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STRUCTURE AND ELECTRICAL PROPERTIES OF THIN ZINC FILMS+

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Thin zinc films of thicknesses ranging from 15 to 75 nm were deposited with rate 0.25 nm/s onto amorphous glass substrates using thermal evaporation method. Nucleation, growth and structure of the films were investigated by transmission electron microscopy diffraction techniques. The films were of polycrystalline structure. The grain size and density were varied with film thickness. The values of bulk resistivity, mean free path and temperature coefficient of resistance of the films were calculated and correlated with film structure. Other physical data were estimated from the theoretical formula.

1. Introduction

The structure of some transition metallic films, especially with respect to zinc films, have been investigated by Kikuchi et al.[1] using vacuum-arc evaporation. Robertson and Unvala[2] studied very thin layers of zinc and other metals deposited on polished copper and on insulating substrates at low temperatures.

+This is a part of M. Sc. Thesis of H. M. T. Aly earned from Faculty of Science, Al-Azhar University in 1981.

Measurements of the resistance of these layers indicated an irreversible change in resistance on annealing concomitant with the diffraction change. The electrical stabilization of resistant films of sputtered zinc has been shown by Tosser et al.[3].

The aim of the present work is to study the structural changes of thin zinc films and their electrical properties as a function of film thickness.

2. Experimental

The different thicknesses of zinc films in the range from 15 to 75 nm were prepared by thermal evaporation of spec. pure zinc (Zn) of 99.999% under vacuum of 1.33×10^{-3} Pa. The films were deposited onto cleaned amorphous glass substrates with deposition rate of 0.25 nm/s. The substrates were maintained at room temperature. The thickness of the film was measured by multiple beam Fizeau fringes method of Tolansky[4]. The structural investigation* was carried out by using Carl Zeiss *ELMI D2* transmission electron microscope (at 45 KV). The electrical measurements** were done by using potentiometric circuit and the films were contacted with electrodes.

3. Results and discussion

The electron diffraction patterns of Zn films were composed of continuous rings indicating that the structure of these films is polycrystalline with no preferred orientation (See Figs. 1 and 2 for Zn films of thicknesses 30 and 75 nm, respectively). These results agree with the work done by Kikuchi et al.[1] on thin metal films of Zn together with other transition metals using the vacuum arc evaporation method. They pointed out that the electron diffraction patterns of the films of metals with low melting points were composed of relatively sharp Debye rings,

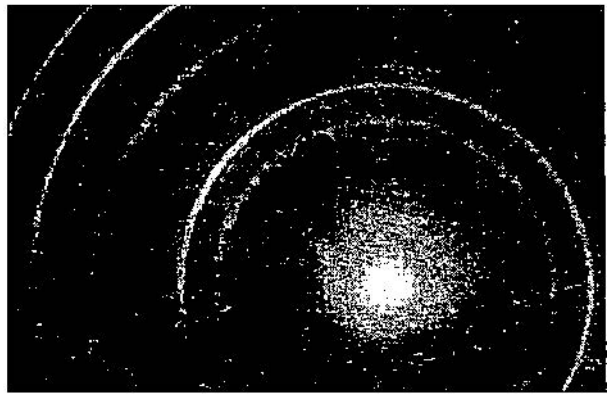

Fig. 1. Electron diffraction pattern of Zn film of thickness 30 nm.

*This part was carried out in the Electron Microscope and Thin Films Laboratory at NRC, Cairo.
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while, the films of metals with high melting points gave structureless image in transmission micrographs and halos in electron diffraction patterns. The halo pattern showed that the structure of the film was amorphous or composed of very small crystallites.

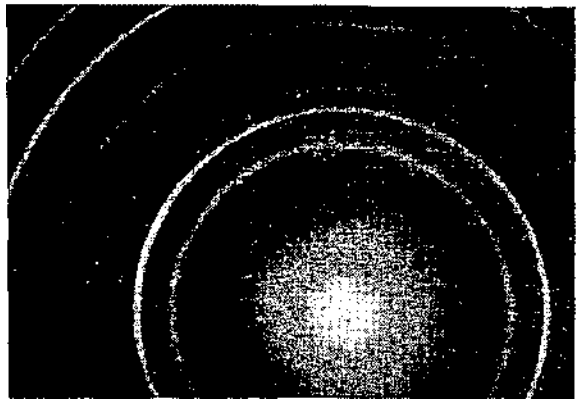

Fig. 2. Electron diffraction pattern of Zn film of thickness 75 nm.

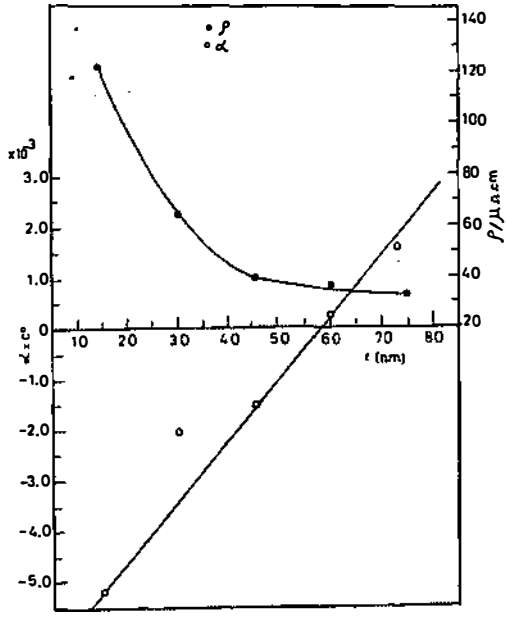

Fig. 3. Relation between resistivity ϱ, temperature coefficient of resistance α and film thickness t.

With respect to the electrical measurements of Zn films, it was found that the resistivity ϱ decreased with increasing film thickness as in the usual behaviour of thin metallic films (See Fig. 3). However, the curve reached a satura-

tion dependence in the thickness range from 60 to 75 nm. Therefore, the intrinsic (bulk) resistivity ϱ_0 and the mean free path of the conduction electrons l_0 were calculated according to the following equation:

$$\varrho/\varrho_0 = 1 + \frac{3(1-P)\,l_0}{8\,t}, \tag{1}$$

where P is the specular parameter varying between 0 and 1, l_0 is the intrinsic mean free path and t is the film thickness (Fuchs[5] and Sondheimer[6]). Consequently, a relation between ϱ and $1/t$ was plotted and shown in Fig. 4. Taking into account that $P = 0$, the values of ϱ_0 and l_0 were found to be 9 $\mu\Omega$ cm and and 509 nm, respectively, which are comparatively greater than the values of bulk Zn (Ashcroft and Mermin[7]).

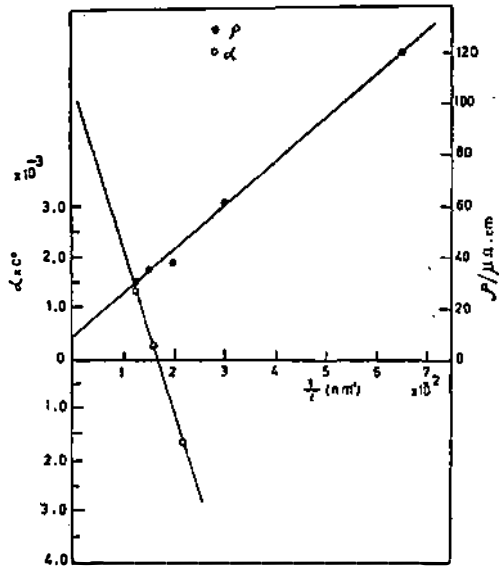

Fig. 4. Relation between resistivity ϱ, temperature coefficient of resistance α and reciprocal of thickness $1/t$.

The resistances of Zn films were measured as functions of heating temperature. It was found that the resistance decreased with increasing temperature for film thicknesses 15, 30 and 45 mm, while for film thicknesses 60 and 75 nm the resistance increased (See Fig. 5a for film thickness 60 nm). From the calculation of temperature coefficient of resistance $\alpha = 1/R \cdot \mathrm{d}R/\mathrm{d}T$, it was found that two regions were exhibited from the relation between α and t as shown in Fig. 3. The first one represents negative values of α for t equal to 15, 30 and 45 nm, while the second region of positive values of α corresponds to the thicknesses 60 and 75 nm. The intrinsic (bulk) values of α_0 was calculated according to Fuchs-Sondheimer equation[5,6]:

$$\alpha = \alpha_0 \left[1 - \frac{3\,l_0\,(1-P)}{8}\right]. \tag{2}$$

By plotting a relation between α and $1/t$, a straight line was obtained as shown in Fig. 4. The values of α_0 and l_0 were found to be 0.0225 °C^{-1} and 308.14 nm, respectively. The value of α_0 is in reasonable agreement with published values. Moreover, the values of l_0 were calculated at each heating temperature. Accordingly, the relations between ϱt and t were plotted and the values of l_0 were estimated using equation (1). Fig. 6 shows one of these relations at heating tempe-

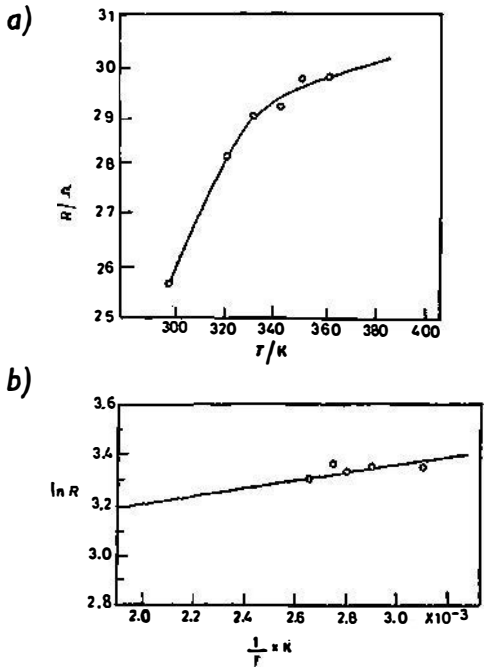

Fig. 5a. Relation between resistance R and temperature T for Zn film of thickness 60 nm.
Fig. 5b. Relation between ln R and $1/T$ for Zn film of thickness 75 nm.

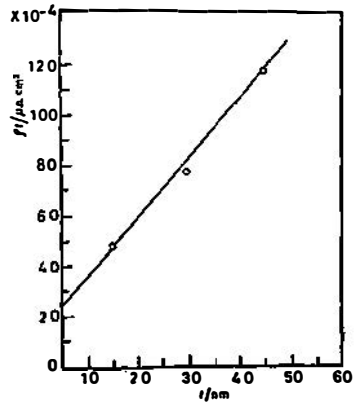

Fig 6. Relation between the product of resistivity and thickness ϱt and thickness t at temperature 323 K.

rature 323 K. It was found that the mean free path decreased with increasing temperature. By plotting a relation between $\log l_0$ and temperature it was possible to calculate the constants from the formula:

$$l_0 = a\, T^{-S}. \tag{3}$$

The value of S was found to be 15 which is the normal value of the metals.

It was possible to calculate the mobility μ from the values of the mean free path by using an expression based on the Lorentz-Sommerfeld theory (Goswami and Koli[8]) as follows:

$$\mu = \frac{l_0\, e}{225 \sqrt{2\pi m^* KT}} \frac{\mathrm{cm}^3}{\mathrm{V \cdot s}} \tag{4}$$

where e is the electron charge, and

m^* is the effective mass,

K is the Boltzmann constant and

T is the heating temperature.

All the obtained data are given in Table 1. It is shown that the mobility of charge carriers decreased with increasing temperature.

TABLE 1.

Temperature T/K	300	323	343	363
Mobility $\mu / \frac{\mathrm{cm}^3}{\mathrm{V \cdot s}}$	955.418	134.1104	124.9554	97.8499

Values of mobility μ of Zn films.

Besides, the activation energy of the charge carriers was estimated from the relation:

$$\ln R = \frac{\Delta E}{KT}. \tag{5}$$

Fig. 5b shows the relation between $\ln R$ and $1/T$ for film thickness 75 nm (as an example), from which ΔE was calculated. It was found that ΔE decreased from 0.0258 to 0.00507 eV with increasing film thickness from 15 to 45 nm, while ΔE decreased from 0.2415 to 0.1725 eV with increasing film thickness from 60 to 75 nm. The concentration of the charge carriers n_i of the different film thicknesses were calculated from the estimated values of the activation energy and the values of heating temperatures. The following relation was used (Goswami and Koli[9]):

$$n_i = 2\,(2\pi m^* KT/h^2)^{3/2} \exp\,(-\Delta E/2KT) \tag{6}$$

where h is Planck's constant.

It was found that n_i increased with increasing heating temperature, while it increased with increasing film thickness up to 45 nm and then irregularly behaved. All the calculated data are given in Table 2.

TABLE 2.

Temp. T/K	Concentration of charge carriers n_1				
	Film thickness 15 nm ($\Delta E = 0.025$ eV)	Film thickness 30 nm ($\Delta E = 0.086$ eV)	Film thickness 45 nm ($\Delta E = 0.005$ eV)	Film thickness 75 nm ($\Delta E = 0.24$ eV)	Film thickness 75 nm ($\Delta E = 0.1728$ eV)
300	3.31×10^{20}	4.62×10^{20}	4.95×10^{20}	5.18×10^{18}	1.95×10^{19}
323	3.83×10^{20}	5.22×10^{20}	5.22×10^{20}	8.00×10^{18}	2.76×10^{19}
343	4.31×10^{20}	5.77×10^{20}	6.12×10^{20}	1.12×10^{19}	3.61×10^{18}
363	4.81×10^{20}	5.81×10^{20}	6.15×10^{20}	1.53×10^{19}	4.625×10^{18}

Values of charge concentration of n_i and she corresponding values of heating temperature and film thickness.

Therefore, the application of Fuchs theory to the present experimental data yielded the determination of the unknown quantities ϱ_0 and l_0. However, Mayadas and Shatzkes[10] following the lines of Fuchs calculation, have taken into consideration grain boundary scattering and calculated the total resistivity of a thin metal film from a model in which three types of electron scattering mechanisms are simultaneously operated: (1) an isotropic background scattering (due to the combined effects of phonons and point defects), (2) scattering due to the external surface and (3) scattering due to a distribution of planar potentials (grain boundaries). For a single-crystal film ϱ_0 defined by the equation:

$$\varrho = \varrho_0 [f(\alpha) - A]^{-1} \tag{7}$$

must be identical to the equation:

$$\sigma_0/\sigma = \varrho/\varrho_0 = \left[1 - \frac{3}{2} \nu \int_1^\infty (a^{-3} - a^{-5})(1 - e^{-\nu a})\, da\right] = \frac{\Phi(\nu)}{\nu} \tag{8}$$

where $a = 1/\cos\Theta$,

σ_0 is the bulk conductivity value,

σ is the film conductivity and

$\nu = t/l_0$ = film thickness / mean free path. (9)

As ϱ is the resistivity of an infinitely thick single crystal[11], equation (8) has generally been used for monocrystalline films (in which the grain size a_g is equal to the film thickness t) by making a substitution $t = a_g$. However, for thin poly-

crystalline films whose grain size is constant (in other words, independent of thickness) equation (8) is commonly rewritten in the form[12]:

$$\varrho/\varrho_g = \left[1 - \frac{A}{f(\alpha)}\right]^{-1} \tag{10}$$

where ϱ_g is the resistivity of an infinitely thick polycrystalline film. From equations (8) and (10), the resistivity ϱ_g of an infinitely thick polycrystalline film is given by

$$\varrho_g = \varrho_0 f(\alpha)^{-1}. \tag{11}$$

The Mayadas-Shatzkes theory[10] concluded that the major portion of the total resistivity in polycrystalline films comes from electron scattering at grain boundaries. This could be taken into account in case of Zn films for their polycrystalline structure. Mayadas and Shatzkes[10] stated also that the surface scattering parameter P used in the Fuchs theory has about the same values for both single-crystal and polycrystalline films of a given material. The grain boundary reflection coefficient r can therefore be determined from measurements on polycrystalline films if the parameters P and l_0 are independently obtained from single-crystal films of the same class. Moreover, an effective intrinsic mean free path l_g for a polycrystal can be defined by using the relation:

$$l_g \approx (\varrho_0/\varrho_g)\, l = f(\alpha)\, l \tag{12}$$

where

$$f(\alpha) = 1 - \frac{3}{2}\alpha + 3\alpha^2 - 3\alpha^3 \ln(1 + 1/\alpha) \tag{13}$$

and

$$\alpha = \frac{l_g}{a_g} \cdot \frac{r}{1 - r} \tag{14}$$

where a_g is the grain size of the films. From the structural invesigation by electron microscopy, it was found that a_g increased with the increase of film thickness of Zn films. This means that α should be decreased and $f(\alpha)$ increased with the increase of film thickness. Consequently, the ratios ϱ_0/ϱ_g and l_g/l were increased with the increase of film thickness and grain size of polycrystalline Zn films. This means that ϱ_g was decreased and l_g was increased with the increase of film thickness. The increase of l_g should lead to the increase of mobility μ of charge carriers with increasing of film thickness. This is in agreement with the increase of grain size since the mobility always arises from grain size of thin films.

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STRUKTURA I ELEKTRIČNA SVOJSTVA TANKIH FILMOVA CINKA

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Tanki filmovi cinka debljine od 15 do 75 nm su napareni na amorfne staklene substrate brzinom od 0.25 nm/s pomoću metode termalne evaporacije. Nukleacija, rast i struktura tankih filmova istraživana su pomoću transmisionog mikroskopa i tehnikom difrakcije. Veličina i gustoća zrna mijenjala se sa debljinom filma. Izračunate su i korelirane vrijednosti volumne otpornosti, srednji slobodni put i koeficijent termalnog otpora filma sa strukturom filma.