

## Laser-Ablation-Synthesized Ag/Zno-Dye Nanocomposite for Linear and Nonlinear Optical Applications

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#### **Research Article**

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### Abstract

In this research, the effect of zinc oxide nanoparticles ZnO, Ag\_ZnO on the optical properties of absorption and emission was studied, as the emission spectrum of the rhodamine B dye was fluorescent with the addition of nanoparticles of ZnO, Ag\_ZnO, and the nonlinear optical properties of three dyes: rhodamine B, methylene blue, and saffron dye. The nonlinear optical response of saffron dye with Ag\_ZnO B compound showed the highest nonlinear response upon impact. For the 405 nm laser, the nonlinear refractive index is large. This coincides with the maximum absorption of the dye, which explains the increased nonlinear refractive index. Interestingly, the addition of zinc nanoparticles increased the nonlinear response of all dyes and demonstrated greater nonlinear properties for Ag\_ZnO. The addition of silver improved the nonlinear response of the dyes, as the Ag\_ZnO structure strongly absorbs most of the visible spectrum and even the near-infrared region of the electromagnetic spectrum. Which gives greater absorption efficiency. The nonlinear refractive index was good when adding ZnO and Ag\_ZnO nanoparticles.

**Keywords:** Zinc Oxide-Silver Nanoparticles; Optical Properties; Nonlinear Optical Response; Rhodamine B; Methylene Blue; Nonlinear Refractive Index

**Abbreviations:** NLO: Nonlinear Optical Properties; LAL: Laser Ablation in Liquid; SPR: Surface Plasmon Resonance; EMW: Electromagnetic Wave.

### Introduction

Metals and their oxide nanoparticles at the nanoscale Have a significant interest across various applications due to their interesting properties resulting from quantum confinement effects, which delete alter their physicochemical characteristics compared to bulk materials [1,2]. These metal nanoparticles have been used in diverse fields, including optics, sensors, and photocatalysis, owing to their unique features such as optical properties, large surface areas, and crystalline structures, metal nanoparticles are used wide range of applications [3,4]. For instance, they can be utilized as optical limiter materials to protect a highly sensitive optical components from the intense energy of lasers and keep their nonlinear optical properties (NLO). Nanostructured materials have these properties where they can act as passive optical limiters by reducing or blocking optical transmittance when expose to laser energy that exceed a specific thresholds [5]. Zinc oxide (ZnO) is an inorganic material and recognized for their interesting properties like particle size, morphology, surface charges and UV illumination; therefore, they have been used in many applications [6]. Incorporating silver (Ag) into ZnO nanostructures had a significant attention because they offer unique attributes [7,8], especially for biomedical applications due to their antimicrobial properties [9]. Moreover, ZnO nanoparticles (NPs) are effective photocatalysis and can degrade hazardous industrial wastewater dyes [10]. Silver nanoparticles (Ag NPs) share similar properties with ZnO NPs, and their combination enhance their biomedical and therapeutic effects. For instance, bimetallic nanoparticles exhibit antimicrobial activity and excellent photocatalytic properties for wastewater treatment [11]. Laser ablation in liquid (LAL) technique allows synthesis of metal and metal oxide nanoparticles without the need for metal precursors, reductants, or capping agents [12,13]. Heterocatalysts, including various metal oxides and metal sulfide heterojunction photocatalysts (e.g., SnO2, Bi2WO6, CuO2, TiO2, CdS), have been employed for color degradation [14]. Semiconductor photocatalysts, like ZnO, are favored due to their stability and tunable bandgap. ZnO, with its 3.2 eV bandgap, has gained widespread recognition as a potent photocatalyst, offering strong oxidation capabilities and being readily available and non-toxic compared to alternative materials [15]. ZnO Has high photo oxidation capabilities make it a preferred choice for photocatalytic applications [16]. On the other hand, zinc oxide (ZnO) as a photocatalyst has limitations due to Having a large band gap, which necessitates ultraviolet (UV) light for photon absorption with wavelengths below 387 nm, and rapid charge carrier recombination, Leading to limit its utility in wastewater treatment [17]. In this study, we investigated the linear and nonlinear optical properties of ZnO and ZnO-Ag nanoparticles and their effects on the linear and nonlinear optical properties of organic dyes, such as rhodamine B, methylene blue, and saffron. Linear properties were

examined through absorption and fluorescence spectroscopy techniques, while nonlinear properties were studied using the Z-scan technique [18]. Laser dyes are complex molecules with diverse ring structures, leading to complex absorption and emission spectra.

#### **Experimental Part**

Two different colloidal solutions were synthesized in order to investigate the effect of the ZnO and Ag-ZnO nanoparticles on the optical properties of Rhodamine B, methylene blue, and saffron dyes. First, ZnO nanoparticle colloid was prepared by laser ablation of Zn plate with a thickness of 1 mm and a purity of 99.99% in distilled water. Then, Ag-ZnO NPs were synthesized by ablation of Ag target in freshly prepared ZnO NPs colloidal solution. The laser ablation process was carried out using an Nd: YAG laser that provides 5 ns laser pulses with a wavelength of 1064 nm and a repetition rate of 4 Hz. The laser beam was focused horizontally to the surface of the target kept inside the distilled water. The laser energy was 700 mJ and the ablation process was carried out for 700 number of pulses. Each dye of Rhodamine B, methylene blue, and saffron were mixed with ZnO and Ag-ZnO NPs separately. The linear optical properties, fluorescence emission and nonlinear optical properties of dyes and prepared NPs were analyzed by UV-Vis spectroscopy, fluorescence emission spectrometry and z-scan techniques respectively. Morphology and article size of ZnO and Ag-ZnO NPs were analyzed using TEM (Figures 1-3).





### **Results and Discussion**

# Characterization of Colloidal ZnO and Ag\_ZnO Solution

During the laser ablation process of Zn plate, the deionised water at first appeared colorless and transparent but after a few minutes it began to change to light grayish-white which confirms the generation of ZnO nanoparticles (Figure 4). This is due to the surface plasmon resonance (SPR) Of ZnO nanoparticles, after introducing Ag metal through laser ablation, the color of the colloidal material changes to yellow-brown. This color change is attributed to the formation of Ag\_ZnO. This was also confirmed by TEM analysis as shown in Figure 5 which gave the structural morphology of the Ag\_ZnO core–shell particles. The darker part in image was considered to be the Ag core particles and the light color place is the ZnO shell layer. Shows the

statistical analysis of particle size distribution, and that both ZnO and Ag-ZnO nanoparticles are spherical in shape and are not agglomerated. The average size of ZnO nanoparticles measures approximately 5.77 nanometers, while the average size of Ag\_ZnO NPs is around 8.39 nanometers.



Figure 4: ZnO and Ag\_ZnO nanoparticle solution.



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## Linear Optical Studies: Absorption and Fluorescence Emission

**Linear absorption of ZnO, Ag\_ZnO NPs:** The UV-vis absorption spectra of Ag \_ ZnO nanoparticles are shown in Figure 6. The spectrum of pure ZnO nanoparticles shows a broad spectrum from 300 to 400 and exhibits a distinct

peak in the absorption spectrum at 248 nm. Pure ZnO nanoparticles show a UV absorption edge at 325 nm. Another spectrum shows a Ag\_ZnO solution and the absorption spectrum appears to contain two absorption peaks, the first at a wavelength of 243 nm and the other at a wavelength of 404 nm.



We can distinguish characteristic ZnO peak in the UV region between 300–400 nm due to intrinsic absorption when electrons transition from the valence band to the conduction band [19]. The Figure 6 shows the absorption spectrum of nanoparticles of ZnO and Ag\_ZnO composition based on the absorbed wavelength. It is clear from Table 1 that the composite ZnO nanocore has an absorption peak in the UV region. While the Ag\_ZnO structure strongly absorbs most of the visible spectrum and even the near-infrared region of the electromagnetic spectrum, the absorption spectrum of coreshell zinc oxide with silver (Ag-ZnO) can be explained by the presence of silver nanoparticles on the surface of the zinc oxide core. The introduction of silver nanoparticles enhances the absorption properties of the composite material, allowing it to absorb a wider range of wavelengths, including UV to IR regions of the electromagnetic spectrum [20,21]. The formation of a silver shell over the zinc oxide core creates a synergistic effect between the two materials, resulting in improved electromagnetic wave (EMW) absorption performance [22].

NPs	Absorption Peak Value	λ <sub>max</sub> (nm)	α(cm <sup>-1</sup> )
ZnO	0.784287	248	60.21
Ag_ZnO	0.269627	404	20.7

**Table 1:** Absorption spectrum characteristics of ZnO, Ag\_ZnO NPs.

**Absorption of rhodamine B, methylene blue and saffron dyes before and after mixing with ZnO and Ag\_ZnO NPs:** The effect of delete ZnO and Ag\_ ZnO nanoparticles (NPs) on the absorption spectra of rhodamine B, methylene blue and saffron dyes is shown in Figure 7.

It is obvious from Figure 7 below that both ZnO and Ag-ZnO nanoparticles affect the absorption spectrum peaks of all dves where the absorption spectra of all dyes were decreased by adding both ZnO and Ag-ZnO nanoparticles. The absorption spectra of Rdh B, MB and SAF dyes were decreased by adding both ZnO, Ag\_ZnO nanoparticles, which indicates that the presence of nanoparticles affects the absorption spectrum peaks of the dyes. This can be attributed to the interaction between the Ag nanoparticles and the dye molecules, leading to a decrease in the intensity of the absorption peaks. When the nanoparticle concentration is higher than the dye concentration, the dye absorption spectrum decreases due to the interaction between the plasmon absorption of silver nanoparticles and the absorption of dye molecules. This reaction results in a decrease in the absorption spectrum of the dye, as evidenced by an increase in optical density. The exact mechanism of this interaction and its impact on the absorption spectra would require A variety of processes can result in quenching, such as excited state reactions, energy transfer, complex-formation and collisional quenching (Table 2) [23].



Figure 7: Optical absorption spectra of dyes before and after addition of ZnO NPs, Ag\_ ZnO NPs.

Samples	Absorption Peak Value	λ <sub>max</sub> (nm)	α(cm <sup>-1</sup> )
rhodamine B	2.903	549	222.86
rhodamine B + ZnO NPs	2.706	553	207.716
rhodamine B + Ag_ZnO NPs	2.642	545	202.804
methylene blue	2.789	658	214.113
methylene blue +ZnO NPS	2.204	665	169.203
methylene blue + Ag_ZnO NPS	1.909	664	146.513
Saffron	2.097	249	160.973
Saffron +ZnO NPS	1.406	250	107.919
Saffron + Ag_ZnO NPS	1.792	252	137.539

**Table 2:** Absorption Spectra of Dyes in the Presence of ZnO and Ag\_ZnO NPs.

**Effect of ZnO, Ag\_ZnO NPs on the Fluorescence of Rhodamine B, Methylene Blue and Saffron dyes**: Rhodamine B dye dissolved in water showed a fluorescence emission when excited with 587 nm light. It is clear from

Figure 8 that the fluorescent emission intensity of the dyes Enhanced by adding ZnO, Ag\_ZnO nanoparticles summarize the fluorescence of Rhodamine B dye before and after addition of nanoparticles.



It is clear from Table 3 that the fluorescence of dyes was increased by adding ZnO and Ag-ZnO nanoparticles. The fluorescence spectra showed that the highest fluorescence intensities of both pure rhodamine B dye and the dye solution with Ag\_ZnO nanoparticles were observed at wavelengths between 550 and 650 nm. The higher fluorescence intensity of the dye solution with Ag\_ZnO nanoparticles can be explained by FRET. FRET occurs when a donor molecule in an excited state transfers its energy to an acceptor molecule in the ground state. In this case, the Ag\_ZnO nanoparticles act as the donor molecules, while rhodamine B dye molecules act as the acceptor molecules. The excited rhodamine B dye molecules transfer their energy to nearby Ag\_ZnO nanoparticles, which can then re-emit the energy as photons of light. This leads to a higher fluorescence intensity in the presence of ZnO-Ag nanoparticles [24]. Through the above interpretation, the results confirm that nanoparticles of ZnO and Ag\_ZnO can enhance the fluorescence emission intensity of the rhodamine B dye, through plasmon interaction of the metal surface. This result indicates that the ZnO shell was used to maintain a certain distance between the fluorescent

mole cule and the metal core, which caused the interaction between the surface plasmon and excitation light field to produce a FL emission enhancement. Furthermore, the ZnO shell layer also could enhance the FL emission of dye, which was caused by the change in photonic mode density and/or reduction in self-quenching of fluorophores for ZnO nanostructure [25].

Dye	λ <sub>max</sub> (nm)	Fluorescence
rhodamine B	584	284.88
rhodamine B + ZnO NPs	587.8	752.02
rhodamine B + Ag_ ZnO NPs	586.9	413.81

**Table 3:** Fluorescence Spectroscopy rhodamine B Data (a)ZnO NPs, (b) ZnO\_Ag NPs.

Methylene blue dye that dissolved in water showed fluorescence emission upon excitation with 664 nm light. It is clear from Figure 9 that the fluorescent emission intensity of the dyes was decreased by adding ZnO, Ag-ZnO NPs only.



The reduced fluorescence of the dye solution with nano\_Ag\_ZnO can be explained by a phenomenon called "fluorescent polarization". Fluorescence polarization was observed in the dye solution with nano-ZnO-Ag due to the attachment of methylene blue dye molecules to the Ag\_ZnO

nanoscale surface. When methylene blue absorbs light, it moves to a higher energy level. As the electrons in methylene blue are bound to the Ag\_ZnO nanosurface, they can absorb the absorbed light, preventing the electrons from returning to the base energy level quickly. As a result, less fluorescent light is released, leading to the lower fluorescence of the dye solution with nano-Ag\_ZnO (Table 4).

Dye	λ <sub>max</sub> (nm)	Fluorescence
methylene blue	697	95.54
methylene blue + ZnO NPs	695.9	94.82
methylene blue + Ag_ZnO NPs	693.1	71.6

**Table 4:** Emission spectrum characteristics of methyleneblue dye before and after adding (a) ZnO NPs, (b) Ag\_ZnONPs.

### **Nonlinear Optical Analysis**

The results in Table 5 showed that both ZnO and Ag\_ZnO nanoparticles have negative nonlinear refractive index. At a wavelength of 405 nm, the nonlinear refractive index (NNRI) of Rhodamine B and methylene blue And saffron dyes was studied before and after the addition of ZnO and Ag\_ZnO nanoparticles. The results showed a nonlinear refractive index and third-order nonlinear sensitivity at 405 nm for all dyes. The results confirmed that the addition of ZnO and Ag\_ZnO improved the nonlinear properties of rhodamine B and methylene blue dye. As shown for the refractive index values listed in Figure 10.



As can be seen from Figure 7b, the NNRI of Rdh B and methylene blue dye was increased by adding ZnO NPs because the wavelength of laser (405 nm) lies in the absorption spectrum of ZnO NPs. This led to an increase in NNRI of rhodamine B and methylene blue dye by heat transfer from ZnO NPs to the dye solution which enhances the thermal Kerr effect [26]. As seen from Figure 10, the NNRI of Rdh B and MB dye was increased after adding Ag\_ZnO NPs under the influence of 405 nm. Laser wavelength can be absorbed by Ag\_ZnO NPs because its wavelength (405 nm) lies within the surface Plasmon band of Ag\_ZnO NPs added to Rdh B and MB dye, this led to an increase in NNRI of MB dye by heat transfer from ZnO NPs to the dye solution. This means that the photon energy was converted to heat when the surface Plasmon band of Ag nanoparticles in dye solution was excited by irradiation of a 405 nm laser leading to increasing the NNRI [27]. As shown in Table 5 of the absorption spectrum, the introduction of silver nanoparticles enhances the absorption properties of the composite material, allowing it to absorb a wider range of wavelengths, including UV to IR regions of the electromagnetic spectrum. This led to the enhancement of the nonlinear properties of the dyes by adding Ag\_ZnO NPs more than ZnO NPs.

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Samples	P(Mw)	$I_0 (MW/m^2)$	L <sub>eff</sub> (m)	ΔΤ	n <sub>2</sub> (m2/W)
ZnO NPs	13.5	91.056	0.00096743	2.103448	3.91*10 <sup>-12</sup>
Ag_ZnO NPs	12.74	85.9301	0.00096961	1.537669	3.02*10 <sup>-12</sup>
Rdh B	13.5	91.056	0.00098994	1.60559	2.92*10 <sup>-12</sup>
Rdh B + ZnO	13.5	91.056	0.00098301	1.832727	3.36*10 <sup>-12</sup>
Rdh B +Ag_ZnO	13.27	89.505	0.00099022	1.834064	3.39*10-12
MB	13.3	89.707	0.00099672	0.877841	1.61*10 <sup>-12</sup>
MB +ZnO	13.27	89.505	0.00098909	1.092205	2.02*10-12
MB +Ag_ZnO	13.27	89.505	0.00099839	0.958917	1.76*10 <sup>-12</sup>
saffron	2.75	18.548	0.0009572	1.591954	1.47*10 <sup>-11</sup>
SAF+ ZnO	2.75	18.548	0.00097957	1.645325	1.48*10 <sup>-12</sup>
SAF+ Ag_ZnO	2.75	18.548	0.0007244	2.021482	1.84*10-11

 Table 5: Nonlinear properties of rhodamine B, methylene blue And saffron dye before after adding ZnO, Ag\_ZnO NPs.

### **Conclusions**

The study investigated the absorption, fluorescence, and nonlinear optical properties of rhodamin B, methylene blue, and saffron dves in the presence of ZnO and ZnO-Ag nanoparticles. The addition of nanoparticles significantly affected the optical properties of the dyes. For instance methylene blue showed decreased absorption due to fluorescent polarization. The presence of ZnO, ZnO-Ag nanoparticles enhanced the fluorescence emission of rhodamin B, the behaviour showed that fluorescence polarization and fluorescence enhancement can be achieved by attaching dye molecules to nanoscale surfaces and nanoparticles, respectively. The fluorescence properties of the dye solutions were found to be influenced by their interactions with the nano-ZnO-Ag and ZnO-Ag nanoparticles. The addition of ZnO, Ag\_ZnO nanoparticles to rhodamine dye random lasers leads to improved characteristics and a lower lasing threshold. The plasmonic enhancement induced by nanoparticles increases energy transfer and fluorescence quenching for MB. Saffron did not exhibit significant fluorescence emission. The study also found that ZnO and ZnO-Ag nanoparticles improved the nonlinear optical properties of all three dyes. The presence of silver nanoparticles in ZnO-Ag nanoparticles induced surface Plasmon resonance, leading to enhanced nonlinearity at 405 nm for all dves. The study concludes that the addition of nanoparticles can significantly modify the optical properties of dyes, making them more suitable for various applications, including photovoltaic devices, solar energy conversion, and random lasers media.

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