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Models, Methods and Measurements of Thin Films Resistivities of Ni/Cu Bi layers and Ni/Pd/Cu Tri layers Films.

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Abstract: In this study we compared resistivity measurements ρ_I and film thicknesses obtained on bi layers Ni/Cu films and tri layers Ni/Pd/Cu films with the values of resistivities and thicknesses calculated according to the model of *Schumann and Gardner applied to a multi-layera system. The experimental and calculated values agree within a few %.*

Keywords: Thin film resistivity, bi layers Ni/Cu films, tri layers Ni/Pd/Cu films, *Schumann and Gardner model*.

1 Introduction

The pratical interest in the study of these materials is the selective hydrogen detection. The general public aspect is that of safety in a context of an increase in the production and use of hydrogen. The PdxNi1-x alloys have a low solubility for hydrogen and have the characteristic being far from oxydable and can used in a broad range of temperatures and hydrogen pressures. This solubility can be controlled by the adjustment of the film composition. These materials whose function is that of a sensor can be used in the form of thin films or into multi-layer deposited on an insulating substrate (α-alumina, glass). In the multi-layer version the functionality of the sensors will depend on the physicochemical modifications of the layer of palladium buried. The reactivity of the system will be followed by the modification of its electric properties to the active interfaces produced by the controlled insertion of atomic hydrogen. The samples studied are nickel, copper, palladium films, bi layers Ni/Cu films, and tri layers Ni/Pd/Cu films.

2 Preparations of Thin Films

2.1 Operational Mode for Obtaining Pure Metal Films

The thin films are prepared by cathode sputtering (RF) Under vacuum ($1.3*10^{-4}$ Pa) in the presence of a low pressure of argon-U [1]. The pure metal films are deposited on square plates of 35 mm side of glass-Corning-1737 and 1 mm thickness in the shape of a disc of diameter equal to 30.5 mm.

The deposit speeds of nickel, copper and palladium films are respectively: VNi = 1.94 Å.s^{-1}, VCu = 3.54 Å.s^{-1}, VPd= 7.11 Å.s^{-1}.

2.2 Thickness Measurements Films

Thickness measurements are obtained using an interference microscope.

The thicknesses of films are deferred on the Table-1.

Table1: Thicknesses of films of copper, nickel and Ni/Cu bi layers.

Thicknesses of copper films (nm)	Thicknesses of nickel films (nm)	Thicknesses of the bilayered Ni/Cu (nm)
$h_{1Cu} = 53 \pm 5$	$h_{1Ni} = 24 \pm 4$	$h_{1Ni/1Cu} = 24/53$
$h_{2Cu} = 85 \pm 5$	$h_{2Ni} = 70 \pm 4$	$h_{2Ni/2Cu} = 70/85$
$h_{3Cu} = 170 \pm 10$	$h_{3Ni} = 140 \pm 4$	$h_{3Ni/3Cu} = 140/170$

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3 Principles of Electric Measurements of Thin Film Properties and Model of Calculations of the Resistivity

3.1 Procedure of Measurement of an Electric Resistance by the Method of the four collinear Probes

The principle of the method consists in putting the four tips in ohmic contact with the surface of the sample; to impose for a short time a constant current via the two external tips, and to measure after a delay the potential difference between the two inner tips. The four tips are in straight line and separated from the same distance s (s = 2.54 mm). The pressure applied at the end of the four retractable tips is controlled using a micrometric screw.

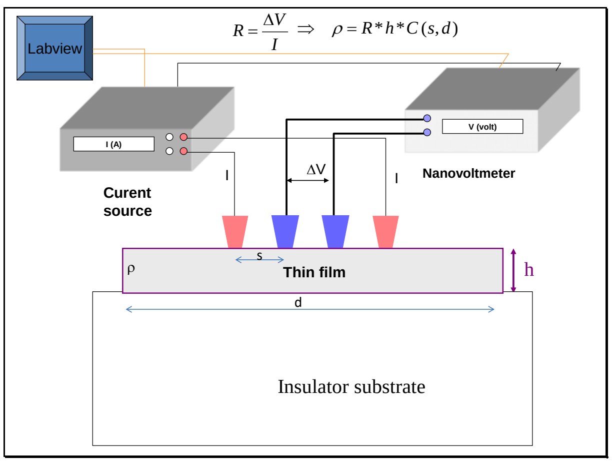

Fig. 1: General diagram of the measurement technique of the resistance of a thin film, in the case of simple layers and multi-layers.

To provide in D.C. current (I) the two external probes we used a gauged source KEITHLEY-6220 and to measure the potential difference between the two internal probes (ΔV). Fig-1 we used a nanovoltmeter KEITHLEY-2182A.
Each test of measurement is carried out at the ambient temperature (T = 293 K) by imposing automatically a sweeping of pulses of current, of increasing values varying in the interval, I = - 2 mA to I = 2 mA. The duration of one pulse of current is equal to 40 ms. Measurements of the tensions between the two inner probes are taken after a delay of approximately 36 ms. Resistance R of the film between the two inner probes is given by the Ohm law,

$$\Delta V = R.I.$$

3.2 Model of Calculation of the Resistivity of a Uniform Layer

In the case of a homogeneous film deposited on an insulating substrate, the calculation of the resistivity is deduced from the electric resistance measured by applying the model developed by Smits [1].
In the case of thin films of thickness h, the resistivity of a sample whose shape is a disc of diameter d Fig.-1, is connected to the resistance R by the relation of Smits:

$$\rho = R.h.C \qquad (1)$$

C is the geometrical factor of correction given by the relation (2):

$$C = \frac{\pi}{\ln 2 + \ln\left(\dfrac{\left(\dfrac{d}{s}\right)^2 + 3}{\left(\dfrac{d}{s}\right)^2 - 3}\right)} \qquad (2)$$

The resistivities measurements are reported in

Figures-2a-2b-2c

3.3 Model of Schumann and Gardner, [2,3] to Determine the Surface Resistance of a Multi-Layer System Deposited on an Insulating Substrate

Historically this model was developed in order to evaluate the surface resistance of a layer of diffusion obtained according to the planar process. The layer of diffusion is single-phase and presents a gradient of resistivity according to the thickness.

We transposed this model to the case of Ni/Cu bi layers and we extracted, starting from the surface resistance RS, the intrinsic resistivities ρi and the thicknesses hi of the two studied layers in Ni/Cu bi layers.

In this model the system is divided into N layers Fig.-3, the electric potential at a point M(r, z) of the $N^{ième}$ layer was calculated in cylindrical co-ordinates. In the reference system having the extremity of the probe source as origin and the free surface of the first layer as plane (x, y), each point of the system is defined by its distance r to the source and by depth z. The end of the probe of current is of finite size having, with the first layer, a circular surface of contact of radius a.

The potential in a point of $N^{ième}$ layer is given by the expression (3):

$$V_n(r,z) = \int_0^\infty \theta_n(\lambda) J_0(\lambda r) \exp^{(-\lambda z)}\, d\lambda + \int_0^\infty \psi_n(\lambda) J_0(\lambda r) \exp^{(\lambda z)}\, d\lambda \qquad (3)$$

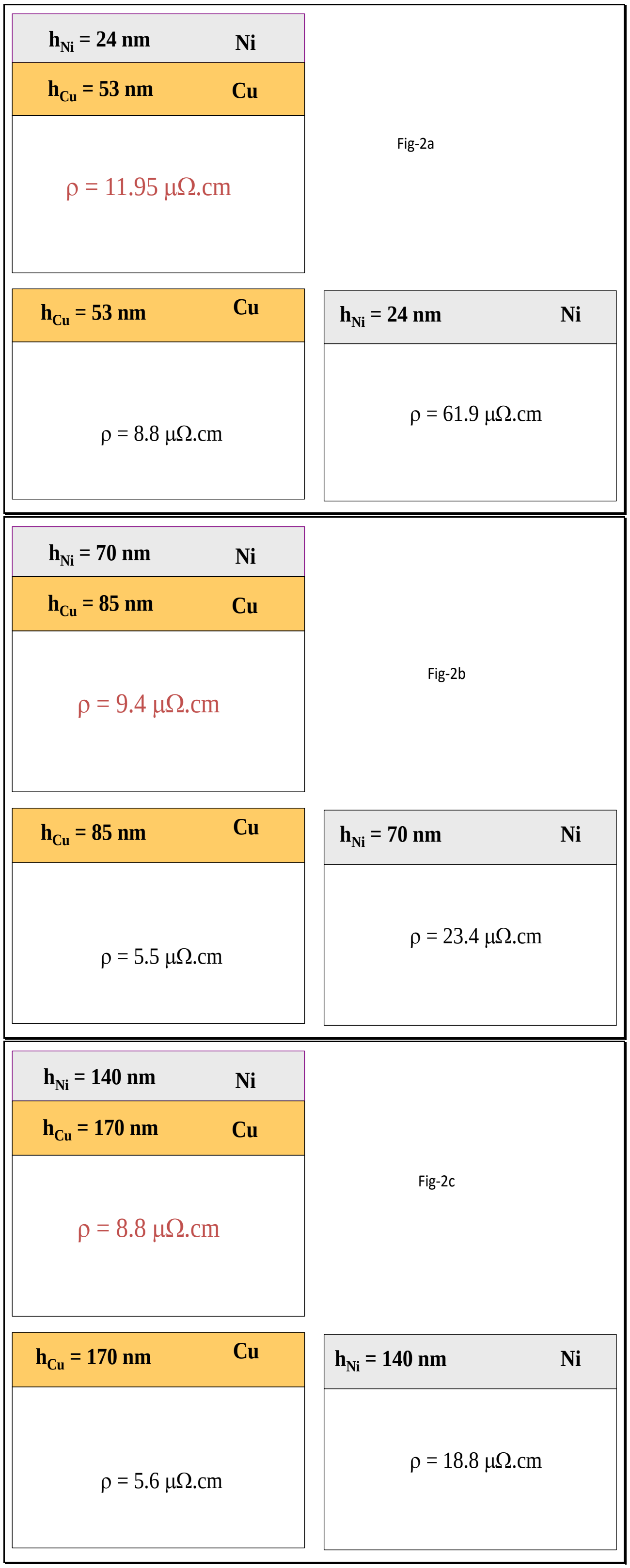

Fig. 2a-2b-2c: Resistivities and thicknesses of Ni/Cu bilayers films, and of Ni and Cu films obtained by experiment.

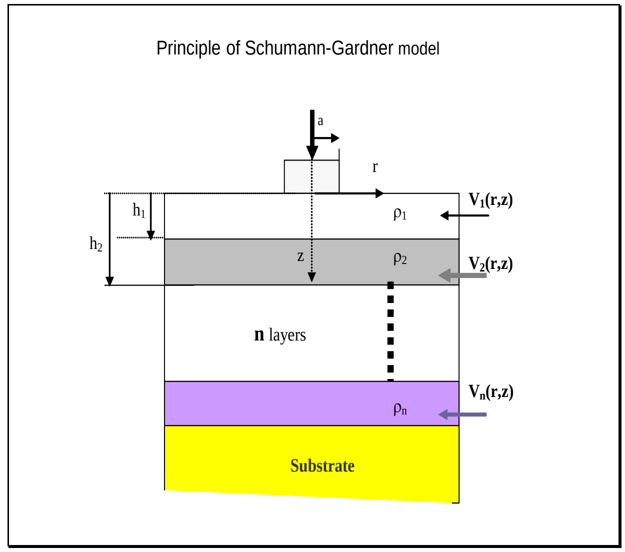

Fig. 3: Parameters characteristic of multi-layer in the model of Schumann-Gardner (S-G).

J0(λr) represents the function of Bessel of first kind of order zero,ψn(λ) and θn(λ) two functions which depend on the boundary conditions and the initial conditions (the number of layers, the thicknesses hi of the layers, the resistivities ρi of the various layers, the function of form of the distribution of the density of current at the end of each probe of current).

In the case of Ni/Cu bi layers the potentials at the two points of measurements at the free surface located respectively at the distances r = s and r = 2s of the ends of the probes of current are given by the expressions (4a-4b):

$$V_1(r=s,z=0)=\frac{I_0\rho_1}{2\pi a}\int_0^\infty \frac{(1+2\theta_1(\lambda))\sin(\lambda a)(J_0(\lambda s))}{\lambda}d\lambda$$

$$V_1(r=2s,z=0)=\frac{I_0\rho_1}{2\pi a}\int_0^\infty \frac{(1+2\theta_1(\lambda))\sin(\lambda a)(J_0(2\lambda s))}{\lambda}d\lambda$$

(4a, 4b)

The expression of the function $\theta_1(\lambda)$ calculated from S-G model, as well as the parameters defining the bi layers are given in the figure-4. With, I_0 the intensity of the current delivered by each of the two sources ρ_1 the resistivity of the first layer of nickel located at the depth z = 0, ρ_2 the resistivity of the second layer of copper, ρ_3 the resistivity of the substrate ($\rho_3 = \infty$), in case of bi layers Ni/Cu films, h_1 and h_2, are respectively the depths of the internal interfaces limiting the two layers, a = 30 µm, the radius of probe contact for z = 0.

The potential difference measured between the two inner probes is given by the relations (5, 6):

$$\Delta V = 2(V_1(s,0) - V_1(2s,0)), \quad (5)$$

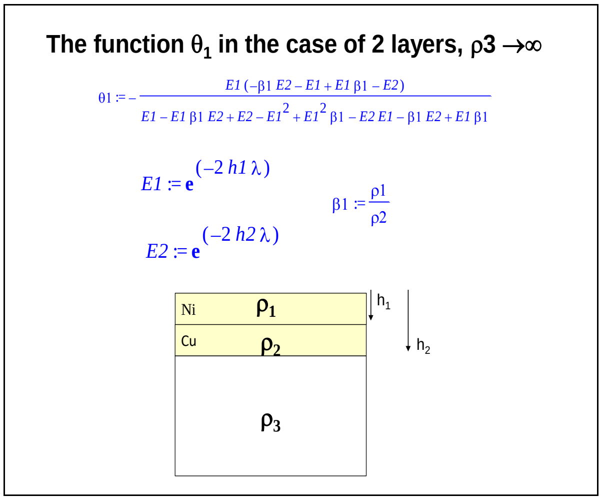

Fig. 4: Expression of the functionθ1(λ) relating to Ni/Cu bi layers according to the S-Gmodel.

$$\Delta V = \frac{I_0 \rho_1}{\pi a} \int_0^{\infty} \frac{\sin(\lambda a)(1 + 2\theta_1(\lambda))(J_0(\lambda s) - J_0(2\lambda s))}{\lambda} d\lambda \quad (6)$$

The electric resistance of surface RS between these two probes is equal t

$$R_S = \frac{\Delta V}{I_0} \quad (7)$$

To calculate the integral numerically *(6)* we used the method of approximation of Simpson, using Maple software. The calculation of the adjustment between the calculated resistance (Rs- calculated) and experimental resistance (Rs-experimental) is considered as satisfactory when:

$$\left| \frac{R_{S-calculated} - R_{S-\text{experimental}}}{R_{S-\text{experimental}}} \right| * 100 \leq 0.5\% \quad (8)$$

3.4 Adjustment of the Resistivities (ρi) and the Thicknesses (hi) of Ni/Cu Bi Layers According to the Model of Schumann and Gardner (S-G).

From electric resistances RS measured for the free face of the bi layers, and by applying model S-G, we calculated the intrinsic resistivities ρi and the thicknesses hi of the various layers composing the Ni/Cu bi layers. The results of these adjustments corresponding to the three bi layers studied are given in Table-2. The agreement between the experimental values h exp, ρ exp those calculated ρi, hi is a few %.

Table. 2: Experiment and calculated of adjustments ρi, hiRSof Ni/Cu bi layers.

Experimental parameters of the three bilayers Ni/Cu/Glass	h_{1Cu} = 53 nm ρ_{1Cu} = 8.8 μΩ.cm h_{1Ni} = 24 nm ρ_{1Ni} = 61.9 μΩ.cm R_{S1} = 0.3666 Ω $\rho_{Bi1.EXP}$ = 12.05 μΩ.cm	h_{2Cu} = 85 5 nm ρ_{2Cu} = 4.57 μΩ.cm h_{2Ni} = 70 5 nm ρ_{2Ni} = 22.5 μΩ.cm R_{S2} = 0.142 Ω $\rho_{Bi2.EXP}$ = 9.40 μΩ.cm	h_{3Cu} = 170 nm ρ_{3Cu} = 5.6 μΩ.cm h_{3Ni} = 140 nm ρ_{3Ni} = 18.8 μΩ.cm R_{S3} = 0.0658 Ω $\rho_{Bi3.EXP}$ = 8.71 μΩ.cm
Parameters calculated of bilayers Ni/Cu/glass h_{Cu}, ρ_{Cu}, h_{Ni}, ρ_{Ni}, R_{cal} $\rho_{Bi1.CAL}$	h_{Cu} = 49.5 nm (7%) ρ_{Cu} = 8.7 μΩ.cm (1%) h_{Ni} = 22.5 nm (6%) ρ_{Ni} = 60 μΩ.cm (3%) R_{cal} = 0.3658 Ω (0.2%) $\rho_{Bi1.CAL}$ = 11.25 μΩ.cm (7%)	h_{Cu} = 82 nm (4%) ρ_{Cu} = 5.5 μΩ.cm (20%) h_{Ni} = 69 nm (1%) ρ_{Ni} = 23. μΩ.cm (2%) R_{cal} = 0.1426 Ω (0.4%) $\rho_{Bi2.CAL}$ = 9.19 μΩ.cm (2%)	h_{Cu} = 168 nm (1%) ρ_{Cu} = 6.3 μΩ.cm (13%) h_{Ni} = 138 nm (2%) ρ_{Ni} = 18.8 μΩ.cm (0%) R_{cal} = 0.0652 Ω (0.9%) $\rho_{Bi3.CAL}$ = 8.52 μΩ.cm (2%)

3.5 Adjustment of the Resistivities ρi and the Thicknesses hi of Ni/Pd/Cu Trilayers According to the Model of Schumann and Gardner (S-G).

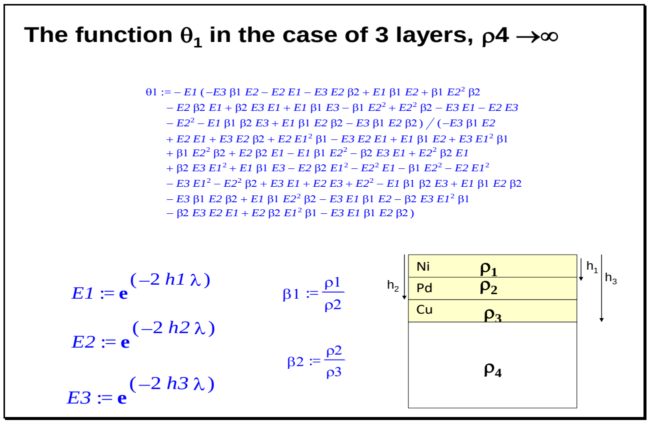

Fig. 5: Expression of the functionθ1(λ) relating to Ni/Pd/Cu tri layers according to the S-G model.

The same technique was used for adjusting the resistivities *ρI and the thicknesses hi of the various layers. In the case of the tri layers Ni/Pd/Cu, the results are given in figure-6 and table-3. The agreement between the experimental and calculated parameters is few %.*

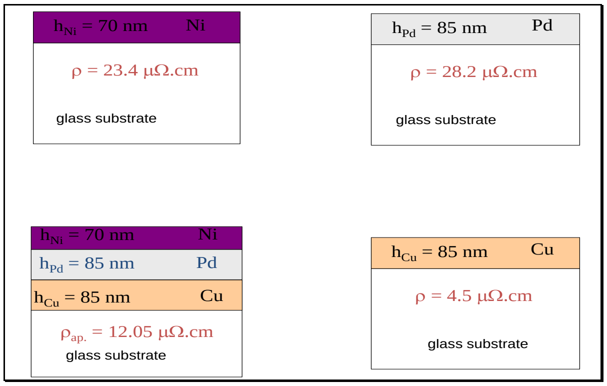

Fig. 6: Experimental resistivities (ρi) and thicknesses (hi) of Ni/Pd/Cu tri layers films, and of Ni, Pd and Cu films.

Table 3: Parameters of adjustments ρi, hi and Rs of tri layers Ni/Pd/Cu film.

Experimental parameters of each of the 3 layers in the trilayers Ni/Pd/Cu film	Thicknesses of pure metal films (nm)		
	h_{Cu} = 85 5	h_{Ni} = 70 5	h_{Pd} = 85 5
	Resistivities o pure metal films (μΩ.cm)		
	ρ_{Cu} = 4 .5	ρ_{Ni} = 23.3	ρ_{Pd} = 28.2
Resistances measured at the surface of the trilayers Ni/Pd/Cu films	$R_{s(experiment)}$ = 0.1176 Ω		
Parameters (h_i, ρ_i) calculated characterizing the trilayers Pd/Ni/Cu films	Thicknesses of films calculated (nm)		
	h_{Cu} = 64 (25%)	h_{Ni} = 72.7 (4%)	h_{Pd} = 95 (12%)
	Resistivities of films calculated (μΩ.cm)		
	ρ_{Cu} = 4 .78 (6%)	ρ_{Ni} = 23.4 (0.4%)	ρ_{Pd} = 30.5 (8%)
Calculations of the Surface resistances of the trilayers Ni/Pd/Cu	$R_{s(Calculeted)}$ = 0.1173 Ω (0.3%)		

4 Conclusions

From the boundary conditions we calculated the functionsθ1(λ) for a system of Ni/Cu bi layers and a system of Ni/Pd/Cu tri layers according to the *Schumann-Gardner* model. *From measurements of surface resistances obtained on bi layers films and tri layers films, we calculated the individual resistivities (ρi) and thickness (hi) of various films composing the studied systems. The agreement between experimental and calculated results is a few %*. In a more general way starting from measurement of the surface resistance of a multi-layer system and knowing the chemical composition of the system one can predict the thicknesses and the resistivities of the layers composing the multi-layer system.

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