P-34: Cu Doping of Atomic Layer Epitaxy SrS via a Rapid Thermal Anneal Process

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Abstract
A process for Cu doping of atomic layer epitaxy (ALE) deposited SrS for thin-film electroluminescent devices is presented. The process involves the evaporation of dopant-containing material onto undoped ALE SrS thin films and rapid thermal annealing of the films. Green SrS:Cu,Na, greenish-blue SrS:Cu,Cl, and blue SrS:Cu,Ag,Cl phosphors are prepared using this processing methodology.

Technical Summary
SrS:Cu is emerging as a bright and efficient phosphor for blue and green alternating-current thin-film electroluminescent (ACTFEL) devices. [1, 2, 3, 4, 5] The development of SrS:Cu ACTFEL displays for commercial use could be achieved using an atomic layer epitaxy (ALE) process. A major obstacle in the realization of an ALE SrS:Cu device has been the lack of a suitable Cu precursor. The purpose of this work is to report an approach for doping ALE SrS films with Cu via thermal evaporation and rapid thermal annealing.

Beginning with an un-doped SrS thin film deposited by ALE, the addition of Cu dopant is accomplished by thermal evaporation of a Cu containing source, such as CuS, CuCl2, or CuF2, followed by a rapid thermal anneal of the sample. Additional dopants are also sometimes incorporated into the phosphor by depositing additional layers prior to the anneal.

The ACTFEL devices employed in this study are fabricated as follows. An un-doped SrS film with a thickness of ~1000 nm is deposited onto a glass substrate coated with layers of indium tin oxide and aluminum-titanium oxide, which serve as the bottom transparent contact and the bottom insulator, respectively, of the ACTFEL device. The SrS layer is ALE deposited with H2S and Sr(thd)2 precursors, where thd stands for 2,2,6,6-tetramethyl-3,5-heptanediionate. Next, thin layers of the appropriate dopant species are thermally evaporated onto the SrS film. Rapid thermal annealing in an Ar atmosphere is then performed in order to diffuse the dopants into the SrS film and enhance the crystallinity of the phosphor. A top insulating layer of silicon oxynitride is deposited by plasma-enhanced chemical vapor deposition, and aluminum dots are thermally evaporated as the top contact.

Electroluminescence spectra for three devices prepared in this manner are shown in Fig. 1. Note that the color may be tuned from green to blue via appropriate dopant incorporation. [5] Various anneal temperatures have been explored. Evaporated and
sputtered SrS films are typically annealed at 810°C. It is found that the electrical stability of the ALE SrS ACTFEL devices degrades significantly when annealed at temperatures above ~750°C, resulting in frequent burn-outs during electrical operation. Figure 2 shows the luminance at 40 volts above threshold for SrS:Cu,Na devices as a function of the anneal temperature. The luminance maximizes at an anneal temperature of ~700°C.

Green SrS:Cu,Na devices are prepared by evaporating a layer of Cu2S followed by a layer of NaF onto the SrS thin film. The luminance and efficiency as a function of applied voltage for a device rapid thermal annealed in Ar at 700°C are shown in Fig. 3. All measurements reported are performed using a bipolar trapezoidal waveform with 5, 30, and 5µs rise, peak, and fall times applied at a frequency of 60 Hz. At 40 volts over threshold a brightness of \( L_{40} = 50 \text{cd/m}^2 \) and efficiency of \( E_{40} = 0.57 \text{ lm/W} \) with \( \text{CIE}x = 0.34 \) and \( \text{CIE}y = 0.58 \) is achieved.

Blue SrS:Cu,Cl devices are prepared by evaporating a CuCl2 layer onto the SrS followed by a rapid thermal anneal in Ar at a temperature of 700°C. For this device \( L_{40} = 7.8 \text{ cd/m}^2 \) and \( E_{40} = 0.074 \text{ lm/W} \) with \( \text{CIE}x = 0.16 \) and \( \text{CIE}y = 0.275 \) are obtained. Blue SrS:Cu,Ag,Cl phosphors are fabricated by depositing a layer of AgCl in addition to the CuCl2 layer. The SrS:Cu,Ag,Cl CIE color coordinates are closer to those of an ideal blue, \( \text{CIE}x = 0.19 \), \( \text{CIE}y = 0.14 \), but the luminance decreases to \( L_{40} = 1.7 \text{ cd/m}^2 \).

A significant improvement in luminance, efficiency, and color of the blue-emitting devices is obtained when a layer of GaF3 is deposited in addition to the CuCl2 and AgCl layers. Luminance and efficiency results for SrS:Cu,Cl,Ga and SrS:Cu,Ag,Cl,Ga are shown in Figs. 4 and 5, respectively. Brightness and efficiency for SrS:Cu,Cl,Ga are \( L_{40} = 17.4 \text{ cd/m}^2 \) and \( E_{40} = 0.14 \text{ lm/W} \) at \( \text{CIE}x = 0.157 \) and \( \text{CIE}y = 0.248 \). The values obtained for SrS:Cu,Ag,Cl,Ga are \( L_{40} = 8.3 \text{ cd/m}^2 \) and \( E_{40} = 0.072 \text{ lm/W} \) at \( \text{CIE}x = 0.165 \) and \( \text{CIE}y = 0.166 \).

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References