Indian J. Phys. **76A** (2), 141-145 (2002)

IJP A
an international journal

Surface polaritons in a semi-infinite composite medium using the Bruggeman dielectric function

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Received 18 December 2000, accepted 11 October 2001

Abstract We present a study of surface polaritons at the boundary of a semi-infinite composite medium bounded by vacuum The effective dielectric function of the composite medium is modeled using the Bruggeman theory. The dispersion curves of the surface polaritons of the composite medium are found to be strongly dependent on a filling factor, which is a measure of the volume content of constituents of the composite medium Numerical results for attenuated total reflection (ATR) spectra due to surface polaritons propagating along the boundary of the semi-infinite composite medium are presented The theory is illustrated by a system consisting of silicon-vacuum-Ag/KCl composite

Keywords : composites, optical properties, polaritons, surfaces

PACS Nos. 71 36 78.30

1. Introduction

There has been an increasing interest in recent years in understanding the science and technology of composite systems [1, 2]. The properties of composite systems differ strikingly from those of the constituent media. Several techniques such as reflectance spectroscopy [3], Raman spectroscopy [4-6] and attenuated total reflection (ATR) spectroscopy [7], amongst others, can be used to study composite systems. Our earlier work in the study of composite systems includes the study of ATR by surface polaritons of such systems [8], and recently a theory of Raman scattering by bulk polaritons of a composite medium was presented [9]. In this paper, our objective is to further the understanding of composite systems by studying the surface polaritons propagating along the boundary of a semi-infinite composite medium, with an effective dielectric function, $\varepsilon_{eff}(\omega)$, using Bruggeman theory (to be referred to as BT) [10]. The composite medium consists of spherical grains of a dielectric function $\varepsilon_s(\omega)$ embedded in a medium with a background dielectric function $\varepsilon_b(\omega)$, and BT is such that the effective dielectric function is symmetric in the materials constituting the composite medium, and is of the form [10]

$$f\frac{[\varepsilon_s(\omega)-\varepsilon_{eff}(\omega)]}{[\varepsilon_s(\omega)+2\varepsilon_{eff}(\omega)]}+(1-f)\frac{[\varepsilon_b(\omega)-\varepsilon_{eff}(\omega)]}{[\varepsilon_b(\omega)+2\varepsilon_{eff}(\omega)]}=0 \quad (1)$$

where f is a filling factor, usually small, and such that

$$0\le f\le 1 \quad (2)$$

and $f = 0$ corresponds to the case when there is only the host material and no spherical grains, while $f = 1$ corresponds to the case when the host constituent has been replaced by the grains constituent.

It is well known that eq. (1) is quadratic in $\varepsilon_{eff}(\omega)$, with two solutions given by

$$\varepsilon_{eff}^{B\pm}(\omega)=\frac{1}{4}[\varepsilon_b(\omega)(2-3f)+\varepsilon_s(\omega)(3f-1)]$$

$$\pm\frac{1}{4}\sqrt{[\varepsilon_b(\omega)(2-3f)+\varepsilon_s(\omega)(3f-1)]^2+8\varepsilon_b(\omega)\varepsilon_s(\omega)]} \quad (3)$$

where $\varepsilon_{eff}^{B+}(\omega)$ and $\varepsilon_{eff}^{B-}(\omega)$ corresponds to the positive root and negative root respectively on the right hand side. The frequency dependence of the Bruggeman dielectric function is

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further discussed in Section 3, and illustrated in Figures 1(a) and 1(b). In presence of damping, the Bruggeman dielectric function is complex, of the form

$$\varepsilon_{eff}^{B\pm}(\omega) = \varepsilon_{eff}^{B\pm'}(\omega) + i\varepsilon_{eff}^{B\pm''}(\omega). \tag{4}$$

The geometry studied in this paper, consists of a composite medium with an effective dielectric function $\varepsilon_{eff}(\omega)$ occupying the half space $z < 0$ and a surface inactive medium with a positive dielectric constant ε in the other half space $z > 0$. The explicit forms for the dielectric functions $\varepsilon_b(\omega)$ of the background medium and $\varepsilon_s(\omega)$ of the spherical grains are given below.

$$\varepsilon_b(\omega) = \varepsilon(\infty) + \frac{S\omega_T^2}{\omega_T^2 - \omega^2 - i\omega\Gamma_b} \tag{5}$$

where $\varepsilon(\infty)$ is the high frequency dielectric constant, S gives the strength of the resonance, ω_T is the TO phonon frequency, and Γ_b is the damping constant for phonons.

$$\varepsilon_s(\omega) = 1 - \frac{\omega_p^2}{\omega^2 + i\omega\Gamma_s} \tag{6}$$

where ω_p is the plasma frequency, and Γ_s is the damping constant for plasmons.

The geometry described above can sustain surface polaritons along the interface of the composite medium and the surface inactive medium. By applying electromagnetic boundary conditions, the dispersion relation for the surface polaritons is given by (See, for example, [11-14]

$$\frac{c^2 q_x^2}{\omega^2} = \frac{\varepsilon\varepsilon_{eff}(\omega)}{\varepsilon + \varepsilon_{eff}(\omega)}, \tag{7}$$

where q_x is the tangential wavevector along the interface. Dispersion curves for surface polaritons propagating along the boundary of the composite medium for varying values of f are given in Section 3.

2. ATR spectra for surface polaritons

Surface polaritons of the composite system can be studied by the ATR technique [7] which has proved to be very successful in probing surface excitations. The ATR setup considered here consists of a coupling prism of dielectric constant ε_1 in the region $z > d$, a surface inactive medium of dielectric constant ε in the region $0 < z < d$ and the composite medium with the dielectric function $\varepsilon_{eff}(\omega)$ in the region $z < 0$. Application of electromagnetic boundary conditions at the interfaces at $z = 0$ and $z = d$, enables one to obtain the reflection coefficient R for ATR spectra (see for example, [11], [15]) given by

$$R = 1 - \frac{4P''}{(1+P'')^2 + P'^2}, \tag{8}$$

where

$$P' = \frac{D_2}{D_1}\left[1 - \frac{(1-T)N}{(D_2 + D_3'T)^2 + D_3''^2T^2}\right] \tag{9}$$

$$P'' = D_2^2 \frac{D_3''}{-}\left[\frac{(1-T^2)}{(D_2 + D_3'T)^2 + D_3''^2T^2}\right], \tag{10}$$

with

$$N = [D_2 + D_3'T][D_2 - D_3'] - D_3''^2T, \tag{11}$$

$T = \tanh(k_{2z}d)$,

$$D_1 = \frac{\varepsilon_1}{k_{1z}}, \tag{13}$$

$$D_2 = \frac{\varepsilon}{k_{2z}}, \tag{14}$$

$$D_3 = \frac{\varepsilon_{eff}(\omega)}{k_{3z}} = D_3' + iD_3'', \tag{15}$$

where k_{1z}, k_{2z} and k_{3z} are the n wavevectors, and D_3 is complex since the effective dielectric function appearing in eq. (15) is complex when there is finite damping, as was mentioned in eq. (4). Numerical results for ATR spectra due to surface polaritons propagating along the boundary of the composite medium are plotted and discussed in Section 3, *first* in the low frequency region, where the effect of varying the filling factor f for a fixed angle of incidence is studied in Figure (2a), while the effect varying the angle of incidence for a fixed filling factor f is studied in Figure (2b), and *secondly* in the high frequency region.

3. Numerical results and discussion

The main objective of this paper was to illustrate numerically the characteristics of surface polaritons along the boundary of a composite medium bounded by vacuum where the effective dielectric function of the composite medium is modeled using the Bruggeman theory. This paper complements an earlier study [8] using the Maxwell-Garnett theory [16, 17], where the effective dielectric function $\varepsilon_{eff}(\omega)$ satisfies

$$f\frac{[\varepsilon_s(\omega) - \varepsilon_b(\omega)]}{[\varepsilon_s(\omega) + 2\varepsilon_b(\omega)]} - \frac{[\varepsilon_{eff}(\omega) - \varepsilon_b(\omega)]}{[\varepsilon_{eff}(\omega) + 2\varepsilon_b(\omega)]} = 0, \tag{16}$$

which is inherently asymmetric in the constituents of the composite medium, and does not show the percolation transition.

Numerical results for the ATR spectra, using eq. (8), are presented. The composite medium is modeled as consisting of silver and potassium chloride, with the following parameters obtained from the work of Cummings *et al* [13]. For KCL, $\varepsilon(\infty) = 2.1$, $\omega_T = 141$ cm^{-1}, $S = 2.13$ and for Ag, $\omega_p = 73100$ cm^{-1}. Cummings *et al* [3] were investigating the bulk reflectance measurements of a Ag/KCl composite and it is important to note that their results are outside the frequency range for surface polaritons considered in this paper. For the ATR configuration, a silicon coupling prism with $\varepsilon_1 = 11.696$ is used, and the gap is of thickness $d = 30000A$ or $d = 300A$ and $\varepsilon = 1$ for vacuum. The damping constants are taken as $\Gamma_b = 0.005\omega_T$ and $\Gamma_s = 0.005\omega_p$ for KCl and Ag respectively.

In order to understand the properties of surface excitations of the composite medium, it is important to understand the frequency dependence of the effective dielectric function given in eq. (3). In Figure (1a), the frequency dependence of the effective dielectric function in the low frequency region is plotted

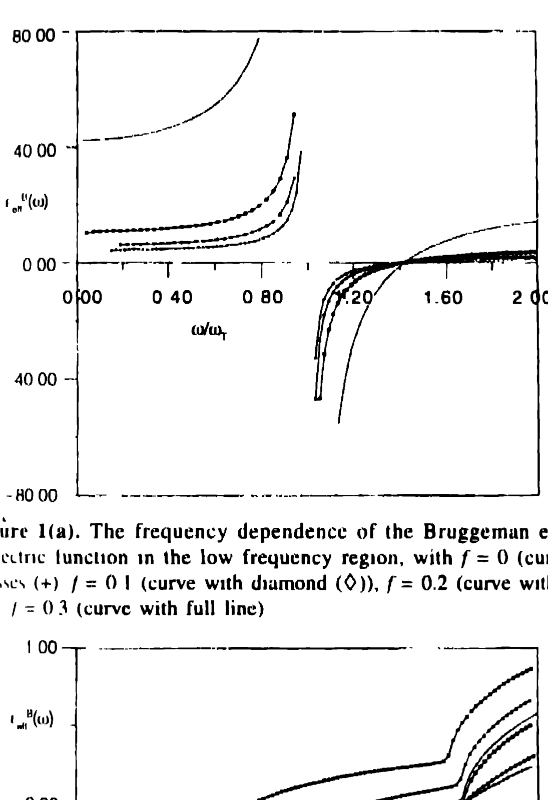

Figure 1(a). The frequency dependence of the Bruggeman effective dielectric function in the low frequency region, with $f = 0$ (curve with crosses (+)) $f = 0.1$ (curve with diamond (◇)), $f = 0.2$ (curve with circles (o)) $f = 0.3$ (curve with full line)

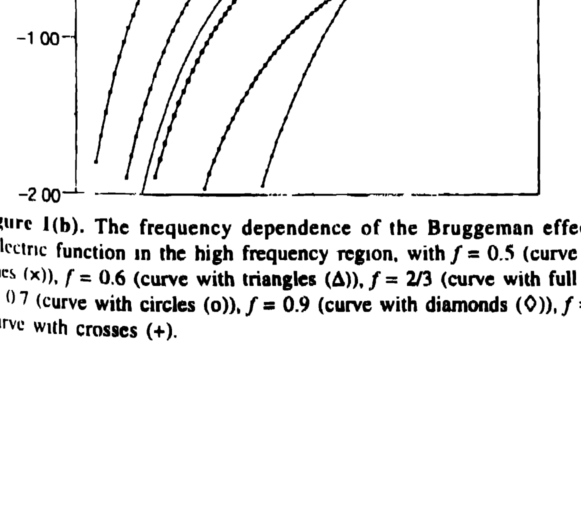

Figure 1(b). The frequency dependence of the Bruggeman effective dielectric function in the high frequency region, with $f = 0.5$ (curve with times (x)), $f = 0.6$ (curve with triangles (Δ)), $f = 2/3$ (curve with full line) $f = 0.7$ (curve with circles (o)), $f = 0.9$ (curve with diamonds (◇)), $f = 1.0$ (curve with crosses (+).

as a function with varying values of f. Figure (1a) shows a dominant insulator type dielectric function, implying that at low frequencies the composite medium is dominated by particles with the dielectric function given in eq. (5), which in this case implies KCl. At higher frequencies, as shown in Figure (1b), the composite medium is dominated by particles with the dielectric function given in eq. (6), that is metallic Ag particles.

The dispersion curves plotted in Figures (2a) and (2b) show that as f increases the upper limiting frequency for surface polaritons increases for a fixed tangential wavevector, while as $cq_x / \omega_T \rightarrow 1$ all the curves converge. In all the dispersion

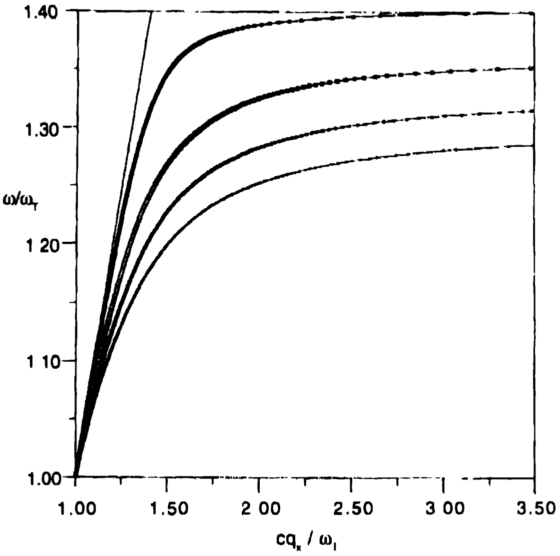

Figure 2(a). Dispersion curves for surface polaritons of a composite medium using the Bruggeman effective dielectric function, with $f = 0$ (curve with crosses (+)), $f = 0.1$ (curve with diamonds (◇)), $f = 0.2$ (curve with squares (□)), $F = 0.3$ (curve with circles (o)), $f = 1/3$ (full curve).

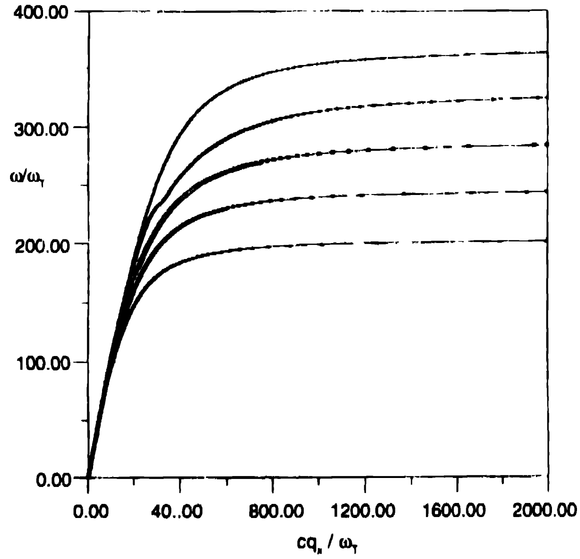

Figure 2(b). Dispersion curves for surface polaritons of a composite medium using the Bruggeman effective dielectric function, with $f = 0.6$ (curve with triangles (Δ)), $f = 0.7$ (curve with circles (o)), $f = 0.8$ (curve with squares (□)), $f = 0.9$ (curve with diamonds (◇)), $f = 1.0$ (curve with crosses (+)).

curves, when $f = 0$, the insulator background is dominant, the upper limiting frequency ω_{sb} for surface polaritons is given by

$$\omega_{sb} = \left[\frac{\varepsilon + \varepsilon(\infty) + S}{\varepsilon + \varepsilon(\infty)}\right]^{1/2} \omega_T \tag{17}$$

while the case when the host background has been replaced by the metallic particles, *i.e.* $f = 1$, the upper limiting frequency ω_{ss} in all the dispersion curves is given by

$$\omega_{ss} = \frac{\omega_p}{\sqrt{1+\varepsilon}}. \tag{18}$$

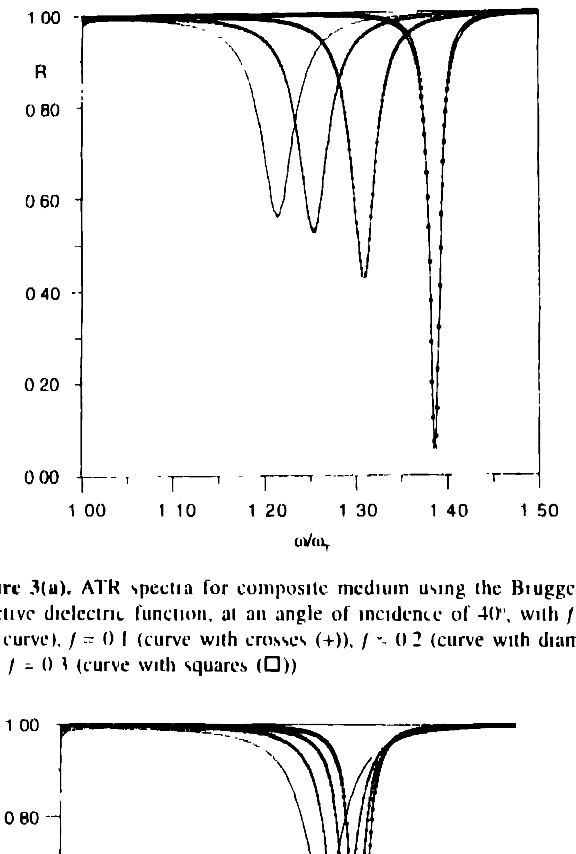

Figure 3(a). ATR spectra for composite medium using the Bruggeman effective dielectric function, at an angle of incidence of 40°, with $f = 0$ (full curve), $f = 0.1$ (curve with crosses (+)), $f = 0.2$ (curve with diamond (◊)), $f = 0.3$ (curve with squares (□))

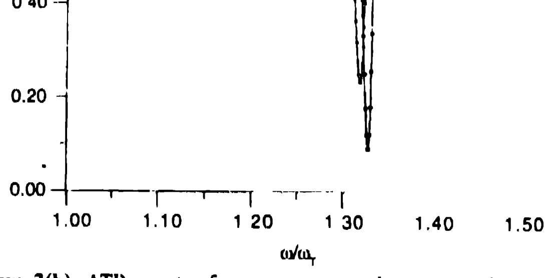

Figure 3(b). ATR spectra for composite medium using the Bruggeman effective dielectric function at a filling factor, $f = 0.2$ at angles of incidence of $\phi = 35°$ (full curve), $\phi = 40°$ (curve with crosses (+)), $\phi = 45°$ (curve with diamond (◊)) and $\phi = 50°$ (curve with circles (o)).

In Figure (3a), a numerical plot of the ATR spectra is shown, illustrating the effect of varying the filling factor f for a fixed angle of incidence, $\phi = 40°$. The effect of varying the angle of incidence at $\phi = 35°$, $\phi = 40°$, $\phi = 45°$ and $\phi = 50°$ for a fixed filling factor of $f = 0.2$ is illustrated in Figure (3b). The physical significance of a minimum in the ATR spectrum is that at this frequency there is matching between the tangential wavevectors of the incident beam and the surface mode, and that energy is removed from the incident beam by the surface mode. From the ATR spectra in Figures (3a) and (4a), it can be noted that as f increases, the minimum in the ATR spectra shifts to higher

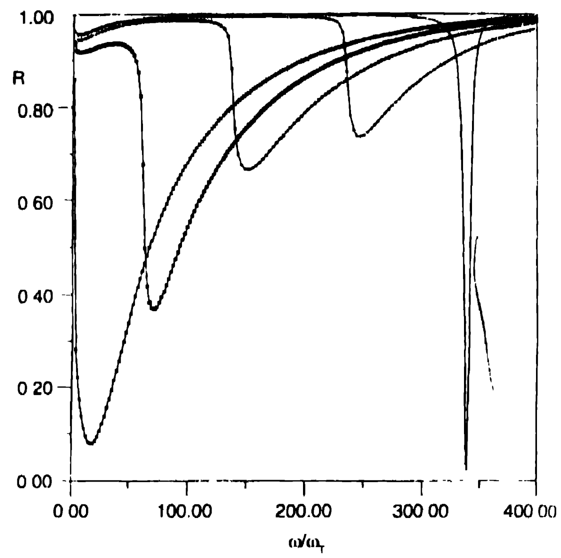

Figure 4(a). ATR spectra for composite medium using the Bruggeman effective dielectric function, in the high frequency region, at an angle of incidence of 40°, with $f = 1/3$ (curve with times (×)), $f = 0.5$ (curve with circles (o)), $f = 0.7$ (curve with diamonds (◊)), $f = 0.9$ (curve with crosses (+)), $f = 1.0$ (full curve), at $d = 300$ A

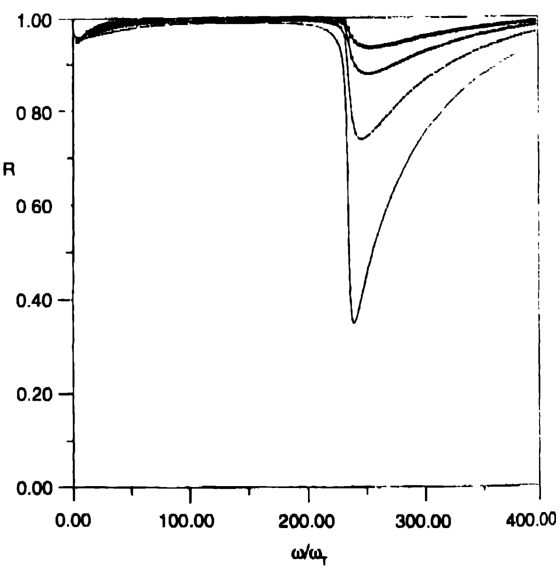

Figure 4(b). ATR spectra for composite medium using the Bruggeman effective dielectric function, in the high frequency region, at a filling factor of $f = 0.9$, at angles of incidence of $\phi = 35°$ (full curve), $\phi = 40°$ (curve with crosses (+)), $\phi = 45°$ (curve with diamond (◊)) and $\phi = 50°$ (curve with circles (o)), at $d = 300$A.

frequencies and also, the frequency where there is a minimum in the ATR spectra, increases with increasing angles of incidence. It is of interest to note that in order to be able to probe the higher frequency surface polaritons as shown in Figures (4a) and (4b), it is necessary to go to reduced gap thicknesses, of the order $d = 300A$.

4. Conclusions

Our main objective in this paper was to extend our earlier work [8] for further understanding of surface polaritons propagating along the boundary of a semi-infinite composite medium comprising of Ag and KCl. This has been achieved by studying the frequency dependence of the Bruggeman effective dielectric function satisfying eq. (1), which is symmetric in the materials constituting the composite medium, and shows a percolation transition at $f = 1/3$. The study carried out in this paper can be further extended by using other formulations of effective dielectric functions, for example, those due to Kirkpatrick [18] and Zhang [19].

In Figures (2a) and (2b), the dispersion curves for surface polaritons of the semi-infinite composite medium are plotted using the Bruggeman effective dielectric function, and all the curves are found to be strongly dependent on the filling factor f, converging as the frequency of the surface polaritons approaches the Transverse Optical (TO) phonon frequency, and the upper limiting frequency is found to increase as f increases.

The numerical ATR spectra of the composite medium have been calculated for the Bruggeman dielectric functions in Figures (3a) (3b), (4a) and (4b) respectively. All the ATR spectra show a strong dependence on the filling factor f and each spectrum shows a minimum which signifies matching between the tangential wavevector of the incident beam and the surface mode, and reduced reflectivity.

Acknowledgments

We would like to acknowledge support from the University of Botswana. Also, we would like to thank Dr. M Masale for some interesting discussions. One of us (NM) would also like to thank ANSTI and the DAAD Foundation of Germany for a fellowship at the University of Botswana.

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