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ESRF

# Experiment Report Form

**The double page inside this form is to be filled in by all users or groups of users who have had access to beam time for measurements at the ESRF**.

Once completed, the report should be submitted electronically to the User Office using the **Electronic Report Submission Application:**

*http://193.49.43.2:8080/smis/servlet/UserUtils?start*

***Reports supporting requests for additional beam time***

Reports can now be submitted independently of new proposals – it is necessary simply to indicate the number of the report(s) supporting a new proposal on the proposal form.

The Review Committees reserve the right to reject new proposals from groups who have not reported on the use of beam time allocated previously.

***Reports on experiments relating to long term projects***

Proposers awarded beam time for a long term project are required to submit an interim report at the end of each year, irrespective of the number of shifts of beam time they have used.

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All users must give proper credit to ESRF staff members and proper mention to ESRF facilities which were essential for the results described in any ensuing publication. Further, they are obliged to send to the Joint ESRF/ ILL library the complete reference and the abstract of all papers appearing in print, and resulting from the use of the ESRF.

Should you wish to make more general comments on the experiment, please note them on the User Evaluation Form, and send both the Report and the Evaluation Form to the User Office.

## Deadlines for submission of Experimental Reports

- 1st March for experiments carried out up until June of the previous year;
- 1st September for experiments carried out up until January of the same year.

## Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report, in English.
- include the reference number of the proposal to which the report refers.
- make sure that the text, tables and figures fit into the space available.
- if your work is published or is in press, you may prefer to paste in the abstract, and add full reference details. If the abstract is in a language other than English, please include an English translation.

| ESRF | **Experiment title:** In-situ High Resolution Diffraction and Topography of the Early Stages of Relaxation in III-V Semiconductors | **Experiment number**: HS571 |
|---|---|---|
| **Beamline**: | **Date of experiments**: from: March 1999 to: June 2001 | **Date of report**: 24.8.01 |
| **Shifts:** 4 runs x 30 | **Local contact(s)**: Dr A Freund, E Ziegler | *Received at ESRF:* |

**Names and affiliations of applicants** (* indicates experimentalists):

**Prof B K Tanner, University of Durham, U.K.**

**Dr P J Parbrook, Prof C Whitehouse, University of Sheffield, U.K.**

**Mr B Lunn, Dr J H C Hogg, University of Hull, U.K.**

**Dr A D Johnson, Mr A M Keir, Mr J Jones, Mr D Wallis, Mr L M Smith, DERA, Great Malvern, U.K.**

## Report:

Strained layers of compound semiconductors such as $In_xGa_{1-x}As$ grown epitaxially on the (001) surface of GaAs form the basis of optoelectronics devices on which the much of the modern communications industry is based. For such devices to operate reliably over extended periods, the strained layer must not relax by creation of misfit dislocations at the interface as these act as local heating and deformation sites. Over the past two years, we have undertaken at the ESRF, some of the most demanding high resolution x-ray diffraction topography (imaging) experiments ever attempted. Using a purpose-built molecular beam epitaxy reactor (Figure 1), that was transported from DERA Malvern and installed on beamline BM5 of the ESRF, high-resolution x-ray diffraction topography (imaging) has been used to observe *in-situ*, the nucleation and propagation of the very first misfit dislocations in fully-strained epilayers of $In_{0.04}Ga_{0.96}As$ on (001) GaAs.

Use of a (second) channel-cut 004 Si beam conditioner after the primary water-cooled 111 channel-cut Si monochromator/beam conditioner has enabled us to record topographs on nuclear emulsion plates with exposure times of the order of seconds while retaining high spatial resolution and contrast[1]. An x-ray sensitive TV imager was used for setting up the experiments, but the individual narrow misfit dislocation images were only visible on the nuclear emulsion plates. By using pre-loaded cassettes (Figure 2) we were able to record sufficient data for each growth run, the run time being determined by the growth processes and procedures, rather than the x-ray exposure times. The highly coherent beam does produce strong phase contrast, particularly from the Be windows of the MBE chamber and this problem was only ever partially overcome. The area imaged in each topograph was typically 3 cm by 3 cm, limited by the x-ray optics associated with the secondary monochromator bandpass.[2]

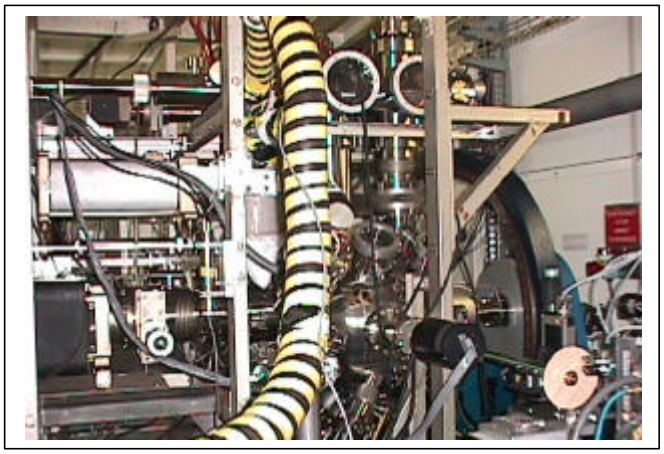


Figure 1
The MBE reactor looking down the beamline

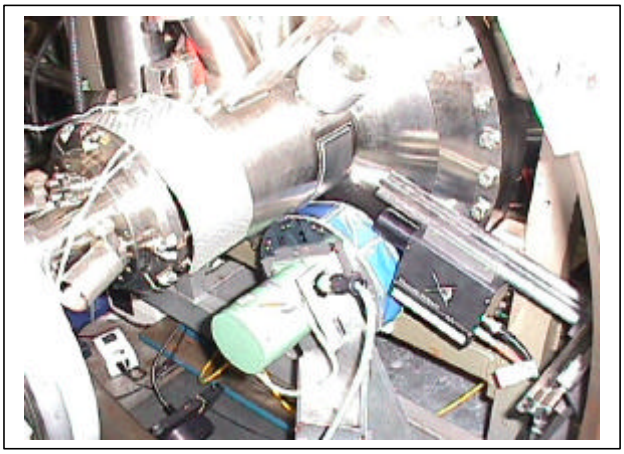


Figure 2
The beryllium window, detector circle and detectors

The reduction in exposure time in comparison with the Daresbury SRS synchrotron radiation source permitted us to grow layers at significantly higher temperatures than at Daresbury. (The new growth temperatures were more in line with growth temperatures used in industry for growth of layers for device fabrication.) Experiments on $In_{0.04}Ga_{0.96}As$ epilayers, grown 60°C below the transition temperature (Tt = 632°C) for the change in reconstruction from c(4x4) to (2x4), confirmed the anisotropy of the initial relaxation seen in the Daresbury experiments. This anisotropy is associated with the two types of dislocation found in the zincblende structure. The fast B(g) dislocations (Fig 3), running in the [110] direction, are nucleated before the slow A(g) set, running in the orthogonal $[1\bar{1}0]$ direction. The ESRF measurements gave a thickness for nucleation of the fast B(g) set of 90 ± 5nm, somewhat higher (Fig 4) than the prediction of the Matthews-Blakeslee model (70 nm using anisotropic elasticity theory and a dislocation core energy factor $\alpha = 2.7$). The thickness for nucleation of the slow $[1\bar{1}0]$ A(g) set was 165 ± 5nm and this again coincided with the thickness at which multiplication of the fast [110] B(g) set was observed. The onset of multiplication of the fast misfit set at the thickness for nucleation of the slow set has proven to be a general result. The mobility of the misfit dislocations was significantly higher than those observed previously, a feature attributed to a lower level of residual gas in the chamber. As a result, at Tt - 60°C, almost no misfit dislocation lines were observed to terminate in the sample in any of the *in-situ* topographs at any stage in the growth process. After nucleation from threading dislocations, all ran fully across the 50 mm diameter wafer to its edge. Recent experiments has shown that nucleation occurs at regions of residual preparation damage at the wafer edge[3]. Only a few examples of cross-slip were observed in the whole field of view (< 1 $cm^{-2}$).

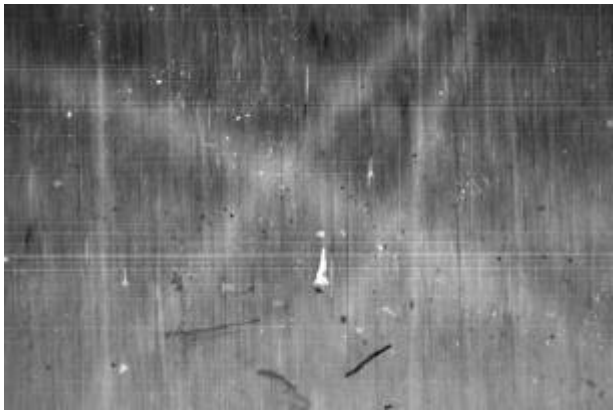


Figure 3 B(g) Misfit dislocations

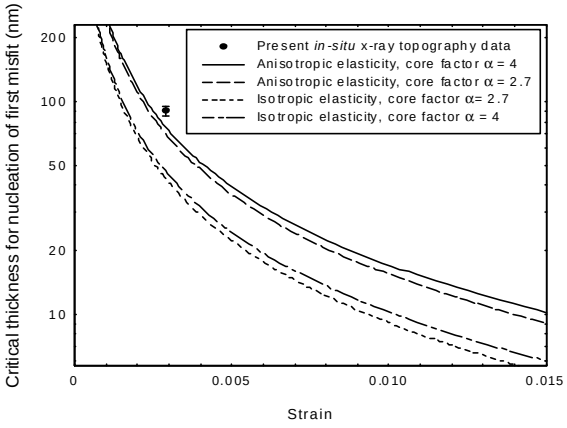
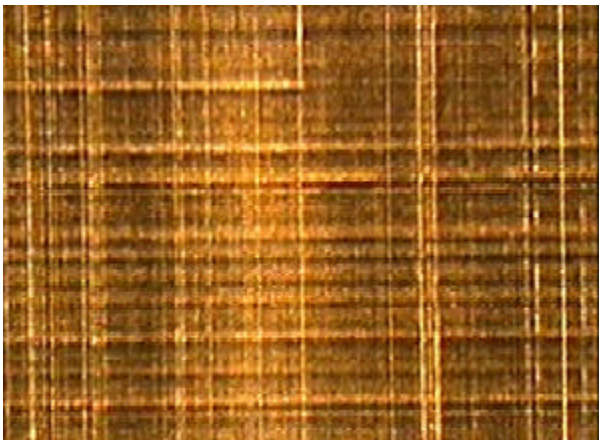



Figure 4 Comparison of undoped 4% In data with theory

As doping bulk crystals results in pinning of dislocations, we have to used much of the ESRF beamtime in studying whether we can use doping techniques to suppress the motion of misfit dislocations and hence raise the critical thickness at which strained epitaxial layers relax. On doping with Si, an increase in critical thickness was observed for both dislocation sets. An example of this is shown in Figure 5.

When a dislocation in the substrate crosses into the strained layer, there is a lateral force on the segment in the epilayer as a result of the biaxial strain present. As a consequence, the segment glides on an inclined slip plane, leaving behind the misfit dislocation segment at the interface. The line tension of the misfit segment results in a retarding force on the threading segment. In equilibrium, these are equal and this results in the classic Matthews-Blakeslee equation for the critical thickness at which misfit dislocations are formed. The Matthews-Blakeslee model has been extended to include an anisotropic Peierls lattice friction and a further frictional force varying linearly with the dopant concentration[4]. For the nucleation of the (initial) fast dislocation set, there is a very good agreement between the predictions of the extended model and experiment (Figure 6). Substantial lattice friction is present in the undoped samples for both dislocation sets and the increase in frictional force with dopant concentration is the same for both fast and slow dislocations.



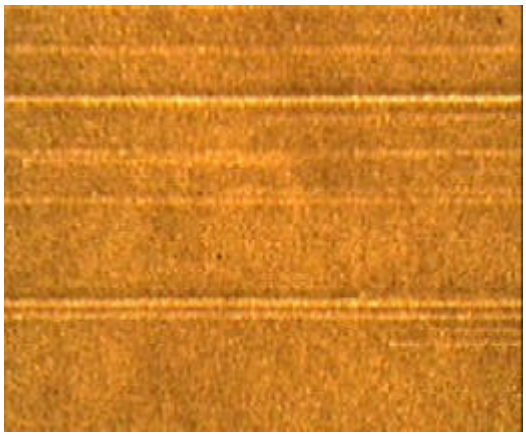
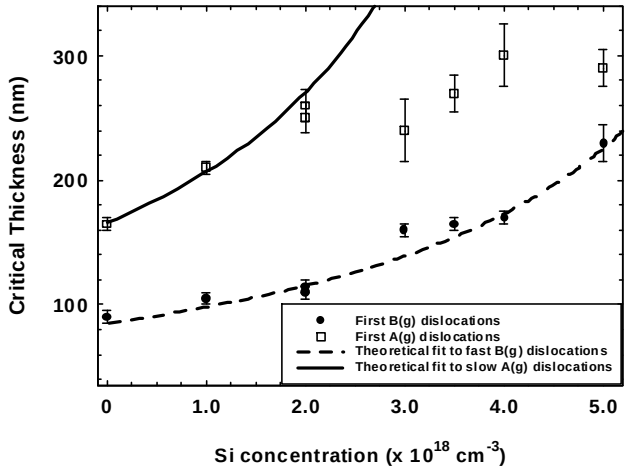


Figure 5 Undoped layer 260 nm thick $4x10^{18}$ $cm^{-3}$ Si doped layer 260 nm thick



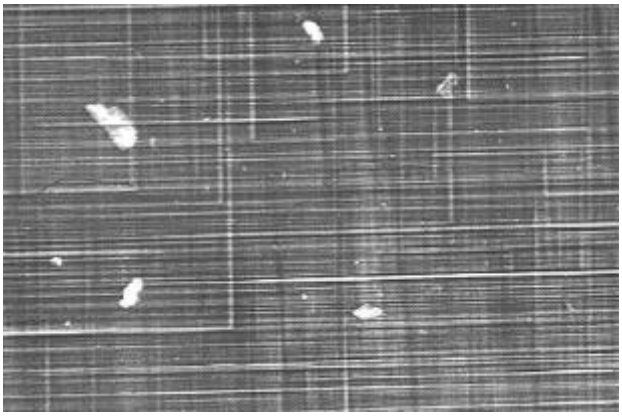



Figure 6 Comparison between measured critical thickness and the predictions of the modified Matthews-Blakeslee model

Figure 7 Topograph of $3.5x10^{18}$ $cm^{-3}$ doped InGaAs showing cross slip

For the slow A(g) dislocation set, the model breaks down for Si doping levels above about $2x10^{18}$ $cm^{-3}$. At higher Si concentration, the critical thickness becomes almost independent of concentration. The change in behaviour is seen from the topographs (Figure 7) to be associated with the initiation of cross slip.

In undoped samples, a small change was observed in the critical thickness as a function of growth temperature. Inclusion of a temperature dependent term in the Matthews-Blakeslee model enabled an activation energy 0.3 ± 0.2eV to be determined for the undoped layers[1]. This value, which is consistent with the observation of enhanced misfit dislocation mobility, is lower than measured previously and probably arises from the improved vacuum conditions and elimination of the long growth pauses necessary to expose the photographic plates at the less intense Daresbury SRS.

Unfortunately, due to difficulties associated with control of the In Knudsen cell, we were unable to obtain data over a very wide growth temperature range and we were not able to gather a full set for the Si doped samples. The data is good, but incomplete. With doping, the drop in critical thickness with growth temperature rise becomes more significant (figure 8). The effect is greatest for the slow A(g) dislocation set.

A general result to emerge from the Long Term Project is that, whether or not cross-slip is significant, the critical thickness for nucleation of the slow A(g) dislocations is that at which multiplication of the fast B(g) set occurs. Despite the evidence for nucleation at surface damage, the result implies that in all cases, the slow dislocation set is in some way associated with the rapid generation of a large number of B(g) dislocations.

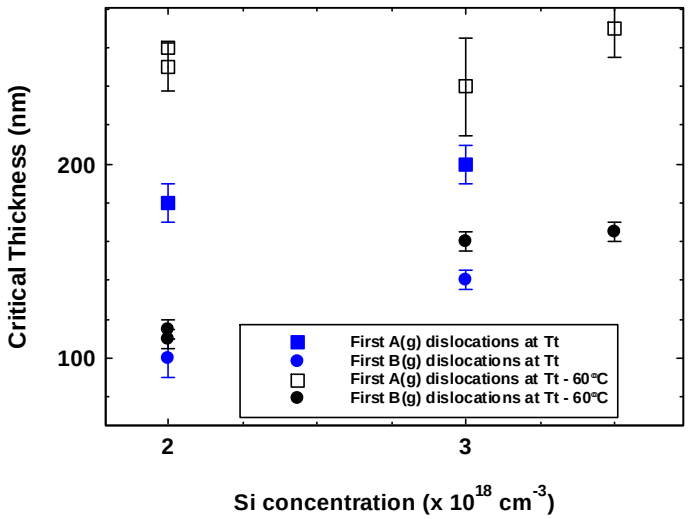



Figure 8 Drop in critical thickness with increased growth temperature for doped epilayers.

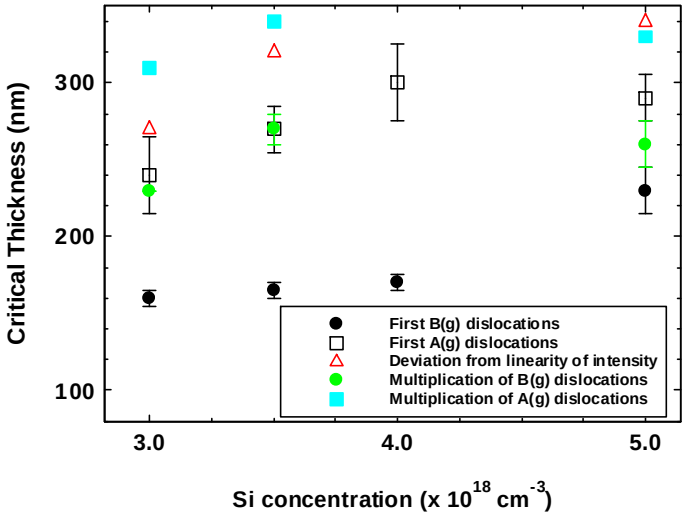



Figure 9 Correspondence of thickness for nucleation of A(g) & multiplication of B(g) misfits

Attempts at raising the critical thickness, and hence suppressing the onset of relaxation by growth of "virtual substrates" of Si doped material of lower strain were partially successful. Our strategy was, using *in-situ* high resolution x-ray diffractometry to monitor In composition, to grow a Si doped buffer layer of $In_{0.03}Ga_{0.97}As$ prior to growth of the $In_{0.04}Ga_{0.96}As$ undoped layer. As evidenced by the example results shown in the Table, we succeeded in raising the total thickness of InGaAs layer for both A(g) and B(g) nucleation. There was, however, a significant reduction in the thickness of the $In_{0.04}Ga_{0.96}As$ that could be growth prior to misfit nucleation. The changes appear independent of Si doping concentration in the virtual substrate. Close study of the images in the topographs indicates that there is significant relaxation occurring at the interface between the substrate and the virtual substrate.

| | **Total thickness for B(g) nucleation (nm)** | **Total thickness for A(g) nucleation (nm)** |
|---|---|---|
| ***Undoped InGaAs (4%)*** | 90 (± 10) | 165 |
| ***Undoped InGaAs (4%) on 80nm 3 x $10^{18}$/cc Si doped InGaAs(3%)*** | 150 (± 10) | 180 |
| ***Undoped InGaAs (4%) on 80nm 5 x $10^{18}$/cc Si doped InGaAs(3%)*** | 140 (± 10) | 180 |

Overall, the ESRF experiments not only show that the Matthews-Blakeslee model is capable of being extended successfully but also show that the critical thickness can be raised substantially by doping at levels that are consistent with device requirements.

During the *in-situ* monitoring of the x-ray optical configuration, we have developed a novel probe for measurement of relaxation in layers with high misfit dislocation density. The technique involves use of a large aperture detector covering the whole of the illuminated area of the sample. Curvature of the substrate, due to the strain in the epitaxial layer results in a reduction in the area of sample illuminated. It is straightforward to show that the change in the maximum intensity is proportional to the change in strain[5]. Figure 10 shows one example of the relaxation process as measured by this highly sensitive method. This is, we believe, the first time that strain hardening in epilayers has been measured quantitatively. We note from Figure 10 that there are three stages. The first, is elastic, or quasi elastic, where there is a linear fall in intensity, corresponding to a linear decrease in radius of curvature and linear increase in the elastic strain in the layer. At a thickness corresponding to the thickness for multiplication of the A(g) dislocation set (the result appears general) region occurs in which almost no further strain is accommodated. Complete relaxation of the additional strain appears to be taking place. Subsequently, hardening occurs and a new linear region is entered, with a different slope. This corresponds to partial relaxation. High resolution rocking curves taken at a thickness of 1μm show that the layer of Fig 10 is only 20% relaxed. We believe that the technique opens up the possibility of studying

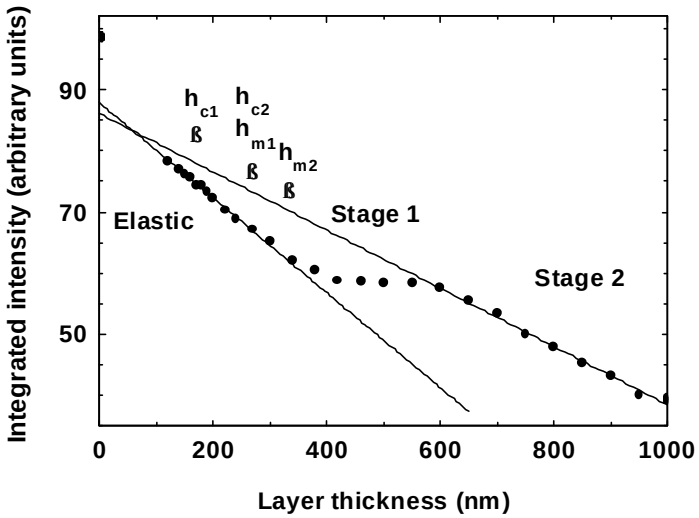



Figure 10 Plot of intensity versus epitaxial layer thickness for an $In_{0.04}Ga_{0.96}As$ layer doped with 3.5 x $10^{18}$ $cm^{-3}$ of Si. $h_{c1}$, $h_{c2}$, $h_{m1}$, $h_{m2}$ correspond to the critical thickness for nucleation and multiplication of B(g) and A(g) misfit dislocations determined from the x-ray topographs.

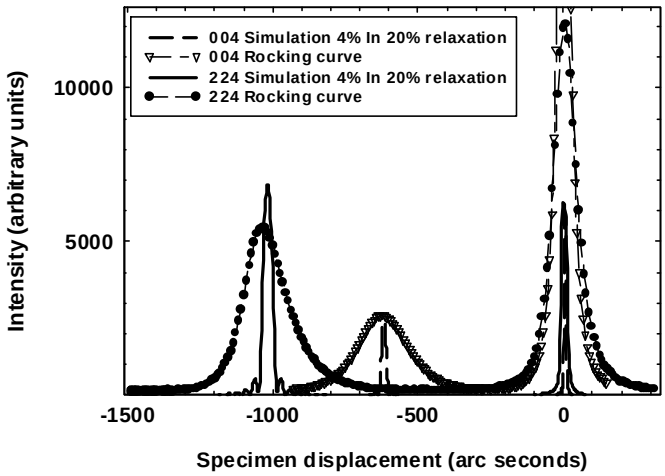



Figure 11 Rocking curves showing that the layer of Fig 10 only relaxed 20% at thickness of 1 μm.

device-relevant epilayers of high mismatch with a view to devising techniques for avoidance of this incomplete relaxation. Particularly in the growth of relaxed graded buffer layers for virtual substrates, this is of paramount importance.

In summary, the high resolution *in-situ* x-ray topography experiments have been difficult but successful. We have developed a good understanding of the effects of doping on the initial stages of relaxation in strained layer epitaxial structures. Our results have been extremely well received in the form of two key invited papers (XTOP2000 in Jasowiec[6] and the American Association for Crystal Growth Conference, Burlington VT, 2001).

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[1] B K Tanner, A. M. Keir, P. Möck, C. R.Whitehouse, G. Lacey, A. D. Johnson, G. W. Smith and G. F. Clark, J. Phys. D: Appl. Phys. 32 A119 (1999)

[2] B.K.Tanner, P.J.Parbrook, C.R.Whitehouse, A.M.Keir, A.D.Johnson, J.Jones, D.Wallis, L.M.Smith, B.Lunn and J.H.C.Hogg, J Phys. D: Appl Phys 34 (2001) A109-A113

[3] P.J.Parbrook, B.K.Tanner, B.Lunn, J.H.C.Hogg, A.M.Keir, and A.D.Johnson, submitted to Appl Phys Lett

[4] B.K.Tanner, P.J.Parbrook, C.R.Whitehouse, A.M.Keir, A.D.Johnson, J.Jones, D.Wallis, L.M.Smith, B.Lunn and J.H.C.Hogg, Appl. Phys. Letts. 77 (2000) 2156-8

[5] B.K.Tanner, P.J.Parbrook, B.Lunn, J.H.C.Hogg, A.M.Keir, and A.D.Johnson, submitted to J Appl Phys

[6] J Gronkowski, J Phys. D: Appl Phys 34 (2001), Guest Editor's introduction.