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Influence of Size, Shape, and Scattering on Electrical Resistivity of Metal Nanowires

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A simple quantitative model has been proposed for exploring the combined effect of size, shape, and electron scattering on the electrical resistivity of metallic nanowires. In the present model, the effect of different cross-sectional shapes of nanowires has been comprised on the surface and grain boundary scattering. For understanding electrical behavior at the nanolevel, the incorporation of specularity parameter (p) with different cross-sectional shapes of nanowires is essential. It is responsible for the reduction in the mean free path of electrons; which generates the favorable condition for enhancing the surface scattering, consequently contributing to increment of electrical resistivity. The applicability of the proposed model has been investigated for copper, nickel, silver, and aluminum metallic nanowires of four different cross-sectional shapes (rectangular, triangular, square, and spherical) along with different values of reflection coefficient (R). Calculated results have been compared with the available experimental data and it is observed that the results are in close agreement, which proves the validity of the proposed model. The proposed model shows the collective effect of size, scattering, and cross-sectional shape factor (δ) on electrical resistivity in a very simple and straightforward manner and able to reduce the complexity of existing models up to great extent.

Keywords: Low-dimension materials; Electrical resistivity; Surface scattering; Grain-boundary scattering; Specularity parameter; Reflection coefficient

1 Introduction

The unique structural, thermal, optical, catalytic, and electrical properties of low-dimensional materials have attracted global attention for many years[1-3]. Modification in the electrical properties at nanolevel is the consequence of discrete electronic energy levels, formation of electric dipoles, and spatial confinement of electrons and holes. Researchers have taken interest in the study of electrical conduction measurements for decades[4-6]. Since low-dimensional materials are basic components for next-generation electronics, therefore, miniaturization of electronic components has made an electrical study of materials important.[7]. Confinement in size of materials into two dimensions (nanowires) exhibit many unique properties which differ from the bulk materials as well as from the one-dimensional constrained case (thin films)[8-12].

Metallic nanowires (NWs) have received special interest from researchers[13-17] due to variation in conductivity with the wire's diameter, which can be used as a transparent conductive material for many devices, such as solar cells, flexible light-emitting devices, stretchable electrodes, flexible transparent thin-film heaters.

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According to Matthiessen's rule the total electrical resistivity (ρ_T) of a metal is a combination of the contribution of individual and independent scattering[18]:

$$\rho_T = \rho_{Th} + \rho_D \quad \text{... (1)}$$

Where ρ_{Th} and ρ_D are thermal and defect resistivity respectively.

The thermal resistivity is due to thermal or phonon contribution which comes from the electron-phonon collisions and it is temperature-dependent. The defect resistivity is an outcome of the impurity atoms, defects (vacancies and grain boundaries), and is temperature independent.

Conductivity measurements of metal NWs have been explained by many models including Dingle's model for circular cross-section wires[19], MacDonald and Sarginson's model for rectangular cross-section[20], and Chamber's model for arbitrary cross-section[21]. Due to the lack of grain boundary scattering for polycrystalline metal wires, these theories are not widely used today. When the size of the metallic nanostructure is comparable or smaller to the electron mean free path (λ), the scatterings of the coherent oscillating electrons at the particle surface (surface scattering) and scattering from the interface between the two grains (grain boundary scattering) both become important[22-23]. An undesirable impact is observed in the electrical transmission properties of these interconnects at room temperature[10].

In recent years researchers have done wonderful work on the combined effect of surface and grain boundary scattering mechanisms on the electrical resistivity of metal nanowires [10,24-26]. Mayadas and Shatzke's theory described that a major contribution in total electrical resistivity is due to the grain-boundary scattering in polycrystalline materials and this phenomenon was also verified by different experiments[27]. C. Durkan and M. E. Well experimentally studied the diameter dependence of the conductivity for polycrystalline gold nanowires and found that the conductivity is affected by the wire's diameter only when it is of mean grain size order{40-45nm}[28]. For practical applications at the nanolevel integration of surface and grain boundary scattering with cross-sectional shape is essential. Existing models are not explaining the integrated effect of surface scattering and cross-sectional shape, and are too complex in application. So, the present study is an effort to formulate a simple straightforward theoretical model to analyze the size and shape-dependent electrical resistivity along with the surface and grain-boundary scattering effect. The model proposed for thermal conductivity by Arora *et al.*[29] has been modified in the light of Wiedemann-Franz law for getting surface scattering and then it is combined with grain boundary scattering[27], and nanowires are considered defects free. The proposed model has been applied for metal nanowires of different cross-sectional shapes such as spherical, rectangular, square, and triangular, and also checked for different values of reflection coefficient (R).

2 Methods

The size effect manifests itself due to the scattering processes of the conduction electrons at the external and internal interfaces. Several scattering mechanisms can contribute to the total electrical resistivity but surface scattering and grain boundary scattering are of great interest:

i. **Surface scattering:**

The Wiedemann-Franz law states that the ratio of the thermal conductivity (K) to the electrical conductivity (σ) of metal is proportional to temperature (T):

$$\frac{K}{\sigma} = LT \qquad \text{... (2)}$$

The proportionality constant, L is known as the Lorentz number.

Electrical conductivity is reciprocal to electrical resistivity (ρ), therefore, at a constant temperature, the relative resistivity for nanomaterials is given as:

$$\frac{\rho}{\rho_n} = \frac{K_n}{K} \qquad \text{... (3)}$$

Where K_n and ρ_n respectively denote the thermal conductivity and electrical resistivity of nanomaterials.

Arora *et al.*[29] proposed a thermodynamic model to study the collective effect of size and dimension dependent specific heat capacity along with phonon surface scattering on thermal conductivity as:

$$\frac{K_n}{K} = \mathrm{p} \times \exp\left[-\frac{\lambda}{D}\right] \times \frac{C_n}{C} \times \left(1 - \frac{N}{2n}\right)^{\frac{3}{2}} \qquad \text{... (4)}$$

Where the value of 'N/2n' is 4d/3D for nanowire and d, D is the atomic diameter and diametric size of the nanowire respectively. C_n and C denote the specific heat for nano and their bulk counterpart respectively. The authors considered the phonon scattering mechanism to visualize the effect of phonon mean free path on thermal conductivity and incorporated the term 'p exp (-λ/D)' in the expression. Where (λ/D) is a dimensionless parameter known as the Knudsen number and 'p' is the specularity parameter which has a constant value lying between 0 and 1 for partially specular and diffuse surfaces.

Electronic conduction is proportional to the background mean free path and the presence of surfaces provides efficient scattering regions[30]. An electron can undergo either elastic (specularly scattered) or inelastic (diffusely scattered) scattering. Specularity parameter (p) and the distance between two roughness features (i.e. correlation length L) are the two variables used to characterize the surface scattering[31]. In nanowires, electrons go through continuous internal and boundary scattering consequently it shows the reduction in the mean free path, and is written as [30]:

$$\lambda = \lambda_0 \left[1 - \frac{(1-p)\exp\left(-{L_1}/{\lambda_0}\right)}{1-p\exp\left(-{L_2}/{\lambda_0}\right)}\right] \qquad \text{... (5)}$$

Where L_1 and L_2 are correlation lengths and L_2 is considered two times that of L_1.

In the present work, equation (4) have been modified by considering surface scattering of electrons in place of phonon scattering for metals so mean free path reduction term 'p exp ($-\lambda/D$)' is replaced by mean free path reduction for electrons [*i.e.* equation (5)] $\exp\left(-\frac{\lambda_0\left[1-\frac{(1-p)\exp\left(-L_1/\lambda_0\right)}{1-p\exp\left(-L_2/\lambda_0\right)}\right]}{D}\right)$ is used in equation (4).

Since the specific heat of nanomaterials is size and dimension dependent quantity, therefore, in this study an expression has been developed for specific heat with the help of previously existing studies[32-33] as:

$$\frac{C_n}{C} = \left(1 - \frac{T_0}{T_{mb}}\right)\left\{1 + \frac{\left(T_0/T_{mb}\right)}{\left(1-\frac{4d}{3D}\right)}\right\} \quad \text{... (6)}$$

T_0 and T_{mb} are the reference temperature and melting temperature of bulk material respectively.

Incorporating the effect of surface scattering and size-dependent specific heat in equation (4) then using it in equation (3) the size-dependent electrical resistivity due to surface scattering can be written as:

$$\rho_n = \frac{\rho}{\exp\left(-\frac{\lambda}{D}\right)\left(1-\frac{4d}{3D}\right)^{3/2}\left(1-\frac{T_0}{T_{mb}}\right)\left\{1+\frac{T_0/T_{mb}}{\left(1-\frac{4d}{3D}\right)}\right\}} \quad \cdots (7)$$

In the present work, the shape factor (δ) has also been incorporated in the above equation as the cross-sectional shape is an important factor that affects the properties of nanowires[34].

$$\rho_n = \frac{\rho}{\exp\left(-\frac{\lambda\delta}{D}\right)\left(1-\delta\frac{4d}{3D}\right)^{3/2}\left(1-\frac{T_0}{T_{mb}}\right)\left\{1+\frac{T_0/T_{mb}}{\left(1-\delta\frac{4d}{3D}\right)}\right\}} \quad \ldots (8)$$

The modified equation (8) describes the effect of surface scattering along with size and shape on electrical resistivity.

ii. Grain boundary scattering:

In the MS grain-boundary scattering model the enhancement in grain boundary resistivity is given by[27]:

$$\rho_{gr} = \frac{\rho}{f(\alpha)} \quad \ldots (9)$$

Where ρ_{gr} denotes the electrical resistivity due to grain boundary scattering of electrons and $f(\alpha)$ is given as:

$$f(\alpha) = 1 - \frac{3}{2}\alpha + 3\alpha^2 - 3\alpha^3 \ln\left(1 + \frac{1}{\alpha}\right) \quad \ldots (10)$$

Where $\alpha = \frac{\lambda_0}{D_g}\frac{R}{1-R}$ ... (11)

In the above equations, D_g is the average grain diameter or the smaller wire width and height. R is the reflection coefficient of the electron striking at the grain boundaries ($0 \leq R \leq 1$).

The total electrical resistivity for metallic nanowires can be given by adding the expressions obtained due to surface scattering equation (8) and grain boundary scattering equation (9):

$$\rho_T = \frac{\rho}{\left[\exp\left(-\frac{\lambda\delta}{D}\right)\left(1-\delta\frac{4d}{3D}\right)^{3/2}\left(1-\frac{T_0}{T_{mb}}\right)\left\{1+\frac{T_0/T_{mb}}{\left(1-\delta\frac{4d}{3D}\right)}\right\}\right]} + \frac{\rho}{\left[1-\frac{3}{2}\alpha+3\alpha^2-3\alpha^3\ln\left(1+\frac{1}{\alpha}\right)\right]} \quad \ldots (12)$$

Equation (12) gives the size and shape-dependent total electrical resistivity due to surface and grain boundary scattering for metals.

The values of different shape factors for different cross-sectional shaped nanowires are given in Table 1 and the input parameters used for the present study are given in Table 2.

3 Results and Discussion

Nowadays, the demands for smart materials increasing with the developments taking place around us particularly in the field of electronics. To make the devices more flexible, stretchable, and portable, conductive parts should have bendable and deformable properties. Recently, nanowires and other two-dimensional nanostructures such as nanorods, nanobelts, nanoribbons, *etc*. have gained the attention of scientists due to their high potential applications in molecular electronics, diagnostic biosensors, energy storage, and optoelectronic devices[40-42].

Table 1 — Shape factor for the different cross-sections of nanowires.

Cross-sectional shape	Shape factor (δ) [34]
Spherical	1
Rectangular	$\frac{n+1}{\sqrt{\pi n}}$, (t=nb); t, b are thickness and width of rectangular nanowires and n is any real integer
Triangular	1.286
Square	1.128

Table 2 — Input parameters used in the present study.

Metals/ Parameter	Bulk electrical resistivity (ρ_0) (Ω-m)[35]	Atomic diameter (d) (nm)	Bulk electron mean-free path (λ_0) (nm)	Bulk melting temperature (T_{mb}) (K)
Cu	$1.678*10^{-8}$	0.256[36]	39.9 [35]	1358 [36]
Ni	$6.93*10^{-8}$	0.2492[36]	14 [39]	1728 [36]
Ag	$1.587*10^{-8}$	0.288[37]	53.3 [35]	1234 [37]
Al	$2.65*10^{-8}$	0.246[38]	18.9 [35]	933.47 [37]

Metallic nanowires are promising candidates for a range of device applications and high reliability interconnects due to their excellent electrical properties, transparency, and flexibility[43]. There have been several sizes and shape-dependent studies on the electrical resistivity of metal nanowires e.g. for Cu NW[44-45], Ni NW[39], Ag NW[24,46], and Au NW[25]. The electrical resistivity is related to the transport of charge carriers which depends on the type of solids say metal, insulator, or composite, and also on the grain size, defects, doped impurities, etc. The presence of contact resistance makes electrical measurements harder[47]. Nevertheless, because of the importance of the Ohmic losses in electronics, many studies already investigated the change of resistivity with the size and the crystalline quality of interconnect structures[48]. The results demonstrate that the two main contributions to the resistivity increment are surface scattering and grain-boundary scattering along with changing the size of metal interconnects. The Matthiessen rule according to which the several contributions add up to the total resistivity[49] was proven to be a valid approximation in most cases.

In the present work an accessible theoretical model has been developed for electrical resistivity of metal nanowires by adding two types of resistivities; first electrical resistivity due to surface scattering, has been obtained by modifying the thermal conductivity model of Arora *et al*. (2018) with the combination of Wiedemann-Franz law and the second one is defect scattering due to grain boundaries[27].

This model incorporates the size and shapes effect on electrical resistivity for copper (Cu), nickel (Ni), silver (Ag), and aluminum (Al) metallic nanowires. Four different cross-sectional shapes of nanowires spherical, rectangular, square, and trigonal have been considered for the application of the model. Figs. 1, 2, 3, and 4 represent the size and shape dependency of electrical resistivity calculated with the help of equation (12) along with available experimental data for Cu, Ni, Ag, and Al metallic nanowires respectively. It is observed that electrical resistivity increases with the decrement in diameter of nanowires and the results are in close agreement with the available experimental data[24,39,43,49]. The electrical resistivity of metallic nanowires is more than that of bulk metals. This is because of the reduction in size which leads to structural electron scatterings; like grain boundary scattering and surface scattering. For bulk metals, the surface scattering dominates at room temperature and structural scatterings are rare, but for metallic nanowires, both the surface and

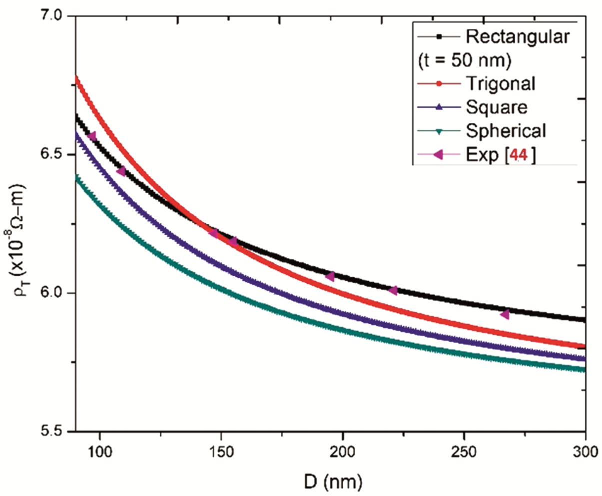

Fig. 1 — Electrical resistivity variation of Cu nanowire with diameter for different cross-sections (p=0.5, R=0.34)

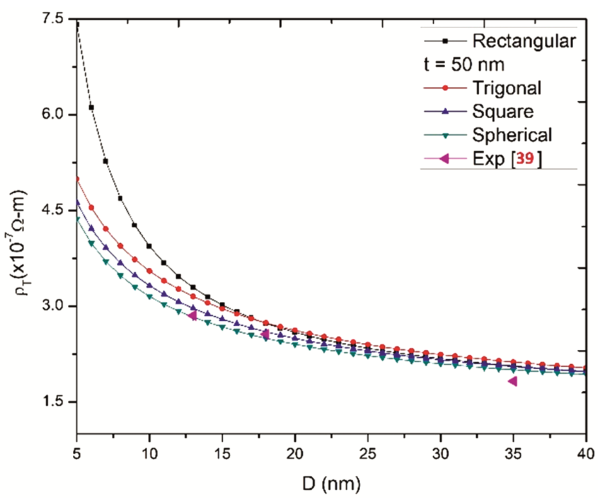

Fig. 2 — Electrical resistivity variation of Ni nanowire with diameter for different cross-sections (p=0.5, R=0.04)

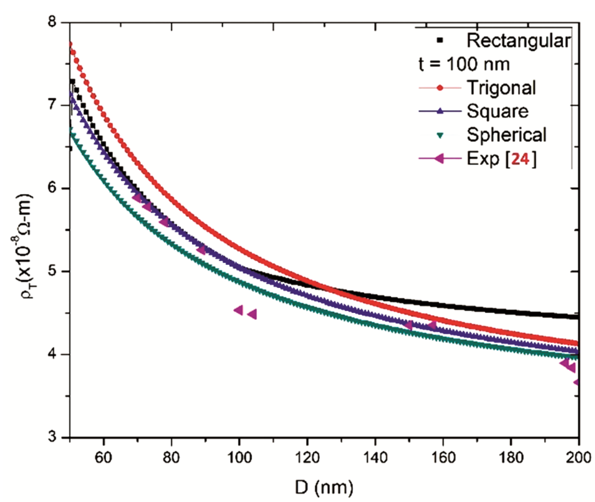

Fig. 3 — Electrical resistivity variation of Ag nanowire with diameter for different cross-sections (p=0.5, R=0.3)

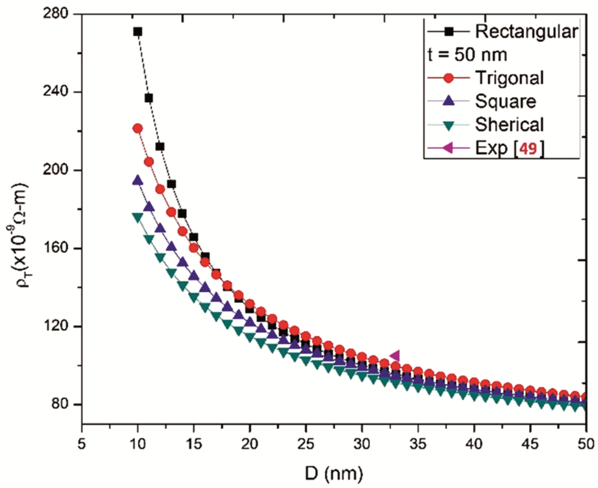

Fig. 4 — Electrical resistivity variation of Al nanowire with diameter for different cross-sections (p=0.5, R=0.4)

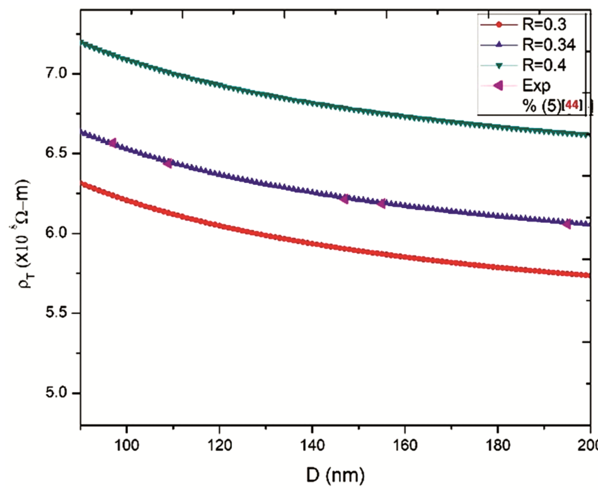

Fig. 5 — Electrical resistivity variation of rectangular Cu nanowire with diameter for different values of R

grain boundary scattering contribute significantly to the electrical resistivity[45].

The electron's motion is interrupted by collisions with the surface when the diameter of nanowires is comparable or smaller than the electron mean free path. In diffused scattering, the electron mean free path is terminated by impinging on the surface and the electron loses its velocity along the direction parallel to the surface due to inelastic scattering, hence conductivity decreases[22]. Nanostructures consist of a large number of grain boundaries and surfaces among them because of forming by nanocrystals. Back reflected electrons exchange their energy with the local phonons after scattering from the grain boundaries. Phonons can transfer more readily across the grain boundaries than electrons hence, on the other side of the grain boundaries exchange of energy takes place between electrons and phonons. Therefore, instead of charge transport, a part of the electron energy transfers through the grain boundaries[45]. This leads to a great reduction in electrical conductivity hence, electrical resistivity is enhanced.

It has also been found that the electrical resistivity varies with different cross-sectional shapes of nanowires. Electrical resistivity has a greater value for the cross-section with the larger value of shape factor, therefore, triangular or rectangular shapes of the cross-section have the largest value (depending on the height and width of the rectangular nanowire) and for the spherical shape, it has the lowest value of electrical resistivity.

The variation of electrical resistivity of Cu, Ni, Ag, and Al metallic nanowires with different values of reflection coefficient (R) along with available experimental data has also been plotted in Figs. 5, 6, 7 and 8 respectively.

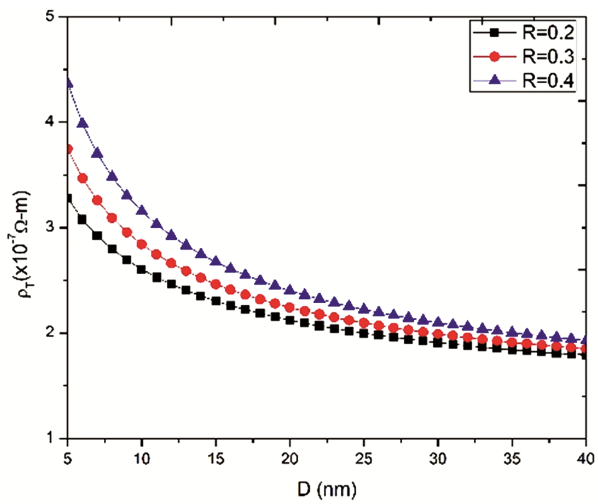

Fig. 6 — Electrical resistivity variation of Spherical Ni nanowire with diameter for different values of R

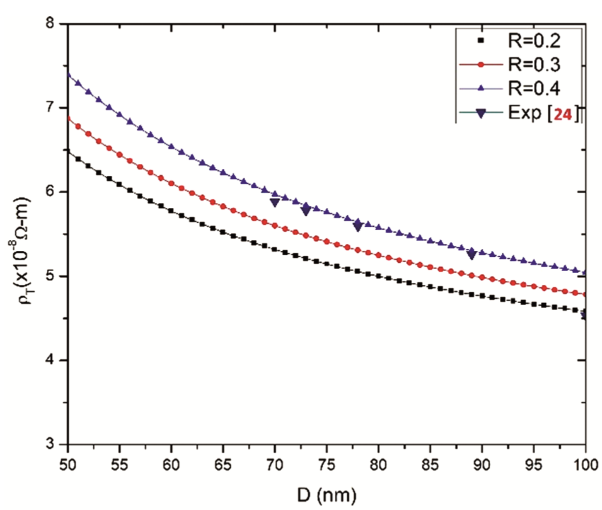

Fig. 7 — Electrical resistivity variation of rectangular Ag nanowire with diameter for different values of R

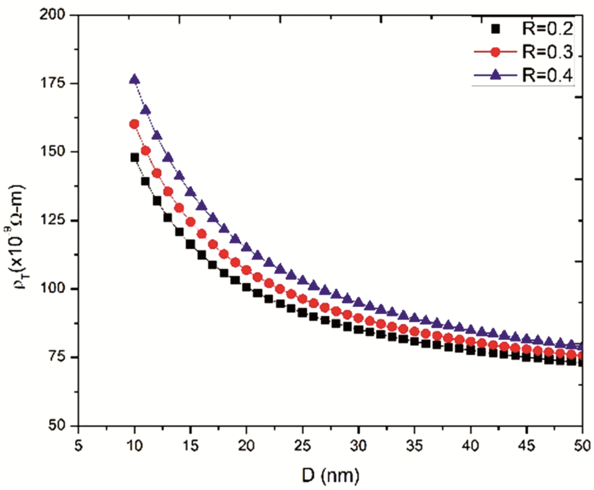

Fig. 8 — Electrical resistivity variation of Spherical Al nanowire with diameter for different values of R

Small variations in R cause large changes in the electrical resistivity. An enhancement in the value of electrical resistivity is observed with the increment in reflection coefficient and the outcomes are in agreement with the available experimental data [24-25,44]. This can be understood simply as the higher the reflection probability at the grain boundary, the higher the resistivity[10].

4 Conclusion

An uncomplicated theoretical model to investigate the integrated effect of size, cross-sectional shape, surface roughness, and reflection probability on the electrical resistivity of metal nanowires has been proposed the first time . Electrical resistivity varies with the variation in size and cross-sectional shape of nanowires. Electrical resistivity shows increment with a decrement in size. Its value is higher for large shape factors due to surface effect; hence the cross-sectional shape of wires also plays an eminent role with size. The present model shows the contribution of two scattering mechanisms: surface scattering and grain boundary scattering to the total electrical resistivity. It is found that both the scattering mechanisms subsidies to the enhancement of total electrical resistivity. The obtained results are in good agreement with the available experimental data, which supports the validity of the proposed model. The collective effect of size, cross-sectional shape, and scattering for nanowires is important to understand the actual electrical behaviour of small-sized materials. Therefore, the present study can be helpful for the fabrication of low electrical conductivity material which can be used as the filaments for incandescent lamps, heating elements, furnaces, space heaters and many electronic devices.

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