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Residual stress gradients along ion implanted zones – cubic crystals

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Large internal strains and stresses can be produced by low temperature implantation over small distances from the free surface in a thick substrate. These are typically non-uniform and have large composition gradients. In dilute b c c solutions, containing unclustered interstitial implants, the residual macroscopic strains may be treated as isotropic. The calculation of residual strain (or stress) is based upon anisotropic elasticity theory and internal stress is given in terms of the dipole tensor for individual defects in single crystal films. In a completely elastic zone, forces act to maintain a rigid outside surface and cause the strain distribution to be zero along directions parallel to the free surface. This produces a strain magnification along the perpendicular direction from Poisson contractions. If the implanted zone is completely relaxed by plastic deformation, the strains are described by the free expansion strains due to both implants and lattice damage. There is no angular dependence of the free expansion strain in this extreme condition. One can determine whether a zone is completely elastic, completely relaxed by plastic deformation, or in some intermediate state from plots of strain against $\sin^2\chi$, where χ is the angle of tilt relative to the surface normal. These results may be obtained from X-ray Bragg intensity data by measuring shifts and line broadening from (hkl) planes at different tilt angles. Theoretical results are given for both single crystal and polycrystalline materials in terms of residual strain and stress.

1. Introduction

Low temperature ion implantation into a metal lattice can produce a disturbed region with a large strain gradient, near the free surface. A moderately thick underlying region, which goes undisturbed, constrains the implanted zone from expanding parallel to the surface of the zone. This results in the development of a biaxial residual strain gradient perpendicular to the surface. It is possible that such biaxial elastic strains may be partially or totally relieved by plastic deformation [1–3].

Residual stress calculations are presented for cubic single crystals with or without the presence of plastic deformation. Anisotropic elasticity theory is required for single crystal films and isotropic theory is used for polycrystalline materials.

2. Residual Stress Theory

The overall implanted zone is considered to respond elastically without the presence of relaxations associated with plastic deformation. The disturbed region is assumed to be contained within semi-rigid walls. Later, this constraint is removed thereby allowing for relaxations due to plastic deformation.

In treating the purely elastic problem, imagine that the implanted zone is dissected into elementary, uncoupled slabs of constant concentration C_i with infinitesimal thickness, dx_3 (Fig. 1a). Allow each slab to undergo a free expansion (or contraction) due to the pressure exerted by the implanted atoms. We know that a concentration of C_i of one kind of point defect and orientation produces a bulk expansion of the lattice given by [4]

$$\frac{C_i}{V}\frac{\partial V}{\partial C_i} = \sum_{i=1}^{3} \varepsilon_{ii} = \frac{C_i \sum_{i=1}^{3} P_{ii}}{(C_{11} + 2C_{12})V} \quad (1)$$

where P_{ii} and ε_{ii} are the diagonal components of the dipole stress and strain tensors, respectively, expressed in terms of the cubic coordinate system. C_{11}, and C_{12} are the elastic constants and V, the volume of a host lattice atom. If the defects occupy positions in the host lattice with less than cubic symmetry, then, in general, $\varepsilon_{11} \neq \varepsilon_{22} \neq \varepsilon_{33}$, and the shear components, ε_{12}, ε_{13} and ε_{23} need not be zero. Cubic symmetry is maintained by allowing each of the axes of the principal strain to be oriented along mutually perpendicular directions in equal numbers. Consequently, the average strains are

$$\langle\varepsilon_{11}\rangle = \langle\varepsilon_{22}\rangle = \langle\varepsilon_{33}\rangle = \frac{C}{3V}\frac{\partial V}{\partial C} \quad (2a)$$

$$\langle\varepsilon_{12}\rangle = \langle\varepsilon_{13}\rangle = \langle\varepsilon_{23}\rangle = 0 \quad (2b)$$

where $C = \Sigma_i C_i$ is the total concentration for all orientations of one kind of defect. It is more convenient to adopt sample axes and write the average free expansion strain of Equations 2a and b in terms of the notation

$$\varepsilon'_{f11} = \varepsilon'_{f22} = \varepsilon'_{f33} \quad (3)$$

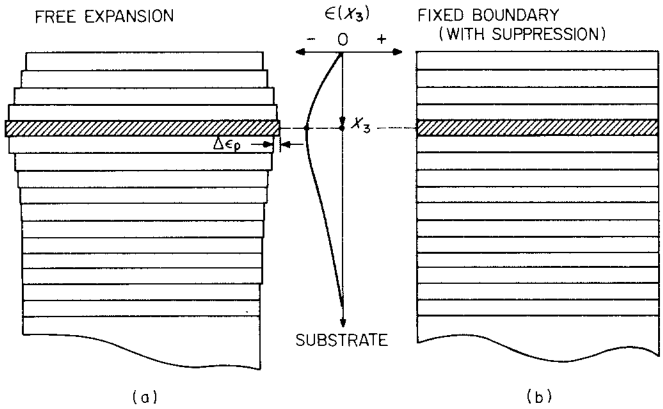

Figure 1 Schematic drawings of implanted zone (a) uncoupled or freely expanded (b) coupled by incremental strains $\Delta\varepsilon_p$ forming rigid surfaces.

and

$$\varepsilon'_{f12} = \varepsilon'_{f13} = \varepsilon'_{f23} = 0$$

In the freely expanded state, Equations 2 and 3 are equivalent due to statistical isotropy in a cubic system. Isotropy is normally lost in the next step when the slabs are joined continuously (Figs 1a and 1b) and the plane stress varies according to the elastic constants and crystal directions.

Forces exerted by the underlying material do not allow these slabs to expand freely. The actual residual stresses which develop along the implanted zone can be calculated using the method of strain suppression. In this method, the slabs are joined continuously by first applying stress σ'_{11}, σ'_{22} on the outer surfaces such that the net elastic strain resulting from the internal stresses exactly cancel the free expansion strain of Equation 3 in each slab along the 1′ and 2′ sample directions. It has been shown, using cubic anisotropic elasticity theory, that [6]

$$\sigma'_{ii} = f_i\varepsilon \quad i = 1 \text{ or } 2 \tag{4}$$

with

$$\sigma'_{33} = \sigma'_{12} = \sigma'_{13} = \sigma'_{23} = 0$$

where

$$f_i = [C_{11} + (A + 1)C_{12} + C_{an}(A - 1)\Omega_i]$$
$$i = 1 \text{ or } 2$$
$$\varepsilon = -\varepsilon'_{f11} = -\varepsilon'_{f22}$$
$$C_{an} = C_{44} - 1/2(C_{11} - C_{12})$$
$$A = \frac{-2C_{12} + C_{an}\Omega}{C_{11} + C_{an}\Omega}$$

with

$$\Omega = 4(\alpha_3^2\beta_3^2 + \alpha_3^2\gamma_3^2 + \beta_3^2\gamma_3^2)$$
$$\Omega_1 = 4(\alpha_1\beta_1\alpha_3\beta_3 + \alpha_1\gamma_1\alpha_3\gamma_3 + \beta_1\gamma_1\beta_3\gamma_3)$$
$$\Omega_2 = 4(\alpha_2\beta_2\alpha_3\beta_3 + \alpha_2\gamma_2\alpha_3\gamma_3 + \beta_2\gamma_2\beta_3\gamma_3)$$

and $(\alpha_1, \beta_1, \gamma_1)$, $(\alpha_2, \beta_2, \gamma_2)$, $(\alpha_3, \beta_3, \gamma_3)$ are the direction cosines of the sample coordinate system (1′, 2′, 3′), with respect to the cubic coordinate system. The application of normal stresses, σ'_{11}, σ'_{22} according to Equation 4 have generated additional elastic strains in the slab, i.e.

$$\varepsilon'_{e11} = \varepsilon'_{e22} = -\varepsilon'_{f11} = -\varepsilon'_{f22}$$

$$\varepsilon'_{e33} = A\varepsilon = \left(\frac{2C_{12} - C_{an}\Omega}{C_{11} + C_{an}\Omega}\right)\frac{C}{3V}\frac{\partial V}{\partial C} \tag{5}$$

The elastic strain profile with depth is directly proportional to the variation of C with depth. Slabs are joined together continuously making the total strain along the 1′ and 2′ directions essentially zero throughout the implanted zone.

The overall sample, including the substrate, must be free of any net external stresses. Therefore, additional reactionary stresses, $\sigma'_{11}(x_3)$, $\sigma'_{22}(x_3)$ must exist which make the net external stresses and moments identically zero. The strain distribution in the sample, due to these reactionary stresses is obtained by applying St Venant's principle. We replace these stresses by an equivalent stress pattern, which gives an identical result expressed as a planar stress distribution acting along the 1′–2′ directions,

$$\Delta\sigma'_{ii}(x_3) = f_i\frac{1}{3V}\frac{\partial V}{\partial C}\left(\frac{1}{t}\int_0^{t_0} C(x_3)\mathrm{d}x_3 + \frac{12}{t^3}(x_3 - t/2)\int_0^{t_0}(x_3 - t/2)\ C(x_3)\mathrm{d}x_3\right)$$
$$i = 1 \text{ or } 2 \tag{6}$$

where t is the thickness of the overall sample and t_0, the thickness of the implanted zone. The elastic strain resulting from the stress distribution of Equation 6 is given by [4]

$$\Delta\varepsilon'_{e11}(x_3) = \Delta\varepsilon'_{e22}(x_3) = \frac{1}{3V}\frac{\partial V}{\partial C}\left(\frac{1}{t}\int_0^{t_0} C(x_3)\mathrm{d}x_3 + \frac{12}{t^3}(x_3 - t/2)\int_0^{t_0}(x_3 - t/2)\ C(x_3)\mathrm{d}x_3\right) \tag{7a}$$

$$\Delta\varepsilon'_{e33}(x_3) = A\Delta\ \varepsilon'_{e11}(x_3) = A\Delta\ \varepsilon'_{e22}(x_3) \tag{7b}$$

An examination of the first and second terms of Equation 7a show that the first is constant while the second varies linearly with x_3. Normally, the thickness of the sample, t is very much larger than the thickness of the implanted zone t_0. Consequently, the additional stresses and strains included in Equations 6 and 7 can be neglected. This is true for samples examined by the authors but is clearly not true for thinned sections used for transmission electron microscopy. For thick samples, the total elastic stresses and strains, can be obtained from Equations 4 and 5. The normal elastic strain, at an angle χ to the x_3 direction, is obtained from [6]

$$\varepsilon'_{e\chi}(x_3) = (\varepsilon'_{e11}(x_3) - \varepsilon'_{e33}(x_3))\sin^2\chi + \varepsilon'_{e33}(x_3) \quad (8)$$

Substituting Equation 5 into Equation 8

$$\varepsilon'_{e\chi}(x_3) = \frac{1}{3V}\frac{\partial V}{\partial C}C(x_3)\,(-\beta^{el}\sin^2\chi - A) \quad (9)$$

where

$$\beta^{el} = \frac{C_{11} + 2C_{12}}{C_{11} + C_{an}\Omega} = 1 - A \quad (10)$$

The total strain gradient in the implanted zone, at tilt angle χ, includes the free expansion strain of Equation 2 and the elastic strain of Equation 9, i.e.

$$\varepsilon'_\chi(x_3) = \beta^{el}\frac{C(x_3)}{3V}\frac{\partial V}{\partial C}(1 - \sin^2\chi) \quad (11)$$

Clearly, the measured strain goes to zero smoothly with a $\sin^2\chi$ or $\cos^2\chi$ dependence, at $\chi = 90°$ (see Fig. 2). The ratio of the total strain at $\chi = 0°$, to the free expansion strain is β^{el} which is a magnification factor that can be as large as 2 to 3 [5]. Therefore, strain enhancement produced by a substrate which constrains the implanted zone can produce a large effect. At an angle $\chi = \chi_0$, the total strain is identically equal to the free expansion. This is obtained by equating Equation 9 to zero [6], giving

$$\cos^2\chi_0 = 1/\beta^{el} \quad \text{or} \quad \sin^2\chi_0 = -A/\beta^{el} \quad (12)$$

For angles $\chi < \chi_0$, the total strain is larger than the free expansion gradient whereas for $\chi > \chi_0$, the reverse is true. This behaviour is shown in Fig. 2 plotted conventionally in terms of $\sin^2\chi$.

For the isotropic case, we set $C_{an} = 0$; and use the relations

$$C_{11} = \frac{(1 - \nu)E}{(1 + \nu)(1 - 2\nu)}$$

$$C_{12} = \frac{\nu E}{(1 + \nu)(1 - 2\nu)} \quad (13)$$

where E is Young's modulus and ν Poisson's ratio of the host lattice. In this limit, the results for $\varepsilon'_\chi(x_3)$ are independent of the crystallographic orientation and are applicable to an untextured polycrystalline material. The strain distribution $\varepsilon'_\chi(x_3)$ and $\sin^2\chi_o$ become

$$\varepsilon'_\chi(x_3) = \frac{C(x_3)}{3V}\frac{\partial V}{\partial C}\left(\frac{1 + \nu}{1 - \nu}\right)(1 - \sin^2\chi) \quad (14a)$$

$$\cos^2\chi_0 = \frac{1 - \nu}{1 + \nu} \quad \text{or} \quad \sin^2\chi_0 = \frac{2\nu}{1 + \nu} \quad (14b)$$

A measurement of the strain at various tilt angles χ, allows one to separate out the free expansion from the elastic strain. X-ray intensity band analysis is used to determine the overall strain distribution and is discussed in later papers [6–9].

Residual stress can be determined from the slope of the individual strain curves shown in Fig. 2 using Equations 4 and 11

$$\frac{d\varepsilon'_\chi(x_3)}{d\sin^2\chi} = \frac{-\sigma'_{ii}(x_3)\beta^{el}}{f_i} \qquad i = 1 \text{ or } 2 \text{ (anisotropic)} \quad (15a)$$

$$= \frac{-\sigma'_{ii}(x_3)}{E/(1 + \nu)} \qquad i = 1 \text{ or } 2 \text{ (isotropic)} \quad (15b)$$

If the concentrations are sufficiently small, these results can be applied to more than one type of point defect by superimposing their fields. Summing over defect species, j, the free expansion becomes

$$\varepsilon'_{f11}(x_3) = \varepsilon'_{f22}(x_3) = \varepsilon'_{f33}(x_3) = \sum_j\left(\frac{C(x_3)}{3V}\frac{\partial V}{\partial C}\right)_j \quad (16)$$

The free expansion term in Equation 16, represents the strain that one would measure after complete relaxation due to plastic deformation. This is independent of χ as is illustrated by the horizontal line in Fig. 2. The next section discusses the intermediate case of partial relaxation of the elastic strain.

3. Residual stress relaxation due to plastic deformation

Residual elastic strains can be relaxed by plastic deformation along an implanted zone. Consider the zone to be divided into slabs parallel to the surface and

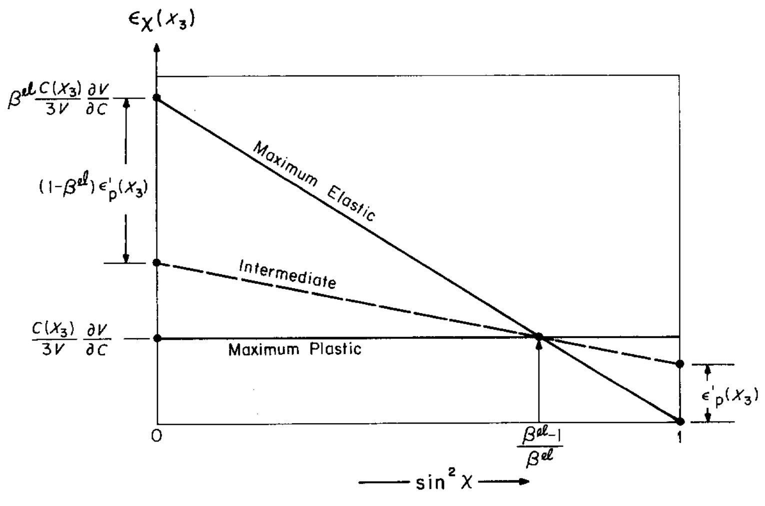

Figure 2 Three strain curves for an internal slab containing internal sources of strain. Note extremes: Completely rigid surface–maximum elastic strain, and freely expanded slab–maximum plastic deformation. Intermediate: Some plastic deformation – incomplete strain suppression. $\beta^{el} = (C_{11} + 2C_{12})/(C_{11} + C_{an}\Omega)$.

of infinitesimal thickness dx_3. If the plastic strain increment is $\Delta \varepsilon_p(x_3)$ at x_3 (Fig. 1) the total strain is the sum of all strain increments extending back to the undisturbed interface. This cummulative sum is designated by $\varepsilon'_p(x_3)$ in the sample coordinate system. Removing the elastic strain suppression stresses allows a slab to expand freely parallel to the surface. This expansion is reduced by the plastic strain, i.e.

$$\varepsilon'_{e11} = \varepsilon'_{e22} = \frac{C(x_3)}{3V}\frac{\partial V}{\partial C} - \varepsilon'_p(x_3) \quad (17)$$

In order to restore the residual elastic strain to the original value prior to the free expansion, biaxial surface stresses σ'_{11}, σ'_{22} are applied. This results in an elastic deformation that is opposite in sign but equal to the strain given by Equation 17. The elastic strain along $x_3(\chi = 0)$ includes the magnification factor, β^{el}, and this must be combined with the free expansion to obtain the observed effective strain at a depth, x_3. If the tilt angle, χ, is included,

$$\varepsilon'_\chi(x_3) = \varepsilon'_p(x_3) + \beta^{el}\left(\frac{C(x_3)}{3V}\frac{\partial V}{\partial C} - \varepsilon'_p(x_3)\right)(1 - \sin^2\chi) \quad (18)$$

Again, the strain varies linearly with $\sin^2\chi$ (or $\cos^2\chi$) as seen in Fig. 2. There are three points of special interest located at $\chi = 0$, χ_0 and 90° with the following strains

$$\varepsilon'_0(x_3) = \left(\beta^{el}\frac{C(x_3)}{3V}\frac{\partial V}{\partial C} + (1 - \beta^{el})\,\varepsilon'_p(x_3)\right) \quad (19a)$$

$$\varepsilon'_{\chi_0}(x_3) = \frac{C(x_3)}{3V}\frac{\partial V}{\partial C} \quad (19b)$$

$$\varepsilon'_{90}(x_3) = \varepsilon'_p(x_3) \quad (19c)$$

The invariant point Equation 19b at χ_0, obtained from Equations 12 or 14b, gives the free expansion strain while the extrapolation point at 90° gives the accumulation of plastic deformation strain between a strain free substrate and the point x_3.

4. Discussion

The previous developments have considered elastic and plastic deformations for only one out of many slabs. However, an ion implanted zone normally does not have a uniform distribution of implants. Therefore, the preceding developments must be extended to a distribution of slabs with different strains (Fig. 1). Before considering such an extension, our findings are summarized for one slab.

Consider a system of finite slabs with each in a state of biaxial uniform strain. The amount of strain suppression required for each was assumed to be given by a strain curve like the one in Fig. 2. The states of strain for one can be summarized as follows.

(I) Residual elastic strain without plastic deformation. A maximum absolute strain is found perpendicular to the free surface as given by $\beta^{el}\ (C(x_3)/3V)(\partial V/\partial C)$. With complete strain suppression, zero strain should be observed along all lines parallel to the free surface ($\chi = 90°$). For anisotropic substances, the plane stresses σ'_{11} and σ'_{22} are not equal.

(II) Residual elastic strain completely relaxed by plastic deformation. Neglecting any defect substructure, the macroscopic elastic condition is equivalent to an ideal free expansion without constraining forces. This results in a constant macroscopic strain for all directions within a slab.

(III) Residual strain is only partially relaxed. The strain is intermediate between extremes I and II. The parameters of interest may be obtained by extrapolating $\chi \to 90°$ to obtain the total accumulated plastic deformation and by interpolating to χ_0 to obtain the free expansion strain.

In each case, the point χ_0 is invariant and behaves as a pivot point for linear strain curves. For Case I, the value of ε'_0 and the slope are the same except for a change in sign. Also, the volume changes can be positive or negative depending upon the nature of the solid solution.

These results are simply extended to a system of thin slabs of constant thickness having average strains that correspond to points along the implant profile (Fig. 2). If each slab is taken to be of uniform composition, there will be one line per slab having a linear angular variation in strain. Typically, the distribution of implants is Guassian-like causing the distribution of lines to cluster near the peak. The distributions for the extreme cases i.e. completely elastic (I) or completely plastic (II) strains are shown schematically in Fig. 3 as

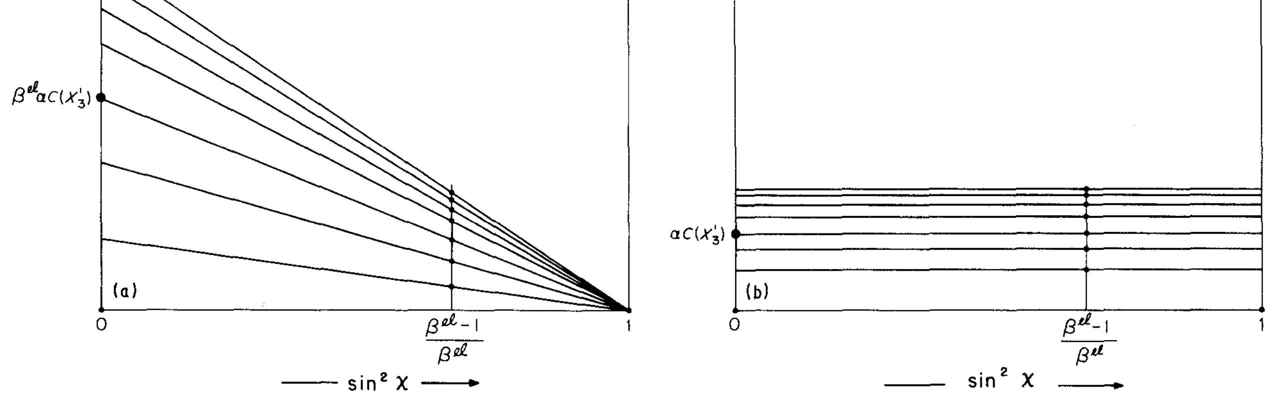

Figure 3 Schematic of (a) completely elastic and (b) freely expanded distribution of stepped strain in a non-uniform sample.

(a) and (b) with $\alpha = (1/3V)/(\partial V/\partial C)$ = constant. These are stepped distributions of strain that become increasingly magnified in the elastic case as $\chi \rightarrow 0°$ and shrink to a point as $\chi \rightarrow 90°$. When the deformation is completely plastic or the sample has freely expanded, the distribution of strains remains unchanged with χ.

At this point, one additional refinement should be considered. Figs 3a and b, at $\chi = 0°$, show results for stepped rather than a continuous change in concentration. This requires $C(x_3)$ to be stepped if both β^{el} and α are constants for a given sample. $C(x_3)$ can be made continuous with a linear variation in each slab. This additional complication would cause each line to expand into a fan-shaped element that converges to a point at $\chi = 90°$ and opens fully at $\chi = 0$. The range of free expansion strain is seen at χ_0. The introduction of fan-shaped elements into Fig. 3a, produces a continuous change in $C(x_3)$ and therefore $\varepsilon_\chi(x_3)$. At this point, Fig. 3a may be redefined as the distribution of mean strains in a system of slabs of constant thickness.

A consideration of the extreme of complete elastic relaxation by plastic deformation, with a continuous change in $C(x_3)$, becomes complicated by the details of the dislocation substructure. However, if one plots mean lines of strain through each subgrain, one would expect clustering as shown in Fig. 3b. The fine details of elastically interacting dislocation substructures [1–3] go beyond the intent of this paper which deals with continuum elasticity theory.

Preliminary results for 5% nitrogen implanted into niobium indicate that the implanted zone remains completely elastic [7]; however, this one case cannot rule out the possibility of having plastic deformation at higher fluences or for different systems.

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References

1. J. H. VAN DER MERWE, in "Treatise on Materials Science and Technology", edited by H. Herman, Vol 2, (Academic Press, New York, 1973) p. 1.
2. P. G. WINCHELL, J. BOAH and P. S. AYRES, *J. Appl. Phys.* **42** (1971) 2612.
3. J. S. VERMAK and J. H. VAN DER MERWE, *Phil. Mag.* **10** (1964) 785.
4. P. H. DEDERICHS, *J. Phys. F.* **3** (1973) 471.
5. S. P. TIMOSHENKO and J. N. GOODIER, "Theory of Elasticity", (McGraw-Hill, New York, 1970).
6. S. I. RAO and C. R. HOUSKA, *J. Appl. Phys.* **52** (1981) 6322.
7. *Idem*, submitted for publication.
8. S. L. RAO, BAO-PING HE and C. R. HOUSKA, submitted for publication.
9. S. I. RAO and C. R. HOUSKA, submitted for publication.

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