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## اندازه گیری ضریب شکست غیر خطی گرافن تولید شده به روش کندگی لیزری در محیط نیتروژن مایع

سارا حسامی افشار، پرویز پروین، حمیدرضا حبیبیان، علی بوالی، طیبه محمدی، علی دیده بان، مریم دزفولی، کامران شایان

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چکیده - تحت تابش قوی نور لیزر، گرافن چند لایه تغییر فاز غیرخطی از خود نشان می دهد. در این گزارش مقدار این تغییر فاز که ضریب شکست غیر خطی نامیده می شود محاسبه خواهد شد. بر این اساس نشان داده می شود که گرافن تولید شده به روش کندگی لیزری مقدار بزرگتری از ضریب شکست غیرخطی را در قیاس با سایر گرافن های تولید شده به روش شیمیایی از خود نشان می دهد.

کلیدواژه - کندگی لیزری، گرافن، نمار شکست غیر خطی

## Z -scan Measurement Of Nonlinear Refractive Index Of Graphene Based On Q-switched Nd:YAG Laser Ablation Of Graphite Target In Liquid Nitrogen

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*Abstract: Under strong laser illumination, few-layer graphene exhibits a nonlinear phase shift. Here, we distinguish this nonlinear optical effect which used to be named nonlinear refractive index. We show that fabricated graphene based on Q-switched Nd:YAG laser ablation possesses a giant nonlinear refractive index in comparison with graphene powders fabricated by chemical method.*

**Keywords:** graphene, laser ablation, nonlinear refractive index.

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## 1 Introduction

Graphene, a single sheet of carbon in a hexagonal lattice, exhibits many interesting electrical and optical properties [1,2] that arise due to its particular bandgap structure  $E(p)=V|p|$ , where the sign corresponds to electron (respectively, hole) band,  $p$  is the quasi-momentum, and  $V \simeq 10^6 \text{ ms}^{-1}$  is the Fermi velocity. In particular, a single graphene layer absorbs a fraction  $\pi\alpha \simeq 2.3\%$  of the incident light across the infrared and visible range, where  $\alpha$  is the fine structure constant. Graphene also has a broadband ultrafast saturable absorption, due to valence band depletion and conductance band filling. This saturable absorption has been used, among other applications, for laser mode locking. The massless bandgap structure of graphene has led to the prediction of other strong nonlinear optical effects, including frequency multiplication [3,4] and frequency mixing [5]. Recently, broadband four-wave mixing in few-layer graphene samples has been reported [6]. This allowed the determination of the absolute value of the third order susceptibility of a single graphene layer  $|\chi_{gr}^{(3)}| \simeq 1.5 \cdot 10^{-7} \text{ esu}$ , approximately 8 orders of magnitude larger than in bulk dielectrics. Four-wave mixing in graphene deposited on optical ferrules has also been observed [7]. In section 2 material will be discussed, in section 3 test setup will be illustrated, and in section 4 results of experiments will be analyzed.

## 2 Materials

The experimental measurement of the nonlinear optical refractive index  $n_2$  of loosely stacked few-layer graphene using Z-scan technique is discussed in this section. The nonlinear refractive index of graphene generated by Laser Ablation in Cryogenic media is investigated in this

experiment. This sample was prepared based on the pulsed laser ablation of graphite target inside the cryogenic liquid using the pulsed nanosecond Q-switched Nd:Y3Al5O12 (Nd:YAG) laser at 1064 nm. Nitrogen is used as cryogenic liquid in the proposed method. Synthesized Graphene through this method which is analyzed by Raman spectroscopy, AFM and other measurements indicates the existence of few layer graphene. TEM and SEM were used for imaging the sheets of graphene. [8]

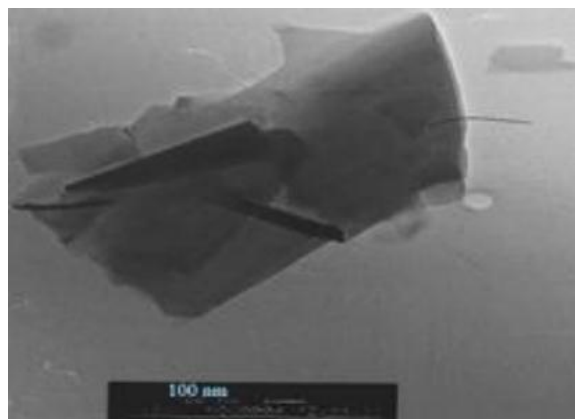


Figure 1: TEM image of graphene synthesized in nitrogen

## 3 Z-Scan

Nonlinear refractive index measurement was carried out by Z-scan, which is a sensitive single-beam technique for measuring both the nonlinear refractive index and nonlinear absorption coefficient for a wide variety of materials. We describe the experimental details and present a comprehensive theoretical analysis including cases where nonlinear refraction is accompanied by nonlinear absorption. In these experiments, the transmittance of a sample is measured by the propagation path ( $z$ ) of a focused Gaussian beam. The sign and magnitude of the nonlinear refraction are easily deduced from such

a transmittance curve (Z-scan). In cases where nonlinear refraction is accompanied by nonlinear absorption, it is possible to separately evaluate the nonlinear refraction as well as the nonlinear absorption by performing a second Z scan with the aperture removed. Previous measurements of nonlinear refraction have used a variety of techniques including nonlinear interferometry, degenerate four-wave mixing, nearly degenerate three-wave mixing, ellipse rotation, and beam distortion measurements. The first three methods, namely, nonlinear interferometry and wave mixing, are potentially sensitive techniques, but all require relatively complex experimental apparatus. Beam distortion measurements, on the other hand, are relatively insensitive and require detailed wave propagation analysis. The technique reported here is based on the principles of spatial beam distortion, which offers simplicity as well as very high sensitivity [7].

#### 4 method

Linear refractive index of graphene can be obtained via different experiments like reflectivity spectroscopy, spin Hall effect of light and Abbe refractometry. And it's near  $2.27 \pm 0.02$ . In this report for reaching nonlinear refractive index, the Z-scan setup was adapted to our graphene, suspended in deionized water. In general, nonlinearities of any order can be considered; however, for simplicity, we first examine only a cubic nonlinearity where the index of refraction  $n$  is expressed in terms of nonlinear indexes  $n_2$ (esu) or  $\gamma$  ( $m^2/W$ ) through

$$n = n_0 + \frac{n_2}{2}|E|^2 = n_0 + \gamma I \quad (1)$$

Where  $n_0$  is the linear index of refraction,  $E$  is the peak electric field (cgs), and  $I$  denotes the irradiance (MKS) of the laser beam within the sample.  $n_2$  and  $\gamma$  are related through the conversion formula

$$n_2(esu) = (cn_0/40\pi)\gamma(m^2/W) \quad (2)$$

where  $c(m/s)$  is the speed of light in vacuum.

According to [9], we can write relation between phase shift and difference between the normalized peak and valley transmittance as

$$\Delta\phi_0 = \frac{2\pi}{\lambda} n_2 I_0 L_{eff} \quad (3)$$

$$\Delta T_{pv} \cong 0.406(1-S)^{0.25} |\Delta\phi_0| \quad (4)$$

Where  $\lambda$  is wave length of incident laser,  $I_0$  is the on-axis irradiance at focus,  $L_{eff}$  is sample length and  $S$  is aperture linear transmittance. In order to optimize signal to noise ratio,  $S$  is usually considered to be 0.4.

Figure 2, shows the experimental setup of closed aperture Z-scan.

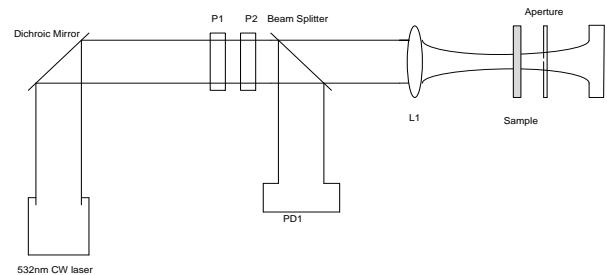


Figure 2 : experimental setup of closed aperture Z-scan

In this work we used laser with wave length at 532nm, with 200mw power in the condition which aperture is removed, 2 polarisers for controlling power of the laser, a lens for focusing the laser beam, and an aperture near the powermeter. Figure 3, shows Z-scan trace for our sample. As seen in this figure, difference between the normalized peak and valley transmittance ( $\Delta T_{pv}$ ) is equal to 0.384 that led  $n_2$  to be  $0.091 \cdot 10^{-3} cm^2 W^{-1}$ .

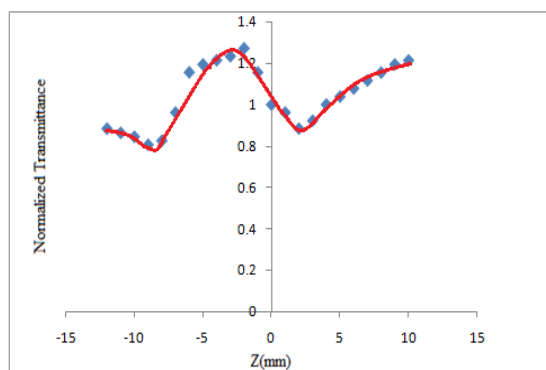


Figure 3: Z-scan trace for the proposed sample

## 5 Conclusion

In this work we distinguished nonlinear optical effect which used to be named nonlinear refractive index. We showed that fabricated graphene based on Q-switched Nd:YAG laser ablation possesses a giant nonlinear refractive index in comparison with graphene powders fabricated by chemical method.

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