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High magnetic fields and oxide single crystals*

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Abstract

We present here the design and development of a 15-tesla cryocooled superconducting magnet by employing superconducting Nb_3Sn single-coil $Bi_2Ca_2Sr_2Cu_3O_{10}$ high-T_c leads and an optical window for irradiation purpose Temperature variation of magnetoresistance for a thin film as well as for a polycrystalline pellet is measured to test the performance of the magnet Floating zone melting crystal growth facility is established by using infrared image furnace to grow high melting oxide single crystals for colossal magnetoresistance measurements Typical results like the growth of lanthanide manganite single crystals and their resistivity and magnetization properties are presented

Keywords Colossal magnetoresistance lanthanide manganites, oxide single crystals cryocooled superconducting magnet floating zone melting

1 Introduction

1 1 *Cryocooled superconducting magnet*

The recent discovery of colossal magnetoresistance (CMR) in distorted perovskite manganites[1 3] $Ln_{1-x}A_xMnO_3$ (Ln rare-earth ion, A divalent cations such as Ca, Sr, Ba, Pb) has attracted considerable attention as they exhibit several novel features including CMR For the past few years, a variety of manganites and cobaltites have been investigated in the form of polycrystalline powders, films as well as single crystals

Superconducting magnets employing Nb_3Sn coils immersed in liquid helium have been designed and fabricated to yield very high (above 10 tesla) magnetic fields [4] Copper wire is used as connecting leads in these magnets Due to heat losses through the leads, the magnets consume large quantities of liquid helium After the discovery of high-T_c superconducting materials such as $YBa_2Cu_3O_7$, $Bi_2SrCa_2Cu_3O_8$ and $Bi_2Sr_2Ca_2Cu_3O_{10}$, copper-connecting leads are being replaced by high-T_c ceramic superconductor leads to reduce thermal losses up to 90% Close-cycle refrigerator systems reaching 4 2 K with a cooling capacity of 1 0 Watt at 4 2 K have been fabricated by employing solid-state conduction cooling From the engineering point of view, one can design a superconducting magnet by employing the conventional Nb_3Sn coil and high-T_c superconductor leads so that the magnet can work without liquid helium Room-temperature bore magnets with variable magnetic fields, essential for researchers working at higher temperatures, are difficult to obtain with the conventional immersion-type superconducting magnet technologies, where the consumption of liquid helium is high It is therefore

*Text of the lecture delivered at the Annual Faculty meeting of the Jawaharlal Nehru Centre for Advanced Scientific Research at Bangalore, on October 21, 1999 most useful to have magnets to produce high magnetic fields without liquid helium to minimize running expenditure and also to alleviate problems encountered due to the non-availability of liquid helium in many countries To this end, there have been efforts to produce cryocooled superconducting magnets with NbTi wires to achieve fields of the order of 9 tesla or lower[5, 6] A key element in designing such a magnet is the use of high-T_c superconductor leads along with a cryocooled system such as the Gifford–McMahon (GM) refrigeration cycle to cool the superconducting coil to 4 2 K and high-T_c leads to below 60 K

There have been some efforts in design,[7–10] but in all of them a three-coil system has been employed since currents of the order of 240 amp could not be passed to achieve a field of 15 tesla and above In these designs, the size of the magnet and the thermal loss were high To compensate the losses, two cryocooled refrigerators of 1 0 W at 4 2 K have been employed, but this increases the weight as well as the cost of the magnet considerably

1 2 *Floating zone melting crystal growth*

Single crystals of oxide materials are widely used in electronic, magnetic and optical communication industries Since they contain no grain boundaries, they give maximum performance in functional devices such as laser oscillators, surface acoustic wave devices, optical switches, second harmonic generators, etc Therefore, it is necessary to establish techniques to grow large and high-quality single crystals of these oxide materials One of the best crystal growth techniques is the floating zone melting (FZM) crystal growth[11, 13] This technique does not use crucible and seed and is a unique method of growing very high melting point materials It is very ideal for oxide materials exhibiting superconductivity and colossal magnetoresistance These oxides may evaporate slowly over a period of time resulting in compositional inhomogeneity in the grown crystal if techniques like Czochralski are employed In the FZM technique the molten portion will be converted into crystalline form within a short time and a new portion will be passing into the hot zone continuously Therefore, the loss of different elements in the molten zone due to evaporation will be minimal compared to Czochralski technique

2 Design and fabrication of superconducting magnet

We have designed and fabricated a versatile magnet giving fields up to 15 tesla by employing Nb_3Sn wire, high-T_c $Bi_2Sr_2Ca_2Cu_3O_{10}$ (2223) leads and a close-cycle helium refrigerator of 1 W at 4 2 K which has several unique features[14]

In the design of the magnet, we have adapted a single-coil system by carefully choosing the Nb_3Sn multifilamentary core of 1 0-mm dia fabricated by modified jelly-roll process with 53% copper content to allow easy heat dissipation We have chosen this wire to pass more current (up to 240 amp) as well as to minimize the coil temperature below 60 K when the coil quench occurs at high currents The coil was wound on stainless-steel drum by adapting wind and react process The specifications of the wire and full details of the system are given elsewhere[14]

Leads made of 2223 high-T_c bismuth cuprate with a current-carrying capacity 300 amp at 77 K were connected between the Nb_3Sn coil and the copper-end terminals The first stage

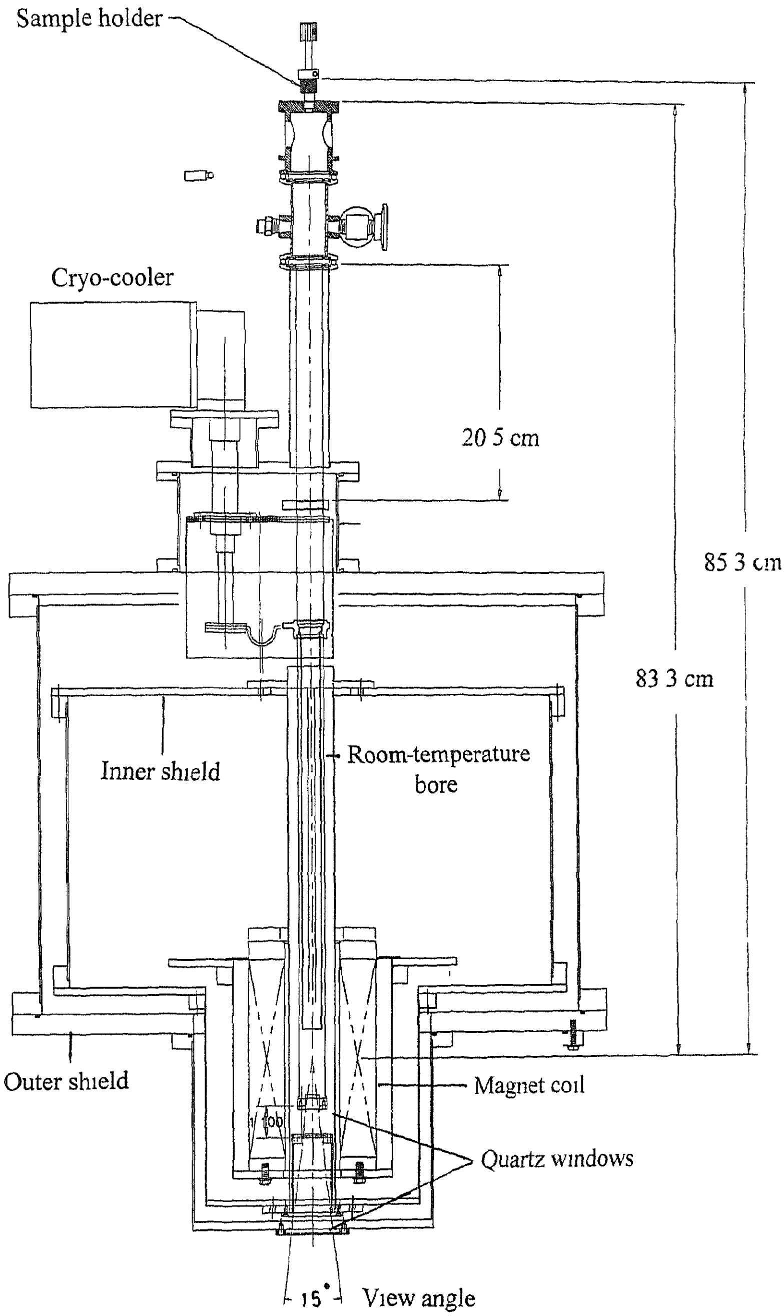
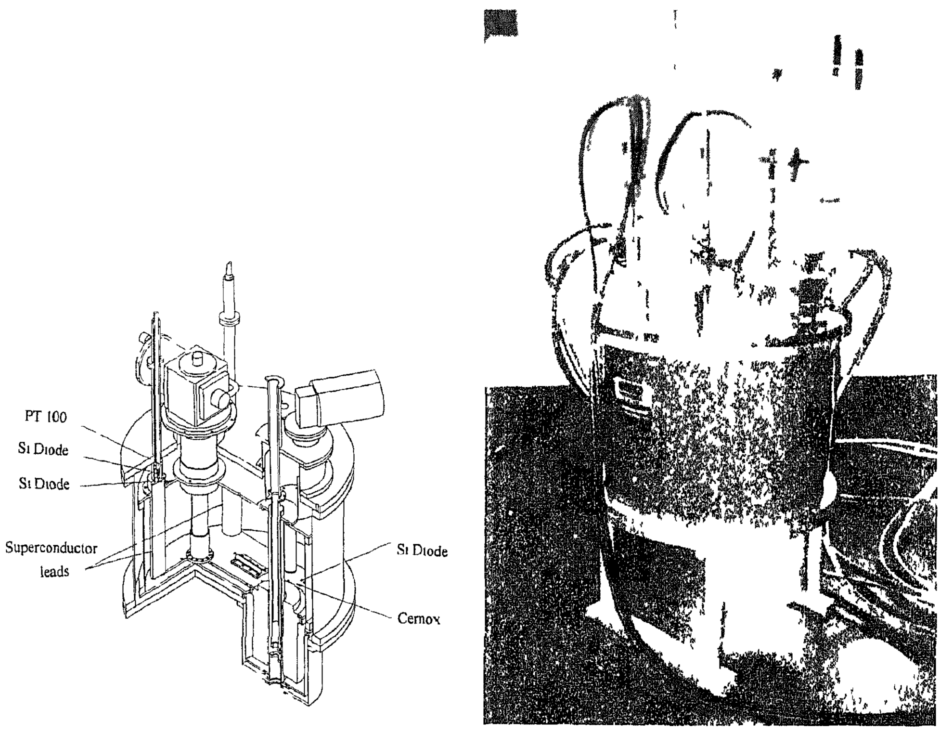


FIG 1 Schematic diagram of the cross-sectional view of a 15-tesla cryocooled superconducting magnet with the resistivity attachment

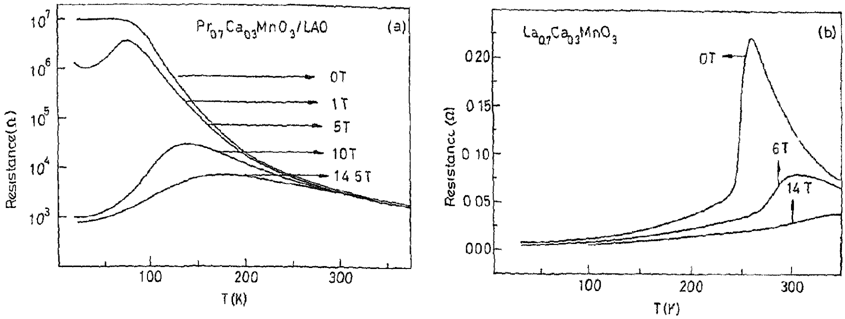
of a GM Sumitomo cryocooler (model SRDK-408 with 1 0-Watt cooling capacity at 4 2 K) was coupled to the high-T_c leads and the second stage was connected to the Nb_3Sn magnet coil The temperature of the coil was maintained below 4 2 K while energizing or de-energizing the magnet at a rate 0 04 amp/s or lower We have also used superinsulation of 15 layers around the superconducting coil to eliminate thermal losses The cryostat has been provided with a 52-mm bore with a quartz window of 20-mm dia incorporated at the bottom of the coil to enable light to pass through (Fig 1)

With this design we have achieved the superconducting coil temperature below 3 5 K and high-T_c leads temperature below 42 K before energizing the magnet When the magnet was energized to 15 tesla, the coil temperature went up to 4 0 K and the leads temperature to 47 K We could operate the magnet at this field for a few hours before it quenched The magnet works continuously without any interruption, when the field is reduced to 14 5 tesla (220 amp) The temperatures of the magnet coil and the high-T_c leads at this field strength were 4 and 45 K, respectively The three-dimensional cross-sectional view of the magnet and the photograph of the fully assembled and working magnet are shown in Fig 2 We have used silicon diodes, cernox and Pt sensors at crucial places to monitor the temperature of each section and also to control the temperature of the sample from15 to 500 K



(a) (b)

FIG 2 (a) A three-dimensional cross-sectional view of the cryocooled superconducting magnet and (b) the assembled 15-tesla magnet



(a) (b)

FIG 3 Temperature variation of magnetoresistance of (a) $Pr_{0.7}Ca_{0.3}MnO_3$ film on $LaAlO_3$ single crystal substrate and (b) $La_{0.7}Ca_{0.3}MnO_3$ pellet for different field strengths

We have carried out colossal magnetoresistance measurements at different fields and at different temperatures starting from 375 to 15 K by using CTI cryocooler model D22-8200

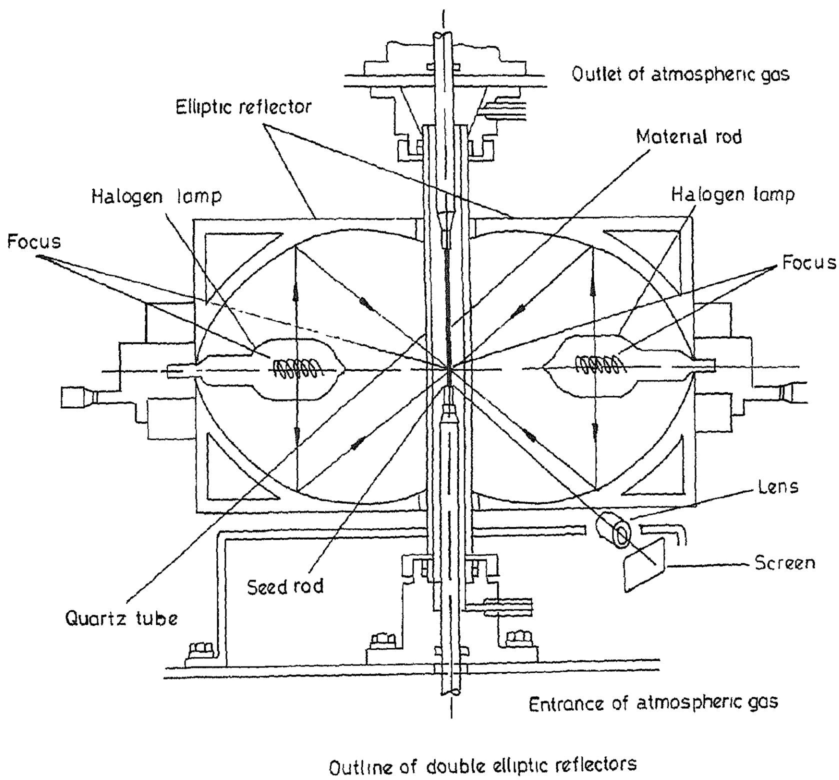


FIG 4 Schematic diagram of the infrared image furnace with two parabolic mirrors and the floating zone melting crystal growth arrangement

Typical magnetoresistance measurements at different temperatures and at different fields for $Pr_{0.7}Ca_{0.3}MnO_3$ thin film deposited on $LaAlO_3$ (100) substrate and $La_{0.7}Ca_{0.3}MnO_3$ polycrystalline pellet are shown in Fig 3 The performance of the system is excellent and we are using the magnet for the past 5,000 hours without any hindrance

Watanabe *et al* [15] have achieved 15-tesla field by employing three coils and two cryocooled systems The precooling time of the coil from room temperature to 4 K was around 110 h We have employed a single coil and a single cryocooled system and achieved the same field and also lowered the precooling time to 13 h The performance of our magnet is superior to Watanabe's magnet in relation to precooling time and cost

3 Floating zone melting crystal growth

The FZM instrument model SC-M35HD of Nichden Machinery has provision to carry out crystal growth at high pressures and at different environments The maximum melting point achievable is about 2400 K by infrared radiation Two parabolic reflectors with halogen lamps were employed in this infrared imaging furnace (Fig 4)

In the FZM crystal growth technique, the stoichiometric composition of the starting materials of high purity are weighed and mixed thoroughly followed by calcination, grinding and re-calcination process The powders are made in the form of rods by hydraulic press and sintered to high density The well-sintered rods are employed as seed and feed rods The inter-

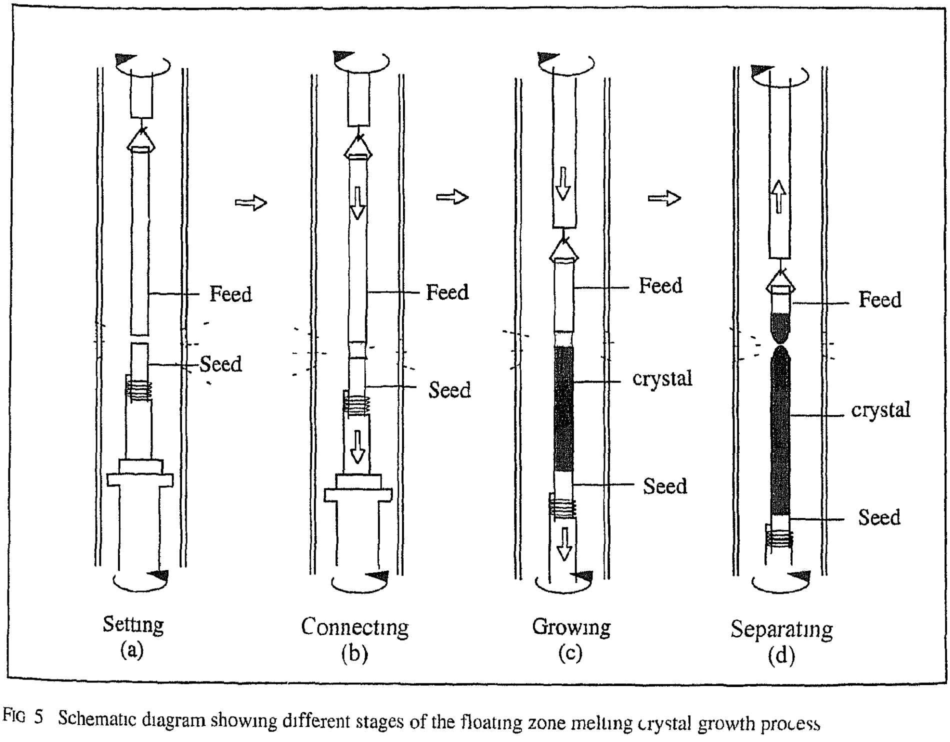
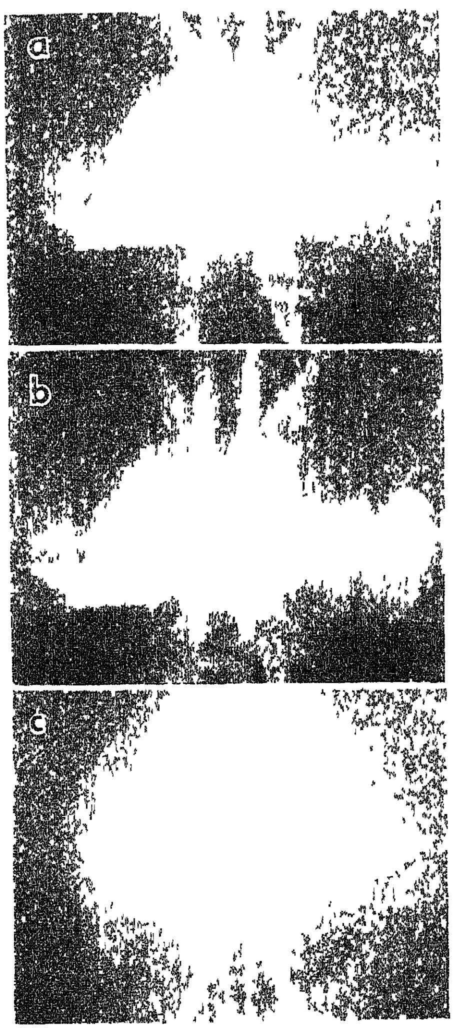


FIG 5 Schematic diagram showing different stages of the floating zone melting crystal growth process

face of the seed rod and the feed rod is melted initially and this molten zone is passed through the full feed rod with a constant speed. The feed rod and the seed rod will keep rotating mutually in opposite direction throughout the crystal growth process to keep the molten zone mix homogeneously (Fig. 5). The diameter of the crystals can be reduced by rotating the upper feed rod faster than the seed rod.[13] The growth direction can either be up or downwards but we have used the downward direction as the molten volume is more stable in this configuration. Crystals as big as 12-mm dia and 170-mm long can be grown at or below 2400 K. Photographs of the growth process at different stages for one of the crystals grown in this laboratory are shown in Fig. 6. Single crystals of $La_{0.9}MnO_3$, $Nd_{0.5}Ca_{0.5}MnO_3$ and $Nd_{0.45}Sr_{0.55}MnO_3$ were grown by using this technique. The temperature variation of resistance and magnetization for an $La_{0.9}MnO_3$ single crystal is shown in Fig. 7. The insulator metal



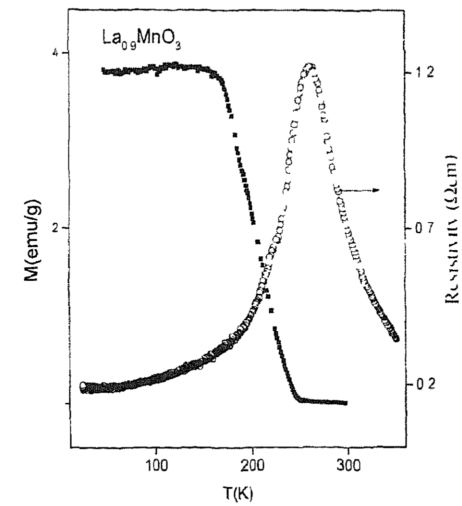


FIG. 7. Temperature variation of resistance and magnetization for $La_{0.9}MnO_3$ single crystal grown by floating zone melting technique.

FIG. 6. Floating zone melting crystal growth process (a) initial stage just before melting (b) melting stage and (c) crystal growing stage.

transition is very sharp as revealed by the magnetization curve and clearly demonstrates that the crystal grown is of very good quality Further work in this direction to grow lanthanide manganite single crystals is in progress

4 Conclusions

A cryocooled superconducting magnet with close-cycle helium refrigerator and an optical window for irradiation studies was designed and fabricated The performance of the magnet is excellent The infrared imaging furnace installed is working well Single crystals of $La_{0.9}MnO_3$, $Nd_{0.5}Ca_{0.5}MnO_3$, $Nd_{0.45}Sr_{0.55}MnO_3$ were grown successfully by employing floating zone melting crystal growth technique

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References

1 Rao, C N R and Raveau B (eds) *Colossal magnetoresistance, charge ordering and related properties of manganese oxides* World Scientific, 1998

2 Rao C N R, Cheetham A K and Mahesh, R *Chem Mater*, 1996, **8**, 2421–2432

3 Urushibara A *et al* *Phys Rev B*, 1995, **51**, 14103–14109

4 Suenaga M *Superconductor material science* (S Foner and B B Schwartz eds), Plenum Press 1981 p 201

5 Jikihara, K *et al* *IEEE Trans Appl Supercon*, 1997 **7**, 423–426

6 Watanabe K *et al* *Jap J Appl Phys*, 1993 **32**, L488–L490

7 Watanabe K, Awaji S and Kimura, K *Jap J Appl Phys*, 1997 **36**, L673–L675

8 Hasebe K *et al* *IEEE Trans Appl Supercon* 1997, **7**, 707–710

9 Dew-Hughes, D *Phil Mag*, 1974, **30**, 293–298

10 Turowski, P and Schneider, Th *Physica B*, 1989, **155**, 87–91

11 Pfann, W G *Solid state physics 4*, Academic Press, 1957, p 423

12 Moritomo Y Asamitsu, A Kuwahara, H and Tokura Y *Nature (Lond)*, 1996, **380**, 141–144

13 Kloc Ch Cheong, S W and Matl, P *J Crystal Growth*, 1998, **191**, 294–297

14 George, S Leong, Y Raju A R and Rao, C N R *Curr Sci*, 1999, **77**, 574–576

15 Watanabe K *et al* *Jap J Appl Phys*, 1998 **37**, L1148–L1150