Scattering by rough surfaces

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We have used the first Born approximation to derive analytic expressions for the scattering by an oriented solid-on-solid rough surface. The surface roughness is described by a vertical height profile $h(\mathbf{r})$ which is a random function of the positional vector $\mathbf{r}$ in the horizontal plane. The mean-square height difference for such surfaces is assumed to diverge either as a power law or a logarithm of r, i.e., $w_r^2 \propto r^{2\alpha}$ or $w_r^2 \propto \ln r$. We derived expressions for both the specular reflection and the diffuse scattering. Effects due to the surface density profile and to atoms absorbed onto the surface, and relevance to the study of the roughening transition are discussed. We also show that the angular average of our result over all directions of the wave vector $\mathbf{q}$ gives a $1/q^{3+\alpha}$ dependence, in agreement with an earlier calculation for inhomogeneous systems with randomly oriented internal surfaces.

I. INTRODUCTION

The scattering of a plane wave by a rough surface can give useful information about its geometrical properties. Submicrometer-level structures can be probed by x-ray and neutron scattering experiments. There are two general classes of problems. The first is scattering by inhomogeneous materials with randomly oriented internal surfaces,[1,2] e.g., colloidal suspensions,[3] porous glass,[4] sedimentary rocks,[5] cements,[6] oil-water emulsions,[7] ferromagnetic and ferroelectric materials with domain walls,[1,8,9] etc. The second is scattering by a single-oriented surface,[10,11] e.g., the air-water interface,[12] a metal surface in vacuum,[13,14] etc. If the interfaces are smooth, the problems are well understood. Scattering by inhomogeneous materials obeys the well-known Debye-Porod law[15,16] and scattering by single surfaces obeys the optical Fresnel law of reflection.[17] In recent years, there has been much interest in studying systems with rough surfaces, and it has become neccessary to understand how these classical results are modified. In a previous paper[1] (hereafter called Paper I), we have developed the scattering theory for inhomogeneous materials with rough internal surfaces. Specifically, we have shown that if the surfaces have a power-law roughness (defined below), the scattering intensity $I(q)$ has a $1/q^{3+\alpha}$ dependence on the wave vector transfer $\mathbf{q}$, with $0 \le \alpha \le 1$. This type of surface is also known as a *self-affine* fractal[8] and has fractal dimension $D = 3-\alpha$. Thus the $1/q^{3+\alpha}$ behavior can also be written as $1/q^{6-D}$, which was independently derived by Bale and Schmidt[19] for *self-similar* fractal surfaces. Such a behavior was observed in many experiments.[2] In this paper, we treat the problem of scattering by a single-oriented rough surface and show how the roughness affects both the specular reflection ($\mathbf{q}$ parallel to surface normal) and diffuse scattering ($\mathbf{q}$ has a component $\mathbf{q}_\perp$ perpendicular to the surface normal). Our work is motivated mainly by the growing interest in using synchrotron-generated high-intensity x-rays to study single surfaces.[11-14]

As in Paper I, the type of rough surface we consider is the so-called solid-on-solid model[20] in which the surface is defined by a vertical-height profile above a horizontal xy plane and can be represented by a single-valued random function $z = h(\mathbf{r})$, where $\mathbf{r} = (x,y)$ is a positional vector in the xy plane. The mean-square interface width w_r^2 is defined by

$$w_r^2 \equiv \langle [h(\mathbf{r}) - h(0)]^2 \rangle_0 , \tag{1.1}$$

where $\langle\ \rangle_0$ denotes an average over all choices of the origin. The surface is assumed to be sharp in the sense that the scattering amplitude changes discretely like a step function across the surface. (This assumption can be relaxed, see Sec. IV.) Two types of roughness are treated in our calculation. The first is a power-law roughness characterized by

$$w_r = b(r/b)^\alpha , \tag{1.2}$$

with α between 0 and 1. As mentioned above, this type of surface is also known as a self-affine fractal, and its fractal dimension is given by $D = 3-\alpha$. b is a microscopic length scale that characterizes the magnitude of the roughness. It is called the crossover length[18] because $w_r > r$ for $r < b$ and $w_r < r$ for $r > b$. As a result, even though w_r diverges at large r, $w_r/r \to 0$ as $r \to \infty$, and the surface is macroscopically smooth. Such surfaces have been used as models for fracture surfaces of metals,[21] rock formations,[22] etc. The second type of roughness we consider is a logarithmic one

$$w_r^2 = b^2 \ln(r/a) , \tag{1.3}$$

where a is the lattice constant. These surfaces arise in the context of roughening transitions[20] which have received much attention recently.[13,14,23,24] Although Eq. (1.3) can be regarded as a type of limiting case ($\alpha=0$) of Eq. (1.2), the calculations have to be performed separately.

The organization of the paper is as follows. In Sec. II, we derive the general expressions for wave-vector transfer parallel and perpendicular to the surface normal ($q_\parallel$ and $\mathbf{q}_\perp$, respectively) and apply them to the power-law rough surfaces. We also show that the result we obtained in Paper I for inhomogeneous systems can be obtained by angular averaging the expressions for single surfaces. Section III describes the results for logarithmic roughness and possible applications to the study of roughening transition. In Sec. IV we summarize our findings and discuss some extensions of our results.

II. GENERAL RESULTS AND POWER-LAW ROUGHNESS

A. General results

A general scattering theory for surfaces has been given previously by Andrews and Cowley.[10] We present here a different formulation which can be easily applied to different types of roughness such as those described by Eqs. (1.2) and (1.3).

Using Cartesian coordinates, we assume the surface is macroscopically oriented in the xy plane with its normal along the z direction. Microscopically, the rough surface is defined by a set of points (x_0,y_0,z_0) which satisfy an equation $z_0=h(x_0,y_0)$, where h is a single-valued function. The assumption of a sharp interface implies that the scattering-length density (amplitude per unit volume) at a point $\mathbf{R}=(x,y,z)$ can be written as $\eta(\mathbf{R})=\rho\Theta(h_R-z)$, where Θ is the step function and $h_R\equiv h(x,y)$. In other words, η is a finite constant ρ below the surface and zero above it. For wave-vector transfer $\mathbf{q}$, the first Born approximation gives the scattered intensity (cross section) as

$$I(\mathbf{q})=\int d\mathbf{R}'\int d\mathbf{R}\, e^{i\mathbf{q}\cdot\mathbf{R}}\langle\eta(\mathbf{R}')\eta(\mathbf{R}'+\mathbf{R})\rangle$$
$$=\rho^2\int d\mathbf{R}'\int d\mathbf{R}\, e^{i\mathbf{q}\cdot\mathbf{R}}\times\langle\Theta(h_{R'}-z')\Theta(h_{R+R'}-z-z')\rangle\ , \tag{2.1}$$

where $h_{R'}\equiv h(x',y')$ and $h_{R+R'}\equiv h(x+x',y+y')\equiv h_{R'}+h_r$. Thus h_r is the height difference between two points on the surface which have their horizontal coordinates separated by $\mathbf{r}=(x,y)$. It is a random variable with an average value of zero and a probability distribution $P(h_r/w_r)$, where w_r is the width of the distribution and $\int_{-\infty}^{\infty}P(v)dv=1$. Assuming that the surface is isotropic in the xy plane, w_r depends only on the distance $r=(x^2+y^2)^{1/2}$. By a change of variable, letting $(h_{R'}-z')\to z'$, the above integral becomes

$$I(\mathbf{q})=\rho^2 A\int dz'\int dz\, e^{iq_\parallel z}\int d\mathbf{r}\, e^{i\mathbf{q}_\perp\cdot\mathbf{r}}\int d(h_r/w_r)P(h_r/w_r)\Theta(z')\Theta(h_r+z'-z)\ , \tag{2.2}$$

where $A=\int dx'\,dy'$ is the macroscopic area of the surface. The statistical average $\langle\ \rangle$ over the random configurations is replaced by the h_r integral over $P(h_r/w_r)$. By rewriting $e^{iq_\parallel z}=e^{iq_\parallel(z-z'-h_r)}e^{iq_\parallel z'}e^{iq_\parallel h_r}$, we have

$$I(\mathbf{q})=\rho^2 A\int dz'\,e^{iq_\parallel z'}\Theta(z')\int dz\, e^{iq_\parallel(z-z'-h_r)}\Theta(h_r+z'-z)\int r\,dr\int d\phi\, e^{iq_\perp r\cos\phi}\int d(h_r/w_r)e^{iq_\parallel h_r}P(h_r/w_r)\ . \tag{2.3}$$

Using the identity

$$\lim_{\epsilon\to 0}\int_{-\infty}^{+\infty}dz\,\Theta(z)e^{-(ik+\epsilon)z}=\frac{1}{ik}$$

we can see that the first two integrals give a factor of $1/q_\parallel^2$, which is equivalent to the square of the surface form factor in the z direction. The ϕ integral gives $2\pi J_0(q_\perp r)$ where J_0 is the zero-order Bessel function of the first kind. The h_r integral gives generally a function $\tilde{P}(q_\parallel w_r)$. Together, we have the general result for any q:

$$I(\mathbf{q})=\frac{\rho^2 A}{q_\parallel^2}\int 2\pi r\,dr\,J_0(q_\perp r)\tilde{P}(q_\parallel w_r)\ . \tag{2.4}$$

To proceed further, we shall *assume* a Gaussian probability distribution function

$$P(h_r/w_r)=\frac{1}{\sqrt{2\pi}}e^{-h_r^2/w_r^2} \tag{2.5}$$

which gives

$$\tilde{P}(q_\parallel w_r)=e^{-q_\parallel^2 w_r^2/2}\ . \tag{2.6}$$

For $\mathbf{q}_\perp=0$, $J_0(0)=1$ in Eq. (2.4) and the specularly reflected intensity has the form

$$I(q_\parallel)=\frac{\rho^2 A}{q_\parallel^2}\int 2\pi r\,dr\,e^{-q_\parallel^2 w_r^2/2}\ . \tag{2.7}$$

We note that if the surface is perfectly smooth, then $w_r=0$ and the integral gives a factor A. Equation (2.7) is then reduced to

$$I(q_\parallel)=\frac{\rho^2 A^2}{q_\parallel^2}\ . \tag{2.8}$$

Contrary to this expression, the Fresnel law in optics has a $q_\parallel^{-4}$ tail for $I(q_\parallel)$. The reason is that the beam size A_0 is usually much smaller than the reflecting surface in optics. As a result, the reflecting area is given by $A=A_0/\sin\theta$, where 2θ is the scattering angle. Since $q_\parallel=2Q\sin\theta$, where Q is the magnitude of the wave vec-

tor for the incident and reflected beams, we can rewrite Eq. (2.8) as $I(q_\parallel)=4\rho^2 A_0^2 Q^2/q_\parallel^4$, which is the familiar form of Fresnel reflectivity. Therefore, Eq. (2.7) can now be understood simply as the Fresnel reflectivity reduced by a Debye-Waller-like factor due to the surface roughness. This change in the reflected intensity becomes the diffuse scattering in other directions

$$I(q_\perp,q_\parallel)=\frac{\rho^2 A}{q_\parallel^2}\int 2\pi r\, dr\, J_0(q_\perp r)e^{-q_\parallel^2 w_r^2/2} . \tag{2.9}$$

Note that $q_\perp$ is coupled to the in-plane distance r and $q_\parallel$ is coupled to the vertical fluctuation w_r.

B. Power-law roughness, $\alpha>0$

The above results are general in the sense that no specific form for w_r has been assumed. Let us now consider the power-law roughness defined in Eq. (1.2). If the exponent $\alpha=0$, w_r can either be independent of r or dependent on the logarithm of r. In the first case, the surface roughness has no spatial correlations. The exponential factor in Eqs. (2.7) and (2.9) can simply be factored out of the integrals. The logarithmic case will be treated in Sec. III. Here we treat the $\alpha>0$ problem.

For specular reflection, Eq. (2.7) gives

$$I(q_\parallel)=\frac{\rho^2 A}{q_\parallel^2}\int 2\pi r\, dr\, e^{-q_\parallel^2 b^{2-2\alpha} r^{2\alpha}/2} .$$

With a change of variable $k=(q_\parallel b)^{1/\alpha} r/b$, we obtain

$$\begin{aligned} I(q_\parallel)&=\frac{\rho^2 b^4 A}{(q_\parallel b)^{2+2/\alpha}}\int_0^\infty 2\pi k\, dk\, e^{-k^{2\alpha}/2} \\ &=\frac{\rho^2 b^4 A}{(q_\parallel b)^{2+2/\alpha}}\frac{\pi 2^{1/\alpha}}{\alpha}\Gamma(1/\alpha) , \end{aligned} \tag{2.10}$$

where Γ denotes the gamma function. This expression predicts an algebraic falloff faster than the $q_\parallel^{-4}$ behavior of a planar surface. Equation (2.10) allows both b and α to be determined from the specular reflection. For the diffuse scattering, we substitute k into Eq. (2.9) and use the fact that J_0 is an even function,

$$\begin{aligned} I(q_\perp,q_\parallel)&=\frac{\rho^2 b^4 A}{(q_\parallel b)^{2+2/\alpha}} \\ &\quad\times\int_0^\infty 2\pi k\, dk\, J_0[kbq_\perp(bq_\parallel)^{-1/\alpha}]e^{-k^{2\alpha}/2} \\ &\equiv\frac{\rho^2 b^4 A}{(q_\parallel b)^{2+2/\alpha}}2\pi G_\alpha\left[\frac{b^2 q_\perp^2}{(bq_\parallel)^{2/\alpha}}\right] . \end{aligned} \tag{2.11}$$

The functional form of G_α depends on the value of α. For special values of α, the integral in Eq. (2.11) can be evaluated. For examples, for $\alpha=1$, using Eqs. (6.631), (9.210) and (9.215) in Ref. 25 gives a Gaussian

$$G_1=\exp\left[-\frac{q_\perp^2}{2q_\parallel^2}\right] .$$

For $\alpha=\frac{1}{2}$, using Eq. (6.623) in Ref. 25, we find

$$G_{1/2}=4\left[1+\frac{4q_\perp^2}{b^2 q_\parallel^4}\right] .$$

In general, G_α is a symmetric peak that centers at $q_\perp=0$ with a width proportional to $(bq_\parallel)^{1/\alpha}/b$. Therefore, in principle b and α can also be determined by diffuse scattering measurements.

C. Angular average

We next compute the angular average of Eq. (2.11) to show that it gives the same results in Paper I which were derived for inhomogeneous systems with randomly oriented internal surfaces. Using spherical coordinates, let θ be the angle between the scattering vector $\mathbf{q}$ and the surface normal $\mathbf{n}$, then $q_\parallel=q\cos\theta$ and $q_\perp=q\sin\theta$. Since Eq. (2.11) is independent of the direction of $\mathbf{q}_\perp$, it is not neccessary to average over the azimuthal angle ϕ. For the average over θ, we let $p=\cos\theta$ in Eq. (2.11) and integrate over p,

$$I(q)=\frac{\rho^2 b^4 A}{(qb)^{2+2/\alpha}}2\pi\int_0^1\frac{dp}{p^{2+2/\alpha}}G_\alpha\left[\frac{b^2 q^2(1-p^2)}{(bqp)^{2/\alpha}}\right] . \tag{2.12}$$

For $qb\gg 1$, the main contribution to the integral comes from small values of p such that the argument of G_α is of order unity. Thus we define a new variable $u=p(qb)^{1-\alpha}$ and transform the integral to

$$\begin{aligned} I(q)&=\frac{\rho^2 b^4 A}{(qb)^{3+\alpha}}2\pi \\ &\quad\times\int_0^{(qb)^{1-\alpha}}\frac{du}{u^{2+2/\alpha}}G_\alpha\left[\frac{1-u^2(qb)^{2\alpha-2}}{u^{2/\alpha}}\right] . \end{aligned} \tag{2.13}$$

We note that the integrand is suppressed at the lower limit by the peak function G_α, and at the upper limit by the algebraic denominator. The product of the two factors peaks near $u\approx 1$ and the integral can be approximated by a constant

$$\begin{aligned} C_1&=\int_0^\infty\frac{du}{u^{2+2/\alpha}}G_\alpha\left[\frac{1}{u^{2/\alpha}}\right] \\ &=\alpha 2^\alpha\left[\frac{\pi}{2}\right]^{1/2}\frac{\Gamma\left[1+\frac{\alpha}{2}\right]}{\Gamma\left[1-\frac{\alpha}{2}\right]} . \end{aligned} \tag{2.14}$$

In the evaluation of this integral, we have used the definition of G_α given in Eq. (2.11). We have also verified that using the results in Paper I to calculate the amplitude of this term gives exactly the same expression for C_1. Therefore, the dominant behavior for $qb\gg 1$ is a $1/q^{3+\alpha}$ power law, in agreement with Paper I. In the opposite limit $qb\ll 1$, the function G_α in Eq. (2.12) is appreciable only for $p\approx 1$. Hence we define another new variable $t=(qb)^{1-1/\alpha}(1-p^2)^{1/2}$ and transform Eq. (2.12) to

$$I(q)=\frac{\rho^2 b^4 A}{(qb)^4}2\pi\int_0^{(qb)^{1-1/\alpha}}\frac{tdt}{[1-(qb)^{-2+2/\alpha}t^2]^{3+2/\alpha}}G_\alpha\left[\frac{t^2}{[1-(bq)^{-2+2/\alpha}t^2]^{1/\alpha}}\right] . \tag{2.15}$$

The integral converges at the lower limit due to the numerator t, and it is suppressed at the upper limit by the function G_α. The main contribution comes from $t\approx 1$, and it can be estimated by a constant

$$\begin{aligned} C_2&=\int_0^\infty t\,dt\,G_\alpha(t^2/2)=\int_0^\infty k\,dk\,e^{-k^{2\alpha}/2}\int_0^\infty t\,dt\int_0^{2\pi}\frac{d\phi}{2\pi}e^{ikt\cos\phi}\\ &=\int_0^\infty k\,dk\,e^{-k^{2\alpha}/2}\frac{1}{2\pi}\int d\mathbf{t}\,e^{i\mathbf{k}\cdot\mathbf{t}}\\ &=\int_0^\infty k\,dk\,e^{-k^{2\alpha}/2}2\pi\delta(\mathbf{k})=\int d\mathbf{k}\,e^{-k^{2\alpha}/2}\delta(\mathbf{k})=1 . \end{aligned} \tag{2.16}$$

Hence the scattering for $qb\ll 1$ is a q^{-4} power law, as found in Paper I. Substituting C_2 into Eq. (2.15) gives exactly the smooth-surface result derived by Debye, Anderson, and Brumberger in Ref. 15. This is due to the fact that the roughness is not observable at a length scale $r\sim 1/q\gg b$. For the same reason, we note that while C_1 depends on α, C_2 does not. It is also interesting to note that while the integrand in Eq. (2.16) contains α, the end result does not.

III. LOGARITHMIC ROUGHNESS AND ROUGHENING TRANSITION

The solid-on-solid model predicts that the surface of a solid can undergo a roughening transition at some temperature T_R below the melting point of the solid.[20,24] The interface width is described by Eq. (1.3) for temperatures $T>T_R$. Below T_R, the width is finite, and there is a finite correlation length ξ. Although there is no specific prediction in the literature, we expect Eq. (1.3) to hold for $r\ll\xi$, and for $r\gg\xi$, w_r^2 should tend to a constant value $b^2\ln(\xi/a)$. In other words, the mean-square width should obey a scaling form

$$w_r^2=b^2\ln\left[\frac{r}{a}f(r/\xi)\right] , \tag{3.1}$$

where $f(r/\xi)$ is a scaling function with the properties

$$f(0)=1 \text{ and } f(r/\xi)=\xi/r \text{ as } r/\xi\to\infty .$$

For convenience of calculation, we shall assume

$$f(r/\xi)=\frac{1}{r/\xi}(1-e^{-r/\xi}) . \tag{3.2}$$

This will affect our result in the region where $q\xi\sim 1$, but not for $q\xi\gg 1$ or $q\xi\ll 1$.

A. Specular reflection ($\mathbf{q}_\perp=0$) in the rough phase ($T>T_R$)

A direct substitution of Eq. (1.3) into Eq. (2.7) gives

$$I(q_\parallel)=\frac{\rho^2 A}{q_\parallel^2}\int_a^L 2\pi r\,dr\,(r/a)^{-q_\parallel^2 b^2/2} , \tag{3.3}$$

where L is the linear dimension of a single-crystal surface. Assuming $L\gg a$ and $A=\pi L^2$, the integral is easily carried out and yields

$$I(q_\parallel)=\frac{\rho^2 A^2}{q_\parallel^2}\frac{(L/a)^{-q_\parallel^2 b^2/2}-(L/a)^{-2}}{1-q_\parallel^2 b^2/4} . \tag{3.4}$$

Note that the first factor is the reflectivity of a perfectly flat surface as given by Eq. (2.8). The second factor is the reduction due to the roughness. For $qb\ll 1$ and $L\gg a$, this factor tends to unity because the roughness is negligible for $r\sim 1/q\gg b$. For $qb\gg 1$, this factor tends to $a^2/L^2b^2q_\parallel^2$ which is less than 10^{-14} for a typical a/L ratio of 10^{-7}. Hence we expect the effect to be unobservable in this limit in an experiment. The effect of roughness should only be observable in a region where $qb\lesssim 1$ where Eq. (3.4) can be approximated by

$$I(q_\parallel)=\frac{\rho^2 A^2}{q_\parallel^2}e^{-(1/2)q_\parallel^2 b^2\ln(L/a)} . \tag{3.5}$$

We note that for $L/a=10^7$, the Debye-Waller factor is greater than $1/e$ for $q_\parallel b\lesssim\frac{1}{3}$. If the reflecting surface consists of domains of different sizes, then the above expression has to be averaged over the size distribution.

B. Specular reflection in the smooth phase ($T<T_R$)

Using Eqs. (3.1) and (3.2) in Eq. (2.7), we obtain

$$\begin{aligned} I(q_\parallel)&=\frac{\rho^2 A}{q_\parallel^2}\int 2\pi r\,dr\left[\frac{\xi}{a}(1-e^{-r/\xi})\right]^{-q_\parallel^2 b^2/2}\\ &=\frac{\rho^2 A}{q_\parallel^2}(\xi/a)^{-q_\parallel^2 b^2/2}\xi^2\int_{a/\xi}^{L/\xi}2\pi s\,ds\,(1-e^{-s})^{-q_\parallel^2 b^2 2}\\ &=\frac{\rho^2 A}{q_\parallel^2}(\xi/a)^{-q_\parallel^2 b^2/2}\left[A+\xi^2\int_0^\infty 2\pi s\,ds[(1-e^{-s})^{-q_\parallel^2 b^2/2}-1]\right] , \end{aligned} \tag{3.6}$$

where we have replaced r/ξ by the variable s and assumed $a \ll \xi \ll L$. If $qb \lesssim 1$ in an experiment, we can rewrite

$$(1-e^{-s})^{-q_\parallel^2 b^2/2}-1=\exp[-\tfrac{1}{2}q_\parallel^2 b^2\ln(1-e^{-s})]-1$$
$$\approx -\tfrac{1}{2}q_\parallel^2 b^2\ln(1-e^{-s})$$
$$=\tfrac{1}{2}q_\parallel^2 b^2\sum_{n=1}^{\infty}\frac{e^{-ns}}{n}\ .$$

Substituting into the integral in Eq. (3.6) and evaluating it term by term we find the leading term in $q_\parallel^2 b^2$ to be

$$\pi q_\parallel^2 b^2\sum_{n=1}^{\infty}\int_0^\infty ds\, s e^{-ns}/n=\pi q_\parallel^2 b^2\zeta(3)\ , \tag{3.7}$$

where $\zeta(3)=\sum_{n=1}^{\infty}n^{-3}$ is the Riemann's ζ function. Combining with Eq. (3.6) gives

$$I(q_\parallel)=\frac{\rho^2A^2}{q_\parallel^2}e^{-(1/2)q_\parallel^2 b^2\ln(\xi/a)}\times\left[1+\frac{\xi^2}{A}\pi\zeta(3)q_\parallel^2 b^2+\cdots\right]\ . \tag{3.8}$$

If $\xi^2 \ll A$ and $qb<1$, we can ignore the high-order terms. We recall that for a reduced temperature $\tau \equiv 1-T/T_R$ below the roughening transition, the correlation length diverges as[20]

$$\xi/a \sim e^{c/\sqrt{\tau}}\ , \tag{3.9}$$

where c is a constant. Thus the temperature dependence of the intensity at fixed $q_\parallel$ is given by

$$I(q_\parallel)=\frac{\rho^2A^2}{q_\parallel^2}e^{-cq_\parallel^2 b^2/2\sqrt{\tau}}\ . \tag{3.10}$$

This expression implies that $I(q_\parallel)$ vanishes at T_R with a zero temperature derivative. In reality, this behavior is cut off by the finite domain size L according to Eq. (3.5). This result seems to be in qualitative agreement with the recent data obtained by Mochrie[13] for the Cu(110) surface, and Held *et al.*[14] for the Ag(110) surface.

C. Diffuse scattering

It is instructive to first carry out the calculation for the smooth phase ($T<T_R$) and then take the $\xi \to L$ limit for the rough phase. We begin by rewriting Eq. (2.9) as

$$I(q_\perp,q_\parallel)=\frac{\rho^2A}{q_\parallel^2}\int 2\pi r\,dr\,J_0(q_\perp r)(e^{-q_\parallel^2 w_\infty^2/2}+e^{-q_\parallel^2 w_r^2/2}-e^{-q_\parallel^2 w_\infty^2/2})\ , \tag{3.11}$$

where $w_\infty^2 \equiv b^2\ln(\xi/a)$. Factoring out $e^{-q_\parallel^2 w_\infty^2/2}$ from the integrand, we note that the first term in the integral gives $A\delta_{q_\perp,0}$ which is just the specular reflection. Using Eqs. (3.1) and (3.2), we have $w_r^2-w_\infty^2=b^2\ln(1-e^{-r/\xi})$. Thus, the remaining terms in the integral can be written as

$$\int_a^L 2\pi r\,dr\,J_0(q_\perp r)(e^{-q_\parallel^2(w_r^2-w_\infty^2)/2}-1)=\int_a^L 2\pi r\,dr\,J_0(q_\perp r)[(1-e^{-r/\xi})^{-q_\parallel^2 b^2/2}-1]$$
$$=2\pi\xi^2\int_0^\infty s\,ds\,J_0(q_\perp\xi_s)[(1-e^{-s})^{-q_\parallel^2 b^2/2}-1]$$
$$\equiv\xi^2 g(q_\perp\xi,q_\parallel b)\ , \tag{3.12}$$

where we have replaced r/ξ by s and assumed $a \ll \xi \ll L$. The form of the diffuse scattering is contained in the function g. Its arguments show that $q_\perp$ is coupled to the in-plane correlation length ξ and that $q_\parallel$ is coupled to the out-of-plane roughness amplitude b, which is what one would expect. Similar to Eq. (3.6), we can calculate g in the small $q_\parallel b$ limit by expanding the integrand in Eq. (3.12) as a power series in e^{-s}. The leading term in $q_\parallel^2 b^2$ is

$$g(q_\perp\xi,q_\parallel b)=\pi q_\parallel^2 b^2\sum_{n=1}^{\infty}\int_0^\infty s\,ds\,J_0(q_\perp\xi r)\frac{e^{-ns}}{n}=\pi q_\parallel^2 b^2\sum_{n=1}^{\infty}(n^2+q_\perp^2\xi^2)^{-3/2}\ , \tag{3.13}$$

where we have used Eq. (6.623) in Ref. 25. We note that this result is a consequence of the scaling function f we assumed in Eq. (3.2). Therefore, while it might be useful as a guide in analyzing $q_\perp$ scans at fixed $q_\parallel$, it is not expected to be completely accurate in the $q_\perp\xi\sim 1$ region. On the other hand, it should give the correct limiting behavior for $q_\perp\xi \ll 1$ and $q_\perp\xi \gg 1$. Combining Eqs. (3.11)–(3.13), we obtain for $T<T_R$,

$$I(q_\perp,q_\parallel)=\frac{\rho^2A^2}{q_\parallel^2}(\xi/a)^{-q_\parallel^2 b^2/2}\left[\delta_{q_\perp,0}+\pi\frac{\xi^2}{A}q_\parallel^2 b^2\sum_{n=1}^{\infty}(n^2+q_\perp^2\xi^2)^{-3/2}+\cdots\right]\ , \tag{3.14}$$

where δ denotes the Kronecker delta. A simple physical understanding of this expression is the following. The first term represents the Bragg scattering and the second term represents the diffuse scattering. Both are attenuated by a Debye-Waller factor due to the surface roughness. The leading $1/q_\parallel^2$ factor comes from the *form factor* $F(q_\parallel)$ of the surface, because it results from the Fourier transform of the scattering density profile in Eq. (2.3). We should note that for $q_\perp=0$, Eq. (3.14) reduces to Eq. (3.8) exactly. Thus the two terms in Eq. (3.8) are understood to be the Bragg and

diffuse scattering in the specular direction. It should be noted that the distinction between Bragg and diffuse scatterings does not exist in Eq. (2.11) for power-law roughness. There, both the specular and off-specular scatterings are described by a single continuous peak function G_α.

Finally, for $T \geq T_R$, if we let $\xi = L$ and $\pi L^2 = A$ in Eq. (3.14), then the Bragg scattering term recovers Eq. (3.5). For the diffuse scattering, since $\xi \to \infty$, the condition $q_\perp \xi \gg 1$ is always satisfied. The discrete sum in Eqs. (3.13) and (3.14) can be replaced by an integral

$$\sum_{n=1}^{\infty} (n^2 + q_\perp^2 \xi^2)^{-3/2} = \int_0^\infty dn\,(n^2 + q_\perp^2 \xi^2)^{-3/2} = (q_\perp \xi)^{-2} . \tag{3.15}$$

Thus, to first order in $q_\parallel^2 b^2$, the decay of the diffuse scattering exhibits a $q_\perp^{-2}$ behavior which should be valid for all values of $q_\perp$ at temperatures above T_R and for $q_\perp \xi \gg 1$ below T_R. For general values of $q_\parallel^2 b^2$, however, this result has to be modified. Substituting Eq. (1.3) directly into Eq. (2.9), we have

$$\begin{aligned} I(q_\perp, q_\parallel) &= \frac{\rho^2 A}{q_\parallel^2} 2\pi \int_0^\infty r\,dr\, J_0(q_\perp r)(r/a)^{-q_\parallel^2 b^2/2} \\ &= \frac{\rho^2 A a^2}{q_\parallel^2 (q_\perp a)^{2 - q_\parallel^2 b^2/2}} 2\pi \lim_{\epsilon \to 0} \int_0^\infty d\nu\, e^{-\epsilon \nu} J_0(\nu) \nu^{1 - q_\parallel^2 b^2/2} \\ &= \frac{\rho^2 A a^2}{q_\parallel^2 (q_\perp a/2)^{2-\tilde{\eta}}} \Gamma^2 \left(1 - \frac{\tilde{\eta}}{2}\right) \sin\frac{\tilde{\eta}\pi}{2} , \end{aligned} \tag{3.16}$$

where $\nu = q_\perp r$, $\tilde{\eta} = q_\parallel^2 b^2/2$ and we have used Eqs. (6.621), (8.756), (8.335), and (8.334) in Ref. 25 to evaluate the integral. The leading order $q_\perp^{-2}$ behavior found in Eq. (3.15) is now modified to a $q_\perp^{-2+\tilde{\eta}}$ power law. This result is intermediate between Eqs. (2.11) and (3.14) in the sense that there is no sharp distinction between Bragg and diffuse scattering, and yet there is a divergence at $q_\perp = 0$. It is interesting to note that the $q_\perp^{-2+\tilde{\eta}}$ behavior in Eq. (3.16) is similar to the $q_\perp^{-2+\eta}$ critical behavior of the two-dimensional (2D) XY model except that, while η is a constant, $\tilde{\eta}$ varies with $q_\parallel$. The reason is that the exponent η in the 2D XY model is proportional to the amplitude b^2 in the solid-on-solid model and b is coupled to $q_\parallel$ in a scattering experiment. This understanding may be useful in analyzing the recent experimental studies of the roughening transitions on Cu(110) and Ag(110) surfaces.[13,14]

IV. SUMMARY AND DISCUSSIONS

To summarize, we have calculated the scattering from a single-oriented rough surface within the first Born approximation. The results should be useful for x-ray and neutron scattering experiments, where the scattering is weak and multiple scattering can be ignored. The most general result is given by Eq. (2.4). With the assumption of a Gaussian probability distribution in Eq. (2.5), we obtained the expression in Eq. (2.9) which should be adequate for most practical purposes. The assumption of a power-law roughness leads to Eq. (2.11) which allows one to determine the amplitude b and the exponent α. The angular average of Eq. (2.11) agrees with the results first derived in Paper I for inhomogeneous systems with randomly oriented surfaces. The calculations for the logarithmically rough surface are carried out with an assumed scaling function defined by Eq. (3.2). This leads to the expressions in Eqs. (3.14) and (3.16), which can in principle be compared with experimental studies of the roughening transition.

It is important to point out that Eqs. (3.14), (3.16), and (2.11) represent three classes of behavior. Equation (3.14) has a clear distinction between Bragg and diffuse scatterings. There is a discontinuity at $q_\perp = 0$. The physical reason is that the height fluctuations are only correlated over a finite distance ξ, and they are bounded. The Bragg intensity is that of a perfectly flat surface modified by a Debye-Waller factor. We expect this general behavior to hold for all surfaces with finite w_r^2. Equations (3.16) and (2.11) apply to situations where w_r^2 diverges. Neither of them contains a Debye-Waller factor and there is no discontinuity at $q_\perp$ to distinguish Bragg and diffuse scatterings. However, the weak logarithmic divergence of w_r^2 in Eq. (1.3) leads to a $q_\perp^{-2+\tilde{\eta}}$ divergence in Eq. (3.16), whereas the strong power-law roughness of Eq. (1.2) leads to a smooth peak function G_α in Eq. (2.11). If the surface consists of many domains of size L, then Eqs. (2.11) and (3.16) should only hold for $q_\perp L > 1$. For $q_\perp L < 1$, the scattering should evolve into separate Bragg and diffuse components, and they can be calculated by Eqs. (2.7) and (2.9) with w_L. If there is a distribution of domain size, the result has to be averaged over L.[26]

Throughout our calculation, two other assumptions have been made for the sake of convenience. Both of them can be relaxed. First, we have assumed a continuum scattering density in Eq. (2.1) which gives the ρ^2 factor in all our results. Modification for a lattice structure is a simple procedure. One simply replaces ρ by

$$\rho_{\mathbf{q}} \equiv \sum_i f_i(\mathbf{q}) e^{i\mathbf{q}\cdot\mathbf{r}_i} \tag{4.1}$$

where the sum is performed over a unit cell, f_i is the form factor for the ith atom, and $\mathbf{r}_i$ its position. As usual, the discrete nature of the lattice also changes $\mathbf{q}$ to

$\mathbf{q}-\mathbf{G}$ in all the results ($\mathbf{G}$ is any reciprocal lattice vector with a bulk scattering Bragg peak) so that all the predictions can be tested near a Bragg peak. The second assumption we made is the infinite sharpness of the density profile, which is represented by the step functions in Eq. (2.1). This leads to the $1/q_\parallel^2$ prefactor that appears in all the expressions, and it can be understood as due to the form factor $F(q_\parallel)$ of the surface along the normal direction. In general, the interface can have a finite internal width W over which the density changes gradually. For such cases, we can replace the $1/q_\parallel^2$ factor by $|F(q_\parallel)|^2$ in all the expressions above. For $q_\parallel < 1/W$, the internal structure cannot be probed and $F(q_\parallel)$ must tend to $1/q_\parallel$. For larger values of $q_\parallel$, $F(q_\parallel)$ should fall off faster than $1/q_\parallel$, and this can give information about the density variation profile across the interface. To give an explicit example, we consider a Fermi-function type density profile $\rho(1+e^{z/W})^{-1}$. Using Eq. (3.222) in Ref. 25, it is easy to show that

$$F(q_\parallel)=\int_{-\infty}^{\infty}\frac{dz\,e^{iq_\parallel z}}{(1+e^{z/W})}$$
$$=W\int_0^{\infty}\frac{du\,u^{iq_\parallel W-1}}{1+u}$$
$$=\frac{\pi W}{\sin iq_\parallel W\pi}=\frac{\pi W}{i\sinh\pi q_\parallel W}\,, \tag{4.2}$$

where we have used $u\equiv e^{z/W}$. For $q_\parallel W\ll 1$, expanding the sinh function recovers the sharp interface form factor $F(q_\parallel)=1/iq_\parallel$ exactly. For any other values of $q_\parallel$, $|F(q_\parallel)|<1/q_\parallel$ and hence the scattering intensity is always less than that by a sharp interface. There are many systems for which the knowledge of $F(q_\parallel)$ is useful, e.g., Bloch walls of magnetic domains, surfactant layers oil and water, heterojunctions in semiconductor devices, etc.

Finally, we should point out that all of the above calculations apply to the general situation where there is a scattering density difference across the interface. A closely related problem is to have strong scatterers lining a rough interface with weak scatterers on both sides, e.g., x-ray scattering by Xe or Kr atoms absorbed on beryllium or graphite surfaces. This problem is also related to the scattering at the anti-Bragg points in the previous (clean surface) problem.[14] The reason is that the anti-Bragg vector $\mathbf{q}$ correspond to $\rho_{\mathbf{q}}=0$ in Eq. (4.1) and hence only the partially-filled unit cells at the surface layer contribute to the scattering. Although the scattering amplitude for each sell is different, there is an average value $\langle\rho_{\mathbf{q}}\rangle$ which gives rise to a coherent scattering and the fluctuations about $\langle\rho_{\mathbf{q}}\rangle$ gives an incoherent background. The solution to the coherent scattering is quite simple. We only have to replace the step-function scattering densities in Eqs. (2.1)–(2.3) by delta functions. The end result is that the $q_\parallel^{-2}$ factor in Eq. (2.4) drops out and ρ is redefined to be $\langle\rho_{\mathbf{q}}\rangle$ for the surface layer. This simple change applies to all the subsequent expressions. For example, Eq. (2.11) would have its prefactor $q_\parallel^{-2-2/\alpha}$ replaced by $q_\parallel^{-2/\alpha}$, Eqs. (3.14) and (3.16) would simply not have the $1/q_\parallel^2$ prefactor.

Another interesting variation on the same theme is to have step-function scattering density profile with infinitely strong amplitude. This "hard wall" model applies to atomic beam scattering by the surface,[23] because the atoms interact so strongly that they cannot penetrate the surface. The first Born approximation is clearly inadequate. This problem has been studied by Villain, Grempel, and Lapujoulade.[24]

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