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EFFECTIVE NONLINEAR COEFFICIENT OF STRONGLY NONLINEAR SPHERICAL DIELECTRIC COMPOSITES

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นายจตุพร ทองศรี: สัมประสิทธิ์ความไม่เชิงเส้นยังผลของไดอิเล็กทริกคอมโพสิตทรงกลมไม่เชิงเส้นอย่างแรง (EFFECTIVE NONLINEAR COEFFICIENT OF STRONGLY NONLINEAR SPHERICAL DIELECTRIC COMPOSITES) อ. ที่ปรึกษา: รศ. ดร.มยุรี เนตรนภิส, 80 หน้า. ISBN 974-53-1040-9.

งานวิจัยนี้ได้ขยายงานของ ยู และ หยวน (1996) ซึ่งศึกษาการตอบสนองทางไฟฟ้าของไดอิเล็กทริกคอมโพสิตทรงกลมไม่เชิงเส้นอย่างแรงโดยใช้วิธีการแยกส่วนซึ่งคอมโพสิตนี้ประกอบด้วยไดอิเล็กทริกทรงกลมไม่เชิงเส้นอย่างแรงฝังกระจายแบบสุ่มในตัวกลางไดอิเล็กทริกไม่เชิงเส้นอย่างแรงอีกชนิดหนึ่ง ซึ่งมีสัมประสิทธิ์ความไม่เชิงเส้นแตกต่างกัน เราได้ประยุกต์ทฤษฎีตัวกลางยังผลในการจำลองทางทฤษฎีและคำนวณขอบเขตของค่าสัมประสิทธิ์ความไม่เชิงเส้นยังผล (χ_e) ของคอมโพสิตนี้สำหรับค่าสัดส่วนโดยปริมาตรของสารฝังกระจายเป็นค่าใดๆ จากผลการคำนวณของเราแสดงว่าค่าขอบเขตบนและขอบเขตล่างของสัมประสิทธิ์ความไม่เชิงเส้นมีช่องว่างเพิ่มขึ้นเมื่อเพิ่มความต่างกันระหว่างสัมประสิทธิ์ความไม่เชิงเส้นของอนุภาคสารฝังกระจายและของตัวกลาง ถ้าคอมโพสิตมีตัวกลางที่มีค่าสัมประสิทธิ์ความไม่เชิงเส้นสูงค่า χ_e มีค่ามากกว่ากรณีที่สารค่าสัมประสิทธิ์ความไม่เชิงเส้นสูงเป็นอนุภาคฝังกระจายสำหรับคอมโพสิตที่มีสัดส่วนโดยปริมาตรเหมือนกัน ค่า χ_e ของเราที่คำนวณโดยทฤษฎีตัวกลางยังผลใช้ได้ทั่วไปกว่างานวิจัยของ ยู และ หยวนที่มีการประมาณให้สารฝังกระจายมีค่าสัดส่วนโดยปริมาตรน้อยมาก และการคำนวณโดยใช้ทฤษฎีตัวกลางยังผลนี้สอดคล้องกับผลงานของ ยู และ หยวน เมื่อสัดส่วนของอนุภาคสารฝังกระจายมีค่าน้อยกว่า 0.1 นอกจากนี้เรายังได้คำนวณค่า χ_e โดยใช้วิธีการแปรผันแบบพื้นฐานเพื่อยืนยันผลการคำนวณโดยใช้วิธีการแยกส่วนทั้งหมด และพบว่าผลของค่า χ_e ที่คำนวณโดยวิธีการแยกส่วนมีค่าน้อยกว่าผลที่ได้จากวิธีการแปรผันซึ่งสอดคล้องกับทางทฤษฎีที่ได้มีการคาดคะเนไว้โดย ยู สุย และ ลี (1996)

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KEY WORDS: EFFECTIVE NONLINEAR COEFFICIENT/ DECOUPLING TECHNIQUE/ VARIATIONAL METHOD/ DIELECTRIC COMPOSITES

JATUPORN THONGSEE: EFFECTIVE NONLINEAR COEFFICIENT OF STRONGLY NONLINEAR SPHERICAL DIELECTRIC COMPOSITES. THESIS ADVISOR: ASSOC. PROF. MAYUREE NATENAPIT, PH.D., 80 pp. ISBN 974-53-1040-9.

This research is an extension of the work of Yu and Yuen (1996) in studying the electric field response of strongly nonlinear dielectric composites by using the decoupling technique. These composites consist of spherical strongly nonlinear dielectric inclusions randomly embedded in different strongly nonlinear dielectric host media. The effective medium theory (EMT) is applied for theoretical modeling and bounds of the effective nonlinear coefficient (χ_e) of the composite are determined for arbitrary inclusion packing fractions. The results show the lower and upper bounds which the gap increases with increasing the contrast between the nonlinear coefficients of the inclusion and host medium. If the composite has material of higher nonlinear coefficient being the host medium instead of the inclusions, the higher χ_e is obtained for the composite of the same packing fractions. Our results of χ_e based on the EMT are more general than those of Yu and Yuen which are valid only at low inclusion packing fraction limit. The EMT results agree very well with the results of Yu and Yuen at inclusion packing fractions less than 0.1. Moreover, we also apply the simple variational method to calculate χ_e in order to confirm all decoupling technique results. It is found that the decoupling technique results are less than those obtained by using the variational method, satisfying the theoretical prediction of Yu, Hui and Lee (1996).

Department .....Physics.....	Student's signature ....จตุพร ทองสี.......
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List of Symbols

α	atomic polarizability
ε_0	permittivity of free space
ε	permittivity or linear coefficient
ε_r	relative permittivity or dielectric constant
ε_e	effective linear coefficient of composite
χ'	electric susceptibility
χ_α	nonlinear coefficient of material α
χ_e	effective nonlinear coefficient of composite
ρ_f	free charge density
φ	electric potential
v_α	volume packing fraction of material α
c'	ratio between inclusion volume and cell volume
$W[\varphi]$	energy functional
W_s	surface energy
$\overrightarrow{p}$	electric dipole moment
$\overrightarrow{E}$	electric field
$\overrightarrow{D}$	electric displacement
$\overrightarrow{P}$	polarization

Chapter I

Introduction

Nowadays, the physics of nonlinear composites has attracted much attention because of their applications in engineering and physics [1, 2, 3]. The optical composite materials, one type of nonlinear composites, play important roles in developing photonic devices [4], laser [5], and optoelectronic technologies [6]. Therefore, it is useful to study the electric field response of strongly nonlinear composite.

The effective response of nonlinear dielectric composites obey a local electric displacement - field ($\overrightarrow{D} - \overrightarrow{E}$) relation of the form $\overrightarrow{D} = \varepsilon\overrightarrow{E} + \chi\left|\overrightarrow{E}\right|^2\overrightarrow{E}$. The strongly nonlinear behavior occurs when the second term ($\chi\left|\overrightarrow{E}\right|^2\overrightarrow{E}$) is much larger than the first term ($\varepsilon\overrightarrow{E}$), then the electric displacement can be written in terms of $\overrightarrow{D} = \chi\left|\overrightarrow{E}\right|^2\overrightarrow{E}$. Because the boundary-value problem of strongly nonlinear media is extremely difficult to solve. As the nonlinearity appears as the leading form of the behavior rather than correction to a predominant linear response, the conventional perturbation method fail. Nevertheless, substantial progress has been made with the aid of various approximate analytical methods and numerical methods over the past few years [7-14].

Blumenfeld and Bergman [7, 8] developed a small contrast expansion for the effective dielectric response of strongly nonlinear composites. Ponte Castaneda [9, 10] proposed a general variational procedure for establishing optimal bounds and estimates for the electric response of nonlinear composites in terms of the effective behavior of linear composites with identical structure. In 1992, Yu and

Gu [11] used the perturbation method to obtain the effective nonlinear coefficient for a small concentration of spherical inclusions embedded in a host medium but this method can not be used for strongly nonlinear composites.

The variational method (variational energy method) has been applied to various fields in science and engineering, as examples, such method has been applied to boundary-value problems in electrostatics, magnetostatics, and electric conduction. This method is suitable not only for weakly nonlinear composites but also for strongly nonlinear composites. Moreover, in 1994-95, Yu and Gu [12, 13] adopted a simple variational method to study the composite which consists of two different nonlinear media. In an attempt to extend the validity of the dilute-limit expression to larger volume fraction, Yu and Lee [14] used a self-consistency condition and Bruggeman-type effective medium approximation (EMA) for strongly nonlinear composites.

Recently, Janthon [15] applied the variational method to study the effective response of linear and nonlinear dielectric composites of spherical inclusions in the dilute limit. Next, Chaiprapa [16] applied the variational method to study the effective response of linear and nonlinear cylindrical dielectric composites and obtained the effective nonlinear coefficient (χ_e) for arbitrary inclusion packing fractions.

Furthermore, Yu and Yuen [17] applied the decoupling technique to strongly nonlinear composites of spherical inclusions in the dilute inclusion packing fraction by using the single inclusion model. They obtained an approximate results for the effective response which are compared with those of the variational approach. However, their results have a limit on the practical application because the single inclusion model is unsuitable in the determination on χ_e of the composites for arbitrary inclusion packing fractions.

In this research, the work of Yu and Yuen [17] is extended to arbitrary inclusion packing fractions. The effective medium theory (EMT) proposed by Hashin [18] is applied for theoretical modeling and studying the electric field response of strongly nonlinear spherical dielectric composites. Then, the effective nonlinear coefficients (χ_e) of the composites are determined for arbitrary inclusion packing fractions by using the decoupling technique. Our results based on the EMT and the work of Yu and Yuen are compared. Moreover, our results are also compared with those obtained by the variational method of which the results are reliable for arbitrary inclusion packing fractions, in order to determine the validity and reliability of the decoupling technique.

In Chapter 2, the details of the variational method and the decoupling technique are presented. In Chapter 3, the effective medium theory (EMT) was proposed by Hashin in studying of effective conductivities of two-phase composite materials is reviewed. In Chapter 4, the simple variational method is applied in solving electrostatic boundary value-problem of the strongly nonlinear composites and the effective nonlinear coefficients (χ_e) are determined. In Chapter 5, the same problems as calculated in chapter 4 are now determined by using the decoupling technique. Then both results are compared, in order to determine the validity and reliability of the decoupling technique. The last chapter is conclusions of this research.

Chapter II

Theoretical Background

In this chapter, the response of dielectric composites in an external electric field will be investigated. The methods which will be applied to study effective dielectric properties of nonlinear composites are the variational method and the decoupling technique. The details of both methods will be presented and applied to determine the nonlinear coefficients of nonlinear dielectric composites in Chapters 4 and 5.

2.1 Polarization

When a dielectric material is placed in an electric field, a slight displacement of the negative and positive charges of the dielectric's atoms or molecules occur and they behave like very small dipoles. The dielectric is said to be polarized when the dipoles exist. For example, a polarized atom of a dielectric material is represented by an electric dipole, i.e., a positive point charge (nucleus) and a negative charge representing the electrons, the two charges being separated by a small distance. When the atom is unpolarized, the cloud surrounds the nucleus symmetrically, as in Fig. (2.1), and the dipole moment is zero. When an external electric field $\overrightarrow{E}$ is applied, the electron cloud becomes slightly displaced or asymmetrical, as in Fig. (2.2), and the atom is polarized having a tiny dipole moment $\overrightarrow{p}$, which points in the same direction as $\overrightarrow{E}$. Typically, this dipole moment is approximately proportional to the field

$$\overrightarrow{p} = \alpha \overrightarrow{E}, \tag{2.1}$$

where α is atomic polarizability. Therefore, when the dielectric is polarized, a convenient measure of this effect is called polarization $\left(\overrightarrow{P}\right)$ which is dipole moment per unit volume [19].

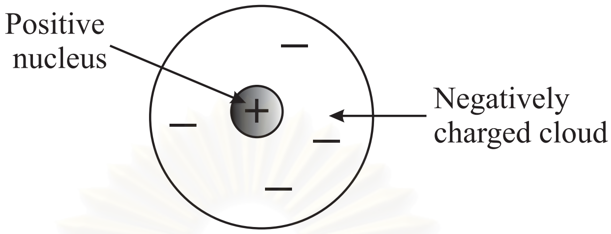

Figure 2.1: An unpolarized atom.

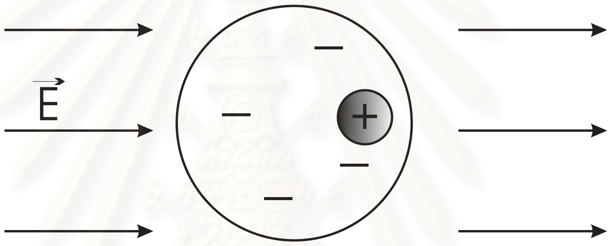

Figure 2.2: A polarized atom.

2.2 Dielectric Media

2.2.1 Linear Dielectrics

Consider the relation between electric displacement ($\overrightarrow{D}$) and polarization ($\overrightarrow{P}$)

$$\overrightarrow{D} = \varepsilon_0 \overrightarrow{E} + \overrightarrow{P}, \tag{2.2}$$

where ε_0 is called the permittivity of free space.

Generally, the dielectric materials in which $\overrightarrow{P}$ is proportional (in magnitude) and parallel (in direction) to $\overrightarrow{E}$, are said to be linear and isotropic. In case of $\overrightarrow{E}$ is not too strong, the dependence of $\overrightarrow{P}$ on $\overrightarrow{E}$ can be written as

$$\overrightarrow{P} = \varepsilon_0 \chi' \overrightarrow{E}, \tag{2.3}$$

where χ' is called the electric susceptibility which depends on the microscopic structure of the medium.

If $\overrightarrow{P}$ and $\overrightarrow{E}$ are related by Eq. (2.3), then the relation between $\overrightarrow{D}$ and $\overrightarrow{E}$ can be obtained by substituting Eq. (2.3) into Eq. (2.2) to give

$$\begin{aligned} \overrightarrow{D} &= (1+\chi')\,\varepsilon_0 \overrightarrow{E} \\ &= \varepsilon \overrightarrow{E}, \end{aligned} \tag{2.4}$$

where $\varepsilon \equiv \varepsilon_0(1+\chi')$ is called the permittivity of the material and

Therefore, the electric displacement is linearly proportional to the electric field in linear dielectric media.

2.2.2 Nonlinear Dielectrics

At large field intensities of about $10^6\ V/m$ or higher, deviation of relation (2.3) becomes noticeable [20], the non-linear effects of the materials are occurred. They arise from the interaction of the external electric fields $\overrightarrow{E}$, with the molecular dipole moment, which rotates those dipole and creates a polarization field $\overrightarrow{P}$. The polarization field is linearity dependant on the magnitude of the external fields so long as they are small, this linearity eventually breaks down and higher order terms are needed to describe the polarization field. The polarization in this case are given by [21]

$$\overrightarrow{P} = \varepsilon_0 \chi' \overrightarrow{E} + \varepsilon_0 \chi'^{(3)} \left|\overrightarrow{E}\right|^2 \overrightarrow{E} + \varepsilon_0 \chi'^{(5)} \left|\overrightarrow{E}\right|^4 \overrightarrow{E} + ..., \tag{2.5}$$

where χ', $\chi'^{(3)}$ and $\chi'^{(5)}$ are the nonlinear first, third and fifth order electric susceptibilities, respectively.

It must be noted that the series development of $\overrightarrow{P}$ contains only odd powers of $\overrightarrow{E}$, because a reversal of the direction of $\overrightarrow{E}$ lead to reversal of direction of $\overrightarrow{P}$,

$$\overrightarrow{P}(\overrightarrow{E}) = -\overrightarrow{P}(-\overrightarrow{E}). \tag{2.6}$$

If the polarization is nonlinear in the field strength, the dependence of the dielectric displacement $\overrightarrow{D}$ on the field strength will also be nonlinear,

$$\overrightarrow{D} = \varepsilon \overrightarrow{E} + \chi \left| \overrightarrow{E} \right|^2 \overrightarrow{E} + ...,$$

where ε and χ are called the linear and nonlinear coefficients, respectively.

The electric displacement $\overrightarrow{D}$ and electric field $\overrightarrow{E}$ relation of the form

$$\overrightarrow{D} = \varepsilon \overrightarrow{E} + \chi \left| \overrightarrow{E} \right|^2 \overrightarrow{E}, \tag{2.7}$$

will be considered. From Eq. (2.7), $\chi \left| \overrightarrow{E} \right|^2 \ll \varepsilon$ is the case of weakly nonlinear dielectrics and strongly nonlinear behavior occurs when $\chi \left| \overrightarrow{E} \right|^2 \gg \varepsilon$ in which the electric displacement can be written as

$$\overrightarrow{D} = \chi \left| \overrightarrow{E} \right|^2 \overrightarrow{E}. \tag{2.8}$$

This equation indicates that the electric displacement is proportional to the electric field to the third power and will be used to describe strongly nonlinear composites in this research

2.2.3 Strongly Nonlinear Dielectric Composites

Consider a two-phase composite [22] with strongly nonlinear property which consists of two dielectrics with nonlinear coefficients χ_1 and χ_2, as shown in Fig. 2.4.

This composite is replaced by a homogeneous and isotropic medium of effective nonlinear coefficient (χ_e), which is an unknown to be specified later. Fig. 2.4 shows the model, the composite that represents the original one and called the effective strongly nonlinear composite.

The effective nonlinear coefficient (χ_e) is defined such that the energy integral of the original composite has to be equal to the energy of the effective

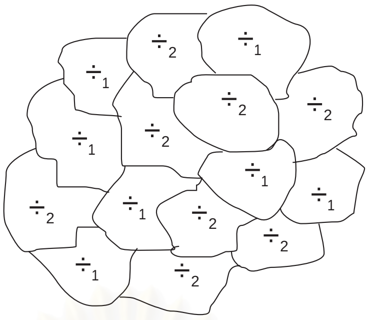

Figure 2.3: A two-phase of strongly nonlinear dielectric composite.

Figure 2.4: The effective strongly nonlinear composite.

nonlinear composite. That is

$$\chi_e E_0^4 V = \int_V \overrightarrow{D}(x) \cdot \overrightarrow{E}(x) dV, \tag{2.9}$$

where $\overrightarrow{E}_0$ is a uniform applied electric field and V is the composite volume.

2.3 Basic Equations in Electrostatics

2.3.1 Laplace's Equation

We now consider the Maxwell equations in electrostatics of dielectric media [23]:

$$\overrightarrow{\nabla} \cdot \overrightarrow{D} = \rho_f \tag{2.10}$$

and

$$\overrightarrow{\nabla} \times \overrightarrow{E} = 0, \text{ or } \overrightarrow{E} = -\overrightarrow{\nabla}\varphi, \tag{2.11}$$

where ρ_f, φ are free charge density and electric potential, respectively.

Replacing Eq. (2.4) into Eq. (2.10), gives

$$\overrightarrow{\nabla} \cdot (\varepsilon \overrightarrow{E}) = \rho_f. \tag{2.12}$$

Substituting $\overrightarrow{E}$ from Eq. (2.11), Eq. (2.12) is replaced by

$$\overrightarrow{\nabla}^2 \varphi = -\frac{\rho_f}{\varepsilon}. \tag{2.13}$$

If $\rho_f = 0$ in some region of the media, then Eq. (2.13) becomes

$$\overrightarrow{\nabla}^2 \varphi = 0. \tag{2.14}$$

Eq. (2.14) is called Laplace's equation and replaced the basic equations in linear dielectric media.

2.3.2 Nonlinear Partial Differential Equations

In case of nonlinear dielectric media, Eq. (2.7) is substituted into the Maxwell equation (2.10), for the case of $\rho_f = 0$, hence

$$\overrightarrow{\nabla} \cdot (\varepsilon \overrightarrow{E} + \chi \left| \overrightarrow{E} \right|^2 \overrightarrow{E}) = 0. \tag{2.15}$$

From $\overrightarrow{E} = -\overrightarrow{\nabla} \varphi$, we get

$$\overrightarrow{\nabla} \cdot (\varepsilon \overrightarrow{\nabla} \varphi + \chi \left| \overrightarrow{\nabla} \varphi \right|^2 \overrightarrow{\nabla} \varphi) = 0. \tag{2.16}$$

For strongly nonlinear dielectric media ($\chi \left| \overrightarrow{E} \right|^2 \gg \varepsilon$), then Eq. (2.16) becomes

$$\overrightarrow{\nabla} \cdot (\chi \left| \overrightarrow{\nabla} \varphi \right|^2 \overrightarrow{\nabla} \varphi) = 0. \tag{2.17}$$

Eqs. (2.16) and (2.17) are the basic equations in nonlinear dielectric media and they are nonlinear partial differential equations which can not be solved exactly. According to the complication of these equations, several methods are applied, these include Perturbation Method [11], Variational Method [12, 13, 14, 15, 16, 24, 25] and Decoupling Techniques [17, 26, 27], but the suitable methods depend on the nature of each problem.

2.3.3 Boundary conditions

The boundary conditions are essential to specify in solving for electric potentials in the composite. The first boundary condition on $\overrightarrow{E}$ at the interface between different media, any surface separating two regions, is that the tangential component of $\overrightarrow{E}$ at any surface is continuous [28],

$$\overrightarrow{E}_{1t} = \overrightarrow{E}_{2t}, \tag{2.18}$$

where $\overrightarrow{E}_{1t}$ and $\overrightarrow{E}_{2t}$ are the tangential components of $\overrightarrow{E}$ in media 1 and 2 evaluated at the interface, respectively. The second, the normal component of $\overrightarrow{D}$ is continuous at the interface,

$$\overrightarrow{D}_{1n} = \overrightarrow{D}_{2n}, \tag{2.19}$$

where $\overrightarrow{D}_{1n}$ and $\overrightarrow{D}_{2n}$ are the normal components of $\overrightarrow{D}$ in media 1 and 2 evaluated at the interface, respectively.

These boundary conditions will be used to determine φ and $\overrightarrow{E}$ in media 1 and 2 of the two-phase composite in Chapters 3 and 4.

2.4 Variational Method

Variational method (or variational energy method) has been applied to various fields in science and engineering. As examples, such method has been applied to boundary-value problems in electrostatics, magnetostatics, and electric conduction. In previous works, Janthon [15] applied the variational method to study the bulk effective response of linear and nonlinear dielectric composites of spherical inclusions in the dilute limit. Recently, Chaiprapa [16] applied the variational method to study the bulk effective response of linear and nonlinear cylindrical dielectric composites and obtained the effective nonlinear coefficient (χ_e) for arbitrary inclusion packing fractions. According to the importance of variational method; consequently, in this section, the variational method of nonlinear dielectric composites will be reviewed and applied to determine the effective nonlinear coefficient in Chapter 4.

Consider a class of nonlinear dielectric composites that obey a displacement-field response of the form $\overrightarrow{D} = \varepsilon\overrightarrow{E} + \chi\left|\overrightarrow{E}\right|^2\overrightarrow{E}$ where the linear and nonlinear coefficients are ε and χ, respectively. The governing equations $\overrightarrow{\nabla}\cdot\overrightarrow{D} = 0$ and $\overrightarrow{\nabla}\times\overrightarrow{E} = 0$ lead to the following nonlinear partial differential equation,

$$\overrightarrow{\nabla}\cdot[\varepsilon(x)\overrightarrow{\nabla}\varphi(x) + \chi(x)\left|\overrightarrow{\nabla}\varphi(x)\right|^2\overrightarrow{\nabla}\varphi(x)] = 0, \tag{2.20}$$

as shown in Eq. (2.20), where $\varphi(x)$ is the electric potential and $\overrightarrow{E} = -\overrightarrow{\nabla}\varphi$. Together with the boundary conditions of the continuity of the tangential component of $\overrightarrow{E}$ and the normal component of $\overrightarrow{D}$ on the interface. Eq. (2.20) forms a nonlinear partial differential equation that cannot be solved exactly. Nevertheless, one can invoke the variational principle by minimizing the energy functional [15, 16],

$$W[\varphi] = \frac{1}{2}\int_V \varepsilon(x)\left|\overrightarrow{\nabla}\varphi(x)\right|^2 dV + \frac{1}{4}\int_V \chi(x)\left|\overrightarrow{\nabla}\varphi(x)\right|^4 dV, \tag{2.21}$$

with respect to an arbitrary variation $\delta\varphi(x)$ away from the solution of Eq. (2.20), provide that $\delta\varphi$ vanishes at the interface. For convenience in subsequent discussion, we denote the linear and nonlinear parts of the energy functional by $W_2[\varphi]$ and $W_4[\varphi]$, respectively,

$$W_2[\varphi] = \int_V \varepsilon(x)\left|\overrightarrow{\nabla}\varphi(x)\right|^2 dV, \tag{2.22}$$

and

$$W_4[\varphi] = \int_V \chi(x)\left|\overrightarrow{\nabla}\varphi(x)\right|^4 dV, \tag{2.23}$$

so that $W[\varphi] = \frac{1}{2}W_2[\varphi] + \frac{1}{4}W_4[\varphi]$. When the minimum condition is satisfied by the solution $\widetilde{\varphi}$, then the effective energy function Eq. (2.21) can be obtained,

$$\widetilde{W} = \frac{1}{2}\int_V \varepsilon(x)\left|\overrightarrow{\nabla}\widetilde{\varphi}(x)\right|^2 dV + \frac{1}{4}\int_V \chi(x)\left|\overrightarrow{\nabla}\widetilde{\varphi}(x)\right|^4 dV. \tag{2.24}$$

It is important to choose a proper trial potential function φ, evaluate the integral in Eq. (2.21), minimize it with respect to φ, and generate explicit formulas for the effective linear and nonlinear coefficients in Chapter 4.

2.5 Decoupling Technique

In this section, the review of the decoupling technique originally developed by Stroud and Wood [27] is presented and will be employed to study the effective response of strongly nonlinear composites in Chapter 5. Consider a class of strongly nonlinear composites which obeys electric displacement-field relation of the form $\overrightarrow{D} = \chi \left|\overrightarrow{E}\right|^2 \overrightarrow{E}$. The nonlinear coefficient χ takes on different values in materials 1 and 2 described by χ_1 and χ_2, respectively. The governing equations for electric displacement $\overrightarrow{\nabla} \cdot \overrightarrow{D} = 0$ and $\overrightarrow{\nabla} \times \overrightarrow{E} = 0$ lead to the following differential equation,

$$\overrightarrow{\nabla} \cdot [\chi(x) \left|\overrightarrow{\nabla}\varphi(x)\right|^2 \overrightarrow{\nabla}\varphi(x)] = 0, \tag{2.25}$$

which is special case of Eq. (2.16) for the first term is negligible. It is convenient to avoid the complication in solving Eq. (2.25) by using decoupling technique. The effective strongly nonlinear dielectric composite which was defined in Fig. 2.4 with the effective nonlinear coefficient (χ_e) given by Eq. (2.9), will be considered.

When a trial electric field $\widetilde{E}(x)$ is used to generate an approximate formula for the effective nonlinear coefficient (χ_e) of Eq. (2.9),

$$\chi_e E_0^4 V = \int_V \chi(x) \left|\widetilde{E}(x)\right|^4 dV, \tag{2.26}$$

or

$$\begin{aligned} \chi_e &= \frac{1}{E_0^4 V}[\int_{V_1} \chi_1(x) \left|\widetilde{E_1}(x)\right|^4 dV + \int_{V_2} \chi_2(x) \left|\widetilde{E_2}(x)\right|^4 dV] \\ &= \frac{v_1 \chi_1 \left\langle \widetilde{E_1}^4 \right\rangle}{E_0^4} + \frac{v_2 \chi_2 \left\langle \widetilde{E_2}^4 \right\rangle}{E_0^4}, \end{aligned} \tag{2.27}$$

where $v_1 = V_1/V$ and $v_2 = V_2/V$ are volume packing fractions of materials 1 and 2, respectively. Let $\left\langle \widetilde{E}_\alpha^4 \right\rangle$ represents the spatial average of trial electric fields to the fourth power in materials $\alpha = 1$ and 2,

$$\left\langle \widetilde{E}_\alpha^4 \right\rangle = \frac{1}{V_\alpha} \int_{V_\alpha} \left|\widetilde{E}(x)\right|^4 dV, \quad \alpha = 1,\ 2. \tag{2.28}$$

where V_α is the volume of α^{th} component. Since $\widetilde{\mathbf{E}}(x)$ can not be solved exactly, Yu, Hui and Lee [26] used linear field $\overrightarrow{E}(x)$ to give an estimate of the effective nonlinear coefficient. From Eq. (2.27), we obtain

$$\chi_e = \frac{v_1\chi_1 \left\langle \overrightarrow{E}_1^4 \right\rangle}{E_0^4} + \frac{v_2\chi_2 \left\langle \overrightarrow{E}_2^4 \right\rangle}{E_0^4}, \tag{2.29}$$

where $\overrightarrow{E}_\alpha(x)$, $\alpha = 1,\, 2$ is the solution of the linear composite satisfy the same boundary conditions and the same microstructure. For the linear response

$$\overrightarrow{D}(x) = \varepsilon(x)\overrightarrow{E}(x), \tag{2.30}$$

where $\varepsilon(x)$ is the linear coefficient described by ε_1 and ε_2 in materials 1 and 2, respectively. The effective linear coefficient of the composite (ε_e) can be derived in similar to Eq. (2.27). The result is

$$\begin{aligned}
\varepsilon_e &= \frac{1}{E_0^2 V}\int_V \varepsilon(x)\left|\overrightarrow{E}(x)\right|^2 dV, \\
&= \frac{1}{E_0^2 V}[\int_{V_1} \varepsilon_1(x)\left|\overrightarrow{E}_1(x)\right|^2 dV + \int_{V_2} \varepsilon_2(x)\left|\overrightarrow{E}_2(x)\right|^2 dV], \\
&= \frac{v_1\varepsilon_1 \langle E_1^2\rangle}{E_0^2} + \frac{v_2\varepsilon_2 \langle E_2^2\rangle}{E_0^2},
\end{aligned} \tag{2.31}$$

where E_1 and E_2 are electric fields in materials 1 and 2, respectively, and $\langle\ \ \rangle$ is the volume spatial average with $\langle E_\alpha^2\rangle = \dfrac{1}{v_\alpha}\int_{v_\alpha}\left|\overrightarrow{E}_\alpha(x)\right|^2 dV,\ \ \alpha = 1,\, 2.$

We invoke the decoupling approximation [27] by ignoring the fluctuations of the local electric fields,

$$\left\langle (E_\alpha^2 - \langle E_\alpha^2\rangle)^2\right\rangle = \left\langle E_\alpha^4\right\rangle - \left\langle E_\alpha^2\right\rangle^2 \cong 0 \tag{2.32}$$

or $\langle E_\alpha^4\rangle$ is approximated by

$$\left\langle E_1^4\right\rangle = \left\langle E_1^2\right\rangle^2, \tag{2.33}$$

and also

$$\left\langle E_2^4\right\rangle = \left\langle E_2^2\right\rangle^2. \tag{2.34}$$

Now, Eq.(2.29) is replaced by using Eqs. (2.33) and (2.34), hence

$$\chi_e = \frac{v_1\chi_1 \langle E_1^2\rangle^2}{E_0^4} + \frac{v_2\chi_2 \langle E_2^2\rangle^2}{E_0^4}. \tag{2.35}$$

To obtain the mean square of the electric field $\langle E_1^2 \rangle$, the derivative of Eq. (2.31) is evaluated for $\dfrac{\partial \varepsilon_e}{\partial \varepsilon_1}$, which immediately gives

$$\left\langle E_1^2 \right\rangle = \frac{1}{v_1} \frac{\partial \varepsilon_e}{\partial \varepsilon_1} E_0^2, \tag{2.36}$$

Similarly for the derivative $\dfrac{\partial \varepsilon_e}{\partial \varepsilon_2}$ of Eq. (2.31), we obtain

$$\left\langle E_2^2 \right\rangle = \frac{1}{v_2} \frac{\partial \varepsilon_e}{\partial \varepsilon_2} E_0^2. \tag{2.37}$$

Comparing Eqs. (2.31) and (2.35), χ_e, χ_2 and χ_1 are written in terms of ε_e, ε_2 and ε_1. We refer to the previous work of Yu, Hui and Lee [26]:

$$\varepsilon_e = \chi_e E_0^2, \tag{2.38}$$

$$\varepsilon_1 = \chi_1 \left\langle E_1^2 \right\rangle, \tag{2.39}$$

$$\varepsilon_2 = \chi_2 \left\langle E_2^2 \right\rangle, \tag{2.40}$$

According to the microstructure of the linear dielectric composites, ε_e can be written as a function of its constituent properties,

$$\varepsilon_e = F(\varepsilon_1, \varepsilon_2, v_2), \tag{2.41}$$

for strongly nonlinear composites, Yu et. al. [26] replaced the linear coefficients from Eqs. (2.38), (2.39) and (2.40) into Eq. (2.41), then χ_e may be expressed in the form

$$\chi_e = F(\chi_1 \left\langle E_1^2 \right\rangle, \chi_2 \left\langle E_2^2 \right\rangle, v_2)/E_0^2. \tag{2.42}$$

Note that the effective linear coefficient (ε_e) and effective nonlinear coefficient (χ_e) are independent of the external electric field $\left(\overrightarrow{E}_0\right)$.

Eqs. (2.41) and (2.42) imply that with the established results from the linear dielectric composite, the established effective linear coefficient χ_e is obtained. This approach gives results with are in good agreement with numerical simulations [26]. Consequently, the deviation of effective linear coefficient will be given in Chapter 3 in order to determine the effective nonlinear coefficient in Chapter 5.

Chapter III

Effective Linear Coefficient

The effective medium theory (EMT) was proposed by Hashin [18] in studying the effective conductivities of two-phase composite materials. In his work, the lower and upper bounds of the effective linear conductivities are determined.

According to the similarities of basic equations for electric conduction and electrostatics for dielectric media, Hashin theory can be applied to determine the effective dielectric constants (or linear coefficient) of linear dielectric composites, and therefore will be reviewed in this chapter. Then further studies extend to nonlinear dielectric composites based on the EMT incorporation with the variational method and decoupling technique to obtain the effective nonlinear coefficient which will be given in Chapters 4 and 5.

3.1 Effective Medium Model

To consider the response of a linear dielectric composite when a uniform external electric field $\left(\vec{E}_0\right)$ is applied. Let the composite be composed of spherical inclusions randomly distributed in a dielectric medium with different linear coefficients. The theoretical model proposed by Hashin called effective medium treatment (EMT) [18] will be applied to determine the effective dielectric constant.

In the EMT, the composite is considered to be composed of spherical cells. Each cell contains only one of the inclusions which is surrounded by the medium. The linear coefficients of the inclusion and the medium are ε_2 and ε_1, respectively.

The ratio of the inclusion volume to the cell volume is $\frac{a^3}{b^3}$. In this model, only a representative cell is considered (see Fig. 3.1), while the other cells are replaced by a homogeneous medium which has the effective linear coefficient ε_e to be specified.

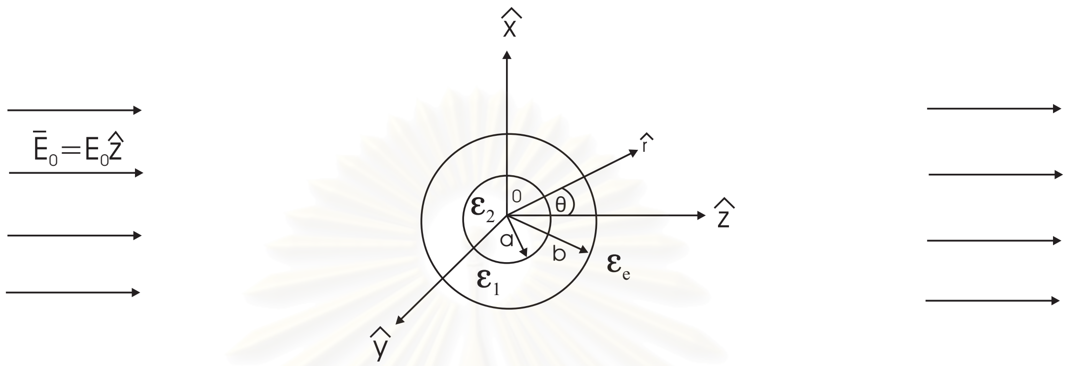

Figure 3.1: A representative cell is composed of a spherical inclusion of radius a having linear coefficient ε_2 surrounded by a concentric shell of radius b having nonlinear coefficient ε_1.

3.2 Electric Potentials

To determine the electric potentials in the cell, according to Eq. (2.14), the basic equation of linear dielectric media is Laplace equation in spherical coordinate, which is

$$\overrightarrow{\nabla}^2\varphi = 0, \tag{3.1}$$

where φ is the electric potential.

In general, the solution of Eq. (3.1) depends on variables r, θ and ϕ. In this theoretical model as shown in Fig. 3.1, the external uniform electric field is applied in the $z-axis$, then the potential has azimuthal symmetry depending on variables r and θ.

The solution of Laplace equation in this case is [28]

$$\varphi(r,\theta) = \sum_{n=0}^{\infty}[A_n r^n + B_n r^{-(n+1)}]P_n(\cos\theta), \tag{3.2}$$

where $P_n(\cos\theta)$ is called Legendre polynomials.

From the boundary conditions:

• At the inclusion center ($r = 0$), the electric potential is finite, then $B_n = 0$.

• Very long distance from the inclusion ($r \to \infty$) , the electric potential of Eq. (3.2) becomes $-E_0 r\cos\theta$.

In cooperation between the boundary conditions and the Legendre polynomials $n = 1$, the electric potentials have the simple forms

$$\varphi_2(r,\theta) = Ar\cos\theta,\ 0 \le r \le a \tag{3.3}$$

$$\varphi_1(r,\theta) = (Br + \frac{C}{r^2})\cos\theta,\ a \le r \le b \tag{3.4}$$

$$\varphi_e(r,\theta) = (-E_0 r + \frac{D}{r^2})\cos\theta,\ b \le r \le \infty \tag{3.5}$$

where φ_2 and φ_1 are the electric potentials in the inclusion and the shell region, respectively. φ_e is the electric potential in effective medium.

The constants A, B, C and D in Eqs. (3.3)-(3.5) can be determined by using this boundary conditions at the inclusion and the outer cell surfaces:

• the tangential component of $\overrightarrow{E}$ is continuous ($E_{1t} = E_{2t}$), then the electric potential is also continuous,

$$\varphi_2(r=a,\theta) = \varphi_1(r=a,\theta)$$

$$A = B + \frac{C}{a^3}, \tag{3.6}$$

and

$$\varphi_1(r=b,\theta) = \varphi_e(r=b,\theta)$$

$$B + \frac{C}{b^3} = -E_0 + \frac{D}{b^3}, \tag{3.7}$$

• the normal component of $\overrightarrow{D}$ is continuous ($D_{1n} = D_{2n}$ or $\varepsilon_2 E_{2n} = \varepsilon_1 E_{1n}$), hence

$$\varepsilon_2 \frac{\partial\varphi_2}{\partial r}\ |_{r=a} = \varepsilon_1 \frac{\partial\varphi_1}{\partial r}\ |_{r=a},$$

$$\varepsilon_2 A = \varepsilon_1 (B - \frac{2C}{a^3}), \tag{3.8}$$

and

$$\varepsilon_1 \frac{\partial \varphi_1}{\partial r} \mid_{r=b} = \varepsilon_e \frac{\partial \varphi_e}{\partial r} \mid_{r=b} .$$

$$\varepsilon_1 (B - \frac{2C}{b^3}) = -\varepsilon_e (E_0 + \frac{2D}{b^3}). \tag{3.9}$$

Replacing Eq. (3.6) in Eq. (3.8), we obtain

$$B = \frac{-xC}{a^3}, \tag{3.10}$$

where $x = \dfrac{\varepsilon_2 + 2\varepsilon_1}{\varepsilon_2 - \varepsilon_1}$.

Substituting Eq. (3.10) into Eqs. (3.7) and (3.9), then we obtain

$$(\frac{1}{b^3} - \frac{x}{a^3})C = -E_0 + \frac{D}{b^3}, \tag{3.11}$$

$$-\varepsilon_1 (\frac{2}{b^3} + \frac{x}{a^3})C = -\varepsilon_e E_0 - \frac{2\varepsilon_e D}{b^3}. \tag{3.12}$$

From Eqs. (3.11)-(3.12), the constants C, and D are obtained, which are usable in solving for another constants:

$$A = \frac{-9\kappa E_0}{[2(1-c') + \beta(2c'+1)] + 2\kappa[(2+c') + \beta(1-c')]}, \tag{3.13}$$

$$B = \frac{-3\kappa(\beta+2)E_0}{[2(1-c') + \beta(2c'+1)] + 2\kappa[(2+c') + \beta(1-c')]}, \tag{3.14}$$

$$C = \frac{3\kappa(\beta-1)a^3 E_0}{[2(1-c') + \beta(2c'+1)] + 2\kappa[(2+c') + \beta(1-c')]}, \tag{3.15}$$

where $c' = \dfrac{a^3}{b^3}$, $\beta = \dfrac{\varepsilon_2}{\varepsilon_1}$ and $\kappa = \dfrac{\varepsilon_e}{\varepsilon_1}$.

From Eqs. (3.13)-(3.15), A, B, and C are still given in terms of $\kappa = \dfrac{\varepsilon_e}{\varepsilon_1}$ called relative effective linear coefficient which is the unknown has to be specified.

The effective linear coefficient (ε_e) is defined as [18]

$$\left\langle \overrightarrow{D} \right\rangle = \varepsilon_e \left\langle \overrightarrow{E} \right\rangle, \tag{3.16}$$

where $\langle \ \rangle$ is the volume average.

The effective linear coefficient term $\left\langle \overrightarrow{D} \right\rangle$ of Eq. (3.16) may be written as the sum over inclusion and host medium, so Eq. (3.16) becomes

$$\varepsilon_e \left\langle E_z \right\rangle = v_2 \varepsilon_2 \left\langle E_z^{(2)} \right\rangle + v_1 \varepsilon_1 \left\langle E_z^{(1)} \right\rangle, \tag{3.17}$$

where the subscript z represents the electric field component in $z - axis$, $\left\langle E_z^{(1)} \right\rangle$ is the volume average of the electric field in the host medium only, and $\left\langle E_z^{(2)} \right\rangle$ is the volume average of the electric field over the inclusion, v_2 is the inclusion packing fraction and $v_1 = 1 - v_2$.

With the boundary condition $\overrightarrow{E} = \overrightarrow{E}_0 = E_0 \widehat{z}$, it is true that [31]

$$\left\langle E_z \right\rangle = E_0, \tag{3.18}$$

hence

$$v_2 \left\langle E_z^{(2)} \right\rangle + v_1 \left\langle E_z^{(1)} \right\rangle = E_0. \tag{3.19}$$

$\left\langle E_z^{(1)} \right\rangle$ in Eq. (3.17) is eliminated by using Eq. (3.19), then

$$\varepsilon_e E_0 = \varepsilon_1 E_0 + v_2(\varepsilon_2 - \varepsilon_1) \left\langle E_z^{(2)} \right\rangle. \tag{3.20}$$

This equation indicates that if we know $\left\langle E_z^{(2)} \right\rangle$, ε_e can be calculated.

Calculating $\left\langle E_z^{(2)} \right\rangle$ for this problem, we get

$$\left\langle E_z^{(2)} \right\rangle = -A. \tag{3.21}$$

From Eqs. (3.13), (3.20) and (3.21), the solution κ is given by

$$\kappa = 1 + \frac{9v_2(\beta - 1)\kappa}{[2(1 - c') + \beta(2c' + 1)] + 2\kappa[(2 + c') + \beta(1 - c')]}, \tag{3.22}$$

where $\kappa = \dfrac{\varepsilon_e}{\varepsilon_1}$, $\beta = \dfrac{\varepsilon_2}{\varepsilon_1}$ and $c' = \dfrac{a^3}{b^3}$.

The relative effective linear coefficient $\left(\kappa = \dfrac{\varepsilon_e}{\varepsilon_1}\right)$ is a function of parameter $c' = \dfrac{a^3}{b^3}$ which is the ratio of the inclusion volume to cell volume in Fig. 3.1. The restriction of c' is

$$c \leq c' \leq 1, \tag{3.23}$$

where c is the inclusion packing fraction limiting the maximum cell volume. $c' = 1$ is for $b = a$ or the case of inclusion embedded in the effective medium without surrounding host medium phase 1.

In this research, the effective linear coefficient (ε_e) will be determined for special cases of $c' = 1$ and $c' = c$ (inclusion packing fraction).

3.3 Effective Linear Coefficient

The composite is categorized into three cases. First, inclusions of material 2 embedded in material 1. Second, inclusions of material 1 embedded in material 2. The last, two interdispersed materials.

3.3.1 Inclusions of Material 2 Embedded in Material 1

Refer to Fig. 3.1, it shows the theoretical model for the composite with dielectric inclusions of phase 2 of linear coefficient ε_2 embedded in phase 1 material of linear coefficient ε_1. We now determine the effective linear coefficient for $c' = \dfrac{a^3}{b^3}$ which is equal to the packing fraction of inclusions ($c' = v_2$).

From Eq. (3.22), we replace $c' = v_2$,

$$2[(2+v_2)+\beta(1-v_2)]\kappa^2-[(2-5)v_2+\beta(1+5v_2)]\kappa-[2(1-v_2)+\beta(1+2v_2)] = 0. \quad (3.24)$$

The solution of κ has two real roots of opposite sign, a negative root gives a negative ε_e which is physically meaningless. Only a positive root is considered, which is

$$\kappa = \frac{\varepsilon_e}{\varepsilon_1} = \frac{(2+\beta)+2(\beta-1)v_2}{(2+\beta)+2(1-\beta)v_2}, \quad (3.25)$$

where $\beta = \dfrac{\varepsilon_2}{\varepsilon_1}$.

The effective linear coefficient can now be determined by rearranging Eq.

(3.25), which gives

$$\varepsilon_e = \varepsilon_1 \left[1 + \frac{v_2}{\dfrac{\varepsilon_1}{\varepsilon_2 - \varepsilon_1} + \dfrac{v_1}{3}} \right]. \tag{3.26}$$

3.3.2 Inclusions of Material 1 Embedded in Material 2

In contrary to subsection 3.3.1, we consider inclusions of material 1 with linear coefficient ε_1 embedded in medium of linear coefficient ε_2 . In this case, ε_e is similar to that of Eq. (3.22) with interchanging between ε_2 and ε_1, v_2 and v_1, hence

$$\kappa = 1 + \frac{9v_1(\beta - 1)\kappa}{[2(1-c') + \beta(2c'+1)] + 2\kappa[(2+c') + \beta(1-c')]}, \tag{3.27}$$

where $\kappa = \dfrac{\varepsilon_e}{\varepsilon_2}$, $\beta = \dfrac{\varepsilon_1}{\varepsilon_2}$ and $c' = \dfrac{a^3}{b^3}$. For c' is minimum equal to the volume packing fraction of inclusion ($c' = v_1$), then Eq. (3.27) becomes

$$\varepsilon_e = \varepsilon_2 \left[1 + \frac{v_1}{\dfrac{\varepsilon_2}{\varepsilon_1 - \varepsilon_2} + \dfrac{v_2}{3}} \right]. \tag{3.28}$$

3.3.3 Two Interdispersed Materials

Now we consider composites consisting of two interdispersed materials phases 1 and 2. The theoretical model is that an inclusion with linear coefficient ε_2 (or ε_1) is embedded in an effective medium with effective linear coefficient ε_e. This is the case $c' = 1$ in Fig. 3.1.

For $c' = 1$, Eq. (3.22) becomes

$$\kappa = 1 + \frac{9v_2(\beta - 1)\kappa}{3(\beta + 2\kappa)}. \tag{3.29}$$

Substituting $\kappa = \dfrac{\varepsilon_e}{\varepsilon_1}$ and $\beta = \dfrac{\varepsilon_2}{\varepsilon_1}$, we get

$$-2\varepsilon_e^2 + 2\varepsilon_e\varepsilon_1 - \varepsilon_e\varepsilon_2 - 2v_2\varepsilon_e\varepsilon_1 - v_2\varepsilon_e\varepsilon_1 + 2v_2\varepsilon_e\varepsilon_2 + v_2\varepsilon_e\varepsilon_2 + \varepsilon_e\varepsilon_1 = 0. \tag{3.30}$$

By using the relation between the packing fraction of inclusion (v_2) and the packing fraction of host medium (v_1) with $v_1 + v_2 = 1$, then Eq. (3.30) becomes

$$-2(v_1+v_2)\varepsilon_e^2+2\varepsilon_e\varepsilon_1-\varepsilon_e\varepsilon_2-2(1-v_1)\varepsilon_e\varepsilon_1-v_2\varepsilon_e\varepsilon_1+2v_2\varepsilon_e\varepsilon_2+(1-v_2)\varepsilon_e\varepsilon_2+(v_1+v_2)\varepsilon_1\varepsilon_2 = 0, \tag{3.31}$$

$$v_2(\frac{\varepsilon_2 - \varepsilon_e}{\varepsilon_2 + 2\varepsilon_e}) + v_1(\frac{\varepsilon_1 - \varepsilon_e}{\varepsilon_1 + 2\varepsilon_e}) = 0. \tag{3.32}$$

Symmetrically, we may consider the phase 1 material is embedded in the effective medium.

For $c' = 1$, Eq. (3.27) becomes

$$v_1(\frac{\varepsilon_1 - \varepsilon_e}{\varepsilon_1 + 2\varepsilon_e}) + v_2(\frac{\varepsilon_2 - \varepsilon_e}{\varepsilon_2 + 2\varepsilon_e}) = 0. \tag{3.33}$$

It is observed that Eq. (3.32) is exactly the same as Eq. (3.33) which explains the symmetrically dispersed of materials 1 and 2.

3.4 Results

The schematic plot of the effective linear coefficients (ε_e) calculated from Eqs. (3.26), (3.28), (3.32) and (3.33) against the packing fraction of inclusion (v_2), are shown in Fig. 3.2 for the case of $\varepsilon_2 \rangle \varepsilon_1$.

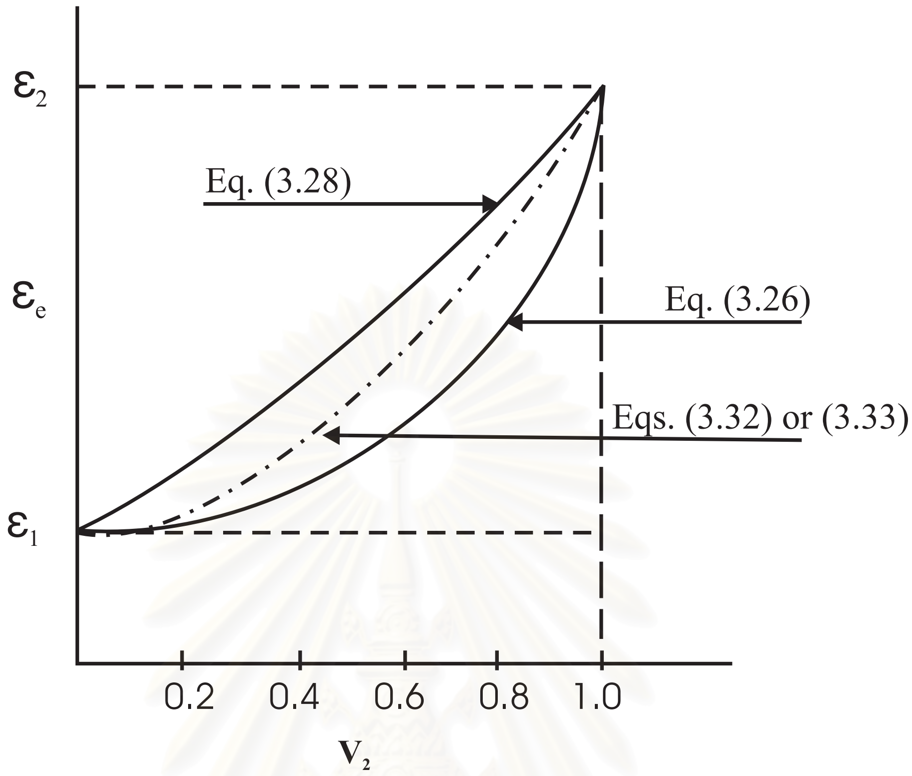

Figure 3.2: Bounds of the effective nonlinear coefficients.

Figure 3.2 shows the lower and upper bounds of the effective linear coefficients obtained from Eqs. (3.26) and (3.28), respectively. The curves representing Eqs. (3.32) and (3.33) coincide and lie between the two bounds.

Chapter IV

Effective Nonlinear Coefficient by Variational Method

In this Chapter, the studying has been extended to strongly nonlinear dielectric composites by employing the effective medium treatment (EMT) and the variational method in solving the electrostatic boundary value problem. Then the effective nonlinear coefficient (χ_e) including the lower and upper bounds are determined.

4.1 Effective Medium Model

To consider the response of a nonlinear dielectric composite when a uniform external electric field $\left(\overrightarrow{E}_0\right)$ is applied. We assume that the composite is composed of two components: inclusions and dielectric medium, which exhibit different nonlinear coefficients. The inclusions are randomly distributed in dielectric medium.

By using EMT, the composite is considered to be composed of spherical cells. Each cell contains only one of the inclusions which is surrounded by the medium. The nonlinear coefficients of the inclusion and the medium are χ_2 and χ_1, respectively. The ratio of the inclusion volume to the cell volume is $\dfrac{a^3}{b^3}$. In this model, only a representative cell is considered (see Fig. 4.1), while the other cells are replaced by a homogeneous medium which has the effective nonlinear coefficient χ_e to be specified.

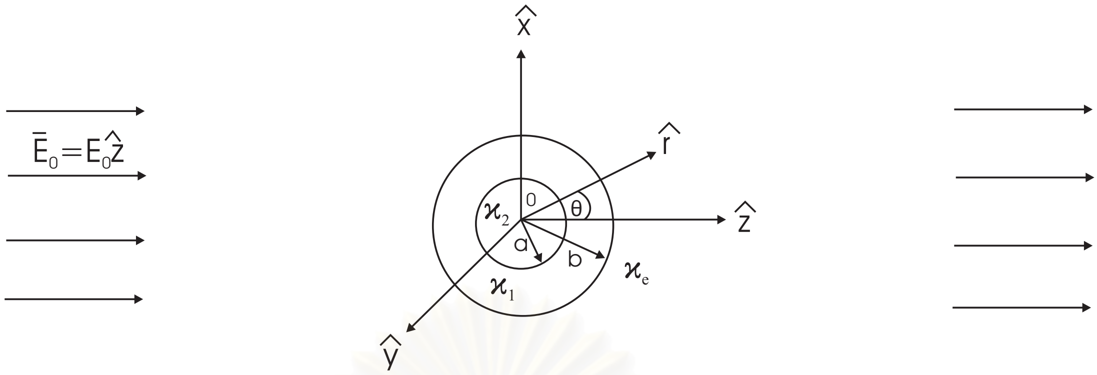

Figure 4.1: A representative cell is composed of a spherical inclusion of radius a having linear coefficient χ_2 surrounded by a concentric shell of radius b having nonlinear coefficient χ_1.

4.2 Effective Nonlinear Coefficient

To obtain the effective nonlinear coefficient of strongly nonlinear composite, three cases of composite materials are considered. First, inclusions of material 2 embedded in material 1. Second, inclusions of material 1 embedded in material 2. The last, two interdispersed materials.

4.2.1 Inclusions of Material 2 Embedded in Material 1

First, we consider the material with inclusions of strongly nonlinear of material 2 embedded in material 1. The theoretical model is shown in Fig. 4.1. To determine the electric potentials in the cell and the effective medium which obey the complicated nonlinear differential Eq. (2.16), the variational method which is explained in section 2.4 will be applied.

We use simple trial potentials:

$$\varphi_2(r,\theta) = -cE_0 r\cos\theta, \quad 0 \le r \le a \tag{4.1}$$

$$\varphi_1(r,\theta) = -E_0(fr - g\frac{a^3}{r^2})\cos\theta, \quad a \le r \le b \tag{4.2}$$

$$\varphi_e(r,\theta) = -E_0(r - d\frac{b^3}{r^2})\cos\theta, \quad b \le r \le \infty \tag{4.3}$$

which were chosen by Yu [24] to predict the strongly nonlinear response of dilute composites with reasonably good results.

The continuity of the potentials at the inclusion and the outer cell surface ($r = b$) are used to determine the relation of constants c, f, g and d in Eqs. (4.1)-(4.3). We get

$$c = f - g, \tag{4.4}$$

$$d = 1 - f + gc', \tag{4.5}$$

where $c' = \frac{a^3}{b^3}$.

We reduce four constants into two variational parameters which are f and g. The other parameters c and d, are related to f and g as shown in Eqs. (4.4) and (4.5).

To determine the two variational parameters f and g with the trial potentials in Eqs. (4.1)-(4.3), the energy functional of Eq. (2.21) is used. For strongly nonlinear composites, the first term of Eq. (2.21) is neglected. Therefore, the energy functional is

$$W[\varphi] = \frac{1}{4}[\int\limits_{v_1} \chi_1(x) \left|\overrightarrow{\nabla}\varphi_1(x)\right|^4 dV + \int\limits_{v_2} \chi_2(x) \left|\overrightarrow{\nabla}\varphi_2(x)\right|^4 dV + \int\limits_{v_e} \chi_e(x) \left|\overrightarrow{\nabla}\varphi_e(x)\right|^4 dV] + W_s, \tag{4.6}$$

where W_s is the surface energy term which is

$$W_s = \chi_e d \frac{b^3}{R^3} E_0^4, \tag{4.7}$$

as pointed out by Bergman [1].

In order to obtain the energy functional $W[\varphi]$, we first determine the potential gradients from Eqs. (4.1)-(4.3),

$$\begin{aligned}
\overrightarrow{\nabla}\varphi_2 &= -cE_0[\cos\theta\widehat{r} - \sin\theta\widehat{\theta}], && (4.8)\\
\overrightarrow{\nabla}\varphi_1 &= -E_0[(f + \frac{2ga^3}{r^3})\cos\theta\widehat{r} - (f - \frac{ga^3}{r^3})\sin\theta\widehat{\theta}], && (4.9)\\
\overrightarrow{\nabla}\varphi_e &= -E_0[(1 + \frac{2db^3}{r^3})\cos\theta r - (1 - \frac{db^3}{r^3})\sin\theta\widehat{\theta}]. && (4.10)
\end{aligned}$$

Then Eqs. (4.8)-(4.10) are substituted into Eq. (4.6), we obtain

$$\begin{aligned} W[\varphi] &= \frac{1}{4}E_0^4[(-\frac{a^3}{R^3}f^4+\frac{b^3}{R^3}f^4+\frac{36}{5}\frac{a^3}{R^3}f^2g^2-\frac{36a^6f^2g^2}{5b^3R^3}+\frac{8}{5}\frac{a^3}{R^3}fg^3 \\ &\quad -\frac{8a^9fg^3}{5b^6R^3}+\frac{8}{5}\frac{a^3}{R^3}g^4-\frac{8a^{12}g^4}{5b^9R^3})\chi_1+\frac{a^3}{R^3}c^4\chi_2+(1-\frac{8b^{12}d^4}{5R^{12}} \\ &\quad -\frac{8b^9d^3}{5R^9}-\frac{36b^6d^2}{5R^6}-\frac{b^3}{R^3}+\frac{36b^3d^2}{5R^3}+\frac{8b^3d^3}{5R^3}+\frac{8b^3d^4}{5R^3})\chi_e] \\ &\quad +\frac{b^3}{R^3}d\chi_eE_0^4, \end{aligned} \tag{4.11}$$

where the composite volume is assumed to be $\frac{4}{3}\pi R^3$. For $R \gg b$, Eq. (4.11) is reduced to

$$\begin{aligned} W[\varphi] &= \frac{1}{4}E_0^4[(-\frac{a^3}{R^3}f^4+\frac{b^3}{R^3}f^4+\frac{36}{5}\frac{a^3}{R^3}f^2g^2-\frac{36a^6f^2g^2}{5b^3R^3}+\frac{8}{5}\frac{a^3}{R^3}fg^3 \\ &\quad -\frac{8a^9fg^3}{5b^6R^3}+\frac{8}{5}\frac{a^3}{R^3}g^4-\frac{8a^{12}g^4}{5b^9R^3})\chi_1+\frac{a^3}{R^3}c^4\chi_2+(1-\frac{b^3}{R^3} \\ &\quad +\frac{36b^3d^2}{5R^3}+\frac{8b^3d^3}{5R^3}+\frac{8b^3d^4}{5R^3})\chi_e]+\frac{b^3}{R^3}d\chi_eE_0^4. \end{aligned} \tag{4.12}$$

Without loss of generality, E_0 is set equal to 1, Eq. (4.12) with $v_2=\frac{a^3}{b^3}$ is

$$\begin{aligned} W[\varphi] &= \frac{1}{4}\frac{b^3}{R^3}[(f^4-f^4v_2+\frac{36}{5}f^2g^2v_2+\frac{8}{5}fg^3v_2+\frac{8}{5}g^4v_2-\frac{36}{5}f^2g^2v_2^2 \\ &\quad -\frac{8}{5}fg^3v_2^3-\frac{8}{5}g^4v_2^4)\chi_1+c^4v_2\chi_2+(1+4d+\frac{36}{5}d^2+\frac{8}{5}d^3 \\ &\quad +\frac{8}{5}d^4)\chi_e]+\frac{1}{4}\chi_e. \end{aligned} \tag{4.13}$$

The constants c and d from Eq. (4.13) are eliminated by using Eqs. (4.4) and (4.5). Minimization of $W[\varphi]$ with respect to the variational parameters f and g gives the following equations:

$$\begin{aligned} \frac{\partial W}{\partial f} &= f^3-x-f^3v_2+\frac{18}{5}fg^2v_2+\frac{2g^3v_2}{5}+(f-g)^3yv_2-\frac{18}{5}fg^2v_2^2 \\ &\quad -\frac{2}{5}g^3v_2^3-\frac{18}{5}x(1-f+gv_2)-\frac{6}{5}x(1-f+gv_2)^2-\frac{8}{5}x(1-f+gv_2)^3 \\ &= 0, \end{aligned} \tag{4.14}$$

and

$$\begin{aligned} \frac{\partial W}{\partial g} &= \frac{18}{5}f^2gv_2+\frac{6}{5}fg^2v_2+\frac{8}{5}g^3v_2+xv_2-(f-g)^3yv_2-\frac{18}{5}f^2gv_2^3-\frac{6}{5}fg^2v_2^3 \\ &\quad -\frac{8}{5}g^3v_2^4+\frac{18}{5}xv_2(1-f+gv_2)+\frac{6}{5}xv_2(1-f+gv_2)^2+\frac{8}{5}xv_2(1-f+gv_2)^3 \\ &= 0, \end{aligned} \tag{4.15}$$

where $x = \dfrac{\chi_e}{\chi_1}$ and $y = \dfrac{\chi_2}{\chi_1}$.

In fact, the functions of $\dfrac{\partial W}{\partial f} = 0$ and $\dfrac{\partial W}{\partial g} = 0$ in Eqs. (4.14) and (4.15) have three roots of f and g which yield the functions extremum. Two roots are complex number which give physically meaningless χ_e. Only real root of f and g will be used.

To solve Eqs. (4.14) and (4.15) for f and g, $c' = \dfrac{a^3}{b^3}$ or inclusion to cell volume is set equal to the inclusion packing fraction. One more condition is given to specify the unknown χ_e. This is the self-consistency condition [18] given by

$$\left\langle E_z^{(e)} \right\rangle = v_1 \left\langle E_z^{(1)} \right\rangle + v_2 \left\langle E_z^{(2)} \right\rangle, \tag{4.16}$$

where subscript z is the electric field component in z-axis. $\left\langle E_z^{(2)} \right\rangle$ and $\left\langle E_z^{(1)} \right\rangle$ are the volume average of the electric field within the inclusion and host medium, respectively.

Eqs. (4.8)-(4.10) are used to calculate $\left\langle E_z^{(1)} \right\rangle$, $\left\langle E_z^{(2)} \right\rangle$ and $\left\langle E_z^{(e)} \right\rangle$. We get

$$\left\langle E_z^{(1)} \right\rangle = fE_0, \tag{4.17}$$

$$\left\langle E_z^{(2)} \right\rangle = cE_0, \tag{4.18}$$

$$\left\langle E_z^{(e)} \right\rangle = E_0. \tag{4.19}$$

By substituting Eqs. (4.17)-(4.19) into Eq. (4.16), we obtain

$$f = 1 + v_2 g. \tag{4.20}$$

From Eqs. (4.14), (4.15) and (4.20), we can solve for f, g and χ_e as in terms of $y = \dfrac{\chi_2}{\chi_1}$ and v_2 which are parameters to specify the effective nonlinear coefficient χ_e. Because of the complication χ_e can not be solved in a closed form. To keep off the complication, χ_e is determined for specific values of $y = \dfrac{\chi_2}{\chi_1}$ and v_2. So, the relative effective nonlinear coefficient $\left(\dfrac{\chi_e}{\chi_1}\right)$ as a function of the inclusion packing fraction (v_2) and the relative nonlinear coefficient $\left(\dfrac{\chi_2}{\chi_1}\right)$ is obtained:

$$\frac{\chi_e}{\chi_1} = F(v_2, \frac{\chi_2}{\chi_1}). \tag{4.21}$$

In this work, the inclusion packing fraction (v_2) is varied from the dilute limit ($v_2 = 0$) to the ideal maximum packing fraction $v_2 = 1$. The parameter $\frac{\chi_2}{\chi_1}$ has been set equal to 10, 100, 1000, 0.1, 0.01, and 0.001. The relative effective nonlinear coefficients $\left(\frac{\chi_e}{\chi_1}\right)$ are determined for arbitrary inclusion packing fractions by using a Mathematica program (see appendix C).

4.2.2 Inclusions of Material 1 Embedded in Material 2

In contrary to subsection 4.2.1 with the theoretical model Fig. 4.1, we now consider the strongly nonlinear dielectric inclusions having nonlinear coefficient χ_1 embedded in the medium of strongly nonlinear coefficient χ_2. The calculation of relative nonlinear coefficient $\left(\frac{\chi_e}{\chi_1}\right)$ is resemblant to the mathematical process in subsection 4.2.1

The relative effective nonlinear coefficient $\left(\frac{\chi_e}{\chi_2}\right)$ is obtained as a function of inclusion packing fraction v_1 and the relative nonlinear coefficient $\frac{\chi_1}{\chi_2}$. For the purpose of reporting in the same figure of those given in subsection 4.2.1, we write the relative effective nonlinear coefficient as these results

$$\frac{\chi_e}{\chi_1} = F'(v_2, \frac{\chi_2}{\chi_1}), \tag{4.22}$$

where v_1 is replaced by $v_2 = 1 - v_1$.

The parameter $y = \frac{\chi_2}{\chi_1}$ has been set equal to 10, 100, 1000, 0.1, 0.01, and 0.001. The relative effective nonlinear coefficients $\left(\frac{\chi_e}{\chi_1}\right)$ are determined for arbitrary packing fractions v_2 by using the Mathematica program (see appendix C).

4.2.3 Two Interdispersed Materials

Now we consider composites consisting of two interdispersed materials phases 1 and 2. The theoretical model is that an inclusion with nonlinear coefficient χ_2 (or χ_1) is embedded in an effective medium with effective nonlinear coefficient χ_e.

In fact, this model is the special case of the EMT with $c' = 1$, it is called the effective medium approximation (EMA). For the case of the composite can not be specified clearly which phase 1 (or 2) is the inclusions as shown in Fig. 4.2.

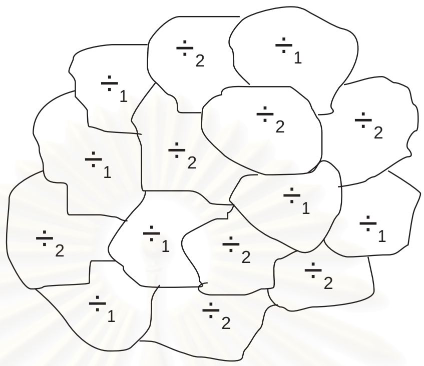

Figure 4.2: Two interdispersed materials.

So the representative cell presented in Fig. 4.1 is replaced by a single particle of phase 1 or 2 having nonlinear coefficient χ_α $(\alpha = 1, 2)$, it is surrounded by a homogeneous medium of effective nonlinear coefficient χ_e (see Fig. 4.3).

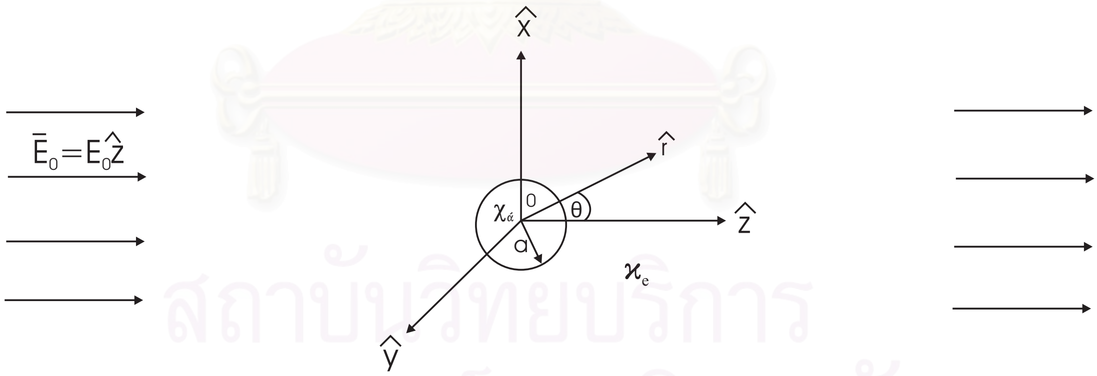

Figure 4.3: A spherical inclusion of radius a with nonlinear coefficient χ_α $(\alpha = 1, 2)$ is surrounded by an effective medium having nonlinear coefficient χ_e.

To determine the electric potentials in the inclusion and the effective medium which obey the complicated nonlinear differential Eq. (2.17), the variational method which is explained in section 2.4 will be applied.

We use simple trial potentials:

$$\varphi_\alpha(r,\theta) = -c_\alpha E_0 r\cos\theta, \quad 0 \le r \le a \tag{4.23}$$

$$\varphi_e(r,\theta) = -E_0(r - b_\alpha \frac{a^3}{r^3})\cos\theta, \quad r \ge a \tag{4.24}$$

where φ_α and φ_e are the electric potentials in the inclusion of material type α ($\alpha = 1, 2$) and the effective medium, respectively. b_α is a variational parameter as yet to be determined.

The continuity of the potentials at the inclusion surface is used in order to determine the relation between parameters c_α and b_α. From Eqs. (4.23) and (4.24), evaluated at $r = a$, we obtain

$$c_\alpha = 1 - b_\alpha. \tag{4.25}$$

To determine the variational parameter b_α with the trial electric potentials, Eqs. (4.23) and (4.24), the energy functional of Eq. (2.21) with the first term is neglected,

$$W[\varphi] = \frac{1}{4}[\int_{v_\alpha} \chi_\alpha(x)\left|\overrightarrow{\nabla}\varphi_\alpha(x)\right|^4 dV + \int_{v_e} \chi_e(x)\left|\overrightarrow{\nabla}\varphi_e(x)\right|^4 dV] + W_s, \tag{4.26}$$

is determined and minimized with respect to the parameter b_α. In this case, the surface energy term (W_s) is [16]

$$W_s = \chi_e b_\alpha \frac{a^3}{R^3} E_0^4. \tag{4.27}$$

From Eqs. (4.23) and (4.24), we get

$$\overrightarrow{\nabla}\varphi_\alpha = -c_\alpha E_0[\cos\theta\widehat{r} - \sin\theta\widehat{\theta}], \tag{4.28}$$

$$\overrightarrow{\nabla}\varphi_e = -E_0[(1 + \frac{2b_\alpha a^3}{r^3})\cos\theta\widehat{r} - (1 - \frac{b_\alpha a^3}{r^3})\sin\theta\widehat{\theta}]. \tag{4.29}$$

Eqs. (4.27)-(4.29) are substituted into Eq. (4.26), hence

$$\begin{aligned} W[\varphi] = {} & \frac{1}{4}E_0^4[(1 - \frac{a^3}{R^3} - \frac{36a^6 b_\alpha^2}{5R^6} + \frac{36a^3 b_\alpha^2}{5R^3} - \frac{8a^9 b_\alpha^3}{5R^9} + \frac{8a^3 b_\alpha^3}{5R^3} \\ & - \frac{8a^{12} b_\alpha^4}{5R^{12}} + \frac{8a^3 b_\alpha^4}{5R^3})\chi_e + \frac{a^3 c_\alpha^4 \chi_\alpha}{R^3}] + \frac{a^3 b_\alpha}{R^3}\chi_e E_0^4, \end{aligned} \tag{4.30}$$

where the composite volume is assumed to be $\frac{4}{3}\pi R^3$. For $R \gg a$, Eq. (4.30) is reduced to

$$\begin{aligned} W[\varphi] &= \frac{1}{4}E_0^4[(1-\frac{a^3}{R^3}+\frac{36a^3b_\alpha^2}{5R^3}+\frac{8a^3b_\alpha^3}{5R^3}+\frac{8a^3b_\alpha^4}{5R^3})\chi_e \\ &\quad +\frac{a^3}{R^3}c_\alpha^4\chi_\alpha]+\chi_e b_\alpha\frac{a^3}{R^3}E_0^4. \end{aligned} \tag{4.31}$$

Without loss of generality, E_0 is set equal to 1, Eq. (4.31) with $v_\alpha = \frac{a^3}{R^3}$ and $c_\alpha = 1 - b_\alpha$ is

$$W[\varphi] = \frac{1}{4}[(1-v_\alpha+\frac{36}{5}v_\alpha b_\alpha^2+\frac{8}{5}v_\alpha b_\alpha^3+\frac{8}{5}v_\alpha b_\alpha^4+4v_\alpha b_\alpha)\chi_e+\frac{1}{4}v_\alpha(1-b_\alpha)^4\chi_\alpha]. \tag{4.32}$$

A dimensionless contrast parameter between the α^{th} component and the effective medium is defined as $y_\alpha = \frac{\chi_\alpha}{\chi_e}$ $(\alpha = 1, 2)$ and c_α is eliminated from Eq. (4.32) by using Eqs. (4.25). Minimization of $W[\varphi]$ with respect to the variational parameter b_α gives

$$\begin{aligned} \frac{\partial W}{\partial b_\alpha} &= v_\alpha\left(1+\frac{18b_\alpha}{5}+\frac{6b_\alpha^2}{5}+\frac{8b_\alpha^3}{5}-(1-b_\alpha)^3 y_\alpha\right), \\ &= 0. \end{aligned} \tag{4.33}$$

Eq. (4.33) is solved analytically for b_α, we obtain three b_α with different roots. Two complex roots give physically meaningless χ_e. Only real root is considered, which is

$$\begin{aligned} b_\alpha &= \frac{-2+5y_\alpha}{8+5y_\alpha} - \\ &\quad \frac{2^{1/3}(396+810y_\alpha)}{3(8+5y_\alpha)(-1296-7020y_\alpha-24975y_\alpha^2+27\sqrt{5}(8+5y_\alpha)\sqrt{1072+5272y_\alpha+6845y_\alpha^2})^{1/3}} + \\ &\quad \frac{(-1296-7020y_\alpha-24975y_\alpha^2+27\sqrt{5}(8+5y_\alpha)\sqrt{1072+5272y_\alpha+6845y_\alpha^2})^{1/3}}{3\ 2^{1/3}(8+5y_\alpha)}. \end{aligned} \tag{4.34}$$

From Eq. (4.34), we note that the parameter b_α is a function of $y_\alpha = \frac{\chi_\alpha}{\chi_e}$ $(\alpha = 1, 2)$. The volume average of local electric field within the spherical inclusion $\left(\left\langle E_z^{(\alpha)}\right\rangle\right)$ is calculated by using Eq. (4.28), we get

$$\begin{aligned} \left\langle E_z^{(\alpha)}\right\rangle &= c_\alpha E_0 \\ &= (1-b_\alpha)E_0. \end{aligned} \tag{4.35}$$

Hence, an approximate expression for $\left\langle E_z^{(\alpha)} \right\rangle$ is obtained by using the solution of b_α calculated in Eq. (4.34). For convenience, we define new parameters which are dimensionless as

$$y_2 = \frac{\chi_2}{\chi_e} = \frac{\chi_2}{\chi_1}\frac{\chi_1}{\chi_e} = \frac{y}{x} \text{ and } y_1 = \frac{\chi_1}{\chi_e} = \frac{1}{x},$$

where $x = \dfrac{\chi_e}{\chi_1}$ and $y = \dfrac{\chi_2}{\chi_1}$.

According to the self-consistency condition of Eq. (4.16), the volume average of local electric field within the spherical material α calculated by Eq. (4.35) is replaced into Eq. (4.16), we get

$$v_2 b(y_2) + (1 - v_2) b(y_1) = 0, \tag{4.36}$$

where $b(y_1)$ and $b(y_2)$ are the solutions of Eq. (4.34) evaluated at $\chi_\alpha = \chi_1$ and $\chi_\alpha = \chi_2$, respectively.

By solving Eq. (4.36), then the relative effective nonlinear coefficient is obtained as a function of v_2 and $y = \dfrac{\chi_2}{\chi_1}$:

$$\frac{\chi_e}{\chi_1} = F(v_2, \frac{\chi_2}{\chi_1}, b(y_1), b(y_2)). \tag{4.37}$$

In this work, the inclusion packing fraction (v_2) is varied from the dilute limit ($v_2 = 0$) to the ideal maximum packing fraction $v_2 = 1$. The parameter $\dfrac{\chi_2}{\chi_1}$ has been set equal to 10, 100, 1000, 0.1, 0.01, and 0.001. The relative effective nonlinear coefficients $\left(\dfrac{\chi_e}{\chi_1}\right)$ are determined for arbitrary inclusion packing fractions by using a Mathematica program (see appendix C).

4.3 Results and Discussion

The relative effective nonlinear coefficients are plotted in terms of $log(\dfrac{\chi_e}{\chi_1})$ against the packing fraction of material 2 (v_2) for various values of relative nonlinear coefficients $\left(y = \dfrac{\chi_2}{\chi_1}\right)$, as shown in Fig. 4.4 for $\dfrac{\chi_2}{\chi_1} = 10$, 100, and 1000, Fig. 4.5 for $\dfrac{\chi_2}{\chi_1} = 0.1$, 0.01, and 0.01.

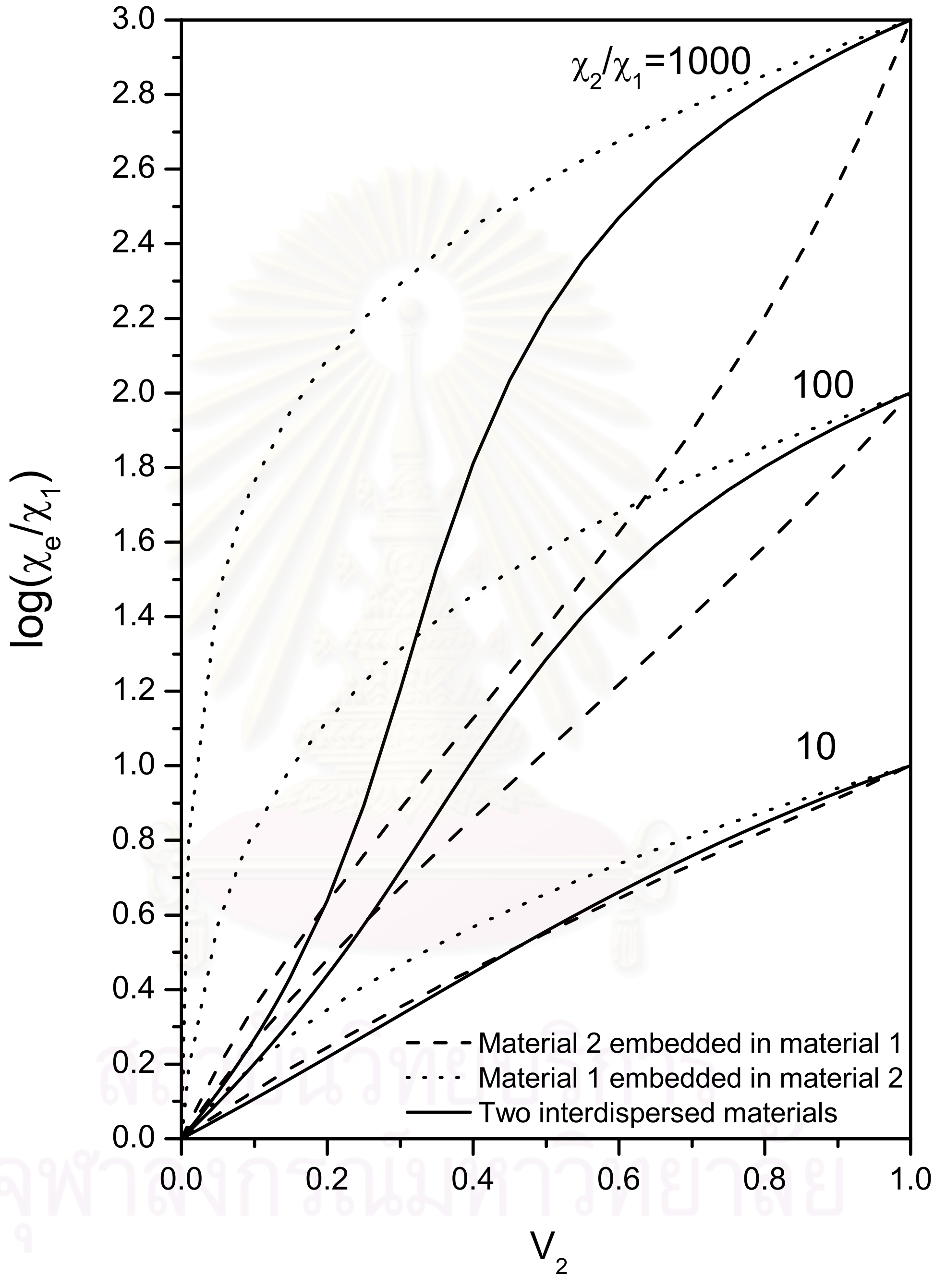

Figure 4.4: Bounds of the effective nonlinear coefficient by the variational method ($\frac{\chi_2}{\chi_1} = 10$, 100 and 1000).

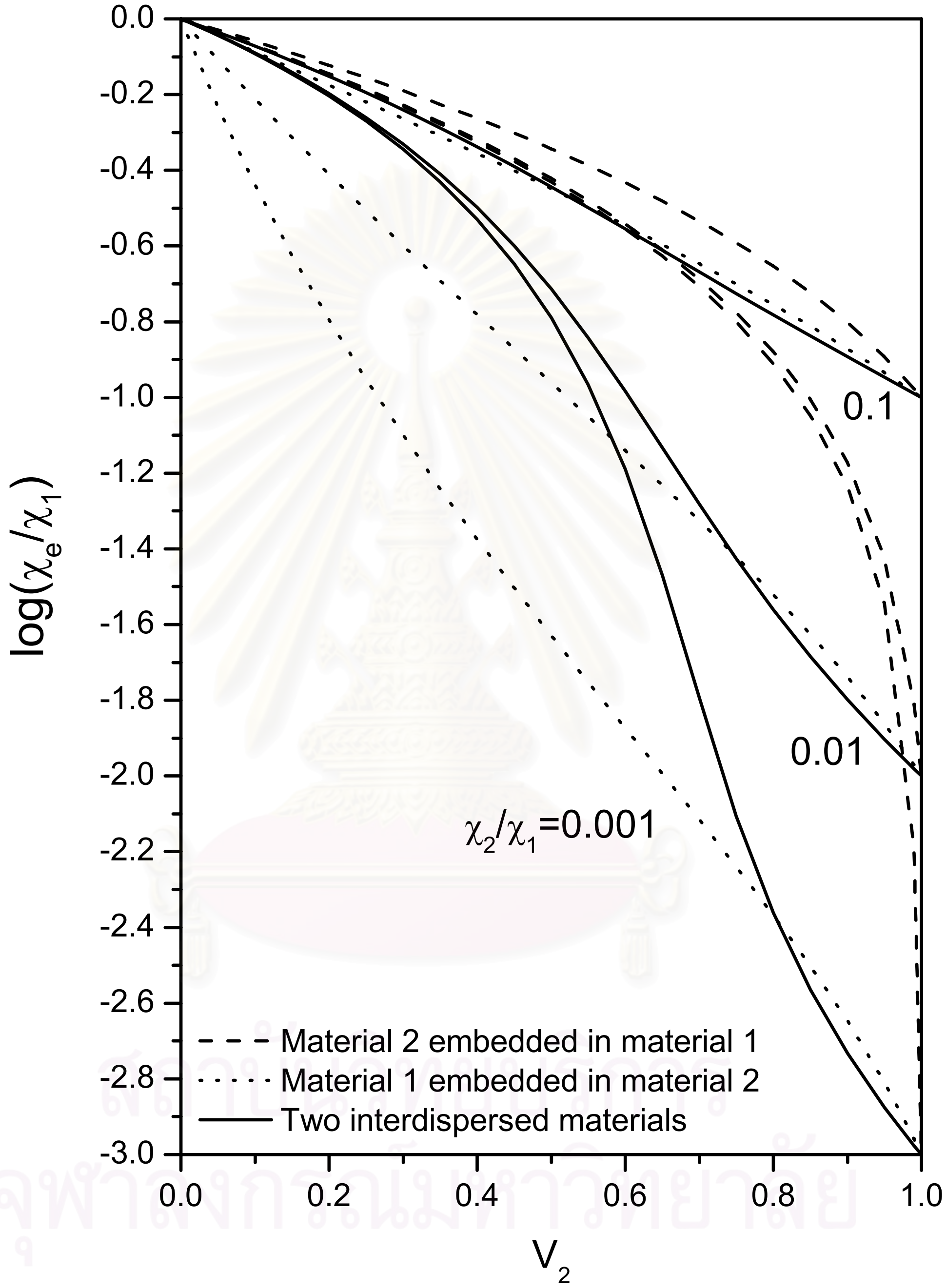

Figure 4.5: Bounds of the effective nonlinear coefficient by the variational method ($\frac{\chi_2}{\chi_1} = 0.1$, 0.01 and 0.001).

Fig. 4.4 and Fig. 4.5 shows the lower (- - -) and upper ($\cdots$) bounds of the effective nonlinear coefficient calculated by Eqs. (4.21) and (4.22), respectively. They are the best possible lower and upper bounds for a statistically homogeneous and isotropic two-phase composite materials, when the only geometrical information available is inclusion packing fractions. In addition, a remainder middle (——) presented for $log(\frac{\chi_e}{\chi_1})$ calculated by Eq. (4.37).

Chapter V

Effective Nonlinear Coefficient by Decoupling Technique

In this chapter, the decoupling technique presented in chapter 2 will be applied to determine the effective nonlinear coefficient (χ_e) of strongly nonlinear dielectric composites. We first begin to investigate the linear response in the range of dilute inclusion packing fractions, then extend to the case of arbitrary inclusion packing fractions. Next, by using the decoupling technique, χ_e including the lower and upper bounds are determined for arbitrary inclusion packing fractions.

5.1 Effective Linear Coefficient

To determine the effective linear coefficient (ε_e) of the composite which contains dilute inclusions, a single inclusion model was assumed [15]. As shown in Fig. 5.1, an inclusion of radius a with linear coefficient ε_2 is embedded in a host medium with linear coefficient ε_1 and an external uniform electric field $\left(\overrightarrow{E}_0\right)$ is applied to study the dielectric response.

To determine the electric potentials, according to Eq. (2.14), the Laplace equation in spherical coordinate is used:

$$\overrightarrow{\nabla}^2\varphi = 0. \tag{5.1}$$

The electric potentials satisfying the boundary conditions at $r = 0$ and

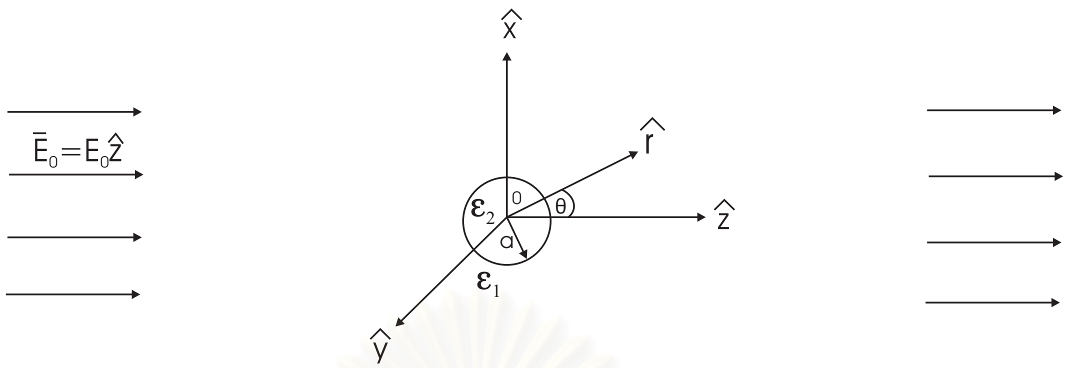

Figure 5.1: The single inclusion model.

$r \to \alpha$ are

$$\begin{aligned} \varphi_2(r,\theta) &= -cE_0 r\cos\theta, \quad 0 \le r \le a & (5.2) \\ \varphi_1(r,\theta) &= -E_0(r - \frac{b}{r^2})\cos\theta, \quad r \ge a & (5.3) \end{aligned}$$

where φ_2 and φ_1 are the electric potentials in the inclusion and the host medium, respectively.

The constants c and b in Eqs. (5.2) and (5.3) are determined by using the continuities of the tangential component of $\overrightarrow{E}$ and the normal component of $\overrightarrow{D}$ at the inclusion surface, hence

$$b = \frac{\varepsilon_2 - \varepsilon_1}{\varepsilon_2 + 2\varepsilon_1} \text{ and } c = \frac{3\varepsilon_1}{\varepsilon_2 + 2\varepsilon_1}. \tag{5.4}$$

In order to determine ε_e, the energy integral is used [15, 3], which is

$$\varepsilon_e = \frac{1}{E_0^2 V}\int_V \varepsilon_\alpha \left|\overrightarrow{E}_\alpha\right|^2 dV, \ \alpha = 1, \ 2, \tag{5.5}$$

where the subscripts 1, and 2 are referred to the host medium and the inclusion, respectively.

From $\overrightarrow{E}_\alpha = -\overrightarrow{\nabla}\varphi_\alpha$ and using Eqs. (5.2)-(5.4), Eq. (5.5) gives

$$\varepsilon_e = \varepsilon_1 + v_2 \frac{3\varepsilon_1(\varepsilon_2 - \varepsilon_1)}{\varepsilon_2 + 2\varepsilon_1}, \tag{5.6}$$

where v_2 is the inclusion packing fraction.

Eq. (5.6) is a well know result which is obtained by assuming that the inclusion volume is much less than the composite volume. Hence, it has a limit on the practical application.

As presentation in Chapter 3, the more general EMT model was used to determined the effective linear coefficient (ε_e) and the result is given by Eq. (3.26) which is valid for composites having arbitrary inclusion packing fractions,

$$\varepsilon_e = \varepsilon_1 \left[1 + \frac{v_2}{\dfrac{\varepsilon_1}{\varepsilon_2 - \varepsilon_1} + \dfrac{v_1}{3}} \right], \tag{5.7}$$

where $v_1 = 1 - v_2$.

The relative effective linear coefficients $\left(\dfrac{\varepsilon_e}{\varepsilon_1}\right)$, calculated from Eqs. (5.6) and (5.7), are plotted against the relative linear coefficient $\left(\dfrac{\varepsilon_2}{\varepsilon_1}\right)$ for various inclusion packing fractions (v_2) in Fig. 5.2.

From Fig. 5.2, the relative effective nonlinear coefficients $\dfrac{\varepsilon_e}{\varepsilon_1}$ calculated by using the single inclusion model with Eq. (5.6) and the EMT model with Eq. (5.7), are presented as dash (- - -) and solid lines (——), respectively. The more inclusion packing fraction (v_2) increases, the greater distinction of $\dfrac{\varepsilon_e}{\varepsilon_1}$ calculated from Eqs. (5.6) and (5.7) are observed. However, for v_2 is less than 0.1, the $\dfrac{\varepsilon_e}{\varepsilon_1}$ values obtained by using both models are approximately corespondent. Therefore, the single inclusion model is for inclusion packing fraction less than 0.1.

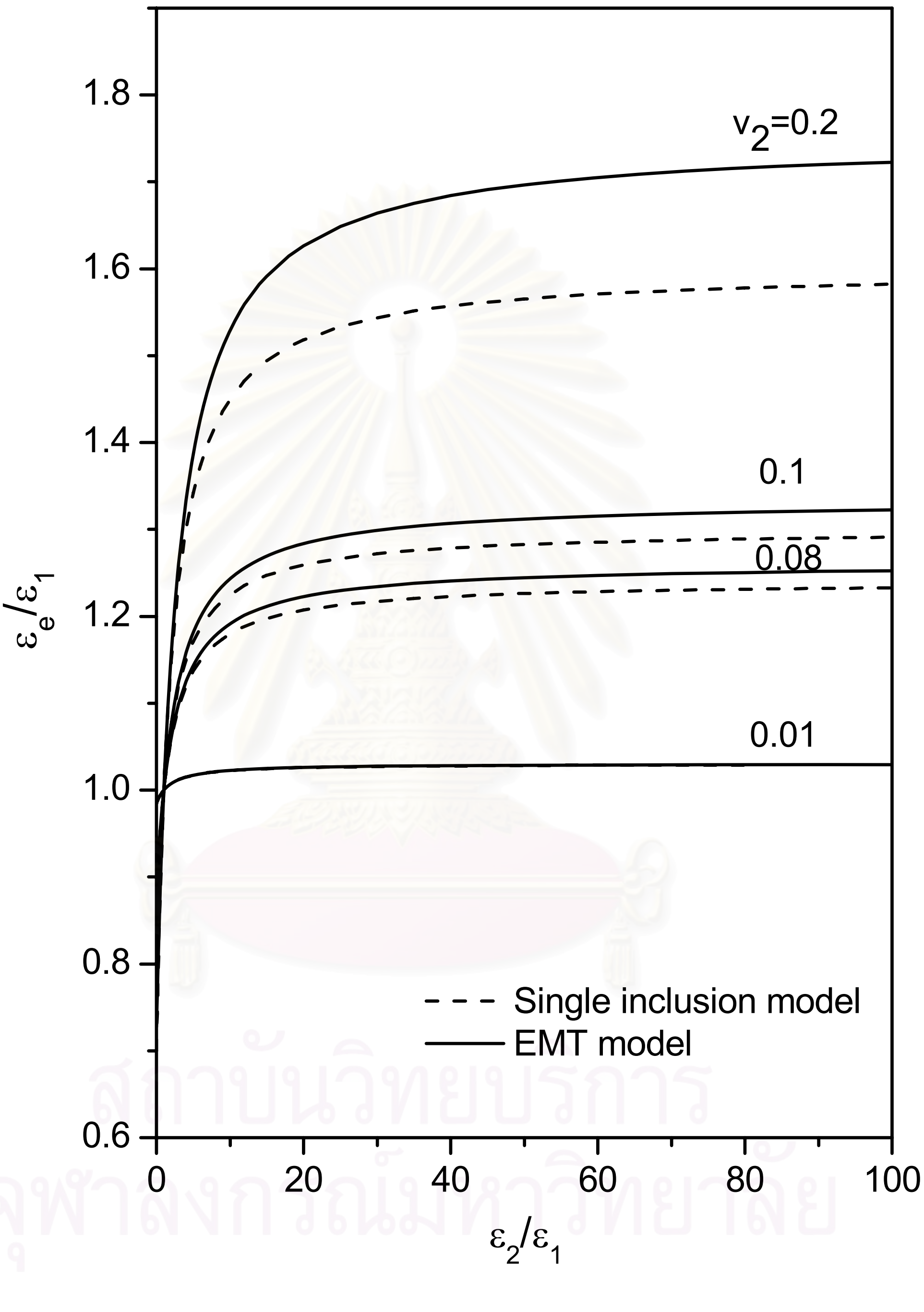

Figure 5.2: Comparison of relative effective linear coefficients $\left(\dfrac{\varepsilon_e}{\varepsilon_1}\right)$ obtained by using the single inclusion model and the EMT model for varying inclusion packing fractions $(v_2 = 0.01,\ 0.08,\ 0.1 \text{ and } 0.2)$.

5.2 Effective Nonlinear Coefficient

Next, extending to a strongly nonlinear dielectric composite, the effective nonlinear coefficient (χ_e) of the composite has been evaluated by using the decoupling technique.

In 1996, Yu and Yuen [17] applied the decoupling technique to strongly nonlinear dielectric composite having dilute inclusion packing fractions. In their work, the single inclusion model was assumed, so the obtained results have limits on practical applications.

To generalize the decoupling technique in determining χ_e for arbitrary inclusion packing fractions, the calculation begins with an estimation of χ_e from Eq. (2.29):

$$\chi_e = \frac{v_1\chi_1 \langle E_1^4\rangle}{E_0^4} + \frac{v_2\chi_2 \langle E_2^4\rangle}{E_0^4}, \tag{5.8}$$

where $\langle E^4\rangle$ is the volume average of electric field to the fourth power, and subscripts $1, 2$ are referred to the host medium and the inclusion, respectively.

We invoke the decoupling technique [26] by ignoring the fluctuations of the local electric field,

$$\left\langle (E_\alpha^2 - \left\langle E_\alpha^2\right\rangle)^2\right\rangle = \left\langle E_\alpha^4\right\rangle - \left\langle E_\alpha^2\right\rangle^2 = 0,\ \alpha = 1, 2,$$

or $\langle E_\alpha^4\rangle$ is approximated by

$$\left\langle E_1^4\right\rangle = \left\langle E_1^2\right\rangle^2, \tag{5.9}$$

and also

$$\left\langle E_2^4\right\rangle = \left\langle E_2^2\right\rangle^2. \tag{5.10}$$

Now, Eq. (5.8) is replaced by using Eqs. (5.9) and (5.10), hence

$$\chi_e = \frac{v_1\chi_1 \left\langle E_1^2\right\rangle^2}{E_0^4} + \frac{v_2\chi_2 \left\langle E_2^2\right\rangle^2}{E_0^4}, \tag{5.11}$$

where E_2^2 and E_1^2 are the mean square of electric fields within the inclusion and host medium, respectively. v_2 and v_1 are the packing fractions of inclusion and host medium, respectively.

Eq. (5.11) implies that if we know $\langle E_1^2 \rangle$ and $\langle E_2^2 \rangle$, the effective nonlinear coefficient χ_e is obtained.

To obtain the mean square of electric fields $\langle E_1^2 \rangle$ and $\langle E_2^2 \rangle$, the derivatives of Eq. (5.7) are evaluated for $\dfrac{\partial \varepsilon_e}{\partial \varepsilon_1}$ and $\dfrac{\partial \varepsilon_e}{\partial \varepsilon_2}$, then substituted into Eqs. (2.36) and (2.37), which give

$$\begin{aligned} \left\langle E_1^2 \right\rangle &= \frac{1}{1-v_2} \frac{\partial \varepsilon_e}{\partial \varepsilon_1} E_0^2, \\ &= \left[1 - \frac{v_2 \varepsilon_1 \left(\frac{\varepsilon_1}{(\varepsilon_2-\varepsilon_1)^2} + \frac{1}{\varepsilon_2 - \varepsilon_1} \right)}{\left(\frac{\varepsilon_1}{\varepsilon_2-\varepsilon_1} + \frac{1-v_2}{3} \right)^2} + \frac{v_2}{\frac{\varepsilon_1}{\varepsilon_2-\varepsilon_1} - \frac{1-v_2}{3}} \right] E_0^2, \end{aligned} \tag{5.12}$$

and also

$$\begin{aligned} \left\langle E_2^2 \right\rangle &= \frac{1}{v_2} \frac{\partial \varepsilon_e}{\partial \varepsilon_2} E_0^2, \\ &= \left[\frac{\varepsilon_1^2}{(\varepsilon_2-\varepsilon_1)^2 \left(\frac{\varepsilon_1}{\varepsilon_2-\varepsilon_1} + \frac{1-v_2}{3} \right)^2} E_0^2 \right]. \end{aligned} \tag{5.13}$$

By using the decoupling technique, the relations between the linear and nonlinear coefficients from Eq. (2.39) with $\varepsilon_1 = \chi_1 \langle E_1^2 \rangle = \chi_1 \beta$ and Eq. (2.40) with $\varepsilon_2 = \chi_2 \langle E_2^2 \rangle = \chi_2 \alpha$, thus the Eqs. (5.12) and (5.13) are modified to be

$$\alpha = \frac{\beta^2}{(\beta - \alpha y)^2 \left(\frac{\beta}{\alpha y - \beta} + \frac{1-v_2}{3} \right)^2}, \tag{5.14}$$

$$\beta = \left(\frac{1}{1-v_2} 1 - \frac{v_2 \beta \left(\frac{1}{\alpha y - \beta} + \frac{\beta}{(\alpha y - \beta)^2} \right)}{\left(\frac{\beta}{\alpha y - \beta} + \frac{1-v_2}{3} \right)^2} + \frac{v_2}{\frac{\beta}{\alpha y - \beta} + \frac{1-v_2}{3}} \right), \tag{5.15}$$

where $\alpha = \langle E_2^2 \rangle$, $\beta = \langle E_1^2 \rangle$, $y = \dfrac{\chi_2}{\chi_1}$ and v_2 is the inclusion packing fraction.

Now, Eqs. (5.14) and (5.15) can be solved self-consistently for $\alpha = \langle E_2^2 \rangle$ and $\beta = \langle E_1^2 \rangle$. We have to determine the unknowns α and β in terms of parameters y and v_2. Because of the complication, α and β can not be solved in closed form. To keep off complication of the determination on α and β in explicit analytical forms, specific values of y and v_2 will be given first, then Eqs. (5.14) and (5.15) are solved numerically for α and β. Let

$$\alpha^* = \alpha(v_2, y), \tag{5.16}$$

$$\beta^* = \beta(v_2, y), \tag{5.17}$$

a^* and β^* represent the numerical solutions of α and β evaluated at given values of v_2 and y.

Without loss of generality, we set $E_0 = 1$ and replace α and β into Eq. (5.11) by a^* and β^*, then the relative effective nonlinear coefficient $\frac{\chi_e}{\chi_1}$ as a function of v_2 and y is obtained:

$$\frac{\chi_e}{\chi_1} = F(v_2, y) = (1 - v_2)\chi_1 (\beta^*)^2 + v_2\chi_2 (\alpha^*)^2 . \tag{5.18}$$

In general, the range of parameter $y = \frac{\chi_2}{\chi_1}$ may varies from very low contrast to vary high contrast, that is from y equal to zero up to thousands. In this work, because at high contrast $\left(\frac{\chi_2}{\chi_1} > 100\right)$, the increase of $\frac{\chi_2}{\chi_1}$ rarely effect the $\frac{\chi_e}{\chi_1}$ value, so we vary $\frac{\chi_2}{\chi_1}$ from 0 to 100. In addition, v_2 is set equal to 0.01, 0.08, 0.1 and 0.2, then the relative effective nonlinear coefficient $\frac{\chi_e}{\chi_1}$ for specific inclusion packing fractions are obtained by using the Mathematica program (see appendix C).

To determine the distinction between the results of the EMT and the work of Yu and Yuen, the relative effective nonlinear coefficients $\left(\frac{\chi_e}{\chi_1}\right)$ calculated by using the EMT and the results of Yu and Yuen [17] are plotted against the relative nonlinear coefficients $\left(\frac{\chi_2}{\chi_1}\right)$ for various values of inclusion packing fraction (v_2). As shown in Fig. 5.3, v_2 is set equal to 0.01, 0.08, 0.1 and 0.2

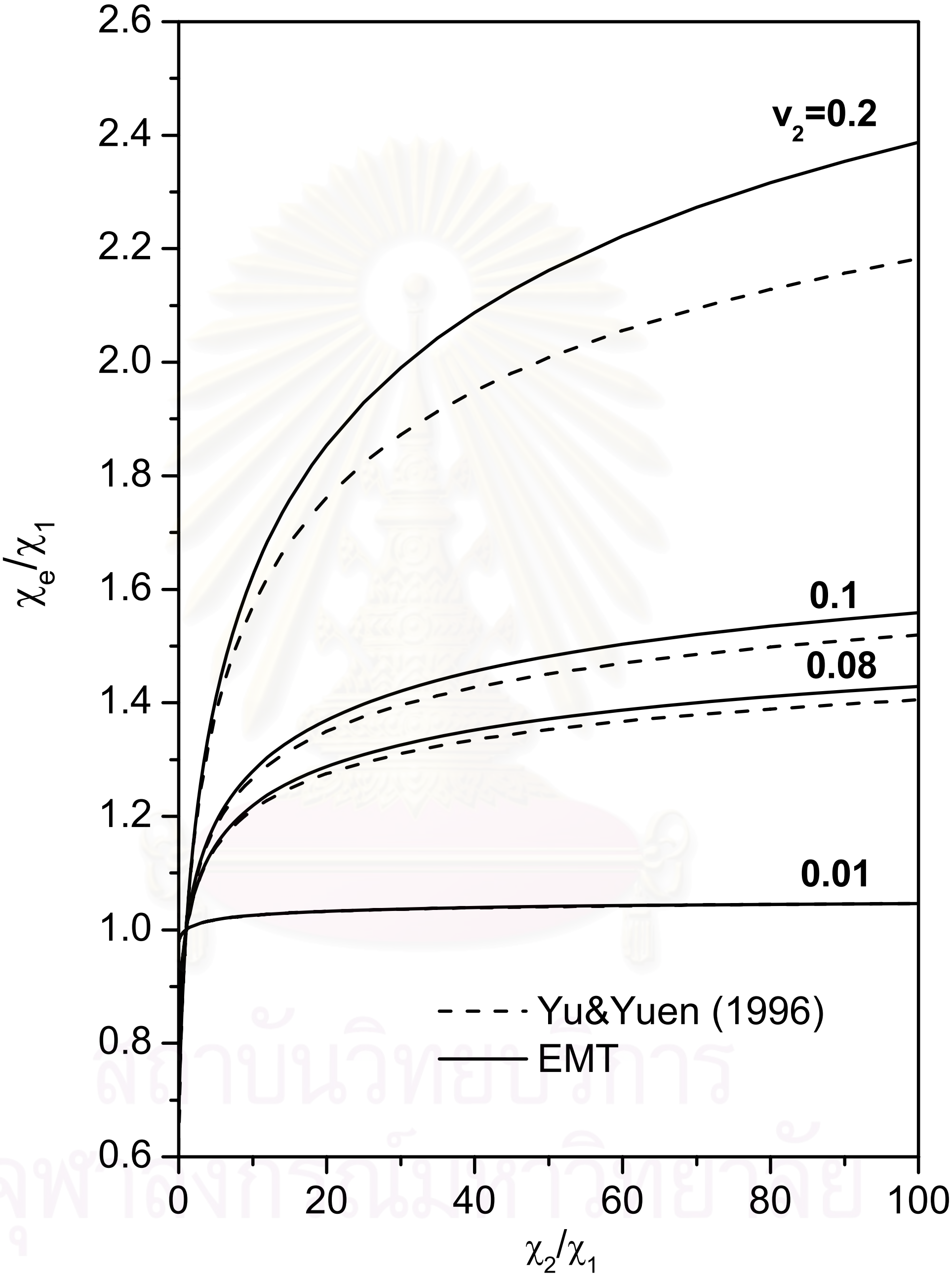

Figure 5.3: Comparison of relative effective nonlinear coefficient for various inclusion packing fractions determined by using the decoupling technique.

From Fig. 5.3, the relative effective nonlinear coefficients $\left(\frac{\chi_e}{\chi_1}\right)$, calculated by Yu and Yuen and the EMT, are presented as dash (- - -) and solid lines (—–), respectively. $\frac{\chi_e}{\chi_1}$ increases rapidly with increasing $\frac{\chi_2}{\chi_1}$, however a higher values of $\frac{\chi_2}{\chi_1}$, the increase of $\frac{\chi_2}{\chi_1}$ rarely affects the $\frac{\chi_e}{\chi_1}$ values. The distinction of $\frac{\chi_e}{\chi_1}$ obtained by Yu and Yuen and the EMT increases with increasing of the inclusion packing fractions (v_2). However, for v_2 is less than 0.1, the $\frac{\chi_e}{\chi_1}$ values obtained by both results are corespondent which confirms our results using the EMT model and also shows the validity of the single inclusion model if $v_2 < 0.1$.

Further, the calculation is separated into three cases based on different kinds of composite microstructure geometry. First, the inclusions of material 2 are embedded in material 1. Second, in contrary, inclusions of material 1 are embedded in material 2 and the last case of two interdispersed materials.

5.2.1 Inclusions of Material 2 Embedded in Material 1

In fact, we has already considered this case in section 5.2. The effective nonlinear coefficient (χ_e) was given by Eq. (5.18) for arbitrary inclusion packing fractions (v_2),

$$\frac{\chi_e}{\chi_1} = F(v_2, \frac{\chi_2}{\chi_1}) = (1 - v_2)\chi_1 \left(\beta^*\right)^2 + v_2\chi_2 \left(\alpha^*\right)^2. \tag{5.19}$$

where v_2 is the inclusion packing fraction with α^* and β^* given by Eqs. (5.16) and (5.17).

5.2.2 Inclusions of Material 1 Embedded in Material 2

This case is opposite to the case considered in subsection 5.2.1. To obtain $\frac{\chi_e}{\chi_1}$, the process of calculations are similar to previous case. As explained in section 4.2.2, we report the results in terms of parameters v_2 and $\frac{\chi_2}{\chi_1}$. We note that in this case v_2 is the packing fraction of material 2 which is the host medium,

$$\frac{\chi_e}{\chi_1} = F'(v_2, \frac{\chi_2}{\chi_1}). \tag{5.20}$$

The effective nonlinear coefficients (χ_e) are calculated for arbitrary values of v_2 by using the Mathematica program (see appendix C).

5.2.3 Two Interdispersed Materials

For two interdispersed materials, inclusions of phase 1 and 2 are randomly mixed together. The composite can not be clearly specified which phase is the inclusion (or host medium). The theoretical model is assumed that an inclusion with nonlinear coefficient χ_2 (or χ_1) is embedded in an effective medium with effective nonlinear coefficient χ_e. This is equivalent to the EMT model with $c' = 1$. To obtain the $\langle E_1^2 \rangle$ and $\langle E_2^2 \rangle$, we have to know ε_e which was derived in Chapter 3. From Eqs. (3.32) or (3.33),

$$v_1(\frac{\varepsilon_1 - \varepsilon_e}{\varepsilon_1 + 2\varepsilon_e}) + v_2(\frac{\varepsilon_2 - \varepsilon_e}{\varepsilon_2 + 2\varepsilon_e}) = 0, \tag{5.21}$$

where v_1 and v_2 are the packing fractions of materials 1 and 2, respectively.

Eq. (5.21) is solved analytically for ε_e. The solution has two real roots of opposite signs, a negative root gives a negative ε_e which is physically meaningless. Only the positive root is considered, which is

$$\varepsilon_e = \frac{1}{4}\left[2\varepsilon_1 - 3v_2\varepsilon_1 - \varepsilon_2 + 3v_2\varepsilon_2 + \sqrt{8\varepsilon_1\varepsilon_2 + (2\varepsilon_1 - 3v_2\varepsilon_1 - \varepsilon_2 + 3v_2\varepsilon_2)^2}\right]. \tag{5.22}$$

To obtain the mean square of electric fields $\langle E_1^2 \rangle$ and $\langle E_2^2 \rangle$. The Mathematica Program is used to determine the derivatives of Eq. (5.22) for $\dfrac{\partial \varepsilon_e}{\partial \varepsilon_1}$ and $\dfrac{\partial \varepsilon_e}{\partial \varepsilon_2}$, then substituted into Eqs. (2.36) and (2.37). We obtain

$$\langle E_1^2 \rangle = \frac{1}{4(1-v_2)}\left[2 - 3v_2 + \frac{8\varepsilon_2 + 2(2-3v_2)(2\varepsilon_1 - 3v_2\varepsilon_1 - \varepsilon_2 + 3v_2\varepsilon_2)}{2\sqrt{8\varepsilon_1\varepsilon_2 + (2\varepsilon_1 - 3v_2\varepsilon_1 - \varepsilon_2 + 3v_2\varepsilon_2)^2}}\right], \tag{5.23}$$

$$\langle E_2^2 \rangle = \frac{1}{4v_2}\left[-1 + 3v_2 + \frac{8\varepsilon_1 + 2(-1+3v_2)(2\varepsilon_1 - 3v_2\varepsilon_1 - \varepsilon_2 + 3v_2\varepsilon_2)}{2\sqrt{8\varepsilon_1\varepsilon_2 + (2\varepsilon_1 - 3v_2\varepsilon_1 - \varepsilon_2 + 3v_2\varepsilon_2)^2}}\right]. \tag{5.24}$$

By using the decoupling technique, the relations between the linear and nonlinear coefficients from Eq. (2.39) and (2.40), with $\varepsilon_1 = \chi_1 \langle E_1^2 \rangle = \chi_1\beta$, and

$\varepsilon_2 = \chi_2 \langle E_2^2 \rangle = \chi_2 \alpha$, thus Eqs. (5.23) and (5.24) become

$$\alpha = \frac{1}{4v_2}\left[-1 + 3v_2 + \frac{8\varepsilon_1 + 2(-1+3v_2)\,(2\beta - 3v_2\beta - y\alpha + 3v_2\alpha y)}{2\sqrt{8y\alpha\beta + (2\beta - 3v_2\beta - y\alpha + 3v_2\alpha y)^2}}\right], \tag{5.25}$$

$$\beta = \frac{1}{4(1-v_2)}\left[2 - 3v_2 + \frac{8\varepsilon_1 + 2(2-3v_2)\,(2\beta - 3v_2\beta - y\alpha + 3v_2\alpha y)}{2\sqrt{8y\alpha\beta + (2\beta - 3v_2\beta - y\alpha + 3v_2\alpha y)^2}}\right]. \tag{5.26}$$

where $\alpha = \langle E_2^2 \rangle$, $\beta = \langle E_1^2 \rangle$, $y = \dfrac{\chi_2}{\chi_1}$ and v_2 is the packing fraction of material 2.

Now, Eqs. (5.25) and (5.26) can be solved self-consistently for $\alpha = \langle E_2^2 \rangle$ and $\beta = \langle E_1^2 \rangle$. We have to determine the unknowns α and β in terms of $\dfrac{\chi_2}{\chi_1}$ and v_2. Because of the complication, α and β can not be solved in closed form. To keep off complication of the determination of α and β in explicit analytical forms, specific values of $\dfrac{\chi_2}{\chi_1}$ and v_2 will be given first, then Eqs. (5.25) and (5.26) are solved numerically for α and β. Let a^* and β^* be the numerical solutions of α and β evaluated at given values of v_2 and $\dfrac{\chi_2}{\chi_1}$. We replace a^* and β^* into Eq. (5.11), then the relative effective nonlinear coefficient $\left(\dfrac{\chi_e}{\chi_1}\right)$ is obtained as a function of packing fraction of material 2 (v_2) and the relative nonlinear coefficient $\dfrac{\chi_2}{\chi_1}$:

$$\frac{\chi_e}{\chi_1} = F(v_2, \frac{\chi_2}{\chi_1}, \alpha^*, \beta^*). \tag{5.27}$$

To determine the bounds of χ_e, the relative effective nonlinear coefficients are plotted as $log(\dfrac{\chi_e}{\chi_1})$ against the packing fractions of material 2 (v_2) for various values of $\dfrac{\chi_2}{\chi_1}$. As shown in Fig. 5.4 and 5.5, $\dfrac{\chi_2}{\chi_1}$ is set equal to 10, 100, 1000, 0.1, 0.01, and 0.001.

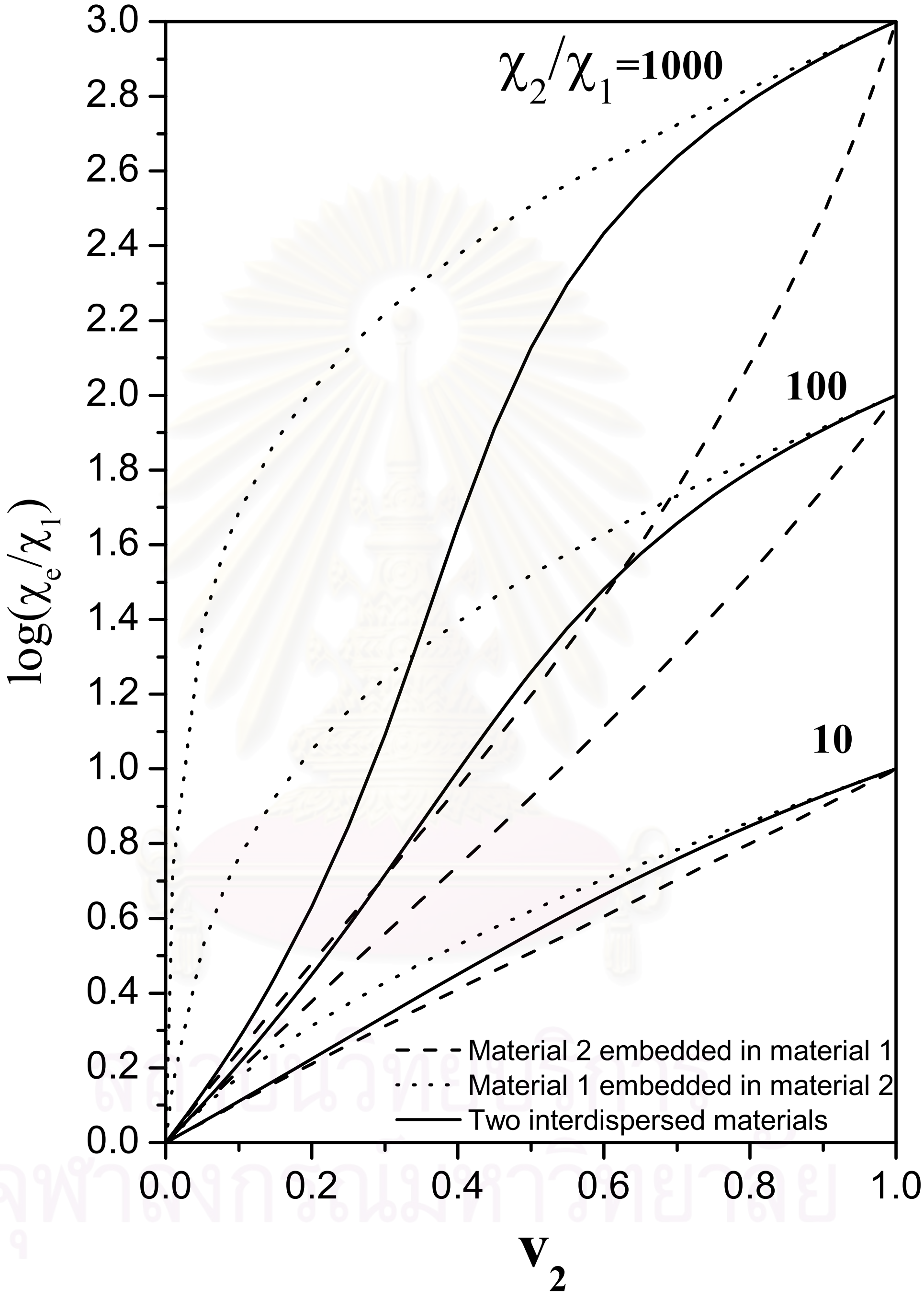

Figure 5.4: Bounds of the effective nonlinear coefficient by the decoupling technique ($\frac{\chi_2}{\chi_1} = 10$, 100 and 1000).

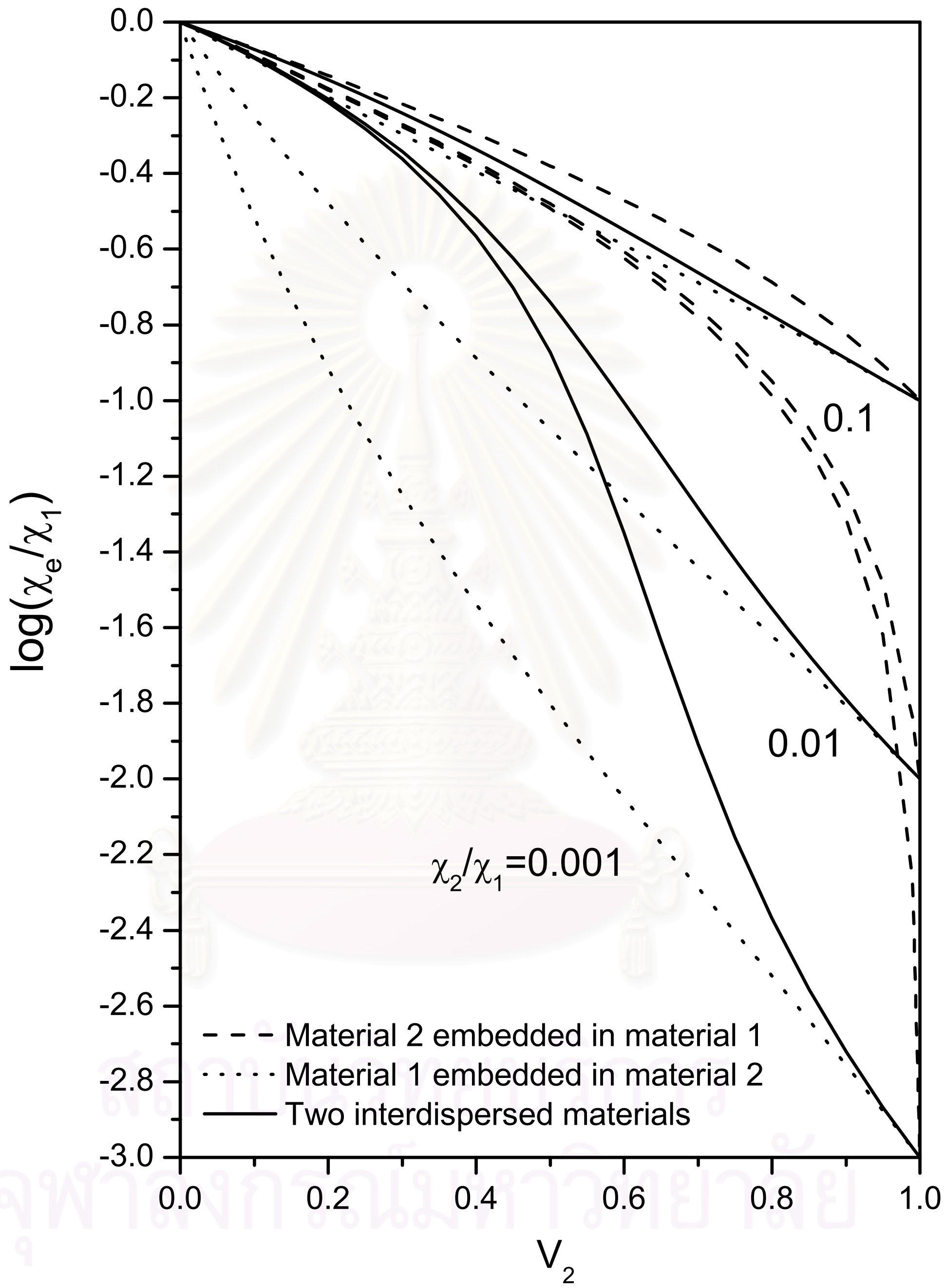

Figure 5.5: Bounds of the effective nonlinear coefficient by the decoupling technique ($\frac{\chi_2}{\chi_1} = 0.1$, 0.01 and 0.001).

Figs. 5.4 and 5.5 show the lower (- - -) and upper ($\cdots$) bounds of χ_e calculated from Eqs. (5.19) and (5.20), respectively. They are the best possible lower and upper bounds for a statistically homogeneous and isotropic two-phase composite materials, when the only geometrical information available is inclusion packing fractions. The remainder middle (—–) calculated from Eq. (5.27), which represents the effective medium approximation. These are similar to the work of Yu, Hui and Lee [26]. The more the difference between the nonlinear coefficients of the materials 1 and 2, the more distinction between two bounds are observed. These results will be compared with those of the variational method for their verification and reliability of the decoupling technique results in the next section.

For detail explanation, we consider the bounds of χ_e for $\frac{\chi_2}{\chi_1} = 100$. As shown in Fig. 5.6, $log(\frac{\chi_e}{\chi_1})$ increases with increasing v_2 because χ_2 is larger than χ_1. For $v_2 = 0$, all curves coincide at $log(\frac{\chi_e}{\chi_1})$ equal to 0 because χ_e becomes χ_1. On the other hand, for $v_2 = 1$, all curves coincide at $log(\frac{\chi_e}{\chi_1})$ equal to 2 because χ_e becomes χ_2 ($log(\frac{\chi_e}{\chi_1}) = log(\frac{\chi_2}{\chi_1}) = log100 = 2$). For $v_2 = 0.4$, the volume ratio of material 2 to material 1 is 4 : 6, $log(\frac{\chi_e}{\chi_1})$ calculated from Eqs. (5.19) and (5.20) are 0.74, and 1.39, respectively. $\frac{\chi_e}{\chi_1}$ calculated from Eq. (5.20) are obtained by assuming that particles or inclusions of material 1 are randomly dispersed in the host medium of material 2. While Eq. (5.19) explains the material with opposite microstructure which is composed of material 2 are randomly dispersed in the host medium of material 1. For the same ratio of materials 1 and 2, and $\chi_2 > \chi_1$, the effective nonlinear coefficient (χ_e) of the composite with material 2 being the host medium is larger than χ_e of the composite case with material 2 being inclusions. The ratio between χ_e of the former to the latter case is about 5.6 for $\frac{\chi_2}{\chi_1} = 100$ and $v_2 = 0.4$.

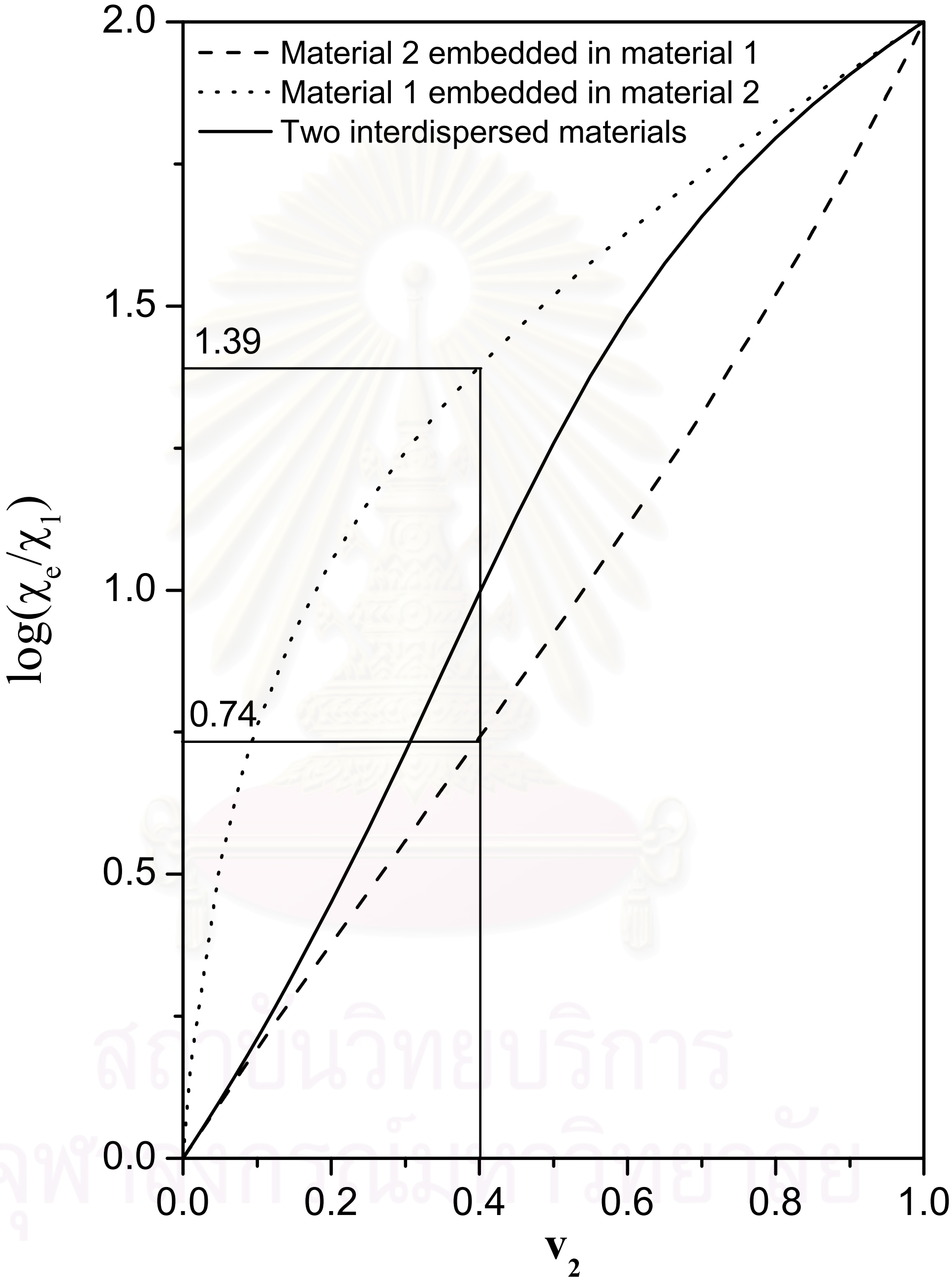

Figure 5.6: Bounds of the effective nonlinear coefficient for $\frac{\chi_2}{\chi_1} = 100$.

5.3 Reliability and Utilization of Decoupling Technique

In order to determine the validity or reliability of the decoupling technique, $log(\frac{\chi_e}{\chi_1})$ calculated by using the decoupling technique are compared with those calculated by using the variational method as shown in Figs. 5.7 - 5.14.

Figs. 5.7 - 5.9 are presented for $\frac{\chi_2}{\chi_1} = 10$, 100, and 1000, respectively. It is found that $log(\frac{\chi_e}{\chi_1})$ calculated by using both methods significantly differ as increasing $\frac{\chi_2}{\chi_1}$. From Fig. 5.9 with $\frac{\chi_2}{\chi_1} = 1000$, the maximum difference reaches to about 30%. Figs. 5.10 - 5.12 are presented for $\frac{\chi_2}{\chi_1} = 0.1$, 0.01 and 0.001, respectively. From Fig. 5.12 with $\frac{\chi_2}{\chi_1} = 0.001$, the maximum difference reaches to about 12%. Therefore, it is concluded that results of decoupling technique are comparable to those of the variational method at only small values of $\frac{\chi_2}{\chi_1}$.

Now, we consider own intersections between $\log(\frac{\chi_e}{\chi_1})$ obtained by using the variational method and the decoupling technique of two interdispersed materials in Fig. 5.7-5.12. Before the intersections, $\log(\frac{\chi_e}{\chi_1})$ from the variational method are less than those from the decoupling technique, after that $\log(\frac{\chi_e}{\chi_1})$ from the variational method are larger than those from the decoupling technique. The intersections in terms of v_2 are approximately $v_2 = 0.5$, 0.3 and 0.2 for $\frac{\chi_2}{\chi_1} = 10$, 100 and 1000, respectively. Therefore, the intersections of $\frac{\chi_2}{\chi_1} > 1$ decrease with increasing $\frac{\chi_2}{\chi_1}$. For $\frac{\chi_2}{\chi_1} = 0.1$, 0.01 and 0.001, the intersections are approximately $v_2 = 0.5$, 0.75 and 0.8, respectively. So, the intersections of $0 < \frac{\chi_2}{\chi_1} < 1$ increase with decreasing $\frac{\chi_2}{\chi_1}$. We expect that the intersections may be caused from the determination on χ_e by using the variational method, which also occur in the work of Jitrin [16].

For material 2 embedded in material 1, we report $\frac{\chi_e}{\chi_1}$ by varying $\frac{\chi_2}{\chi_1}$ for lower values of v_2 in Figs. 5.13. From Fig. 5.13, $\frac{\chi_e}{\chi_1}$ calculated by using the variational method (Eq. (4.21)), and the decoupling technique (Eq. (5.19)) are compared for inclusion packing fractions $v_2 = 0.01$, 0.08, 0.1 and 0.2. It is found that $\frac{\chi_e}{\chi_1}$ calculated by using both methods are in good agreement only at very dilute packing fractions and lower values of $\frac{\chi_2}{\chi_1}$. It is concluded that the decoupling technique is reliable for $v_2 \leq 0.1$ with $\frac{\chi_2}{\chi_1}$ less than about 10 and also for $v_2 = 0.2$ with $\frac{\chi_2}{\chi_1}$ less than about 5 because these are observed that the difference is about 5%.

For inclusions of material 2 embedded in material 1 and inclusions of material 1 embedded in material 2, Figs. 5.7 - 5.13 show χ_e predicted by using the decoupling techniques are all less than those using the variational method. These confirm the theoretical prediction reported by Yu and Yuen [17] that χ_e(decoupling)$\leq$ χ_e(exact)$\leq$ χ_e(variational). It is clear that both methods are indispensable for estimating χ_e of intractable boundary value-problems. If both results coincide, they both give the exact result. On the other hand, if both results are tight, the estimations are good.

Next, we consider the gap between $\chi_e(decoupling)$ and $\chi_e(variational)$ in Fig. 5.13. By using the data of Fig. 5.13, we now report the data on a logarithmic scale in Fig. 5.14. From Fig. 5.14, the gap between $\chi_e(decoupling)$ and $\chi_e(variational)$ depends on v_2 and $\frac{\chi_2}{\chi_1}$. For the range of $\log(\frac{\chi_2}{\chi_1}) < 0$ or $0 \leq \frac{\chi_2}{\chi_1} < 1$, the gap decreases with decreasing the contrast between χ_2 and χ_1 ($\frac{\chi_2}{\chi_1}$ approaches 1) and becomes zero at $\frac{\chi_2}{\chi_1} = 1$. Similarly, for the range of $\log(\frac{\chi_2}{\chi_1}) > 0$ or $\frac{\chi_2}{\chi_1} > 1$, the gap also decreases with decreasing the contrast $\frac{\chi_2}{\chi_1}$.

The gap between $\chi_e(decoupling)$ and $\chi_e(variational)$ increases with increasing v_2. These may be explained by considering the approximation $\langle E_1^4 \rangle \approx \langle E_1^2 \rangle^2$ used in Eqs. (5.9) and (5.10). Therefore, we calculate the percentage of discrepancy between $\langle E_1^4 \rangle$ and $\langle E_1^2 \rangle^2$ $\left(\Delta\% = \left[\frac{\langle E_1^4 \rangle - \langle E_1^2 \rangle^2}{\langle E_1^4 \rangle} \right] \times 100 \right)$ of the field in the medium $\left(\vec{E}_1 \right)$ from the variational method results. $\Delta\%$ are plotted against $log(\frac{\chi_2}{\chi_1})$ for $v_2 = 0.01$, 0.08, 0.1 and 0.2 in Fig. 5.15. From Fig. 5.15, $\Delta\%$ depend on $\frac{\chi_2}{\chi_1}$ and v_2 similar relation with these of Figs. 5.13 and 5.14.

It is found that $\Delta\% \geq 0$ or $\langle E_1^4 \rangle \geq \langle E_1^2 \rangle^2$ for the whole ranges of $\dfrac{\chi_2}{\chi_1}$ and for all values of v_2. Because $\chi_e(decoupling)$ depends on $\langle E_1^4 \rangle$ (see Eq. (5.8)), we therefore expect our results are less than the expect values satisfying the theoretical relation between $\chi_e(exact)$ and $\chi_e(decoupling)$ as shown in Appendix B that $\chi_e(exact) \geq \chi_e(decoupling)$.

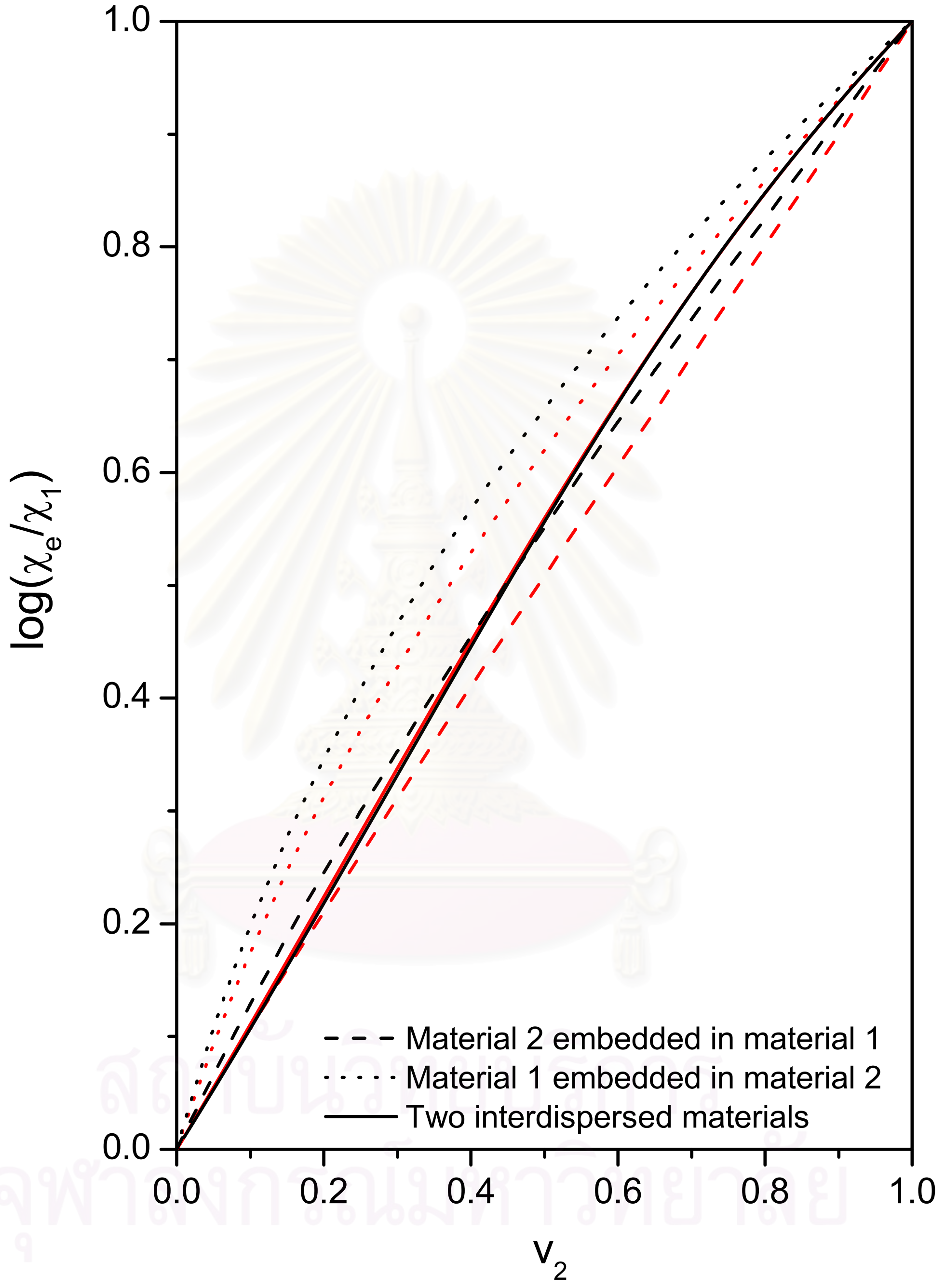

Figure 5.7: Comparison of bounds of $log(\frac{\chi_e}{\chi_1})$ obtained by using the variational method (black lines) and the decoupling technique (red lines) for $\frac{\chi_2}{\chi_1} = 10$.

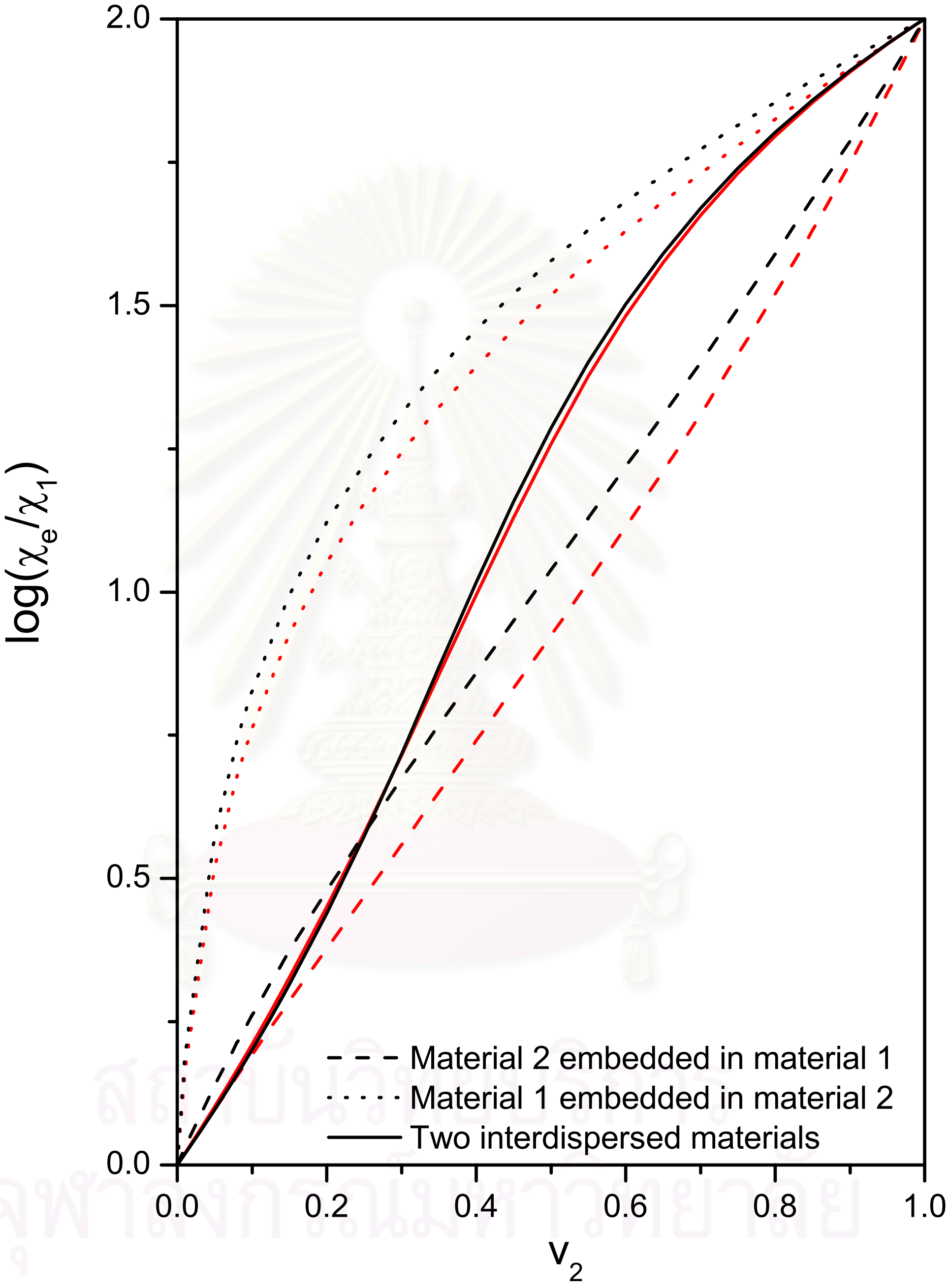

Figure 5.8: Comparison of bounds of $log(\frac{\chi_e}{\chi_1})$ obtained by using the variational method (black lines)and the decoupling technique (red lines) for $\frac{\chi_2}{\chi_1} = 100$.

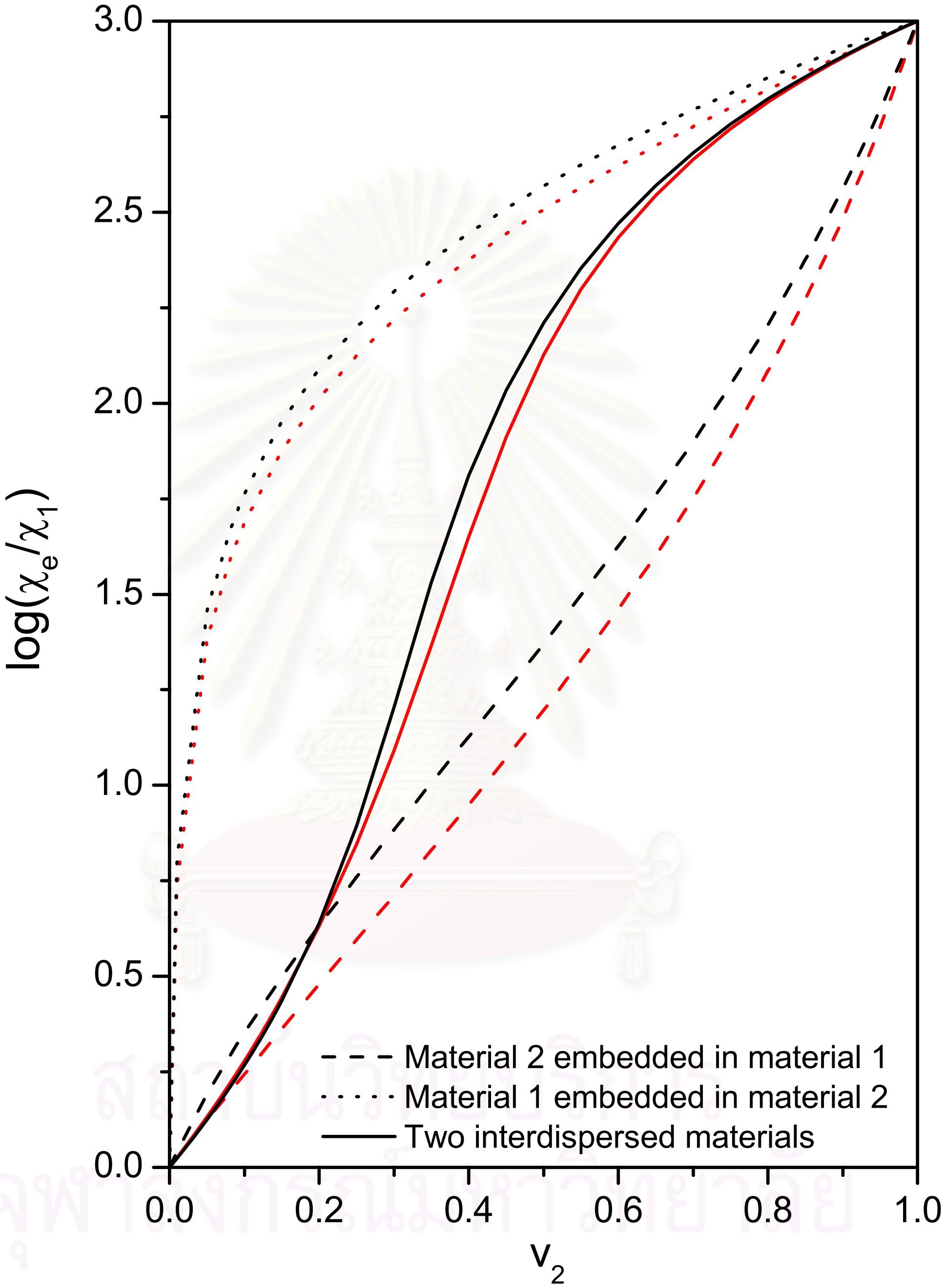

Figure 5.9: Comparison of bounds of $log(\frac{\chi_e}{\chi_1})$ obtained by using the variational method (black lines)and the decoupling technique (red lines) for $\frac{\chi_2}{\chi_1} = 1000$.

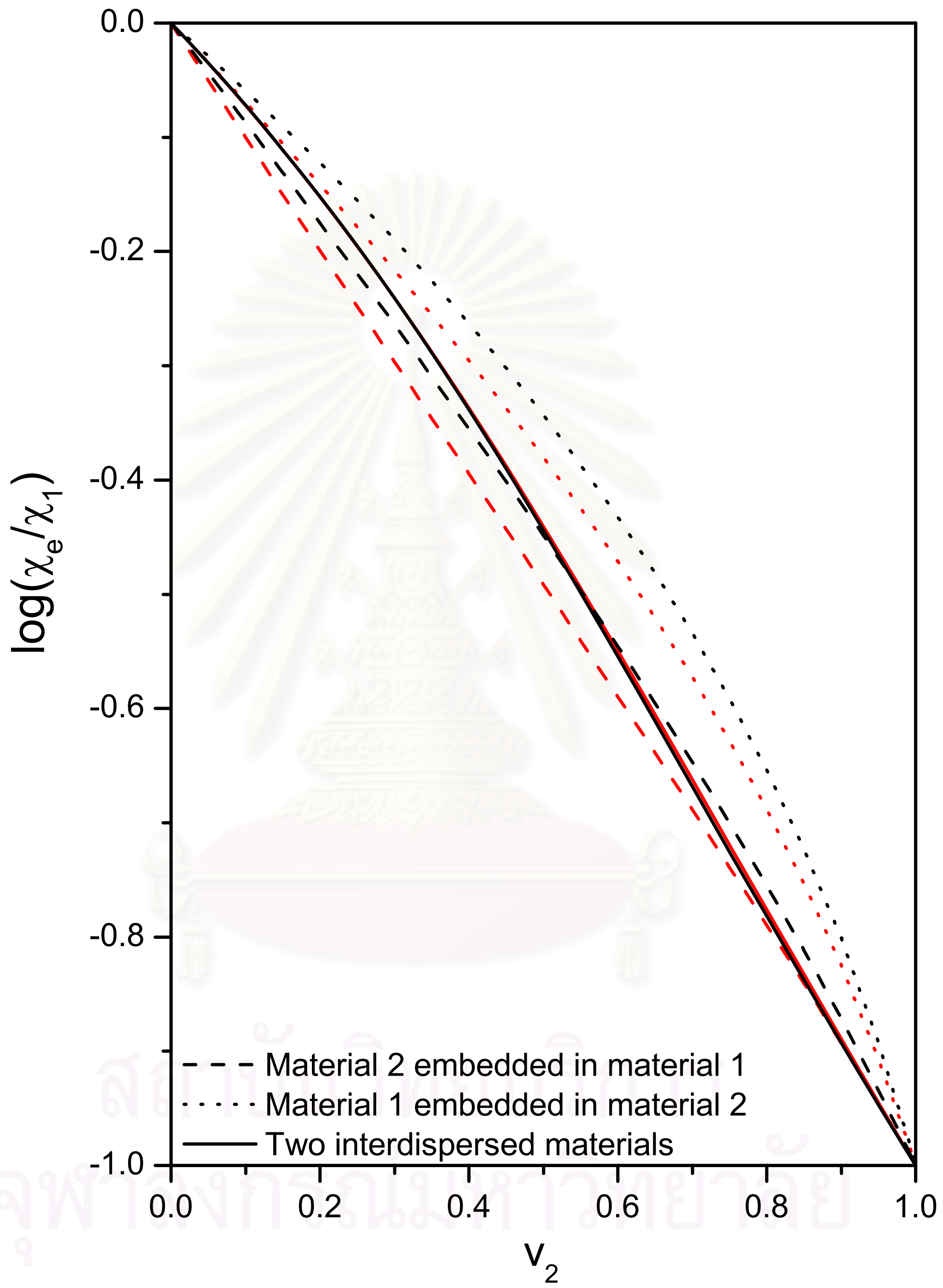

Figure 5.10: Comparison of bounds of $log(\frac{\chi_e}{\chi_1})$ obtained by using the variational method (black lines)and the decoupling technique (red lines) for $\frac{\chi_2}{\chi_1} = 0.1$.

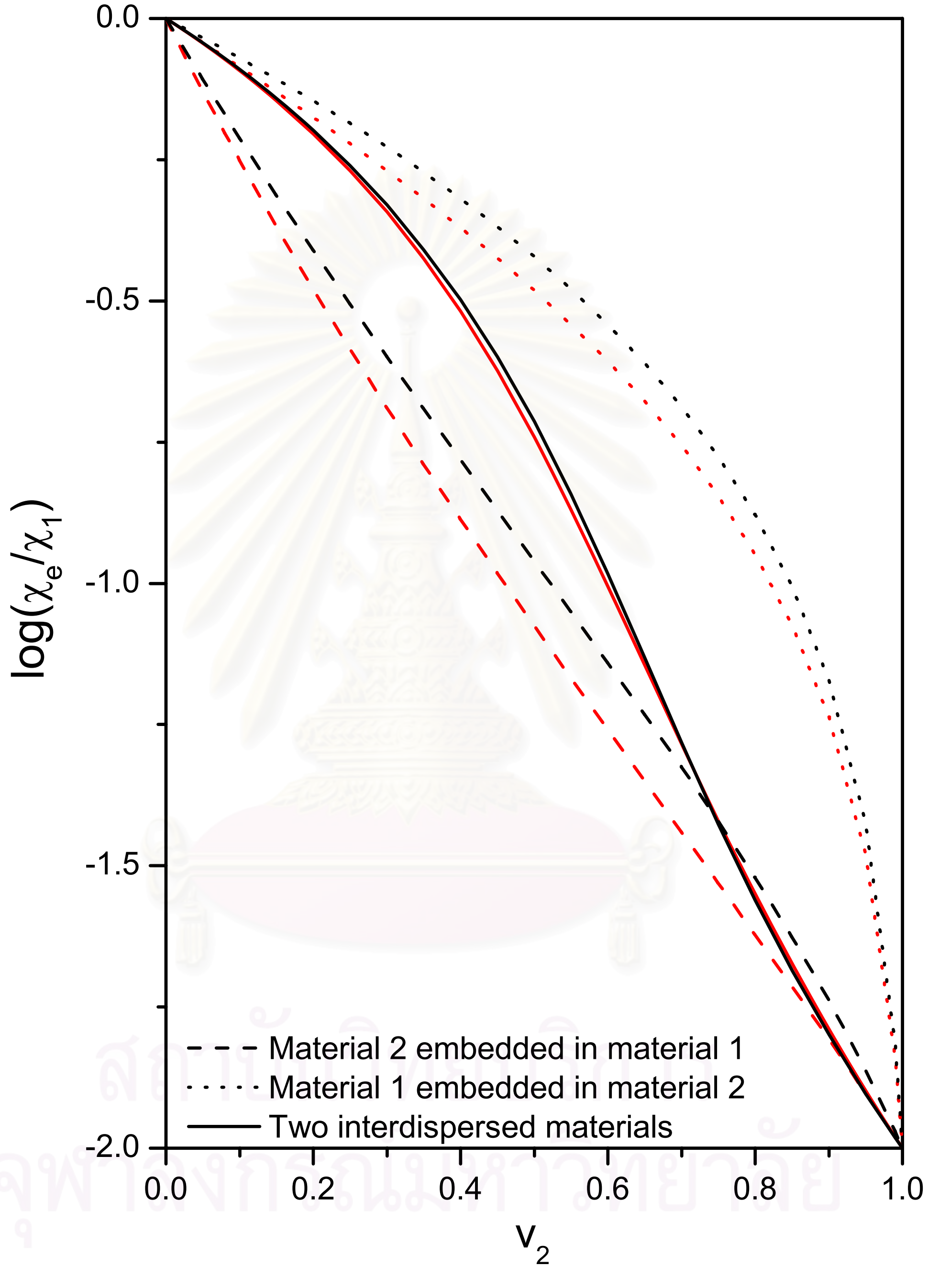

Figure 5.11: Comparison of bounds of $log(\frac{\chi_e}{\chi_1})$ obtained by using the variational method (black lines)and the decoupling technique (red lines) for $\frac{\chi_2}{\chi_1} = 0.01$.

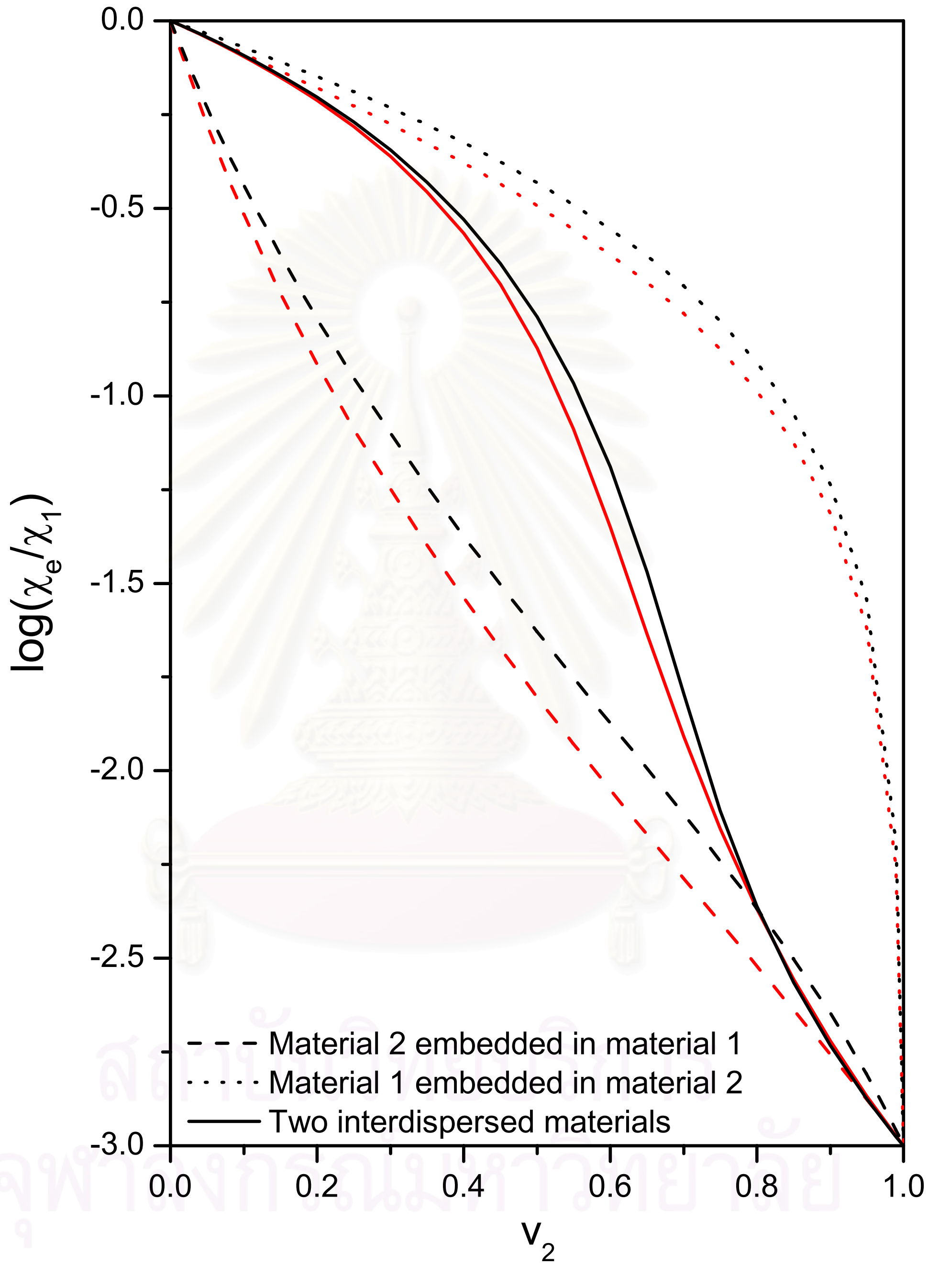

Figure 5.12: Comparison of bounds of $log(\frac{\chi_e}{\chi_1})$ obtained by using the variational method (black lines)and the decoupling technique (red lines) for $\frac{\chi_2}{\chi_1} = 0.001$.

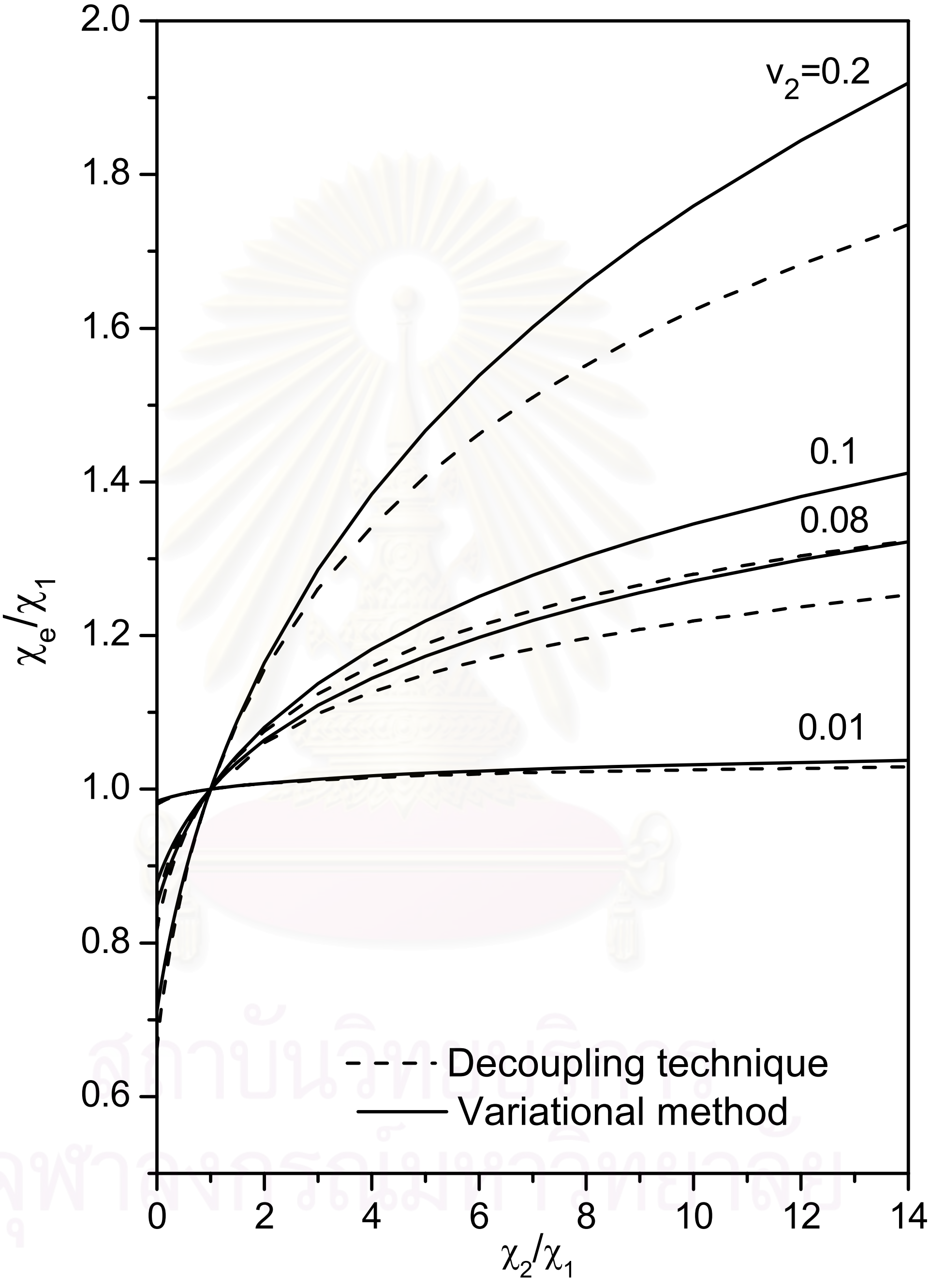

Figure 5.13: Comparison of $\dfrac{\chi_e}{\chi_1}$ obtained by using the variational method and the decoupling technique for $v_2 = 0.01$, 0.08, 0.1 and 0.2.

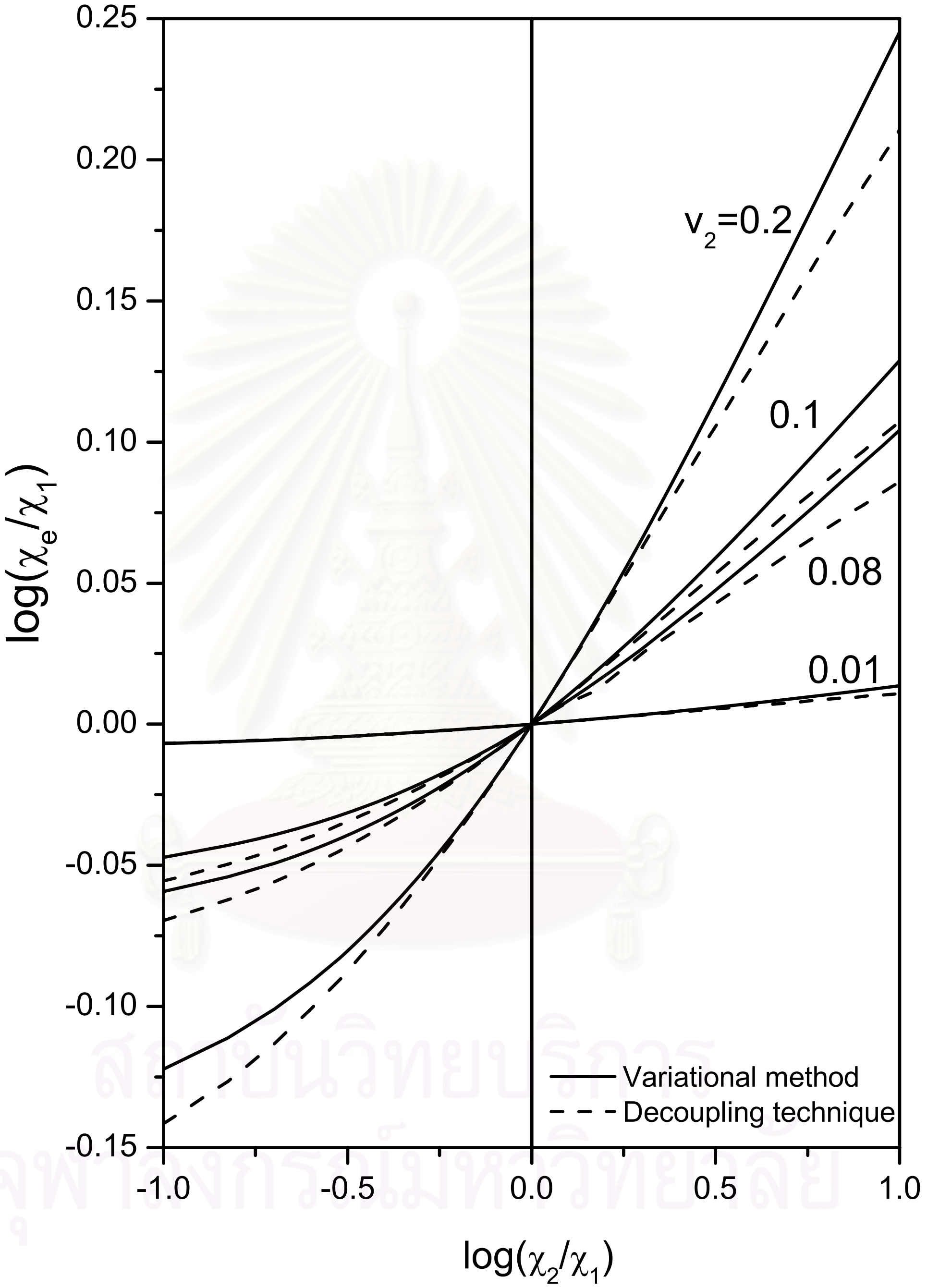

Figure 5.14: Comparison of $log(\dfrac{\chi_e}{\chi_1})$ obtained by using the variational method and the decoupling technique for $v_2 = 0.01$, 0.08, 0.1 and 0.2.

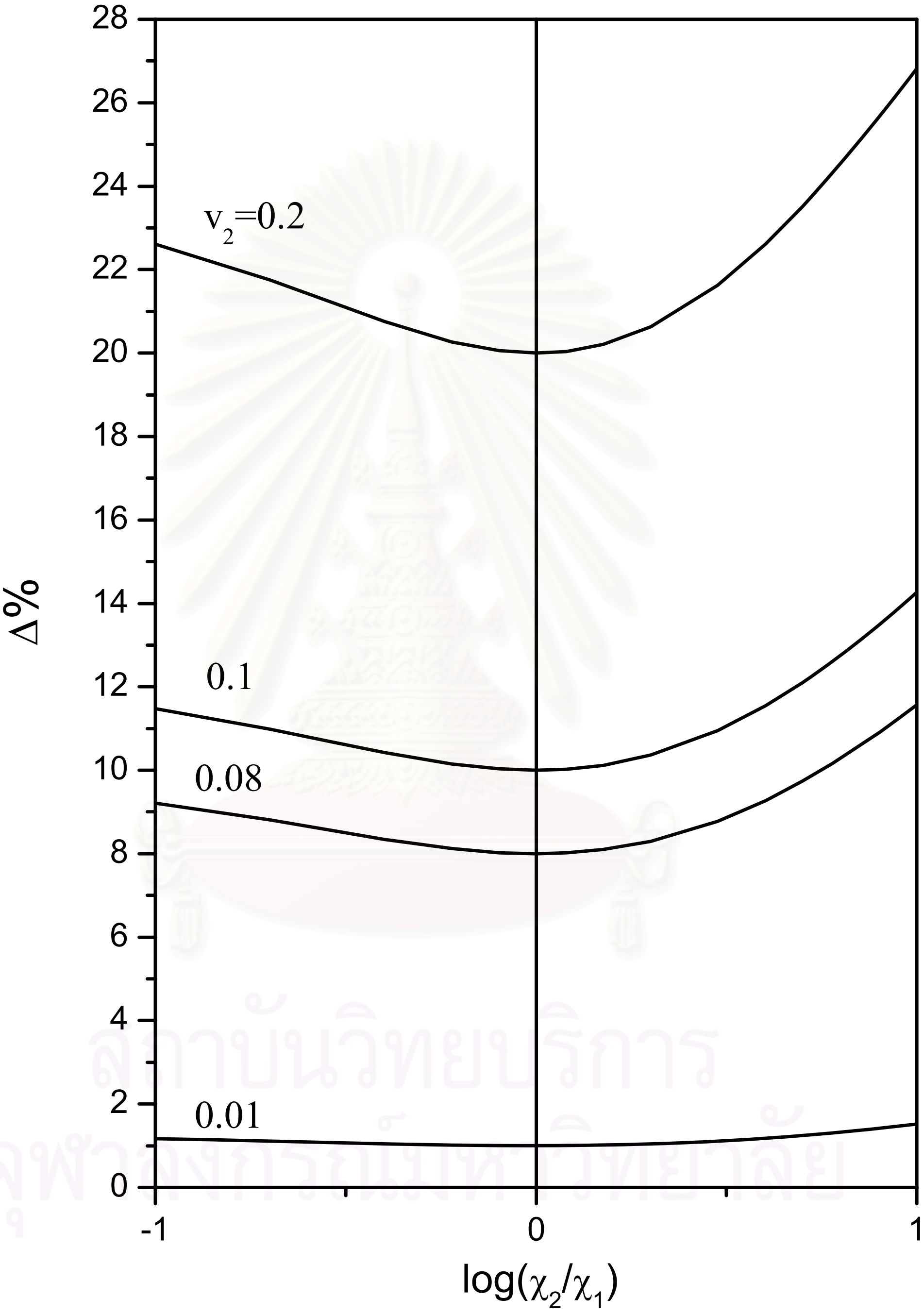

Figure 5.15: The percentage of discrepancy ($\Delta\%$) between $\langle E_1^4 \rangle$ and $\langle E_1^2 \rangle^2$.

5.4 Experimental Effective Nonlinear Coefficients

To test our programs in sections 4.2.3 and 5.2.3, which are used in determination on the effective nonlinear coefficients of the composites, our results are compared with the experimental results of Gehl, Fisher and Boyd in 1997 [32]. In their work, the nonlinear-optical responses of the porous-glass-based composite ones are studied as experimental samples. The samples have two parts: silica glass (72%) and spaces (28%), then the spaces in the sample were saturated and replaced with various nonlinear fluids, such as methanol, carbon tetrachloride and diiodomethane. The ratios between the nonlinear coefficients of the glass and various fluids $\left(\frac{\chi_{glass}}{\chi_{fluid}}\right)$ are 0.62, 0.32 and 0.03, respectively. By using Mach-Zehnder interferometer and analytical process, the relative effective nonlinear coefficients $\left(\frac{\chi_e}{\chi_{fluid}}\right)$ were determined.

In comparison, our results which predict the effective nonlinear coefficient of two interdispersed materials and the experimental results are plotted in Fig. 5.16. Our results using the variational method and the decoupling technique agree very well with the experimental results, while the discrepancies of $\frac{\chi_e}{\chi_{fluid}}$ between the experiment and our result are about 15% for methanol, 13% for carbon tetrachloride and 5% for diiodomethane. Moreover, $\frac{\chi_e}{\chi_{fluid}}$ of carbon tetrachloride and diiodomethane lie between our variational and decoupling results satisfying the theoretical prediction that $\chi_e(decoupling) \leq \chi_e(exact) \leq \chi_e(variational)$; which confirms that our results are reliable.

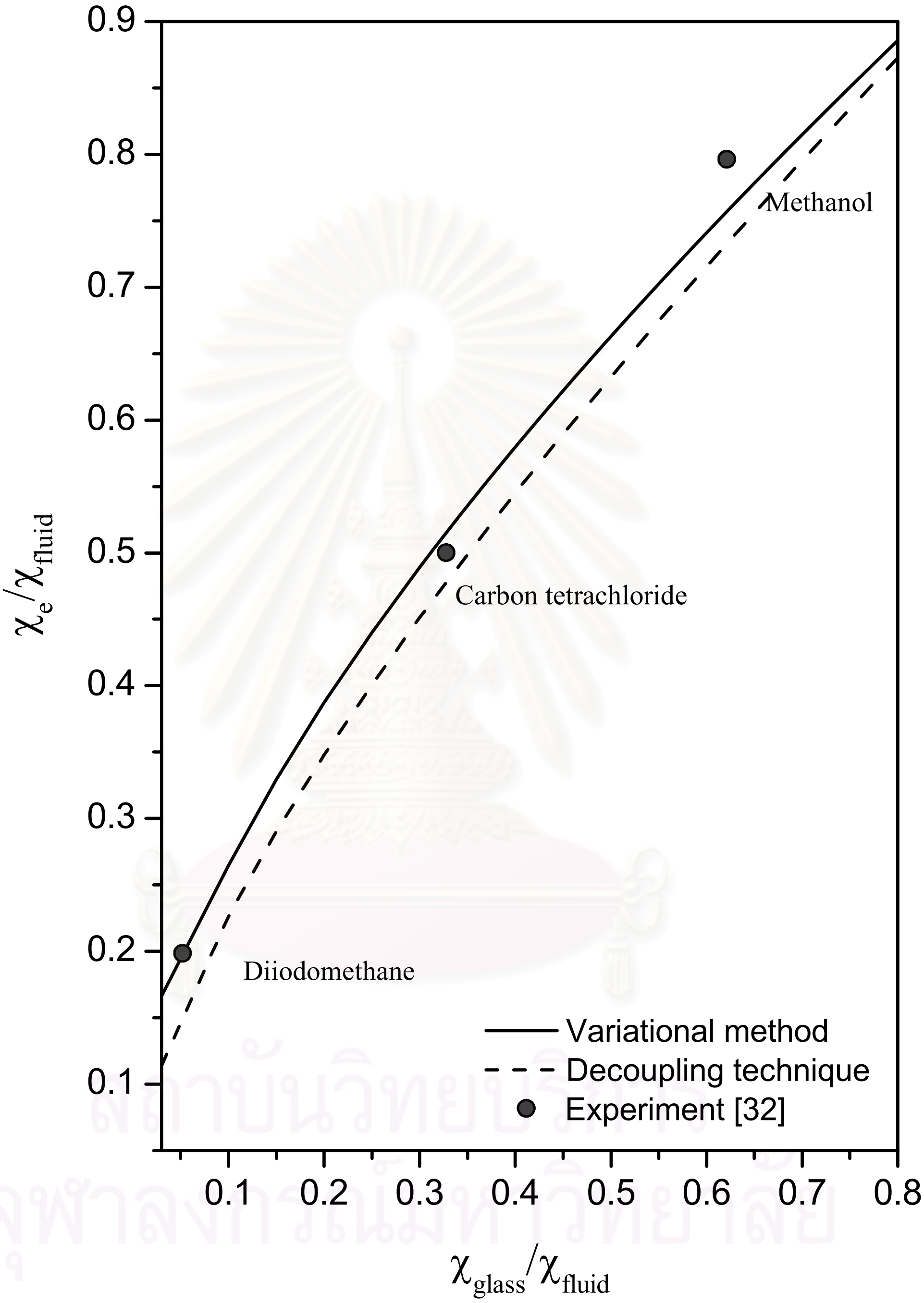

Figure 5.16: Comparison of the relative effective nonlinear coefficient $\frac{\chi_e}{\chi_{fluid}}$, the experimental results are compared with those calculated by using the variational method and the decoupling technique ($v_{fluid} = 0.28$).

Chapter VI

Conclusions

This research is an extension of the work of Yu and Yuen [17] in studying the electric field response of strongly nonlinear dielectric composites. These composites consist of spherical strongly nonlinear dielectric inclusions randomly embedded in a strongly nonlinear dielectric host medium of different nonlinear coefficient. In their work, they assumed that the inclusion volume is much less than the composite volume, then the effective nonlinear coefficient (χ_e) of the composite is determined by using the decoupling technique. Hence, their work has limits on practical applications.

In this research, the effective response of strongly nonlinear dielectric composites has been investigated by using the decoupling technique. The reliability of χ_e in dilute inclusion packing fraction is now extended to arbitrary inclusion packing fractions. The effective medium theory (EMT) originally proposed by Hashin [18] is applied for theoretical modeling and studying the electric field response of these composites.

We consider the composite which is composed of two components, material 1 and material 2, and exhibits nonlinear coefficients χ_1 and χ_2, respectively and determine the bounds of χ_e. The results show the lower and upper bounds in Figs. 5.4 - 5.12 and the higher the contrast between χ_1 and χ_2, the larger the gap between the two bounds are observed. Moreover, if the composite has material of higher nonlinear coefficient being the host medium instead of the inclusions, the higher χ_e is obtained, for the composite of the same packing fraction, as seen in Fig. 5.6, the ratio of χ_e is about 5.6.

In order to confirm the reliability of χ_e based on the EMT, the calculated χ_e is compared to those of the single inclusion model of Yu and Yuen's work. It is found that χ_e based on the EMT is comparable to the result of Yu and Yuen at inclusion packing fraction less than 0.1.

Moreover, in this research, we also apply the simple variational method to calculate χ_e in order to confirm all decoupling technique results. Comparing χ_e calculated by the decoupling technique and χ_e calculated by the simple variational method, we found that both results agree quite well, especially at inclusion packing fraction less than 0.1 for the contrast less than 10 which is the range of the work of Yu and Yuen.

Our results of χ_e calculated by using the decoupling technique are less than those calculated by using the variational method which satisfies the theoretical prediction that $\chi_e(decoupling) \leq \chi_e(exact) \leq \chi_e(variational)$ [17]. Our theoretical results which are obtained by using the decoupling technique and the variational method also agree with the experimental results of Gehl, Fisher and Boyd [32] in the determination on χ_e of the porous-glass-based composite materials.

At the end of this research, we would like to propose that there is a recent method which is applicable to determine χ_e called the effective energy approximation [33]. This approximation is accomplished by the Ponte Castaneda variational principle [34] and Torquato approximation [35]. Moreover, the addition of variational parameters is interested for improvement in χ_e. Therefore, the effective energy approximation and the addition of variational parameters are suggested for further studies.

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Appendices

Appendix A

Energy Functional

In this appendix, we have to show that the determination of the extremum condition of the energy functional as in Eq. (2.22), is Laplace equation (Eq. (2.14)) for linear dielectric media, and nonlinear partial differential equation Eq. (2.16) for nonlinear dielectric media.

A.1 Energy Functional of Linear Media

Consider the relation between the electric displacement $\overrightarrow{D}$ and electric field $\overrightarrow{E}$ of linear dielectric media:

$$\overrightarrow{D} = \varepsilon \overrightarrow{E}. \tag{A.1}$$

By using $\overrightarrow{E} = -\overrightarrow{\nabla}\varphi$, the energy functional of linear dielectric media having volume Ω can be written as

$$W = \frac{1}{2} \iiint\limits_{\Omega} \varepsilon \left| \overrightarrow{\nabla}\varphi \right|^2 dxdydz, \tag{A.2}$$

hence

$$W = \frac{1}{2} \iiint\limits_{\Omega} \varepsilon \left[\varphi_x^2 + \varphi_y^2 + \varphi_z^2 \right] dxdydz, \tag{A.3}$$

where $\varphi_x = \dfrac{\partial \varphi}{\partial x}$, $\varphi_y = \dfrac{\partial \varphi}{\partial y}$ and $\varphi_z = \dfrac{\partial \varphi}{\partial z}$.

From the variational principle [36] of which

$$I = \iiint\limits_{\Omega} F(u, u_x, u_y, u_z, x, y, z) dxdydz \tag{A.4}$$

is the functional of variations with $u(x,y,z)$ being a trial function and $u_x = \dfrac{\partial u}{\partial x}$. Then F satisfying

$$\frac{\partial F}{\partial u} - \frac{\partial}{\partial x}(\frac{\partial F}{\partial u_x}) - \frac{\partial}{\partial y}(\frac{\partial F}{\partial u_y}) - \frac{\partial}{\partial z}(\frac{\partial F}{\partial u_z}) = 0, \tag{A.5}$$

which is Euler-Lagrange's equation for several variables. The solution of Eq. (A.5) yields $u(x,y,z)$ satisfying the functional I has an extremum.

Comparing our functional in Eq. (A.3) with the functional I in Eq. (A.4), we obtain

$$F \equiv \frac{1}{2}\varepsilon(\varphi_x^2 + \varphi_y^2 + \varphi_z^2). \tag{A.6}$$

Replacing Eq. (A.6) into Eq. (A.5) to obtain

$$-\varepsilon(\varphi_{xx} + \varphi_{yy} + \varphi_{zz}) = 0, \tag{A.7}$$

where $\varphi_{xx} = \dfrac{\partial \varphi_x}{\partial x}$, $\varphi_{yy} = \dfrac{\partial \varphi_y}{\partial y}$ and $\varphi_{zz} = \dfrac{\partial \varphi_z}{\partial z}$.

Rearranging Eq. (A.7), thus we obtain

$$-\varepsilon\overrightarrow{\nabla}^2\varphi(x,y,z) = 0,$$

or

$$\overrightarrow{\nabla}^2\varphi(x,y,z) = 0. \tag{A.8}$$

This is Laplace's equation for linear dielectric media. It implies that the extremum condition of the energy functional in Eq. (A.2) gives the solution $\varphi(x,y,z)$ which is also the solution of Laplace's equation in Eq. (A.8).

A.2 Energy Functional of Nonlinear Media

For nonlinear dielectric media with the relation between the electric displacement $\overrightarrow{D}$ and electric field $\overrightarrow{E}$ is

$$\overrightarrow{D} = \varepsilon\overrightarrow{E} + \chi\left|\overrightarrow{E}\right|^2\overrightarrow{E}. \tag{A.9}$$

In this case

$$W = \frac{1}{2}\iiint\limits_{\Omega} \varepsilon \left|\overrightarrow{\nabla}\varphi\right|^2 dV + \frac{1}{4}\iiint\limits_{\Omega} \chi \left|\overrightarrow{\nabla}\varphi\right|^4 dV \tag{A.10}$$

describes the energy functional. Replacement of Eq. (A.10), we obtain

$$\begin{aligned} W &= \frac{1}{2}\iiint\limits_{\Omega} \varepsilon \left[\left(\tfrac{\partial\varphi}{\partial x}\right)^2 + \left(\tfrac{\partial\varphi}{\partial y}\right)^2 + \left(\tfrac{\partial\varphi}{\partial y}\right)^2\right] dxdydz \qquad (\text{A.11}) \\ &+\frac{1}{4}\iiint\limits_{\Omega} \chi \left[\left(\left(\tfrac{\partial\varphi}{\partial x}\right)^2 + \left(\tfrac{\partial\varphi}{\partial y}\right)^2 + \left(\tfrac{\partial\varphi}{\partial y}\right)^2\right)\left(\left(\tfrac{\partial\varphi}{\partial x}\right)^2 + \left(\tfrac{\partial\varphi}{\partial y}\right)^2 + \left(\tfrac{\partial\varphi}{\partial y}\right)^2\right)\right] dxdydz, \end{aligned}$$

or

$$\begin{aligned} W &= \frac{1}{2}\iiint\limits_{\Omega} \varepsilon \left[\varphi_x^2 + \varphi_y^2 + \varphi_z^2\right] dxdydz \\ &+\frac{1}{4}\iiint\limits_{\Omega} \chi \left[\left(\varphi_x^2 + \varphi_y^2 + \varphi_z^2\right)\left(\varphi_x^2 + \varphi_y^2 + \varphi_z^2\right)\right] dxdydz. \qquad (\text{A.12}) \end{aligned}$$

Similar to previous section, we define

$$F \equiv \frac{1}{2}\varepsilon(\varphi_x^2 + \varphi_y^2 + \varphi_z^2) + \frac{1}{4}\chi\left(\varphi_x^2 + \varphi_y^2 + \varphi_z^2\right)\left(\varphi_x^2 + \varphi_y^2 + \varphi_z^2\right), \tag{A.13}$$

where $\varphi\,(x, y, z)$ is the functional. Substituting Eq. (A.13) into the Euler-Lagrange's equation Eq. (A.5), we obtain

$$\begin{aligned} &-\left[\frac{\partial}{\partial x}\left(\varepsilon\varphi_x\right) + \frac{\partial}{\partial y}\left(\varepsilon\varphi_y\right) + \frac{\partial}{\partial y}\left(\varepsilon\varphi_y\right)\right] \\ &-\left[\frac{\partial}{\partial x}\left(\chi\varphi_x\right)\left(\varphi_x^2 + \varphi_y^2 + \varphi_z^2\right) + \frac{\partial}{\partial y}\left(\chi\varphi_y\right)\left(\varphi_x^2 + \varphi_y^2 + \varphi_z^2\right) + \frac{\partial}{\partial z}\left(\chi\varphi_z\right)\left(\varphi_x^2 + \varphi_y^2 + \varphi_z^2\right)\right] \\ = &\;\; 0, \qquad (\text{A.14}) \end{aligned}$$

$$-\left[\nabla\cdot\left(\varepsilon\overrightarrow{\nabla}\varphi\right)\right] - \left[\overrightarrow{\nabla}\cdot\chi\left|\overrightarrow{\nabla}\varphi\right|^2\overrightarrow{\nabla}\varphi\right] = 0,$$

or

$$\left[\overrightarrow{\nabla}\cdot\left(\varepsilon\overrightarrow{\nabla}\varphi + \chi\left|\overrightarrow{\nabla}\varphi\right|^2\overrightarrow{\nabla}\varphi\right)\right] = 0. \tag{A.15}$$

This is a nonlinear partial differential equation describing the potential of the nonlinear media as shown in Eq. (2.21). It implies that the extremum condition of the energy functional in Eq. (A.10) gives the solution $\varphi(x, y, z)$ which is also the solution of the nonlinear partial differential equation in Eq. (A.15).

Appendix B

Theoretical Relation Between $\chi_e(exact)$ and $\chi_e(decoupling)$

In order to show the theoretical relation between $\chi_e(exact)$ and $\chi_e(decoupling)$, the works of Ponte Castaneda [9, 10] are considered. In his work, the theoretical relation between $\chi_e(exact)$ and the composite parameters was derived; as a result,

$$\chi_e(exact) \geq \frac{1}{E_0^4}\left(2\varepsilon_0 E_0^2 - \frac{v_1\varepsilon_1^2}{\chi_1} - \frac{v_2\varepsilon_2^2}{\chi_2}\right). \tag{B.1}$$

From the previous work of Yu et. al. [30], there are the relations $\varepsilon_1 = \chi_1 \langle E_1^2\rangle$ and $\varepsilon_2 = \chi_2 \langle E_2^2\rangle$, which give

$$\langle E_1^2\rangle = \frac{\varepsilon_1}{\chi_1}, \tag{B.2}$$

and

$$\langle E_2^2\rangle = \frac{\varepsilon_2}{\chi_2}. \tag{B.3}$$

According to Eq. (2.31), ε_e can be written as

$$\varepsilon_e = \frac{v_1\langle E_1^2\rangle}{E_0^2} + \frac{v_2\langle E_2^2\rangle}{E_0^2}. \tag{B.4}$$

Then, $\langle E_1^2\rangle$ and $\langle E_2^2\rangle$ in Eqs. (B.2) and (B.3) are substituted into Eq. (B.4), hence

$$\varepsilon_e = \frac{1}{E_0^2}\left(\frac{v_1\varepsilon_1^2}{\chi_1} + \frac{v_2\varepsilon_2^2}{\chi_2}\right). \tag{B.5}$$

Replacing the left-hand side of Eq. (B.5) into Eq. (B.1), we obtain

$$\chi_e(exact) \geq \frac{\varepsilon_e}{E_0^2},$$

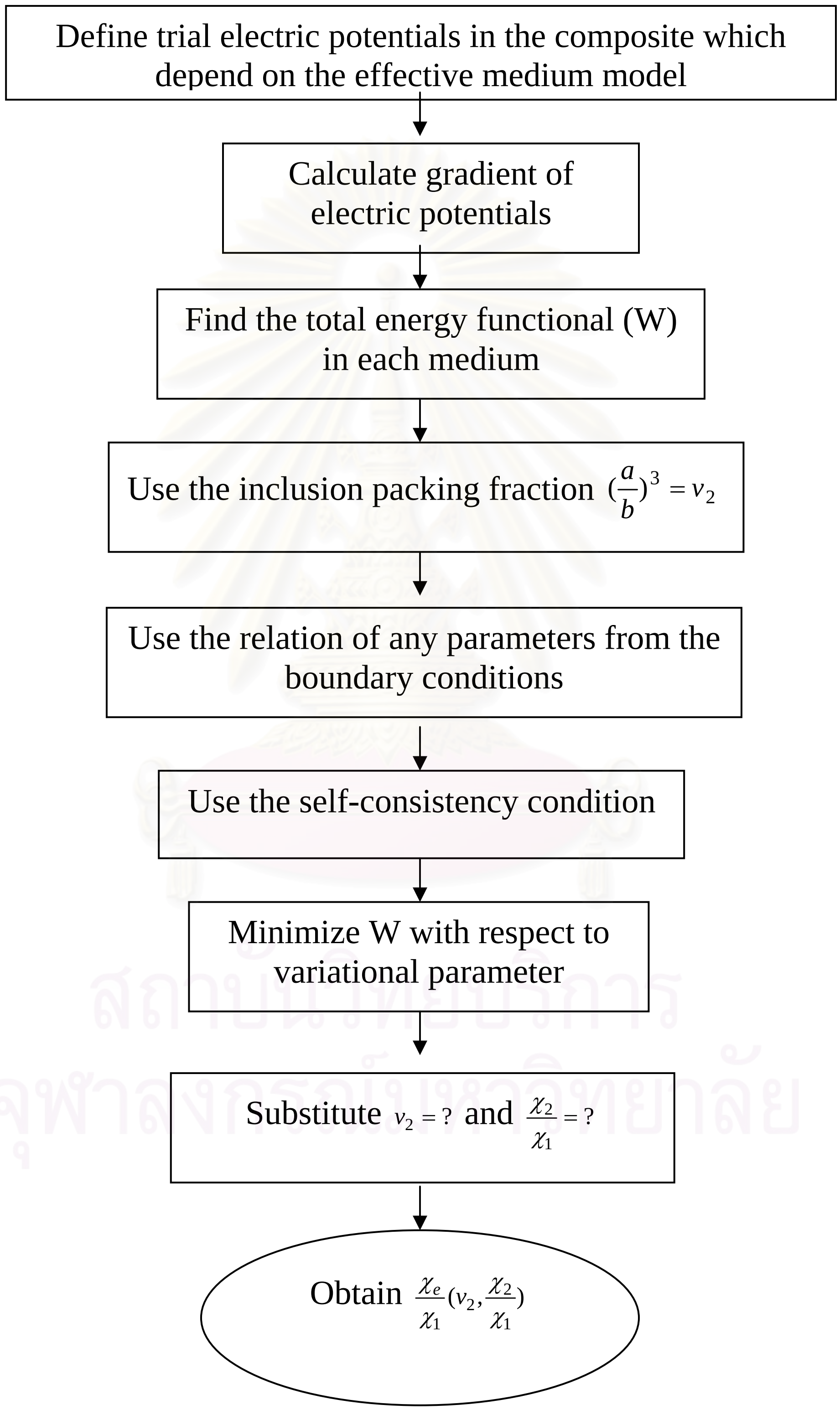
and by using the relation $\varepsilon_e = \chi_e E_0^2$, we also obtain

$$\chi_e(exact) \geq \chi_e(decoupling). \tag{B.6}$$

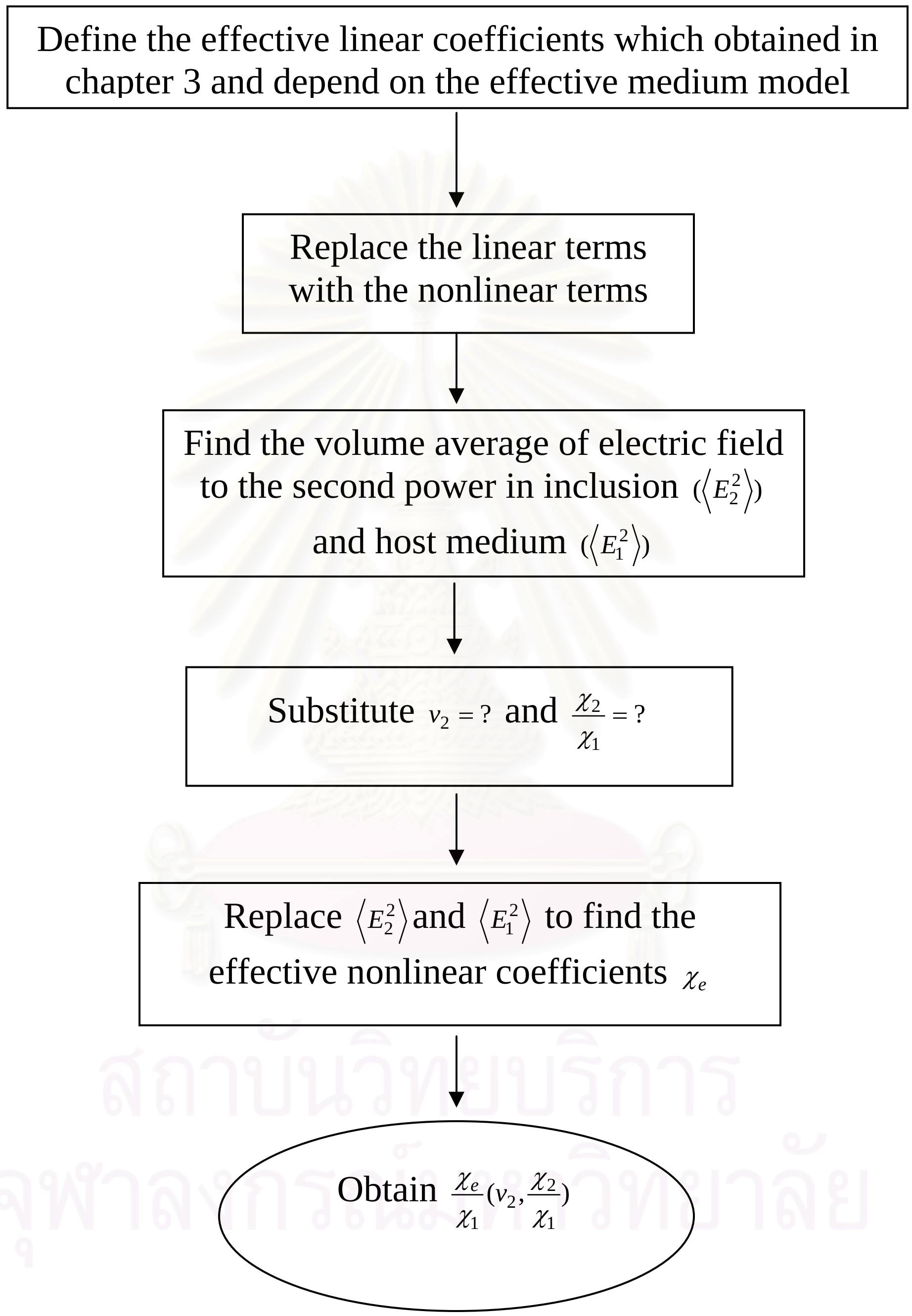
Appendix C

Mathematica Program

(* This flow chart is for the determination on χ_e by using the variational method with the Mathematica Program *)



(* This flow chart is for the determination on χ_e by using the decoupling technique with the Mathematica Program *)



Vitae

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2004 J. Thongsee and M. Natenapit. Effective Nonlinear Coefficient of Strongly Nonlinear Spherical Dielectric Composites. 30^{th} *Congress on Science and Technology of Thailand*, Bangkok, Thailand (19-21 October 2004): D 008