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NONLINEAR OPTICS' EFFECT ON ADVANCED IMAGING TECHNOLOGIES

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Abstract

Nonlinear optics has significantly impacted innovative imaging technologies by enabling various operations such as spectrum transformation, nonlinear absorption, nonlinear dispersion, as well as phase and amplitude modulation. Optical materials including glasses, crystals, nanomaterials, and polycrystalline substances are essential in nonlinear optical processes. Cubic nonlinear optical processes do not need noncentrosymmetric structures, although second-order nonlinear optical effects must. Anisotropic crystals like KDP, LiNbO3, BBO, and KTP are often used for second-order nonlinear optical applications, whereas glasses are suitable for third-order nonlinear optical applications. Polycrystalline compounds, often considered inappropriate for optics because of their strong light scattering, are now being considered as potential nonlinear optical pigments. Nanocrystals, such as upconverting nanoparticles and quantum dots, exhibit altered optical characteristics compared to bulk media and have promise for nonlinear optical uses. Microcrystalline materials, including metal-organic complexes and cooperation polymers, have been studied for their potential as nonlinear optical media. NLO pigments, compounds with nonlinear absorption and extra optical properties, do not need great optical transparency. Novel nonlinear optical (NLO) materials have been used in laser advancements for nonlinear microscopy, optical power restriction, and saturation absorbers. Semiconductor quantum dots are being considered as effective replacements for organic dyes in nonlinear microscopy for luminous marking. Optical power restriction may be achieved by two-photon processes that occur directly or through two-step mechanisms in materials that absorb a third photon after absorbing two photons. Evaluating the nonlinear absorption capabilities of new compounds often depends on their effectiveness as saturable absorbers in laser systems. Understanding the temporal response of saturable absorbers is crucial for laser mode-locking, since they may be categorized as either fast or slow. The recovery time of saturable absorbers determines the duration of mode-locked pulses. Femtosecond lasers and time-resolved pump-probe methods may be used to determine the temporal properties of saturable absorbers. This article explores the impact of nonlinear optics on advanced imaging technologies, focusing on the importance of different optical materials and their use in nonlinear optical processes. Advancements in NLO materials provide potential for improved optical features and imaging technologies.

Keywords: Optical power limitation, nonlinear microscopy, saturable absorbers, mode-locking, microcrystalline substances.

1. *Introduction*

In numerous ways, optical substances, that is, elements that possess some advantageous optical properties can be utilized. Glasses of diverse kinds, such as the most well-known oxide glasses (such as silica glass), additional inorganic glasses (such as chalcogenides), as well as amorphous compounds of different kinds (such as amorphous polymers and mixed substances) are the most significant category of these substances for use in optical devices. An additional category of materials frequently employed in the field of optics comprises crystals, which are usually single-crystalline plates, pieces, prisms, or rods. These crystals comprise particular characteristics that are advantageous in the fabrication of optical components. Some uses may also benefit from the properties of particular species (such as dyes) when present as liquid solutions. The emergence of nanotechnology has broadened the spectrum of optical materials through the implementation of nanosystems, which include both simple nanoparticles such as quantum dots and plasmonic fragments, as well as complicated forms including nanostructured surfaces or drug nanocarriers. Furthermore, contrary to the prevailing belief that polycrystalline substances are largely unusable in optics due to their high scattered light, an emerging class of materials called "transparent ceramics" (Ikesue et al., 1995; Lu et al., 2002) derive their outstanding visibility from the nanosized particles of materials that comprise them.

The aforementioned broad assertions are also applicable to the field of nonlinear optics, which utilizes the intense light produced by laser sources to accomplish various functions, including phase and amplitude modification, spectrum transformation, nonlinear absorption, and nonlinear dispersion (Boyd et al., 2008). Nonlinear optical (NLO) reactions among photons are all facilitated by the medium through which electromagnetic radiation travel. The substance demands vary based on the specific process. For instance, second-order nonlinear optical effects (SHOEs), which are characterized through the second-order nonlinear vulnerability $j^{(2)}$, necessitate substances with noncentrosymmetric constructions for second-harmonic era (SHG). Conversely, cubic NLO procedures, which employ the third-order vulnerability $j^{(3)}$, do not impose this requirement. The illustration of the entirely defective $\chi^{(3)}$, whose actual component is accountable for nonlinear refraction as well as fictitious component is accountable for nonlinear absorption, is the most significant in this context. Isotropic substances, like glasses, are appropriate for third-order NLO uses, whereas anisotropic crystals, including potassium dihydrogen phosphate (KDP), LiNbO3, β-barium borate (BBO), and potassium titanyl phosphate (KTP), are among the most frequently employed second-order substances.

This study aims to explore the potential of nonlinear optics in driving advancements in imaging technology, particularly in scientific investigation, medical diagnostics, and industrial imaging. It will focus on the profound effects of nonlinear optics on imaging resolutions, functionalities, and applications, ultimately aiding in the development of optical imaging technologies and their practical implementations. The research will examine the potential of nonlinear optics methods, including parametric imaging, materials science, biomedicine, and harmonic generation imaging, to significantly transform imaging capabilities in various disciplines, including remote sensing, materials science, and biomedicine.

2. NLO Pigments

The potential for dispersal resulting from the polycrystalline nature of particular substances does not universally preclude their utility. Phosphates, which are solid-state substances capable of emitting light (luminescence), and pigments, which are intractable colored species utilized to dye other substances, are two instances of substances that possess crystallinity and find extensive optical applications. Although polycrystalline substances have been in existence for an extended period of time, their NLO characteristics have seldom been regarded as noteworthy.

Powder SHG, which is employed to semiquantitatively evaluate the second-order NLO merit of crystalline substances (a method established separately by Graja (1986) and Zhang and Halasyamani (2017), is a domain of NLO research where the polycrystalline nature of the sample may be advantageous. However, this does not appear to be a viable route to practical uses. As previously mentioned, a viable approach to producing substances that possess favorable optical clarity and are therefore suitable for use as NLO media is to decrease the particle size of the substance to dimensions significantly smaller than the wavelength of light.

Therefore, nanomaterials composed of crystals (nanocrystals) are generally highly appealing for NLO applications, as their size-dependent phenomena (such as quantum entanglement in the instance of semiconductors) result in altered optical characteristics relative to bulk media and may also contribute to the improvement of optical nonlinearity. The benefits of nanoparticles composed of diverse semiconductors—quantum dots, upconverting nanoparticles incorporating lanthanide ions, metal (plasmonic) nanoparticles, and so forth—are widely recognized (Zhang and Wang, 2017; Olesiak-Banska et al., 2019). These substances are omitted from this evaluation in favor of a concentration on more recent advancements pertaining to materials that are comparatively obscure.

Furthermore, in evaluating the merits of different materials for near-field optics (NLO), the analysis extends beyond nanoscale crystalline substances to encompass the potential of microcrystalline substances, particularly those classified as collaboration polymers, involving their porous variants called metal-organic structures (MOFs), to serve as NLO medium for particular applications. Indeed, we have contended in Medishetty et al. (2017) and Zaręba et al. (2019) that these microcrystalline substances retain the ability to utilize their NLO characteristics (particularly nonlinear absorption and its associated impacts) in a variety of contexts, thereby constituting a category known as NLO pigments (Zaręba et al., 2020). This concept can be additionally broadened to encompass a subcategory of materials that exhibit luminosity when excited nonlinearly (referred to as "NLO phosphors"), in addition to substances that serve other purposes such as photocatalysis, photochemical modifications, photocurrent generation, monitoring, and so forth, that do not inherently demand high optical transparency.

3. Regular Application of Innovative Nonlinear Optical Materials

Historically, the belief that stronger nonlinear compounds could replace well-established substances like inorganic crystals, glasses, and organic pigments was largely unfulfilled due to obstacles like material processing complexities, durability, and wavelength range specifications. However, there are opportunities for embracing nascent NLO materials, such as two- or multiphoton activated luminescence, optical power limitations, and the use of saturated absorbers in laser innovations, which could lead to the widespread adoption of these materials.

The propensity for organic dyes to undergo photobleaching and their restricted functionality in the infrared region has generated considerable attention regarding alternative luminescent markers, such as semiconductor quantum dots, for nonlinear microscopy applications. In such instances, determining which nonlinear characteristic ought to be utilized to evaluate the applicability of novel substances is a crucial concern. The applicable benefit element can differ based on the situation at hand. It could be calculated by multiplying the two-photon absorption cross section by the quantum yield of emission (also known as two-photon luminosity), denoted as $\gamma\sigma_2$, where γ represents the brightness of the quantum output or its worth adjusted to unit volume or mass (Samoc et al., 2012).

Optical power restricting has seen significant progress, with new substances being suggested for their ability to restrict medium transmission to extremely low strength or flow. Direct two-photon procedures can achieve power limitation, but efficiency is typically low, especially with nanosecond laser pulses. Some materials show enhanced power-limiting efficiency through a two-step mechanism, where a third photon is absorbed after two photon absorptions. This may be significant if the high state lifespan is longer than laser pulse duration. RSA is the most commonly used mechanism for power limitation. However, without additional information, it's difficult to draw significant contrasts between different researchers' outcomes. Scholarly articles often focus on transmittance compared to fluency information, neglecting microscopic factors and excited conditions.

The nonlinear absorption characteristics of emerging compounds can be determined by analyzing their efficacy as saturable absorbers. These absorbers, crucial in laser chambers for producing ultrashort pulses, impact the system's efficiency. Continuous lasers often use Q-switching to produce high-energy, sustained pulses in nanosecond intervals. The cavity quality aspect, which represents the proportion of energy depletion per cycle to energy stored within the cavity, is crucial in determining these absorber characteristics.

Pulsed lasers now use active Q-switching rather of the passive technique. Mode-locking produces femtosecond-range pulses, which are far shorter than the cavity's round-trip time, making it an effective way for generating brief impulses. To do this, a strategy has to be put in place to minimize loss during a radiation peak of higher intensity (Zhang et al., 2019). This may be accomplished by using a semiconductor saturable-absorber mirror (SESAM) or Kerr lens mode-locking, which involves using a saturated absorber that is carefully chosen. The saturable absorber's temporal reactivity is a crucial quality in this context, categorized as either "fast" or "slow."

Fast saturable absorbers have shorter relaxation periods than the mode-locked pulse duration, whereas slow saturable absorbers have protracted relaxation times. The time-dependent transparency of a saturable absorber may be determined using time-resolved pump-probe techniques that use ultrafast lasers (Nyk et al., 2014). Quicker relaxation time leads to a shorter recovery period for the absorbent from "bleaching" owing to absorption. The recovery time is crucial in defining the duration of the laser pulse produced when the device operates as a mode locker (Wang et al., 2017). For example, both kinds of absorbers may generate pulses of about the same duration when the modulating depth, which refers to the change in absorbance or reflectivity due to incoming light, is kept constant. For best results, the absorber recovery period should not exceed 10 times the width of the pulse (Wang et al., 2016).

4. Metal-organic frameworks (MOFs) and Other Polymers

Coordinated polymers (CPs) and metal-organic frameworks (MOFs) have attracted significant attention because of their wide range of physicochemical properties and exceptional structural adjustability, which is rarely found in other types of compounds. Therefore, it is not unexpected that research using these chemicals led to significant experimental efforts, often crossing conventional boundaries of chemistry and materials research and extending into areas such as medicine (Cao et al., 2020). Various classification methods developed due to the increasing interest in CPs, resulting in the use of inconsistent naming standards. Therefore, it is crucial to start this part by providing a clear explanation of the types of compounds classified as CPs and MOFs, as well as those that do not fall under this category. We will provide an outline of the recommended terminology produced by IUPAC throughout the process.

The International Union of Pure and Applied Chemistry (IUPAC) defines a coordination compound (CP) as a substance with repeated coordination units spread in one, two, or three dimensions. A coordinating entity is an ion or neutral compound with a central atom, typically a metal atom, and ligands, arrays or collections of atoms. CPs can be conceptualized as an infinite polymeric expansion of metal complex units, extending at least a single dimension. The IUPAC terminology does not restrict the chemical composition of a ligand substance, making any organic or inorganic molecule capable of coordinating metal ions viable for CP synthesis (Batten et al., 2013).

The concept of coordinating polymers (CPs) extends beyond porous coordinating polymers (PCPs) to include MXenes and 2D/3D perovskites. However, the general public views CPs in a more limited manner, primarily recognizing them as one-dimensional, two-dimensional, and three-dimensional coordination compounds containing organic compounds and metal ions as ligands. This classification excludes most inorganic substances, including MXenes. Hybrid organic-inorganic perovskites, composed of protonated organic amines, are often referred to as HOIPs instead of CPs.

5. The NLO Characteristics of CPs and MOFs

Prior to 2015, the primary focus of nonlinear optics for CPs was the estimation of the respective SHG performances of CP powder materials. A significant portion of the prior research in this field was dedicated to examining novel CPs with high SHG efficiencies (Ye et al., 2005). The crystal engineering of the SHG response of CPs, or the investigation of the relationship among structural variables and SHG efficacy, has received considerable interest (Evans and Lin, 2002). However, research pertaining to third-order NLO characteristics, such as nonlinear absorption, was not prevalent during that period. Previous research endeavored to define nonlinear absorption coefficients by employing the Z-scan method. However, a significant obstacle in this regard was the absence of standardized protocols for accurately characterizing insoluble macromolecular substances, including CPs. The aforementioned concerns, at most, resulted in significant incongruities in the documentation of NLO-related amounts for CPs (Medishetty et al., 2017).

The introduction of studies that demonstrated the 2PA and MPA features of microcrystalline CPs in a number of research journals around 2015 marked a crucial turning point in the area of nonlinear optical properties of CPs. Quah et al. (2015) made early studies of the 2PA and MPA properties of microcrystalline MOFs using microscopy-based SSTPEF. The chemical under examination is a pillar-layered Metal-Organic Framework (MOF) made up of zinc ions, trans, trans-9,10-bis(4-pyridylethenyl)anthracene, and trans-4,4'-stilbenedicarboxylic acid. It displayed Nonlinear Optical (NLO) absorption involving three, two, and even four photons. MOFs exhibit strong nonlinear absorption when paired with a high quantum yield of fluorescence and prolonged lifetimes. Can be used to provide low-threshold emission stimulation, as shown in Zn(II) and In(III) MOFs using tetrakis[4-(4-carboxyphenyl)phenyl]ethene, an aggregation-induced emission (AIE) dye (Medishetty et al., 2017).

MOFs that could undergo photochemical processes were also introduced to NLO uses, including two-photon lithography, for the first time (Yu et al., 2015). During the same period, progress has been achieved in the field of methodology. The ISTPEF (internal standard two-photon excited fluorescence) method was introduced by our group to quantify 2PA cross regions in luminous fragments of microcrystalline CPs in substances (Zaręba et al., 2019). This method was subsequently utilized to characterize crosslinked conjugated polymers along with additional two-photon active non-soluble substances (NLO dyes) (Zaręba et al., 2019). For the initial time, visual nonlinearities of CPs in nanoparticle form have been characterized using the Z-scan method, as illustrated for Prussian Blue (ferric hexacyanoferrate(II)) NPs, which absorb three photons extensively, and ZIF-8 NPs, which absorb two photons weakly (Zareba et al., 2016).

Despite the aforementioned and other facets of the nonlinear optical properties of CPs were thematically dispersed, they served as preliminary indicators of what and where to search for in subsequent investigations. Substantial development has been made since then in the study of the NLO properties of CPs, with major contributions focusing on the structure-property connections in nonlinear absorption of CPs as well as innovative uses including two-photon catalysis. As such, the subsequent literature review shall be dedicated to developments that have arisen since our previous examination of that particular domain.

6. Recent Advances in Nonlinear Absorption of CPs

The structural adaptability exhibited by CPs is likely one of the most captivating attributes in the field of nonlinear response engineering. Practically every metal in the periodic table has been employed in the synthesis of CPs, and the potential ligands are virtually limitless. The ligand families that have been the subject of the most extensive research include di- and polytopic carboxylic, phosphonic, and sulfonic acids, N-heterocyclic foundations, compounds belonging to different ligand categories, and ligands that possess multiple functional families (Paz et al., 2011; Chen et al., 2014). Nevertheless, it is significant to mention that polytopic carboxylic acids have attracted the most interest due to their advantageous functional properties and the substantial dependability of the resulting crystal structures—a quality that is fiercely coveted in the realm of materials fabrication (Pang et al., 2019).

While the configuration of CPs can be influenced by both ligands and inorganic ions, it is expected that the latter will facilitate the majority of the nonlinear reaction. A number of design principles for two-photon chromophores have been derived from extensive research on the structure and properties of organometallic and organic two-photon chromophores (Pawlicki et al., 2009; Zhang et al., 2017). The straightforward implementation of these principles in the synthesis of novel CP ligands facilitates the development of CPs with enhanced nonlinear absorption properties. On the other hand, CPs are characterized by their compacted macromolecular architecture, in which the electronic interaction among constituent elements may transpire across space. The extent of porosity and the specific coordination net type determine this phenomenon. An illuminating analogy can be drawn from prior NLO investigations that involved porphyrins and particles of AIE pigments. The collaborative impact of NLO is observed to be influenced by the proximity of aromatic components. Specifically, the 2PA response is greater in the aggregated form compared to the non-aggregated form (Biswas et al., 2012; Justyniarski et al., 2018).

Coordination networks (CPs) are renowned for their ability to implement an extensive variety of crystal structures; this corresponds to the network's architecture imposing an extensive number of potential forms. A modification to the crystal packing motif of CP and associated phenomena, including aggregation and net topology, may therefore have an impact on the resultant nonlinear response. Therefore, it is unsurprising that the intermarriage of AIE molecules and MOFs has been swiftly acknowledged as a fertile ground for structure-property investigations (Zarība et al., 2021).

7. Conclusion and Recommendation

In summary, nonlinear optics has significantly influenced imaging technology, with opaque materials like nanomaterials, glasses, and crystals being used in various optical devices to exploit their properties. These materials enable nonlinear optical reactions, resulting in phase and amplitude changes, spectra transmutation, absorption, and dispersion. The properties and attributes of specific substances are crucial in determining their suitability for various nonlinear optical procedures. Polycrystalline substrates, including pigments and phosphates, have gained attention for their optical transparency and ability to manifest nonlinear optical properties. Nanomaterials, such as crystal nanocrystals, have shown potential in modifying optical properties and enhancing nonlinearity. Novel NLO material implementations have been widely used in various disciplines, including optical power limitation, nonlinear microscopy, and the use of saturated absorbers in laser innovations. Researchers have investigated alternative luminescent markers, such as semiconductor quantum dots, in nonlinear microscopy to address limitations associated with organic dyes. The efficacy of optical power limitation has been improved through direct two-photon processes and two-step mechanisms.

Nonlinear optics has significantly impacted imaging technology, but further research is needed to advance its applications. Future research should focus on developing novel materials with improved nonlinear optical characteristics, such as crystal structures, nanomaterial compositions, and composite materials. Developing characterization techniques, such as femtosecond laser techniques and time-resolved pump-probe methodologies, can provide a better understanding of the nonlinear optical properties of materials. This will aid in material selection and optimization for specific applications. Integration with imaging systems is a potential paradigm shift in imaging technology, allowing for enhanced contrast, depth penetration, and resolution.

Optimizing nonlinear optical processes, such as enhancing nonlinear microscopy methodologies, developing new methods for mode-locking and Q-switching in lasers, and developing advanced power-limiting materials with higher damage thresholds, is crucial for practical applications. Multidisciplinary collaborations among scientists, engineers, and researchers from diverse disciplines are also beneficial in the field of nonlinear optics. Promoting partnerships between engineering, material science, optics, and chemistry can lead to groundbreaking advancements and solutions in nonlinear imaging. In conclusion, nonlinear optics has a significant impact on imaging technology, and further research and advancements in this field can unlock new territories and advancements in fields like medicine, biology, and materials science.

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