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DEPENDENCE OF THE ELECTRICAL CONDUCTIVITY IN GOLD FILMS ON THE MECHANISM OF ELECTRON SCATTERING AT THE SURFACES

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Carrying out measurements of sheet resistivity in polycrystalline gold films we found that their electrical conductivity is higher than in theoretical previsions based on the hypothesis that the scattering of the electrons by the film surfaces is entirely diffuse. For interpreting this behaviour we supposed that the collisions of the carriers against the film boundaries are partially elastic. Since our experiments demonstrate that the film conductivity depends on the characteristics of the surrounding media, we assumed that the fractions of electrons specularly reflected at the film/substrate and film/coating interfaces are different. To evaluate the proportions of carriers elastically scattered we used a method based on the anomalous skin effect. After calculating the conductivity for uncoated gold films deposited on crown glass substrates we compared the results with the experimental data obtaining a satisfactory agreement, which can be improved assuming that the probability of specular reflection increases with the film thickness.

Measuring at room temperature the sheet resistivity of polycrystalline gold films 100–10000 Å thick vacuum deposited by electron bombardment on smooth crown glass substrates at 10^{-6} torr and subsequently annealed at 250°C during 2 hours we found the electrical conductivities given in the last column of Table I.

Each of these data has been obtained by averaging among the results of measurements carried out on several square-shaped samples of equal thickness, which was carefully controlled with a piezoelectric quartz crystal monitor. Comparing the behaviour of specimens evaporated separately we observed that films thicker than 500 Å show a complete reproducibility of the electrical properties, while thinner layers exhibit discrepancies which increase with decreasing thickness. However, even in ultrathin samples 100 Å thick the departures from the mean values listed in the last column of Table I are lower than 5%.

It shall be emphasized that the reproducibility of gold films is much better than that of deposits of less-noble metals vacuum condensed at the same pressure. This may be attributed to the greater susceptibility of these metals to oxidation or adsorption of some other residual gases. Studying the electrical properties of nickel films [1] we noticed that the

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TABLE I

Comparison between calculated and observed values of the electrical conductivity in gold films at room temperature

Thickness (Å)	Electrical conductivity (10^5 mho/cm)		
	Theoretical values		Experimental values
	Completely diffuse reflection	Partially specular reflection	
100	1.54	2.15	0.78
200	1.83	2.56	1.65
400	2.86	3.16	2.94
600	3.36	3.56	3.48
800	3.61	3.75	3.73
1000	3.75	3.87	3.91
1500	3.95	4.03	4.10
2000	4.05	4.11	4.16
2500	4.11	4.16	4.21
3000	4.15	4.19	4.25
4000	4.20	4.23	4.30
5000	4.23	4.25	4.33
7500	4.27	4.29	4.35
10000	4.29	4.30	4.35

attainment of a reproducibility like that of gold layers evaporated in a vacuum of only 10^{-6} torr requires a pressure of the order of 10^{-9} torr.

To interpret the experimental results shown in Table I we expressed the conductivity of the film, σ_f, *versus* its thickness, t, by the known formula:

$$\sigma_f = \sigma_b[1-3a/8t+(3a/2t)\int_1^{\infty}(1/s^3-1/s^5)\exp(-ts/a)ds] \tag{1}$$

derived by Sondheimer [2] from Boltzmann distribution equation on the assumptions that the Fermi surface is spherical and the scattering of the electrons by the surfaces of the film is completely diffuse. Here σ_b indicates the conductivity of the bulk metal and a the mean free path of the electrons at the Fermi level, which may be represented as a function of the carrier concentration, N, by the relation $a = (\hbar\sigma_b/e^2)\,(3\pi^2/N^2)^{1/3}$, obtaining $a = 365$ Å. Solving Eq. (1) through repeated integrations by parts, and making suitable approximations for large and small values of the ratio t/a, we got the results listed in the second column of Table I. Comparing them with the experimental data we note that ultrathin layers ($t = 100$–200 Å) show a resistivity higher than in theoretical previsions, while thicker deposits behave oppositely. To justify the first disagreement we point out that very thin films can condense as arrays of islands separated by dielectric gaps where the charge transport takes place through a mechanism of electron tunneling [3–4] ruled by the equation $\sigma_i = \sigma_f \exp(-4\pi d\sqrt{2m^*\Phi}/h-e^2/\varepsilon rkT)$, where σ_i is the true conductivity of the island deposit; σ_f the conductivity derived from Eq. (1) which would result if the film were continuous; m^* the effective mass of the electrons; Φ the height of the potential barrier between islands; ε the permittivity of the interposed medium; r the average linear dimension of the islands; d their mean distance. During our measurements on layers 100–200 Å thick we observed both negative temperature coefficients of resistance and deviations from Ohm's law, which fit quite well the hypothesis that the film assumes a discrete island structure. Now it must be explained why the conductivity of films thicker than 200 Å, which should be homogeneous and isotropic, is greater than would be expected on the basis of Eq. (1), in contrast with the results of a previous study on the electrical properties of nickel films [1]. This phenomenon could be interpreted by supposing that the collisions of the carriers against the film boundaries are partially elastic [5–6]. However, in nickel deposits the increase in conductivity due to the specular reflection of the electrons should be overbalanced by the contrary effect associated with the oxidation or the adsorption of some other residual gases. To support this hypothesis we emphasize that, while the temperature coefficients of resistance noticed in gold films are close to that of the bulk metal, those exhibited by nickel layers are by far inferior.

Another surprising result of our measurements is the variation of the conductivity observed by substituting the crown glass substrate with plates of quartz, flint glass and glazed alumina, or by depositing onto the gold film a dielectric coating consisting for instance of MgF_2, SiO or CeO_2. This means that the proportion of electrons specularly reflected at the surfaces of the film depends on the characteristics of the surrounding media, so as to be different at the film/substrate and film/coating or film/air interfaces. To take into consideration all these effects we must rewrite Eq. (1) in the modified form [7–9]:

$$\sigma_f = \sigma_b \left\{1 - \frac{3a}{4t} \int_1^{\infty} \left(\frac{1}{s^3} - \frac{1}{s^5}\right) \frac{[1-\exp(-ts/a)]\,[2-p_1-p_2+(p_1+p_2-2p_1p_2)\exp(-ts/a)]}{1-p_1p_2\exp(-2ts/a)}\, ds\right\} \tag{2}$$

where the parameters p_1 and p_2 represent the fractions of electrons elastically scattered at the top and bottom surfaces of the film. For thin and thick layers, provided p_1 and p_2 are small, this equation reduces respectively to:

$$\sigma_f = \sigma_b(3t/4a)(1+p_1+p_2)\,[\log(a/t)+0.4228] \qquad (t < a) \tag{3}$$

$$\sigma_f = \sigma_b[1-3\,(2-p_1-p_2)\,a/16t] \qquad (t > a) \tag{4}$$

To use these formulae we shall know the value of the reflection coefficients p_1 and p_2, which may be determined by a method based on the anomalous skin effect [10–12]. Let us point out that the anomalous skin effect consists in an abnormally large surface resistance arising at low temperature owing to the absorption of high frequency electromagnetic radiations whose penetration into the conductor is limited to a skin depth range shorter than the electron mean free path. In these conditions only the carriers which travel nearly parallel to the surface, so as to remain within the skin region, may contribute effectively to the charge transport. Therefore the decrease in conduction associated with the radiation absorption allows to calculate the proportion of electrons striking the film boundaries at grazing incidence, *i. e.* the fraction specularly reflected [13], that can be expressed through the equation:

$$p_i = 1+4\lambda_i'/3\pi a - 8\lambda_i' c/3\lambda_i'' v_F \qquad (i = 1;2) \tag{5}$$

Here v_F and c represent respectively the velocity of the electrons at the top of the Fermi distribution, whose value for gold is 1.40×10^8 cm/sec, and the speed of light, while λ_i' and λ_i'' denote two parameters which depend on the real (n_i) and imaginary (k_i) parts of the complex refractive index of the film relative to the coating ($i = 1$) or the subtrate ($i = 2$) according to the expressions:

$$\lambda_i' = \lambda\{(A_i - n_i^2 + k_i^2)/[(A_i - n_i^2 + k_i^2)^2 + 4n_i^2 k_i^2]\}^{1/2} \tag{6}$$

$$\lambda_i'' = \lambda(A_i - n_i^2 + k_i^2)/2n_i k_i \tag{7}$$

where λ is the wavelength of a monochromatic radiation in the spectral region of the anomalous skin effect (preferably in the near infrared), while A_i is a suitable constant. Developing the calculations for uncoated gold films condensed on crown glass substrates, and considering waves with $\lambda = 1\mu$, for which the optical indices at the film/air and film/substrate interfaces have the values: $n_1 = 0.24$; $k_1 = 28$; $n_2 = 0.16$; $k_2 = 18.67$, from eqs. (6) and (7) we derived: $\lambda_1' = 5.82\times 10^{-2}\mu$; $\lambda_1'' = 21.93\ \mu$; $\lambda_2' = 7.89\times 10^{-2}\mu$; $\lambda_2'' = 26.88\ \mu$. On substituting back into Eq. (5) we found: $p_1 = 0.16$; $p_2 = 0.24$. Introducing these data in Eqs (3) and (4) we obtained the conductivities listed in the third column of Table I, which are closer to the experimental results than those associated with completely inelastic collisions. Therefore the hypothesis that the surface scattering is not entirely random agrees rather satisfactorily with our measurements. However, a careful comparison shows that, as the film thickens, its conductivity increases more rapidly than in theoretical previsions based on the specular reflection model. This could mean that the parameters p_1 and p_2, which in our approximation were treated as constants, should increase with the film thickness. But a detailed investigation allows to demonstrate that the variations of p_1 and p_2 with t are limited to narrow ranges including $p_1 = 0.16$ and $p_2 = 0.24$, which then may be assumed as reasonable mean values holding for any thickness from 100 Å to 1μ. The thickness dependence of the scattering parameters could be explained by observing that the change of t alters the angular distribution of the electrons striking the surfaces, so as to modify the ratio between diffusely and specularly reflected carriers.

To conclude let us emphasize that the evaluation of the reflection coefficients through a method based on the anomalous skin effect implies that they are quite independent of the frequency of the applied field, so that the values found in the near infrared region can be also used in the limit of low frequency or constant fields.

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