New Approach in Equilibrium Theory for Strained Layer Relaxation

A. Fischer, H. Kühne, and H. Richter
Institute of Semiconductor Physics, Walter-Korsing-Strasse 2, 15230 Frankfurt (Oder), Germany
(Received 20 June 1994)

We present a new approach in equilibrium theory for strain relaxation in metastable heteroepitaxial semiconductor structures, one which includes the elastic interaction between straight misfit dislocations. The free-surface boundary conditions are satisfied by placing an "image dislocation" outside the crystal in such a manner that its stress field cancels that of the real misfit dislocation at the surface. This image method provides an equilibrium theory which correctly predicts critical strained layer thicknesses and completely describes the strain relief via plastic flow in lattice mismatched epilayers.

PACS numbers: 62.20.Dc, 62.20.Fe, 68.55.Jk, 68.60.Bs

Since the introduction of pseudomorphic epitaxy, the mechanical behavior of thin layers and multilayered structures is of considerable interest. The growth of coherent thin layers on rigid crystalline substrates is possible when biaxial compressive or tensile strain in the layer accommodates the lattice mismatch between the film and substrate material. When the stored strain energy exceeds a certain threshold, the heterostructure becomes metastable and the film strain may give way to misfit dislocations. The basic energetic and kinetic parameters describing mismatch accommodation by in-plane strain and misfit dislocation in metastable heterostructures appear to be well described by the framework of Matthews and Blakeslee [1] and Dodson and Tsao [2]. However, it is evident that they cannot adequately explain the point of strain relief onset via plastic flow and the work hardening behavior of strained layers at the end of the thermal relaxation process since they ignore the phenomenon of elastic interaction between straight misfit dislocations. The first includes the problem of developing a relationship between the equilibrium critical thickness at which dislocations form and the bulk lattice mismatch. The latter involves balancing the force required to move misfit dislocations against the elastic stress field due to dislocation interaction [3].

In this Letter, we present a new approach in equilibrium theory for strain relaxation in metastable heteroepitaxial semiconductor structures which includes the elastic interaction between straight misfit dislocations. The free-surface boundary conditions are satisfied by placing an "image dislocation" outside the crystal in such a manner that its stress field cancels that of the real misfit dislocation at the surface. This image method provides an equilibrium theory which correctly predicts critical strained layer thicknesses. Furthermore, by considering the exact solution for the elastic interaction of real and image dislocations, our equilibrium model is suitable for completely describing the strain relief via plastic flow and the phenomenon of work hardening in lattice mismatched epilayers.

It is necessary to introduce notation. At thermodynamic equilibrium, misfit dislocations appear at the interface of strained layer heterostructure when the strained layer is thick enough that it is energetically favorable for the mismatch to be accommodated by a combination of elastic strain and interfacial misfit dislocations, rather than by elastic strain alone [4]. This equilibrium critical thickness h_{crit} has been calculated and discussed by many authors, e.g., [1,2,5–7], in the continuum picture as well as through phenomenological description for dislocation dynamics. However, there have been many reports, e.g., [7,8], of experimental determinations of h_{crit} indicating that coherence apparently persists to thicknesses much greater than that predicted by classical theories. The semiempirical kinetic model of Dodson and Tsao [2] is more appropriate to describe the latter stages of relaxation, where the effective stress is decreasing due to a reduction in misfit strain produced by the high dislocation density. For the case of sufficiently low dislocation content in the strained layer near the point of strain relief onset, this model reduces to the equilibrium form of Matthews and Blakeslee [1]. Thus the challenge remains to develop a predictive model appropriate for strained layer relaxation.

To introduce an appropriate continuum model in response we begin by analyzing the conditions under which the strained layer relaxation should occur in metastable heterostructures, and we then modify the governing models to account for the discrepancies intimated previously. In linear elasticity the superposition principle holds true. The displacements, strains, and stresses caused by a set of forces acting on a body are the sum of those caused by the individual forces. In a finite body, boundary conditions at the surface must be satisfied. For example, no forces can act on a free surface. The image-force method provides a powerful tool to solve such problems in the continuum theory of elasticity [9]. Let us now consider the schematic illustration in Fig. 1. For strained layer case, the free-surface boundary conditions are satisfied by placing an image dislocation outside the crystal in such a manner that its stress field cancels that of the real interfacial misfit dislocation at the surface. Thus the strains and stresses in a finite heterostructure subjected to point and lattice

 0031-9007/94/73(20)/2712(4)$06.00

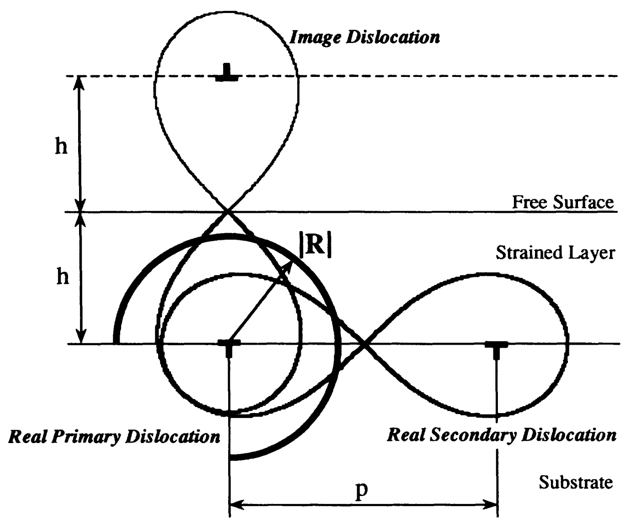

FIG. 1. Schematic illustration of the configuration of real and image misfit dislocations in a strained heteroepitaxial structure. The modulus of complex dislocation semispacing $|\mathbf{R}|$ is indicated.

mismatch forces can be described as a superposition of strains and stresses caused by real internal sources and image sources applied on the external surface of the body. The boundary condition in this case is satisfied if the self-stress of an imaginary misfit dislocation of equal strength and opposite sign at a position $2h$ along the strained interface normal is superposed on the self-stress of the real primary dislocation.

In the continuum picture the presence of dislocations causes strains around the line and, as a response to these, stresses as known from conventional elasticity theory. These stresses are defined by the contact forces transmitted through internal area elements. We speak of self-stresses to distinguish them from the applied misfit stresses. In linear approximation, the self-stress σ_S of a straight dislocation in a region bounded by a coaxial cylinder of radius R is [9]

$$\sigma_S = \{Gb(1 - \nu \cos^2 \theta)/[4\pi R(1 - \nu)]\}[\ln(\alpha R/b)], \quad (1)$$

where G is the shear modulus of the epilayer material, ν its Poisson ratio, b is the magnitude of the Burgers vector, θ is the angle between the dislocation Burgers vector and its line direction, and α is a factor which accounts for the energy in the dislocation core where linear elasticity does not apply. α is generally taken to be in the range from 1 to 4 for covalently bonded semiconductor materials [9]. Because of the logarithmic dependence, the elastic self-stress is insensitive to the precision of choice of a value of α. We set $\alpha = 1$. So, referred to the slip plane surface, the shear component of the self-stress of a straight dislocation τ_S is given by $\tau_S = \cos\phi\, \alpha_S$, where ϕ is the angle between the slip plane and the strained interface normal. In the present case, then, an interfacial 60°-type misfit dislocation on the slip plane causes resolved shear stress, to wit:

$$\tau_S = \{\cos\phi\, Gb[1 - (\nu/4)]/[4\pi R(1 - \nu)]\}[\ln(R/b)]. \quad (2)$$

Notice that only shear stresses in the slip system produce glide forces on a dislocation.

So far we have been considering the image-force and self-stress problem for a misfit dislocation in a metastable heterostructure. As shown in Fig. 1, imagine now a real secondary dislocation lying parallel to the real primary dislocation at a distance of p and moving continuously towards the primary one. For further deformation under the driving force produced by the internal misfit stress, it is necessary that the moving misfit dislocation have to overcome the resistance caused by superposed self-stress field of imaginary dislocation and the real primary dislocation bounded by a virtual cylinder of radius $R = h$. Note that at larger distances from the real primary dislocation the image stresses largely cancel the dislocation stresses. Again, for the elastic stress field extension of the real interfacial misfit dislocations lying parallel to another, a reasonable approximation would be to take roughly one-half the distance p between dislocations for R [3,9]. According to the principle of superposition, we will now combine the imaginary and the real free-surface term of the self-stress of a misfit dislocation, i.e., h and $p/2$, respectively, and get a complex dislocation semispacing $\mathbf{R}$. Its modulus $|\mathbf{R}| = R_{h,p}$ is given by

$$R_{h,p} = (1/h^2 + 4/p^2)^{-1/2}, \quad (3)$$

where h is the thickness of the epilayer, and the subscripts h and p stand for the imaginary and real component, respectively. We can say that the modulus of complex dislocation semispacing $R_{h,p}$ is a complex solution for the stress-free boundary associated with the presence of two free surfaces. The relationship represented by Eq. (3) is plotted in Fig. 2 for different layer thicknesses h. The pursual of Fig. 2 shows some interesting relationships. For dislocation spacings p greater than $5h$, the modulus of complex dislocation semispacing as a measure of the extension of the elastically strained continuum about a misfit dislocation is dominated by this imaginary term, i.e., by the layer thickness h. In this case, Eq. (3) reduces to $R_{h,p} \sim h$. If p diminishes continuously, then the effect of the real term on $R_{h,p}$ is increasing slowly. When the dislocation spacing reaches the value $p \sim 2h$, the imaginary and the real free-surface terms make the same contribution to the modulus of complex dislocation semispacing. Below $p \sim h/5$, as the real misfit dislocations approach one another, the effect of the imaginary component vanishes, and $R_{h,p}$ does not depend on h. Replace now in Eq. (2) R by $R_{h,p}$, the shear component of the total self-stress created by present dislocation content in a finite body becomes

$$\tau_S = \{\cos\phi\, Gb[1 - (\nu/4)]/[4\pi R_{h,p}(1 - \nu)]\}[\ln(R_{h,p}/b)]. \quad (4)$$

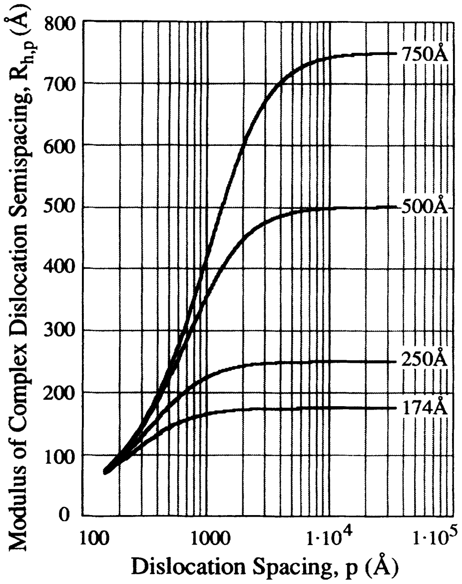

FIG. 2. A plot of the calculated modulus of complex dislocation semispacing $|\mathbf{R}| = R_{h,p}$ as a function of misfit dislocation spacing p for different strained layer thicknesses.

For complete describing of the stain relief via plastic flow in lattice mismatched epilayers, we should now consider the resolved shear stress τ that acts on the slip system as a consequence of the internally applied misfit stress. In an initially misfit dislocation free substrate-epilayer system, the in-plane stain ε is given by $(a_l - a_S)a_S$, where a denotes in-plane lattice parameter, and the subscripts s and l refer to the substrate and the layer, respectively. When the elastic strain is partially relieved by a single array of misfit dislocations created at the interface, the residual in-plane strain becomes $\varepsilon = [(a_l - a_s)/a_s] - (b\cos\lambda/p)$, where λ is the angle between the Burgers vector and the direction in the interface, normal to the dislocation line, and the term $b\cos\lambda/p$ represents the strain relief via plastic flow [10]. Here p is the average distance between the dislocations. Hence it follows that, at equilibrium, the lattice mismatch accommodation would occur without elastic strain, i.e., without tetragonal distortion of the cubic lattice cells. Physically, that cannot be. To overcome this difficulty we will now substitute the modulus of complex dislocation semispacing $R_{h,p}$ for $p/2$ in the residual in-plane stain expression above. So the true residual in-plane strain becomes $\varepsilon = [(a_l - a_s)/a_s] - [b\cos\lambda/2R_{h,p})]$, where the h term accounts for elastic strain and the p term for plastic flow. In this case, the resolved shear stress τ acting on the slip system on a misfit dislocation is defined as

$$\tau = \cos\lambda\cos\phi[2G(1+\nu)/(1-\nu)] \times \{[(a_l - a_s)/a_s] - [b\cos\lambda/(2R_{h,p})]\}. \quad (5)$$

The additional h term reflects the fact that, at equilibrium, the lattice mismatch of a strained layer will not be totally accommodated by misfit dislocations, but that a thickness-dependent elastic strain is retained by the epilayer.

As we have previously argued, the stains and stresses in a finite heterostructure subjected to point and lattice mismatch forces can be described as a superposition of strains and stresses caused by real internal sources and image sources. We should now consider the resolved shear stress τ that acts on the slip system as a consequence of the internally applied misfit stress and the elastic stress field due to dislocation interaction. As shown above, during misfit strain relief via plastic flow, an intrinsic stress field is built up. For further deformation, it is necessary that the moving misfit dislocations have to overcome the resistance caused by the stress field. Consequently, the dislocation self-stress field is in the opposite direction to the applied misfit stress. The excess resolved shear stress required to produce plastic flow will then by given by the difference between the two stress components. Equations (4) and (5) yield $\tau_{\rm exc} = \tau - \tau_S$, where the second term, too, accounts for work hardening of the material. Combining the two terms, we get the expression for the excess resolved shear stress

$$\tau_{\rm exc} = \cos\lambda\cos\phi[2G(1+\nu)/(1-\nu)]\{[(a_l - a_s)/a_s] - [b\cos\lambda/(2R_{h,p})](1+\beta)\}, \quad (6)$$

with

$$\beta = \{[1-(\nu/4)]/[4\pi\cos^2\lambda(1+\nu)]\}\ln(R_{h,p}/b).$$

Here the quantity $[b\cos\lambda/(2R_{h,p})]\beta$ corresponds to the decrease in active shear stress through elastic interaction between dislocations depending on the current equilibrium misfit dislocation density of the crystal.

To demonstrate the physical significance of the new approach in equilibrium theory for the strained layer relaxation proposed here, we have calculated the equilibrium critical thickness of $Si_{1-x}Ge_x/Si$ strained layer structures as a function of the fractional atomic Ge content x. Furthermore, we have compared our results to those predicted by using relaxation models based on elastically noninteracting dislocations. Taking the in-plane misfit strain $\varepsilon = 0.0418x$, Eqs. (3) and (6) and the equilibrium conditions for the point of strain relief onset via plastic flow, i.e., $\tau_{\rm exc} = 0$ and $p \to \infty$, lead to the following expression for the critical strained layer thickness $h_{\rm crit}$:

$$x = [b\cos\lambda/(0.0836h_{\rm crit})] \times (1 + \{[1-(\nu/4)]/[4\pi\cos^2\lambda(1+\nu)]\}\ln(h_{\rm crit}/b)). \quad (7)$$

For elastically noninteracting misfit dislocations, the equilibrium critical thickness of a single strained epilayer upon a substrate of different lattice parameter according to Matthews-Blakeslee [1] is given by

$$x = [b/(0.0836 h_{\rm crit})]\{[1 - (\nu/4)]/[2\pi(1 + \nu)]\} \times \ln(h_{\rm crit}/b). \quad (8)$$

Inserting appropriate material parameters in Eqs. (7) and (8), $\cos\lambda = 0.5$, $\nu = 0.28$, and $b = 3.84$ Å for growth on (001) surface, the equilibrium critical thickness calculated for the two different models are plotted in Fig. 3. It is seen, at first glance, that our values of $h_{\rm crit}$ represented by Eq. (7) are much larger than the values calculated using Eq. (8). Moreover, for a fractional atomic Ge content x greater than 0.5, the Matthews-Blakeslee formulation does not provide any value for $h_{\rm crit}$. Physically, that cannot be. Additionally, we have compared our theoretical results with the published experimental data [7,8] of $h_{\rm crit}$ obtained from $Si_{1-x}Ge_x/Si$ structures, grown by molecular-beam epitaxy at a growth temperature of 750 °C For each composition, there is good agreement between our theoretical results and the experimental data reported in Refs. [7,8]. For example, for $x = 0.2$, $h_{\rm crit}$ was found to be approximately 250 Å. As seen in Fig. 3, for this case, our analysis yields the value of 220 Å. The Matthews-Blakeslee model predicts only an equilibrium critical thickness value of 80 Å.

So far we have been considering the point of strain relief onset via plastic flow. The ramifications of our model for plastic flow and work hardening in $Si_{1-x}Ge_x/Si$ strained layer structures will now be discussed briefly. For further discussion of this point, see Ref. [3]. According to the previous Eq. (5), the in-plane epitaxial film stress σ becomes

$$\sigma = [2G(1 + \nu)/(1 - \nu)]\{0.0418x - [b \cos\lambda/(2R_{h,p})]\}. \quad (9)$$

Putting the values of the material parameters into Eqs. (6) and (9), $\cos\lambda = 0.5$, $\cos\phi = 0.816$, $G = 64$ GPa, $\nu = 0.28$, and $b = 3.84$ Å, assuming a metastable epilayer with $x = 0.25$ and $h = 500$ Å, and recalling Eq. (3), we can evaluate the variation in $\tau_{\rm exc}$ and σ for the complete thermal relaxation process. At the beginning of deformation, where $p \to \infty$, $\tau_{\rm exc} = 0.6$ GPa, and $\sigma = 1.9$ GPa. During plastic flow via misfit dislocation generation and propagation, $\tau_{\rm exc}$ and σ diminishes continuously and then remains unchanged at zero and 1.1 GPa, respectively. At this equilibrium deformation stage, the resolved shear stress that acts on the slip system as a consequence of the internally applied misfit stress and the shear component of elastic stress field due to dislocation interaction compensate one another and the strain relief via plastic flow comes to rest. Thus, as a result of work hardening, the lattice mismatched epilayer will remain in a certain state of strain at the end of the thermal relaxation process. It should be noted that such behavior of incomplete strain relief was found experimentally [3], too.

In summary, suffice it to say that our more refined model not only yields a better agreement between computed and measured values, it also provides a new approach in understanding complex mechanism for strain relaxation and defect propagation in a strained layer on a lattice mismatched substrate.

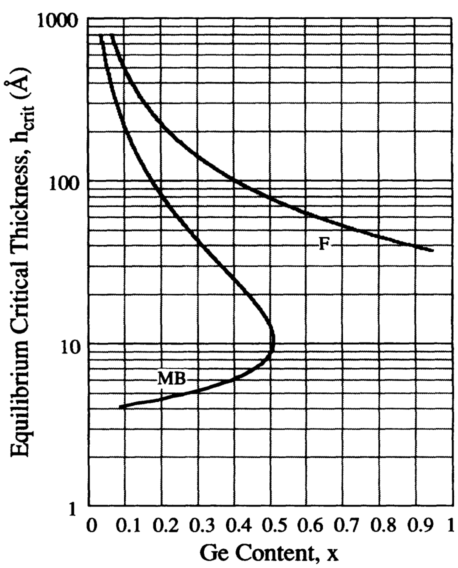

FIG. 3. Comparison of predicted equilibrium critical thickness $h_{\rm crit}$ for relaxation models based on elastically noninteracting (MB: Matthews-Blakeslee) and interacting (F: this study) misfit dislocations as a function of Ge content x.

[1] J. W. Matthews and A. E. Blakeslee, J. Cryst. Growth **27**, 118; **32**, 265 (1974).
[2] B. W. Dodson and J. Y. Tsao, Appl. Phys. Lett. **51**, 1325 (1987); **52**, 852 (1988).
[3] A. Fischer and H. Richter, Appl. Phys. Lett. **64**, 1218 (1994).
[4] G. C. Osbourn, IEEE J. Quantum Electron. **QE-22**, 1677 (1986).
[5] D. C. Houghton, Appl. Phys. Lett. **57**, 1434 (1990); **57**, 2124 (1990).
[6] R. Hull, J. C. Bean, D. J. Werder, and R. E. Leibenguth, Phys. Rev. B **40**, 1681 (1989).
[7] E. Kasper, in *Physics and Applications of Quantum Wells and Superlattices*, edited by E. E. Mendez and K. von Klitzing (Plenum Press, New York and London, 1987), p. 101.
[8] R. People and J. C. Bean, Appl. Phys. Lett. **47**, 322 (1985).
[9] J. P. Hirth and J. Lothe, *Theory of Dislocations* (McGraw-Hill, New York, 1985).
[10] J. R. Willis, S. C. Jain, and R. Bullough, Appl. Phys. Lett. **59**, 920 (1991).