Photoluminescence and photoconduction characteristics of crystalline quasi two-dimensional γ-alumina grown by graphene-assisted Atomic Layer Deposition[1]

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INTRODUCTION

As a transition phase nanocrystal form of alumina, γ-alumina retains its technological advantages of high heat conduction, low production cost, excellent radiation, and corrosion resistance, as well as advanced catalytic, and opto-electronic properties.[2] Our group has developed a two-step synthesis method by utilizing graphene assisted atomic layer deposition (ALD) to fabricate the free-standing quasi-2D crystalline γ-Al_2O_3.[3] Continuing with our investigation of the unique properties of this material, this study concentrates on the optoelectronic properties.

The temperature dependent charge transport mechanisms and wavelength dependent photoluminescence studies of the material indicates a small polaron transport. The polaronic charge transport with two activation energies in high and low temperature regimes points to participation of the bound polarons in the charge transport at temperature above 303K.

METHOD

Thin films of amorphous alumina are first deposited on the graphene foam substrate by ALD using precursors $(Al(Ch_3)_3)$ and H_2O. The sample is then air annealed at $800°C$ during which the alumina undergoes crystallization, and the graphene backbone is removed by reacting with oxygen. The process yields a white scaffold that consists of 2D nanosheets of γ-alumina with the nominal thickness on the order of 10nm.[2]

The as-synthesized material was tested with a Horiba Jobin microPL system to investigate the photoluminescence (PL) properties. Temperature dependent photocurrent measurement was also done by Keithely to study the transport mechanism in the 2D nanosheet network.

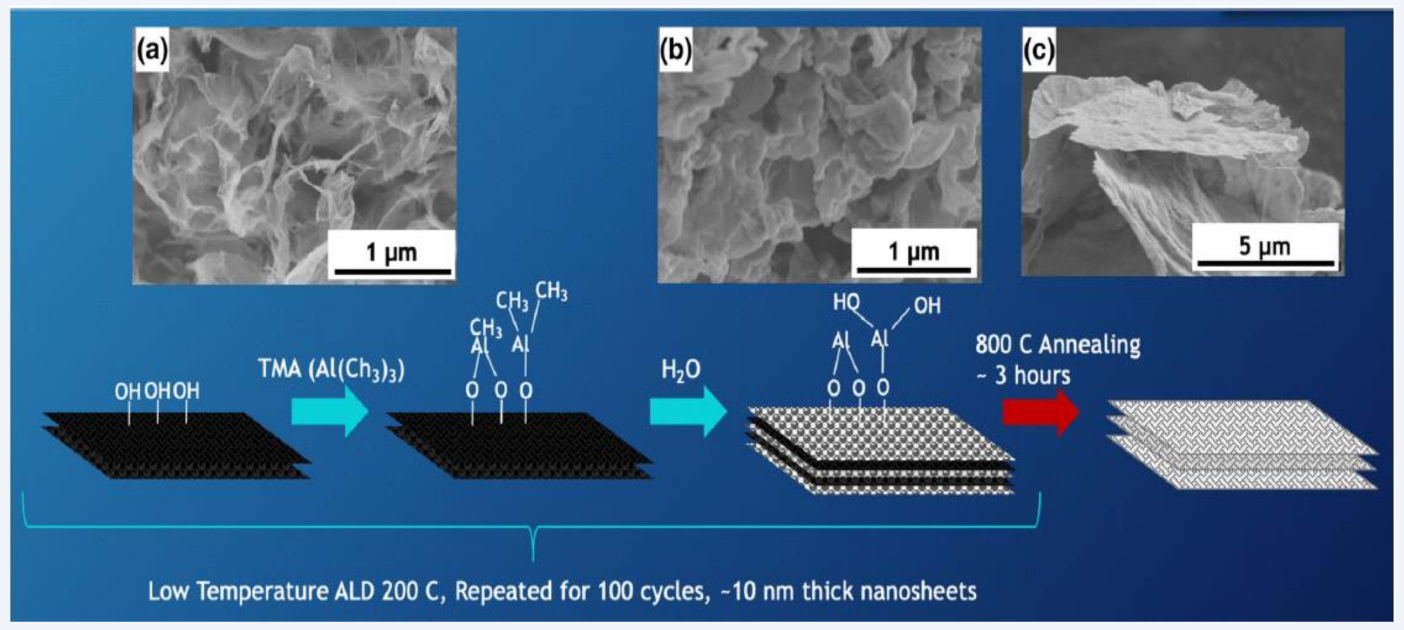

Figure 1. Fabrication schematic of the two-step fabrication process. (a) a SEM image of graphene foam showing the intertwined graphene sheets network, (b) a SEM image of the amorphous alumina deposited on the graphene foam by ALD, and (c) a SEM image of free-standing quasi-2D γ-alumina flakes after thermal annealing and graphene removal.[2]

RESULTS

PL & Temp Dependent Photoconductivity

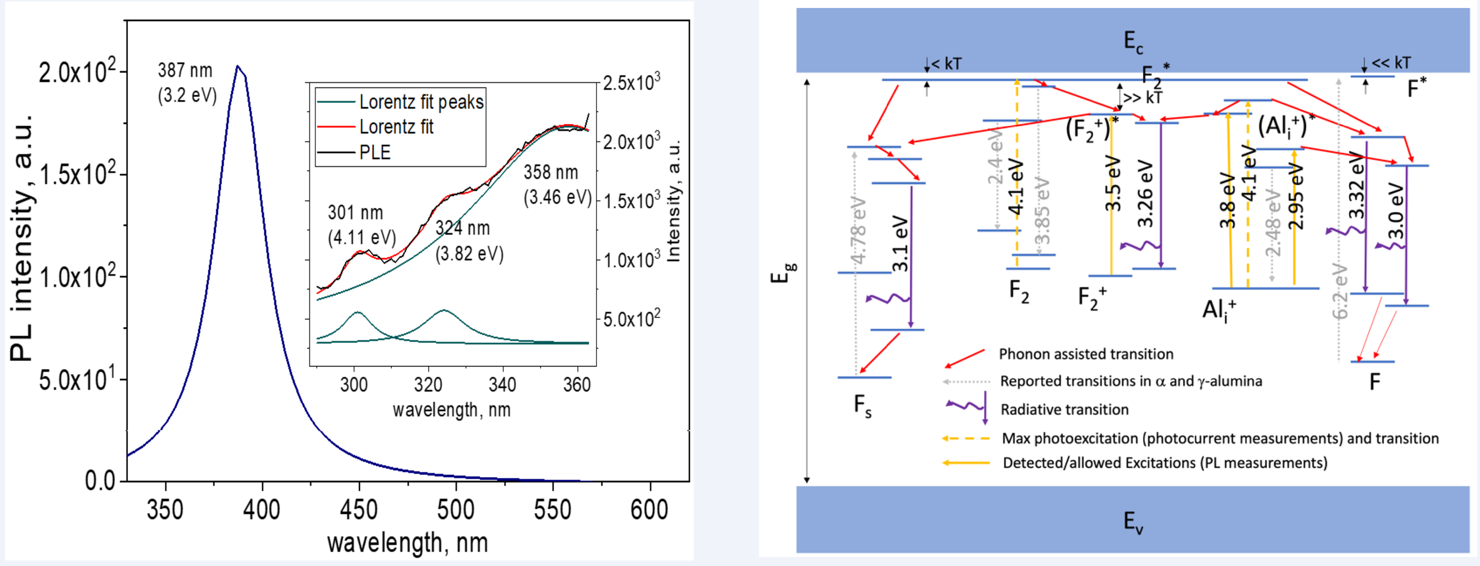

Figure 2. a) Room temperature PL spectra of quasi-2D γ-Alumina and PLE(inset) showing the states contributing to emission at 387nm. b) schematic of excitation and emission in quasi-2D γ-Alumina

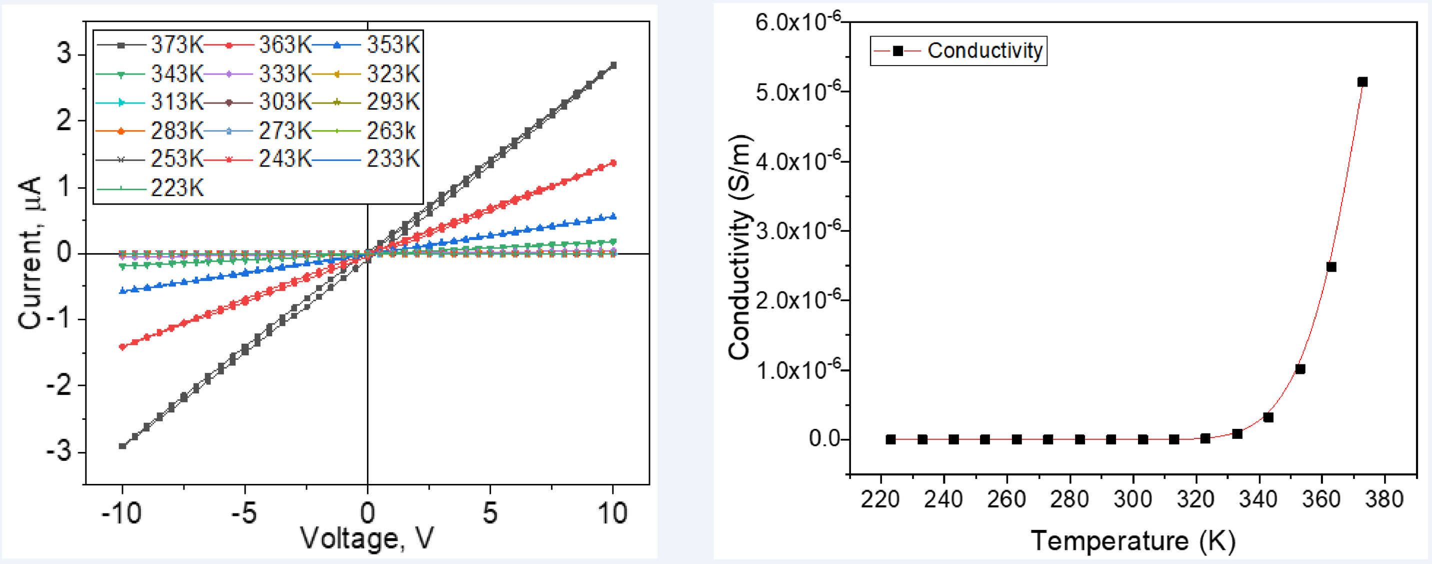

Figure 3. a) Temperature dependent I-V characteristics, b) temperature dependent photo-conductivity of quasi-2D γ-Alumina.

Charge Transport Models

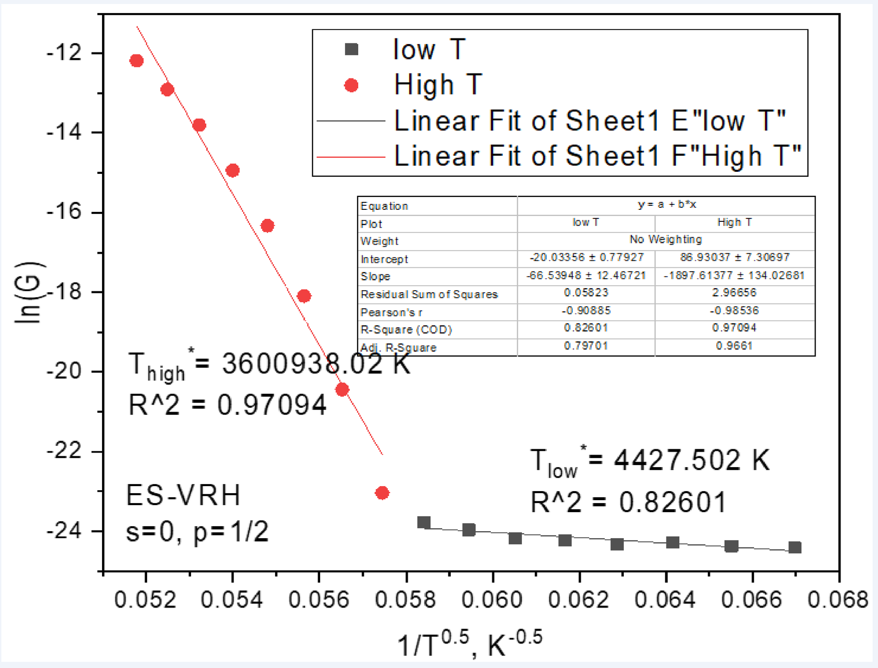

Figure 4. Fitting results of Efros and Shklovskii-Variable Range Hopping (ES-VRH) model

In VRH model, the charge transport occurs by electrons tunneling/hopping between localized states, and an increase in the activation energy at the Neel temperature is expected for the hopping transport of a small polaron. The coulomb interactions between localized states is considered in the ES-VRH model.

ES-VRH model:

$$\sigma(T) = \sigma_0 T^{-s} e^{-\left(\frac{T_0}{T}\right)^p}$$

$E_a = 1.42 \pm 0.10\ eV$ (high T) and $E_a = 0.050 \pm 0.0094\ eV$ (low T)

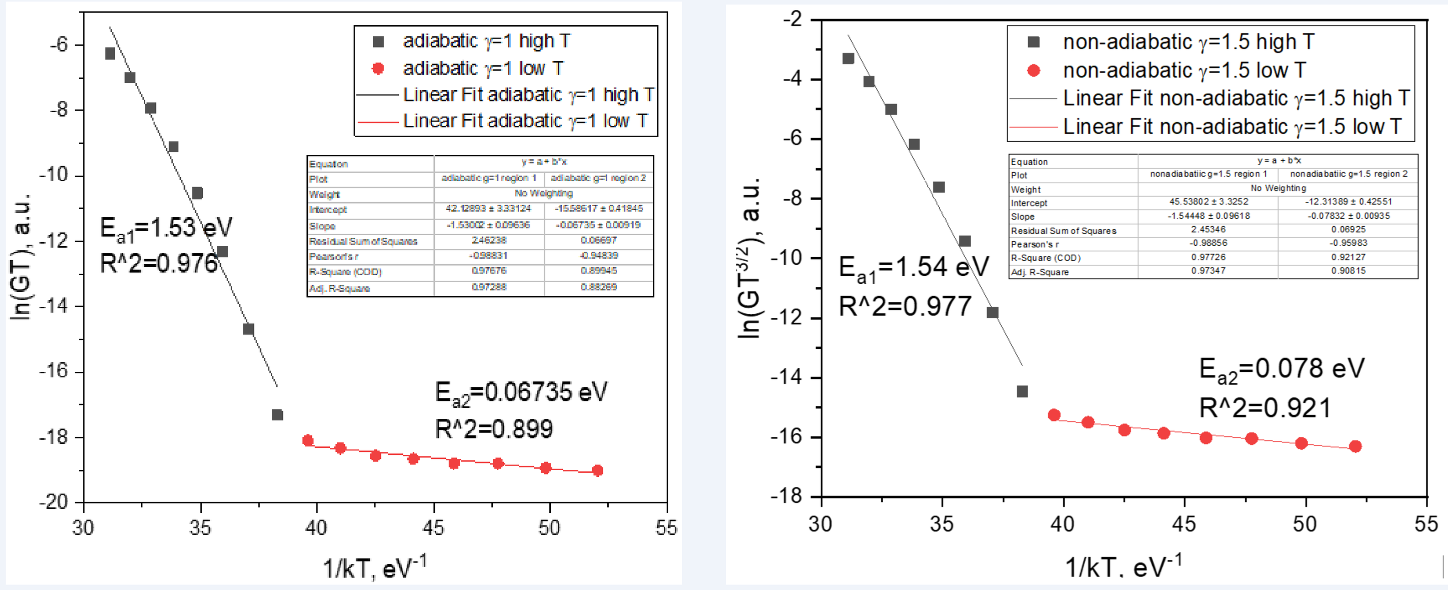

Figure 5. Fitting results of a) Smoothed Particle Hydrodynamics (SPH) adiabatic, b) SPH non-adiabatic model.

In small polaron hopping model, the charge transport is facilitated by hopping or quantum tunnelling between localized states.

SPH model:

$$G = G_0 T^{-\gamma} e^{-\frac{E_a}{KT}}$$

SPH adiabatic: $\gamma = 1$, $E_a = 1.53 \pm 0.10\ eV$ (high T) and $E_a = 0.067 \pm 0.0092\ eV$ (low T)

SPH non-adiabatic: $\gamma = 1.5$, $E_a = 1.54 \pm 0.10\ eV$ (high T) and $E_a = 0.078 \pm 0.0094\ eV$ (low T)

CONCLUSIONS

In conclusion, the PL spectra indicates that this quasi-2D γ-alumina has a strong emission peak at 387 nm, and this emission is contributed by 4.11, 3.82, and 3.46 eV excitations. The temperature dependent photoconductivity result suggests different charge transport mechanisms at the high and low temperature regimes. By fitting the experimental data with charge transport models, we conclude that the low temperature conductivity is controlled by free polarons with a small activation energy of 60-70meV. The increase of the conductivity in the high temperature region could be explained by the participation of the bound polarons in the transport with an activation energy of ~1.5eV.

REFERENCES

1. E. Kheirandish, Y. Lin and N. Kouklin, Photoluminescence and photoconduction characteristics of crystalline quasi two-dimensional **γ**-alumina grown by graphene-assisted Atomic Layer Deposition, in preparation (2021).
2. E. Kheirandish and N. Kouklin, Light emission study of free-standing quasi-2D-**γ**-alumina grown by graphene-assisted Atomic Layer Deposition, *J. Electron. Mater.*(2021).
3. E. Kheirandish, M. Schofield, M. Gajdardziska-Josifovska and N. Kouklin, Quasi-2D crystalline **γ**-alumina grown by graphene-assisted Atomic Layer Deposition, *Adv. Mater. Interfaces*, 7, 2000561 (2020).