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Invited Paper

Resolving the multipolar scattering modes of a submicron particle using parametric indirect microscopic imaging

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ABSTRACT

In this work, we show the spatial distribution of the scattered electromagnetic field of dielectric particles by using a new super-resolution method based on polarization modulation. Applying this technique, we were able to resolve the multipolar distribution of a Cu_2O particle with a radius of 450 nm. In addition, FDTD and Mie simulations have been carried out to validate and confirm the experimental results. The results are helpful to understand the resonant modes of dielectric submicron particles which have a broad range of potential applications, such as all-optical devices or nanoantennas.

1. Introduction

High-permittivity dielectric materials, such as silicon, are widely used in micro and nano-electronics due to their electric properties, and compatibility with CMOS techniques. Also, they are important in the design of optical components, such as nanoantennas [1]. Their low losses in the visible and near-infrared ranges, as well as their capacity to support either electric or magnetic Mie resonances make them unique in the control of light scattering [2]. Directional light scattering has been reported in dielectric nanoparticles, including spherical [3–5] and cylindrical [6,7] structures. These features make dielectric nanoparticles an alternative to plasmonic ones [8,9]. In this sense, they can potentially be the basic element of upcoming all-optical devices in a wide range of applications, ranging from bio-sensing [10] to new communication devices [11,12]. Thus, the interest on light scattering of dielectric nanoparticles has strongly increased in the last years [13–16]. The fabrication of silicon is not trivial because of the difficulty in controlling the shape of the particles [39]. Several works have been reported on other high refractive index materials which support the Mie resonances such as Germanium (Ge), Gallium Arsenide (GaAs), Indium Phosphide (InP) etc. [40]. In this work, we analyze the scattering properties of sub-micron particle made of copper (I) oxide (Cu_2O), whose optical properties are adequate to support Mie scattering resonances in the visible region [41]. This material provides us a good control in fabricating different shaped nanostructures as compared to silicon [38] and already has potential applications in solar energy conversion, catalysis and gas sensors, which may be improved with the emergence of these resonances, enlarging its applicability [39].

The first theoretical and experimental demonstration about the electric and magnetic dipolar resonance from silicon nanoparticle in the visible range was reported [17,18]. Several studies about electric and magnetic resonance in dielectric nanoparticles can be found in the literature [19–23]. Both near- and far-field scattering properties are worthy of investigating due to their importance in the interactions with adjacent structures and their practical implementation, respectively. The far-field analysis is usually performed by dark-field spectroscopy [24]. However, the imaging technique is also quite interesting. Unfortunately, the resolution of conventional optical microscopy is constrained by the diffraction limit and thus cannot resolve the sub-wavelength features of the sample under

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test (SUT) in the far field region. The information about small features of the SUT is included in the evanescent waves, which decay exponentially with distance and are detectable only in the near field region. In the far field region, the loss of these evanescent waves and the information that they contain precludes reconstructing an image of an object with a resolution better than one-half of the incident wavelength [25].

While commonly used dielectric nanoparticles have simple shapes, their arrangements may have sub-wavelength details, like hot-spots in dimer structures [26]. Super-resolution far-field method is deserved to be explored and it is full of challenges. Many super-resolution far-field imaging techniques are based on fluorescence, few non-fluorescent species based far-field imaging can achieve super-resolution [27]. However, these standard far-field techniques offer limited information about subwavelength scattering signal distribution in the far field region especially about the high-order multipolar modes, which provides new opportunities for directional and complex-polarization controlled emission from nano-emitters [28,29]. In fact, we previously reported a polarization-based method which resolves sub-diffraction details in graphene layers [29]. This technique, which is known as Parametric Indirect Microscopic Imaging (PIMI), resolves sub-diffraction features of the SUT by modulating the optical field, fitting the field intensity variation curves of each pixel of the image and extracts the indirect polarization parameters. The imaging process reduces the width of the point spread function (PSF) by filtering off the irrelevant scattering from neighboring source points with different local structure characteristics in the SUT and leads to break the diffraction limit [29]. In this work, we successfully applied PIMI technique to study the electromagnetic (EM) scattering of copper (I) oxide (Cu_2O) particles. In particular, we report the spatial distribution of the scattered localized optical modes of a Cu_2O submicron particle. A comparison between experiments and simulations also shows the accuracy of this method to characterize light scattering of high-permittivity dielectric nanoparticles. The successful results of far-field scattering characterization, even when magnetic modes arise, show that PIMI is a powerful tool to perform future super-resolution imaging of complex arrangements of dielectric resonant nanoparticles.

1.1. *Theoretical and experimental basis*

Before impinging on the SUT, the polarization status of the incoming light is modulated in a controlled and precise manner in PIMI. The linearly polarized light with different polarization angles as shown in Fig. 1(b) excite different scattering information due to the anisotropy present in the SUT. This anisotropy produces near to far field coupling. When this light comes to the CCD after the interaction with the SUT, it can be formulated with the Jones model [42,43] and also we can check whether the derived intensity follows the near to far field coupling principle or not [29]. The spatial points which follow the coupling principle fit well with the fitting criteria such as adjusted-root-square (Adj-R-Square) [29], whereas the others do not follow filtered off as noise. Only the middle portion of the pixel intensity confirms the near to far field coupling and the remaining spatial field points filter off. In this way, the width of the PSF decreases and we become able to sense the scattering signals beyond the diffraction limit. The Fig. 1(a) shows the fitting curve fitting the experimental data points.

In Fig. 1(b), a schematic of the polarization illumination has been shown and Fig. 1(c–h) shows the recorded scattered field of a Cu_2O sphere under the different polarization illuminations. As it can be seen in Fig. 1(c–h), at every different polarization illumination, we get different scattered field distribution and then by using our reconstruction method, we fit every data point as shown in Fig. 1(a) and extracts fine information about the scattering field. Due to this extra scattering sensing, PIMI makes us enable to study the angular scattering distribution of the scatterer in a much better way. By using the anisotropy, $sin\delta$, which is the phase shift between the phase of the two electric field components, Ey and Ex [30], the azimuthal angle or the slow vibration angle, the angle along the slow vibrational axis [2] and I_{dp}, we calculated the Stokes parameters. I_{dp} is the average of all polarization scattered from the SUT. This means that if, for instance, we rotated the linear polarization field with an angle step of 36° i.e. 0°, 36°, 72°, so on, I_{dp} is the average of all scattering field information of these polarizations [43]. By using a relationship between the Jones and Muller models, we previously introduced a model to calculate the Stokes parameter [29].

$$\begin{aligned} S_0 &= I_{dp}(1+\sin\delta) = E_{0x}^2 + E_{0y}^2 \\ S_1 &= I_{dp}(1+\sin\delta)\cos 2\phi = E_{0x}^2 - E_{0y}^2 \\ S_2 &= 2\sqrt{I_{dp}}(1+\sin\delta)\cos 2\phi = 2E_{0x}E_{0y}\sin\delta \\ S_3 &= 2\sqrt{I_{dp}}(1+\sin\delta)\sin 2\phi = 2E_{0x}E_{0y}\cos\delta \end{aligned} \tag{1}$$

Where E_{0x} and E_{0y} are the amplitude of the electric field on the x and y-axis [30], respectively, which can be derived from our experimental data using Eq. (1). The PIMI parameters not just provide us the information about the scattering distribution around the scatterer, instead, they delivered the information about the polarization states, polarization degree, and polarization angle.

2. Results and discussion

An experimental setup of the PIMI system has been displayed in Fig. 2(a) whose details are present in the method section. The considered particles are made of copper (I) oxide (Cu_2O), a dielectric material with a relative high-refractive index (3.22 + 0.033i) at 532 nm [41] to support Mie modes [1–3] and with low losses in the visible and near-IR range [31]. Crystalline Cu_2O nanoparticles were deposited on a silicon (Si) substrate and characterized through SEM (Scanning Electron Microscope) images. A scanning electron micrograph of a Cu_2O particle is shown in Fig. 2(b). It can be seen that it has a well-defined spherical shape and it has an average radius of 450 nm. This particle size allows a simple and more accurate fabrication than using sub-wavelength particles as well as the excitation of several multipolar modes at the considered wavelength. The probed operation of the method in sub-diffraction structures [30] enables to assume that it can also be used in the characterization of a nanoparticle.

Mie calculations were performed to investigate the optical response of these particles and, in particular, the multipolar contributions. Fig. 3(a) shows both the scattering and absorption efficiencies of isolated Cu_2O particles with a radius of 450 nm as a function of the incident wavelength calculated by Mie theory [32]. At the wavelength of interest (solid line at 532 nm in Fig. 3), the scattering effect is dominant; however, a non-negligible absorption is still presented, which limits the observance of pure scattering phenomena. Fortunately, scattering signals have a strong intensity that could be detected by our experimental set-up. Operation at longer wavelengths enables the dominant scattering behavior and reduces the influence of absorption losses. The different multipolar scattering contributions are displayed in Fig. 3(b). The high radius/wavelength ratio of the considered particle supports several multipolar contributions. It should be mentioned that only the dominant ones in the considered wavelength range are shown in this figure. In particular, electric and magnetic dipolar (ED and MD), quadrupolar (EQ and MQ) and octupolar (EO, MO) contributions are displayed. These modes are directly related to Mie coefficients a_1, b_1, a_2, b_2, a_3, and b_3, respectively [33]. At the considered incident wavelength (532 nm), several modes have significant values. The