Saeid Shojaei[1]
Irina V. Antonova[2,3,4,5], Irina Kurkina[2], S. Mehdi Sattari[1]
[1] Photonics Department, Research Institute for Applied Physics & Astronomy (RIAPA), 29 Bahman Blvd, University of Tabriz, Tabriz, Iran

[2]Ammosov North-Eastern Federal University, 677000, Yakutsk, Russia
3Rzhanov Institute of Semiconductor Physics SB RAS, 630090, Novosibirsk, Russia

[4]Novosibirsk State University, 630090, Novosibirsk, Russia
[5]Novosibirsk Technical State University, 630090, Novosibirsk, Russia

s_shojaei@tabrizu.ac.ir, shojaei.sh@gmail.com

Negative differential resistance in fluorographene matrix

In the present study, we demonstrate NDR for films created from a partially fluorinated graphene (FG) suspension. The origin of NDR is analyzed through the increasing in the fluorination degree of the suspension. NDR resulting from the formation of a barrier system in the film is observed for different degrees of fluorination. A step-like increase in current together with negative valley is specific to these films. The formation of graphene islands or graphene quantum dots (GQD) and a fluorinated graphene (FG) network is demonstrated in such films. Theoretical calculations based on effective 2D Dirac equation solution are performed to predict and model the appearance of NDR in these systems.

A graphene suspension was created from natural graphite using dimethylformamide (DMF) intercalation, and sonication. Then, graphene suspension was fluorinated by means of treatment in an aqueous solution of hydrofluoric acid[1,2]. It was found that NDR and a step-like increase in the current are found for films created from the fluorinated graphene suspension with fluorinated degree ≤ 25%.16 In our experiments, we produced three types of FG suspensions treated for 7-20 days in the aqueous solution of hydrofluoric acid. Then, thin films were created with use of spin-processor or drops on the silicon substrates. Films were labeled as FG1, FG2 and FG3. Fig.1 presents the scanning electron microscopy (SEM) images of films created from three suspensions. The main difference between the films is the relation between numbers of small and large flakes. A considerable decrease in the thickness and lateral size of the graphene flakes is found to occur in parallel with the fluorination during the treatment in the aqueous solution of hydrofluoric acid[1].
Fig.2 presents the scanning electron microscopy (SEM) images of films created from three suspensions. The main difference between the films is the relation between numbers of small and large flakes.

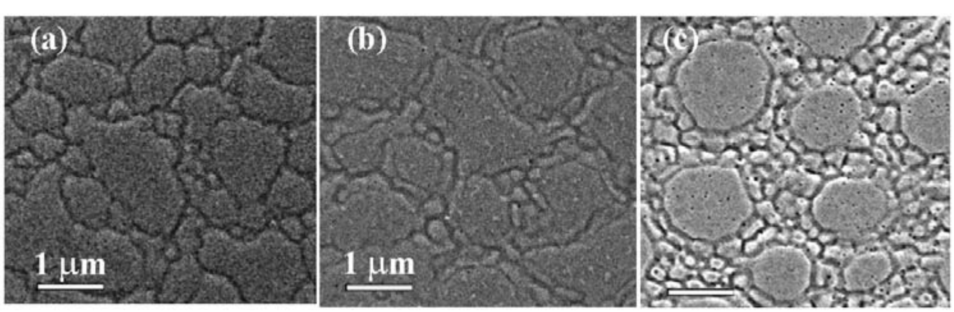

Figure 1: SEM images of the films created from different suspensions (a) FG1, (b) FG2, (c) FG3

Fig.2a presents the experimental current – voltage (I – V) characteristics for all samples together with numerical simulation. In this case one NDR with peak / valley ratio ~ 8 is clearly seen that is considerable value for device application. In the case of films with smaller flake and smaller GQD sizes (FG2 and FG3) a set of NDR peaks are found in I – V curves (Fig.2b). In the latter cases it is worth to mention that the first peak at positive voltage is more pronounced that other peaks at negative voltages.

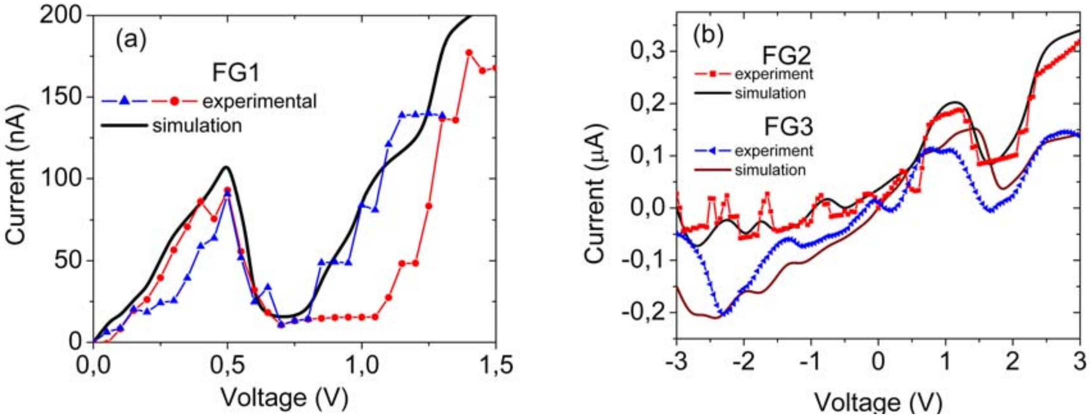

Figure 2: Measured and calculated (solid curve) I-V characteristic of graphene films with GQDs in fluorographene matrix based on sample FG1 (a) and FG2, FG3 (b).

In order to elucidate the effect of quantum size effect and resonant tunneling between adjacent quantum levels of GQDs or weakly fluorinated graphene flakes with FG flakes barriers that leads to NDR, we have carried out numerical calculation on a one dimensional well/barrier model to solve the effective 2D Dirac Hamiltonian as[3]:

$$H = -i\hbar\left(v_F\left(\sigma.p\right)\right) + eV\left(x\right)\hat{1} + \Delta\left(x\right)\sigma_z, \qquad (1)$$

where $\Delta(x)$ is the energy gap in different regions of samples as FG and GQD. $V(x)$ is position dependent external linear voltage, V_f, σ, p are Fermi velocity, Pauli matrix and momentum, respectively. I-V charecteristics of samples are calculated by adopted transfer matrix method within Landauer-Buttiker formalism[3,4]. The result of simulation is depicted in Fig. 2. It can be seen very good agreement between experiments and numerical simulation.

In conclusion, self-organized arrays of GQDs embedded in a matrix of partially fluorinated graphene were formed by chemical functionalization of graphene layers using treatment of samples in a solution of HF in water. The GQD sizes in films were determined from lateral-force AFM measurements roughly evaluated from large sizes (200-300 nm) to small (20–70 nm). One to multi-peak NDR with high peak to valley ratio were observed corresponds to different samples. This observation has very good consistency with that predicted by calculations.

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