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Nonlinear Geometric Phase Gradient Metasurfaces beyond the Dipole Approximation

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Abstract: In this work, we identify the role of higher order antenna's modes on a metasurface's Pancharatnam – Berry phase by investigating second harmonic light scattering from two metasurfaces exhibiting dipolar and quadrupolar radiation.

1. Introduction

Compact nanostructured surface arrays of optical antennas or "metasurfaces", [1] enable near-continuous control of the spatial phase of optical wavefronts formed by the light that they scatter in the linear and nonlinear regime. This is achieved by controlling antenna dispersion, via each antenna's shape and size or engineering a geometric phase i.e. Pancharatnam−Berry (PB) phase via the interplay of antenna orientation and circularly polarized illumination. In the case of nonlinear metasurfaces, the PB phase approach has been studied for metasurfaces exhibiting third-harmonic generation (THG), second-harmonic generation (SHG) and four-wave mixing (FWM). However, in these studies, the variation of the nonlinear phase across a surface, and the resulting diffraction, was understood in terms of the rotational symmetry of each antenna [2] and a reformulation of the grating condition into a generalized law of nonlinear refraction. To predict the location of diffraction orders, some of these studies evaluated the grating equation shifted by the nonlinear PB phase. Yet, the grating equation assumes point dipole-like scattering and optical antennas, dielectric or metal, rarely scatter as simple point dipoles. Moreover, octave spanning nonlinear light scattering must naturally include high-order modes.

2. Results and Discussion

In this work, we aim to identify the role of high-order antenna modes on a metasurface's PB phase. To do this, we investigate the nonlinear scattering from two metasurfaces made of (A) centrosymmetric antennas (gold bar) and (B) non-centrosymmetric composite antennas (a gold bar and two nearby disks), as shown in the scanning electron microscopy (SEM) of figure 1a and 1b. These nanoantennas are known to exhibit "bright" dipole-like and "dark" quadrupole-like SHG. [3] Optical beam can excite the dipolar resonance of the bar (see figure 1c), and induce a quadrupole – like SHG response via the coupling of the nonlinear polarization to second-order modes. Meanwhile, the composite antennas can induce a dipole-like SHG response via near field coupling to additional dipolar resonances, supported by the disks that decorate the bar.

To investigate the PB phase-induced nonlinear diffraction, we incorporate these antennas, labelled by indices (m,n), at positions $\boldsymbol{\rho}_{m,n}$, with azimuthal rotation angles, $\phi_{m,n}$, into square arrays constituting two metasurfaces with a nearest neighbour rotation of $\Delta\phi = \phi_{m,n} - \phi_{m-1,n} = \pi/3$ along the x̂ direction. When illuminated by circularly polarized light, the relative rotation angle of antennas establishes a phase gradient that is dependent on the nonlinear process and scattering polarization. For SHG, each antenna acquires a local phase $\psi_{m,n}^{SHG} = (2\sigma_{in} - \sigma_{out})\phi_{m,n} = \Delta\sigma_{SHG}\phi$. where incident and output light have the helicities, $\sigma_{in}, \sigma_{out} \in \{-1,1\}$ in the circular polarization basis.

In a first approximation , the scattered electric field, $\boldsymbol{E}_{m,n}^{(1)}(\boldsymbol{r})$, from each antenna can be described as a dipole point source field, $\boldsymbol{E}_d(\boldsymbol{r})$, which has a *local* phase, $\psi_{m,n}$, due to each antenna's orientation, $\phi_{m,n}$ such that $\boldsymbol{E}_{m,n}^{(1)}(\boldsymbol{r}) = \boldsymbol{E}_d\{\boldsymbol{r} - \boldsymbol{\rho}_{m,n}\}e^{i\psi_{m,n}}$. To include higher order modes in the analysis, we consider a sum over N dipole contributions for each antenna mode to find the scattered electric field, $\boldsymbol{E}_{m,n}^{(N)}(\boldsymbol{r})$. Example of such decomposition are shown in figure 1d and 1e. In the far field, we find

$$\boldsymbol{E}_{m,n}^{(N)}(\boldsymbol{r}) = \boldsymbol{E}_{m,n}^{(1)}(\boldsymbol{r})\left[\sum_{l=1}^{N} e^{-i\{\boldsymbol{k}\cdot R(\phi_{m,n})\boldsymbol{s}_l + \sigma_{out}\theta_l\}}\right]$$

where $\boldsymbol{\rho}_{m,n}$ is both the position and centre of rotation of each antenna in the 2D array; $\boldsymbol{s}_l$ is the displacement of dipole l relative to $\boldsymbol{\rho}_{m,n}$ in the antenna basis (i.e. for $\phi_{m,n} = 0$); θ_l is the orientation of the l^{th} dipole in the antenna basis; and $R(\phi_{m,n})$ is the rotation matrix applied to $\boldsymbol{s}_l$ about the point $\boldsymbol{\rho}_{m,n}$. Now, the electric field of each

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nanoantenna not only depends on its whole orientation (i.e. PB phase), but also on the local phases acquired by each constituent dipole that describes the antenna's higher order mode.

To illustrate this experimentally, we have considered the Fourier plane imaging of our metasurfaces for SHG pumped at a wavelength of 1470 nm using LCP light ($\sigma_{in} = 1$). Figure 1f shows the diffraction orders of the metasurface B and it matches the prediction by the generalized law of diffraction. This indicates that SHG from each antenna must be dipole allowed. However, Figure 1g shows the diffraction orders of the metasurface A. We now observe two diffraction orders located at $n = \{-4,2\}$ for LCP SHG, and four diffraction orders located at $n = \{-4, -2, 2,4\}$ for RCP SHG and their location cannot be predicted by the law. Instead, we must include the higher second order dark mode of the bar, modelled here as two destructively interfering dipoles, to correctly predict the location of the diffraction orders.

To conclude, while symmetry arguments in nonlinear optics are a powerful predictive tool, a full modal description is required to describe nonlinear metasurfaces. By decomposing each antenna's nonlinear scattering into its constituent dipoles, we have found generalized diffraction rules to account for both structural rotation (i.e. PB phase) and higher order modes. In our main publication, [4] we have also considered the case of FWM in these metasurfaces to reveal the relevance of higher order modes in odd-order nonlinear processes. This study extends the interpretation of PB phase nonlinear metasurfaces to include complex composite antennas and explains the observation from apparently symmetry disallowed nonlinear processes.

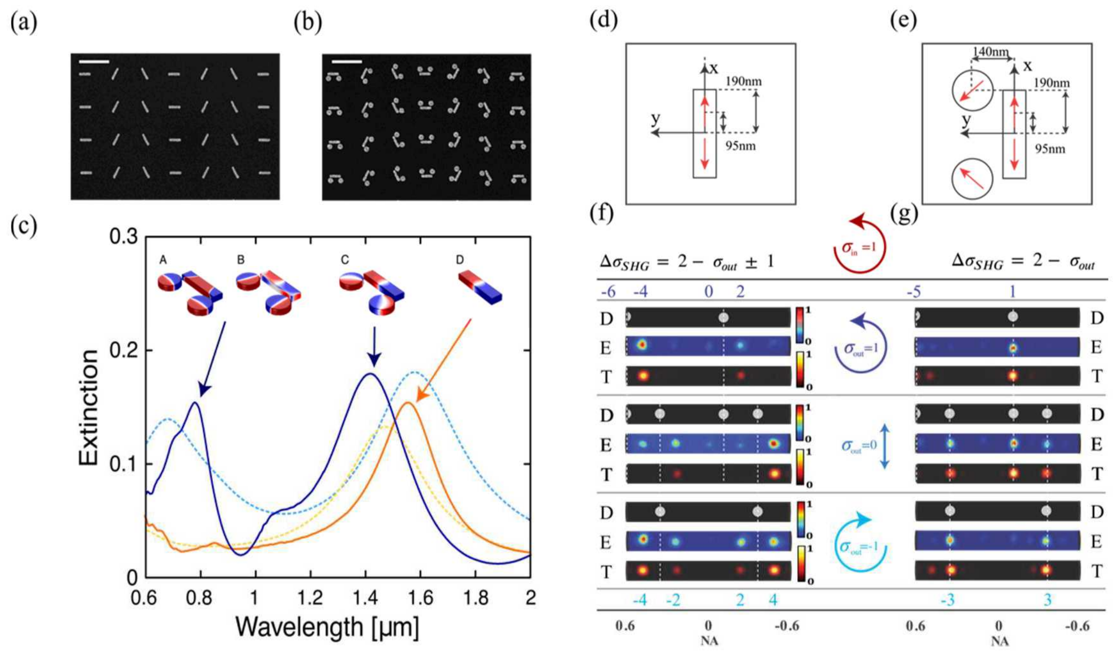

Fig 1. (a, b) Scanning electron microscopy (SEM) images of the two metasurfaces. Dimensions of bars are length = 370 nm, width = 75 nm, height = 40 nm. Disks have the same heights with diameters of 150 nm. Gap widths are 20 nm. The lattice period is 1 μm in both directions. Scale bar is 1 μm. (c) Measured (solid lines) and simulated (dashed colour lines) extinction spectra. Insets represents the charge distribution at 1500 and 750 nm for linear polarization parallel (B, C, D) and perpendicular (A) to the bar. (d, e) Normalized dipole decomposition of the antenna's mode at 735nm for (d) metasurface A, (e) metasurface B. (f, g) Diffraction patterns for SHG at 735nm under left circularly polarized light for (f) metasurface A, (g) metasurface B, predicted by : the generalized law of diffraction (D); the multipole scattering model (T); and measured (E). The location of the diffraction orders are numbered as n multiples of $\pi/3a$ for $-6 < n < 6$ with $n = -(6p - 2 + \sigma_{out} \pm 1)$ for quadrupole-like scattering and $n = -(6p - 2 + \sigma_{out})$ for dipole – like scattering with p an arbitrary integer.

3. References

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