb-doped Al_2O_3, which is of great importance. Atomic layer deposition (ALD) is a versatile technique to deposit thin films with nanometer-scale precision over large substrates. The depositions of Al_2O_3 and RE oxides by ALD are getting mature [13,15]. Demonstrating atomic-layer control allows for locating the Yb^{3+} ions into the more favorable environment for luminescent excitation and opens new routes to the engineering materials with targeted optical and electrical properties.

In this paper, we fabricate the nanolaminate Yb-doped Al_2O_3 films by ALD, and the characteristic 977 nm EL is realized from the Si-based metal-oxide-semiconductor light-emitting devices (MOSLEDs). The ALD nanolaminates offer the possibility of controlling the dopant agglomerate and the distance between adjacent dopant layers, which is beneficial for determining the optimum doping concentration and distance among Yb^{3+} ions that render maximum EL efficiency. This material could find important applications in the integrated photonics where compact light sources are required.

The Al_2O_3:Yb films were grown on 2–5 Ω·cm *n*-Si substrates, which were cleaned through the standard RCA process and then loaded into a 4-inch chamber ALD system. $Yb(THD)_3$ (THD: 2,2,6,6 tetramethyl 3,5 heptanedionate) and Trimethylaluminum [TMA, $Al(CH_3)_3$] were used as the precursors for Yb_2O_3 and Al_2O_3, respectively, with ozone as the oxidant. N_2 was used as the carrier and purge gas with a flow rate of 20 sccm. The growth chamber was firstly evacuated to a base pressure of 20 Pa. During the growth, the TMA source was maintained at room temperature, while the Yb precursor and the precursor delivery lines were heated at 195 °C. One Yb_2O_3/Al_2O_3 cycle consist of 2 s $Yb(THD)_3$/0.015 s TMA pulse, 5 s N_2 purge, 1.8 s ozone pulse, and 9 s N_2 purge. The substrates were maintained at 350 °C, resulting in the growth rate of 0.2 and 1.0 Å/cycle for Yb_2O_3 and Al_2O_3 films, respectively. For exploration of the luminescent Al_2O_3:Yb films, three series of samples were fabricated concerning the Yb dopant layer, Al_2O_3 interlayer, and total thickness, respectively, by

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adjusting the corresponding Yb_2O_3 or Al_2O_3 cycles. After the deposition, the films were annealed at 800 °C or 1100 °C in N_2 atmosphere for 1 h to enable stoichiometry and Yb activation. Afterwards, ~100 nm TiO_2/Al_2O_3 films with 1.94 nm Al_2O_3 and 8.0 nm TiO_2 sublayers were deposited onto the Al_2O_3:Yb films by ALD, acting as the protection layer to enhance the device stability. ~100 nm ZnO:Al electrode films were deposited onto the TiO_2/Al_2O_3 films by ALD, which were lithographically patterned into 0.5 mm circular dots. ~100 nm Al films were subsequently deposited on the backsides of *n*-Si by heat evaporation.

The thickness of the films was measured by an ellipsometer with a 632.8 nm He-Ne laser beam at an incident angle of 69.8°. The crystal structures of Al_2O_3:Yb films were characterized by X-ray diffraction patterns (XRD, D/max 2500/pc, Rigaku, Cu Kα radiation, $\lambda = 1.5406$ Å). Surface morphologies were scanned by atomic force microscopy (AFM, Dimension Icon, Bruker). To activate EL from the MOSLEDs, forward bias was applied with the negative voltage connecting to *n*-Si. In comparison negative biases feature the lower EL at higher voltages. EL and Current-Voltage (I-V) characteristics were recorded by a Keithley 2410 SourceMeter. The EL signal was collected by a 0.5 m monochromator and detected by an InGaAs detector connected to a Keithley 2010 multimeter. The absolute EL power from the device surface was measured by a calibrated optical power-meter (1830-C with 818-IR Sensor, Newport).

XRD patterns of the Al_2O_3:Yb films with 1 nm Al_2O_3 interlayer and 2 Yb dopant cycles are depicted in Fig. 1(a). The 800 °C annealed film shows no diffraction peak, indicating an amorphous Al_2O_3 matrix. In comparison, the 1100 °C annealed film presents some broad diffraction peaks ascribed to nano-crystalline Al_2O_3. No Yb-related phase presents in either patterns. Previous studies on the crystallinity of Al_2O_3 have shown that temperature above 1500 °C is needed for the prominent crystallization. Surface morphologies scanned by AFM for the films are shown in Fig. 1(b). Both the film surfaces are quiet smooth without notable grains. For the Al_2O_3:Yb film annealed at 800 °C, the root-mean-square roughness is 0.35 nm, which increases to 2.27 nm after annealing at 1100 °C. Thus the 1100 °C annealed film presents slight grain-growth, which is in consistent with the XRD patterns that Al_2O_3 nanocrystals emerge after annealing at 1100 °C. Because of the difference of ionic radius between the Al^{3+} (~0.5 Å) and Yb^{3+} ions (0.86 Å), disordered or amorphous Al_2O_3 maintaining vacancy and defect sites supplies a better host to Yb dopant. The current injections of the 1100 °C annealed devices are much limited in comparison with that of the 800 °C ones. Moreover, the precipitation of Yb^{3+} ions pertaining to high temperature annealing causes a reduction of luminescent Yb centers, the EL performance is consequently reduced [16,17]. Thus we focus on the 800 °C annealed Al_2O_3:Yb films for device fabrication.

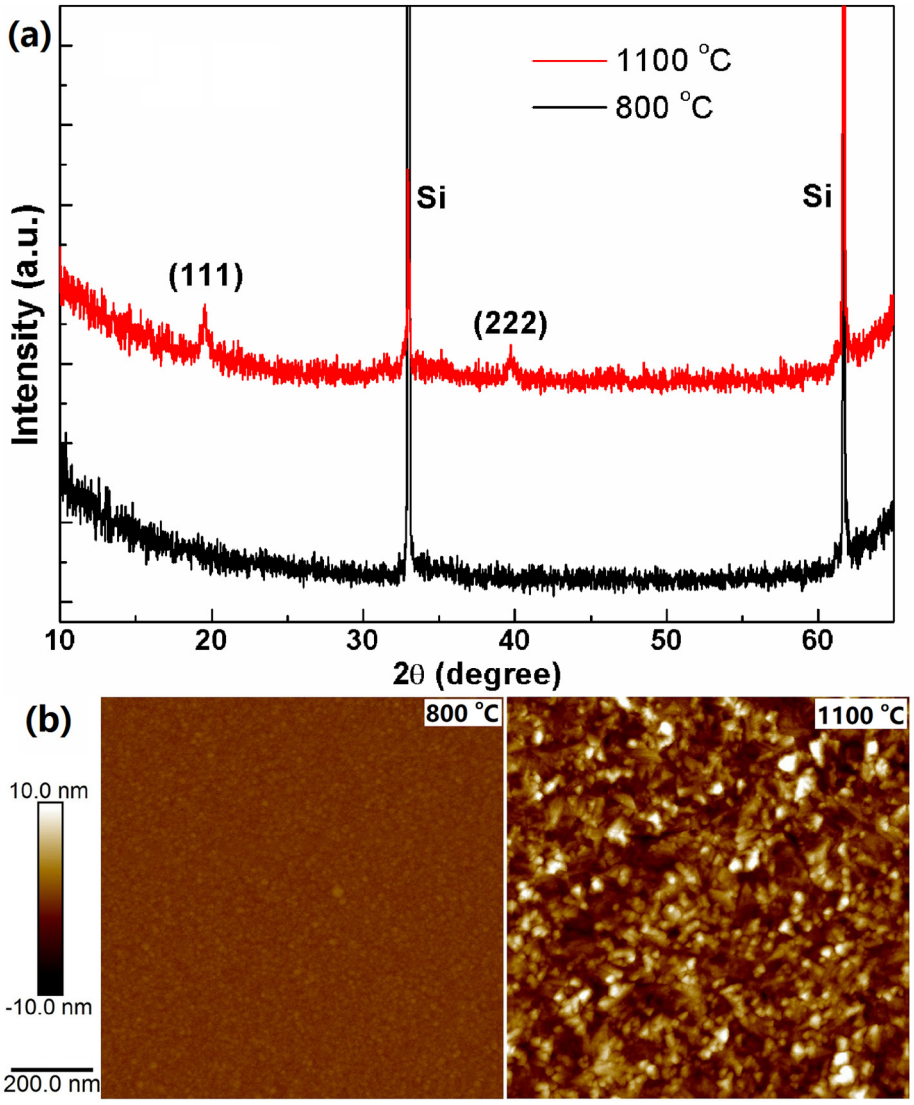

Fig. 1. XRD patterns (a) and 3D AFM images (b) of the Al_2O_3:Yb films with 1 nm Al_2O_3 interlayer and 2 Yb dopant cycles, annealed at 800 °C and 1100 °C, respectively.