Si/Ge films on laterally structured surfaces: An x-ray study of conformal roughness

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X-ray diffraction measurements in the region of small incidence and exit angles on thin amorphous silicon/germanium films on laterally structured surfaces are performed. From fits of the data we obtain directly how the Fourier components of the substrates propagate through the evaporated films without being influenced by the intrinsic statistical roughness of the interfaces. The results show that a replication factor extracted from a given model can be quantitatively tested with our measurements. © *1996 American Institute of Physics.* [S0003-6951(96)02301-7]

A large body of work concerning correlated roughness in multilayers with statistically rough interfaces was done in the past (see, e.g., Refs. 1–7). But the separation of the intrinsic roughness from the conformal part is often not unique and always very difficult.

In this letter we study how the Fourier spectrum of evaporated thin Si/Ge films is influenced by an underlying laterally periodic substrate (i.e., by a surface grating with period d). These substrates can be regarded as a particular kind of "roughness" with only a few enhanced Fourier components. Due to the lateral periodicity the conformal part is now located at positions $q=m\cdot 2\pi/d$ (m integer) in reciprocal space and therefore clearly separated from the statistical microscopic roughness. X-ray scattering experiments from Si/Ge layer systems on Si gratings with a periodicity of $d=9800$ Å and a height of $h\sim 100$ Å in the region of small incidence and exit angles were performed to investigate how the Fourier coefficients of the stepped substrates propagate through the layer stacks (an example for polymer films is shown in Ref. 8).

To explain the x-ray data we consider a system of $N-1$ overlayers on top of a surface described by a periodic height function $f_1(x)$. The locations $z_k(\mathbf{r}_\parallel)$ of the interfaces are described by $z_k(\mathbf{r}_\parallel)=f_k(x)+l_k+\delta f_k(\mathbf{r}_\parallel)$ with a randomly fluctuating quantity $\delta f_k(\mathbf{r}_\parallel)$. The periodic part of the interface k is $f_k(x)=f_k(x+d)$ and l_k denotes the baseline of $f_k(x)$ (note: the baseline of the substrate is $l_1=0$).

Calculating the x-ray scattering intensity for such a system in Born approximation yields:[8]

$$I(\mathbf{q}_r,q_z)\sim\frac{1}{q_z^4}\sum_m 4\pi^2\delta(\mathbf{q}_{r,m})\times\sum_{j,k=1}^{N}\Delta\rho_k\Delta\rho_j e^{iq_z(l_k-l_j)}\mathscr{C}^*_{k,m}(q_z)\mathscr{C}_{j,m}(q_z). \quad (1)$$

The asterisk denotes a conjugate complex quantity and the electron density differences of the overlayers are given by $\Delta\rho_k=\rho_{k+1}-\rho_k$. The quantities $\mathscr{C}_{k,m}(q_z)$ are the Fourier coefficients of the functions $\exp\{-iq_zf_k(x)\}$.[8–11] They can be calculated analytically for a symmetric trapezoidal-like periodic structure with the width of the bars, grooves, and intermediate regions s_k, g_k, and b_k, respectively ($d=s_k+g_k+2b_k$), and the heights h_k,[10,11] under the assumption of a Gaussian distribution of the microscopic roughnesses $\delta f_k(\mathbf{r}_\parallel)$. Finally after averaging in the z-direction [see also Eq. (14) in Ref. 10] the result

$$\mathscr{C}_{k,m}(q_z)=e^{-iq_mg_k/2}\left[r_{g_k}(q_z)\frac{\sin(\frac{1}{2}q_mg_k)}{\frac{1}{2}q_md}+(-1)^m\cdot r_{s_k}(q_z)\frac{\sin(\frac{1}{2}q_ms_k)}{\frac{1}{2}q_md}e^{-iq_zh_k}\right]+o[(q_zh_k)^{-1}], \quad (2)$$

is obtained with the roughness factors $r_{s_k,g_k}(q_z)=\exp(-q_z^2\sigma^2_{s_k,g_k}/2)$ and rms roughnesses σ_{s_k,g_k} of the bars and grooves of layer k. Note that Eq. (2) is an expansion which is correct up to the first order in $(q_zh_k)^{-1}$. This is no restriction because in our work $h_k>100$ Å is always fulfilled and the Born approximation is only valid for $q_z>0.05$ Å^{-1} [an exact analytic expression for trapezoidal grating functions is given by Eq. (3.35) in Ref. 11].

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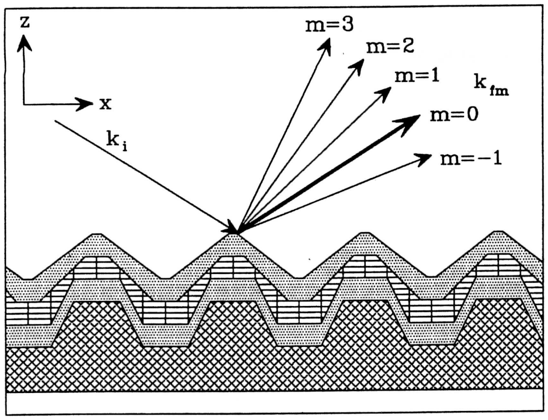

FIG. 1. Schematic drawing of a laterally structured surface with Si/Ge films on top. Due to the lateral periodicity an incoming x-ray wave $\mathbf{k}_i$ is reflected ($m=0$) as well as scattered into nonspecular diffraction orders ($m\neq 0$). Note that the evaporated films follow the substrate periodicity but with different Fourier coefficients (for clarity this effect is exaggerated in this sketch).

The momentum transfer $\mathbf{q}:=\mathbf{k}_f-\mathbf{k}_i$ ($\mathbf{k}_i$ and $\mathbf{k}_f$ are the wave vectors of the incident and scattered x rays, respectively) is decomposed by $\mathbf{q}=(\mathbf{q}_r,q_z)^T$. The delta function $\delta(\mathbf{q}_{r,m})$ in Eq. (1) leads to resolution limited diffraction orders at the positions $\mathbf{q}_{r,m}=(q_x-q_m,q_y)=0$ in reciprocal space which are caused by the periodic structure of the samples (see Fig. 1).[12,13]

The substrates are symmetric trapezoidally shaped surface gratings with a spacing of $d=9800$ Å and $s_1=g_1=4000$ Å, $b_1=900$ Å. They were prepared using lithographic methods.[14] Furthermore, we assume a very thin ($l_2=10$ Å), perfectly conformal $[\mathscr{C}_{2,m}(q_z)=\mathscr{C}_{1,m}(q_z)]$ native SiO_2 layer on the substrates.[15] The evaporated films $k=3,\ldots,N$ are also assumed to be trapezoidal (see Fig. 1) but with other parameters s_k, g_k, b_k, and h_k which means with other Fourier components but the same periodicity d.

We have investigated two samples. Ge and Si layers of a nominal thickness of 50 Å were evaporated on the substrates with a commercial setup (Balzers BAK 550). Due to the preparation conditions amorphous layers are expected with a density of 90% compared to that of bulk single crystals. The first sample consists of a Si grating ($h_1=105$ Å) with only one Ge layer on top. This sample was investigated to check if a homogeneous film builds up on top of the periodic surface. Furthermore we get the values for the thickness of the GeO_2 layer which forms after the evaporation and the values of the Si and Ge densities and rms roughnesses of the Si/Ge and Ge/air interfaces from this measurement and previous investigations.[15] To reduce the number of free fit parameters these results were taken for the fits of the data of the second sample. This sample was a Si grating with $h_1=130$ Å and three evaporated layers (Ge/Si/Ge).

The x-ray measurements were performed at the National Synchrotron Light Source (Brookhaven National Laboratories) on the Exxon beamline X10B using a wavelength of $\lambda=1.131$ Å. The measured q_z resolution for our setup was $\delta_{q_z}=3.5\times 10^{-3}$ Å^{-1}. The resolution in the direction y out of the scattering plane (x,z) was rather coarse and therefore need not be considered (integration over q_y).

We have performed q_z scans in reciprocal space along rods with $q_x=q_m$ which means along the diffraction orders and additionally between the diffraction orders along rods with $q_x=q_m+\pi/d$ to monitor the diffusely scattered intensity stemming from the random fluctuations $\delta f_k(\mathbf{r}_\parallel)$ of the interfaces.[16] This diffuse intensity was subtracted from the data taken along q_z for $q_x=q_m$ to obtain the true specular reflectivity and true intensity of the diffraction orders, respectively. Therefore the periodic conformal part is easily separated from the statistical roughness and can be explained with the aforementioned model. However there are indeed conformal roughness contributions caused by the random roughness fluctuations of the evaporated layers (see Refs. 1, 2, and 8). They are also subtracted from the data by subtracting the intensity obtained along $q_x=q_m+\pi/d$. But we are only interested in the strictly periodic part of the rouhgness, i.e. in that part with wave numbers $q=m\cdot 2\pi/d$, which is perfectly correlated throughout the layer stack.

The analysis of the measurements of the first sample shows that the Ge layer is nearly conformal and homogeneous on the stepped surface with a rather thick oxide layer on top (thicknesses of the Ge and GeO_2 layers: $l_{Ge}=36$ Å and $l_{GeO_2}=26$ Å). Figure 2 shows four q_z scans along the positions $q_x=q_m$ for $m=0,1,2,3$ ($m=0$ reflectivity) for the second sample together with the fit using Eqs. (1) and (2). Due to the oversimplified model of trapezoidal layer structures deviations between the fit and the data are clearly visible. It should also be noted that all curves are fitted simultaneously without introducing any normalization constants and that the inclusion of the roughness of the grooves and bars with the roughness factors in Eq. (2) might be too simple. Nguyen *et al.*[17] have shown that an overgrown sharp pattern can be significantly smoothed out by the evaporation of films. But their films are made out of other materials and they are rather thick so that a direct comparison is difficult. Nevertheless the fit is good enough to yield the parameters $s_k=g_k$, b_k, and h_k of each layer so that the Fourier coefficients of the periodic interfaces as a function of the layer thickness l_k can be calculated. The substrate parameters change from the bottom to the top GeO_2 layer to $h_{GeO_2}=127$ Å, $s_{GeO_2}=g_{GeO_2}=3800$ Å, and $b_{GeO_2}=1100$ Å. The obtained layer thicknesses of the Ge/Si/Ge/GeO_2 system are $l_{Ge}=68$ Å, $l_{Si}=65$ Å, $l_{Ge}=36$ Å, and $l_{GeO_2}=26$ Å. The inset in Fig. 2 shows a transverse scan at fixed $q_z=0.2$ Å^{-1}. The diffraction orders which contain the information about the periodic part of the surface can be seen as resolution limited peaks.

To describe the propagation of the Fourier components of the films more quantitatively we define the replication factor $\chi(q,l)$ according to Spiller *et al.*:[18]

$$\tilde{f}_k(q)=\chi(q,l_k)\cdot\tilde{f}_{k-1}(q). \tag{3}$$

The function $\chi(q,l)$ connects the Fourier transform $\tilde{f}_k(q)$ of layer k with baseline l_k with the Fourier transform $\tilde{f}_{k-1}(q)$

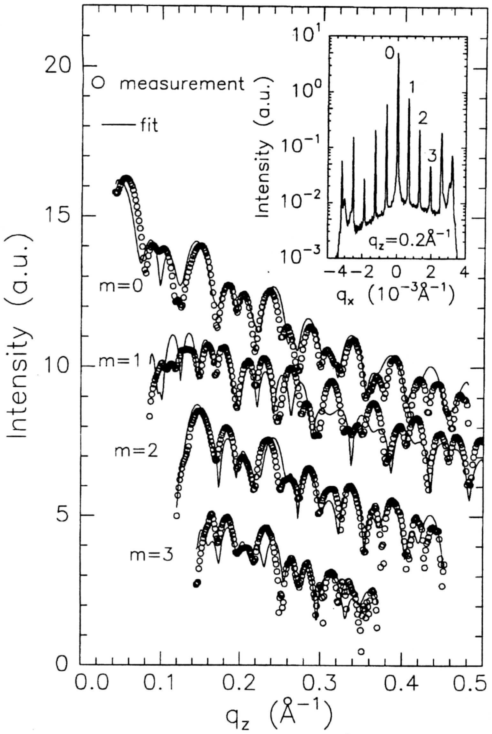

FIG. 2. True specular $m=0$ and true intensity along the diffraction orders $m=1,2,3$ (symbols) and corresponding fits (lines) for a Si grating with evaporated Ge/Si/Ge films. For clarity, all curves are shifted three orders of magnitude against each other. The inset displays a transverse scan obtained at the position $q_z=0.2$ Å $^{-1}$.

of the underlying layer $k-1$. Note that for a laterally periodic surface the continuous variable q has to be replaced by discrete values $q_m=m\cdot 2\pi/d$ and the Fourier transforms have to be replaced by the respective Fourier coefficients. The intrinsic statistical part of the roughness is neglected in this description because it was subtracted from our data.

We are now able to obtain the replication factor at the positions l_k and for the discrete values q_m, i.e., the fits yield directly $\chi(q_m,l_k)$. Calculating the Fourier coefficients with the results of the fit shown in Fig. 2 yield the symbols in Fig. 3. They show the replication factor for $q=1\cdot 2\pi/d,3\cdot 2\pi/d,5\cdot 2\pi/d$. Due to $s_k=g_k$ only odd Fourier coefficients occur. Higher order Fourier coefficients can not be extracted from our data with a sufficient accuracy. The lines in Fig. 3 are fits to the data with the replication factor proposed by Spiller *et al.*:[18] $\chi(q,l_k)=\exp(-q^2\nu_k l_k)$ with the relaxation parameter ν_k of layer k. We have assumed the same $\nu_k=\nu$ for all layers and obtain $\nu=500$ Å from Fig. 3. With this number we can extract the number $N(q)=1/\{2\ \ln(1+q^2\nu l)\}$ which is a measure for the degree of correlation in multilayers (for details see Spiller *et al.*[18]). From our data we get the values $N(q_1)=50$, $N(q_3)=6$, and $N(q_5)=2$. Therefore the investigations show that the (periodic) roughness is nearly completely correlated for the wave number $q_1=2\pi/d$ and partially correlated ($N<1$ would mean uncorrelated) for the higher frequency components $q_3=3\cdot 2\pi/d$ and $q_5=5\cdot 2\pi/d$, respectively.

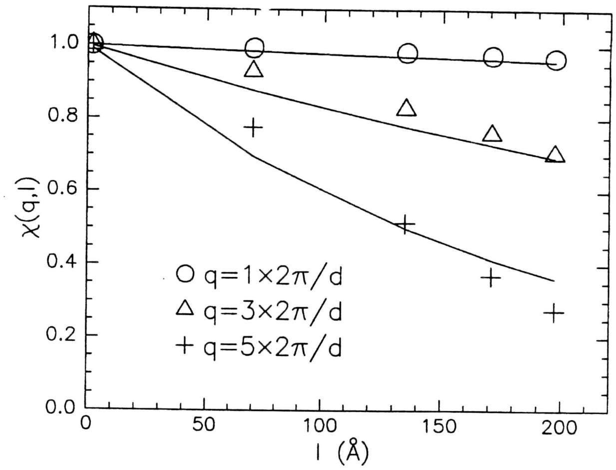

FIG. 3. Replication factor $\chi(q,l)$ for $q=q_m$ with $m=1,3,5$ obtained from the x-ray data (symbols) and a fit (lines) corresponding to a simple diffusion model with $\chi(q,l)=\exp(-q^2\nu l_k)$. Note that the upper Ge layer is split into a pure Ge and a GeO_2 layer.

In summary, we have shown a new way to investigate the phenomenon of correlated roughness. The use of a surface grating as substrate makes it possible to extract the conformal part of the roughness out of the x-ray data in an unique and unambiguous manner. Therefore the replication factor can be directly obtained from the measurements and a quantitative comparison with existing models can be made.

[1] J. B. Kortright, J. Appl. Phys. **70**, 3620 (1991).
[2] D. G. Stearns, J. Appl. Phys. **71**, 4286 (1992).
[3] E. E. Fullerton, J. Pearson, C. H. Sowers, S. D. Bader, X. Z Wu, and S. K. Sinha, Phys. Rev. B **48**, 17432 (1993).
[4] S. K. Sinha, M. K. Sanyal, S. K. Satija, C. F. Majkrzak, D. A. Neumann, H. Homma, S. Szpala, A. Gibaud, and H. Morkoc, Physica B **198**, 72 (1994).
[5] T. Salditt, T. H. Metzger, and J. Peisl, Phys. Rev. Lett. **73**, 2228 (1994).
[6] D. E. Savage, Y.-H. Phang, J. J. Rownd, J. F. MacKay, and M. G. Lagally, J. Appl. Phys. **74**, 6158 (1993).
[7] A. P. Payne and B. M. Clemens, Phys. Rev. B **47**, 2289 (1993).
[8] M. Tolan, G. Vacca, S. K. Sinha, Z. Li, M. Rafailovich, J. Sokolov, H. Lorenz, and J. P. Kotthaus, J. Phys. D: Appl. Phys. **28**, A231 (1995).
[9] M. Tolan, G. König, L. Brügemann, W. Press, F. Brinkop, and J. P. Kotthaus, Europhys. Lett. **20**, 223 (1992).
[10] M. Tolan, W. Press, F. Brinkop, and J. P. Kotthaus, J. Appl. Phys. **75**, 7761 (1994).
[11] M. Tolan, Ph.D. Thesis, Kiel University, 1993.
[12] M. Gailhanou, G. T. Baumbach, U. Marti, P. C. Silva, F. K. Reinhart, and M. Ilegems, Appl. Phys. Lett. **62**, 1623 (1993).
[13] Q. Shen, C. C. Umbach, B. Weselak, and J. M. Blakely, Phys. Rev. B **48**, 17967 (1993).
[14] W. Hansen, J. P. Kotthaus, and U. Merkt, *Nanostructured Systems*, Semiconductors and Semimetals Vol. 35 (Academic, New York, 1992).
[15] J.-P. Schlomka, Diploma Thesis, Kiel University, 1994.
[16] S. K. Sinha, E. B. Sirota, S. Garoff, and H. B. Stanley, Phys. Rev. B **38**, 2297 (1988).
[17] K. B. Nguyen and T. D. Nguyen, J. Vac. Sci. Technol. B **11**, 2964 (1993).
[18] E. Spiller, D. Stearns, and M. Krumrey, J. Appl. Phys. **74**, 107 (1993).