Investigating Thermo-Optical Nonlinearities in Graphene and Graphene Oxide by Spatial Self-Phase Modulation (SSPM) Method

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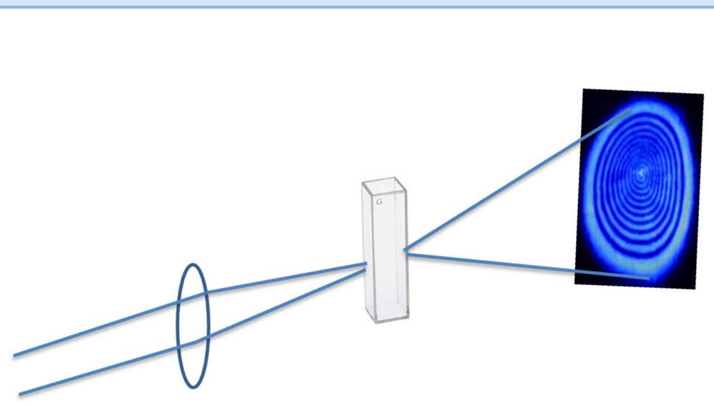
Abstract

The origin of SSPM in liquid suspension of graphene could be due to more than one phenomenon. We calculate here, the nonlinear refractive index of graphene and graphene oxide (GO) liquid suspension by SSPM method, and discuss the possible dominance of thermal effect as origin of SSPM in graphene.



Theory

Refractive index of a (nonlinear) medium depends on the incident intensity (I) as

$$n = n_0 + n_2 I$$

When a laser beam passes through a nonlinear medium, due to its asymmetric radial intensity distribution there arises a change in refractive index of the medium which is experienced by the laser beam itself. This creates spatial phase difference along its radial path. The resultant interference pattern is seen in far field in form of multiple concentric bright and dark rings.

Methods

Graphene (10-20 layers) and graphene oxide (1-2 layers) were prepared using electrochemical exfoliation of graphite and modified Hummer's method, respectively. 0.1mgml^{-1} concentrated solution of both samples in Dimethylformamide (DMF) were used in experiment.

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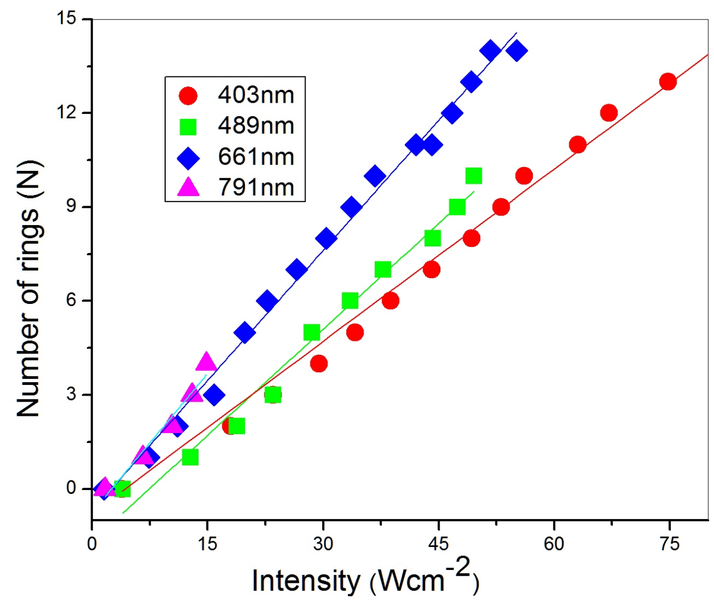
Results and Discussion

Nonlinear refractive index (n_2) of medium is given by

$$n_2 = \left(\frac{\lambda}{2n_0 L}\right)\frac{N}{I}$$

n_0: Medium refractive index

L: Optical path length

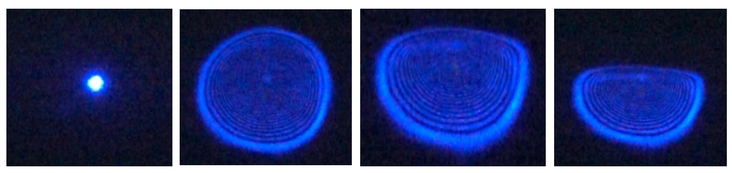


Number of rings vs intensity plot for graphene

Recent works regard SSPM in liquid suspension in graphene due to its high electronic mobility and Dirac cone-like band structure. However, laser induced thermal effect cannot be ruled out as another major contributor.

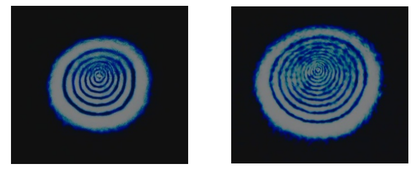
Thermal convection result into temperature gradient and distortion in upper half of ring pattern is observed.

(λ=403nm, power=90mW)



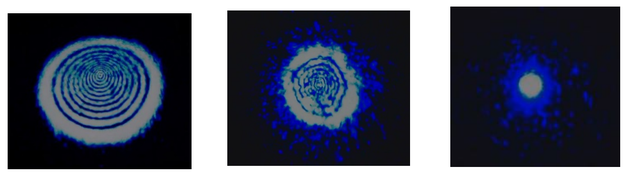
t = 0s t = 0.25s t = 0.4s t = 1s

Despite much lower electron mobility and larger bandgap, graphene oxide exhibits more number of diffraction rings.
(λ=489nm, power=35mW)



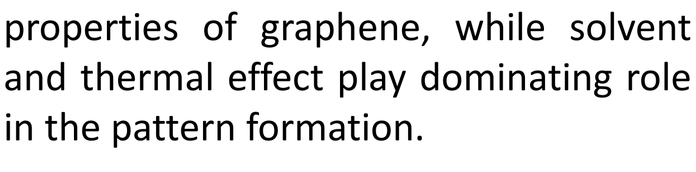
Graphene Graphene Oxide

Strong dependence of diffraction ring patterns on type of solvents casts doubts over nonlinear origin of SSPM.
(λ=489nm, power=35mW)



GO in DMF GO in Acetone GO in DI water

Modulation of CW laser beam produces significant change in ring pattern, supporting the dominance of thermal effect. If it was electronic dominance, number of rings should remain constant regardless of laser modulation.
(λ=489nm, power=40mW)



Without modulation 10 Hz 100 Hz

These observations show that SSPM has little to do with electronic and optical properties of graphene, while solvent and thermal effect play dominating role in the pattern formation.

Summary and Conclusion

1. Value of n_2 is estimated to be 3.8 X $10^{-10\pm1}$ for graphene and 6.2 X $10^{-10\pm1}$ for graphene oxide that match quite well with reported values (in m^2W^{-1} units).
2. Though not completely distinguishable, thermal effect dominates over other modes of origin in SSPM phenomenon in graphene.

References

Yanan Wang *et. al.,* Nanoscale, 9, 3547-3554 (2017).
Rui Wu *et. al.,* Nano Letters, 11, 5159-5164 (2011).