# Crosshatching on a SiGe film grown on a Si(001) substrate studied by Raman mapping and atomic force microscopy

H. Chen,* Y. K. Li, C. S. Peng, H. F. Liu, Y. L. Liu, Q. Huang, and J. M. Zhou
*Institute of Physics, The Chinese Academy of Sciences, Beijing 100080, China*

Qi-Kun Xue
*State Key Laboratory for Surface Physics, Institute of Physics, The Chinese Academy of Sciences, Beijing 100080, China*

(Received 25 March 2002; published 23 May 2002)

The morphology, stress, and composition distributions of the crosshatch pattern on a SiGe film grown on a Si(001) substrate using a low-temperature Si buffer are studied by atomic force and Raman microscopies. Crosshatching is not related to composition fluctuation regardless of the stress undulation associated with strain relaxation in the SiGe film. The crosshatch morphology arises from vertical lattice relaxation induced by piled-up misfit dislocations in the Si buffer layer and substrate. A model for crosshatch formation is proposed.

DOI: 10.1103/PhysRevB.65.233303 PACS number(s): 68.55.−a, 78.30.−j, 81.15.Hi, 68.37.Ef

The growth of mismatched semiconductor materials such as SiGe/Si and InGa(Al)As/GaAs has been the subject of extensive experimental and theoretical studies over past decades,[1–3] driven by the potential of creating novel electrical and optical devices as well as integrating existing devices with different materials. One of the most important issues is to grow a defect-filtering buffer with low density of dislocations between the overlying device structure and the substrate.[4,5] The formation of defects (dislocations) associated with strain relaxation in the buffer layer is the major factor that deteriorates device performances. Attempts to eliminate such defects have led to the development of several novel growth methods. Among these, the compositional grading[4,6,7] and low-temperature buffer[8–10] techniques are the two most successful, leading to reductions in the density of threading dislocations by 4–5 orders of the magnitude.

However, irrespective of the methods used, strain relaxation always results in an undulated surface morphology,[11,12] referred to as a "crosshatch" pattern, as well known for nearly 30 years.[13] The crosshatch is characteristic of the low misfit regime (lattice mismatch less than 2%) and so far has been observed in almost all heterostructures studied, including SiGe/Si, InGaAs/GaAs, AlGaAs/GaAs, InGaP/GaAs, and InGaP/GaP, as long as the dislocation density in the relaxed film is reduced to $10^4–10^6$ cm$^{-2}$. Such crosshatch-related surface roughening can affect carrier mobility and the quality of quantum structures grown on such surfaces. It is not compatible with planar integrated circuit technologies, the major driver for applications. To eliminate the defects it is therefore very important to understand their structure and origin.

Surface morphology evolution during strain relaxation and the mechanism by which dislocations give rise to crosshatching have been studied mainly by atomic force microscopy (AFM) and transmission electron microscopy (TEM), and basically two types of modes have been proposed.[11,12,14–17] In the first model, the formation of crosshatch patterns is caused by surface diffusion during growth in response to the inhomogeneous strain fields arising from the network of misfit dislocations.[12,14,16] The second model suggests that crosshatching results from the surface steps at the intersection of the epilayer surface and the misfit-dislocation glide plane, and is a result of plastic shear displacements.[11,15,17] The two mechanisms are in fact not mutually exclusive.[11] In this paper, we report the results of a combined AFM and micro-Raman mapping study of the crosshatch pattern, and establish the relation between composition and strain distribution. We propose a model that eliminates the major problems of previous models.

Our sample was grown on a Si(001) substrate in a VG Semicon V80S molecular beam epitaxy (MBE) system. A 50-nm low-temperature Si (LT-Si) layer was first grown at 400 °C,[8–10] followed by a 500-nm-thick $Si_{0.75}Ge_{0.25}$ epilayer at 550 °C. A growth rate of 0.1 nm/s was used for both Si and SiGe layers. Raman spectra were measured with a Renishaw 1000 Raman microscope at room temperature, using the 514.5-nm line of an $Ar^+$ laser. A 180° backscattering geometry was employed with a spectrum resolution of 2 cm$^{-1}$. The spatial resolution in the $x$ and $y$ directions is about 1 $\mu$ m. The beam size of the laser is about 10 $\mu$m for Raman spectroscopy and 1 $\mu$m for Raman microscopy. In order to examine the dependence of crosshatch morphology on epilayer thickness, a special chemical mechanical polishing procedure was used to remove certain layers of the film.

The typical surface morphology of the samples measured by *ex situ* AFM is shown in Fig. 1(a). The crosshatching along the two ⟨110⟩ directions due to the formation of underlying misfit dislocations is immediately evident. Surface roughness is mainly due to the crosshatch undulation, and its average amplitude is approximately 6 nm, as determined by sampling an area of $50\times50$ $\mu$m$^2$. The crosshatch lines in Fig. 1(a) are straight and long, which indicates long misfit dislocations and presumably few threading dislocations.[8] Figure 1(b) shows the surface morphology for the sample after 200-nm-thick epilayer was removed and the crosshatching pattern still exists.

Shown in Fig. 2 is the Raman spectrum of the sample. In addition to the high-energy line at the phonon frequency of ~520.5 cm$^{-1}$ for the Si substrate, the spectrum exhibits three distinct peaks located at 290.1, 408.4, and 511.6 cm$^{-1}$, respectively. These peaks are interpreted in terms of atomic vibrations involving Ge-Ge, Si-Ge, and Si-Si bonds in the

 ©2002 The American Physical Society

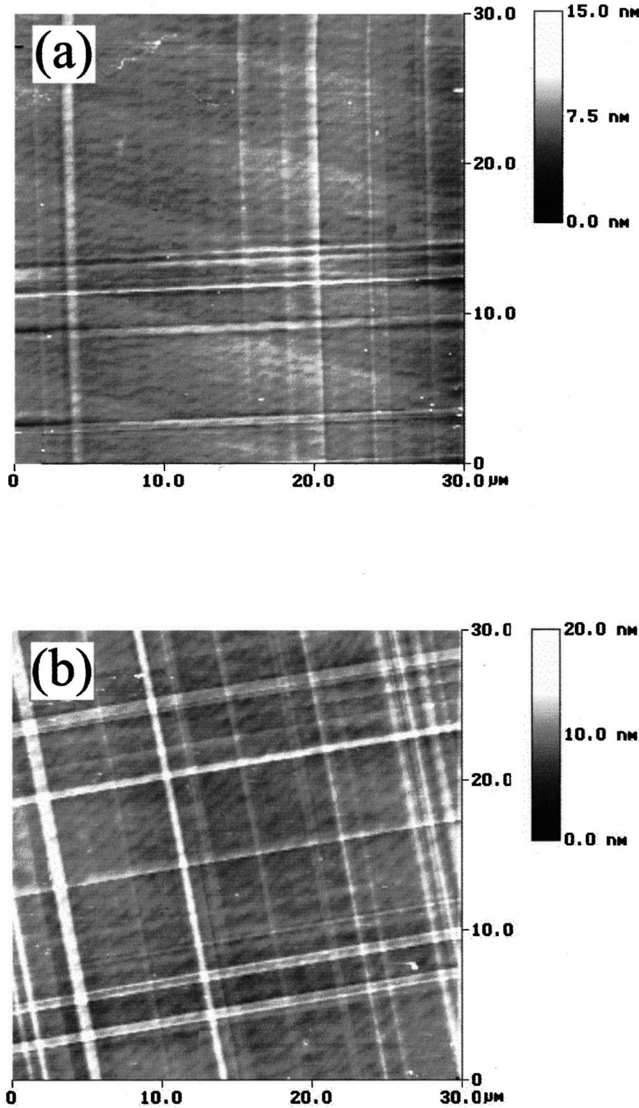


FIG. 1. (a) AFM image of the SiGe film grown on a LT-Si buffer on a Si substrate. Crosshatches on the SiGe surface with an average depth of about 6 nm are observed. (b) AFM image for the sample after 200-nm-thick was removed.

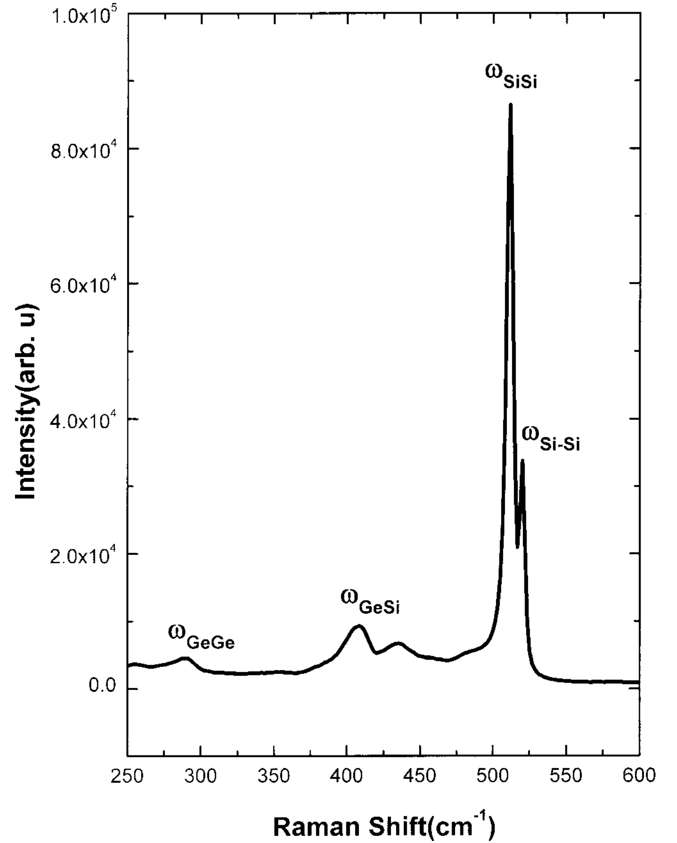


FIG. 2. Raman spectrum from SiGe film of Fig. 1. The additional features between the Si-Ge and Si-Si peaks is attributed to motion of Si atoms in specific local environments.

film, respectively.[18–21] As the relative intensities and energies of the Ge-Ge, Si-Ge, and Si-Si vibrations are dependent of the relative number and the strain-induced distortion of corresponding bonds in the alloy,[18–21] we use the spectrum to determine the alloy composition and strain based on the method by Tsang *et al.*[21] and later by Groenen *et al.*[22] For $0<x<0.5$, the dependences on composition $x$ and in-plane strain $\varepsilon$ of the SiSi, SiGe, and GeGe phonon frequencies $\omega$ can be written as

$$\omega_{\mathrm{SiSi}}=520.5-62x-815\varepsilon, \tag{1}$$

$$\omega_{\mathrm{SiGe}}=400.5+14.2x-575\varepsilon, \tag{2}$$

$$\omega_{\mathrm{GeGe}}=282.5+16x-385\varepsilon. \tag{3}$$

Using the experimental values for the phonon frequencies measured in Fig. 2, we obtain a composition $x=0.244\pm0.003$ and strain $\varepsilon=-0.001$, which indicates 92% stress relaxation. The residual strain measured by our Raman spectroscopy is in good agreement with that of the x-ray diffraction measurement by Li *et al.*[10]

Shown in Fig. 3 are the Raman images of TO-phonon shifts[23] for the SiSi bonds from the substrate [Fig. 3(a)] and for the SiSi [Fig. 3(b)] and SiGe [Fig. 3(c)] bonds from the SiGe epilayer taken in the same spatial region. Cross stripes can be observed in these images. In comparison with the AFM image, we confirm that this type of cross striping corresponds to the crosshatching of the SiGe film.

We next calculate the changes of strain and composition using Eqs. (1)–(3). The results are shown in Fig. 4. Surprisingly, no matter how inhomogeneous the surface topography, there is no observable change in composition. *The Ge is basically distributed uniformly in the epilayer*, as we clearly see in Fig. 4(b). The bright and dark spots that provide the greatest contrast variation in the image correspond to two threading dislocations terminated at the surface. In contrast, the strain in the epilayer follows the undulation of the crosshatch pattern and varies significantly from $-0.0075$ to $-0.0015$ with its lowest value in the crosshatched region where the strain relaxation is most complete [Fig. 4(c)]. In the case of the substrate [Fig. 4(a)], the strain changes sign and varies from $-0.00072$ to 0.00072, which indicates that some regions are compressively strained as well. It is not difficult to understand this result if we reverse the epitaxy

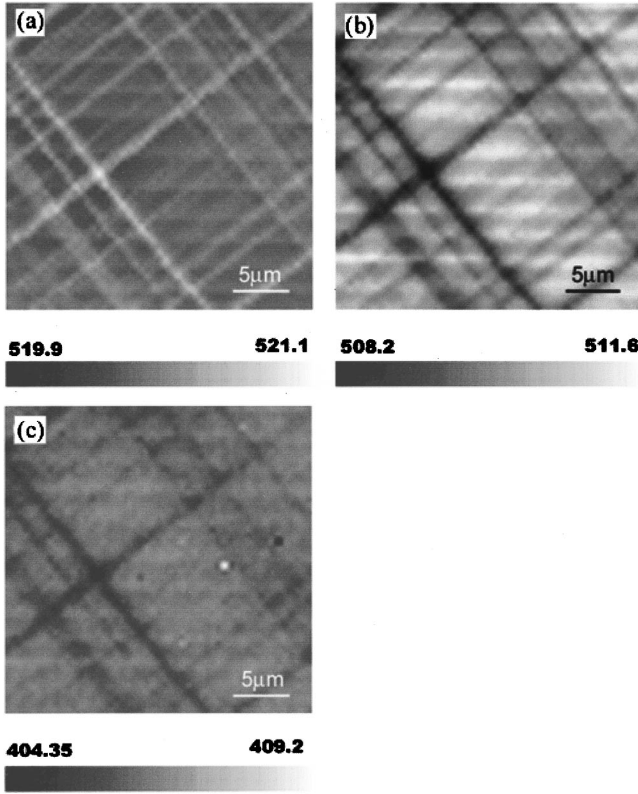


FIG. 3. Raman mapping of TO phonon shifts for the Si-Si bonds of the substrate (a), the Si-Si bonds of the film (b); and the Si-Ge bonds (c) of the SiGe film. The contrast variation of the image indicates the TO phonon shifts.

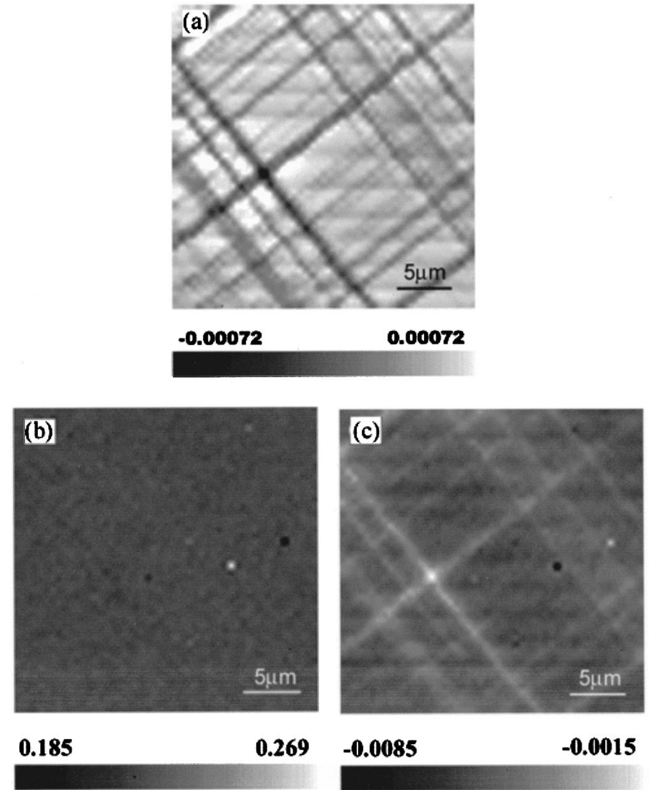


FIG. 4. Distributions of the strain in the Si substrate (a) and Ge composition (b) and strain (c) in the film.

relationship—the Si substrates were "epitaxially grown" on the SiGe. The Si "film" with smaller lattice constant will be tensily strained. Due to the formation and gliding of the misfit dislocations in the Si "film," the local lattice constant could become larger than the normal region, giving rise to a compressive strain in the region beneath the crosshatches.

To estimate the depth of crosshatching, we first calculate the change of the crystal parameters in the direction vertical to the surface in both crosshatched and normal regions, which we denote as $b_\perp$ and $a_\perp$, respectively:

$$b_\perp(a_\perp)=[a_{\mathrm{si}}+x(a_{\mathrm{Ge}}-a_{\mathrm{Si}})](1+\varepsilon_\perp), \tag{4}$$

where $\varepsilon_\perp$ is the component of the strain tensor in the growth plane.

If we ignore the strain variation in the substrate, then the crosshatching depth would be expressed by $l(a_\perp - b_\perp)/a_{\mathrm{SiGe}}$, where $l$ is the thickness of the SiGe layer and $a_{\mathrm{SiGe}}$ is the crystal parameter of the fully relaxed SiGe.[24] Using the calculating method and parameters in Ref. 24, a value of ~30 Å for the depth is obtained. We have also used Hirth and Lothe's classic theory to estimate the surface displacement caused by the dislocation pileups.[25] According to them, the atomic displacements along the two polar coordinate axes ($r$ and $\theta$) are

$$u_r=\frac{b}{2\pi}\left[1-\frac{(2-2\nu)}{2(1-\nu)}\sin\theta\ln r+\frac{\sin\theta}{4(1-\nu)}+\theta\cos\theta\right],$$

$$u_\theta=\frac{b}{2\pi}\left[-\frac{(1-2\nu)}{2(1-\nu)}\cos\theta\ln r-\frac{\cos\theta}{4(1-\nu)}-\theta\sin\theta\right],$$

in which $\nu=0.3$ is the Possion constant, and $b$ is the edge component of the misfit dislocation and takes a form of $(\sqrt{2}/2)\cos 30°a$ ($a=5.431$ Å, the lattice constant of Si). In the present case, $r=1$ $\mu$m and $\theta=0$. Using these parameters and the above expressions, we obtain $u_r=0$ and $u_\theta=1.6$Å, which agrees with our measurement very well.

Our cross-section TEM experiment shows that the strain relaxation of SiGe film with LT-Si buffers is similar to that for compositionally graded SiGe layers and is of the modified Frank-Read (MFR) type as well documented by LeGoues *et al.*[6,7] We also often observe some reversed V-shaped alignment of the dislocations gliding from two sets of ⟨111⟩ planes deep into the substrate generated by the MFR source, which is again similar to that found in compositionally graded SiGe layers (see Fig. 4 in Ref. 6, for example).

In the following, we will show that the present results cannot be explained solely by the two models mentioned above or by their combination. If the crosshatching corresponds to an atomic step of 2.8 Å at the intersection of the associated glide plane and the epilayer from a single 60° dislocation (Burgers vector=$a/2$ [101]),[11] the undulated morphology will be a sum of the individual surface step displacements from the dislocations involved in the MFR pileup process. For the film shown in Fig. 1, typically ~10 dislocations are observed in pileups. Thus a total displacement of ~28 Å from the (001) plane is expected. This is

much smaller than the values observed by AFM and estimated in our calculation. Moreover, it will lead always to a step bunching and thus a dipolelike morphology, rather than the observed V shape. Further, if the crosshatches are indeed due to this plastic displacement, it should be possible to remove them by polishing. However, such a pattern still exists even after our ~200-nm-thick epilayer is removed. On the other hand, if the undulation pattern is driven by the growth kinetics associated with the relaxation-induced inhomogeneous strain fields,[12,14,16] then we will expect a simultaneous composition fluctuation in addition to the strain undulation, due to inequivalent surface diffusion and incorporation of Ge and Si atoms.[26] This is not observed here [see Fig. 4(b)]. This model has also difficulties with the polishing observation.

In our model, crosshatching is mainly related to a nonuniform elastic field caused by the beneath dislocations associated with strain relaxation. When a misfit dislocation forms, it creates a strain field that causes lattice expansion in the (001) plane and contraction of the interlayer distance along the ⟨001⟩ direction. This contraction will lead to a downward movement of the layers above the dislocation. The extent of the movement of an individual layer depends on the distance $z$ from the dislocation along the ⟨001⟩ direction. This is proportional to $z^{-2}$, because the stress of dislocation decays as $z^{-2}$. Thus the total displacement ($\Delta$) at the surface, caused by this interface dislocation, should be an integral of the contraction $\delta_z$ against $z$ for entire thickness of the epilayer.

Meanwhile, in the (001) plane, the relaxation of the neighboring atoms also occurs and decreases with increasing distance from the dislocation. Considering all possible movements involved, we should expect an oval or half-sinusoidal morphology at the surface along the two [110] directions. Due to dislocation pileup, the dislocations gliding deeper into the Si substrate should also make contributions to the morphology change. These contributions would further increase the magnitude of the distortion and make the oval or half-sinusoidal shapes steeper, which would eventually result in a V-shaped crosshatch. According to our model, it will be impossible to remove the crosshatch pattern by polishing unless the underlying dislocations are eliminated.

Our model does not exclude other two models. In reality, two and even all three processes might come into play together. For example, atoms are more easily incorporated into the stress-released regions, as much more energy is gained in this case. Strain-driven adatom diffusion can enhance the growth process for crosshatching.[12,14,16] This might be a good explanation for the discrepancies in morphology measured by different methods. However, as a fundamental process, the nonuniform elastic field caused by the underlying dislocations associated with strain relaxation should be considered in any successful model to explain the formation of crosshatching regardless of the methods and systems in which crosshatch pattern occurs.

In summary, we have investigated surface crosshatching of a relaxed SiGe film grown on a LT-Si buffer on a Si substrate, using AFM and Raman spectroscopy and mapping. The most interesting result is the uniform composition distribution independent of the stress undulation that causes the crosshatch pattern. By comparing the results obtained from different measurement methods and calculations, we show that the nonuniform elastic field caused by the underlying dislocations associated with strain relaxation is essential to explain the crosshatch pattern.

The authors would like to thank Professor Dave E. Aspnes and Dr. Kevin F. Kelly for critical reading and comments of the manuscript. The work is partly supported by the National Science Foundation of China through Grant Nos. 59902013 and 69625608. The authors would also like to thank Ranishaw Beijing Office for help in the Raman mapping measurements.

*Corresponding author. Electronic address: hchen@aphy.iphy.ac.cn

[1] G. Abstreiter *et al.*, Phys. Rev. Lett. **54**, 2441 (1985).
[2] Y. H. Xie *et al.*, J. Appl. Phys. **73**, 8364 (1993).
[3] E. A. Fitzgerald, Mater. Sci. Rep. **7**, 87 (1991).
[4] E. A. Fitzgerald *et al.*, Appl. Phys. Lett. **59**, 811 (1991).
[5] D. P. Malta *et al.*, Appl. Phys. Lett. **60**, 844 (1992).
[6] F. K. LeGoues, B. S. Meyerson, and J. F. Morar, Phys. Rev. Lett. **66**, 2903 (1991).
[7] F. K. LeGoues *et al.*, J. Appl. Phys. **71**, 4230 (1992).
[8] H. Chen *et al.*, J. Appl. Phys. **79**, 1167 (1996).
[9] K. K. Linder *et al.*, Appl. Phys. Lett. **70**, 3224 (1997).
[10] J. H. Li *et al.*, Appl. Phys. Lett. **71**, 3132 (1997).
[11] M. A. Lutz *et al.*, Appl. Phys. Lett. **66**, 724 (1995).
[12] M. Albrecht *et al.*, Appl. Phys. Lett. **67**, 1232 (1995).
[13] S. Kishino, M. Ogirima, and K. Kurata, J. Electrochem. Soc. **119**, 617 (1972).
[14] J. W. P. Hsu *et al.*, Appl. Phys. Lett. **61**, 1293 (1992).
[15] S. Yu. Shiryaev, F. Jensen, and J. W. Petersen, Appl. Phys. Lett. **64**, 3305 (1994).
[16] T. Pinnington *et al.*, Phys. Rev. Lett. **79**, 1698 (1997).
[17] R. Hammond *et al.*, Appl. Phys. Lett. **71**, 2517 (1997).
[18] M. A. Renucci, J. B. Renucci, and M. Cardona, in *Light Scattering in Solids II*, edited by M. Balkanski (Flammarion, Paris, 1971), p. 326.
[19] W. Byra, Surf. Sci. **12**, 253 (1973).
[20] M. I. Alonso and K. Winer, Phys. Rev. B **39**, 10 056 (1989).
[21] J. C. Tsang *et al.*, J. Appl. Phys. **75**, 8090 (1994).
[22] J. Groenen *et al.*, Appl. Phys. Lett. **71**, 3856 (1997).
[23] To record the Raman images, the sample stage is moved in increments of 1 $\mu$m along both $x$ and $y$ directions in an area 50 $\times 50\ \mu m^2$, and thus each image consists of 2500 spectra.
[24] Q. M. Ma, K. L. Wang, and J. N. Schulman, Phys. Rev. B **47**, 1936 (1993).
[25] J. P. Hirth and J. Lothe, *Dislocation Theory*, 2nd ed. (Wiley, New York, 1982), p. 79.
[26] Y.-W. Mo, Ph.D. thesis, University of Wisconsin-Madison, 1991.