

# Second Harmonic Generation of Femtosecond Laser Pulses of Central Wavelength 1000 nm, 1100 nm and 1300 nm Using ZnO Nanorods

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## Research Article

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# Abstract

In this work high density ZnO nanorods were grown on glass substrate by using a two steps method. In the first step drop-casting method was used for growth of seed layer by taking Zinc Acetate Dihydrate as precursor. In the second step, ZnO nanorods were prepared through Chemical Bath Deposition process on this seed layer by taking Zinc Nitrate as precursor. Field Emission Scanning Electron Microscope study was done to study the morphology of grown ZnO nanorods. From this study the average diameter and length of nanorods were found to be 33 nm and 270 nm respectively. These nanorods are successfully used for second harmonic generation (SHG) of femtosecond laser pulses of central wavelengths like 1000 nm, 1100 nm and 1300 nm. Discussion is given on the significance of this result along with its potential application.

## 1. Introduction

Femtosecond (fs) laser pulses are very useful to explain and monitor various ultrafast physical, chemical, biological phenomena (WEINER 2009). They are also useful for Multiphoton Microscopy/NLO Microscopy (Le et al. 2007), Medical Therapeutic Surgery (Chen 2012; Curley et al. 1992) Telecommunication Component growth and characterization (Dorrer and Kang 2002), Micro Machining (Hamad 2016; Slocombe and Li 2000; Zhang) Photonic band gap material generation (Cumpston et al. 1999), Spectral Comb generation (Michel et al. 2003; Wilken et al. 2012), Pump probe spectroscopy (Vigliotti et al. 2004) and frequency metrology (Jones and Diels 2001) etc. These applications can be achieved perfectly only through the precise control of the characteristics of fs laser pulses. So, proper characterization of these pulses is essential. The characterization of these pulse is generally done by various instruments like autocorrelator, Frequency Resolved Optical Gating (FROG), Spectral Phase Interferometry for Direct Electric-field Reconstruction (SPIDER) etc. Second harmonic generation (SHG) is most commonly used phenomena for the characterization of these laser pulses through these instruments. Due to high chemical stability, high damage threshold, low cost and high second order optical nonlinearity, the Zinc oxide (ZnO) nanorods has been found to be very promising for SHG of fs pulses (Dai et al. 2013; Das et al. 2014; Larciprete et al. 2015; Multian et al. 2017; Panda et al. 2016; Panda and Das 2017; Rout et al. 2019; Wang et al. 2019). This material is also found to be very promising for characterization of fs pulses (Panda and Das 2017). However, in all these works SHG have been done with fs pulses of central wavelength 800 nm only. In this work, it is demonstrated that the SHG of fs pulses of other central wavelengths like 1000 nm, 1100 nm and 1300 nm can also be done by using a single ZnO nanorods sample at only one angle of incident. Discussion is given on the significance of this result along with its potential application.

## 2. Experimental Procedure

### 2.1. Growth and Characterization of the ZnO nanorods

A two-step method was used for the growth of ZnO nanorods on glass substrate (Panda et al. 2016). In brief, firstly drop-casting method was used for growth of seed layer. In this, 22 milligrams of zinc acetate dihydrate ( $C_4H_6O_4Zn \cdot 2H_2O$ ) was added with 20 ml of absolute ethanol for obtaining a 5 mM solution. 0.06 ml of prepared solution was equally spread in the area of  $16.5 \text{ cm}^2$  over glass substrate and left it for drying using hot air for next 10 minutes. Then the entire process was repeated for four times on the same substrate. After that, the sample prepared was heated to form the ZnO seed layer using an induction heater. At the surface of sample this induction heater provides the temperature of  $132^\circ\text{C}$  for 2100W power at its maximum. The ZnO seed layer was prepared by calcination for one hour at  $132^\circ\text{C}$ . In the second step, ZnO nanorods were prepared through Chemical Bath Deposition (CBD) process on seed layer. In this step, 20 mM zinc nitrate ( $Zn(NO_3)_2$ ) solution was prepared by adding 0.15 gm of Zinc nitrate hexahydrate (Emplura) with distilled water (25 ml). Then 0.8 M of NaOH solution was prepared by mixing 0.81 gm of NaOH (Himedia) and 25 ml of distilled water followed by stirring. Then 25 ml of both zinc nitrate and NaOH solutions was mixed slowly to prepare 50 ml of mixture solution and this mixture was then stirred for 5 minutes. Then prepared solution was heated at  $70^\circ\text{C}$  and seed layer coated glass substrate was submerged for two hours. After two hours, the sample was removed from the solution, cleaned with water and dried by hot air. Field Emission Scanning Electron Microscope (FESEM) study was done to study the morphology of grown ZnO nanorods.

## 2.2. Experimental work on SHG of fs laser pulses of different wavelengths

The schematic diagram of experimental setup used for this work is shown in the Fig. 2. Fs pulses of different wavelength generated by an amplified fs laser pumped OPA (TOPAS-prime of LIGHT CONVERSION) was used for this purpose. The pump wavelength was at 800 nm and the duration of the laser pulses are 110 fs. There output of the OPA system at wavelength 1000 nm, 1100 nm and 1300 nm where used for the SHG experiment. These ultrafast pulses were focused onto the ZnO nanorods samples using a 5 cm focal length lens to generate the second harmonics. The angle of incident on the sample was  $45^\circ$ . Notch filters of appropriate wavelength were used to cut the fundamental radiation from the SHG. HR 4000 spectrometer was used to detect the SHG radiation.

## 3. Results And Discussion

The result of the morphological study of the ZnO nanorods is shown in Fig. 1. It shows the FESEM image of the ZnO nanorods. From this figure the average diameter and length of nanorods were found to be 33 nm and 270 nm respectively.

The SHG spectrum for the fs pulses of central wavelength 1000 nm, 1100 nm and 1300 nm are shown in Fig. 3a, Fig. 3b and Fig. 3c respectively. As it can be seen from this figure, the central wavelength of SHG pulses are at wavelengths 500 nm, 550 nm, 650 nm respectively. This figure clearly confirms that a single sample of ZnO nanorods can be used for SHG of fs laser pulses of central wavelength 1000 nm, 1100 nm, 1300 nm.

So in this work we have demonstrated that SHG of fs pulses of other central wavelengths other than 800 nm can also be done using the ZnO nanorods. The more interesting fact here is the SHG of the multiple wavelength ultrafast pulses were done using a single ZnO nanorods sample at only one angle of incident. This become possible because of the fact that SHG in the present case has be done with the ZnO nanorods having length much less than the coherence length ( $l_c$ ). This can be easily verified from the Fig. 4. This figure indicates the plot of  $l_c$  versus fundamental wavelength. In particular, the Fig. 4 (a) and Fig. 4 (b) represent the variation of  $l_c$  for broad wavelength and short wavelength range respectively.

The following equation was used to estimate the  $l_c$  (Boyd 2003)

$$l_c = \frac{\lambda}{4\pi(n_{2\omega} - n_{\omega})} \quad (1)$$

Where  $n_{\omega}$ = refractive index for fundamental wavelength,  $n_{2\omega}$ = refractive index for second harmonic wavelength,  $\lambda$  = wavelength of fundamental laser radiation. The  $n_{\omega}$ ,  $n_{2\omega}$  are estimated from the Sellmeier equation (Bond 1965)

$$n_{ZnO} = 1.9148 + \left(\frac{0.0569}{\lambda^2}\right) - \left(\frac{0.0136}{\lambda^4}\right) + \left(\frac{0.002168}{\lambda^6}\right) \quad (2)$$

From Fig. 4, the  $l_c$  for the SHG of laser pulses within the wavelength range 800 nm – 4000 nm is found to be  $\geq 1200$  nm. This indicate if the used ZnO nanorods are having length much less than 1200 nm then they can be easily used for SHG of fs pulses of any possible wavelength in the aforementioned range without going for phase matching process. This is the case in our present study as the length of the used ZnO nanorods are of 270 nm length.

It is also to note here that conventionally, the SHG of ultrafast pulses of various central wavelengths is mostly done by using the angular tuning or temperature method methods in the commercial nonlinear optical (NLO) crystals. One needs sophisticated extra arrangement to realize such tuning. In our case no such tuning is required. So the process reported here is much simpler. There are a few other advantages of our methods. Firstly the angular or temperature tuning supports the SHG of ultrafast pulses in a limited wavelength range. But as the method reported here is based on non-phase matching process, it can have much broader working range.

Finally, it is also to note that the SHG study in this work has been done by taking the outputs of an OPO. As an OPO can generate fs pulses of any wavelength within its working range so the SHG of them can be easily done by a single ZnO nanorods sample at only one angle of incident. It is to mention here that the SHG of ZnO can be easily used for realization of ultrafast laser pulse diagnostics system. This has already been demonstrated for the fs laser pulses of 800 nm (Panda and Das 2017). As a single ZnO nanorods sample can be used for SHG in a single angle of incident, a broad wavelength range ultrafast

pulse diagnostics system can be easily realized using this concept. This will be particularly very useful to characterize the fs pulses generating from the OPO.

## 4. Conclusion

In this work high density ZnO nanorods were grown on glass substrate by using a two steps method. These nanorods are successfully used for second harmonic generation (SHG) of femtosecond laser pulses of central wavelengths like 1000 nm, 1100 nm and 1300 nm. Such method of SHG of various central wavelengths is much simpler and can be used for characterization of fs laser pulses coming out from an OPO. Further work on this direction is under progress.

## Declarations

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**Conflict of interest:** The authors declare that they have no conflict of interest.

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<https://www.industrial-lasers.com/cutting/article/16488567/femtosecond-laser-micromachining-a-backto-basics-primer>

## Figures

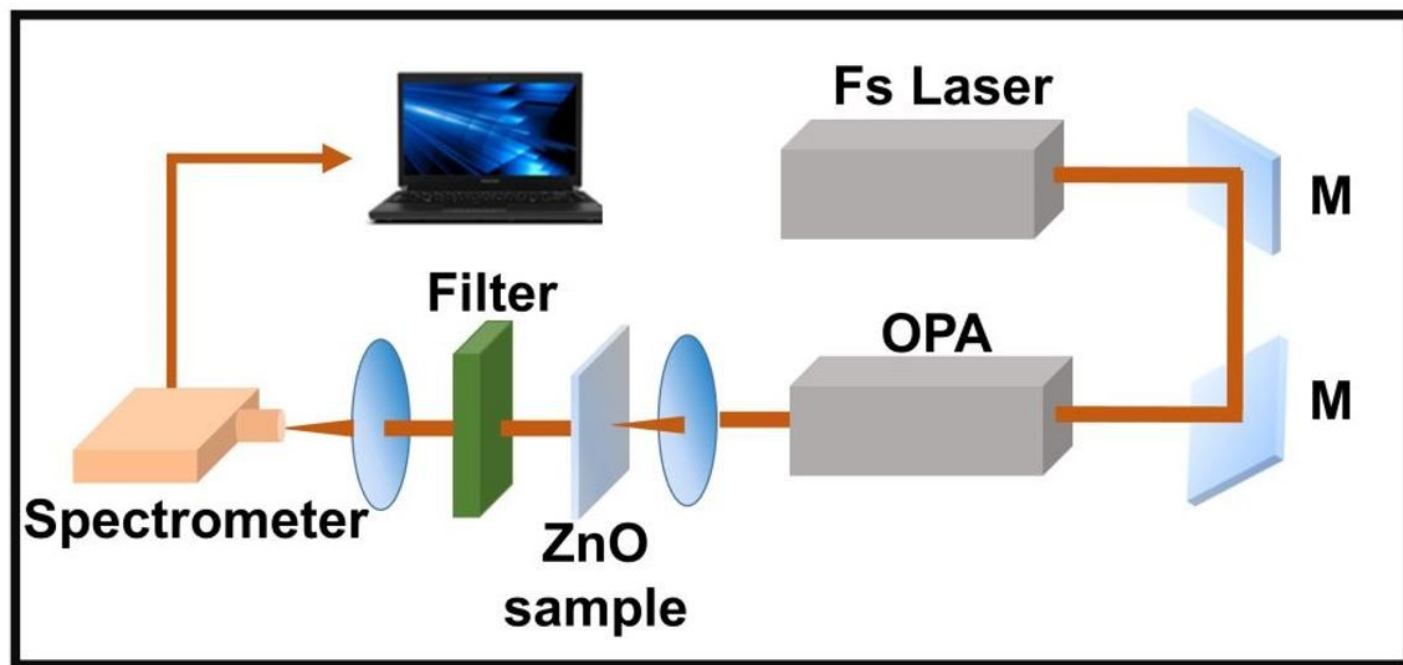


Figure 1

Schematic diagram of experimental setup for SHG.

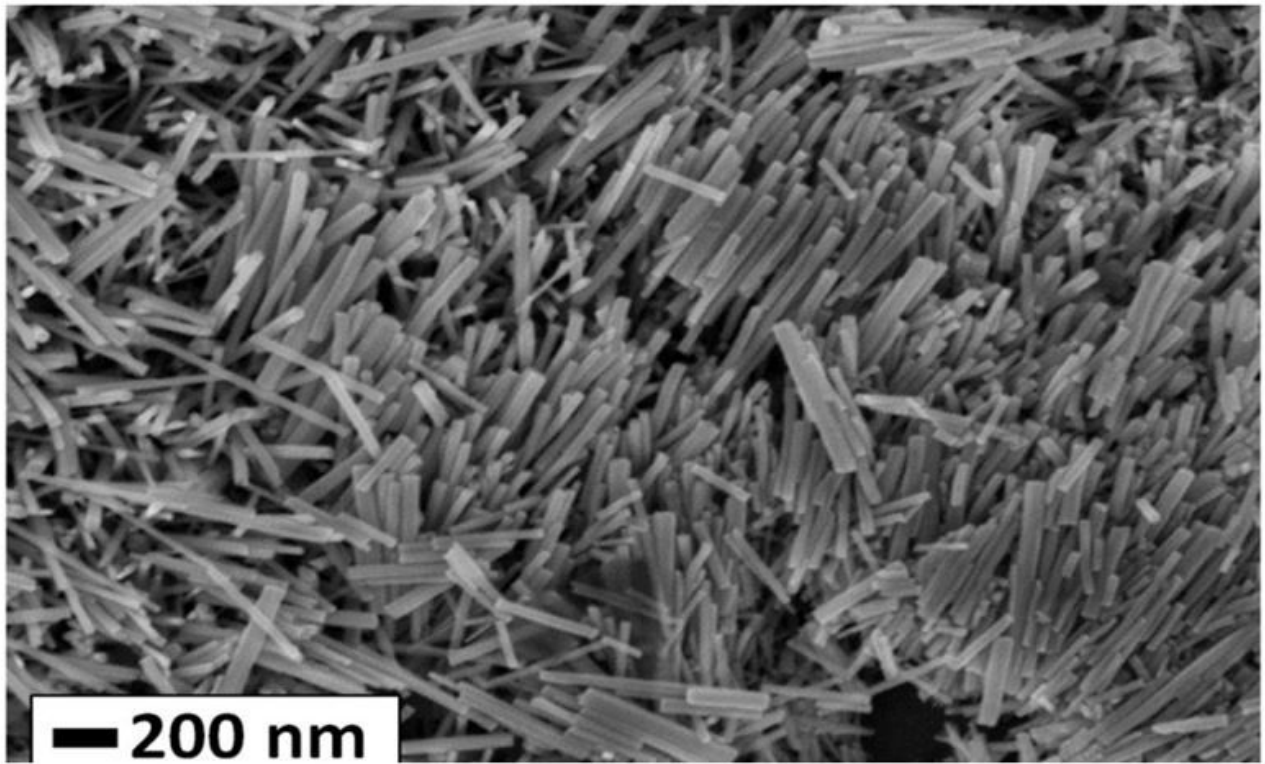


Figure 2

FESEM micrograph of ZnO nanorods grown on glass substrates.



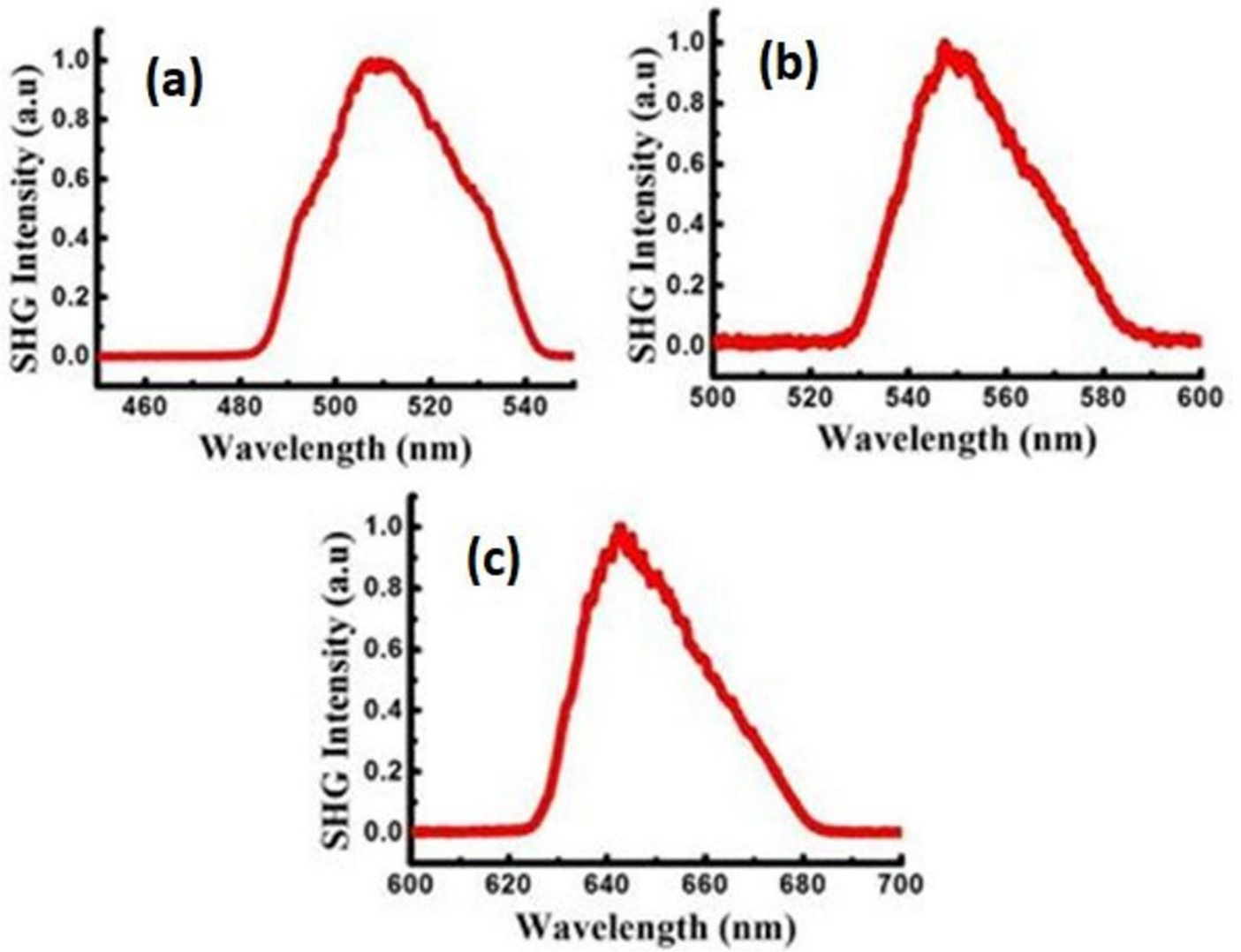


Figure 3

SHG spectra of fs pulses at central wavelengths of (a) 1000 nm, (b) 1100 nm and (c) 1300 nm.

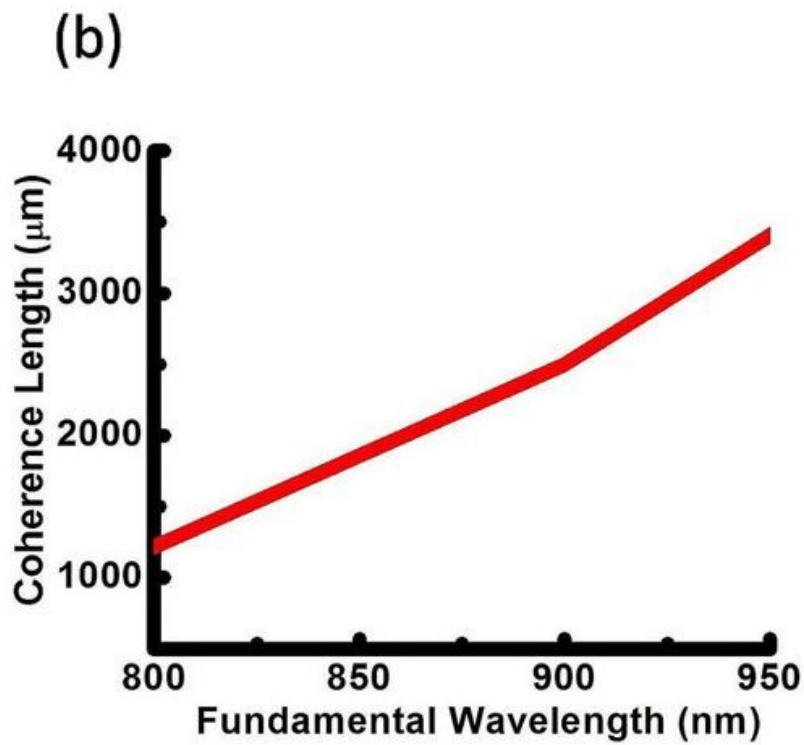
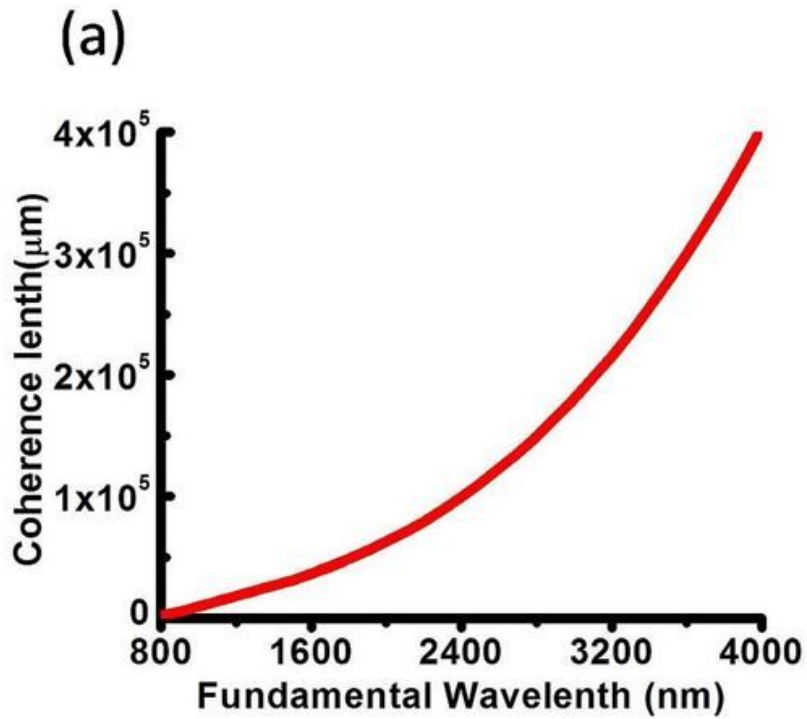


Figure 4

The plot of fundamental wavelength vs. coherence length in (a) long and (b) short wavelength range