

LETTER TO THE EDITORS

GROWTH OF Tl DOPED PbTe SINGLE CRYSTALS BY THE TRAVELLING HEATER METHOD

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Single crystals of Tl doped PbTe were grown using the travelling heater method (THM) and the electrical properties were evaluated at 77 K and found to be p-type.

The travelling heater method (THM) has proved to be a suitable technique to grow highly perfect single crystals in a great variety of compounds, especially of II–VI and III–V compounds. It offers interesting advantages such as higher homogeneity in composition and lower growth temperatures, leading to lower concentrations of lattice defects and impurities.

Only a few attempts at growing lead chalcogenides or lead tin chalcogenides, the most important IV–VI compounds for optoelectronic devices, have been tried by this method [1–4]. There has been a certain interest in thallium doped PbTe as a substrate material for lead tin telluride laser diodes [5,6]. We prepared Tl doped PbTe single crystals of high structural perfection, i.e., without low-angle boundaries which have been shown to usually result with Bridgman grown crystals of the same material [7].

PbTe was synthesized in evacuated silica ampoules from 5N materials at about 1000°C with a tellurium excess of 0.5 at%. It was followed by a normal freezing procedure to prepare the feed material. Together with the synthesized PbTe, the ampoule was charged with 0.5 at% 5N thallium to dope the source material for use by the THM method. During melting the excess of tellurium and the doping element are assumed to form a thallium tellurium compound. After a relatively fast normal freezing process or even after quenching the ampoule, the usual THM arrangement consisting of a seed, a 4 to 5 mm long initial Te

zone and the doped feed material (all 12 mm diameter) was put into a silica ampoule. The evacuated and closed ampoule was placed in a small vertical resistance heater without any after-heater or similar equipment which allowed good observation during growth. With a zone temperature of about 550°C, the initial Te zone doubled its length to about 10 mm according to the phase diagram which is characterized by the PbTe congruent melting point of about 924°C and an eutectic composition with 85.5 at% Te at 405°C.

The average temperature gradient at the solid–liquid interface was measured to be ap-

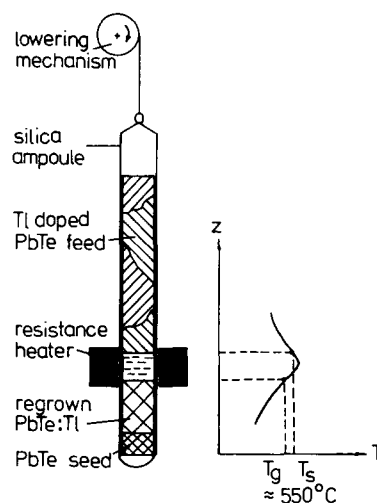


Fig. 1. Schematic representation of the crystal growth arrangement for growing Tl doped PbTe single crystals.

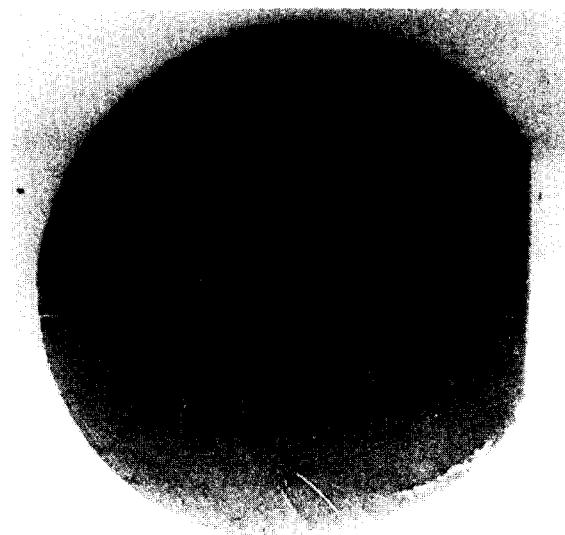


Fig. 2. (420) X-ray reflection topograph of a (100) surface of PbTe doped with thallium (12 mm diameter).

proximately 100 K/cm, depending on the zone position relative to the initial ingot. The velocity of the ampoule, driven by a low-speed synchronous motor with speed reducer, was 2 mm/day. It is considered a usual growth rate for THM [8] which is limited by the diffusion-defined transport and the great amount of excess component in the liquid zone. Single crystals were grown up to a total length of 10 cm, limited only by the length of the source material.

Besides the crystal growth experiments from a PbTe seed (doped or undoped) with (100) orientation, spontaneous nucleation from the lower tip of the ampoule was utilized in some cases. There were no differences with respect to the structural perfection. Whereas all crystals grown from a (100) seed showed the same orientation as the seed,

without seeding no preferred orientation of the growing ingot could be observed. The growth directions were inclined with about 10° to 35° from $\langle 100 \rangle$, but the number of experiments does not allow a statistic analysis.

The thallium doped PbTe crystals grown by THM were highly perfect without low-angle boundaries. Fig. 2 shows a (420) X-ray reflection topograph of a 12 mm diameter slice after spark-erosive cutting and chemical polishing. The etch pit density of (100) surfaces (etched after Ohtsuki et al. [9]) was about 10^4 up to 10^6 cm^{-2} , which is assumed to depend on the relatively high thallium content. All crystals were p-type with a carrier concentration of some 10^{18} to $2 \times 10^{19} \text{ cm}^{-3}$ and an average Hall mobility of about $5 \times 10^3 \text{ cm}^2/\text{V} \cdot \text{s}$ (all values determined at 77 K). Increasing carrier concentrations are due to Tl segregation over the crystal length ($k_0 < 1$).

References

- [1] H. Jäger and E. Seipp, *Verhandl. Deut. Physik. Ges.* 1(1978) 86.
- [2] P. Rudolph, M. Mühlberg, D. Schikora and A. Engel, in: *ICCG-6, Moscow 1980, Extended Abstracts, Vol. III*, p. 224.
- [3] R. Triboulet et al., in: *Proc. 4th Intern. Conf. on the Physics of Narrow-Gap Semiconductors, Linz, 1981*.
- [4] I.P. Kazakov and O.I. Rachmatov, in: *Kosmičeskaja Tehnologija i Materialovedenie (Moscow, 1982)* p. 143.
- [5] S.H. Groves, K.W. Nill and A.J. Strauss, *Appl. Phys. Letters* 25 (1974) 331.
- [6] L.R. Tomasetta and C.G. Fonstad, *Appl. Phys. Letters* 25 (1974) 440.
- [7] M. Mühlberg, *Kristall Tech.* 15 (1980) 565.
- [8] G.A. Wolff and A.I. Mlavsky, in: *Crystal Growth, Theory and Techniques, Vol. 1*, Ed. C.H.L. Goodman (Plenum, London, 1974) p. 193.
- [9] O. Ohtsuki, K. Shinohara and O. Rynzan, *J. Appl. Phys.* 10 (1971) 515.