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ELECTRICAL TRANSPORT PROPERTIES OF THIN POLYCRYSTALLINE BISMUTH FILMS

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Abstract:

Bismuth films were deposited onto glass substrates at three different deposition rates (0.3, 5 and 20 nm s^{-1}). The filmswere found to have polycrystalline structure with a grain size dependent on deposition rate as well as film thickness. The resistivity ρ, TCR, Hall coefficient R_H and magnetoresistance coefficient of the Bi films were measured.

At 300 K the thickness dependence of the resistivity can br roughly fitted by the Fuchs-Sondheimer theory with specularity parameters of 0.678, 0.671 and 0.652 corresponding respectively to the deposition rates 0.3, 5 and 20 nm s^{-1}. The conclusion drawn from the temperature dependence of the resistivity, concerning the diffuseness of the surface scattering of the charge carriers, were confirmed by the dependence of the mean free path on the sample temperature. The Hall coefficient, carrier concentration, Hall mobility seem to be field independent.

Introduction:

Many authors have inestigated the structure and the electrical transport properties of the bismuth films (1-5). Great interest have been paid concerning the temperature dependence of these properties. However, the depsotion rate was not well specified.

In the present work, the primary emphasis was to study the effect of the deposition rate on Bi film structure as well as the electrical transport properties.

Experimental procedure:

In a vacuum of 10^{-5} Torr elemental Bi, nominally 99.999% pure (Balzer's Comp. West Germany), was evaporated from a tungsten boat, typically at a deposition rate of 0.3, 5 or 20 nm s^{-1}, onto glass substrate kept at room temperature. Most of the substrates used in this work were flat microscopic slides. The film thickness was controlled during the deposition process by QSG 101 quartz crystal monitor, then measured by the conventional method (6). Both X-ray diffractograms and electron micrographs were used in investigating the structure of Bi films. The electrical measurements were performed either at room temperature or at elevated temperatures using high resolution multimeter (TE924 Microdigit). The technique utilised in measuring Hall coefficient and magnetoresistance was described elsewhere (7).

Results:

a) Film structure

Typical X-ray diffraction patterns as well as EM diffraction patterns are illustrated in Figs. (1& 2). These patterns indicate that Bi films deposited at room temperature onto glass substrates have polycrystalline structure. Bi films after annealing still have polycrystalline structure. It was found that the size of individual crystallite increases with increasing film thickness and decreases with increasing deposition rate. The crystallites are ten or more times smaller than those prepared on a heated mica (3,4,8). However, the obtained results are in good agreement with those reported by Inoue et al (9).

b) Resistivity

Both the thickness dependence and deposition rate dependence of Bi resistivity were studied at room temperature and at elevated temperatures up to 150°C. It was found that the resistivity of Bi films decreased monotonically as the film thickness increased. It was also found that the resistivity decreased with increasing the sample temperature. For any selected Bi films of the same thickness, the resistivity increased as the deposition rate increased.

Thickness dependence of the resistivity, deposition rate dependence of resistivity, and temperature dependence of resistivity were illustrated in Figs. (3 & 4). The over-all thickness dependence of resistivity o curves for the deposition rates (0.3, 5 and 20 nms^{-1}) can be understood in terms of Fuch-Sondheimer boundary-scattering theory.

$$\rho = \rho_0 [1 + (\frac{3\ell}{8t}) (1 - P)] \qquad (1)$$

where ρ_0 is the resistivity of Bi films of infinite thickness (bulk resistivity of the same structure), t film thickness, ℓ mean free path of the charge carriers and P specularity parameter.

The previous equation predicts a linear dependence of ρt on t. Typical representations for the three above mentioned deposition rates are shown in Fig. (5). The intercepts of these plots on the y-axis give ℓ (1-P) and the slopes give

ρ_0. The mean free path of the charge carriers is given by:

$$\ell = (3\,\pi^2)^{1/3}\,\hbar\ /\ e^2\, n^{2/3} \rho \qquad (2)$$

At 300 K, using the obtained values of ρ_0 and the carrier concentration reported by El-Shazly et al. (10), equation (2) yields ℓ. Using the calculated values of ℓ in conjunction with the determined values of ℓ (1-P), the specularity parameter P can be evaluated. Values of ℓ(1-P), ρ_0, 1 and P for Bi films deposited onto glass substrates at deposition rates of 0.3, 5 and 20 nms^{-1} are given in table (1)

Table (1): Value of ℓ (1-P), $_{o}$ ℓ and P Determined from Fig. (5) & Eq. (2)

Deposition rate, nms^{-1}	ℓ(1-P), nm	ρ μ.Ω.cm	ℓ, nm	P
0.3	238	137.5	739	0.678
5.0	220	152.0	669	0.671
20.0	200	177.0	574	0.652

Following the same procedure, similar quantities representing ρ_0 and ℓ(1-P) at elevated temperatures can be determined for Bi films deposited at various rates. Using these values in conjunction with the corresponding values of P, determined before (assumed to be unchanged for each deposition rate), the mean free path of free charge carriers can be determined. Then assuming that the carrier concentration should be the same for all samples, eq. (2) yields ℓ. The obtained values are given in table (2).

Table(2): Values of 1(1-P), ρ_0 (μ.Ω.cm), 1(nm) measured at elevated temperatures for Bi films deposited at various rates.

Depos rate / T,K	0.3 nm^{-1}			5 nms^{-1}			20 nms^{-1}		
	1(1-P)	ρ_0	ℓ	ℓ(1-P)	ρ_0	ℓ	ℓ(1-P)	ρ_0	ℓ
300	238	137.5	739	220	152	669	200	177	574
340	245	128	761	235	138	714	212	160	609
380	280	116	870	268	123	815	238	144	684
420	301	109	935	294	112	894	270	133	776

It is obvious that with increasing the sample temperature ρ_0 decreases whilst ℓ increases keeping the deposition rate constant. The calculated values of ℓ using eq. (2) are slightly higher than those values determined throughout eq. (1) by 1.3%.

c) Temperature coefficient of resistivity (TCR)

Data representing R = f (T), illustrated in Fig. (4) was used in determination of TCR.

The obtained results for TCR ($= \frac{1}{R}\frac{dR}{dT}$) are shown in Fig. (6) for different deposition rates. It is obvious that TCR increases with increasing the deposition rate and decreases gradually with increasing the sample temperature. However, it is always negative in the temperature range 30-150°C.

d) Hall coefficient

Measurements of Hall coefficient as a function of the applied field intensity are illustrated in Fig. (7). The following two characteristic features are to be noted:

i. Hall coefficient seems to be field independent whatever the deposition rate or the film thickness.

ii. Hall coefficient is always positive for Bi films when measured at room temperature.

The positive sign of the Hall coefficient was believed to be due to the presence of the local acceptor states, which may locate near the top of the hole band through the formation of a band gap (transition from semimetal to semiconductor). The Hall mobility and the carrier concentration were also found to be field independent. The obtained results are in good agreement with those reported before (9, 10).

e) Magnetoresistance in Bi films

Thin bismuth films in the thickness range of 50-800 nm show a significant magnetoresistance when measured at room temperature. It was found that $\Delta\rho/\rho$ is almost proportional to the square of the magnetic field intensity B up to 0.78 Tesla for various films whatever the deposition rate. However, the magnetoresistance coefficient of Bi films deposited either at 5 or 20 nms^{-1} is 3-4 times greater than that of Bi films depositted at 0.3 nms^{-1}.

Discussion:

We have studied the transport properties of glass coated Bi films ($60 \leq t \leq 300$ nm) deposited at three deposition rates (0.3, 5 and 20 nms^{-1}). The substrate was kept at room temperature during the deposition process. Structural investigation showed that films prepared in this way either before or after annealing were made up of small crystallites with different orientations. So, these films were polycrystalline. The size of the crystallites increased with increasing film thickness and decreasing the deposition rate. They have the same magnitude in comparison with those reported by Inoue et al (9) for glass coated Bi films, but of one order magnitude smaller than those prepared on heated mica (3, 4, 8).

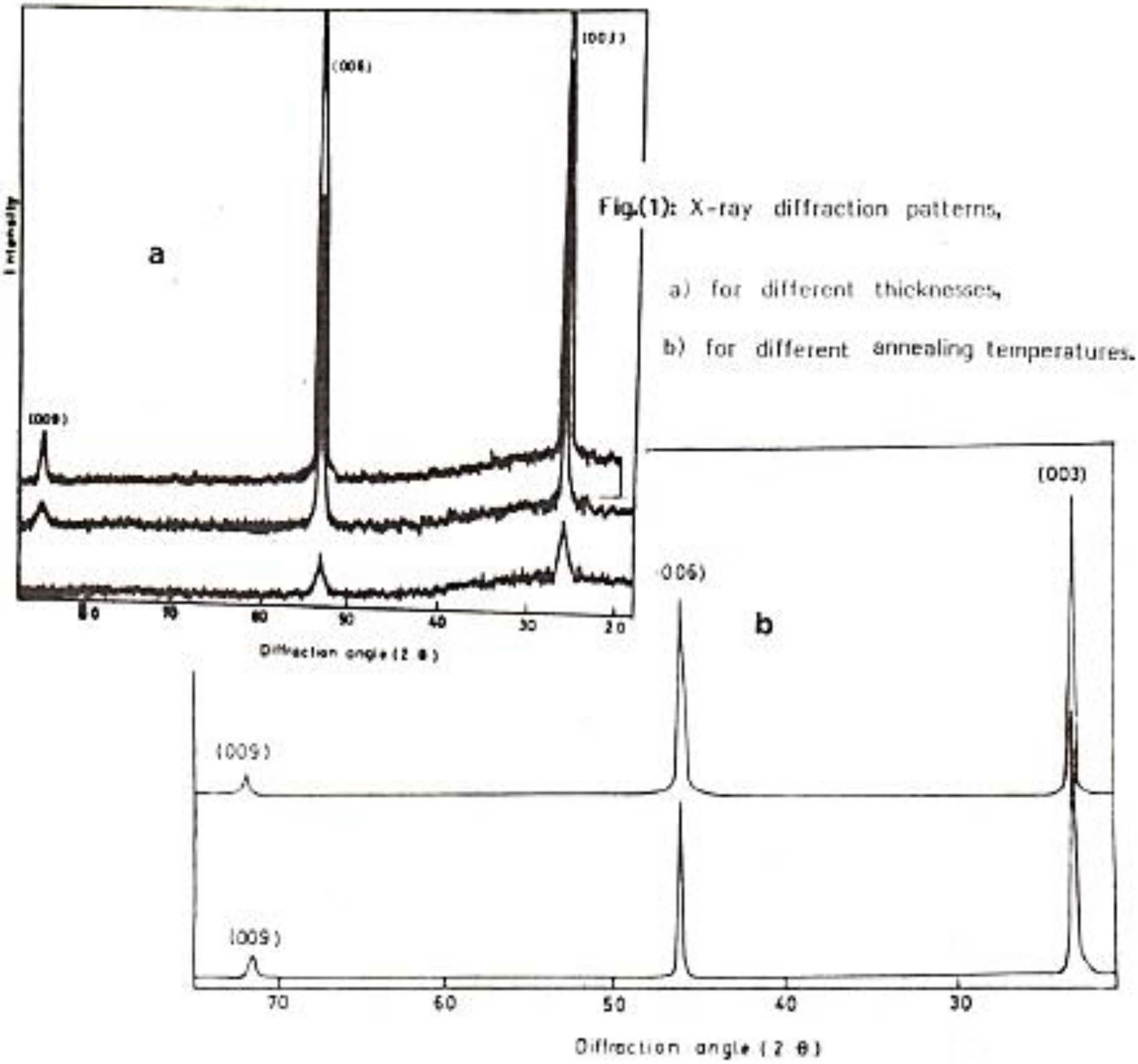


Fig.(1): X-ray diffraction patterns,

a) for different thicknesses,

b) for different annealing temperatures.

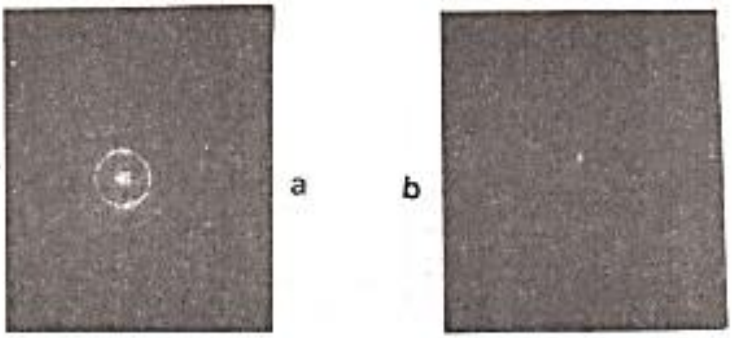


Fig.(2): E.M. diffraction patterns for Bi films on glass substrates, a) before annealing, b) after annealing.

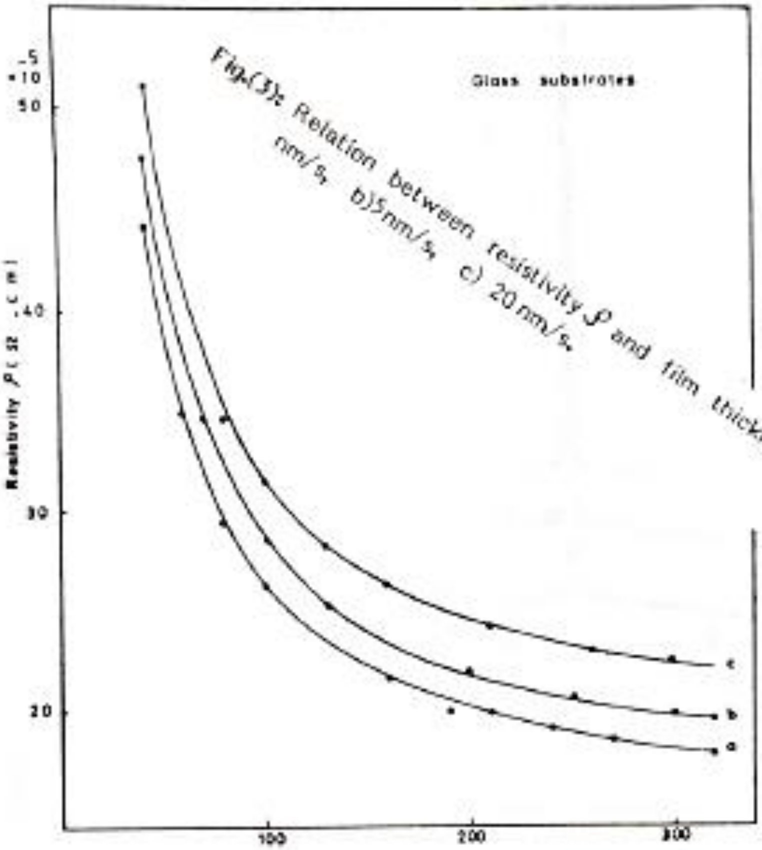


Fig.(3): Relation between resistivity ρ and film thickness t for different rates, a) 0.3 nm/s, b) 5 nm/s, c) 20 nm/s.

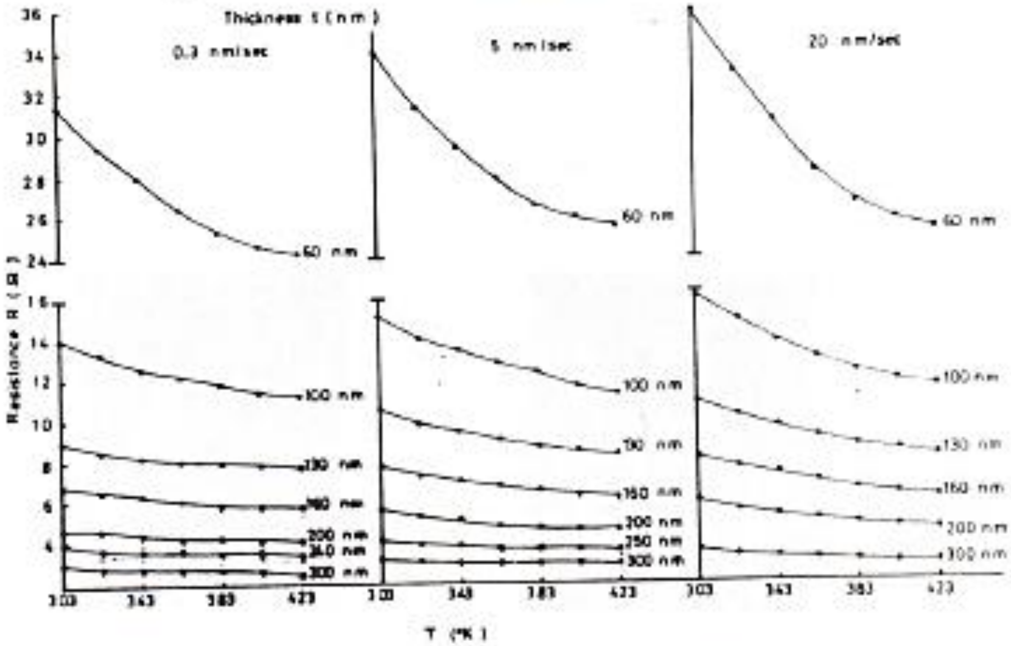
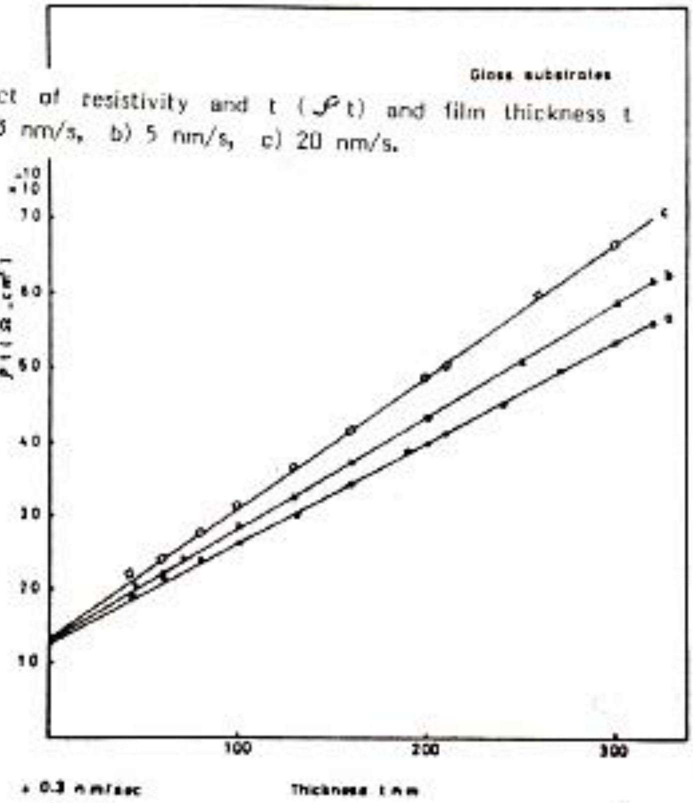


Fig.(4): Relation between resistance R and sample temperature T for different film thickness deposited with different rates on glass substrates.

Fig.(5): Relation between product of resistivity and t (ρt) and film thickness t for different rates: a) 0.3 nm/s, b) 5 nm/s, c) 20 nm/s.



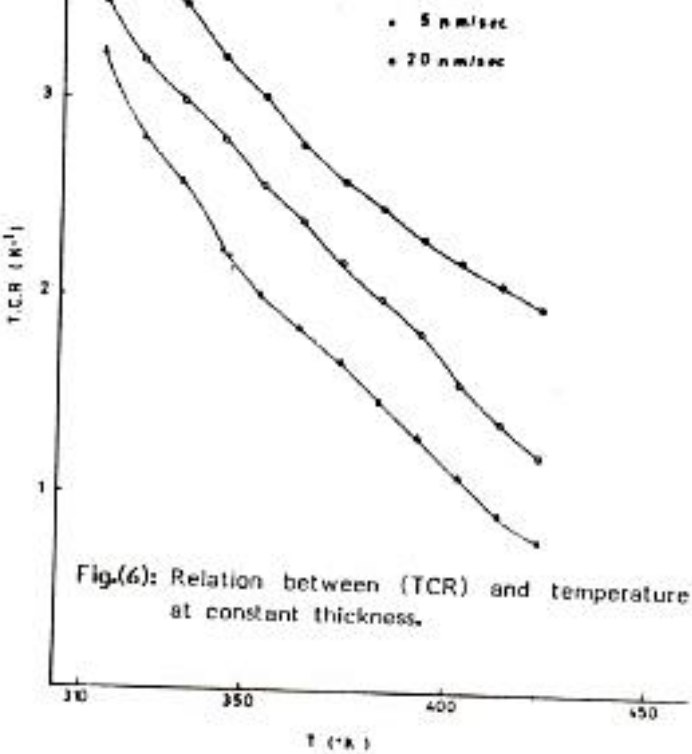


Fig.(6): Relation between (TCR) and temperature T for different deposition rates at constant thickness.

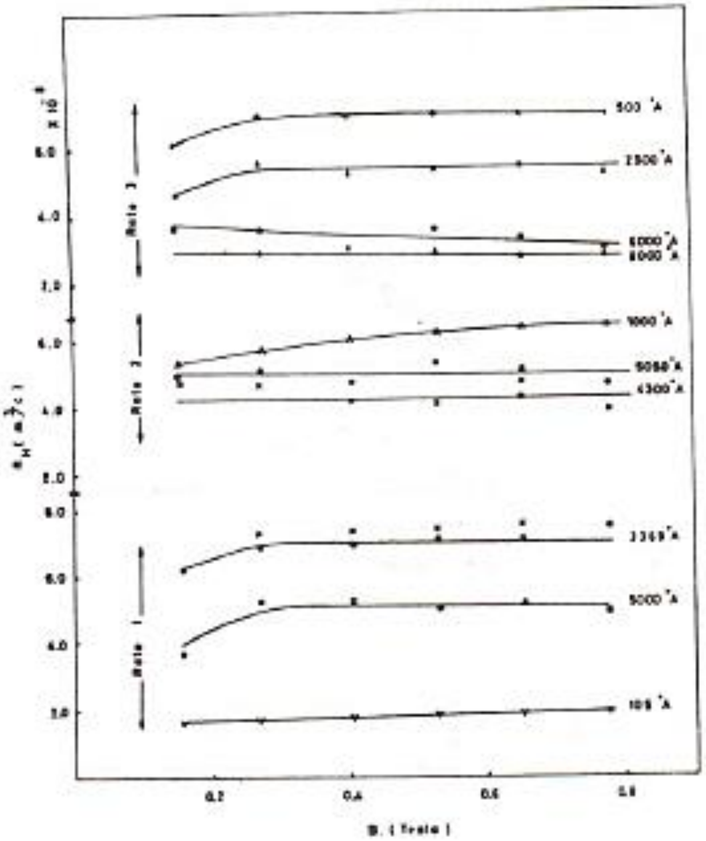


Fig.(7): Relation between Hall coefficient R_H and intensity of magnetic field B for different rates.

The thickness dependence of resistivity either at room temperature or at any elevated temperature, as can be explained in view of film structure, was understood by Fuchs-Sondheimer boundary scattering theory. At 300 K, $P = 0.678$, 0.671 and 0.652 for Bi films deposited respectively at 0.3, 5 and 20 nms^{-1}, in agreement with the previously mentioned findings indicating partially diffuse scattering (8). Hoffman (8) finding at 300 K ($P = 0.56$ and $\ell = 590$ nm) depend upon values of $\rho_0 = 1.2 \times 10^{-4}\,\Omega.cm$ and $n = 2.6 \times 10^{18}\ cm^{-3}$ as reported for bulk material (11, 12). This may be not completely true because of the difference in structure for Bi in thin film form and in bulk form. To overcome this difficulty, we make use of $\rho_\bullet$, which is the resistivity of Bi film of infinite thickness, determined from Fig. (5), as well as the free charge carrier concentration determined for the same films either throughout Hall coefficient measurements or throughout optical constants measurements (10). The conclusions, drawn from the temperature dependence of the resistivity, concerning that P and n should be the same for Bi films prepared with certain deposition rate, were confirmed by the dependence of the mean free path on the sample temperature.

Concerning the semiconductor-like behaviour observed in thin bismuth films, i.e., the negative temperature coefficient of resistivity as suggested by Duggal et al. (3), is due to the uncrossing of the conduction and valence bands predicted by the quantum size effect theory (13).

The Hall coefficient as well as the carrier concentration and Hall mobility seem to be field independent when measured at room temperature. The positive sign of the Hall coefficient was believed to be due to the presence of the local acceptor states, which may locate near the top of the hole band through the formation of the band gap, according to the quantum size effect (transition from semimetal to semiconductor).

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