

Research Article

On the Kerr Effect and Optical Bistability of Graphene Based Spherical ZnO@Ag Core-Shell Nanocomposite

Gashaw Beyene* 

Department of Applied Physics, Adama Science and Technology University, Adama, Ethiopia

Abstract

The drawback of one type of material can be modified by incorporating it with another suitable material. One of the mechanisms to enhance the properties and overcome the drawbacks of a material is by forming a core-shell nanostructure. Core-shell nanocomposites are one of the most preferable structures used to overcome the drawbacks of one material and enhance the properties of another. Graphene monolayer (GML), zinc oxide (ZnO), and silver (Ag) nanomaterials are highly recommended for optical-based applications and for enhancing the properties of other materials. Using a quasi-static approximation framework, we examine the optical response of a graphene monolayer-based ZnO@Ag spherical core-shell nanocomposite embedded in a highly compatible dielectric host matrix. The numerical study revealed that optical factors such as the dielectric function of the host medium, filling factor, volume fraction, and graphene's Fermi energy greatly impact the nanocomposite's local field enhancement factor, optical bistability, and hysteresis response. The graphene monolayer-wrapped ZnO@Ag spherical core-shell nanocomposite exhibits a strong nonlinear optical response in the visible spectral region. A low bistability threshold in the visible range was achieved by adjusting key parameters and utilizing Kerr-nonlinear graphene with silver as the outer shell, which enhances the nonlinear effect. These findings imply that core-shell nanostructures made with two-dimensional materials such as graphene have a unique advantage for plasmonic and optical device optimization, data storage application.

Keywords

Graphene Monolayer, Optical Bistability, Local Field Enhancement Factor, Hysteresis

1. Introduction

Due to their optimized properties, core-shell nanoparticles (CSNPs) have a wide range of potential applications in optics, nanomaterials, biomedicine [1, 2], chemical and biological sensors [2, 3], magnetic nanocomposites, chemical engineering [3, 4], catalysts [3, 4], environmental science [1, 5], solar cells [6, 7], pharmaceuticals [8], ultraviolet detectors, optical switching, and nanoscale electronic systems. Consequently, considerable efforts are being devoted to their study

through both theoretical and experimental investigations.

By combining two or more different materials and tuning key parameters, the structural, optical, and photocatalytic properties can be significantly enhanced. We previously reported that these unique features arise from the confined spatial distribution of polarization charges on the nanostructure's surface, induced by the interaction of the composite with the electromagnetic field. This interaction is strongly amplified

*Corresponding author: gashaw.beyene@astu.edu.et (Gashaw Beyene), gashaw4nuclear@gmail.com (Gashaw Beyene)

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by the surface plasmon resonance (SPR) phenomenon [9-13], which originates from the collective oscillation of conduction electrons in the noble metal shell that effectively couple with the incident electromagnetic field and propagate along the surface [14].

Core-shell nanoparticles (CSNPs) can be composed of metals, semiconductors, dielectrics, organic or inorganic materials, or a combination of these [15-17]. Due to the exciton-plasmon interaction, ZnO@noble-metal CSNPs, in particular, possess a wide range of potential applications in emerging technologies [18]. Surface coating with noble metals such as Ag, Au, Cu, and Pt has been found to significantly modify the structural and optical properties of ZnO nanocrystallites, thereby enhancing their practical applications.

Among various noble metal nanoparticles, silver (Ag) was selected as the shell material for ZnO nanoparticles due to its exceptional electrical, optical, electronic, and catalytic properties, as well as its high bactericidal activity, biocompatibility, efficient solar energy conversion, strong field enhancement, non-toxicity, and cost-effectiveness.

At ambient temperature, wurtzite-structured ZnO exhibits a wide direct band gap ($E_g = 3.37$ eV), a high exciton binding energy (60 meV), and strong transmittance in the near-ultraviolet region. It has attracted extensive research interest due to its size-dependent electronic, optical, photochemical, and luminescent properties, as well as its abundance, tunable morphologies, non-toxicity, flexibility, compatibility, and environmental friendliness [19, 20]. Owing to these properties, ZnO nanoparticles are considered promising materials for various applications, including photocatalysis [21-24], photodetectors [24], gas sensors [24], piezoelectric sensors, light-emitting diodes (LEDs) [22, 24], quantum devices [22], photovoltaic cells, UV optical switches [18], and ultraviolet lasers.

Furthermore, the nonlinear optical (NLO) effects of the composite, arising from the interaction between incident light and nonlinear optical materials, offer a wide range of potential applications. Owing to their relevance in biological imaging, ultrafast optical switching, optical limiting, and optoelectronic devices, third-order optical nonlinearities in various core-shell nanostructures have recently been investigated in considerable depth [25].

Graphene is a carbon-based monolayer material widely used in plasmonic devices, optoelectronic switches, photodetectors [25, 26], optical antennas, light-emitting diodes (LEDs) [25], and solar cells [27, 28]. It possesses outstanding optical and nonlinear properties [27]. In such nanocomposites, a graphene monolayer exhibits a particularly strong optical nonlinear Kerr effect [21, 29]. By integrating monolayer graphene into ZnO@Ag core-shell nanostructures, enhanced nonlinear optical characteristics can be achieved.

For example, in nonlinear optical systems, optically induced bistability (OIB) enables two distinct output light intensities for the same input intensity and holds great promise for applications in optical logic functions [30, 31], met-

amaterials, all-optical switching [32, 33], memories [30, 34, 35], amplifiers [34], optical transistors [33, 35], and low-power lasing. Various types of core-shell nanostructures exhibiting optical bistability have been explored, including nonlinear shell-coated metallic nanoparticles (NPs) [36], nonlinear monolayer graphene-coated dielectric materials [28], nonlinear dielectric cores coated with metallic NPs [37], and graphene-wrapped metal core-dielectric shell structures [38].

To the best of our knowledge, optically induced bistability and local field enhancement in nonlinear monolayer graphene coated on a ZnO core with a noble metal silver outer shell have not yet been reported in the visible spectral range. Therefore, we are motivated to investigate the optical response of graphene-based ZnO@Ag core-shell nanostructures to tailor their properties and expand their potential applications by exploiting the unique characteristics of the ZnO core and Ag shell nanoparticles.

In this work, we investigate the optical bistability and local field enhancement of plasmonic metal-coated ZnO nanoparticles with a graphene monolayer acting as a spacer. We propose a graphene-based composite in which semiconductor ZnO nanoparticles covered by a graphene monolayer are randomly embedded in a linear host medium, with an outer silver shell forming spherical core-shell structures having radii in the range of 25-40 nm. As previously discussed, silver nanoparticles are preferred as a coating material to enhance the local field in ZnO nanoparticles at the resonance frequency through the surface plasmon resonance (SPR) effect.

Using the quasi-static approximation, we calculated the parameters that determine the composite's optical response, including its effective third-order nonlinear coefficient, a key parameter governing the material's nonlinear behavior.

The structure of this paper is as follows: Section 2 employs the electrostatic approximation to provide a theoretical description of the graphene-based ZnO@Ag core-shell nanocomposite. Section 3 presents the numerical analysis and results, and Section 4 concludes the study.

2. Theoretical Model

As illustrated in Figure 1, the proposed theoretical model consists of a spherical ZnO core covered by a graphene monolayer and an outer silver shell, embedded in a non-absorptive, linear host matrix. The radii of the core and the composite (core + shell) are denoted as r_c and r_s , respectively, with a geometric volume ratio defined as $\beta = 1 - (r_c/r_s)^3$. In this configuration, the graphene monolayer (GML) exhibits Kerr-like nonlinear surface conductivity and is positioned between the ZnO core and the Ag shell. The other key parameters depicted in the figure are the dielectric functions of the core, shell, and host matrix, denoted as ϵ_c , ϵ_s , and ϵ_h , respectively.

When an electromagnetic field is incident on the composite core-shell nanoparticle, polarization induces an electric field

within the system. The distribution of the electrostatic potential, Φ , associated with the induced field both inside and outside the nanoparticle can be determined by solving the Laplace equation, $\nabla^2\Phi = 0$, in spherical coordinates under the quasi-static (QS) approximation, assuming that the dimensions of the nanostructure are much smaller than the wavelength of the incident light [17].

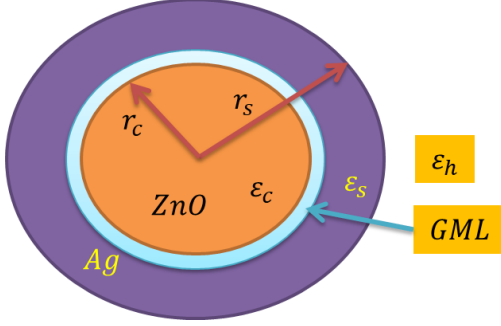


Figure 1. Schematic description of graphene based spherical ZnO@Ag core-shell nanocomposite.

If the spherical core-shell nanoparticle is located at the origin of a spherical coordinate system within a uniform, static electric field polarized along the z -axis, and assuming azimuthal symmetry, the potential distribution in the three regions can be expressed as:

$$\Phi_1(r, \theta) = -A_1 E_0 r \cos\theta, \quad r < r_c \quad (1)$$

$$\Phi_2(r, \theta) = -E_0 \left(A_2 r - \frac{B_1}{r^2} \right) \cos\theta, \quad r_c < r < r_s \quad (2)$$

$$\Phi_3(r, \theta) = -E_0 \left(r - \frac{B_2}{r^2} \right) \cos\theta, \quad r_s < r \quad (3)$$

The graphene monolayer is treated as a conducting film with surface conductivity, as it is a two-dimensional electromagnetic material with an extremely small thickness (≤ 1 nm) relative to the radius of the spherical inclusion. Consequently, for the monolayer graphene-spaced ZnO@Ag spherical core-shell nanostructure, the non-source-free boundary conditions are applied [28]. The boundary conditions at the inner and outer surfaces are used to determine the coefficients A_1 , A_2 , B_1 and B_2 . In particular, the coefficient A_1 characterizes the optical response of the ZnO core. It can be demonstrated using Eqs. (1)-(3) along with the appropriate boundary conditions that:

$$A_1 = \frac{9\epsilon_h \epsilon_s}{C_1 + C_2 \Omega} \quad (4)$$

where

$$C_1 = (\epsilon_c + 2\epsilon_s)(2\epsilon_h + \epsilon_s) + 2\beta(\epsilon_c - \epsilon_s)(\epsilon_c - \epsilon_h),$$

$$C_2 = 2[(\epsilon_c + 2\epsilon_s) - \eta(\epsilon_c - \epsilon_s)],$$

$$\Omega = i \frac{\sigma}{\omega \epsilon_0 r_s}.$$

The surface conductivity of the monolayer graphene can be expressed as follows, treating it as a one-atom-thick structure and accounting for its nonlinear, local tangential field-dependent behavior [39]:

$$\sigma \approx \sigma_0 + \sigma_3 \langle |E_c|^2 \rangle_g. \quad (5)$$

The frequency-dependent surface conductivity of graphene, σ_0 , is expressed as the sum of the intraband σ_{intra} and interband σ_{inter} contributions, given by the following expressions [28, 40], assuming the absence of an external magnetic field and applying the random-phase approximation:

$$\sigma_0 = \sigma_{intra} + \sigma_{inter}, \quad (6)$$

$$\sigma_{intra} = i \frac{e^2 k_B T}{\pi \hbar^2 (\omega + i/\tau)} \left[\frac{E_F}{k_B T} + 2 \ln \left(\exp \left(-\frac{E_F}{k_B T} \right) + 1 \right) \right] \quad (7)$$

$$\sigma_{inter} = i \frac{e^2}{4\pi \hbar} \ln \left[\frac{2E_F - (\omega + i/\tau)\hbar}{2E_F + (\omega + i/\tau)\hbar} \right]. \quad (8)$$

The incident field frequency, Fermi energy, electron-phonon relaxation time, temperature, elementary charge, Boltzmann constant, and reduced Planck constant are denoted by, E_F , τ , T , e , k_B , and $\hbar$, respectively. When the Fermi energy E_F is significantly larger than the photon energy, interband transitions in graphene can be neglected in comparison to the intraband contribution.

Graphene is nonlinear material, so, the nonlinear conductivity coefficient σ_3 is given by [28],

$$\sigma_3 = -i \frac{9}{8} \frac{e^2}{\pi \hbar^2} \frac{(e v_F)^2}{E_F \omega^3}, \quad (9)$$

where v_F Fermi velocity of electron.

The relationship between the local electric field in the core E_c and outside the inclusion E_h in the composite is provided by,

$$E_c = A_1 E_h. \quad (10)$$

From the average field theory of composite materials one can relate E_0 , E_h and E_c by the following relation [28],

$$f \langle E_c \rangle + (1 - f) \langle E_h \rangle = E_0, \quad (11)$$

where $f = 4\pi N a^3 / 3$ is the filling factor and N is the number of NPs per unit volume.

Squaring Eq (11) and plugging its values given by Eqs. (4) and (10) can be written as

$$Y = b_3 X^3 + b_2 X^2 + b_1 X, \quad (12)$$

where

$$Y = \left| \frac{ev_F}{E_F \omega} \right|^2 |E_0|^2, \quad X = \left| \frac{ev_F}{E_F \omega} \right|^2 |E_c|^2, \quad b_3 = \left| \frac{C_2}{8\epsilon_s \epsilon_h} \right|^2 (1-f)^2 \frac{e^4 E_F^2}{\pi^2 \hbar^2 \omega^2},$$

$$b_2 = \frac{2}{|9\epsilon_s \epsilon_h|^2} \text{Re}[C_2 a_1] (1-f) \frac{9e^2 E_F}{8\omega \pi \hbar^2}, \quad b_1 = \left| \frac{a_1}{9\epsilon_s \epsilon_h} \right|^2,$$

$$a_1 = 9\epsilon_s \epsilon_h + C_1(1-f) + C_2(1-f) \left(i \frac{\sigma_0}{\omega r_s \epsilon_0} \right).$$

The field enhancement factor is the ratio of applied electric field intensity to electric field intensity around the core-shell nanoparticle. The local field enhancement factor $|F|^2$ is calculated using Eqs. (1) and (4), as detailed in [15];

$$|F|^2 = |A_1|^2 = \left| \frac{9\epsilon_h \epsilon_s}{C_1 + C_2 \Omega} \right|^2 \quad (13)$$

3. Numerical Analysis

When the frequency of the incident radiation is close to the surface plasmon resonance frequency, the local electric field within the core-shell nanostructure can be significantly enhanced. This phenomenon has been studied in the context of optically induced bistability (OIB), revealing that the amplification occurs predominantly at a single resonant frequency. Additionally, when the frequency of the incident electromagnetic wave approaches the metal's surface plasmon frequency, semiconductor@noble-metal nanoparticles exhibit another notable feature: an anomalous enhancement of the local electric field.

The surface plasmon (SP) is strongly influenced by the size, shape, distribution, and host matrix of metal nanoparticles [15].

3.1. Local Field Enhancement Factor

Figure 2 shows the local field enhancement factor for two

parameters with fixed values of $v_F = 1 \times 10^6$ m/s, $E_F = 0.4$ eV, $T = 300$ K, $\tau = 1 \times 10^{-13}$ /s and composite radius $r_s = 40$ nm.

For this portion, we neglect σ_3 , for the surface conductivity of graphene monolayer $\sigma_0 \gg \sigma_3$ in order to reduce the complexity of the computation.

The difference in dielectric properties between the core and the surrounding medium, together with the surface plasmon resonance of the noble metal shell, are two factors that enhance the local field enhancement factor (LFEF). Figure 2(A) illustrates the effect of the aspect ratio (volume ratio) on the LFEF for a fixed composite radius. As the volume ratio increases, the LFEF decreases, which is associated with the reduction of plasmon resonance on the metal-coated surface.

The electric field within the composite decreases as the volume ratio of the core to the shell increases. The surrounding host medium also influences the optical properties of the core-shell composite; as shown in Figure 2(B), the local field enhancement factor (LFEF) increases with the dielectric constant of the host medium.

For sub-wavelength spherical core-shell nanoparticles, the field enhancement factor is determined by the relative distance from the center of the core or shell, the properties of the surrounding environment, and the volume ratio of the inner to outer material, rather than by the absolute size of the nanoparticle.

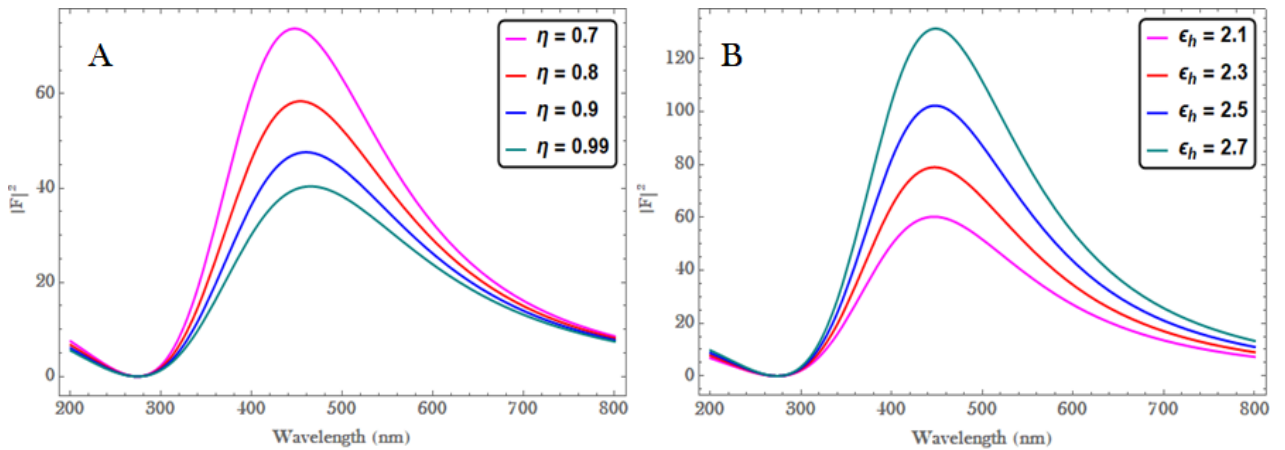


Figure 2. Local field enhancement factor of graphene based spherical ZnO@Ag core-shell nanoparticle for different volume fraction ($\epsilon_h = 2.25$ [10]) A) and host matrix ($\eta = 0.7$) (B).

3.2. Optical Induced Bistability

The nonlinear component of graphene conductivity (σ_3) plays a crucial role in the observation of optically induced bistability (OIB) in graphene-based core-shell nanoparticles [30]. Figures 4 and 5 depict the average local electric field within the core as a function of the externally applied field for different parameter values. The OIB can be controlled and tuned by adjusting these parameters.

Optically induced bistability (OIB) is represented in the $Y - X$ plane by S-shaped curves, showing that a single value of $|E_0|^2$ can correspond to three distinct values of the local field $|E_c|^2$, which is characteristic of OIB.

For the fixed parameters indicated in Figure 2, OIB is sensitive to the Fermi energy, as illustrated in Figure 3(A). Reducing the Fermi energy of the monolayer graphene increases the bistability threshold (unstable region indicated by dashed lines) and enlarges the hysteresis loop for the metal core coated with monolayer graphene and a dielectric shell.

The increase in instability leads to the characteristic switching-up and switching-down behavior of a hysteresis cycle. The Fermi energy of graphene strongly influences OIB, with a decrease in this parameter resulting in a higher bistable switching-up threshold field. The Fermi energy of embedded graphene plays a crucial role in achieving a broader bistability [41]. While various nanostructures with strong local field effects in the nonlinear regime have been extensively explored to achieve low bistable thresholds through enhanced

nonlinearity, here we achieve the desired effect using a graphene monolayer by increasing its Fermi energy in the visible region via plasmonic metal coating.

As the Fermi energy decreases, the hysteresis area (the region between the two arrows of the same color) increases, as shown in the figure. The plasmonic resonance of the metallic shell influences the optical bistability of the composite, and the plasmonic resonance itself is affected by the shell thickness [42]. Moreover, the interaction between bonding and antibonding charges on the structure's surface changes as the thickness of the plasmonic shell increases or decreases.

As shown in Figure 3(B), for a constant aspect ratio, the composite radius increases from 25 nm to 40 nm, corresponding to shell thicknesses of 4.08, 4.90, 5.72, and 6.53 nm, respectively. Consequently, the surface plasmon resonance is enhanced, which in turn amplifies the nonlinear effects of the graphene monolayer.

Reducing the thickness of the metallic shell enhances the strong electron-electron interactions, which are a source of nonlocality, and can significantly lower the optical bistability switching threshold in both the near and far fields, indicating the potential for nonlocality-enhanced nonlinear optical devices [43]. As the sizes of both the core and shell increase, the local electric field within the composite also increases, accompanied by an expansion of the hysteresis area, as illustrated in Figure 2(A). Our results indicate that a low bistable threshold is achieved at a moderate composite size.

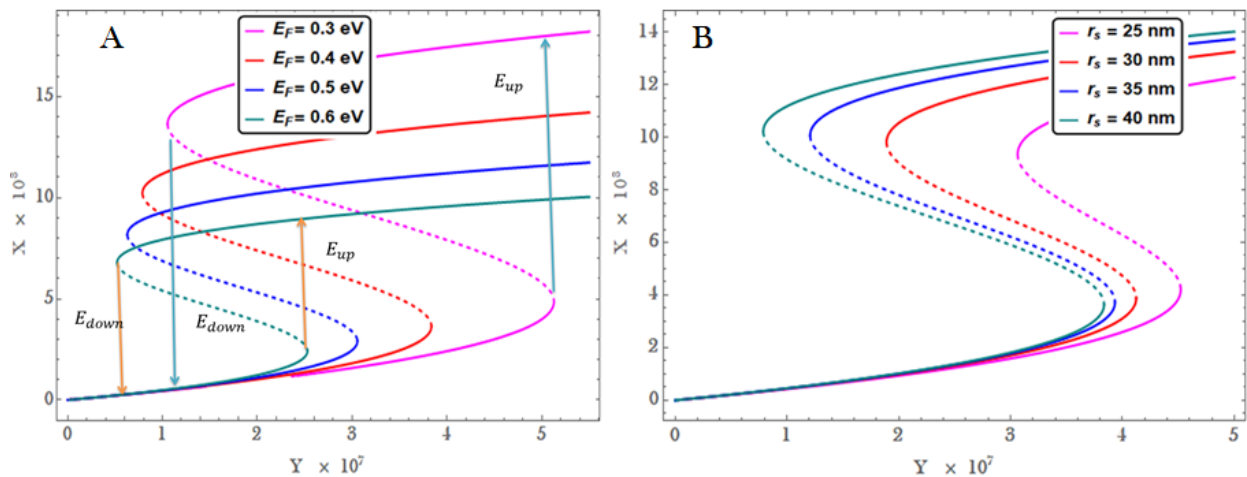


Figure 3. Optical bistability curves $|E_c|^2$ as a function of the incident applied field $|E_0|^2$ for a ZnO@Ag core-shell nanoparticle at 430 nm incident light wavelength for different values of Fermi energy ($r_s = 40$ nm)(A) and size of composite ($E_F = 0.4$ eV)(B).

In addition to the parameters discussed above, the density of composites embedded in a given region of the host medium and the type of medium itself also influence the optical bistability of the composite. Figure 4 illustrates the effects of the host medium and inter-composite interactions at an incident wavelength of 430 nm, with all other parameters held constant

as in Figure 3.

The interaction between arrays of composites embedded in the host medium is strong, as shown in Figure 4(A), due to a reduction of plasmon resonance on the metallic surface. The filling factor represents the number of composites present per unit area of the host medium. At low external field strengths,

the switching thresholds decrease as the filling factor increases; in this case, the local field enhancement within each composite is reduced. As illustrated in Figure 4(B), the

switching thresholds also decrease as the dielectric constant of the host medium increases.

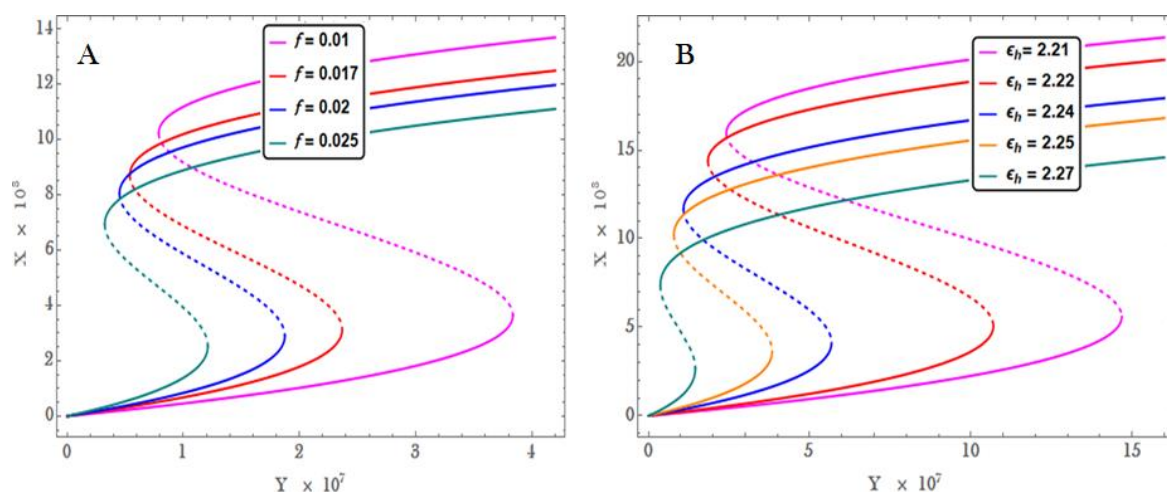


Figure 4. Optical bistability curves $|E_c|$ as a function of the incident applied field $|E_0|$ for a ZnO@Ag core-shell nanoparticle for different values of filling factor (A) and DF of host matrix (B).

According to the result and as reported in different papers, the threshold and hysteresis loops of OIB expand with increasing Fermi energy [30]. A higher Fermi energy corresponds to an increased electron concentration, which is responsible for enhanced absorption in the studied composite [44, 45].

4. Conclusion

Finally, we theoretically examined the effect of Kerr non-linearity on the local field enhancement factor and optical induced bistability of a monolayer graphene-wrapped ZnO core with a silver outer shell spherical nanostructure. Our results indicate that monolayer graphene exhibits a very strong nonlinear optical response in the visible range. The host medium, array concentration, Fermi energy, and metallic shell thickness all influence the local field enhancement factor, optical bistability, and hysteresis behavior of graphene-based ZnO@Ag core-shell nanostructures. A high local field is achieved when these factors are optimized. Specifically, a low bistable threshold is obtained by using a composite with a relatively large shell thickness, increasing the Fermi energy, array concentration, and the dielectric constant of the host medium. Due to the tunable optical response of this composite, it is a promising candidate for applications in optical switching, optical transistors, optical filters, optical memory, and other plasmonic devices.

Abbreviation

LFEEF Local Field Enhancement Factor

NLO	Nonlinear Optics
SPR	Surface Plasmon Resonance
CSNP	Core-shell Nanoparticles
Eg	Band Gap Energy
GML	Graphene Monolayer
OIB	Optically Induced Bistability
QS	Quasi Statics
LED	Light Emitting Diodes

Author Contributions

Gashaw Beyene is the sole author. The author read and approved the final manuscript.

Conflicts of Interest

The author declares no conflict of interest. The funders had no role in the design of the study; in the collection, analysis, or interpretation of data; in the writing of the manuscript, and in the decision to publish the results.

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