Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C

Supplementary Information

Influence of the covalent grafting of organic radicals to graphene on its magnetoresistance

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SI 1. HR-TEM images.

SI 2. Chemical Analysis.

SI 3. EPR of unmodified graphene.

SI 4. I-V conductivity versus voltage.

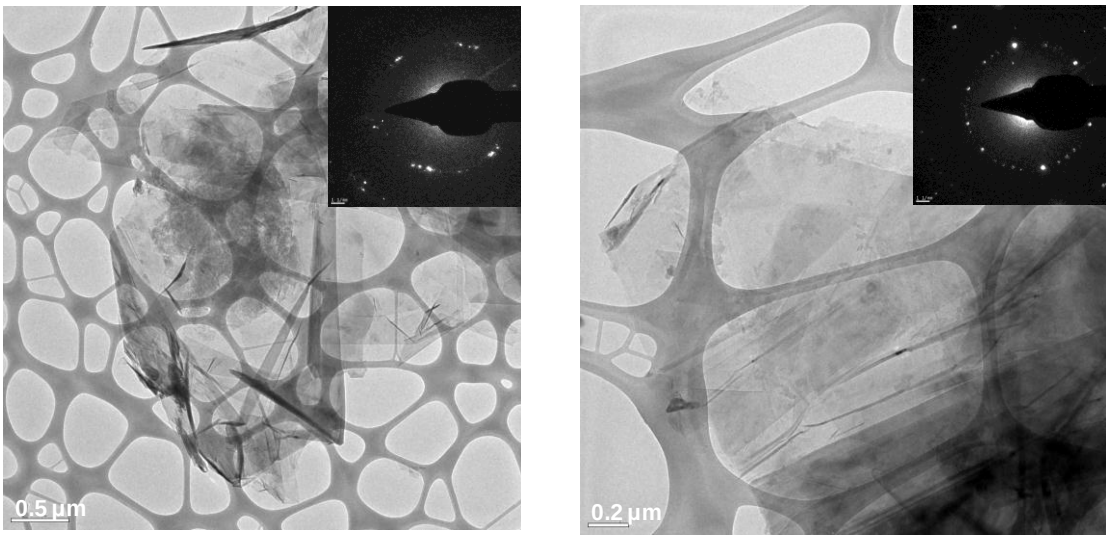
SI 5. MR versus magnetic field.

SI 6. MR versus magnetic field at variable temperature.

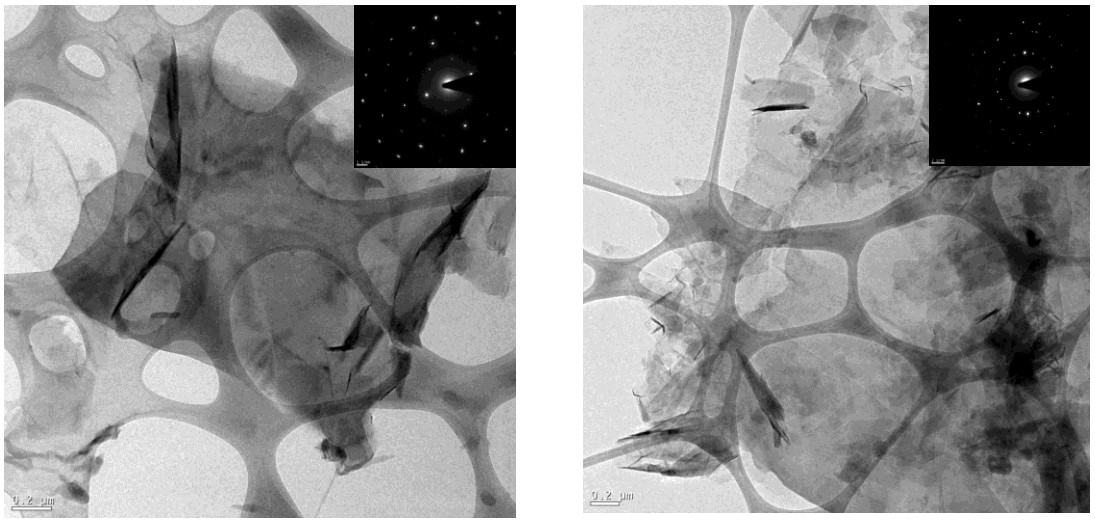
SI 7. Resistance versus field for ethylmalonate molecules.

SI 8. Fitting of the dependence of MR with the magnetic field at H>2T.

SI 1. HR-TEM images of (*top*) exfoliated graphene in oDCB (*left*) and **1-G** dispersed in ethanol (*right*), and (*bottom*) exfoliated graphene in benzylamine (*left*) and **1-G+** dispersed in ethanol (*right*). Insets display the Selected Area Electron Diffraction (SAED) collected for each sample that show the superposition of hexagonal diffraction patterns indicative of the presence of few-layers graphene.



(a)

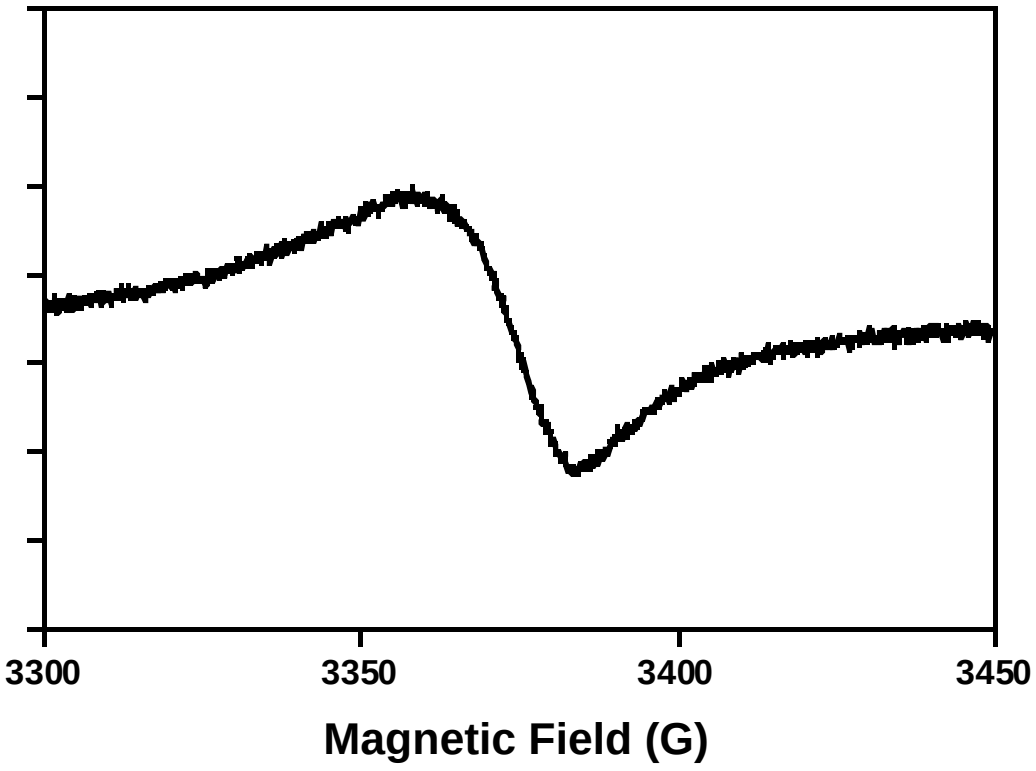


(b)

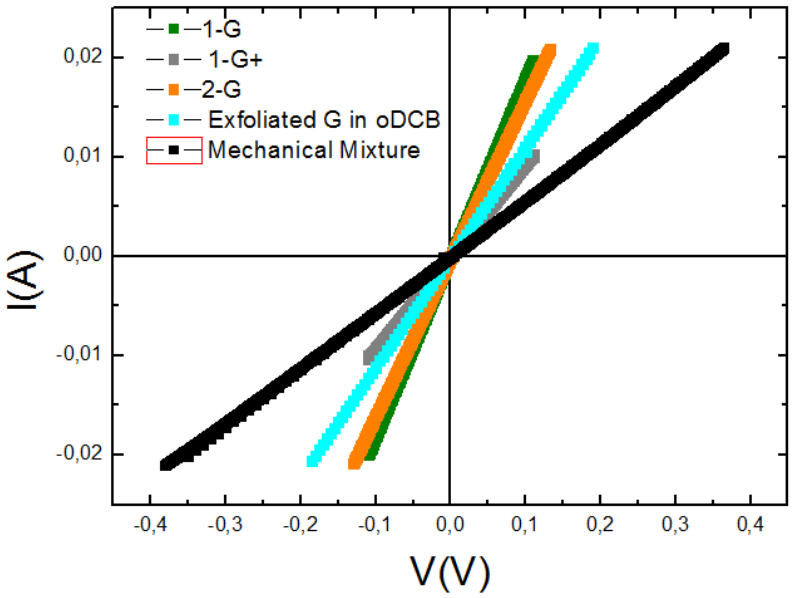
SI 2. CHN analysis performed over pristine graphite, **1-G** and **1-G+**.

	%C	%O	%N
Graphite	97.57	0.05	0.03
1-G	96.17	3.21	0.35
1-G+	93.78	5.64	0.58

SI 3. EPR spectra at 300 K of unmodified graphene

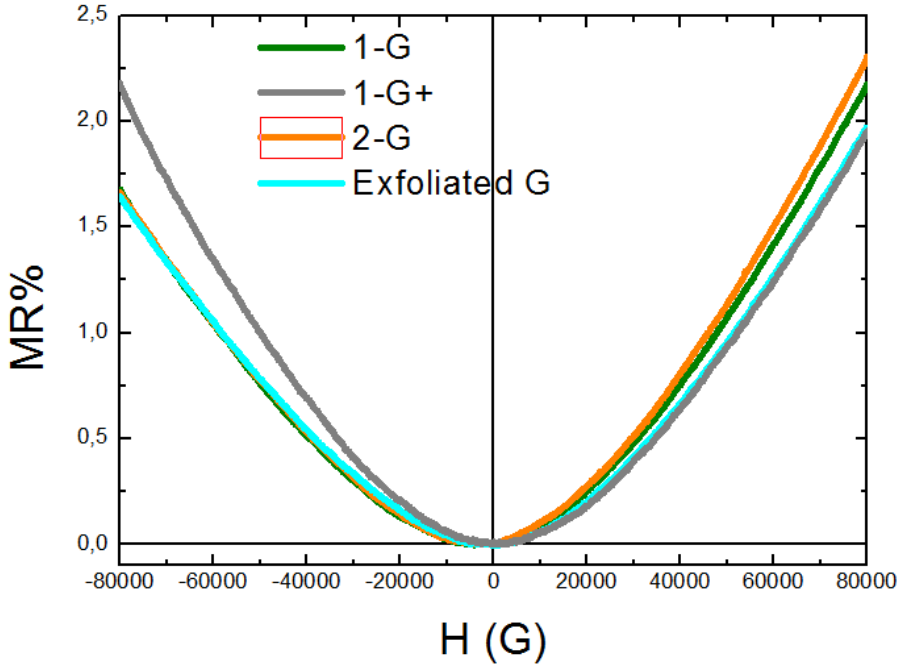


SI 4. I-V conductivity versus voltage at room temperature of **1-G** (green), **1-G+** (grey), **2-G** (orange), exfoliated graphene in oDCB (light blue) and the mechanical mixture of graphite with **1** (black). In the bottom part, a table showing the conductivity values calculated from inverse of the slope of the I-V curves for the different samples in the ohmic region at very low voltages are summarized.

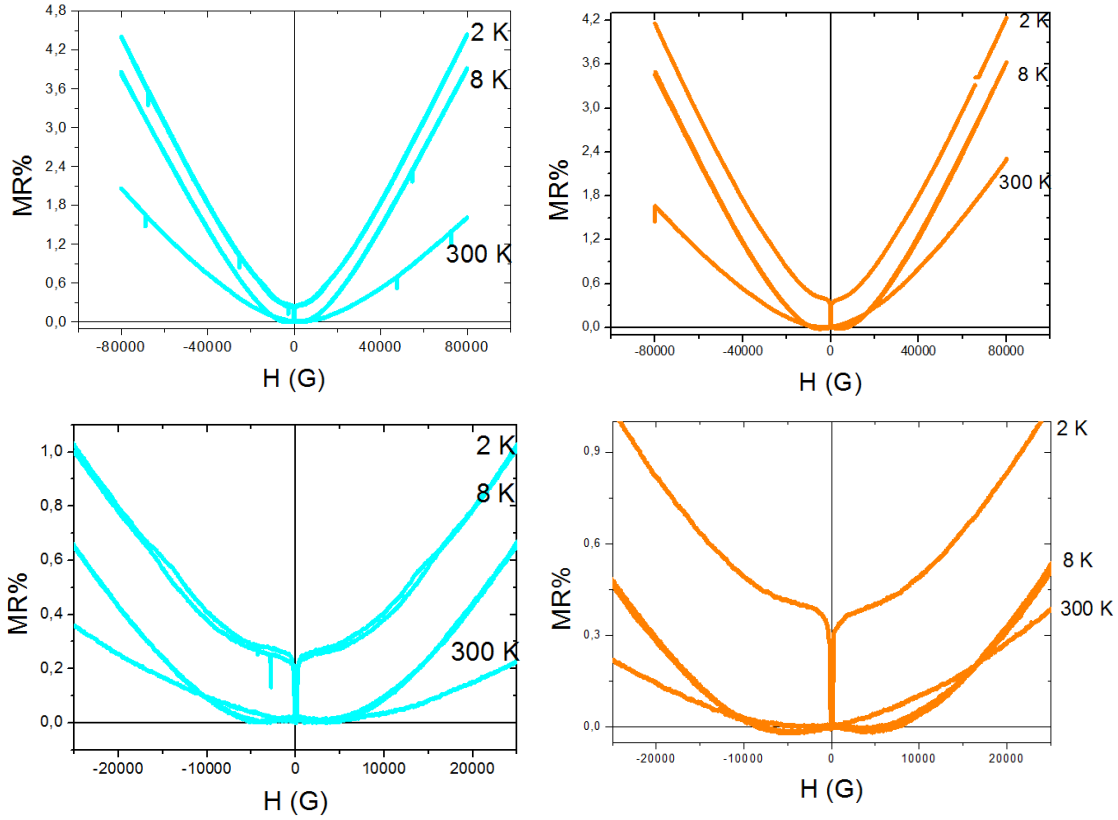


Sample	Conductivity [S·cm^{-1}]	Surface [cm^2]
1-G+	0.93	$1.77 \cdot 10^{-3}$
1-G	24.00	$1.87 \cdot 10^{-5}$
2-G	29.00	$1.53 \cdot 10^{-5}$
Exfoliated G in oDCB	30.00	$8.75 \cdot 10^{-5}$
Mechanical mixture of G and 1	28.22	$1.25 \cdot 10^{-4}$

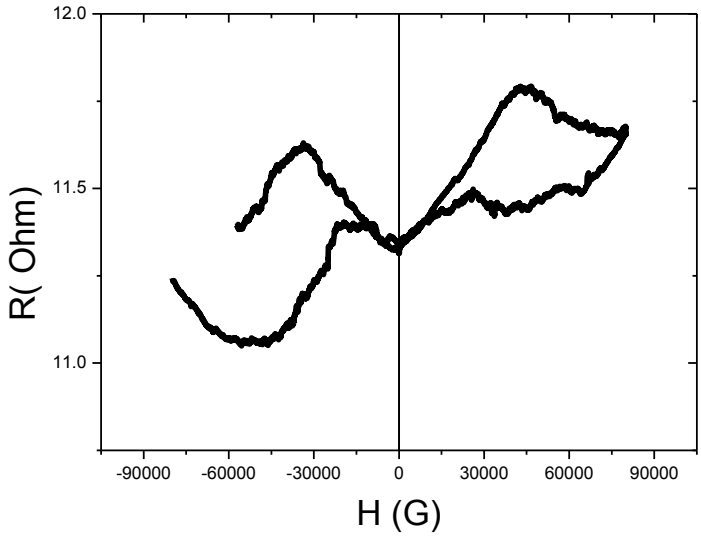
SI 5. MR versus magnetic field at 300K of **1-G** (green), **1-G+** (grey), **2-G** (orange) and exfoliated G in oDCB (blue). The three samples at 300 K show no differences between them at the low magnetic field part.



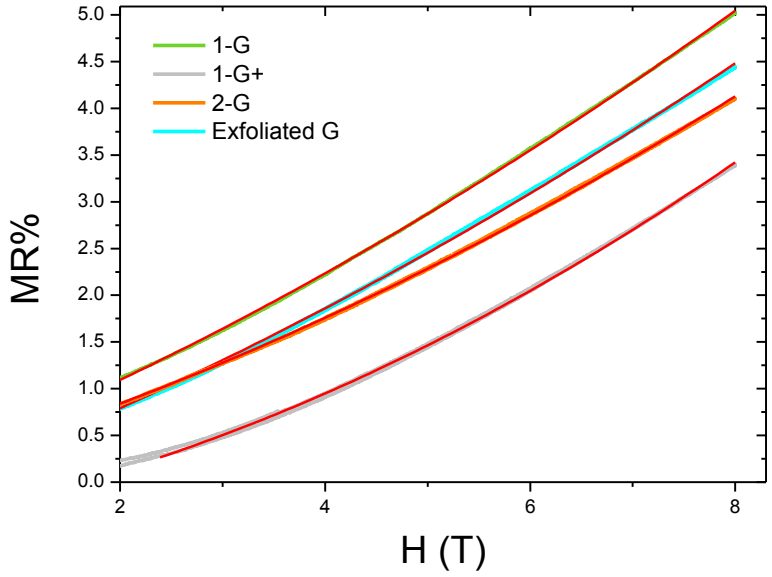
SI 6. MR versus magnetic field at different temperature of G-exfoliated in oDCB (left), **2-G** (right)). The bottom panel shows a zoom-in of the low field area for lower and higher temperature with the appearance of an anti-localization effect at 2 K. The LFMR effect present in 1-G and 1-G+ is not observed for any of them.



SI 7. Resistance versus field for ethylmalonate molecules. As describen in the text, no MR signal was observed.



SI 8. MR versus magnetic field at magnetics fields higher than 2 T were fitted with the following expressions $MR(H)= a\cdot H+b\cdot H^2$, considering both linear and quadratic dependence.[2, 3a]



As observed, the MR behavior at high fields is quite similar for all samples. In fact, all of them exhibit a non-saturating MR as expected for graphene-based materials.[1] In this high field area, for graphene two contributions are usually present. The first one in due to the curving of electron trajectories in the graphene plane which usually results in a positive MR with quadratic magnetic field dependence $MR\sim (\mu\cdot\mu_0\cdot H)^2$.[1a, 2] The second one is a linear MR, which has been observed in many systems.[3] Abrikosov gave with a quantum description for a zero band-gap system to explain the linear MR behavior.[4] Here, all the experimental data have been fitted with the equation proposed in reference 1a:

$$MR(H)=R(H)/R(0)-1=\rho H + ((\mu\cdot\mu_0\cdot H)^2$$

where the MR is proportional to a linear plus a quadratic magnetic field dependence, and where μ_0 is the permeability in vacuum and ρ is the proportional coefficient.

As a parameter the μ the carrier mobility is obtained from the curve fitting. The calculated carrier mobility is 141 $cm^2\cdot V\cdot s$ for the exfoliated graphene at 2 K, and the criterion $\mu(\mu_0\cdot H)<1$ is satisfied for the quadratic MR. The lower values for carrier mobility μ may be very low due

[1] a) Z-M. Liao, H-C. Wu, S. Kumar, G.S. Duesberg, Y-B. Zhou, G.L.W. Cross, I. V. Shvets, D-P. Yu, *Adv. Mater.*,2012, **24**, 1862. b) E. McCann, K. Kechedzhi, V.I. Fal'ko, H. Suzuura, T. Ando, B.L. Altshuler, *Phys. Rev. Lett.* 2006, **97**, 146805

[2] J. L. Olsen, Electron Transport in Metals, Interscience, New York 1962

[3] A.L. Friedman, J.L. Tedesco, P.M. Cambell, J.C. Culberstson, E. Aifer, F. K. Perkins, R.L. Myers-Ward, J. K. Hite, C.R. Eddy Jr., G.G. Jenirgan, D.K. Gaskill, *Nat. Nanotechnol.* 2008, **3**, 408.

[4] A.A.Abrikosov, P.B. Littlewood, *Nature*, 2003, **426**, 162

to the existence of boundaries, defects and impurities. The calculated carrier mobility for the rest is lower as one can expected because the impurities and the defects increase.