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Local-field effects in the nonlinear optical response of metamaterials

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Abstract

We investigate optical second-harmonic generation from metamaterials that consist of two-dimensional arrays of lithographically designed T-shaped gold nanodimers, where the horizontal and vertical bars of the T are separated by a nanogap. The response is shown to depend in a nontrivial way on the gap size and to exhibit an unexpected dependence on the polarizations of the fundamental and second-harmonic light. The experimental results are explained by extensive calculations of the polarized local fields at the fundamental and second-harmonic frequencies. Due to the noncentrosymmetry requirement of second-order nonlinear processes, strong local fields in the gap region alone are not sufficient to drive the response efficiently. Instead, polarization properties of the local field and its asymmetric distribution around the dimer perimeters play the key role and enable efficient interaction with the surface nonlinearity of the metal. A small slant in the relative orientations of the vertical and horizontal bars of the dimer gives rise to second-harmonic signals that are forbidden for the ideal dimer with no slant. These signals are also explained by the local-field distributions, now modified by the slant.

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1. Introduction

Metamaterials are artificial materials that cannot be found in nature and that have been designed to exhibit prescribed electromagnetic properties. Whereas several different types of materials satisfy this basic definition and can be useful in a number of applications, the recent explosive interest in metamaterials has been motivated by the possibility of designing materials with a negative index of refraction [1–3]. Negative-index materials offer the intriguing possibility of imaging with resolution beyond the traditional diffraction limit [2,4].

A negative index of refraction can be achieved at a given wavelength if both the electric permittivity and magnetic permeability are negative at this wavelength [1,2]. The permittivity, permeability, and index of refraction are macroscopic quantities, obtained through averaging the responses of individual building units of the material over distances larger than the units

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themselves but much smaller than the wavelength of radiation. The key prerequisite for using such macroscopic or effective medium concepts is therefore that the material appears homogeneous over the scale of wavelength. In addition, the same quantities, which may possibly be anisotropic, should describe the material for any angle of incidence of radiation into the material.

Negative-index metamaterials often consist of periodic arrays of metal particles [3,5–7]. The metals naturally give rise to negative permittivity (real part), whereas negative permeability is achieved by designing the structure to have a magnetic resonance in the desired wavelength range. The structural features are limited by present nanofabrication techniques to linewidths of a few tens of nm and array periods of a few 100 nm. This is sufficient to achieve the effective medium limit for infrared and longer wavelengths [5]. However, as the operation range of metamaterials approaches visible and even shorter wavelengths [6,7], one must pay special attention to justify effective medium approaches. For example, diffraction in a material with wavelength-scale internal structure may give rise to phenomena reminiscent of those expected from a negative-index material [8]. This is not to say that such materials could still not have interesting and useful properties for a number of applications, only that their description in terms of effective quantities may not be appropriate [9].

From a somewhat different point of view, metal nanoparticles themselves have their own history of interesting properties with no reference to effective medium approaches. The optical properties of such particles are dominated by the plasmon resonances of conduction electrons [10]. The resonances depend on the size and shape of the particles, as well as their mutual ordering and dielectric environment. The resonances are associated with strong local fields in the vicinity of the particles, which can lead to enhanced optical interactions. The prime example of this is surface-enhanced Raman scattering (SERS), which has enabled the detection of single molecules [11]. Such local-field enhancement is also particularly interesting for nonlinear optical processes, which scale with a high power of the field.

Ordering of the particles into a regular array with a period on the order of the wavelength can significantly modify the character of plasmon resonances and the local-field distribution in the structure. For example, variations in the period or the angle of incidence can change a certain diffraction order from evanescent into propagating on either side (e.g., air/vacuum or substrate) of the nanostructure, leading to diffraction anomalies of Rayleigh type [12–14]. In addition, propagating surface modes [13,14], such as surface plasmon polaritons or resonant waveguide modes, can be excited even when the structure apparently has a sub-wavelength period in the sense that only zeroth diffraction orders can propagate in external media. These resonances can exhibit extreme sensitivity to the angle of incidence, and it is clear that the effective medium approaches cannot properly account for such features. Such metamaterials can nevertheless have extremely interesting properties, e.g., high specific rotation of polarization [15,16] supported by plasmonic or waveguide resonances [17]. The long-range coupling between the particles through the grating [12] can significantly influence the spectral properties of the sample [18,19] and the local fields [20] as well as the second-order nonlinear response [21,22].

Small features, such as sharp corners and nanoscale gaps, are thought to be particularly favorable for concentrating electromagnetic energy in metal structures [23–25]. For example, SERS can be enhanced by $\sim$12 orders of magnitude by rough metal surfaces [11]. Random and fractal structures have also been shown to lead to enhanced and highly localized nonlinear interactions such as second-harmonic generation (SHG) [26–28]. Recent progress in nanofabrication has also allowed the development of designer structures, such as nanoscale antennae [29–31], tips for near-field microscopy [32–34], and structures for coherent localization of electromagnetic energy [35,36].

For the optimization of nonlinear optical effects, the goal has usually been to achieve the highest possible local field strength within a nanostructure. However, the enhancements can exhibit a substantial dependence on the symmetry of the structure and on the field polarization. This is particularly true for nanodimers consisting of two nanoparticles separated by a nanogap. In such cases, only the polarization along the interparticle axis is strongly enhanced, whereas the orthogonal polarization yields significantly less enhancement [25,29,37]. Symmetry considerations are particularly important for second-order nonlinear effects that are forbidden in centrosymmetric materials. Two-dimensional designer nanostructures for field enhancement often appear centrosymmetric when investigated at normal incidence. Any second-order responses observed under such conditions [38] must therefore arise from symmetry breaking of the ideal structure because of small-scale defects, which may act as attractors of strong local fields. For example, we have shown that the second-harmonic response of metal nanoparticles is very sensitive to chiral symmetry breaking [39–41].

We have recently shown that second-harmonic generation from T-shaped, noncentrosymmetric nanodimers is