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INFLUENCE OF THE DEFECT DENSITY ON ELECTRICAL RESISTIVITY OF THIN METALLIC FILMS

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It is proposed that the real single metallic films may often be treated as a double-layer film, with an inherent upper layer of higher defect density than that of the base layer. The upper and base layers belong to the same material, differing only in their electronic bulk mean free paths. The corresponding theoretical results were calculated and compared with some experimental data.

ВЛИЯНИЕ ПЛОТНОСТИ ДЕФЕКТОВ НА ЭЛЕКТРИЧЕСКОЕ УДЕЛЬНОЕ СОПРОТИВЛЕНИЕ ТОНКИХ МЕТАЛЛИЧЕСКИХ ПЛЕНОК

В работе предложена двухслойная модель металлической пленки, в которой плотность дефектов в верхнем слое больше, чем в нижнем, причем оба слоя, сделанные из одного и того же материала, отличаются только величиной длины свободного пробега электрона. Проведено срабнение теоретических результатов с экспериментальными данными.

I. INTRODUCTION

The experimental data of electrical resistivity in thin metallic films are commonly analyzed with the aid of the size effect conductivity theory (Fuchs-Sondheimer theory) [1, 2] involving three transport parameters: bulk resistivity ϱ_0 of a material with the same defect density as that of the film, the bulk mean free path l_0 of conduction electrons corresponding to ϱ_0, and the specularity parameter P characterizing the quality of the film surface. The theory takes into account the following assumptions: free electron gas, isotropic and well defined relaxation time of the conduction electrons, spherical Fermi surface, the same value of the mean free path for all film thicknesses, an equal potential along the film surface.

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However, while discussing the validity of the FS theory, one must keep in mind that this theory does not take into account the thickness dependence of the density of defects.

Of course, the density of defects in thin metallic films is an important parameter affecting the resistivity of films. It is clear that if the concentration of point, line and plane defects inside the films is large enough, their contribution to the scattering of conduction electrons is comparable to or prevails over that given by the scattering on the outer surfaces in the FS theory [3—6]. In some annealing studies [7—11], however, there is observed an increase in the film resistivity with decreasing thickness, considerably greater than that according to the FS theory. This discrepancy could be explained by an increase of the concentration of structural defects (e.g. vacancies) in the film. For instance, it was found that for gold [12] and ytterbium [13] films of a thickness of about 100 nm the resistivity is 10 % and 54 % greater, respectively, than the usual bulk value at room temperature. This was attributed to inherent film defects.

The aim of the present paper is to show that some experimental results with single metallic films can be explained using the assumption that the single layer can be imagined as two layers of the same material differing by their density of defects. We assume that the upper layer is much thinner than the base layer. Therefore we call the defects in the outer layer „surface defects“. The dependence of highly concentrated surface defects on the electrical resistivity of thin metallic films represents information which is useful for studying both adsorption or absorption phenomena and catalytic reactions. In [3] it was shown that the oxygen content in e-gun evaporated thin tungsten films is greater than that of rf sputtered ones, which is the reason why the evaporated films have a greater resistivity. Wilson and Sinha [14] applied the impurity scattering theory of Kuzmenko [15] to the conductivity of copper films deposited in argon at 1.33×10^{-4} Pa in an attempt to estimate the number of trapped argon atoms.

II. DOUBLE-LAYER MODEL

The large discrepancy between the theoretical FS and experimental results [4, 13, 16, 17] for thin metallic films indicates that scattering mechanisms other than the diffuse scattering on the film surfaces are responsible for the large resistivity of the films. Moreover, the electron scattering by grain boundaries need not play the major role, since the electron mean free path is often smaller than the average grain diameter in a metal. The remaining mechanisms for explaining the large film resistivity is the diffuse scattering of conduction electrons by surface defects. Annealing and aging of the films may result in a partial removal of these defects, thereby substantially decreasing the resistivity. It is not unreasonable to assume that thinner films contain larger average densities of defects than thicker films.

Let us consider a typical film with surface defects, vacancies or displaced atoms, and physically adsorbed impurity atoms [18, 19]. Thus, we consider the film with the thickness $a+b$ consisting of two layers of the same metal (surfaces and interface lying in the planes $z=0$, $z=a+b$, $z=a$, respectively): the base layer with thickness a, conductivity σ_1, electron mean free path l_1, concentration of defects n_1; the upper layer with thickness b, conductivity σ_2, electron mean free path l_2, concentration of defects n_2, with $a>b$, $n_2>n_1$, $l_1>l_2$. The bulk conductivities of the layers are given as

$$\sigma_{ib}=\frac{Ne^2l_i}{mv_F}, \qquad i=1,2,$$

where N is the number of electrons per unit volume, e is the electron charge, m is the electron effective mass, l_1, l_2 are mean free path lengths of the conduction electrons, and v_F is the velocity of an electron at the Fermi surface.

We must keep in mind that the two layers belong to the same metal; therefore they have equal effective masses, Fermi velocities, and densities of conduction electrons. The surface scattering of conduction electrons on the outer surfaces of the film is characterized by the specularity parameter P. We shall consider only the case when $P=0$ (diffuse surface scattering). There is no potential step at the interface between the layers so that there is no additional scattering occurring on the plane $z=a$. In our further considerations we shall use in fact the double-layer model proposed by Lucas [20]. It should be pointed out that there are special investigations described in literature [20—22] devoted to the superimposition of a metallic layer onto a metallic base film. We assume, in contradiction to these papers, that even the real single metallic film may be treated as a double-layer film with an inherent upper layer of higher defect concentration than that of the base film. The density of defects as well as the thickness of the upper layer depend on how the film was prepared.

An expression for the ratio of the total film (i.e. double-layer) conductivity to the conductivity of the bulk material of the base layer 1 is (c.f. [20]):

$$\begin{aligned}\frac{\sigma_f}{\sigma_{1b}}=\frac{1}{(a+b)l_1}\Big\{ & al_1+bl_2+\frac{3}{2}\int_0^1 \mathrm{d}x(x-x^3)Dl_1l_2(1-A)(1-B)(1+PA)(1+PB)- \\ & -\frac{3}{4}\int_0^1 \mathrm{d}x(x-x^3)\,D(1-P)\,[l_1^2\,(1-A)(1+PAB^2)+l_2^2\,(1-B)(1+PA^2B)]- \\ & -\frac{3}{4}\int_0^1 \mathrm{d}x\,(x-x^3)\,D[l_1^2(1-A)(1+PA)(1-PB^2)+l_2^2(1-B)(1+PB)(1-PA^2)]\Big\}\end{aligned} \tag{1}$$

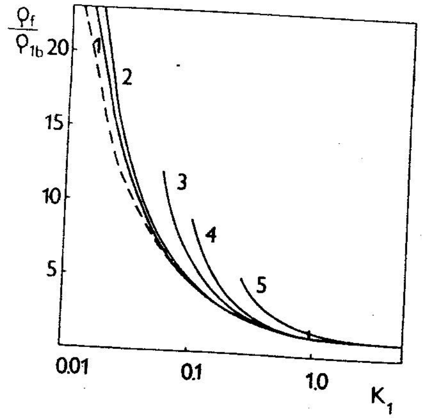

Fig. 1. The film resistivity vs the reduced thickness $K_1 = (a+b/l_1$ for the mean free path ratio $l_2/l_1 = 0.5$. Note: 1, 2, ..., 5 refer to upper layer thickness b/l_1 = 0.005, 0.01. 0.05. 0.1, 0.25, respectively; broken line denotes the single film ($b/l_1 = 0$).

where $D=(1-P^2A^2B^2)^{-1}$, $A=\exp(-a/l_1x)$, $B=\exp(-b/l_2x)$, $x=\cos\Theta$, and Θ is the angle between the incident conduction electron and the normal to the film surface. Note that the bulk material which contains a natural density of defects in layer 1 is exactly the bulk material from which the film was prepared.

III. RESULTS AND DISCUSSION

The dependence of the resistivity ratio ϱ_f/ϱ_{1b} (inverse eq. (1)) on the reduced thickness K_1 (film thickness $a+b$ over the mean free of conduction electron in the base layer 1) for various values of thickness b of the upper layer and some values of the ratio l_2/l_1 was calculated numerically for the diffuse surface scattering ($P=0$). The results as well as the theoretical FS single film curve are drawn in Figs 1—4. The figures show that the greater the upper layer thickenss is, the greater is the resistivity of the film (the total thickness $a+b$ being constant). As expected, for higher density of defects (smaller mean free path l_2) in the upper (surface) layer the shift from the FS curve becomes greater.

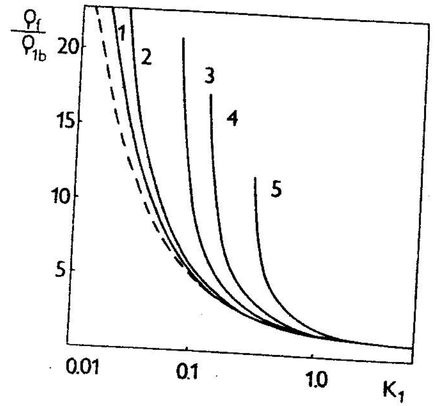

Fig. 2. The film resistivity vs the reduced thickness $K_1 = (a+b)/l_1$ for the mean free path ratio $l_2/l_1 = 0.2$. Notation is the same as in Fig. 1.

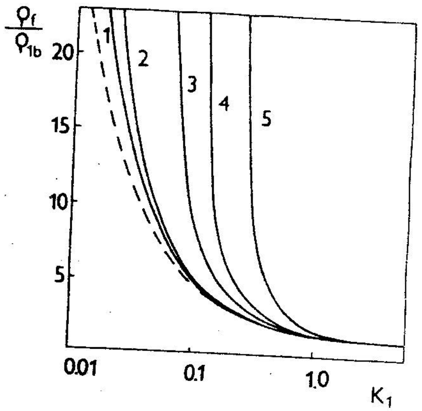

Fig. 3. The film resistivity vs the reduced thickness $K_1 = (a+b)/l_1$ for the mean free path ratio $l_2/l_1 =$ 0.1. Notation is the same as in Fig. 1.

Bist and **Srivastava** [13] studied the electrical resistivity of ytterbium thin film at room temperature. Their experimental results do not lie on the theoretical curve calculated according to the FS (diffuse surface scattering, bulk mean free path about 50 nm). However, applying our model to these experimental data we can find a theoretical curve fitting well the experimental results, assuming the ytterbium film as a double-layer structure, the base layer with the bulk mean path of the conduction electrons of about 50 nm, and the upper layer with the thickness 12.5 nm and the bulk mean free path of the conduction electrons of about 10 nm (Fig. 5). Our model can be successfully applied also to some other experimental data, e.g. [4, 16, 23], where a difference between the FS theoretical curve and the experimental data appeared.

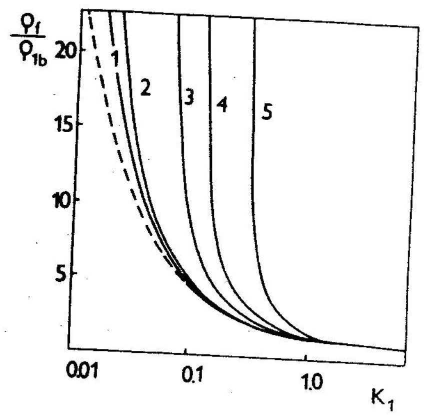

Fig. 4. The film resistivity vs the reduced thickness $K_1 = (a+b)/l_1$ for the mean free path ratio $l_2/l_1 =$ 0.05. Notation is the same as in Fig. 1.

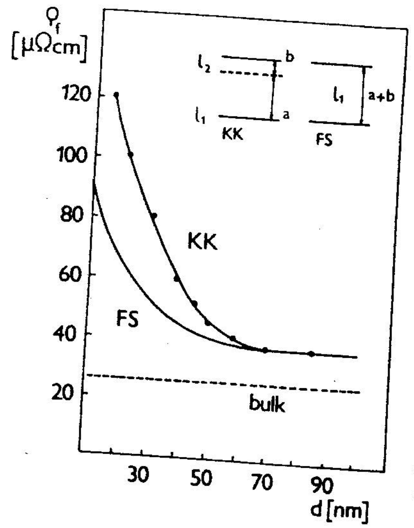

Fig. 5. Thickness dependence of the electrical resistivity of polycrystalline ytterbium films (see [13] for experimental data denoted by dots). Parameters: $d=a+b$, $b=12.5$ nm, $l_1=50$ nm, $l_2=10$ nm. FS – Fuchs-Sondheimer curve, KK – our fit.

IV. CONCLUSION

The Fuchs-Sondheimer conductivity theory is sometimes found to be unsatisfactory for the interpretation of experimental electrical resistivity/conductivity data in a thin metallic film consisting of one-material single layer. In many cases this is due to the non-homogeneity of the films, i.e. the presence of some significant concentration of defects. We suggest that in the case when most defects lie close to the upper surface, the model of the film consisting of two layers, with a higher density of defects in a thin "surface" layer, describes the reality better. This is confirmed by the numerical calculation of resistivity using the electron mean free path and thickness of the upper layer as additional parameters. Taking the values of these parameters in accord with the film history, the experimental data can be well fitted.

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