Effective-medium theory of a nonlinear composite medium using the T-matrix approach: Exact results for spherical grains

N. C. Kothari
School of Physics, University of Hyderabad, Hyderabad 500 134, India
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An effective-medium theory for nonlinear spherical particles embedded in a linear host medium is developed. The formulation is based on a T-matrix approach. Introduction of a field-dependent T matrix enables one to obtain an exact expression for effective dielectric constant of a medium with spherical grains. In the special cases of low-field values, the results obtained previously for the third- and fifth-order nonlinear susceptibilities of the medium are recovered. The resonance behavior of the effective dielectric constant and intrinsic optical bistability are also discussed.

I. INTRODUCTION

The effective dielectric function for a linear composite medium can be calculated using several different methods.[1–5] For a small volume fraction f of the inhomogeneities, the T-matrix formulation reproduces the well-known Maxwell-Garnett result. This formulation was also extended to include the effects due to the nonlocal dielectric function for individual particles.[5] Recently, the attention has shifted to the nonlinear response of these media owing to the possibilities of producing enhanced nonlinearities in metal colloids and semiconductor crystallites. Surface-plasmon resonances of the metal particles and excitonic behavior observable in many semiconductors at low temperature are enhanced by quantum size effects. Interesting experiments performed by Flytzanis and co-workers[6–10] indicate a huge (six to eight orders of magnitude) enhancement of effective nonlinearities by a colloidal medium.

Agarwal and Dutta Gupta[11] have recently calculated the third- and fifth-order nonlinear susceptibilities of a composite medium by generalizing the T-matrix formulation. They assume each spherical particle to be a Kerr-like medium and show that a third-order nonlinearity of the particles can lead to all higher odd-order nonlinearities in the macroscopic behavior of the composite medium. Recently, Haus *et al.*[12,13] have developed the effective-medium theory for nonlinear ellipsoidal particles embedded in a linear or nonlinear host dielectric. The effective-medium dielectric constant in the linear regime for ellipsoidal particles has already been calculated previously.[14]

In the present work, we follow the T-matrix approach and calculate the effective-medium dielectric constant which includes the contributions from all higher odd-order nonlinear susceptibilities. The nonlinear T matrix involved is field dependent and its form cannot be obtained explicitly. However, in the actual calculations the averages of T matrices are involved and hence the explicit forms of T matrices are irrelevant. We consider semiconductor microcrystallites in a vacuum and study the resonance behavior of the effective dielectric constant for this type of composite medium. The enhanced nonlinearity due to resonances, combined with local-field effects in particles arising from dielectric confinement, which is responsible for intrinsic feedback, are shown to produce the bistable behavior of the local field with respect to the applied field and also with Maxwell field. Similar studies have also been reported earlier for a single particle[15,16] and a colloidal medium.[17]

II. T-MATRIX FORMULATION

In this section, the T-matrix formulation is developed for a nonlinear heterogeneous medium. Let the nonlinear medium be characterized by a dielectric function

$$\epsilon=\epsilon_0+\delta\epsilon_M \ , \tag{2.1}$$

where ϵ_0 is the spatially invariant (homogeneous) part and $\delta\epsilon_M$ is the inhomogeneous part. The inhomogeneous part $\delta\epsilon_M$ is made up of linear as well as field-dependent nonlinear contributions

$$\delta\epsilon_M=\delta\epsilon+\frac{A|\mathbf{E}|^2}{1+B|\mathbf{E}|^2} \ . \tag{2.2}$$

We treat $\delta\epsilon_M$ as a perturbation to ϵ_0 and solve Maxwell's equation

$$\nabla\cdot\mathbf{D}=0,\quad \mathbf{D}=\epsilon\mathbf{E} \ , \tag{2.3}$$

where $\mathbf{D}$ is the displacement vector and $\mathbf{E}$ is the electric field. If $\overleftrightarrow{G}(\mathbf{r},\mathbf{r}')$, is the Green's function satisfying the equation

$$\nabla\cdot\epsilon_0\overleftrightarrow{G}=\delta(\mathbf{r}-\mathbf{r}')\overleftrightarrow{I} \ , \tag{2.4}$$

then the solution of Eq. (2.3) can be expressed as

$$\mathbf{E}(\mathbf{r})=\mathbf{E}_0+\int d\mathbf{r}'[\delta\epsilon_M(\mathbf{r}')\mathbf{E}(\mathbf{r}')\cdot\nabla']\overleftrightarrow{G}(\mathbf{r},\mathbf{r}') \ . \tag{2.5}$$

In Eq. (2.5), $\mathbf{E}_0$ is the field inside the medium in the absence of inhomogeneities ($\delta\epsilon_M=0$), and is the solution of the homogeneous equation

$$\nabla\cdot\epsilon_0\mathbf{E}_0=0 \ . \tag{2.6}$$

The field $\mathbf{E}_0$ will be assumed to be constant. Equation (2.5) can be rewritten in a compact form as

$$\mathbf{E}=\mathbf{E}_0+G\delta\epsilon_M\mathbf{E}\ . \tag{2.7}$$

Substituting Eq. (2.2) in Eq. (2.7), we obtain

$$\mathbf{E}=\mathbf{E}_0+G\delta\epsilon\mathbf{E}+G\frac{A|\mathbf{E}|^2\mathbf{E}}{1+B|\mathbf{E}|^2}\ . \tag{2.8}$$

The solution to the integral equation (2.8) is written as

$$\mathbf{E}=\mathbf{E}_0+GT_{\mathrm{L}}\mathbf{E}_0+GT_{\mathrm{NL}}|\mathbf{E}_0|^2\mathbf{E}_0\ , \tag{2.9}$$

where in Eq. (2.9) we have introduced linear and nonlinear T matrices T_{L} and T_{NL}. The explicit expressions for T_{L} and T_{NL} are of no relevance in actual calculations. Only the averages of T matrices are needed. Comparing linear and nonlinear terms of Eq. (2.8) with that of Eq. (2.9), we get

$$\delta\epsilon\mathbf{E}=T_{\mathrm{L}}\mathbf{E}_0\ , \tag{2.10}$$

$$\frac{A|\mathbf{E}|^2\mathbf{E}}{1+B|\mathbf{E}|^2}=T_{\mathrm{NL}}|\mathbf{E}_0|^2E_0\ . \tag{2.11}$$

Equations (2.10) and (2.11) enable us to rewrite the second equation of Eq. (2.3) as

$$\mathbf{D}=\epsilon_0\mathbf{E}+T_{\mathrm{L}}\mathbf{E}_0+T_{\mathrm{NL}}|\mathbf{E}_0|^2\mathbf{E}_0\ . \tag{2.12}$$

Now, we perform an ensemble average (denoted by $\langle\ \rangle$) of Eqs. (2.9) and (2.12) to obtain the following relations:

$$\langle\mathbf{E}\rangle=(1+\langle GT_{\mathrm{L}}\rangle)\mathbf{E}_0+\langle GT_{\mathrm{NL}}\rangle|\mathbf{E}_0|^2\mathbf{E}_0\ , \tag{2.13}$$

$$\langle\mathbf{D}\rangle=\epsilon_0\langle\mathbf{E}\rangle+\langle T_{\mathrm{L}}\rangle\mathbf{E}_0+\langle T_{\mathrm{NL}}\rangle|\mathbf{E}_0|^2\mathbf{E}_0\ . \tag{2.14}$$

Introducing the effective dielectric function ϵ_{eff} as given by

$$\epsilon_{\mathrm{eff}}=\bar{\epsilon}+\frac{\bar{A}\langle|\mathbf{E}|^2\rangle}{1+\bar{B}\langle|\mathbf{E}|^2\rangle}\ , \tag{2.15}$$

we can write the average effective displacement vector $\langle\mathbf{D}\rangle$ as

$$\langle\mathbf{D}\rangle=\bar{\epsilon}\langle\mathbf{E}\rangle+\frac{\bar{A}|\langle\mathbf{E}\rangle|^2\langle\mathbf{E}\rangle}{1+\bar{B}|\langle\mathbf{E}\rangle|^2}\ . \tag{2.16}$$

Substituting Eq. (2.13) into Eqs. (2.14) and (2.16), we obtain two different forms for the mean displacement:

$$\langle\mathbf{D}\rangle=[\epsilon_0(1+\langle GT_{\mathrm{L}}\rangle)+\langle T_{\mathrm{L}}\rangle]\mathbf{E}_0+[\epsilon_0\langle GT_{\mathrm{NL}}\rangle+\langle T_{\mathrm{NL}}\rangle]|\mathbf{E}_0|^2\mathbf{E}_0\ , \tag{2.17}$$

$$\langle\mathbf{D}\rangle=\bar{\epsilon}(1+\langle GT_{\mathrm{L}}\rangle)\mathbf{E}_0+\bar{\epsilon}\langle GT_{\mathrm{NL}}\rangle|\mathbf{E}_0|^2\mathbf{E}_0+\frac{\bar{A}P_0|\mathbf{E}_0|^2\mathbf{E}_0}{1+\bar{B}Q_0|\mathbf{E}_0|^2}\ , \tag{2.18}$$

where

$$P_0=(1+\langle GT_{\mathrm{L}}\rangle)|1+\langle GT_{\mathrm{L}}\rangle|^2+\langle GT_{\mathrm{NL}}\rangle^*(1+\langle GT_{\mathrm{L}}\rangle)^2|\mathbf{E}_0|^2+2\langle GT_{\mathrm{NL}}\rangle|1+\langle GT_{\mathrm{L}}\rangle|^2|\mathbf{E}_0|^2$$
$$+2(1+\langle GT_{\mathrm{L}}\rangle)|\langle GT_{\mathrm{NL}}\rangle|^2|\mathbf{E}_0|^4+(\langle GT_{\mathrm{NL}}\rangle)^2(1+\langle GT_{\mathrm{L}}\rangle)^*|\mathbf{E}_0|^4+\langle GT_{\mathrm{NL}}\rangle|\langle GT_{\mathrm{NL}}\rangle|^2|\mathbf{E}_0|^6\ , \tag{2.19}$$

$$Q_0=|1+\langle GT_{\mathrm{L}}\rangle|^2+\langle GT_{\mathrm{NL}}\rangle^*(1+\langle GT_{\mathrm{L}}\rangle)|\mathbf{E}_0|^2+\langle GT_{\mathrm{NL}}\rangle(1+\langle GT_{\mathrm{L}}\rangle)^*|\mathbf{E}_0|^2+|\langle GT_{\mathrm{NL}}\rangle|^2|\mathbf{E}_0|^4\ . \tag{2.20}$$

A comparison of Eqs. (2.17) and (2.18) yields the following:

$$\bar{\epsilon}=\epsilon_0+\frac{\langle T_L\rangle}{1+\langle GT_{\mathrm{L}}\rangle}\ , \tag{2.21}$$

$$\frac{\bar{A}P_0}{1+\bar{B}Q_0|E_0|^2}=\langle T_{\mathrm{NL}}\rangle-(\bar{\epsilon}-\epsilon_0)\langle GT_{\mathrm{NL}}\rangle\ . \tag{2.22}$$

The quantities $\bar{A}$ and $\bar{B}$ are to be determined for an effective medium. This will be done in the next section for spherical grains embedded in a linear homogeneous medium.

III. SPHERICAL PARTICLES IN THE HOST MEDIUM

Let the nonlinear heterogeneous medium consist of nonlinear spherical particles embedded in a linear dielectric with dielectric constant ϵ_0. The particles will be assumed to have sharp size distribution with sizes much smaller than the wavelength of light. Let the particles be characterized by the nonlinear dielectric function ϵ_p given by

$$\epsilon_p=\epsilon_1+\frac{\alpha|\mathbf{E}_{\mathrm{L}}|^2}{1+\beta|\mathbf{E}_{\mathrm{L}}|^2}\ , \tag{3.1}$$

where the first term ϵ_1 gives the linear part and the second term gives the nonlinear contribution to ϵ_p with α and β constants. The field $\mathbf{E}_{\mathrm{L}}$ is the local field inside the particle. For a sphere the local field $\mathbf{E}_{\mathrm{L}}$ is related to the applied field $\mathbf{E}_0$ by a relation

$$\mathbf{E}_{\mathrm{L}}=\frac{3\epsilon_0}{\epsilon_p+2\epsilon_0}\mathbf{E}_0\ . \tag{3.2}$$

Using Eq. (3.1) in Eq. (3.2), we obtain

$$\mathbf{E}_{\mathrm{L}}=\mathbf{E}_0X-\frac{a|\mathbf{E}_{\mathrm{L}}|^2\mathbf{E}_{\mathrm{L}}}{1+\beta|\mathbf{E}_{\mathrm{L}}|^2}\ , \tag{3.3}$$

where we have defined

$$X=\frac{3\epsilon_0}{\epsilon_1+2\epsilon_0},\quad a=\frac{\alpha}{\epsilon_1+2\epsilon_0}\ . \tag{3.4}$$

Equation (3.3) has the solution given by

$$\mathbf{E}_{\mathrm{L}}=\mathbf{E}_0Xs\ , \tag{3.5}$$

where s is a function of the applied field $\mathbf{E}_0$, and satisfies the following transcendental equation:

$$s=1-\frac{a|\mathbf{E}_0|^2|X|^2|s|^2}{1+(a+\beta)|\mathbf{E}_0|^2|X|^2|s|^2} . \tag{3.6}$$

For a single particle in the host medium, the local field $\mathbf{E}_L$ is the propagating Maxwell field. Comparison of Eq. (3.5) with Eq. (2.13), after using Eq. (3.6), gives

$$1+\langle GT_L\rangle = X , \tag{3.7}$$

$$\langle GT_{NL}\rangle = -\frac{aX|X|^2|s|^2}{1+(a+\beta)|\mathbf{E}_0|^2|X|^2|s|^2} . \tag{3.8}$$

Now, we calculate the local displacement vector $\mathbf{D}_L(=\epsilon_p\mathbf{E}_L)$ inside the particle. Using Eqs. (3.1), (3.5), and (3.6), we obtain

$$\mathbf{D}_L=\epsilon_1\mathbf{E}_0X+\left[1-\frac{\epsilon_1 X}{3\epsilon_0}\right]\times\left[\frac{aX|X|^2|s|^2|\mathbf{E}_0|^2\mathbf{E}_0}{1+(a+\beta)|\mathbf{E}_0|^2|X|^2|s|^2}\right] . \tag{3.9}$$

Comparison of Eq. (3.9) with Eq. (2.17), and using Eqs. (3.7) and (3.8) gives

$$\epsilon_0X+\langle T_L\rangle=\epsilon_1X , \tag{3.10}$$

$$-\frac{\epsilon_0 aX|X|^2|s|^2}{1+(a+\beta)|\mathbf{E}_0|^2|X|^2|s|^2}+\langle T_{NL}\rangle = \left[1-\frac{\epsilon_1X}{3\epsilon_0}\right]\left[\frac{aX|X|^2|s|^2}{1+(a+\beta)|\mathbf{E}_0|^2|X|^2|s|^2}\right] . \tag{3.11}$$

From Eqs. (3.7), (3.8), (3.10), and (3.11), we have

$$\langle GT_L\rangle = X-1 , \tag{3.12}$$

$$\langle GT_{NL}\rangle = -\frac{aX|X|^2|s|^2}{1+(a+\beta)|\mathbf{E}_0|^2|X|^2|s|^2} , \tag{3.13}$$

$$\langle T_L\rangle=(\epsilon_1-\epsilon_0)X , \tag{3.14}$$

$$\langle T_{NL}\rangle=\frac{\alpha X^2|X|^2|s|^2}{1+(a+\beta)|\mathbf{E}_0|^2|X|^2|s|^2} . \tag{3.15}$$

Equations (3.12)–(3.15) hold good for a single nonlinear sphere inside the host medium. Equations (3.13) and (3.15) show that the averages of the nonlinear T matrix and its product with Green's function G are field-dependent quantities. The above results can be generalized to the case of a colloidal medium if we neglect such features as size dispersion, correlation effects arising due to multiple scattering, etc. Thus, for a small volume fraction f of the particles in the composite medium, the T matrix can be written as the sum of the T matrices of the individual particles. Thus, for a composite medium, we have

$$\langle GT_L\rangle=f(X-1) , \tag{3.16}$$

$$\langle GT_{NL}\rangle=-\frac{faX|X|^2|s|^2}{1+(a+\beta)|\mathbf{E}_0|^2|X|^2|s|^2} , \tag{3.17}$$

$$\langle T_L\rangle=f(\epsilon_1-\epsilon_0)X , \tag{3.18}$$

$$\langle T_{NL}\rangle=\frac{f\alpha X^2|X|^2|s|^2}{1+(a+\beta)|\mathbf{E}_0|^2|X|^2|s|^2} . \tag{3.19}$$

Now, we apply the results obtained in this section to find the effective-medium parameters derived in Sec. II. Equation (2.21) can be written, using Eqs. (3.16) and (3.18), as

$$\bar{\epsilon}=\epsilon_0+\frac{f(\epsilon_1-\epsilon_0)X}{1+f(X-1)} . \tag{3.20}$$

Equation (3.20) is a standard Maxwell-Garnett result for a linear composite. From Eq. (2.22) we obtain, after using Eqs. (3.17) and (3.19),

$$\frac{\bar{A}P_0}{1+\bar{B}Q_0|\mathbf{E}_0|^2}=\frac{[f\alpha X^2|X|^2+f(\bar{\epsilon}-\epsilon_0)aX|X|^2]|s|^2}{1+(a+\beta)|\mathbf{E}_0|^2|X|^2|s|^2} . \tag{3.21}$$

The quantities P_0 and Q_0 given by Eqs. (2.19) and (2.20) can be simplified after some algebraic manipulations into the following forms by using the results of this section:

$$P_0=[1+f(X-1)]|1+f(X-1)|^2\times\left[1-\frac{f}{P}(1-s)\right]\left|1-\frac{f}{P}(1-s)\right|^2 , \tag{3.22}$$

$$Q_0=|1+f(X-1)|^2\left|1-\frac{f}{P}(1-s)\right|^2 , \tag{3.23}$$

where s is given by Eq. (3.6), and we have defined P as

$$P=\frac{1}{X}[1+f(X-1)] . \tag{3.24}$$

Equation (3.21) is a single equation for two unknown parameters $\bar{A}$ and $\bar{B}$. We make the following choices for $\bar{A}$ and $\bar{B}$:

$$\bar{A}=\frac{f\alpha X^2|X|^2+f(\bar{\epsilon}-\epsilon_0)aX|X|^2}{[1+f(X-1)]|1+f(X-1)|^2} , \tag{3.25}$$

$$1+\bar{B}Q_0\frac{|\mathbf{E}_L|^2}{|X|^2|s|^2}=\frac{1}{|s|^2}[1+(a+\beta)|\mathbf{E}_L|^2]\times\left[1-\frac{f}{P}(1-s)\right]|1-\frac{f}{P}(1-s)|^2 . \tag{3.26}$$

In Eq. (3.26), Eq. (3.5) is used. It should be mentioned here that the final result for the effective-medium dielectric constant ϵ_{eff} does not depend upon any arbitrary choices of $\bar{A}$ and $\bar{B}$. Thus the procedure of determining ϵ_{eff} is consistent. The parameters $\bar{A}$ and $\bar{B}$ in Eqs. (3.25) and (3.26) are chosen such that the comparison of results with Ref. 11 for small-field values can be made directly in terms of $\bar{A}$. In fact, we shall see that $\bar{A}$ is nothing but the third-order susceptibility $4\pi\bar{\chi}^{(3)}$ in the expansion of ϵ_{eff} into a power series of $|\mathbf{E}|^2$.

Equation (3.25) can be put into the following form after

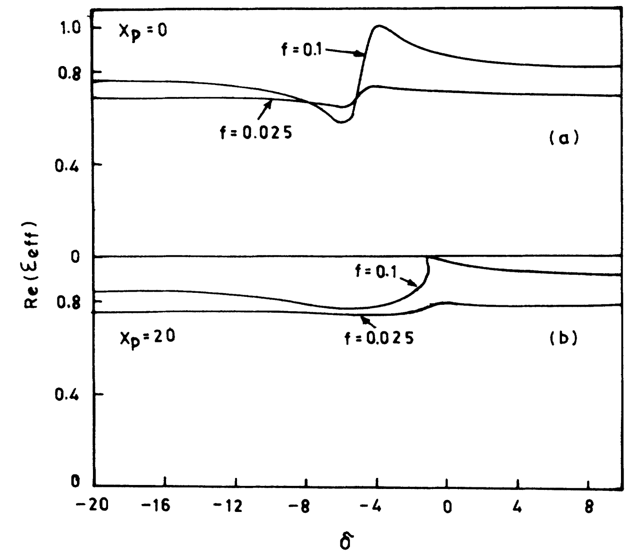

FIG. 1. (a) Plots of the real part of ϵ_{eff} vs δ for $x_p=0$, and $f=0.025$ and 0.1. Composite medium consists of CdS particles dispersed in space. (b) Same as in (a) except $x_p=20$.

using Eqs. (3.20) and (3.24):

$$\overline{A}=\frac{f\alpha}{P^2|P|^2} \ . \tag{3.27}$$

From Eq. (3.26), the quantity $\overline{B}|\mathbf{E}_{\text{L}}|^2$ can be found as

$$\overline{B}|\mathbf{E}_{\text{L}}|^2=\frac{1}{|P|^2}\left[1+a_0|\mathbf{E}_{\text{L}}|^2-\left|\frac{1+\beta|\mathbf{E}_{\text{L}}|^2}{1+a_0|\mathbf{E}_{\text{L}}|^2}\right|^2\right] , \tag{3.28}$$

where we have introduced a_0, given by

$$a_0=a+\beta-f\frac{a}{P} \ . \tag{3.29}$$

In a composite medium, when more than one spherical particle is embedded into the host medium, the local field $\mathbf{E}_{\text{L}}$ of a single particle is not the same as the propagating Maxwell field $\langle \mathbf{E}\rangle$. The relation between $\langle \mathbf{E}\rangle$ and $\mathbf{E}_{\text{L}}$ can be obtained by using Eqs. (2.13), (3.5), (3.16), and (3.17), and is given by

$$\langle \mathbf{E}\rangle=P\mathbf{E}_{\text{L}}\left[\frac{1+a_0|\mathbf{E}_{\text{L}}|^2}{1+\beta|\mathbf{E}_{\text{L}}|^2}\right] . \tag{3.30}$$

For a single particle in the host medium $f\to 1$, and from Eq. (3.30), we see that $\langle \mathbf{E}\rangle\to\mathbf{E}_{\text{L}}$. For $f\to 0$, $\langle \mathbf{E}\rangle\to\mathbf{E}_0$, given by Eq. (3.2). Now, we calculate the effective-medium dielectric constant ϵ_{eff} using Eq. (2.15). Writing $1+\overline{B}|\mathbf{E}|^2$ as $1+\overline{B}|\mathbf{E}_{\text{L}}|^2|\mathbf{E}|^2/|\mathbf{E}_{\text{L}}|^2$ and using relations

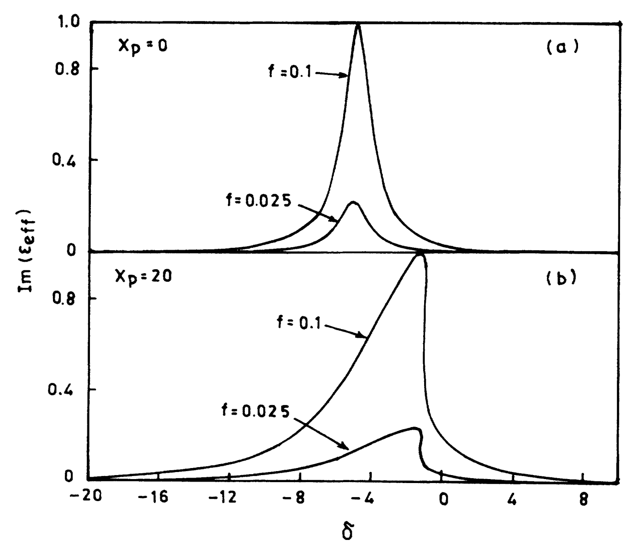

FIG. 2. (a) Plots of the imaginary part of ϵ_{eff} vs δ for $x_p=0$, and $f=0.025$ and 0.1. Composite medium same as in Fig. 1. (b) Same as in (a) except $x_p=20$.

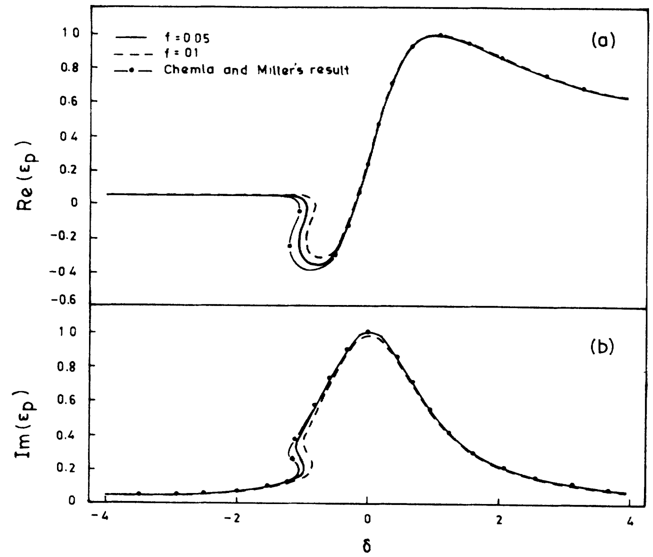

FIG. 3. (a) Plots of the real part of ϵ_p vs δ for $f=0.05$ and 0.1, and $x_p=20$. A curve from Fig. 2 of Ref. 16 for $y=20$ is also included for comparison. (b) Plots of the imaginary part of ϵ_p vs δ for the same values of parameters as in (a).

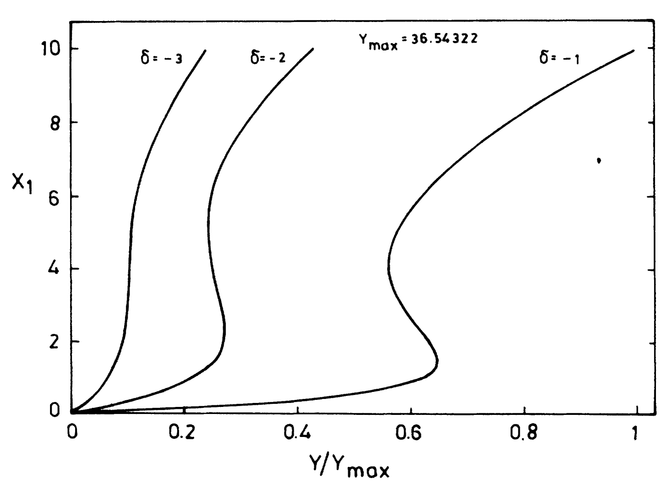

FIG. 4. Plots of x_1 vs y for $\delta=-1$, -2, and -3. Composite medium same as in Fig. 1.

(3.28) and (3.30) into Eq. (2.15), we obtain

$$\epsilon_{\text{eff}}=\bar{\epsilon}+\frac{\bar{A}|P|^2|\mathbf{E}_{\text{L}}|^2}{1+a_0|\mathbf{E}_{\text{L}}|^2}\ , \tag{3.31}$$

where again $\mathbf{E}_{\text{L}}$ is related to $\langle \mathbf{E} \rangle$ by Eq. (3.30). It can be easily verified that for $f \to 0$, $\epsilon_{\text{eff}} \to \epsilon_0$ (linear host medium), and for $f \to 1$, $\epsilon_{\text{eff}} \to \epsilon_1+\alpha|\mathbf{E}|^2/1+\beta|\mathbf{E}|^2$ (single particle). For small-field values, Eq. (3.31) can be expanded into a power series of $|\mathbf{E}|^2$. The result for $\beta=0$ is

$$\epsilon_{\text{eff}}=\bar{\epsilon}+4\pi\bar{\chi}^{(3)}|\mathbf{E}|^2+4\pi\bar{\chi}^{(5)}|\mathbf{E}|^4+4\pi\bar{\chi}^{(7)}|\mathbf{E}|^6+4\pi\bar{\chi}^{(9)}|\mathbf{E}|^8+\cdots\ , \tag{3.32}$$

where

$$4\pi\bar{\chi}^{(3)}=\bar{A}\ , \tag{3.33}$$

$$4\pi\bar{\chi}^{(5)}=-\bar{A}\eta_0\ , \tag{3.34}$$

$$4\pi\bar{\chi}^{(7)}=\bar{A}(\eta_1+\eta_0^2)\ , \tag{3.35}$$

$$4\pi\bar{\chi}^{(9)}=-\bar{A}(\eta_2+2\eta_1\eta_0+\eta_0^3)\ , \tag{3.36}$$

with

$$\eta_0=\frac{1}{|P|^2}(2a_0+a_0^*)\ , \tag{3.37}$$

$$\eta_1=\frac{1}{|P|^4}[a_0^2+(a_0^*)^2+|a_0|^2]\ , \tag{3.38}$$

$$\eta_2=\frac{1}{|P|^6}[a_0^3+a_0^2a_0^*+a_0(a_0^*)^2+(a_0^*)^3]\ . \tag{3.39}$$

Equations (3.33)–(3.39) are consistent with the perturbation approach to the T-matrix formulation given by Agarwal and Dutta Gupta.[11] It should be noted here that if we replace ϵ_1 by ϵ_p in Eq. (3.20), $\bar{\epsilon}$ becomes ϵ_{eff} given by Eq. (3.31).

IV. RESONANCE BEHAVIOR OF ϵ_{eff} AND INTRINSIC OPTICAL BISTABILITY

In this section we study the resonance behavior of the effective-medium dielectric constant ϵ_{eff} and optical bistability of local field $\mathbf{E}_{\text{L}}$ with respect to the applied field $\mathbf{E}_0$ and Maxwell field $\mathbf{E}$ in the absence of external feedback. We consider the nonlinear composite medium made up of spherical particles of CdS dispersed in space. For this medium ϵ_p is given by[16]

$$\epsilon_p=\epsilon_\infty+\beta'\frac{\delta+i}{1+\delta^2+(|\mathbf{E}_{\text{L}}|^2/I_s)}\ , \tag{4.1}$$

where ϵ_∞ is the dielectric constant of the background, $\delta=(\Omega-\omega)/\Gamma$ is the normalized detuning, ω is the optical frequency, Ω is the resonance frequency, Γ is the width of resonance, β' is the resonance contribution to the dielectric constant, and I_s is the saturation intensity. From Eq. (4.1) we can write

$$\epsilon_1=\epsilon_\infty+\beta'\frac{\delta+i}{1+\delta^2}\ , \tag{4.2}$$

$$\alpha=-\frac{\beta'(\delta+i)}{I_s(1+\delta^2)^2}\ , \tag{4.3}$$

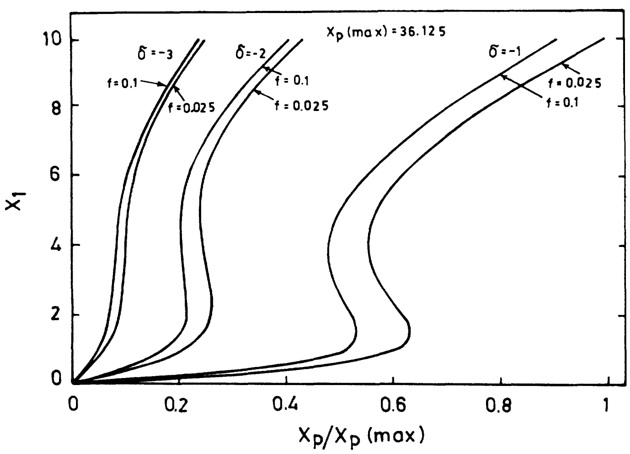

FIG. 5. Plots of x_1 vs x_p for $\delta=-1$, -2, and -3, and $f=0.025$ and 0.1.

$$\beta=\frac{1}{I_s(1+\delta^2)}\ . \tag{4.4}$$

For CdS particles, we take[16] $\epsilon_\infty=6$, $\Gamma=0.4$ meV, $\Omega=2.555$ eV, and $\beta'=40$. In Figs. 1 and 2, we have plotted the frequency dependence of real and imaginary parts of ϵ_{eff} for $f=0.1$ and 0.025, and $x_p=|\langle \mathbf{E} \rangle|^2/I_s=0$ and 20 using Eqs. (3.30) and (3.31). It can be noticed that for $x_p=20$ the peaks of the curves have red-shifted compared with the curves for $x_p=0$. This feature is similar to that observed in a nonlinear oscillator model.[18] Also, increasing the values of the volume fraction f increases effective absorption of the medium. The peak values of the real parts of ϵ_{eff} also increase with increasing f. In Fig. 3, we have plotted the frequency dependence of real and imaginary parts of ϵ_p for $f=0.1$ and 0.05, and $x_p=20$ using Eqs. (4.1) and (3.30). The kinks representing the bistable behaviors of real and imaginary parts of ϵ_p with respect to δ are clearly seen. Similar results were also obtained by Chemla and Miller[16] for a single spherical particle in vacuum. Their plots are also included for comparison.

Next, we shall discuss intrinsic optical bistability. To

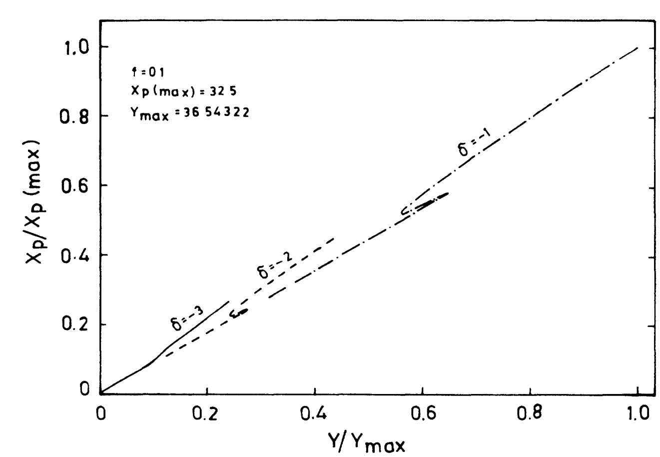

FIG. 6. Plots of x_p vs y for $\delta=-1$, -2, and -3, and $f=0.1$.

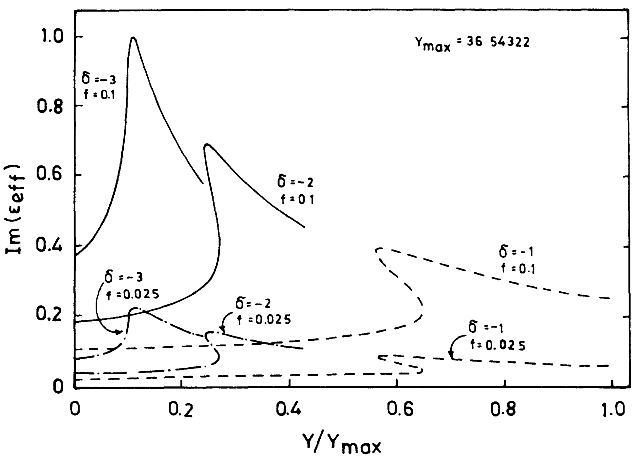

FIG. 7. (a) Plots of the imaginary part of ϵ_{eff} vs y for $\delta=-1$, -2, and -3, and $f=0.025$ and 0.1.

observe optical bistability one generally requires the combination of strong optical nonlinearity and a feedback mechanism. Most optical bistable devices use an external cavity to provide the necessary feedback. However, internal feedback in some cases without mirrors can also be possible. The generation of internal feedback is possible if the intrinsic properties of the nonlinear medium are strongly modified by incident fields. In the composite media, the local-field effects in the particles arising from dielectric confinement gives rise to an internal feedback. Together with enhanced nonlinearity due to the excitonic (surface plasmon) resonances in semiconductor (metal) particles, this internal feedback mechanism produces the bistable behavior for the local field $\mathbf{E}_L$ with respect to the applied field $\mathbf{E}_0$ and also with the propagating Maxwell field $\mathbf{E}$. Correspondingly, one also has the bistable behavior in real and imaginary parts of ϵ_p and ϵ_{eff}. In Fig. 4, we have shown $x_1=|\mathbf{E}_L|^2/I_s$ versus $y=|\mathbf{E}_0|^2/I_s$ plots for $\delta=-1$, -2, and -3. For $\delta=-3$ the detuning is too large and the cure shows only kink. In Fig. 5, x_1 versus x_p plots are shown for $\delta=-1$, -2, and -3, and $f=0.1$ and 0.025. It may be noticed that for larger values of f, the bistability threshold reduces. In Fig. 6, x_p versus y plots for $f=0.1$ and $\delta=-1$, -2, and -3 are shown. Figure 7 shows plots of imaginary parts of ϵ_{eff} versus y for $\delta=-1$, -2, and -3, and $f=0.1$ and 0.025. In all the figures 1–7, the scales are normalized to unity.

V. CONCLUSIONS

In conclusion, we have developed an effective-medium theory for nonlinear spherical particles embedded in a linear host medium by using T-matrix approach. Our results are exact in the sense that for small volume fractions f, the effective-medium dielectric constant includes the contributions from all higher odd-order nonlinear susceptibilities. For small field values, our results reproduce previously obtained results by Agarwal and Dutta Gupta[11] for third- and fifth-order nonlinear susceptibilities of the composite medium. The averaged nonlinear T matrix involved in our calculations is field dependent. The results were applied to a model composite medium made up of CdS particles dispersed in space. The resonance peak of the imaginary part of effective dielectric constant is found to be red-shifted. The amount of red shift increases with increasing the values of x_p. This feature is similar to that observed in a nonlinear oscillator model.[18] Increasing the value of the volume fraction f increases the absorption inside the medium.

We have also discussed the intrinsic optical bistability. The local-field effects in particles arising from dielectric confinement gives rise to an internal feedback. Together with enhanced nonlinearity due to resonances, this internal feedback mechanism produces bistability for local field inside the particle with respect to the applied field and also the propagating Maxwell field. This leads to a bistable behavior in effective dielectric constant also.

The propagation of intense light beams through a composite medium was addressed earlier under certain simplifying assumptions in both the stationary and nonstationary regimes.[19–22] Recently, Haus *et al.*[17] solved the wave propagation equation numerically in a composite medium. They found that there exists a boundary separating a low-absorbing regime from a high-absorbing regime in the material. This also shows the similarity of this problem to the nonlinear oscillator model.[18] Similar results have also been obtained by Li *et al.*[23] These authors take into account the saturation of the two-level system in their expression for the dielectric constant of the particle which is also taken into account in this work [see Eq. (4.1)], whereas in Ref. 17 this saturation of the two-level system of the particle was ignored. Furthermore, they use the self-consistent field approximation and take the effective dielectric function based on this approximation, which is better than the approximate form of the Maxwell-Garnett effective dielectric function used in Ref. 17. Moreover, in Ref. 17 the wave-propagation equation was solved under the slowly varying envelope approximation (SVEA), whereas besides obtaining the results with the SVEA, Li *et al.*[23] also solve the wave-propagation equation exactly as well as under the mean-field approximation. In all these cases the qualitative results are the same as those obtained in Ref. 17. The results obtained in Refs. 17 and 23 are based on numerical solution of the wave equation. In the present work, the exact analytical expression for the nonlinear effective dielectric constant ϵ_{eff} has been obtained within the framework of Maxwell-Garnett formulation.

Finally, it should be mentioned in concluding that the results of Chemla and Miller,[16] and Leung[15] are for a single particle embedded in a linear host medium, whereas the results presented in Figs. 1–7 in this paper are for multiple particles in the host medium. The results of this paper are accurate in all the orders of electric field while assuming the volume fraction f to be small. The results of Ref. 11 are accurate up to the fifth-order in the electric field while assuming f small, whereas the results of Refs. 15 and 16 are good for all field values, but only for a single particle in the linear host medium.

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[1]See, for example, R. Landauer, in *Electrical Transport and Optical Properties of Inhomogeneous Media* (Ohio State University, 1977), Proceedings of the First Conference on the Electrical Transport and Optical Properties of Inhomogeneous Media, AIP Conf. Proc. No. 40, edited by J. C. Garland and D. B. Tanner (AIP, New York, 1978), p. 2.
[2]E. Gubernatis, in *Electrical Transport and Optical Properties of Inhomogeneous Media* (Ohio State University, 1977) (Ref. 1), p. 84.
[3]I. Webman, J. Jortner, and M. H. Cohen, Phys. Rev. B **15**, 5712 (1977).
[4]W. Lamb, D. M. Wood, and N. W. Ashcroft, Phys. Rev. B **21**, 2248 (1980).
[5]G. S. Agarwal and R. Inguva, Phys. Rev. B **30**, 6108 (1984).
[6]K. C. Rustagi and C. Flytzanis, Opt. Lett. **9**, 344 (1984).
[7]D. Ricard, P. Roussignol, and C. Flytzanis, Opt. Lett. **10**, 511 (1985).
[8]F. Hache, D. Ricard, and C. Flytzanis, J. Opt. Soc. Am. B **3**, 1647 (1986).
[9]P. Roussignol, D. Ricard, J. Lukasik, and C. Flytzanis, J. Opt. Soc. Am. B **4**, 5 (1987).
[10]D. Ricard, in *Nonlinear Optics: Materials and Devices,* edited by C. Flytzanis and J. L. Oudar (Springer, Berlin, 1986).
[11]G. S. Agarwal and S. Dutta Gupta, Phys. Rev. A **11**, 5678 (1988).
[12]J. W. Haus, R. Inguva, and C. M. Bowden, Phys. Rev. A **40**, 5729 (1989).
[13]J. W. Haus, N. Kalyaniwalla, R. Inguva, M. Bloemer, and C. M. Bowden, J. Opt. Soc. Am. B **6**, 797 (1989).
[14]L. K. H. Van Beek, *Progress in Dielectrics,* edited by J. B. Birks (CRC, Cleveland, 1967), Vol. 7, p. 69.
[15]K. M. Leung, Phys. Rev. A **33**, 2461 (1986).
[16]D. S. Chemla and D. A. B. Miller, Opt. Lett. **11**, 522 (1986).
[17]J. W. Haus, N. Kalyaniwalla, R. Inguva, and C. M. Bowden, J. Appl. Phys. **65**, 1420 (1989).
[18]J. W. Haus, L. Wang, M. Scalora, and C. M. Bowden, Phys. Rev. A **38**, 4043 (1988).
[19]G. B. Al'tshuler, V. S. Ermolaev, K. I. Krylov, and A. M. Prokhorov, J. Opt. Soc. Am. B **3**, 660 (1986).
[20]N. C. Kothari and C. Flytzanis, Opt. Lett. **11**, 806 (1986). In all the figures of this paper the values of z' should be twice the values mentioned.
[21]N. C. Kothari and C. Flytzanis, Opt. Lett. **12**, 492 (1987).
[22]N. C. Kothari, J. Opt. Soc. Am. B **5**, 2348 (1988). In Fig. 4 of this paper the values of z' should be twice the values mentioned.
[23]Y. Q. Li, C. C. Sung, R. Inguva, and C. M. Bowden, J. Opt. Soc. Am. B **6**, 814 (1989).