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Two-scale Kirchhoff theory: comparison of experimental observations with theoretical prediction

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Abstract. We introduce a non-perturbative two-scale Kirchhoff theory, in the context of light scattering by a rough surface. This is a two-scale theory which considers the roughness both in the wavelength scale (small scale) and in scales much larger than the wavelength of the incident light (large scale). The theory can precisely explain the small peaks which appear at certain scattering angles. These peaks cannot be explained by one-scale theories. The theory was assessed by calculating the light scattering profiles using atomic force microscope (AFM) images, as well as surface profilometer scans of a rough surface, and comparing the results with experiments. The theory is in good agreement with the experimental results.

Keywords: new applications of statistical mechanics

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Contents

1. Introduction

Wave scattering by rough surfaces has been extensively studied both analytically and experimentally. For analytical approaches two methods have been generally considered: rigorous electromagnetic theory and approximate methods. The Kirchhoff theory is among the electromagnetic theories and is known as a 'tangent plane theory'. This theory is most widely used to calculate the distribution of the specular and diffuse parts of the reflected light. The Kirchhoff theory treats any point on a scattering surface as a part of an infinite plane, parallel to the local surface tangent. The theory is therefore exact for an infinite, smooth and planar scatterer, but is approximate for scatterers that are finite sized, non-planar or with rough surfaces [1]. Due to the computational limitations, most studies have been done for one-dimensional data of the surfaces. There are only a few cases of the analysis of two-dimensional surface data. One- and two-dimensional exact approaches have been successfully applied to dielectric, metallic or perfectly conducting surfaces [2, 3], deterministic surfaces [4, 5], dielectric films on a glass substrate [6] and dielectric films [7, 8]. Such exact calculations have been compared with experimental results and approximate models [6, 9]. Also some authors have studied wave scattering from random layers with rough interfaces [10, 11].

The joint probability density function (PDF) of surface slopes and heights $P(\partial_x h, h)$ is a key function in the estimation of the main parameters of wave scattering by a rough surface [12]–[16]. This is more obvious in a geometrical optics approach, when the angular distribution of the scattered power is proportional to the specular reflecting slope PDF. The slope PDF has also been introduced in [12, 15, 16] in the context of Bragg scattering. They have shown that the Bragg scattering results must be averaged by the proper slope PDF of the rough surface. This is also true for the estimation of the thermal emission from rough surfaces at small grazing angles [15, 16].

In the present paper, we introduce a non-perturbative two-scale Kirchhoff theory. The theory is applied to explain the small peaks observed in the scattering profile of a rough surface, at certain scattering angles. The theory employs the data obtained from the rough surface in two different scales. To check the theory we have measured the scattered light intensity as a function of the scattering angle, $I(\theta)$, using a setup consisting of a He–Ne laser (632.8 nm), a photo-multiplier tube (PMT) detector and a computer-controlled micro-stepper rotation stage. The resolution of the micro-stepper was 0.5 min. Alumina sheets were used as the rough samples. The surface topography of the alumina samples

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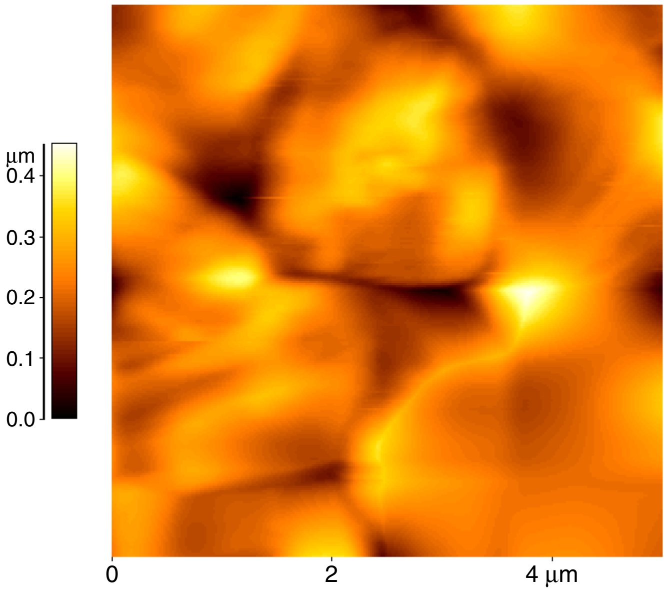

Figure 1. AFM image of the alumina surface in the length scale 5 μm $\times$ 5 μm (small scale).

in small scale (<5 μm) was obtained using an atomic force microscope (AFM) (Park Scientific Instruments). The images in small scale were collected in a constant force mode and digitized into 256×256 pixels. A commercial standard pyramidal Si_3N_4 tip was used. A variety of scans, each with size L, were recorded at random locations on the surface. The large-scale (<5 mm) morphology line scans of the alumina samples were recorded using a surface profilometer (Taylor Hobson). Figures 1 and 2 show typical AFM image and surface profile data with resolutions of about 20 nm and 0.25 μm, respectively.

2. Non-perturbative two-scale Kirchhoff theory

The Kirchhoff theory is based on three major assumptions [1]:

(a) The surface is observed from far field.

(b) The surface is regarded as flat, and the optical behaviour is locally identical at any given point on the surface. Therefore the Fresnel laws can be locally applied.

(c) The amplitude of the reflection coefficient, R_0, is independent of the position on the rough surface.

The field scattered by the rough surface, $\psi^{\mathrm{sc}}(r)$, is obtained by an integration over the mean reference plane S_{M} [1] (the geometry is displayed in figure 3),

$$\psi^{\mathrm{sc}}(r) = \frac{\mathrm{i}k \exp(\mathrm{i}kr)}{4\pi r} \int\int_{S_{\mathrm{M}}} \left(a\frac{\partial h}{\partial x_0} + b\frac{\partial h}{\partial y_0} - c \right) \exp(\mathrm{i}k(Ax_0 + By_0 + Ch(x_0, y_0)))\, \mathrm{d}x_0\, \mathrm{d}y_0 \tag{1}$$

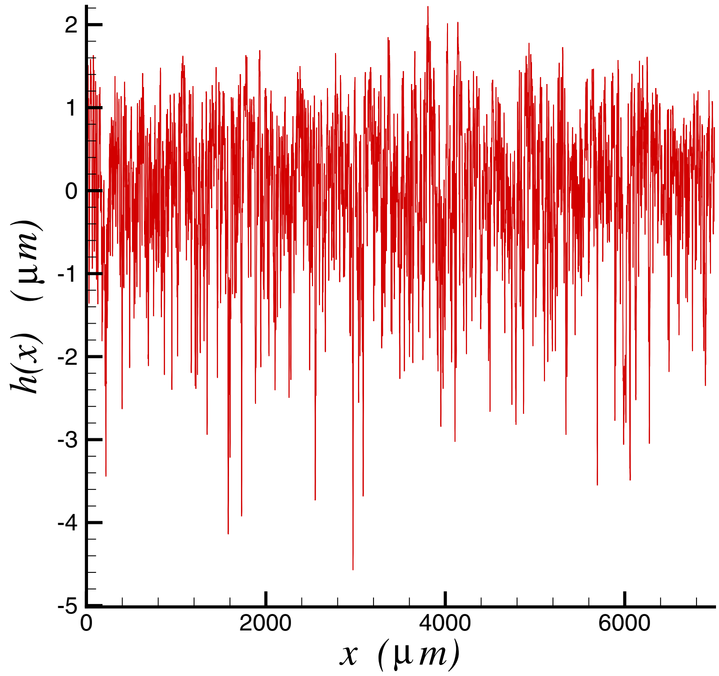

Figure 2. Profilometer scans of the alumina surface with resolution 0.25 μm (large scale).

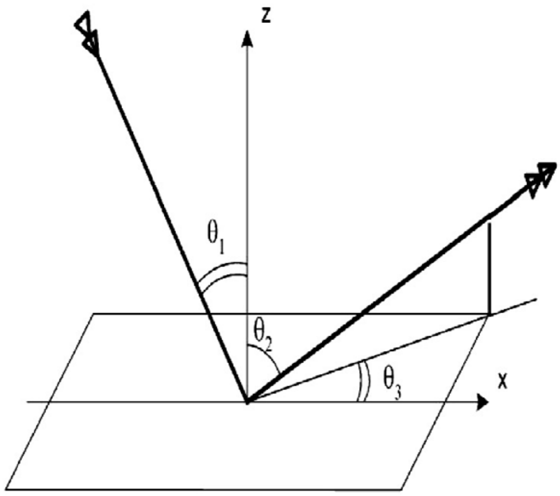

Figure 3. The geometry of the scattering angles θ_1, θ_2 and θ_3.

where

$$
\begin{aligned}
A &= \sin\theta_1 - \sin\theta_2\cos\theta_3,\\
B &= -\sin\theta_2\sin\theta_3,\\
C &= -(\cos\theta_1 + \cos\theta_2),\\
a &= \sin\theta_1(1 - R_0) + \sin\theta_2\cos\theta_3(1 + R_0),\\
b &= \sin\theta_2\sin\theta_3(1 + R_0),\\
c &= \cos\theta_2(1 + R_0) - \cos\theta_1(1 - R_0).
\end{aligned}
$$

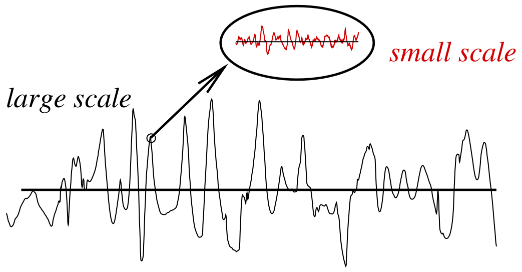

Figure 4. Two-scale observation model of alumina surface.

In the derivation of the equation (1), it is assumed that the incident wave $\psi^{\rm in}$ is a plane wave with a wavevector $\mathbf{k}$ as $\psi^{\rm in}(r) = \exp(\mathrm{i}\mathbf{k}\cdot\mathbf{r})$.

In most cases, the wave scattering models from rough surfaces implicitly assume that the surface is rough on a single scale. However, in practice all surfaces are rough on several scales, ranging from the atomic scale to the scale determined by the length of the surface. Nevertheless, only a finite range of scales is important in scattering of waves from a surface, i.e. the range covering the wavelength of the incident radiation. Models have been developed for describing surfaces that consist of high-frequency fluctuations superimposed on a slowly varying roughness [1]. These models use perturbation theories to describe the scattering from the high-frequency roughness, and this is modified in some manner by the low-frequency component [17]. All of the perturbative methods deal with the effect of the large-scale fluctuations as perturbation to the small-scale height fluctuations. Here, we intend to observe the surface in two scales with nanometre and micrometre resolutions. Figure 4 shows schematically the modulation of small-scale height fluctuations by large-scale variations. Various statistical parameters such as the joint height and height gradient PDF, surface roughness σ, correlation function $C(R)$, and correlation length τ, were measured in two scale.

In what follows, we are going to describe the non-perturbative two-scale Kirchhoff theory. We first calculate the contributions of the coherent and the diffuse fields by the Kirchhoff theory in small scale. The coherent field with a Gaussian height distribution will be [1]:

$$\langle\psi^{\rm sc}\rangle\langle\psi^{\rm sc}\rangle^* = I_0\exp(-g) \tag{2}$$

where $g = k^2\sigma^2C^2$. Also k, σ and I_0 are the norm of the wavevector, surface roughness in small scale and the scattered reflected intensity of the corresponding smooth surface. For isotropic surface and for samples with sizes much larger than the correlation length $L \gg \tau$ (and for a slightly rough surface i.e. $g \ll 1$), the diffuse field intensity for Gaussian height distribution will be given by [1]:

$$\langle I_{\rm d}\rangle = \frac{k^2F^2\tau^2}{4\pi r^2}g\exp(-g)A_{\rm M}\exp\left(-\frac{k^2(A^2+B^2)\tau^2}{4}\right) \tag{3}$$

where $F = \frac{1}{2}(Aa/C + Bb/C + C)$ and $A_{\rm M}$ is the effective area of the rough surface which experiences the incident radiation. Therefore, the overall scattered intensity is written as [1]:

$$\langle I\rangle = I_0\exp(-g) + \langle I_{\rm d}\rangle. \tag{4}$$

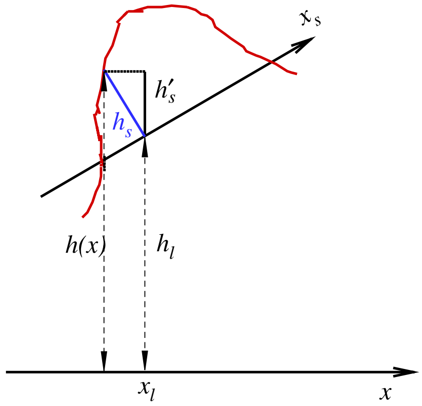

Figure 5. Definition of the height field in terms of h'_{s} and h_{l}. The field h'_{s} can be found from the field h_{s} via Euler matrices.

So far, we have expressed the results of the light scattering from the surface in small scale. Now we divide the whole surface into many small pieces (meshes), the length of which is smallest scale of our observation. In each mesh we can apply one-scale (small-scale) Kirchhoff theory. Therefore for each mesh we have a similar expression for the coherent field to equation (2), but with different angles which depend on the positions of the small mesh. In the small scale, we denote the height field in position x_{s} and y_{s} with h_{s}. Therefore, one can write the height field in any position, $\vec{x}$, as follows:

$$\begin{aligned} h &= h_{\mathrm{l}} + h'_{\mathrm{s}} \\ x &= x_{\mathrm{l}} + x'_{\mathrm{s}} \\ y &= y_{\mathrm{l}} + y'_{\mathrm{s}}. \end{aligned} \tag{5}$$

The subscripts l and s denote the large and small scales, respectively. The vector $(x'_{\mathrm{s}}, y'_{\mathrm{s}})$ is the position of h'_{s} in the small-scale coordinates. In figure 5 we have shown $(h'_{\mathrm{s}}, x'_{\mathrm{s}}, y'_{\mathrm{s}})$ and $(h_{\mathrm{s}}, x_{\mathrm{s}}, y_{\mathrm{s}})$, schematically. We note that the AFM images will give us $h_{\mathrm{s}}(x_{\mathrm{s}}, y_{\mathrm{s}})$ and via the large scale topography we will find $h_{\mathrm{l}}(x_{\mathrm{l}}, y_{\mathrm{l}})$. The vectors $(h'_{\mathrm{s}}, x'_{\mathrm{s}}, y'_{\mathrm{s}})$ and $(h_{\mathrm{s}}, x_{\mathrm{s}}, y_{\mathrm{s}})$ can be related to each other, via a rotational Euler matrix, with three rotational angles α, β and γ, i.e. $A(\alpha, \beta, \gamma) = R_h(\gamma) R_y(\beta) R_x(\alpha)$.

The local angles $(\theta_1, \theta_2, \theta_3)$ are defined by the average plane in the small scale. Therefore, all a, b, c, A, B, C are constant for all points within the small piece. In each small-scale element h_{l} is fixed so that $\partial h_{\mathrm{l}}/\partial x_{\mathrm{s}} = 0$. Hence, the total scattered field has the following expression:

$$\begin{aligned} \psi^{\mathrm{sc}}(r) &= \sum_{x_{\mathrm{l}}, y_{\mathrm{l}}} \Bigg[\frac{\mathrm{i}k \exp(\mathrm{i}kr)}{4\pi r} \int\int_{s_{\mathrm{M}}} \left(a_{\mathrm{s}} \frac{\partial h'_{\mathrm{s}}}{\partial x'_{\mathrm{s}}} + b_{\mathrm{s}} \frac{\partial h'_{\mathrm{s}}}{\partial y'_{\mathrm{s}}} - c_{\mathrm{s}} \right) \\ &\qquad \times \exp \mathrm{i}k(A_{\mathrm{s}} x'_{\mathrm{s}} + B_{\mathrm{s}} y'_{\mathrm{s}} + C_{\mathrm{s}}(h'_{\mathrm{s}}(x'_{\mathrm{s}}, y'_{\mathrm{s}}))) \, \mathrm{d}x'_{\mathrm{s}} \, \mathrm{d}y'_{\mathrm{s}} \Bigg] \exp \mathrm{i}k(A_{\mathrm{l}} x_{\mathrm{l}} + B_{\mathrm{l}} y_{\mathrm{l}} + C_{\mathrm{l}} h_{\mathrm{l}}) \\ &= \sum_{x_{\mathrm{l}}, y_{\mathrm{l}}} \psi^{\mathrm{sc}}_{\mathrm{s}}(r) \exp(\mathrm{i}k(A_{\mathrm{l}} x_{\mathrm{l}} + B_{\mathrm{l}} y_{\mathrm{l}} + C_{\mathrm{l}} h_{\mathrm{l}})). \end{aligned} \tag{6}$$

We note that $\partial h'/\partial x'_{\mathrm{s}} = \partial h/\partial x_{\mathrm{s}}$ and the summation is over the small-scale samples modulated by the large-scale fluctuations. If we assume that the joint PDF of heights and its slope of two scales are independent, then the average of the field scattered in any direction will be given by:

$$\begin{aligned}\langle\psi_{-e}^{\mathrm{sc}}(r)\rangle &= N\sum_{h_l,\partial_x h_l}\sum_{x_l,y_l}\langle\psi_{\mathrm{s}}^{\mathrm{sc}}(r)\rangle\exp(\mathrm{i}kC_l h_l)\exp(\mathrm{i}k(A_l x_l+B_l y_l))P(h_l,\partial_x h_l)\\ &= N\sum_{h_l,\partial_x h_l}\langle\psi_{\mathrm{s}}^{\mathrm{sc}}(r)\rangle\exp(\mathrm{i}k(C_l h_l))P(h_l,\partial_x h_l)\sum_{x_l,y_l}\exp((\mathrm{i}k(A_l x_l+B_l y_l)))\\ &= N\frac{\sin(kL_x)}{kL_x}\frac{\sin(kL_y)}{kL_y}A_{\mathrm{M}}\sum_{h_l,\partial_x h_l}\langle\psi_{\mathrm{s}}^{\mathrm{sc}}(r)\rangle\exp(\mathrm{i}kC_l h_l)P(h_l,\partial_x h_l).\end{aligned} \tag{7}$$

The subscript $(-e)$ denotes scattering from the surface without the edge terms. The rough surface has been assumed to be rectangular with extent $-X \le x_0 \le X, -Y \le y_0 \le Y$. Also L_x and L_y are length scales in the scattering area (the effective area of light incidence), and $S_{\mathrm{M}} = (\sin(kL_x)/kL_x)(\sin(kL_y)/kL_y)A_{\mathrm{M}}$ is the constant term in all observation angles. The quantity $NP(h_l,\partial_x h_l)$ is the number of points with height h and slope $\partial_x h_l$. It is noted that for a homogeneous surface $p(h,x)$ is independent of position along the surface, x. In order to perform analytical calculations, it is necessary to assume that the edge effects are non-stochastic, i.e. $\langle\psi_e\rangle = \psi_e$ [1]. Based on this assumption, the coherent part becomes:

$$\langle I_{\mathrm{coh}}\rangle = \langle\psi^{\mathrm{sc}}\rangle\langle\psi^{\mathrm{sc}}\rangle^* = N^2\sum_{h_1,\partial_x h_1,h_2,\partial_x h_2}|\langle\psi_{\mathrm{s}}^{\mathrm{sc}}\rangle|^2 P(h_1,\partial_x h_1)P(h_2,\partial_x h_2)\exp(\mathrm{i}k(C(h_2-h_1))) \tag{8}$$

where $\psi^{\mathrm{sc}} = \psi_e + \psi_{-e}$. It is noted the non-stochastic assumption of the edge effect leads to the cancellation of all terms containing edge effects. In cylindrical coordinates, for an isotropic surface, the substitutions $x_2 - x_1 = R\cos\theta$ and $y_2 - y_1 = R\sin\theta$ can be made. Since the heights PDF and the heights difference PDF are independent (we will confirm this assumption in the next section), i.e. $P(h_1,\partial_x h_1) = P(h_1)P(\partial_x h_1)$, and defining

$$\sum_{h_1,h_2}\mathrm{d}h_1\,\mathrm{d}h_2\exp(\mathrm{i}k(C(h_2-h_1)))P(h_1)P(h_2) = \chi(kC,-kC,R)$$

we then find:

$$\langle I_{\mathrm{coh}}\rangle = S_{\mathrm{M}}^2\left|\sum_{\partial_x h_1}NP(\partial_x h_1)\langle\psi_{\mathrm{s}}^{\mathrm{sc}}\rangle\right|^2\chi(kC,-kC,R). \tag{9}$$

It is known that the total average scattered field in small scale is $\langle\psi_{\mathrm{s}}^{\mathrm{sc}}\rangle = \chi(kC_{\mathrm{s}})\psi_0^{\mathrm{sc}}$.

For a Gaussian height distribution, the one- and two-dimensional characteristic function is given by:

$$\chi(kC_{\mathrm{s}}) = \frac{1}{\sigma_{\mathrm{s}}\sqrt{2\pi}}\int_{-\infty}^{+\infty}\exp\left(-\frac{h^2}{2\sigma_{\mathrm{s}}^2}\right)\exp(\mathrm{i}kC_{\mathrm{s}}h_{\mathrm{s}})\,\mathrm{d}h_{\mathrm{s}} = \exp(-k^2C_{\mathrm{s}}^2\sigma_{\mathrm{s}}^2/2), \tag{10}$$

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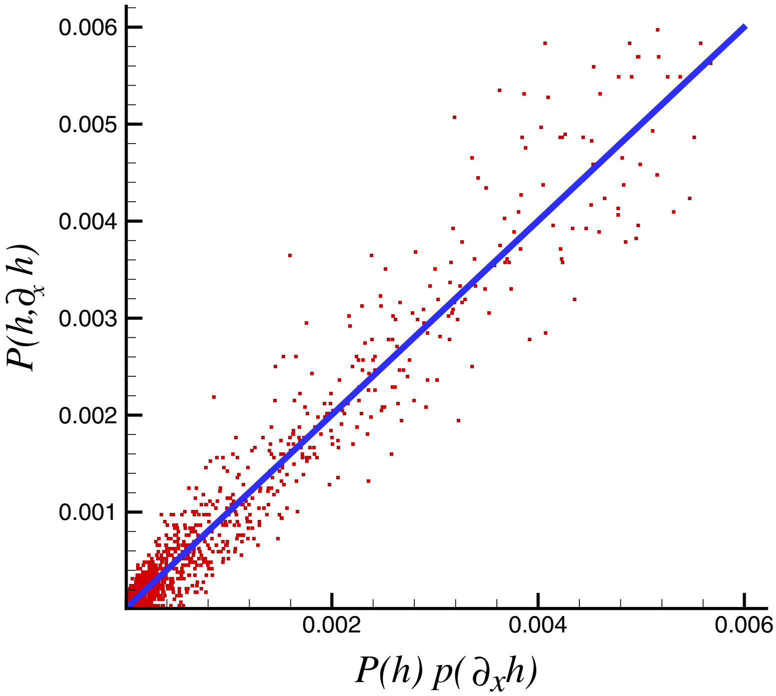

Figure 6. Joint PDF versus $P(h)P(\partial_x h)$, that shows that the height and slope PDFs are almost independent.

and

$$\chi(kC, -kC, R) = \exp(-k^2 C^2 \sigma_1^2 (1 - C(R))), \tag{11}$$

where $C(R) = \langle h(r)h(r+R)\rangle/\sigma_1^2$ is the surface correlation function in the large scale. Also the average of total intensity are given by:

$$\begin{aligned}\langle I_{\mathrm{tot}}\rangle &= \langle \psi^{\mathrm{sc}}\psi^{\mathrm{sc}^*}\rangle \\ &= N\sum_{h_1,\partial_x h_1}\sum_{x_0,y_0}\sum_{x_1,y_1} P(h_1,\partial_x h_1)\langle \psi_{\mathrm{s}}^{\mathrm{sc}}\psi_{\mathrm{s}}^{\mathrm{sc}^*}\rangle \exp(\mathrm{i}k(A(x_2-x_1)+B(y_2-y_1))).\end{aligned} \tag{12}$$

Performing the summation, we find $\sum_{h_1} = N$, where N is the number of points on the surface. So, the average total intensity becomes:

$$\langle I_{\mathrm{tot}}\rangle = S_{\mathrm{M}}^2 N \sum_{\partial_x h_1} N(\partial_x h_1)\langle \psi_{\mathrm{s}}^{\mathrm{sc}}\psi_{\mathrm{s}}^{\mathrm{sc}^*}\rangle. \tag{13}$$

Finally, the diffuse field intensity is obtained as:

$$\langle I_{\mathrm{d}}\rangle = \langle I_{\mathrm{tot}}\rangle - \langle I_{\mathrm{coh}}\rangle. \tag{14}$$

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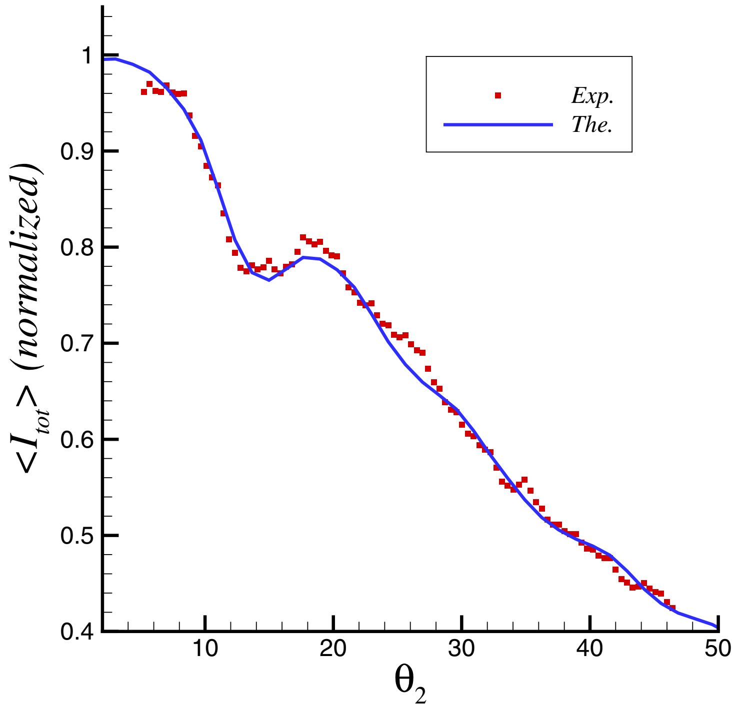

Figure 7. Comparison of theoretical prediction via two-scale Kirchhoff theory and experimental results for scattered field (bold symbols).

3. Comparison with experiments

Here we test the non-perturbative two-scale Kirchhoff theory with experiment. For this purpose, we obtain the height profile of alumina sheets as the rough samples, using the profilometer in large scale and the AFM images in small scales. Indeed we intend to observe the surface in two scales: nanometre and micrometre. To use the two-scale theory the surface must possess two conditions. First, the PDF of the height and its slope must be independent at small and large scales, i.e. $P(h_\mathrm{l}, \partial_x h_\mathrm{l}, h_\mathrm{s}, \partial_x h_\mathrm{s}) = P(h_\mathrm{l}, \partial_x h_\mathrm{l}) P(h_\mathrm{s}, \partial_x h_\mathrm{s})$. The homogeneous rough surfaces possess this condition. Indeed the statistical parameters in small scale (roughness, exponents, etc) are similar at any point of the sample (large scale). This means that the two PDFs are independent. The second condition is that the height and height gradient fluctuation must be independent in the large scale. This means that the joint PDF of the height and height gradients can be decomposed as $P(h_\mathrm{l}, \partial_x h_\mathrm{l}) = P(h_\mathrm{l}) P(\partial_x h_\mathrm{l})$. This assumption needs confirmation. In figure 6, we have plotted the joint PDF $P(h_\mathrm{l}, \partial_x h_\mathrm{l})$ versus $P(h_\mathrm{l}) P(\partial_x h_\mathrm{l})$. It is obvious that the joint PDF versus multiplication of single PDFs fits with a line with slope one. Considering its statistical error we observe that the height and height gradient PDFs are independent. For large values of h and $\partial_x h$, our assumption becomes poor, and thus uncertainty increases.

To compare the experimental observation with those of the theoretical prediction, we need to estimate the several types of PDFs in small and large scales. In equation (8), we need to evaluate the quantity $\langle \psi_\mathrm{s}^{\mathrm{sc}} \psi_\mathrm{s}^{\mathrm{sc}^*} \rangle$ in the small scale and the PDF of the height gradients in the large scale. To evaluate the intensity $\langle \psi_\mathrm{s}^{\mathrm{sc}} \psi_\mathrm{s}^{\mathrm{sc}^*} \rangle$, we have to use

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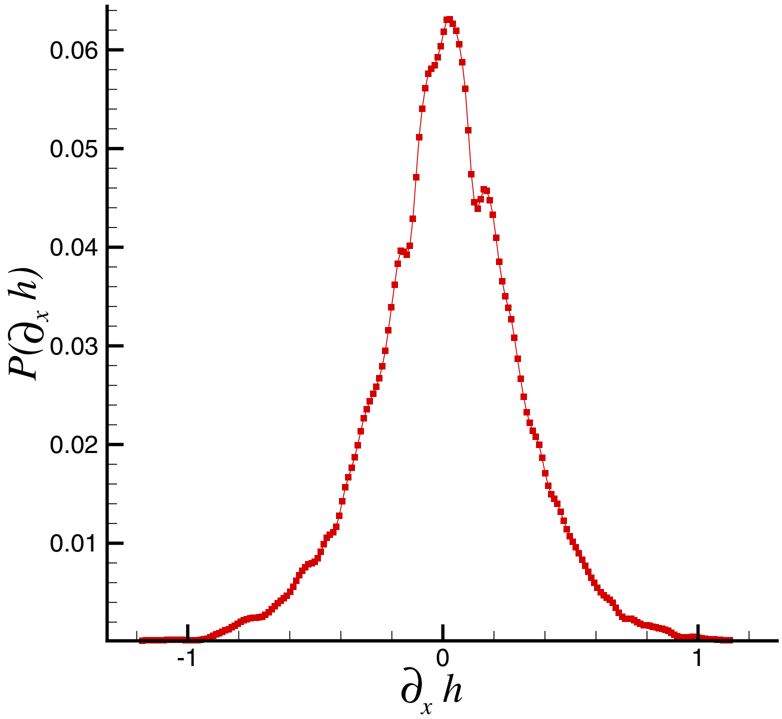

Figure 8. The PDF of height gradient in large scale.

equation (2), where the averaging is done in the small scale. Therefore we need the PDF of height fluctuation in the small scale. Also we need other statistical quantities, such as the surface roughness σ, correlation function $C(R)$, correlation length τ, in small and large scales. We evaluate the height–height correlation function $\langle h(x+R)h(x)\rangle$ versus radial distance R for large-scale fluctuations. We find the following expressions for the alumina surface: $C(R) = 2.14\exp(-R^2/608)$ and $1.27\exp(-0.58R)$, for small and large scales, respectively. Also the roughness exponent, variance and scaling length for the small (large) scale have been found as 0.85 $(0.85, 031)$, 0.31 μm (1.33 μm) and 1.5 μm (19.4 μm), respectively. It is found that the height PDFs in the two scales are almost Gaussian. Indeed we checked that the fourth moment is related to the second moment via $\langle(h-\bar{h})^4\rangle \simeq 3\langle(h-\bar{h})^2\rangle^2$. The equality holds for the Gaussian height fields. The estimated statistical quantities enable us to predict the average total intensity. In figure 7, we have plotted the experimental observation and theoretical prediction of total intensity. It is evident that the theoretical prediction fits with the experimental observation. We observe that the theory is able to predict the small peak at angle $\simeq 18°$ in the variation of the total intensity versus angle scale θ_2. We note that if one plots the PDF of the height gradient, one finds that the PDF also has small peaks at angle scale $\tan^{-1}(\partial_x h) \simeq 9°$. This means that the gradient PDF is responsible to have a small peak in the variation of the total intensity in terms of angle scale (we note that the slope $\alpha = \tan^{-1}(\partial_x h)$ produces 2α contribution in the reflection of the light from the surface). In figure 8, the behaviour of the slope PDF $(\partial_{x_l} h_l)$ in terms of $\partial_{x_l} h_l$ has been given. Also, as shown in figure 7, the two-scale Kirchhoff theory is able to predict the small peak in the variation of the total intensity in terms of angle scale. As we observe, there are other peaks in figure 7, where the theory cannot predict the peaks for large angle scales. Indeed for these angle scales we should take into account the shadowing effect [15, 16]. In [12], the validity range of geometrical shadow functions has been investigated for a randomly rough surface for which the shadowed Kirchhoff approximation has been shown to give good results for the scattered intensity distribution. We will discuss the modification of the two-scale Kirchhoff theory by the shadowing effect elsewhere.

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