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13. ABSTRACT (Maximum 200 words) Initial experiments aimed at determining the utility of adding Al to the growth surface showed no apparent advantage, thus the program was focused on synthesis of compounds containing Ga, Ti and As. Experiments using elemental group III sources and (100) GaAs substrates were performed at various temperatures ranging from 200°C-450°C. Attempts to increase the growth temperature while achieving similar Ti-incorporation behavior through the use of GaAs/TlAs and GaAs/TkGaAs superlattices were not successful. Analysis of the non-superlattice samples by electron microprobe (EMPA) showed evidence of Tl incorporation in all films grown at 400°C, as shown in Table I.			
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SYNTHESIS AND CHARACTERIZATION OF AlGaTlAs

GRANT NO. F49620-96-1-0344

FINAL REPORT
30 Sep 1999

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OBJECTIVES

The purpose of this program was to study the synthesis and material properties of AlGaTlAs for IR applications. It was hoped that the lower vapor pressures above these alloys would allow them to be grown lattice matched to InP substrates, and at higher growth temperatures than we have shown to be necessary for the InTlV alloys. The same questions of miscibility, environmental stability and interfacial control were to be explored for AlGaTlAs as described in the original proposal for InTlP and InTlAs. Layers were grown by metalorganic molecular beam epitaxy (MOMBE) using a combination of gaseous and elemental sources. Optimization of the growth process also included investigation of the effects of growth temperature and V/III ratio. The layers were characterized structurally (x-ray diffraction, scanning electron microscopy, transmission electron microscopy and atomic force microscopy), electrically (Hall measurement), chemically (electron microprobe analysis and Auger electron spectroscopy) and optically (photoluminescence and absorption). The goal was to make air stable alloys and then determine the lattice constant and bandgap as a function of Tl content.

RESULTS

Initial experiments aimed at determining the utility of adding Al to the growth surface showed no apparent advantage, thus the program was focused on synthesis of compounds containing Ga, Tl and As. Experiments using elemental Group III sources and (100) GaAs substrates were performed at various temperatures ranging from 200°C - 450°C. As for the InTlAs and InTlP systems, higher growth temperatures reduced the amount of Tl deposited at the surface, making ~375°C the upper limit for growth. However, unlike the InTlAs system, metallic Tl droplets are not observed, at least for lower Tl cell temperatures. In spite of the absence of Tl droplets, the surface morphologies for films grown at > 200°C are quite rough, and suggest 3D growth resulting in polycrystalline material, as shown in Figure 1. By contrast, films grown at 200°C are remarkably smooth. Imaging of this surface, and of those deposited at higher temperatures, shows no phase contrast suggesting that there is no spatial variation in the distribution of the Tl atoms. Increasing the Tl cell temperature from 485 to 545°C for a

growth temperature of 200°C produced a slightly rougher surface, also shown in Figure 1. This type of structure is more like that observed in the InTlV systems and usually suggests the presence of metallic Tl. This is supported by the appearance of one of the metallic Tl peaks in the powder x-ray diffraction scan of this sample. This peak is not evident in the sample grown at the lower Tl cell temperature. Even more surprising than the smooth morphology of the low Tl sample grown at 200°C is the absence of an oxidation induced change in the surface structure even after exposure to air for ~one week. All previous Tl-containing samples grown by GSMBE were found to oxidize quite rapidly, sometimes even in minutes, resulting in the appearance of a forest of faceted crystallites on the surface. The lack of oxidation in this sample further suggests that the bonding of the Tl in the low T GaTlAs is different than in the InTlV systems.

Attempts to increase the growth temperature while achieving similar Tl-incorporation behavior through the use of GaAs/TlAs and GaAs/TlGaAs superlattices were not successful. The Tl segregated to the surface during growth and oxidized upon removal from the system. This is similar to the behavior observed in the InTlV systems and is believed to be due to the inability of the Tl bonding orbitals to hybridize at the growth surface. As discussed in the final report for the parent grant (Grant No. F49620-96-1-0001), the most likely method of controlling the Tl behavior, particularly at higher growth temperatures will be the use of chemical precursors in which the Tl is already hybridized, such as triethylthallium (TETl).

Analysis of the non-superlattice samples by electron microprobe (EMPA) showed evidence of Tl incorporation in all films grown at < 400°C, as shown in Table I. The two Tl-containing samples grown at 200°C show Tl atomic fractions of ~ 5% and 15%. If these are zincblende alloys this would correspond to Tl mole fractions (X_{Tl}) of 12.5% and 31% respectively. Again unlike the case for the InTlV system, EMPA shows a uniform distribution of Tl across the GaTlAs samples, even those deposited at higher temperatures. In the InTlV materials, the Tl was always present in the form of pure metallic droplets. Some of the Tl incorporation, particularly at low growth temperatures, may be due to the incorporation of excess As in the films. None of the low T samples shows evidence of oxygen contamination.

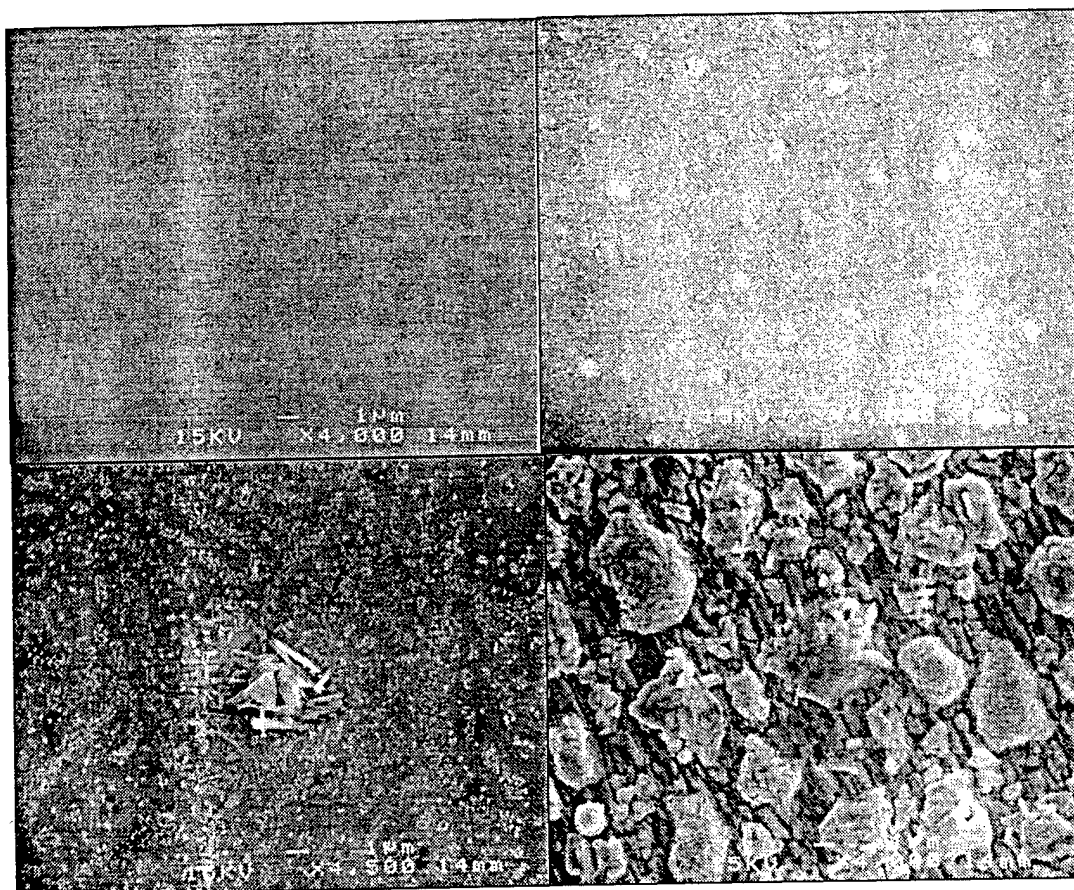


Figure 1. SEM micrographs of surfaces of Tl-containing samples. Upper left: GaTlAs grown at 200°C at a Tl cell temperature (T_{Tl}) of 485°C; lower left: GaTlAs grown at 200°C, $T_{\text{Tl}} = 545^\circ\text{C}$; upper right: GaTlAs grown at 275°C; $T_{\text{Tl}} = 505^\circ\text{C}$; lower right: InTlAs grown at 200°C for comparison.

TABLE I. Compositions of GaTlAs samples as determined by EMPA. All concentrations are in atomic percent.

Substrate Temperature (°C) /Tl Cell Temperature (°C)	Tl	Ga	As	O
200/0	0	42	58	0
200/485	4-7	35	60	0
200/545	14-16	33-35	55-57	0
275/505	7	46-47	46-47	0
325/515	4	48-49	46-48	0
375/530	~0.05	50-52	47-49	0
450/630	0	50	50	0
400/0	0	50	50	0

The air stability of the films deposited at low temperature remains excellent, as reflected in the surface morphologies shown in Figure 2, even after several months. This again indicates that the Tl is bonded in a manner different than that obtained in the InTlV alloys. Unfortunately, this alteration in bonding also results in an undesirable crystal structure. XTEM analysis, shown in Figure 3, indicates that the material is amorphous with small crystallites at the film/substrate interface. This is most likely due to the high As/Group III ratio in these films (as shown in Table I), and is similar to the situation obtained in low temperature GaAs which also shows a high As/Group III ratio. It is interesting to note that films grown at slightly higher temperatures (275° vs. 200°C) do not contain excess As and do not exhibit the air stability of the samples grown at lower temperatures. This suggests that the excess As is needed to induce the Tl to bond. This is in agreement with previously reported results on the growth of TlAs where only As-rich phases of TlAs_x could be obtained.

The optical properties of the GaTlAs films have been investigated using optical absorption. The absorbance data was converted to absorption coefficient, α , using Beer's Law. As shown in the plot of α vs. energy in Figure 4, the addition of Tl to the growth surface at low temperatures does appear to shift the band edge. The shift does not appear to be related to the amount of Tl in the film, with lower X_{Tl} producing a smaller bandgap. In fact, when control samples of GaAs grown under the same conditions without Tl are examined for comparison, it is clear that the shift in the absorption edge is due primarily to the presence of the excess As/amorphous GaAs structure since the same bandgap can be obtained with or without Tl. Thus even though the Tl seems to be stably bonded, it is not clear what role, if any, the Tl is playing in the band structure of the material.

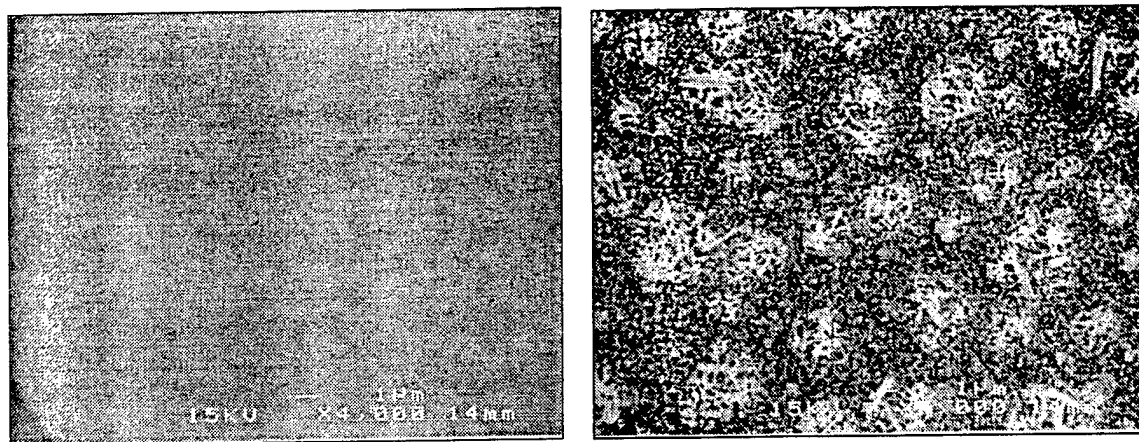


Figure 2. SEM micrographs of the surface of GaTlAs grown at 200°C (left) or 275°C (right) after exposure to air for one week.

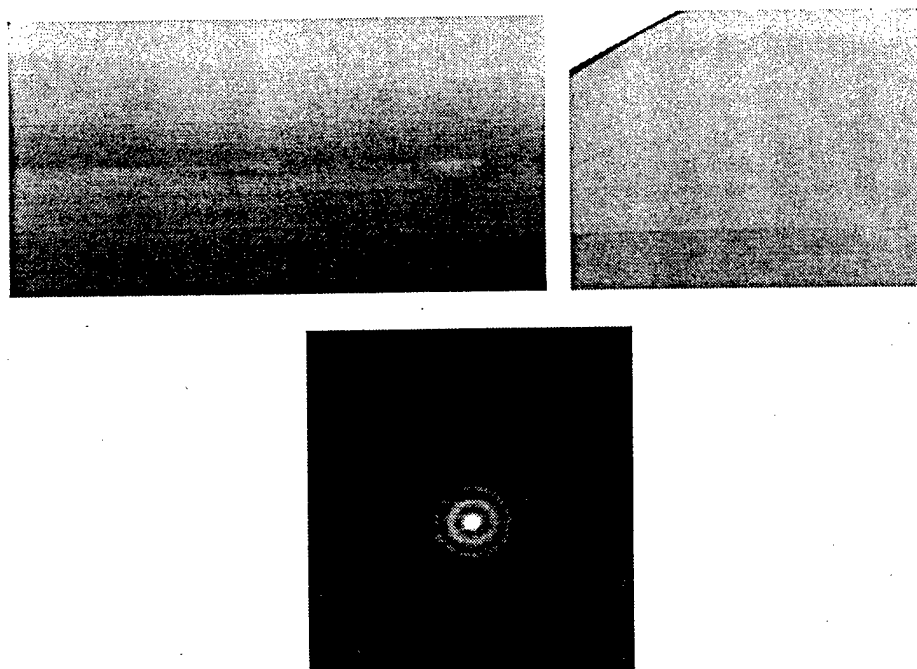


Figure 3. Cross-sectional TEM (XTEM) micrographs of 123 nm GaTlAs film grown at 200°C: (upper left) 100K mag. and (upper right) 300K mag. Selected Area Diffraction Pattern (bottom) shows the material to be amorphous except for the small crystallites located near the film/substrate interface.

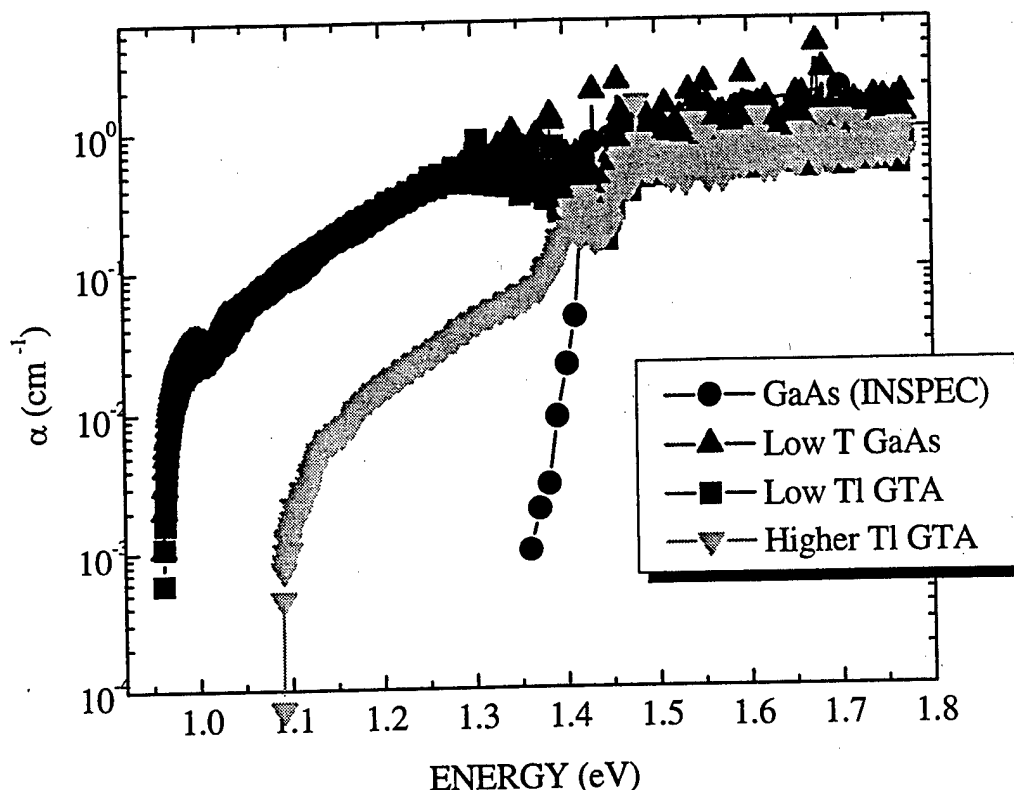


Figure 4. Absorption coefficient as a function of energy for various GaTlAs samples. Data for single crystal GaAs is shown for comparison (Data taken from INSPEC).

ACCOMPLISHMENTS

This program produced the first chemical evidence of an apparent single phase Tl-containing III-V compound which is air stable. The optical and structural characteristics of this material were determined and compared with similarly prepared GaAs.

PERSONNEL SUPPORTED

This program has supported one graduate student, S. M. Donovan, and has graduated one Ph.D. student, M. A. Antonell.

INTERACTIONS

We have been working closely with an AFOSR funded modeling program at SRI, Int. headed by Dr. A. Sher. The close interaction between these two efforts resulted in

refinement of the SRI model, while results of the SRI model have been used to narrow the experimental conditions to be investigated, thus saving considerable time and money.

PUBLICATIONS

"Growth of Tl-Containing III-V Materials by GSMBE," M. J. Antonell, C. R. Abernathy, W. A. Acree, M. A. Berding, A. Sher, M. Van Schilfgaarde, A. Sanjuro and K. Wong, J. Crystal Growth, **188**, 113 - 118, 1998.

"Synthesis and characterization of In-Tl-Sb compounds grown by molecular beam epitaxy," M. J. Antonell, C. R. Abernathy, W. A. Acree, M. A. Berding and A. Sher, J. Vac. Sci. & Tech. A, Vol. 17, pp. 338-341 (1999).

"Growth of Ga-Tl-As by Gas Source Molecular Beam Epitaxy," M.J. Antonell, B. Gila, K. Powers and C.R. Abernathy, submitted for publication.