Observation compensation effect and crossover between Percolation and variable-range-hopping regimes for dc conductivity in germanium irradiated with large fluencies fast neutrons

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The dependence of dc resistivity on temperature in the range 1.7 k-300 k, of the structural disordered by irradiation fast neutrons with fluencies 10^{16} cm^{-2} $\leq \varphi \leq 1.2\times10^{17}$cm^{-2} is measured. From the analysis of dependence of dc resistivity on temperature the Meyer – Neldel Rule (MNR) is found to be applicable in these samples. The E_{MN} is found to be greater than 0 for the intermediate and hopping mechanisms of conduction. But for the band to band mechanism of conduction E_{MN} is less than 0. A correlation between ρ_{oo} and $(E_{MN})^{-1}$ is detected. The crossover between Efros-Shklovskii (ES) percolation mechanism of electrical conductivity and Mott variable range hopping (VRH) regime of conductivity is noticed and investigated. Both approaches, the percolation criterion of equivalent random resistor network [20] approach and the phenomenological energy additivity approach [33], are suitable to explain the crossover between the above mentioned two regimes of conductivity. Also coulomb gap (Δ) in the density of states (Dos) in vicinity of Fermi level is calculated. The coulomb gap is found to be inversely proportional with the irradiation fluency.

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1. Introduction

In the study of disorder materials, various properties, e.g., conductivity and diffusion are found to exhibit exponential thermally activated (Arrhenius) behavior [1]:

$$Z = Z_o \exp\left(\frac{-\Delta E}{KT}\right). \qquad (1)$$

Here Z is the absolute rate of a thermally activated process, Z_0 the preexponential factor, ΔE the activation energy and k the Boltzmann constant. Meyer and Neldel observed that Z_0 is related to ΔE by the following relation:

$$Z_o = Z_{oo} \exp\left(\frac{\Delta E}{E_{MN}}\right), \qquad (2)$$

Where Z_{00} and E_{MN} are positive constants. E_{MN} is known as the Meyer- Neldel energy for the process in question. This empirical relation is known as the MN relation or the compensation effect. The compensation effect has been observed in a wide variety of materials for a number of phenomena [2]. In the class of amorphous semiconductors, the MN rule has been reported in a Si:H films in which ΔE is varied by doping, by surface absorption, light soaking or by preparing films under different conditions [3-5].

Germanium irradiated with large fluency of fast neutrons can serve as a convenient object for experimental studies of hopping resistivity and nonmetal-metal transition [6]. The electric properties of neutron irradiated crystalline germanium are determined by disordered regions near an n- p- conductivity inversion, and by acceptor-like radiation defects with shallow energy levels near the valence band far from an n- p-conductivity inversion [7]. Transport of charge carriers in neutron irradiated germanium has been studied over wide range of temperature 1.6K<T<300K and for concentrations of acceptor-like radiation defects, $6\times10^{15} < N_{RD} < 2\times10^{16}$cm^{-3} [8]. It was found that the activation energy of hopping conduction at shallow levels of acceptor- like radiation defects is equal to 16 meV in the case of low concentration and compensation.

Variable-range-hopping (VRH) of conduction has recently received renewed significant attention due to considerable theoretical and experimental advances [9]. The variable-range-hopping (VRH) resistivity of three-dimensional disordered systems was shown by Mott [10, 11] to behave as ln $\rho\alpha(T_M/T)^{1/4}$. Efros and Shklovskii (ES) [12] argued that the Coulomb interactions create a gap that leads to ln $\rho\alpha(T_{ES}/T)^{1/2}$ at low temperatures. The crossover from Efros-Shklovskii (ES, percolation) behavior $\rho\ \alpha\ T^{-1/2}$, to Mott law (VRH) $\rho\ \alpha\ T^{-1/4}$ of the three dimensional (3D) VRH, resistivity with increasing temperature has been observed in different materials [14]. Popescu et. al. [15] attempted to relate the characteristics of the density of states (DOS) at the Fermi level to the Meyer-Neldel relation. The aims of the present work are to investigate the applicability of Meyer-Neldel empirical formula [1] on various regions of the temperature dependence of electrical resistivity [15] of the given samples. And investigate the crossover low temperature conductivity from Efros Shklovskii percolation model to variable range hopping mechanism argued by Mott.

2. Theoretical basis

2.1 Meyer –Neldel rule

The temperature dependence of dc conductivity (σ) of a semiconducting material generally follows the empirical Arrhenius type equation [1] of the form

$$\sigma(T) = \sigma_0 \exp. (-\Delta E/kT) \tag{3}$$

If ln (σ) is plotted on the ordinate against 1/T on the abscissa, a straight line is obtained over a certain temperature range, the slope of the line determines the activation energy ΔE. The point intersection of this line with the ordinate yields the value of the pre-exponential factor σ_0. For many classes of materials [16], experimental evidence suggests that a correlation exist between the activation energies and pre-exponential factors of the form

$$\mathrm{Ln}\ (\sigma_0) = b\Delta E + \ln \sigma_{00} \tag{4}$$

Where $b = (kT_o)^{-1}$ and σ_{00} are constants, this relation can be written also as:

$$\sigma_0 = \sigma_{00} \exp.(\Delta E/kT_0) \tag{5}$$

Where, the parameter $E_{MN} = kT_0$ is often called the Meyer-Neldel characteristic energy. The relation (5) gives the dependence of the pre-exponential factor σ_0 on the activation energy ΔE and represents the Meyer-Neldel rule [17]. This rule is valid in disorder materials even when ΔE is varied by doping.

2.2 The crossover from Efros Shklovskii (percolation) model to Mott (VRH) model

The temperature dependence of DC resistivity of disorder or amorphous materials has three mechanisms of conduction which we may expect to find in appropriate ranges of temperature. The three regions of resistivity are apparent, satisfying the thermally activated resistivity relation [15]:

$$\rho = \rho_o \exp.(E_C-E_F)/KT + \rho_1 \exp.(E_A-E_F+W_1)/KT + \rho_2 \exp. (W)/KT \tag{6}$$

Where the term ($\rho_o \exp.(E_C-E_F)/KT$) represents transport by carriers excited beyond the mobility edges into non-localized (extended) states at E_C or E_V. And ρ is the resistivity, ρ_o is the pre-exponential factor. A plot of lnρ versus 1/T will yield a straight line if E_C-E_F is a linear function of T over the temperature range measured. As the temperature decreases transport carriers excited into localized states at the band edges and the resistivity is given by $\rho_1 \exp.(E_A-E_F+W_1)/KT$, where W_1 is the activation energy for carriers hopping, W_1 should decrease with decreasing temperature on the account of variable - range nature of the hopping transport. However, as the temperature dependence is through the carrier activation term, an approximately linear dependence of lnρ versus 1/T is again expected. As the temperature lowered more, there will be a contribution from carriers with energies near E_F which can hop between localized states. This contribution is described by $\rho_2 \exp. (W)/KT$, where $\rho_1 \leq \rho_2$ and W is the hopping energy, of the order of half the width of the band of the states. At temperatures such that kT is less than the band width, hopping will not be between nearest neighbors and variable-range hopping of the form $\rho = \rho_2 \exp. (B/T^{1/4})$ with $B=2\{\alpha^3/k\ N(E_F)\}^{1/4}$, where α is a constant, k is the Boltzmann's constant and $N(E_F)$ is the density of states at Fermi level. Electronic interactions are known to play an important role in the strongly localized regime. Pollak [18], and Efros and Shklovskii [19] pointed out that the long-range nature of the interactions leads to a dip in the single particle density of states N (ε) at the Fermi level. Efros and Shklovskii argued this soft gap is of the form $N(\varepsilon) \sim \varepsilon^{d-1}$ where ε is the energy measured from the Fermi energy and d is the space dimension.

A study of the corresponding details of the mechanism of hopping conductivity obviously requires a comparison of the experimental data, in particular, on the temperature dependence of resistance, with the results of theoretical calculations over a wide range of temperatures. For materials with typical parameters (intermediate impurity concentration and intermediate compensation) in the temperature range that typical for the experiments (~0.01-10 K). A crossover is observed from conductivity with variable - range hopping (VRH) of Mott Type [$\rho(T)\ \alpha\ \exp.(T_M/T)^{1/4}$] to conductivity over states in the Coulomb gap - the Efros – Shklovskii low [$\rho(T)\ \alpha\ \exp.(T_{ES}/T)^{1/2}$] [18,19]. The nature of the conductivity in the two indicated regimes differs both in its numerical parameters and, possibly, in its physical nature [20]. The crossover region turns out to be quit wide; therefore, a comparison of the predictions of the theory with experiment is quite complicated. Thus, in order to be able to compare the experimental values of the parameters T_M and T_{ES} with their calculated values,

$$T_M = \beta_0/N(E_F)a^3 \tag{7}$$

$$T_{ES} = \beta_1 e^2/\kappa\ a \tag{8}$$

Where β_0 =21 and β_1 =2.8 are numerical coefficients, a is a localization radius and κ is the dielectric constant, It is important to have a valid description of the crossover region.

3. Experimental

Five samples of non-doped germanium with carrier density $n_0 \approx 3x10^{13}$ cm^{-3} were irradiated with fast neutrons with energies of E ≥ 0.1 MeV in the range of $6x10^{16}$ cm^{-2} ≤ Φ ≤ $1.2x10^{17}$ cm^{-2}. As a result of irradiation, all the original samples become disordered p-type [8]. In order to reduce the transmutation doping effect all samples were placed in 1mm thick cadmium containers. During irradiation in the reactor the ratio between thermal neutrons fluency and the fluency of fast neutrons was about ten. So, it was possible to obtain samples of germanium "doped" with acceptor-like radiation defects {Ge (RD)}. To ensure that the electrical properties were controlled by the transmutation doping, a complete annealing at 450 °C for 24 h was performed. For electrical conductivity measurements a special double wall glassy cryostat is designed. This cryostat is attached with vacuum pump its evacuation rate is faster than the evaporation of He^4 gas, thus the pressure inside the cryostat is decreases and hence the temperature. The conventional four probe method is used for electrical resistivity measurements. Ni electrode is participated in the desired position on the samples using electrochemical deposition technique (cold method). Thin Cu wires are fixed above the Ni electrodes using In. The samples were in parallelepiped shape with length about 8-12mm, thickness about 1 - 2 mm, width about 2-3 mm and the electrode apart about 3-4 mm. Resistivity of Ge (RD) was measured in the temperature range from 1.7 K up to 300 K. The temperature was determined with a semiconductor thermistor in the interval 77.4 - 4.2K, from saturated vapor pressure of He^4 in the interval 4.2-1.5 K. The voltage across samples always less than 1 volt and the current across the sample decreases from μA to nA order as the temperature lowered. The electrical properties of Ge (RD) are determined solely by acceptor-like radiation defects [21]. The least square method fitting, using a computer program Excel 2010, is used to analysis the data extracted and to calculate the uncertainty in the experimental results.

4. Results and discussion

4.1 Meyer – Neldel rule (MNR)

Fig. 1.a shows ln (ρ_0) = f (ΔE) for the three mechanisms of conductivity of the five samples as obtained from ln (ρ) =f (1/T) of Fig. 2. The slope of the straight lines is $(E_{MN})^{-1}$ and ρ_o is the intercepts. The lines of dependence of ln ρ = f (ΔE) of conductivity of second mechanism of conductivity (in the temperature rang 4.2 K ≤T≤ 14 K) and third mechanism of conductivity (in the temperature range 1.88K≤T 5.5K) have positive slopes. The E_{MN} for the hopping range of conductivity is 0.065meV and ρ_o for this range of conductivity is equal to $4.3x10^{-5}$ Ω.cm. And for the intermediated range of conductivity the E_{MN} is equal to 0.31meV and preexponential factor ρ_0 is equal to 0.35Ω. cm. But the slope of the first mechanism of conductivity (temperature range, 12K ≤ T ≤ 100K) has a negative one. The negative slope in the ln (ρ) = f (ΔE) of the first mechanism of conduction,(temperature range, 12K ≤ T ≤ 100K), means the existence of reverse MN rule in this mechanism of conduction since ΔE<0. Many authors observed E_{MN} <0 and referred to this as anti or reverse MNR [22, 23]. For the band to band mechanism of conduction in the temperature range 12K ≤ T ≤ 100K, the slope of $\ln\rho_o$ Vs. ΔE is small and negative, the value of E_{MN} is equal to -2.35 meV and ρ_o is equals 1.65 Ω.cm. Abtew et al [24] reported inverted MN rule in hydrogenated amorphous silicon. High temperature causes the optical gap to shrink for both crystalline and amorphous material. Abtew [24] reported that as at high temperatures Materials have an exceedingly weak MNR or don't exhibit the MNR at all [25]. The slopes and the intercepts of three above mentioned lines are given in figure (1.b). The three points extracted from that lines of the three stages of conductivity have a straight line behavior with fitting equation of the form

$$\ln\rho_{00} = 1.2737 - 0.0007(E_{MN})^{-1} \quad (9)$$

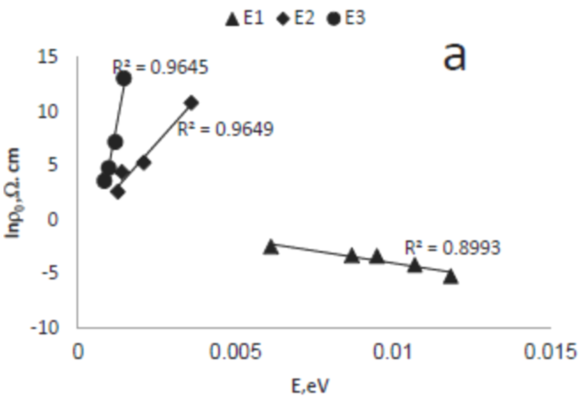

Fig. 1.a Shows ln (ρ_o) = f(ΔE) for the three mechanisms of electrical conductivity which are satisfies equation No. 6.

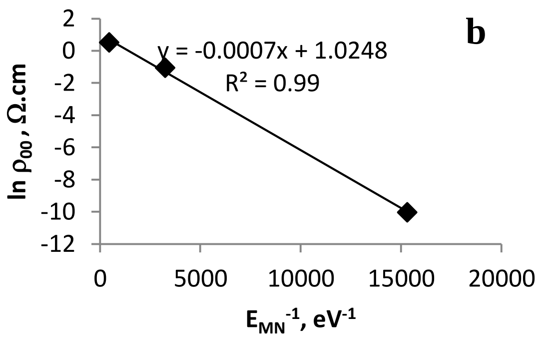

Fig. 1.b. Shows ln (ρ_{oo}) = f (1/E_{MN}) as obtained from the least square method applied on the data represented in Fig. (2).

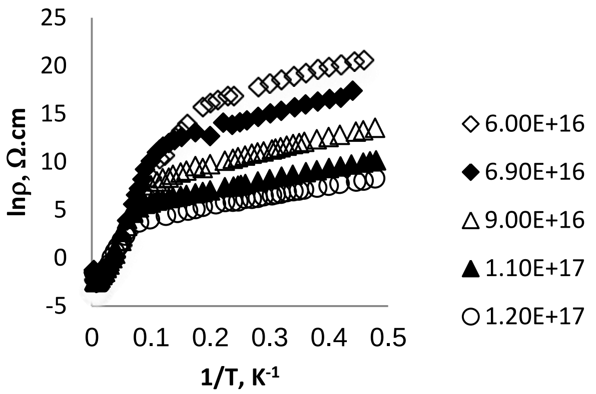

Fig. 2. Shows ln (ρ) = (T^{-1}) for different irradiated samples.

Also the correlation between ρ_{00} and E_{MN} were observed by Wang and Chen [26] for C_{60} at different stages of growth process and at different gate voltages of the field effect transistor. It has also been reported in a-Si: H produced by different technique [27]. Recently, Mehta Kumar and co-workers reported the "further MNR" in various Chalcogenide glasses paying heed to thermally activated photoconduction, high field conduction, and non-isothermal crystallization [28]. Several models have been proposed to explain the MN rule [29]. Busch [30] suggested that the MN rule in extrinsic broad band semiconductors is due to freezing of the donor concentration during cooling after the preparation. The MN rule in amorphous or polycrystalline semiconductors may derive from an exponential tailing of majority band states, or may be due to long- ranged electrostatic random potential. The discussion of whether there is one universal explanation for MNR in different systems is not yet settled [31].

4.2 The crossover from Efros Shklovskii model to Mott model

Fig. 2 shows ln (ρ) as a function of 1/T, where T is the temperature, for the five irradiated samples, the three regions of resistivity are apparent, satisfying the thermally activated resistivity relation (6). At high temperature range, 12K $\leq$ T $\leq$ 100K the first term of equation (6), which represents transport by carriers, excited beyond the mobility edges into non-localized (extended) states at E_V. Fig. 3 shows the dependence of electrical resistivity on negative square root of temperature as argued by the percolation theory of Efros-Shklovskii [19, 32]. Fig. 3. a shows the dependence of electrical resistivity on $T^{-0.25}$, from which the Mott characteristic temperature T_M is determined. Also Fig. 3.b shows the dependence of electrical resistivity on $T^{-0.5}$, from which the Efros Shklovskii characteristic temperature T_{ES} is obtained. Table 1 indicates the values of electrical conductivity activation energies E_i, i =1, 2, 3, of different conductivity mechanisms, T_{ES} and T_M as related to irradiation fluency φ.

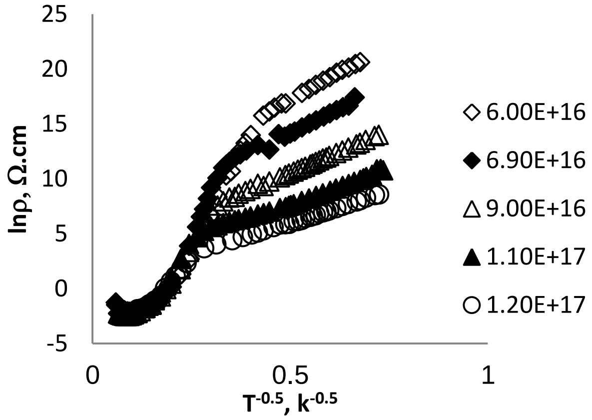

Fig. 3. Shows ln (ρ) = ($T^{-0.5}$).

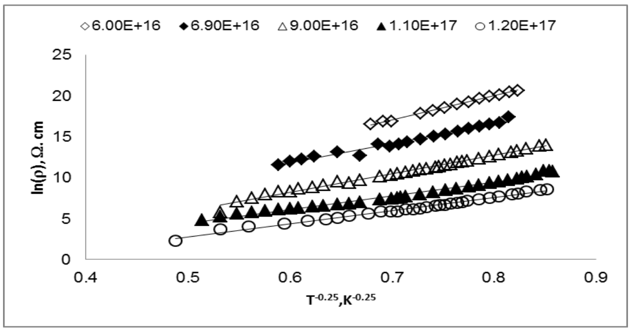

Fig. 3.a. shows ln (ρ) = ($T^{-0.25}$) Mott variable range hopping mechanism of conduction.

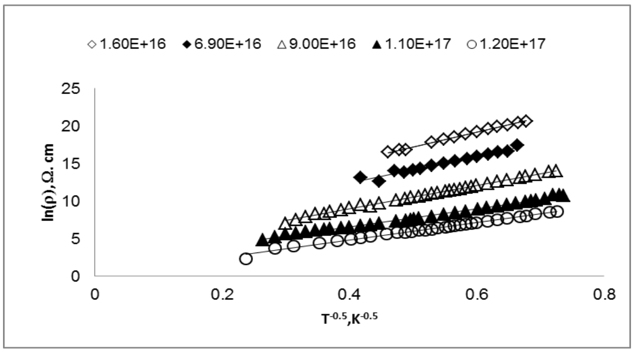

Fig. 3.b Shows ln (ρ) = ($T^{-0.5}$) Efros-Shklovskii Percolation mechanism of conduction.

Table 1. Values of electrical conductivity activation energies E_1, E_2, E_3, Efros Shklovskii characteristic temperatures T_{ES} and Mott characteristic temperature T_M as obtained from the data represented in Fig's 2 and 3 for different fluency of irradiation.

Sample No.	φ, n.cm^{-2}	E_1,eV	E_2,eV	E_3 ,eV	T_{ES}, k	T_M, k
1	6.00E+16	0.011±0.01	0.002±0.0003	0.001±0.0005	388±2	7.70E+05
2	6.90E+16	0.012±0.01	0.006±0.0003	0.0015±0.0005	321±2	3.28E+05
3	9.00E+16	0.012±0.01	0.004±0.0003	0.001±0.0005	243±2	2.58E+05
4	1.10E+17	0.007±0.01	0.0014±0.0003	0.0006±0.0005	146±2	7.95E+04
5	1.20E+17	0.009±0.01	0.0012±0.0003	0.0008±0.0005	135±2	6.56E+04

Pollak [18], and, Efros and Shklovskii [19, 32], have predicted that long –range electron electron interaction reduces the density of states (DOS) at Fermi level (FL). Forming the so called Coulomb gap (CG), for which g (ε) = $g_0\ \varepsilon^2$, where $g_0 = (3/\pi)\ (e^2/\kappa)^{-3}$, the energy $\varepsilon = E_F - E$ is measured from the Fermi level E_F, and κ is the dielectric constant, where $T_{ES} = 2.8e^2 /(\kappa\ a)$. The half widthΔ, of the CG can be roughly determined by,

$$\Delta \sim [(T_{ES})^3/T_M]^{1/2} \qquad (10)$$

Fig. 4 shows the dependence of Δ on the irradiation fluency, the coulomb's gab decreases with increasing irradiation fluency, it has been reported that [33], Δ collapses at the critical concentration N_c , at which insulator metal transition occurs. Also Shlimak et. al. [34] proved that the CG smears by temperature. The smearing of CG either by temperature or by increasing charge carriers concentration lead to crossover mechanism of conduction from percolation mechanism (ES) to Mott variable range hopping mechanism. Aharony et al. [32] gave the first attempt to describe the temperature behavior of the conductivity in the crossover region with the aid of a universal expression:

$$\mathrm{Ln}\ (\rho/\rho_0) = A\ f\ (T/T_x) \qquad (11)$$

Where ρ_0, A, T_x are material dependent scaling factors and f(x) is a universal function, given by

$$f(x) = \frac{1 + [(1+x)^{1/2} - 1]/x}{[(1+x)^{1/2} - 1]^{1/2}} \qquad (12)$$

Under these conditions, in the asymptotic resistivity limits [35], the following relations applied in the present samples

$$T_M = =A^4 T_x, \qquad (13)$$

$$\text{and} \qquad T_{SE} = 4.5A^4 T_x \qquad (14)$$

Using these relations, together with the obtained experimentally T_M &$T_{ES,}$ to determine the values of T_x and A. From the universal scaling equations No.11, No. 12 and the relations No.13, No.14 Fig. (5) shows the normalized conductivity ($\ln\rho/\rho_0$) vs. the normalized temperature (T / T_x). The data for all samples collapse via normalization factor (N. F.) on the universal curve which corresponds to f(x), (relation 12), as seen in Table 2.

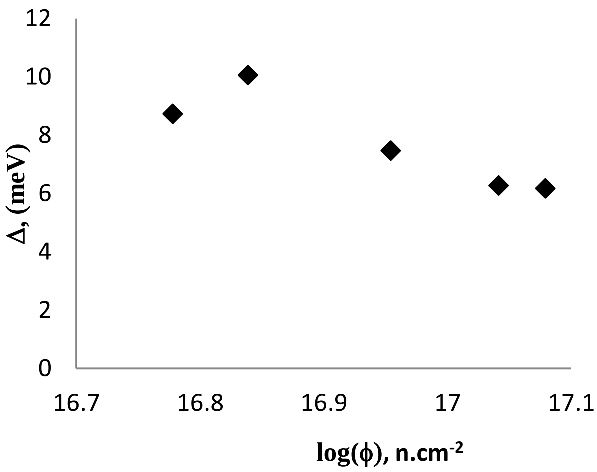

Fig. 4. Shows the dependence of coulomb gap on the irradiation fluency Δ = f ($\log_{10}$ φ).

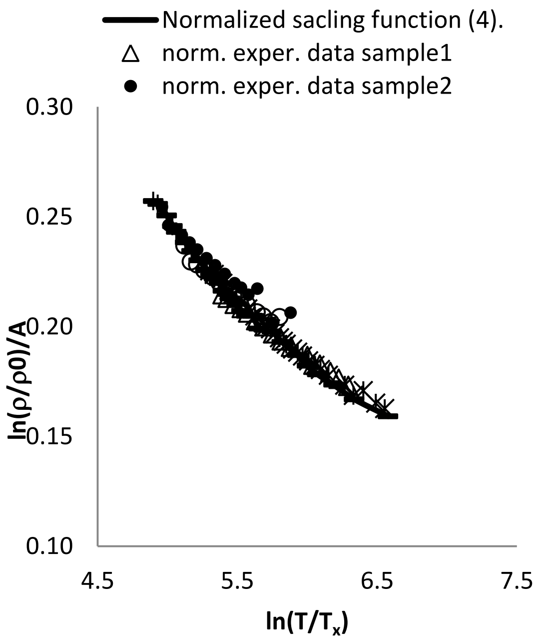

Fig. 5. Shows the fitting of experimental data which is consistent with the normalized equation No.11.

Table 2. Shows the values of A, T_x, and N.F.(normalization factor) of different samples.

Sample No.	A	T_x	N. F
1	93.9±0.1	.010±0.02	0.80±0.01
2	76.4±0.1	0.016±0.02	0.75±0.01
3	72.3±0.1	0.010±0.02	0.88±0.01
4	49.2±0.1	0.010±0.02	0.48±0.01
5	46.4±0.1	0.014±0.02	1±0.01

On the basis of the percolation criterion of equivalent random resistor network [36] consisting of randomly placed sites, Agrinskaya et. al. [20] proposes approach to describe the crossover from percolation mechanism (ES) of electrical conduction to the variable range hopping mechanism (Mott). The theoretically derived equation is

$$T\xi - 0.6\frac{T_M^{3/2}}{T_{ES}^{1/2}}\arctan\frac{\xi T T_M^{1/2}}{0.6T_{ES}^{3/2}}\xi^3 = T_M \qquad (15)$$

Where ξ is the critical value of the argument of the of the exponential in the temperature dependence of the hopping resistance ln R (T)= B ξ (T) where B is a coefficient. A computer program Mathematica 5 is used for solving equation no. (15). Fig. 6 shows the comparison of the ξ with the experimental extracted data of, ln (ρ*$T^{-0.5}$) = f($T^{-0.5}$), dashed line for sample no.5 and continuous one for sample no.3. The figure shows consistence of the obtained experimental data with the values of ξ obtained from equation no. (15), via value of the coefficient B equal 2/3 for sample no. 3 and 1/2 for sample no.5. Thus, both approaches, Aharony et. al. [18] approach which argued the crossover is described by phenomenological energy additivity. And the approach of Agrinskaya et al [20] which depend on the percolation criterion of equivalent random resistor network, are suitable in low temperature to explain the crossover from percolation mechanism of conduction to the variable range hopping conduction in the temperature range lower than 10K.

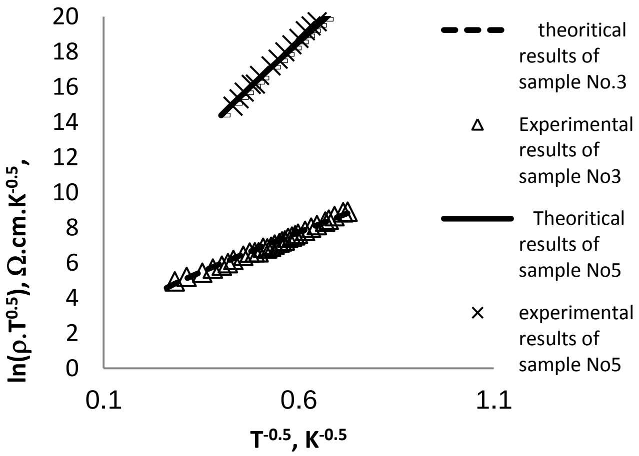

Fig. 6. Shows the fitting of experimental results of samples no. 3 (dashed line) and sample no.5 (continuous line) with the normalized equation No. 15.

5. Conclusions

The dc conductivity is measured in the temperature range (1.7k - 300k). The overall results show that the Meyer Neldel Rule is applicable in the three mechanisms of dc conductivity of germanium samples disordered with large fluencies reactor neutrons from 6×10^{16} cm $^{-2}$ up to 1.2×10^{17}cm^{-2} . Also the slopes and intercepts of the three straight lines satisfies MNR are correlated this indicates the existence of further MNR in germanium irradiated fast neutrons. Crossover between Percolation regime of conductivity and variable range hopping is observed. Both approaches of phenomenological energy additivity approach and the percolation criterion of equivalent random resistor network approach suitable to explain the crossover between the above mentioned mechanisms of conductivity. The Coulomb gap in the density of states in vicinity of Fermi level is calculated. The width of Coulomb gap is found to decrease with increasing radiation fluency.

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