A work-hardening based model of the strain relief in multilayer graded-buffer structures

D. González, D. Araújo, G. Aragón, and R. García
Departamento de Ciencia de los Materiales e IM y QI, Universidad de Cádiz, Apartado 40, 11510 Puerto Real, Spain

(Received 2 July 1997; accepted for publication 25 September 1997)

This paper describes the modeling of the dislocation distribution and of the strain relief in linearly and step-graded structures, based on work-hardening considerations. The model considers the energy variation in an InGaAs/GaAs system upon introducing a new dislocation into the interfacial fixed array of misfit dislocations. An analytical expression for the strain relaxation in graded-buffer structures is proposed. Transmission electron microscopy observation confirms the model predictions and reveals that the saturation state of relaxation is reached in the buried layers. © *1997 American Institute of Physics.* [S0003-6951(97)02247-X]

In the last decades bandgap engineering of II–VI and III–V semiconductor materials has permitted the fabrication of semiconductor devices such as LEDs lasers, and transistors. Nevertheless, the materials used for the device fabrication generally have physical properties (such as thermal expansion and lattice parameter) which differ from those of commercial substrates. Consequently, a plastic relaxation can occur during growth and the dislocation generated can drive into the active layer causing a worsening of the device performance. Therefore, the possibility for mismatched layers to grow without the generation of defects passing through the heteroepitaxial structure is of great interest. However, as no general theory of the strain relief in heteroepitaxial multilayers is available, it is difficult to design a buffer to obtain such a result. Only for some special cases such as single layers[1,2] or linearly graded layers,[3,4] can acceptable predictions of the strain relief be obtained. The necessity of a general theory for the design of relaxing buffers applied to II–VI, III–V, and III–N materials motivate the present study. To help address this need, we present here a model of strain relaxation in heteroepitaxial InGaAs/GaAs (001) multilayers using work-hardening based calculations.[5]

Relaxation in a single layer has been widely studied in the literature. Three stages can be distinguished.[6] In the first one, the relaxation is slow since only the bending of existing dislocations coming from the substrate occur as described by Matthews and Blackeslee[7] (region A in Fig. 1). In the second one, when the layer is thick enough for the multiplication of dislocations[8] (region B), a fast relaxation occurs.[9] In the third stage, an inhibition of the relaxation occurs for much thicker layers (region C), due to a work-hardening process in the material.[10] We successfully explained this third relaxation regime in single layer structures by dislocation interactions in a recently published model.[5,11] We extend here the model to the more complex compositionally graded structures including step- and linearly graded layers usually used as buffer layers to change the lattice parameter.[12]

In the design of buffer layers, used to adapt lattice parameters of substrates to that of grown-in devices two key requirements must be considered: (i) to grow a composition profile that induces a relaxation to just run into the work-hardening regime, i.e., that introduces the maximum number of misfit dislocations (MDs) per unit of thickness without the formation of threading dislocations (TDs), (ii) to predict the lattice parameter reached at the end of the buffer and the alloy composition that matches perfectly with that of the grown-in device.

The aim of this work is to answer both points. With this purpose in mind, we shall extend our recently published work-hardening based model[5] of relaxation for single layers to graded multilayered buffers. The model is first compared to experimental strain relaxation studies on four different graded InGaAs/GaAs structures with In composition steps arising between $x=0$ and $x=0.3$. The studied samples consist in one linearly graded structure of 1 μm thickness and three step-graded structures with 5%, 7.5%, and 10% In-steps, each one with a thickness of 120 nm. The samples are labelled A, B, C, and D, respectively.

During the growth of the graded structures, the density of misfit dislocations in the buried layers increases until the saturation state is reached.[13] The transmission electron microscopy (TEM) observations show constant interfacial MD density for all the samples. This regular distribution (see Fig.

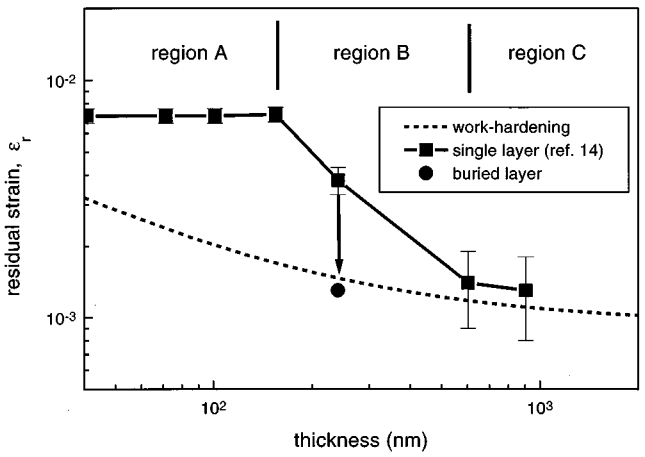

FIG. 1. Residual strain vs layer thickness for single $In_{0.1}Ga_{0.9}As/GaAs$ layers. The residual strain of buried layers of step-graded structures (solid circle), estimated by TEM, has reached the relaxation state of a thick single layer, i.e., when the work-hardening process controls the strain relief (region C). The residual strain calculated by the work-hardening model (dashed line) is in agreement with the experimental data for buried layers of the step-graded structures (solid point).

Downloaded 20 Oct 2006 to 150.214.231.66. Redistribution subject to AIP license or copyright, see http://apl.aip.org/apl/copyright.jsp

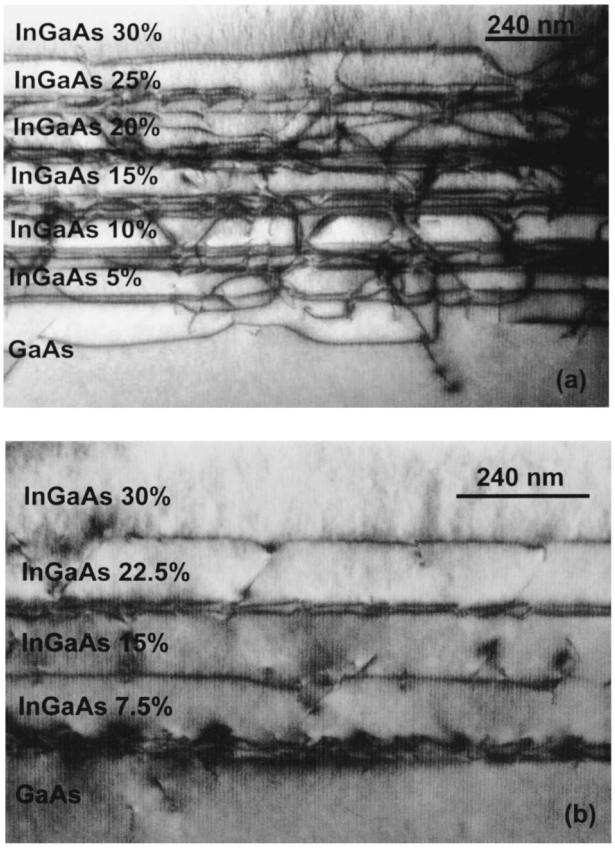

FIG. 2. (a) Bright field g004 XTEM micrographs of sample B; (b) Bright field g004 XTEM micrographs of sample C. Nearly constant MD densities around 1×10^5 cm^{-1} and 1.6×10^5 cm^{-1} are measured at buried interfaces for samples B and C, respectively. The sample structures are described at the left side of the micrographs. The layer thickness, as for all the samples studied, is 120 nm. Note that no dislocation threads across the structure.

2) of MD throughout the structure suggests that a work-hardening process must be dominant at buried interfaces. To verify this, we applied the work-hardening model previously published[5] for single layers. Briefly, the model considers the energetic state of the system during the introduction of a new dislocation into the preexisting fixed array of MDs, without changing the internal spacing. While in simple layers the lack of strain energy means that the nucleation sources are inactive for small thicknesses, in the case of step-graded multilayers, the sources at the buried interfaces remain active due to the tension accumulated by the successive grown layers with higher misfit. The saturation state is reached very fast, and the strain relaxation follows a work-hardening behavior. The work-hardening energy, $\Delta E_{W\text{-}H}$, is defined as the difference in the total energy of the system before E_1 and after E_2 the introduction of new dislocation:

$$\Delta E_{W\text{-}H}=E_2-E_1=E_s+E_{\text{int }m}+E_{\text{int }d}, \tag{1}$$

where E_s is the self energy of a new dislocation, $E_{\text{int }m}$ the interaction energy between the dislocation and the lattice misfit and $E_{\text{int }d}$ the interaction energy between the new dislocation with the fixed array of misfit dislocations at the interface. The detailed mathematical expression of each term has been described elsewhere.[5,11] In the first steps of the growth, i.e., when few MDs are present, $\Delta E_{W\text{-}H}$ is negative, meaning that the system tends to relax. We defined the saturation state of relaxation as the maximum MD density, $\rho_{W\text{-}H}$, energetically favorable, that the system allows. This state corresponds to the zero energy value of $\Delta E_{W\text{-}H}$. Assuming that all the dislocations relax in the same way (identical 60° Burger vector), $\rho_{W\text{-}H}$ is estimated solving numerically the following equation:

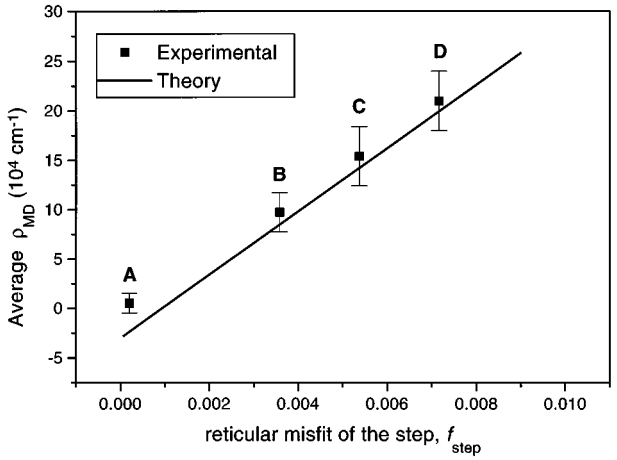

FIG. 3. Average misfit dislocation densities for each structure determined by statistical counting of cross-section TEM observations (open circle with statistical error bars). The predicted work-hardening density (full line) vs the steps of composition are shown to fit very well with the experimental data and can be analytically expressed by the Eq. (3) linear regression of the calculated data.

$$\Delta E_{W\text{-}H}[h,f_n,\rho_{W\text{-}H}(h,f_n)]=0, \tag{2}$$

where f_n is the reticular misfit of the n layer with respect to the $n-1$ layer and h is the individual layer thickness. Figure 1 shows the strain thickness diagram with previously published experimental data[14] and the model predictions for $In_{0.1}Ga_{0.9}As/GaAs$. The dashed curve corresponds to the strain relaxation model predictions when $\Delta E_{W\text{-}H}=0$ (deduced applying $\varepsilon=f-\rho_{W\text{-}H}b/2$).

In the case of single epilayers, the model only describes correctly the relaxation for high thickness (region C) where the work-hardening process limits the strain relief. In region B (see arrow), for single layers, the relaxation is governed by kinetic processes that affect the nucleation and multiplication mechanisms of new dislocations. This is not the case for compositionally graded structures. Indeed, these relax much faster than single layers due to the strain energy accumulated in the highly mismatched upper layers. This energy allows the generation of supplementary dislocations and the layer relaxes faster. The solid circle point in Fig. 1 corresponds to the relaxation state of the buried layers of sample D. It lies rather below the empirical curve of Dunstan[1] and is in good agreement with our work-hardening model predictions.

The MD densities deduced for different misfit steps are calculated using Eq. (2). In Fig. 3 their values are shown to fit the experimentally measured MD density perfectly. Each density data point is the average of values for all the buried interfaces of the sample. The upper layer has a lower MD since it is still at the second stage of relaxation and is therefore not considered in the displayed MD density average. A good prediction of the MD density is obtained by the linear regression of the calculated data

Downloaded 20 Oct 2006 to 150.214.231.66. Redistribution subject to AIP license or copyright, see http://apl.aip.org/apl/copyright.jsp

$$\rho_{W\text{-}H} = 0.42 \times 10^4 + 3.8 \times 10^7 f, \quad (3)$$

where $\rho_{W\text{-}H}$ is the MD density (expressed in cm^{-1}) predicted at the saturation state. The proposed model allows the prediction of the distribution of MD and the strain in buffer structures with constant graded composition. Each mismatched step corresponds to a different MD density as shown in Fig. 3.

This behavior shows that, in contrast to the Dunstan predictions,[14] the reticular misfit is not totally relieved in the buried layer. The displayed MD values would only allow a relaxation of 75%–85% of the reticular misfit of the underlying layers. As a consequence of work-hardening processes, residual strain of 15%–25% remains and is added to the layer above inducing a linear increase in strain versus thickness. As a result, at the end of the structure, a relaxation in the order of 70%–80% is obtained in all the samples. This is not better than that obtained in a single layer, but as shown in the micrographs of Fig. 2, the MDs are distributed between all the interfaces, causing a strong limitation of the threading dislocation formation. Independent of the number of layers in the stack, the integrated MD densities, ρ_t (sum of all interfaces), measured by TEM amount to $5-6\times10^5$ cm^{-1}. The same total number of MDs can be introduced into single layers or graded buffer layers. The corresponding densities are determined by the work-hardening model predictions in such a layer thickness range. Thus, in a step-graded structure of n layers, due to the well-organized distribution of MDs between all the interfaces, a density of $\rho_t/n(\approx 6/n \times 10^5$ $\mathrm{cm}^{-1})$ is introduced at each interface. Assuming that the density of TDs has a squared behavior regarding the MD density ($\rho_{TD} \propto \rho_{MD}{}^2$), basically due to MD interactions,[15] the formation of TDs falls in graded structures by a factor of $1/n^2$ with respect to single layers. This fact, together with the high residual strain of the last layer, makes possible the observed low threading dislocation density.[3] Indeed, the samples studied have TDs densities lower than the detection limit of the PVTEM technique (10^5 cm^{-2}).

In conclusion, we have modeled the strain relaxation in step-graded stacks of multilayers. This allows us to estimate the saturation state of relaxation reached by such structures and thus a prediction of the extent of strain relief is possible. The density of misfit dislocations at individual interfaces, that increases linearly with the step composition, is well predicted by the work-hardening model presented. The repulsion of a new dislocation by a fixed array of preexisting MDs is shown to be the dominant relaxation process at this stage. Our calculations demonstrate that the internal layers never reach a complete relaxation, causing an accumulation of the residual strain from layer to layer. The presented model permits a good prediction of relaxation along the buffer thickness and can be applied to buffer layer design for device fabrication.

This work was made possible through grants from the National Materials and Science program (MAT Project No. 94/0538), and from the Andalucian government (Group No. TEP-0120). The work was carried out at the Electron Microscopy Center of Cádiz University. The authors wish to thank Dr. P. Kidd for the x-ray measurements and J. M. Geraldía and Dr. R. Baker for technical support during this study. Thanks are due to Dr. E. Calleja and Dr. A. Sacedón from Universidad Politecnica de Madrid and to Dr. L. González and Dr. Y. González from Instituto Nacional de Microelectrónica de Madrid for growing the samples.

[1] D. J. Dunstan, P. Kidd, L. K. Howard, and R. H. Dixon, Appl. Phys. Lett. **59**, 3390 (1991).
[2] V. Krishnamoorthy, Y. W. Lin, L. Calhoun, H. L. Liu, and R. M. Park, Appl. Phys. Lett. **61**, 2680 (1992).
[3] J. Tersoff, Appl. Phys. Lett. **62**, 693 (1993).
[4] S. I. Molina, F. J. Pacheco, D. Araújo, R. García, A. Sacedón, E. Calleja, and P. Kidd, Appl. Phys. Lett. **65**, 2460 (1994).
[5] D. González, D. Araújo, G. Aragón, and R. Garcia, Appl. Phys. Lett. (to be published).
[6] D. D. Perovic, Inst. Phys. Conf. Ser. **146**, 117 (1995).
[7] J. W. Matthews and A. E. Blakeslee, J. Cryst. Growth **27**, 118 (1974).
[8] R. Beanland, J. Appl. Phys. **72**, 4003 (1992).
[9] B. W. Dodson and J. Y. Tsao, Appl. Phys. Lett. **51**, 1325 (1987).
[10] B. W. Dodson, Appl. Phys. Lett. **53**, 37 (1988).
[11] T. Y. Zhang, J. E. Hack, and L. J. Guido, J. Appl. Phys. **75**, 2358 (1994).
[12] F. J. Pacheco, D. González, S. I. Molina, M. P. Villar, A. Sacedón, E. Calleja, and R. Garcia, Inst. Phys. Conf. Ser. **146**, 211 (1995).
[13] P. M. Mooney, J. L. Jordan-Sweet, J. O. Chu, and F. K. LeGoues, Appl. Phys. Lett. **66**, 3642 (1995).
[14] D. J. Dunstan, P. Kidd, P. F. Fewster, N. L. Andrew, L. González, Y. González, A. Sacedón, and F. González-Sanz, Appl. Phys. Lett. **65**, 841 (1994).
[15] L. B. Freund J. Appl. Phys. **68**, 2073 (1990).

Downloaded 20 Oct 2006 to 150.214.231.66. Redistribution subject to AIP license or copyright, see http://apl.aip.org/apl/copyright.jsp