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REVIEW

of the doctoral dissertation of Ameneh Mikaeeli, M.Sc.

entitled

"Thermal and Electrical Properties of Nanolayer Polymers"

The dissertation was prepared within a Double Degree Ph.D. programme at Nicolaus Copernicus University in Toruń, Faculty of Physics, Astronomy and Informatics, and Ruhr University Bochum, Faculty of Physics and Astronomy, under the supervision of Dr. hab. Michał Pawlak, Professor at NCU, and Prof. Dr. Andreas D. Wieck.

The doctoral dissertation concerns the electrical and thermal transport properties of conducting polymer thin films, with particular emphasis on PEDOT:PSS. The work combines investigations of electrical conductivity, thermal conductivity, transport anisotropy, substrate and thickness effects, as well as nanoscale charge-transport behaviour. An important part of the study is also devoted to the influence of a lateral electric field applied during spin coating on the formation and directional electrical properties of PEDOT:PSS thin films.

The subject of the dissertation is timely and scientifically relevant, as the understanding of coupled charge and heat transport in conducting polymers is important both from the point of view of fundamental condensed-matter physics and for potential applications in conducting-polymer electronics and thermoelectric technologies.

The dissertation is predominantly experimental in character, complemented by analytical transport modelling, numerical simulations of charge-transport models, and numerical fitting of thermal-transport data. The Candidate investigates PEDOT:PSS films of different thicknesses deposited on glass, Si-doped GaAs, and ITO-coated glass substrates, and also considers modified PEDOT:PSS formulations and processing conditions. Electrical properties are studied using temperature-dependent four-probe measurements in the Van der Pauw configuration and conductive atomic force microscopy (C-AFM), whereas thermal transport is analysed using photothermal infrared radiometry (PTR) and thermoreflectance (TR). The experimental data are additionally analysed using theoretical transport models and numerical fitting procedures. An important feature of the work is the combination of macroscopic and nanoscale measurements and the investigation of both in-plane and cross-plane transport in PEDOT:PSS thin-film systems.

The dissertation is organized into seven chapters. The introductory chapter presents the scientific background of conducting polymers and PEDOT:PSS, formulates the research motivation, hypothesis and objectives, and introduces the general framework for the subsequent discussion of electrical and thermal transport. Chapters 2 and 3 describe the materials, sample-preparation procedures and experimental techniques used for electrical and thermal characterization, while Chapter 4 presents the theoretical background and numerical simulations of the charge-transport models applied later in the analysis. The main experimental results are concentrated in Chapters 5–7.

Chapter 5 is devoted to electrical transport and examines the influence of substrate, film thickness and temperature. The Candidate compares transport descriptions based on variable-range hopping and fluctuation-induced tunnelling, analyses sheet resistance and, for films deposited on glass, surface roughness, discusses cross-plane transport, studies PEDOT:PSS on ITO-coated substrates and considers the influence of additives.

Chapter 6 addresses thermal transport. The Candidate determines and analyses in-plane and cross-plane thermal conductivity, discusses thermal anisotropy as a function of temperature, and attempts to correlate electrical and thermal transport using the Wiedemann–Franz framework and effective Lorenz numbers.

Chapter 7 concerns electrically induced in-plane anisotropy. A custom spin-coating system was developed to apply a lateral electric field during deposition, solvent evaporation and solidification while monitoring the current through the forming film in real time. The resulting films were subsequently characterized by macroscopic directional electrical measurements and C-AFM.

Overall, a major strength of the dissertation is the broad and complementary experimental approach applied to the study of electrical and thermal transport in PEDOT:PSS thin films. The Candidate combines temperature-dependent electrical measurements, investigations of substrate and thickness effects, cross-plane electrical characterization, nanoscale current mapping by C-AFM, and directional thermal-transport measurements. These experimental studies are further supported by transport-model analysis, numerical simulations and numerical fitting procedures. This combination of methods allows transport properties to be examined at different length scales and along different directions in PEDOT:PSS thin-film systems.

The systematic character of the electrical measurements is an important positive aspect of the work. The Candidate investigates F HC Solar films of different thicknesses and deposited on different substrates, and explicitly evaluates the reproducibility of electrical measurements for one representative series of films. For the F HC Solar samples analysed in Chapter 5, the fluctuation-induced tunnelling model provides a very good description of the temperature-dependent conductivity and is identified by the Candidate as the preferred transport model within the investigated range.

Another valuable element of the dissertation is the investigation of thermal transport. For a 480 nm PEDOT:PSS film deposited on glass, the Candidate determines and analyses both in-plane and cross-plane thermal conductivity over the investigated temperature range and evaluates the corresponding thermal anisotropy. The work also attempts to relate thermal and electrical transport using the Wiedemann–Franz framework and effective Lorenz numbers.

One of the clearest original elements of the dissertation is the development and implementation of a custom electric-field-assisted spin-coating setup. The system enables the application of a lateral electric field during spin coating and film solidification, together with real-time monitoring of the electrical current. This experimental approach makes it possible to follow the electrical evolution of the film during formation rather than only after deposition.

The field-assisted films exhibit a consistent directional trend in their in-plane resistance: for all investigated samples, the resistance measured parallel to the applied field is higher than that measured perpendicular to it. Although the magnitude of the anisotropy varies substantially between individual samples, the direction of the effect is systematic. This represents one of the clear experimental findings of the dissertation.

The real-time current monitoring and the subsequent C-AFM measurements further strengthen the experimental part of the work. The current-time measurements provide direct information on changes in the electrical response during film formation, while C-AFM reveals spatial variations in the local current distribution of the final films. These measurements complement the macroscopic transport data and provide additional information on the heterogeneity of charge transport at the nanoscale.

Despite these strengths, several aspects of the theoretical interpretation would benefit from further clarification.

In Chapter 4, transport is discussed mainly in terms of localization and thermally activated processes associated with structural disorder. This description is relevant to disordered conducting polymers, although the formulation could be more nuanced with respect to the relationship between structural disorder, electronic disorder and carrier localization. In particular, the discussion of Anderson localization would benefit from a clearer explanation of the physical origin of the disorder responsible for localization, rather than relating it mainly to the absence of long-range crystalline order.

A related point concerns the physical meaning of the parameter n in the variable-range hopping model. In the thesis, n is linked to the dimensionality of transport, but lower values of n are also associated with increasing anisotropy or structural ordering. I would therefore ask the Candidate to clarify how n should be interpreted in the present work. Does it represent the dimensionality of the hopping process in the conventional Mott sense, or is it treated as an effective fitting parameter describing the degree of anisotropy or structural organization?

The reproducibility test performed for 160 nm films on glass is a useful element of the work and shows good consistency for this particular sample series. Since the test concerns one film thickness and one substrate, the conclusions on reproducibility may be most appropriately related to this specific set of samples.

Since sheet resistance is expected to decrease with increasing film thickness, it would be useful to clarify to what extent the observed trend reflects this geometrical effect and to what extent it may indicate changes in the intrinsic transport properties of the films. In this context, resistivity or conductivity may provide a clearer basis for comparison.

The cross-plane electrical analysis would also benefit from some clarification of the notation and data treatment. In Section 3.5, the quantities used in Eq. 3.26 are described as electrical resistances, while the expression also contains an explicit thickness weighting. It would therefore be helpful to clarify more explicitly how these quantities are defined and how the PEDOT:PSS contribution was extracted from the measured PEDOT:PSS/GaAs:Si structure. A clearer distinction between resistance, resistivity and conductivity would make this part of the analysis easier to follow.

A question concerns the interpretation of the Wiedemann–Franz analysis. The relation introduced in the thesis refers to the electronic contribution to thermal conductivity, whereas the experimentally determined thermal conductivity represents the total heat transport in the film. I would therefore ask the Candidate to clarify what assumptions are made about the non-electronic contribution when the slope of thermal conductivity versus sigma T is used to determine an effective Lorenz number. In particular, how could a temperature-dependent phonon contribution affect the extracted value, especially in the in-plane direction, where the effective Lorenz number is approximately 60.6 L0?

The directional electrical anisotropy observed in Chapter 7 is a clear and interesting experimental result. The microscopic interpretation in terms of field-induced reorganization of the PEDOT:PSS microstructure is physically plausible and consistent with the discussion presented in the thesis. However, the electrical measurements themselves do not directly determine molecular or domain orientation. I would therefore ask the Candidate how the proposed structural mechanism could be experimentally verified. Which complementary structural or spectroscopic techniques would be most suitable to distinguish between the observed transport anisotropy and the underlying microscopic reorganization?

The preparation conditions used for electric-field-assisted spin coating would benefit from clarification. Section 7.6 states that a DC voltage of 30 V was applied, corresponding to an electric field of approximately 3 V/mm, whereas Section 7.7 refers to a constant lateral bias of 10 V during film fabrication. Could the Candidate clarify whether these values refer to different sample series or whether one of them requires correction? Since the applied field is an important experimental parameter, it would be useful to state unambiguously which preparation conditions correspond to the samples discussed later in Chapter 7.

The procedure used to derive cross-plane conductivity from the C-AFM maps would benefit from further clarification. The thesis defines the current used in the calculation as the sum of the currents recorded over all pixels within the scanned area. Since C-AFM measures current locally at individual tip positions, I would ask the Candidate to explain the physical basis for combining the summed pixel current with the total scanned area in the conductivity calculation. How should the resulting conductivity values be interpreted, particularly given their dependence on the scan size for the same sample?

Several minor editorial and internal consistency issues are present in the thesis. These include the use of 1/n in the interpretation of the VRH exponent although the standard Mott VRH relation is formulated in terms of 1/(n+1), an incorrect equation reference in the discussion of the fluctuation-induced tunnelling model, an R^2 value of 3.87 reported in Table 5.5, and an inconsistency between the stated temperature range of 8–300 K and the 8–200 K range given for Figure 5.8. These points do not affect the overall conclusions of the dissertation, although greater consistency in notation and numerical reporting would improve the clarity of the work.

Despite the comments listed above, I assess the dissertation positively. The Candidate has carried out a broad and technically demanding experimental programme and has demonstrated familiarity with a wide range of electrical, thermal and nanoscale characterization methods. The work shows that she is able to prepare samples, develop and adapt experimental procedures, analyse temperature-dependent transport data, apply physical models and compare information obtained at different length scales.

The dissertation provides evidence that the Candidate possesses the required general theoretical knowledge in the field of conducting polymers and thin-film transport. Some parts of the theoretical discussion would benefit from more rigorous formulation, but this does not alter the overall positive assessment of her scientific competence. The scope and complexity of the experimental work also support the assessment that she is capable of conducting independent scientific research.

The most evident original elements of the work are the development of the electric-field-assisted spin-coating system, the real-time monitoring of the electrical evolution during film formation, and the observation of systematic directional in-plane anisotropy. The investigation of anisotropic thermal transport and the attempt to combine electrical, thermal and nanoscale transport information also constitute valuable elements of the dissertation.

The critical comments concern mainly the strength of selected microscopic interpretations, the justification of several quantitative procedures, and some inconsistencies in presentation. In my view, they do not undermine the central experimental results or the main original elements of the work.

Conclusion

In conclusion, I consider the doctoral dissertation of Ameneh Mikaeeli, M.Sc., entitled "Thermal and Electrical Properties of Nanolayer Polymers" to satisfy the requirements for a doctoral dissertation specified in Article 187 of the Act of 20 July 2018 – Law on Higher Education and Science, as well as the applicable regulations of Nicolaus Copernicus University in Toruń, including Resolution No. 38 of the Senate of Nicolaus Copernicus University in Toruń of 26 September 2023, as amended. Overall, the dissertation provides evidence that the Candidate possesses the theoretical background and research skills required at the doctoral level. The work includes original experimental contributions and presents a substantial body of experimental results on electrical and thermal transport in PEDOT:PSS thin films. I therefore assess the dissertation positively and recommend that Ameneh Mikaeeli, M.Sc., be admitted to the public defence of her doctoral dissertation.

Reviewer:
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