Decreased group velocity in compositionally graded films

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A theoretical formalism is presented that describes the group velocity of electromagnetic signals in compositionally graded films. The theory is first based on effective medium approximation or the Maxwell-Garnett approximation to obtain the equivalent dielectric function in a z slice. Then the effective dielectric tensor of the graded film is directly determined, and the group velocities for ordinary and extraordinary waves in the film are derived. It is found that the group velocity is sensitively dependent on the graded profile. For a power-law graded profile $f(x)=ax^m$, increasing m results in the decreased extraordinary group velocity. Such a decreased tendency becomes significant when the incident angle increases. Therefore the group velocity in compositionally graded films can be effectively decreased by our suitable adjustment of the total volume fraction, the graded profile, and the incident angle. As a result, the compositionally graded films may serve as candidate material for realizing small group velocity.

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I. INTRODUCTION

The physics of inhomogeneous composite media has received much attention in recent years because of its potential applications in laser physics and optical technology [1,2]. Due to the presence of both inhomogeneity and nonlinearity in random composites, large optical nonlinearity enhancement was already predicted through the local field effect [3,4]. On the other hand, the study of the group velocity of electromagnetic signals in nanocomposite materials was performed [5–8]. Actually, in femtosecond nanophotonics, one of the key problems is to realize the tunability of the group velocity [8]. For electromagnetic wave propagation in composite materials, Sølna *et al.* [5,6] showed that the composites can exhibit a larger (or smaller) group velocity of electromagnetic signals than the one in the components, if we combine one component with a high refractive index and low dispersion with the another component with a low refractive index and high dispersion. More recently, based on the well-established Bruggeman effective medium approximation, an enhancement of group velocity in two-phase granular composite media was demonstrated [7]. In addition, we reported that the suitable adjustments to the particles' shapes or shape distributions are helpful to realize enhanced group velocity of electromagnetic signals in two-phase composite media [9]. Such an enhancement phenomenon was also observed in a uniaxial composite medium [10]. Note that most of above works concentrate on the problem of how to realize the enhancement of the group velocity in composite materials.

Parallel to the study of random composite media, graded composite materials attracted much interest in various engineering applications due to their physical properties [11]. For instance, it was experimentally observed that compositionally graded barium strontium titanate thin films have better electric properties than a single-layer barium strontium titanate film with the same composition [12]. In nature, the graded materials are abundant ranging from biological cells to liquid-crystal droplets. Theoretically, nonlinear differential effective dipole approximation and the first-principles approach have been put forward to predict nonlinear optical properties such as third-order nonlinear susceptibility, second, and third harmonic generations in composite media of particles with spatially varying physical properties [13–15], and with compositionally graded profiles [16–18]. It was found that the graded composite media or compositionally graded films can be used as an optical material for producing large enhancements of nonlinear optical properties. Although a lot is known about the effective nonlinear optical properties of graded composite media or compositionally graded films, as far as we know, little is known about propagation of pulses through graded composite media or compositionally graded films. In this paper, we will investigate the group velocity of electromagnetic signals in compositionally graded films. We shall show that the group velocity in the graded composites will be smaller than the one in the components. The decreased magnitude can be adjusted by the suitable choice of the graded profile. Moreover, since the graded films are physically anisotropic, we are able to take one step forward to study the group velocity as a function of the incident angle.

Our paper is organized as follows. In Sec. II, we derive the effective dielectric tensor in the compositionally graded films, and present the formulas for the ordinary and the extraordinary group velocities. In Sec. III, we present our numerical results. Finally, a summary of our results will be given in Sec. IV.

II. THEORETICAL DEVELOPMENT

We consider a two-phase, functionally graded film with width L along the x axis. In each x slice (x is in the range from 0 to L), spherical particles with permittivity ϵ_1 are randomly embedded in the host medium with permittivity ϵ_2 (see Fig. 1). Without loss of generality, we assume both components to be nonmagnetic with permeability $\mu=1$. To char-

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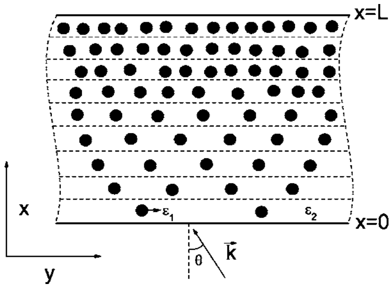

FIG. 1. The model for our system with compositional gradient.

acterize the gradation effect, we take the volume fraction of component 1 to be $f(x)$ at position x.

To investigate the group velocity of pulse propagation in such compositionally graded film, we shall first obtain the effective dielectric tensor of the system. In this connection, we would like to take a two-step solution.

In the first step, we solve the equivalent (local) dielectric constant $\bar{\epsilon}(x)$ for the x slice. For this purpose, we adopt the Maxwell-Garnett approximation (MGA), which is valid for the asymmetric system in which the granular inclusions are randomly embedded in the host medium [19],

$$\bar{\epsilon}(x) = \epsilon_2 + 3\epsilon_2 \frac{f(x)(\epsilon_1 - \epsilon_2)}{\epsilon_1 + 2\epsilon_2 - f(x)(\epsilon_1 - \epsilon_2)}. \quad (1)$$

However, if two components are randomly mixed with each other, we shall use the Bruggeman effective medium approximation (EMA) instead of the Maxwell-Garnett approximation, which admits [20],

$$f(x)\frac{\epsilon_1 - \bar{\epsilon}(x)}{\epsilon_1 + 2\bar{\epsilon}(x)} + [1 - f(x)]\frac{\epsilon_2 - \bar{\epsilon}(x)}{\epsilon_2 + 2\bar{\epsilon}(x)} = 0. \quad (2)$$

In the second step, we are able to estimate the effective permittivity tensor $\overleftrightarrow{\epsilon}_e$ as follows:

$$\overleftrightarrow{\epsilon}_e = \epsilon_{xx}^e \mathbf{e}_x \mathbf{e}_x + \epsilon_{yy}^e \mathbf{e}_y \mathbf{e}_y + \epsilon_{zz}^e \mathbf{e}_z \mathbf{e}_z, \quad (3)$$

where

$$\frac{1}{\epsilon_{xx}^e} = \frac{1}{L}\int_0^L \frac{dx}{\bar{\epsilon}(x)}, \quad (4)$$

$$\epsilon_{yy}^e = \epsilon_{zz}^e = \frac{1}{L}\int_0^L \bar{\epsilon}(x)dx. \quad (5)$$

Then, we aim to study the pulse propagation through the compositionally graded films. The starting point for the group velocity is the macroscopic Maxwell equations [21,22]

$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t}, \quad \nabla \times \mathbf{H} = \frac{\partial \mathbf{D}}{\partial t}. \quad (6)$$

From these equations, we can easily derive

$$\nabla \times (\nabla \times \mathbf{E}) = -\mu_0 \frac{\partial^2 \mathbf{D}}{\partial t^2}, \quad (7)$$

where $\mathbf{D} = \epsilon_0 \overleftrightarrow{\epsilon}_e \cdot \mathbf{E}$.

Assuming solution of the form

$$\mathbf{E}(\mathbf{r},t) = \mathbf{E}_0 e^{i\mathbf{k}\cdot\mathbf{r}} e^{-i\omega t}, \quad (8)$$

Eq. (7) takes the form

$$\mathbf{D} = \frac{1}{\mu_0 \omega^2}[\mathbf{k}(\mathbf{k}\cdot\mathbf{E}) - k^2\mathbf{E}], \quad (9)$$

with $k^2 \equiv \mathbf{k}\cdot\mathbf{k} = k_x^2 + k_y^2 + k_z^2$.

Hence the components of $\mathbf{D}$ are written as

$$D_x = -\frac{1}{\mu_0\omega^2}\left[k_x(\mathbf{k}\cdot\mathbf{E}) - k^2\frac{D_x}{\epsilon_0\epsilon_{xx}^e}\right], \quad (10)$$

$$D_{y(z)} = -\frac{1}{\mu_0\omega^2}\left[k_{y(z)}(\mathbf{k}\cdot\mathbf{E}) - k^2\frac{D_{y(z)}}{\epsilon_0\epsilon_{yy}^e}\right]. \quad (11)$$

According to Eqs. (10) and (11), we obtain three linear homogeneous equations for three components of $\mathbf{D}$,

$$\left(\frac{k^2 - k_x^2}{\epsilon_{xx}^e} - \frac{\omega^2}{c^2}\right)D_x - \frac{k_x k_y}{\epsilon_{yy}^e}D_y - \frac{k_x k_z}{\epsilon_{yy}^e}D_z = 0,$$

$$-\frac{k_x k_y}{\epsilon_{xx}^e}D_x + \left(\frac{k^2 - k_y^2}{\epsilon_{yy}^e} - \frac{\omega^2}{c^2}\right)D_y - \frac{k_y k_z}{\epsilon_{yy}^e}D_z = 0,$$

$$-\frac{k_x k_z}{\epsilon_{xx}^e}D_x - \frac{k_y k_z}{\epsilon_{yy}^e}D_y + \left(\frac{k^2 - k_z^2}{\epsilon_{yy}^e} - \frac{\omega^2}{c^2}\right)D_z = 0. \quad (12)$$

In order to get nonzero $\mathbf{D}$, the determinant of their coefficients should vanish [22],

$$\begin{vmatrix} \frac{k^2 - k_x^2}{\epsilon_{xx}^e} - \frac{\omega^2}{c^2} & -\frac{k_x k_y}{\epsilon_{yy}^e} & -\frac{k_x k_z}{\epsilon_{yy}^e} \\ -\frac{k_x k_y}{\epsilon_{xx}^e} & \frac{k^2 - k_y^2}{\epsilon_{yy}^e} - \frac{\omega^2}{c^2} & -\frac{k_y k_z}{\epsilon_{yy}^e} \\ -\frac{k_x k_z}{\epsilon_{xx}^e} & -\frac{k_y k_z}{\epsilon_{yy}^e} & \frac{k^2 - k_z^2}{\epsilon_{yy}^e} - \frac{\omega^2}{c^2} \end{vmatrix} = 0. \quad (13)$$

As a consequence, one yields the eigenvalue equation,

$$g(\mathbf{k},\omega) \equiv -\frac{\omega^2}{c^2}\left(\frac{k^2}{\epsilon_{yy}^e} - \frac{\omega^2}{c^2}\right)\left(\frac{k_x^2}{\epsilon_{yy}^e} + \frac{k_y^2 + k_z^2}{\epsilon_{xx}^e} - \frac{\omega^2}{c^2}\right) = 0. \quad (14)$$

In other words, Eq. (14) gives two equations, which support two types of waves in the compositionally graded films. The first type corresponds to the usual ordinary wave and the other corresponds to the extraordinary solutions, i.e.,

$$g_o(\mathbf{k},\omega) \equiv \left(k^2 - \frac{\omega^2}{c^2}\epsilon_{yy}^e\right) = 0, \quad (15)$$

$$g_e(\mathbf{k},\omega) \equiv k_x^2\epsilon_{xx}^e + (k_y^2 + k_z^2)\epsilon_{yy}^e - \frac{\omega^2}{c^2}\epsilon_{xx}^e\epsilon_{yy}^e = 0. \quad (16)$$

The group velocity $\mathbf{v}_g$ of pulse propagation is defined as the gradient of the angle frequency ω with respect to $\mathbf{k}$; that is [10]

$$\mathbf{v}_g = \nabla_{\mathbf{k}}\omega|_{\omega_0} = \left.\frac{\partial\omega}{\partial k_x}\right|_{\omega_0}\mathbf{e}_x + \left.\frac{\partial\omega}{\partial k_y}\right|_{\omega_0}\mathbf{e}_y + \left.\frac{\partial\omega}{\partial k_z}\right|_{\omega_0}\mathbf{e}_z, \tag{17}$$

where ω_0 is the central frequency of the initial wave packet. According to Eq. (14), the ordinary and the extraordinary group velocities can be redefined as,

$$\mathbf{v}_g^{o(e)} = \nabla_{\mathbf{k}}\omega|_{\omega_0} = -\frac{1}{\partial g_{o(e)}/\partial\omega}\nabla_{\mathbf{k}}g_{o(e)}|_{\omega_0}. \tag{18}$$

Carrying out some tedious calculations, we arrive at

$$\mathbf{v}_g^{o} = \frac{2c^2\mathbf{k}}{\omega\left(2\epsilon_{yy}^e + \omega\frac{d\epsilon_{yy}^e}{d\omega}\right)} \tag{19}$$

for ordinary waves, and

$$\mathbf{v}_g^{e} = \frac{2c^2(\epsilon_{xx}^e k_x\mathbf{e}_x + \epsilon_{yy}^e k_y\mathbf{e}_y + \epsilon_{yy}^e k_z\mathbf{e}_z)}{\omega\left[2\epsilon_{xx}^e\epsilon_{yy}^e + \omega\left(\frac{d\epsilon_{yy}^e}{d\omega}\epsilon_{xx}^e + \frac{d\epsilon_{xx}^e}{d\omega}\epsilon_{yy}^e\right)\right] - c^2\left[\frac{d\epsilon_{xx}^e}{d\omega}k_x^2 + \frac{d\epsilon_{yy}^e}{d\omega}(k_y^2 + k_z^2)\right]} \tag{20}$$

for extraordinary waves.

So far, we have successfully obtained the general formulas for the group velocity of electromagnetic signals in compositionally graded films. In view of the fact that the wave vectors $\mathbf{k}$ lie in the xy plane (as shown in Fig. 1), one yields $\mathbf{k}=k\cos\theta\mathbf{e}_x+k\sin\theta\mathbf{e}_y$. As a result, the magnitudes of group velocities for ordinary and extraordinary waves are expressed as

$$v_g^0 = \frac{2c\sqrt{\epsilon_{yy}^e}}{2\epsilon_{yy}^e + \omega\frac{d\epsilon_{yy}^e}{d\omega}}, \tag{21}$$

$$v_g^e = \frac{2c\sqrt{\frac{\epsilon_{xx}^e\epsilon_{yy}^e}{\epsilon_{xx}^e\cos^2\theta + \epsilon_{yy}^e\sin^2\theta}}\sqrt{(\epsilon_{xx}^e\cos\theta)^2 + (\epsilon_{yy}^e\sin\theta)^2}}{2\epsilon_{xx}^e\epsilon_{yy}^e + \omega\left(\frac{d\epsilon_{yy}^e}{d\omega}\epsilon_{xx}^e + \frac{d\epsilon_{xx}^e}{d\omega}\epsilon_{yy}^e\right) - \omega\frac{\epsilon_{xx}^e\epsilon_{yy}^e}{\epsilon_{xx}^e\cos^2\theta + \epsilon_{yy}^e\sin^2\theta}\left(\frac{d\epsilon_{xx}^e}{d\omega}\cos^2\theta + \frac{d\epsilon_{yy}^e}{d\omega}\sin^2\theta\right)}. \tag{22}$$

It is easy to check that v_g^e is incident angle dependent, and when θ is zero, i.e., $\mathbf{k}\|\mathbf{e}_x$, v_g^e reduces to v_g^o. Therefore, we shall only perform numerical calculations on v_g^e. In this connection, we must get the derivatives of ϵ_{xx}^e and ϵ_{yy}^e with respect to ω, which are given by

$$\frac{d\epsilon_{xx}^e}{d\omega} = \frac{(\epsilon_{xx}^e)^2}{L}\int_0^L \frac{dx}{[\bar{\epsilon}(x)]^2}\frac{d\bar{\epsilon}(x)}{d\omega}, \quad \frac{d\epsilon_{yy}^e}{d\omega} = \frac{1}{L}\int_0^L \frac{d\bar{\epsilon}(x)}{d\omega}dx, \tag{23}$$

where $d\bar{\epsilon}(x)/d\omega$ is given by

$$\frac{d\bar{\epsilon}(x)}{d\omega} = \frac{9f(x)\epsilon_2^2\frac{d\epsilon_1}{d\omega} + [1-f(x)][(\epsilon_1+2\epsilon_2)^2 + 2(\epsilon_1-\epsilon_2)^2 f(x)]\frac{d\epsilon_2}{d\omega}}{[\epsilon_1 + 2\epsilon_2 + (\epsilon_2-\epsilon_1)f(x)]^2} \tag{24}$$

for MGA, and

$$\frac{d\bar{\epsilon}(x)}{d\omega} = \frac{f(x)[\epsilon_2 + 2\bar{\epsilon}(x)]^2\bar{\epsilon}(x)\frac{d\epsilon_1}{d\omega} + [1-f(x)][\epsilon_1 + 2\bar{\epsilon}(x)]^2\bar{\epsilon}(x)\frac{d\epsilon_2}{d\omega}}{f(x)[\epsilon_2 + 2\bar{\epsilon}(x)]^2\epsilon_1 + [1-f(x)][\epsilon_1 + 2\bar{\epsilon}(x)]^2\epsilon_2} \tag{25}$$

for EMA.

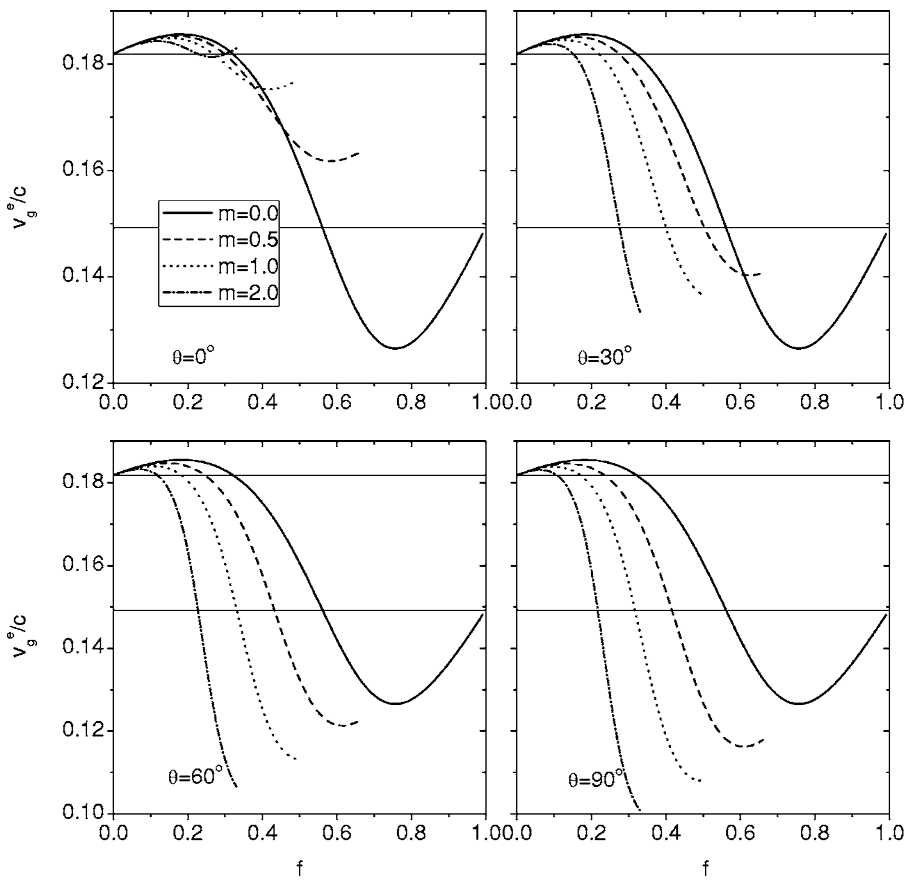

FIG. 2. The group velocity for extraordinary waves v_g^e/c in compositionally graded films as a function of the total volume fraction f for power-law graded profile $f(x)\sim ax^m$ with various m and for various incident angles θ. In each slice, we adopt effective medium approximation.

III. NUMERICAL RESULTS

In what follows, we shall do some numerical calculations. We choose $\epsilon_1=1.44$ and $\epsilon_2=5$. Moreover, to observe the decreased group velocity easily, we set $d\epsilon_1/d\omega|_{\omega=\omega_0}=13.2/\omega_0$ and $d\epsilon_2/d\omega|_{\omega=\omega_0}=5/\omega_0$ in previous works [7,9,10].

In Fig. 2, we plot the group velocity for extraordinary wave v_g^e against the total the volume fraction $f\equiv\int_0^L f(x)dx/L$ in compositionally graded films for various incident angle $\theta=0°$, 30°, 60°, and 90°, and for power-law gradient profile $f(z)=ax^m$ with various $m=00.5,1.0,2.0$. In each x slice, we assume two granular components to be symmetric in microstructure, whose local dielectric constant is described by the Bruggeman EMA. For the nongraded case $m=0$, the composite film is indeed isotropic with $\epsilon_{xx}^e=\epsilon_{yy}^e$. As a consequence, the group velocity is found to be independent of the incident angle. Actually, such a case has been examined in [7,9]. However, when the compositional gradient is taken into account, we find that in the total volume fraction region $f<0.3$, the group velocity is decreased monotonically with increasing m. We understand this as follows. The introduction of the compositional gradient indicates that the composite film acts as a multilayer one, and the existence of many interfaces leads to strong scattering of the energy, resulting in more slow group velocity than in the nongraded case. Note that the decreased tendency is more significant for larger θ. For instance, for small θ (see the case $\theta=0°$), although the group velocity for graded case is smaller than the one for nongraded one, it is still larger than the group velocity in component 1. With the increase of incident angle (see the case $\theta=30°$), one can observe comparatively smaller group velocity than the one in both components. It should be mentioned here that the so-called small group velocity for the graded case is still larger than the minimum achieved in random composites without gradations. For large θ (see the cases $\theta=60°$ and 90°), we realize the smallest group velocity in the compositionally films indeed by the suitable adjustment of the graded profile. For example, for $m=2$ and $\theta=90°$, the group velocity in the compositionally films is decreased to 40% in comparison with that in component 1.

Next, we aim at the compositionally graded film in which Hashin-Shtrikman microgeometry exists in each x slice. In this situation, MGA is a good approximation to estimate the local dielectric constant. Numerical results for v_g^e are shown in Fig. 3. For the nongraded profile, there exists a maximal peak in the whole volume faction region from 0 to 1, and the minimal group velocity in the composite films is the same as the one in component 1. When the graded profile is introduced, the position of maximal peak shifts to small volume fractions. In this situation, to achieve a small group velocity, one can choose large m in the case of the wave vector perpendicular to the x axis. Comparing Fig. 2 with Fig. 3, we can conclude that in addition to the graded effect and the incident angle, the microgeometric information plays also an

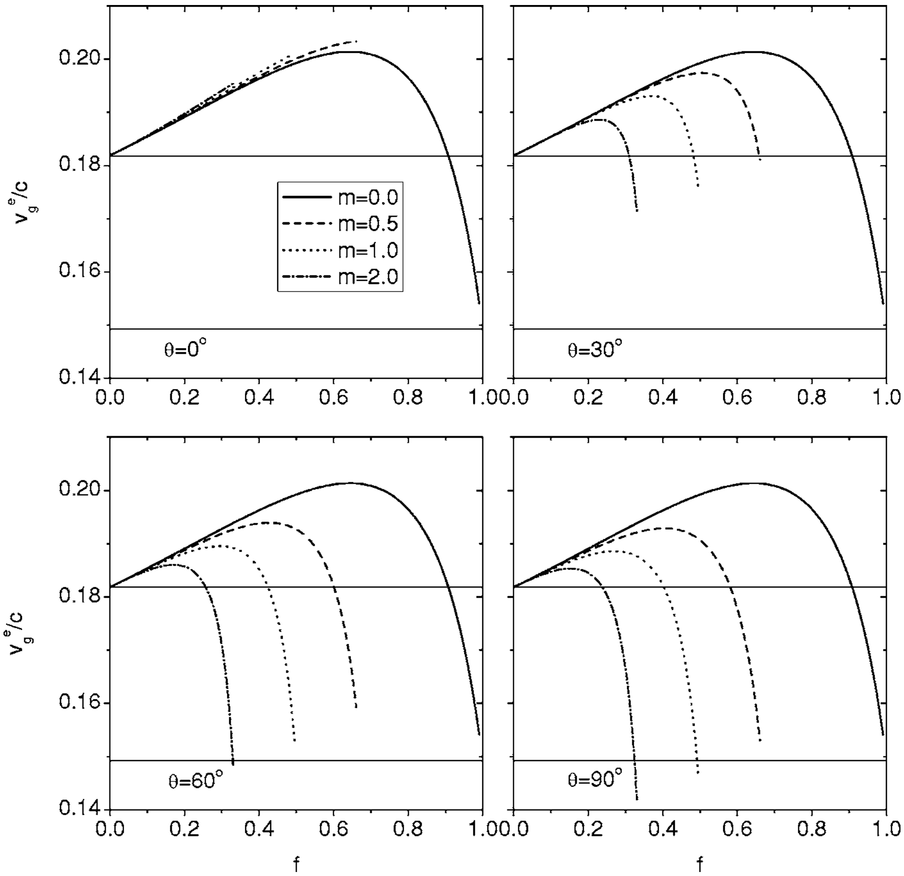

FIG. 3. Same as Fig. 1, but for the Maxwell-Garnett approximation used in each slice.

important role in determining the group velocity in the composite media.

In the end, the dependence of v_g^e on the incident angle is shown in Fig. 4. Both theories predict that the group velocity exhibits monotonic decrease with the increasing incident angle for a given compositional gradient. It is known that for the ordinary wave vector, the effective dielectric constant is smaller than the one for extraordinary wave with $\theta=90°$, resulting in a fast group velocity. On the other hand, for large m, the physical properties at each x slice becomes quite different especially in high x region, and multilayer scattering becomes strong. Therefore the effective group velocity becomes weak.

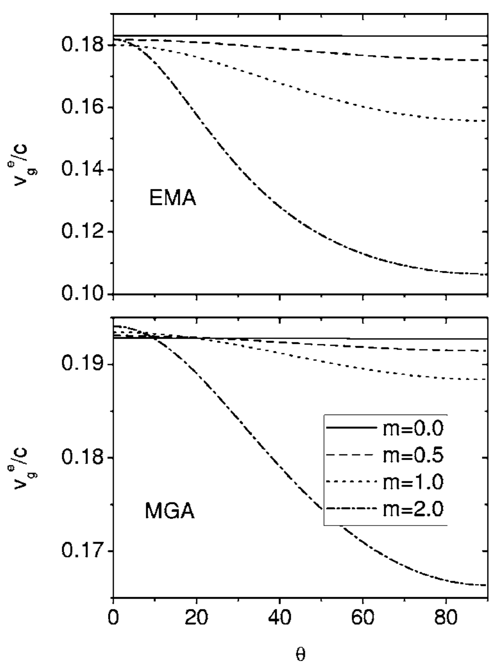

FIG. 4. v_g^e/c as a function of the incident angle for various m.

IV. SUMMARY

In this paper, we have investigated the angle dependent group velocity in compositionally graded films. At each x slice, both the Maxwell-Garnett approximation and the effective medium approximation are adopted to derive the local dielectric function. We have found that the compositional gradient plays an important role in determining the group velocity of electromagnetic signals through the film. For power-law compositional profile $p(x)\sim x^m$, it was predicted that large m leads to low group velocity. Furthermore, the group velocity can be further decreased for a large incident angle. Therefore by the suitable choice of the volume fraction, the compositional gradient and the incident angle, the group velocity can be effectively decreased.

Since our formulas are derived in the quasistatic approximation, where the spatial scale of optical inhomogeneities is much lower than the radiation wavelength, the dispersion can be described within the framework of the Maxwell-Garnett model and effective medium model. In this connection, the role of interference phenomena is indeed insignificant. Once the spatial scale can be compared to the radiation wavelength, new physical phenomena shall appear [8]. On the other hand, due to simplistic treatments of the distributional statistics of the components, we do not take into account the effect of the scattering losses. For this purpose, some sophisticated approaches such as those provided by the strong-property-fluctuation theory [10] can be applied.

ACKNOWLEDGMENT

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