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An enhanced method is developed for analysis of third-order nonlinearities in optical nanostructures with a scalar magnetic field frequency-domain formulation; it is shown to produce fast and accurate results for 2D problems without a superfluous vector electric field formalism. While a standard TM representation using cubic nonlinear susceptibility results in an intractable implicit equation, our technique alleviates this problem. In addition to a universal approach, simpler, more efficient solutions are proposed for media having solely either a real (lossless Kerr-type medium) or an imaginary (nonlinear absorbing medium) nonlinearity. Combining these solutions with a finite-element method, we show simulation examples validated with alternative approaches. © 2009 Optical Society of America

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In the past few years, a number of unusual nonlinear (NL) wave interactions were predicted to occur in metamaterials. In a majority of theoretical studies to date, NL metamaterials were considered uniform media with a prescribed dielectric permittivity and magnetic permeability, and the nanostructured nature of these artificial materials was not taken into account. Local field enhancement and other effects are likely to alter NL interactions of electromagnetic waves with metamaterials. Therefore, the availability of efficient and reliable numerical modeling tools taking into account an actual nanostructure and near-field effects are essential for developing future applications of metamaterials.

Several advanced approaches have been demonstrated for solving the problem of strongly NL media [1–5], where the solution of the wave equation in an arbitrary layer of a lamellar structure is based on a method of single expression and where the more traditionally accepted superposition of counterpropagating waves is not exploited. The method of single expression uses integration starting from the shadow side, thereby reducing a complex multiboundary problem to a Cauchy problem, with no approximation in the boundary conditions for the electromagnetic field and with no constrains on the form of the wave equation solution that is required by more traditional, earlier methods [6–8].

Unfortunately, none of the above references [1–8] deal with the finite-element (FE) analysis of light propagation in subwavelength lamellar structures with nanoscale plasmonic inclusions. At the same time, the FE method is widely used for detailed linear and NL simulations of complex scattering geometries in the microwave and optical range [9]. The difficulties of linear FE modeling of dispersive optical metamaterials has been discussed, for example, in [10], but the caveats of NL FE modeling of 2D structures for TM waves utilizing a scalar magnetic field (H-field) wave equation have not been considered yet.

In this Letter, an efficient method is developed for introducing third-order nonlinearities in optical nanostructured materials, including photonic metamaterials. The method uses scalar H-field frequency-domain formulation; it is shown to produce fast and accurate results without a superfluous vector electric field (E-field) formalism. A standard TM representation using cubic NL susceptibility is problematic because of an intractable implicit equation. To alleviate this problem, simplified solutions are derived for a lossless Kerr-type medium and an NL absorbing medium. For a lossless Kerr-type medium a comprehensive simulation example is validated and discussed.

Consider the propagation of a monochromatic wave of frequency ω (time-dependent term $e^{-\iota\omega t}$ is omitted) in a medium with a third-order susceptibility $\chi^{(3)}$ (or coefficient n_2) given by

$$\varepsilon_r = \varepsilon_l + 3\chi^{(3)}|\vec{E}|^2 = \varepsilon_l(1 + 4n_2\eta^{-1}|\vec{E}|^2). \quad (1)$$

Here, ε_r is the relative permittivity, ε_l is the linear permittivity, $\eta=(\mu_0/\varepsilon_0)^{1/2}$ is free-space impedance, with ε_0 and μ_0 being the permittivity and permeability of vacuum, and $\vec{E}$ is the E-field strength vector.

In a frequency-domain formulation and a 2D geometry, the Maxwell equations reduce to a single scalar wave equation for the out-of-plane field component. For TE modes, where only a scalar E-field is employed, incorporating the nonlinearity, Eq. (1), is quite simple. However, for TM modes, relevant for resonant plasmonic structures, the wave equation for the out-of-plane H-field, $\vec{H}=\hat{\mathbf{z}}h(x,y;\omega)$, is $\nabla\cdot(\varepsilon_r^{-1}\nabla h)+k^2\mu_r h=0$, where μ_r is the relative permeability and $c=1/\sqrt{\mu_0\varepsilon_0}$ and $k=\omega/c$ are the speed of light and the

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wavenumber in vacuum, respectively. Since the E-field components depend on h through the curl equation

$$\vec{E} = \iota(\omega\varepsilon_0\varepsilon_r)^{-1}\nabla \boldsymbol{h} \times \hat{\mathbf{z}}, \tag{2}$$

combining Eqs. (1) and (2) and utilizing $f=3|\chi^{(3)}||\nabla h \times \hat{\mathbf{z}}|^2(\omega\varepsilon_0)^{-2}$, $\varphi=\arg\chi^{(3)}$, yields an implicit form,

$$\varepsilon_r = \varepsilon_l + fe^{\iota\varphi}/|\varepsilon_r|^2, \tag{3}$$

(or $\varepsilon_r|\varepsilon_r|^2-\varepsilon_l|\varepsilon_r|^2=fe^{\iota\varphi}$), which substantially restrains any straightforward numerical implementation of the third-order NL effects in a standard scalar frequency-domain simulation scheme.

We note that this problem does not appear in a 3D frequency-domain formulation, with a 2D geometry treated by using a vector E-field formulation [9], nor in a 3D time-domain formulation with a coupled system of partial differential equations for E and H fields. However, 3D methods exhibit significant redundant complexity compared with the scalar frequency-domain formulation of 2D geometries and are substantially less efficient for computationally expensive problems, especially with multiphysics content, where coupled NL phenomena are solved consistently. To circumvent this redundancy, we solve the resulting cubic equation (3) for ε_r in terms of the H-field and $\chi^{(3)}$.

Although a general solution of Eq. (3) can be readily obtained, it requires a detailed *ad hoc* analysis depending on the linear loss (or gain) and a specific type of NL effect, and it is not presented here. In this Letter, we discuss two major H-field formulations for the most frequent situations in optical metamaterials, when the linear absorption of a given nanostructured elemental NL material is negligible, $\mathrm{Im}(\varepsilon_l)=0$, and the elemental NL material exhibits either a purely real (lossless Kerr-type medium, $\varphi=0$) or purely imaginary (NL absorber, $\varphi=\pi/2$) third-order nonlinearity. These assumptions allow Eq. (3) to be solved with the Chebyshev (trigonometric) approach [11].

We define $\varepsilon_r=(1+u)v$, $\Delta=2+fv^{-3}$, and $v=\varepsilon_l/3$. Substituting into Eq. (4) yields a Chebyshev form, $u^3-3u=\Delta$ [2]. The real root for $\Delta\in[2,\infty)$ is $u=2\cosh(\frac{1}{3}\cosh^{-1}(\frac{1}{2}\Delta))$ so that

$$\varepsilon_r = v + 2v\cosh\left(\frac{1}{3}\cosh^{-1}\frac{\Delta}{2}\right) = (1+a+a^{-1})v, \tag{4}$$

where $a=[\frac{1}{2}(\Delta+\sqrt{\Delta^2-4})]^{1/3}$.

Separating the real part $\varepsilon_r'=\varepsilon_l$ and the imaginary part ε_r'' of the permittivity yields the depressed cubic formula $(\varepsilon_r'')^3+\varepsilon_l^2\varepsilon_r''=f$. After substituting $\varepsilon_r''=vu$, $\Delta=fv^{-3}$, and $v=\varepsilon_l/\sqrt{3}$, the latter equation arrives at another Chebyshev form, $u^3+3u=\Delta$, with the only relevant root being $u=2\sinh[\frac{1}{3}\sinh^{-1}(\frac{1}{2}\Delta)]$ [12]. Hence,

$$\varepsilon_r = \varepsilon_l + \iota 2v\sinh\left(\frac{1}{3}\sinh^{-1}\frac{\Delta}{2}\right) = \varepsilon_l + \iota(b-b^{-1})v. \tag{5}$$

Here $b=[\frac{1}{2}(\Delta+\sqrt{\Delta^2+4})]^{1/3}$.

The simulation results, performed by using a commercial FE solver (COMSOL Multiphysics), were validated in two stages. First, we considered a plane wave normally incident on a uniform NL film. The solution obtained with the 2D scalar H-field formulation, $\mathrm{d}(\varepsilon^{-1}\mathrm{d}h/\mathrm{d}x)/\mathrm{d}x+k^2\mu_0 h=0$, was found to be in complete agreement with the solution obtained for the 2D scalar E-field formulation, as well as with the reference solution from an alternative NL 1D solver [13]. Second, the solution for the 2D nanostructures described below was found to be in excellent agreement with simulations performed for the same 2D geometries by using the 3D vector E-field formulation with the nonlinearity defined directly as in Eq. (1).

We now use the scalar H-field formulation to examine the NL response of metamagnetics, well-studied magnetically resonant arrays of coupled nanostrip pairs [14]. This structure consists of a sandwich of two thin metallic strips separated by a thin dielectric spacer and can exhibit a negative magnetic response ranging from the mid-IR to the visible. The exact geometry and material parameters used in the current example are the same as sample D of [14] except that the strips are covered by a 160 nm layer (cladding) of a Kerr material ($\mathrm{Im}(n_2)=0$), and hence Eq. (4) was embedded in the FE simulations. The transmission and reflection scattering data of the structure are shown in Fig. 1(a). The structure exhibits a resonance of a magnetic nature at 698 nm.

Field maps show that, across the visible and near-IR range, the E-field between the strips is *locally* enhanced by up to $\rho\sim5-15$, where $\rho\equiv|E|/|E_{\mathrm{inc}}|$ is the near-field enhancement factor. In addition, the E-field at the boundary of the spacer is enhanced by a more modest *average* value of $\rho_2\equiv(\int\rho^2\mathrm{d}l/\int\mathrm{d}l)^{1/2}\sim2-4$; see Fig 1(c). Thus, since the wavelength of the magnetic resonance, λ_m, is sensitive to the value of the permittivity at the boundary of the spacer [15], the latter enhancement can be exploited in order to modify λ_m through the Kerr response of the cladding. Indeed, for a nonlinearity of $n_2I_{\mathrm{inc}}=0.0013$, we observe a shift of $\sim$6 nm in the location of the maximum (minimum) in the reflectance (transmittance) spectrum [Fig. 1(b)]. For this nonlinearity, the change in the index of refraction is $\Delta n\equiv\rho_2^2n_2I_{\mathrm{inc}}\cong0.006-0.02$. Interestingly, we note that the maximal *average* enhancement is about 50 nm

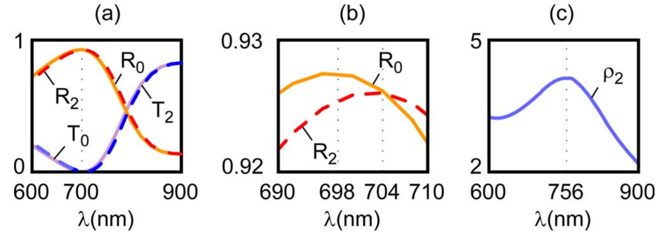

Fig. 1. (Color online) Simulation results for coupled nanostrip pairs covered with a $n_0=1.53$ Kerr material. (a) Transmission $\mathrm{T}_{0,2}$ and reflection $\mathrm{R}_{0,2}$ spectra for the linear (T_0, R_0, solid) and NL (T_2, R_2, dashed, $n_2I_{\mathrm{inc}}=0.0013$) cases. (b) Same as (a); enlargement of the resonance range. (c) Average enhancement ρ_2 over the spacer boundary.

away from the resonance, where maximal *local* enhancement of the H-field is attained.

For the same structure, a comparable shift was obtained experimentally by thermal tuning of the index of refraction of a liquid crystal cladding [15]. Tuning of the magnetic-resonance frequency of an analogous structure, split-ring arrays covered with a Kerr material, was studied in [16,17] by using a quasi-static analysis. In particular, it was shown that for nonlinearities as small as 10^{-4}–10^{-5}, the magnetic-resonance frequency may be tuned by a few percent and may even exhibit a bistable behavior. Our full-wave simulations thus provide a qualitative validation of the quasi-static analysis.

The above simple model using our new approach has also been compared with a corresponding model utilizing a vector E-field formulation with the same number of elements (934 FEs) and an identical mesh topology. As a result, the scalar H-field formulation requires only 8815 variables (degrees of freedom) with third-order (cubic) scalar elements or 24,031 degrees of freedom for fifth-order (quintic) scalar elements. The vector E-field formulation requires as many as 95,325 degrees of freedom with cubic vector elements [10]. Using the same nonparallelized solver for the above example, the scalar H-field formulation gives the solution approximately four times faster between the resonances (and about five times faster near the resonances) in comparison with the corresponding vector E-field models. Such computational efficacy is especially useful for evolution-driven optimization techniques [18] and coupled multiphysics problems. The nonlinearities used in our simulations are available either from fast electronic nonlinearities of organic materials [19] and semiconductors or alternatively, from slow reorientation (e.g., in liquid crystal) and thermal nonlinearities [20].

The universal approach built on the solution of the implicit equation for the nonlinear dielectric function is shown in [21], where all important features of the method are analyzed using as an example a more general cubic nonlinearity.

In this Letter, the basic case of lossless linear susceptibility in combination with either purely real or purely imaginary cubic susceptibility has been derived and tested.

In addition to the example shown in the Letter in [21] detailed simulation results are obtained for a nonlinear focusing device with optically controlled isotropic Kerr-type nonlinearity (embedding a NL material in the subwavelength slits of the thick silver film is proposed as a new method of all-optical control of the output beam). The device geometry and material parameters are adapted from [22]. The core principles, activation of the slit polaritonic modes and consequent tunable beam focusing, are obtained in [21] also using the proposed approach in a finite-element frequency-domain modeling environment. As in this Letter, the simulation results prove the predicted functioning of the device tested with our scalar H-field formulation.

In summary, we have proposed a versatile numerical approach for modeling third-order NL interactions in optical nanostructures. This method eliminates the need for time-consuming 3D full-wave simulations in many cases of practical interest such as TM wave interaction with plasmonic nanostructures.

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