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TEMPERATURE DEPENDENCE BEHAVIOR OF ELECTRICAL RESISTIVITY IN NOBLE METALS AT LOW TEMPERATURES

Muhammad A. AL-Jalali
Physics Department, Faculty of Science, Taif University, Taif, AL-Haweiah, P. O. Box 888, Zip code 21974, Kingdom of Saudi Arabia
* Corresponding author; E-mail address: aljalaliphys@gmail.com

ABSTRACT

Resistivity temperature – dependence and residual resistivity concentration-dependence in pure noble metals(Cu, Ag, Au) have been studied at low temperatures. Dominations of electron – dislocation and impurity, electron-electron, and electron-phonon scattering were analyzed, contribution of these mechanisms to resistivity were discussed, taking into consideration existing theoretical models and available experimental data, where some new results and ideas were investigated.

Indexing terms/Keywords

Resistivity; electron –impurity scattering; electron-electron scattering; electron-phonon scattering; temperature dependence.

Academic Discipline And Sub-Disciplines

Theoretical Physics(solid state), Spin Glass Theory

SUBJECT CLASSIFICATION

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TYPE (METHOD/APPROACH)

Full research article – Theoritical along with experimental analysis

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1. Introduction

Portable devices on satellites greatly affected by fluctuations in temperature in outer space, especially the electrical resistance that it needs to adjust with very high accuracy to receive high quality information on the earth stations.

Since 1864, Matthiessen's rule of the total resistivity of metals [1, 2, 3] was discovered by Augustus Matthiessen as an empirical rule.The total resistivity is the sum of many mechanisms results from the scattering of conduction electrons by thermal lattice vibrations and imperfections such as impurity atoms, interstitials, dislocations, and grain boundaries. Noble metals resistivity have been developed in a very large, across very advanced technologies to estimate the purity of these elements and their alloys [4, 5, 6, 7].Matthiessen's rule is a basis for understanding the resistivity behavior of metals and alloys at high and low temperatures.

In general, the electrical resistivity (ρ (T)) of pure noble metals [8,9,10,11] consists of the residual resistivity (ρ_0) (originating mainly from imperfections within the structure of the material) and the temperature-dependent resistivity ($\rho_L(T)$) arising due to electron–phonon (lattice) interactions, which may be written by an empirical form :

$$\rho(\mathrm{T,c}) = \rho_0 + \rho_L(T) \qquad (1)$$

A single crystal of noble metals would have a lower residual resistivity than polycrystalline that cause an increase in the scattering of electrons, and reducing the mean free time between collisions. The amplitude of the second term increases with increasing temperature, and the more they interfere with conduction .

In magnetic noble metals alloys [12], the dynamics of the spin system are usually such that the spins remain in thermal equilibrium down to very low temperatures. Magnetic impurities play a double contribution, the first in residual resistivity as imperfections, the second as temperature – dependent disorder magnetic resistivity, in which case it is more usual to express total resistivity as:

$$\rho(\mathrm{T,c}) = \rho_0 + \rho_L(T) + \rho_m(T) \qquad (2)$$

Where ρm(T)is the temperature-dependent magnetic contribution resulting from spin-disorder.

Matthiessen's rule is an approximation and is not universally valid ,because not only residual resistivity and resistivity of the pure ideal metal(electron-phonon scattering contribution) but also many mechanisms contribute to resistivity which cause a deviation from Matthiessen's rule(DMR)[13,14,15,16],for this reason DMR may be added as this form:

$$\rho(\mathrm{T,c}) = \rho_0 + \rho_L(T) + \Delta\rho(T) \qquad (3)$$

Where Δρ (T) represents all contributions to resistivity temperature and concentration dependent, which come from DMR.

The purity of noble metals could be calculated from residual resistivity ratio (RRR)[17,18] Where the RRR is defined as the ratio of the electrical resistivity at two temperatures: 273K (the ice point) and 4.2 K (the liquid helium normal boiling point) and may be written as(RRR =ρ(273K)/ρ_{extrap}(4.2K). Sometimes, the room temperature resistivity ρ(T=300 K) is used instead of the resistivity at ice point ρ(T=273K). RRR values serve as a convenient measure of the purity of the metal and is often used as a material specification for superconductors.

In addition, the Debye temperature θ_D (the temperature of a crystal's highest normal mode of vibration), Size dependence of θ_R and size effects, generally cause a DMR because the phonon spectrum changes by physical defects [19,20,21].

The aim of this paper is to comparison between predictions of theoretical models and the experimental data and trying to check an excellent agreement with each other.

2. Theoretical Quantum Mechanics Background

In general, the resistivity of pure noble metals [22] is composed of the residual resistivity, resistivity due to electron-phonon interactions and the resistivity due to electron-electron scattering. The total resistivity ρ can be written as:

$$\rho = \frac{m}{ne^2\tau} = \rho_{electron-\mathrm{impurity}} + \rho_{electron-\mathrm{defects}} + \rho_{electron-\mathrm{phonon}} + \rho_{eectron-electron} + \ldots\ldots \qquad (4)$$

Where n is the free electron density, e the electron charge and m its mass, scattering time τ is the main problem in resistivity because more than one source of scattering is present; Matthiessen's rule can also be stated in terms of the scattering time:

$$\frac{1}{\tau} = \frac{1}{\tau_{\mathrm{mpurity}}} + \frac{1}{\tau_{\mathrm{defects}}} + \frac{1}{\tau_{\mathrm{phonon}}} + \ldots\ldots \qquad (5)$$

Where τ the true average scattering time and $\tau_{\mathrm{impurities}}$, τ_{defects} are the scattering time by impurities, lattice faults, the surface, or other defects is expected to be temperature-independent , that depends on how the sample was made. This scattering rate can vary greatly between samples of the same material made in different labs or in different

ways. Unfortunately, all these mechanisms give temperature independent contributions [23], Residual resistivity of noble metals may be written as:

$$\rho_{\text{residual resistivity}} = \rho_{electron-\text{impurity}} + \rho_{electron-\text{defects}} \qquad (6)$$

Where defects: dislocation, vacancies, Frenkel and Schottky defect, chemical impurity… etc..

Scattering by lattice vibrations (phonons) is temperature dependent and the rate $1/\tau_{\text{phonon}}$ depends on the intrinsic at high temperatures (above the Debye temperature) the number of phonons per unit volume is proportional to the temperature and ρph is proportional to T. At low temperatures, two effects come into play. The number of phonons falls as T^3 because of phonon drag and U-processes; moreover, the energy and momentum of these phonons become small, and phonon drag contribution sometimes neglected because noble metals have spherical Fermi surfaces so that they are ineffective in scattering electrons. Consequently, the resistivity falls more quickly than the phonon density as T^5 because of N-processes.

In simple metals such as noble metals (bulk), the temperature-dependent part $\rho_{electron-\text{phonon}}(T)$ is well described by the following Bloch– Gruneisen (BG) function (n=5) or Bloch–Wilson (BW) formula (n=3) [24]:

$$\rho_{electron-\text{phonon}}(T) = \frac{C}{M\,\theta_R}\left(\frac{T}{\theta_R}\right)^5 \int_0^{\frac{\theta}{T}} \frac{z^5}{\left(e^z-1\right)\left(1-e^{-z}\right)}dz = \frac{C}{M\,\theta_R}\left(\frac{T}{\theta_R}\right)^5 J_5\left(\frac{T}{\theta_R}\right)$$

$$\rho^{N-process}_{electron-\text{phonon}}(T) = 4A\left(\frac{T}{\theta_R}\right)^5 J_5\left(\frac{T}{\theta_R}\right)$$

$$\rho^{U-process}_{electron-\text{phonon}}(T) = BT^3 J_3\left(\frac{T}{\theta_R}\right)$$

$$where \qquad J_n\left(\frac{T}{\theta_R}\right) = \int_0^{\frac{\theta}{T}} \frac{z^n}{\left(e^z-1\right)\left(1-e^{-z}\right)}dz = \int_0^{\frac{\theta}{T}} \frac{z^n e^z}{\left(e^z-1\right)^2}dz \qquad (7)$$

Where A is a constant, proportional to the square of the electron-lattice interaction constant C (constant of the metal), θ_R is the Debye temperature obtained from resistivity measurements, $J_n\left(\frac{T}{\theta_R}\right)$ a transport integral [25, 26, 27].

At high and low temperature relation (7) has exact solution yielding the following proportionalities:

$$\rho_{electron-\text{phonon}}(T) \to 142.431\frac{C}{M\,\theta_R}\left(\frac{T}{\theta_R}\right)^5 \quad as \quad T \ll \theta_R$$

$$\rho_{electron-\text{phonon}}(T) \to \frac{C}{4M\,\theta_R}\left(\frac{T}{\theta_R}\right) \quad as \quad T > \theta_R \qquad (8)$$

At the lowest temperatures $\rho_{electron-\text{phonon}}(T)$ goes to zero and the overall resistivity reduces to the residual resistivity.

At low temperature, the normal electron- electron scattering does not contribute to the electrical resistivity because in such collisions, the charge and the momentum are conserved. Whereas, Umklapp processes most of momentum impart to the lattice as a whole and they can thus contribute to the electrical resistivity. In addition, Umklapp processes contribution is greatly reduced due to the operation of the Pauli's exclusion principle between initial and final states. The expression for resistivity $\rho_{ee}(T)$ due to electron- electron Umklapp scattering processes can be written as [28,29,30,31,32,33]:

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$$\rho_{e-e} = \left(\frac{\pi^3 z}{16}\right)\left(e^2/\upsilon_F E_F^3\right)k_B^2\left(G^2/\beta^2\right)T^2 = A_{ee}T^2$$

$$where \quad \beta = \frac{k_c}{k_F} = 0.353 r_s^{1/2}$$

$$and \quad G = 1/\Omega \int_{cell} u_{k_1}^*(r)u_{k_3}(r)\exp(ig.r)d^3r$$

$$g = k_1 + k_2 - k_3 - k_4 \qquad (9)$$

(For g ≈2k_F), where z is the coordination number of the reciprocal lattice, v_F is the Fermi velocity (= hk_F/m), E_F is the Fermi energy, k_F is the Fermi wave vector and k_B is the Boltzmann constant, G overlap integral , g is the reciprocal lattice vector and Ω is the atomic volume, where r_s is the radius of the Wigner-Seitz sphere, In the expression for A_{ee} all the values are known except G, it will proceed to evaluate this important factor, which plays a crucial role in the e-e Umklapp scattering processes.

Finally, at low temperatures, the total theoretical resistivity in pure noble metals as expected may be written as:

$$\rho = \rho_0 + AT^2 + BT^3\int_0^{\frac{\theta}{T}}\frac{z^3}{\left(e^z-1\right)\left(1-e^{-z}\right)}dz + CT^5\int_0^{\frac{\theta}{T}}\frac{z^5}{\left(e^z-1\right)\left(1-e^{-z}\right)}dz \qquad (10)$$

Whereas at high temperature, resistivity will become:

$$\rho = \rho_0 + dT \qquad (11)$$

3. Experimental database

In a previous study by AL-Jalali [34, 35]; the papers have concentrated only on magnetic resistivity in magnetic dilute alloys. But this paper will treat all other resistivities,and will concentrate on total resistivity in the pure noble metals. Review experimental data from crude results [36,37,38,39,40,41,42], for Copper,Silver and Gold, which belong to resistivity as a function of temperature will be analyzed to discover all mechanisms that contribute in noble metals resistivity, and calculate more than constant by comparison between theoretical and experimental expressions.

4. Results and Discussion

At low temperatures, analysis of experimental data for normal metals resistivity has a general expression as a sum of many contributions(additive law) and take a general form:

$$\rho(T,C) = \rho_0 + AT^2 + BT^3 + CT^5 + ... \qquad (11)$$

Where A, B, C are very important constants to calculate many theoretical constants, which is difficult to know them without comparison between these experimental and theoretical formulae.

Also at high temperatures all contributions vanish except phonons and impurity resistivity, and resistivity will become:

$$\rho(T,C) = \rho_0 + AT \qquad (12)$$

Figure (1) shows a general diagram between experimental noble metals resistivity as a function of low and high temperatures.

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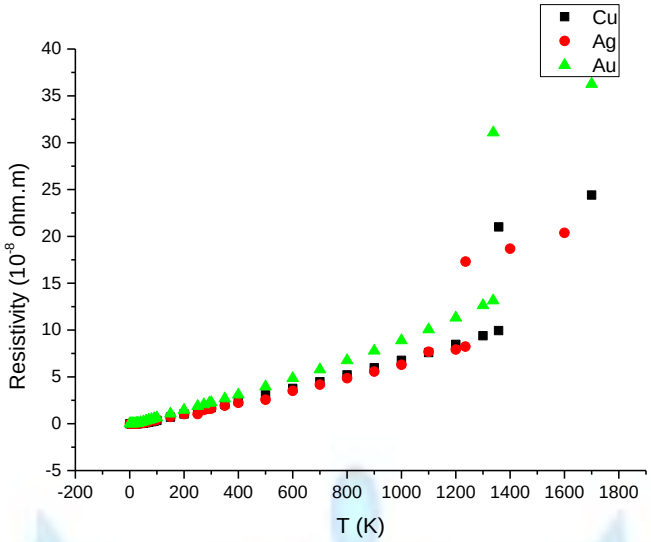

Figure (1): noble metals resistivity as a function of low and high temperatures.

Purity of noble metals was calculated from the ratio of β =ρ (273K)/ρ (4.2K) =RRR, and a suitable fitting between RRR and concentration show that purity highly decrease with concentration increasing, figures (2, 3, 4).

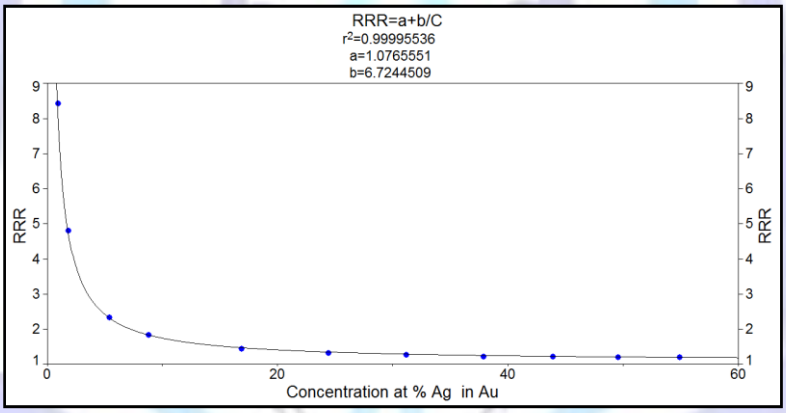

Figure (2): RRR as a function of Ag concentration in Au

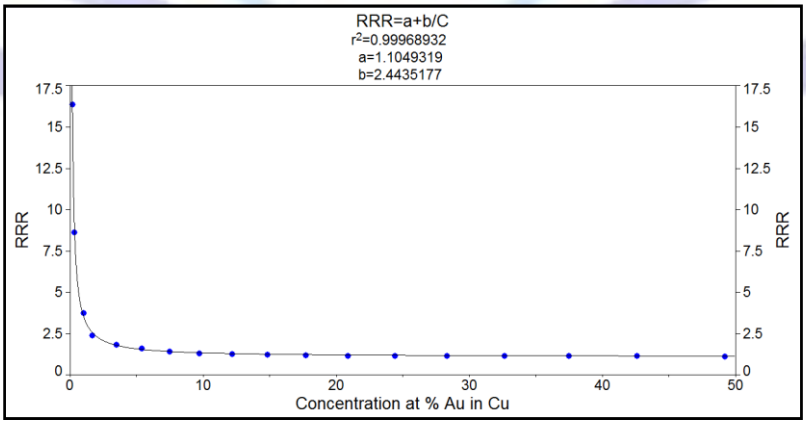

Figure (3): RRR as a function of Au concentration in Cu

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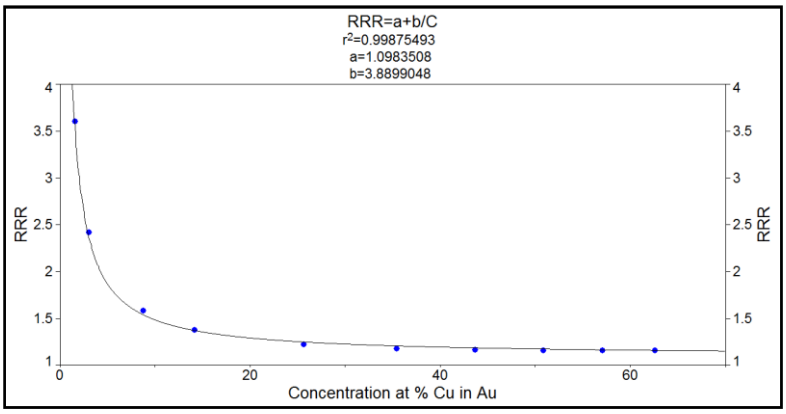

Figure (3): RRR as a function of Cu concentration in Au

It is found that; Residual resistivity as a function of concentration is a mixed from Nordheim's rule [43] for one kind of normal impurity:

$$\rho_0 \propto c(1-c) \qquad (13)$$

In addition, Mott's rule for tow kind of normal impurity [44]:

$$\rho_0 = A(1-c)^2(1-c)c^2 + B(1-c)c \qquad (14)$$

Where A, B are constants. Figures (4, 5) show a changing between residual resistivity and concentration; it is expected that the samples contain more than one kind of impurities.

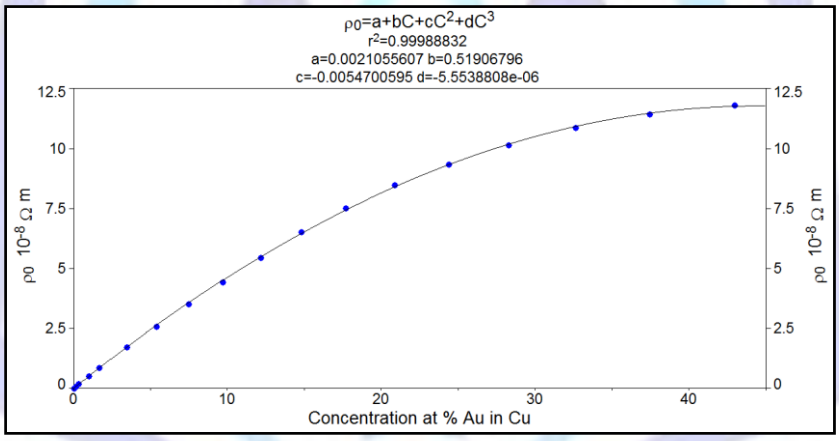

Figure (4): Gold residual resistivity versus Concentration of Gold in Cu-Au alloy

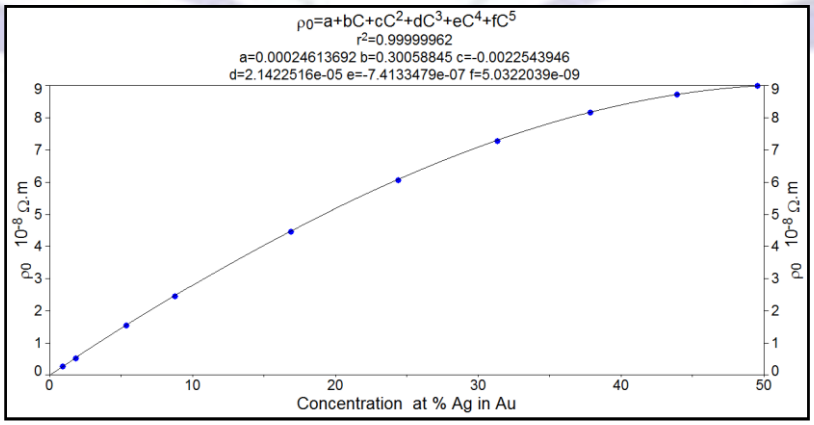

Figure (5): Silver residual resistivity versus Concentration of Silver in Cu-Au alloy

However, in magnetic alloys the situation will be complicated, especially in dilute magnetic alloys.by return to my papers [34,35] which belong to Cu-Mn dilute alloys (Mn in ppm= part per million), where Mn residual resistivity did not calculate.

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By recalculation this resistivity, it is found logarithm residual resistivity dependence of Mn concentration, and the relationship takes the following form:

$$Ln\rho_0 = 0.877\ln(c) - 25.42 \quad Ln(f\ \Omega.m) \qquad (14)$$

In addition, for Cu-Fe dilute alloy (Fe in ppm) has the following form:

$$Ln\rho_0 = 0.945L\ \mathrm{n}(c) - 22.81 \quad Ln(f\ \Omega.m) \qquad (16)$$

Unfortunately, the experimental results calculate the total residual resistivity and could not discriminate between the origins of its mechanisms. firsr Theoretical attempt was made by blatt [45] to calculate resistiveties due to vacancies and interstitials and found that:

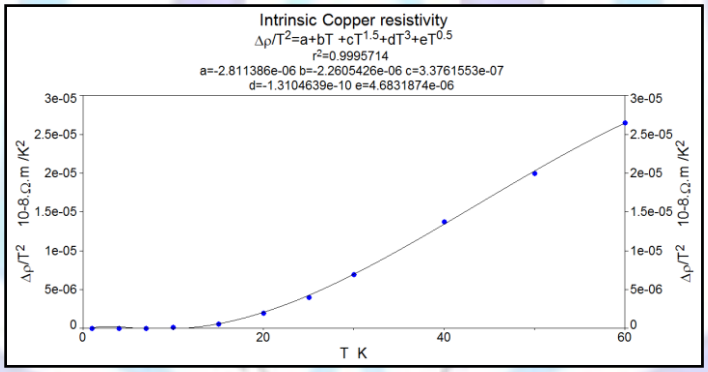
$$\rho_{\mathrm{int}} = (0.5\,to\,10)\times 10^{-8}\Omega.m\ /\ at.\%$$
$$\rho_{vac} = (1\,to\,1{,}5)\times 10^{-8}\Omega.m\ /\ at.\% \qquad (17)$$

However, many attempts were made by neutron scattering, which need deeper investigations.

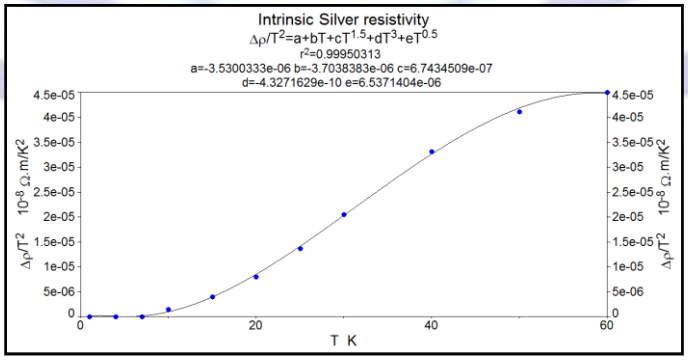
In order to calculate intrinsic resistivity, the basic step is to subtract residual resistivity from total resistivity, as the following relation:

$$\Delta\rho = \rho(T,C) - \rho_0 = AT^2 + BT^3 + CT^5 + ...$$
$$\Delta\rho/T^2 = A + BT + CT^3 + ... \qquad (18)$$

Figures (6, 7, 8) show intrinsic resistivity for noble metals (Cu, Ag, Au) as a function of temperature. Precise mathematical analysis shows the existence of new (terms) mechanisms were not taken into account , like $T^{2.5}$, $T^{1.5}$,$T^{0.5}$,…etc., These mechanisms do not appear, but the presence of magnetic impurities, which appear from spin waves at low temperatures.



Figures (6): intrinsic resistivity for Copper versus temperature



Figures (7): intrinsic resistivity for Silver versus temperature

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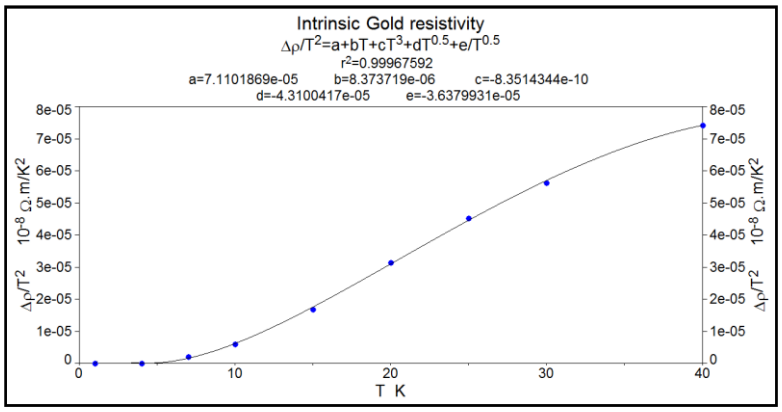

Figures (8): intrinsic resistivity for Gold versus temperature

Debye temperature for face centered cubic (fcc) (Ag, Au, Cu,) metals, near absolute zero and in terms of the elastic constants of the solid. The Debye characteristic temperature is given by [46]:

$$\theta_0^D = \frac{h}{k}\left(\frac{9N}{4\pi v^2}\right)^{\frac{1}{2}}\left[\frac{2}{\upsilon_t^3}+\frac{1}{\upsilon_\ell^3}\right]^{-\frac{1}{3}} \qquad (19)$$

Where h = Planck's constant, k = Boltzmann's constant, N = Avogadro's number, V = atomic volume, υ_ℓ = longitudinal sound velocity, υ_t = transversal sound velocity which mean that Debye temperature is related to the speed of sound in the crystal.

There are agreement between values calculated from (19) and experimental results,for Cu(Exp. θ_D =345.9K,Calcu. θ_D =345.6k),Ag (Exp. θ_D ==227.1k, Calcu. θ_D = 226.6K) and Au(Exp. θ_D = 161.5k, Calcu. θ_D =162.4 I 2k). This indicates that cubic solids can be very well represented by Debye's model and that they show, to a certain extent, polycrystalline behaviour at absolute zero.

Another method to calculate Debye temperature, where the Debye temperature define as a part of the integration of the heat capacity [47] where ω_D is the Debye frequency:

$$C_v = 9N_A K_B\left(\frac{T}{\theta}\right)^3\int_0^{\frac{\theta}{T}}\frac{z^3}{\left(e^z-1\right)\left(1-e^{-z}\right)}dz$$

$$\theta_D = \frac{\hbar\omega_D}{k_B} = \frac{h\vartheta_D}{k_B} = \frac{h\upsilon_{mean}}{\lambda_{\min}} \qquad (20)$$

υ_{mean} the mean acoustic velocity calculated from the longitudinal and transverse velocities, $\lambda_{\min}$ The minimum wavelength corresponding to the Debye temperature, This wavelength should be equivalent with the length of the smallest unit cell.At frequencies higher than the cut off frequency the “lattice” unable to “see” the vibration because the wavelength of the vibration is smaller than the basic unit of the atomic arrangement; therefore, the vibration becomes independent from the lattice. The Debye cut off frequency or temperature separates the collective thermal lattice vibration from the independent thermal lattice vibration.Figure (9) shows Debye temperature as a function of temperature from specific-heat results.

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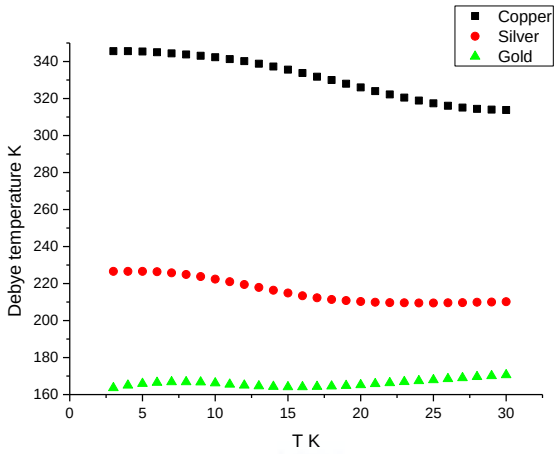

Figure (9): Debye temperature versus temperature from specific-heat results (After David R. Smith and F. R. Fickett, 1995) [48].

Debye temperature can be estimated from the melting point (T_m) of the crystal using the Lindemann relation [49, 50]:

$$\theta_D = C\left[T_m / MV^{\frac{3}{2}}\right]^{\frac{1}{2}} \qquad (21)$$

Where C is a constant depending on the X-ray intensity data structure. V unit volume was estimated from the lattice constant, M Molecular weight . Debye temperature estimated from resistivty less than 4% from estimated from specific heat .

From theoretical equation (8) and experimental relation (18), the electron-lattice interaction constant C (constant of the metal) could be estimated, it is found that:

C=2.265x10-5 Ω.m.K for Cu, C=2.609x10-3 Ω.m.K for Ag, C=2.823x10-3 Ω.m.K for Au.

Which will be useful to estimate other constants. Moreover, from equation (9) and (18), G overlap integral was calculated and its values as follows: G_{Cu}=0. 063, G_{Ag}=0. 075 and G_{Au}=0. 097.

Finally, the agreement between theoretical and experimental was somewhat acceptable.

5. Conclusion

Purity of pure Copper was RRR=772, Silver was RRR=1465 and Gold was RRR=93. 2, which means that many impurities may be found in the samples. This result explains why residual resistivity has a power series equation.

New terms have been found in total resistivity during analysis data, like T^3, which belong to phonons U-processes, and $T^{2.5}$, $T^{1.5}$, $T^{0.5}$, which belong to magnetic origin.In addition, Debye temperature and some theoretical constant were treated.

Most of all predictions of theoretical models was an agreement with analyzing experimental data, but need more investigation to reach to an agreement with each other.

Acknowledgement

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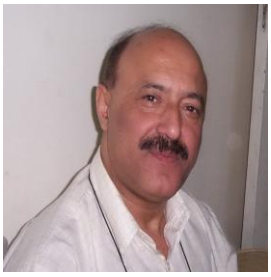
Author' biography with Photo



Muhammad Ahmad Ali AL-JALALI (Syrian) , Is Associate Professor in department of Physics Taif University, Kingdom of Saudi Arabia. B.Sc. in Physics & Chemistry, High Diploma in Modern Physics, M. Sc. in Modern Physics, Ph.D. in Theoretical Physics (solid state). His field of interest is Spin Glasses, memories and nuclear memories, modern magnetism,NMR, new & renewable energy, scientific cooperation between south and north states.

His site: **https://sites.google.com/site/maljalali/**