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# Efficient second-harmonic and terahertz generation from single $BiB_3O_6$ crystal using nanosecond and femtosecond lasers

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**The paper reports the efficient UV and terahertz generation from a 1.29 mm thick and Type I, $\theta = 28.9°$ cut $BiB_3O_6$ (bismuth triborate, BIBO) crystal using femtosecond and nanoseconds laser pulses. We have employed 800 nm wavelength pulses of 50 and 140 fs obtained from a Ti:sapphire laser amplifier and oscillators at 1 kHz and 80 MHz repetition rates, respectively. The conversion efficiency of second-harmonic generation (SHG) was ~50% while that obtained for terahertz (THz) generations was of the order of $1.85 \times 10^{-5}$%. In addition, LDS-698 dye laser radiation tunable between 650–700 nm was also used as a source for SHG between the 325–350 nm range. The dye laser was pumped by SHG (532 nm) radiation from an electro-optically *Q*-switched Nd:YAG laser having a pulse repetition rate of 10 Hz and a pulse width of 10 ns. A conversion efficiency of 4.01% was obtained for generation of UV at 343.5 nm. Finally, we have measured the transmission, refractive index, absorbance, and conductivity properties of BIBO crystal in the THz domain. We also ascertained the coherence length, relative permittivity and reflectivity of the crystal.** 

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## 1. INTRODUCTION

There is a continuous need for growing new nonlinear optical materials with improvised linear and nonlinear optical properties which can cover a wide range of transmission between the deep-UV to far-infrared range. It is possible to generate efficient deep-UV to mid-IR radiation by employing different types of nonlinear frequency conversion processes such as second-harmonic generation (SHG), sum-frequency mixing and difference-frequency mixing (DFM), etc. Some of the well-known nonlinear crystals such as $KH_2PO_4$ (KDP), KD * P, $\beta$-$BaB_2O_4$ ($\beta$-BBO), $LiB_3O_5$ (LBO), LAP, $KTiOPO_4$(KTP), BNA, DAST, etc., are widely used for making efficient nonlinear devices. In addition, some of the selected inorganic and organic crystals such as BBO, KTP, DAST, BNA, etc., can also be used for the generation of powerful terahertz (THz) radiation using DFM, plasma generated from filamentation of femtosecond laser, and optical rectification techniques [1–6]. Among all the BBOs is the first very promising borate group crystal which has found potential applications in the generation of deep-UV to terahertz radiation due to its excellent optical and nonlinear properties. Bhar *et al.* have reported the best use of the phase-matching condition for efficient deep-UV–vis radiation using frequency mixing techniques. They have used *Q*-switched nanosecond pulse in their study and employed sum frequency generaton (SFG) and SHG techniques [7–9]. There are many more groups who have used femtosecond pulse for powerful UV generation from BBO crystal using the 800 nm wavelength [10,11]. The same femtosecond laser is also used for THz generation and measurement of refractive indices in the THz domain [12,13].

The $BiB_3O_6$ (bismuth triborate, BIBO) crystal was introduced by Hellwig *et al.* in 1998 [14–17]. It is a highly promising negative biaxial nonlinear optical crystal that belongs to monoclinic group $C_2$ and apart from having a large nonlinear optical coefficient and laser high damage threshold, it is nonhygroscopic as well. It offers an optical transparency between the 160 nm to 2.7 μm region. The UV transmission cutoff of BIBO is at much deeper wavelength than BBO and it offers large effective nonlinearity ($d_{\text{eff}} = 3.2$ pm/V). Its nonlinear coefficient is 3.5–4 times higher than that of LBO and 1.5–2 times higher than that of BBO. These attractive properties have recently been explored to demonstrate the potential of BIBO for efficient SHG using continuous wave [18,19], long pulse, nanosecond, picosecond [20,21], and femtosecond pulsed lasers [22–26]. For example, conversion efficiencies of SHG at 532 nm in the

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Full Length Article

# Tunable and low-threshold random lasing emission in waveguide aided Rhodamine-6G dye incorporated silica embedded thin films

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ABSTRACT

A simple and low-cost approach for producing tunable and low threshold RL emission in the visible region is reported by using a fluorescent laser dye as gain medium and light scattering is achieved in silica nanospheres (SNS) under the pump light of 532 nm, obtained by second harmonic generation of a Q-switched Nd: YAG laser fundamental radiation of 1064 nm wavelength. Depending upon the size ($a$) of the scatterer particles in comparison to the wavelength ($\lambda$) of the pump light, the scattering mechanism can be classified into different categories. However, to demonstrate various RL parameters for scatterer particles residing in Rayleigh scattering ($a < \lambda$) and Geometrical optics regime ($a >> \lambda$) in Rh6G dye doped PVA film, we have deliberately synthesized two different sized SNS (notably, 400 nm and 1000 nm). Also, to demonstrate the tuning in the RL emission by enhancement in pump photon density the gain medium has been enclosed within two glass slides. The performances of developed three RL systems, one made with bare film (S1), one cover with one glass slide (S2) and another one in which the gain medium is enclosed between two glass slides (S3) have been compared. It has been demonstrated experimentally that in the developed RL system with 400 nm SNS particles, RL emission in the incoherent regime is obtained. On the other hand, in the case of 1000 nm SNS particles, RL emission in the coherent regime is demonstrated. The tunable random lasing emission covering 585–592 nm wavelength regions with the lowest emission line-width of 4.2 nm and the lowest RL threshold of 1.59 mJ/cm$^2$ is obtained from the developed RL systems. The demonstrated low cost and simple strategy for the development of tunable RL devices provided here will find novel applications in laser-based imaging, RL based sensing, and other optoelectronic devices.

## 1. Introduction

Thanks to the recent progress in the industrial applications of CW and pulsed laser sources. Although conventional laser sources are proved to be very effective for their various applications in fundamental and applied research fields, but still there exist a lot of challenges [1–6]. Therefore, there is a recent surge in the development of laser sources and particularly a lot of impetus has been given in the development of random lasers (RLs) by employing various luminescent materials as gain media [1–4]. However, unlike conventional laser sources, which include (i) a pump source, (ii) a gain medium, and (iii) optical resonators, RLs use a different approach for its operation. Random scattering of light provided by jumbled nano/microstructures present inside a gain medium offers a substitution of optical cavity to achieve lasing [2–6]. The generation of light in RLs mainly relies on several factors, like geometrical configurations, optical properties of gain/scattering media, structural distinctions of the scattering particles etc. However, enclosing the whole random medium into a waveguide feedback configuration gives additional degrees of freedom to simultaneously tune the emission regimes along with decreasing the lasing threshold through various approaches, such as through pump volume amplification techniques, increasing the pump photon density via wave guiding, changing the cavity length ($l_c$), or by changing scattering strength [3–8]. Further, the optical feedback mainly depends on the scattering mean free path ($l_{sc}$) and transport mean free path ($l_t$) of a randomized system. Any change in $l_{sc}$ may also cause the transition of RL emission from incoherent to coherent regime and vice versa [5–9]. However, three distinct scattering regimes can be defined [7], as given below, depending upon the size of the nanoparticles/microparticles used as scatterer particles in RL systems.

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OPTICAL PHYSICS

# Emission peak shifted incoherent random laser through the combined effects of coupling of surface plasmons in a triangular shaped silver nanostructure, microbubbles, and the waveguiding mechanism

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**The random laser (RL) is now becoming an essential tool for various photonics applications, and a plethora of research advancements in RL coupled with developments in the field of techniques of syntheses of various nanostructured materials is taking place. But the realization of tuning the peak emission wavelength of RL is still very challenging. However, in this report we have demonstrated an emission peak shifted tunable low threshold incoherent RL in the visible region in a gain medium of a commercially available dye laser material and by employing the rarely used scatterer materials of triangular silver nanoparticles (TSN), microbubbles, and the waveguiding mechanism. The scattering properties of trapped microbubbles, along with the localized surface plasmon resonance property of TSN of appropriate concentration within waveguided thin films composed of glass substrates, have been methodically investigated to demonstrate the reduction in lasing threshold and tunability in the peak emission wavelength. A two-fold reduction in RL threshold by addition of TSN in the disordered system, along with a considerable narrowing down of the emission spectra to a few nanometers, are obtained. Furthermore, the peak emission wavelength shift of 6 nm is reported by suitably changing the system configuration by the addition of an optimum concentration of TSN along with trapped microbubbles. The as-developed system shows high-quality laser performance with the maximum value of $\eta = 0.64$, a quantity describing the ratio of the number of stimulated radiative photons within RL and the total number of emissive photons. We propose that the total internal reflections from the microbubble surface, along with plasmonic enhancement and scattering from the TSN, mediate the waveguided RL to achieve the low threshold. Therefore, this report is an early step towards demonstrating efficient RL in a ternary scattering system. Many more avenues for investigating this developing research issue may be helpful for the future development of affordable and robust optoelectronic devices.** 

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## 1. INTRODUCTION

The fascinating realm of random lasers (RLs), also designated as "mirror-less lasers," has inspired remarkable curiosity in the scientific community, leading to deep dives into the novel territory of the interaction of light with matter having randomness [1,2]. Since the first theoretical prediction given by Letokhov [3], random lasing action has been investigated in varieties of physical arrangements, gain media, and scatterer particles. Lasing action in a random medium is generally achieved by significantly expanding the trajectory of the light inside a volume, and amplification occurs due to multiple scattering, raising the gain length $l_g$ ($l_g$ is defined as the length over which the intensity of light is amplified by a factor $e$), owing to which the gain surpasses the overall loss in the system [4]. In contrary to conventional lasers, in RLs, strong light scattering by random scatterers present within the gain medium could improve the confinement of light that may result in the development of closed optical loops within the gain medium that are inherent to coherent RLs [5]. On the other hand, while light gets scattered multiple times by the scatterers placed within the gain medium, some selected modes may obtain a higher dwell time within the medium. Consequently, they can sustain over longer scattering

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Research Article

# Employment of nonlinear optical properties of GO/Ag nanocomposite scatterer materials for achieving random lasing in the visible region in the gain medium of a commercially available dye

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ABSTRACT

Graphene oxide (GO) and silver nanoparticles (AgNPs) are recently been widely employed in various sectors including in photonics applications due to their fascinating properties. On the other hand, huge interests are shown by the researchers for development of scatterer materials for achieving random laser (RL) with improved performances. In this work, the GO/Ag nanocomposite material comprising of GO nanosheets and AgNPs have been prepared and its remarkable nonlinear optical (NLO) properties are employed for the first time to demonstrate RL in the visible region by the introduction of disorder and multiple scattering within the amplifying medium of Rhodamine-B dye. Interestingly by changing the cuvette path lengths, here we have shown that the gain volume within the amplifying media can be varied effectively, which directly influence the lasing threshold. The larger value of NLO coefficients in GO/Ag scatterer induced greater refractive index contrast ($\Delta n$) between the scatterer and surrounding medium and thus enhances the light-matter interactions in the GO/Ag nanocomposite and consequently the lasing threshold for RL generation is reduced significantly by 50 % than that of bare GO as scattering center. This report opens an exciting prospect of using NLO properties of GO/Ag nanocomposite for achieving enhanced scattering in different gain media for demonstration of low threshold RL, which may revolutionize the future development of RLs.

## 1. Introduction

In recent decades, the field of photonics has witnessed a remarkable advancement, with a growing emphasis on the development of novel light sources and laser technologies. Among these developments, the phenomenon of random lasing has emerged as a captivating and promising area of research [1]. In random lasers (RLs), amplification of light is provided from multiple scattering events within the disordered configuration of dye and scattering medium [2]. Since, the theoretical prediction by Letokhov in 1967 [3], RLs have gained considerable attention in the scientific community. After the first experimental demonstration of coherent RL in ZnO [4], rigorous theoretical and experimental investigations on RLs have been carried out in different gain media [5–9]. The intrinsic properties of RL, such as low threshold, robustness against external perturbations, and potential for compact and versatile designs for chip-scale optoelectronic devices, have picked up the interest of researchers of inter-disciplines. Most importantly, the efficacy of RL depends on various nanostructures present within the disordered media. In this regard, scattering centers present within an active media is significant in determining its RL characteristics such as the lasing threshold, modal characteristics etc. Particularly, the size, geometrical shape, concentration of scatterers within the gain medium greatly impacts its emission characteristics. In recent years, there has been a growing demand of employing discrete categories of nano/micro-structures in gain media. Particularly, these scattering media including metal [10], semiconductor [11], metal organic framework [12], biological structures [13,14], liquid crystals [15], external feedbacks [16,17], micro-bubbles [18] etc., which have been extensively employed for generation of coherent [19] or white [20] RL emission, however with some drawbacks. Interestingly, the intrinsic nonlinearity in two-dimensional (2D) materials enables the manipulation of gain and absorption profiles, which plays a pivotal role in controlling the threshold for RLs. These 2D materials can be tailored to exhibit tunable and highly efficient nonlinear responses, affecting the

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# Enhanced optical power limiting and visible luminescence in colloidal dispersion of ultra-small Au nanoclusters synthesized by single-pot chemical technique

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ABSTRACT

Here, we present a detailed investigation on the synthesis and nonlinear multiphoton absorption properties of the colloidal solution of Au nanoclusters (AuNCs) which contains the atomic clusters, with the number of atoms per cluster (NAC) of only one ($Au_1$NC) and two ($Au_2$NC). These AuNCs are synthesized by an easy single step one-pot simple chemical process and by using dimethylformamide (DMF) both as reducer and stabilizer. The presence of both $Au_1$NC and $Au_2$NC are found in the sample by their distinct signature in the UV–Vis. absorption spectrum as well as in the high-resolution mass spectrum. The synthesized material has been found to exhibits a strong and stable blue-luminescence with a moderately high quantum yield (QY) of 12.4% when excited with UV light. The nonlinear optical two-photon absorption (2PA) properties of $Au_1$, $Au_2$NC solutions are being reported here by *Z*-scan studies, for the first time, by using both 10 *ns* and 100 *fs* pulse laser radiations having wavelength of 532 nm. It is significantly noted here that the synthesized AuNCs are found to exhibit reverse saturable absorption (RSA) when excited either by *ns* or by *fs* laser pulses. A high third-order nonlinear susceptibility ($\chi^{(3)}$) of the order of $10^{-13}$ (esu) of the synthesized materials are obtained under *fs* laser excitation and it is attributed to the 2PA through electronic band to band transition. In contrast, the variation of the 2PA coefficient ($\beta$) with input intensity ($I_0$) represents the footprint of free carrier involvement in the enhanced nonlinear absorption in the case of *ns* excitation. Furthermore, through the non-saturable nonlinear multiphoton absorption, the synthesized AuNCs exhibit excellent optical power limiting phenomenon with the limiting threshold ($F_{th}$) of 9.1 mJ/cm$^2$ (*fs* excitation) and 2.02 J/cm$^2$ (*ns* excitation) owing to their enhanced 2PA coefficient. Therefore, we believe that our synthesized ultra-small colloidal AuNCs can be used as the promising candidate material for advanced photonics application in the future.



## 1. Introduction

Thanks to the high polarizability of sub-nanometer AuNCs, having molecular-like electronic structures, due to the appearance of strong quantum size effect. Hence, a high-luminescence along with a very high nonlinear optical (NLO) response can be achieved in AuNCs due to the interaction with the strong electromagnetic field of an incident laser light [1–9]. However, the optical properties of AuNCs can be tuned elegantly by playing with the number of atoms per cluster (NAC). Hence, AuNCs have stimulated considerable interest in the research community having importance not only in fundamental research [1,2] but also for their potential applications in photonics [3], optical limiting [4], and optical switching [5]. Previously, Thomas et al. have demonstrated the enhancement in NLO response in AuNCs in comparison to that of Au nanoparticles (AuNPs) [4]. Mendez et al. have successfully demonstrated that the polarizability per atom for $Au_{34}$NC would be 5.59 Å$^3$, whereas it would increase to 7.15 Å$^3$ in case of $Au_6$NC [6]. In another study, Brevet et al. have reported that the value of the hyperpolarizability for $Au_{25}$NC is $109 \times 10^{-30}$ esu, which has been increased to $509 \times 10^{-30}$ esu for $Au_{15}$NC [7]. Therefore, one can expect that by reducing the value of NAC in AuNCs stronger NLO response can be achieved. Previously, nonlinear multiphoton absorption in Au and AgNCs with different NAC (=10, 15, 25, 38, 144, *etc.*) have been reported [8–11]. But till now there is no report on NLO properties of AuNCs with NAC being less than 10. The synthesis of stable AuNCs continues to remain a considerable challenge in compared to the synthesis of gold nanoparticles (AuNPs). Previously, syntheses of PAMAM [1], thiol [2], polyethylenimine [10] stabilized AuNCs have been reported.

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Research Paper

# Syntheses of flower and tube-like $MoSe_2$ nanostructures for ultrafast piezocatalytic degradation of organic dyes on cotton fabrics

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ABSTRACT

The synthesis of few-layered transition metal dichalcogenides (TMDCs) with abundant exposure of the active site, *viz.*, is an important key to achieve excellent dye degradation performance. Here, we have reported synthesis and ultrafast dye degradation performance of flowers-like $MoSe_2$ nanostructure (FMN) with ~230 nm in diameter and its transformation to tube-like $MoSe_2$ microstructure (~1 $\mu$m in length) by tuning the solvothermal reaction time. The piezoelectric devices are developed using the FMNs delivers the highest open-circuit voltage of ~ 2.12 V, which is ~21 times higher than that of the developed device with the tube-like $MoSe_2$ microstructure. The piezoelectric property of the synthesized samples has been judiciously utilized further for ultrafast degradation of organic dyes within 60–120 s only under the low-frequency (40 kHz) ultrasonication vibration in the dark. The estimated dye degradation efficiencies of the FMNs-based piezocatalyst are found to be ~86% and 85% for degradation of Rhodamine B (RhB) and methylene blue (MB) dye within the 60 s, respectively. Also, the FMN has exhibited an excellent piezocatalytic dye degradation capability for RhB-MB dye mixture and dye loaded on a cotton fabric with an efficiency of ~98% (60 s) and 84% (120 s), respectively. The piezocatalytic dye degradation mechanism of FMNs has also been explained theoretically.

## 1. Introduction

The existing water is being polluted gradually by different toxic dyes coming from the different textiles industries and environmental pollutants. Therefore, the treatment of organic dyes present in wastewater has fascinated significant and long-term consideration (Borgarello et al., 1981; Kabra et al., 2004). The wastewater treatment is also a major problem area in energy research (Borgarello et al., 1981; Kabra et al., 2004). Recently, researchers are extensively utilizing semiconductor nanoparticle-based photocatalysts to degrade organic pollutants by creating strong oxidizing free species under light illumination (Sharma et al., 2009; Xiao et al., 2021). But due to the limitation of the band gap matching of the semiconductor photocatalysts with the illuminated light irradiation, the photocatalytic dye degradation is not applicable at all times. Additionally, for fast degradation, the semiconductor nanomaterial must show a high capability to generate and separate electron–hole ($e^-$–$h^+$) pairs under optical irradiation. So far, a number of strategies such as doping, use of high reaction temperature, and designing of heterostructured materials have been tried to enhance photocatalytic dye degradation (Ajmal et al., 2014). However, these materials and methods are also limited due to their low solar energy conversion efficiency (<20%), and low light transmission in intensely dyed toxins (Banin et al., 2021). Therefore, novel environment-friendly, recyclable, highly efficient methods and materials are necessary for forthcoming wastewater purification techniques. Mechanical energy is a sustainable abundant natural energy that can be harvested by employing piezoelectric materials (Wu et al., 2018, 2016, 2017; Mushtaq et al., 2018; Lan et al., 2017; Lin et al., 2017). Piezoelectric materials can produce an electric field in reaction to an external force. A built-in electric field powerfully increases the separation of free carriers (Wu et al., 2018, 2016, 2017; Mushtaq et al., 2018; Lan et al., 2017; Lin et al., 2017). Thus, piezoelectric materials have been widely used in photocatalytic dye degradation. Thus, the innovative studies associated to piezoelectric water treatment are paid significant attention (Wu et al., 2018, 2016, 2017; Mushtaq et al., 2018; Lan et al., 2017; Lin et al., 2017). Recently, researchers have investigated the piezocatalytic

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