

Laser gain media based on nanocomposite materials

Ksenia Dolgaleva* and Robert W. Boyd

The Institute of Optics, University of Rochester, Rochester, New York 14627, USA

**Corresponding author: ksenia@optics.rochester.edu*

Received February 6, 2007; accepted February 23, 2007;
posted March 8, 2007 (Doc. ID 79758); published July 19, 2007

A new way of controlling the laser properties of optical materials by designing composite materials that exploit local-field effects is proposed. It is shown that the basic laser properties, such as the radiative lifetime of the upper laser level, small-signal gain coefficient, and saturation intensity can be controlled independently by means of local-field effects. These ideas could be used to design laser systems with significantly improved properties. © 2007 Optical Society of America

OCIS codes: 160.3380, 160.4670, 160.4760.

1. INTRODUCTION

Nanocomposite optical materials are nanoscale mixtures of two or more homogeneous constituents in which the individual particles are much smaller than the optical wavelength but still large enough to have their own dielectric identities. The optical properties of composite materials can be adjusted by controlling the constituents and morphology of the composite structure. Properly tailored composites can display the best qualities of each of their constituents, or, in certain cases, can display properties that even exceed those of their constituents. These features render composite materials valuable for applications in photonics and laser engineering.

It is well known that in dense media the electric field acting on emitters (the local field) is generally different from both the external field and the average (or Maxwell) field within the medium [1,2]. Local-field effects have been shown to be significant in uniform media [3,4]. They can also be a valuable tool in tailoring laser properties of composite materials.

The optical properties of composite materials have been the subject of many studies (see, for example, Refs. [5–8]). In particular, the modification of the radiative lifetime of composite materials caused by local-field effects was addressed in many publications both theoretically [9–13] and experimentally [14–22]. The influence of the local-field effects on the nonlinear optical properties of composite materials is even more significant, as the material response scales as several powers of the local-field correction factor (i.e., the quantity equal to the ratio of the local field acting on a typical emitter to the average field in the medium). Theoretical modeling of the nonlinear optical response has been reported for many different geometries of composite materials [23–26]. In particular, rigorous theories for Maxwell–Garnett-type composite materials [23] and layered composite materials [25] have been developed. It was shown that a significant enhancement of the nonlinear optical response is possible under certain conditions. A number of experiments in the field

yielded promising results [27–30]. Thus, a composite-material approach has proven to be a valuable tool in designing optical materials with enhanced nonlinear response.

While there have been numerous studies of nonlinear composite materials, there has not been a systematic study yet of their laser properties. In this paper, we present a study of the influence of the local-field effects on the laser properties of composite materials. Although local-field effects can come into play differently in different composite geometries, separate theoretical investigations of the laser properties of various composite geometries is outside the scope of this paper. The goal of this paper is to present a general picture and to show the significance of the local-field effects in modifying laser properties. Thus, we limit ourselves with a simple treatment of the local field based on the Lorentz model [1,31]. We believe that the analysis done in this paper will help in further development of new materials for laser applications.

2. COMPOSITE GEOMETRIES

There are three types of composite geometries mainly discussed in the literature: Maxwell–Garnett composites [5,6,23], Bruggeman composites [30,32,33], and layered composites [25,26,29] (see Fig. 1).

The Maxwell–Garnett type of composite geometry is a collection of small particles (the inclusions) distributed in a host medium. The inclusions are assumed to be spheres or ellipsoids of a size much smaller than the optical wavelength; the distance between them must be much larger than their characteristic size and much smaller than the optical wavelength. Under these conditions, one can treat the composite material as an effective medium, characterized by an effective (average) dielectric constant, ϵ_{eff} , which satisfies the relation [5,6]

$$\frac{\epsilon_{\text{eff}} - \epsilon_h}{\epsilon_{\text{eff}} + 2\epsilon_h} = f_i \frac{\epsilon_i - \epsilon_h}{\epsilon_i + 2\epsilon_h}. \quad (1)$$

Here ϵ_h and ϵ_i are the dielectric constants of the host and inclusion materials, respectively, and f_i is the volume frac-

tion of the inclusion material in the composite.

In the Maxwell–Garnett model, the composite medium is treated asymmetrically. It is assumed that the host material completely surrounds the inclusion particles, and the result for the effective dielectric constant of the composite will be different if we interchange the host and inclusion dielectric constants in expression (1). This problem is eliminated in the Bruggeman composite model, in which each particle of each constituent component is considered to be embedded in an effective medium characterized by ϵ_{eff} . The corresponding equation defining the effective dielectric constant thus has the form [30]

$$0 = f_a \frac{\epsilon_a - \epsilon_{\text{eff}}}{\epsilon_a + 2\epsilon_{\text{eff}}} + f_b \frac{\epsilon_b - \epsilon_{\text{eff}}}{\epsilon_b + 2\epsilon_{\text{eff}}}. \quad (2)$$

Here ϵ_a and ϵ_b are the dielectric constants of the constituent components a and b and f_a and f_b are the volume fractions of the components.

The third composite model shown in Fig. 1 is a layered structure consisting of alternating layers of two materials (a and b) with different optical properties. The thicknesses of the layers should be much smaller than the optical wavelength. Materials of this sort are anisotropic. For light polarized parallel to the layers of such a composite, the effective dielectric constant is given by a simple volume average of the dielectric constants of the constituents:

$$\epsilon_{\text{eff}} = f_a \epsilon_a + f_b \epsilon_b. \quad (3)$$

The electric field in this case is spatially uniform, as the boundary conditions require continuity of its tangential part on the border between two constituents. However, for the light polarized perpendicular to the layers, the effective dielectric constant is given by

$$\frac{1}{\epsilon_{\text{eff}}} = \frac{f_a}{\epsilon_a} + \frac{f_b}{\epsilon_b}. \quad (4)$$

In the latter case, the electric field is nonuniformly distributed between the two constituents in the composite, and local-field effects are of particular interest.

The composite geometries that we described above are those studied most often in the design of composite optical materials. In this paper, we limit ourselves to a simple treatment of a composite material as a uniform medium characterized by a dielectric constant ϵ_{eff} . At the present level of approximation, the Lorentz model can be used to describe the effects of the local field on the laser properties of the medium [11]. More detailed treatment of laser properties of composite materials requires developing separate theoretical models for each composite geometry, which is beyond the scope of this paper.

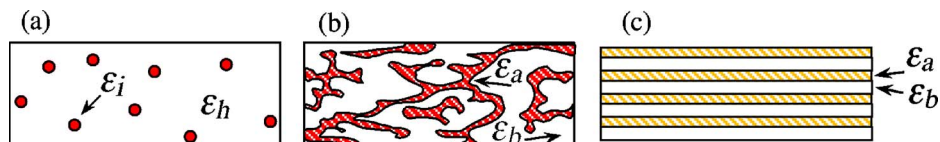


Fig. 1. (Color online) Composite material structures: (a) Maxwell–Garnett geometry, (b) Bruggeman geometry, (c) layered geometry.

3. LOCAL FIELD: LORENTZ MODEL

In this paper, we use the Lorentz model of the local field to account for the modification of the linear laser properties, such as small-signal gain, radiative lifetime, and saturation intensity, by local-field effects. In this section, we briefly review the Lorentz theory of the local field.

Let us assume for now that the medium is lossless and dispersionless. We represent the dipole moment induced in a typical molecule (or atom) of the medium as

$$\tilde{\mathbf{p}} = \alpha \tilde{\mathbf{E}}_{\text{loc}}, \quad (5)$$

where α is the microscopic polarizability and $\tilde{\mathbf{E}}_{\text{loc}}$ is the local field acting on the molecule. (The tilde denotes quantities oscillating at an optical frequency.) The expression

$$\tilde{\mathbf{E}}_{\text{loc}} = \tilde{\mathbf{E}} + \frac{4\pi}{3} \tilde{\mathbf{P}}, \quad (6)$$

relating the local field to the average (macroscopic) polarization $\tilde{\mathbf{P}}$ and the average electric field $\tilde{\mathbf{E}}$ in the medium, is derived in many textbooks (see, for example, Refs. [1,2,34]). The textbook model used for deriving Eq. (6) is known as the virtual-cavity model, because the medium is treated as a cubic lattice of point dipoles, and a fictitious sphere is introduced as a trick for calculating the local field acting on a typical dipole in the medium. An alternative, more elegant, derivation of the relationship (6), which does not require introducing an imaginary sphere, was described by Aspnes [31].

The macroscopic polarization of the material is given by the equation

$$\tilde{\mathbf{P}} = N \tilde{\mathbf{p}}, \quad (7)$$

where N denotes molecular (or atomic) number density. Using Eqs. (5)–(7), we find that the polarization and macroscopic field are related by

$$\tilde{\mathbf{P}} = N \alpha \left(\tilde{\mathbf{E}} + \frac{4\pi}{3} \tilde{\mathbf{P}} \right). \quad (8)$$

We assume that the polarization $\tilde{\mathbf{P}}$ is linear in the average field:

$$\tilde{\mathbf{P}} = \chi^{(1)} \tilde{\mathbf{E}}, \quad (9)$$

where $\chi^{(1)}$ is the linear optical susceptibility of the medium. Substituting expression (8) into Eq. (9), solving for $\chi^{(1)}$, and eliminating the field $\tilde{\mathbf{E}}$, we find that

$$\chi^{(1)} = \frac{N\alpha}{1 - \frac{4\pi}{3}N\alpha}. \quad (10)$$

Expressing the left-hand side of Eq. (10) as $\chi^{(1)} = (\epsilon^{(1)} - 1)/4\pi$ ($\epsilon^{(1)}$ is the dielectric function of the medium), we can derive the well-known Lorentz–Lorenz (or Clausius–Mosotti) relation

$$\frac{\epsilon^{(1)} - 1}{\epsilon^{(1)} + 2} = \frac{4\pi}{3}N\alpha. \quad (11)$$

Through rearrangement of Eq. (11), we can express the linear susceptibility as

$$\chi^{(1)} = \frac{\epsilon^{(1)} + 2}{3}N\alpha. \quad (12)$$

Substituting expression (12) into Eq. (9), then Eq. (9) into Eq. (7), and using the relationship (5) between the local field and the dipole moment, we obtain the equation relating the local field to the average field:

$$\tilde{\mathbf{E}}_{\text{loc}} = \frac{\epsilon^{(1)} + 2}{3}\tilde{\mathbf{E}}. \quad (13)$$

The local field given by Eq. (13) is known in literature as the Lorentz local field, and the factor

$$L = \frac{\epsilon^{(1)} + 2}{3} \quad (14)$$

is known as Lorentz local-field correction factor. Expression (14) for the local-field correction factor is valid in the case of homogeneous media, where all the particles (molecules or atoms) are of the same sort. It is also valid in materials where the emitters enter as interstitial inclusions [10,18].

We use the Lorentz model summarized in this section to phenomenologically study the influence of local-field effects on the laser properties of composite laser media. As we pointed out in Section 2, composite materials can be treated as effective media, as the sizes of the particles of the constituent components are much smaller than the optical wavelength. Under this condition we can consider a composite material as an effective medium characterized by an effective (average) dielectric constant ϵ_{eff} . Thus, at the present level of approximation, we can use Eq. (14) for the local-field correction factor with the effective dielectric constant in place of $\epsilon^{(1)}$.

4. INFLUENCE OF LOCAL-FIELD EFFECTS ON LASER PROPERTIES OF DIELECTRIC MATERIALS

In this section, we describe the modification of laser properties, such as the radiative lifetime, the small-signal gain coefficient, and the saturation intensity by the local-field effects. We undertake our analysis based on a simple argument of the validity of the Lorentz model for treating the local-field effects.

A. Radiative Lifetime

The radiative lifetime τ of emitters in a dielectric material depends on the dielectric constant of the material. It is inversely proportional to the Einstein A coefficient, which, in turn, can be expressed through Fermi's golden rule as

$$A = \frac{1}{\tau} = \frac{2\pi}{\hbar} |V_{12}(\omega_0)|^2 \rho(\omega_0). \quad (15)$$

Here $V_{12}(\omega_0)$ is the energy of interaction between the emitter and the electric field in the medium, and $\rho(\omega_0)$ is the density of states at the emission frequency ω_0 . In a medium with refractive index n_{eff} in which the local-field effects are significant, the interaction energy scales as

$$V_{12,\text{loc}} \propto \frac{L}{\sqrt{n_{\text{eff}}}}. \quad (16)$$

The factor L enters the expression for the local-field-corrected interaction energy $V_{12,\text{loc}}(\omega_0)$ because the local field acting on an individual emitter differs from the macroscopic average field. The factor $\sqrt{n_{\text{eff}}}$ in the denominator of Eq. (16) comes from mode normalization and thus appears in the expression for the electromagnetic energy density in a dielectric medium [11]. The density of states in the medium is proportional to the square of the effective refractive index:

$$\rho(\omega_0) \propto n_{\text{eff}}^2. \quad (17)$$

Using expressions (15)–(17), we can conclude that the local-field-corrected spontaneous emission rate A_{loc} in the medium with refractive index n_{eff} is related to the spontaneous emission rate in the medium of unit refractive index (we call it A_{vac}) as

$$A_{\text{loc}} = n_{\text{eff}} |L|^2 A_{\text{vac}}. \quad (18)$$

The relation (18) has been shown to hold also when the effect of dispersion is included in $V_{12,\text{loc}}$ and in the density of states [11]. The corresponding relation for the local-field-corrected radiative lifetime τ_{loc} in terms of the “vacuum” lifetime τ_{vac} takes the form

$$\tau_{\text{loc}} = \frac{\tau_{\text{vac}}}{n_{\text{eff}} |L|^2}. \quad (19)$$

Here and in all later sections of this paper we assign the “vac” subscript to the variables denoting quantities in a medium with the same chemical environment as that of the dielectric medium under consideration, but with the refractive index equal to unity. The variables marked with the “loc” subscript denote the local-field-corrected quantities.

B. Small-Signal Gain

In this paper, we focus on laser gain media. Most laser gain media can be modeled as collections of two-level atoms, regardless of what the actual level diagram of the active medium is, because in most cases the nonradiative transitions are much faster than the radiative transition from the upper laser level. We use the two-level-atom model to derive the expression for the local-field-corrected small-signal gain.

We start from the driven wave equation

$$-\nabla^2 \tilde{E} + \frac{1}{c^2} \frac{\partial^2 \tilde{E}}{\partial t^2} = -\frac{4\pi}{c^2} \frac{\partial^2 \tilde{P}}{\partial t^2}. \quad (20)$$

(We limit ourselves to considering scalar fields for simplicity.) Here

$$\tilde{E} = E(z)e^{-i\omega t} + \text{c.c.} = A_0 e^{i(\bar{k}z + \omega t)} + \text{c.c.} \quad (21)$$

is the average electric field. In the general case of a lossy or amplifying medium, the parameter \bar{k} is complex: $\bar{k} = k + i\alpha_0/2$. The real part of \bar{k} is the wavenumber:

$$\text{Re}(\bar{k}) \equiv k = \frac{\omega n_{\text{eff}}}{c}, \quad (22)$$

where n_{eff} is the effective refractive index of the medium, while the imaginary part of \bar{k} characterizes amplification or attenuation in the medium, according to

$$\text{Im}(\bar{k}) = \frac{1}{2} \alpha_0 = -\frac{1}{2} g_0. \quad (23)$$

Here α_0 and g_0 are the small-signal intensity absorption and gain coefficients, respectively. We seek the solution for the local-field-corrected small-signal gain coefficient, which we denote as $g_{0,\text{loc}}$.

We take the polarization \tilde{P} entering the wave equation (20) to be linear:

$$\tilde{P} = \chi_{\text{loc}}^{(1)} \tilde{E}, \quad (24)$$

where $\chi_{\text{loc}}^{(1)}$ is the local-field-corrected linear susceptibility. For a collection of two-level atoms [35],

$$\chi_{\text{loc}}^{(1)} = -\frac{c \alpha_{0,\text{vac}}(\Delta)}{4\pi\omega_{ba}} L(T_2\Delta - i), \quad (25)$$

where ω_{ba} is the frequency of the atomic transition, L is the local-field-correction factor, $\Delta = \omega - \omega_{ba}$ is the detuning of the optical field with respect to the atomic transition frequency, and T_2 is the coherence relaxation time. The vacuum absorption $\alpha_{0,\text{vac}}(\Delta)$ experienced by a weak optical wave detuned from the resonance is given by [35],

$$\alpha_{0,\text{vac}}(\Delta) = -\frac{4\pi\omega_{ba} N w^{\text{eq}} |\mu_{ba}|^2 T_2}{c \hbar (T_2^2 \Delta^2 + 1)}, \quad (26)$$

where w^{eq} is the equilibrium value of the population inversion, N is the atomic number density, and μ_{ba} is the transition dipole moment of the two-level atom.

The local-field-correction factor L , given by Eq. (14) as $L = (\epsilon_{\text{eff}} + 2)/3$, can be shown to take the form

$$L = \frac{T_2\Delta + i}{T_2(\Delta - \Delta_L) + i}, \quad (27)$$

if one chooses to express ϵ_{eff} in terms of atomic parameters as [4,36]:

$$\epsilon_{\text{eff}} = 1 - \frac{f r_e \lambda_0 c N}{\Delta - \Delta_L + i/T_2}. \quad (28)$$

Here $\Delta_L = -(f r_e \lambda_0 c N)/3$ is the Lorentz redshift [4] (f is the oscillator strength of the atomic transition, $r_e = e^2/mc^2$ is the classical electron radius, and λ_0 is the vacuum transition wavelength). The Lorentz redshift appears in the expression for ϵ_{eff} as a consequence of the local-field effects and leads to $L \neq 1$ (note that if $\Delta_L = 0$, then $L = 1$, and there are no local-field effects).

Substituting the electric field given by Eqs. (21)–(23) and the polarization in the form of Eq. (24) into the wave equation (20), then taking the time and space derivatives and dropping the electric field that appears as a multiplicative factor on both sides of the resulting equation, we end up with the equation

$$g_{0,\text{loc}} = -\frac{4\pi k}{n_{\text{eff}}^2} \text{Im}[\chi_{\text{loc}}^{(1)}] \quad (29)$$

for the gain coefficient. Substituting the factor L given by Eq. (27) into expression (25) for the linear susceptibility, and then Eq. (25) into Eq. (29), we arrive at the expression

$$g_{0,\text{loc}}(\Delta) = -\frac{\alpha_{0,\text{vac}}(\Delta)}{n_{\text{eff}}} \frac{\omega}{\omega_{ba}} |L|^2 = \frac{g_{0,\text{vac}}(\Delta)}{n_{\text{eff}}} \frac{\omega}{\omega_{ba}} |L|^2 \quad (30)$$

for the local-field-corrected small-signal gain in terms of the vacuum absorption coefficient $\alpha_{0,\text{vac}}(\Delta)$, or vacuum gain coefficient $g_{0,\text{vac}}(\Delta)$ (i.e., the absorption or gain coefficient in a medium with the unit refractive index). Assuming that the optical wave is in resonance with the atomic transition ($\Delta = 0$), we find the following expression for the local-field-corrected gain coefficient:

$$g_{0,\text{loc}}(\Delta) = -\frac{g_{0,\text{vac}}(0)}{n_{\text{eff}}} |L|^2. \quad (31)$$

It is in agreement with that obtained by Milonni [11].

C. Saturation Intensity

In this section, we use the following convention regarding the way we denote different variables. We assign the vac subscript to the variables describing quantities in a medium with unit refractive index, no subscript to variables denoting non-local-field-corrected quantities in a medium with the refractive index n_{eff} , and the loc subscript to variables denoting local-field-corrected quantities.

The gain g for a homogeneous atomic transition saturates with increasing signal intensity $I = (n_{\text{eff}} c |E|^2)/(2\pi)$ according to [37]:

$$g = \frac{g_0}{1 + I/I_s}. \quad (32)$$

The saturation intensity is the intensity that reduces the small-signal gain coefficient to a half of its value; it is given in terms of the atomic transition parameters by [35]:

$$I_s = \frac{cn_{\text{eff}} \hbar^2}{2\pi 4|\mu_{ba}|^2 T_1 T_2}. \quad (33)$$

Here T_1 is the population relaxation time.

To account for local-field effects in Eq. (32) for saturated gain coefficient, one has to substitute the local-field-corrected counterparts of the quantities entering the equation. We obtained an equation for the local-field-corrected small-signal gain coefficient $g_{0,\text{loc}}$ in the previous section [see Eq. (31)]. The local-field-corrected intensity can be written as

$$I_{\text{loc}} = \frac{n_{\text{eff}} c}{2\pi} |E_{\text{loc}}|^2 = \frac{n_{\text{eff}} c}{2\pi} |L|^2 |E|^2 = |L|^2 I. \quad (34)$$

Expression (33) for the saturation intensity contains two quantities that may be affected by local-field effects, namely, T_1 and T_2 . We assume that T_2 does not depend on the local-field correction factor, as would be true for many line-broadening mechanisms. Thus, we can simply retain the vacuum value of T_2 in the equation for the saturation intensity. T_1 is the lifetime of the upper laser level, which we assume to be purely radiative. Thus, the result Eq. (19) obtained earlier applies:

$$T_{1,\text{loc}} = \frac{T_{1,\text{vac}}}{n_{\text{eff}} |L|^2}. \quad (35)$$

Making use of Eq. (35) and the assumption that T_2 does not introduce local-field correction to the expression for the saturation intensity, we find that

$$I'_s = \frac{cn_{\text{eff}} \hbar^2}{2\pi 4|\mu_{ba}|^2 T_{1,\text{loc}} T_{2,\text{loc}}} = \frac{cn_{\text{eff}} \hbar^2 n_{\text{eff}} |L|^2}{2\pi 4|\mu_{ba}|^2 T_{1,\text{vac}} T_{2,\text{vac}}}. \quad (36)$$

In Eq. (36), we denote the saturation intensity I'_s in order to discriminate it from both the non-local-field-corrected saturation intensity I_s and the local-field-corrected saturation intensity $I_{s,\text{loc}}$, as we did not account for all the local-field corrections affecting the saturation intensity yet.

The local-field-corrected saturated gain coefficient can be written as

$$g_{\text{loc}} = \frac{g_{0,\text{loc}}}{1 + \frac{I_{\text{loc}}}{I'_s}} \quad (37)$$

in terms of the quantities given by Eqs. (31), (34), and (36). We define the local-field-corrected saturation intensity so that, if written in terms of it, Eq. (37) takes the form

$$g_{\text{loc}} = \frac{g_{0,\text{loc}}}{1 + \frac{I}{I_{s,\text{loc}}}}. \quad (38)$$

Making use of Eqs.(36)–(38), we express the local-field-corrected saturation intensity in terms of the vacuum saturation intensity as

$$I_{s,\text{loc}} = n_{\text{eff}}^2 I_{s,\text{vac}}. \quad (39)$$

5. ANALYSIS

The basic operation of lasers can be characterized most simply in terms of the upper-level spontaneous emission lifetime τ , the laser gain coefficient g , and the gain saturation intensity I_s . All three of these parameters can be controlled through use of composite material geometry. In the simplest formulation of local-field effects, we can assume that these laser parameters depend only on the effective value n_{eff} of the refractive index of the host material. We showed in the previous sections of this paper that the basic laser parameters scale with the effective refractive index according to

$$A_{\text{loc}} = n_{\text{eff}} |L|^2 A_{\text{vac}}, \quad (40a)$$

$$g_{0,\text{loc}} = \frac{|L|^2}{n_{\text{eff}}} g_{0,\text{vac}}, \quad (40b)$$

$$I_{s,\text{loc}} = n_{\text{eff}}^2 I_{s,\text{vac}}. \quad (40c)$$

The quantities marked with vac denote the values of the parameters in a medium with the same chemical environment but with the refractive index equal to unity.

Treating the composite laser gain medium as an effective medium and assuming that the amplification (and loss) at the laser transition frequency is small enough to neglect the imaginary part of the effective dielectric constant ϵ_{eff} , we can express the local-field correction factor, given by Eq. (14), as

$$L = \frac{n_{\text{eff}}^2 + 2}{3}. \quad (41)$$

Making use of expression (41) for the factor L , we plot the local-field-corrected basic laser parameters, given by Eq. (40), in Fig. 2. We choose the range of refractive indices available in dielectric composite materials. Clearly, significant control over the laser parameters is available through use of a composite geometry.

Control of the three laser parameters is crucial for the development of laser systems for the following reasons. (1) The upper-state lifetime controls how large the pumping rate of the laser needs to be in order to establish a population inversion. (2) The gain coefficient determines

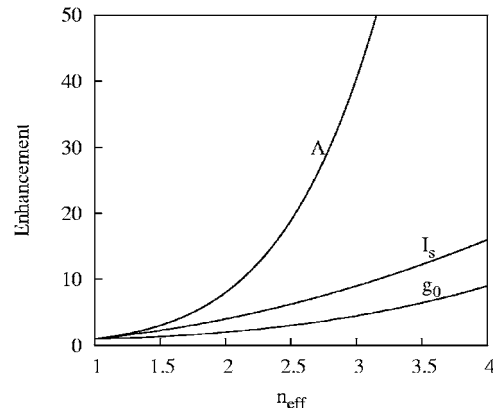


Fig. 2. Variation of the principal parameters that controls the basic operation of a laser with the effective refractive index of the composite material.

the laser threshold condition. The gain needs to be large enough for the laser to reach the threshold, but it is not desirable for the gain to be too large because excessive gain can lead to the development of parasitic effects such as amplified spontaneous emission. (3) The saturation intensity (and its related energy quantity, the saturation fluence) determines the output power of a laser, from the point of view that the output power is determined by the condition that the saturated round-trip gain must equal the round-trip loss. In practice, the output intensity of most lasers is typically a factor of several times the saturation intensity.

Our assumption that local-field effects in composite laser gain media can be accounted for using the Lorentz model with the effective refractive index entering the expression for the factor L is good for conceptual understanding of how the local-field effects help one to manipulate the laser parameters. For a more precise and detailed analysis of a particular composite geometry, a more sophisticated model needs to be developed. This portion of work is in progress.

6. CONCLUSIONS

We propose a method for controlling and tailoring the basic laser properties of laser gain media, such as the radiative lifetime of the upper laser level, the small-signal gain coefficient, and the saturation intensity. The idea behind the method is designing new composite materials by mixing two or more materials on a nanoscale to obtain new laser gain media with the laser properties enhanced compared with those of the constituents. The enhancement can be achieved by implementing local-field effects that can significantly modify optical properties of materials. The composite-material approach has been used for enhancing the nonlinear properties of dielectric [23–27,29,30] and metal-dielectric [28,38] composites. However, we are unaware of any previous systematic study of the modification of the laser properties of composite materials by the local-field effects.

In this paper, we report a preliminary study of how local-field effects can help one to change the laser properties of composite materials in a desired way. This proof-of-principle study shows that it is possible to independently control the radiative lifetime, gain coefficient, and saturation intensity. The equations demonstrating how these basic laser characteristics scale with the effective refractive index and the local-field correction factor were derived [see Eqs. (40a)–(40c)]. We believe that the equations can be used for different composite geometries. Although our approach is general to all composite geometries for the linear optical regime and can be used with a good precision for predicting the linear laser properties, more complete theories, unique to different composite geometries, are needed. We are currently working in this direction.

ACKNOWLEDGMENTS

We thank Nick Lepeshkin, Peter Milonni, and John Sipe for valuable discussions of the topic described here. This work was supported by the National Science Foundation.

REFERENCES

1. H. A. Lorentz, *Theory of Electrons*, 2nd ed. (Teubner, 1916).
2. J. D. Jackson, *Classical Electrodynamics* (Wiley, 1962).
3. J. Guo, J. Cooper, and A. Gallagher, "Selective reflection from a dense atomic vapor," *Phys. Rev. A* **53**, 1130–1138 (1996).
4. J. J. Maki, M. S. Malcuit, J. E. Sipe, and R. W. Boyd, "Linear and nonlinear optical measurements of the Lorentz local field," *Phys. Rev. Lett.* **67**, 972–975 (1991).
5. J. C. Maxwell Garnett, "Colours in metal glasses and in metallic films," *Philos. Trans. R. Soc. London, Ser. A* **203**, 385–420 (1904).
6. J. C. Maxwell Garnett, "Colours in metal glasses, in metallic films, and in metallic solutions," *Philos. Trans. R. Soc. London, Ser. A* **205**, 237–288 (1906).
7. V. M. Shalaev and M. I. Stockman, "Fractals: optical susceptibility and giant Raman scattering," *Z. Phys. D: At., Mol. Clusters* **10**, 71–79 (1988).
8. R. J. Gehr and R. W. Boyd, "Optical properties of nanostructured optical materials," *Chem. Mater.* **1996**, 1807–1819 (1996).
9. R. J. Glauber and M. Lewenstein, "Quantum optics of dielectric media," *Phys. Rev. A* **43**, 467–491 (1991).
10. P. de Vries and A. Lagendijk, "Resonant scattering and spontaneous emission in dielectrics: microscopic derivation of local-field effects," *Phys. Rev. Lett.* **81**, 1381–1384 (1998).
11. P. W. Milonni, "Field quantization and radiative processes in dispersive dielectric media," *J. Mod. Opt.* **42**, 1991–2004 (1995).
12. M. S. Tomas, "Local-field corrections to the decay rate of excited molecules in absorbing cavities: the Onsager model," *Phys. Rev. A* **63**, 053811 (2001).
13. H. T. Dung, S. Y. Buhmann, and D.-G. Welsch, "Local-field correction to the spontaneous decay rate of atoms embedded in bodies of finite size," *Phys. Rev. A* **74**, 023803 (2006).
14. G. L. J. A. Rikken and Y. A. R. R. Kessener, "Local field effects and electric and magnetic dipole transitions in dielectrics," *Phys. Rev. Lett.* **74**, 880–883 (1995).
15. F. J. P. Schuurmans, D. T. N. de Lang, G. H. Wegdam, R. Sprik, and A. Lagendijk, "Local-field effects on spontaneous emission in a dense supercritical gas," *Phys. Rev. Lett.* **80**, 5077–5080 (1998).
16. G. M. Kumar, D. N. Rao, and G. S. Agarwal, "Measurement of local field effects of the host on the lifetimes of embedded emitters," *Phys. Rev. Lett.* **91**, 203903 (2003).
17. G. M. Kumar, D. N. Rao, and G. S. Agarwal, "Experimental studies of spontaneous emission from dopants in an absorbing dielectric," *Opt. Lett.* **30**, 732–734 (2005).
18. C.-K. Duan, M. F. Reid, and Z. Wang, "Local field effects on the radiative lifetime of emitters in surrounding media: virtual- or real-cavity model?," *Phys. Lett. A* **343**, 474–480 (2005).
19. P. Lavallard, M. Rosenbauer, and T. Gacoin, "Influence of surrounding dielectrics on the spontaneous emission of sulphorhodamine B molecules," *Phys. Rev. A* **54**, 5450–5453 (1996).
20. G. Lamouche, P. Lavallard, and T. Gacoin, "Optical properties of dye molecules as a function of the surrounding dielectric medium," *Phys. Rev. A* **59**, 4668–4674 (1999).
21. S. F. Wuister, C. de Mello Donega, and A. Meijerink, "Local-field effects on the spontaneous emission rate of CdTe and CdSe quantum dots in dielectric media," *J. Chem. Phys.* **121**, 4310–4315 (2004).
22. K. P. Dolgaleva, R. W. Boyd, and P. W. Milonni, "Influence of local-field effects on the radiative lifetime of liquid suspensions of Nd:YAG nanoparticles," *J. Opt. Soc. Am. B* **24**, 516–521 (2007).
23. J. E. Sipe and R. W. Boyd, "Nonlinear susceptibility of composite optical materials in the Maxwell Garnett model," *Phys. Rev. A* **46**, 1614–1629 (1992).
24. J. E. Sipe and R. W. Boyd, "Nanocomposite materials for nonlinear optics based on local field effects," in *Optical Properties of Nanostructured Random Media*, Vol. 82 of

- Topics in Applied Physics, V. M. Shalaev, ed. (Springer-Verlag Berlin, 2002), pp. 1–19.
25. R. W. Boyd and J. E. Sipe, “Nonlinear optical susceptibilities of layered composite materials,” *J. Opt. Soc. Am. B* **11**, 297–303 (1994).
 26. R. W. Boyd, R. J. Gehr, G. L. Fischer, and J. E. Sipe, “Nonlinear optical properties of nanocomposite materials,” *Pure Appl. Opt.* **5**, 505–512 (1996).
 27. G. L. Fischer, R. W. Boyd, R. J. Gehr, S. A. Jenekhe, J. A. Osaheni, J. E. Sipe, and L. A. Weller-Brophy, “Enhanced nonlinear optical response of composite materials,” *Phys. Rev. Lett.* **74**, 1871–1874 (1995).
 28. D. D. Smith, G. Fischer, R. W. Boyd, and D. A. Gregory, “Cancellation of photoinduced absorption in metal nanoparticle composites through a counterintuitive consequence of local field effects,” *J. Opt. Soc. Am. B* **14**, 1625–1631 (1997).
 29. R. L. Nelson and R. W. Boyd, “Enhanced electro-optic response of layered composite materials,” *Appl. Phys. Lett.* **74**, 2417–2419 (1999).
 30. R. J. Gehr, G. L. Fischer, and R. W. Boyd, “Nonlinear-optical response of porous-glass-based composite materials,” *J. Opt. Soc. Am. B* **14**, 2310–2314 (1997).
 31. D. E. Aspnes, “Local-field effects and effective-medium theory: a microscopic perspective,” *Am. J. Phys.* **50**, 704–709 (1982).
 32. R. Landauer, “The electrical resistance of binary metallic mixtures,” *J. Appl. Phys.* **23**, 779–784 (1952).
 33. X. C. Zeng, D. J. Bergman, P. M. Hui, and D. Stroud, “Effective-medium theory for weakly nonlinear composites,” *Phys. Rev. B* **38**, 10970–10973 (1988).
 34. M. Born and E. Wolf, *Principles of Optics*, 6th ed. (Cambridge U. Press, 1997).
 35. R. W. Boyd, *Nonlinear Optics*, 2nd ed. (Academic, 2003).
 36. H. A. Lorentz, *The Theory of Electrons*, 2nd ed. (Dover, 1965).
 37. A. E. Siegman, *Lasers* (University Science, 1986).
 38. R. S. Bennink, Y. K. Yoon, and R. W. Boyd, “Accessing the optical nonlinearity of metals with metal-dielectric photonic bandgap structures,” *Opt. Lett.* **24**, 1416–1418 (1999).