## Electrical behavior of reduced graphene oxide thin films. Interplay of lateral size and chemical composition of single sheets.

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Structural and electrical properties of Reduced Graphene Oxide thin films (RGO-tf) strongly depend on such properties of the single RGO sheets.In particular, the same sheet shows a wide range of electrical properties because of the different reduction procedure and the final oxidation degrees.[1] In recent years, interest has been mainly devoted to study how the properties of RGO-tfdepend on the chemical composition of the single RGO sheet[2] while a systematic study on the effect of the lateral size and the shape of the sheets is still lacking. We recently demonstrated that the electrical and magnetic transport properties of thermallyRGO-tf strongly depend on the lateral size of the single sheets.[3] In this work, for the first time we combined the morphological and the chemical composition aspects of the RGO sheets in order to study how the interplay of the lateral size and the oxidation degree of the single layer affect the comprehensive electrical charge transport of the thin film.

Samples are prepared by spin coatinga few nanometer thick film of pristine graphene oxide (GO) on ultra-flat surface of silicon oxide, with three different lateral sizes: (S1)  $15\pm9 \mu m$ , (S2) $360\pm190 nm$  and (S3)  $150\pm44 nm$ . Solution with different GO size are prepared by sonicating at three times: 0, 2 and 20 hours. Mean values and corresponding error bar are calculated by the lateral size distributions of the GO sheetsachieved by the analysis of images acquired using Atomic Force (AFM) and Scanning Electron (SEM) microscopies. Each sample is then thermally reduced in vacuum ( $10^{-6}$  mbar) at three temperatures: 300, 600 and 900 °C. The oxidation degree is monitored with X-Ray Photoemission Spectroscopy (XPS) in ultra-high vacuum (pressure  $2 \cdot 10^{-10}$ mBar). Resistance measurements on RGO-tf are performed sweeping the temperature between  $2^{\circ}$ K and  $300^{\circ}$ K in vacuum.

Monitoring the chemical composition, we found that:i) the sonication does not modify the oxidation degree of the pristine GO and ii) the lateral size of the sheets does not roughly affect the reduction mechanisms as shown in figure 1a where the C/O ratio is plotted vs the annealing temperature. Despite the chemical properties do not vary, the measured electrical behaviour of the thin film show a strong dependence on the lateral size of the sheets for all the annealing temperatures as shown in Figure 1b for Ta = 900° where the transport mechanism is described by the2D Mott Variable Range Hopping (VRH)[4]model in the temperature range between 20°K and room temperature. For all the annealing temperatures, we observe that both the slope and the y-intercept increase with the decreasing of the lateral size of the single sheet. Larger the sheet size lower the electrical resistance of the film.Larger lateral size also corresponds to a lower number of sheets between source and drain electrodes suggesting that the contact region between two sheets can be treated as "defects" which affect the charge transport corresponding to an increasing of the electrical resistance with the number of the RGO sheets.



Figure 1 – (a) C/O ratio obtained from C 1s spectra for RGO of different size and reduction temperature. (b) Electrical resistance vs  $1/T^{1/3}$  for different RGO size, reduced at 900 °C.

## References

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