LONG WAVELENGTH PbSnTe LASERS WITH CW OPERATION ABOVE 77K

Koji Shinohara, Mitsuo Yoshikawa,
Michiharu Ito and Ryuiti Ueda
Kansai Research Center
Fujitsu Laboratories Ltd., JAPAN

SUMMARY

Lead tin telluride diode lasers with emission wavelengths of 6 to 9 μm easily operate continuously at temperatures above 77K. These lasers have the $Pb_{1-y}Sn_yTe/Pb_{1-x}Sn_xTE/Pb_{1-y}Sn_yTe/PbTe$ (substrate), ($x > y$) double heterostructure.

To prepare this structure by LPE, the growth temperature must be below 600°C to suppress diffusion of the tin during the epitaxial growth.

When the heterojunctions are formed by the usual LPE method, the junction boundaries become irregular in the case for the lasers with wavelengths of over 10 μm at 77K.

In this paper, the mechanism by which the heterojunction boundaries become irregular is cleared and a new LPE method which prevents the irregularity is explained. The lasers prepared from the wafers grown by the new method have demonstrated CW operation at wavelengths longer than 10 μm above liquid nitrogen temperature.

INTRODUCTION

Lead tin telluride diode lasers are well known as excellent radiation sources in the wavelengths longer than 6 μm[1]. The diode lasers should be of double heterostructures to reduce the threshold current densities or to operate at temperatures above 77K[2]. Lasers with wavelengths from 6 to 10 μm operate easily above 77K and they are in practical use.

On the other hand, it is very difficult for the lasers to operate on CW with wavelengths over 10 μm above liquid nitrogen temperature. The reason for this is the difficulty of preparing flat heterojunctions for the lasers with wavelengths over 10 μm. The heterojunction boundaries become irregular if the junctions are made by the usual LPE method.

A new LPE method has been developed which overcomes the problem of irregular heterojunction boundaries.

By the new method, lasers capable of CW operation have been made at wavelengths longer than 10 μm at temperatures above 77K.

EXPERIMENTAL

Highly reliable lasers with mesa strata structure have been developed[3][4]. The structure of the laser is shown in Figure 1. The heterojunctions have been prepared by the liquid phase epitaxial growth method using graphite sliding boat. To confine the photons and carriers in the active layer, the proportion of SnTe which is denoted as X in the active layer is higher than Y which is the proportion of two confining layers. The insulating films have been made by the anodic oxidation[4].

The wavelength of the lasers is determined by the composition of the active region and the temperature. To make the wavelength longer than 10 μm at 77K, the value of X must be more than 0.2, while for wavelengths of 6 to 10 μm X is less than 0.2.

When the double heterojunctions are formed by the usual LPE technique, the shapes of the heterojunction boundaries vary obviously whether the composition of the active layer is over 0.2 or not.

Two photomicrographs of etched cross sections of double heterowafers are shown in Figure 2. Both are grown by the same LPE method. One is an example of the wafers for the 6 to 10 μm lasers, and the others is an example of the wafers for over 10 μm.

Heterojunctions contain many dislocations, which is due to the misfit of the lattice constant. The misfit dislocations between the substrate and the first layer spread in the direction of the crystal growth. This is due to the mutual self diffusion of Pb and Sn[5]. The growth temperature of the second and the third layers must be low enough to suppress mutual diffusion. Therefore, the active region is grown at a temperature of 600°C.

A photomicrograph for the long wavelength laser shows that the heterojunction boundaries between the first and second layers and between the second and third layers are irregular. When the heterojunction boundaries are irregular, the scattering loss that results becomes so serious that lasers made from wafers shown in Figure 2A do not operate even at 20K. But lasers with wavelengths from 6 to 10 μm, prepared from the wafer with flat heterojunctions shown in Figure 2B, give good CW operation above 77K.

To confirm the cause of irregularity in wafers for long wavelength lasers, another experiment was performed. The result is shown in Figure 3. The first layer of Y = 0.13 was grown all over the surface of the PbTe substrate. After removing the solution for the first layer, the solution for the second layer was kept on just half of the first epitaxial layer for 1 minute isothermally at 600°C. Then the solution was removed. The surface where the second solution was kept on is uneven, while the remaining half of the surface is smooth. The cross sectional view shows that at some places, the melting back occurs as was expected, and moreover, epitaxial growth occurs near the melted back area.

From the measurement of the lattice constant by the X-ray diffraction technique, the composition of the precipitated layer was determined to be $x = 0.25$.

From this experiment, the cause of irregularity is found to be due not only to the melting back but also the precipitation. Of course, the saturated solutions were used for the epitaxial growth, but even with these saturated solutions, both melting back and precipitation occurred.

The mechanism of the melting back and precipitation using saturated solutions can be explained by the phase diagram.

The ternary phase diagram of Pb, Sn and Te was investigated in detail by Harris, et al.[6].

The temperature of the epitaxial growth of the second and third layers is below 600°C, so the region below the 600°C isothermal liquidus line must be considered. Note that the isocompositional solidus lines curve sharply where the composition is over $x = 0.2$.

First, the irregularity of the heterojunction boundary between the first and second epitaxial layers is considered.

The ternary phase diagram which is related to the first and second layers is shown in Figure 4. When the first layer is already grown and the solution for the second layer makes contact with the first layer, the first layer is in equilibrium with the solution whose composition is denoted by the point B where $Y = 0.13$ solidus intercepts the 600°C liquidus. So, the first layer is not in equilibrium with the solution at the point A where $x = 0.25$ solidus intercepts the 600°C liquidus, which is the composition of the second solution. The first composition and this second composition are so different that a disequilibrium emerges instantaneously when the second solution is held on the first epitaxial layer. To restore equilibrium between the solid and the liquid, the composition of the second solution changes from the point A toward the point B along the 600°C isothermal liquidus line.

In order to change the composition of this solution, the molecular interchange between the solid and the liquid of different composition is required.

By the local melting back of the first layer of $Y = 0.13$, the composition of the solution moves from the point A toward the point C which is the composition of the first layer. This compositional change is indicated by the vector a. On the other hand, by the precipitation of $x = 0.25$, the composition of the solution moves from the point A against the point D which is the composition of the second layer. This compositional change due to the precipitation is indicated by the vector b.

When the melting back and precipitation occur simultaneously, the motive force of the compositional change along the 600°C isothermal liquidus emerges. This is the reason why the interface of the first and second layers becomes irregular.

The mechanism of the irregularity between the second and the third layers is just the reverse mechanism of the aforementioned irregularity at the interface of the first and second layer; that is, the composition of the third solution moves toward the composition of the second solution along the 600°C liquidus line.

All this explains why the heterojunctions become irregular when double heterowafers are produced by the usual LPE method.

However, a way has been found to produce flat heterojunction boundaries on double heterowafers for the 6 to 10 μm lasers.

The reason this can be done can be understood from the phase diagram. The isocompositional lines are straight below $x = 0.20$. So, the compositional difference of the solutions between the first layer and the second layer is not so large, the result being that serious disequilibrium does not occur. Of course, the solution composition for the second layer moves toward that of the first solution, but the motive force is so small that the boundaries do not become irregular.

From the aforementioned analysis, it is seen that to obtain flat heterojunction boundaries for the long wavelength laser, it is necessary to produce precipitation only, while suppressing the melting back. So, supercooled solutions for the growth of the second and third layers were used in order to allow no time for compositional change.

The temperature profile for the usual LPE growth method is shown in Figure 5. The first layer is grown slowly to assure the quality of the epitaxial layer, then the second and third layers are grown continuously at the cooling rate of 1°C per minute.

The modified temperature profile in order to achieve the supercooled condition is shown in Figure 6. The beginning half of the first layer is grown under the usual conditions, but in the middle point of the first layer growth, the cooling rate is changed to as much as 3°C per minute. While the last half of the first layer is growing, the solutions for the second and third layers are sufficiently supercooled prior to making contact with the substrate. Then the second and the third layers are grown continuously.

The cooling rate of 3°C per minute is sufficient to keep the solutions supercooled.

A photomicrograph of the cross section grown by the new LPE method is shown in Figure 7. Notice how flat the heterojunction boundaries are.

Lasers prepared from the wafers like this easily operate on CW with wavelengths longer than 10 μm above 77K.

In Figure 8, one example of the lasing spectrum is shown demonstrating CW operation on an 11.45 μm wavelength with single mode. The temperature of the heat sink is 77K.

CONCLUDING REMARKS

The mechanism by which heterojunction boundaries become irregular was shown and a new method was explained of liquid phase epitaxial growth which prevents this irregularity.

Lasers prepared from the wafers grown by this new LPE method have demonstrated CW operation at wavelengths longer than 10 μm above 77K.

We assume that lasers with emission wavelengths longer than 16 μm can also be operated continuously above 77K if necessary.

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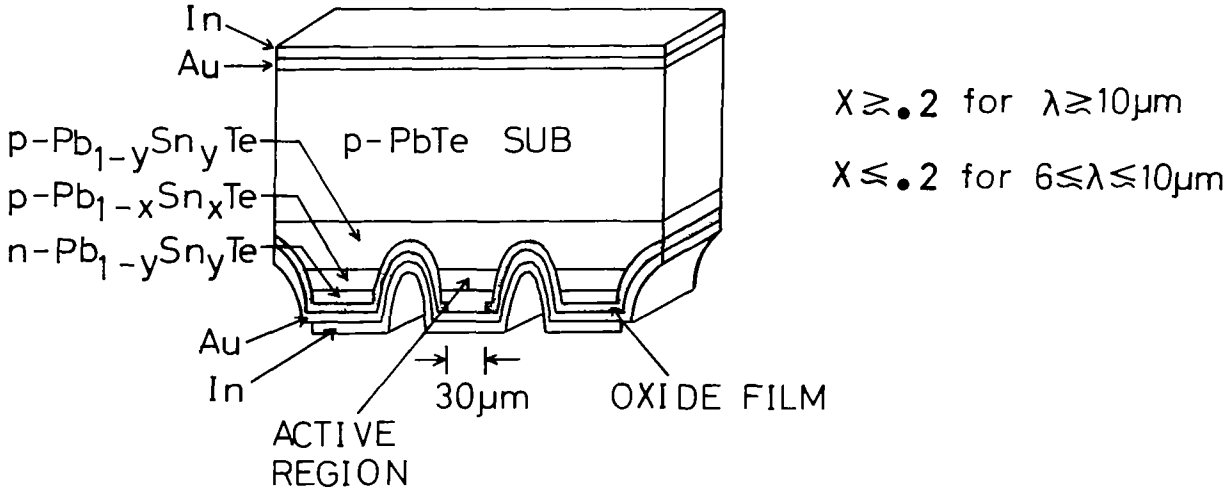
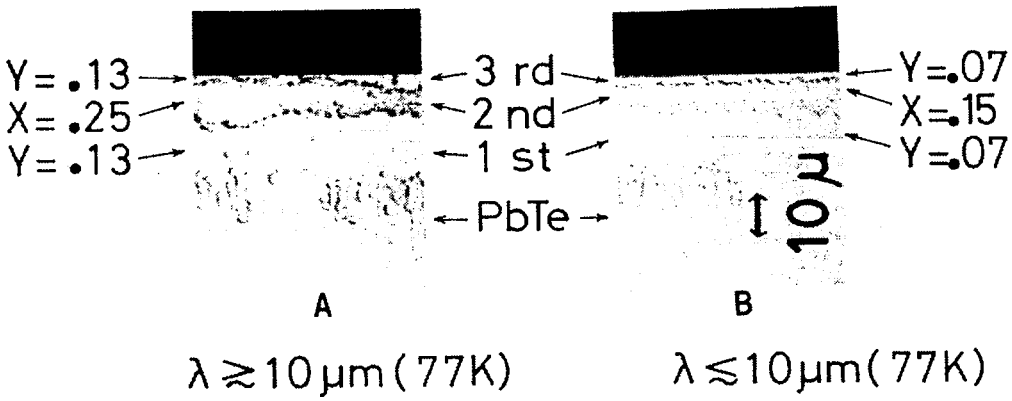


Figure 1.- Schematic structure of the laser diode.



A is an example of the wafers for over 10 μm at 77K

B is an example of the wafers for 6 to 10 μm

Figure 2.- Etched cross sections of double heterowafers.

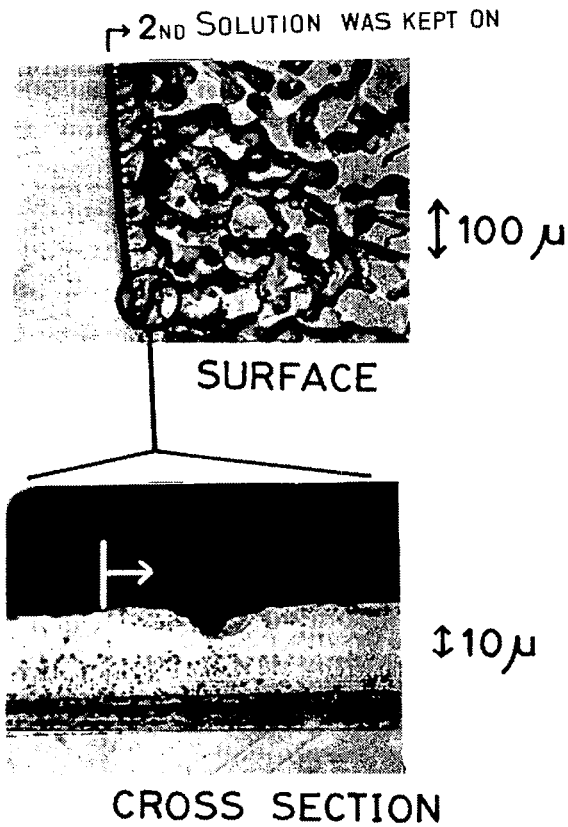
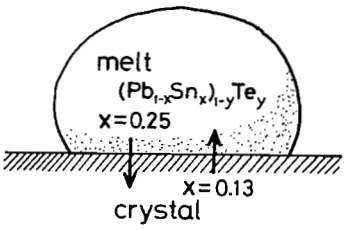


Figure 3.- Heterointerface morphology.



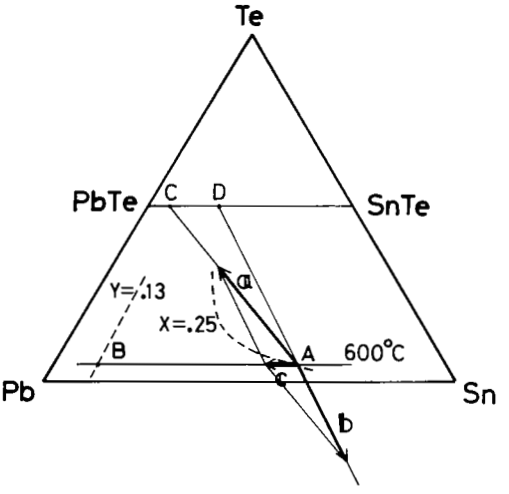


Figure 4.- Ternary phase diagram of Pb-Sn-Te explaining the heterojunction boundary's irregularity.

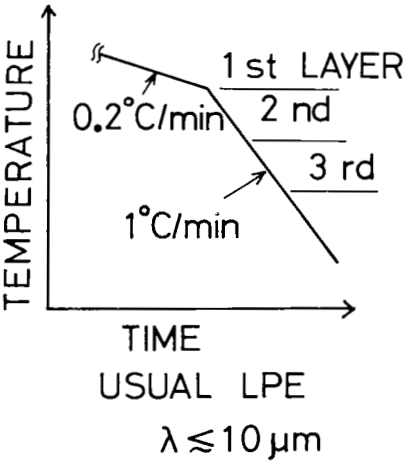


Figure 5.- The temperature profile of usual LPE method.

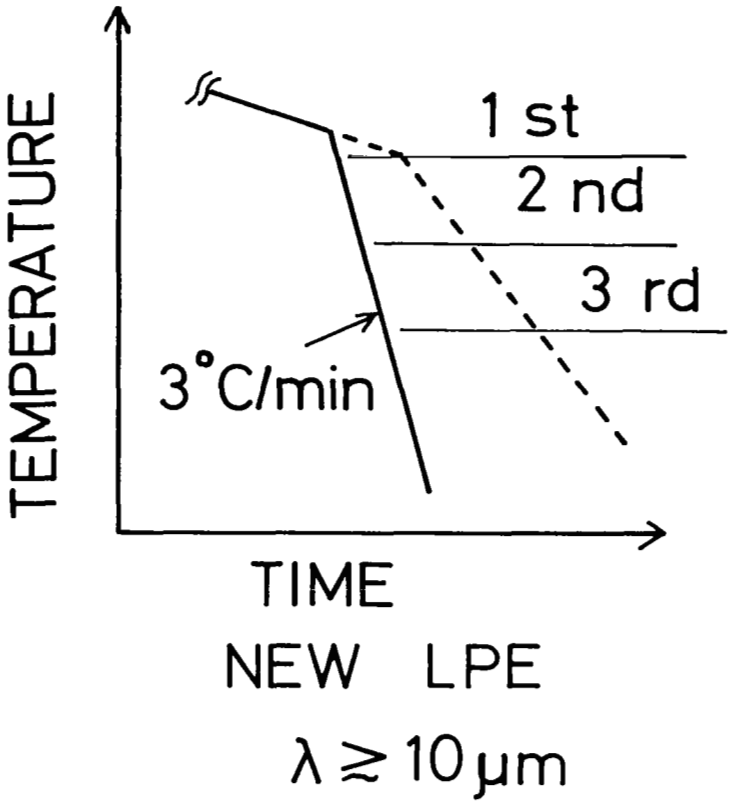


Figure 6.- The temperature profile of the new LPE method.

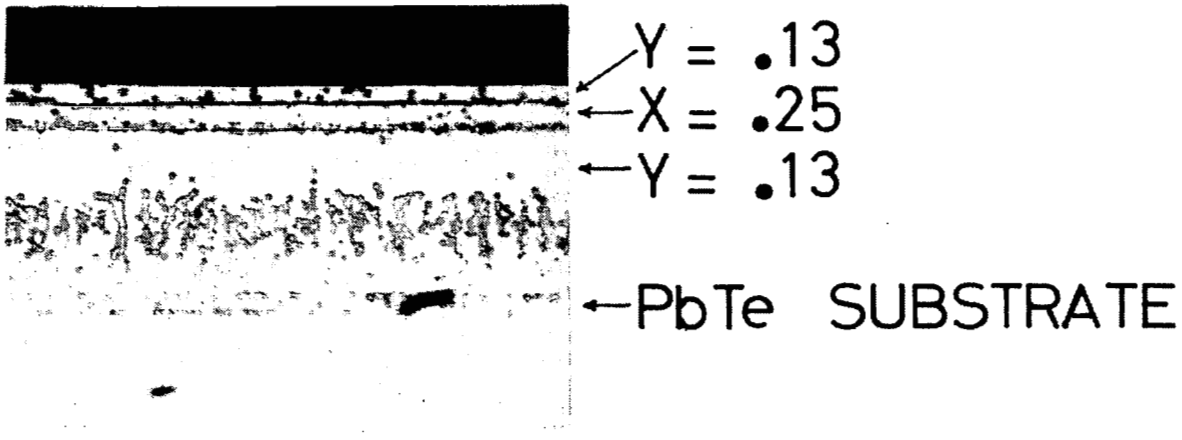


Figure 7.- Etched cross section of the double heterowafer for the long wavelength lasers grown by the new LPE method.

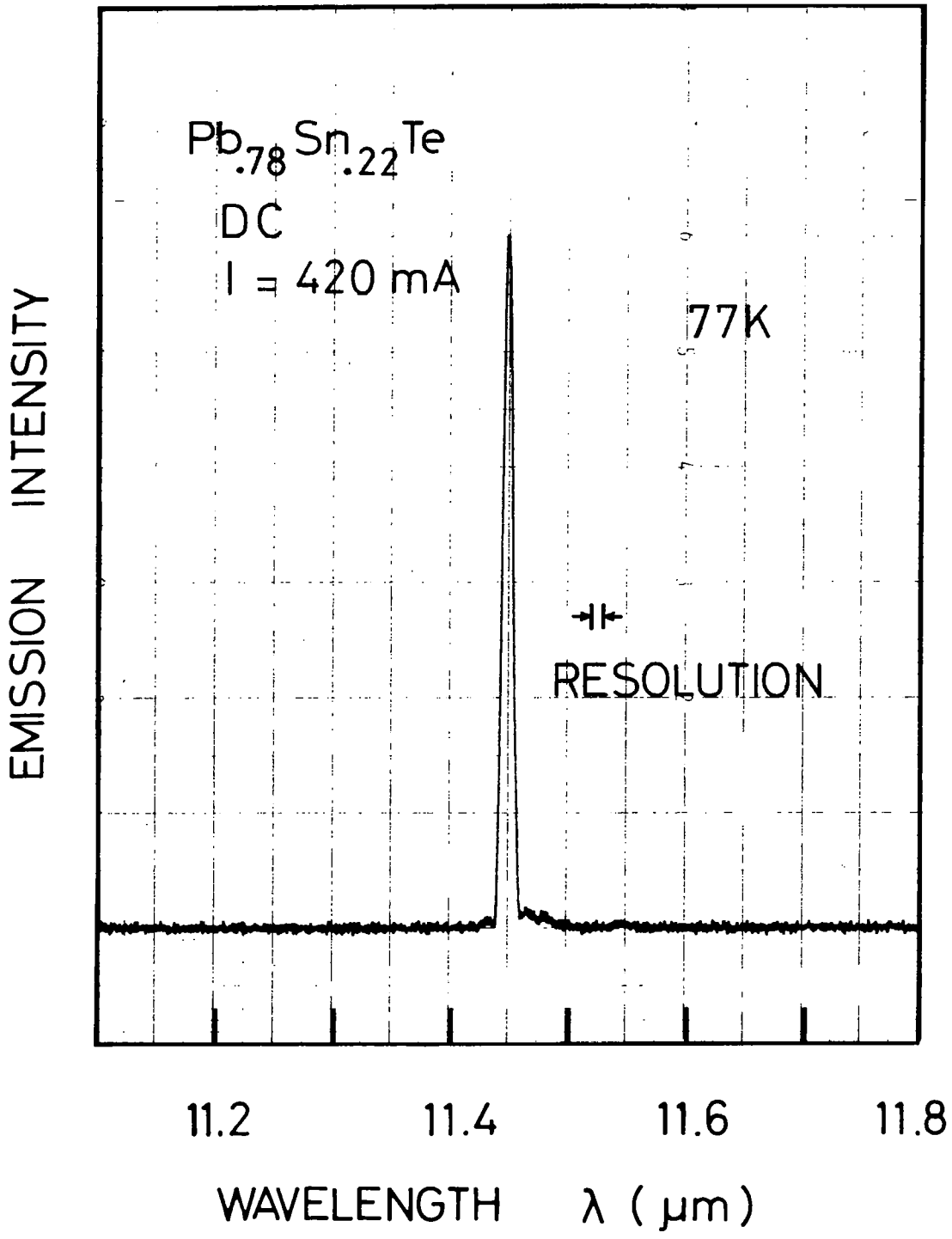


Figure 8.- One example of the lasing spectrum.