Double-resonant optical materials with embedded metal nanostructures

Ildar R. Gabitov

Department of Mathematics, University of Arizona, 617 North Santa Rita Avenue, Tucson, Arizona 85721, and Theoretical Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545

Robert A. Indik

Department of Mathematics, University of Arizona, 617 North Santa Rita Avenue, Tucson, Arizona 85721

Natalia M. Litchinitser

Department of Electrical Engineering and Computer Science, University of Michigan, 1301 Beal Avenue, Ann Arbor, Michigan 48109

Andrei I. Maimistov

Department of Solid State Physics, Moscow Engineering Physics Institute, Kashirskoe sh. 31, Moscow, 115409 Russia

Vladimir M. Shalaev

School of Electrical and Computer Engineering, Purdue University, West Lafayette, Indiana 47907

Joshua E. Soneson

Program in Applied Mathematics, University of Arizona, 617 North Santa Rita Avenue, Tucson, Arizona 85721

Received November 3, 2005; accepted November 6, 2005; posted December 8, 2005 (Doc. ID 65763)

We derive equations modeling the resonant interaction of electric and magnetic components of light fields with metal nanostructures. This paired resonance was recently shown to produce negative refractive index. The model equations are a generalization of the well-known Maxwell–Lorentz model. We demonstrate that in the case of nonlinear polarization and linear magnetization, these equations are equivalent to a system of equations describing the resonant interaction of light with plasmonic oscillations in metal nanospheres. A family of solitary wave solutions is found that is similar to pulses associated with self-induced transparency in the framework of the Maxwell–Bloch model. The evolution of incident optical pulses is studied numerically, as are the collision dynamics of the solitary waves. These simulations reveal that the collision dynamics vary from near perfectly elastic to highly radiative, depending on the relative phase of the initial pulses.

OCIS codes: 190.4400, 260.5740, 250.5530.

1. INTRODUCTION

The resonant interaction of light with plasmonic oscillations in metal nanostructures has recently become a focus of considerable interest owing to the potential of harnessing strong, locally generated fields for a wide variety of applications.[1,2] In addition, it has been recently demonstrated that such structures produce a negative refractive index in the optical domain.[3] The negative refractive index is a consequence of simultaneous resonant electric and magnetic interactions with the nanostructures, which is reminiscent of arrays of LC circuits that demonstrate negative refractive index in the microwave regime.[4–6] The equations that describe the interaction of coherent light with a medium consisting of molecules (considered as harmonic oscillators) is known as the Maxwell–Lorentz model.[7] Taking into account the quantum nature of the atoms leads to the Maxwell–Bloch equations in the two-level approximation.[8] The Maxwell–Bloch equations are nonlinear and describe many important phenomena, such as self-induced transparency, optical pulse amplification, and superfluorescence.

In this study we extend the Maxwell–Lorentz system to account for plasmonic and magnetic resonances. These new resonances are due to interaction with plasmonic oscillations in the metal nanostructures. The interaction of the electric and plasmonic fields leads to strong electric resonance. In addition, nanostructures can mimic the effect of LC circuits to produce a resonant interaction with the magnetic field. Just as nonlinear effects in the Maxwell–Bloch equations (a single resonance) have been

a rich source of important physical phenomena, these simultaneous magnetic and electric resonances combined with nonlinear effects may well lead to new physics.

In what follows we first investigate the simplest nanostructures: nanospheres. In this case, there is no magnetic resonance, though there is nonlinearity in the polarization response to the electric field.[9,10] We derive equations describing this interaction that are of Maxwell–Duffing form. Then we consider the case of simultaneous magnetic and electric resonance, where the magnetic susceptibility is linear, but the electric polarization is nonlinear. We show that in the latter case, the equation for the magnetic field decouples, and the study of this system reduces to that of the Maxwell–Duffing equations we derive for nanospheres.

We find a family of solitary wave solutions whose behavior is analogous to self-induced transparency in Maxwell–Bloch. We numerically study the evolution of incident optical pulses as well as the collision dynamics of the solitary waves. These results are relevant to both the system of nanospheres and the more complex systems of nano-LC circuits.

2. SINGLE-RESONANT MEDIUM: NANOSPHERES

Quantum effects in metal nanoparticles driven by a resonant optical field play an important role in inducing a strong nonlinear response, as was recently shown.[9,10] Here we consider the nonlinear resonant interaction of ultrashort optical pulses with metal nanoparticles distributed uniformly in a host medium. We restrict to the case of composite materials for which the resonance frequencies of the host medium are well separated from those of the nanoparticles. Examples include silver or gold spherical or spheroidal nanoparticles embedded in SiO_2. In these cases, the plasmonic resonance frequencies are in the visible part of the spectrum, whereas the resonance of the host is in the ultraviolet. For spheroidal particles with large aspect ratios, the longitudinal plasmon oscillations are in the infrared so that the difference between the resonant frequencies of the host and the plasmon oscillations is even larger.

Light interaction with metal nanoparticles can be described by a system consisting of Maxwell's equations for the electric field and an oscillator equation describing the displacement of conduction electrons in the metal nanoparticles from equilibrium (plasmonic oscillations). The nanoparticles are much smaller than the optical carrier wavelength λ_0. This allows light scattering and spatial effects in the nanoparticles to be neglected. As shown by Rautian[9] and Drachev *et al.*,[10] who further developed the earlier work by Hache *et al.*,[11] the response of the conduction electrons in the metal nanoparticles to an external electric field induces a leading-order cubic nonlinearity. The interaction of the electric field with plasmonic oscillations in nanoparticles with resonance frequency ω_r in the presence of this cubic nonlinearity can be described by the forced Duffing equation

$$\tilde{\mathcal{Q}}_{TT} + \omega_r^2 \tilde{\mathcal{Q}} + \varkappa \tilde{\mathcal{Q}}^3 = (e/m)\tilde{\mathcal{E}}. \tag{1}$$

In this expression $\tilde{\mathcal{Q}}$ represents plasmon displacement from equilibrium, T is time; $\varkappa$ is the coefficient of nonlinearity; e and m are the electron charge and rest mass, respectively; and $\tilde{\mathcal{E}}$ is the electric field. The tilde is used to denote rapidly varying quantities. The nonlinear coefficient $\varkappa$ can be estimated by comparing off-resonance nonlinear response in Eq. (1) with the Drude nonlinearity for (nonresonant) conduction electrons in metal nanoparticles. That susceptibility is characterized by[9] $\chi^{(3)} \simeq Ne^4a^2/(m\hbar^2\omega_0^4)$. Here a and N are the radius of the nanoparticle and the conduction electron density of the metal, respectively, and ω_0 is the optical carrier frequency. This results in the estimate $\varkappa \simeq (ma\omega_0^2/\hbar)^2$.

We are interested in pulse dynamics that vary on a much slower scale than the plasmonic, host atom, and carrier wave oscillations and can be described using a slowly varying envelope approximation. In this approximation Eq. (1) becomes

$$i\mathcal{Q}_T + (\omega_r - \omega_0)\mathcal{Q} + (3\varkappa/2\omega_0)|\mathcal{Q}|^2\mathcal{Q} = -(e/2m\omega_0)\mathcal{E}, \tag{2}$$

where the slowly varying envelopes of the electric field and plasmonic oscillations are represented by $\mathcal{E}$ and $\mathcal{Q}$, respectively. Maxwell's equation couples to the material polarization induced by the plasmonic oscillations. The equation for the electric field envelope is

$$i\left(\mathcal{E}_Z + \frac{1}{v_g}\mathcal{E}_T\right) = -\frac{2\pi\omega_0 N_p e}{cn_0}\langle\mathcal{Q}\rangle - \frac{2\pi\omega_0 N_a|d|^2}{cn_0\hbar\Delta_a} \times\mathcal{E} - \frac{2\pi i\omega_0 N_a|d|^2}{cn_0\hbar\Delta_a^2}\mathcal{E}_T, \tag{3}$$

where Z is the propagation coordinate, v_g is group velocity, c is the speed of light, n_0 is the refractive index evaluated at the carrier frequency ω_0, and N_p is the product of the conduction electron density N and the metal-filling factor p (the fraction of the composite occupied by metal). N_a is the concentration of host atoms, d is the projection of the dipole matrix element in the direction of the electric field polarization, and $\Delta_a = \omega_a - \omega_0$ is detuning from the resonance frequency of host atoms. The last two terms in Eq. (3) represent corrections to the, refractive index and group index due to the off-resonance interaction with the host medium, which, for illustration, is considered as an ensemble of two-level atoms. This equation is derived from the Maxwell–Bloch equations in the nonresonant case by considering Δ_a as a large parameter and applying the adiabatic following approximation.[12] Additional resonances would produce similar terms. We consider the case in which optical pulse intensity and duration as well as composite material parameters are such that the characteristic length of resonant light interaction with plasmonic oscillations is much smaller than the characteristic lengths for both group velocity dispersion and nonlinearity induced by the host medium. Therefore the terms representing these effects are omitted from Eq. (3).

In a composite material, the sizes and shapes of metal nanoparticles vary owing to limited fabrication tolerances. It is known that the plasmon resonance in spherical metal nanoparticles depends weakly on size in the range between 10 and 50 nm,[13] so variations in size are not important. However, variations in the shape and orientation of the nanoparticles can significantly change plasmonic resonance frequencies. This results in a broad-

ening of the resonance line of the bulk composite. The angle brackets $\langle \mathcal{Q}(t,z,\omega)\rangle = \int_{-\infty}^{\infty} \mathcal{Q}(t,z,\omega) g(\omega) \mathrm{d}\omega$ denote averaging over the distribution $g(\omega)$ of the resonance frequencies (line shape). Defining

$$\mathcal{E} = -E\frac{2m\omega_0^3}{e}\sqrt{\frac{2}{3\varkappa}}\exp(ik_s Z), \quad \mathcal{Q} = Q\omega_0\sqrt{\frac{2}{3\varkappa}}\exp(ik_s Z), \tag{4}$$

where $k_s = 2\pi\omega_0 N_a |d|^2 / cn_0\hbar\Delta_a$, and introducing the copropagating coordinate system $z = (\omega_p^2/4cn_0\omega_0)Z$, $t = \omega_0(T - Z/u)$, u here is shifted group velocity defined as $u^{-1} = v_g^{-1} + 2\pi\omega_0 N_a |d|^2 / cn_0\hbar\Delta_a^2$, $\omega_p^2 = 4\pi N_p e^2/m$, $\omega = (\omega_r - \omega_0)/\omega_0$, Eqs. (2) and (3) can be reduced to the simpler form

$$iE_z = \langle Q\rangle, \quad iQ_t + \omega Q + |Q|^2 Q = E. \tag{5}$$

These equations represent a generalization of the classical Maxwell–Lorentz model. In the case of identical nanoparticles, the averaging in Eq. (5) can be reduced to a single dimensionless frequency $\bar{\omega}$ [i.e., detuning frequency distribution $g(\omega) = \delta(\omega - \bar{\omega})$].

3. DOUBLE-RESONANT MEDIUM: NANO-LC CIRCUITS

To illustrate the equations governing nonlinear wave dynamics in the presence of both electric and magnetic resonance, we consider the simplest case of nonlinear polarization response and linear magnetic susceptibility. Maxwell's equations in the general case have the form

$$\nabla \times \mathbf{E} = -c^{-1}\mathbf{B}_t, \quad \nabla \times \mathbf{H} = -c^{-1}\mathbf{D}_t,$$

$$\mathbf{B} = \mathbf{H} + 4\pi\mathbf{M}, \quad \mathbf{D} = \mathbf{E} + 4\pi\mathbf{P}. \tag{6}$$

In real experiments electric and magnetic fields should be properly aligned with respect to the nanostructure to maximize the resonance interaction. As an example, electric and magnetic fields of the form $\mathbf{E} = (E,0,0)$, $\mathbf{H} = (0,H,0)$ are considered. In the linear case, the Fourier transformation may be applied to obtain

$$\hat{E}_z = i\omega c^{-1}\hat{\mu}(\omega)\hat{H}(\omega), \quad \hat{H}_z = i\omega c^{-1}\hat{\epsilon}(\omega)\hat{E}(\omega),$$

thus

$$\hat{E}_{zz} = i(\omega/c)^2\hat{\mu}(\omega)\hat{\epsilon}(\omega)\hat{E}(\omega). \tag{7}$$

Assume that polarization is defined by the plasma oscillation electron density so that the equation for polarization has the form

$$P_{tt} = \frac{\omega_p^2}{4\pi}E - \kappa P^3, \tag{8}$$

where ω_p is effective plasma frequency and κ is the nonlinearity coefficient. The origin of this nonlinearity could be quantum effects in the nanostructures or a nonlinear dielectric coating. For a description of the magnetization we use the simplest model of magnetic resonance as an illustration:

$$M_{tt} + \omega_T^2 M = -\frac{\beta}{4\pi}H_{tt}, \tag{9}$$

where ω_T is the Thomson frequency and β is determined by the geometry of the nanostructures. In the linear case we obtain

$$\hat{\epsilon}(\omega)\hat{\mu}(\omega) = \frac{(\omega^2 - \omega_p^2)(\omega_T^2 - \omega^2 + \beta\omega^2)}{\omega^2(\omega_T^2 - \omega^2)}, \tag{10}$$

and the linear dispersion relation for wave propagation is given by

$$k^2 = \left(\frac{\omega}{c}\right)^2 \hat{\epsilon}(\omega)\hat{\mu}(\omega). \tag{11}$$

Therefore the spectrum of linear waves contains a gap. The high-frequency branch corresponds to plasmons, and the low-frequency branch corresponds to waves in a left-handed material (LHM),[14] i.e., if the phase velocity is positive, then the group velocity is negative. Near $\omega = \omega_p$, $\omega > \omega_p$ we have the plasmon dispersion relation

$$\omega^2(k) = \omega_p^2 + \frac{\omega_p^2 - \omega_T^2}{(1-\beta)\omega_p^2 - \omega_T^2}c^2k^2. \tag{12}$$

For this branch of the dispersion relation both the phase velocity and group velocity are positive. Near $\omega = \omega_0 = \omega_T/\sqrt{1-\beta}$, $\omega < \omega_0$, we have

$$\omega^2(k) = \omega_0^2 - \frac{\beta\omega_T^2}{(1-\beta)^2(\omega_p^2 - \omega_0^2)}c^2k^2. \tag{13}$$

For this branch of the dispersion relation the phase velocity is positive; however, the group velocity is negative one. Thus in this spectral region waves in a linear approximation are propagated as waves in a LHM. Now that we have seen the linear LHM scenario we return to the nonlinear case.

Let us consider a plane electromagnetic wave propagating in the z direction. For an isotropic medium the Maxwell equations have the following scalar form:

$$E_z + c^{-1}H_t = -4\pi c^{-1}M_t, \quad E_x = 0,$$

$$H_z + c^{-1}E_t = -4\pi c^{-1}P_t, \quad H_y = 0. \tag{14}$$

This system is closed by two additional equations describing the interaction of the electric and magnetic fields with the metal nanostructures. To account for dimensional quantization due to confinement of the plasma in the nanostructures the additional term $\omega_D^2 P$ is included in Eq. (8):

$$P_{tt} + \omega_D^2 P + \kappa P^3 = \frac{\omega_p^2}{4\pi}E,$$

$$M_{tt} + \omega_T^2 M = -\frac{\beta}{4\pi}H_{tt}. \tag{15}$$

Normalization of the variables

$$\tau = t/T, \quad \xi = z/L, \quad q = P/P_0, \quad m = M/M_0,$$
$$e = E/E_0, \quad h = H/H_0,$$

$$T = \omega_p^{-1}, \quad L = c\omega_p^{-1}, \quad P_0 = \omega_p/\sqrt{|\kappa|},$$
$$\epsilon = \kappa/|\kappa|, M_0 = P_0 = E_0/4\pi$$

transforms Eqs. (14) and (15) to the following dimensionless nonlinear plasmon-LC circuit model:

$$\epsilon_\zeta + h_r = -m_r, \quad h_\zeta + e_r = -q_\tau,$$

$$q_{\tau\tau} + \gamma^2 q + \epsilon q^3 = e, \quad m_{\tau\tau} + \mu_T^2 m = -\beta h_{\tau\tau}, \tag{16}$$

where $\gamma^2=(\omega_D^2/\omega_p^2)$ and $\mu_T^2=(\omega_T^2/\omega_p^2)$.

To find a slowly varying envelope approximation of Eqs. (16) the Fourier transform is applied, resulting in

$$\bar{e}_\zeta - i\omega\bar{h} = i\omega\bar{m}, \quad \bar{h}_\zeta - i\omega\bar{e} = i\omega\bar{q},$$

$$(\gamma^2 - \omega^2)\bar{q} + \epsilon F[q^3](\omega) = \bar{e}, \quad (\mu_T^2 - \omega^2)\bar{m} = -\beta\omega^2\bar{h}, \tag{17}$$

where $F[f]$ denotes the time Fourier transform of the function f.

Magnetic susceptibility can be excluded from Eqs. (17) by introducing the magnetic permeability

$$\hat{\mu}(\omega) = 1 + \frac{\beta\omega^2}{\mu_T^2 - \omega^2}. \tag{18}$$

After substituting Eq. (18), Eqs. (17) reads

$$\bar{e}_{\zeta\zeta} + \omega^2\hat{\mu}(\omega)\bar{e} = -\omega^2\hat{\mu}(\omega)\bar{q},$$

$$(\gamma^2 - \omega^2)\bar{q} + \epsilon F[q^3](\omega) = \bar{e}. \tag{19}$$

Equations (19) describe wave propagation in a medium whose nonlinear properties are determined only by the polarization. However, the dispersion relation also takes the magnetic properties of the medium into account. Further, we consider fields in the Eqs. (19) as quasimonochromatic waves:

$$q(\zeta,\tau) = \tilde{q}(\zeta,\tau)\exp(-i\omega_0\tau + ik_0\zeta) + \text{c.c.},$$

$$e(\zeta,\tau) = \tilde{e}(\zeta,\tau)\exp(-i\omega_0\tau + ik_0\zeta) + \text{c.c.} \tag{20}$$

It can be shown that the Fourier component $A(k,\omega) = \int_{-\infty}^{\infty} A(x,t)\exp(i\omega t - ikx)\mathrm{d}t\mathrm{d}x$ of any quasimonochromatic wave $A(x,t)=a(x,t)\exp(-i\omega_0 t + ik_0 x)$ can be expressed via Fourier amplitude of the envelope $a(k,\omega) = \int_{-\infty}^{\infty} a(x,t)\exp(i\omega t - ikx)\mathrm{d}t\mathrm{d}x$ in the form of $A(k,\omega)=a(k - k_0, \omega - \omega_0)$. If the envelope is slowly varying in time and space compared with the carrier wave then its Fourier amplitude is localized in the domain of small wave numbers and frequencies $(k-k_0/k_0 \ll 1, \omega-\omega_0/\omega_0 \ll 1)$. Therefore the system of equations describing the evolution of the electric and polarization fields together with the magnetic field can be presented as

$$[(k_0+k)^2 - (\omega_0+\omega)^2\hat{\mu}(\omega_0+\omega)]\tilde{e}(k,\omega) = (\omega_0+\omega)^2\hat{\mu}(\omega_0+\omega)\tilde{q}(k,\omega),$$

$$[(k_0+k)^2 - (\omega_0+\omega)^2\hat{\mu}(\omega_0+\omega)]\tilde{h}(k,\omega) = (\omega_0+\omega)(k_0+k)\tilde{q}(k,\omega),$$

$$[\gamma^2 - (\omega_0+\omega)^2]\tilde{q}(k,\omega) + \epsilon F[q^3](k,\omega) = \tilde{e}(k,\omega). \tag{21}$$

Prior to proceeding further it is useful to consider the linear case, in which the medium polarization is described by

$$(\gamma^2 - \omega^2)\bar{q}(k,\omega) = \bar{e}(k,\omega).$$

The equations for $\bar{e}$ and $\bar{h}$ in the linear case have the following form:

$$[k^2 - \omega^2\hat{\mu}(\omega)]\bar{e}(k,\omega) = \omega^2\hat{\mu}(\omega)(\gamma^2 - \omega^2)^{-1}\bar{e}(k,\omega),$$

$$[k^2 - \omega^2\hat{\mu}(\omega)]\bar{h}(k,\omega) = \omega k(\gamma^2 - \omega^2)^{-1}\bar{e}(k,\omega).$$

Defining the dielectric permittivity as $\hat{\epsilon}(\omega)=[1+1/(\gamma^2 - \omega^2)]$ we rewrite the equation for $\bar{e}$ as

$$[k^2 - \omega^2\hat{\epsilon}(\omega)\hat{\mu}(\omega)]\bar{e}(k,\omega) = 0,$$

which gives the standard dispersion relation $k^2 = \omega^2\hat{\epsilon}(\omega)\hat{\mu}(\omega)$.

It should be noted that the variable q corresponds to medium polarization in Eqs. (16) and in the equations that follow. Let the variable $\bar{q}(k,\omega)$ describe the contribution of resonance structures–nonlinear oscillators and introduce the host medium dielectric permittivity $\hat{\epsilon}_{\text{host}}(\omega)$ to take dispersion properties of the host medium into account. Hence Eqs. (21) should be modified as follows:

$$[(k_0+k)^2 - (\omega_0+\omega)^2\hat{\epsilon}_{\text{host}}(\omega_0+\omega)\hat{\mu}(\omega_0+\omega)]\tilde{e}(k,\omega) = (\omega_0+\omega)^2\hat{\mu}(\omega_0+\omega)\tilde{q}(k,\omega),$$

$$[(k_0+k)^2 - (\omega_0+\omega)^2\hat{\epsilon}_{\text{host}}(\omega_0+\omega)\hat{\mu}(\omega_0+\omega)]\tilde{h}(k,\omega) = (\omega_0+\omega)(k_0+k)\tilde{q}(k,\omega),$$

$$[\gamma^2 - (\omega_0+\omega)^2]\tilde{q}(k,\omega) + \epsilon F[q^3](k,\omega) = \tilde{e}(k,\omega). \tag{22}$$

Taking into account that the arguments of the functions $\tilde{q}(k,\omega)$, $\tilde{e}(k,\omega)$, and $\tilde{h}(k,\omega)$ are much smaller than ω_0 and k_0, we consider only a second-order approximation with respect to ω/ω_0. Let us introduce

$$k^2(\omega) = (\omega_0+\omega)^2\hat{\epsilon}_{\text{host}}(\omega_0+\omega)\mu(\omega_0+\omega). \tag{23}$$

The expression in brackets on the left-hand side of the first two of Eqs. (22) can be replaced by

$$2k_0\left[k - \left(\frac{\partial k}{\partial\omega}\right)_{\omega=\omega_0}\omega - \frac{1}{2}\left(\frac{\partial^2 k}{\partial\omega^2} + \frac{1}{2k_0}\frac{\partial k}{\partial\omega}\frac{\partial k}{\partial\omega}\right)_{\omega=\omega_0}\omega^2\right] = 2k_0\left[k - \frac{1}{v_{g0}}\omega - \frac{1}{2}D_0\omega^2\right].$$

Here the group velocity v_{g0} and second-order dispersion

D_0 are introduced in a standard way. The equations for the electric and magnetic fields are then

$$2k_0\left[k-\frac{1}{v_{g0}}\omega-\frac{1}{2}D_0\omega^2\right]\tilde{e}(k,\omega)=\omega_0^2\hat{\mu}(\omega_0)\tilde{q}(k,\omega),$$

$$2k_0\left[k-\frac{1}{v_{g0}}\omega-\frac{1}{2}D_0\omega^2\right]\tilde{h}(k,\omega)=\omega_0 k_0\tilde{q}(k,\omega).$$

The equation for the magnetic field is unnecessary owing to the fact that in the approximation being considered the magnetic and electric fields are connected through the relation $k_0\tilde{e}(k,\omega)=\omega_0 k_0\tilde{h}(k,\omega)$.

In the spatiotemporal system of coordinates the equation for the slowly varying electric field envelope reads

$$i\left[\frac{\partial}{\partial\zeta}+\frac{1}{v_{g0}}\frac{\partial}{\partial\tau}-\frac{i}{2}D_0\frac{\partial^2}{\partial\tau^2}\right]\tilde{e}(\zeta,\tau)=-\frac{\omega_0^2}{2k_0}\hat{\mu}(\omega_0)\tilde{q}(\zeta,\tau). \tag{24}$$

The envelope approximation for $q(\zeta,\tau)$ reads

$$i\frac{\partial\tilde{q}}{\partial\tau}+(\gamma-\omega_0)\tilde{q}-3(\epsilon/\omega_0)|\tilde{q}|^2\tilde{q}=-\tilde{e}/\omega_0. \tag{25}$$

If we consider the strongest resonance effects and neglect effects due to chromatic dispersion, Eqs. (24) and (25) are similar to the Maxwell–Duffing equations derived for the resonance interaction of an optical field with metal nanoparticles. If we set $\hat{\mu}(\omega_0)=1$ we obtain the Maxwell–Duffing model considered above.

The properties of the dielectric permittivity in the model result from the equation for the polarization evolution. The magnetic permeability is explicitly given as $\hat{\mu}(\omega_0)$. The refractive index is a function of both properties for a steady-state situation. In this case, the setting is more general. We have a dynamical system that cannot be described in terms of refractive index.

In the case where both the polarization and magnetization are nonlinear, the equations for electric and magnetic fields are coupled. The system-modeling double resonance in this case consists of four equations. The derivation of these equations and analysis of their solutions will be published elsewhere.

4. SOLUTIONS OF THE MAXWELL–DUFFING SYSTEM

The Maxwell–Duffing system [Eqs. (5)] has solitary wave solutions if all oscillators are identical:

$$E(t,z)=\frac{v^{3/4}\exp[i\varphi+i\Omega t-iK\xi-i\chi(\xi)]}{\xi_0[\cosh(\xi/\xi_0)+K]^{1/2}},$$

$$Q(t,z)=E(t,z)\frac{\exp[-2i\chi(\xi)]}{\sqrt{v}}, \tag{26}$$

where $\xi=[z-v(t-\tau)]/\sqrt{v}$, $\chi(\xi)=\arctan[\Gamma\tanh(\xi/2\xi_0)]$, $\xi_0=1/2(1-K^2)^{1/2}$, $\Gamma=[(1-K)/(1+K)]^{1/2}$, and $K=(\bar{\omega}-\Omega)/2\sqrt{v}$. These solutions are parameterized by velocity v, frequency Ω, phase shift φ, and position τ. The velocity v is the amount by which the wave is slowed from the copropagating frame velocity u, so in the laboratory frame, the actual pulse velocity is $u-v$. The quantity ξ_0 must be real, hence $1-K^2>0$. Thus the condition for existence of these solutions is $|\bar{\omega}-\Omega|<2\sqrt{v}$. This choice of parameters provides relatively simple mathematical expressions for the solitary waves. In practice it is easier to both control and measure peak amplitude, $A=2v^{3/4}(1-K)^{1/2}$, than the pulse velocity, therefore A, Ω, φ, and τ form a more suitable set of parameters. Given the pulse amplitude A, the corresponding velocity parameter depends on the value of the quantity $\bar{\omega}-\Omega$. If $\bar{\omega}=\Omega$, then $v=(A/2)^{4/3}$ trivially. For the case when $\bar{\omega}\neq\Omega$, we can write the amplitude as $A=2v^{3/4}(1-\sigma|\bar{\omega}-\Omega|/2\sqrt{v})^{1/2}$, where the parameter $\sigma=\mathrm{sgn}(\bar{\omega}-\Omega)$. Then defining $\bar{v}=(2\sqrt{v}/|\bar{\omega}-\Omega|-\sigma)^{1/2}$ and $\bar{A}=\sqrt{27/2}|\bar{\omega}-\Omega|^{-3/2}A$ leads to an expression for the rescaled velocity $\bar{v}=(y^{-1/3}-\sigma y^{1/3})/\sqrt{3}$, where $y=\sigma[(\bar{A}^2+\sigma)^{1/2}-\bar{A}]^{1/2}$. In this calculation, the appropriate branches have been chosen so that the expressions are consistent with reality and positivity conditions on the parameters. Figures 1(a) and 1(b) show the amplitude and argument of E, respectively, for three different values of velocity: $v=0.25$ (dotted curve), $v=1$ (dashed curve), and $v=2$ (solid curve).

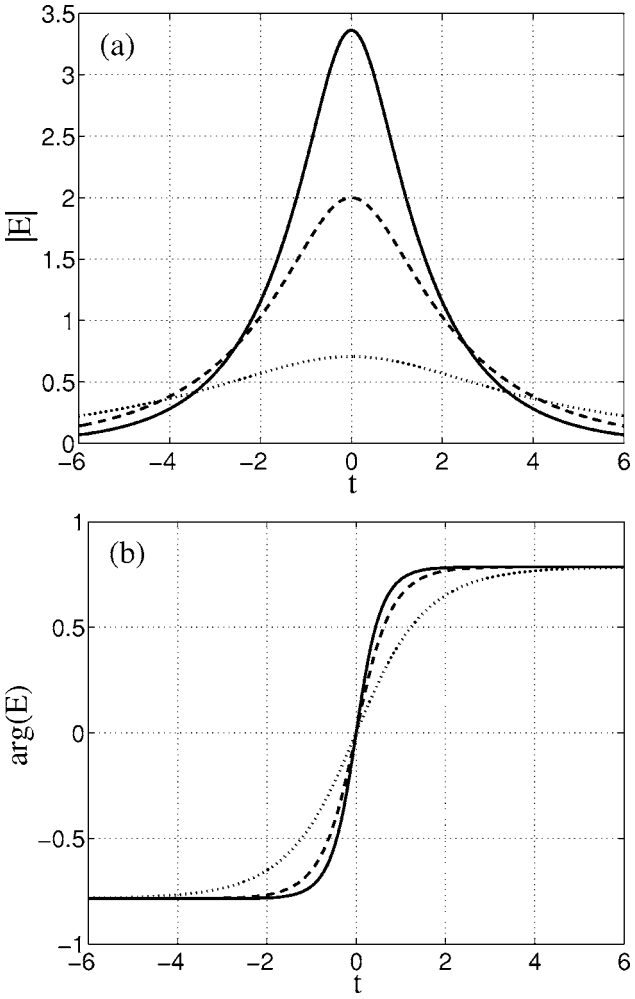

Fig. 1. (a) Electric field amplitude and (b) argument of solitary waves of the Maxwell–Duffing model, plotted for three values of velocity: $v=0.25$ (dotted curve), $v=1$ (dashed curve), and $v=2$ (solid curve).

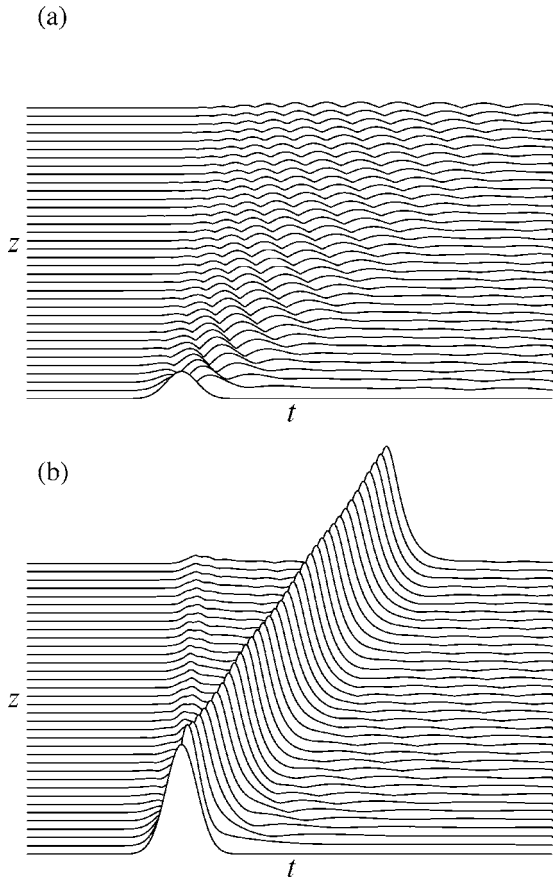

Fig. 2. Evolution of electric field amplitude with initial conditions (a) $\exp(-t^2/2)/2$ and (b) $2\exp(-t^2/2)$.

In optics it has become standard practice to refer to certain solutions of nonintegrable systems as solitons. These solutions are characterized as solitary waves that are robust to external perturbations, including collisions with other solitary waves. In addition, arbitrary initial data for these "soliton" supporting systems tend to evolve into a sum of solitary waves and continuous radiation. The remainder of this paper details our numerical investigation of these properties, in which Eqs. (5) are integrated in the case of delta-distributed resonance frequencies and zero detuning.

Numerical simulations of the evolution of Gaussian initial data are presented in Figs. 2(a) and 2(b). In Fig. 2(a) the initial condition $E(t,0)=\exp(-t^2/2)/2$ simply evolves into continuous radiation, whereas in Fig. 2(b) the initial condition $E(t,0)=2\exp(-t^2/2)$ emits some radiation but also achieves energy confinement and persists as a soliton. This behavior is similar to the self-induced transparency exhibited by the Maxwell–Bloch equations,[15] which describe optical pulse interaction with resonant two-level media. As the amplitude of the initial pulse is increased, the pulse splits into two [illustrated in Fig. 3(a) for the initial condition $E(t,0)=5\exp(-t^2/2)$] or more solitons and emits continuous radiation. Figure 3(b) shows the amplitude(s) of these solitons as a function of input pulse amplitude. As the input pulse amplitude increases, the output soliton amplitude also increases until a bifurcation occurs and a new soliton emerges. An increase in the input pulse amplitude results in the further production of more solitons, along with continuous radiation. Figure 4 shows a comparison between the analytic form of the solitary waves of Eq. (26) for E and numerical simulations. The velocity and frequency of the solitary waves are obtained from measurements of their amplitudes and half widths in our simulation. The plot shows that the numerical solution differs from the analytic forms only inasmuch

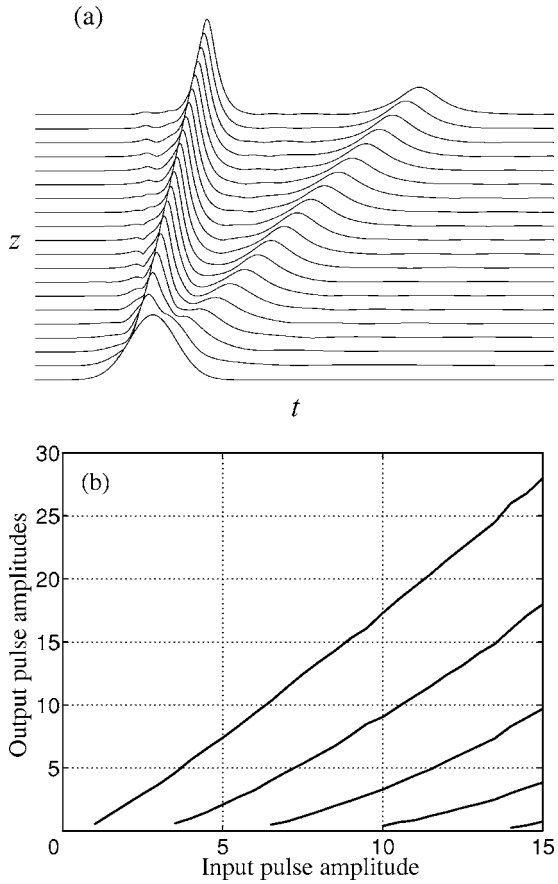

Fig. 3. (a) Evolution of electric field amplitude with initial condition $5\exp(-t^2/2)$, (b) output solitary wave amplitude(s) as a function of Gaussian input pulse amplitude A_0, where the initial condition is given by $A_0\exp(-t^2/2)$.

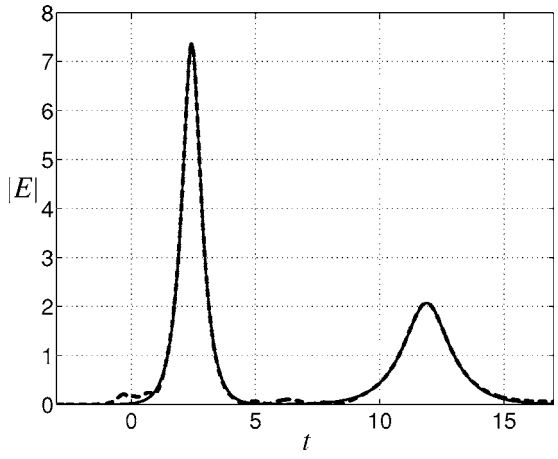

Fig. 4. Comparison of the analytical form of the solitary wave solutions (solid curve) with the results of numerical simulations (dashed curve) under conditions identical to those of Fig. 3(a).

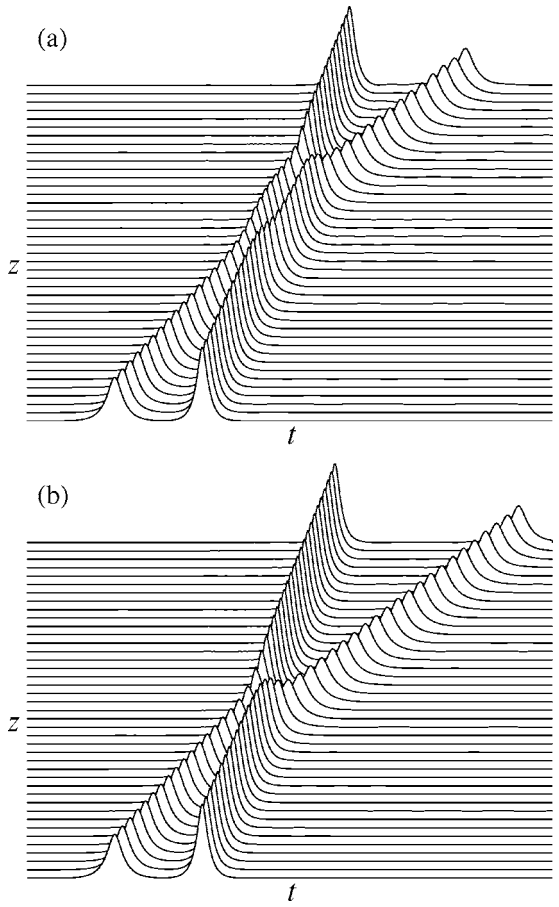

Fig. 5. Electric field amplitude showing collision dynamics of solitons for different values of relative phase; (a) $\Delta\phi=0$, (b) $\Delta\phi=\pi$.

as excessive radiation is emitted in the numerical simulation. The agreement of the analytic forms with the simulation results indicates that the system self-selects the solitary waves presented in Eqs. (26).

Two simulations illustrating collision dynamics are presented in Figs. 5(a) and 5(b), where the sum of two well-separated solitary waves is used as the initial condition. Figure 5(a) illustrates an in-phase collision, in which the relative phase $\Delta\varphi=\varphi_1-\varphi_2=0$, where the subscripts identify the soliton. The other initial soliton parameters are $v_1=1$, $\Omega_1=0$, $\tau_1=-10$, $v_2=2$, $\Omega_2=0$, and $\tau_2=0$. In Fig. 5(b) an out-of-phase $\Delta\varphi=\pi$ collision is illustrated. The same parameters are used, except for a shift in the relative phase. This results in a much faster collision. In both simulations the solitary waves persist after the interaction, although their characteristic parameters undergo shifts and radiation is emitted during the collision. A numerical study indicates that the collisions are quasielastic for values in the approximate interval $\Delta\varphi\in(\pi,2\pi)$. For some $\Delta\varphi$ values away from this interval, simulations show that one of the solitons is completely destroyed, whereas the other persists. A detailed analysis of the dependence on initial parameters will be presented elsewhere.

The soliton phenomena described above occur at light intensities such that the dimensionless field amplitude E is at least of order one. The intensity at which E is order one is estimated as $I\simeq(c/\varkappa)(m\omega_0^3/e)^2\simeq c(\hbar\omega_0/ea)^2$. For the particle radius $a=20$ nm and the carrier wavelength $\lambda_0=500$ nm, this results in light intensity of $I\sim 10$ GW/cm^2, which can be easily obtained with ultrashort laser pulses. The optical pulse durations for which this model is valid are limited by the condition $\Delta\omega\ll\omega_0$ (the spectral width of the pulse must be much smaller than the carrier frequency), required by the slowly varying envelope approximation. The pulse duration should also be much shorter than the characteristic plasmonic oscillation damping time, which is determined by the time required for electron thermalization in the metal nanoparticles ($\sim$400 fs[16,17]). The envelope approximation is appropriate for pulses with width $\tau\gtrsim 20$ fs.

The Maxwell–Duffing system is relevant for both the nanospheres and the more complex system with magnetic resonance. However, in the latter case, the sign of the magnetic permeability can be controlled by tuning the carrier frequency and shifting from the left-handed to the right-handed propagation regime.

5. CONCLUSION

In summary, equations are derived describing the interaction of electromagnetic fields with metal nanostructures with simultaneous resonances for the electric and magnetic medium response. In the case where the electric medium response includes nonlinearity, while the magnetic response is linear, this leads to the same Maxwell–Duffing system we derive for the case of metal nanospheres. A family of solitary wave solutions is presented in an envelope approximation for the Maxwell wave and Duffing oscillator equations, showing that energy confinement is possible for resonant optical pulse interaction with plasmonic oscillations in that system. The existence condition for these solutions is presented. Numerical simulations show that stable solitary waves evolve from Gaussian initial pulses with sufficient amplitudes and exhibit behavior analogous to self-induced transparency in Maxwell–Bloch. Simulations also reveal that the collision dynamics are highly dependent on initial soliton parameters, behaving quasi elastically in some regimes but having radically different behavior in others.

The authors are grateful to V. P. Drachev for helpful discussions. In addition, we would like to acknowledge funding under National Science Foundation grant DMS-0509589, Department of Energy/Advanced Scientific Computing Research Applied Mathematical Research Program at Los Alamos National Laboratory, State of Arizona Proposition 301.

REFERENCES

1. J. B. Pendry, "Negative refraction," Contemp. Phys. **45**, 191–202 (2004).
2. G. V. Eleftheriades and K. G. Balmain, *Negative-Refraction Metamaterials: Fundamental Principles and Applications* (Wiley, 2005).
3. V. M. Shalaev, W. Cai, U. Chettiar, H.-K. Yuan, A. K. Sarychev, V. P. Drachev, and A. V. Kildishev, "Negative index of refraction in optical metamaterials," Opt. Lett. **30**, 3356–3358 (2005).

4. R. A. Shelby, D. R. Smith, and S. Schultz, "Experimental verification of a negative index of refraction," Science **292**, 77–79 (2001).
5. A. K. Iyer, P. C. Kremer, and G. V. Eleftheriades, "Experimental and theoretical verification of focusing in a large, periodically loaded transmission line negative refractive index metamaterial," Opt. Express **11**, 696–708 (2003).
6. K. Aydin, K. Guven, M. Kafesaki, L. Zhang, C. M. Soukoulis, and E. Ozbay, "Experimental observation of true left-handed transmission peaks in metamaterials," Opt. Lett. **29**, 2623–2625 (2004).
7. H. A. Lorentz, *The Theory of Electrons* (Dover, 1952).
8. L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Dover, 1987).
9. S. G. Rautian, "Nonlinear saturation spectroscopy of the degenerate electron gas in spherical metallic particles," JETP **85**, 451–461 (1997).
10. V. P. Drachev, A. K. Buin, H. Nakotte, and V. M. Shalaev, "Size dependent χ^3 for conduction electrons in Ag nanoparticles," Nano Lett. **4**, 1535–1539 (2004).
11. F. Hache, D. Ricard, and C. Flytzanis, "Optical nonlinearities of small metal particles: surface-mediated resonance and quantum size effects," J. Opt. Soc. Am. B **3**, 1647–1655 (1986).
12. A. M. Basharov, and A. I. Maimistov, "Propagation of ultrashort electromagnetic pulses in a Kerr medium with impurity atoms under quasi-resonance conditions," Quantum Electron. **30**, 1014–1018 (2000).
13. V. M. Shalaev, ed., *Optical Properties of Random Nanostructures*, Vol. 82 of Springer-Verlag Topics in Applied Physics (Springer-Verlag, 2002).
14. V. G. Veselago, "The electrodynamics of substances with simultaneously negative values of ϵ and μ," Sov. Phys. Usp. **10**, 509–514 (1968).
15. S. L. McCall and E. L. Hahn, "Self-induced transparency by pulsed coherent light," Phys. Rev. Lett. **18**, 908–911 (1967).
16. V. Halt, J. Guille, J.-C. Merle, I. Perakis, and J.-Y. Bigot, "Electron dynamics in silver nanoparticles: comparison between thin films and glass embedded nanoparticles," Phys. Rev. B **60**, 11738–11746 (1999).
17. C. Voisin, D. Christofilos, N. Del Fatti, F. Valle, B. Prvel, E. Cottancin, J. Lerm, M. Pellarin, and M. Broyer, "Size-dependent electron–electron interactions in metal nanoparticles," Phys. Rev. Lett. **85**, 2200–2203 (2000).