Components of the Nanocomposite Photoanisotropic Materials

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A new approach has emerged that has been extensively studied by many research groups, namely doping azobenzene-containing materials with nanoparticles with various compositions, sizes, and morphologies. The resulting nanocomposites have shown significant enhancement in their photoanisotropic response, including increased photoinduced birefringence, leading to a higher diffraction efficiency and a larger surface relief modulation in the case of polarization holographic recordings.

Keywords: nanocomposite materials ; nanoparticles ; photoanisotropic materials ; azopolymers

1. Introduction

The demand for new materials to develop modern technologies is constantly growing. Although nanocomposite (NC) materials have been in use for a long time, they are constantly being improved to enable increasingly advanced applications. The new materials, obtained by combining two or more materials on nanoscale level, in most cases not only combine the qualities of their components, but also yield new, advantageous properties. In many cases, the nanoparticles (NPs) incorporated in nanocomposite materials modify and improve the optical, mechanical, and electrical properties of the matrix.

The optical properties of nanocomposite materials have been used since ancient times. Stained glass windows and ancient works of art exemplify their applications. A popular example of the intriguing optical features of nanocomposites is the Lycurgus cup from the 4th century AD [1]. In this case, the basis is the effect obtained from the excitation of the surface plasmon resonance due to the presence of gold and silver nanoparticles in the glass, which is expressed in a change in the color of the cup depending on whether it is viewed in transmission or reflection.

The combination of organic and inorganic materials in nanocomposites opens up a new and exciting area in applied optics. Inorganic components modify the optical properties of the organic components. In addition, the organic matrix may provide a flexible and ordered structure to the nanocomposite materials. For example, polymers can be hybridized with nanoparticles, modifying the refractive index, birefringence, and other characteristics of the composite material [2][3][4][5][6][7].

From the point of view of holographic applications, in recent years, special attention has been paid to nanocomposite materials composed of photopolymers and nanoparticles [8][9][10][11][12][13][14][15][16][17][18][19][20][21][22][23][24][25][26][27][28][29][30][31][32][33][34][35][36]. The ability of these materials to modify their structure and surface through light-induced polymerization is used. Problems like the creation of practically non-shrinkable holographic recording media have been solved by using various types of nanoparticles. Many applications of these materials in holography have been outlined by Tomita et al. in their topical review on photopolymerizable nanocomposite photonic materials [12]. The modern trends in the development of light-sensitive media for holography applications are presented in a review article by Barachevsky [37]. Other type of hybrid organic/inorganic (or nanocomposite) materials include polymer-dispersed liquid crystal (PDLC) materials or liquid crystals doped with nanoparticles of photosensitive polymers, which have been intensively studied by many research groups [38][39][40][41][42][43][44][45][46][47][48][49][50][51][52][53].

They most often represent an azopolymer matrix doped with metallic or non-metallic nanoparticles. In order to use a given material for polarization holographic recording, it must be photoanisotropic, or in other words, it must be able to register and record the polarization of light. The higher the value of the photoinduced birefringence, the greater the diffraction efficiency of the polarization diffraction gratings recorded in this material. Azobenzene-containing materials are often the preferred recording media for polarization holography due to their high values of photoinduced birefringence, allowing for the inscription of highly efficient polarization-selective holographic gratings. These gratings have specific polarization properties [54][55][56][57] that enable various applications in the fields of polarization-selective optical elements, high-capacity data storage, and many others [58][59][60][61][62]. A polarization diffraction grating is the key component in the design of a spectrophotopolarimeter, which can measure simultaneously in real time the spectra of all four Stokes parameters of light, as reported by Todorov and Nikolova [58][60]. Provenzano et al. reported the application of anisotropic gratings for circular dichroism measurements using a new configuration that is simpler than the conventional one [63]. Particularly important are the applications of polarization holographic gratings as polarization-selective diffractive optical elements (PSDOEs). Specially designed gratings can be used as circular polarization beam splitters [64] or to convert a circularly polarized incident beam into a linearly polarized beam [65]. They can act as bifocal, spherical, cylindrical, and tunable Fresnel lenses or microlens arrays [66][67][68][69], or they can be designed to generate asymmetric diffraction [70].

The implementation of high-density and high-capacity volumetric data storage is another important application of photoanisotropic materials. Polarization holography enables polarization multiplexing, as shown by Nikolova and Ramanujam [55][71]. Lin et al. designed and experimentally demonstrated a polarization multiplexing holographic memory with an increased storage capacity using a circular polarization recording configuration [72]. Similarly, polarization holographic gratings were applied for polarization multichannel multiplexing, vector beam storage, and fabrication of polarization multiplexing diffracting optical elements [73], self-interference incoherent digital holography [74][75], virtual reality displays [76][77], and also 4G optical elements with spatially modulated birefringence across the surface [78][79]. Yang et al. and Xia et al. applied the diffraction characteristics of anisotropic gratings to demonstrate logic operations using Boolean algebra for all-optical diffraction elements using an azo-dye doped polymer film [80][81].

In 1995, Rochon et al. [82] and Kim et al. [83] first reported a very important effect in azopolymers: the formation of surface relief during polarization holographic recordings. They found that in this case together with the polarization grating in the volume of the media, a surface relief grating (SRG) is formed in the azobenzene-containing polymer films. The surface relief grating is produced by an interference pattern of light and is due to a photoinduced mass transport of the azopolymer. Since SRG formation is an effective and simple nanofabrication process, it has provoked significant interest in many research areas [84][85][86][87][88][89][90]. Various applications based on its unique features have been reported so far in optics [12][28][70][91][92][93], sensors [24][88][94][95][96][97][98], mechanical applications [99][100][101][102][103], optically controlled alignment [104][105][106], etc.

The development of azobenzene-containing nanoparticles and the possibility for optical manipulation of their shape and size also opens several new fields of applications [107][108][109][110]. A very significant elongation of the azopolymer-containing drops has been observed when illuminated with linearly polarized light—up to six times their initial diameter [109]. It has also been demonstrated that their shape can be controlled by the direction of light polarization and the laser irradiation time. Potential applications in optical signal control as well as mechanical motion control of photosensitive soft materials at the microscale and nanoscale were also suggested [110].

All these applications require an efficient photoanisotropic media with high photoinduced birefringence. To date, photoinduced anisotropy has been observed in various photosensitive materials. Amongst them, the azobenzene-containing materials have been extensively used due to the high values of photoinduced birefringence and diffraction efficiency achieved in them when recording polarization diffraction gratings [111][112][113][114][115][116][117][118]. Under irradiation with polarized light within their absorbance band, the azobenzene groups undergo a series of *trans-cis-trans* photoisomerizations until they reorient perpendicularly to the polarization of the pump light. This anisotropic orientation of the azobenzene groups leads to birefringence, which furthermore can be spatially controlled with high resolution over the area of the optical element. A probe beam, with a wavelength outside of the absorption band of the used material, can non-destructively read this birefringence. Diverse applications have inspired the investigation of large number of azopolymers [117][118][119][120]. A commercially available azopolymer, commonly denoted as PAZO (poly[1-4-(3-carboxy-4-hydrophenylazo)benzensulfonamido]-1,2-ethanediyl, sodium salt]), is often the preferred material for polarization holography by many research groups due to the high values of photoinduced birefringence and large amplitude of the surface relief gratings inscribed in it [121][122][123][124][125][126][127][128][129][130][131][132]. In addition, its solubility in water and methanol allows for the easy preparation of nanocomposite materials using water suspensions of nanoparticles, for example gold or silver NPs.

Many approaches have been implemented in order to obtain polarization-sensitive materials with the highest possible photoinduced birefringence. Most often, new azopolymer architectures have been synthesized and tested with various substituents for the azochromophores, different spacer length between the main and the side chain of the azopolymer, etc. Alternatively, some methods are aimed at improving the performance of already existing azopolymers, for example, via thermally assisted recording [133], or by doping the azopolymer matrix with nanoparticles.

2. Components of the Nanocomposite Photoanisotropic Materials

Two most important components of the nanocomposite photoanisotropic materials: (i) nanoparticle dopants with various chemical compositions, sizes, and morphologies, and (ii) photoanisotropic matrices based on azodyes or azopolymers. Due to their nanoscale dimensions, the nanoparticles are usually characterized using transmission electron microscopy (TEM) or scanning electron microscopy (SEM).

Furthermore, these techniques allow us to determine the size distribution of the NPs and also the way they are dispersed within the nanocomposite thin film samples. The azobenzene-containing NC components (azodyes or azopolymers) are presented with their chemical structures that give the essential information about the azochromophores, the substituents used, the length of the side-chain spacer, etc.

2.1. Nanoparticles

Nanoparticles with different compositions have attracted the attention of researchers as possible dopant components, such as quantum dots (QDs) [134][135][136], carbon nanotubes (CNTs), and carbon nanofibers (CNFs) [137][138], ZnO [128][139][140][141][142], SiO_2 [143][144], TiO_2 [145][146][147][148], semiconductor nanoparticles like tellurium containing chalcogenide system $(GeTe_4)_{100-x}Cu_x$ [149] and goethite (α-FeOOH) nanorods [129][150], nanozeolites [151], and upconverting nanoparticles (UCNPs) [152]. Gold (Au) and silver (Ag) NPs are the most commonly used metallic nanoparticles [132][146][153][154][155][156][157][158][159][160][161]. Studies have also been conducted with bioactive metals, such as copper and nickel (Cu, Ni) [162][163]. Nanocomposites with various sized nanoparticles have also been investigated. Very small nanoparticles with sizes in the range 2.5–10 nm have been used by some researchers. There are more studies with medium-sized NPs from 10 nm to 50 nm, whereas fewer studies focus on the larger NPs with sizes in the range 150–600 nm. Nanoparticles of different shapes have also been used. The most commonly used shape is spherical. There are also several studies of nanorods, as well as of nanoparticles with hexagonal, cubic, or rectangular shape.

2.2. Commonly Used Azo-Containing Matrices for Nanocomposites

A very commonly used azopolymer, as researchers have already noted, is the commercially available azopolymer PAZO (poly[1-4-(3-carboxy-4-hydrophenylazo)benzensulfonamido]-1,2-ethanediyl, sodium salt]). PAZO was used as the base to obtain photoanisotropic NCs by many authors, like Berberova et al. [128][141][158], Nedelchev et al. [129][140][150], Falcione et al. [132], Fernandez et al. [146], Mateev et al. [147][162], Nazarova et al. [148][156], Stoilova et al. [149][163], and others.

Some other azopolymers have also been employed as components of NPM and are denoted by the authors as follows: P_1—Nedelchev et al. [139], Nazarova et al. [164], P_{1-2}—Nedelchev et al. [139], Nazarova et al. [164], P_2—Nedelchev et al. [139], p4VP(DY7)$_{1.0}$ Hautala et al. [161], PDR19—Kang et al. [145], PEPC-co-DO—Achimova et al. [135] and P1, P2, P3—Vijayakumar et al. [165].

In other studies, nanocomposites are composed from photoanisotropic azo dyes, nanoparticles and a non-photoanisotropic polymer matrix [142][166][167][168][169].

2.3. Main Optical Parameters of the Nanocomposite Photoanisotropic Materials

As was already mentioned in the Introduction, an essential parameter that characterizes the optical response of any photoanisotropic media, including the NPM, is the maximal value of the photoinduced birefringence, denoted as Δn_{max}. It is easily measured by a simple *pump-probe* optical scheme, which does not require coherent lasers or vibration isolation, and for this reason it is the most commonly used parameter to evaluate the enhancement of the nanocomposites' performance in comparison with the non-doped azobenzene-containing material.

In case of polarization holographic recording, two more parameters are used to quantify the behavior of the nanocomposite photoanisotropic materials, namely the diffraction efficiency (DE) and height of the formed surface relief grating (h_{SRG}).

These three main parameters are studied and compared throughout to demonstrate the effect of adding different nanoparticles to the azodye/azopolymer matrix.

3. Conclusions

Photoanisotropic nanocomposites based on an azodye or azopolymer matrix with added nanoparticles are an attractive and relatively easy to produce materials with improved optical characteristics. Due to the wide variety of simple synthesis techniques that can be used to obtain nanoparticles of different compositions, shapes and sizes, the optical properties of nanocomposites can be precisely tuned and adjusted for specific applications.

Most of the results show an increase in one or all of the parameters: birefringence, diffraction efficiency, or surface relief height of the polarization holographic gratings recorded in the nanocomposites compared to the undoped material. Other optical parameters, such as absorption, response time, stability, etc., were also investigated. Nanocomposites with different compositions were studied, varying both the type of nanoparticles and the type of matrix. Both metallic and non-metallic particles have been studied, with gold and silver nanoparticles being the most commonly used metallic ones, and zinc oxide and quantum dots being the most frequently used non-metallic ones. The preferred photoanisotropic matrix amongst the azopolymers is the commercially available polymer PAZO, and amongst the azodyes, the most common are Methyl Orange and Disperse Red 1.

The optimal concentrations of the nanoparticles were determined, as well as in some cases their optimal size. The influence of the shape of the nanoparticles on the optical properties of the nanocomposites was also investigated.

Nanocomposite materials, unlike other optical materials, have a much wider range of applications due to their composite nature. One of their components, namely nanoparticles, can be implemented in different ways; for example, they can be dispersed in a liquid or applied as a thin film. This opens up the possibility of a wide range of potential applications. New opportunities are being discovered for the emergence of new products that exploit the unique optical advantages provided by nanocomposite materials.

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