

Research Article

Island size distribution study for rubrene thin films on muscovite mica and SiO_2 using scaling theory

Kamila Rehman^{a,b}, Aaliya Rehman^{a,*}, Shaimaa M. Abdalbaqi^b, Helmut Sitter^b

^a*Department of Physics, Govt. M.A.O. Graduate College, Lahore 54000, Pakistan*

^b*Institute of Semiconductor Physics, University of Linz, 4040 Linz, Austria*

Abstract

For the application of various theories to find a growth model for rubrene, it is essential to analyze the nucleation process during initial stages of growth. Here we present a study of rubrene deposited by Hot Wall Epitaxy on mica and SiO_2 using different substrate temperatures and calculating the island size distribution for each temperature. In order to find a growth model for rubrene we apply scaling theory during these initial stages of growth.

Keywords:

Nucleation, Scaling theory.

1. Introduction

Impressive new developments using organic materials has resulted in organic light emitting diodes (OLEDs) [1–4] as well as flexible devices [5] with the future being of organic solar cells [6, 7] and organic field effect transistors (OFETs) [8, 9]. However, the efficiency of any such device is compromised by the low charge carrier mobility. There needs to be intensive research into the charge transport system in crystalline form of organic materials [10–18] as well as the defining of models governing their growth. The attempt to predict the conditions of optimum growth of rubrene thin films on muscovite mica requires finding of a growth model for rubrene deposition. For this it is necessary to understand the nucleation process during the initial stages of island formation followed by application of different existing theories to the results. Here the scaling theory [19, 20], first shown by Venables et. al. [21], which has been successfully applied for formulating a growth model for pentacene [22] is selected and applied to rubrene during the initial stages of its growth.

2. Materials and Methods

An organic material called rubrene was acquired from Aldrich and subjected to thermal sublimation for additional purifica-

tion. The substance was 98% pure. After being transferred into the Hot Wall Epitaxy device, Rubrene was put within a quartz tube. Acquired from Segliwa GMBH, $15 \times 15\text{mm}^2$ Muscovite Mica substrates were manually split in the air prior to being put into the HWE vacuum chamber. Once a vacuum of 10-6 mbar was reached, the substrate deposition temperature was preheated for 15 minutes. With this in situ heat treatment, all materials that have adsorbed onto the substrate surface are completely eliminated. Rubrene was then applied on mica (001) and freshly cleaned SiO_2 wafer (SiO_2 coated Si wafer) substrates at a vacuum of 10-6 mbar. For mica, substrate temperatures of 90°C and 120°C were used, while for SiO_2 , substrate temperatures of 120°C. In addition to the source temperature, the wall temperature was always kept at 180°C. The growth durations of the samples were 1, 2, 4, and 8 minutes. Making use of atomic force microscopy was utilized to perform morphological examinations on regions with dimensions of $10 \times 10\mu\text{m}^2$ and $50 \times 50\mu\text{m}^2$, using SiC tip. (AFM) images of the deposited organic thin films with a Digital Instruments Dimension 3100 microscope in the tapping mode. Fig. 1 shows these AFM pictures of rubrene applied to SiO_2 and muscovite mica surfaces. This series of AFM scans was subjected to the analysis that follows. One way to determine the island density would be to count the grains per $10 \times 10\mu\text{m}^2$. After a large number of cross sections were analyzed, the average height of these islands was found. The distribution of island heights was then computed using this information.

*Corresponding Author:

aaliya.rehman@gmail.com (Aaliya Rehman)

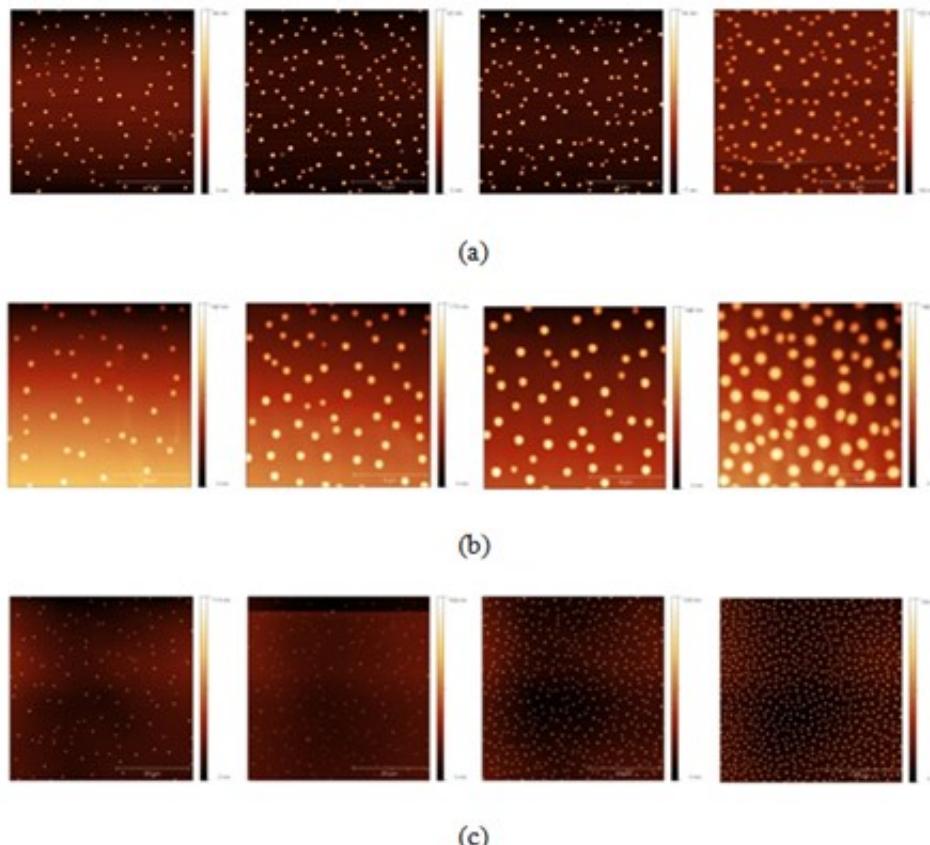


Figure 1. $10 \times 10 \mu\text{m}^2$ AFM images of rubrene grown on mica for 1, 2, 4 and 8 minutes (left to right) at (a) $T_{\text{sub}} = 90^\circ\text{C}$ and (b) $T_{\text{sub}} = 120^\circ\text{C}$ and $50 \times 50 \mu\text{m}^2$ AFM images of (c) SiO_2 for 0.5, 1, 2 and 4 minutes (left to right) at $T_{\text{sub}} = 120^\circ\text{C}$.

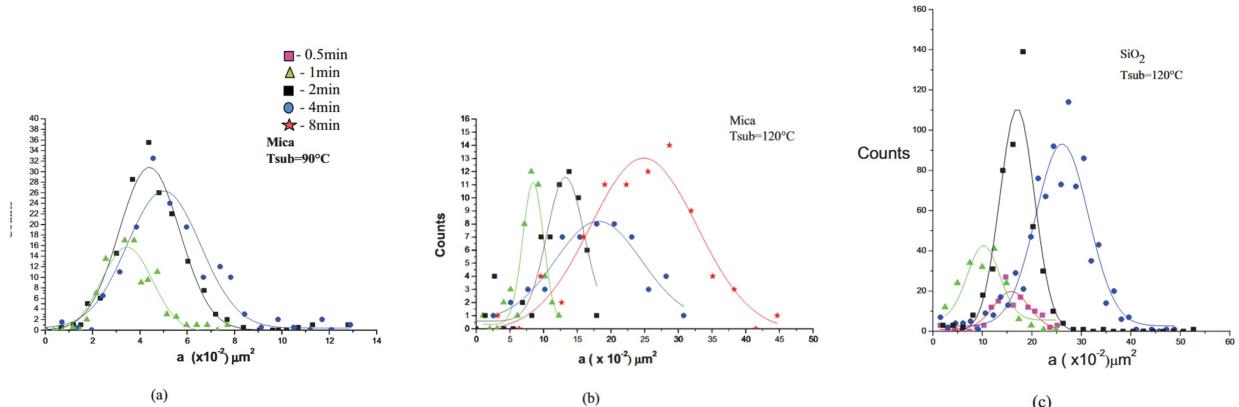


Figure 2. Island size distribution density for rubrene coverages of a few monolayers on mica at (a) $T_{\text{sub}} = 90^\circ\text{C}$, (b) $T_{\text{sub}} = 120^\circ\text{C}$ and (c) SiO_2 at $T_{\text{sub}} = 120^\circ\text{C}$.

3. Results and Discussion

The growth rate in the two series of samples was found by plotting the measured film thickness versus the time of growth and which gave the corresponding growth rate in monolayers (ML) per minute. The coverages for all three series of samples calculated from their growth rates ranged from less than a ML to around 4 ML's. Hence the scaling theory which holds for the first few monolayers of growth, was applied to these samples.

The island size distribution density is calculated from the AFM micrograph's frequency count analysis and is plotted for the first few monolayers of rubrene growth in Fig. 2

We have used the scaling law here and apply it to the island size distribution density and observe that all the curves collapse into $f_i(u)$ where $f_i(u)$ is the scaling function and i is the critical nucleus size. A critical island size of 2 for $T_{\text{sub}} = 90^\circ\text{C}$ and about 30 for all other cases was obtained by the application of the rate equation and scaling theory. However, the question

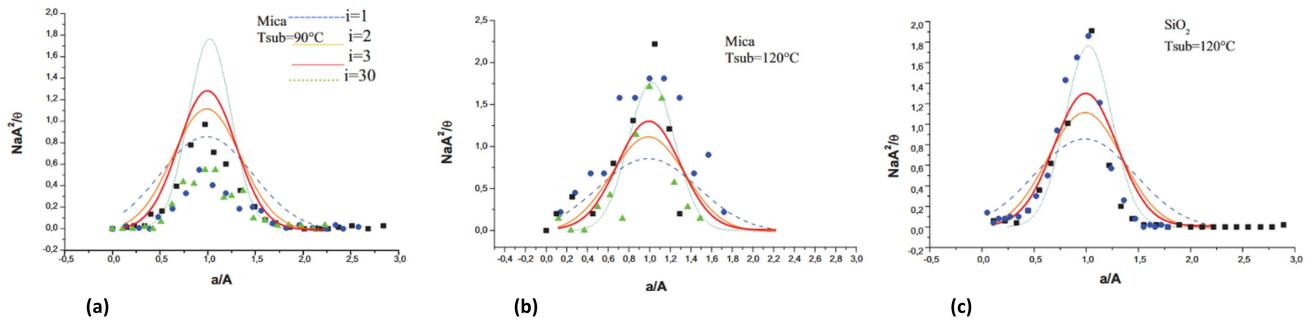


Figure 3. Scaled Island size distribution of rubrene grown on mica (a) $T_{sub} = 90^\circ\text{C}$, (b) $T_{sub} = 120^\circ\text{C}$ and (c) SiO_2 at $T_{sub} = 120^\circ\text{C}$.

remains whether this theory can explain the growth mechanism of rubrene if island heights reach many monolayers

4. Conclusion

We have presented the study of the nucleation stage of rubrene layers which were grown by Hot Wall Epitaxy (HWE) on two substrates at different temperatures. The plots show an agreement between model and data in cases of fig. 3b and 3c where $f_i(u) = 30$. However, this seems too high a value for critical island size for rubrene. Although we restricted our investigations to the first few monolayers of rubrene, it is important to mention that the island heights were much more than that. Hence, we conclude that the scaling theory cannot be applied to the investigation of materials which have island heights exceeding the first few monolayers.

References

- [1] C. W. Tang, S. A. VanSlyke, Organic electroluminescent diodes, *Applied physics letters* 51 (12) (1987) 913–915.
- [2] M. Muccini, A bright future for organic field-effect transistors, *Nature materials* 5 (8) (2006) 605–613.
- [3] S. Hirata, Y. Sakai, K. Masui, H. Tanaka, S. Y. Lee, H. Nomura, N. Nakamura, M. Yasumatsu, H. Nakanotani, Q. Zhang, et al., Highly efficient blue electroluminescence based on thermally activated delayed fluorescence, *Nature materials* 14 (3) (2015) 330–336.
- [4] S. Scholz, D. Kondakov, B. Lussem, K. Leo, Degradation mechanisms and reactions in organic light-emitting devices, *Chemical reviews* 115 (16) (2015) 8449–8503.
- [5] T. Someya, Z. Bao, G. G. Malliaras, The rise of plastic bioelectronics, *Nature* 540 (7633) (2016) 379–385.
- [6] H. Sirringhaus, 25th anniversary article: organic field-effect transistors: the path beyond amorphous silicon, *Advanced materials* 26 (9) (2014) 1319–1335.
- [7] B. Lussem, C.-M. Keum, D. Kasemann, B. Naab, Z. Bao, K. Leo, Doped organic transistors, *Chemical reviews* 116 (22) (2016) 13714–13751.
- [8] A. J. Heeger, 25th anniversary article: bulk heterojunction solar cells: understanding the mechanism of operation, *Advanced materials* 26 (1) (2014) 10–28.
- [9] R. A. Street, Electronic structure and properties of organic bulk-heterojunction interfaces, *Advanced Materials* 28 (20) (2016) 3814–3830.
- [10] S. R. Forrest, The path to ubiquitous and low-cost organic electronic appliances on plastic, *nature* 428 (6986) (2004) 911–918.
- [11] R. W. I. de Boer, M. Gershenson, A. Morpurgo, V. Podzorov, Organic single-crystal field-effect transistors, *physica status solidi (a)* 201 (6) (2004) 1302–1331.
- [12] G. Malliaras, R. Friend, An organic electronics primer, *Physics Today* 58 (5) (2005) 53–58.
- [13] H. Najafov, B. Lee, Q. Zhou, L. C. Feldman, Podzorov, V, Observation of long-range exciton diffusion in highly ordered organic semiconductors, *Nature materials* 9 (11) (2010) 938–943.
- [14] I. G. Lezama, M. Nakano, N. A. Minder, Z. Chen, F. V. Di Girolamo, A. Facchetti, A. F. Morpurgo, Single-crystal organic charge-transfer interfaces probed using schottky-gated heterostructures, *Nature materials* 11 (9) (2012) 788–794.
- [15] S. Z. Bisri, K. Sawabe, M. Imakawa, K. Maruyama, T. Yamao, S. Hotta, Y. Iwasa, T. Takenobu, Organic single-crystal light-emitting transistor coupling with optical feedback resonators, *Scientific Reports* 2 (1) (2012) 985.
- [16] S. Illig, A. S. Eggeman, A. Troisi, L. Jiang, C. Warwick, M. Nikolk, G. Schweicher, S. G. Yeates, Y. Henri Geerts, J. E. Anthony, et al., Reducing dynamic disorder in small-molecule organic semiconductors by suppressing large-amplitude thermal motions, *Nature communications* 7 (1) (2016) 10736.
- [17] M. Kikuchi, K. Takagi, H. Naito, M. Hiramoto, Single crystal organic photovoltaic cells using lateral electron transport, *Organic Electronics* 41 (2017) 118–121.
- [18] X. Ren, M. J. Bruzek, D. A. Hanifi, A. Schulzenberg, Y. Wu, C.-H. Kim, Z. Zhang, J. E. Johns, A. Salleo, S. Fratini, et al., Negative isotope effect on field-effect hole transport in fully substituted 13c-rubrene, *Advanced Electronic Materials* 3 (4) (2017) 1700018.
- [19] R. Ruiz, B. Nickel, N. Koch, L. C. Feldman, R. F. Haglund Jr, A. Kahn, F. Family, G. Scsoles, Dynamic scaling, island size distribution, and morphology in the aggregation regime of submonolayer pentacene films, *Physical review letters* 91 (13) (2003) 136102.
- [20] J. G. Amar, F. Family, Critical cluster size: Island morphology and size distribution in submonolayer epitaxial growth, *Physical Review Letters* 74 (11) (1995) 2066.
- [21] J. Venables, G. Spiller, M. Hanbucken, Nucleation and growth of thin films, *Reports on progress in physics* 47 (4) (1984) 399.
- [22] B. Stadlober, U. Haas, H. Maresch, A. Haase, Growth model of pentacene on inorganic and organic dielectrics based on scaling and rate-equation theory, *Physical Review B* 74 (16) (2006) 165302.