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COMPARISON STUDY OF THE NONLINEAR OPTICAL PROPERTIES OF Al AND Cu NANOPARTICLES PREPARATION VIA PULSE LASER ABLATION

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Abstract. The nonlinear optical properties of aluminum (Al) and copper (Cu) nanoparticles synthesized via pulsed laser ablation in liquid were systematically investigated to determine their suitability for advanced photonic applications, particularly optical limiters. Understanding the differences in their nonlinear responses is crucial for selecting the most effective material for laser protection and optical switching devices. Experimental results revealed that Cu nanoparticles exhibit a negative nonlinear refractive index, indicating a strong self-defocusing effect, along with pronounced reverse saturable absorption. These characteristics contribute to efficient suppression of high-intensity laser beams. In contrast, Al nanoparticles demonstrated a positive nonlinear refractive index, corresponding to self-focusing behavior, with nonlinear absorption mechanisms differing significantly from those of Cu. The comparative analysis confirmed that Cu nanoparticles deliver superior optical limiting performance due to their favorable absorption and transmission behavior under varying laser intensities. Overall, the study highlights Cu nanoparticles as a highly promising candidate for optical limiter instruments and related nonlinear optical applications.

Keywords: Nonlinear properties, Al and Cu Nanoparticles, Pulse Laser Ablation.
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1 Introduction

Nonlinear optics is one of the Subspecialty of modern optics (Corless et al., 1997), with the important application of solid-state dye lasers that eliminate many of the common issues associated with static or flowing liquid systems (Mahdi et al., 2023). Pulsed laser ablation is a method that is used for the preparation of nanoparticles in which specific parameters are used to synthesize nanoparticles from pure metallic materials, such as the number of pulses, wavelength, and energy of pulse (Patra et al., 2018). In recent years, pulsed laser ablation in liquid (PLAL) has gained popularity as a top-down method for generating nanoparticles (Ibrahim et al., 2023). Laser ablation of metal targets dissolved is free from pollution in contrast to chemical routes to the synthesis of metal nanoparticles in solution (Al-Helaly et al., 2025). Laser parameters play a major role in determining the shape and size of the nanoparticles produced for a particular purpose (Irhayyim & Hussain, 2023). These parameters include wavelength, laser power, and pulse duration (Hussain et al., 2020). In recent years, many studies have been conducted to prepare nanoparticles using pulsed laser ablation techniques from noble metals such as gold, silver, and copper, and to study their optical and structural properties. Thus, for their affordability, high conductivity, and widespread availability compared to noble metals, copper and aluminum nanoparticles have garnered significant attention (Musa et al., 2016; Mahdi et al., 2023). These metal nanoparticles are primarily utilized in optical, electronic, and catalytic applications due to their distinctive properties. Additionally, they demonstrate excellent surface plasmonic characteristics within the ultraviolet and visible spectrum range (Chiad et al., 2023; Chiad et al., 2021). There are several works of literature on copper and aluminum nanoparticles fabricated by laser ablation technique. Miranda et al. produced copper nanoparticles by laser ablation in liquid and investigated the surface-enhanced Raman scattering of the nanoparticles (Hu et al., 2015). In this study, the nonlinear optical properties of copper and aluminum nanoparticles prepared in distilled water by pulsed laser ablation will be compared. The study aims to determine which of these nanoparticles is the best for optical applications.

2 Non-Linear optical properties

Sheik-Baha, in 1990, employed a measuring technique in order to find the nonlinear refraction of thin samples. It is referred to as Z-scan technique (Globe & Hussain, 2020). If a high-intensity laser beam travels through an example, this alters the refractive index induced such that the laser beam acts as self-focusing or defocusing. Nonlinear refractive index was calculated based on the peak-valley change of the normalized transmittance using the following equations (Shanon & Sh, 2016):

$$n_2 = \Delta\Phi_o / I_o \, L_{\text{eff}} k \tag{1}$$

where $k = 2\pi/\lambda$, λ represents the wavelength of beam. L_{eff} represents the specimen's effective length and $\Delta\Phi_o$ represents the nonlinear phase.

$$\Delta\Phi_o = \frac{\Delta T_{p-v}}{0.406} \tag{2}$$

Intensity at focal spot I_o determined by:

$$I_o = \frac{E}{\Delta t \, \pi \omega_o^2} \tag{3}$$

where E is the pulse energy, Δt represents the pulse duration. L_{eff} is the sample effective length and can be estimated by:

$$L_{\text{eff}} = \frac{1 - e^{-\alpha_o L}}{\alpha_o}, \tag{4}$$

where L is sample length and α_o the linear absorption coefficient.

The normalized rate in transmitted intensity can be approximated using the following equation (Shannon & Sh, 2016):

$$\beta = \frac{2\sqrt{2}\Delta T}{I_o L_{\text{eff}}}. \tag{5}$$

3 Z scan technique

Zee scan is a very common method for the investigation of nonlinear optical properties and used in the estimation of the sign and magnitude of the coefficient of nonlinear absorption and the nonlinear refractive index. Two processes are involved in the method: The closed aperture is indicated in the Fig. 1. The beam converges in the pre-focal area and approaches the aperture if the sample's nonlinear refractive index n2 is negative (Al-Helaly et al., 2025). The coefficient of nonlinear absorption β is measured using the open aperture. Multiple photons being absorbed when the laser intensity is big adequate or due to other nonlinear phenomena naturally results in nonlinear absorption (Hussain et al., 2020; Shanon, 2023)

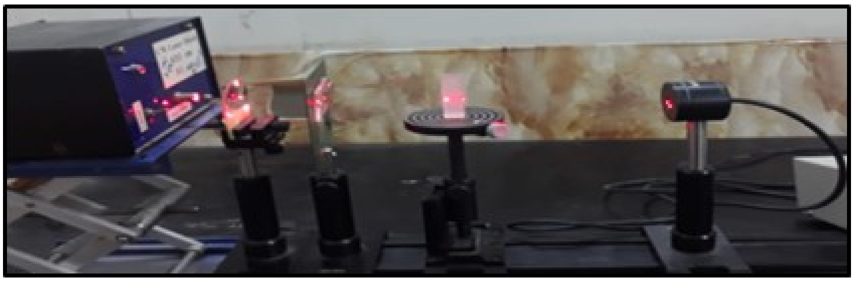

Figure 1: Three-point bending test results of hybrid FMLs

4 Results and Discussions

4.1 Preparation of Cu, Al nanoparticles in distilled

In this method, Cu and Al nanoparticles were prepared via pulsed laser ablation technique. A Q-switched Nd-YAG laser system (Fig. 2.) was used, delivering pulses at a wavelength of 1064 nm with a maximum pulse energy of 800 mJ, a pulse width of 10 ns, a repetition rate of 6 Hz, and the diameter of effective beam equal to 8 mm. The number of pulses applied for Cu and Al nanoparticles was 300, 500, and 1000. A high-purity (99.99) copper and aluminum target was placed at the bottom of a glass vessel containing 10 mL of distilled water for the ablation process. The absorption spectra (SPE spectra) of the nanoparticle solution were analyzed by dual-beam UV-VIS spectrophotometers. Fig. 3. presents the FESEM images of Cu and Al nanoparticles, showing particle sizes ranging from 31 to 55 nm.

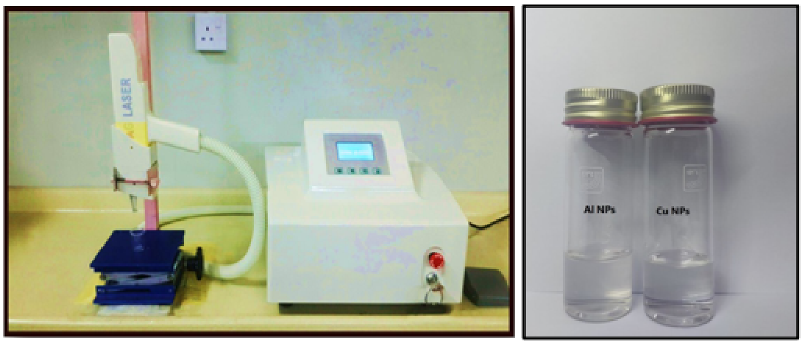

Figure 2: Experimental setup for nanoparticles synthesis by PLAL

4.2 Fourier Transform Infrared Spectroscopy FTIR

Fourier Transform Infrared Spectroscopy (FTIR) is a powerful technique for identifying the chemical composition of materials. When a molecule is exposed to infrared radiation, it absorbs specific energy levels corresponding to the vibrational frequencies of its chemical bonds. These absorptions appear as peaks in the spectrum, with each peak representing a distinct vibration of a particular bond. Typically, peaks in the 3500–3200 cm-1 region indicate the presence of hydrogen bonds (O–H), while peaks in the 3000–2800 cm-1 range correspond to C–H bond vibrations. Additionally, peaks appearing in the 1700–1600 cm-1 region usually signify the

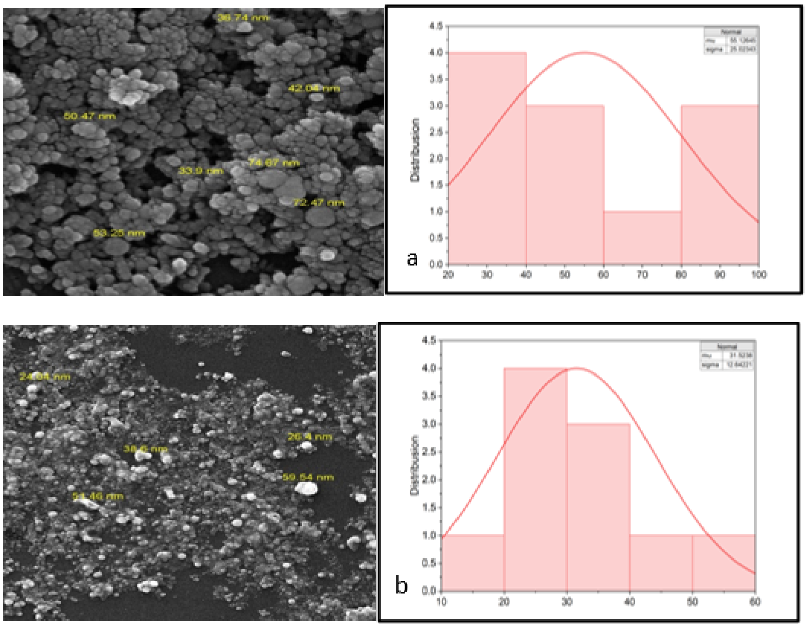

Figure 3: FFSEM and Distribution for a. Cu b. Al Nanoparticles

presence of a carbonyl group (C=O). These characteristic peaks are observed in both copper and aluminum nanoparticles, as illustrated in Fig. 4.

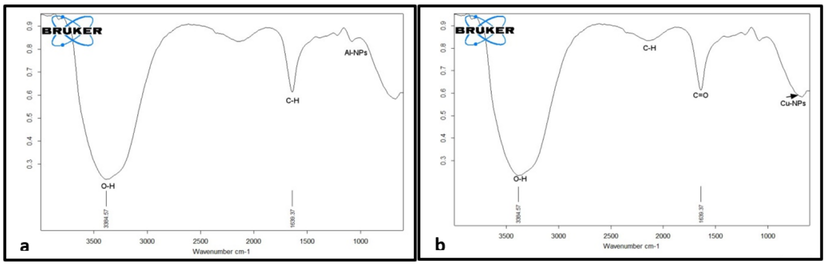

Figure 4: Fourier Transform Infrared Spectroscopy a. Al b. Cu Nanoparticles

4.3 Linear optical properties

The UV and visible spectra were determined by SPR. The absorption and transmittance spectra of the copper Fig. 5a. and aluminum Fig. 5b. nanoparticles colloids were measured using a quartz cell. The maximum wavelength (λ max) of maximum absorption was measured. While the absorption coefficient shown in Fig. 6.

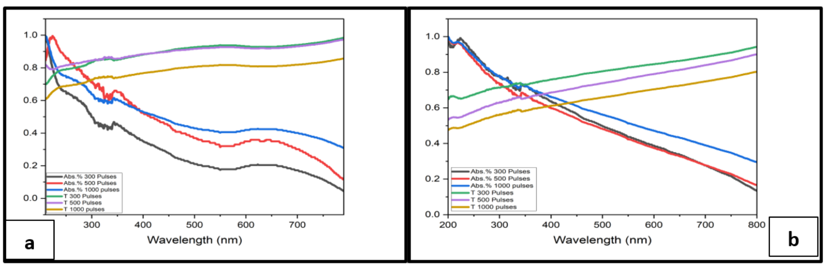

Figure 5: UV and visible absorption spectra Vs Transmission for a) Cu NPs. b) Al NPs

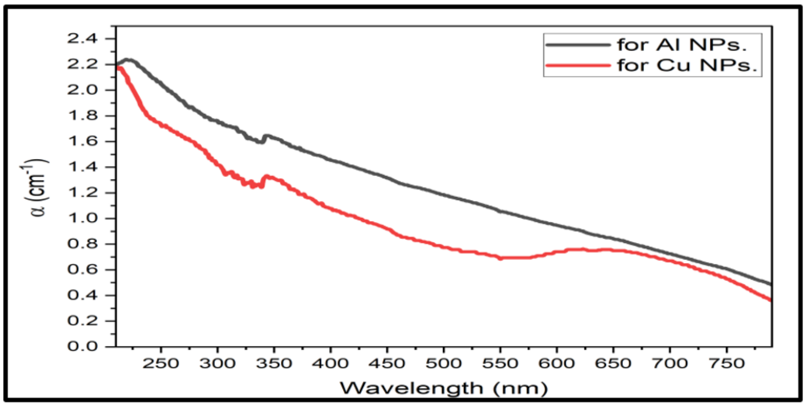

Figure 6: Absorption coefficient for Cu and Al NPs

As illustrated in (Fig. 4a.) and (Fig. 4b.), the Increasing in concentration of nanoparticles (number of laser pulses) conducive to an increase in the plasmonic resonance, where the absorption spectra peaks appear in the ultraviolet range due to quantum size effects. The figure shows the absorption and plasmonic resonance peaks position for Cu at 325 nm and 650 nm, and for Al nanoparticles at 350 nm, in which the absorption curve is shifted to the shorter wavelength of the blue color for Cu NPs, which confirms the formation of Cu nanoparticles, while the absorption curve for Al shows a shift of the spectrum towards the light-yellow wavelength. Fig. 6. shows the difference in the absorption coefficient curve of Cu and Al nanoparticles. The peak in the curve for Cu nanoparticles is at a longer wavelength compared to Al nanoparticles. This shows that the surface electrons of Cu nanoparticles resonate at a lower frequency in response to red and orange light, while the surface electrons of Al particles resonate at a higher frequency in response to blue and violet light. This is very important in optical applications such as sensors or solar cells.

4.4 Non-Linear optical properties

In these examinations, the particles size of the Cu nanoparticles achieved, are known to be around (50.25, 32.04, 23.4) nm at (300,500,1000) pulses. While the particles size of the Al nanoparticles are (73.67, 60.69, 50.04) nm at (300,500,1000) pulses The above results indicate in Table 1. The Transmission of close and open aperture Zscan was measured by Diode laser with wavelength 650 nm and power 50 mW.

Table 1. The nonlinear parameters for Cu and Al nanoparticles

NPs.	No. of Pulse	$\Delta\Phi_o$,rad	$n_2 \times 10^{-12}$ cm^2/W	$B \times 10^{-7}$ cm/W	NPs.	No. of Pulse	$\Delta\Phi_o$,rad	$n_2 \times 10^{-13}$ cm^2/W	$B \times 10^{-7}$ cm/W
	1000	1.293	1.34	2.42		1000	0.95	9.91	2.85
Cu	500	1.066	8.06	2.31	Al	500	0.76	5.79	1.95
	300	1.231	9.06	2.05		1000	0.67	5.03	2.01

Figure 7,8 and 9 explained the normalized transmission of both Cu and Al nanoparticles specially fig 9a explained the difference between the results it was showed that the Cu nanoparticles peak to valley exhibited negative nonlinear refractive index (self-defocusing), while the curve of normalize transmission of Al nanoparticles exhibited positive nonlinear refractive index (self-focusing). In the other hand Fig 9b explained the curve of normalize transmission exhibited saturation absorption for Cu NPs. and two photon absorption for Al NPs.

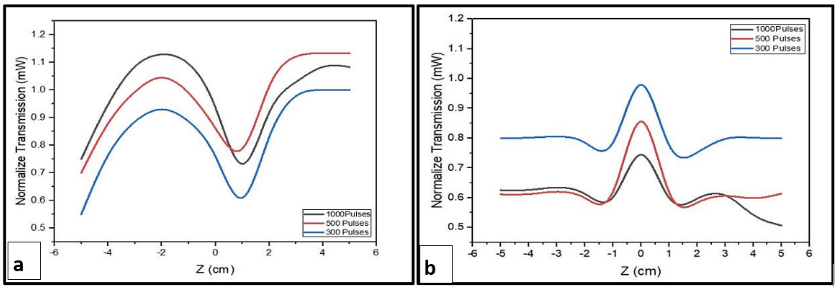

Figure 7: Z scan at 1000, 500 and 300 pulses for Cu NPs. a. Close aperture b. Open aperture

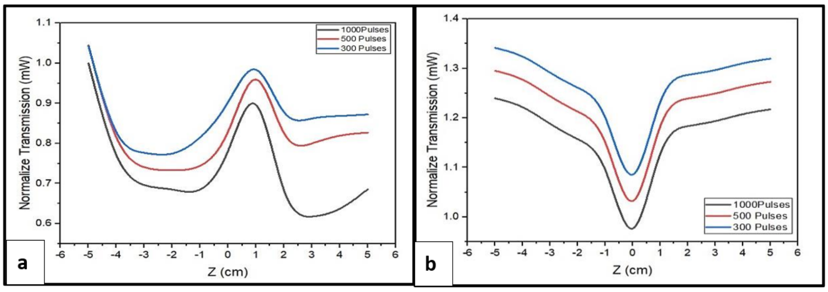

Figure 8: Results by Z scan at 1000, 500 and 300 pulses for Al NPs. a. Close aperture b. Open aperture

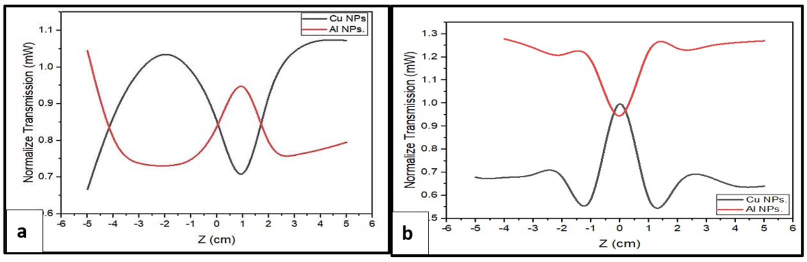

Figure 9: Comparison the results of Z scan for Al NPs. a. Close aperture b. Open aperture

5 Conclusion

In this study, copper and aluminum nanoparticles were successfully synthesized using pulsed laser ablation in distilled water and characterized to compare their linear and nonlinear optical responses. UV–Vis measurements confirmed the formation of well-dispersed nanoparticles with size-dependent plasmonic resonance peaks, where Cu nanoparticles exhibited SPR bands at 325 nm and 650 nm, while Al nanoparticles showed a dominant band near 350 nm. These spectral positions reflect the different electronic structures of Cu and Al and demonstrate the strong influence of laser pulse number on particle concentration and quantum size effects.

FESEM analysis showed that Cu nanoparticles decreased in size from 50 nm to 23 nm with increasing pulse number, whereas Al nanoparticles ranged from 74 nm to 50 nm. These variations in particle size directly influenced the optical behavior. FTIR results further confirmed the presence of characteristic vibrational bands associated with O–H, C–H, and C=O groups on the nanoparticle surfaces, indicating partial oxidation and surface functionalization.

The nonlinear optical investigation using the Z-scan technique revealed a clear contrast between the two materials. Cu nanoparticles exhibited a negative nonlinear refractive index, indicating self-defocusing behavior, accompanied by saturable absorption in the open-aperture scans. In contrast, Al nanoparticles showed a positive nonlinear refractive index (self-focusing) with two-photon absorption as the dominant nonlinear absorption mechanism. These differences arise from variations in particle size, electronic structure, and plasmonic resonance of the two metals.

Overall, the comparative results demonstrate that Cu nanoparticles are more suitable for applications requiring saturable absorbers, optical switching, and laser pulse shaping, while Al nanoparticles exhibit strong nonlinear refraction and two-photon absorption, making them promising candidates for photonic devices, optical power limiting, and super-resolution imaging systems. These findings confirm that PLA-synthesized metal nanoparticles offer tunable and material-dependent nonlinear optical properties, making them valuable for advanced optical and optoelectronic applications.

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