Journal of Kerbala University , Vol. 6 No.1 Scientific .March. 2008

Cole –Cole diagrams of gold doped germanium films

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Abstract:

The gold doped germanium films with different gold concentration CAE(0,0.08,0.15, 0.2,0.3) weight %have been prepared by thermal evaporation under vacuum of (10^{-5} Toor) with thickness (1 μm) at substrates temperature (553K)and annealed at(793K)to achieve good diffusion of gold in the germanium films texture. Measurements of The dielectric measurements were carried out over frequency range (10^2-10^7 Hz) for all pure and doped Ge films .It was found that all samples displayed dielectric dispersion thus the curves log (ε_1)versus log (w),log (ε_2) versus log (w)gave direct evidence of the existence of Debye-type relaxation have a wide distribution of relaxation times. Cole –Cole diagrams had been used to determine the distribution parameters (α) and the relaxation time (τ).

Introduction:

Dielectric materials are those materials whose electrons are strongly bounded to the atomic structures by restoring forces[1]

Polarization is a phenomenon includes the appearance of electric charges in the surface of the dielectric [2]. On the macroscopic scale , the polarization (P) is defined as [2]:

$P=\varepsilon_o(\varepsilon_r-1)E$.............(1)

Where ε_o is the vacuum permittivity , ε_r is the real part of permittivity an E is applied field .There are several types of polarization present when an external electric field is applied on the dielectric material. The total polarization (P_t) will be considered to be the sum of the individual polarization vectors [3]:

$P_t=P_e+P_i+P_0+P_s$.......(2)

Where the terms on the right hand represents the individual electronic, ionic, oriantation,and the space charge vector respectively.

Dielectric relaxation is defined as the exponential decay of the polarization (P) with time (t) ,when an external field is removed [4]]. However, when an external field (E) is applied through dielectric material the dipoles does not attain its orientation immediately, which is mean that there is phase difference between dipoles polarization and applied field [5,6].Also the dipoles polarization required amount of time after removing the external field ,such that the rise and the decay of polarization can be expressed in exponential form (for rise and decay respectively):

$P(t)=P_o(t)\ (1-e^{t/\tau})$........(3)

$P(t)=P_o(t)\ e^{t/\tau}$..............(4)

Where (τ) is the relaxation time which is defined as the time required to reducing the dipole polarization to 1/2 from it is maximum value ,P_o is orientation polarization .

There are several models discussing dielectric relaxation to fined out the causes of dielectric dispersion such as Debye dispersion,Fuoss-Kirkwood and Cole –Cole respectively.Debye proposed that insulating material is usually used over the wide range of frequency and predict that when external alternating field is applied at vary high frequencies the molecular dipoles have no time enough to charge their orientation in keeping with the applied field since its period to orientate is less than the relaxation time and the dielectric comprises only the contributions from the electronic and ionic polarization .The existence dielectric constant is called optical dielectric constant(ε_∞) [6,7] ,while at very low frequencies the period of applied field is very large relative to the (τ) ,here the polarization is go to its maximum value ,and the dielectric constants contain the full contribution from the dipoles polarization. and the power loss is zero[6,8] ,here dielectric constant called the static dielectric constant (ε_s) as shown in Fig (1).

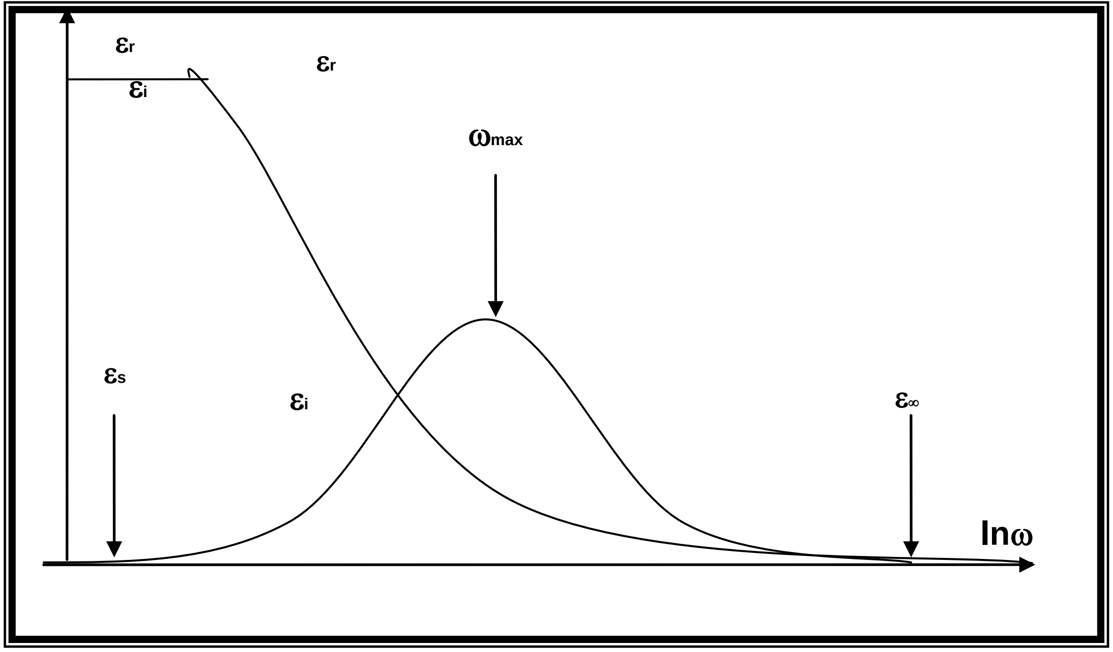

Figure (1) Debye dielectric dispersion curves.

At intermediate values of frequencies of constant relative permittivity a step occurred centered around the maximum angular frequency ($w_{max}=1/\tau$) .The dipoles polarization dose not keep in phase with the external field and the

power loss has maximum value at $w_{max}=1$ [6,8] .The tow components of the dielectric constant ε_1, ε_2 give the physical meaning of the dissipation factor ($\tan\delta$)which is the (energy dissipated per cycle /energy started per cycle or,:-

$\tan\delta= \varepsilon_1/ \varepsilon_2$...........................(5)

also ε_1, ε_2 can be written as :-

$\varepsilon_1= \varepsilon_\infty+ \varepsilon_s- \varepsilon_\infty/1+w^2 \tau^2$...............(6)

$\varepsilon_2= (\varepsilon_s- \varepsilon_\infty)w\tau /1+w^2 \tau^2$...............(7)

Equations (6,7) are called Debye dispersion equations from these equations the maximum value of ε_2 occurred when $w=w_m=1/\tau$, where the polarization keep the applied field and resonance is occurred .

The mechanism of the A.C conductivity (σ a.c(w)) gives informatics about the nature of polarization mechanisms in dielectric ,also provide information about the capacitance, interface and the amount of conductivity present[9].A frequency dependence on a.c conductivity (σ a.c(w)) has been observed in many amorphous semiconductors has the form[10]:-

$\sigma_{a.c}(w) =A w^s$..................(8)

Where s, is the exponent factor ,A is the proportional factor .

The total measured conductivity (σ t(w)) at a given frequency (w) is separable in to d.c and a.c components ,namely

$\sigma_t(w)= \sigma_{a.c}(w)+ \sigma_{d.c}$.........(9)

$\sigma_t(w)= A w^s + \sigma_{d.c}$

Where (σ d.c) is the d.c component at low frequency, while

(σ a.c(w) is due to relaxation processes[11].

For non metallic materials the electrical conductivity depends on the temperature (T) according to the Arrhenius equation [12] :-

σ a.c(w)= σ_0 exp(-E_{ac}/k_BT)............(10)

When an electric field is applied through the dielectric material ,the dipoles in the dielectric tend to follow the field ,flipping back and forth as the field reverses its direction during each cycle ,because the dipoles experiences some fraction due to its collision with other molecules .This means that some fraction due to its collision with other molecules ,This means that some energy is absorbed from the field ,which called dielectric loss[13]. In practice its convenient to specify the absorption losses in dielectric at a given frequency and temperature by the loss tangent (tan (δ)) ,where δ is angle between the displacement current and its loss component .The behavior of the dielectric loss under an alternating current can be expressed in an equivalent circuit by combination of capacitor (C) with resistance (R) connected in parallel or in series[14].

When sinusoidal voltage ($V=V_oe^{iwt}$)) is applied through the dielectric ,then resultant current is the sum of conduction current (I_C) which is in phase with the voltage (V) and displacement current (I_r) which is(90^0) out of phase which (V) [16]:-

$I=\varepsilon^*C_odV/dt=jw\ \varepsilon^*C_oV$.................(11)

Or

$I= wC_o\ (\varepsilon_i+jw\ \varepsilon_r\ )V$......................(12)

The conduction current component is:-

$I_c= wC_p\ \varepsilon_rV$......................(13)

And the displacement component is:-

$I_r= wC_p\ \varepsilon_iV$.......................(14)

From the obove equations the real and the imaginary parts of dielectric constant can be expressed in the following relations:-

The reciprocal of the resistance in the equivalent parallel circuit for a given frequency is called an alternative conductance (G_P) .If the specimen had effective area of (A) and thickness (d) the conductance can be expressed by the relation[14]:-

$G_P= \sigma$ a.c(w)A/d................(15)

From equation (12)

$\varepsilon_1 =\ \ Gp/ C_0\ w$......................(16)

And by substitution the value of (G_P)from equation (15)in equation (16) one can get:-

$\varepsilon_2=\ \ \sigma$ a.c/ $\varepsilon_0\ w$.......................(17)

In the past and recently considerable interest has been devoted to the gold doped germanium because of their potential application as infrared detector ,Also alloying Ge-Au with other elements provides the basis of infrared detectors over wide of range of wavelength [17,18,19] .

In this paper the real (ε_1)and imaginary part (ε_2) of dielectric constant for gold doped germanium films with various gold concentrations C_{Au} were measured ,the relation between them were discussed .An attempt had also made to interpret the results in terms of Maxwell- Wagner model. Cole-Cole diagrams were plotted and used to estimate the values of polaizability (α)or distribution parameter and the relaxation time(τ).

Experimental Part:-

The Ge –Au films of thickness 1 µm have been prepared by thermal evaporation of Ge from tungsten boat .The prepared films were doped with gold(Au) in different ratios (0,0.08,0.15, 0.2, 0.3) by thermal evaporation of Au from another tungsten boat at substrate temperature 553K .thermal diffusion of Au in the Ge films texture will be achieved by annealing the prepared samples at temperature 793K for three hours . To study measure the effect of gold concentration and frequency of the applied electric field on the real (ε_1)and imaginary part (ε_2) of dielectric constant of gold doped germanium films, the (LRC) meter (model HP-4274A) and (HP-4275A)are used which are able to measure several parameters like specimen capacitance ,dissipation factor ,resistance and phase angle in the frequency range (10^2-10^7Hz).

Results and Discussion:

A direct evidence of the existence of multi-relaxation time in the gold doped germanium films is obtaining by plotting Cole-Cole diagrams as shown in Figs(2A,B,C, D and E).It has been observed that for all films reported here (ε_1)versus(ε_2) curves represent the arc of circles having their centers lying below the absicca axis .This confirms the existence of distribution of (τ) in all films. By measuring the angles ($\alpha\pi/2$)the vales of the polarizability (α) had been determined and were listed in table(1).We can notice that the values of (α)systematically decreased with the increase in gold concentrations C_{Au} ,this in in agreement with the concept of molecular relaxation ,the decrease of (α) with the increase in C_{Au} results from rise of the forces of the intermolecular [20],while the increase of (α)value at C_{Au}=0.2 came from the weaken the forces as result of formation of barrier between gold and germanium atoms[21].

Another evidence of Debye-type relaxation is obtaining by plotting the curves(log ε_1)versus (log w) and (log ε_2)versus (log w) which were drown in Fig .(3 and 4).According to Deby's of intrinsic relaxation time (ε_1 and ε_2) can be written as in equations (6 and 7),these equations predict that these curves must be symmetrical around (w_D) ,where the angular frequency (w_D) corresponds to maximum absorption (according to the equation $w_D=1/\tau$) where (τ) represent the most probable value of a spread of relaxation times ,

The dielectric constant (ε_1) of gold doped germanium films with various concentrations namely (0,0.08,0.15,0.2 and 0.3) were measured within the employed frequency range(10^2-10^7Hz) ,from the spectrum of (log ε_1) versus (log w) it is obvious that (ε_1) tends to increase with increase gold concentration while it decreased with increase frequency to reach lower values this ascribed to the fact that electrode blocking layer is dominated thus the dielectric behavior is affected by the electrode polarization[22] while at high frequency the dielectric signal is not affected by electrode polarization ,also it can be noticed that the values of (ε_1) are affected greatly by the gold percentage ,moreover (at frequency =10^2 Hz) (ε_1) increased rapidly to three order of magnitude and to five order of magnitude of its value of pure Ge at C_{Au}=0.08 ,0.15 respectively, then at C_{Au}=0.2 (ε_1) decreases sharply to value lower than that of pure Ge followed by another increase to about five order of magnitude of its value of pure Ge at at C_{Au}=0.3 .The increase of(ε_1) with Au addition can be explained by symmetry changes which take place in the Ge lattice due to the replacement of Ge by Au atoms while the decrease in (ε_1) value may be attributed to the formation of a conductor or non linear capacitor with high energy barrier[21] . On the other hand in heterogeneous materials like gold doped germanium or multiphase materials the motion of charge carriers take place through one phase and the some charge carriers may trapped and accumulated at interfaces and defects as a results the electric field will be distorted and the dielectric constant increased. this effect depends on the conductivity of the present phases .this type of polarization called Maxwell-Wagner effect.

Fig.(5) showed the variation of (ε_2)with frequency with different C_{Au} one can be seen the dielectric loss absorption bands appear within the frequency range employed at different doping concentrations, indeed(ε_2) at the absorption bands increased from 94.74 to 215.1 to 24350 corresponding to $G_{Au=}$0,0.08,0.15 however (ε_2)decreased to 0.891 at $G_{Au=}$0.2 which attain to increase to 3911 at $G_{Au=}$0.3 ,the relaxation time were calculated as usual from frequencies corresponding to the loss maxima using equation($w_D=1/\tau$) and were listed in table (1).The observed energy maxima shifted to higher frequency at C_{Au} =0.08,0.15 and 0.3 this can be understood by considering the relaxation process which is in this case β-relaxation evidence from the particular molecular mechanism and local nature originating mainly from the motion of charge carriers ,also due structural defects[20].on the other hand It is clear that (τ) values for all C_{Au} namely (0,0.08,0.15 and 0.3)decrease with increase gold addition except C_{Au}=0.2,this result was estimated since the addition of gold rises the force of intermolecular ,while the increase of (τ) value attributes to the formation of barrier as mentioned before.

Conclusions:

Measurements of dielectric constants of gold doped germanium films with various concentrations had been conducted and several significant results have emerged from this experimental study:-

1- The values of (ε_1 and ε_2)affected strongly by the gold addition .

2-The values of (α and τ) are found to decreases with the increase of Au addition when the doping rises the force of the intermolecular while (α and τ) are found to increase when Au addition reduces the force of the intermolecular i.e. forms a barrier
with germanium atoms.

3-The big jump in the (ε_1 and ε_2) values explained in the light of Maxwell-Wagner model .

Table (1) Illustrates the values of (α and τ) for gold doped germanium films.

Doping concentration wt.%	α	τ (sec)
0	0.1887	$5.222x10^{-6}$
0.08	0.1660	$5.017x10^{-6}$
0.15	0.1449	$2.837x10^{-6}$
0.2	0.4166	$5.385x10^{-4}$
0.3	0.3888	$0.559x10^{-6}$

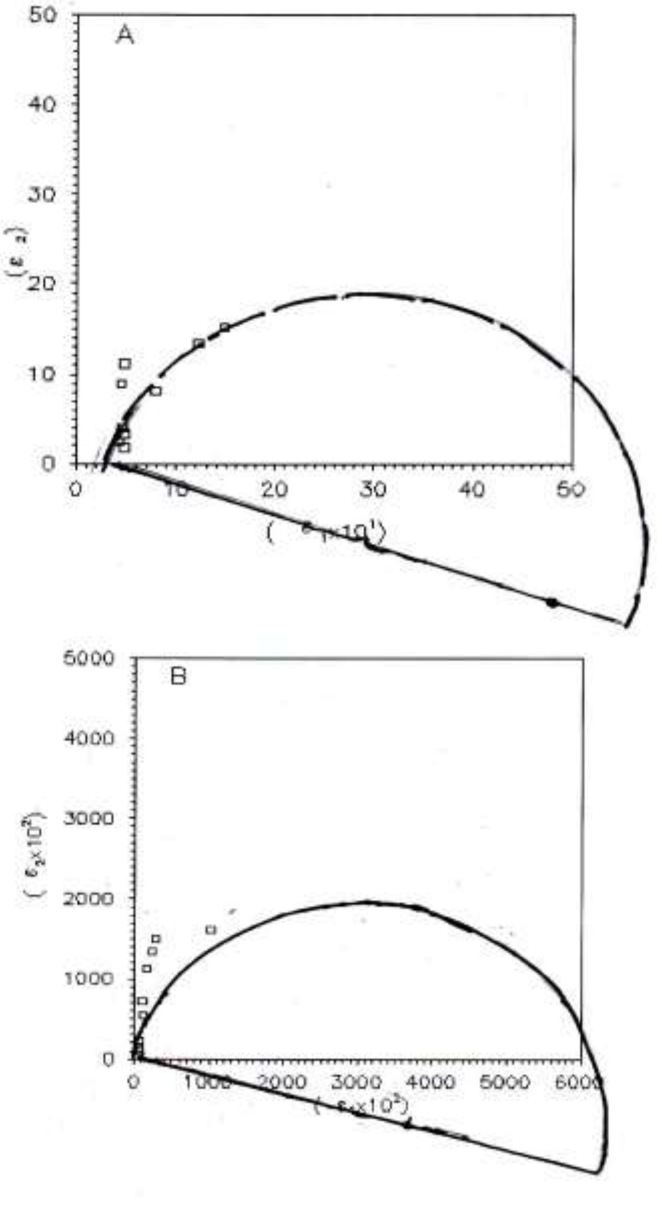
A
50
40
30
20
10
0
0
10
20
30
40
50
(ε 2)
(ε1×10¹)
B
5000
4000
3000
2000
1000
0
0
1000
2000
3000
4000
5000
6000
(ε2×10²)
(ε1×10²)

Fig.(1A,B)

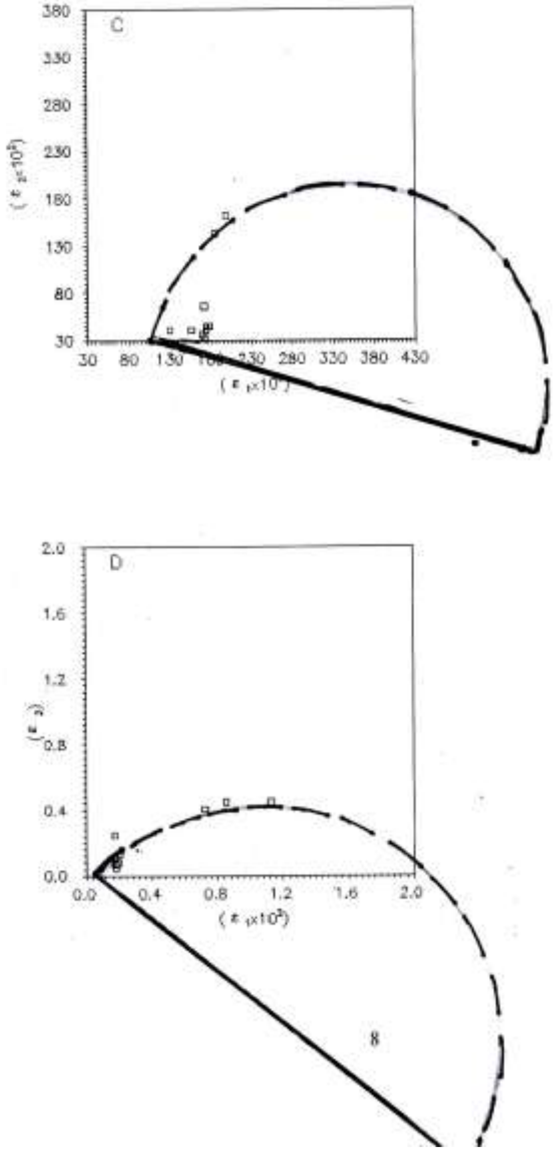
C
380
330
280
230
180
130
80
30
30 80 130 180 230 280 330 380 430
(ε2x10^2)
(ε1x10^2)
D
2.0
1.6
1.2
0.8
0.4
0.0
0.0 0.4 0.8 1.2 1.6 2.0
(ε2)
(ε1x10^2)
8

Fig.(1C,D)

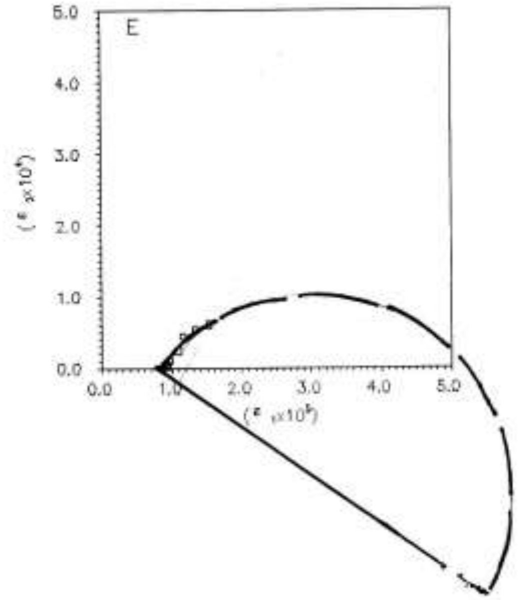

Fig.(1)Cole –Cole digrams for gold doped germanium films with different gold concentrations. (A)0, (B)0.08%, (C)0.15% ,(D)0.2% and (E)0.3%.

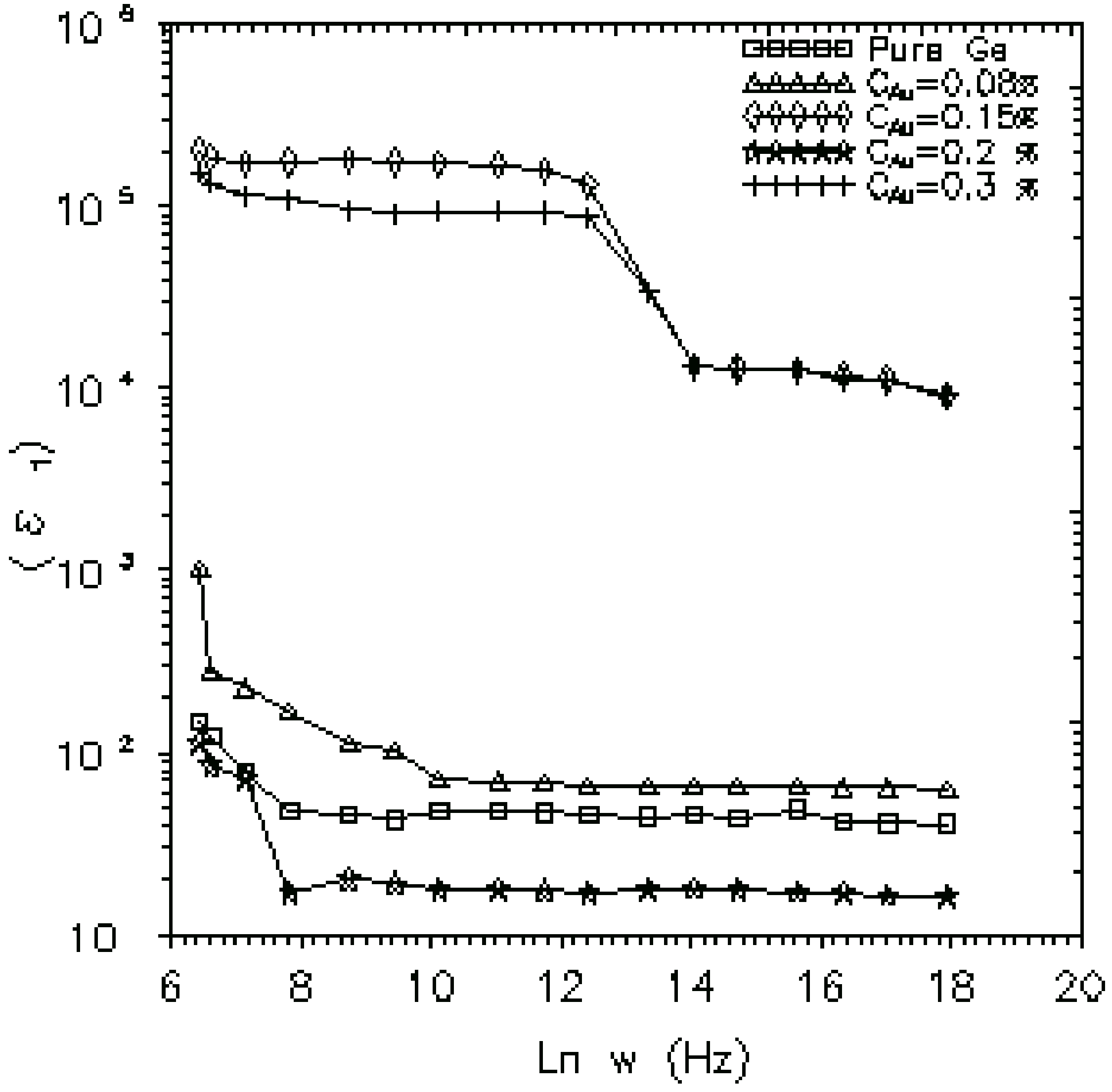

Fig. (3) The variation of log (ε_1) with log (w) for gold doped germanium films.

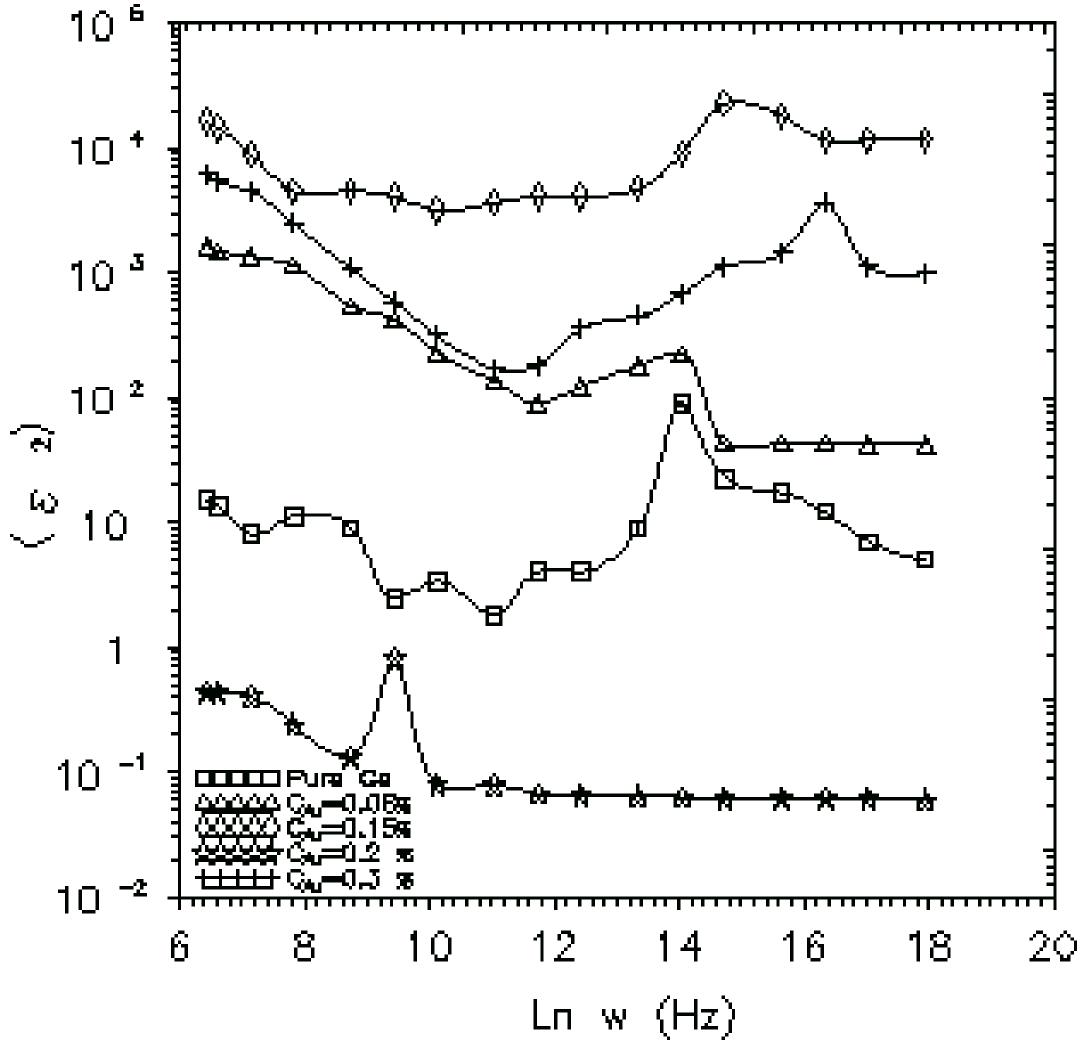

Fig. (4) The variation of log (ε_2) with log (w) for gold doped germanium films.

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