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Nonlinear Optical Properties of Silver Nanoparticles: A Comprehensive Review

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Abstract

Nonlinear optics has become an essential area of study for understanding and harnessing the complex light-matter interactions that occur in molecules, nanostructures, and advanced materials. This review introduces a summary of the latest advancements, with a particular focus on the nonlinear optical characteristics of noble metal nanoparticles, especially silver nanoparticles (AgNPs). These nanomaterials have gained significant interest due to their typical plasmonic features and size-dependent behavior. Silver nanoparticle colloids demonstrate enhanced light absorption at high intensities, known as reverse saturable absorption, along with a self-defocusing optical response characterized by a negative nonlinear refractive index. It has been observed that the size of noble metal nanoparticles significantly influences their nonlinear optical properties. The thermal lens phenomenon was addressed utilizing a unique approach that considers the

cumulative thermal lensing effect induced in the sample caused by high-repetition-rate femtosecond laser pulses. The article highlights current challenges and prospects in the field, particularly in the development of nonlinear optical materials for photonic, sensing, and optical limiting applications.

Keywords: Nonlinear optics; silver nanoparticles; nonlinear absorption; nonlinear refractive index

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1. Introduction

The nonlinear optical (NLO) response of materials arises from changes in their optical characteristics under the influence of high-intensity electromagnetic fields (Bano et al., 2021; Boyd, 2020). Within the broader scope of electromagnetism, and optics in particular, the study of NLO properties in specific material classes remains a key area of research. Such optical investigations not only enhance our understanding of the fundamental mechanisms governing strong light–matter interactions but also support a broad range of applications in photonics (Boutton et al., 1997). NLO materials, whether in bulk, microstructured, or nanoscale forms, have become integral components of numerous photonic devices. They play essential roles across various disciplines, including biology (Kachynski et al., 2008), optical computing (Nelson et al., 1991), industrial processing (Correa et al., 2011), data storage (Ditlbacher et al., 2000), and medicine (Henari & Henari, 2016). Additionally, they are utilized in optical systems for functions such as optical limiting to regulate light intensity and protect sensors and human eyes (Perry et al., 1996), passive mode-locking

(Fermann et al., 1993), and pulse compression (Krebs et al., 2013; Gaida et al., 2017).

2. Nonlinear optics

The study of processes where light alters a molecule's optical characteristics is known as NLO. In general, only high-intensity light, such as that produced by lasers, is sufficient to induce such changes, even in materials with relatively weak nonlinear responses. Consequently, NLO effects were not observed until the advent of laser technology. The first experimental evidence of 2nd harmonic generation (SHG), widely regarded as the beginning of modern NLO studies, was reported by Franken et al. in 1961, shortly after Maiman's invention of the first laser in 1960 (Maiman, 1960; Franken et al., 1961). Since then, numerous nonlinear optical phenomena have been experimentally demonstrated, including the optical Kerr effect (Wijayan, 2019; Nurhuda et al., 2008), optical solitons (Sulaiman, 2020), optical parametric amplification (Brosnan & Byer, 1979), self-phase modulation (Stolen & Lin, 1978), sum-frequency generation (Guyot-Sionnest et al., 1987), and harmonic light scattering (Terhune et al., 1965).

At its core, NLO explores how intense light interacts with matter (Bano et al., 2021; Boyd, 2020). As electromagnetic waves propagate through a medium, they interact with the material's electrons and atomic structure, causing spatial and temporal redistributions of electrical charges. The primary mechanism involves the displacement of valence electrons from their equilibrium positions owing to the light wave's electric field. This displacement induces electric dipoles, which collectively give rise to macroscopic polarization. When the interaction is sufficiently strong, the resulting polarization leads to the generation of different electromagnetic fields that differ from the original in frequency, phase, amplitude, or polarization (Franken et al., 1961).

3. Origin and Fundamental Theory of Nonlinear Optical Phenomena

When a monochromatic electric field E interacts with a medium, it causes a displacement of the medium's electric charges, namely, electrons within atoms and molecules, either collectively or relatively. This displacement induces a secondary electric field known as electric polarization P, which explains the medium's response to the external field. In the regime of linear optics, the induced polarization is directly proportional to the amplitude of the applied electric field (Franken et al., 1961; Li, 2017; Sutherland, 2003):

$$P = \varepsilon_o \chi^{(1)} E \tag{1}$$

where εo is the vacuum permittivity and χ(1) is the linear susceptibility of the medium.

In the case of NLO; the electric field is extremely intense, typically in the range of 107 to 1010 V/cm as produced by lasers, the medium's polarization response becomes nonlinear and is expanded as a power series in the electric field (Franken et al., 1961; Li, 2017; Sutherland, 2003):

$$P = \varepsilon_o \chi^{(1)} E + \varepsilon_o \chi^{(2)} E^2 + \varepsilon_o \chi^{(3)} E^3 + \cdots \tag{2}$$

Here, χ(2) and χ(3) are the second-order and third-order nonlinear susceptibilities, respectively. The first term matches to linear polarization, while the higher-order terms represent nonlinear polarization.

Equation (2) can be expressed in a more compact form:

$$P = P_L + P_{NL} \tag{3}$$

where:

$P_L = \varepsilon_o \chi^{(1)} E$ is the linear polarization

$P_{NL} = \varepsilon_o \chi^{(2)} E^2 + \varepsilon_o \chi^{(3)} E^3 + \cdots$ is the nonlinear polarization

Breaking these down:

$$P^{(1)} = \varepsilon_o \chi^{(1)} E \quad (4)$$

$$P^{(2)} = \varepsilon_o \chi^{(2)} E^2 \quad (5)$$

$$P^{(3)} = \varepsilon_o \chi^{(3)} E^3 \quad (6)$$

In the linear regime, polarization varies linearly with the strength of the field. However, in nonlinear regimes, 2nd-order polarization is proportional to E2, and 3rd-order polarization scales with the cube of the field. This nonlinear response leads to phenomena like self-focusing and frequency doubling, and optical Kerr effects. Notably, in strong-field conditions, the relationship between polarization and field strength becomes increasingly complex and non-proportional.

4. Nonlinear optical properties of silver nanoparticles and their applications

The origin of NLO is commonly traced back to the epochal experiment by Franken and colleagues in 1961, which demonstrated second harmonic generation (Franken et al., 1961). In their study, ruby laser light with a wavelength of 6493 Å was passed through a quartz crystal, resulting in emitted light in the ultraviolet region. The frequency of this transmitted light was found to be twice that of the incident beam (i.e., 2ω), corresponding to half the original wavelength. Since then, wide study has been devoted to the enhancement and characterization of NLO materials, their nonlinear properties, and their diverse applications. A wide range of NLO-active materials—like organic crystals, metallic nanoparticles, polymeric thin films, and chiral surfaces, have been investigated through both experimental

and theoretical approaches (Boyd, 1992; Zyss & Chemla, 1987; Zyss & Ledoux, 1994; Zyss, 2013; Nalwa & Miyata, 1996; De La Torre et al., 2004; Ostroverkhova & Moerner, 2004; Eisenthal, 2006; Asselberghs et al., 2004; Coe, 2006).

Noble metal nanoparticles, particularly silver nanoparticles (AgNPs), have garnered considerable interest as viable materials for all-optical signal processing, optical limiting, and photonic devices due to their strong third-order NLO responses. These responses are primarily attributed to surface plasmon resonance (SPR) and the quantum size effect (Zhang & Wang, 2017; Ara et al., 2011; Ganeev et al., 2001; Henari & Manaa, 2018). The quantum size effect enhances the optical performance of AgNPs compared to other noble metals, while their relatively low intrinsic plasmonic energy loss at visible wavelengths makes them particularly effective in supporting SPR (Li et al., 2017). Furthermore, research has demonstrated that both SPR and the corresponding NLO characteristics of metallic nanoparticles are heavily reliant upon the surrounding dielectric environment, as well as particle shape and size (Alesenkov et al., 2015; Chen et al., 2016; Hua et al., 2015; Sato et al., 2014).

Numerous studies have examined the NLO behavior of AgNPs (Shahriari et al., 2015; Al-Isawi et al., 2020; Aleali & Mansour, 2010; Dudhani & Kosaraju, 2010; Salloom, 2017; Rativa et al., 2008; Gurudas et al., 2008; Unnikrishnan et al., 2003; Anija et al., 2003; Deng et al., 2008; Ganeev & Ryasnyansky, 2006). For example, Shahriari et al. employed a 532 nm diode laser to study the NLO response of AgNPs, revealing a negative nonlinear refractive index (n2) and positive nonlinear absorption coefficient, which is indicative of self-defocusing behavior and saturable absorption (SA) effect, respectively. It is worth noting that negative nonlinear refractive index n2 refers to a self-defocusing effect in which the refractive index of the medium decreases with increasing light intensity. This behavior is characterized by a negative value of the n2, and in silver nanoparticles, it is primarily attributed to the thermal lensing effect and electron cloud distortion under

intense laser fields (Boyd, R. W. ,2020). On the other hand, SA is a competing nonlinear process where the absorption of light decreases with increasing light intensity. This occurs when the ground-state electrons are depleted faster than they can relax back, leading to a reduction in overall absorption. Reported values for n2 and the absorption coefficient were on the order of 10−8cm2/W and 10−4 cm/W, respectively. Both of which were strongly influenced by nanoparticle size (Shahriari et al., 2015). Al-Isawi et al. used a femtosecond laser at 800 nm and observed reverse saturable absorption (RSA) along with a negative n2 in AgNPs (Al-Isawi et al., 2020). In contrast to SA, RSA is a nonlinear optical phenomenon where the absorption of light increases with increasing light intensity. This occurs when absorption of the excited state is large compared with absorption of the ground state. In the case of silver nanoparticles, RSA typically occurs due to excited-state absorption mechanisms, where photoexcited electrons absorb additional photons. This leads to an increase in absorption at high intensities, making AgNPs suitable for optical limiting applications (Boyd, R. W. ,2020). Gurudas et al. investigated Ag nanodots using 532 nm picosecond laser pulses and measured their nonlinear refractive and absorptive behaviors. The broad SPR absorption profile indicated a distribution of nanoparticle sizes and shapes, with RSA and SA effects depending on sample characteristics. Such tunability suggests that properly engineered AgNPs could be optimized for diverse photonic applications, such as passive mode-locking and protecting the eyes and sensors with optical limiting (Gurudas et al., 2008). Unnikrishnan et al. studied the nonlinear absorption of Ag nanodots at 532 nm, outside their SPR band, and observed an intensity-dependent transition from SA to RSA behavior (Unnikrishnan et al., 2003). Anija et al. (2003) and Deng et al. (2008) reported similar switching behaviors for AgNPs embedded in ZrO2 and PMMA, respectively, using nanosecond pulses at 532 nm. Rativa et al. (2008) employed femtosecond pulses at 800 nm to study aqueous AgNP

suspensions, showing that both n2 and the nonlinear absorption coefficient depended on nanoparticle concentration.

Aleali & Mansour (2010) examined the optical limiting efficiency of AgNPs irradiated with nanosecond pulses at 532 nm and found that nonlinear scattering could enhance their limiting efficiency. Zhang et al. (2017) (Dudhani & Kosaraju, 2010) used picosecond pulses from an Nd:YAG laser at 532 nm and identified an RSA response in AgNP colloids, attributed to two-photon absorption (TPA). According to Salloom (2017), the nonlinear absorption of AgNPs is also influenced by self-focusing effects. It is important to note that most of the above-mentioned studies were conducted at excitation wavelengths significantly different from the SPR peak of around 400 nm. However, Ganeev and Ryasnyansky (2001) investigated NLO absorption at the SPR resonance, reporting that AgNPs exhibit SA when excited with 1.2-ps, and RSA under 8-ns pulses. These findings underscore the critical effect of wavelength and pulse duration on the NLO behavior of AgNPs.

The unique nonlinear optical properties of AgNPs, such as RSA, SA, and a negative n_2, make them promising candidates for a wide range of advanced photonic and optoelectronic applications. One of the most notable applications is in optical limiting devices, which protect sensitive optical sensors and human eyes from laser-induced damage. Due to their strong RSA response at high intensities, AgNPs act as effective limiters by attenuating excessive laser power (Al-Isawi et al., 2020; Aleali & Mansour, 2010; Boyd, 2020). AgNPs are also used in passive mode-locking and pulse shaping within ultrafast laser systems. Their SA behavior under specific excitation conditions allows for the generation of short pulses, essential in high-speed optical communication and ultrafast spectroscopy (Fermann et al., 1993; Shahriari et al., 2015; Gurudas et al., 2008). In all-optical switching, AgNPs enable intensity-dependent modulation of light without converting optical signals to electronic form. This is especially valuable in the development of integrated photonic circuits and optical computing

platforms (Nelson et al., 1991; Zhang & Wang, 2017). Furthermore, silver nanoparticles are widely applied in surface-enhanced Raman scattering (SERS) and plasmonic sensing, where their size- and shape-tunable surface plasmon resonance enhances local electromagnetic fields, boosting sensitivity for molecular detection (Henari & Manaa, 2018; Hua et al., 2015). In the biomedical field, AgNPs are employed in nonlinear optical imaging, leveraging their strong third-order susceptibility for contrast enhancement in multiphoton microscopy and diagnostic applications (Kachynski et al., 2008; Boyd, 2020). These multifaceted applications underscore the importance of AgNPs in both fundamental research and emerging optical technologies, highlighting their potential for future innovations in nanoscale light manipulation.

5. Conclusion

NLO is fundamentally concerned with phenomena arising from higher-order nonlinear polarization within a medium. Nobel Laureate Nicolaas Bloembergen, one of the pioneers in the field, offered a widely accepted definition of NLO: “Optical phenomena are considered nonlinear when a material interacts with an electromagnetic field in a way that its response does not scale linearly with the field's intensity” (Bloembergen, 1977). It is important to know that an electromagnetic field—commonly referred to as a light field— includes both magnetic and electric elements. Whereas the magnetic polarization of a medium can be influenced by the magnetic component of light, this effect is typically negligible in most materials due to its extreme weakness. However, in media containing asymmetric chiral molecules, magnetic interactions may become more prominent. Despite this, standard NLO literature generally focuses on the electric field component of light, as it is the primary driver of NLO effects in most materials. In the context of silver nanoparticles (AgNPs), their NLO properties are governed by a combination of particle size and laser parameters. Smaller nanoparticles tend to exhibit stronger nonlinear

responses due to enhanced surface area and quantum confinement effects. Additionally, variations in laser power, wavelength, and energy fluence can significantly impact the nanoparticles' interaction with light. These parameters collectively influence their ability to absorb and scatter light, modify its propagation direction, and enhance optical limiting performance, making AgNPs valuable for applications in photonic and optoelectronic systems.

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