Thermal roughening of Cu(115): An energy-resolved helium-atom-beam scattering study

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The structure of the Cu(115) surface has been investigated in the temperature range between 160 and 700 K with energy-resolved helium-atom-beam scattering. The "power-law" diffraction profiles found in previous helium-scattering experiments without energy analysis are shown to be the result of inelastic (phonon) scattering. In our energy-resolved measurements, the line shapes of the elastically scattered intensity can be described by a two-dimensional Lorentzian. This asserts that on this surface a possible crossover of the height difference correlation function to the expected logarithmic behavior might only occur for distances larger than about 100 Å (perpendicular to the average step direction).

INTRODUCTION

Thermal roughening of single-crystal surfaces has been widely studied both theoretically and experimentally. It is related to a range of phenomena: For example, by altering the density of defect sites (e.g., step edges), thermal roughness can affect chemical reactivity, catalytic properties or growth processes.

The concept of a "thermodynamic" roughening phase transition on surfaces has been introduced a long time ago.[1] Below the sharply defined roughening transition temperature T_R, thermally induced defects exist, but the defect density is low enough to have a well-defined surface plane. At the roughening transition temperature, the defect density achieves a critical level, and the interfacial width goes to infinity. Specifically, for a two-dimensional (2D) system, the height difference correlation function is predicted to diverge logarithmically for a thermodynamical large sample.[1-3]

On an atomic level, roughening is signaled by the presence of defects, the proliferation and/or meandering of steps. While this "atomic" roughening is a necessary condition for "thermodynamic" roughening, it is not sufficient. In fact, at any temperature defects are always present, but their configuration must not necessarily lead to a logarithmically diverging height correlation function. In order for a low index surface to roughen, atomic steps must be thermally induced. Since the energetic cost of creating a step is high, roughening is expected only at temperatures near melting. For a vicinal surface steps already exist, and the roughening transition only requires them to meander, by formation and displacement of atomic kinks. Since the energetic cost of creating kinks in a preexisting step is less than the cost of creating a step, the roughening temperature of vicinal surfaces is expected at considerable lower temperatures as compared to low index surfaces of the same material.

This reasoning has been confirmed experimentally. The thermal roughening of many crystal surfaces has been analyzed with x-ray, electron, or He scattering (see Ref. 3 for an overview). In some cases, a logarithmically

diverging height correlation function has indeed been detected. The roughening of Cu(11n) vicinal surfaces has been extensively studied using helium-atom-beam scattering,[3-5] and among others the (113) and (115) surfaces have been shown to undergo a roughening transition of the Kosterlitz-Thouless-type, evidenced by a power-law line shape of diffraction peaks, due to a logarithmically diverging height difference correlation function.[4,5] This finding for the Cu(113) surface stands in contrast to the results of a recent high-resolution electron-scattering study of the same surface. Wollschläger, Luo, and Henzler[6] find that the line shapes can be much better described by a 2D Lorentzian form. In earlier helium-atom-beam scattering experiments of Cu vicinal surfaces, no energy resolution was available, so it was necessary to assume that scattering was predominantly elastic. However, as pointed out by Lapujoulade and others,[7,8] the He diffraction line shapes could suffer from distortions, due to inelastic-scattering processes (single- and multiphonon events), especially at high surface temperatures, and obscure the information on the surface structure contained in the elastically scattered intensity. In fact, energy resolved helium-atom-beam scattering measurements by Hofman, Toennies, and Manson[9] on a singular surface of the same material, Cu(001), reveal a significant, under certain conditions overwhelming contribution of single- and multiphonon events in the diffracted intensities, with respect to elastic scattering. We show here experimentally that it is imperative to discern inelastic scattering for a correct determination of the structure of the Cu(115) surface.

The paper is organized as follows. The experimental setup and the characteristics of the sample are presented. The line shape analysis of both the "total intensity" (no energy analysis) and "elastic only" measurements under antiphase conditions are then discussed. Using "total intensity" measurement, the previous evidence for a Kosterlitz-Thouless roughening transition near 400 K is reproduced. However, time-of-flight (TOF) spectra demonstrate that much of the diffuse scattering is inelastic. The elastic-scattering line shapes can be described by

2D Lorentzians, which asserts that logarithmic behavior of the height difference correlation function does not occur on our sample for distances smaller than about 100 Å.

EXPERIMENTAL SETUP

This experiment was performed with the Ramses II helium-atom-beam spectrometer. In comparison with previous experiments at this laboratory,[4] the major refinement is the availability of TOF analysis (single chopping model, Ref. 9 and references therein) to measure the energy distribution of the scattered beam. The spectrometer includes extensive differential pumping and the detector includes mass filtering. The resulting reduction in background level allows us to easily measure scattering intensities six orders of magnitude lower than the direct beam intensities. Finally, the angular resolution of the spectrometer is higher than in previous studies. Our instrument allows for rotation of both the beam source and the crystal. Generally, rotating the beam source provides better momentum-transfer resolution. Nevertheless, we have chosen to access different parallel momentum transfers in this experiment by rotating the crystal, because the power-law exponent (see below) is generally a function of perpendicular momentum transfer. Changing the parallel momentum transfer, while scanning through a particular diffraction peak, introduces slight changes also in perpendicular momentum transfer: By rotating the crystal and not the beam source, this effect is minimized.

The sample preparation was similar as in previous experiments.[4] The crystal was cut and polished to the (115) face, as determined by x-ray Laue diffraction. The sample was desulfurized at 1170 K for a month under a purified H_2 flow. The surface was then cleaned *in situ* by repeated cycles of argon-ion bombardment and annealing to 750 K. This produced a clean, well-ordered Cu(115) surface, as determined by helium diffraction.

At a surface temperature of 160 K, the in phase and antiphase diffraction peaks (elastically scattered part only) show the same full width at half maximum (FWHM) (0.045 Å^{-1} parallel to the steps and 0.074 Å^{-1} perpendicular to the steps), the only difference being a slightly different peak shape. These halfwidths are found to be larger than the resolution of the spectrometer itself and are, therefore, imposed by the average terrace size of the sample. The residual-defect free region is estimated to about 100 Å perpendicular and 150 Å parallel to the steps. The antiphase peak shape in each azimuthal direction could well be described by a Lorentzian, which has been used as the instrument/sample response for the detailed line-shape analysis at high temperatures. As in previous experiments using helium-atom-beam scattering from the Cu(115) surface,[4] the diffraction peaks were found to broaden slightly asymmetrically with temperature. The steeper slopes could be shifted to the right or the left side of the peak just by probing different areas of the surface. This asymmetry is, therefore, likely due to mosaicity. For the detailed analysis of the diffraction line shapes, we have used the steeper sides of the diffraction peaks.
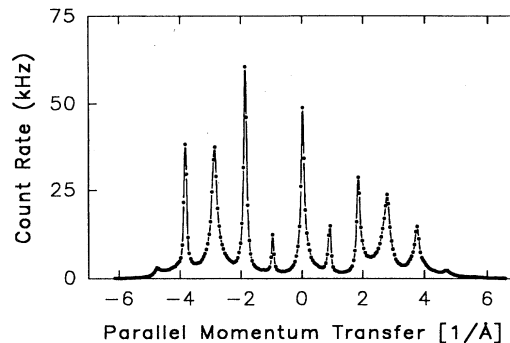

FIG. 1. Helium-atom-beam diffraction pattern from Cu(115). The scattering plane is perpendicular to the steps, and the incident energy is $E_i = 64$ meV.

DIFFRACTION PATTERN AT ROOM TEMPERATURE

The Cu(11n) vicinal faces are composed of (001) terraces separated by steps, the edges of which are parallel to the close-packed $\langle \bar{1}10 \rangle$ direction. The nearest-neighbor distance is $a \cong 2.55$ Å, and the terraces of the (115) surface are $2\frac{1}{2}$ atoms wide. The periodicity in the macroscopic surface plane perpendicular to the step edges is 6.625 Å. Diffraction is allowed along crystal truncation rods parallel to the $\langle 115 \rangle$ direction.

The corrugation of the surface perpendicular to the average step direction is large, producing intense helium-atom-beam diffraction peaks through fifth order, see Fig. 1. By contrast, only the specular reflection is visible in the azimuthal direction parallel to the steps. No spurious diffraction features have been observed in either azimuth.

Both the width and intensity of the diffraction peaks are sensitive to interference effects between regions of the surface, which are separated by thermally excited defects. The phase difference for the helium scattered by surface regions separated by these defects is $\phi = Qa$, where $Q = k_f - k_i$ is the momentum transfer and a is the
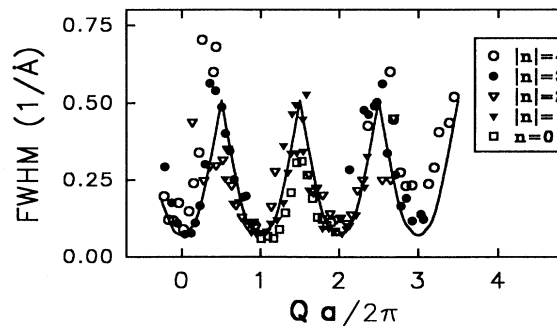

FIG. 2. Full width at half maximum (FWHM) of the diffraction peaks versus phase $Qa/2\pi$ inferred from angular distributions taken at $T = 680$ K. n denotes the diffraction peak of nth order. The observed periodicity asserts that, in the temperature range covered by our experiment, the dominant thermally excited defect is the creation of kinks in preexisting steps.

translation characteristic of a surface defect. The dominant defect is expected to be a kink in a step edge, resulting in a terrace, which is either an atom too wide or too narrow, thus $a = a\mathbf{n}_{110}$ for a vicinal $(11n)$ surface of a fcc metal with nearest-neighbor distance a. For the specular reflection, for which $\mathbf{Q}$ is perpendicular to the surface, the phase reduces to $\phi = Q_z c$, where $c = (\frac{2}{27})^{1/2} a \cong 0.694$ Å is the height difference between (115) surfaces separated by "secondary steps." Figure 2 shows the peak widths of the diffraction peaks as a function of phase inferred from (not energy resolved) measurements at about 680 K. The observed periodicity confirms that at this temperature, the predominant defect is still the excitation of single kinks in preexisting steps.

DIFFRACTION PEAK LINE SHAPES

A convenient way to describe the morphology of a multilevel interface in real space is in terms of the height difference correlation function, which we define here as $H(\mathbf{r}) \equiv \langle [h(\mathbf{r}) - h(0)]^2 \rangle$, where $h(\mathbf{r})$ is the height of the
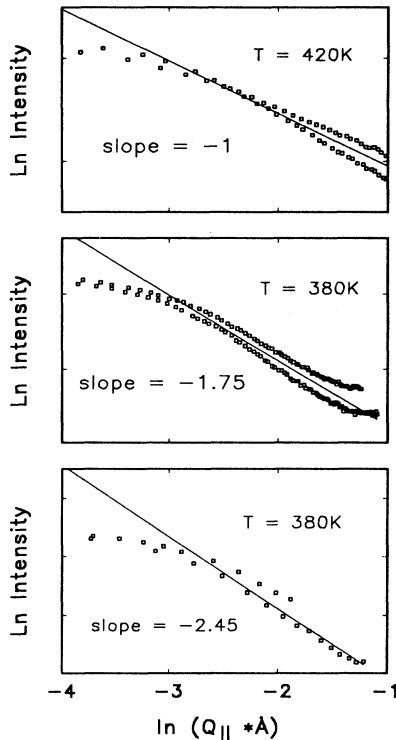

FIG. 3. Logarithm of the intensity of the specular peaks vs logarithm of parallel momentum transfer. The scattering plane is perpendicular to the steps. The peaks are seen to be slightly asymmetric. Lines are fits to straight sections in the wings of the peak profile chosen by eye inspection. Upper panel: The incident energy is $E_i = 64$ meV. Middle panel: The incident energy is $E_i = 23$ meV. Lower panel: Elastic only angular distribution obtained by time-of-flight analysis in order to reject inelastic scattering. The effect upon the slope and thus upon the apparent roughening exponent $\tau = (2\text{-slope})$ is considerable.
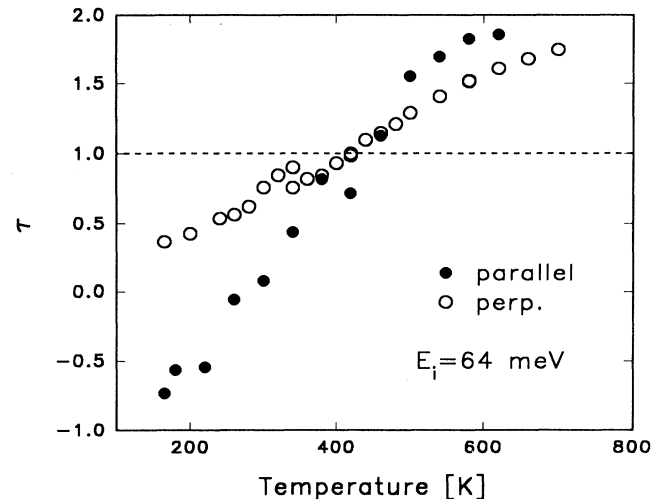
surface relative to a reference plane. We shall assume that the relative height depends only upon the distance r. Then, the structure factor $S(\mathbf{Q})$ may be written[2,10]

$$S(\mathbf{Q}) = 2\pi \int_0^\infty \langle \exp\{iQ_z c[h(r) - h(0)]\} \rangle J_0(Q_\parallel r) r \, dr ,$$

$$(1)$$

which is the two-dimensional Fourier transform of the pair-correlation function. J_0 is the Bessel function of zeroth order. The pair-correlation function may be related to the height correlation function within the Gaussian approximation[2,3] by

$$\langle \exp\{iQ_z c[h(r) - h(0)]\} \rangle \approx \exp[-(Q_z c)^2 H(r)/2] , \quad (2)$$

where Q_z is the perpendicular momentum transfer, and c the lattice unit perpendicular to the substrate as defined above.

Quite generally, the height difference correlation function $H(r)$ has the value 0 for $r = 0$ (by definition), while it increases for increasing r. In most cases, H is bound for large values of r, crossing over to a saturation value w ("the width of the interface") at the lateral distance $r = \xi$, which is called the correlation length.[10]

A remarkable exception to a bound height difference correlation function constitutes the correlation function of a Kosterlitz-Thouless type rough surface above its roughening temperature T_R. Here, the lateral correlation length ξ is infinite, and so is the width of the interface w.[2,3,10] The height correlation function is predicted to exhibit a logarithmic divergence,

$$H(r) = A(T)\ln(r) + \text{const} \qquad (3)$$

for large lateral distances r. $A(T)$ is a temperature-

FIG. 4. Roughening exponent $\tau[\tau = (2\text{-slope})]$ vs temperature obtained from the simplified analysis as shown in Fig. 3, for both azimuthal directions. The incident He beam energy is $E_i = 64$ meV. Based on this analysis, the criterion for the thermodynamically rough state ($\tau \geq 1$) would be fulfilled for $T \geq 420$ K, thus the roughening temperature T_R ($\tau = 1$, dashed line) would be at 420 K.

dependent prefactor. It has been shown[2,7] that in order to account for the anisotropy of a vicinal surface, r can be expressed as $r = (x/\eta_n)^2 + (y/\eta_y)^2$, where η_x/η_y describes the anisotropy. This very specific form of the height correlation function gives rise (for not too large values of A) to power-law divergencies in the scattering.[2] In the following, we will demonstrate the impact of energy-resolved measurements on the He diffraction line shapes and discuss the elastic only line shapes, with respect to the expected power-law behavior for a Kosteritz-Thouless roughening of this vicinal surface.

DIFFRACTION LINE SHAPES WITHOUT ENERGY ANALYSIS

In order to make contact with previously published results,[4] at first the same experimental conditions as in Ref. 4 have been applied. In particular, we used a high-energy incident He beam (64 meV), and the diffraction pattern was recorded without time-of-flight analysis. According to Villain, Grempel, and Lapujoulade,[2] the logarithmic divergence of the height correlation function leads to line shapes, which can be approximated for not too large coefficients $A(T)$ by a power law,

$$S(\mathbf{Q}) \sim [(Q_x \eta_x)^2 + (Q_y \eta_y)^2]^{\tau/2 - 1} . \tag{4}$$

This expression is valid, when the roughening exponent τ is smaller than 2. τ is related to $A(T)$ as $\tau = (\pi^2/2)A(T)$ for antiphase conditions. At the roughening temperature T_R, one finds $\tau = 1$. The functional form of the line shape suggests that a rapid and convenient way to extract the exponent τ is to plot the logarithm of the intensity vs the logarithm of the parallel momentum transfer.[7] Then, the exponent τ is simply $\tau = 2 - \Delta \ln I / \Delta \ln Q_\parallel$. Figure 3 shows an example. This simple analysis is only valid within a certain momentum-transfer window, since close to $Q_\parallel = 0$ the diffraction is strongly influenced by instrumental effects, while at large momentum transfers, care must be taken not to run into neighboring diffraction peaks. Figure 4 shows the result of this type of analysis from a series of diffraction line shapes taken at antiphase condition with a high-energy beam. It is seen that the behavior of τ as a function of temperature is nearly identical as compared to that found in earlier experiments;[4] the apparent roughening temperature T_R is slightly higher, at about 420 K instead of 380 K.

We have performed this type of measurement and this analysis in addition for a number of different experimental conditions, in particular, with a low-energy He beam (23 meV) under glancing incident angle corresponding to the minimum momentum transfer $Q_z \cong 4.52 \text{ Å}^{-1} \approx \pi/c$ needed to observe the specular reflection under antiphase condition. Reducing the incident energy and increasing the incident angles will both decrease the excitation probability for inelastic scattering, due to the reduction in perpendicular momentum transfer.[9,11] The middle graph of Fig. 3 shows that the effect on the line shape is quite pronounced by significantly altering the slope of the wings, and therefore, the roughening exponent τ. The behavior of τ vs T is now quite different in comparison to
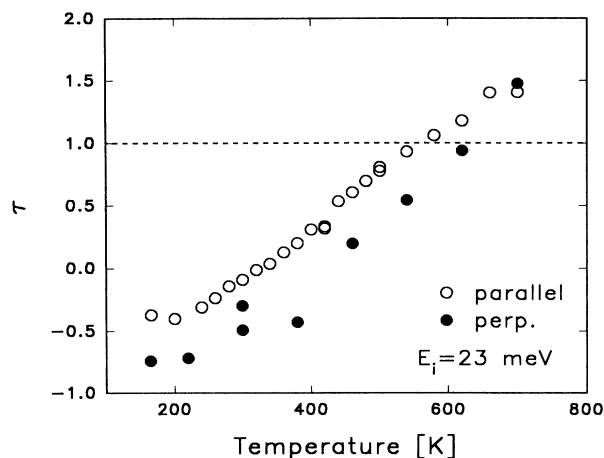

FIG. 5. Same as Fig. 4, but the incident energy is $E_i = 23$ meV. The apparent roughening temperature is now at $T = 580$ K.

the result, which has been inferred from angular distribution recorded with a high-energy incident beam, and the apparent roughening temperature is now located at around 580 K, as seen in Fig. 5. These findings point clearly to a significant contribution of inelastic scattering and the necessity of a TOF analysis in order to separate the elastic part of the scattered intensity for this system.

DIFFRACTION LINE SHAPES WITH ENERGY ANALYSIS

The results reported in the previous section indicate that the use of a low-energy beam in order to minimize distortions due to inelastic scattering is an advantage, if one is interested in a line-shape analysis of diffraction
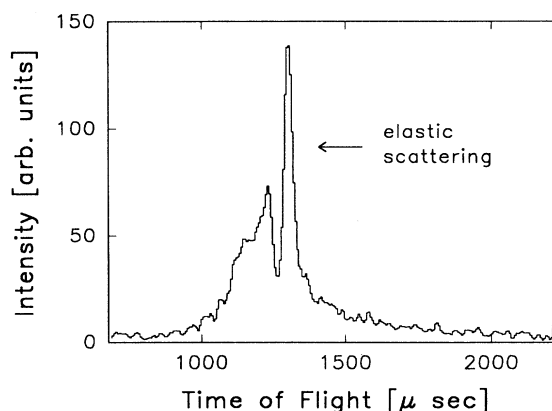

FIG. 6. TOF analysis of the scattered intensity in the wings of the specular peak profile. The incident energy is $E_i = 23$ meV, the surface temperature is $T = 700$ K, and the scattering plane is perpendicular to the steps. The measuring time is 3 min. Although the amplitude of the elastic peak dominates, its integrated intensity amounts to only a minor part of the total; thus most of the scattering is inelastic.

peaks. Moreover, in addition to reducing inelastic effects, fewer diffraction channels are available and the specular intensity is substantially larger. Finally, by reducing the magnitude of Q, the resolution in reciprocal space is improved. However, for an accurate line-shape analysis, these conditions are not sufficient. Figure 6 shows a typical intensity distribution scattered into the wing of the specular diffraction peak, energy resolved by TOF analysis and recorded at 700 K, with an incident energy of 23 meV: Strong multiphonon excitation is already noticeable at this temperature, and although the amplitude of elastic scattering (FWHM ≈ 0.5 meV) still dominates, the integrated intensity of the elastic channel amounts to only a minor part of the total.[12]

The consequences for the line shapes by considering the elastic only scattering are demonstrated in Fig. 7 for the specular reflection perpendicular to the steps under antiphase condition. It is seen that the main effect of rejecting inelastic scattering at this temperature is an overall decrease in intensity, due to a nearly constant inelastic intensity over the parallel momentum range probed. In view of this experimental finding, one might be tempted to subtract simply an inelastic background[4] estimated from ordinary angular distributions (without energy analysis). However, we find it quite impossible to infer the *correct* amount of elastic with respect to inelastic scattering from an inspection of the angular distribution. While this uncertainty would have only little influence on Gaussian or Lorentzian line shapes, dramatic changes in *presumed* power-law line shapes are expected, and seen. This is obvious from Fig. 3 (lower panel), which shows the elastic only data on logarithmic scales. Once again, the slope of the wings dramatically changes even in comparison to the data taken with a low-energy beam, but without TOF analysis. The impact on the
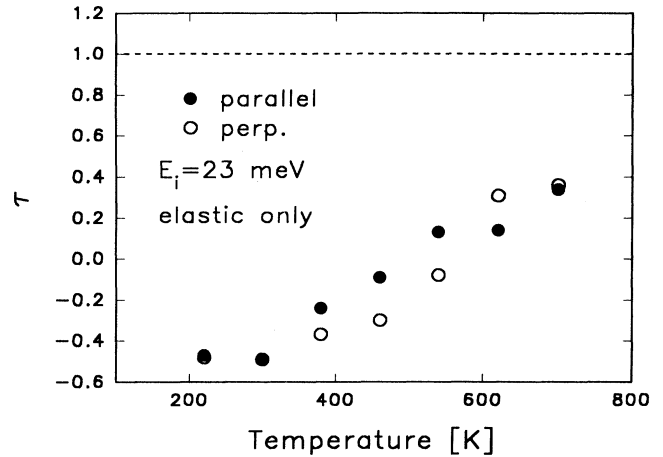
FIG. 8. Apparent roughening exponent τ vs temperature obtained by analyzing the elastic only scattering as in Fig. 3, lower panel. The roughening temperature T_R ($\tau = 1$) for this surface would be far above 700 K, when the roughening exponent is determined this way.

roughening exponent is pronounced. As seen in Fig. 8, the behavior of the apparent roughening exponent τ vs T is now substantially different from what has been inferred with the same analysis of the diffraction peaks without energy-resolved measurements. In particular, in the whole temperature range covered by our experiment τ does not increase much with temperature, and even at 700 K it is still far away from reaching $\tau > 1$, the criterium (under antiphase conditions) for a Kosterlitz-Thouless type rough state with logarithmically diverging height difference correlation function.

This discrepancy to previous conclusions, reached from measurements without TOF analysis,[4] cannot be attributed to this simplified procedure to extract the exponent τ. In the following we show the results of a reanalysis of the elastic only data, first in terms of the structure factor derived by Dutta and Sinha,[13] and second in terms of a direct numerical integration of the pair-correlation function.

The Dutta-Sinha structure factor describes the scattering for a surface, which has a logarithmically diverging height difference correlation and includes the effects of a long-range cutoff, which is approximated by a Gaussian factor, $\exp(-Q_\parallel^2 L^2/4\pi)$. This cutoff is determined by instrumental resolution and the finite-size effects of the crystal due to a finite average domain size introduced by cleaning or polishing procedures. For this purpose, we have replaced the empirically found Lorentzian peak shapes at low temperatures by the sum of two Gaussians. The structure factor contains two adjustable parameters, the exponent τ and the cutoff length, L.
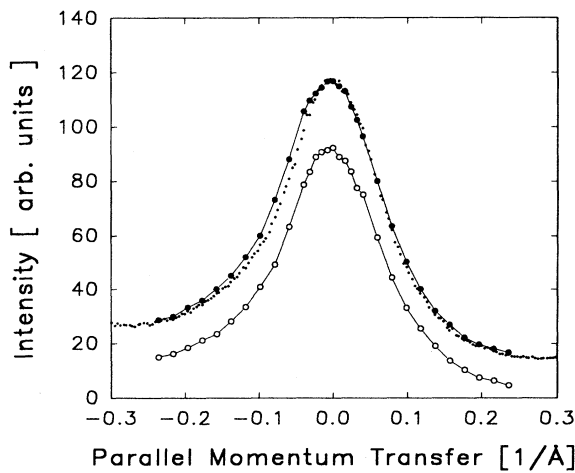

FIG. 7. Peak profiles of the specular scattering without TOF analysis (small circles), and with TOF analysis: Filled large circles denote data obtained from the integration of the TOF spectra (which reproduces of course the line shape obtained in ordinary angular scans), open large circles from the integration of the elastic component only.

$$S(Q_x) \cong \left[\frac{L}{a}\right]^{2-\tau} \Gamma(1-\tau/2) \left[\frac{2}{\gamma\sqrt{\pi}}\right]^{\tau}$$

$$\times \Phi\left[1-\tau/2, 1, -\frac{Q_x^2 L^2}{4\pi}\right], \qquad (5)$$

where

$$\Phi(a_0,1,z)= \sum_{n=0}^{\infty} \frac{\Gamma(a_0+n)}{\Gamma(a_0)} \frac{z^n}{(n!)^2} \qquad (6)$$

is the "Kummer" function; Γ denotes the gamma function; $a_0=3.61$ Å is the bulk lattice constant for copper; $\gamma=1.781\ldots$ is a constant. This structure factor may be readily calculated, since the sum for the Kummer function converges rapidly.

Within this description, the value of L should remain fixed, since both the domain size and the instrumental resolution do not change, and the evolution in peak shape as the temperature increases above the roughening temperature should be governed only by an increase in τ. However, we found that the elastic only data at high temperatures could not be fitted by locking L to the instrument/sample response obtained from fits to the line shapes at 160 K. Acceptable fits were obtained only by treating both L and τ as adjustable parameters at each temperature, see Fig. 9. However, there is no justification on physical grounds for letting L float. A detailed analysis reveals that in this procedure L is effectively a parametrization of the "central part" to "wing" ratio, with large L corresponding to a high central part. We find that the combination of τ's and L's obtained from those fits keeps in fact the "central part" to "wing" ratio nearly constant with temperature; this indicates that the diffraction peak simply broadens without changing its form.
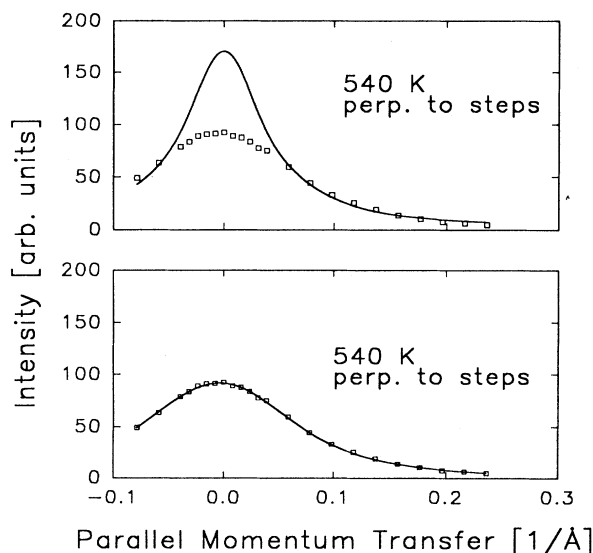
An alternative method to determine the line shapes is

FIG. 9. Fit to the elastic only specular diffraction profile with the Dutta-Sinha structure factor [Eq. (5)]. In this analysis the Lorentzian instrument/sample response has been replaced by the sum of two Gaussians. If the wings of the profile are forced to fit, the center part is severely overestimated (upper panel). The profile can be made to fit excellently, when the instrument/sample response length L is arbitrarily decreased (lower panel).

to numerically integrate the structure factor using the pair correlation function in Eq. (1). This allows one to introduce nearly any desired height correlation function, but care must be taken in order to assure numerical stability, due to the rapidly oscillating Bessel function. By comparing the numerical integration with the Dutta-Sinha structure factor, we could demonstrate the numerical stability of the integration routine. Of course, this procedure is more time consuming, but it has the advantage that the roughening exponent τ is not limited to values smaller than 2, as in Eq. (4). It is worth noting that roughening exponents larger than 2 lift the singularity in the power-law behavior [Eq. (4)] and produce a "dome-shaped" structure factor.

For comparison with the experimental data, we included a convolution with our sample/instrument response. Using the logarithmic height correlation function [Eq. (3)], we encountered exactly the same situation as with the fits to the Dutta-Sinha structure factor: Acceptable fits could only be obtained by increasing artificially the half width of the instrument/sample response. Locking this half width to the low-temperature value for the high-temperature data led to very poor fits.

On the basis of the foregoing analysis, we are led to conclude that the previously observed power-law line shapes for the Cu(115) surface[4] have been the result of inelastic scattering. However, in our opinion, it is not legitimate to conclude from this observation alone that the surface does not undergo a Kosterlitz-Thouless type roughening transition with a logarithmically diverging height difference correlation function in the temperature range covered by our experiment. Indeed, we would like to stress that the divergent logarithmic height difference correlation function is meant to describe only the asymptotic (large distance) behavior of the rough surface. We assumed so far, as it has been done frequently before[3,7] and in the previous analysis of helium-atom-beam diffraction line shapes of Cu(115) as well,[4] that the logarithmic law is valid also for very small distances. This may, however, not be the case. The consequences for the diffraction line shapes by dropping this assumption are discussed in the following sections.

SENSITIVITY OF THE EXPERIMENTAL DATA TO A LOGARITHMICALLY DIVERGING HEIGHT DIFFERENCE CORRELATION FUNCTION

For the diffraction line-shape analysis, the Dutta-Sinha formulation of the structure factor or the convolution with the instrument/sample response are quite reasonable attempts to incorporate the upper cutoff of logarithmic behavior, either due to a finite transfer width of the instrument or due to a limited average terrace width of the crystal under investigation. However, it is clear that there must also be a lower cutoff of logarithmic behavior of the correlation function. By definition, $H(r=0)=\langle[h(r=0)-h(0)]^2\rangle=0$, however, for Eq. (3), $H(r=0)=-\infty$. A conceivable way to correct the short-range behavior is to replace $H(r)=A(T)\ln(r)$ by $H(r)=\frac{1}{2}A(T)\ln[(r/\lambda)^2+1)]$, as suggested by Mochrie
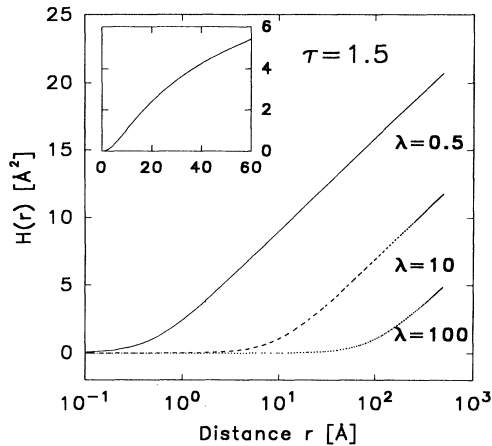
FIG. 10. Calculated correlation function $H(r) = 1/2A(T)\ln[(r/\lambda)^2 + 1]$ for different values of the crossover length λ (Å). Logarithmic behavior is obtained for distances larger than about 5λ. The inset shows that this ansatz leads to an almost linear behavior in the crossover region.

FIG. 12. Fits to the elastic only peak profiles for $T = 540$ K (upper part perpendicular to the steps, lower part parallel) based on $H(r) = \tau/\pi^2\ln[(r/\lambda)^2 + 1]$ and convoluted with our instrument/sample response.

et al.,[14] where λ characterizes the short-range limit to the logarithmic behavior. This functional form of the height difference correlation function recovers essentially its logarithmic behavior at lateral distances $r > 5\lambda$, as shown in Fig. 10. The effect of this correction to logarithmic behavior at short range on the resulting diffraction line shape is illustrated in Fig. 11, where we have used Mochrie's height difference correlation function in a numerical integration of the structure factor convoluted with our instrument/sample response for different values of the crossover distance λ.

For very small values of λ ($\to 0$), the resulting peak shape maintains power-law behavior, as expected. For very large λ ($\to \infty$), the peak shape is expected and seen

to be Gaussian. However, for intermediate λ, the peak shape becomes almost Lorentzian. With our sample/instrument response, we find peak shapes practically indistinguishable from a simple 2D Lorentz for $\lambda \sim 10$ Å.

Figure 11 demonstrates as well that an analysis of such line shapes in terms of the simplified procedures described above (slope of log-log plots or pure power-law line shapes) may lead to a substantial underestimation of the roughening exponent τ as compared to the actual one. One can still find reasonably straight lines in the wings of the line shapes, but their slope depends sensitively on the value of the crossover distance λ. Generally, such a simplified analysis thus appears to be only appropriate, if the crossover distance is very small.

For the analysis of our data in terms of Mochrie's correlation function, we have used a three parameter
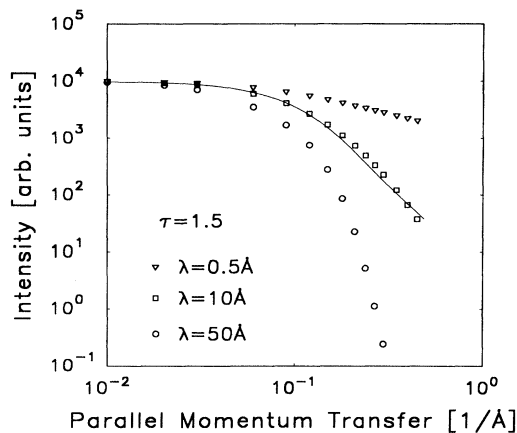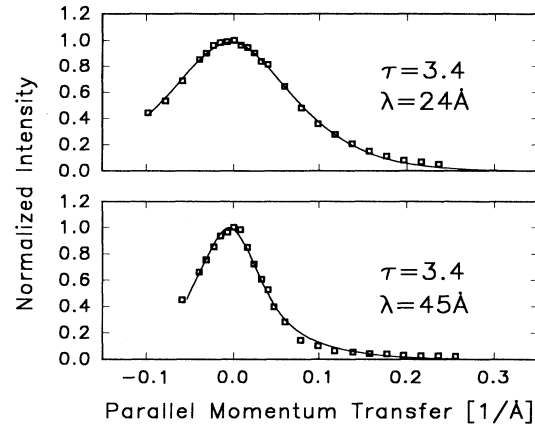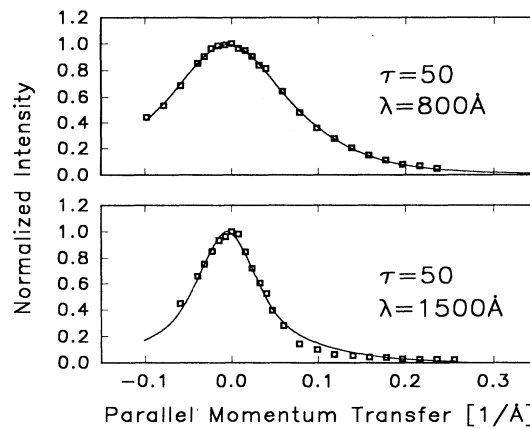

FIG. 11. Calculated peak profiles under the antiphase condition based on $H(r) = 1/2A(T)\ln[(r/\lambda)^2 + 1]$ for $\tau = 1.5$ and different values of the crossover length λ, convoluted with our sample/instrument response. For $\lambda \sim 10$ Å, we find peak profiles practically indistinguishable from a 2D Lorentzian (solid line).

FIG. 13. Fits to the elastic only diffraction profiles for $T = 540$ K based on $H(r) = 2\tau/\pi^2\ln[(r/\lambda) + 1]$, convolution with our sample/instrument response included. The large values for τ and the λ's demonstrate that possible logarithmic behavior of the correlation function is not reflected in our data, but the entire profile can be described by a 2D Lorentz alone.

description in terms of the roughening exponent and two crossover lengths parallel and perpendicular to the steps $(\tau, \lambda_{pa}, \lambda_{perp})$. The fits to the line shapes were done simultaneously for both azimuthal directions and included the sample/instrument response. Figure 12 shows that Mochrie's height difference correlation function can reasonably well describe our experimental peak shapes. The resulting large value for τ, if relevant, would suggest actually that the Cu(115) surface would be very rough already at 540 K, with τ's substantially larger than those deduced by analyzing line shapes assuming logarithmic behavior of the correlation function down to very small distances. However, the interpretation of these values for τ and its relation to a logarithmically diverging height difference correlation function must be taken with caution. It appears that these fit parameters are determined primarily by the *short-range* behavior of the empirically constructed height correlation function and they are not very sensitive to its long-range logarithmic behavior.

As a general rule, the structure factor maintains the power-law behavior for $Q_\parallel < 1/\lambda$, with $Q_\parallel = |\mathbf{Q}_\parallel|$. That means, the $Q_\parallel$ window, which is sensitive to the long-range logarithmic behavior is reduced to very small $Q_\parallel$ values, close to the origin. But just there, for $Q_\parallel < 2\pi/L$, the long-range cutoff L, due to the sample/instrument response, dominates the line shape. Thus, it is only the range $1/\lambda > Q_\parallel > 2\pi/L$ of the peak profile, that can be sensitive to the roughening exponent τ. If L, however, is on the order of λ, the long-range behavior of the correlation function is not reflected in the peak profile. Since the values for λ are comparable to L in our case (we recall that logarithmic behavior is recovered for distances larger than 5λ, see Fig. 10), τ is here reduced to a simple fitting parameter that has no physical significance.

This is substantiated by the following reasoning. Mochrie's height difference correlation function that corrects the short-range behavior in terms of a parabolic law shows a positive curvature at very short distances, but a negative curvature at large r, see Fig. 10. With the τ's and λ's deduced from the fits to our experimental data, the point of inflection is located at about 30 Å. Thus, the correlation function shows no logarithmic behavior nor a preferential curvature in the distance range set by our instrument/sample response, but winds around a linear correlation function. This implies that we should be able to describe the line shapes also with

$$H(r) = \frac{2}{\pi^2}\tau \ln[r/\lambda + 1] \ . \tag{7}$$

This correlation function is linear at short distances and recovers logarithmic behavior for distances larger than about three times the crossover length λ. A description of our data with this functional form produces good fits, however with τ and λ being extremely large, see Fig. 13.

These large values of τ and λ have of course no physical significance, but simply reflect the fact that the peak
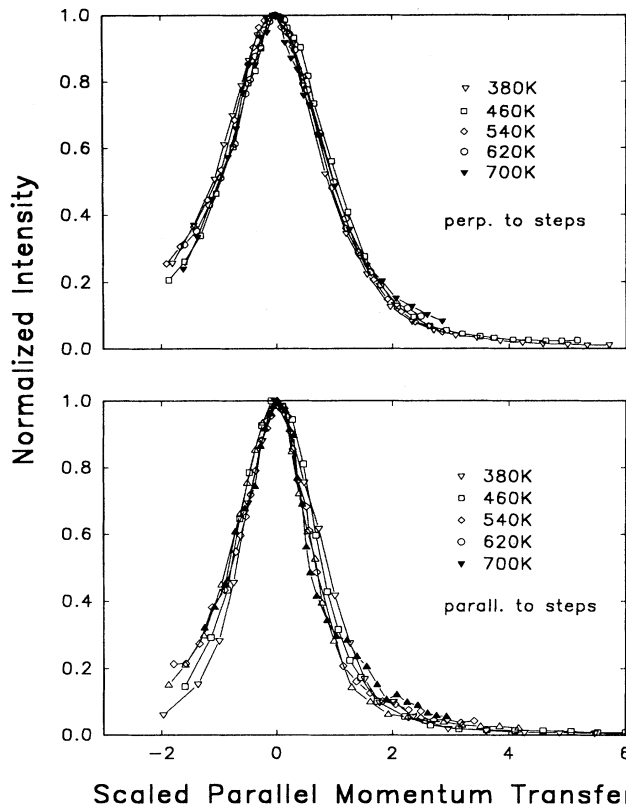

FIG. 14. Scaled antiphase angular distributions (elastic only scattering) in the temperature range between 380 and 700 K, upper panel perpendicular to the steps, lower panel parallel to the steps. The angular distributions are seen to fall upon each other. The absolute FWHM's in this temperature range vary by a factor 2 perpendicular and by a factor 5 parallel to the steps. If there were power-law components in the line shapes, this type of scaling would fail.

shapes are determined by the linearized version of Eq. (7), i.e.,

$$H(r) = \frac{2}{\pi^2}\frac{\tau}{\lambda}r \ . \tag{8}$$

This linear correlation function implies that the peak shape is described by a 2D Lorentzian, whose halfwidth is simply set by *one single* parameter, the ratio τ/λ (λ is much larger than our sample/instrument response length). Thus, this analysis reveals that the *entire* profile can be described by a one-parameter description in terms of a 2D Lorentzian; logarithmic behavior is not reflected in our data.

This finding holds for all high-temperature data. Figure 14 shows the peak profiles for different surface temperatures, each scaled with their respective FWHM. It is seen that in both symmetry directions the peak profiles can be made to fall upon each other with this procedure;

if the peak profiles showed power-law behavior, this type of scaling would fail.

CONCLUSION AND DISCUSSION

The thermal roughening of Cu(115) has been examined with energy-resolved helium-atom-beam scattering. The diffraction peak shapes are found to be substantially altered when TOF analysis, in order to reject inelastic scattering, is performed. The elastic only diffraction line shapes can well be described in both azimuthal directions by a 2D Lorentzian peak form for surface temperatures up to 700 K, in contrast to previous experiments without TOF analysis, in which a power-law line shape has been found. This result underlines the necessity to reject inelastic scattering in helium-scattering experiments (and presumably also in experiments performed with other diffraction techniques), if one is interested in a detailed line shape analysis. This conclusion has also been reached from energy resolved helium-scattering experiments by Zeppenfeld et al.[15] for the Cu(110) surface and by Cvetko et al. and Bracco et al.[16] for the Au(110) and Ag(110) surface, respectively.

The observed Lorentzian line shape in our experiment asserts that the surface morphology can be described by a linear height difference correlation function, in contrast to the expected logarithmic behavior. The question then arises, if the surface in the temperature range covered by our experiment is in the "thermodynamically" rough state at all. Clearly, this question cannot be answered on the basis of our findings for the shape of the peak profiles, since the criterium for the roughening temperature, $\tau = 1$,[2,3] is not applicable any more. An alternative way to determine the roughening temperature is to focus on the width of the interface, which is predicted to be infinite in the rough state.[1-3] A finite width of the interface would give rise to a second, resolution limited component in the structure factor.[6,10] We found that this component disappears in the temperature range between 350 and 400 K. However, this observation is not a sufficient criterion to establish the roughening temperature, because this second component must be shown to disappear at the same temperature for all phases (except in phase), and not only under antiphase conditions.

Thus, on the basis of the present data set, it is not cogent that the surface is not in a Kosterlitz-Thouless type rough state, but we do assert that a possible crossover to a logarithmically diverging height difference correlation function does not occur on our Cu(115) sample for distances up to our sample/instrument response length, i.e., about 100 Å. This finding stands in contrast to Monte Carlo simulations using nearest-neighbor interaction lattice models, which suggest[5,17] that the logarithmic behavior is obtained whenever r exceeds the nearest-neighbor distance perpendicular to the steps. A very large crossover length to logarithmic behavior of about 200 Å has been found in a recent x-ray scattering experiment on the thermal behavior of Pt(100) as well.[14]

These results provoke the seemingly trivial statement that both an instrument with a large transfer width and

samples with large average terrace widths are needed in order to answer the question of how the height correlation function behaves at large distances. While considerable improvements with respect to the performance of the instruments in helium, electron, and x-ray scattering with transfer widths of thousands of Å's have been reached, average terrace widths of, in particular, metallic samples are, with few exceptions such as Pt(111),[18] Pt(100),[14] or Cu(100),[19] often limited to a few hundred Å's or less. In our opinion, it is not excluded that for such surfaces the behavior of the correlation function is imposed by finite-size effects.

Semiconductor surfaces, in particular Si(100) can be prepared with much larger average terrace widths than most metal surfaces and the phase transition should, therefore, be much less susceptible to finite-size effects. However, a recent scanning tunneling microscopy and LEEM study reveals the surprising result that no logarithmic behavior of the correlation function for distances up to a few micrometers has been detected either.[20] A recent determination of the correlation function by reflection electron microscopy on vicinal Si (111) (Ref. 21) led to the conclusion that logarithmic behavior is observed only at very *short* distances (up to seven interstep distances), but a much steeper increase than logarithmic is observed for large distances, there, where one would expect logarithmic behavior.

Given this diversity of experimental results it is, in our opinion, fair to say that the existence of logarithmically diverging correlation functions on "real" vicinal surfaces in their rough state is not generally experimentally well established.[22] The difficulties to produce high-quality, residual defect-free surfaces could certainly be one of the causes. One might also ask to what extent a vicinal surface is actually a realization of a true two-dimensional system, for which logarithmic behavior is expected. In fact, the experimentally found linear correlation function is reminiscent of a 1D system (which is thermodynamically rough at any nonzero temperature).[2] On the other hand, the theory of Rys[23] questions even the very existence of a Kosterlitz-Thouless-type phase transition with a logarithmically diverging height difference correlation function on vicinal surfaces. He argues that long-range (beyond nearest-neighbor) interactions between steps destroy the Kosterlitz-Thouless-type phase transition. While on singular surfaces, the existence of long-range, many-body interactions (and, in particular, the inclusion of corner interactions,[23] which break the hill-and-valley symmetry) introduce different types of phase transitions and push the Kosterlitz-Thouless phase transition to higher temperatures,[24] its implications for the behavior of vicinal surfaces have not yet been, to the best of our knowledge, explicitly investigated. Theory[2] and Monte Carlo simulations[5,17] for vicinal surfaces are based on approximate Hamiltonians with repulsive nearest-neighbor interactions so far. Interactions of long-range nature are certainly conceivable, in particular for vicinal metal surfaces. In fact, there is growing experimental[25] and theoretical[26] evidence that interactions between steps can even be attractive for certain interstep distances.

Long-range and many-body interactions could set

different internal length scales other than the interstep separation perpendicular and the collision length[17] parallel to the steps, so that the expected logarithmic behavior of the correlation function will be shifted to large distances, not accessible to most experiments. Therefore, it would be helpful to elucidate in theory and simulations the behavior of the correlation function at short and intermediate range, when both finite-size effects and a more realistic treatment of interactions between steps are taken into account.

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