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Title(English)	Materials exploration of amorphous oxide semiconductor thin film phosphors and application to light-emitting devices
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Light-emitting diodes (LEDs) are essential component for opto-electronic devices, however the present LEDs have issues to be solved, such as high-temperature growth process & expensive single-crystal substrates for inorganic emission layers and chemical instability for organic ones. To overcome these issues, I focused on wide-bandgap amorphous oxide semiconductor (AOS). A representative AOS of amorphous IGZO (*a*-IGZO) has high electron mobility (~10 cm^2/Vs) and low defect density even when deposited at room temperature (RT), leading to high-mobility thin-film transistor. These characteristics are suitable for thin-film phosphor with low-temperature process, but *a*-IGZO doped with Eu ion showed low luminous efficiency because of auger-recombination by high carrier density (~10^{15} cm^{-3}).

In this thesis, amorphous gallium oxide (*a*-GO) with ultra-wide bandgap (4.12 eV) is selected for the host material for AOS-based thin-film phosphor, because it has much lower carrier density ~2×10^{14} cm^{-3} as well as high electron mobility ~8 cm^2/Vs. I developed AOS-based phosphor thin films and LEDs using rare earth (*RE*) and transition metal (*TM*) ions doped *a*-GO (*a*-GO:RE_x, *a*-GO:TM_x) on glass substrate with low temperature process. Main results are summarized as follows.

1. Fabrication and PL property characterization of *a*-GO:Eu_x

I fabricated *a*-GO:Eu_x films at RT on silica-glass substrates by pulsed laser deposition and investigated the effect of dopant concentration (x), deposition condition (oxygen pressure (P_{O2})), and post-annealing (annealing temperature (T_a)) under O_2 on PL property. PL intensity of *a*-GO:Eu_x films was increased with increase of x, while it was deteriorated when a certain threshold x of 0.8 was exceeded because of concentration quenching. By controlling P_{O2} during deposition, it was found that maximum PL intensity was only obtained at optimized P_{O2} of 11 Pa, where PL intensity deteriorated in the lower P_{O2} and higher P_{O2} region because of oxygen deficiency formation and introduction of weakly-bonded oxygen, respectively. Additionally, T_a was also an important factor to improve PL intensity, where it increased with increase of T_a up to 600 ºC because the annealing effectively remove the weakly bonded oxygen and reduce defects via structural relaxation. On the other hand, the deterioration of PL intensity was observed at T_a = 700 ºC because of crystallization with grain boundary defects. I realized *a*-GO:Eu_x films with clearly visible red-light emission from Eu^{3+} by optimizing x, P_{O2}, and T_a due to the reduction of non-radiative recombination centers.

2. Effect of *RE* (*RE*=Pr, Sm, Tb, Eu, and Dy) dopants on PL properties of *a*-GO:RE_x

I investigated PL properties of *a*-GO:RE_x films doped with various kinds of *RE* ions (*RE*=Pr, Sm, Tb, Eu, and Dy). The RT-deposited *a*-GO:RE_x films emited various colors; i.e., blue & red from Pr^{3+}, red light from Sm^{3+} and Eu^{3+}, green from Tb^{3+}, and blue & yellow from Dy^{3+}. Especially, Strong light emissions visible by human eyes were observed from the *a*-GO:Pr_x, *a*-GO:Eu_x, and *a*-GO:Tb_x films. From these results, I demonstrated the multi-color light-emission in *a*-GO:RE_x films prepared with low temperature process.

3. Fabrication and PL property characterization of *a*-GO:*TM*$_x$

I first screened *TM* ions as active emission centers using β-Ga_2O_3 polycrystalline bulks doped with *TM* = Cu, Mn, Cr and found that only $(Ga_{1-x}Cr_x)_2O_3$ ceramics emit red light originating from Cr^{3+} ion. The most intense red PL from *a*-GO:Cr_x films deposited at RT was achieved by optimizing x = 0.001 and P_{O2} = 5 Pa. Post-annealing at T_a = 400 ^oC further improved PL intensity. Crystallization started being observed at T_a = 500 ^oC, and further improved PL intensity due to crystallization. The effect of T_a in the *a*-GO:Cr_x films is opposite to that of *a*-GO:*RE*$_x$ films. This result originate from difference in their crystal phaess; i.e., $(Ga,RE)_2O_3$ do not form stable single phases while $(Ga,Cr)_2O_3$ do and it is already-known as a good crystalline phosphor. The diffrences in the stable solid-solution phases are explained by the ion radii of the constituent elements.

4. Demonstration of LEDs using *a*-GO:*RE*$_x$ thin films with low-temperature process

I fabricated a bottom-emission type LED structure with ITO/*a*-GO:*RE*/*α*-NPD/*a*-MoO_x/Ag using *a*-GO:*RE*$_x$ emission layers. Multiple color emission was achieved using *a*-GO:*RE*$_x$ with *RE* = Eu, Pr, and Tb. The current efficiency of *a*-GO:Eu (0.22 cd/A) was 2 orders of magnitude higher than that of *a*-GO:Tb and *a*-GO:Pr (~10^{-3} cd/A); the difference is plausibly explained by the different electronic structure observed by RPES and HAXPES. i.e., Eu^{3+} 4*f* states are buried in the valence band of *a*-GO host material, so it seems impossible to directly inject holes from the VBM to the Eu^{3+} 4f states. Hence *a*-GO:Eu would show the red-emission by impact ionization, resulting high emission efficiency, while *a*-GO:Tb and *a*-GO:Pr show light emission by electron-hole recombination.