

# THICKNESS DEPENDENCE OF THE RESIDUAL RESISTIVITY AND TRANSPORT PROPERTIES OF SUPERCONDUCTING TIN FILMS

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## Abstract

We have studied the variation of residual electrical resistivity of superconducting tin films by Joule evaporation on glass substrates at room temperature ( series I ) and cooled down to 150 K ( series II ) and 77 K ( series III ) as a function of their inverse thickness. The extrapolation to infinite thickness allows to determine the residual resistivity corresponding to the bulk materials of the same series. The transport properties are interpreted in the framework of the Fuchs-Sondheimer's theory taking into account the value of the product  $\rho \ell$  given by the free electron model. We have analysed the measured residual resistivity dependence of  $T_c$  in terms of valence effects and Markowitz-Kadanoff's theory.

## 1 INTRODUCTION

In numerous papers devoted to problem of the electrical resistivity ( [ 1 ], [ 2 ] ) much attention was directed into the thickness dependence of electrical resistivity. It is well known the residual resistivity of thin films increases when the thickness decreases [ 3 ].

The corresponding interpretation deals mostly with scaling models [ 4 ], the resistivity in the polycrystalline films is strongly dominated by grain boundary scattering ( the crystallite size increased with film thickness and decreased with substrate temperature ), or with the influence of mean free path. [ 5 ] obtained from Fuch's theory. In this paper we have analyzed the measured the thickness dependence of the residual resistivity of pure tin in terms Fuchs-Sondeheimer theory [ 6 ]. Further extrapolation to infinite thickness gives the bulk material residual resistivity. On the other hand, we have studied and evaluated the influence on  $T_c$  of the residual resistivity by using Markowitz-Kadanoff's theory [ 7 ].

In section 2 we describe the experimental procedures and sample preparations techniques. Section 3 shows that our measurements extrapolate naturally the values of  $\rho_\infty$  in agreement with theory

and previous experiments. In section 3 we discuss in particular the interest of different extrapolation procedures and show that our measurements extrapolate naturally the values of  $\rho_\infty$ . The relevant theoretical predictions are recalled in each section.

## 2. SAMPLE CHARACTERIZATION AND MEASUREMENTS

Tin ( 99,999% purity ) is vacuum-evaporated from a resistively heated crucible under a pressure of about  $10^{-6}$  mmHg . Films are simultaneously deposited on several glass substrates at room temperature. They are then covered with a photoresist layer, light exposed through a photographic mask, and chemically etched. We have studied 3 series of films with thickness from 0,25 to 5  $\mu\text{m}$  ( series I ), 2,50 to 5  $\mu\text{m}$  ( series II ) and 0,25 to 2,50  $\mu\text{m}$  ( series III ).

Each series is characterized by its thickness, residual resistivity, electron mean free path, and critical temperature. The thickness is determined from interferometric and resistive measurements. In the latter case, us is made of the well-known formula



$$d \approx \frac{\rho_{295} \cdot L}{(R_{295} - R_{4,2}) \cdot W} \quad (1)$$

For a film of length L and width W, showing resistances R(295) and R(4,2) à T=295 and 4,2 K, respectively. We have taken  $\rho(295)=11 \times 10^{-6} \Omega\text{-cm}$  as for bulk material [ 8 ]. Both methods given the same results to within 10%.

### 3. EXPERIMENTAL RESULTS

#### 3.1 Residual resistivity and mean free path

The resistance measurement allows the

$$\rho_d \approx \rho(4,2) = \frac{\rho_{295} \cdot R_{4,2}}{R_{295} - R_{4,2}} \quad (2)$$

determination of the residual resistivity

And therefrom the value of the effective electron mean free path  $l_d$  satisfying  $\rho_d l_d = 0,22 \times 10^{-11} \Omega\text{-cm}^2$ . We have taken here the value that is compatible with our measurements of penetration depth and coherence length [ $\rho_d l_d = 4\pi k_B \xi_0 T_c \lambda_L^2(0)/0,18 \hbar^2 c^2$ ] and which coincides with that obtained from a free-electron model calculation with three conduction electrons per atom. The characteristics of our films are given by Fuchs' theory [5]. The residual resistivity  $\rho_d$  of a film of thickness d is evaluated by the Fuchs-Sondheimer description [ 6 ]. Nevertheless, practical application of this theory to extract information about  $l_\infty$  is hindered by the fact that published values of  $\rho_\infty$  differ by factors of up to three ([ 6 ], [ 9 ]). A consistent interpretation of our results is obtained when  $\rho l$  is calculated from the measured superconducting quantities  $T_c$ ,  $\xi_0$ ,  $\lambda_L(0)$ . This gives  $\rho_d l_d = \rho_\infty l_\infty = 0,22 \times 10^{-11} \Omega\text{-cm}^2$ , coinciding with results from a simple free-electron model calculation for Sn. We have used the exact Fuchs' expression to extract values of  $l_\infty$  and  $\rho_\infty$  for each film from the above  $\rho l$  value and the measured resistivities  $\rho_d$ . In the region of thickness and mean free path of our samples ( $l_\infty \sim 3d$ ) Fuchs' expression is very well approximated by the linear equation

With  $\gamma = 0,46$ . The subscript  $\infty$  refers to the bulk material having the same volume properties and texture as the film. In fact, the Fuchs' expression can be linearized over a large range of d values ( strictly speaking when  $d \gg l_\infty$  ) by Eq.(3); if diffuse scattering at the surface is assumed,  $\gamma = 3/8$ , close to the value found for  $\rho_\infty \sim 3d$ , our case.

$$\rho_d = \rho_\infty + \gamma \rho_\infty l_\infty / d \quad (3)$$

Equation (3) is sometimes used [ 3 ] to obtain the product  $\rho l$ . In our case, since  $\rho l$  is a material constant, a plot of  $\rho_d$  as a function of  $d^{-1}$  should thus give for our three series of films three straight lines of the same slope. Actually, as shown in Fig.1, this is not the case. To extract information about the  $\rho l$  value from Eq. (3) it is necessary for  $\rho_\infty$  to remain constant for all values of d considered. In fact, when  $\rho_\infty$  is obtained from the exact Fuchs' expression [ 5 ], we find that it satisfies quite closely a linear expression.

$$\rho_\infty = \rho_b + c/d \quad (4)$$

where  $\rho_b$  is the resistivity of hypothetical bulk material, obtain by extrapolation to  $1/d = 0$ . The values of  $\rho_b$  and c are different for the I, II and III series. The  $\rho_\infty$  thickness dependence can be understood if, as shown by Mayadas et al. [ 4 ], the resistivity in polycrystalline films is strongly dominated by grain boundary scattering

Since the crystallite size increased with film thickness [ 4 ] and decreased with substrate temperature, observation of these grain boundary effects must be expected in our polycrystalline samples. So, differences in the coefficients [ Eq.(4)] between I, II and III series can be ascribed [ 10 ] to the anisotropy of the Fermi surface in Sn, which would make its contribution to the current in the plane of the film dependent on crystallite orientation. Furthermore, the intergrain resistance should increase with misalignment of neighboring grains. Finally, Eqs.(3) and (4) explain why the measured film resistivities can be reasonably well fitted ( Fig.1) by straight lines.

We can then say that each series of films defines by extrapolation to  $1/d = 0$  (Fig.1) a hypothetical bulk material whose transport properties are represented by the values  $\rho_{bI} = 1,3 \times 10^{-7} \Omega\text{-cm}$ ,  $\rho_{bII} = 2,5 \times 10^{-7} \Omega\text{-cm}$  and  $\rho_{bIII} = 4,210^{-7} \Omega\text{-cm}$ .

#### 3.2. Critical temperature

Previous extrapolation procedures can be applied to obtain other bulk parameters. The preceding discussion suggests that the critical temperature  $T_b$  of the bulk material of resistivity  $\rho_b$  characterizing a series of films evaporated under similar conditions can be defined by

$$T_c = T_b + C_1 (\rho_d - \rho_b) \quad (5)$$

where  $T_b$  is the critical temperature of a hypothetical bulk material obtained when  $\rho_d \approx \rho_b$ . In fact, the residual



resistivity  $\rho_b$  of each bulk material is obtained from linear extrapolation toward zero of quantity  $1/d$ . We find that  $T_c$  is decreasing function of the thickness.

Toxen [ 11 ] ascribes such variation to the effect of stress resulting from differential thermal contraction of film and substrate. According to Toxen's prescription [11],  $T_c$  would be given by

$$T_c = T_b + C_2 \left( \frac{1}{d} \right) \quad ( d \text{ in nm } ) \quad (6)$$

Where  $C_2 = \gamma \cdot \rho_b \cdot \mathcal{L}_b \cdot C_1$ , involves the reflection conditions on the surface of sample and grains boundaries and contrary to  $C_1$ , depends upon substrate temperature. In Fig. 2,  $T_c$  is plotted vs.  $\rho_d$  and the values of  $T_b$ ,  $C_1$  and  $C_2$  are shown in Table I, where it can be seen that the critical temperature and mean free path of bulks materials corresponding to each series decrease ( $T_{bIII} < T_{bII} < T_{bI} < T_0$ ) where  $T_0 = 3,722$  [ 3 ], ( $\mathcal{L}_{bIII} < \mathcal{L}_{bII} < \mathcal{L}_{bI}$ ). A similar behaviour has been observed ([12],[13]) in bulks supraconductors weakly doped with impurities. Markowitz and Kadanoff [ 7 ] explain it considering in a pure materials, the electron-electron interaction is anisotropic : the authors show ( if the pairing potential depends upon orientation of the supraconducting electrons moments as compared to cristalligraphic directions) that addition of impurities lead to depression of this anisotropy which induce ( if  $\mathcal{L} \gg \xi_0$  ) a rapid decrease of  $T_c$ . The impurities can produce another's effects ( excepted anisotropy ) whose collective contribution called "valence effects" ( because dominated by valence effect ) induce on contrary an increase ( linear function of  $\rho$  ) of the critical temperature. So, if we consider that the

anisotropy effect is responsible of the behaviour of these three bulks materials I, II and III, the values of the relative variation of the critical temperature obtained by applying the M K theory to tin are 0,35, 1,8 and 3,5%. Experimentally, the global variations of  $T_c$  are 0,24, 0,43 and 0,51% respectively ; the values of  $T_{bII}$  and  $T_{bIII}$  are very strongly influenced by valence effects contrary to value of  $T_{bI}$  which is practically insensitive to them. Thus, the valence effects provide an explanation for discrepancy previously observed between the values obtained by the MK theory and our experimental results. In the same way, the increase of the critical temperature of our films as a function of their resistivity  $\rho_d$  ( Fig. 2 ) can be interpreted in terms of valence effects ( on meaning of MK ) as bulks and isotropic materials.

### 4.CONCLUSION

Measurements of residual resistivity and critical temperature of Sn films condensed on cold substrates show that they can be safely extrapolated to obtain the parameters characteristic of bulk material. However, a careful definition of the condition of such extrapolation is necessary.

Depending on the conditions of evaporation, different series of films correspond to different bulk materials. The transport properties are correctly described by Fuchs-Sondheimer's theory. MK theory and valence effects have made complementary contributions to the understanding the behaviour in terms of temperature of our films.

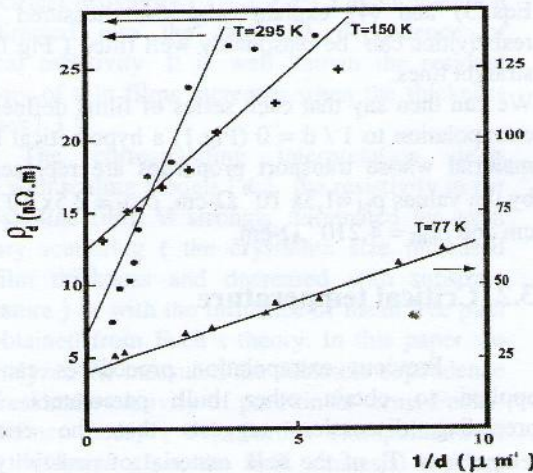


Fig. 1.

Fig. 1. Residual resistivity of the films as a function of inverse thickness.

(●) The I series (films condensed at room temperature),

(◻) the II series (films condensed at 150 K),

(π) the III series (films condensed at 77 K)

The solid lines are the best fits to the data.

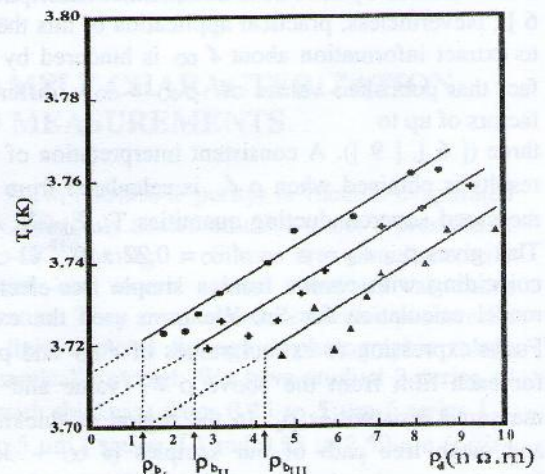


Fig. 2.

Fig. 2. Critical temperature films of series I (●), II (◻) and III (π) as function their residual resistivity.



TABLE I. Characterising parameters of bulk materials corresponding to I, II and III series.

	$\rho_b$ ( $n\Omega \cdot m$ )	$\ell_b$ ( $\mu m$ )	$T_b$ (K)	$C_1$ ( $mK / n\Omega \cdot m$ )	$\gamma$	$C_2$ ( $mK \cdot \mu m$ )
Series I	1,3	0,33	3,713	9,91	0,72	3,06
Series II	2,5	0,17	3,707	9,82	1,39	5,80
Series III	4,2	0,10	3,703	9,25	2,79	10,83

## REFERENCES

1. P.A Badoz, A. Briggs, E. Rosenberg, F. Arnaud d'Avitaya, C. d'Anterrosches, *App. Phys. Lett.* **51**, 169 (1987).
2. G. Fishman, D. Calecki, *Phys. Rev. Lett.* **62**, 1302 (1989).
3. D. C. Larson, in *Physics of Thin Films* (Academic Press, New York, 1971), Vol. 6, p81.
4. A. F. Mayadas, *J. Appl. Phys.* **39**, 4241 (1968).
5. K. Fuchs, *Proc. Camb. Phil. Soc.* **34**, 100 (1938); E.H. Sondheimer, *Adv. Phys.* **1**, 1(1952).
6. A.M. Toxen, M.J. Quinn, *Phys. Rev.* **138A**, 1145 (1965).
7. D. Markowitz and L. P. Kadanoff, *Phys. Rev.* **131**, 563 (1963).
8. G. K. White and S. B. Woods, *Rev. Sci. Instr.* **28**, 638 (1967).
9. K. R. Lyall and J. F. Cochran, *Phys. Rev.* **159**, 517 (1967).
10. B. N. Aleksandrow and M. I. Kaganov, *Zh. Eksp. Teor. Fiz.* **41**, 1333 (1961).
11. A. M. Toxen, *Phys. Rev.* **123**, 442 (1961).
12. G. Chanin, E.A. Lynton and B. Serin, *Phys. Rev.* **144**, 719 (1959).
13. D. P. Seraphim, C. Chiou and D. J. Quin, *Acta Metall.* **9**, 861 (1961).

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