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Optical Properties of Metal–Dielectric Nanocomposites thin film (Al_2O_3:Cu)Produced by Pulsed Laser Deposition

Amer S. Al-Khafagi[2], Sahib N. Abdulwahid[3]*, Ghaleb A. Al-Dahash[1]

[1,2]University of Kufa, College of Education for girls, Department of Physics, IRAQ.
[3]University of Babylon, College of Science for girls, Department of Laser Physics, IRAQ.

Abstract : We prepared Metal–Dielectric Nanocomposites thin film (Al_2O_3:Cu) Produced by Pulsed Laser Deposition with two different methods ,first is by doping Cu in Al_2O_3 with different concentrations of Cu .Second is embedded Cu in Al_2O_3 matrix , where deposited on glass substrate with different temperatures and different laser parameters (energy and pulses).Optical properties of thin films such as absorption, absorption coefficient, complex refractive index of the films have been estimated .The absorption coefficient was determined from absorption measurements at room temperature in the wavelength range 300-500 nm. We study the effect of each of two methods on theses optical properties. We showed a new absorption peak at (590nm).

Key words : Al_2O_3:Cu, Metal–Dielectricnanocomposite, refractive index, PLD, optical Properties.

Introduction

The discovery and use of copper throughout the history of mankind has been as old and large they have found numerous applications, such as microelectronics, becoming indispensable to our civilization material. Because this has an increased interest in the analysis of thin films of copper and copper oxide.

Copperoxide prepares a material in monocrystalline, polycrystalline or amorphous, there are a variety of techniques. In recent years, laser ablation has been used for numerous applications, standing out as a versatile technique for growth of thin films, production of nanoparticles and clusters[1,2]. In the technique of pulsed laser deposition (PLD), the white, which is located in a vacuum chamber is irradiated by a high power laser, allowing evaporate or ablate the material and create around a plasma. The first to use this technique were Smith and Turner in 1965 by a ruby laser a vacuum chamber[3]. The most common techniques for growing thin films are ALD (Atomic Layer Deposition)[4], unbalanced magnetron sputtering.

A new technique for production of thin film by using laser beam is metallic nanoparticles (NPs) embedded in a dielectric host which Physical and chemical properties of nanocomposite materials formed are strongly dependent on their morphological features such as size, shape or size dispersion as well as on their distribution within the matrix.

Thus, the control of these features during the production has been a challenge for long time and, in fact, the lack of production methods with the required degree of control has been one of the major drawbacks for the development of practical applications based on this type of materials. A strong experimental effort has been made in recent years to develop methods with the required level of control, the most widely used ones being ion implantation and thin film technologies. While the former introduces the metal into a bulk dielectric, the second is based on the production of both the host and the NPs using the same deposition technique. A wide variety of wavelengths and pulse durations have been used for thin film deposition by PLD. The reports published in the literature on material ablation and deposition include laser pulses ranging from infrared to ultraviolet and from continuous to femtosecond pulses. Nevertheless, the conditions that have demonstrated up to date to be more successful for material deposition and for which more results are available are what we would refer to as classical PLD, in which a laser with wavelength in the ultraviolet range (≈193–351 nm) and laser pulses in the nanosecond regime is used as ablation source. This research was carried out using a laser pulsed Nd: YAG operating in the infrared and uses a glass substrate to produce thin films of Alumina-copper Nanocomposites. The Nanocomposites thin film produced is analyzed by optical spectroscopy and films are characterized by X-Ray, Scanning electron Microscopy (SEM) and Atomic Force Microscopy (AFM).

Experimental Technique

A Nd: YAG 1064 nm, 10 ns duration by pulse, pulse energy of 250mJ, repetition rate 10 Hz is focused by a lens focal length 30 cm through one of the ports of the vacuum chamber. On a holder the sample is located Cu, 95% purity. Vacuum Pressure about 10^{-5} mbar is varied; the substrate temperature is kept at 150 Ċ. We have used two methods to preparation of thin film, the first was Adding Cu to Al_2O_3 with different concentrations and Pressed in tablets, and the second was by embedded Cu particles in dielectric host (Al_2O_3) by applied laser pulses in Al_2O_3 tablet and Cu tablet alternately. It was used two tablets the first of alumina (Al_2O_3) and the second of copper (Cu) on a rotating base so that spins 360 degrees on the pirate where alternating laser beam to fall on each of the two tablets. The experimental detail growth technique and making laser ablation produced diagrammed in Fig. (1).

Fig.(1) PLD system

Results and Discussion

The optical properties which have been obtained from the deposition process by using two ways of deposition, first method is Add impurities way and the second was by embedded Cu particles in dielectric host (Al_2O_3) where the results have been found and the calculation of the optical properties of the above-mentioned cases.

The figure (2) represents a change in absorbance with wavelength of the range of (300-800nm) for pure deposited thin films without impurities or embedded either copper or alumina on a glass substrates with different preparation conditions. we noted that the value of the absorbance decreases with increasing wavelength and we also noted that the absorbance value decrease with low rate at high wavelengths range (low energy range) while decreasing absorbance significantly in the short wavelengths range (high energies range), this behavior due to increasing transmittance in the high wavelengths region because the excitation and absorption processes needs high energy. We also noted that absorbency of copper thin film greater than the absorbency of the (Al2O3) thin film due to increased bulk density of copper more than twice than it is alumina.

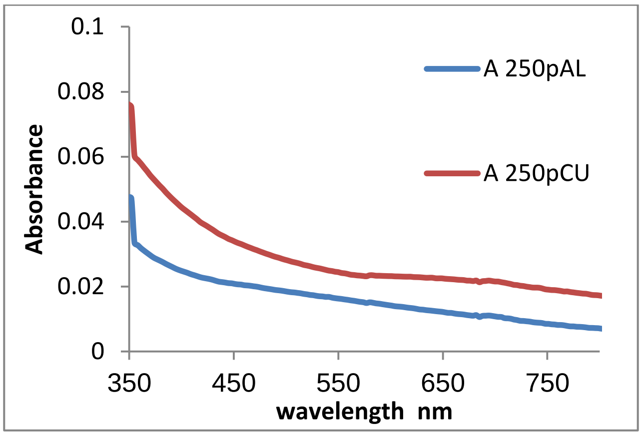

Fig.(2)absorbance of pure materials for Cu & for Al_2O_3 at 250 Ċ & 250 pulse

Forms (3), (4), (5) and (6) describes the effect of additive of copper with different ratios, as well as the effect of increasing the number of pulses to change absorbency of (Cu-Al2O3) thin films with constant substrate temperature where we noted that absorbance edges appears a simple shift toward the infrared region (red shift) with different preparation conditions due to an increase in particle size with increased of Cu additive because of coalescences .We also noted an increase of absorbance value with the increasing number of pulses due to an increase in the thin films thickness and this is consistent with the researcher[5].With stay the same behavior of decreasing in absorbance value at high and low energies.

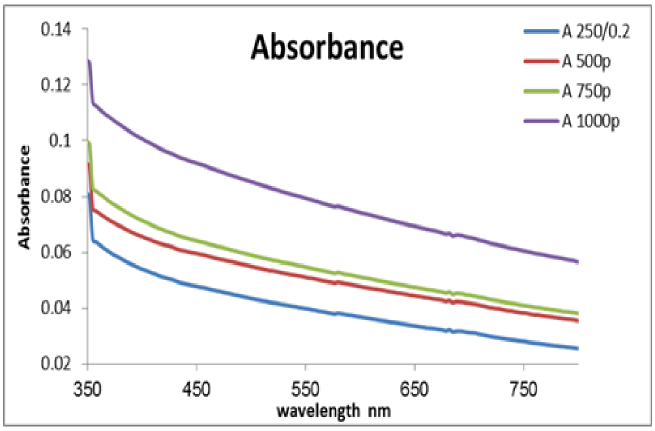

Fig.(3) absorbance of doped Al_2O_3 thin film with(0.02 Cu) at 250 Ċ & (250,500,750,1000) pulse at laser energy (140mJ)

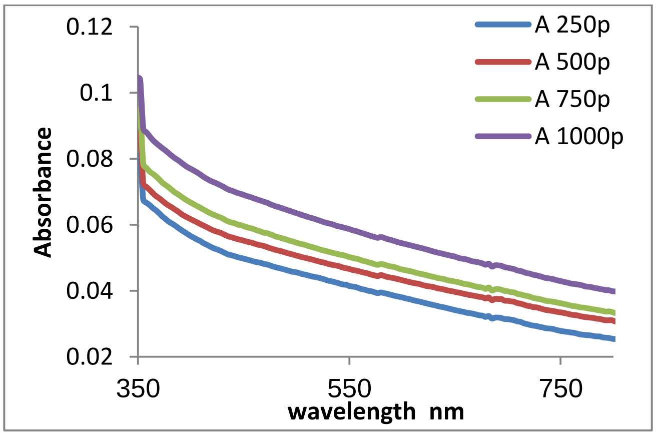

Fig.(4) absorbance of doped Al_2O_3 thin film with(0.05 Cu) at 250 Ċ & (250,500,750,1000) pulse at laser energy (140mJ)

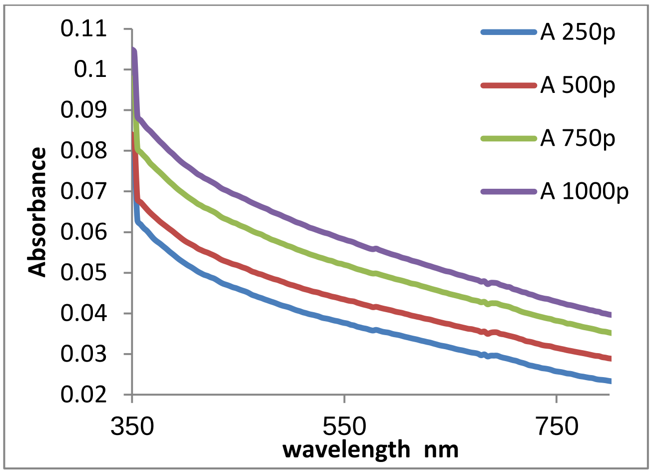

Fig.(5) absorbance of doped Al_2O_3 thin film with(0.02 Cu) at 250 Ċ & (250,500,750,1000) pulse at laser energy (140mJ)

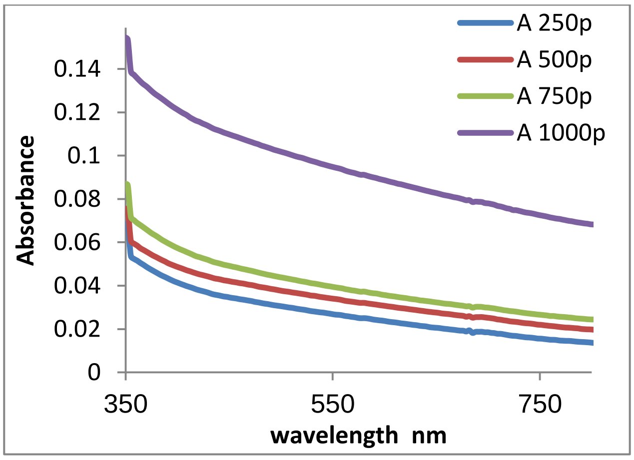

Fig.(6) absorbance of doped Al_2O_3 thin film with(0.1 Cu) at 250 Ċ & (250,500,750,1000) pulse at laser energy (140mJ)

While the second way the implant (embedded) and also shown he as in figures (7) and (8) where we noted a difference in the value of the absorbance change with the increasing of substrate temperature, this increasing of absorbance due to the decrease in transmission with appear a new absorption peak (590nm) wavelength , at (150 Ċ) substrate temperature where this value increases with increasing pulses number, this is because of when the particle size of the copper is much smaller than the wavelength of applied beam (visible and infrared zone), so that the electric component of electromagnetic wave will Polarize free carriers of copper nanoparticles which leads to attract the carriers to this polarization, which will appear as a form effective dielectric constants for thin film nanocomposite material (Al2O3: Cu) as we will see in the dielectric constant debate that this effect is called (dielectric or classical confinement) where one application of this effect is the coloring of some types of glass process at work planting for some types of nanomaterials in it. This is consistent with the researcher (Kreibig U)[6]. The small size of the copper granules compared to the wavelength of this leads to the possibility of raising the surface plasmon resonance (SPR) at the surface (Metal-dielectric).

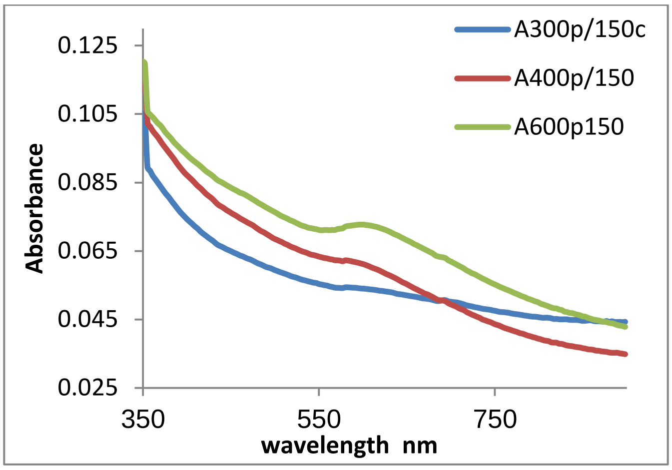

Fig.(7) absorbance of embedded of Cu in Al_2O_3 thin film at 150 Ċ & (300,400,600) pulse at laser energy (140mJ)

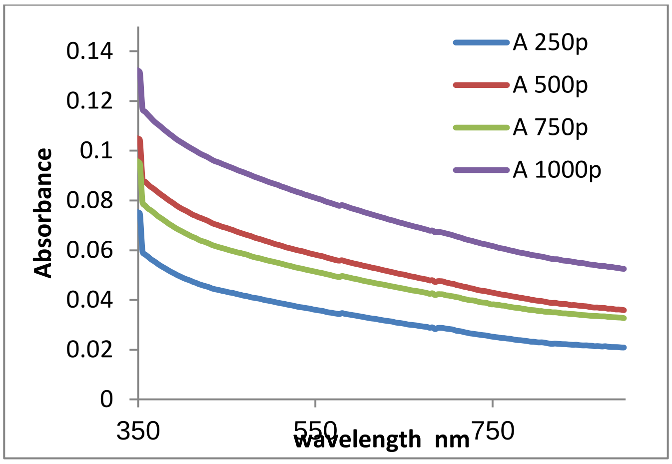

Fig.(8) absorbance of embedded of Cu in Al_2O_3 thin film at 250 Ċ & (250,500,750,1000) pulse at laser energy (100mJ)

To calculate the absorption coefficient of the methods used and comparison with every way in terms of increase and decrease in the absorption coefficient. We notde that the absorption coefficient increases in the ultraviolet region along at the beginning of the visible region. This indicates the presence of energy bands is a capability to absorb such energies that will carry the photon wavelength associated with this and we observed that the absorption coefficient increases with the absorbance. It is also known that the absorption coefficient refers to the loss in the light of falling through a certain thickness. Where Figures (9), (10), (11), (12) and (13), showed the changing the absorption coefficient with the wavelength of the doped thin film (Al_2O_3: Cu) with Different preparation conditions. We noted the absorption coefficient around ($10^4 cm^{-1}$) at range of wavelengths (300-800nm), this shows that the electronic transfer directly but increases (the values of absorption coefficient) with an increased number of pulses (increase of the thickness) and this helps to increase the expectation of transitions directly in over the short wavelengths because of direct transfer need to be enough energy to occur ,also we noted slight increased absorption with the increase in the proportion of doped coefficient and this is attributable to the increased particle size with increasing the proportion of doping. While in the case of the implant (embedded) we noted an increase absorption coefficient for rates in the case of doping suggest the possibility of a transfer directly based on temperature ($150C^0$) and be surface plasmon resonance (SPR) and on copper nanoparticles, which have around (590nm) as in figures (14)and (15). This is agreement with (Seona, R)[7].

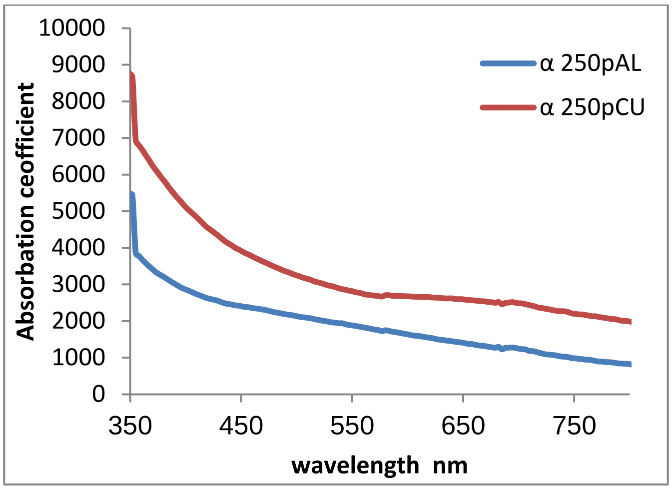

Fig (9) shows Absorption coefficient of pure Al_2O_3 and pure Cu thin films at 250 Ċ & (250) pulse at laser energy (140mJ)

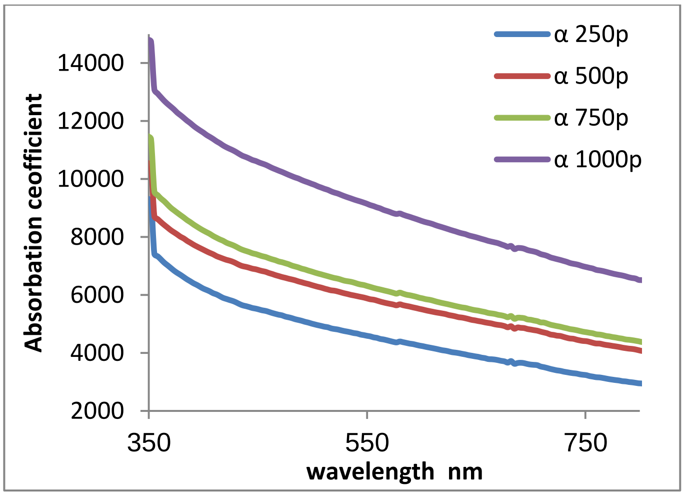

Fig.(10) shows Absorption coefficient of doped Al_2O_3 thin film with(0.02 Cu) at 250 Ċ & (250,500,750,1000) pulse at laser energy (140mJ)

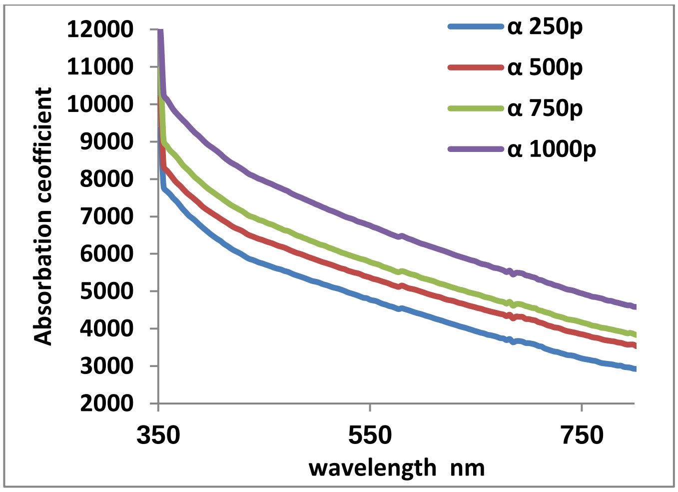

Fig.(11) shows Absorption coefficient of doped Al_2O_3 thin film with(0.05 Cu) at 250 Ċ & (250,500,750,1000) pulse at laser energy (140mJ)

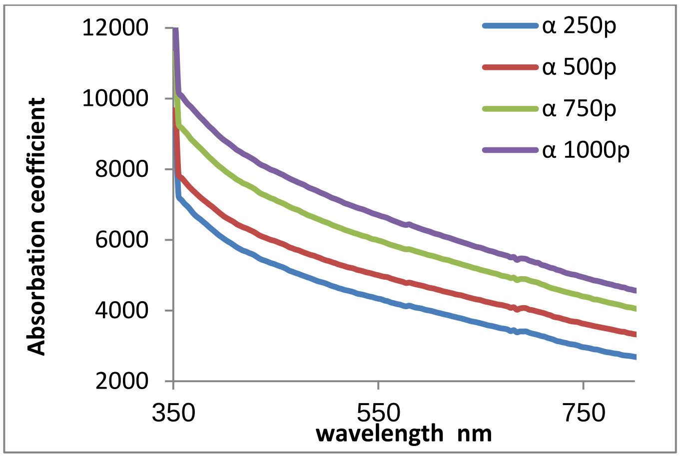

Fig.(12) shows Absorption coefficient of doped Al_2O_3 thin film with(0.07 Cu) at 250 Ċ & (250,500,750,1000) pulse at laser energy (140mJ)

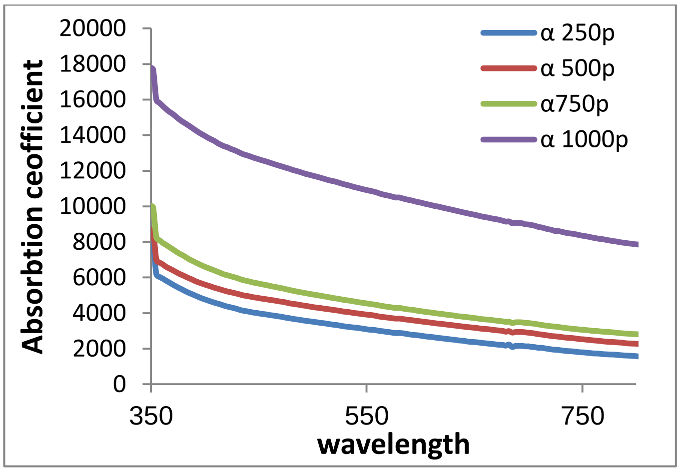

Fig.(13) shows Absorption coefficient of doped Al_2O_3 thin film with(0.1 Cu) at 250 Ċ & (250,500,750,1000) pulse at laser energy (140mJ)

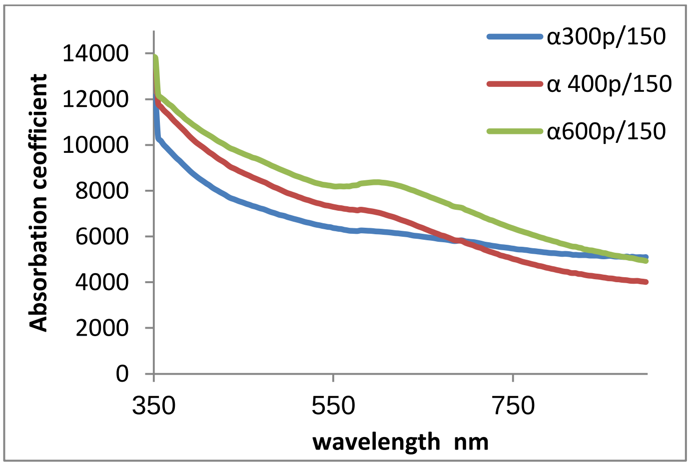

Fig.(14) shows Absorption coefficient of embedded thin film of Al_2O_3 by (Cu) at 150 Ċ & (300,400,600) pulse at laser energy (140mJ)

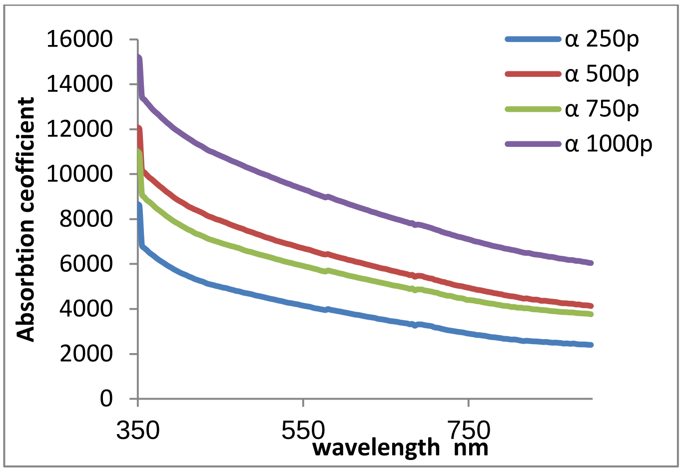

Fig.(15) shows Absorption coefficient of embedded thin film of Al_2O_3 by (Cu) at 250 Ċ & (300,400,600) pulse at laser energy (100mJ)

The figure (16) represents a change in the refractive index for each of the copper thin film and alumina (host) with a wavelength where we note that they possess the same behavior, but differ in the value of (n) where the n of copper thin film greater than (n) of the host thin film(alumina) because of the difference in the density of materials where the atomic-sized of copper greater by 2.3 times than it is of host(alumina). We also noted that the refractive index of the Host and added decreased with increasing wavelength due to increased Transmission at higher wavelengths.

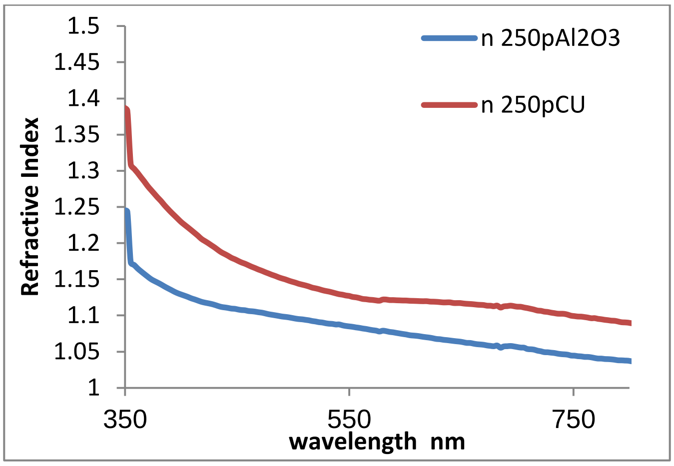

Fig.(16) shows refractive index of pure Cu And pure Al_2O_3 at ($250C^0$),(250) pulses and energy (140 mj)

The figures (17), (18), (19) and (20) showed the influence of both doping ratio and pulses number to change the behavior of the refractive index (n) where we first noted that the refractive index of (Cu: Al2O3) is much higher than the host due to the presence of copper inside the composite, and this increase also depends on the increase of thickness due to increased number of pulses and a second noted a gradual decrease of the refractive index with increasing wavelength due to increase the of transmission for all doping ratios, where this is consistent with the theory of Mie for scattering conductive spheres in dielectric host[8-11]. In addition, we noted an increase the value of the refractive index when increasing copper doping ratios up to (n=1.75) at the absorbent edge at doping ratio (0.1). This is mean possibility to control the value of a refraction index by selecting doping ratios as well as it can be controlled to specify the number of pulses and As this behavior is new behavior as regard the host material (Al_2O_3) is amorphous materials and this behavior is agreement with the researcher Del Coso, R.[12].

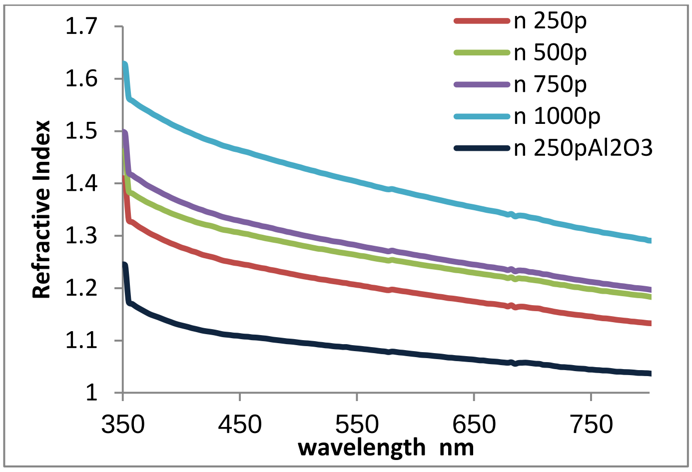

Fig.(17) shows refractive index of doped (Al_2O_3:Cu)by(0.02Cu) at ($250C^0$),(250,500,750,1000) pulses and energy (140 mj)

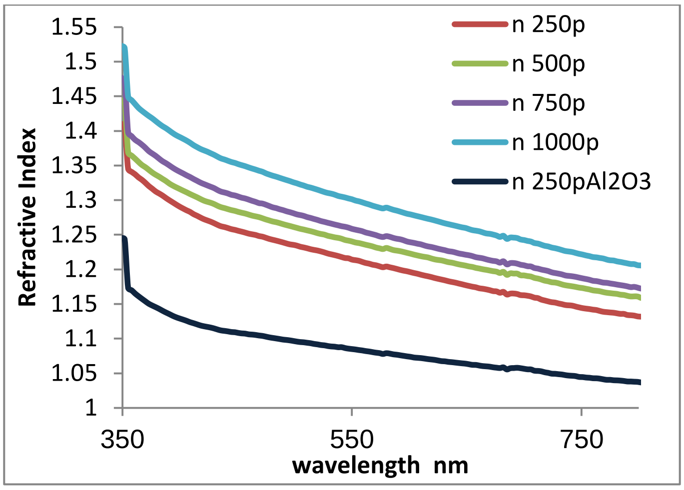

Fig.(18) shows refractive index of doped (Al_2O_3:Cu)by(0.05Cu) at ($250C^0$),(250,500,750,1000) pulses and energy (140 mj)

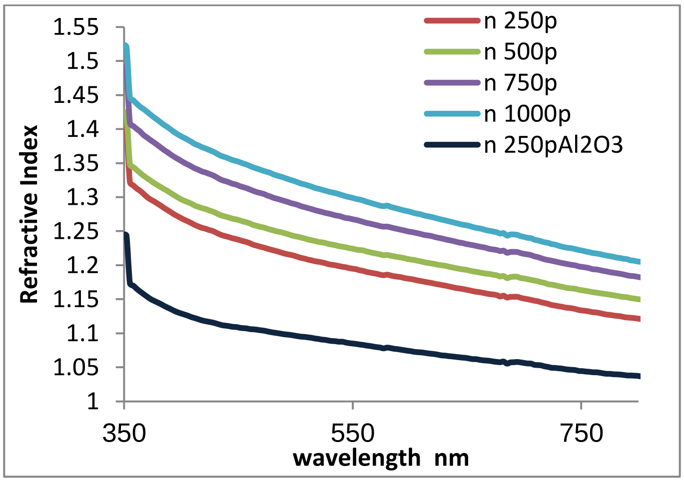

Fig.(19) shows refractive index of doped (Al_2O_3:Cu)by(0.07Cu) at ($250C^0$),(250,500,750,1000) pulses and energy (140 mj)

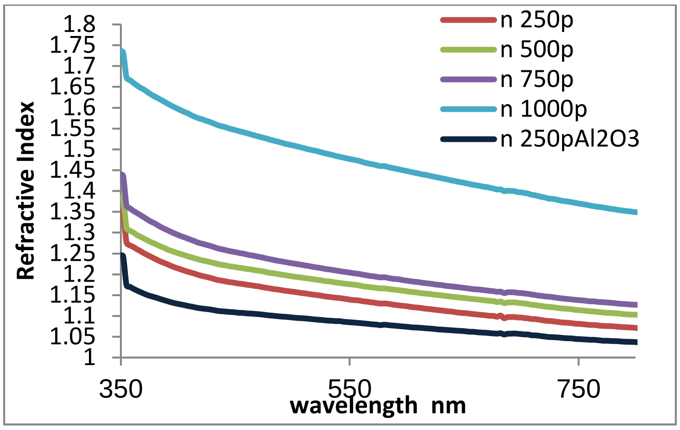

Fig.(20) shows refractive index of doped (Al_2O_3:Cu)by(0.1Cu) at (250C^0),(250,500,750,1000) pulses and energy (140 mj)

The figures (21) and (22) shows the change in refractive index (n) with the wavelength for thin films prepared by implantation method (embedded) where we noted appear of SPR within the range (590-600nm) wavelength which appeared very clear at 150C^0 temperature and when the number of pulses 400P and 500P was not clear when the temperature 250C^0 due to the formation of nucleation processes and growth optimal and appropriate to form (shells) from the host on the copper grains or may be oxidized which leads to generate the SPR within this range and also agree with previous research[8, 13].

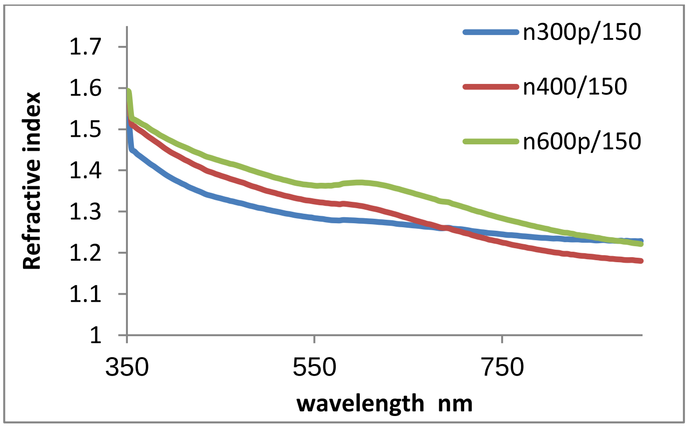

Fig.(21) shows refractive index of (Al_2O_3:Cu)embedded (Cu) at (150C^0),(300,400,600) pulses and energy (140 mj)

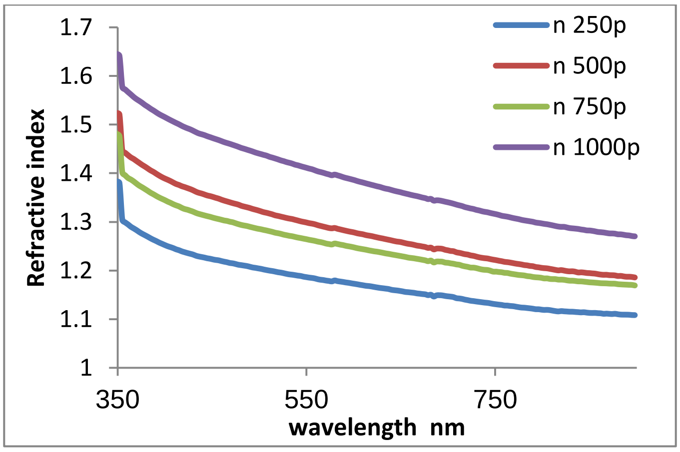

Fig.(22) shows refractive index of (Al_2O_3:Cu)embedded (Cu) at (250C^0),(250,500,750,1000) pulses and energy (100 mj)

The extinction coefficient (k) represents the amount of energy absorbed in a thin film or possible to represent attenuation of electromagnetic wave inside the material, which is a part of the imaginary refractive index of the complex according to the equation (N=n+ik) and the associated absorption coefficient (α) according to the equation ($k= \alpha\lambda/4\pi$).

We Noted that extinction coefficient for doping case as in Figures (23), (24), (25), which depends as a function of wavelength, noted that in the ultraviolet region and in the low concentrations be low extinction coefficient while the coefficient increases with increasing of Cu concentrations where it is higher than in the embedded process in order to increase absorption coefficient, because the relationship between them is a direct correlation. While in the visible region, the extinction coefficient is small because low absorption in this area, as well as in the embedded process is more than other cases and thus increases all other cases. As shown in Figures (26), (27).

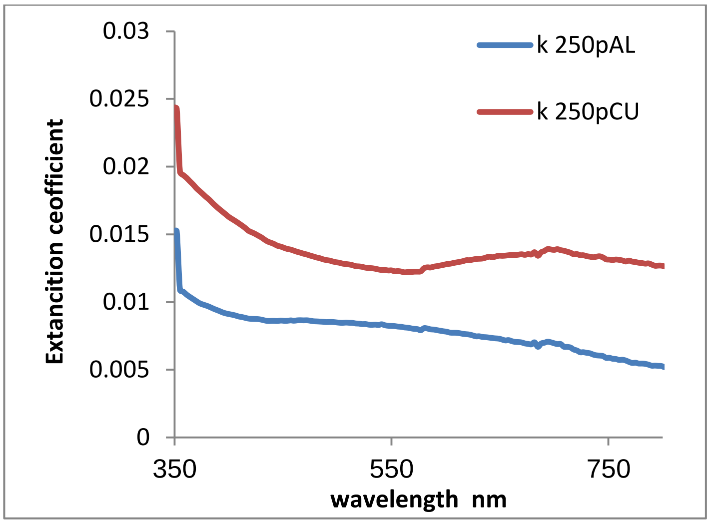

Fig.(23) shows extinction coefficient of pure Cu And pure Al_2O_3 at ($250C^0$),(250) pulses and energy (140 mj)

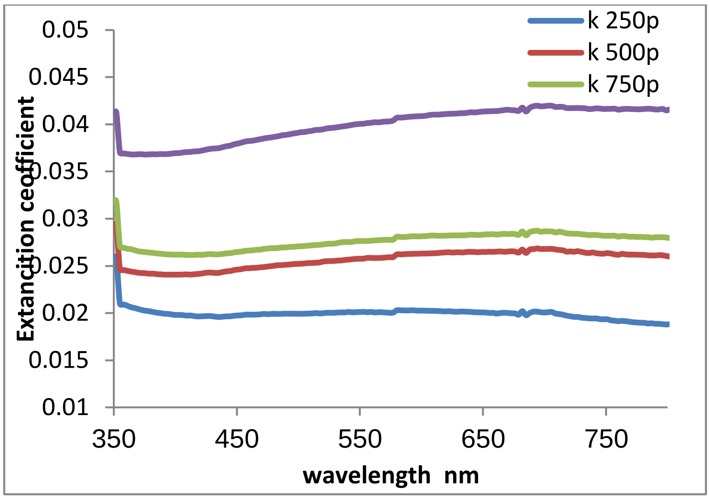

Fig.(23) shows extinction coefficient of doped Al_2O_3 with(0.02Cu)at ($250C^0$),(250,500,750,1000) pulses and energy (140 mj)

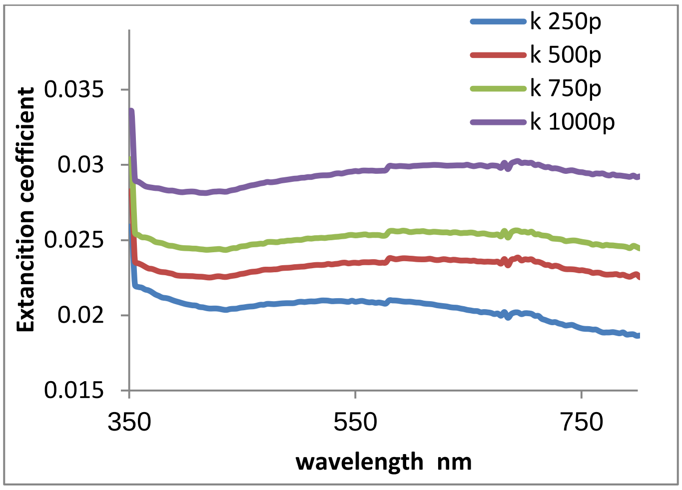

Fig.(24) shows extinction coefficient of doped Al_2O_3 with(0.05Cu)at ($250C^0$),(250,500,750,1000) pulses and energy (140 mj)

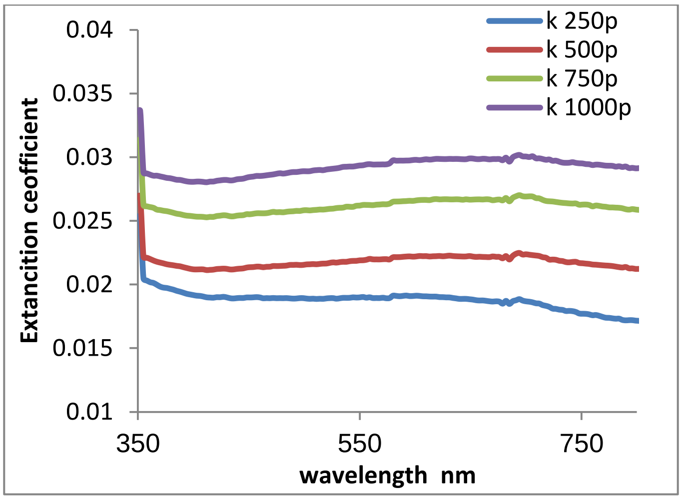

Fig.(25) shows extinction coefficient of doped Al_2O_3 with(0.07Cu)at ($250C^0$),(250,500,750,1000) pulses and energy (140 mj)

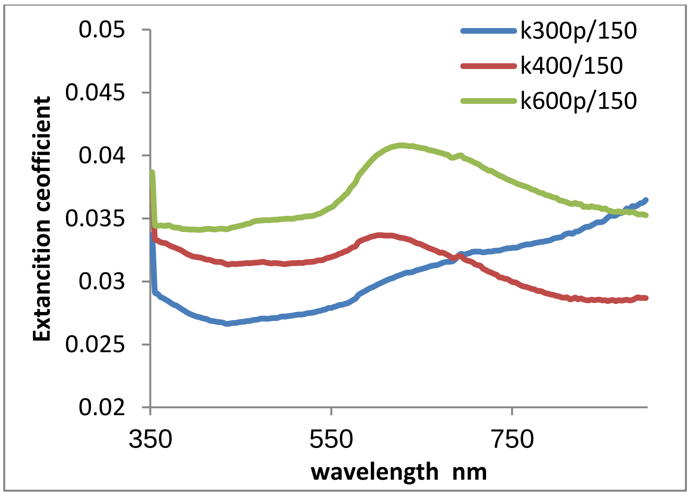

Fig.(26) shows extinction coefficient of embedded Al_2O_3:Cu)at ($150C^0$),(300,400,600) pulses and energy (140 mj)

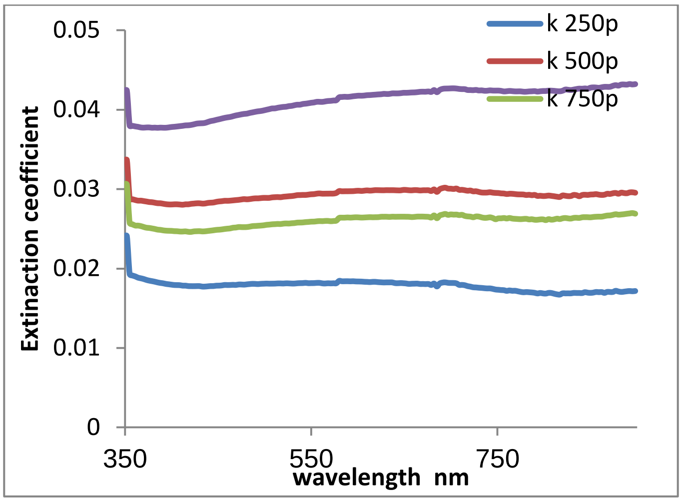

Fig.(27) shows extinction coefficient of embedded Al_2O_3:Cu)at ($250C^0$),(250,500,750,1000) pulses and energy (100 mj)

Conclusions

1. Pulsed laser deposition method proven in the preparation of highly efficient nanocomposite in the form of thin films composed of nanocrystals of metal implanted (embedded) in dielectric host.
2. The possibility to control the particle size by controlling the number of pulses of laser for the metal in the case of the implant (embedded) in addition to the control of the ratio of doping.
3. The grains produced by the implant (embedded) are performed by nucleation, growth and coleasenses at the surface of the base operations, leading to the largest distribution when increasing the number of pulses.
4. The refractive index of a material is the key parameter that affects all optical properties.
5. Index changes produced by presence of metal particles in a dielectric because of strong effects accompanying excitation of surface plasmons. This allows manufacturing highly selective filters and mastering the colors.
6. The surface plasmon resonance of pulsed laser deposited nanocomposite films formed by Cu NCs embedded in an Al_2O_3host shifts towards the red when the effective aspect ratio of the NCs decreases as a consequence of an increase in their anisotropy.

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