

論文 / 著書情報
Article / Book Information

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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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Category(English)	Doctoral Thesis
種別(和文)	論文要旨
Type(English)	Summary

論文要旨

THESIS SUMMARY

系・コース : Department of, Graduate major in	材料 材料	系 コース	申請学位 (専攻分野) : Academic Degree Requested	博士 (工学) Doctor of Engineering
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要旨 (英文 800 語程度)

Thesis Summary (approx.800 English Words)

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 cm²/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 ($\sim 10^{15}$ 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 $\sim 2 \times 10^{14}$ cm⁻³ as well as high electron mobility ~ 8 cm²/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 O₂ 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 °C 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 °C because of crystallization with grain boundary defects. I realized *a*-GO:Eu_x films with clearly visible red-light emission from Eu³⁺ 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 emitted various colors; i.e., blue & red from Pr³⁺, red light from Sm³⁺ and Eu³⁺, green from Tb³⁺, and blue & yellow from Dy³⁺. 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-Ga₂O₃ polycrystalline bulks doped with *TM* = Cu, Mn, Cr and found that only (Ga_{1-x}Cr_x)₂O₃ ceramics emit red light originating from Cr³⁺ 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 °C further improved PL intensity. Crystallization started being observed at *T*_a = 500 °C, 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)₂O₃ do not form stable single phases while (Ga,Cr)₂O₃ do and it is already-known as a good crystalline phosphor. The differences 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/*a*-NPD/*a*-MoO₃/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 ($\sim 10^{-3}$ cd/A); the difference is plausibly explained by the different electronic structure observed by RPES and HAXPES. i.e., Eu³⁺ 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 Eu³⁺ 4*f* 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.

備考 : 論文要旨は、和文 2000 字と英文 300 語を 1 部ずつ提出するか、もしくは英文 800 語を 1 部提出してください。

Note : Thesis Summary should be submitted in either a copy of 2000 Japanese Characters and 300 Words (English) or 1copy of 800 Words (English).

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