Bunches of misfit dislocations on the onset of relaxation of Si$_{0.4}$Ge$_{0.6}$/Si(001) epitaxial films revealed by high-resolution x-ray diffraction

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The experimental x-ray diffraction patterns of a Si$_{0.4}$Ge$_{0.6}$/Si(001) epitaxial film with a low density of misfit dislocations are modeled by the Monte Carlo method. It is shown that an inhomogeneous distribution of 60° dislocations with dislocations arranged in bunches is needed to explain the experiment correctly. As a result of the dislocation bunching, the positions of the x-ray diffraction peaks do not correspond to the average dislocation density but reveal less than a half of the actual relaxation.

Si$_{1-x}$Ge$_x$ films on Si substrate constitute a heteroepitaxial system that finds numerous applications in the whole compositional range [1] and, at the same time, is a model system that demonstrates the whole spectrum of strain relaxation mechanisms. When either thickness or Ge content is small, the layers stay strained, by accepting the lateral lattice spacing of the substrate and expanding vertically due to the Poisson effect. Larger strain is relaxed by one of the two mechanisms, plastic relaxation in planar layers by introduction of misfit dislocations at the interface [2-5] or development of three-dimensional islands in the Stranski–Krastanov growth mode [6]. Planar films with controlled strain state are required for various applications. Particularly, the high compressive strain in the Si$_{0.4}$Ge$_{0.6}$ films, studied in the present work, ought to enhance a room-temperature two-dimensional hole gas mobility, which is important for application of such films in the field effect transistors [7].

X-ray diffraction is a well established technique to characterize relaxed epitaxial films. Positions of the x-ray peaks provide the lattice parameters of a relaxed film and hence the density of misfit dislocations [8,10]. An application of the same analysis at the onset of relaxation suffers from the peak broadening due to a small layer thickness [11]. Moreover, we show below that the inhomogeneity in the dislocation distribution plays an essential role in the x-ray diffraction analysis. The position of the coherent peak is given by the less strained regions of the film and hence underestimates the relaxation. The diffuse x-ray intensity occurs more sensitive to both the presence of misfit dislocations and their distribution.

We have chosen for a detailed x-ray diffraction study a 27 nm thick Si$_{0.4}$Ge$_{0.6}$ film on Si(001), demonstrating an early stage of the relaxation. Thinner unrelaxed and thicker relaxed films of the same series of samples were studied earlier [12]. The samples were grown by reduced pressure chemical vapor deposition in an industrial ASM Epsilon 2000 system. Germane and disilane precursors were used to grow Si$_{0.4}$Ge$_{0.6}$ epilayers at the growth temperature of 450°C. The critical thickness for plastic relaxation at a Ge content $x = 0.6$ is 10 nm [11,13,14]. The low growth temperature allows us to obtain 2.7 times thicker layer possessing a small relaxation.

High-resolution x-ray diffraction measurements were performed using a 9 kV SmartLab Rigaku diffractometer with a rotating anode. The diffraction setup included a two-crystal Ge monochromator in the 400 setting and a one-dimensional high-speed position-sensitive detector D/teX Ultra from Rigaku.

Figures 1(a-d) present experimental reciprocal space maps in the symmetric 004 and several asymmetric reflections, in a sequence of increasing asymmetry. The wave vectors are represented in the dimensionless units of the product of the components of the reciprocal space vector ($q_x, q_z$) and the film thickness $d = 27$ nm. Each map comprises a coherent scattering streak extended along the surface normal, and diffuse scattering. The presence of the coherent and the diffuse intensities is an indication of a weakly distorted film, possessing a low density of misfit dislocations. A closer inspection of the symmetric 004 map in Fig. 1(a) reveals that the positions of the coherent and the diffuse maxima do not coincide: with the origin $q_z = 0$ chosen at the position of the coherent peak, the diffuse intensity is maximum at $q_z d \approx 5.5$.

Figure 2(a) shows line scans extracted from the maps perpendicular to the scattering vectors (ω-scans) at the intensity maxima of the respective maps. For the 004 reflection, two scans are presented, one through the maximum of the coherent intensity (gray line) and the other through the maximum of the diffuse intensity (green line). Evidently, the former scan shows a larger peak intensity, while the latter has a higher diffuse scattering intensity. In asymmetric reflections, the scans through the coherent and the diffuse maxima reveal a less pronounced (albeit present) difference, and we present only the scans through the coherent maxima.

The reciprocal space maps in asymmetric reflections in Figs. 1(b-d) reveal a strong asymmetry of the diffuse intensity distributions. For each reflection, the coherent streak at $q_x = 0$ separates the diffuse intensity in two lobes, the one at $q_x < 0$ possessing notably higher intensity in comparison with the other at $q_x > 0$. This
The directions of the scattering vectors are shown by white arrows. The insets in (a-d) show full reciprocal space maps comprising the substrate and the film peaks.

Reciprocal space maps calculated by the Monte Carlo method in Figs. 1(e-h) and the diffraction profiles shown by black lines in Fig. 2 demonstrate a quantitative agreement with the experimental maps and curves. Now we describe the Monte Carlo model of the dislocation distribution that we have used. We assume two arrays of straight misfit dislocations with the dislocation lines in the two orthogonal (110) directions. For dislocations with the lines normal to the scattering plane \((x, z)\), Burgers vectors \(\mathbf{b} = \frac{1}{2}(011)\) have the same component \(b_x = -a/2\sqrt{2}\) releasing the misfit, while the signs of two other components, screw \(b_y = \pm a/2\sqrt{2}\) and edge \(b_z = \pm a/2\), are chosen on random and uncorrelated (here \(a\) is the lattice parameter of the substrate). The position of the Bragg peak corresponding to the average relaxation is given by \(q_0x = \rho Q_x b_x\) and \(q_0z = -\frac{Q_z}{\nu} \rho Q_x b_z\), where \(\rho\) is the linear density of misfit dislocations and \(Q_x, Q_z\) are the components of the reciprocal lattice vector. The shift \(q_0z\) is taken into account in Figs. 1(e-h), but the \(q_0x\)-shift is not made, as discussed below.

To reach an agreement between the experimental and the calculated curves in Figs. 1 and 2 we varied the dislocation density and the distribution of dislocations. The dislocation density \(\rho d = 0.5\) used in the Monte Carlo calculations corresponds to a relaxation degree \(R = 0.05\). The positions of the dislocations are modeled as a Markov chain, with the probability \(P\) to have a distance \(\rho^{-1}P\) between two subsequent dislocations possessing a lognormal distribution. The probability density is generated as
The standard deviation of the lognormal distribution is the standard deviation of the distribution, 

\[ \sigma = \exp \left( \mu + \frac{\sigma^2}{2} \right) - 1 \]

where \( \mu \) and \( \sigma^2 \) are the mean and variance of the lognormal distribution, respectively. The parameters \( \mu \) and \( \sigma \) are calculated for the dislocation distribution. On a mesoscopic scale, the surface relief exhibits rather sharp peaks caused by dislocation bundles separated by relatively flat regions. In calculating surface displacements from dislocations, we assume that the slip steps are eliminated by surface diffusion, as proposed by Andrews et al.\(^\text{[23–25]}\), so that the surface displacement due to each dislocation is a continuous function of the coordinate \( x \). The x-ray diffraction profile, calculated for this distribution of the dislocation positions, is shown in Fig. 3(d) by the black line. It is calculated at \( q_z = q_{0z} \), i.e., at the \( q_z \) position in between the ones presented in Fig. 3(a), for the same model of the dislocation distribution.

Figures 3(b,c) present, for a comparison, more homogeneous dislocation distributions modeled in the literature\(^\text{[23–25]}\). The dislocation positions in Fig. 3(b) are chosen on random independently from each other, and the signs of the tilt components of their Burgers vectors are also not correlated. The dislocation density is the same, \( \rho d = 0.5 \). The surface displacements (red line) are qualitatively similar to those in Fig. 3(a) but have less pronounced peaks and smaller flat areas. However, the x-ray diffraction profile calculated for this model by

\[ P = \exp \left[ \mu + \sigma N(0,1) \right], \]

where \( N(0,1) \) is the standard normal distribution with the mean 0 and dispersion 1. The standard deviation of the lognormal distribution is taken to be \( s = 10 \) times larger than its mean value. Explicitly, the parameters of the lognormal distribution are \( \sigma = \sqrt{\ln(1 + s^2)} = 2.15 \) and \( \mu = -\sigma^2 / 2 = -2.31 \).

Figure 3(a) shows an example of the dislocations distributed according to our model, and the surface displacement caused by these dislocations. The positions of the dislocations are marked by vertical bars. Blue and green bars correspond to dislocations with opposite signs of the tilt component of the Burgers vector \( b_z \). The large standard deviation of the distribution gives rise to bunches of dislocations separated by dislocation-free regions. The black curve in Fig. 3(a) is the surface displacement calculated for this dislocation array. On a mesoscopic scale,
the same Monte Carlo method [red curve in Fig. 3(d)]
looks qualitatively different. It possesses the side max-
ima described theoretically for uncorrelated random mis-
fit dislocations [16] and observed experimentally for
InP/Ga0.9As/GaAs(001) [15], Si0.75Ge0.25/Si(001) [16],
and ZnSe/GaAs(001) [17] epitaxial films.

Figure 3(c) shows a model with random dislocation
positions but correlated Burgers vectors. Alternating
regions containing a given number of dislocations with
the same tilt component of the Burgers vector b were
considered in Ref. [25]. In our model, the number of dis-
locations in a group is taken on random: the sign of
b is changed with the probability p = 0.1, so that there are
on average 10 dislocations in a group. The surface profile
(blue line) varies over a larger lateral scale and a larger
height. The diffraction profile calculated for this model
[blue line in Fig. 3(c)] exhibits the same side maxima as
the one for the case of uncorrelated positions and Burg-
ers vectors above. Thus, the bunching of dislocations is
required to explain our experimental diffraction profiles.

Figure 3(c) presents the surface relief of the investi-
gated Si0.4Ge0.6/Si(001) film as measured by atomic force
microscopy (AFM). The cross-hatch pattern observed in
this micrograph is a well-known manifestation of plastic
relaxation [23,29]. With the dislocation density pd = 0.5
as determined from the x-ray data, the number of disloca-
tions on a 10 µm interval is 185, while only about 35 ran-
domly spaced parallel lines are seen in Fig. 3(e). Hence,
a single line in the AFM image corresponds to a group of
dislocations, rather than a single dislocation. This is in a
good agreement with our analysis and the calculated pro-
file in Fig. 3(a). The formation of the cross-hatch pattern is a
result of a complicated interplay between surface dif-
fusion and dislocation generation. It remains debatable,
if the dislocations cause the surface undulations or, oppo-
sitely, the undulations due to surface diffusion are sources
of dislocations [29]. Hence, we do not make a quantita-
tive comparison of the measured and the modeled surface
profiles.

The expected shift q0z of the coherent 004 peak due to
an average strain calculated by the expression given
above for the dislocation density of our sample (pd = 0.5)
is equal to q0z ≈ 4.4. This shift is taken into account
in Fig. 2(b), so that the intensity maximum is expected to
be at the origin, qz − q0z = 0. We have verified this
prediction by additional Monte Carlo calculations (not
shown) of similar diffraction profiles for uncorrelated or
more ordered dislocations, which give the intensity max-
ima at the expected position. However, the peak of the
calculated curve in Fig. 2(b) is at (qz − q0z)d ≈ −2.65.
Hence, the coherent peak position corresponds to less
than half of the actual film relaxation.

The difference between expected and calculated peak
positions can be explained by the dislocation bunching,
which gives rise to regions with large and small strains,
as it is reflected in the surface profile in Fig. 3(a). The
dislocation-rich regions possess large strain and large
strain inhomogeneity, so that they contribute mostly
to the diffuse scattering. In contrast, the dislocation-
depleted regions possessing small strain and small strain
gradients contribute to the coherent intensity. As a re-
sult, the positions of the coherent and the diffuse in-
tensity maxima on the calculated reciprocal space maps in
Figs. 4(e-h) do not coincide: the coherent peak re-
fects the areas in the sample which are less strained than
the average, while the diffuse peak represents the more
strained ones.

The coherent peaks in Figs. 4(f-h) remain at the same
lateral position qz = 0 as they are in an elastically rel-
exed dislocation-free film. This peak position has been
analyzed in Ref. [10] [see discussion after Eq. (27)] and
in Ref. [30]. One can also see from the experimental
maps in the insets in Figs. 4(a-d), that the qz-positions
of the substrate and the film peaks coincide. The in-
tensity maxima move to qz = q0x when the dislocation
density is increased, the coherent peak weakens, and the
diffuse peak dominates.

Summarizing, diffuse x-ray intensity from misfit dislo-
cations can be revealed at the very early stages of relax-
ation of the epitaxial films, when the shift of diffraction
peaks due to these dislocations is not yet visible. The
diffuse intensity distribution is sensitive to the spatial
arrangement of misfit dislocations. We model the dislo-
cation distribution by the Monte Carlo method and find
that the diffraction pattern from a Si0.4Ge0.6/Si(001) epi-
taxial film on the onset of relaxation is due to a very in-
homogeneous dislocation distribution. Distances between
dislocations vary very broadly, so that the standard de-
velopment of the dislocation spacings is 10 times larger than
the mean distance between dislocations. In other words,
dislocations form bunches, as a result of the action of
small number of dislocation sources. These bunches are
seen as cross-hatch patterns in the AFM images of the
film.

The inhomogeneous dislocation distribution results in
peculiar features of the diffraction patterns. The posi-
tions of the coherent and the diffuse peaks do not co-
icide, since the former is mostly due to undisturbed
regions between dislocation bunches while the latter is
due to the inhomogeneous strain at the bunches. More-
over, since the coherent peak represents the undisturbed
regions, rather than the strain averaged over the whole
film, its position at the onset of relaxation does not cor-
respond to the actual density of misfit dislocations and
underestimates relaxation by more than a factor of 2.

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