Structured Light by Rotating Au Nanoparticles in a Dynamic Distribution

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Abstract. Herein is reported the potential impact of the optical Kerr effect exhibited by rotating plasmonic nanoparticles on the generation of structured light. The third-order nonlinear optical properties exhibited by a round continuously variable distribution of metal nanoparticles incorporated in a dielectric substrate were analysed. The nanosystems were studied by using the finite difference method and TiO2-supported Au nanoparticles explored by a vectorial two-wave mixing technique. Attractive applications for developing quantum functions assisted by mechano-optical effects can be considered.

1. Introduction

The concept of structured light has emerged from the discovery of a fascinating characteristic in the orbital angular momentum (OAM) associated with electromagnetic waves [1]. Some propagation modes of electromagnetic radiation can correspond to a particular OAM [2]. In order that an optical beam can be related to an OAM, the beam must be characterized by a specific kind of phase wavefront. Specially, in structured light, the angular location around the center of its wavefront follows a pattern; and in the peculiar case of twisted light, the phase would either monotonically increases or decreases as a spiral shape goes around from the center trajectory of the beam [3]. The phase wavefront related to twisted light describes a helix geometry as the beam propagates [4]; outstanding applications for spin-to-orbital angular momentum conversion in all-optical systems have been envisioned [5]. Moreover, circular polarized light carries an angular moment of rotation that represents a possibility for developing momentum transmission and quantum communication [6].

A simple method for generating an orbital angular momentum of light is based on an induced shift in the perpendicular electric vectorial components of a circularly polarized beam in propagation through a wave retarder [7]. However, several techniques can be useful for the generation of structured light by including spiral phase plates [8], holograms [9], stimulated Raman scattering [10], plasma interactions [11], or nonlinear phenomena [12].

Remarkably, the nonlinear optical properties exhibited by nanoparticles have been pointed out to be good candidates for designing ultrafast photonic functions regarding their powerful surface plasmon resonances [13]. Furthermore, the third-order nonlinear optical properties of metal nanoparticles can be easily controlled by multi-functional signals that may involve quantum magneto-conductive effects [14], fractional thermal phenomena [15], or mechanical actions [16]. In this direction, in this work an attempt
has been made to further investigate the generation of structured light by the assistance of mechano-optical effects together to plasmonic nanoparticles.

2. Materials and methods

The propagation of optical waves through a round continuously variable distribution of metal nanoparticles incorporated in a dielectric substrate was analyzed. Gold nanoparticles embedded in a TiO$_2$ film were considered as a nonlinear media with an average nanoparticle size of 20 nm and a maximum nanoparticle density 2$\times$10$^{10}$ particles/cm$^2$. The linear decrease in the distribution of the nanoparticle density in the thin film sample with a disc shape reached a minimum nanoparticle density 1$\times$10$^7$ particles/cm$^2$. The thickness of the film was 700 nm. A 532 nm wavelength provided by a laser system was selected as an optical source taking into account the excitation of the localized surface plasmon resonance of the Au nanoparticles around this wavelength. The transmittance of plane waves were evaluated taking into account a fixed position of the sample and the centre of the spot size of the beam coincides with the centre of the sample in disc shape. The incident polarization of the beams in the two-wave mixing was linear with irradiance for each beam of 500 MW/cm$^2$. We consider pulses of 4 nanoseconds focused in a spot diameter of 1 mm. Comparatively, we analysed the propagation of the beam through the sample in rotation and with the beam along a non-centred section of the disc. A two-wave mixing with the rotating sample was proposed in order to control an induced birefringence in the sample with potential application for dynamic structuration of light by this simple method.

The distribution of electric fields involved in a two-wave mixing process can be described by using the Fresnel-Kirchhoff diffraction integral [17],

$$U(x,y,z) = \frac{1}{i\lambda} \int_S U(x_0,y_0) \left[ \frac{\cos(\vec{n} \cdot \vec{r}) - \cos(\vec{n} \cdot \vec{r}')}{2} \right] e^{i\kappa r} ds$$

where $\lambda$ is the wavelength, $U(x_0,y_0)$ describes the amplitude distribution located at the $z = 0$ position $S$, with a normal direction $\vec{n}$, $r$ and $r'$ correspond to the vectors located between the point $z = 0$ and a normalized point in the plane $z$, otherwise $\kappa = 2\pi/\lambda$ is the wavevector used for the numerical simulations.

The numerical estimation of the induced birefringence was approximated by the finite difference method considering the wave equation governing a two-wave mixing method [18]:

$$\nabla^2 E_{\pm} = -\frac{n_0^2 c^2}{c^2} E_{\pm}$$

where $\omega$ is the angular frequency of light, $c$ is the free space speed of light, and the refractive index was approximated by [18]:

$$n_{\pm}^2 = n_0^2 + 4\pi \left| A \right| E_{\pm}^2 + (A + B) E_{\pm}^2$$

where $n_0$ is the linear refractive index, $A = \chi^{(3)}_{1112}$ and $B = \chi^{(3)}_{1121}$ are corresponding to the third-order nonlinear optical susceptibility tensor $\chi^{(3)}$. It is possible to calculate a phase-change, $\theta$, derived from the optical Kerr effect of the sample governed by [18]:

$$\theta = \Delta n L = n_2 I_f L$$

where $\Delta n$ is the change in refractive index, $n_2$ represents the nonlinear refractive index, $I_f$ corresponds to optical irradiance in propagation through the sample and $L$ is described by the optical path involved in the nonlinear optical processes of the study. The angle of rotation, $\phi$, can be estimated by [18],

$$\phi = \frac{1}{2} \Delta n \frac{\sigma}{c} L$$
here $\sigma$ describes the optical frequency and $c$ is assumed to be the speed of the light.

3. Results and discussion

In order to see the potential coupling in the nanoparticles by increasing the nanoparticle density or decreasing the interparticle distance, we used the Mie theory and Fröhlich condition for modeling an excited pair of nanoparticles interacting with an electric field below 70 KV/m. We were taking into account that the ablation point for optical damage of the Au nanoparticles in TiO$_2$ is close to 1 GW/cm$^2$.

It can be stated that the shape and size of the nanoparticles also play a crucial role in the local field enhancement through the excitation of the surface plasmon resonances. In figure 1a are plotted numerical results of the electric field intensities attributed to the nanoparticles governed by equation 1. From figure 1a can be observed an intensification of the electromagnetic field together to a quenching of the electromagnetic interaction in the surrounding of the nanoparticles studied.

![Figure 1](image1.png)

Figure 1. (a) Numerical results of the electric potential distribution field for two gold nanoparticles interacting with a near resonance optical excitation. (b) Calculated optical-phase change evolution of one beam in a two-wave mixing as a function of irradiance and nanoparticle density of the studied sample.

The transmitted beams in the two-wave mixing show important changes derived by the participation of the collective response of the nanoparticles. From a two-wave mixing configuration interacting with a nonlinear media, an interference irradiance pattern is generated in the superposition of the beams. Because of this interference effect in high-intensity conditions, a modulation of the nonlinear refractive index according to equation 3 can be originated in the sample. The modulation in the refractive index as a birefringence grating also produces a modification of the polarization of the incident beams as described in a standard optical Kerr gate [18]. In figure 1b is shown the evolution of the optical-phase change as a function of the nanoparticle density of the nanoparticles. The estimated nonlinear refractive index for $1 \times 10^{10}$ particles/cm$^2$ was $n_2=9.1 \times 10^{-11}$ cm$^2$/W, while the nanoparticles with a density $1 \times 10^7$ particles/cm$^2$ exhibit an estimated $n_2=3.7 \times 10^{-15}$ cm$^2$/W according to comparative samples experimentally measured [19]. The optical-phase change was calculated by using equations (2-5).

In figure 2a is schematized the sample with one of the beams involved in a two-wave mixing interaction, considering its potential to generate twisted light from a strong modification in nonlinear refractive index and optical phase-change in the different regions with a modulated nanoparticle density. This result is in good agreement with the proposed method that obtains twisted light beyond the visible by an induced grating [20]. The modification in the OAM of light results in our case from the
participation of the optical Kerr effect that depends on optical irradiance. However, the contribution of the collective behavior of the nanoparticle density in the sample represents a strong influence in the optical-phase change that generates the structured light. An additional aspect for modulating the optical-phase change and the orbital angular momentum can be given by different spatial distributions of the nonlinear nanostructures which can be obtained by the rotation of the sample. The rotation of the sample is proposed to be conducted by a servomotor with the sample interacting in a two-wave mixing configuration. In figure 2b is illustrated the potential modulation of structured light from a rotating sample with a round continuously variable distribution of metal nanoparticles incorporated in a dielectric substrate.

The experimental verification of the phase-change by a two-wave mixing in rotating bimetallic nanocomposites has previously reported in comparative samples [21]. Plane wavefronts in the system proposed in figure 2b can result in structured light controlled by the irradiance and speed of the sample.

4. Conclusions

Modification in the optical-phase change by the dynamic collective response in plasmonic nanoparticles incorporated in a rotating sample was analyzed. The third-order nonlinear optical properties induced by a two-wave mixing configuration were proposed for generating structured light by rotating nanostructures dispersed in a gradient distribution. Coupling effects and powerful third-order nonlinear optical behavior in metal nanoparticles can take a crucial role in the modulation of angular momentum of ultrafast laser pulses. The sharp selectable surface plasmon resonance phenomena in dynamic mechano-optical functions can be useful for developing future all-optical systems and quantum information processing devices.

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