Acta metall. mater. Vol. 40, No. 6, pp. 1287–1292, 1992
Printed in Great Britain.
0956-7151/92 \$5.00 + 0.00

CRACKING OF BRITTLE FILMS ON ELASTIC SUBSTRATES

M. D. THOULESS, E. OLSSON† and A. GUPTA
IBM Research Division, T. J. Watson Research Center, Yorktown Heights, NY 10598, U.S.A.

(Received 25 September 1991; in revised form 6 November 1991)

Abstract—An analysis is presented that relates the crack spacing in a brittle film on an elastic substrate to the stress in the film, its thickness and fracture toughness. This analysis differs from an earlier one presented by one of the authors in that it considers the effect of a sequential, rather than a concerted, propagation of cracks, and predicts a larger crack spacing. The validity of the present analysis was confirmed by experimental results from a model system consisting of epitaxial $PrBa_2Cu_3O_{7-x}$ films on $SrTiO_3$ substrates. The experimentally observed relationship between the crack spacing and the film thickness was in excellent agreement with the theory.

Résumé—On présente une analyse qui lie l'espacement des fissures dans un film fragile sur une substrat élastique à la contrainte dans le film, à son épaisseur et à sa ténacité à la rupture. Cette analyse diffère d'une précédente présentée par l'un des auteurs en ce qu'elle considère l'effet d'une propagation séquentielle, plutôt que concertée, des fissures et prévoit un plus grand espacement des fissures. La validité de la présente analyse est confirmée par les résultats expérimentaux pour un sytème modèle formé de films de $PrBa_2Cu_3O_{7-x}$ sur des substrats de $SrTiO_4$. Les relations observées expérimentalement entre l'espacement des fissures et l'épaisseur du film sont en excellent accord avec la théorie.

Zusammenfassung—Es wird eine Analyse vorgelegt, die den Rißabstand in einem spröden Film auf einem elastischen Substrat mit der Spannung im Film, der Filmdicke und der Bruchzähigkeit verbindet. Diese Analyse unterscheidet sich von einer früheren Analyse eines dieser Autoren darin, daß sie den Einfluß eines sequentiellen statt eines konzertierten Fortschreitens von Rissen betrachtet; sie sagt größere Rißabstände voraus. Die Gültigkeit der jetzigen Analyse wird von Experimenten an einem Modellsystem bestätigt, welches aus epitaktischen $PrBa_2Cu_3O_{7-x}$-Filmen auf $SrTiO_3$-Substraten besteht. Der experimentell beobachtete Zusammenhang zwischen Rißabstand und Filmdicke stimmt ausgezeichnet mit der Theorie überein.

1. INTRODUCTION

A brittle film attached to a substrate and subjected to a tensile stress may fail by the propagation of a series of cracks along the film (Fig. 1). When the stress state within the film is one of biaxial tension, and there is no preferred cleavage direction, the cracks will divide the film into a series of islands in a pattern often seen in drying mud. In this paper, attention is focused on situations in which there is a directionality associated with the crack pattern, either because there is some preferred cleavage direction, or because there is some degree of uniaxiality in the stress. Under such conditions the cracks form in a series of parallel lines. The spacing between the cracks is not uniform, but there is a mean value that is dependent on the stress in the film [1].

When the substrate is ductile, the crack spacing is dictated by the yield stress of the substrate [2, 3]. A shear-lag analysis predicts that the cracks can be no closer than

$$\lambda^* = 2\sigma_c h/\tau \tag{1}$$

where σ_c is the minimum stress in the film required to propagate a crack, h is the film thickness and τ is the yield stress [2]. If the substrate is elastic, and no delamination occurs at the interface, an analysis by one of the present authors [4] has shown that the crack spacing is dictated by, h, the film stress and the fracture resistance of the film. A thermodynamic minimum spacing was obtained by equating the energy changes between a cracked and uncracked film to the energy associated with an array of cracks. It was emphasised that this minimum spacing was a lower bound that could be obtained only in the presence of a sufficient density of initial flaws. Statistical effects associated with the initial flaw population have been studied recently in a series of experiments by Delanney and Warren [5]. However, for reasons that are discussed in the present paper, it is now recognised that the minimum spacing presented in Ref. [4] is probably unobtainable in practice, even in the presence of a suitable density of flaws. The analysis was limited to the concerted propagation of an array of cracks; it will be seen that a rather different result is obtained when the cracks are assumed to propagate sequentially, so that the array evolves over time. Two recent papers [6, 7] have

†Present address: Department of Physics, Chalmers University of Technology, 412 96 Göteborg, Sweden.

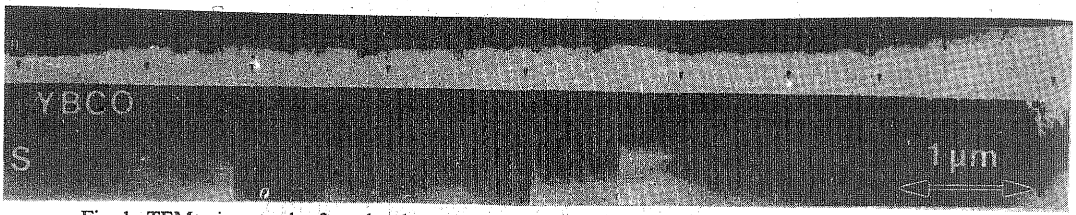

Fig. 1. TEM micrograph of cracks along the (001)-planes in a $YBa_2Cu_3O_{7-\delta}$ (YBCO) film deposited on a [110]-$SrTiO_3$ substrate (S) at 625 °C.

considered the mechanics of a cracked film incorporating the effects of an elastic mismatch between the film and substrate. However, results for the crack density were not explored in detail. In this paper, a simple result is obtained for the crack spacing in a homogeneous system, and is verified by observations on a ceramic film deposited on a ceramic substrate.

2. THEORY

Figure 2 shows a schematic illustration of the geometry considered in this paper. A semi-infinite plane of modulus E and Poisson's ratio ν is covered by a brittle film of thickness h with identical elastic properties. The film contains a series of semi-infinite and parallel cracks equally spaced at a distance l apart. These cracks are assumed to extend through the entire thickness of the film but are arrested, with no slip, at the interface. In addition, the film is assumed to be subject to a strain with a component orthogonal to the array of cracks that is uniform, tensile and of magnitude ϵ_0. This can be considered either to be a residual strain, or to result from the application of an external load. A previous calculation [4] has shown that the difference in strain energy (per unit area of interface between the film and substrate) between a cracked and uncracked portion of the film is given by

$$\Delta U = 1.98(h/l)\hat{E}\epsilon_0^2 h, \qquad l/h \gtrsim 8$$
$$= [0.5 - 0.0316(l/h)]\hat{E}\epsilon_0^2 h, \quad l/h \lesssim 8 \qquad (2)$$

where $\hat{E}$ is Young's modulus in plane stress and $E/(1-\nu^2)$ in plane strain. Since the strain energy in the uncracked portion of the film is

$$U_0 = 0.5\hat{E}\epsilon_0^2 h, \qquad (3)$$

it can be shown that the strain energy in the cracked portion of the film is

$$U_l = [0.5 - 1.98(h/l)]\hat{E}\epsilon_0^2 h, \quad l/h \gtrsim 8$$
$$= 0.0316(l/h)\hat{E}\epsilon_0^2 h, \qquad l/h \lesssim 8. \qquad (4)$$

†It is recognised, of course, that this can be considered only as an approximation for analytical purposes since cracks, unlike dislocations, cannot adjust their spacing to maintain uniform arrays.

A final expression required for the calculations of this section relates the energy associated with the array of cracks to the mode I fracture resistance of the film, Γ_f

$$U_c = \Gamma_f h/l. \qquad (5)$$

These four equations allow various bounds to be placed on the crack spacing that can be expected for given values of h, ϵ_0 and Γ_f. For example, if $\Delta U < U_c$, it is thermodynamically impossible for an array to advance across the film. Use of the first expression in equation (2) sets the bound on the parameters required for a single crack to propagate across the film [4, 8]

$$\hat{E}\epsilon_0^2 h/\Gamma_f \gtrsim 0.5. \qquad (6)$$

The second expression in equation (2) sets the thermodynamic limit on the minimum crack spacing, $l = \lambda_{min}$, that can be obtained when the cracks are close enough to interact [4]

$$(\lambda_{min}/h) = 8[1 - \sqrt{1 - 0.5\Gamma_f/(\hat{E}\epsilon_0^2 h)}]. \qquad (7)$$

An array of cracks more closely spaced than this limiting value will not propagate. To achieve this minimum spacing, not only must there be a sufficient density of initial flaws, but the cracks must propagate in a concerted fashion across the film [4]. The following analysis will show that a larger spacing is predicted when the cracks develop in a sequential manner.

As an approximation for investigating the sequential development of a crack array, the energy associated with uniform arrays of increasing density is considered.† It is assumed that the maximum crack density is obtained when the change in elastic energy between a film with a crack spacing l and one with a

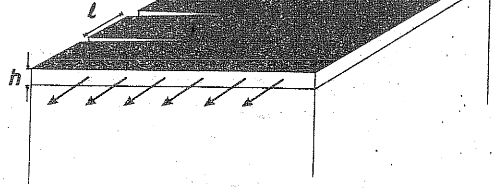

Fig. 2. Schematic illustration of a thin film of thickness h, with a series of semi-infinite and parallel cracks spaced at a distance l apart.

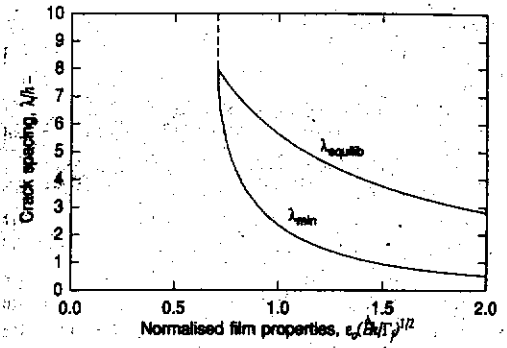

Fig. 3. Crack spacing plotted as a function of the normalised properties of the film. The curve for the thermodynamic minimum [equation (7)] is compared to the equilibrium results of equation (9).

crack spacing $l - \delta l$ is insufficient to provide the energy required for the additional cracks. This condition can be expressed as

$$\frac{\partial U_l}{\partial l} + \frac{\partial U_c}{\partial l} \geqslant 0. \qquad (8)$$

Differentiation of equations (4) and (5) then yields the following expression for the crack spacing

$$\lambda_{\text{equilib}}/h \simeq 5.6\sqrt{\Gamma_f/(\hat{E}\epsilon_0^2 h)}, \qquad (9)$$

provided that $\hat{E}\epsilon_0^2 h/\Gamma_f \gtrsim 0.5$. This expression is plotted and compared with equation (7) in Fig. 3. It is perhaps of some interest to note that equations (8) and (9) can also be obtained by a slightly different argument since, as illustrated in Fig. 4, λ_{equilib} corresponds to the spacing that results in minimising the total energy, $U_{\text{total}} = U_l + U_c$, of the cracked film. The analysis of Hutchinson and Suo [6] gives a spacing that is intermediate to the two limits given here.

Beyond the absolute magnitude of the two expressions, an important difference between equations (7) and (9) is the predicted relationship between the thickness and the spacing. Use of equation (7) suggests that the crack spacing should decrease as the film thickness increases. Conversely, equation (9) predicts that the crack spacing increases as $h^{1/2}$†. It will be seen that this latter relationship is confirmed by the experimental results of the following section. Furthermore, the two expressions indicate an interesting feature about the stability of an array of cracks propagating with a density greater than that given by equation (9). The condition for fracture can be satisfied for all the cracks in such an array if their tips remain co-linear. However, the results suggest that if a perturbation occurs whereby some cracks advance ahead of the array, the lagging cracks may be shielded to such an extent that the energy-release rate associated with them drops below the value required for fracture. A less dense array would then be created.

Finally, it is possible to use the equations of this section to compute an effective modulus for the cracked film. This can be used to calculate, for example, the curvature of a film-substrate system containing an array of cracks [7]. The effective modulus, $\hat{E}_{\text{eff}}$, is related to U_l by

$$U_l = 0.5\hat{E}_{\text{eff}}\epsilon_0^2 h, \qquad (10)$$

so that, by comparison with equation (4) it can be seen that

$$\hat{E}_{\text{eff}}/\hat{E} = 1 - 3.96(h/l), \quad l/h \gtrsim 8$$
$$= 0.0632(l/h), \quad l/h \lesssim 8. \qquad (11)$$

Simple beam theory can then be used to determine the curvature of a film on a substrate. If it is assumed that the film is very thin compared to the substrate, the curvature of the cracked system is

$$\kappa = (6\epsilon_0 h/H^2)(\hat{E}_{\text{eff}}/\hat{E}) \qquad (12)$$

where H is the thickness of the substrate.

Beuth [7] used an elegant Reciprocal-Theorem analysis to calculate the change in curvature between a cracked and uncracked system for the general case when the film and substrate have different elastic constants. However, the solutions presented in Ref. [7] are limited to geometries for which the cracks are sufficiently far apart that they do not interact. The first part of equation (11) is completely consistent with the solution in Ref. [7] for the special case of a homogeneous system. The second part of equation (11) allows the change in curvature to be calculated when the cracks do interact.‡

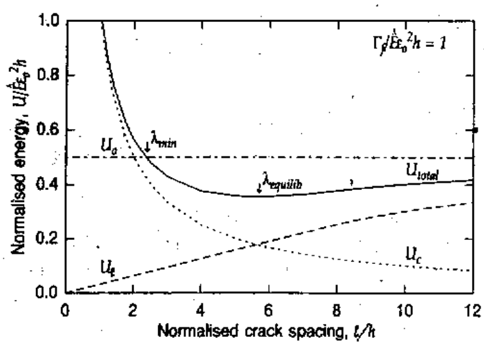

Fig. 4. A plot showing how the various terms for the energy of a cracked film depends on the crack spacing; U_0 is the energy (per unit area of interface) of the uncracked film, U_l is the energy density of the cracked film, U_c is the energy density associated with the cracks, and $U_{\text{total}} = U_l + U_c$.

†Close to the conditions defined by equation (6), the crack spacing increases to infinity as the thickness of the film is decreased to its critical value. The details of this region are lost in the approximate analytical expressions used in the analysis; however, the dotted line in Fig. 2 indicates the asymptotic behaviour.

‡This expression can also be obtained by using the second part of equation (2) in the Reciprocal-Theorem analysis of Ref. [7].

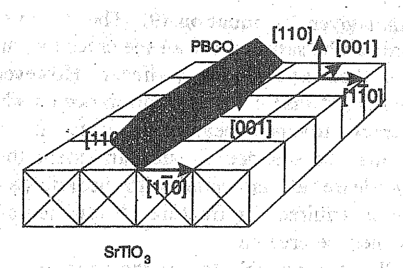

Fig. 5. A schematic illustration of the relationships between the crystallographic orientation of a [110]-$PrBa_2Cu_3O_{7-x}$ film and a [110]-$SrTiO_3$ substrate.

3. EXPERIMENTS

Epitaxial [110]-$PrBa_2Cu_3O_{7-x}$ (PBCO) films grown by a technique of laser ablation on [110]-$SrTiO_3$ substrates proved to be an ideal model system to study the cracking phenomenon analysed in the previous section. Although the films are not themselves superconducting, they are of interest because they can serve as templates for growing films of the high-T_c superconductor, $YBa_2Cu_3O_{7-\delta}$ [9–11]. However, owing to a large difference in the thermal expansions of the film and substrate along the [001] direction, the films have a tendency to crack along the (001) planes during cooling from the deposition temperature. It was this feature that made the films of interest for this particular study. The details of the film preparation are given elsewhere [11]. The important points to note are that the films were deposited at 625 °C on 1 mm thick [110]-oriented $SrTiO_3$ substrates, and that the only parameter varied between the films was the thickness which ranged from 0.08 to 0.50 μm.

Epitaxy in this system is obtained between the (003) planes of the $PrBa_2Cu_3O_{7-x}$ film and the (001) planes of the $SrTiO_3$ substrate, as shown in Fig. 5. However, at the deposition temperatures, the [003]- and [1$\bar{1}$0]-lattice parameters of the $PrBa_2Cu_3O_{7-x}$ are larger than the [001]- and [1$\bar{1}$0]-lattice parameters of the $SrTiO_3$ [11, 12]. The resultant compressive strain in the film is relaxed by the formation of ⟨103⟩-misfit dislocations at the interface between the film and substrate, and by stacking defects along the (001) planes of the film (Fig. 6). Transmission electron microscopy (TEM) showed that the dislocation density did not vary over the range of film thicknesses studied; this indicates that these thicknesses were far above the critical value required for the formation of a misfit dislocation, and implied that the dislocation densities were approaching the asymptotic limit. The major effect of the misfit dislocations is to accommodate the strain in the [1$\bar{1}$0] direction. They do, however, relieve some of the strain in the [001] direction; measurements made by TEM showed that the strain relaxed by the dislocations in this direction was 0.3% for all the samples.

Stacking defects also contribute to strain relaxation in the [001] direction. Again, the density was independent of thickness. This implies that the strain in the [001] direction, as well as in the [1$\bar{1}$0] direction, was almost totally relaxed at the deposition temperature. The strain accommodated in the [001] direction by stacking defects was determined to be 1.8%. The total strain relaxed in this direction by both the stacking defects and the misfit dislocations was therefore 2.1%. This value compares very favourably with the 2.2% predicted from the lattice parameters at 625°C [11].

Upon cooling, the film contracts more than the substrate in the [001] direction. At room-temperature there is only a 0.08% mismatch between the [003] spacing of the $PrBa_2Cu_3O_{7-x}$ films grown under the conditions described in Ref. [11] and the [001] spacing of the $SrTiO_3$ [12, 13]. Consequently, if the compressive, misfit strain between these lattices is completely relaxed at the deposition temperature, a tensile strain of (2.1 − 0.08%) will be generated in the films when they are cooled to room temperature. This tensile strain, which is equated to ϵ_0, is responsible for the development of the cracks.

After fabrication, the surfaces of the films were etched in a solution of 5% bromine diluted in ethanol. They were examined by scanning electron microscopy, of which an example is shown in Fig. 7. It was observed that the films contained two types of crack: a series of large cracks that had propagated over large distances, and a much finer distribution of small cracks. This observation suggests that the system satisfies the assumption of the theoretical treatment that the final crack spacing is not limited by the defect density. The average crack spacing for the large cracks was obtained from micrographs such as Fig. 7; at least 100 measurements were made on each film. The data is summarised in Table 1.

4. DISCUSSION

It can be seen from equation (9) that, to make a comparison between the predicted and observed crack spacings, five parameters must be known: the

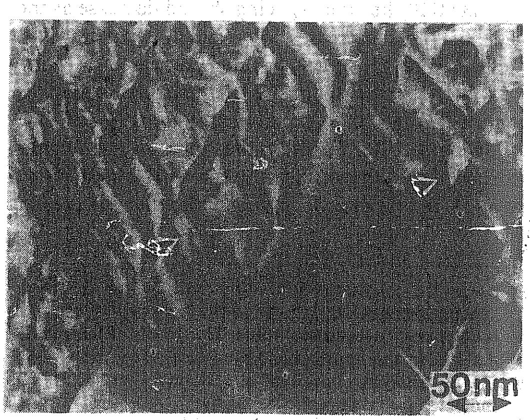

Fig. 6. ⟨103⟩-misfit dislocations and stacking defects (arrowed) at the interface of the $PrBa_2Cu_3O_{7-x}$ film and $SrTiO_3$ substrate.

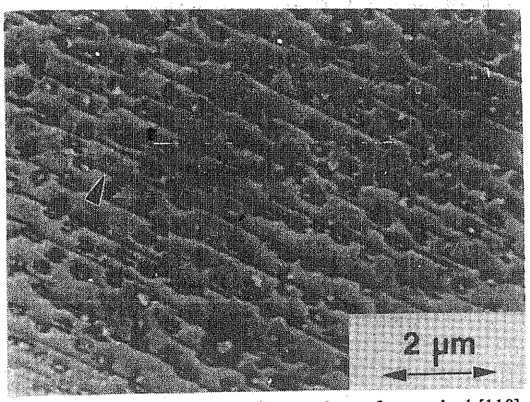

Fig. 7. SEM micrograph of the surface of a cracked [110]-film of $PrBa_2CuO_{7-x}$.

film thickness, h, the mean crack spacing, λ, the residual strain in the film, ϵ_0, the modulus of the film $\hat{E}$, and the appropriate fracture resistance Γ_f. The first two parameters, h and λ, were measured directly from the samples (Table 1). The residual strain in the film was estimated, as outlined in the previous section, to be $\epsilon_0 = 2.0\%$. The appropriate modulus of the film was estimated by using published data for the elastic stiffness constants, C_{ij}, of $YBa_2Cu_3O_{7-\delta}$ [14], and assuming that they are not significantly different for $PrBa_2CuO_{7-x}$. The modulus of interest is that of the [001] direction under conditions of zero strain in the [1$\bar{1}$0] direction and zero stress in the [110] direction. This modulus can be obtained by the methods described in Ref. [15], and, from the data of Ledbetter *et al.* [14], has a value of 102 GPa.

This leaves only the fracture resistance of the (001) planes as an unknown quantity. Unfortunately, there does not appear to be any reliable data for this parameter. Therefore, the expedient was resorted to of fitting the data to equation (9) and, hence, deducing its value. The resultant value of $\Gamma_f = 6.5\ \mathrm{Jm^{-2}}$ is very reasonable. For example, a fracture toughness of 0.8 $\mathrm{MPa\sqrt{m}}$ is obtained if the modulus is assumed to be 102 GPa. This is slightly lower than the 1.1 $\mathrm{MPa\sqrt{m}}$ estimated by Cook *et al.* [16] for the (100) and (010) planes of $YBa_2Cu_3O_{7-\delta}$, and is consistent with the notion that, because of their structure, the (001) planes should be cleavage planes. With a value assigned to all the parameters in equation (6), a prediction of 0.09 μm can be made for the critical film thickness below which no cracks will propagate. It is therefore satisfying that no cracks were observed in a film 0.08 μm thick (Table 1). Finally a graphical comparison of the theory and experimental data is shown in Fig. 8. It will be observed from this figure that the form of the relationship between the crack spacing and the film thickness is represented very well by the theory.

Table 1. Observed crack spacings along the [001] direction as a function of the film thickness for films deposited at 625 °C

Film thickness (μm)	Average crack spacing (μm)
0.08	No cracks
0.10	0.71
0.17	0.91
0.26	1.08
0.33	1.29
0.50	1.53

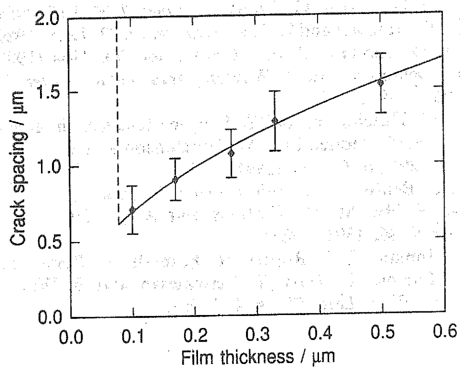

Fig. 8. A comparison of the experimental results for the crack spacing as a function of film thickness with the predictions of equation (9). The dashed portion of the theoretical curve indicates the minimum thickness below which a single crack cannot propagate across the film. The error bars on this figure indicate one standard deviation of the observed crack spacings about the mean values.

5. CONCLUSIONS

Some simple mechanics have been presented for the formation of an array of cracks within a film. It has been assumed for the purposes of the analysis that no delamination or slip occurs at the interface, and that there is a sufficient density of nuclei from which cracks can form, so that the spacing is not limited by the statistics of flaw distributions. The analysis predicts that, in an elastically homogeneous system, the crack spacing is proportional to the square root of the film thickness and is dependent on the tensile strain in the film, its fracture resistance and elastic constants. The assumptions of the model appear to be well-satisfied in a system consisting of $PrBa_2Cu_3O_{7-x}$ deposited by laser ablation on $SrTiO_3$ substrates, and oriented so that the cleavage planes lie perpendicular to the interface. Experimental observations on the system appear to follow very well the trends predicted by the analysis.

Acknowledgements—This work was supported in part by DARPA under contract No. N00014-89-C-0112. EO was also partially supported by the Swedish board for technical development. The authors would like to acknowledge the contributions of P. R. Duncombe, D. R. Clarke and T. M. Shaw.

REFERENCES

1. G. Gille, in *Current Topics in Materials Science* (edited by E. Kaldis), Vol. 12, pp. 421–472. North-Holland, Amsterdam (1985).

2. M. S. Hu and A. G. Evans, *Acta metall.* **37**, 917 (1989).
3. D. C. Agrawal and R. Raj, *Acta metall.* **37**, 1265 (1989).
4. M. D. Thouless, *J. Am. Ceram. Soc.* **73**, 2144 (1990).
5. F. Delannay and P. Warren, *Acta metall. mater.* **39**, 1061 (1991).
6. J. W. Hutchinson and Z. Suo, in *Advances in Applied Mechanics* (edited by J. W. Hutchinson and T. Y. Wu), Vol. 29, pp. 63–191 (1991).
7. J. L. Beuth, *Int. J. Solids Struct.* In press.
8. M. S. Hu, M. D. Thouless and A. G. Evans, *Acta metall.* **36**, 1301 (1988).
9. A. Inman, C. T. Rogers, R. Ramesh, K. Remschnig, L. Farrow, D. Hart, T. Venkatesan and B. Wilkins, *Appl. Phys. Lett.* **57**, 2484 (1990).
10. E. Olsson and A. Gupta, to be published.
11. E. Olsson, A. Gupta, M. D. Thouless, A. Segmüller and D. R. Clarke, *Appl. Phys. Lett.* **58**, 1682 (1991).
12. M. E. Lopez-Morales, D. Rios-Jara, J. Taguena and R. Escudero, *Phys. Rev. B* **41**, 6655 (1990).
13. *Thermophysical Properties of Matter*, Vol. 13. The TRCP Data Series, (edited by Y. S. Touloukian and C. Y. Ho). IFI/Plenum, New York (1975).
14. H. Ledbetter and M. Lei, *J. Mater. Res.* In press.
15. J. F. Nye, *Physical Properties of Crystals*. University Press, Oxford (1979).
16. R. F. Cook, T. R. Dinger and D. R. Clarke, *Appl. Phys. Lett.* **51**, 454 (1987).