Multiscale Network Models of Nano-Materials: Structure and Dynamics

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Abstract. We study two models of multi-scale structures and demonstrate how their magnetic and conduction properties depend on the structure. (a) in a 3D network structure with power-law inhomogeneous connectivity, representing a generalized "superlattice" of magnetic nano-particles, we simulate the magnetization reversal driven by the external magnetic field. The hysteresis curves depend on the network connectivity and clustering. (b) In a 2D graph with a given distribution of voids, representing the assembled structure of conducting nano-particle films on substrate we simulate the conduction processes with single-electron tunneling between nodes driven by the voltage difference across the network. We show how the non-linear current–voltage characteristics emerge above the threshold voltage in different random and regular nano-particle arrays.

INTRODUCTION

Systems of nano-size objects (quantum dots, nano-particles, bio-molecules) assembled into large-scale structures and functional devices are termed *nano-networks* to stress their structural irregularity, which is caused by the nonlinear assembly processes. Robust multiscale structures often emerge in the self-assembly processes [1]. On the other hand, the assembled large-scale structures of nano-objects posses a range of physical properties that can not be found at the level of an individual nano-object and in the bulk material. The properties of nano-network are thus interesting for new nano-technologies. Here we discuss single-electron tunneling through the nano-particle films [2] under the conditions of Coulomb blockade between nano-particles [3], which is interesting for the microelectronics [4]. Another example is the magnetic memory in models of complex semiconductor nanostructure with an effective exchange interaction [5].

In the network theory the topology of the underlying network may considerably affect the dynamic processes (spin dynamics, diffusion, electron tunnelings) taking part on the network [6–8], by involving different time scales and other collective dynamic phenomena. The objectives of the numerical modeling of such structures and processes, which is the subject of our work, is to reveal the structure–dynamics interdependences and to determine the range of parameters where the processes can be optimized.

In this report we present two multi-scale structures and certain properties of the driven dynamic processes on them. In particular, we study a scale-free network, that can be regarded as a model of generalized "superlattice" of magnetic nanostructures with an effective exchange interaction [5], and a planar graph with a given distribution of voids, representing the assembled nano-particle films on a substrate [2, 8]. Here we highlight

only two striking features of the physical processes on these network structures:

- Hysteresis loop properties in the spin-reversal dynamics on the scale-free network which is driven by the external magnetic field;
- Current–Voltage characteristics in the single-electron tunneling conduction on the planar graph which is driven by the voltage difference across the network.

In the following we first briefly describe the structures and their characteristics that are relevant for the respective processes and show the numerical results for the hysteresis and current–voltage curves. More detailed study can be found in the Refs. [1, 6, 8–12].

SPIN REVERSAL ON SCALE-FREE STRUCTURES

We grow the clustered scale-free graph by the algorithm introduced in Ref. [13]. An example is shown in Fig. 1, left. The relevant topological properties are the power-law inhomogeneity of the connectivity q_i (number of links attached to a node i), which varies as a power of node rank $q_i \sim r_i^{-\gamma}$, with $\gamma = 0.68$. Similarly, the inhomogeneity of the number of triangles attached to a node $\Delta_i \sim r_i^{-1}$. Each node of the graph represents a

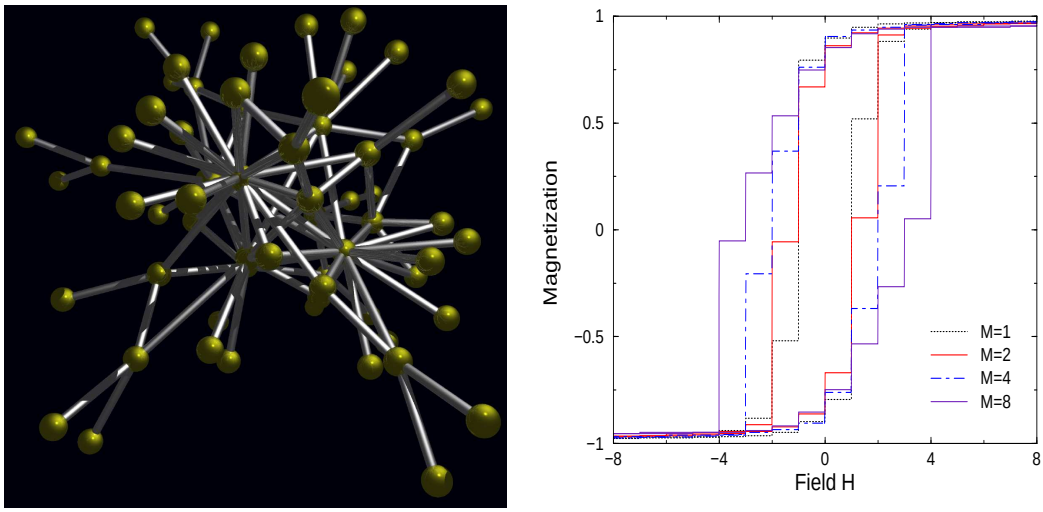

FIGURE 1. Model of the clustered scale-free structure of nano-particles in 3D rendering (left) and simulated hysteresis loops for different clustering parameter M, from [1], (right).

nano-particle with spin $S = \pm 1$. We assume *anti-ferromagnetic* exchange interaction along the network links, the geometry of which is exactly given by the network's adjacency matrix C_{ij}:

$$H \equiv -\sum_i h_i S_i = -J \sum_{i,j>i} C_{ij} S_i S_j - h_{ext} \sum_i S_i \, . \tag{1}$$

The system is driven by the external field h_{ext}, starting form large negative value and slow increase by integer values till a large positive value through the hysteresis loop (shown in Fig. 1, right). During the field ramps avalanches of the reversed spins occur,

in analogy to the classical domain-wall motion in disordered magnets. However, the concept of the domain walls is entirely lost in the network models. Similarly, avalanches exhibit a distribution with characteristic oscillations, related to the size of giant cluster and following-up subclusters on the graph [1]. Consequently, the hysteresis curve shows the critical jump of the magnetization at the coercive field H_c, which appears to *increase* with the size of the giant cluster. By increasing the average number of links per node, parameter M, the graph's giant cluster and at the same time the number of elementary triangles attached to a node increase, leading to increased spin *frustration*. However, the coercive field increases with M, in full contrast with the classical memory materials.

CONDUCTING NANO-PARTICLE FILMS

The model nano-particle films on a substrate is grown by the algorithm of cell aggregation, introduced in Ref. [10]. Alternatively, we use the empirical data [8] of nano-particle positions on substrate and make a network by connecting pairs of nano-particles at distances smaller than a *tunneling radius r*. An example of such structure is shown in Fig. 2, left. The characteristic features are planar geometry and r-dependent connectivity. Driven by the voltage difference the conduction with *single-electron tunneling* through

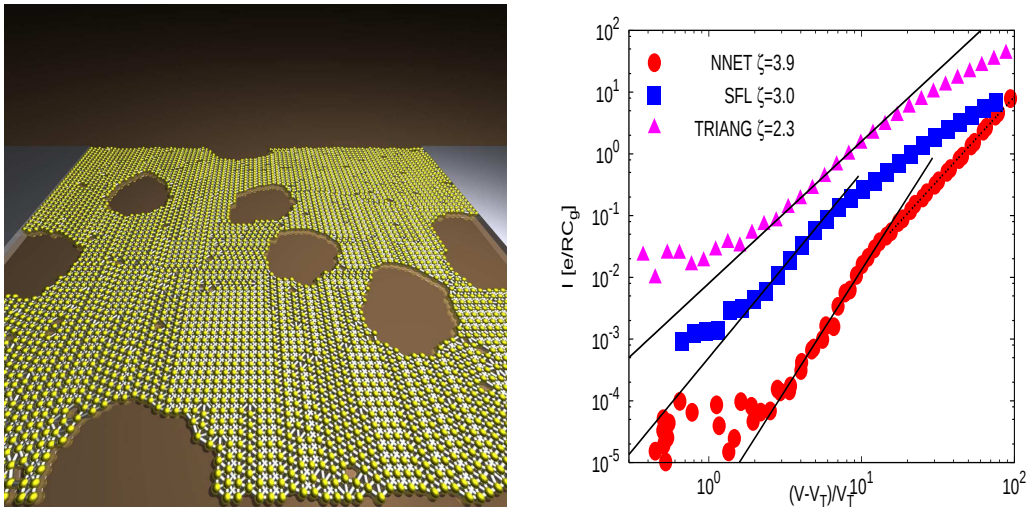

FIGURE 2. Conducting network NNET of nano-particles on substrate [8] with links representing possible tunnelings (left) and simulated $I(V)$ curves for NNET, SFL, and triangular topologies (right).

these networks is enabled by the Coulomb blockade effects due to small nanoparticle capacitance C. The electrostatic energy of the array is given by:

$$E = \frac{1}{2}\mathbf{Q}^{\dagger}M^{-1}\mathbf{Q} + \mathbf{Q}\cdot V^{ext} + Q_{\mu}\Phi^{\mu}, V^{ext} = M^{-1}C_{\mu}\Phi^{\mu}\ ; \qquad (2)$$

where $\{Q_i\}\ i = 1,2,\cdots N$ are nanoparticle charges, C_{ij} capacitances, Φ^{μ} ($\mu = +,-,gate$) potentials. The network structure, given by its adjasency matrix elements A_{ij}, contributes through the capacitance matrix $M_{ij} = \delta_{ij}\left(\sum_k CA_{ik} + \sum_{\mu} C_{i\mu}\right) - CA_{ij}$. Tunneling of a single electron along a junction $i \rightarrow j$ between two nanoparticles induces the energy

change ΔE_{ij}, which is felt in a large area. The tunneling rate $\Gamma_{ij} = -\frac{\Delta E_{ij}/e^2R}{1-\exp(\Delta E_{ij}/k_BT)}$ also depends on nature of the processes at each junction, which gives the quantum tunneling resistan R. In our numerical implementation [9], R appears as a parameter which scales the current. For the network shown in Fig. 2, left, the simulated $I(V)$ curve is given on Fig. 2, right, exhibiting a large nonlinearity $I(V) \sim \left(\frac{V-V_T}{V_T}\right)^{\zeta}$, $\zeta = 3.9$ [8]. The exponent ζ depends on the topology of the sample via the collective effects of many *conducting paths* opening between the electrodes. For comparison, we also show the simulated $I(V)$ curves for the regular triangular array with charge disorder, with the exponent in a good agreement with measured $\zeta = 2.3$ [14], and for our model network with a scale-free distribution of loops (SFL) where $\zeta \approx 3$, demonstrating the topology effects.

CONCLUSION

Using two examples of the network models of the nano-particle assemblies (scale-free structure of magnetic nano-particles and conducting nano-particle arrays on substrates) we have demonstrated that the topology of nano-particle assemblies can boost their emergent physical properties. The quantitative dynamic–topology interdependence can be studied systematically within the graph theory and numerical simulations of the physical processes on networks. Our numerical implementations leave ample space for the process and topology adjustments, more details are given in review Refs. [1, 12].

Acknowledgments: We thank for the financial support from the EC FP6 project MRTN-CT-2004-005728 and from the program P1-0044 of the MVZT (Slovenia).

REFERENCES

1. Tadić, B., *From Microscopic Rules to Emergent Cooperativity in Large-Scale Patterns*, in Systems Selfassembly: Multidisciplinary Snapshoots, edited by N. Krasnogor *et al.*, Elsevier, 2007.
2. Moriarty, P., . Taylor, M.D.R, Brust, M., Phys. Rev. Lett. **89**, 248303 (2002).
3. Fery,D.K., Goodnick, S.M., *Transport in Nanostructures*, Cambridge Univeristy Press, 1997.
4. Guo, L., leobandung, E., and Chou, S.Y., *A Silicon Single-Electron Transistor Memory Operating at Room Temperature*, Science **275**, 649 (1997).
5. Archer, P.I., Santangelo, S.A., and Gamelin, D.R., *Nano Letters*, **7**, 1037-1049 (2007).
6. Tadić, B., Malarz, K., and Kulakowski, K., *Physical Review Letters*, **94**, 137204 (2005).
7. Tadić, B., Rodgers, G.J., and Thurner, S., *Int. J. Bifurcation and Chaos*, **17**, July (2007).
8. Blunt, M.O., Šuvakov, M., Pulizzi, F., Martin, C.P., Pauliac-Vaujour, E., Stannard, A., Rushfort,A. W., Tadić, B., and Moriarty,P., *Nano Letters*, **7**, 855-860 (2007).
9. Šuvakov, M. and Tadić, B., *Simulation of the Electron Tunneling Paths in Networks of Nano-Particle Films'*, Y. Shi *et al.* Eds., Lecture Notes in Computer Science, **4488**, pp. 641-648, Springer (2007).
10. Šuvakov, M. and Tadić, B., *Topology of Cell-Aggregated Planar Graphs*, V.N. Alexandov *et al.* Eds., Lecture Notes in Computer Science Part III, **3993**, pp. 1098-1105, Springer, (2006).
11. Šuvakov, M. and Tadić, B., Physica A **372**, 354 (2006).
12. Šuvakov, M. and Tadić, B., *Charge Transport in Assembled Nano-Particle Structures*, chapter in "Statistical mechanics Research Focus", Nova Science Publishers (in preparation)
13. Tadić, B., Physica A, **293**, 273-284 (2001).
14. Parthasarathy, R., Lin, X., Elteto, K., Rosenbaum, T.F., Jeager, H.M., Phys. Rev. Lett. **92**, 076801 (2004).