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Inhomogeneous transport property of Alq<sub>3</sub> thin films: Local order or phase separation?

## Thurzo, I. AMM. Méndez, H. A, Iacovita, C. A, Zahn, D.R.T. A

Institute of Physics, TU Chemnitz, D-09107 Chemnitz, Germany

## Abstract

Steady-state current-voltage (I-V) and impedance-voltage (Z-V) measurements were performed on in situ (UHV) prepared metal (Ag, Al)/Alq<sub>3</sub>/indium-tin oxide (ITO) devices after exposure to air. When increasing the positive bias on the top metal electrode to a relatively well-defined critical value, a transition from semiconducting to semi- or even insulating behavior of the contacted Alq<sub>3</sub> thin film is observed by means of I-V measurements. The final insulating state remains stable when applying negative bias to the Ag electrode. In the case of the Al electrode, there is a voltammetric current wave under a well-defined negative bias indicating a redox reaction of mobile ions at the Al electrode. The Z-V measurements reveal a peculiar feature of ac transport through the Alq<sub>3</sub> thin films, namely the equivalent series capacitance is equal to its parallel counterpart in the frequency range from 100 to 1 MHz and amounts to only a fraction (0.3-0.5) of the expected geometrical capacitance of the device. An equivalent electrical circuit has been developed, based on the existence of two parallel transport paths: an insulating (amorphous) Alq<sub>3</sub>-phase shunted by a semiconducting (semi-insulating) one, both running into the impedance of the back contact. The equivalent circuit model composed exclusively of frequency independent elements is useful for predicting the maximum frequency for retaining the full geometrical capacitance. Even though the model is capable of describing the bias dependence of the impedance correctly, it does not shine light on the nature of the (ordered) phase or domain responsible for the dielectric loss. The possibility of local order connected with dipole-dipole interaction in the metal/Alq<sub>3</sub> interface zone is discussed. In any case, the ordered portion of the organic material seems to form the huge interface dipole of about 1 eV with Ag or Al [M.A. Baldo, S.R. Forrest, Phys. Rev. B 64 (2001) 085201], the direction of the dipole promoting electron injection to Alq<sub>3</sub>. Then the semiconductor-to-insulator transition could be initiated by a damage of the interface dipole under a critical positive dc bias of the metal, preventing the flow of both dc and the real component of low-frequency ac current. The transition is not accompanied by any significant change in the impedance of the back contact common to both phases. © 2006 Elsevier B.V. All rights reserved.

## **Author Keywords**

Conduction; Dielectric loss; Inhomogeneity; Organic semiconductor