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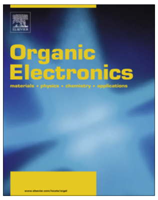

Charge trapping models of resistance switching in organic bistable devices with embedded nanoparticles

Francesco Santoni [a],*, Alessio Gagliardi [b], Matthias Auf der Maur [a], Aldo Di Carlo [a]

[a] *Department of Electronic Engineering, University of Rome "Tor Vergata", 00133 Rome, Italy*
[b] *Technische Universität München, Electrical Eng. and Information Tech., Arcisstr. 21, 80333 München, Germany*

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ABSTRACT

We discuss three different models of switching between the high conductivity and low conductivity state in organic bistable devices (OBD) with embedded nanoparticles. All models assume the same basic mechanism: charge trapping and de-trapping in metal nanoparticles. We show trapped charges can both induce an increase or a reduction of the total current depending on device configurations. The influence of energy disorder is investigated.

1. Introduction

In recent years, organic devices with embedded metal nanoparticles (NPs) have received much attention for the possibility of being used as non-volatile memory devices [14,4]. Organic memories have been considered as a replacement to flash memories, especially when characteristics such as flexibility and low costs are required. Several organic materials and device architectures have been investigated. In particular, two architectures have been studied: single layer devices with metallic NPs dispersed in organic material matrices [3,5], and three layer devices where two organic regions are separated by a layer of deposited NPs surrounded by metal oxide [11,21]. All these devices show bistability and switch at voltage threshold V_{th} between ON and OFF currents, corresponding to low resistance state (LRS) and high resistance state (HRS) respectively. The state of the device can be reversibly changed by applying a negative or positive voltage beyond the threshold. The memory state is retained for hours or days.

Although encouraging experimental results, still the theory describing the electrical behavior of such devices is an open issue. Two main mechanisms have been recognised in literature to account for the resistive switching: (1) highly conductive and localized pathways (usually called *filaments*) can be formed inside the organic matrix through migration of metal atoms [8,20,15]; (2) nanoparticles can act as trap sites (or induce trap states in the organic around them), and bistability is an effect of the trapped charge [21,11,3]. Charges can be trapped and de-trapped controlling the applied voltage, thus switching the device. In this work we deal with the second mechanism from a theoretical and simulations point of view.

Experimentally different behaviors have been observed. In Refs. [5,23] it is proved that NPs are essential to OBDs: if control devices with the same structure, but without NPs, are fabricated, they show no bistability. When NPs are inserted in the organic material, devices are bistable and the ON current is the same as in control devices without NPs. A different behavior is reported for the OBDs with

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* Corresponding author.
E-mail address: francesco.santoni@uniroma2.it (F. Santoni).

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embedded NPs in Refs. [3,11], where the OFF current is the same as the current in not-bistable control devices without NPs. We will discuss later in more details these experimental evidences. The different behavior of various devices indicates that bistability can be due to different mechanisms.

We have identified three different effects that trap states can have on the electrical behavior:

1. Space charge (Section 3). The formation of a space charge potential limits the transport of charge carrier of the same sign, thus reducing the current.
2. Induced doping (Section 4). Trapped charges dope the organic, thus increasing the density of carrier of the opposite sign.
3. Shockley–Read–Hall recombination (Section 5). Trap states act as recombination centers. Once a fixed space charge is present around recombination centers, SRH recombination is suppressed, hence the current is raised.

These effects are always concurrent, and the global effect is determined by the prevailing one. In the following each mechanism will be described in detail. Using numerical simulations we will provide the evidence for the relevance of each mechanism on the observed bistable behavior of devices.

2. Simulation environment

All charging effects have been investigated using the simulation tool TiberCAD [12] and the complete model for charge injection and transport in organic semiconductors presented in Ref. [18]. We use an effective drift–diffusion model:

$$\begin{cases} \nabla \cdot (\varepsilon \nabla \varphi) = e(n - p - N_d^+ + N_a^- + N_t) = -\rho \\ \nabla \cdot j_n = \nabla \cdot (\mu_n n \nabla \phi_n) = -R + G \\ \nabla \cdot j_p = \nabla \cdot \left(\mu_p p \nabla \phi_p\right) = R - G \end{cases} \quad (1)$$

The first is the Poisson equation, where ε is the dielectric constant of the material, n and p are the electron and hole densities respectively, N_d^+ and N_a^- the densities of ionized donors and acceptors, and the term N_t indicates in general any other distribution of charged traps. μ_n and μ_p are electron and hole mobilities. There are two continuity equations for currents: j_n and j_p are electron and hole current densities, proportional to the gradients of their respective electro-chemical potentials ϕ_n and ϕ_p. R and G are recombination and generation terms.

The equation system can be solved on 1, 2 and 3-dimensional domains, using the finite elements method (FEM) [18], allowing simulations of many different device structures. Different regions can be defined and their physical properties – such as carrier densities, mobility models, recombination models – configured independently. One can also introduce in each region different distributions of fixed electron and hole traps, setting their density and energy distribution (e.g. single-level, constant, exponential or gaussian).

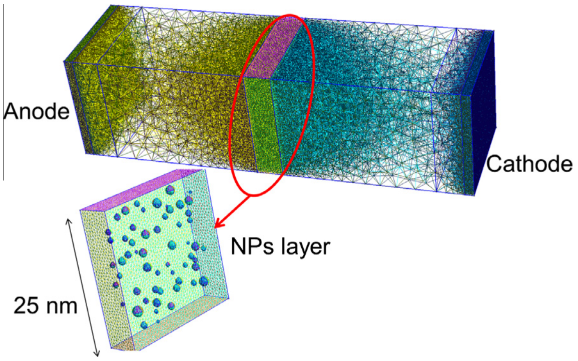

Fig. 1. Example of three-dimensional mesh for a device with embedded metal NPs.

We performed simulations of 2D and 3D structures, assuming metal NPs can be approximated as spheres. We developed scripts for automatic generation of 2D and 3D meshes with adjustable parameters for distribution and size of NPs (an example is shown in Fig. 1). Charge trapping models assume that charge carriers can be trapped inside NPs via tunneling [21,3]. Trap states can be also present in the organic surrounding the NPs, or in the metal oxide that in some devices is formed around NPs [11,21,3]. In the drift–diffusion model it makes no difference if charges are trapped directly inside the NPs or in the surrounding medium; in any case we model trapped charges introducing in the Poisson equation a suitable distribution of charge traps on the surface of NPs. Temperature is always set to 300 K.

Simulations are not time-dependent, the model describes only the steady state of the physical system, thus we cannot simulate dynamically the resistance switching, but we can only study the electrical behavior for fixed states of trap charging. We used our simulation environment to investigate the different models presented in the introduction. For every model we propose which device reported from literature suggests that physical mechanism in its bistability.

3. Space charge

3.1. Fixed charge distribution

3.1.1. Evidences for space charge limited currents

A strong evidence for space charge is found in the results of Ref. [5]. The reported device structure is Al/PMMA/Al. C_{60} NPs are dispersed in the whole volume of PMMA with different densities (0 wt%, 5 wt%, 10 wt%). The control device without NPs (0 wt%) is not bistable. Device with embedded NPs are bistable and the ON/OFF ratio raises as NPs density is increased. The current of the control device is a maximum. The ON state of the 5 wt% device is comparable with the control current. The ON state of the 10 wt% device is one order of magnitude lesser than the control current. In both devices the OFF state is sensibly lesser than the control current. These evidences suggests that NPs act as trapping sites, producing a space

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