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Electroluminescent Yb₂O₃:Er and Yb₂Si₂O₇:Er nanolaminate films fabricated by atomic layer deposition on silicon



Zhongtao Ouyang, Yang Yang*, Jiaming Sun**

School of Materials Science and Engineering, Nankai University, Tianjin 300350, China

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ABSTRACT

Atomic layer doped Yb_2O_3 :Er and $Yb_2Si_2O_7$:Er nanolaminate films are fabricated on silicon by atomic layer deposition, and $\sim 1530\,\mathrm{nm}$ electroluminescence (EL) is obtained from the metal-oxide-semiconductor light-emitting devices (MOSLEDs) based on these films. The Yb_2O_3 films transfer to $Yb_2Si_2O_7$ phase after annealing above $1000\,^\circ\mathrm{C}$. Intense photoluminescence from $Yb_2Si_2O_7$ film confirms high efficiency and energy transfer under optical excitation, but the limited electron conduction restricts the EL performance. EL from the Yb_2O_3 :Er MOSLED outperforms, presenting an external quantum efficiency up to 8.5% and the power efficiency of 1×10^{-3} . The EL is derived to result from the impact excitation of Er^{3+} ions by hot electrons, which stem from Fowler-Nordheim tunneling mechanism under sufficient bias voltage. The critical distance for the cross relaxation of doped Er^{3+} ions in nanolaminate Yb_2O_3 matrix is experimentally determined to be $\sim 3\,\mathrm{nm}$. Such devices manifest the technological potential of Er-doped Yb-oxides for applications in silicon-based optoelectronics.

1. Introduction

Research on the realization of electrical-driven silicon-based light sources holds promise for the full integration of optoelectronics, which would take advantage of the mainstream CMOS technology and overcome contemporary technological issues in microelectronics and telecom [1,2]. Among various emitters, Er³⁺ ions have attracted considerable interest since their near-infrared (NIR) emission at 1.53 µm coincides with the minimum loss window of optical telecommunication [3]. However, Er³⁺ ions undergo significant quenching due to non-radiative energy back-transfer in silicon. Strategies to avoid such demerit consist of using wide bandgap hosts like SiO2 or Si3N4. Although good platforms for photoluminescence (PL), their insulating properties impede electroluminescence (EL) performance [4-6]. Attempt to increase luminescent efficiency by increasing the doping density encounters the concentration quenching due to clustering or interaction of Er³⁺ ions when the density is over $10^{20} \, \text{cm}^{-3}$ [7,8]. The RE compounds are possible alternatives, in which the Er concentration can reach $10^{22}\,\mathrm{cm}^{-3}$ and the reduction of clustering is expected due to periodic arrangement of the dopant atoms [9]. Therefore, it is promising to inspect RE oxides as host for Er^{3+} ions to yield the 1.53 μ m luminescence.

PL enhancement for the Er-related 1.53 μm emission has been reported for a range of sensitizers, among which Yb³⁺ ions is of great

In this work, we fabricate metal-oxide-semiconductor light-emitting devices (MOSLEDs) based on nanolaminate Yb_2O_3 :Er and $Yb_2Si_2O_7$:Er films, which are deposited using atomic layer deposition (ALD). The

E-mail addresses: mseyang@nankai.edu.cn (Y. Yang), jmsun@nankai.edu.cn (J. Sun).

importance due to the much broader Yb3+ absorption band with the 10-fold larger cross section than Er³⁺ at 900-1000 nm [10-12]. These enable a potential to excite Er3+ ions more effectively in Yb-related matrix via the energy transfer from the ${}^2F_{5/2}$ level of $Yb^{3\,+}$ to the ${}^4I_{11/2}$ level of Er³⁺ ions [13]. This sensitizing process has long been researched for homogeneously doped films, focusing on the up-conversion PL characteristic [14,15]. However, a high EL efficiency imposes strict requirements to sufficient electron injection, a difficulty that results in few reports on EL from such compounds to date. Even the reported results in the literatures are conflicting due to the disparities in electrical and optical properties between different RE oxides as well as the fabrication-dependent properties observed for similar materials [4,16,17]. In order to taking advantage of the high doping possibility and sensitizing properties of Yb-related oxide (such as Yb2O3 and Yb₂Si₂O₇) to realize Er³⁺ EL, the nanolaminate films are preferable, which allow for locating the Er³⁺ ions in more favorable environment for excitation and supply new routes to achieve the targeted optical and electrical properties. The realization of EL from Er³⁺ ions in the Yb₂O₃ and Yb2Si2O7 nanolaminate films is expectable to be an important alternative in optoelectronics applications.

^{*} Corresponding author.

^{**} Corresponding author.

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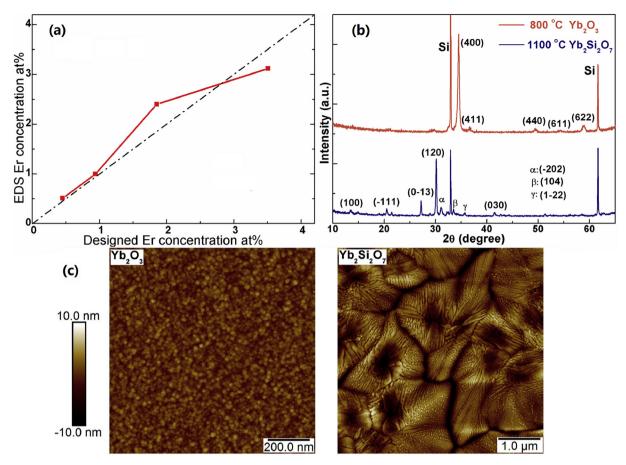


Fig. 1. (A) The Er-doping concentrations detected by EDS, in different Yb_2O_3 :Er films. (b) The XRD patterns of Yb_2O_3 :Er and $Yb_2Si_2O_7$:Er films deposited with 1 nm Yb_2O_3 interlayer, representing the deposited films annealing at 800 °C and 1100 °C. (c) Surface morphologies scanned by AFM for the Yb_2O_3 :Er films annealed at 800 °C and $Yb_2Si_2O_7$:Er films annealed at 1100 °C.

unique growth mechanism of ALD, based on successive self-limiting gas-surface reactions, simultaneously realizes the precise control of the thickness and composition with excellent homogeneity [18–20]. By alternating deposition sequence of $\rm Er_2O_3$ and $\rm Yb_2O_3$, nanolaminate $\rm Yb_2O_3$:Er films with varying interlayers are formed. Annealing above 1000 °C turns the films into $\rm Yb_2Si_2O_7$. Intense PL from $\rm Yb_2Si_2O_7$ film shows high efficiency and energy transfer under optical excitation, but the limited electron conduction restricts the EL performance. MOSLED based on $\rm Yb_2O_3$:Er film is superior in EL, presenting an external quantum efficiency (EQE) up to 8.5% and a power efficiency (PE) of 1×10^{-3} . The critical distance for the cross relaxation of doped $\rm Er^{3+}$ ions in nanolaminate $\rm Yb_2O_3$ matrix is experimentally determined to be ~ 3 nm. These results provide an avenue for RE-doped nanolaminates to further optoelectronic applications.

2. Experiments

The luminescent films were grown on <100>-oriented n-Si with a resistivity of 2–5 Ω cm, which were cleaned through the standard RCA process and then loaded into the 4-in. chamber ALD system. N₂ 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. Yb (thd)₃ and Er (thd)₃ (thd = 2,2,6,6-tetramethyl-3,5-heptanedionate) were used as the precursors for Yb₂O₃ and Er₂O₃, respectively, with ozone acting as the oxidant. During the ALD process, the Yb and Er sources were maintained at 195 °C and 200 °C, respectively, while the precursor delivery lines were heated at 200 °C. One Yb₂O₃/Er₂O₃ cycle consists of 2 s Yb(thd)₃/Er (thd)₃ pulse, 5 s N₂ purge, 1.8 s ozone pulse, and 9 s N₂ purge. The substrates were maintained at 350 °C. Based on

the former research, 2 dopant cycles are preferable concerning both the efficient doping and the absence of RE clustering [21]. The growth rate for the Yb_2O_3 and Er_2O_3 films are 0.2 and 0.22 Å/cycle respectively, which agree well with the previous reports [22]. For exploration of the Yb_2O_3 :Er films, the growth cycles of Yb_2O_3 vary from 25 to 200, corresponding to a Yb_2O_3 interlayer of 0.5–4 nm. After the deposition, the films were annealed at 800 °C or 1100 °C in N_2 atmosphere for 1 h to enable stoichiometry. Subsequent device procedures are as previously mentioned [21], resulting in the multilayer-structured MOSLEDs of $ZnO:Al/TiO_2-Al_2O_3/Yb_2O_3:Er$ or $Yb_2Si_2O_7:Er/Si/Al$. The top ZnO:Al electrodes were lithographically patterned into 0.5 mm circular dots.

The film thickness was measured by an ellipsometer with a 632.8 nm He-Ne laser at an incident angle of 69.8°. The film composition was detected by energy-dispersive X-ray spectroscopy (EDS, X-Max^N 50, Oxford). The crystal structures of the films were characterized by X-ray diffraction patterns (XRD, D/max 2500/pc, Rigaku, Cu Kα radiation, $\lambda = 1.5406 \,\text{Å}$). Surface morphologies were scanned by atomic force microscopy (AFM, Dimension Icon, Bruker). The PL spectra were collected by an Edinburgh FLS920P Spectrometer, under different laser excitations. To activate EL from the MOSLEDs, appropriate forward bias was applied with the negative voltage connecting to the *n*-Si substrates. In comparison negative biases feature a 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 using a calibrated optical power meter (1830-C with 818-IR Sensor, Newport). The high-frequency (1 MHz) capacitance-voltage (C-V) characteristics were measured by an automatic

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