Vol. A37 (1970) *ACTA PHYSICA POLONICA* Fasc. 4

EFFECT OF EVAPORATION CONDITIONS ON ELECTRICAL PROPERTIES OF InSb THIN LAYERS

By M. Oszwałdowski

Industrial Institute of Electronics, Division in Toruń*

(Received June 4, 1969)

The effect of the conditions under which InSb thin layers are obtained on their electrical properties is discussed. The layers were produced by the three temperature and by flash evaporation methods. Particular attention was paid to the effect of the chemical composition of the vapours and its fluctuation during the evaporation process on the magnitude of conductivity, Hall coefficient and electron mobility in the layers. The results obteined imply the formation of two kinds of defects which act as donors and acceptors. The number of defects and the degree of their compensation depends on the chemical composition of the vapours during the evaporation process. It was found that the number of defects and their type influences the mobility and its temperature dependence.

1. Introduction

A number of papers [1—6] deal with the methods of obtaining InSb thin layers of high electron mobility and their physical properties. The principal difficulty in obtaining InSb thin layers of stoichiometric composition is the dissociation of indium antimonide which leads to fractional evaporation.

Currently several techniques of obtaining InSb thin layers are known which allow the effects of this dissociation to be avoided. The most commonly used are the three temperature method [1, 2] and the flash evaporation method [5]. The layers produced in these methods are polycrystalline and the electron mobility in them is of the order of 10 to 15×10^3 cm^2/Vsec. The experimental results of many authors [2, 4, 6, 7] indicate that the main mechanism which limits electron mobility in InSb thin layers is scattering at the grain boundaries of crystallites. Mobility is seen to increase with an increase in crystallite volume.

A characteristic feature of InSb thin layers in the appearance of a maximum in the electron mobility *versus* temperature curve at high temperatures. This peak appears independently of the way in which the InSb thin layers are obtained and it lies near 100 °C [2, 3]. This phenomenon does not show up in bulk InSb, in which the electron mobility decreases sharply with increased temperature.

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* Address: Przemysłowy Instytut Elektroniki, Toruń, Młodzieżowa 29/37, Polska.

2. Preparation of InSb thin layers

Thin layers of indium antimonide for use in studies of electrical properties were obtained by the three temperature method or the flash evaporation technique in a vacuum of 10^{-5} to 10^{-6} torr. Masks were used during evaporation which enabled the preparation of five separate layers each of area 8×2 mm^2 or twelve of area 7×15 mm^2 in a single evaporation procedure.

In the three temperature technique the indium and antimony were vaporized from tungsten furnaces, into which mobile crucibles of Al_2O_3 were placed. The crucibles, together with the indium and antimony, were weighed before and after evaporation, allowing the quantity of evaporated material to be determined. The arrangement of the vapour sources and substrates is shown in Fig. 1. The distance betwen the vapour sources, *d*, and the distance between the sources and substrates, *h*, could be adjusted as required.

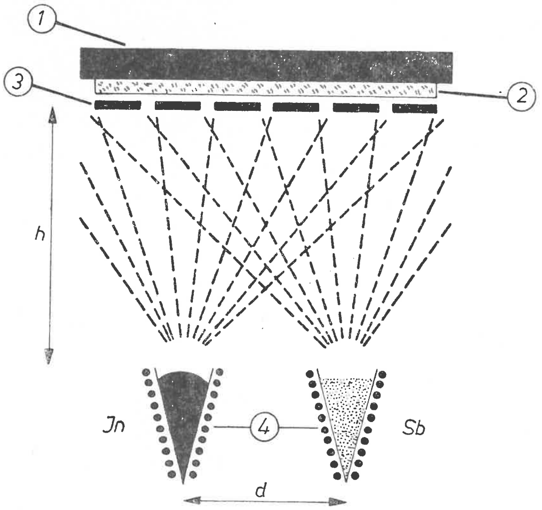


Fig. 1. Arrangement of vapour sources for obtaining a variable chemical composition of vapour stream along the substrate. *1* — **substrate furnace,** *2* — **substrate,** *3* — **mask,** *4* — **vapour source**

Flash evaporation of thin InSb layers was accomplished with the use of equipment described in [8]. The starting material was powdered indium antimonide of concentration $n \approx 10^{16}$ cm^{-3}. The grain size of the powder was 150 to 200 μm. In order to develop the required excess of antimony in the vapour phase during evaporation of layers onto substrates at temperatures higher than 400 °C, a predetermined amount of powdered Sb was added to the InSb powder. Both powders, of the same grain size, were thoroughly mixed before being placed in the vacuum apparatus.

3. The effect of layer preparation conditions on electron mobility

In order to establish the influence on electron mobility of the conditions under which InSb thin layers are prepared, the electrical properties of these layers were examined for dependence on substrate temperature, annealing time, vapour chemical composition and type of substrate used. Of these factors only the type of substrate did not have any real effect on the magnitude of electron mobility. Glass, quartz, and mica were used as substrates in these experiments.

The condition of InSb thin layer growth most intensely studied in this work was the chemical composition of the vapours from which the layers were obtained. There is very little information available at present on the influence of vapour composition on the electrical properties of thin layers of indium antimonide.

The composition of the vapour stream was altered either by selecting appropriate temperatures of the vapour sources or by displacing the two sources by such a distance ($d \sim h$) that layers of different composition were obtained along the substrates. During evaporation onto hot substrates (>400 °C) the required vapour composition was acquired by appropriately selecting the temperatures of both vapour sources, while the sources themselves were brought closer together ($d \ll h$). In this way it was possible to obtain during one evaporation procedure a number of samples evaporated under identical conditions for measurements of electrical properties.

If the ratio of the number of antimony atoms to that of indium atoms falling per unit time per substrate area is denoted as Sb/In, then depending on the substrate temperature there exists a certain interval of Sb/In values within which stoichiometric InSb layers are obtained. Table I gives the optimum values of the Sb/In ratios for different substrate temperatures at which stoichiometric InSb layers were obtained. The same table also includes the average magnitudes of specific conductivity σ, Hall coefficient R_H and Hall mobility μ_H for the cited evaporation conditions. These data show that with increasing substrate temperature the interval of Sb/In ratios at which layers of high mobilities may be achieved becomes broadened. This may be justified by the higher efficiency of reevaporation of the excess antimony from the InSb layers at the higher substrate temperatures.

The substrate temperature also has a serious effect on the electrical properties of InSb thin layers. The choice of substrate temperature must depend on the vapour chemical composition in the evaporation process. As is seen from Table I, the higher the substrate temperature applied, the higher the antimony excess should be in the vapour phase in order to achieve stoichiometric layers. As is well known [2, 9], the advantages gained by maintaining the substrates near the melting point of InSb (525 °C) lie in the creation of good crystallization conditions and in the increased efficiency of evaporation of the excess antimony. The highest temperature at which continuous InSb layers were obtainable was 490 °C. Above this temperature the layers obtained consisted of crystallites loosely bound to one

TABLE I

Substrate temperature [°C]	σ [Ω^{-1} cm^{-1}]	R_H [cm^3 C^{-1}]	μ_H [cm^2 V^{-1} s^{-1}]	Sb/In
400—410	50—100	40—60	$2-5\times10^3$	1.1—1.4
460—470	40—100	70—140	$5-11\times10^3$	1.1—1.6
480—490	40—80	80—230	$6-12\times10^3$	1.2—2.2

another. The InSb layers evaporated at temperatures from 480 to 490 °C had the highest mobilities, reaching $\mu_H = 12 \times 10^3$ cm^2/V sec.

A rise in electron mobility was also observed in layers submitted to annealing. The layers were annealed for 5 to 90 minutes at the same temperature in which they were evaporated. Table II gives the properties of several InSb thin layers annealed for different periods of time. Apart from the rise in mobility, an increase in the Hall coefficients in the annealed layers is also seen. It should be emphasized, however, that in order to obtain a rise in mobility the annealing time should be chosen according to the layer composition after it is evaporated, *i.e.* to the Sb/In ratio. In layers evaporated with a vapour stream of approximately stoichiometric composition (Sb/In ≈ 1), long annealing caused some loss of antimony, *i.e.* it disturbed the stoichiometric composition of the layer. A distinct drop in mobility was observed in these layers.

TABLE II

Layer	Sb/In	d [μm]	σ [Ω^{-1} cm^{-1}]	R_H [cm^3 C^{-1}]	μ_H [cm^2 V^{-1} s^{-1}]	Substrate temperature [°C]	Annealing time (min.)
A	1.35	1.8	56	133	7400	490	5
B	1.35	1.8	41	220	9000	490	30
C	1.5	2.8	85	70	6900	470	5
D	1.35	2.5	48	208	10000	470	30

In accord with the results obtained earlier [1—4, 7] a rise in electron mobility in layers with larger crystallites was found. An increase in crystallite size was observed for the higher substrate temperatures and in layers submitted to annealing. It was also found that crystallite size increases with layer thickness. However, for layers thicker than 2 to 3 μm the crystallite size is virtually independent of their thickness. In these layers the effect of grain growth boundaries on electron mobility decreases, and the principal factors diminishing mobility are defects of the crystal lattice.

The cardinal role of crystal lattice defects, probably associated with local departures from stoichiometry, is easily seen in layers obtained by flash evaporation. This technique produces InSb thin layers with electron mobilities lower than those obtained by the three temperature method [9].

In the present work, a mixture of powders, InSb+Sb, was used in the flash evaporation method in order to make feasible a comparison of the electrical properties obtained by means of both methods and to achieve high electron mobilities. In this way it was possible to evaporate stoichiometric layers of InSb onto substrates kept at the maximum temperature.

The electrical properties of InSb thin layers obtained by the flash evaporation method depended to the highest extent on the continuity and uniformity of the evaporation process. If during evaporation of the layers there were large fluctuations in the vapour chemical composition due to the mechanism supplying the powder to the vapour source, then no later thermal processing could produce layers of high electron mobility. Such layers, no matter what kind of annealing was applied, had low mobilities. The influence of fluctuations in the vapour composition on the properties of InSb thin layers is shown in Table III.

TABLE III

Series No.	μ_H [cm^2 V^{-1} s^{-1}]	R_H [cm^3 C^{-1}]	σ [Ω^{-1} cm^{-1}]	m
		Inhomogeneous vapour stream		
I	3300	29	86	6
II	2700	150	18	4
		Homogeneous vapour stream		
III	8400	290	29	5
VI	7500	154	49	4

This table gives the average values of the Hall coefficient, specific conductivity and mobility for four series of InSb thin layers. Each of the series had been obtained in a single evaporation procedure, m given in the last column defines the number of samples in the series, which were used for calculating the average values of R_H, σ, and μ_H. All samples were evaporated at substrate temperatures from 480 to 490 °C and Sb/In = 1.3 to 1.5. Series I and II were evaporated with large fluctuations of density and composition of the vapour stream, series III and IV with small fluctuations. The decrease in electron mobility in the series I and II cannot be associated with scattering at crystallite boundaries, for the samples in all four series had the same crystallite size.

It is an interesting fact that the layers evaporated with high vapour stream fluctuations can possess large Hall coefficients, even though their electron mobility is low. This means that during evaporation two kinds of defects may be formed, in approximately equal quantities, which act as donor and acceptor centers. These defects may arise because of local departures from stoichiometric composition. As is well known, in indium antimonide excess antimony atoms form donor centers, whereas excess indium atoms form acceptor centers. Of course, other defects are also possible, but the fact that the Hall coefficient in InSb thin layers of high mobility depends on the vapour composition during the evaporation process strongly supports this cause. The dependence of the electrical properties of InSb thin layers obtained by the three temperature method at substrate temperatures 480 and 490 °C on the chemical composition of the vapours is shown in Table IV.

The conditions under which the layers cited in this table were obtained are analogous to the previous ones, except for the chemical composition of the vapours. It is clear that although these layers had similar mobilities, they differed quite distinctly as regards specific conductivity and Hall coefficient. This may be explained by the increase in the compensation of donor and acceptor levels with a change in the vapour composition. It is evident that with decreasing Sb/In ratio the Hall coefficient increases and the specific conductivity falls.

TABLE IV

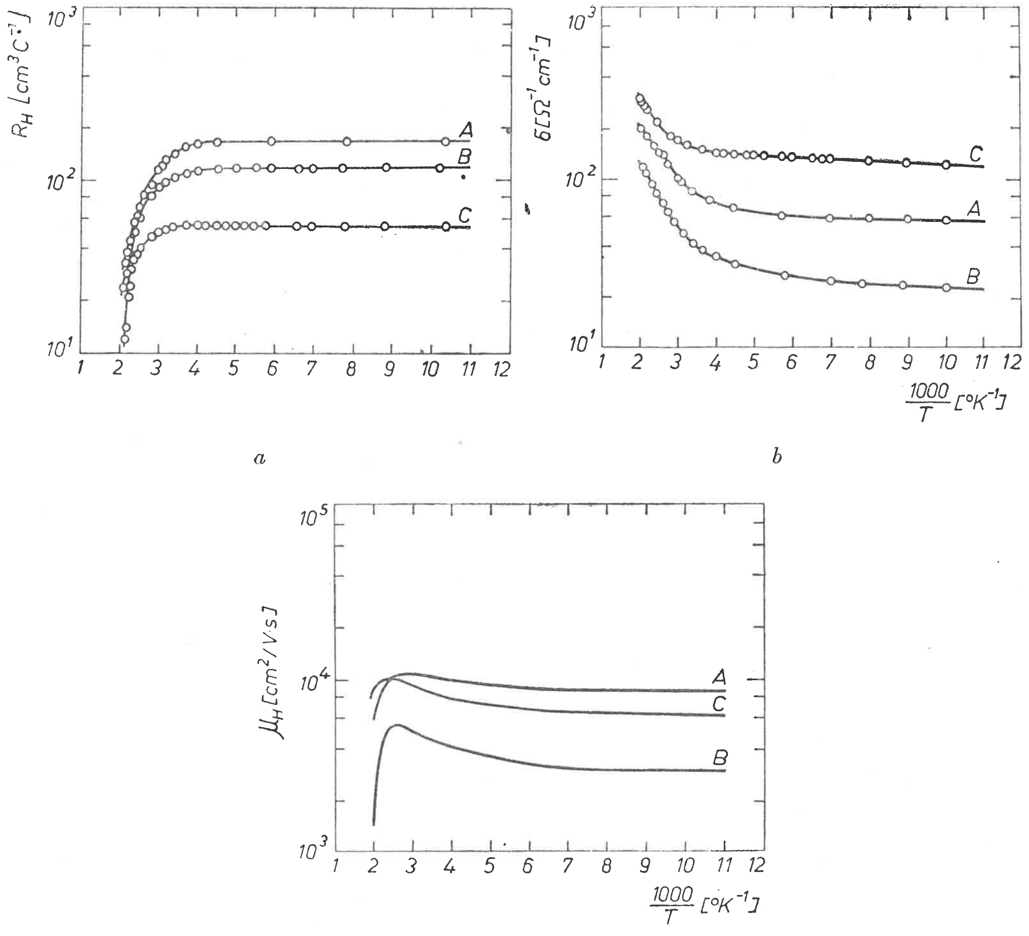
Layer	d [μm]	μ_H [cm^2 V^{-1} s^{-1}]	σ [Ω^{-1} cm^{-1}]	R [cm^3 C^{-1}]	Sb/In
			480°C		
G	2.8	5900	85	70	1.5
H	2.1	5700	60	94	1.35
I	2.1	6100	58	107	1.12
			490°C		
F	1.9	12300	86	145	2.2
B	1.8	9000	41	220	1.35
J	2.4	9400	41	230	1.24

4. *Temperature dependence of electron mobility*

Figure 2 presents the temperatures dependences of the Hall coefficient, conductivity and electron mobility for three InSb thin layers. These samples, labelled A, B, and C, had thicknesses of 1.9 μm, 2.8 μm, and 4.0 μm and electron concentrations at 100 °K of 3.7×10^{16} cm^{-3}, 5.2×10^{16} cm^{-3}, and 1.2×10^{17} cm^{-3}, respectively.

A comparison of the shapes of the temperature dependences of electrical properties of InSb thin layers with corresponding data for bulk InSb [10] shows that the dependence of R_H on temperature for thin layers and bulk n-InSb are analogous. Differences do appear, however, in the temperature dependence of the conductivity. In InSb thin layers a continuous rise in conductivity is observed at the transition from extrinsic conduction to intrinsic conduction, instead of a minimum of conductivity. This type of σ *versus* temperature dependence is characteristic of InSb thin layers independently of the technique used in their production.

Generally speaking, however, a well marked out transition from extrinsic to intrinsic conduction appears, only for layers of high electron mobility. In layers of low mobility there is no distinct transition between one and the other type of conduction, while the slope of the $\sigma = f(T)$ curve at low temperatures depends on the degree of compensation. This is shown in Fig. 3, where the results of measurements of conductivity are given as a function of temperature for three layers obtained at substrate temperatures of 400 °C. These layers had an electron mobility $\mu_H \approx 1000$ cm^2/V sec. The numbers beside the curves in Fig. 3 are the Sb/In ratios. The dependence $\sigma = f(T)$ on the chemical composition of the vapours, *i.e.* on the degree of compensation is shown. In contradistinction to the InSb layers of high mobility, the conductivity in layers of low mobility may vary within the 100 to 500 °K range by several orders of magnitude. A characteristic feature of InSb thin layers is the appearance of a maximum in the temperature dependence of mobility at high temperatures and a drop in mobility at low temperatures. A typical mobility *versus* temperature curve for these layers is shown in Fig. 2.



a

b

c

Fig. 2. Temperature dependence of: *a* — Hall coefficient, *b* — conductivity, *c* — electron mobility in InSb thin layers

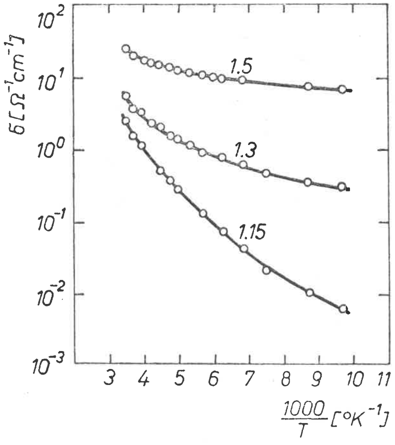


Fig. 3. Conductivity *versus* temperature of InSb layers of different degrees of compensation obtained at 400°C. The numbers beside the curves are the Sb/In ratios

Wieder [11] interpreted the dependence of μ_H on temperature using the Dexter–Seitz model which describes the process of scattering by edge dislocations. The crystallite boundaries were treated as high angle edge dislocations. In this picture the mobility restricted by edge dislocation scattering, μ_D, is a linear function of temperature: $\mu_D \sim \beta T$. The slope of the curve, β, depends on the temperature at which the mobility reaches a maximum value.

The studies of the temperature dependence of the mobility InSb thin layers made in this work, however, did not corroborate, the linear dependence between the slope of the curve $\mu_H = f(T)$ at low temperatures and the position of the mobility maximum. The results of measurements by Kasyan and Kot [3] also seem to confirm the lack of this type of dependence.

The results of the present research rather imply that the shape of the temperature *versus* electron mobility curve for thin layers of indium antimonide depends on the degree of compensation and the magnitude of the electron mobility.

REFERENCES

[1] K. G. Günther, *Z. Naturforsch.*, **13a**, 1081 (1958).
[2] K. G. Günther, H. Freller, *Z. Naturforsch.*, **16a**, 279 (1961).
[3] V. A. Kasyan, M. V. Kot, *Trudy Fiz. Poluprov.*, Kishinev. Gosuniver., **1**, 57 (1962).
[4] V. A. Kasyan, M. V. Kot, *Izv. Akad. Nauk SSSR*, **28**, 993 (1964).
[5] E. K. Müller, J. L. Richards, *J. Appl. Phys.*, **35**, 1233 (1964).
[6] W. J. Williamson, *Solid-State Electron.*, **9**, 213 (1966).
[7] R. Koike, R. Ueda, *Japan J. Appl. Phys.*, **3**, 191 (1964).
[8] M. Oszwałdowski, *Przegląd Elektroniki*, **7**, 329 (1968).
[9] H. Freller, K. G. Günther, *The Radio and Electron. Engineer.*, **9**, 97 (1967).
[10] V. V. Galavanov, D. N. Nasledov, A. S. Filipchenko, *Izv. Akad. Nauk SSSR*, **28**, 963 (1964).
[11] H. H. Wieder, *Solid-State Electron.*, **9**, 373 (1966).