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Dependence of low temperature resistivity of iron films on thickness, substrate temperature and rate of deposition

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ABSTRACT

Under high vacuum conditions, using electron beam gun evaporation method iron films of varied thicknesses were deposited onto the glass substrates. These films have been investigated for electrical resistivity in the temperature range 80K to 300K. The residual resistance ratio, RRR, and the temperature coefficient of resistance, TCR, were determined and discussed. The power laws for the temperature dependence of resistivity have been established. The thickness dependence of electrical resistivity has been considered in terms of Fuchs-Sondheimar theory and the resistivity of infinitely thick film, ρ_0, and electron mean free path, l, were determined. The influence of deposition rate of the film on resistivity has been studied. The resistivity decreased with increasing substrate temperature and that is attributed to the formation of larger crystallites with increase of substrate temperature. The deposition rate of 2 Å/s is observed to be the optimum rate at which the iron films of minimum resistivity can be produced. It is for the first time that iron films have been investigated in single measuring setup for low temperature resistivity and its dependence on thickness of the film, substrate temperature and rate of deposition.

KEYWORDS

Thin films;
Electrical resistivity;
Fuchs-Sondheimar theory;
Electron mean free path.

INTRODUCTION

The study of thin films of magnetic materials has always been a subject of great interest to many physicists due to their interesting properties such as low temperature coefficient of resistivity (TCR), low thermo electric power (TEP) etc., and applications as thin film resistors[1,2]. The interesting physics associated with the properties of the thin films has also been of attraction. The electrical resistivity studies on several materials as a function of temperature, thickness of the film, rate of deposition, substrate temperature, annealing temperature, vacuum levels were reported[3-7]. For example, the resistivity of manganese films was investigated as a function of substrate temperature, thickness and annealing temperature[1,3]. The electrical transport studies on chromium films were reported[4,5]. Though, most of the metal thin films exhibit temperature dependence of resistivity similar to that of their bulk forms, interestingly, few interesting aspects have been found in some metal films. For example, large deviations from the Matthiessen's rule have been detects in copper films of about 100nm

thick[6]. Nickel films showed anomalous resistivity dependence on temperature[7]. The manganese films measured negative temperature coefficient around its Neel's temperature[8] and low temperature resistivity of these films was found to follow a T^2 law. The power law behaviour of low temperature resistivity in many metals differs from each other.

The standard theory used for accounting the electrical conductivity of thin films is that of Fuchs and Sindheimer, which we will refer to as the F-S theory[9] where, the scattering of electrons within the film is described by a relaxation time τ, and that at the surface of the film by a specularity parameter, p.

In view of the fact that low temperature resistivity behaviour of transition metal films is not completely understood yet, here, we report low temperature resistivity studies on iron films deposited using electron beam gun under high vacuum conditions. Further, we report here the residual resistance ratio (RRR), temperature coefficient of resistance (TCR), resistivity dependence on thickness, substrate temperature, rate of deposition and temperature of the films produced under the same vacuum conditions measured in single setup. Very few of studies of this kind can be traced in the literature.

EXPERIMENTAL

Three sets of iron (of purity 99.5%) films were deposited on to the glass substrates by electron beam gun method under the vacuum conditions of $1x10^{-5}$ Torr in a standard Hindhivac coating unit. These films include, (i) four films of thickness 100nm, 200nm 300nm and 400nm deposited at a substrate temperature of 200 ^{0}C and the rate of deposition to be 1 Å/sec, (ii) four films of thinness 100 nm each deposited at the substrate temperatures of 100 ^{0}C, 150 ^{0}C, 200 ^{0}C and 250 ^{0}C and rate of deposition to be 1 Å/sec and (iii) three films of thickness 100 nm each deposited at a substrate temperature of 200 ^{0}C and the rates of deposition to be 1 Å/s, 2 Å/s and 5 Å/s. The thicknesses of the film were monitored during deposition with the help of quartz crystal digital thickness monitor.

The samples were cut to the appropriate sizes and electrical resistance measurements were carried out by following four point method in a liquid nitrogen cryostat in the temperature range from 80 to 300K. A constant current of 1mA has been passed parallel to the plane of the film from a Scientific Instruments make constant source and the voltage developed across the voltage measuring leads was measured using a Keithley nanovoltmeter (Model 2182A). The currents and voltages were measured with in the accuracy of 2%. The temperature was controlled using a local make temperature controller and is measured with the help of pt-100 sensor. The error on the measured temperature was ±1K.

RESULTS

The resistivity, ρ, of the films has been determined as per, $\rho=(Rtb)/l$, where R is the sheet resistance, t the thickness, b the breadth and l the distance between the voltage measuring leads on the film, and are found to be of the order of 10^{-7} (Ωm) for all three series of films. The residual resistivity ration, RRR, of the films was worked out using the expression $\rho(300K)/\rho(80K)$ as, the lowest and highest temperatures used in the present experiments were 80K and 300K respectively. The RRR values thus obtained are recorded in TABLE 1. The temperature coefficient of resistance (TCR) has been determined in the temperature range from 80K to 300K as $TCR = [(d\rho/dT)/\rho_s]$ where ρ_s is the room temperature resistivity. The obtained TCR values are in the order of $10^{-3}(K^{-1})$ and are recorded in TABLE 1.

TABLE 1 : Residual resistivity ratio (RRR) and temperature coefficient of resistivity, (TCR) for different thickness, t, for series (i), for different substrate temperature, T_S, for series (ii) and for different deposition rates, t_d, for series (iii) iron films.

Series (i)			Series (ii)			Series (iii)		
t (nm)	RRR	TCR x10^{-3} (K^{-1})	T_S (^{0}C)	RRR	TCR x10^{-3} (K^{-1})	t_d (Å/s)	RRR	TCR x10^{-3} (K^{-1})
100	1.356	1.230	100	1.251	0.962	0.5	1.122	0.492
200	1.434	1.414	150	1.273	1.046	1	1.356	1.230
300	1.548	1.701	200	1.356	1.230	2	1.305	1.002
400	1.596	1.768	250	1.320	1.209	3	1.320	1.080

The resistivity of the film decreased with increasing thickness at all temperatures of interest. The plots of resistivity versus thickness at three different temperatures are shown in Figure 1(a). The resistivity decreased with increasing substrate temperature in the entire range of temperature of study. The resistivity variation with

substrate temperature is shown in Figure 1(b). With increasing rate of deposition, the resistivity of the films decreased up to 2 Å/s and increased for 5 Å/s as shown in Figure 1(c).

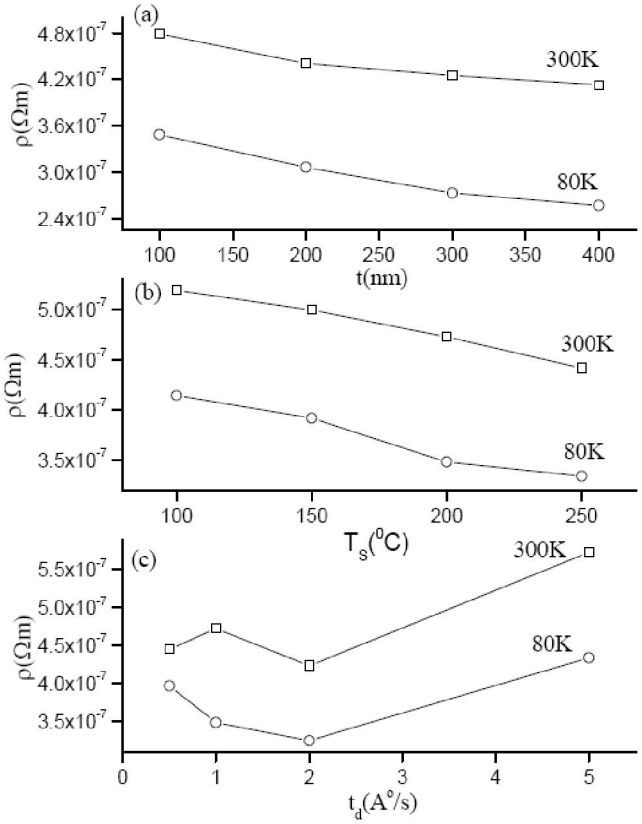

Figure 1 : (a) Resistivity, ρ, versus thickness, t, for series (i) films, (b) Resistivity, ρ, versus substrate temperature, T_d, for series (ii) films and (c) Resistivity, ρ, versus deposition rate, t_d, for series (iii) films.

DISCUSSION

The resistivity of the films increased slowly with increasing temperature and are in the range of 10^{-7} (Ωm), which agree with the ranges reported for manganese and nickel film[7,8]. The RRR values of the films are close to unity suggesting that the mean free path of the electrons is broadly constant with temperature. This also implies that impurity (defects) and/or grain boundary scattering rather than phonon scattering is the dominant mechanism. The TCR increased with increasing thickness of the films and are in the range of 1.230×10^{-3} K^{-1} to $1.768 \times 10^{-3} K^{-1}$.

Resistivity dependence on thickness

The higher resistivity for thinner films may be due to the presence of an island structure with a large density of defect sites. The size of islands increases with increase of film thickness and eventually attach to each other leading to a continuous film and that reduces the density of defect sites. Therefore, electrical resistivity decreases with increasing film thickness[10,11]. The resistivity decrease with increase of film thickness is considered in view of Fuchs-Sondheimar (FS) theory. According to FS theory, the electrical resistivity of a metallic thin film is expressed as $(\rho / \rho_0) = 1 + (3l / 8t)(1-p)$, where ρ is resistivity of the film, ρ_0 the resistivity of infinitely thick film, t the thickness of the film, l the electron mean free path and p specularity parameter. The plot of (ρt) versus t is plotted and shown in Figure 2. The least square linear fit through the data gave ρ_0 and l(1-p). The ρ_0 and l(1-p) values obtained are $\rho_0 = (2.30 \pm 0.1) \times 10^{-7}$ (Ωm) and $l(1-p) = (1.38 \pm 0.27) \times 10^{-5}$ (Å) at temperature 100K, $\rho_0 = (3.04 \pm 0.08) \times 10^{-7}$ (Ωm) and $l(1-p) = (1.14 \pm 0.22) \times 10^{-5}$ (Å) at 200K and, $\rho_0 = (3.864 \pm 0.05) \times 10^{-7}$ (Ωm) and $l(1-p) = (9.27 \pm 1.4) \times 10^{-6}$ (Å) at 300K respectively. Since the range of specularity parameter which is a measure of surface smoothness is 0 to 1, the electron mean free path, l, has been calculated for different values of p in the range 0 to 1. The l values thus obtained at three different temperatures are listed in TABLE 2 and they are in close agreement with l values quoted for thin manganese films[3]. From the TABLE 2, it can be seen that l decreases with increasing temperature. This implies that when the temperature is increased more phonons are generated and which enhance electron-phonon scattering leading to reduced mean electron free paths.

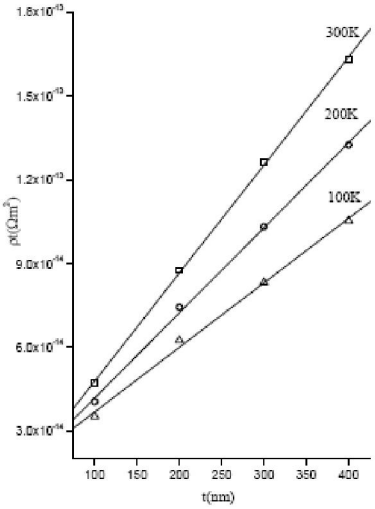

Figure 2 : The plots of (ρt) versus t at three different temperatures. The solid lines are the least square linear lines fit as per F-S theory.

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TABLE 2 : The electron mean free paths, l, at different temperature, T, for different values of specularity parameter, p, between 0 and 1, for series (i) films.

T = 100 K		T = 200 K		T = 300K	
p	l (Å)	p	l (Å)	p	l (Å)
0.2	799	0.2	1248	0.2	2001
0.4	1065	0.4	1664	0.4	2669
0.6	1598	0.6	2496	0.6	4003
0.8	3196	0.8	4993	0.8	8007

Resistivity dependence on substrate temperature and rate of deposition

The decrease of resistivity with increase of substrate temperature (Figure (1(b)) may be due to the presence of larger crystallites in the films that were grown at higher substrate temperatures than that produced at lower substrate temperatures. When the film is deposited at higher substrate temperature, the clusters or adatoms will exhibit higher surface mobility and hence produce larger crystallites[11-13]. However, our hypothesis needs confirmation by the structural studies. The resistivity of the films decreased up to the deposition rate of 2 Å/s and increased for higher deposition rates. It may be that in the present films up to the deposition rate of 2 Å/s stable and relatively larger crystallites are formed and for higher deposition rates the size of the crystallites formed become small leading to larger electrical resistivities. This implies that under the present vacuum conditions the deposition rate of 2 Å/s is an optimum rate at which a film of minimum electrical resistivity can be produced.

Resistivity dependence on temperature

The resistivity-temperature data indicated two regimes of variations. As an example, The resistivity variation with temperature for series (i) films is shown in Figure 3. Similar nature of variations are observed for series (ii) and series (iii) films. Therefore, the temperature dependence of resistivity of the films has been considered by dividing the entire temperature range into two intervals that is, T=80K to 125K and T= 125K to 300K. The power law of the type, $\rho(T) = A + B T^n$ has been fit to the data in the temperature range from 80K 125K and $\rho(T) = C + D T^m$ in the temperature range 125K to 300K[14]. The best fit coefficients have been extracted in both the temperature ranges and are shown in TABLE 3. The exponent m was found to be slightly more than

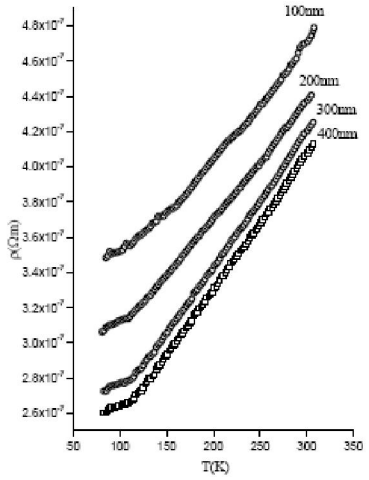

Figure 3 : The plots of resistivity, ρ, versus temperature, T, for series (i0 films.

TABLE 3 : The fit parameters A, B and exponent n values for all the three series of films for the temperature range from 80K to 125K.

Series (i)				Series (ii)				Series (iii)			
t (nm)	A (Ωm)	Bx10^{-7} (ΩmK^{-n})	n	T_S (^{0}C)	A (Ωm)	Bx10^{-7} (ΩmK^{-n})	n	t_d (Å/s)	A (Ωm)	Bx10^{-7} (ΩmK^{-n})	n
100	6.1x10^{-18}	3.45	4.5	100	1.58x10^{-10}	4.02	2.0	0.5	7.55x10^{-11}	3.81	2.2
200	8.72x10^{-16}	3.03	3.5	150	5.53x10^{-16}	3.88	3.5	1	6.17x10^{-18}	3.45	4.5
300	4.29x10^{-20}	2.72	5.5	200	6.17x10^{-18}	3.45	4.5	2	2.38x10^{-10}	3.05	2.0
400	9.08x10^{-14}	2.48	2.6	250	1.83x10^{-25}	3.32	8.0	3	4.37x10^{-18}	4.33	6.9

The fit parameters C, D and exponent m values for all the three series of films for the temperature range from 125K to 300K.

Series (i)				Series (ii)				Series (iii)			
t (nm)	C (Ωm)	Dx10^{-7} (ΩmK^{-m})	m	T_S (^{0}C)	C (Ωm)	Dx10^{-7} (ΩmK^{-m})	m	t_d (Å/s)	C (Ωm)	Dx10^{-7} (ΩmK^{-m})	m
100	2.85x10^{-11}	3.21	1.5	100	1.14x10^{-10}	3.74	1.25	0.5	2.18x10^{-10}	3.79	1.0
200	1.86x10^{-10}	2.62	1.2	150	1.58x10^{-10}	3.49	1.20	1	2.85x10^{-11}	3.22	1.5
300	4.02x10^{-10}	2.05	1.1	200	2.85x10^{-11}	3.21	1.50	2	3.39x10^{-13}	3.23	2.2
400	4.02x10^{-10}	2.15	1.1	250	8.66x10^{-11}	2.98	1.30	3	4.88x10^{-13}	4.30	2.2

one in all the three series of films establishing a non-linear relation between resistivity and temperature in the temperature range 125K to 300K, which is contrary to the linear behavior observed previously in other metallic films in this range of temperature. The coefficient A is found to be in the range 10^{-14} to 10^{-18} (Ωm) for series (i), 10^{-10} to 10^{-25} (Ωm) for series (ii) and 10^{-10} to 10^{-18} (Ωm) for series (iii) films respectively. The coefficient C is found to be in the range of 10^{-10} to 10^{-11} (Ωm) for series (i), 10^{-10} to 10^{-11} (Ωm) for series (ii) and 10^{-10} to 10^{-13} (Ωm) for series (iii) films respectively. The coefficients B and D are found to be of the order of 10^{-7} (ΩmK^{-n}) for all the films. The exponent n is found to be much greater than one.

Using the best fit values of n and m the plots of resistivity versus T^n or T^m as the case may be, were plotted. For example, using the best fit n value of 3.5, the plot of resistivity versus $T^{3.5}$ in the temperature range 80 K to 125 K for a film of thickness 200 nm belonging to series (i) is shown in Figure 4 (a). Similarly for the same film, using the best fit m value of 1.2, the plot of resistivity versus $T^{1.2}$ in the temperature range 125 K to 300K is shown in Figure 4 (b). In both the Figures 4(a) and 4(b), the solid lines drawn are the least square linear lines. Similar plots for one film from each of the series (ii) and series (iii) are shown in Figures 5 & 6 respectively.

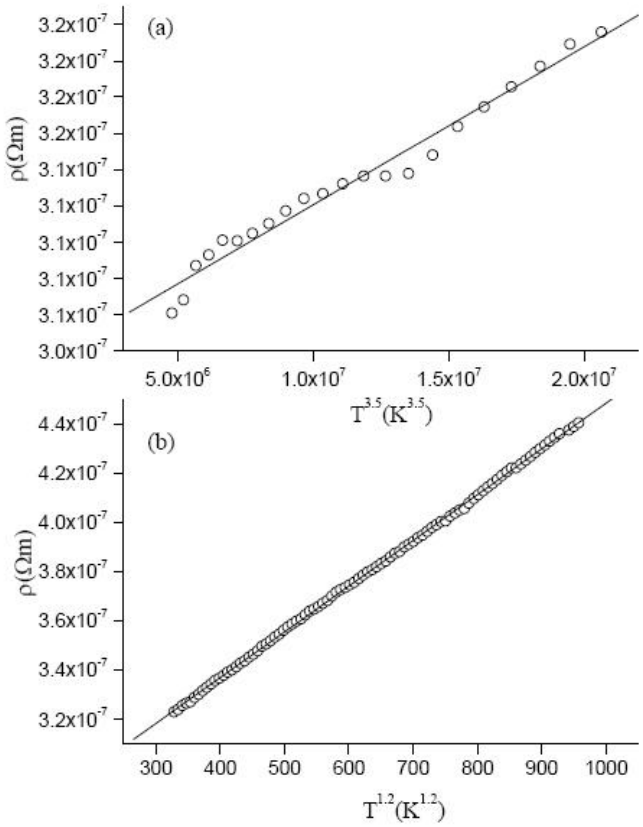

Figure 4. (a) The resistivity, ρ, versus $T^{3.3}$ for the temperature range from 80K to 125K. (b) The resistivity, ρ, versus $T^{1.2}$ for the temperature range from 125K to 300K for a film of thickness 200 nm in series (i). The solid lines are the least square linear lines.

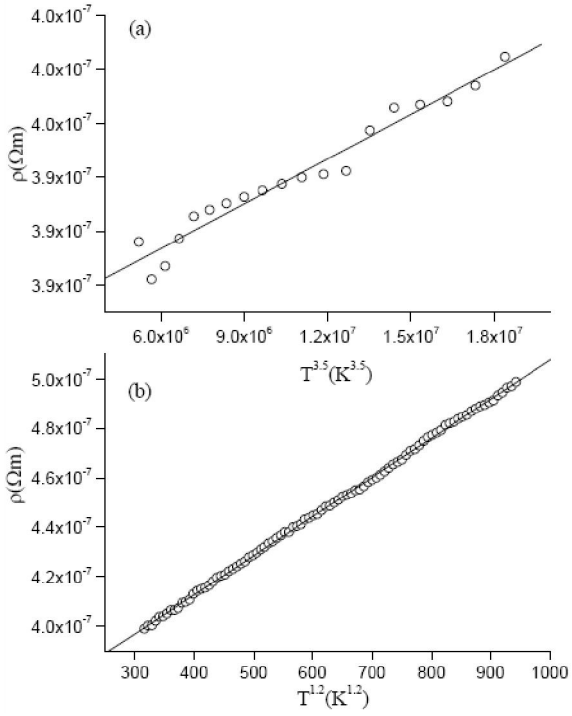

Figure 5. (a) The resistivity, ρ, versus $T^{3.5}$ for the temperature range from 80K to 125K. (b) The resistivity, ρ, versus $T^{1.2}$ for the temperature range from 125K to 300K for a film deposited at the substrate temperature of 150 ^{0}C in series (ii). The solid lines are the least square linear lines.

In the low temperature range above 20K or so, the electron-phonon s-d scattering[14,15] T^3 term called Block-Wilson term begins to dominate over the electron-magnon T^2 term. In our samples, the exponent n is between 2 and 5 for most of the samples. These large n values indicate smooth interfaces between the grains in the films which yielded reduced electron-phonon scattering. White and woods observed[16] n value of 3.3 in bulk Fe and concluded that the interfaces between the grains got smoothened and larger grains have been formed at those temperatures. At higher temperatures ie, between 125K and 300K, linear relation between resistivity and temperature is expected. However, the present m values deviate from unity. This may be due to the magnetic contribution. In this high temperature range, there is a possibility of an additional inter-band s-d electron-magnon scattering where, the s electrons scatter by magnons in to d band holes.

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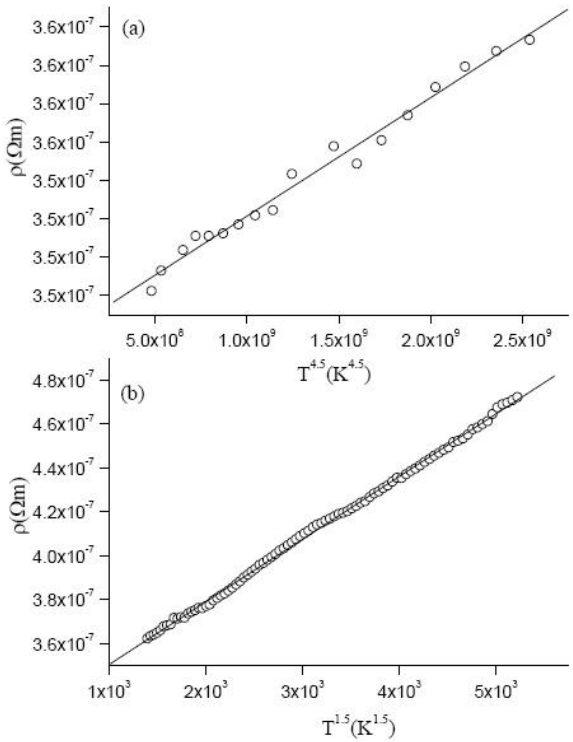

Figure 6 (a) The resistivity, ρ, versus $T^{4.5}$ for the temperature range from 80K to 125K. (b) The resistivity, ρ, versus $T^{1.5}$ for the temperature range from 125K to 300K for a film deposited at a rate of 1 (Å/s) in series (iii). The solid lines are the least square linear lines.

CONCLUSIONS

Three series of Fe films deposited under high vacuum conditions were investigated for resistivity in the temperature range 80 K 300K. The temperature coefficient of resistance and residual resistance ratio were determined. The data has been analyzed in terms of thickness, substrate temperature and rate of deposition.

The electron mean free paths were determined by applying F-S theory to resistivity versus thickness data.

The decrease of resistivity with increasing substrate temperature is attributed to the formation of larger crystallites with increase of substrate temperature.

Under the present vacuum conditions, the deposition rate of 2 Å/s is observed to be the optimum rate at which the iron films of minimum resistivity can be produced.

Temperature dependence of resistivity has been thoroughly analyzed and from that it is noted that in the temperature range from 80K to 125K, a functional relation of the type, $\rho(T) = (A + B T^{n})$ holds with n grater than 3. For the temperature range 125K to 300K a functional relation of the type, $\rho(T) = (A + B T^{m})$ holds with m just grater than unity.

It is for the first time that iron films deposited by electron beam gun method were investigated for low temperature resistivity and the data subjected to thorough analysis in terms of thickness, rate of deposition, substrate temperature and empirical relations between resistivity and temperature are established.

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