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**A RESEARCH ABOUT ENHANCEMENT OF LIQUID
CRYSTAL NANOPARTICLE COMPOSITES:
ELECTRICAL AND OPTICAL STUDIES**

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A Research about Enhancement of Liquid Crystal Nanoparticle Composites: Electrical and Optical Studies

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Abstract – Liquid crystals possess large dielectric and electro-optical properties owing to their large anisotropy coupled with the collective molecular reorientation. Doping dyes and nanoparticle into liquid crystals increases their optical responses significantly due to increased induced intermolecular torque, and other guest-host effects. The guest-host mixtures can be employed in display applications, optical storage devices.

We investigate the nonlinearity of a liquid crystal cell doped with gold nanoparticles by considering their selective absorption. Such nonlinearities are promising for optical processing applications and optical limiters. Systems displaying thermal nonlinearities are particularly attractive as the maximum nonlinearity may occur in the absence of an applied field and additionally this nonlinearity can be controlled by the reorientation of the liquid crystal.

Due to the increased popularity of fast response display, the driving force to make high-speed switching liquid crystal (LC) material is raised. Doping nanoparticles such as carbon nanotubes (CNTs) in LC material is one of the methods to improve the display performance. Recently, many studies reported that enhancement of electro-optical (EO) performance was achieved by LC nanocomposites. The attraction of the hybrid system is not only the advance in EO performance, but also the alignment of CNTs. The disordered nature of CNTs makes it hard to expose the excellent properties and the self-ordered nature of LC material perfectly fit in this problem. Moreover, with LC acts as host, the CNTs orientation can be manipulated by external electric or magnetic field. This creates a promising future for novel nano-devices. On the other hand, there are doubts or questions on the stability of the nanocomposites and optimization of the sonication parameters and CNTs concentration that needed to be addressed before the mass production in the industry.

Nanoparticles (NPs) have emerged as extremely promising materials to alter and improve the properties of liquid crystals (LCs) used, for example, in device applications. In this paper, we summaries recent work from our lab that aims to provide a fundamental understanding of structure–property and composition–property relationships governing LC–NP interactions, which may point to new directions for major improvements in the efficiency of LCs used in display applications. A variety of LC hosts (phases) doped with surface-functionalized gold NPs have been systematically studied ranging from one-dimensionally ordered nematic over two-dimensionally ordered smectic to three-dimensionally ordered columnar phases. Significant progress with respect to LC–NP interactions was made for NP-doped nematic phases. Here, the observation of an unusual texture for Au NP-doped nematic LCs, that is, the formation of birefringent stripe textures and the induction of homeotropic alignment of the nematic LC similar to chiral finger (or fingerprint) textures, provided the basis for numerous experimental studies using Au NPs with different core sizes and surface functionalities.

INTRODUCTION

Research on liquid crystal (LC) has involved in Chemistry, Physics, Biology, electrical engineering, electronic engineering and many other fields. The

study of liquid crystals began in 1888 by Austrian Botanist F. Reinitzer when he prepared cholesteryl benzoate (the first liquid crystal); justified him to be called the grandfather of liquid crystal science. Liquid crystal materials are unique in their properties and

uses. In addition to display applications they are also promising materials for the photonic applications.

The discovery of liquid crystals is thought to have occurred nearly 150 years ago although its significance was not fully realized until over a hundred years later.

Around the middle of the last century Virchow, Mettenheimer et al. have found that the nerve fiber they were studying formed a fluid substance when left in water which exhibited a strange behavior when viewed using polarized light. They did not realize this was a different phase but they are attributed with the first observation of liquid crystals. Later, in 1877, further investigations of this phenomenon were carried out by the German physicist O. Lehmann who observed and confirmed, using the first polarized optical microscope designed by him, the existence of crystals which can exist with softness that one could call them nearly liquid. He found that one substance would change from a clear liquid to a cloudy liquid before crystallizing but thought that this was simply an imperfect phase transition from liquid phase to crystalline phase. The first reported documentation of the LC state was through an accidental observation by an Austrian botanist, Friedrich Reinitzer in 1888, working in the Institute of Plant Physiology at the University of Prague. He observed "double melting" behavior of cholesteryl benzoate. The crystals of this material melted at 145.50C into a cloudy fluid, which upon further heating to 178.50C became clear. This discovery represented the first recorded documentation of the LC phase.

He was the first to suggest that this cloudy fluid was a new phase of matter. He has consequently been given the credit for the discovery of the liquid crystalline phase. Presently, the liquid crystalline (LC) composites, filled with nano-scale colloidal particles, attract ever more and more interest. It was demonstrated that nanomaterial dopants with highly anisotropic (rod- or disc-like) shape can affect and improve the distinctive photonic and electro-optic characteristics of LC used for optical device and display applications.

Particularly interesting are LC composites based on chiral nematic liquid crystals (cholesterics). These materials exhibit selective reflection and giant optical activity that can easily be regulated by electric field and temperature. Doping of cholesteric liquid crystals (CLC) by nanoparticles can dramatically enhance their optical and opto-electronic characteristics. Introduction of ferroelectric particles in a CLC results in significant increase of the birefringence and dielectric anisotropy, as well as expansion of band reflection, and allows reduction of the driving voltage of switching between bistable textures. Addition of SiO₂ nanoparticles to the mixture of 5CB (39.75%) and cholesterol oleyl carbonate (COC, 60.25%) substantially affects the helical structure of the system, and introduced disorder leads to decrease in the phase transition temperature

and loss of the ability of selective reflection. The composites with magnetic nanoparticles dispersed in a chiral nematic LC are of great interest due to the possibilities of making onedimensional photonic crystals.

Doping of LC by highly anisotropic carbon nanotubes (NTs) allows reduction of the response time and driving voltage, as well as suppressing of the parasitic back flow and image sticking typical for LC cells. Also, remarkable electromechanical and electro-optical memory effects, as well as ultra-low percolation thresholds, were discovered. The previous experiments with NTs dispersed in cholesteric mixtures have demonstrated the impact of NTs on the selective reflection spectra, and it was suggested that NTs could destroy the translational order in the smectic phase of a CLC. Increasing of the concentration of chiral additive (cholesterol nonanoate) in the nematic LC did also affect the stability of 0.01% NTs dispersion accelerating the aggregation and sedimentation of NTs. The observed destabilization effect was explained by strong interactions of NTs with the helical structure of cholesteric LC and by the decrease of the helical pitch. The dielectric studies of the cholesteric LC (mixture of chiral additive ZLI-811 with nematic E7), filled by 0.5% of NTs, also have demonstrated the presence of interactions between NTs and the LC director. The chiral hybrid composites, based on the mixture of CLC and NTs, may be promising for construction of a gas sensor with high dynamic range. The functional ability of such chiral hybrid composites is determined by the nature of integration of NTs networks into the cholesteric structure that provides strong sensitivity of the optical and electrical properties of material to the external chemical and physical factors. However, the nature of such integration is still unexplored, and little is known about the properties of chiral hybrid composites on the basis of NTs and CLC.

The doping of liquid crystals (LCs) with nanoparticles has become a common method of improving their optical, magnetic, electrical and physical properties. For example, ferroelectric nanoparticles have been shown to decrease the Fréedericksz threshold and increase the birefringence of nematics. Similarly, ferromagnetic nanoparticles have been used to increase the sensitivity of the host liquid crystals to an applied magnetic field.

Gold nanoparticles have also been investigated as possible dopants for liquid crystals. Initially this was due to their potential to form a tunable, self-organizing, three-dimensional (3D) metamaterial suitable for the visible and infrared regions of the optical spectrum. However gold nanoparticles have since also been shown to improve the electro-optical properties of their host liquid crystals. Such suspensions display increased dielectric anisotropy and birefringence, lowered optical and electrical Fréedericksz thresholds, and increased thermal

stability. Moreover, even low concentration suspensions have increased the nonlinearity of hybrid photorefractive liquid crystal cells. Liquid crystals have also been shown to cause gold nanoparticles to spontaneously form linear self-assemblies and large 3D plasmonic crystals have been formed in an LC host using optical tweezers. Gold nanoparticles functionalized with liquid crystalline surfactants have been demonstrated to form large, complex, self-assembled structures, which exhibit anisotropic absorption. Furthermore these samples also show pseudo-liquid-crystal phases, with mesogenic functionalized gold nanorods showing a magnetically controllable nematic phase. Finally gold nanoparticle liquid crystal suspensions have been shown to exhibit large thermal nonlinearities with n_2 coefficients of up to $1.9 \times 10^{-5} \text{ cm}^2 \text{W}^{-1}$.

Gold nanoparticles are of particular interest due to their plasmon resonances, which occur in the optical spectrum. We are specifically interested in the decay of these plasmons, which leads to heating of the surrounding material, an effect that has already been investigated as a potential cancer treatment. In this paper we focus on the large thermal nonlinearity induced by the local heating of gold nanoparticle suspensions in a liquid crystal. While other absorbing systems such as dye-doped liquid crystals can exhibit giant nonlinearities of up to $1000 \text{ cm}^2 \text{W}^{-1}$, these require the application of a bias field. Gold nanoparticles are preferable to dyes as absorbers for several reasons: their absorption cross section is greater by over an order of magnitude and, due to their excellent thermal stability, they do not bleach.

The nonlinearity of gold nanoparticle liquid crystal composites has been investigated experimentally and theoretically by Ouskova et al. with an aim towards optical processing applications. They doped a nematic liquid crystal with small gold nanospheres in order to enhance its thermal nonlinearity. This paper shows a schematic diagram of their experimental setup. Two pump beams write a refractive index grating on the liquid crystal cell, which is probed by a third beam. They also modeled the thermal nonlinearity by solving the thermal diffusion equation. The best theoretical estimates they obtain are larger than the experimental values. In particular, the overshoot is larger in the system with greater absorbance of the light beams. In order to explain this difference we extend here their model to include the effect of pump and probe absorbance.

LITERATURE REVIEW

Composite materials are engineered or naturally occurring materials made from two or more constituent materials with significantly different physical or chemical properties. They remain separate and distinct at the macroscopic or microscopic scale within the

finished structure. In terms of nanocomposite, it is a multiphase material where one of the phases has one, two or three dimensions of less than 100 nanometers (nm), or structures having nano-scale repeat distances between the different phases that make up the material. The properties of composite material are highly depends on the ratio of the constituent materials. For example, high stiffness can be achieved by adding larger mass fraction of carbon fiber into carbon-fiber reinforced materials. But when material is in the nano scale, the mechanical, electrical, thermal, optical, electrochemical, catalytic properties will differ markedly from that of the original materials and this situation fully adapt to nanocomposite. The properties of nanocomposites differ from conventional composite materials because of the extraordinarily high surface to volume ratio or aspect ratio. The area of the interface between the matrix and reinforcement phase is typically an order of magnitude greater than the conventional composite materials and the percentage by weight of the nanoparticles introduced can remain very low (on the order of 0.5wt%-5wt%). In general, composite or nanocomposite materials are designed to improve the certain properties of the matrix material by adding a small amount of second phase into the matrix.

The liquid crystalline nature of cholesterol was first found by an Austrian physiologist called Friedrich Reinitzer by extraction from carrots in 1888. He found that cholesteryl benzoate does not melt in the same manner as other compounds but with two melting points. At 145.5°C it melts into a cloudy liquid and at 178.5°C it melts again into a clear liquid. Remarkably, the phenomenon is reversible. By further examination in collaboration with Otto Lehmann & von Zepharovich, the intermediate cloudy fluid was found to be crystalline. By that time, Reinitzer had described three important features of cholesteric liquid crystals: 1) the existence of two melting points, 2) the reflection of circularly polarized light, and 3) the ability to rotate the polarization direction of light.

After Reinitzer's accidental discovery, he did not study liquid crystals further. But it was continued by Lehmann as he realized that he had encountered a new phenomenon. Lehmann started a systematic study, the first material is cholesteryl benzoate, and then of related compounds which exhibited the double-melting phenomenon. He was able to make observations in polarized light. Using his microscope equipped with a hot stage maintained at high temperature. The intermediate cloudy phase clearly sustained flow, but other features under a microscope, convinced Lehmann that he was dealing with a solid.

Lehmann's work was significantly expanded by a German chemist called Daniel Vorländer, who had

synthesized most of the liquid crystals known from the beginning of the 20th century until his retirement in 1935. However, liquid crystals were not popular among scientists and the material remained a pure scientific curiosity for about 80 years.

Liquid crystal is a state of matter that is intermediate between the crystalline solid and the amorphous liquid. It can be also regarded as a liquid with ordered arrangement of molecular orientation and it has anisotropic mechanical, electric, magnetic and optical properties. A LC molecule has a rod-like structure with rigid core and flexible alkyl chains. It has an aspect ratio from around 3 to around 10 and the rod-like molecular shape is responsible for the anisotropic properties.

In general, phase transformation between crystalline, anisotropic and isotropic phases is reversible and it can be obtained by either dissolving in some solution (lyotropic) or by heating or cooling (thermotropic). There are three phases of liquid crystal: Nematic, smectic, and cholesteric. Nematic liquid crystals, most widely used phase, have threads distributed all over the area. The molecules should be more or less parallel to each other and it can move in all directions with free rotation along the long molecular axes. The average direction of the long axes of the molecules is called director. Nematics have fluidity similar to that of ordinary (isotropic) liquids but they can be easily aligned by an external magnetic or electric field. Aligned nematics have the optical properties of uniaxial crystals and this makes them extremely useful in displays. Smectic phase have layered structure and the layers can slide over each other and give rise to the flow characteristics. It can be divided into several subclasses by different molecular arrangement inside the layers. In the Smectic A phase, the molecules are oriented along the layer normal, while in the Smectic C phase they are tilted away from the layer normal. These phases are liquid-like within the layers. There are many different smectic phases, all characterized by different types and degrees of positional and orientational order. Cholesteric phase can be viewed as the nematic state superimposed with the natural twist. That means the average direction of the molecules in every layer is unidirectional skewed to the layer just below it.

Like most liquids and solids, liquid crystals exhibit curvature elasticity. The elastic constants of a liquid crystal determine the restoring torques that arises when the system is perturbed from its equilibrium configuration. In LCD, electric field is often applied to cause a reorientation of the molecules. The static deformation of LC is determined by the balance between the electric torque and the elastic restoring torque and it can be divided into a combination of three basic deformations: splay, twist and bend.

GOLD NANOPARTICLE-LIQUID CRYSTAL COMPOSITES

Over the past few decades, research on nanomaterials has greatly influenced the emerging field of nanoscience and nanotechnology. The huge impact of nanomaterials in presentday technology is solely based on their size-dependent electronic, optical, magnetic, and chemical properties. Under this framework, research into micro/nano-scale colloids dispersed in liquid crystals (LCs) has facilitated enormous interest, due to their potential scientific and technological relevance. LCs are the best examples of anisotropic fluids and have proven to be an appropriate medium to study the interaction among colloidal particles and LCs. Several theoretical and experimental investigations on colloidal particles dispersed in LCs have been carried out, wherein the shape/size of colloids and the selfassembling nature of the LCs are widely taken into consideration.

Topological defects (Saturn rings, hedgehog space, and boojums) are produced due to the elastic deformation of the LC by the dispersed colloidal particles, which further mediate the long-range anisotropic interactions among colloids, leading to their controlled organization within the LC matrix. Furthermore, the formation of topological defects also provides a platform to better understand the dynamics of alignment of LC molecules in the vicinity of colloidal particles.

More importantly, the multibillion dollar LC technology has recently tied up with nanotechnology to modify the physical properties of LCs through nanomaterials and vice versa. For example, the use of metal nanoparticles (MNPs) to tailor the properties of LCs and LCs to tune the properties of MNPs has opened up several avenues for these LC-MNP composites to be utilized in several opto-electronic devices.

Among all MNPs, gold nanoparticles (GNPs) dispersed in LCs have been widely studied and found to be pivotal for technological applications. In this article, we review (a) SPR formation/behaviour in GNPs, and (b) GNP-LC nanocomposites: tuning the physical features of GNPs by using LCs and vice versa.

The GNP-LC composites are usually prepared by dispersing capped GNPs into the LCs at an isotropic temperature, followed by ultrasonication and evaporation. Wherein, the use of capped GNPs (Fig. 1) is very significant since capping (non-mesogenic, mesogenic, or both): (i) controls the miscibility of the GNPs into the LC, (ii) prohibits the agglomeration of GNPs into the LC, and (iii) maintains the long-lasting stability of the GNP-LC composites. Detailed information about the capping of GNPs can be readily obtained from very thoughtful reviews and other reports. Despite the numerous advantages of the capping strategy, certain issues like: (i) tedious chemical reactions for functionalization of the GNPs,

(ii) the need for removal of unwanted by-products, (iii) their limited purity and production, and (iv) the prohibited direct interaction between the GNP and LC, still remain. In view of these limitations, a novel method 'Sputter Deposition' has been recently devised for uniform dispersion of GNPs into the LC host. It is reported that gold is sputter deposited uniformly in the form of nanoparticles into the LC host having a vapor pressure of less than 1 Pa. It is speculated that certain parameters, such as the wettability and elastic constants of the LC, are responsible for controlling the size of the GNPs. Such sputtered nanocomposites have been found to be stable for even more than three months. More importantly, this method is (i) completely physical and hence provides an easier, faster, and more pure production as compared to other techniques, and (ii) also applicable to the fabrication of dielectric, semiconductor, ferroelectric, and magnetic nanoparticles. However, the disadvantages of method are that (i) only the use of hosts (liquids, oils, etc.) with a low vapour pressure is feasible, and (ii) the size of nanoparticles depends upon the physical properties of the hosts.

Last but not the least, it is of worth to point out that the original dispersing medium (i.e. the solvent) of the GNPs should be chemically non-reactive with the LCs. Otherwise, the medium may affect the physical properties of the LC before its evaporation from the composite. Therefore, it is strongly advised to choose solvents that are negligibly reactive with LCs and can be easily evaporated from the GNP-LC composites.

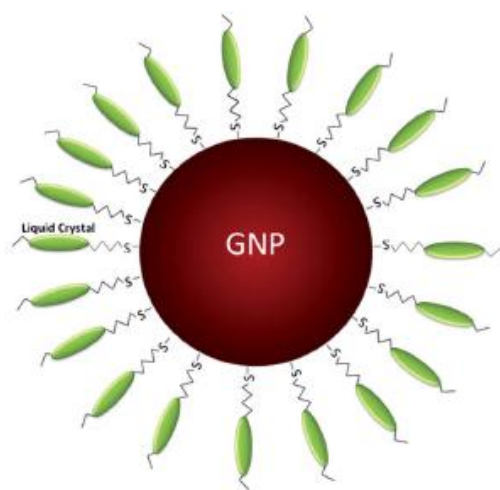


Fig. 1 The schematic shows a mesogenic monolayer-capped GNP.

Now we review both aspects of GNP-LC composites: (i) tuning of the GNP's properties by LC (nematic, smectic, and their chiral analogues) and (ii) tuning of LC's properties by the GNP.

Tuning of gold nanoparticle properties by liquid crystals-

Nematic and chiral nematic (or cholesteric) liquid - crystals. Nematic liquid crystals (NLCs), composed of rod-like mesogens, are found to be the simplest anisotropic dielectric systems in terms of easier understanding of their molecular (i.e. director) alignment and related physical properties. For example, the dielectric constant of the NLC can be altered through variation of the temperature and externally applied electric/magnetic field and hence has proved to be one of the best anisotropic dielectric media to tune the optical properties of GNPs in GNP-LC composites.

Tuning of liquid crystal properties by gold nanoparticles-

Nematic and cholesteric blue phase liquid crystals - Prasad et al. investigated alkylthiol-capped GNP (~15–20 nm, ≤5 wt%) doped NLC composites and observed that the electrical conductivity and nematic-to-isotropic phase transition temperature (T_{N-ISO}) of the NLC are dependent on the concentration of GNPs. The T_{N-ISO} of the composite decreases almost linearly with increasing concentration of GNPs. The electrical conductivity increases with doping concentration of GNPs, whereas the conductivity anisotropy decreases very minutely.

The authors believed that the observed results could be due to the presence of GNPs and not their capping agent (i.e. alkylthiol). Sridevi et al.⁸¹ also studied GNR-doped NLC composites and observed enhanced anisotropic conductivity, elastic and dielectric constants, but no measurable changes in T_{N-ISO} of the composite as compared with the undoped sample, unlike Prasad et al. The results were explained on the basis of the shape anisotropy of the GNRs. On the other hand, Khatua et al. reported an increase in T_{N-ISO} and a decrease in the threshold voltage for the Fréedericksz transition of NLC doped with SOPB-capped GNP (~6 nm, ~0.2 wt%) composites.

Smectic liquid crystals - The doping of citrate ioncapped GNPs is mostly experimented with the chiral Smectic C phase (SmC*) among all smectic liquid crystalline phases, due to the technological importance of the SmC* phase. Kaur et al. reported the enhanced optical and electro-optical properties in GNP-doped ferroelectric liquid crystal (FLC) composites. The five-fold increase in optical tilt, reduction of the threshold voltage by one-tenth, and the enhanced memory effect (~half an hour) was monitored in doped FLC samples.

EXPERIMENTAL METHODS

The liquid crystal (LC) materials have proved themselves the most promising candidates in the field of highly growing electro-optical display devices as well as various non-display devices having tremendous impact on the scientific society and human beings. The liquid crystal has very interesting physical and optical properties. Therefore, a systematic study is very important to know the physical and optical chemical, electro-optical and dielectric properties of these materials.

Instruments and Experimental techniques –

Instruments user :

1. Spin coater
2. Science Tech India Hot Air Oven
3. Physical Balance (Sartorius)
4. Polarizing Microscope (Radical, RXLr-5)
5. Instec HCS 302 Hot Plate
6. Impedance Gain/Phase Analyzer HP4194A
7. Impedance/Gain Phase Analyzer Solartron (SL 1260)
8. UV Visible Spectrometer ELICO SL164
9. Storage Oscilloscope HM407
10. Temperature Controller Julabo F-25
11. Self-Designed 3-In-1 Analyzer
12. Textronics Oscilloscope (Model Number TDS 2024C)
13. Textronics Function Generator (Model Number AFG3021B)
14. Photo Detector (Model PD02LI)
15. Hydraulic Press (MP -15)
16. Shimadzu Fourier Transform Infrared Spectrophotometer
17. Differential Scanning Calorimeter (DSC 200 F3 MAIA)
18. Photomultiplier Tube

The dielectric, electro-optical properties and materials constants of LCs can be determined with help of the above mentioned instruments. The sample cell is mounted onto the sample holder of the hot/cold stage

and kept on the rotating table of the polarizing microscope. The sample can be viewed through the microscope.

Experimental Identification of LC -

In the course of research and commercial manufacturing, it is vitally important to be able to identify the types of liquid crystals phases that are exhibited by compound or a mixture of compounds.

The most widely used techniques of liquid crystal phase identification are optical polarizing microscopy, which reveals that each different liquid crystal phase has a distinct optical texture. However this identification of liquid crystal phases through optical polarizing microscopy is often difficult and requires a lot of experience.

However the ultimate technique for the identification and classification of mesophases is X-ray analysis. X-ray analysis of a liquid crystal will map the positions of the molecules within the phase and hence determine the phase structure and classification to which the particular phase belongs. However to maximize the information aligned samples are required. Miscibility study is another method of identifying the mesophase. The material with unknown mesophase is mixed with a known material that possesses mesophases that have already been identified. If a particular mesophase of the unknown material is completely miscible with a known mesophase, then both the mesophases are identical. Other technique used to identify the structure of mesophase and liquid crystalline mesophase include neutron scattering and nuclear magnetic resonance.

Optical Transmittance -

The optical transmittance measurement has been done by placing the cell between two-crossed polarizers of polarizing microscope model (Radical, RXLr-5) fitted with a hot stage. The complete experimental arrangement for the optical transmittance measurement is shown in figure 2.



Figure 2. Optical transmittance measurement and texture study setup used in present work. The optical texture showing the texture clicked.

The most fascinating thing about this method is the visualization and clicking of sample texture during measurement by camera (Model Prog Res CT3) fitted on one of the eyepieces of the microscope. The light intensity coming through one of the eyepieces has been measured by Photo Detector (Model PD02LI). The optical transmittance obtained from the photo detector (Instec-PD02LI) is directly fed to a digital storage oscilloscope (Tektronix TDS-2024C) in an electrical form. The output wave form intensity is then used to determine the transmittance.

CONCLUSION

We have modeled the optical thermal nonlinearity of gold nanoparticle liquid crystal suspensions by extending previous models to include the attenuation of the pump and probe beams. As our model gives a much better fit to existing experimental data we can conclude that the consideration of this attenuation is key to accurately predicting the behavior of the thermal nonlinearity. Due to the attenuation of the beams, the scaling of the induced refractive index grating with the number of absorbers is no longer trivial.

Every aspect of GNPs such as the formation/behaviour of SPRs, role of capping, uniform dispersion into LCs, tuning of GNP properties by LCs and vice versa are widely covered in this article. The SPR peak of GNPs can be tuned by changing either the dielectric constant of the surrounding medium or the size of the GNPs. LCs are recognized as one of the best surrounding mediums for GNPs, because the dielectric constant of LCs can be easily changed by the external electric field as compared to other dielectrics. The alignment of LC molecules around GNPs also provides a good platform to understand the dynamics of interaction between GNPs and LC molecules and hence the affected SPR peak, finally leading to ordering of GNPs and sensing applications.

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