Available online at http://www.journalijdr.com

IJDR

ISSN: 2230-9926

International Journal of Development Research
Vol. 14, Issue, 02, pp. 65004-65010, February, 2024
https://doi.org/10.37118/ijdr.27867.02.2024

RESEARCH ARTICLE OPEN ACCESS

HIGHLY DOPED METAL OXIDES SUBSTITUTE THE CONVENTIONAL METALS IN RECENT PLASMONIC MATERIALS

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ARTICLE INFO

Article History:
Received 02nd January, 2024
Received in revised form
14th January, 2024
Accepted 06th February, 2024
Published online 28th February, 2024

Key Words:

Surface Plasmon Polariton, Surface Plasmon resonance, Trans conducting oxides.

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ABSTRACT

Highly doped metal oxides like Indium Tin Oxide can have metal like optical properties in the NIR frequency range. This paper aims to report the optical properties of highly doped metal oxides and its efficient use as substitute to metals in the excitation and propagation of surface plasmon polariton wave with the help of MATLAB simulations.

Citation*: *Sukla Rajak, 2024. *"Highly doped metal oxides substitute the conventional metals in recent plasmonic materials". International Journal of Development Research*, 14, (02), 65004-65010.

INTRODUCTION

Conventional metals have their potential uses as plasmonic material in recent Plasmonic applications and optoelectronic devices. In the rapidly growing fields of Plasmonics and metamaterials operating in near IR and IR region and opto-electronic device applications and solar cells [1-3], new plasmonic materials like Indium tin oxide (ITO-In_2O_3: Sn), Aluminum and Gallium doped Zinc Oxide (ZnO: Al and ZnO: Ga) become alternatives to conventional metals. Conventional metals pursue high losses in visible and IR region [4]. They have very large negative real permittivity [5] and their optical properties cannot be tuned. Due to these undesirable properties, many novel applications in Plasmonics face some limitations while using conventional metals as plasmonic materials. Transparent conducting oxides like ITO, Al and Ga doped ZnO on the other hand draw special attention of recent investigations on the propagation of surface plasmon polariton (SPP) related plasmonic study Transparent conducting oxides have low intrinsic loss in near IR and IR region having advantages of low intrinsic loss. They have some other advantages like semiconductor-based designs, compatibility with standard nanofabrication processes [6] can be achievable. As, their optical properties are dependent on the charge carrier concentrations, their optical properties can be tuned and the devices using them have tunability [7] in frequencies. Transparent conducting oxides are non-stoichiometric in nature. They have electrical resistivity ~10^{-5} Ω cm, absorption coefficient 10^4 cm^{-1} in the near UV and visible range and optical band gap~3 eV. Highly doped metal oxides, TCOs having charge carrier concentration around 10^{21} cm^{-3} can show metallic properties and behave as plasmonic in NIR wavelength region [8] Among all the transconducting oxides, Indium tin oxide (90% wt indium oxide with 10% wt tin oxide) has been mostly used as a potential plasmonic material in NIR [9-14], IR region [15] and optical frequency ranges. With variation of the doping concentrations of Sn in In_2O_3 , the optical constants changes drastically in the optical frequency range from near infrared to ultraviolet region[16,17]. Rhodes et al have experimentally demonstrated the propagation of surface plasmon polaritons (SPPs) at the interface of ITO and glass substrate. However, the scarcity and high price of pure Indium necessitate the replacement of ITO by alternative oxides. This is the main reason why ZnO thin films with advantages of lower cost, resource availability and non toxicity draws the special attention of recent investigations on plasmonic study. Pure intrinsic Zinc oxides are stoichiometric in nature. Intrinsic ZnO is an n-type wide-bandgap semiconductor. Undoped ZnO thin films have low carrier density and this feature leads it to have high resistivity. But he addition of impurities (Group III elements like Al and Ga) in wide bandgap semiconductors (ZnO) offers a measurable dramatic change in the electrical and optical properties of Zinc oxide. In comparison to pure metals and ITO, Al- and Ga- doped ZnO thin films possess some extraordinary advantages, like they have low cost, thermal stability, and comparatively low deposition temperature with greater stability under hydrogen plasma bombardment [18]. Moreover, they are found to exhibit tunable optical properties [7] and are compatible with standard fabrication and integration procedures [6].

In the near IR and IR region of wavelength region, Transparent conducting oxides can exhibit metallic properties when heavily doped. Due to doping, charge carrier concentration increases to some extent so that it results in large plasma frequency. And a large plasma frequency results Drude metal like optical properties in materials.

As the plasma frequency is related to the charge carrier concentration by the equation

$$\omega_p{}^2 = \frac{ne^2}{\varepsilon_0 m^*}$$

According to classical Drude theory, the complex electrical permittivity or dielectric function is defined as

$$\varepsilon = \varepsilon' + i\varepsilon'' = \varepsilon_\infty - \frac{\omega_p{}^2}{\omega(\omega + i\gamma)}$$

where, ε_∞ is the high frequency dielectric constant, γ is damping coefficient of free electrons and ω_p is plasma frequency. Materials should have small to γ value to have low loss.. In order to have negative ε in the optical range, the material should have large plasma frequency ω_p and small high frequency dielectric constant ε_∞ . A large plasma frequency results Drude metal like optical properties in materials. Present study aims to report the efficient use of two transparent conducting oxides, namely, aluminium-doped and gallium doped zinc oxide in the excitation and propagation of surface plasmon polariton wave.

In this paper we wish to find the dual resonance properties supported by the TCO thin films. In search of optical response of these TCOs with the exciting incident radiation three phase Kretschmann configuration is taken for the Mathematical simulations. The MATLAB simulation curves of the surface plasmon resonance is presented in this paper. The investigation of the condition of angle of incidence and thickness of the metal oxide thin films at which they show dual peak surface plasmon resonance is reported here.

Mathematical Background: When light travels in X direction with frequency ω then electric field will be

$$E_x = E_0 e^{\left[i\omega\left(\frac{nx}{c}\right)-t\right]} e^{-\frac{\omega kx}{c}} \dots\dots\dots\dots\dots\dots\dots\dots. (1)$$

Where n= refractive index and k is extinction coefficient.
And complex refractive index is $N = n + ik$

Conducting metal oxides (CMO) have contributions only from free carriers and no transition among bound states, in the spectral region of interest [19].So the response of free carriers to the electromagnetic radiation can be described by Drude model [20]. According to Drude theory the complex refractive index depend on the number of free electrons, *Ne*, an electronic charge, *e*, and an effective mass, *m** as follows

$$N = 1 - \frac{4\pi N_e e^2}{m^* \omega^2} \qquad \dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots (2)$$

The plasma frequency is the critical frequency separating a highly reflecting and totally transparent region and is defined by the frequency at Zero complex refractive index.

$$\omega_p{}^2 = \frac{4\pi N_e e^2}{m^*} \qquad \dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots (3)$$

The reflecting region is the region with large extinction coefficient and zero-approaching refractive index. The typical electron density of metals is approximately 10^{22} cm^{-1} leading to 5×10^{10} per s of the plasma frequency (the ultraviolet).

Transparent conducting oxides (TCOs) are regularly used in liquid-crystal displays.

These heavily doped TCO materials can exhibit metallic properties in the near-IR[8].

From equation 1 ($\omega p \propto \sqrt{N}$) it depicts that they have large plasma frequency which results in Drude-metal-like optical properties [21] .According to generalized Drude theory the permittivity of a material

$$\varepsilon = \varepsilon'(\omega) + \varepsilon''(\omega) = \varepsilon_{int} - \frac{\omega_p{}^2}{\omega(\omega + i\Gamma)} \qquad \dots\dots\dots (4)$$

ε_{int} is high frequency static dielectric constant and it is due to the screening effect of bound electrons in the material.

$\Gamma = 1/\gamma$

Where γ is mean Drude-relaxation time and Γ is damping coefficient of the free carriers. For low loss material Γ should be small. From Equ(4) it can be said that ω_p should be large and ε_{int} should be small to get negative ε' in the optical range of interest.

But many semiconductors suffer from solid –solubility limits when doping concentration become> 10^{21} cm^{-3}. However, zinc oxide and indium oxide overcome this limit [22] and can be heavily doped to be metallic substitutes in the near-IR. If the doping concentration increases more than 10^{22} cm^{-3} then they can response in visible ranges but suffer from solid solubility limits.

In this work, we have studied the optical properties of aluminium-doped zinc oxide (AZO), gallium-doped zinc oxide (GZO), and indium-tin-oxide (ITO)

At which frequency the real permittivity of the material crosses zero is called the cross over frequency (ω_c) .The highest cross over frequency for ITO and GZO is about 0.9 eV whereas it is 0.7 eV for AZO due to improved crystallinity structure. This differentiates the efficiency of the materials [22,23]. Notably, AZO offers the lowest Drude damping, but it also has the lowest ω_c (and hence the longest cross-over wavelength). GZO and ITO can produce cross-over wavelength as low as 1.2 μm . However, Drude damping in GZO is slightly higher than that in AZO and lower than that in ITO. The surface plasmon polariton response of TCO thin films depends on the film thickness [24,25] because the net carrier concentration depends on the volume to surface ratio of the film [26,27] Again as scattering is dependent on the microstructure of the film and microstructure is thickness dependent [24,25]. The surface plasmon waves have *p*-character because the surface charge induces the discontinuity of the electric field in the surface normal *z*-direction, but *s*-waves has only *Ey* component (no *Ez* component). In the Kretschmann configuration to excite a surface plasmon polariton by p-polarized light incident on a planar metal oxide surface from the adjacent dielectric medium, the in-plane component of the photon wavevector in the prism should coincide with the SPP wave vector at the air- metal oxide interface [28] i.e.

$$K_{sp} = \frac{\omega}{c}\sqrt{\varepsilon_{prism}}\,\sin\theta \qquad \dots\dots\dots\dots\dots\dots\dots\dots (5)$$

The reflectivity of a material with normal incidence of light, is a function of material properties and it increases with the number of free carriers. However when light coupled to SPPs with almost 100% efficiency then a sharp minimum in the reflectance profile is observed. The tunnelling distance decreases with the increase of the film thickness. As a result the efficiency of the SPP excitation (and the field enhancement) decreases. Under the condition of total internal reflection as shown in Fig.1 the coupling between the p-polarised light and the surface plasmons occurs when the energy ($h\omega_i$) and momentum (hk_i) of the incident light matches with the energy ($h\omega_{sp}$) and momentum (hk_{sp}) of the surface plasmons. If the TCO film is too thin the SPP will strongly be damped because of radiation damping into the glass. And if the film is too thick the SPP no longer be efficiently excited due to absorption in the oxide film. So the occurrence of a minimum in the reflectivity curve for the transparent conducting oxide thin films is sensitive to the thickness of the metal oxide film and also to the angle of incidence at which the exciting light impinges on the surface which is shown in following figures.

RESULTS AND DISCUSSIONS

Surface plasmon polaritons are generated when the incident light gets coupled to the exciting light incident on the Kretschmann configuration [Fig. 1]. Optical responses of these materials with their film thickness and also with the angle of incidence is described in the study which confirms the response of the material to Surface Plasmon resonance. So, it is necessary to investigate the optical response of these materials with their film thickness and also with angle of incidence of the impinging light on the metal oxide surface. In this paper theoretical investigations on SPR resonance properties of ITO is done by MATLAB simulation technique. The graphical plots correspond to the TCO films with the lowest cross-over wavelengths and lowest losses. For this case, the thickness of the metal oxide is taken in the range from 10nm to 500nm and the angle of incident radiation is in between 40^0 to 70^0 and the wave number variation is taken upto 16000cm^{-1} and the permittivities of air and BK7 glass prism are 1(ε_{air}) and 2.31(ε_{prism}) respectively. For the theoretical simulations required high frequency dielectric constant (ε_{int}), plasma frequency(ω_p), and damping coefficient (Γ) of the Drude Dispersions for ITO, ZnO:Al and ZnO:Ga (2 at %) are given in Table 1.below

Table 1.

TCO Film	ε_{int}	ω_p (rad/sec)	Γ(rad/sec)
ITO	3.9	2.9×10^{15}	1.8×10^{14}
ZnO:Al	3.5	1.4×10^{15}	2.3×10^{14}
ZnO:Ga(annealed)	4.0	0.837×10^{15}	0.39×10^{14}

The variation of Reflectance with the wave no of the incident light in the 1.55 micron wavelength window also with different increasing angle of incidence from 45^0 to 69^0 is shown in Fig 2. At the minimum reflectivity position, the missing light is assumed to having been totally converted to surface plasmons at the interface which carry away the energy along the interface such that it cannot reach the detector. For 50 nm thick Al dopped ZnO thin film here we see that the surface Plasmon polariton responds to the excitation at 3873 cm^{-1} position of frequency. Here we can tell it asymmetric type resonance. The asymmetric peak occurred at 1273 cm^{-1} position. We can call it also Fano resonance. Resonance deep is greater for lower value of incident angle in the angular range 45^0 to 70^0. Where as in case of ITO we see that resonance occurred at frequency 7692 cm^{-1} for 50 nm thickness of the TCO film. From fig we can describe it a well known sharp resonance curve. Here we also see that for increasing incident angle the response curve is less deep.

From the figures it can be concluded that as per we increase thickness of TCO material film the resonant peak is much deeper for both ITO and ZnOAl and minimum reflectivity position shifts to higher wave no i.e lower wavelength region. So resonance will occurred at higher frequencies. For thickness 500 nm we see that both ZnOAl and ITO both responds very fruitfully to the predictions as stated earlier. Both the response figures are symmetric here. The behaviour of surface plasmons in ITO in the wave number dimension is presented in the following figures. The top left panel of Fig.1 shows the cross sectional spectral view of ITO from 42^0 to 69^0 in incremental steps of 3^0. Here it is noticed reflectance intensity is reduced in two regions of frequency which are apparent. The lower wave number position where Reflectance intensity reduced to 5.23 % observed at 3395 cm^{-1} (eV) which corresponds to the surface plasmon frequency ω_{sp}. The second dip in the reflectivity curve corresponds to the plasma frequency ω_p at 7533 cm^{-1} (eV) where reflectivity reduced almost 100% i.e. there is an absorption of light. The fact will be interpreted as the missing light is assumed to having been totally converted to surface plasmons at the interface which carry away the energy along the interface such that it cannot reach the detector. And the critical frequency is observed at 2228 cm^{-1} after which the reflectivity reduction occurred and the critical angle is 42^0 above which light is totally internally reflected. The variation of Reflectance with the angle of incidence is clearly shown in fig 1(b) followed by the contour image in fig 1(c). In this fig contour shows that around 45^0 of angle of incidence dual resonance takes place.

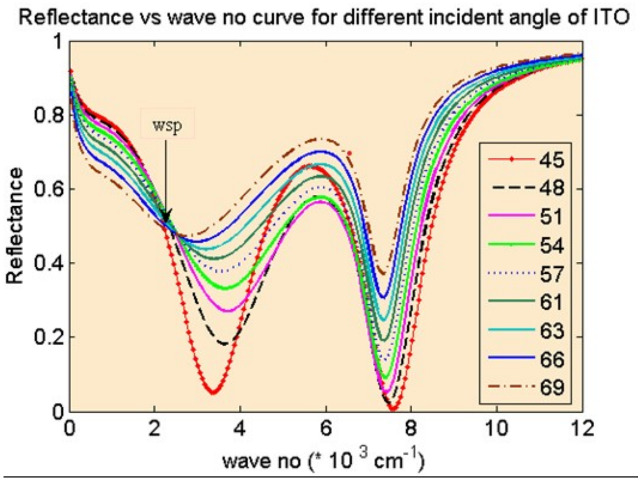

Fig. 1(a).

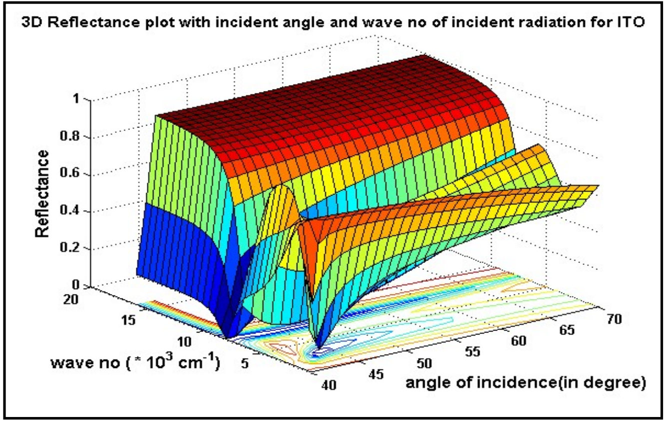

Fig 1(b).

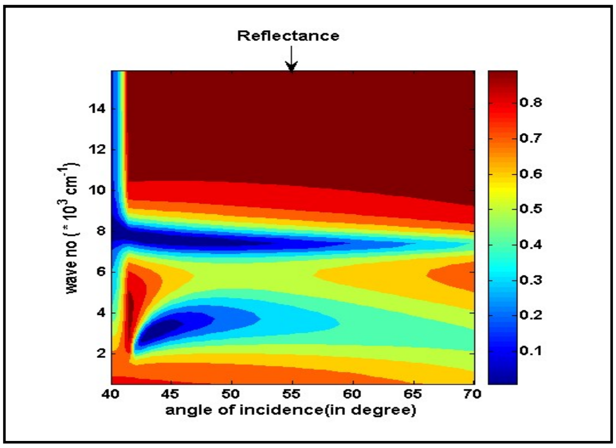

Fig. 1(c).

From the data calculated from the theoretical investigation it can be predicted that 50nm thick ITO film at 44^0 angle of incident light supports dual resonance at the frequency 3183 cm^{-1} and 7639cm^{-1} which is also supported by contour Fig 1(c) and 2(c). The variation of Reflectance (Rp) spectra for increasing thickness d with a fixed 45^0 angle of incidence is shown in Fig 2(a). From this fig it is clear resonant wave no position shifted towards higher region with increasing d, and also dual resonance can be predicted to occur for 50nm thick ITO film for 44^0 angle of incidence at the aforesaid frequency position which again justified by the contour Fig 2(b) and

2(c). However beside the perfect dual resonance position, in the fig every reflectance profile for below 200nm thickness shows asymmetric type nature with dual dips in the whole spectral region upto 16000 cm^{-1}.

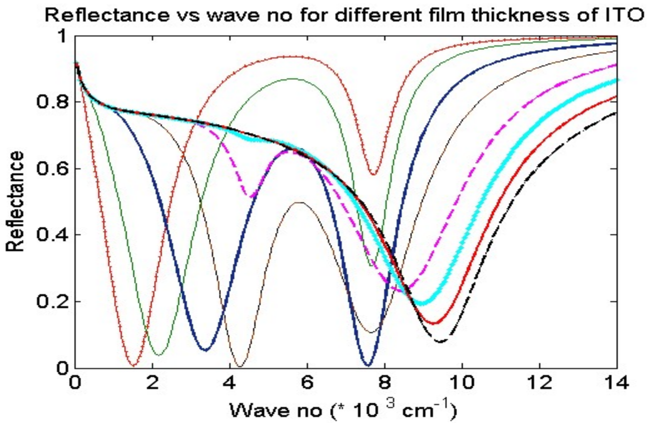

Fig. 2(a).

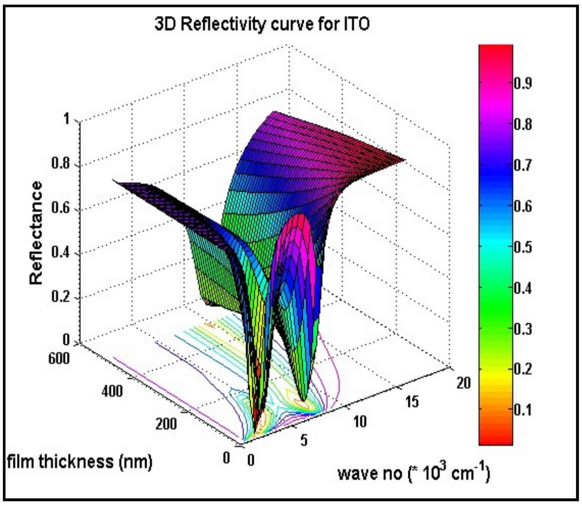

Fig. 2(b).

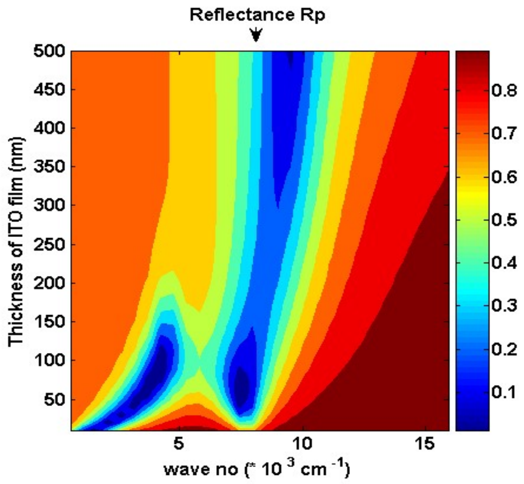

Fig. 2(c).

The 2nd dip position which is likely to be occurred at 7639 cm-1 for upto 100nm thick conducting metal oxide plotted in brown color. And for more thick film, resonance in the range 4000cm^{-1} to 12000 cm^{-1} shifted towards 9496cm-1 for 500nm film and profile became symmetric type with single dip and absorption at the same point increases with thickness. Fig2(b) describes the three dimensional reflectivity curve with x dimension as wave number and y dimension as thickness of the CMO film. This diagram follows the contour fig 2(c). Here also from color bar the variation of reflectance with film thickness and also the occurrence of dual resonance around 50 nm thickness is prominent. Here one important feature is that with increasing thickness the at the resonance position there 100% absorption is not happened. There is some sort of incident light reflects (≈7%) from the CMO surface. The point wsp signifies the wave no position where a longitudinal surface plasmon can be excited. At this position $\omega_{sp}=\omega_p/\sqrt{(1+\varepsilon_{air})}$ [16 er 18] $=\omega_p/\sqrt{2}$, as $\varepsilon_{air}=1$. It is well known that, below the plasma frequency incident radiation is reflected as shown in fig A and the reflected radiation will be in infrared region. And above the plasma frequency incident radiation is transmitted in the transparent conducting oxides which is supported by fig 1(a). Here for ITO, wsp=2228 cm^{-1} which corresponds to this frequency.

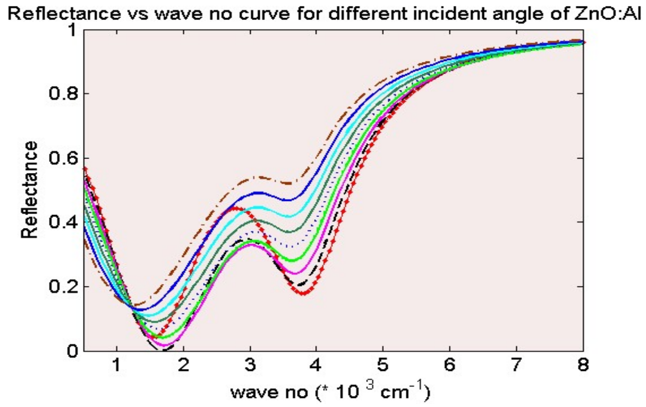

Fig. 3(a).

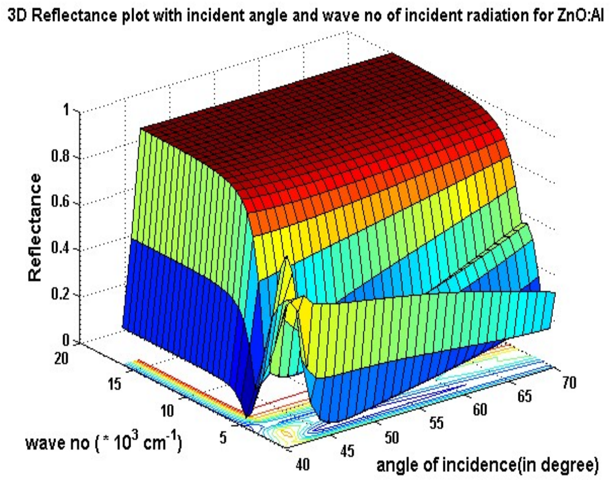

Fig. 3(b).

Now for aluminium doped ZnO film, the graphical variation of Reflectivity spectra observed from the CMO film with the wave number as a function of the incident angle from 45^0(red line) to 69^0(brown line) in 3^0 increments is given in Fig 3(a). The spectra is asymmetric type with dual dips (but not so much specified dual resonance in this thickness) occurring at 1698 cm^{-1} and 3820 cm^{-1}. So, at this situation Al doped ZnO film can be considered as fully transparent to the incident light. However in fig 3(b) followed by fig 3(c), reflectivity increases to some extinct (10%) and incident light coupled to surface plasmon polariton at single resonant frequency for the higher incident angle. Now the performance of ZnO: Al for SPR effect on the basis of the thickness of the CMO film is depicted in fig 4(a) and also three dimensional variation of reflectance with wave no and thickness described in 4(b) and (c) at 45^0 fixed incident angle. Similar to ITO, for below 100nm thick film reflectivity curve has two peaks and the minimum reflectivity is not closer to zero i.e some light may reflect back to detector. But for more thick film SPR effect can be seen with single resonant peak shifted from 3820 cm^{-1} (for 100nm) to 4934 cm^{-1} (for 500nm).

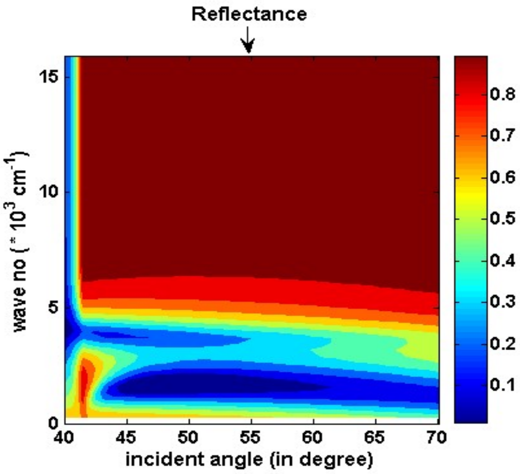

Fig. 3(c).

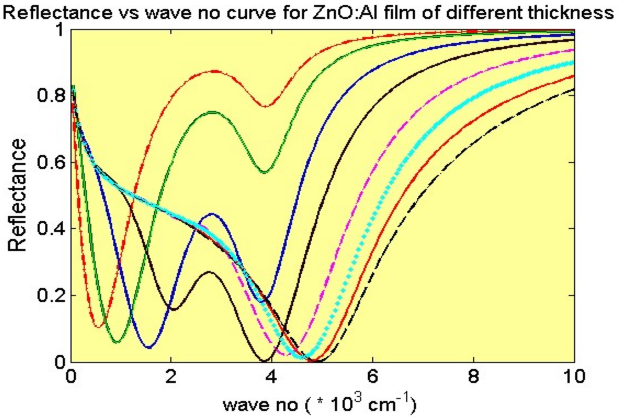

Fig. 4(a).

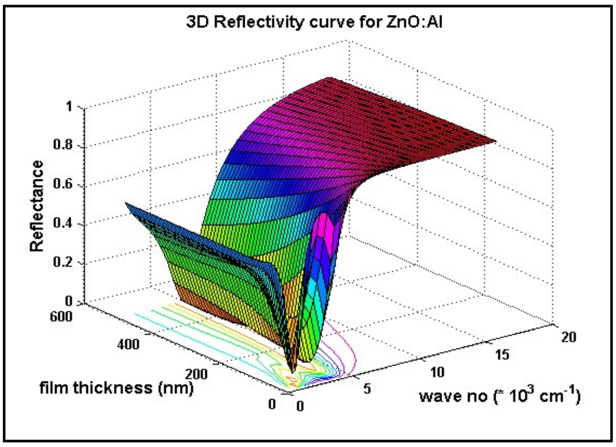

Fig. 4(b)

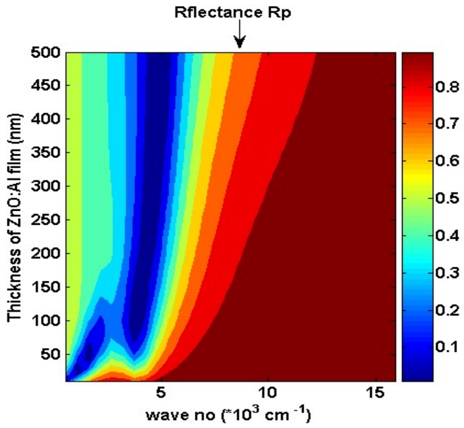

Fig. 4(c).

When the exciting light incident at an angle 46^0 on 73nm thick ZnO:Al film coated on the BK7 glass substrate, the film become transparent and almost fully absorbing to the incident light and causes for SPR effect at 1963 cm^{-1} and 3767cm^{-1} frequencies. This theoretical prediction is supported by the graphical contour plot 4(c). Now if the taken CMO film be Ga doped ZnO film then the response of the film to SPR effect with increasing incident angle is shown in fig 5[(a),(b) and (c)]. In fig 5(a), wsp(742cm^{-1}) represents the wave no corresponding to plasma frequency above which surface plasmon can response to exciting radiation.

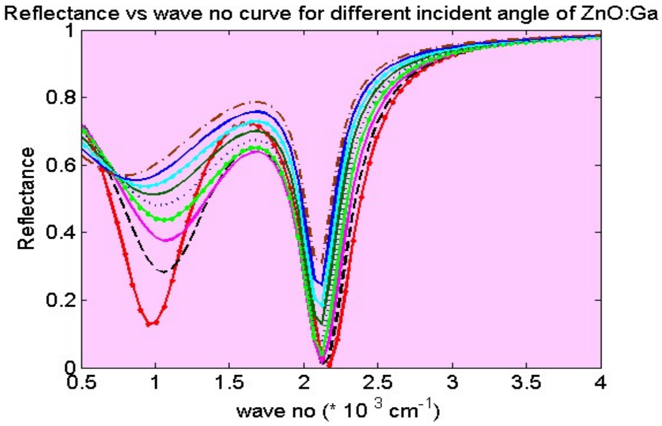

Fig. 5 (a)

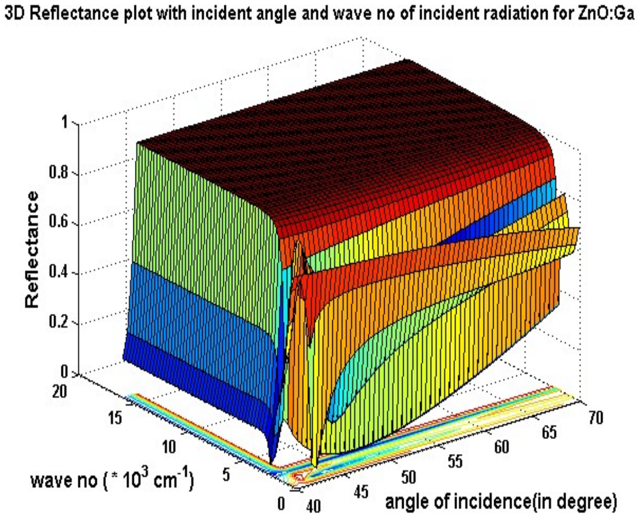

Fig. 5 (b).

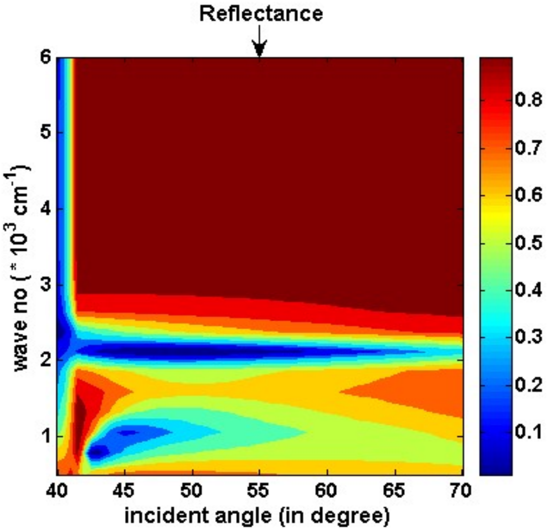

Fig. 5 (c).

From this contour fig we can see that there may be a chance of dual resonance at $\theta=43^0$. The minimum reflectivity position occurs at frequencies 901 cm^{-1} and 2175 cm^{-1} for 50 nm thick ZnO:Ga film.

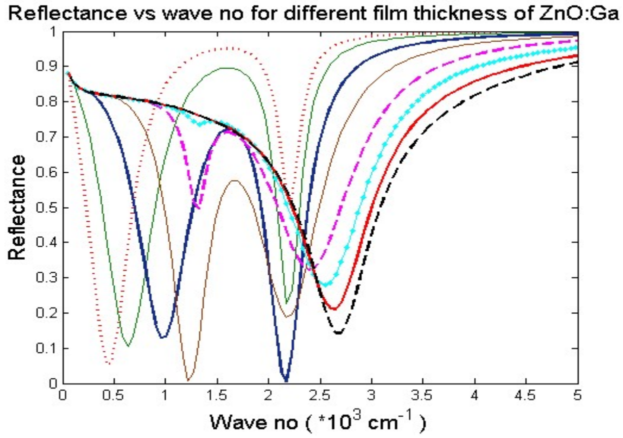

Fig. 6(a)

Fig 6 (a) is the plot of reflectivity detected back from the ZnO:Ga film coated on glass substrate with the frequency for different film thickness varying from 10nm to 500nm, which is more justified in the next figures 6(b) and 6(c). Here three dimensional variation of Rp is given at a time with wave no and also thickness. The spectral reflectance curve for 500nm become symmetric and having single resonant frequency at 2706 cm-1 .Above 250nm value of d, the profile become symmetric with single dip. But the film remains to some extent (~ 15%) opaque to the incident radiation.

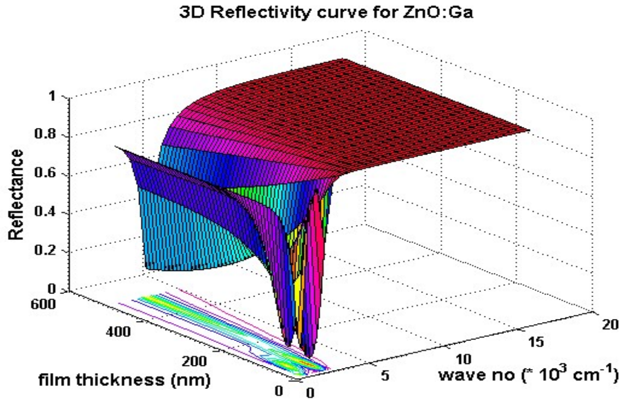

Fig. 6(b)

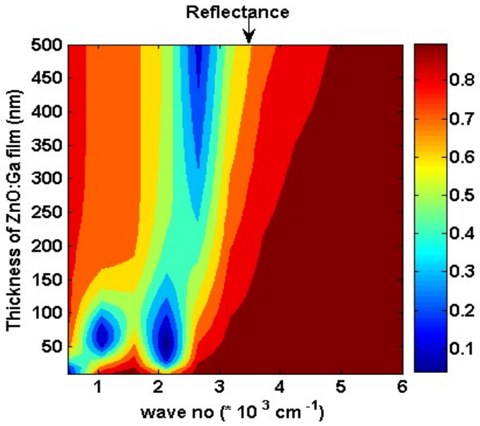

Fig. 6(c).

From these figures it is seen than Al doped ZnO give better sensitivity for the whole thickness range. In the minimum reflectivity position of wave no for ZnO: Al the reflectance is more minimum than others though the frequency range of resonance are different for the different film materials.

CONCLUSIONS

For the dual resonance is concerned both the three materials can support the feature under some constraints of incident angle θ, thickness of the TCO film. Different TCO film have different resonant frequencies in perfect dual resonance condition. This particular behaviour of the TCO thin film can be used as filtration technique in opto electronic devices.

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