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ELECTRICAL CONDUCTIVITY AND THE STRUCTURE OF THIN BISMUTH FILMS

A. H. ABOU EL ELA, S. MAHMOUD*, M. A. MAHMOUD and A. H. EID*†

Physics Department, Islamic Girls College, Nasr City Cairo, and Physics Department National Research Centre, Cairo, Egypt*

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Measurements of the electrical conductivity and the observations of the accompanying changes of structural properties of bismuth films of different thicknesses were carried out at room temperature. The electrical conduction mechanism was correlated with structural changes occuring during growth of the film. For small thickness $d < L$, where L is the electron mean free path, the film resistance varied as d^{-n}, where $n \simeq 2$. However, for $d > L$ the value of n become smaller than 2.

1. Introduction

The investigation of thin films is of interest both from the purely scientific point of view and from the practical one, since new specific effects, not observed in bulk samples, can appear in the film state. Measurements of the electrical properties of thin films could offer a suitable means of characterizing structural changes, if sufficient data relating the film structure to the electrical properties were available. Several investigators[1-4] have pointed out that changes in the electrical properties may arise from scattering of the conduction electrons at the surfaces of

† Now at the Intermediate College, Elkasim, Saudia Arabia.

the crystallites. These effects may be attributed to the structural changes occuring during growth or annealing processes. The structure of the deposited films depends on physical parameters which could significantly affect the electrical conduction[5,6]; the film thickness, the rate of evaporation, the nature and the state of the substrate.

The aim of the present contribution is to investigate the electrical conductivity, the mechanism of growth and the microstructure of bismuth films of different thicknesses prepared by vacuum evaporation.

2. *Experimental*

Bismuth films of different thicknesses were prepared by vacuum evaporation of highly pure bismuth (99.999 %) from a tungsten filament simultaneously onto copper supported thin carbon film and quartz substrate at room temperature in a vacuum of 10^{-4} Pa at an evaporation rate of about 3 nm/s. Samples were also deposited onto thin cleaved mica. Predeposited gold layers were used as electrodes for electrical measurements, where a small current passed along the film and the potential difference was measured by a potentiometer circuit. Film thickness was measured by Tolansky interference method[7].

3. *Results and discussion*

The dependence of the film resistance on thickness is shown in Fig. 1, at three different temperatures. The film resistance $R(d)$ rapidly decreases from a value which is determined practically by the substrate resistance to a value of the order of few ohms when d decreases. This behaviour of the $R(d)$ dependence is determined wholly by those structural changes which occur during the initial and subsequent stages of the formation of the condensate.

When the atoms are condensed in vacuum on the substrate they are captured by crystallization centres such as point defects, cleavage steps etc. and the resistance in this case is that of the dielectric substate, Fig. 1, region 1.

For higher film thickness (12 nm), on amorphous carbon substrate, very small and thin nuclei are formed, Fig. 2 a. The electron diffraction pattern recorded consists of continuous rings which indicates that the film has a polycrystaline structure and consists of aggregates of bismuth crystals randomly oriented on the carbon substrate.

When the nuclei start to coalesce, Fig. 2b (18 nm), the number of voids decreases and the resistance falls sharply and the effective cross section of the current conducting films increases with the average thickness. As the film thickness increases, the nuclei tend to become small crystallites of different shapes. Some islands are formed from several nuclei (dark areas), which either represent thicker parts of the film or due to group of crystals in the reflecting position,while others consist of few nuclei (transparent areas). The continuous increase in the effective cross section of the current conducting film explains why the resistence continued to fall rapidly. In double logarithmic coordinates this part of the dependence $R(d)$ is given by a straight line whose slope is about — 2, region 2 in Fig. 1.

Larger crystals of sharp geometrical shape are formed with the deposition of further metal atoms, Fig. 2c. Moreover, small irregular holes are observed for thicker films (45 nm). Andrew and Krasavec[9] have attributed these holes to bubbles of residual gasses in deposited thin films of gold.

Thicker films ($d \sim 70$ nm), Fig. 2 d, contain transparent areas, dark grains and grain boundaries. It appears that transparent areas are grains or portions of grains out of Bragg condition, dark grains are grains or portions of grains in the Bragg condition for diffraction. The average grain size is 1 micron and increases with thickness. Continous paths are formed from colaesced islands having a low electrical impedance which control the conduction in the film and lead to further decrease of film resistance. The gaps in the quasi-continuous structure are filled to give a continuous film. The slope of $R(d)$ curve decreases with increasing thickness and for $d \sim 90$ nm the slope is about -1.05 and a relation $R \sim d^{-1}$ holds for film thickness > 90 nm.

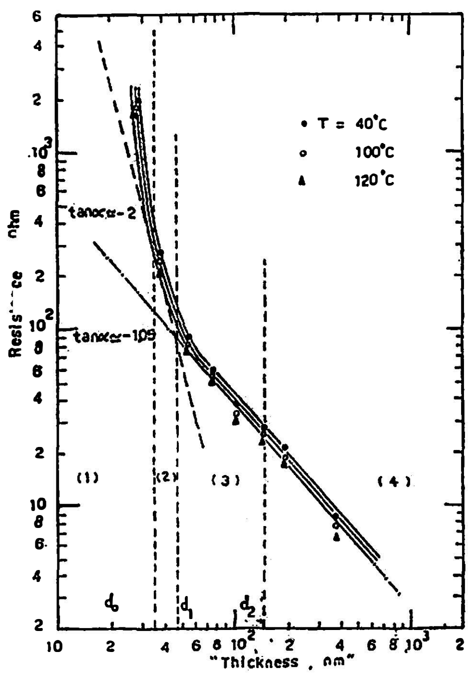
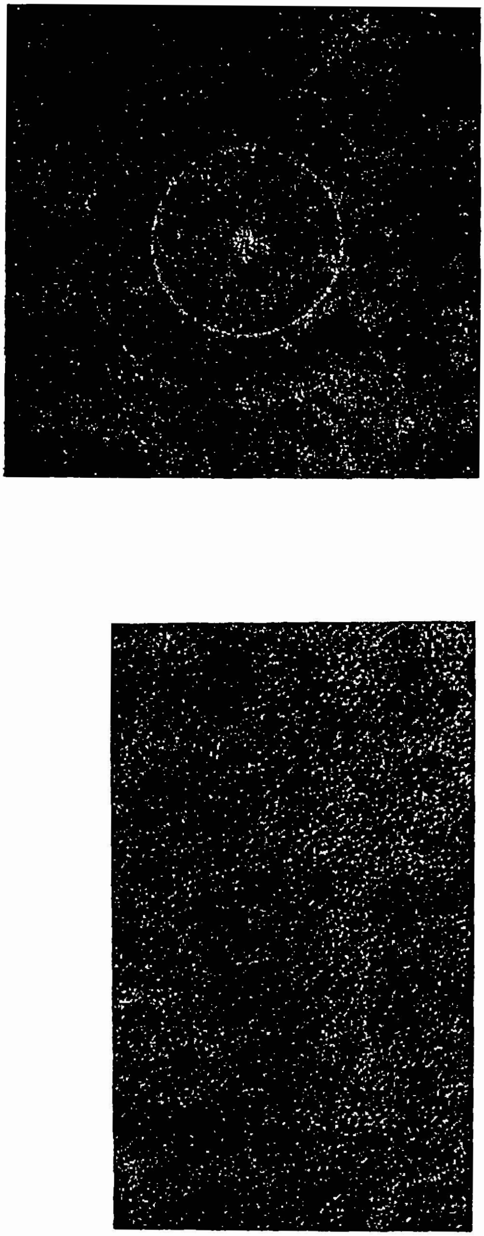
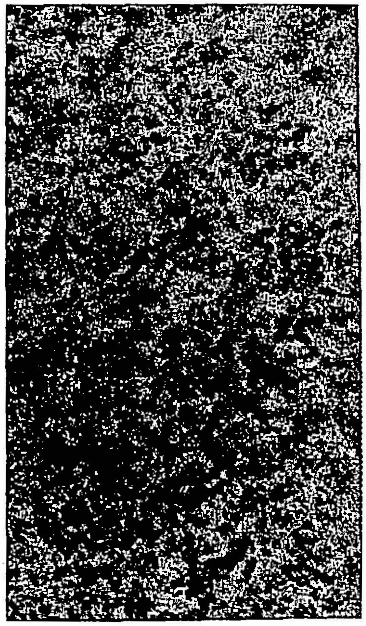


Fig. 1. The dependence of the film resistivity on the film thickness at different temperatures.

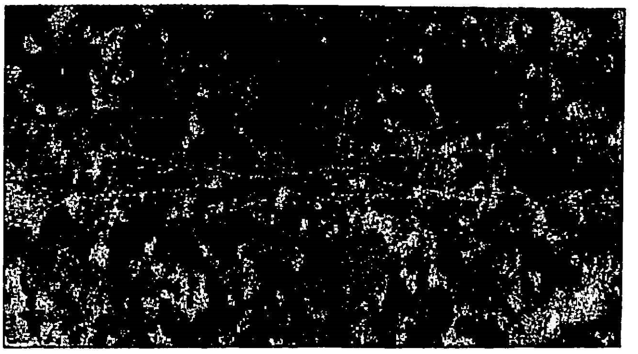
Theoretical models[2,4,8,10,11] have shown, that the electrical resistance of thin films should in some way depend on the film thickness (d). From these models it follows that the electrical conductivity for very thin films, of discontinuous structure, is attributed to a tunnelling mechanism[8,11]. Moreover, for thicker films, when the scattering of electrons at the film boundaries is mainly of the diffuse type and



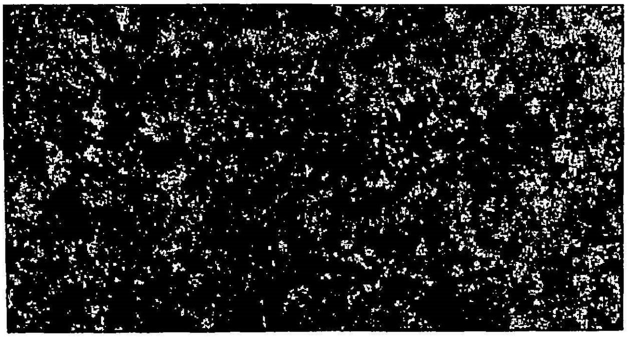
(a) 12 nm, 8000 X



(b) 18 nm, 6000 X



(c) 45 nm, 6000 X



(d) 70 nm, 6000 X

Fig. 2. Electron micrographs of bismuth films deposited onto amorphous carbon substrate.

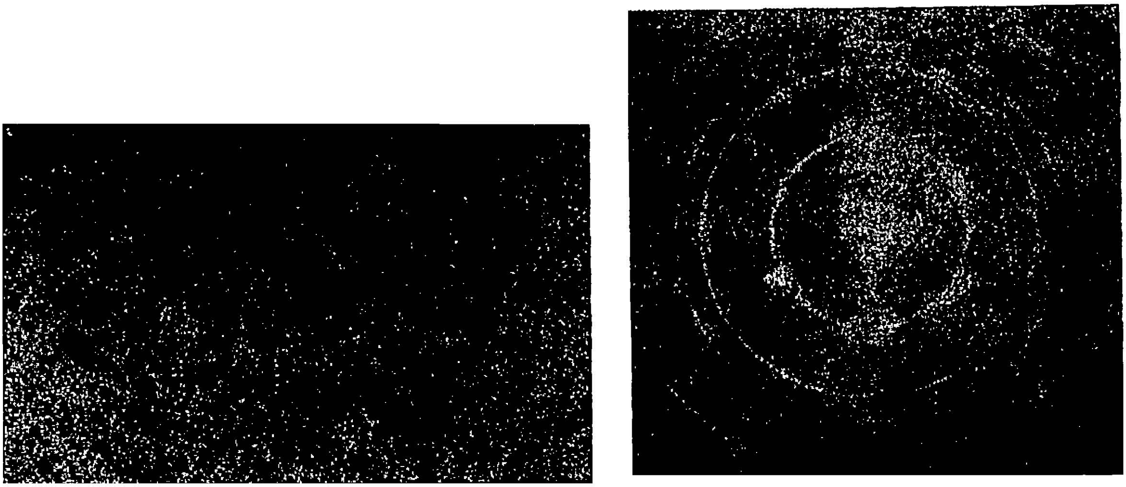
$d < L$, where L is the electron mean free path in a bulk sample at a given temperature, the resistance thickness dependence should be[1-3]

$$R(d) = \frac{4}{3} \varrho_0 \left(\frac{a L}{b d^2}\right) \left(\frac{1-p}{1+p}\right) \left(\ln \frac{L}{d}\right)^{-1} \tag{1}$$

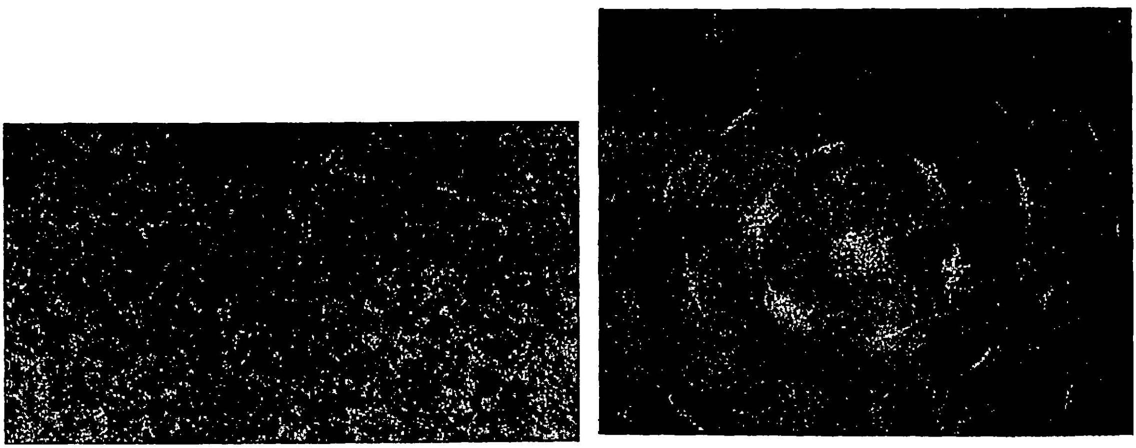
where ϱ_0 is the resistivity of the bulk crystal, p is the coefficient of specular reflection of electrons from the film boundaries ($0 \leq p \leq 1$). and a, b are the length and width of the film. For $d > L$, according to Fuchs-Sondheimer model[1-2], the $R(d)$ dependence is

$$R(d) = \varrho_0 \left(\frac{a}{bd}\right) \left[1 + \frac{3}{8}(1-p)\right] \frac{L}{d} \tag{2}$$

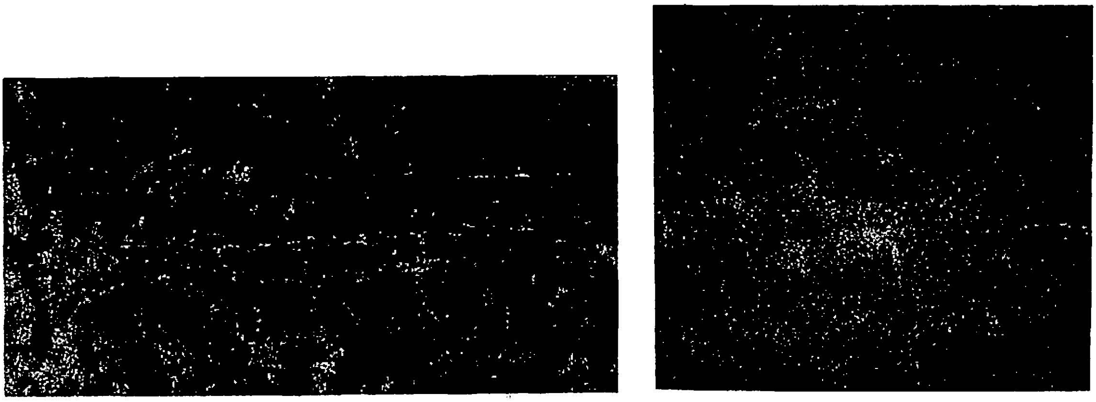
for $d < L$ the resistance $R(d)$ should decrease as d increaes closely in accordance with d^{-2} (due to the factor L/d^2 which decreases much faster than the factor $(\ln L/d)^{-1}$ increases). While for $d > L$, the resistance should vary as $R \sim d^{-1}$. Deviation from Eq. 2 is observed for film thickness < 36 nm, where the slope of the $R(d)$ dependence becomes larger than 2 (region 2), and the abrupt increase in



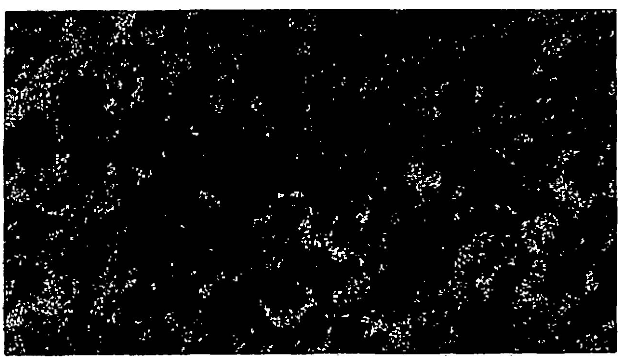
(a) 12 nm, 6000 X



(b) 40 nm, 6000 X



(c) 60 nm, 6000 X



(d) 80 nm, 6000 X

Fig. 3. Electron micrographs of bismuth films deposited onto cleaved mica substrate.

resistance of films thinner than 36 nm could not be explained in terms of Fuchs--Sondheimer model, which neglects the scattering due to grain boundaries. Our calculations[12] for the resistance of thin films < 36 nm show that the experimental data is best fitted to Maydas and Shatzkes model[4], for the resistance of a thin film in which three types of scattering mechanisms are simultaneously operative; an isotropic background scattering (due to phonons and point defects), scattering due to grain boundaries, and scattering due to external surfaces.

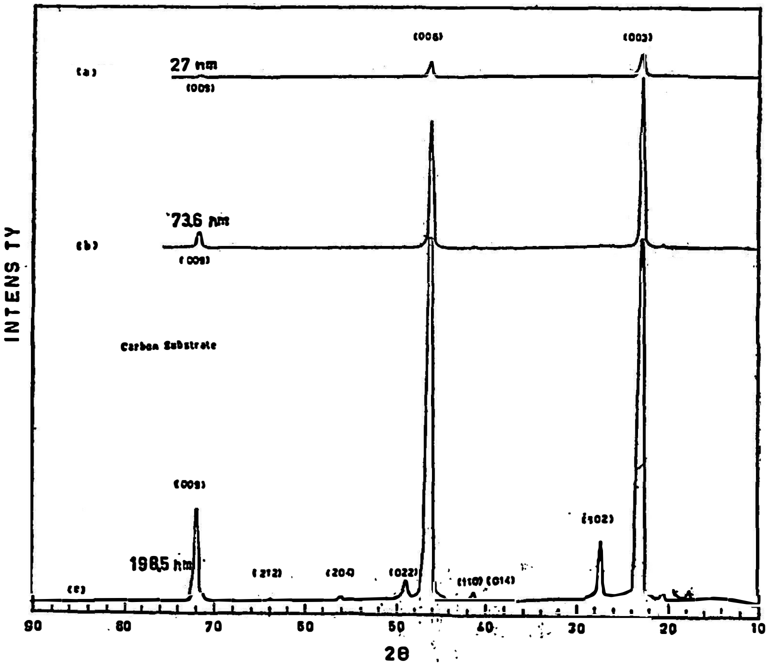


Fig. 4. The effect of the film thickness on its crystal structure, X-ray diffraction patterns for different film thickness: (a) 27 nm; (b) 73.6 nm c) 198.5 nm deposited onto amorphous carbon substrate,

Electron transmission micrographs and diffraction patterns for thin bismuth films deposited onto a cleaved mica substrate are shown in Fig. 3. For small film thickness $\sim$ 12 nm (Fig. 3a) bismuth nuclei are small crystallites of different orientations which predominantly coalesced to small islands and aggregates. This indicates that the stage of coalescence between nuclei is more pronounced than in the case of carbon substrate.

For thicker films (40 nm) the particles grow in size and coalescence of nuclei and formation of grains is observed (Fig. 3b). At this stage electron diffraction patterns consist of arcs having the most intense blackening in the center. The arcy appearance of the rings in the diffraction patterns shows that the film consists of many crystals which are preferentially oriented in respect to the electron beam, due to the influence of the mica substrate.

With further increase of the film thickness large crystals with sharp geometrical shapes produced by coalescence are formed (Fig. 3d) and the size of crystallites increases with the thickness of the film.

For thick films $d > 80$ nm nearly continuous film appears (Fig. 3d) with aggregates of thick particles superimposed on the transparent thin film. Black areas in Fig. 3d represent grains and/or parts of grains in reflecting position.

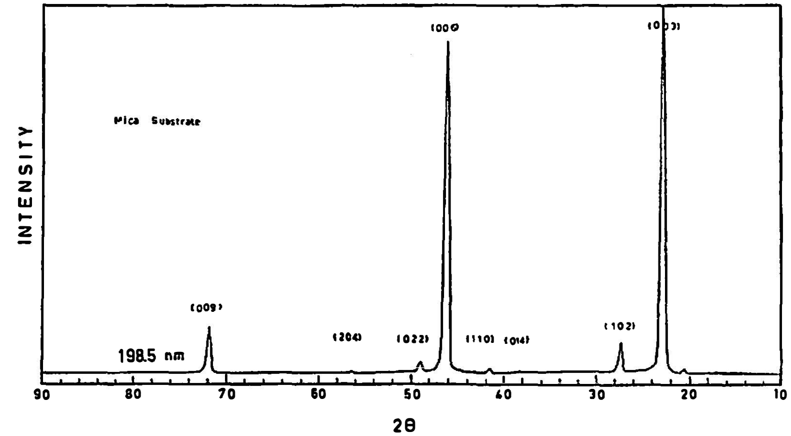


Fig. 5. X-ray diffraction pattern for 198.5 nm film deposited onto cleaved mica substrate.

X-ray diffraction patterns for bismuth films deposited on amorphous substrate (Fig. 4) and on cleaved mica substrate (Fig. 5) showed that the crystallites have an arbitrary azimuthal orientation. Moreover, the degree of crystal perfection remains unchanged with a decrease in film thickness down to about 30 nm.

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ELEKTRIČKA VODLJIVOST I STRUKTURA TANKIH SLOJEVA BIZMUTA

A. H. ABOU EL ELA, S. MAHMOUD, M. A. MAHMOUD i A. H. EID

Physics Department, Islamic Girls College, Nasr City Cairo, i

Physics Department, National Research Centre, Cairo, Egypt

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Načinjena su mjerenja električke vodljivosti na tankim slojevima bizmuta različitih debljina sa usporednim praćenjem strukturalnih promjena. Mjerenja su načinjena na sobnoj temperaturi. Mehanizam električke vodljivosti je povezan sa strukturalnim promjenama koja nastaju za vrijeme rasta filma. Za male debljine uzoraka $d < L$ otpor filma se mijenja prema izrazu d^{-n} u kojem za n treba uzeti približno vrijednost 2. Sa L je označena slobodna duljina puta elektrona. Za $d > L$ vrijednost za n je manja od 2.