Supporting Information

Purely Coherent Nonlinear Optical Response in Solution Dispersions of Graphene Sheets

Rui Wu†, Yingli Zhang†, Shichao Yan†, Fei Bian†, Wenlong Wang†,*, Xuedong Bai†, Xinghua Lu†, Jimin Zhao†,*, Enge Wang‡

† *Beijing National Laboratory for Condensed Matter Physics and Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China*

‡ *International Center for Quantum Materials, School of Physics, Peking University, Beijing 100871, China*

* E-mail: (W.W) wwl@iphy.ac.cn, (J.Z.) jmzhao@iphy.ac.cn

S1

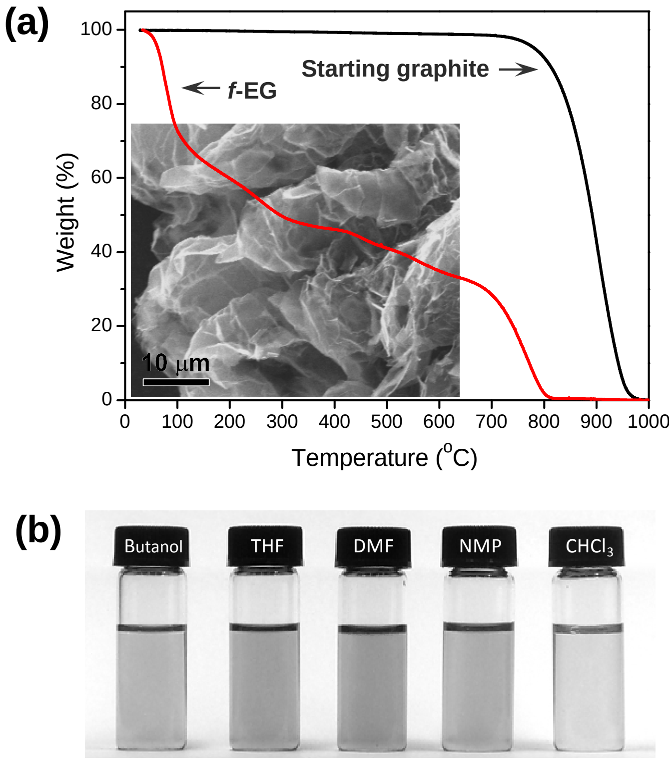

Figure S1. (a) Thermogravimetric analysis, performed under dry air with a temperature ramp rate of 10° C/min, of f-EG and the starting pure graphite. A representative SEM image of f-EG is shown as inset. (b) Picture of the prepared solution dispersions of the octylamine functionalized graphene sheets in different organic solvents of butanol, THF, DMF, NMP, and $CHCl_3$.

S2

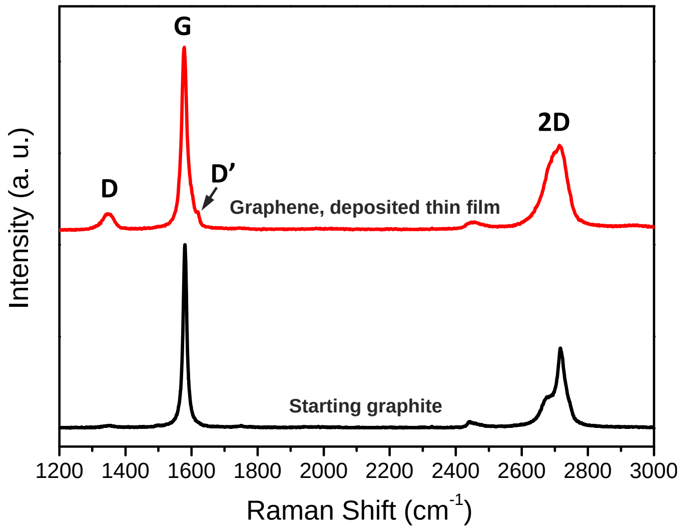

Figure S2. Raman spectra at 532 nm for the starting bulk graphite and the graphene thin film deposited on alumina filter.

S3

Two tables are used to elucidate such a huge optical nonlinearity in our graphene sample. In Table 1 we compare the nonlinear refractive index n_2 within carbon-related materials. It is seen that the n_2 of our graphene dispersion is larger than most of the other carbon-related materials, such as the carbon nanotube and C_{60}. In Table 2 we compare the threshold intensity I_{th} for observing diffraction rings in our experiment with those reported values of many other materials. From Fig. 4 we obtain that I_{th} is 12.5 W/cm^2 and indeed we find that I_{th} is 0.6 W/cm^2 in our parallel experiment if we do not focus the laser beam before it incident on the sample (Fig. 4). As shown the intensity threshold is almost the lowest among the reported values within the scope of our limited knowledge. For the above two properties n_2 and I_{th}, the solution density and sample thickness are two factors that need to be considered. We did not include them in the table, but we tried to include all the relevant reported values for the other materials.

Table S1. Effective nonlinear refractive index n_2 of carbon-related materials.

Material	Effective n_2 (m^2/W)	Wavelength (nm)
Graphene-NMP	3×10^{-9}	532
C_{60}-benzene [S1]	10^{-17} *	1064
C_{60}-benzene/toluene[S2]	-10^{-19}*	532
Carbon nanotube [S3]	10^{-18} *	532
	10^{-17} *	1064

* Derived from the reference paper.

Table 2. Reported low intensity thresholds I_{th} for observing diffraction rings

Material	Threshold I_{th} (W/cm^2)	Wavelength (nm)
Graphene-NMP	0.6	532
C_{60}-benzene [s1]	10	632.8
$Sr_{0.61}Ba_{0.39}Nb_2O_6$ [S4]	30*	514.5
Nematic liquid crystal [S5]	130	514.5
Roselle-Hibiscus Sabdariffa [S6]	700*	514.5

* Estimated using the threshold powers given in the reference papers.

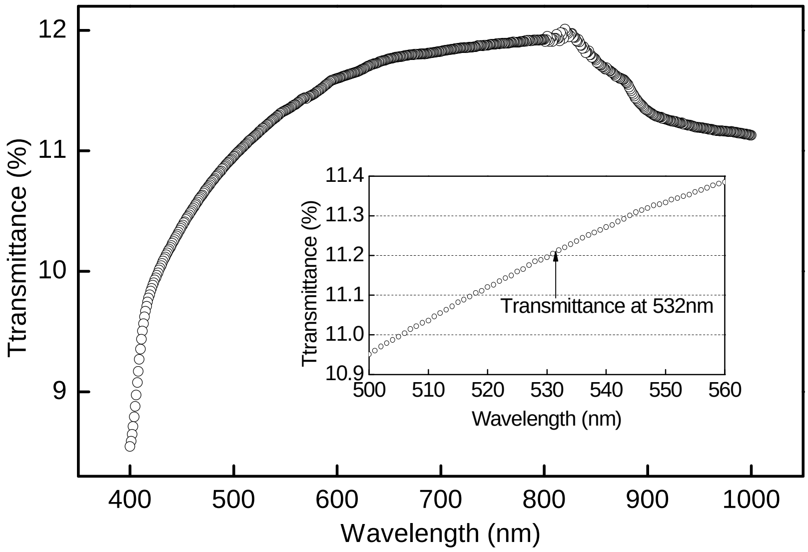

Figure S3. Transmittance spectrum of graphene suspension measured in the wavelength range from 400nm to 1000nm, which excluded the influence of solvent NMP and the cuvette. Transmittance of the graphene suspension at 532nm is 11.21% and the one of monolayer graphene is about 97.7% [S7]. Thus, the number of effective graphene layers in the suspension that laser passed through is evaluated to be about 94.

S5

For the 532 nm and 800 nm experiments, the laser power was relatively high. When light passed through the dispersion solution, light was absorbed and the solution was heated, where tiny gas bubbles could be seen moving upward in a speed of a few centimeters per second. The net result was that the upper part of the liquid has a lower density, which resulted in a smaller effective n_0 for the upper part. The further away, the smaller n_0, giving an effect similar to that of a convex lens. The net effect is like an additional refraction bending down the upper beams. For the upper rings, with identical number of rings to the lower part of the rings, the corresponding diffraction angle became very small, leading to deformed ring patterns.

References

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